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Sr^{90} MONITORING AT THE SAVANNAH RIVER PLANT

by

F. E. Butler

Health Physics Section
Savannah River Plant

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ABSTRACT

Radioassays of milk, soil, grass, and other material showed no detectable Sr^{90} due to Savannah River Plant wastes. These assays indicated that the Sr^{90} found in this geographical area (25-mile radius of SRP) was nuclear weapons debris. Sr^{90} was separated from milk by absorption on cation exchange resin. An extraction technique was used in assaying Sr^{90} in all types of samples. Cs^{137}/Sr^{90} ratios were found to be constant in soil samples. This indicates that Sr^{90} in soils may be estimated by the simpler Cs^{137} analysis.

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Sr^{90} MONITORING AT THE SAVANNAH RIVER PLANT

INTRODUCTION

Two hundred and six nuclear explosions were reported between 1945 and October 1958. Fallout debris from these explosions resulted in widespread contamination of soil, water, and biological materials. The heaviest deposition occurred in the Northern Hemisphere,(1-4) particularly in North America.

Sr^{90} is one of the most hazardous fission products because it is a bone-seeker with a long biological half-life. The amount of this nuclide in soil and milk doubled between 1957 and 1959 in many locations. Most authorities agree that the amount of Sr^{90} in foods is still far below that considered harmful.(5,6) The most pessimistic case is milk, which recently increased in Sr^{90} content, at some locations, to 60% of the 33 μμc/ℓ level set as the maximum permissible amount by the National Committee on Radiation Protection and Measurements.

Worldwide Sr^{90} monitoring was undertaken by a number of laboratories during the past decade. Analysis is complicated because Sr^{90} must be separated from large quantities of chemically similar calcium contained in most samples.

Methods of analysis formerly differed at each laboratory, but most methods were standardized by the World Health Organization in 1958. Recently, smaller samples have been assayed due to increased levels of Sr^{90}. Most topsoils and milks may now be assayed accurately if 500-gram samples are used.

The study at the Savannah River Plant was initiated to determine whether Plant wastes contributed measurably to Sr^{90} contamination found in this geographical area (25-mile Plant radius) or whether this contamination was attributed solely to fallout from weapons tests. Procedures developed during this work, as well as data accumulated, contribute to the general fund of knowledge concerning Sr^{90} monitoring.

SUMMARY

Milks, soils, grasses, and other material collected during 1958 and 1959 were analyzed for Sr^{90}. Data indicate that there is no more Sr^{90} on or near the Savannah River Plant than there is at other locations in the southeastern United States.

Analytical procedures were developed that improve on those in routine use at other locations. These improvements are:

Sr^{90} in milk is absorbed on cation resin and eluted from the resin with acid. This procedure avoids lengthy evaporation and ashing of whole milk.

The final Sr^{90} assay of all types of samples is obtained by separating Y^{90} from dilute acid solution by di-alkyl phosphoric acid extraction. Carrier-free Y^{90} is counted; this method displays good decontamination factors for all other nuclides.

These procedures offer good precision and accuracy (or tracer recovery) along with reduction in the number of procedural steps.

Local milk samples analyzed from April 1958 to September 1959 were comparable in Sr^{90} content to those samples obtained from Atlanta milk by the U. S. Department of Public Health, Education and Welfare. An abrupt increase of Sr^{90} in all types of samples in the Spring of 1959 was apparently the result of increased rainout due to the 1958 weapons tests carried out by the USSR.

A preliminary study indicates that milk can be assayed within a work day, prior to its public consumption as fresh milk.

Soils collected in October 1958, in the Plant vicinity (25-mile radius) contained Sr^{90} at the same level as reported by the Health and Safety Laboratory (HASL) of the Atomic Energy Commission for October collections of soils from Atlanta, Georgia and Jacksonville, Florida. Local soils resampled in June 1959 contained less Sr^{90} than the October 1958 soils.

A study of Sr^{90} penetration in local soils was made at four locations. These samples, and samples taken from 66 other locations within a three-day period, show a variable Sr^{90} content. There is also a definite correlation between calcium and Sr^{90} contents of soils; this indicates that Sr^{90} retention is in proportion to calcium retention.

Cs^{137} and Sr^{90} results are compared in samples of milk, soil, and grass. Only soil contains a constant ratio. The Cs^{137}/Sr^{90} ratio of 2.0 ±0.3 in soil indicates that Sr^{90} may be estimated by the simpler Cs^{137} analysis.

DISCUSSION

BACKGROUND INFORMATION

The importance of fallout analyses and the interpretation of results are indicated by the Congressional hearings conducted in 1957 and 1959.(7,8) Most of the leading scientists in this field testified; a complete summary of fallout data was compiled.

Data presented at these hearings as well as other information published recently is summarized.(1-6,8,10)

The level of fallout has continued to rise as the result of bomb tests with increased amounts of Sr^{90} having been detected in air, rain, soil, milk, and human bones.

Stratospheric residence time is now believed to be 1 to 3 years, rather than 6 years. Increases in fallout, particularly in 1959, were due to short stratospheric residence of debris introduced in the North Temperate Zone.

The majority of fallout occurred in latitudes 40 to 50° North.

Methods of analyses have improved although they are still too complex for the desired worldwide coverage of fallout. Some methods have now been standardized.

A greater emphasis has been placed on fundamental studies. Methods for purging Sr^{90} from soil and milk have been considered.

Milk as a source for the uptake of Sr^{90} has been the subject of many studies. The importance of these studies is reflected by the following:

Milk is one of the chief sources of body calcium.

Sr^{90} is chemically similar to calcium.

Milk is an especially critical source of Sr^{90} in the formation of the bones of children.

Milk is a universal food that is produced throughout the year.

Worldwide bone studies were made at Columbia University; the most recent study is one concerning Sr^{90} analyzed in 1958.(4) It was determined that bones of children under five years of age contain about 10 times as much of this fission product as adult bones.

Future assays are expected to show a rise in Sr^{90} content in bones, due to increases of Sr^{90} in milk and other foods during 1957 to 1959. Sr^{90} results in soil, grass, milk, and other samples, which contribute toward bone assimilation, are reported in "sunshine units" (micromicrocuries of Sr^{90}/grams of calcium). This method of reporting is convenient, since the samples contain large amounts of chemically similar calcium, which is easily analyzed. Kornberg(11) has summarized the objections to indiscriminate use of the Sr/Ca ratios for assessing the potential hazards of Sr^{90}.

Experiments at Hanford and the University of California have demonstrated that assimilations of calcium (Ca^{45}) and strontium (Sr^{89}, Sr^{90}) are not proportional to the contents of the nuclides in animal diets.(12-16) Bone depositions are influenced by a number of factors: (1) chemical states, (2) relative amounts of phosphates and other bone nutrients, and (3) metabolism. Twentyfold increases in calcium or phosphate in rat diet are required to reduce Sr^{89} bone deposition by 50%.(16) In one study(12), an increase in calcium in milk actually promoted Sr^{90} uptake in bones.

Several recent studies concerned the mechanism of Sr^{90} fixation in human bones.(17-19) It was determined that Sr^{90} eventually becomes incorporated within bone by attachment or exchange with calcium. The mechanism for calcium phosphate formation is reported to be so specific that strontium cannot replace calcium in the formation of nuclei from which bone and tooth crystals grow.(19) Organic chelating agents will remove Sr^{90} from bones prior to its incorporation within the bone.(19)

Selective removal of Sr^{90} from milk has been accomplished in recent work at the University of Tennessee.(20) Calcium-based resins used in a laboratory-scale ion exchange removed 94% of strontium from skimmed milk samples.

Soil discrimination factors have been studied, particularly with regard to the calcium and Sr^{90} passage from soil to vegetation.(11,21,22) Addition of several tons of calcium per acre of soil reduces Sr^{90} in vegetation by a negligible amount. Plants grown in soils containing extremely small amounts of calcium contain more Sr^{90}.(21)

Analyses were developed by each group engaged in Sr^{90} work.(23-25) In September 1958, the World Health Organization met to review existing analytical procedures; as a result procedures used at Atomic Energy Research Establishment(26) (AERE) and at the Health and Safety Laboratory of the N. Y. Office of the AEC(24) (HASL) were adopted as standard methods.(9)

The initial purpose of soil analysis at SRP was to compare soils in the vicinity of the Plant by a procedure which would yield accurate, reproducible data. The procedure used by the Sunshine Group(23), with some modifications, was found to be adequate for detection of Sr^{90} in Plant wastes. Since most soils in this area are sandy loams, the ammonium acetate (available) leach method was used to remove most of the Sr^{90} and calcium. Several soils were reassayed for Sr^{90} content by the hydrochloric acid (total) extraction method, in order to compare results from this area with those obtained at other locations.

Procedures are described in detail in the Appendix. Major departures from the recommended methods are found in the milk separation procedure and in the final Sr^{90} determination. The accuracy of each result is determined by recovery of $Sr-Y^{90}$ tracer added to identical samples.

To compare results with those obtained by other methods of analysis and to assure accuracy, the following controls were maintained:

Interlaboratory sample results were compared (Table IV).

Duplicate samples, tracers, and reagent blanks were run.

All samples were recounted to determine Y^{90} purity.

The low background counter was calibrated frequently. Overnight and weekend backgrounds were determined.

MILK

SAMPLING

Milk samples were collected singly and in groups over the period from April 1958 to September 1959. Most of the samples were obtained in the immediate vicinity of SRP as shown in Figure 1. One group of milk samples was collected in five southeastern states during May 1958. Fresh milk and raw milk from small dairies as well as homogenized and skimmed milk from larger dairies were collected in gallon containers. Several condensed and powdered samples were also diluted to whole milk consistency and analyzed.

EXPERIMENTAL

Methods used for separation of Sr^{90} and calcium from milk have involved evaporation and ashing of milk samples.(9,23,24,26) This treatment generally requires about two days, although a rapid ashing technique is described by Campbell.(27) The ash is dissolved in dilute acid prior to strontium or Y^{90} precipitation.(23) An intermediate phosphate precipitation is performed in one procedure.(26) In all procedures, the chemical yield is determined by the recovery of a stable strontium carrier which is added to the milk or to the first acid solution of ash.

Nervik, Kalkstein, and Libby(28) report that nearly 90% of the calcium and strontium from milk is removed by treatment with "Dowex" 50W resin in the sodium form. In initial experiments at SRP batches of "Dowex" 50W resin were added to milk containing $Sr-Y^{90}$ traces. After the milk was adjusted to pH 6 to 7 and stirred, greater than 90% of the tracer was recovered in the resin.

Further testing resulted in removal of 99.1 and 99.6% of Sr^{85} tracer from two 500-ml samples of milk after 30-minute batch stirrings with 15 grams of resin. The resin was allowed to settle and 5-ml aliquots of milk were counted in a gamma well counter.

Dairy No.	Sample Date	Background Count on Original Milk, c/m	Sr^{85} Tracer Added Before Resin Treatment, c/m	Sr^{85} Tracer Added After Resin Treatment, c/m
1	4/7/58	240	3740	271
2	4/7/58	244	3666	257

The procedure for separating Sr^{90} from milk is described in detail in Appendix II. Briefly, the procedure includes the following steps:

After adjustment of the pH to 5 to 7, 500-ml aliquots of milk are stirred with 30 grams of pretreated resin.

The milk is discarded and the resin is washed with distilled water, which is also discarded.

Sr^{90} is eluted from the resin with 8N HNO_3.

The acid is evaporated and the milk residues are dissolved in 0.08N HCl.

The procedure in Appendix III is followed for final Sr^{90} determination.

The entire procedure requires about two weeks, including Y^{90} buildup time.

Experiments were also made to determine a quick method for Sr^{90} determination. After the initial 20% HDEHP (di-2-ethylhexyl-phosphoric acid) extraction of the 0.08N HCl solution was completed, as described in Appendix III, Y^{90} was back-extracted with acid and counted.

Cs^{137} was determined in aliquots of dilute HCl solutions prepared for Sr^{90} analysis. The procedure is described in Appendix III. To test for interference of K^{40}, one gram of potassium was added to several milk samples prior to analysis. No increase in counting rate was detected after Cs^{137} analysis.

CONCLUSIONS

All regional milk samples analyzed over the eighteen-month period are listed in Table I.

Over-all tracer recovery for forty samples was 85.2%, with a standard deviation of 10.3%; on the average, 38 duplicate milk samples agreed within ±8% (1.3 d/m of Sr^{90}). This precision was good for both raw and homogenized milk samples.

Table IV summarizes interlaboratory comparisons of milk samples (method at other locations included evaporation and ashing of milk). Comparisons were good, even for milks with relatively low Sr^{90} content.

In the following table samples are grouped to show the rising trend in Sr^{90} and for comparison with results from other cities. Although milk samples are analyzed at several laboratories, only U. S. Department of Health, Education, and Welfare (USDHEW) results are shown for comparison.

			USDHEW Milk Samples(a)	
Sample Period	No. of SRP Milk Samples	Sr^{90}, $\mu\mu c/\ell$	Location	Sr^{90}, $\mu\mu c/\ell$
4/58	4	13.0	St. Louis	12.3
5/58(b)	10	15.1	Atlanta	13.7
9/58, 10/58	11	12.0	Atlanta	13.8
4/59	5	21.1	Atlanta	20.8

(a) Monthly summaries published by the U. S. Department of Health, Education, and Welfare, Cincinnati, Ohio. These results have also appeared in the Health and Safety Quarterly Summaries.(6) Atlanta milk was not analyzed in April 1958.

(b) May 1958 samples were collected from five southeastern states.

Most of the SRP samples in these four groups are from large dairies and are in good agreement with the single monthly composite sample collected in Atlanta, Georgia and assayed at Cincinnati, Ohio.

Unfortunately, it was not possible to resample the same milk source each time during the period from April 1958 to September 1959. Several locations were sampled twice or more and were compared for Sr^{90} increase as follows:

Milk Resamples, Sr^{90}, $\mu\mu c/\ell$

	Sample Date										
Dairy No.(a)	4/58	5/58	10/58	11/58	12/58	1/59	2/59	4/59	6/59	7/59	8/59
1 & 2(b)	13.0	18.0		15.5				22.8			
21					13.9	14.3	15.1		17.3	23.5	
20			19.1	20.5					32.4		24.1
14			10.0						27.8	22.7	
18			10.6						37.1	22.4	

(a) See Table I for location of dairy

(b) Dairies exceeding 50 cows

The highest Atlanta result was 22.8 μμc of Sr^{90}/ℓ in May 1959 and was followed by gradual monthly decreases in Sr^{90}; local milk samples were highest in May through June 1959. Brogdon, Talatha, and Snelling milk samples (from one-cow dairies) were high in both Sr^{90} and calcium content.

The abrupt increases of Sr^{90} in all milk samples during May through June is explained by increased rainout of the nuclide observed in the spring of 1959.(1) This rainout was caused by short stratospheric holdup of October 1958 Russian bomb debris. Temporary increases of Sr^{90} in grass, and thus in milk, may have been due to a stem-base absorption of Sr^{90}.(37) The mechanism postulates temporary holdup of fallout in surface roots and organic matter around the base of grass stems and, therefore, makes Sr^{90} contamination available to the grass through normal metabolic processes.

Sr^{90} determined by the rapid assay procedure was comparable to the final results obtained by standard analysis.

Milk Sample	Sr^{90}, μμc/ℓ Rapid Assay	 Standard Assay
Dairy No. 20 sampled 7/59	23.5	24.1
USDHEW	12.8	11.5
USDHEW	4.6	4.5

Samples were counted several times and Y^{90} decay was established. One uncertainty in a rapid assay on fresh milk is the possibility that complete equilibrium of Sr^{90} and Y^{90} has not been reached. The abbreviated milk assay procedure is useful if an analysis were required prior to public consumption. Samples exceeding the maximum permissible limit of 33 μμc of Sr^{90}/ℓ can be analyzed in smaller volumes, thereby reducing acid evaporation time. The analysis can be performed in 4 to 6 hours, using 200-ml milk aliquots.

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The Cs^{137}/Sr^{90} ratios shown in Table I average 8.2 with maximum and minimum ratios of 16.2 and 3.2. These results are in agreement with those found at USDHEW. Since the ratio is not constant, Sr^{90} cannot be predicted by the simpler analysis of Cs^{137}.

Harley, in dietary Sr^{90} estimates for the United States(6), reports an average per capita milk diet of 233 quarts during 1958, representing an average intake of 1860 μμc of Sr^{90}. Milk samples shown on Table I for the SRP area, during the twelve months ending August 1, 1959, contained an average of 16.5 μμc of Sr^{90}/ℓ. This represents, for comparision, the intake of 3658 μμc of Sr^{90} per person. The sources of milk in this group are: (1) 60% from larger dairies; (2) 23% from one-cow dairies; and (3) 17% powdered or condensed milk. Brogdon milk was omitted from this group. For further comparison, the maximum permissible amount of Sr^{90} in milk is 33 μμc/ℓ. This is based on the recommendation of the National Committee on Radiation Protection and Measurements and amounts to 7336 μμc of Sr^{90} intake per person each year over a lifetime. Local milk samples during the twelve-month period contained 50% of the MPA for Sr^{90}.

SOIL AND GRASS

SAMPLING

Soil and grass samples were collected over a 2000-square-mile area within a three-day period in October 1958. The sampling procedure is described in Appendix I. Samples were taken on four perimeters at increasing distances from the Plant Separations Areas as shown in Figure 1.

Perimeter	Number of Sample Locations	Avg. Distance from Separations Areas	Avg. Distance Between Samples
F and H	24	1 mile	0.5 mile
Inner	14	6 miles	2 miles
Outer	14	10 miles	4 miles
25-mile	14	25 miles	11 miles

Soil samples were collected at 0 to 2 and 2 to 4-inch depths at each of the 66 sample locations.

In June 1959, soils were resampled on the outer perimeter at depths of 0 to 2 inches and 2 to 6 inches respectively. To determine Sr^{90} penetration at four locations, 1-inch soil layers were collected down to the 6-inch level, followed by a 6 to 12-inch sample.

All of the soil samples were acidic and over 80% were classified as the sandy loam variety. Soils from the outer perimeter varied most, with 5 of 14 soil samples ranging from fine sand to silt loam. All of the inner perimeter soils were sandy loam.

EXPERIMENTAL

Ammonium Acetate Extraction of Soil. This type of extraction removes the loosely bound calcium and strontium considered to be available for plant growth. The procedure has been used, with variations, at other laboratories.(23,24,25) Procedures used at SRP and Hanford(29) were adopted from the Sunshine Method(23) and are similar.

The "available" procedure is described in detail in Appendix II. The general steps are as follows:

1. Stir 500 grams of dry soil with 1N ammonium acetate and allow to settle overnight.

2. Filter mixture and wash soil with additional extraction solution.

3. Evaporate and oxidize filtrate with HNO_3 and H_2O_2.

4. After removal of iron by hydroxide scavenging, dissolve residual solids in dilute HCl prior to analysis for Sr^{90} and calcium.

Precision and accuracy obtained by this method are demonstrated by the results from 17 assays on a composite inner perimeter sample.

Analysis	Number of Assays	Results
Calcium	17	0.118 ±0.005 g/kg
Sr^{90}	9	29.7 d/m/aliquot (Maximum = 30.7, minimum = 28.8)
Tracer Sr^{90}	8	78.2 ±5.0% recovery

Hydrochloric Acid Extraction of Soil. HCl extraction of soil is one of several methods used for removal of total calcium and strontium. This procedure is applicable for measuring accumulated Sr^{90} from fallout, including the portion which is complexed to a greater degree or absorbed within soil particles. Other methods of complete removal of Sr^{90} include the heating of soil in a muffle furnace, followed by sodium carbonate or sodium hydroxide fusion,(9) and electrolytic separation.(23,30)

The "total" procedure used at SRP was patterned after that used at HASL(24) and is described in Appendix II. The general steps are as follows:

1. Stir 300 g of dry soil with 1:1 HCl and allow to settle overnight.

2. Remove acid by filtration and wash soil with additional acid.

3. Precipitate calcium and Sr^{90} from the acid extract as oxalates.

4. Reduce the oxalates to oxides by heating in a muffle furnace and dissolve in dilute HCl prior to analysis for Sr^{90} and calcium.

Precision and accuracy were comparable to the "available" method. To determine whether the extraction of Sr^{90} and calcium was complete, six soils were re-extracted with hot 1:1 HCl. Calcium carrier was added to the filtrate prior to oxalate precipitation. On analysis no additional calcium or Sr^{90} could be detected.

Additional Sr^{90} was recovered from the original extracts of the above soils. After addition of calcium carrier, and oxalate scavenging, 10 to 20% more Sr^{90} was coprecipitated. The incomplete Sr^{90} coprecipitation was due to the relatively low calcium content in soils in this area. More than one oxalate coprecipitation is normally required for complete recovery of strontium from solutions.(31)

Cs^{137} determinations were made on aliquots of the 1:1 HCl filtrate, following the procedure in Appendix III. Tracer recoveries for Cs^{137} were only 40 to 50%, but precision obtained for duplicate samples was good. Purity of all precipitates was tested by gamma pulse height analysis. The only peak detected was that of Cs^{137}.

Preparation of Grass Samples. Grass samples collected in October 1958 were prepared according to Appendix II:

1. Fifty grams of grass was fired in a muffle furnace in a "Vycor" beaker.
2. The ash was refluxed with HNO_3 three times.
3. The HNO_3 was evaporated and the residues were dissolved in dilute HCl prior to Sr^{90} and calcium analysis.

The precision and tracer recovery were poor for grass samples compared to soil analyses, although the average assay for each perimeter was considered to be reliable.

CONCLUSIONS

The fourteen 0 to 2-inch soils collected on the inner perimeter (Figure 1) in October 1958 were analyzed by two methods of soil leaching. These results are shown in Table II.

Duplicate results by the ammonium acetate "available" method are shown for each location. On the average, duplicates agreed within 7% or 2.0 d/m per 180 grams of soil. After correction for recovery of tracer samples, the results are shown in Table II as μμc Sr^{90} per kg of soil. The average of the 14 results was 93 μμc Sr^{90} per kg, compared with 95 μμc Sr^{90} per kg obtained by analysis of a composite sample made from equal amounts of soil from each location. Good precision was obtained for all Sr^{90}, calcium, and Cs^{137} soil analyses.

All 1958 and 1959 soil results are summarized in Table III. The "available" analyses satisfy the initial purpose of this investigation. Since all 0 to 2-inch perimeter samples contained 94 to 100 μμc Sr^{90} per kg, there was no detectable influence of Plant waste (Sr^{90}) over fallout contamination.

"Total" analyses, also shown on Table III, were run for comparison with results obtained at the Health and Safety Laboratory (HASL). The weight of several dried 12x12x1-inch soil plots was found to be 7.6 kg. This number was used to convert "total" μμc Sr^{90} per kg to mc Sr^{90} per square mile.

October 1958 Soils, Sr^{90}, mc/mile^2

SRP Results			HASL Results[6]		
	Soil Depth, in.			Soil Depth, in.	
Sample Location	0 - 2	2 - 4	Sample Location	0 - 2	2 - 8
Inner Perimeter Only	22.3	9.6	Atlanta	23.5	13.5
All Perimeters	29.1	10.6	Jacksonville	26.2	9.8

Soils in the SRP area compare favorably with those in Atlanta and Jacksonville. The highest result obtained by HASL in October 1958 was 78 mc Sr^{90} per square mile for Salt Lake City soil 0 to 7.5 inches deep.

Inner perimeter soil analyses are compared graphically in Figure 2. Analyses of these sandy loam, acidic, low calcium soils by both methods of leaching gave similar results. Good intermethod comparisons were found for some English soils of the same composition.[31] Soils collected at most of the other SRP perimeters contained more organic matter than did soils collected from the inner perimeter. Results in Table III indicate a much higher "total" Sr^{90} content for these perimeters at both the 0 to 2 and 2 to 4-inch depths.

Outer perimeter soils at both depths were analyzed by the "available" method. Results are summarized in Figure 3. Note that the calcium content at 0 to 2 and 2 to 4 inches is almost identical, while the Sr^{90} content in 2 to 4-inch soils is proportional, but lower.

In June 1959, outer perimeter soil samples decreased in Sr^{90} content, as shown on Table III (footnote b). Samples for determining Sr^{90} penetration were also taken in June 1959. These were analyzed by the "total" method and are reported in Figure 4.

Each group of soils in Figures 2, 3, and 4 shows a large variation in Sr^{90} content, even though all samples from a given perimeter were collected on the same day. Although some locations are only two miles apart, local differences in fission product rainout and variations in exchange capacities may explain the nonuniformity. Each location may have received equal rainfall over an extended period, but some received rain while a larger portion of atmospheric bomb debris was available. In all three figures, there is a correlation between calcium and Sr^{90}. Very high or very low calcium content (e.g., locations 9 and 10, Figure 2) is accompanied by high or low Sr^{90}. Sr^{90} in local soils appears to be fixed or adsorbed in proportion to calcium content.

wms Table III

Cs^{137} analysis for twenty soils, including those on Table II, showed the Cs^{137}/Sr^{90} ratio to be 2.0 ±0.3. The ratio determined for five soils at Argonne(32) was 1.9. Cs^{137} in these soils was determined by pulse height analysis. A 2.0 ratio was also found in air samples, regardless of latitude or apparent age of fission product debris collected.(33) Additional comparisons will be made for local soils, but an estimate of Sr^{90} appears to be possible from Cs^{137} content of unknown soils.

Grass samples from the 25-mile radius, the outer perimeter, and the H-Area perimeters contained 1490, 1414, and 1330 μμc of Sr^{90} per kg, respectively, in October 1958. Although precision for duplicate samples was not as good for grass samples as that obtained for soil samples, perimeter results are considered to be reliable. Calcium was fairly consistent at 4 grams per kg at each location.

Comparison of Cs^{137} and Sr^{90} content in grasses from one perimeter showed very erratic ratios. Calcium and Sr^{90} results obtained for grass and by "available" analysis of soils also showed no correlation at the same sampling locations. Some grasses grown in very low calcium soils were relatively high in Sr^{90} content. These results may be explained by factors discussed in the Background Information and Milk sections, including stem-base absorption(37) and Plant calcium-Sr^{90} discriminations.(21)

MISCELLANEOUS

The three types of samples discussed here are oyster, river water, and urine. Other samples were also analyzed, but not in sufficient numbers to be related to the regional survey program. These include bones and flesh of fish, animal bones, and deer antlers. All samples were assayed by the HDEHP extraction method (Appendix III).

Oysters were collected in 1958 at the mouths of the Savannah and Edisto Rivers. Eighteen separate samples of shells and flesh were dried, fired in a muffle furnace, and dissolved in 0.08N HCl prior to analysis.

Shells from both locations contained 1.3 d/m Sr^{90} per gram (or 1.5 μμc of Sr^{90} per gram of calcium). Although counting statistics were relatively poor, comparisons of these results indicated no contamination at the mouth of the Savannah River resulting from SRP operations. All flesh samples contained less than 0.1 d/m Sr^{90} per gram.

River water from above and below the Plant was analyzed for Sr^{90}. Soluble Sr^{90} was determined by filtering water through No. 42 Whatman paper prior to evaporation, wet ashing, and dissolving of the residue in 0.08N HCl. The Sr^{90} contained in water and in particulate matter was determined by addition of 100 mg of calcium carrier and oxalate coprecipitation, starting with step 3 in the Total Soil Method (Appendix II).

Eleven samples collected in August and September 1959 contained from 1 to 2 μμc of Sr^{90}/ℓ. At this level, 3500-ml samples were counted with less than 10% counting error. Duplicate and tracer recoveries were good and distilled water reagent blanks were negative.

Eight individual urine samples and a separate composite urine sample were collected in August 1959 from SRP employees whose duties do not involve work with radioactive materials. Sr^{90} was quantitatively precipitated as an ammoniacal phosphate and the precipitate was fired in a muffle furnace(36) and dissolved in 0.08N HCl.

The average Sr^{90} content of the individual urine samples was 3.6 d/m/ℓ (maximum and minimum were 5.2 and 2.3 d/m/ℓ, respectively); the composite urine contained 2.4 d/m Sr^{90}/ℓ. Eighteen pooled and individual urine samples analyzed at HASL(5) in 1956 contained an average of 1.1 d/m Sr^{90}/ℓ. The threefold increase in Sr^{90} in urine is comparable to increases found in milk and other samples in the three-year period. Analysis by this method is simple and accurate. The 90-minute counting deviation for a 1-liter sample is less than 15%.

FUTURE PROGRAM

The following program is planned for monitoring Sr^{90} in the Savannah River Plant area.

Milk samples will be collected quarterly at the eight locations shown in Figure 1. Milk from the larger dairies in this region will also be analyzed on a quarterly basis.

Water samples from above and below the Plant will be analyzed weekly. Rainwater will be continuously collected for analysis.

Soil and vegetation samples will be collected from the Plant outer perimeter in June of each year.

Deciduous and adult teeth have been collected from the families of Plant employees and from residents of Augusta, Georgia and Anderson, South Carolina. Preliminary results indicate that single teeth may be assayed for Sr^{90} content. The future program will be based on results of additional experiments.

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APPENDIX I

COLLECTION PROCEDURE FOR SOIL AND GRASS

A. SELECTION OF SAMPLE LOCATIONS

Locations for sampling soil and grass should be chosen according to the following requirements:

1. The soil should be undisturbed.

2. Select an open area that is not covered by trees or bushes.

3. Select an area that will allow sampling of both grass and soil.

4. Do not sample in a ditch or on uneven ground that may have accumulated rain abnormally.

5. The location should be easily accessible from a paved road.

B. SAMPLE IDENTIFICATION

1. Tag each soil and grass sample, recording the date, regional number, and perimeter. Tag the soil sample for 0-2 or 2-4 inch depth.

2. Place the grass and the two soil samples from each location into a larger bag.

C. GRASS SAMPLING

1. Select fresh bermuda grass samples when possible.

2. Using the grass cutters, cut the grass to ground level over an area of several square feet.

3. Cut the grass from five or more plots with a total area of several hundred square feet.

4. Accumulate the grass cuttings by placing them into a paper bag. Collect two or more pounds of grass, estimating the weight by comparison with a weighed sample.

D. SOIL SAMPLING

1. Select plots from which the grass was cut to ground level.

2. Take 0-2 inch soil from three plots at each location, using the flat-bladed spade. First cut an entry trench about 1 foot long, and 4 inches wide. Then, using a ruler for accurate measurement, insert the shovel blade at the 2-inch depth for sampling.

3. Accumulate two or more pounds of 0-2 inch soil onto a polyethylene sheet. Sort out the grass and other material and pour the soil into a polyethylene bag.

4. Collect the 2-4 inch soil at the same locations, being careful not to allow "fall-in" of the top soil.

APPENDIX II

SAMPLE PREPARATION PROCEDURES

A. MILK

Resin Pretreatment - "Dowex" 50-W, X-8, medium porosity, 200-400 mesh resin is obtained in the hydrogen form. To remove the finer particles and the excess HCl, the resin is washed with an equal volume of water and allowed to settle. The water is poured off and the resin is transferred to a Buchner funnel fitted with a No. 41 Whatman filter paper. After three distilled water rinses, the slightly damp resin is stored.

1. Add 30 g of pretreated resin to 500 ml of milk contained in a beaker. Stir the milk for one minute and adjust the pH to 6(±1) with 6M NaOH (Note 1). Stir the milk for 30 minutes and allow the resin to settle.

2. Remove the milk with suction and wash the resin with 200 ml of distilled water. Remove the water with suction and discard the milk and water.

3. Add 200 ml of 8N HNO_3 to the resin and stir for 30 minutes. Slurry the solution and, using a Buchner vacuum filter equipped with two No. 42 Whatman filter papers, filter the solution. Wash the resin three times with 50-ml volumes of 8N HNO_3, allowing each wash solution to filter completely.

4. Pour the combined acid washes to a beaker and evaporate them to dryness. Add 50 ml of 30% H_2O_2 and bake the residue (Note 2).

5. Cool and dissolve the white residue in 30-60 ml of 0.08N HCl.

Note 1 - Most milks will curdle after initial stirring with resin. The samples will return to liquid at about pH 6.

Note 2 - Oxidize the solids with additional HNO_3 and H_2O_2 if necessary.

B. SOIL

Each sample is mixed, screened, and dried according to the Sunshine Procedure(23) prior to extraction.

Available Method

1. Add 500 ml of neutral 1N ammonium acetate to 300 g of soil contained in a beaker. Stir for 30 minutes and allow the sample to sit overnight.

2. Slurry the solution and filter, using a Buchner vacuum filter equipped with a No. 42 Whatman filter paper. Wash the soil with ammonium acetate solution in volumes of 200, 200, and 100 ml, allowing each rinse to filter completely.

3. Combine the solutions and evaporate them to dryness. Wet ash the solids twice with 100 ml of HNO_3. Add 50 ml of 30% H_2O_2 and bake the residue to dryness.

4. Dissolve the residue in 3 ml of HCl and 50 ml of distilled water. Heat the solution to 80°C and add 1:1 NH_4OH, while stirring, to pH 7. Filter the solution immediately onto a prepared gravity filter funnel fitted with a No. 41 Whatman filter paper. Wash the precipitate with 25 ml of hot 1% NH_4Cl and four 25-ml volumes of hot distilled water. Evaporate the filtrate to dryness.

5. Dissolve the residue in 0.08N HCl and transfer and dilute to volume in a 100-ml volumetric flask.

Total Method

1. Stir 300 g of soil with 300 ml of 6N HCl for 30 minutes. Allow the sample to stand overnight.

2. Slurry the solution and filter, using a Buchner vacuum funnel equipped with No. 42 Whatman paper. Wash the soil with two 50-ml volumes of hot distilled water, allowing complete filtration between rinses. Dilute the rinse solution to 600 ml in a polyethylene bottle.

3. Pour a 400-ml aliquot into a beaker and add 400 ml of distilled water and 20 g oxalic acid. Heat the solution to 80°C and add 1:1 NH_4OH, while stirring, until the pH is adjusted to 5.

4. Allow the oxalates to settle overnight. Filter the solution, using a Buchner vacuum funnel fitted with a No. 42 Whatman filter paper. Wash the precipitate onto the filter with distilled water. Transfer the precipitate and filter paper to a porcelain crucible. (Note 1)

5. Dry the precipitate in a muffle furnace set at 100°C for one hour, and then fire at 650°C over a four-hour period. Remove the crucible and allow it to cool to room temperature. Add 10 drops of HNO_3 and H_2O_2, being careful not to splatter the precipitate. Place the crucible on a hot plate and bake the residue to dryness.

6. Dissolve and transfer the residue to a 50-ml volumetric flask, using 0.08N HCl.

Note 1 - Additional Sr^{90} may be precipitated from the solution by a second calcium carrier oxalate precipitation.

Add 40 ml of HCl to the filtrate from Step 4. Then add 50 mg of calcium (as calcium acetate), heat the solution to 80°C, and precipitate oxalates by adjustment of pH to 5. Repeat Step 4, combining the precipitates in the original crucible.

C. VEGETATION

1. Using a paper cutter, cut 200 g of vegetation. Dry the mixed vegetation in an oven set at 105°C.

2. Add 50 g of vegetation to a "Vycor" beaker and fire it overnight at 600°C.

3. Cool the beaker, add 40 ml of HNO_3, and reflux with low heat for 30 minutes. Cool the solution and separate the acid by centrifuging.

4. Repeat Step 3 using two additional 40-ml volumes of HNO_3. Combine the leach solutions and evaporate to dryness. Add 50 ml of 30% H_2O_2 and evaporate to dryness.

5. Dissolve the residue in 0.08N HCl and dilute to volume in a 200-ml volumetric flask.

APPENDIX III

ANALYTICAL PROCEDURES

After preparation of samples in 0.08N HCl, described in Appendix II, aliquots are taken for specific analyses.

A. Sr^{90}

The procedure was adapted from the work of Peppard and associates (34). Di-2-ethylhexyl phosphoric acid (HDEHP) is obtained from Union Carbide Chemical Company. All dilutions are made in toluene.

Two 20% HDEHP extractions are performed with the 0.08N HCl aliquot. Interfering nuclides, including $Ce\text{-}Pr^{144}$, $Zr\text{-}Nb^{95}$, Zn^{65}, as well as Y^{90}, are removed from the sample solution. The sample is stored for buildup of Y^{90} for about two weeks. A third extraction, using 5% HDEHP, removes the Y^{90}, which is finally counted in a low background counter.

The following procedure applies to 30-ml solutions of 0.08N HCl. Samples contained in 30 to 60 ml of solution have been extracted with HDEHP, using proportional increases in all extraction solutions.

1. Pour 30 ml of 0.08N HCl sample solution into a 125-ml separatory funnel. Extract the sample with 30 ml of 20% HDEHP, shaking vigorously for two minutes. Allow the phases to separate and drain off the bottom sample layer.

2. Extract the sample again with a fresh solution of 20% HDEHP. Record the time of the second extraction (T_1) and store the sample in a small bottle for two weeks.

3. Extract the sample, after Y^{90} buildup, with 30 ml of 5% HDEHP. Note the time of extraction (T_2).

4. Drain the sample solution to the bottle. Wash the 5% HDEHP solution by shaking with two 30-ml volumes of 0.08N HCl. Discard the wash solutions.

5. Extract Y^{90} from the 5% HDEHP solution with two 30-ml volumes of 3N HNO_3. Shake two minutes for each extraction and combine the 3N HNO_3 solutions in a small beaker.

6. Evaporate the 3N HNO_3 to small volume and quantitatively transfer the solution to a shallow 1-1/4-inch stainless steel planchet. Evaporate the solution to dryness under an infrared lamp and count for Y^{90}.

7. Apply the buildup factor (T_1-T_2 time) and decay factor of Y^{90} (T_2-midpoint of counting time) for a final calculation of Y^{90} d/m/sample.

B. CALCIUM

Calcium was determined after calcium oxalate precipitation. The precipitate was filtered and excess oxalic acid was removed by washing the precipitate with hot distilled water. The precipitate was dissolved in dilute H_2SO_4 and calcium was determined by titration of oxalate with $KMnO_4$.

Aliquots of all sample solutions except milk were taken for calcium analysis. Separate whole milk samples were evaporated and oxidized prior to analysis.

C. Cs^{137}

Cs^{137} was precipitated with cesium carrier and phosphotungstic acid(35). The precipitate was separated and counted in the low background counter.

1. Evaporate an 0.08N HCl aliquot of sample to dryness. Dissolve the residue in 6N HCl - 0.05N HF solution. Transfer the solution to a conical centrifuge tube and dilute to 40 ml with 6N HCl - 0.05N HF.

2. Add a carrier solution containing 1 mg each of strontium, zirconium, ruthenium, and cesium. Swirl the tube to mix the solution.

3. Add 1 ml of 0.05N phosphotungstic acid and allow the sample to stand 10 minutes. Centrifuge the solution for 5 minutes at 2000 rpm and discard the supernate.

4. Wash the precipitate with 5N HNO_3, centrifuging and discarding the wash solution.

5. Transfer the precipitate to a shallow 1-1/4-inch stainless steel planchet, using dilute HNO_3 acid. Evaporate the solution under an infrared lamp and count the sample in a low background counter.

APPENDIX IV

LOW BACKGROUND COUNTER

All samples analyzed for Sr^{90} and Cs^{137} were counted in a Tracerlab CE-14 low background counter. The counter was modified by addition of an electric timer. "Lucite" acrylic resin planchet holders were also constructed to permit sample counting to within 1/8 inch of the two detectors. Shallow 1-1/4-inch stainless steel planchets were used.

Y^{90} was extracted from NBS Sr-Y^{90} solution and counted at 47% efficiency. Frequent background counts were made, including overnight and weekend counts. Twenty consecutive background counts were as follows:

Detector 1	Detector 2
0.51 ±0.04 c/m	0.48 ±0.03 c/m

Sample counting and recounting (to verify Y^{90} purity) was performed to reduce counting error to a *minimum*:

c/m/sample	Length of Count
5 to 10	60 minutes
$>$ 10	30-45 minutes
$<$ 5	$\geq$ 90 minutes

All sample aliquots were adjusted for 60-minute counting when possible.

TABLE I

RESULTS OF MILK SAMPLES COLLECTED FROM APRIL 1958 TO AUGUST 1959

Location of Dairy	Dairy Number	Sample Date	Sr^{90}, d/m(a)	μμc Sr^{90}(b) Per Liter	Gram Calcium(c) Per Liter	μμc Cs^{137}(d) Per Liter	Ratio Cs^{137}/Sr^{90}
Aiken, S. C.	1	4/7/58	(e)	11	(f)		
Aiken, S. C.	2	4/7/58	(e)	15	(f)		
Aiken, S. C.	1	4/25/58	(e)	11	(f)		
Aiken, S. C.	2	4/25/58	(e)	15	(f)		
Dalton, Ga.	3	5/30/58	(e)	16	(f)		
Alabama	4	5/30/58	(e)	19	(f)		
Charleston, S. C.	5	5/30/58	(e)	12	(f)		
Williston, S. C.	6(g)	5/30/58	(e)	9	(f)		
Anderson, S. C.	7	5/30/58	(e)	19	(f)		
Jacksonville, Fla.	8	5/30/58	(e)	5	(f)		
Aniston, Ala.	9	5/30/58	(e)	13	(f)		
Lake City, S. C.	10	5/30/58	(e)	16	(f)		
Charlotte, N. C.	11	5/30/58	(e)	24	(f)		
Augusta, Ga.	1	5/30/58	(e)	18	(f)		
Charleston, S. C.	5	9/1/58	11.6 12.2	11.8	1.06		
Morganton, N. C.	12	9/15/58	16.8 13.7	15.4	1.12		
Lakeview, S. C.	13	10/12/58	13.4 15.3	14.4	1.12		
Williston, S. C.	6(g)	10/16/58	16.2 16.9	16.8	1.06		
Talatha, S. C.	14(g)	10/16/58	10.6 9.7	10.0	1.38		
Aiken, S. C.	15	10/16/58	13.0 14.5	14.2	1.09		
Waynesboro, Ga.	16	10/17/58	8.7 9.1	8.5	.92		
North Augusta, S. C.	17	10/17/58	8.4 6.8	7.4	1.08		
Snelling S. C.	18(g)	10/17/58	10.9 10.7	10.6	1.19		
Sylvania, Ga.	19	10/17/58	4.6 5.7	4.2	1.06		
Brogdon, S. C.	20(g)	10/26/58	20.3 20.2	19.1	1.14		
Brogdon, S. C.	20(g)	11/16/58	19.3 15.1	22.1	1.14	71	3.2
Brogdon, S. C.	20(g)	11/30/58	18.7 16.9	18.9	1.29	118	6.2
Brogdon, S. C.	20(g)	1/25/59	20.3 18.0	19.5			
New Ellenton, S. C.	1	11/21/58	16.3 15.1	15.5	.93	76	4.9
Langley, S. C.	21	12/2/58	13.2 13.1	17.1	1.17	81	4.7
Langley, S. C.	21	12/9/58	11.0 10.6	13.1	1.11	61	4.7
Langley, S. C.	21	12/16/58	8.9	11.5	1.08	46	4.0
Los Angeles, Calif.	22, Condensed		19.1 18.1	26.3	1.19	221	8.4
New York, N. Y.	23, Condensed	Purchased	10.9 11.9	16.1	1.08	226	14.0
Columbus, Ohio	24, Condensed	4/21/59	4.0 2.9	6.0	.33	91	15.2
St. Louis, Mo.	25, Condensed		17.9	21.6	1.07	98	4.5
Los Angeles, Calif.	26, Powdered		4.5 5.4	6.3	1.04	102	16.2
New York, N. Y.	27, Powdered		8.1 9.4	12.4	.96	101	8.1
New Ellenton, S. C.	1	4/21/59	15.1 15.3	25.8	1.09	176	6.8
New Ellenton, S. C.	2	4/21/59	11.6	19.7	.94	155	7.9
New Ellenton, S. C.	2, Skim	4/21/59	13.1 12.1	21.3	.98	223	10.5
New Ellenton, S. C.	28	4/21/59	10.2 12.2	19.0	.94	244	12.8
Langley, S. C.	21	4/21/59	10.8 12.3	19.6	.92	142	7.2
Langley, S. C.	21	1/6/59	14.0 13.4	14.3	1.19		
Langley, S. C.	21	2/3/59	12.8 16.0	15.1	1.05		
Talatha, S. C.	14(g)	6/29/59	26.0 25.7	27.8	1.36		
Snelling, S. C.	18(g)	6/29/59	34.2 35.0	37.1	1.36		
Langley, S. C.	21(g)	6/29/59	16.2 17.1	17.3	1.11		
Williston, S. C.	6(g)	6/29/59	20.2 21.4	21.7	.97		
Aiken, S. C.	15	6/29/59	11.3 13.1	12.6	1.15		
Talatha, S. C.	14(g)	7/20/59	20.6 22.0	22.7	1.33		
Snelling, S. C.	18(g)	7/20/59	20.4 21.0	22.4	1.20		
Langley, S. C.	12	7/20/59	21.2 22.2	23.5	1.12		
Brogdon, S. C.	20(g)	7/20/59	44.6 45.5	32.4	1.13		
Brogdon, S. C.	20(g)	8/30/59	23.4 27.4	24.1	1.02		

(a) Most duplicate samples were 500 ml. All were recounted to verify for Y^{90}.

(b) Corrected for recovery of $Sr\text{-}Y^{90}$ tracer added to the third samples. The recovery of 40 tracers was 85.2 ±10.3%.

(c) Average of two or more determinations.

(d) Analyzed in duplicate and corrected for Cs^{137} tracer recovery.

(e) Single 1 liter samples assayed.

(f) Over-all average of Ca in these samples = 1.12 g/ℓ.

(g) Raw milk from an individual cow. Most others were homogenized, pasteurized milks from dairies of greater than 50 cows.

TABLE II

ZERO TO TWO INCH INNER PERIMETER SOIL SAMPLE RESULTS

October 1958

	Available Leaching Method			Total Leaching Method			
Type of Soil	Sr^{90}, d/m/180 g	Sr^{90}, μμc/kg(a)	Calcium, g/kg(b)	Sr^{90}, μμc/kg(c)	Calcium, g/kg(b)	Cs^{137}, μμc/kg(c)	Ratio, Cs^{137}/Sr^{90}
Sandy Loam	37.6 39.9	120	.155	122.0	.177	266	2.2
Sandy Loam	28.2 29.3	89	.104	68.5	.112	199	2.9
Sandy Loam	30.1 28.9	93	.271	140.9	.335	216	1.5
Sandy Loam	27.4 25.6	82	.037	126.2	.045	228	1.8
Sandy Loam	15.6 16.8	50	.014	76.0	.035	142	1.9
Sandy Loam	30.5 31.0	95	.029	100.3	.039	196	2.0
Sandy Loam	20.7 18.1	60	.131	73	.192	149	2.0
Sandy Loam	27.0 24.6	80	.056	78.6	.095	266	3.4
Fine Sandy Loam	61.0 54.2	179	.191	203.8	.249	362	1.8
Sandy Loam	23.5 23.3	72	.054	51	.078	192	3.8
Sandy Loam	34.3 34.8	107	.151	127	.189	222	1.8
Sandy Loam	38.7 35.4	115	.126	133	.188	300	2.3
Sandy Loam	24.8 23.1	74	.204	99.8	.266	206	2.1
Sandy Loam	26.9 29.0	84	.170	96.1	.205	180	1.9
Average		93	.119	107	.157	223	2.3
Composite		95	.118	103	.154	236	2.3

(a) Corrected for average tracer recovery of 80.9%.

(b) Average of 3 analyses. These were aliquots from the duplicates and tracer samples.

(c) Corrected for average tracer recovery. Duplicate sample precision was comparable to precision shown by ammonium acetate leach method.

TABLE III

SUMMARY OF OCTOBER 1958 SOIL ANALYSES BY AVAILABLE AND TOTAL LEACHING METHODS

Sampling Perimeter	Number of Samples	Available Method		Total Method	
		Sr^{90}, μμc/kg	Ca, g/kg	Sr^{90}, μμc/kg	Ca, g/kg
		0-2-inch Soils			
25-Mile Radius	14	95(a)	.220	171	.348
Outer	14	96(a,b)	.305	150(b)	.361
Inner	14	94(a)	.119	105(a)	.154
F and H	24	100(a)	.215	124	.177
		Average			
		98	.215	137	.280
		2-4-inch Soils			
25 Mile Radius	14	30	.182	50	.259
Outer	14	37(a)	.307	64(b)	.333
Inner	14	42	.092	45	.108
F and H	24	31	.089	40	.112
		Average			
		35	.167	50	.203

(a) Each soil from the perimeter was analyzed. All others were determined from four analyses of composite samples from each perimeter.

(b) In June 1959, the outer perimeter soils were resampled. The following results show a decrease in Sr^{90} at the 0-2-inch soil levels. Sr^{90} also decreased in the 2-6-inch soil level.

0-2-inch sample averages Available Method = 75 μμc/kg
Total Method = 119 μμc/kg
2-6-inch sample averages Total Method = 55 μμc/kg

TABLE IV

INTERLABORATORY COMPARATIVE ANALYSES

Sample	Origin of Samples	Other Sites		SRP	
		Sr^{90}	Ca, g	Sr^{90}	Ca, g
Milk, powdered	USDHEW(a)	7.9 ±0.7 μμc/100 g	1.17/100 g	7.6 μμc/100 g	1.18/100 g
Milk, fresh	USDHEW	17.4 μμc/ℓ	1.15/ℓ	11.7 μμc/ℓ	1.03/ℓ
Milk, fresh	USDHEW	9.1 μμc/ℓ	1.129/ℓ	11.5 (12.0 & 11.0) μμc/ℓ	1.09/ℓ
Milk, fresh	USDHEW	4.2 μμc/ℓ	1.023/ℓ	4.5 (4.3 & 4.7) μμc/ℓ	1.03/ℓ
Milk, fresh	Hanford(b)	9.4 & 7.6 μμc/ℓ		10.1 & 10.2 μμc/ℓ	
Milk Ash	HASL(c)	1.41 ±0.16 d/m/g		1.61 d/m/g	
$Sr\text{-}Y^{90}$ Solution	HASL	91.1 ±6.0 d/m/vial		91.1(a) d/m/vial	
Animal Bone Ash	HASL	11.5 ±0.8 d/m/g		13.3 & 13.4 d/m/g	
$Ca_3(PO_4)_2$ - Blank	HASL	≤0.047 d/m/g		<0.3 d/m/g	

(a) U. S. Department of Health, Education, and Welfare, Robert A. Taft Center, Cincinnati, Ohio.

(b) Hanford Atomic Products Operations, Richland, Washington.

(c) Health and Safety Laboratory, AEC, New York.

BIBLIOGRAPHY

1. Libby, W. F. Stratospheric Fallout Particularly from the Russian October Series. Atomic Energy Commission, Washington, D. C. and Carnegie Institute of Washington, D. C. TID-5556, 19 pp. (1959).

2. Eisenbud, M. "Deposition of Strontium-90 Through October 1958". Science 130, pp. 76-80 (1959) July, No. 3367.

3. Holland, J. Z. Summary of New Data on Atmospheric Fallout. Division of Biology and Medicine, Atomic Energy Commission. TID-5554, 15 pp. (1959).

4. Kulp, J. L., A. R. Shulert, and E. J. Hodges. "Strontium-90 in Man, III". Science 129, pp. 1249-1255 (1959) May, No. 3358.

5. Environmental Contamination from Weapon Tests. New York Operations Office. Health and Safety Lab., Atomic Energy Commission Research and Development Report. HASL-42, 370 pp. (1958).

6. Hardy, E. P., Jr. and S. Klein. Strontium Program Quarterly Summary Report. New York Operations Office, Health and Safety Lab., Atomic Energy Commission Research and Development Report. HASL-51, 97 pp. (Nov. 1958); HASL-55, 109 pp. (Feb. 1959); HASL-65, 204 pp. (May 1959).

7. U. S. Congress. Joint Committee on Atomic Energy. The Nature of Radioactive Fallout and Its Effects on Man. Part 2. Washington: U. S. Govt. Print. Off. (1957).

8. U. S. Congress. Joint Committee on Atomic Energy. Fallout from Nuclear Weapons Tests. Washington: U. S. Govt. Print. Off. (1959) 2618 pp.

9. Hursh, J. B., ed. Chemical Methods for Routine Bioassay. Rochester, N. Y. Univ. AECU-4024, 104 pp. (1958).

10. Dunham, C. L. Radioactive Fallout - A Two-Year Summary Report. Division of Biology and Medicine, Atomic Energy Commission. TID-5550, 112 pp. (1959).

11. Kornberg, H. A. On the Passage of Pairs of Elements Through Food Chains. General Electric Co., Hanford Atomic Products Operations, Richland, Wash. AEC Research and Development Report HW-60127, 37 pp. (1959).

12. MacDonald, N. S., et. al. The Effects of Calcium and Phosphates in Foods on Radiostrontium Accumulation. Atomic Energy Project, Univ. of Calif., Los Angeles. AEC Research and Development Report. UCLA-317, 20 pp. (1954).

13. Palmer, R. F., R. C. Thompson, and H. A. Kornberg. "Effect of Calcium on Deposition of Sr^{90} and Ca^{45} in Rats". Science 127, pp. 1505-1506 (1958). June, No. 3313.

14. Durbin, P. W., et. al. Metabolic Studies with Strontium-90 in the Rhesus Monkey. Preliminary Report. California Univ., Berkeley, Radiation Lab. AEC Research and Development Report. UCRL-3634, 26 pp. (1957).

15. Alexander, G. V., and R. E. Nusbaum. The Relative Retention of Strontium and Calcium in Human Bone Tissue. California Univ. Los Angelos, AEC Research and Development Report. UCLA-426, 23 pp. (1958).

16. Palmer, R. F., R. C. Thompson, and H. A. Kornberg. "Factors Affecting the Relative Deposition of Strontium and Calcium in the Rat". Science 128, pp. 1505-1506 (1958) Dec., No. 3337

17. Boyd, J., W. F. Neuman, and H. C. Hodge. On the Mechanism of Skeletal Fixation of Strontium. Rochester, N. Y. Univ. Atomic Energy Project. AEC Research and Development Report, UR-512, 16 pp. (1957).

18. Gietelman, H. J., and W. F. Neuman. Lead-Hydroxy Apatite Interaction. Rochester, N. Y. Atomic Energy Project. AEC Research and Development Report. UR-551, 25 pp. (1959).

19. Sobel, A. E., et. al. "A Way Out for Unwelcome Sr". Chemical & Eng. News 37, 42-3 (1959) Sept., 28, No. 39.

20. "Removal of Sr^{89} and Ca^{45} from Milk by Means of Ion Exchange Columns". Semi-Annual Progress Report for July 1, 1958 to December 31, 1958. Tenn. Univ., Knoxville, Agricultural Research Lab., AEC Research and Development Report. ORO-183, 45 pp. (1959) p. 9.

21. Hungate, F. P., R. L. Uhler, and J. F. Cline. Hanford Biology Research Annual Report for 1957. General Electric Co. Hanford Atomic Products Operation, Richland, Washington. AEC Research and Development Report. HW-53500, 227 pp (1957) pp. 7-16.

22. Comar, C. L., R. S. Russel, and R. H. Wasserman. "Strontium-Calcium Movement from Soil to Man". Science 126, pp. 485-492 (1957) Sept., No. 3372.

23. Martell, E. A. The Chicago Sunshine Method: Absolute Assay of Strontium-90 in Biological Materials, Soils, Waters, and Air Filters. Chicago Univ. Enrico Fermi Inst. for Nuclear Structure. AEC Research and Development Report. AECU-3262, 65 pp. (1956).

24. Harley, J. H., and I. B. Whitney. Health and Safety Laboratory Analytical Methods. NYO-4700 (1957). New York Office of the AEC.

25. Bryant, R. J, et. al. Radiostrontium Fallout in Biological Materials in Britain. Great Britain, Atomic Energy Research Establishment, Harwell, Berks, England. AERE-HP/R-2056, 49 pp. (1956).

26. Ibid, With Amendments (1957).

27. Campbell, J. E., et. al. "The Occurrence of Sr^{90}, I^{131}, and Other Radionuclides in Milk". Presented at annual meeting of the American Public Health Association on October 7, 1958.

28. Nervik, W. E., M. I. Kalkstein, and W. F. Libby. Purification of Milk for Calcium and Strontium with "Dowex" - 50W Resin. California. Univ., Berkeley. Radiation Lab., AEC Research and Development Report. UCRL-2674, 13 pp. (Aug. 1954)(declassified Nov. 1955).

29. Silker, W. B. Strontium-90 Concentration in the Hanford Environs. General Electric Co. Hanford Atomic Products Operation, Richland, Washington. AEC Research and Development Report. HW-55117, 28 pp. (1958).

30. Graham, E. R. "Determining Strontium-90 in Soil by Electrodialysis and Ion Exchange Method". Soil Sci. 88, 11-15 (1959) July, No. 1.

31. Bryant, F. J., et. al. Radiostrontium in Soil, Grass, Milk, and Bone in the United Kingdom: 1956 Results. Great Britain. Atomic Energy Research Establishment, Harwell, Berks, England. AERE-HP/R-2353, 33 pp. (1957).

32. Radiological Physics Division Semiannual Report - January Through June 1958. Argonne National Lab., Lemont, Ill. AEC Research and Development Report. ANL-5919, 143 pp. (1958).

33. Baus, R. A., et. al. Radiochemical Analyses of Air-Filter Samples Collected During 1957. Naval Research Lab., Washington, D. C. NRL-5239, 19 pp. (1958).

34. Peppard, D. F., G. W. Mason, and S. W. Moline. "The Use of Dioctyl Phosphoric Acid Extraction in the Isolation of Carrier-Free Y^{90}, La^{140}, Ce^{144}, and Pr^{143}." J. Inorg. Nucl. Chem. 5, pp. 141-6 (1957) No. 2.

35. Mizzan, E. Phosphotungstate Precipitation Method of Analysis of Radioactive Cesium in Solutions of Long Lived Fission Product Activities. Atomic Energy of Canada, Ltd., Chalk River Project, Canada. PDB-128, 14 pp. (1954).

36. Boni, A. L. "Rapid Determination of Mixed Beta-Gamma Radionuclides In Urine". Health Physics 2, 186 p. (1959) Oct., No. 2.

37. Russell, R. S. "Deposition of Strontium-90 and Its Content in Vegetation in Human Diet in the United Kingdom". Nature 182, 834-839 (1958) September, No. 4639.

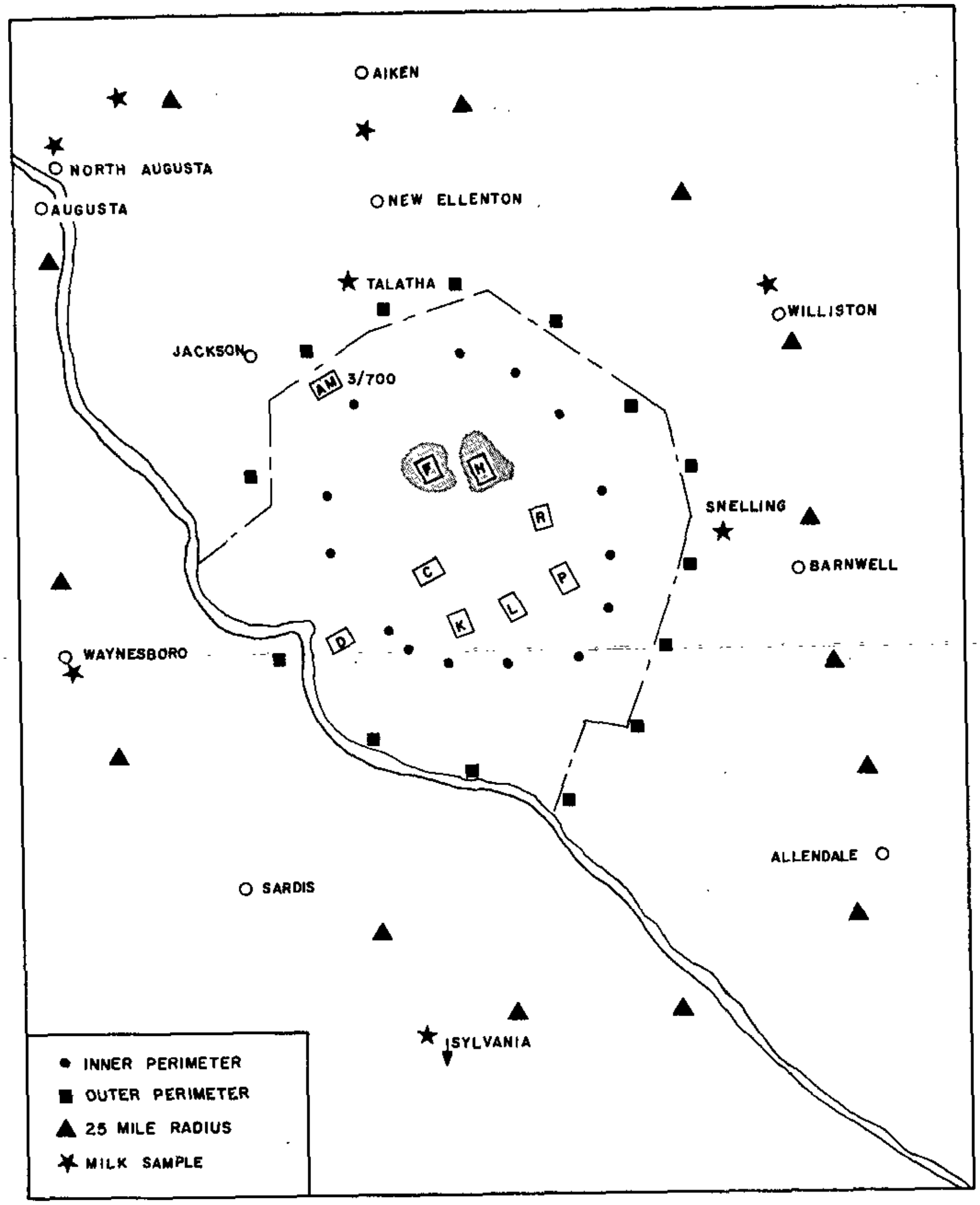

FIG. 1 SAMPLE LOCATIONS

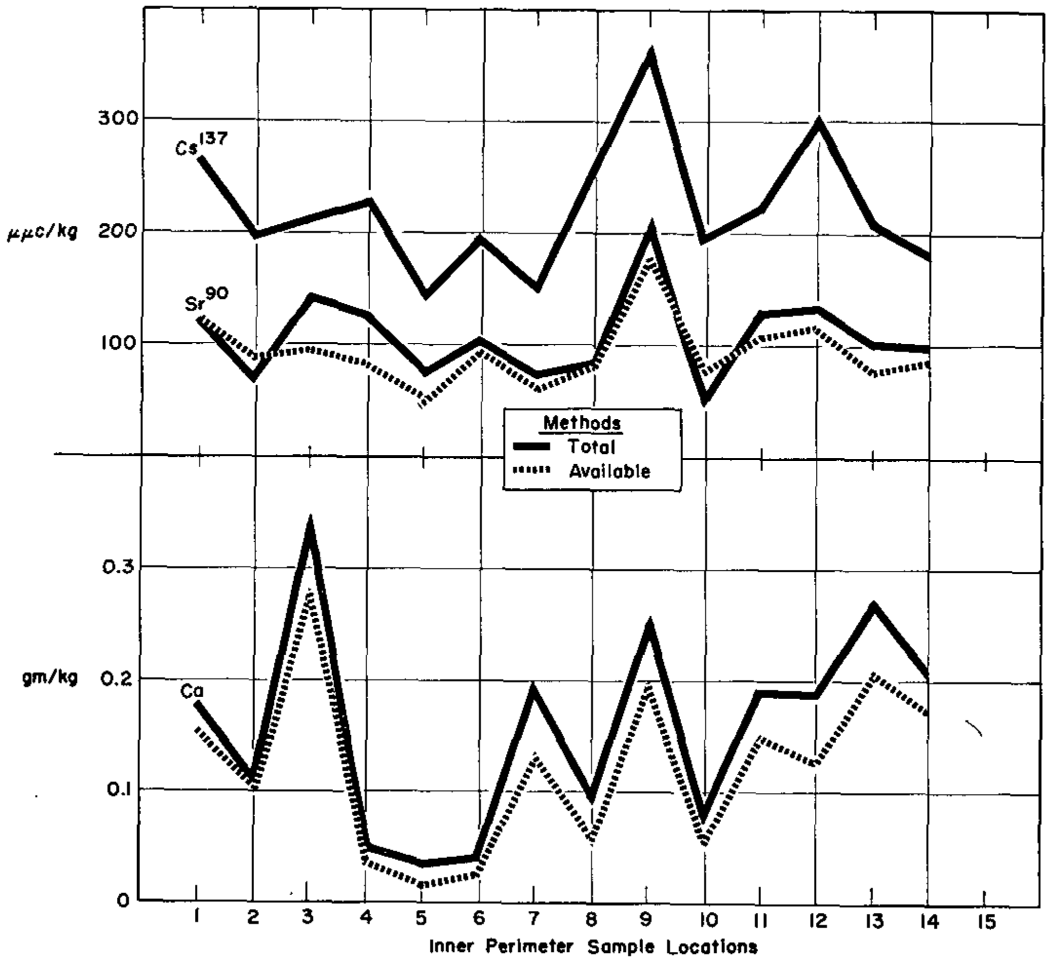

FIG. 2 COMPARISON OF RESULTS BY TWO METHODS OF SOIL EXTRACTION

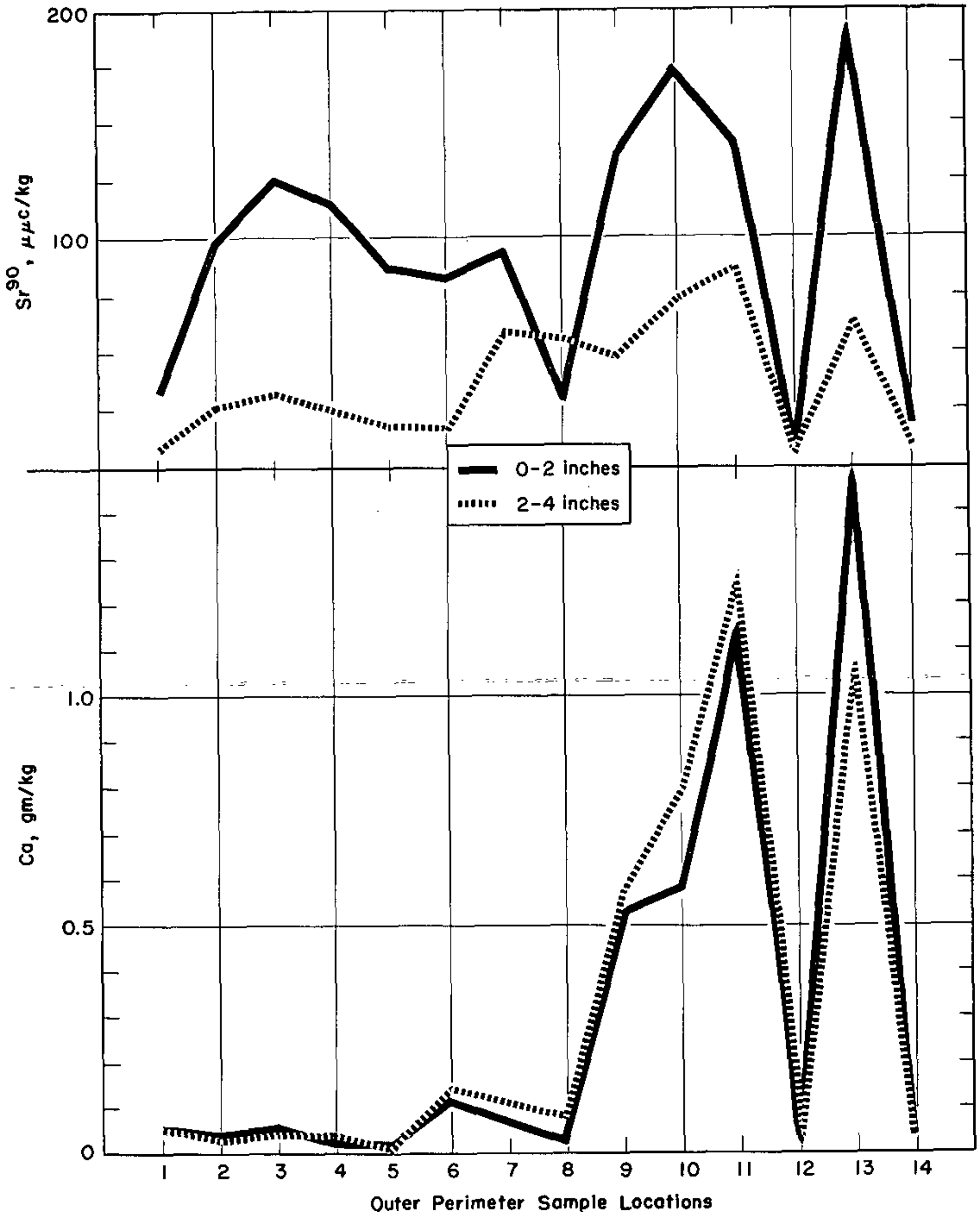

FIG. 3 AMMONIUM ACETATE EXTRACTION OF 0-2 INCH AND 2-4 INCH SOILS

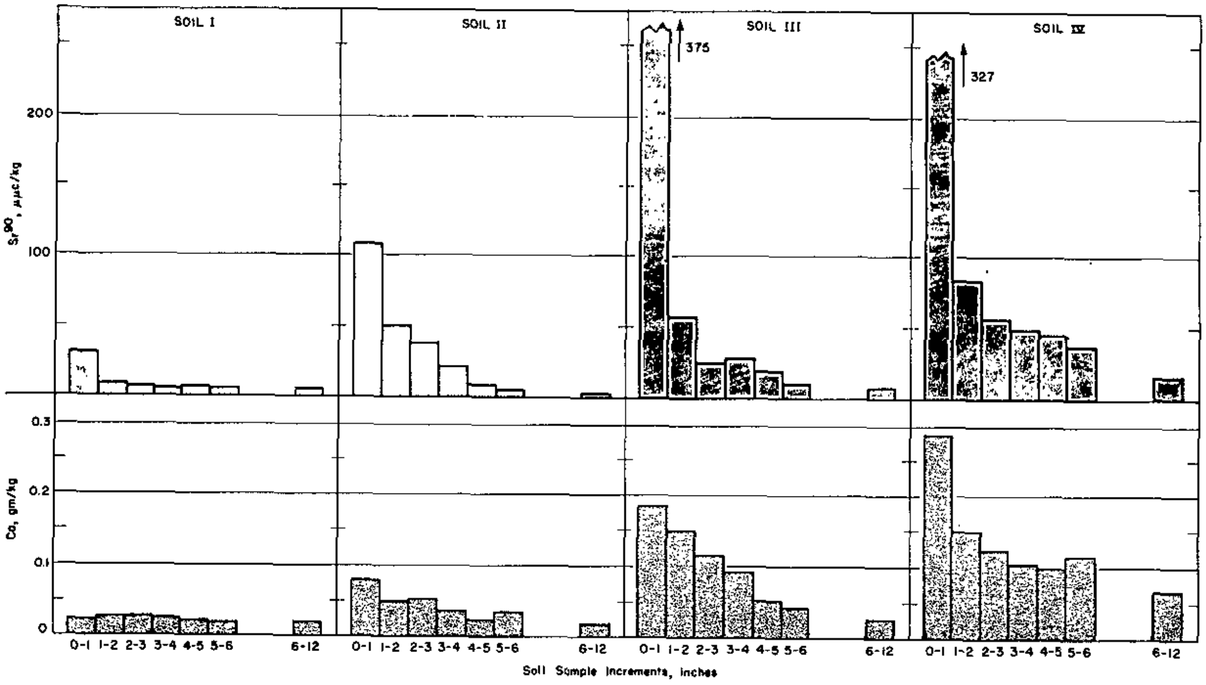

FIG. 4 PENETRATION OF Sr^{90} IN OUTER PERIMETER SOIL SAMPLES
(June 1959)