doses ranging from several thousand down to 25 roentgens. If the dose-response function continues linear and passes through the origin, any amount of radiation (no matter how small) will carry a certain probability of producing a detrimental effect and a certain fraction of the natural mutation rate must be attributed to unavoidable background exposure. The fraction of natural frequency that results from background radiation is not known. The most outstanding recent results regarding the potential genetic effects of background and fallout radiation are those reported by Russell (*3*), who found that mutation frequency in spermatogonia and oöcytes of mice was influenced by dose rate. From this extremely important discovery, it may be concluded that the 30-year dose from background and from fallout may have less genetic consequences than had been predicted previously. The discovery that mutation frequency is dose-rate dependent, however, does not invalidate the linear hypothesis, and it cannot be assumed from present knowledge that further lowering of the dose rate will result in still less potential genetic risk.

In any event, the predicted dose rate and the integrated 30-year gonadal dose from present levels of biospheric contamination are between 1 and 2% of that from natural background; therefore, the potential genetic hazard to the world population from weapon tests to date will not be more than 1 to 2% of the unavoidable burden from natural sources of radiation.

Estimation of the potential somatic consequences of biospheric contamination from weapon testing is even more difficult and uncertain than the estimation of genetic effects. The absolute somatic hazards imposed on the world population by the bone and bone marrow doses estimated in Table II are critically dependent on the shape of the dose-response curves for leukemia and bone sarcoma and, as with mutation frequency, on the extent to which they may be dose-rate dependent. If somatic response shows a curvilinear or threshold relationship with dose (Figure 5), a factor (or factors) other than 70-year integrated exposure must be important, and biological effect may drop to insignificant levels at the very low doses and dose rates received from natural background radiation and fallout. Although existence of an absolute threshold may never be proved empirically, existence of a practical one may be inferred from a curvilinear dose-response relationship. If, however, the linear proportionate hypothesis is accepted, any amount of radiation to the bone and bone marrow carries a finite probability of producing bone sarcoma and leukemia, and a fraction of the natural population incidence of these diseases must be attributed to natural background. In this case also, the controversy over population distribution of Sr^{90} bone concentrations about the mean loses much of its significance, as it is necessary to average the potential hazards over the population at risk. Only in the event of a threshold in the dose-response relationship is it of primary importance to know the fraction of the population that exceeds the mean by 2, 5, or 10 times. Figure 5 presents the alternative possibilities in a controversy which cannot be resolved at present and which may not be resolved unequivocally for many years. The answer is not known, even by the most vocal opponents or proponents of nuclear weapon testing. At present it is possible only to say from the data in Table II that if a fraction of the normal incidence of leukemia and bone cancer indeed is caused by natural radiation, weapon tests to date may increase the incidence of these diseases in the generation born during the time of maximum biospheric contamination by about 5 to 10% of that due to unavoidable natural background exposure.

Literature Cited

The proceedings of the two Congressional Subcommittee Hearings on Fallout from Nuclear Weapon Tests (referenced below) contain 4683 pages of testimony and documented material furnished for the record by dozens of participants and investigators. The reader is referred to these proceedings for additional information, for support material, and for specific credit to the individuals cited. No attempt is made to cite individual references except in those cases where publication has occurred since the hearings.

(1) Burton, J. D., Milbourn, G. M., Russell, R. S., *Nature* **185,** 498 (1960).
(2) Kulp, J. L., Schulert, A. R., Hodges, E. J., *Science* **132,** 448 (1960).
(3) Special Subcommittee on Radiation, "Fallout from Nuclear Weapons Tests," Joint Committee on Atomic Energy, 86th Congress, 1st Session, Vol. 1, 2, and 3, U. S. Govt. Printing Office, Washington, D. C., May 5–8, 1959.
(4) Special Subcommittee on Radiation, "The Nature of Radioactive Fallout and Its Effects on Man," Joint Committee on Atomic Energy, 85th Congress, 1st Session, U. S. Govt. Printing Office, Washington, D. C., May 27–29, June 3, 1957 (Part 1), June 4–7, 1957 (Part 2).

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PLANT UPTAKE OF RADIONUCLIDES

Availability of Exchangeable and Nonexchangeable Strontium-90 to Plants

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A PORTION of the strontium-90 in soils is not readily exchangeable with neutral salts, such as ammonium acetate or strontium nitrate (*2, 12, 14*). Since the strontium-90 in world-wide fallout is essentially water-soluble (*6*), the portion that is not now exchangeable has become so by reactions in the soil. Soluble and exchangeable cations in soils are usually readily available for uptake by plants (*10*); therefore, it is of interest to attempt to determine whether or not the nonexchangeable portion of strontium-90 in soils contributes significantly to plant uptake.

Discrimination Factor

Discrimination factors are a measure of the relative availabilities of two similar ions to plants grown in a nutrient medium. For the uptake of strontium-90 and calcium from soils, the discrimination factor is defined as:

$$\frac{\mu\mu\text{c. available } Sr^{90}}{\text{grams available Ca in soil}} \Big/ \frac{\mu\mu\text{c. } Sr^{90}}{\text{grams Ca in plant}}$$

While the measurement of strontium-90 and calcium in plants is unequivocal, the measurement of available cations in

Exchangeable and nonexchangeable fractions of strontium-90 were determined in soil samples taken from the plow layer of cultivated fields in the coastal plain of North Carolina in June 1955 and December 1958. Exchangeable strontium-90 contents averaged about 10 and 50 μμc. per kg. of soil on the two sampling dates, respectively. Nonexchangeable strontium-90 contents averaged 4 and 7 μμc., respectively. Lower amounts of both fractions of strontium-90 were recovered in samples extracted after dry storage for 1 year. The uptake of strontium-90 and calcium from these soils was studied by growing cowpeas in the greenhouse. From 8 to 18% of the exchangeable strontium was taken up, depending on the uptake of exchangeable calcium. Use of discrimination factors to determine availability of nonexchangeable strontium-90 to plants is discussed. Nonexchangeable strontium-90 apparently made little or no contribution to uptake.

Table I. Exchangeable and Nonexchangeable Ca and Sr^{90} Contents of Soils Collected from Pitt County, N. C.

Soils	Exchangeable Ca, Meq./Kg.	Exchangeable Sr^{90}, μμc./Kg.	Nonexchangeable Sr^{90}, μμc./Kg.	Nonexchangeable Sr^{90} as % of Total Sr^{90}
		JUNE 1955		
Lynchburg-Pitt No. 1	16.6	13.2	2.5	15.9
Dunbar-Pitt No. 2	36.7	13.3	1.8	11.9
Dunbar-Pitt No. 3	15.0	10.8	3.6	25.0
Lynchburg-Pitt No. 4	26.7	7.1	7.2	50.3
		DECEMBER 1958		
Lynchburg-Pitt No. 1	12.9	45.9	2.5	5.2
Dunbar-Pitt No. 2	42.3	50.1	2.6	4.9
Dunbar-Pitt No. 3	19.2	57.1	9.6	14.4
Lynchburg-Pitt No. 4	27.2	51.2	13.0	20.2

the soil is not. Available nutrients in a soil may be defined as those belonging to a pool which contributes to cation uptake by plants grown on that soil. Plants reflect the integrated value of the pool, and this value is difficult to duplicate with a chemical extraction. Measurement of the specific activity of a nutrient element in plants grown in soil containing a radioactive isotope of the element has been very useful in determining the suitability of various chemical extractants for measuring available nutrients (*5*). For similar ions, the discrimination factor approximates a specific activity measurement, and in the case of fission product elements, may be more easily measured than the specific activity.

Dilute $CaCl_2$ extracts of soils gave nearly the same ratio of strontium and calcium as was found in plants grown on these soils (*11*)—i.e., the discrimination factor was approximately 1. Discrimination factors of approximately 1 have also been found for uptake of strontium and calcium by plants grown in nutrient solutions over a range of strontium and calcium atom ratios from 1:10 to 1:1000 (*1*, *8*). At least when the strontium concentration is much less than calcium concentration, as it normally is in soils, dissolved strontium and calcium are equally available to plants.

The exchangeable cations of soils have shown ratios of strontium and calcium from 1 to 2.5 times greater than was found in plants grown on the soils (*7*, *9*)—i.e., the discrimination factors were 1.0 to 2.5. This means that the availability of exchangeable strontium in some soils is less than that of exchangeable calcium. The observed discrimination factors are within the range of exchange equilibrium constants for replacement of calcium by strontium on different clay minerals (*4*). The results are consistent with the view that plants absorb cations from the soil solution, and the solution is replenished by cation exchange reaction with the solid phase. Exchangeable calcium in noncalcareous soils has been shown to correspond closely with the pool of calcium available to plants (*3*).

Acid-extractable cations have not generally been used to calculate discrimination factors. However, uptake of rubidium and cesium was more closely correlated with acid-soluble potassium than with exchangeable potassium in soils (*7*). Strong acids are known to extract much more than the exchangeable calcium from soils, especially calcareous soils (*13*). However, it is possible that nonexchangeable strontium-90 (acid-extractable), which was comparatively recently in a soluble form, is nearly as available as exchangeable strontium-90 or calcium. If this is so, the "total" (exchangeable plus nonexchangeable) strontium-90 should be reflected in plant uptake, and discrimination factors for similar soils, with similar clay minerals, should be nearly constant when total strontium-90 is used to calculate them. If nonexchangeable strontium-90 is not available, the discrimination factors should be nearly constant when exchangeable strontium-90 is used to calculate them.

Methods

Preparation of Soil. Discrimination factors (see Discussion) were used to assess the relative availabilities of Sr^{90} and calcium to cowpeas grown on four soil samples in the greenhouse. The soil samples were taken from the plow layer of cultivated Dunbar and Lynchburg sandy loams in eastern North Carolina in December 1958. These soils varied considerably in amount of nonexchangeable strontium-90 (Table I). The soils are noncalcareous, with exchangeable strontium and calcium atom ratios of about 1 to 1000. The soils have similar clay mineral composition and texture in the plow layer so that soil differences were not expected to affect the discrimination factors. Fifty pounds of each soil sample was dried on greenhouse benches. After breaking the lumps, the soils were sieved and mixed. Separate aliquots were taken for the green house experiment and for the soil analyses.

Culture of Plants. The greenhouse experiment with cowpeas was replicated three times. Each replicate consisted of two No. 10 cans containing 5 pounds of soil per can. Before potting, 0.13 gram of N, 0.13 gram of P, 0.20 gram of K, as $NH_4H_2PO_4$ and KNO_3, and smaller amounts of the micronutrients were mixed with the soil for each pot. The pots were seeded February 16, 1959. After good growth was established, the plants were thinned to four per pot. The aboveground portions of the plants were harvested May 11 at floral initiation. The total yield of aboveground portions from each replicate was weighed and dry ashed in an oven at 500° C.

Table II. Ca and Sr^{90} Contents of Cowpeas Grown in Greenhouse in Soils Listed in Table I

(Average of three replicates)

Soils	Yield, Grams Dry Wt.	Ca, Meq./Gram	Sr^{90}, μμc./Gram	$\frac{Sr^{90}}{Ca}$, μμc./Meq.	% Exchangeable Sr^{90} in Soil Taken Up by Plants
Lynchburg-Pitt No. 1	26.5	0.47	0.89	1.89	11.4
Dunbar-Pitt No. 2	36.6	0.79	0.47	0.59	7.6
Dunbar-Pitt No. 3	46.4	0.66	1.02	1.55	18.2
Lynchburg-Pitt No. 4	43.2	0.59	0.55	0.93	10.2
Error, std. dev.	1.7	0.011	0.034		

Table III. Sr^{90} Contents of Soil Samples Collected from Greene and Edgecomb Counties, N. C., December 1958

Soils	Exchangeable Ca, Meq./Kg.	Exchangeable Sr^{90}, μμc./Kg.	Nonexchangeable Sr^{90}, μμc./Kg.	Nonexchangeable Sr^{90} as % of Total Sr^{90}
Lynchburg-Greene No. 2	27.8	35.0	3.8	9.8
Dunbar-Greene No. 3	24.8	38.6	7.4	16.1
Dunbar-Greene No. 4	24.5	30.6	3.0	8.9
Lynchburg-Edgecomb No. 1	33.1	48.7	4.4	8.3
Lynchburg-Edgecomb No. 3	29.3	44.1	5.4	10.9
Dunbar-Edgecomb No. 4	21.9	28.8	6.6	18.6

Determination of Ca. Exchangeable Ca in the soil was determined by extraction with ammonium acetate. Twenty-five grams of soil was shaken with 100 ml. of 1N neutral ammonium acetate, and allowed to stand overnight. The suspension was filtered, and an additional 150 ml. of 1N neutral ammonium acetate was leached through the soil. The leachate was dried on a hot plate, organic matter was destroyed with HNO_3 and $HClO_4$, and the residue was dissolved in 0.1N HNO_3. Calcium was determined by titration with (ethylenedinitrilo)tetraacetic acid (EDTA).

Determination of Sr^{90}. Successive extractions were made to determine the amounts of exchangeable and nonexchangeable Sr^{90} in the soil. Exchangeable Sr^{90} was removed by extraction with neutral 0.75N $Sr(NO_3)_2$. The reagent displaced Sr^{90} from the soil by isotopic exchange, and furnished a large amount of carrier strontium to minimize possible losses of Sr^{90} in the later chemical treatments. Nonexchangeable Sr^{90} was removed by subsequent extraction with hot 4N HCl.

The extractions were carried out by suspending 1 kg. of soil in 1 liter of 0.75N $Sr(NO_3)_2$, and allowing to stand overnight. The suspension was filtered and the soil was resuspended in a second liter of 0.75N $Sr(NO_3)_2$. After standing 4 to 6 hours, the suspension was filtered and the soil was leached with a third liter of 0.75N $Sr(NO_3)_2$. The $Sr(NO_3)_2$ extracts were combined, dried on a hot plate, and treated with concentrated HCl to destroy organic matter. The soil was resuspended in 1.5 liters of 4N HCl and allowed to stand overnight on a hot plate at 80° C. The suspension was filtered and the soil leached with 1 liter of 1N HCl. The HCl extracts were combined, dried on a hot plate, and treated with concentrated HNO_3 to destroy organic matter.

The HCl extract required further treatment to separate strontium from large amounts of iron, aluminum, and silicon. After destruction of organic matter, the residue was digested in HCl and filtered. Sulfuric acid was added to the filtrate to precipitate $SrSO_4$. The $SrSO_4$ was filtered, dried, and fused with Na_2CO_3 at 900° C. The fusion cake was then digested in hot water to remove the sulfate, and the $SrCO_3$ was dissolved in HCl.

Both extracts were scavenged four times to remove interfering radioactive contaminants, chiefly radium, thorium, their daughters, and rare-earth fission products. The scavenges were made by precipitating, in order, $Y(OH)_3$, $BaCrO_4$, $BaCrO_4$, and $Y(OH)_3$ in the extracts. $Y(OH)_3$ was precipitated by adding 20 mg. of yttrium carrier and then adding NH_4OH to pH 8. $BaCrO_4$ was precipitated homogeneously by adding 20 mg. of barium carrier, an excess of dichromate, and then hydrolyzing KCNO slowly at 5° C. to pH 5. After the four scavenges, yttrium carrier was again added to the extracts. They were stored for 2 weeks to allow the equilibrium amount of yttrium-90 to build up.

Determinations of Sr^{90} were made by separating and counting its radioactive daughter, Y^{90}. Yttrium was first precipitated as the hydroxide and then reprecipitated as the oxalate before it was mounted for counting under an end window Geiger-Müller tube. Each sample was counted at least three times to follow its radioactive decay. Two sets of counting equipment were used, with efficiencies of 20 and 31%, as determined by comparable analyses of a National Bureau of Standards sample of Sr^{90}-Y^{90}. Background corrections ranged from 2.2 to 2.5 counts per minute. The Sr^{90} content of each extract was checked at least twice.

The contents of Sr^{90} and Ca in plant material from each replicate were determined separately. The ashed plant material was dissolved in 1N HCl and filtered. A small aliquot was taken for determination of calcium by titration with EDTA. The remaining solution was scavenged twice by precipitating $Y(OH)_3$, omitting the $BaCrO_4$ scavenges, and Sr^{90} was determined in the same way as it was for soil extracts.

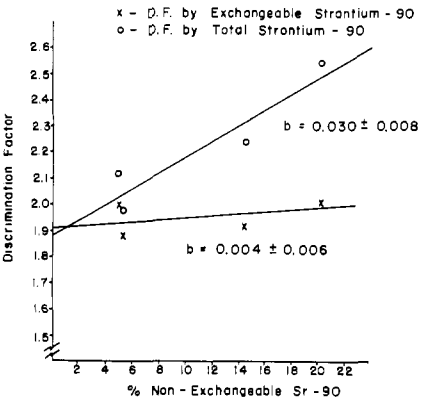

Figure 1. Effect of nonexchangeable Sr^{90} on discrimination factors calculated using exchangeable or total Sr^{90} in the soil

Results and Discussion

The Ca and Sr^{90} contents of the soils and cowpeas are shown in Tables I and II. Sr^{90} contents of the soils were determined on a single soil sample, so no direct measure of analytical error is available. Replicates from the greenhouse experiment showed coefficients of variation of about 5% for yield and contents of Sr^{90} and calcium. Variation between replicates includes the errors of aliquoting soil, growing plants in the greenhouse, and analyzing plant materials.

The discrimination factors were nearly constant when they were calculated using exchangeable Sr^{90} in the soil, but not when they were calculated using total Sr^{90} (Figure 1). This indicates that nonexchangeable Sr^{90} was not taken up appreciably by the cowpeas. The regression lines were fitted to the experimental points by the method of least squares. The slope, b, of each regression line is shown together with its standard error of estimate. Owing to the small number of experimental points, the difference between slopes is barely significant at the 95% confidence level.

Table IV. Ca and Sr^{90} Contents of Cowpeas Grown in Greenhouse in Soils Listed in Table III

(Average of three replicates)

Soils	Yield, Grams Dry Wt.	Ca, Meq./Gram	Sr^{90}, μμc./Gram	$\frac{Sr^{90}}{Ca}$, μμc./Meq.
Lynchburg-Greene No. 2	41.0	0.89	0.89	1.00
Dunbar-Greene No. 3	44.9	0.77	0.88	1.14
Dunbar-Greene No. 4	33.5	0.55	0.55	1.00
Lynchburg-Edgecomb No. 1	43.2	0.48	0.56	1.17
Lynchburg-Edgecomb No. 3	45.3	0.79	1.03	1.30
Dunbar-Edgecomb No. 4	44.8	0.69	0.65	0.94
Error, std. dev.	1.8	0.027	0.045	

Possibly the HCl extraction did not remove all of the Sr^{90} remaining in the soil after extraction with $Sr(NO_3)_2$. Up to 15 or 20% more Sr^{90} has been found in extracts obtained by fusing soil samples with Na_2CO_3 than in extracts obtained by treatment with 6*N* HCl at room temperature (*15*). If part of the Sr^{90} in soils is not extracted with HCl, it would not be expected to be available to plants, especially since the evidence indicates that nonexchangeable Sr^{90} is not available to plants.

The possibility that Sr^{90} is incompletely extracted from soils by HCl digestion was emphasized by analyses of six Lynchburg and Dunbar soil samples obtained from eastern North Carolina at the same time that the four samples from Pitt County were taken. These samples were also used in the greenhouse experiment with cowpeas, and were treated in the same way as those from Pitt County, except that the soil extractions were not made until December 1959. The samples from Pitt County had been extracted in February 1959 at nearly the same time as the aliquots of soil were prepared for the greenhouse experiment. An average of only 38 μμc. of exchangeable Sr^{90} per kg. of soil was found in the soils extracted at the later time (Table III), compared to an average of 52 μμc. in the soils extracted at the earlier time. Yet the plant uptake of calcium and Sr^{90} indicated that all of the soils contained about the same amount of available Sr^{90} (Table IV), that is, with equal calcium contents in the soil, the plants contained the same ratios of Sr^{90} and Ca. The efficiency of the HCl extractions is now being checked by fusing the soils with Na_2CO_3.

The results indicate that about 15 μμc. of Sr^{90} per kg. of soil was fixed during storage in the laboratory and that it was not extracted by HCl digestion. If the exchangeable Sr^{90} analyses for all 10 soils had been comparable, a more definite statement about the difference between slopes of the regression lines in Figure 1 could probably have been made. At present, all that can be said is that nonexchangeable Sr^{90} appears not be taken up appreciably by cowpeas.

Literature Cited

(1) Bowen, H. J. M., Dymond, J. A., *J. Exptl. Botany* **7,** 264–72 (1956).
(2) Brown, I. C., Menzel, R. G., Roberts, H., Beltsville, Md., unpublished data, 1958.
(3) Davis, D. E., MacIntire, W. H., Comar, C. L., Shaw, W. M., Winterberg, S. H., Harris, H. C., *Soil Sci.* **76,** 153–63 (1953).
(4) Krishnamoorthy, C., Overstreet, R., *Ibid.*, **69,** 41–53 (1950).
(5) Lathwell, D. J., Sanchez, N., Lisk, D. J., Peech, M., *Agronomy J.* **50,** 366–9 (1958).
(6) Martell, E. A., *Science* **129,** 1197–206 (1959).
(7) Menzel, R. G., *Soil Sci.* **77,** 419–25 (1954).
(8) Menzel, R. G., Heald, W. R., *Ibid.*, **80,** 287–93 (1955).
(9) Menzel, R. G., Heald, W. R., *Soil Sci. Soc. Am. Proc.* **23,** 110–112 (1959).
(10) Russell, E. J., "Soil Conditions and Plant Growth," 8th ed., revised by E. W. Russell, pp. 441–2, Longmans, Green and Co., New York, 1950.
(11) Russell, R. S., Schofield, R. K., Newbould, P., Proc. Second International Conference on Peaceful Uses of Atomic Energy, Vol. **21,** pp. 146–8, Pergamon Press, London, 1958.
(12) Schulz, R. K., Overstreet, R., Babcock, K. L., *Hilgardia* **27,** No. 13, 333–42 (1958).
(13) Sigmond, A. A. J., von, "Handbuch der Bodenlehre," **VIII,** 148–174, Julius Springer, Berlin, 1931.
(14) U. S. Atomic Energy Commission, Rept. **HASL-42,** pp. 7–8 (1958).
(15) *Ibid.*, Strontium Program, Quarterly Summary Rept. **HASL-77,** pp. 44–45 (1960).

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Depth of Feeding as It Affects the Concentration of Radioactivity within the Plant

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IN THE early reports of Project Sunshine, Libby and coworkers (*4*) expressed a belief that strontium-90 from fallout was concentrated in the top $2^1/_2$ inches of soil. Since that time, the nuclides have moved downward and it has been shown by others (*1*) that they have penetrated to greater depths probably even in virgin soil. Plowing and other agricultural practices have certainly placed surface concentrations to depths of 6 to 8 inches. Percolation of strontium-90 through the soil occurs more rapidly when the calcium content of the soil is high (*3*).

Root penetration varies markedly with the crop, rainfall, type of soil, locale of growth, food, and other factors, and ranges from 10 inches for some garden plants to well over 10 feet for alfalfa (*5*).

The depth to which a plant extends its root system and obtains nutrients should have a direct effect upon the plant's concentration of a radionuclide if that nuclide is concentrated at or near the soil surface. In a sense the nuclide absorbed near the surface is diluted by elemental species obtained at greater depths. Since both top and root growth respond to the presence of nutrients, the placement of fertilizers will stimulate root development in the zone of application and an increase in nutrient absorbed from that zone will result. A fertilizer placed in a zone which contains a radionuclide will increase the uptake of that radionuclide because of stimulation of plant growth. A calculated discrim-