sulfate determinations are given in Table V.

Sulfur recoveries of 100% were obtained from sodium sulfite, sodium thiosulfate, and the mineral zinc sulfide concentrate should assure the complete sulfur retention in the nitric-perchloric acid method of oxidation of biological materials.

Active boiling in perchloric acid of standard quantities of potassium sulfate for 1, 2, and 4 hours showed identical sulfate recoveries.

Effect of Perchloric Acid Evaporation on Sulfate Recovery. It is often necessary or desirable to evaporate the residual perchloric acid before precipitation of sulfates as barium sulfate. In such instances sodium chloride has been used as the fixative for sulfate. Hillebrand *et al.* (*3*) however, maintain that evaporation of sulfates in perchloric acid solution can be carried to dryness without loss of sulfuric acid, provided the solution contains a relatively large excess of an element such as calcium. The calcium excess necessary to obtain full recovery of sulfate after evaporation to dryness in perchloric acid of such materials as the sulfur containing amino acids was determined (Table VI).

Evaporation of potassium sulfate to dryness from perchloric acid solution showed losses of 50 to 20% of sulfate content. With calcium additions in the calcium to sulfur weight ratios of 1 to 1 up to 2 to 1, the maximum sulfur recovery (98%) was obtained with the 1.5 to 1.0 ratios. On the other hand, an addition of sodium as the chloride in the weight ratio of 20 to 1 showed a loss of 22.6% of the 10 mg. sulfate sulfur.

To retain all the sulfate upon evaporation in perchloric acid it is only necessary to have a calcium content to give a calcium-sulfur weight ratio of 1.5 to 1.0.

Literature Cited

(1) Assoc. Offic. Agr. Chemists, "Official Methods of Analysis," 8th ed., 1955.

(2) Evans, R. J., St. John, J. L., *Ind. Eng. Chem., Anal. Ed.* **16,** 630 (1944).

(3) Hillebrand, W. F., Lundell, G. E. F., Bright, H. A., Hoffman, J. F., "Applied Inorganic Analysis," 2nd ed., Wiley, New York, 1953.

(4) Horn, M. J., Jones, D. B., Blum, A. E., *J. Biol. Chem.* **166,** 321 (1946).

(5) McChesney, E. W., Banks, W. F., *Anal. Chem.* **27,** 987 (1955).

(6) Richards, L. A., U. S. Dept. Agr. Agricultural Handbook 60, 1954.

(7) Shaw, W. M., *Anal. Chem.* **30,** 1682 (1958).

(8) Youden, W. J., "Statistical Methods for Chemists," Wiley, New York, 1951.

Received for review January 29, 1959. Accepted August 24, 1959.

CESIUM-137 UPTAKE BY PLANTS

Factors Affecting Uptake of Radioactive Cesium by Lettuce, Grass, and Alfalfa

ERIC B. FOWLER and C. W. CHRISTENSON

Health Division, Los Alamos Scientific Laboratory, University of California, Los Alamos, N. M.

Cesium-137 forms about 6% of the products released on fission. It enters the biosphere through fallout and, hence, may find its way into plants and eventually into man. A study of soil factors which affect the uptake of cesium and its possible control are reported. An increase in the available potassium in the soil is reflected by a decrease in the cesium-137–potassium ratio in the plant. Discriminations by plants against cesium-137 were in the range of 300 to 1400, dependent upon the soil and the plant. Discrimination may, in part, be related to the ion exchange capacity of the soil and in part to the exchangeable potassium content of the soil. Potassium fertilizers may decrease the cesium-137–potassium ratio in the plant, but may mobilize cesium in some soils and thus render it more available to the plant.

An observed effect of potassium on the uptake of cesium by corn and millet has been reported by Menzel (*7*). A few widely varied discrimination factors against cesium by plants have been reported and reviewed by Langham and Anderson (*6*).

Cesium-137 forms about 6% of the products released on fission. It enters the biosphere through fallout and, hence, may find its way into plants and eventually into man. Unlike strontium, the biological half life of cesium is relatively short; however, a study of factors affecting the uptake of cesium-137 by plants is important because the widespread prevalence of this isotope in the biosphere effects its continued intake by man.

This paper reports the results of a study of soil factors which affect the uptake of cesium and its possible control.

Methods

The naturally occurring exchangeable potassium content of the four soils chosen for this study varied from 0.092 to 0.376 meq. per 100 grams of soil. They were collected from the plow layer of tilled soils on four midwestern farms. The soils were mixed with cesium-137 at a level of 1000 c.p.m. per gram. About 5 kg. of soil were dispensed into polyethylene containers. Each soil was planted in triplicate to lettuce, grass, and alfalfa. The containers were placed in steel trays in a greenhouse. All water was applied from the bottom, except for that applied during the germination period when the soil was watered from the top with a hand sprinkler.

Four cuttings of each crop were made in the 1958 growing season. The cuttings were weighed, dried, and ashed at 400° F. for 8 hours. The cut weight per pot of each crop varied from 48 to 96 grams for lettuce, from 15 to 40 grams for grass, and from 23 to 50 grams for alfalfa. Plant ash was digested in 6*N* nitric acid. Radioassay for cesium-137 was performed on the supernatant liquid after oxalate precipitation of calcium. Exchangeable soil cations were determined on the extracted portion after the soil had been leached with 1*N* ammonium acetate. Ion exchange capacities were determined by the method of Frysinger and Thomas (*4*) and do not represent the sum of the exchangeable cations as determined by leaching with ammonium acetate.

Results

Results of chemical analyses of soils are shown in Table I. Average concentrations of cesium-137 and total

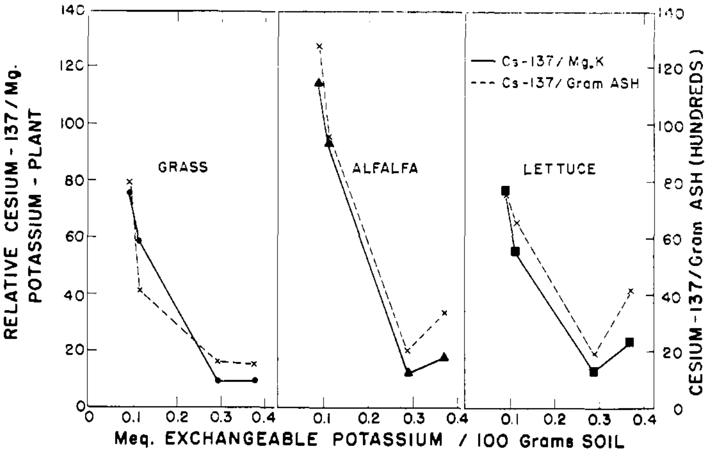

Figure 1. Effect of increments of exchangeable soil potassium on cesium-137 uptake by the plant

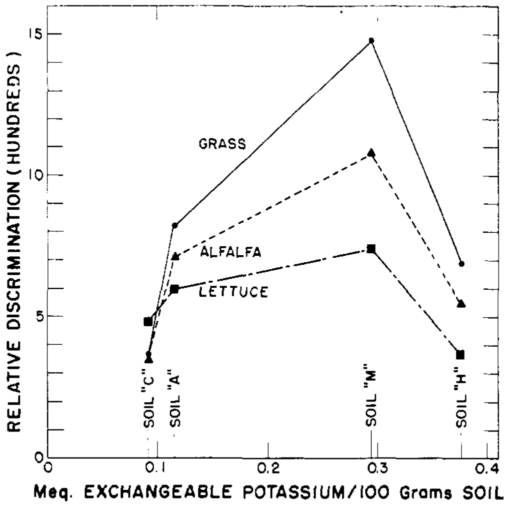

Figure 2. Plant discrimination against cesium-137 at various potassium levels

Table I. Soil Analyses

	Soil C, Sandy	*Soil A, Loamy Sand*	*Soil M, Loam*	*Soil H, Sandy Loam*
Ion exchange capacity, meq. cesium/100 g. soil	1.7	4.9	19.2	9.6
Kjeldahl nitrogen, %	0.092	0.149	0.335	0.210
Organic carbon, %	0.86	1.81	3.35	2.54
Potassium[a]	0.092	0.115	0.294	0.376
Magnesium[a]	6.6	7.5	18.0	10.7
Calcium[a]	4.39	7.68	31.74	12.77
Iron[a]	2.52	1.45	2.52	2.74
pH	6 8	5.8	7.3	6.4

[a] Meq./100 grams soil extracted with 1*N* ammonium acetate.

Table II. Cesium-137 and Total Potassium per Gram Plant Ash

	Soil C[a]		*Soil A*		*Soil M*		*Soil H*	
	Cs-137, c.p.m.	*K, mg.*	*Cs-137, c.p.m.*	*K, mg.*	*Cs-137, c.p.m.*	*K, mg.*	*Cs-137, c.p.m.*	*K, mg.*
Grass	7,900	105	4100	71	1600	178	1500	171
Lettuce	7,500	97	6500	118	1900	144	4100	180
Alfalfa	12,700	112	9500	102	2000	165	3300	196

[a] Exchangeable potassium values of soils as listed in Table I.

potassium per gram of plant ash are shown in Table II. In general, the cesium-137 concentration decreased and the potassium concentration in the plant increased as the exchangeable potassium of the soil increased. Soil H does not follow the pattern set by the other three soils. Soil H has the highest exchangeable potassium content, but a relatively low ion exchange capacity. The implication of these results will be developed later.

The effect of available soil potassium on the uptake of cesium-137 is shown graphically in Figure 1. The cesium-137 in the plant per unit of plant potassium has been plotted against the exchangeable potassium content of the soil. The dotted curve is a plot of total cesium-137 per gram of plant ash *vs.* soil potassium. As cesium follows potassium in metabolism, the cesium-137–potassium ratio is of interest from this standpoint. Cesium-137 per gram of ash is important with regard to absolute cesium content. The two curves are very similar.

The most marked effect of potassium was noted in the case of alfalfa and grass, where an increase in available soil potassium by a factor of 3.2 decreased the cesium-137–potassium ratio in the plant to about 12% or $^1/_8$ of the original. A calculation for lettuce shows a decrease to about 19% or $^3/_{16}$ of the original.

Even though soil H has the highest exchangeable potassium content (0.376 meq. per 100 grams), the effect on cesium-137 uptake is less than that found in soil M (0.294 meq. per 100 grams). This anomaly suggests that other factors may be responsible for the decreasing cesium-137–potassium ratio in the plant.

The discrimination factors against cesium-137 by the plant were calculated for three crops and four soils. These values are derived from the ratio $\frac{\text{Cs-137}}{\text{exchangeable K}}\ (\text{soil}) \Big/ \frac{\text{CS-137}}{\text{K}}\ (\text{plant})$. The results are shown graphically in Figure 2 where discrimination values have been plotted against exchangeable soil potassium. Each plotted point represents an average of seven to nine values. The standard deviation within a set of values varied from 14 to less than 6% of the mean. A value greater than 1 indicates discrimination against cesium by the plant. Discrimination was evidenced by the three plants on all four soils varying with the plant and soil from about 300 to 1400. The values were higher than those expected in the light of previous experience with calcium and strontium-90 (*3*).

Of the four soils, the high potassium soil, H, is out of its expected position, if one assumes exchangeable soil potassium to be the sole controlling factor affecting the uptake of cesium. Hence, a correlation between each of the following, their ratios, and discrimination was sought: soil calcium, soil magnesium, soil iron, and soil organic matter. No correlation could be found. However, a plot of soil ion exchange capacity *vs.* plant discrimination indicated a more linear relationship between the ion exchange capacity of the soil per unit of soil exchangeable potassium and plant discrimination (Figure 3).

Discrimination values vary with the plant and with the ratio of ion exchange capacity of the soil to its exchangeable potassium content. It is apparent that a statement of plant discrimination must be accompanied by a statement of soil characteristics and type of plant. It can be generally stated, with these four soils, that as the ratio of ion exchange capacity to exchangeable potassium in the soil increases, there will be an increase in discrimination against cesium on the part of the plant.

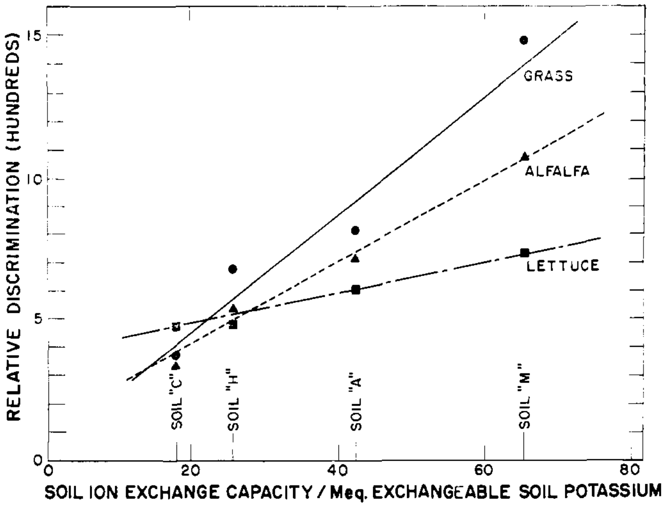

Figure 3. Relation of discrimination to ion exchange capacity per unit of exchangeable soil potassium

Discussion

It is realized that all of the factors affecting the uptake of radionuclides by plants cannot be adequately resolved with only four different types of soils. The discussion which follows is based on only four soils and the authors admit that the data are limited. Further investigation will be needed to resolve completely the complex effects of ion exchange capacity and degree of potassium saturation on the discrimination against cesium-137.

The discrimination factors against cesium-137 reported here are greater than those reported for strontium-90 (*3*) by two to three orders of magnitude.

Auerbauch (*1*) reports a discrimination factor against cesium of about 200 based on the amount of cesium found in the soil. The present results are based on the amount of cesium added to the soil and would probably drop by a factor of 4 to 5 if based on the amount of cesium leached by 1*N* ammonium acetate. On the other hand, if plants were growing under natural field conditions where cesium-137 is present in a surface layer, the discrimination factors might be higher than those reported here. Unpublished results which were obtained by the authors on plant depth of feeding indicate that only about 20 to 25% of a plant's nutrient is obtained from the surface 5 inches. Dilution of cesium-137 by feeding in a zone free of the radionuclide, as occurs in nature, would tend to increase the discrimination factor. This value would be an apparent discrimination, but would not reflect a true discrimination on the part of the plant.

A high discrimination factor could result from one or both of the following: Possibly the plant rejects cesium, or cesium is too tightly bound to the soil colloid to be available to the plant. There is some evidence which supports the latter proposition.

The uptake of cesium by plants grown hydroponically was reported to be higher by a factor of 1000 than when plants were grown in soil (*5*). The fractional plant nutrition method was used in this study in which the plants were grown in a nutrient solution and were then transferred to and from a tap water solution of cesium-137. This method probably exaggerates the uptake of cesium-137. However, the study did show that plants absorb cesium if it is available. In studies reported here from 20 to 25% of the cesium-137 in the soil was leached by 1*N* ammonium acetate. If the plant could remove comparable amounts, the discrimination factors should be lower than reported. The extremely high discrimination factors reported here, indicate that other factors are involved. Some reaction might occur in the soil between its components and cesium, thus rendering cesium unavailable to the plant but available in part to the ammonium acetate. Further evidence of the unavailability of cesium is indicated by the fact that cesium is tightly bound to soils and volcanic tuff and is not easily removed by leaching with solutions of the mono-, di-, and trivalent cations; potassium and ammonium ions are exceptions in this case (*2*).

Langham and Anderson (*6*) have calculated the over-all ratio from soil to milk for cesium and found that this theoretical cesium content is less than that which is shown by direct counting in the large volume counter. This difference is, in part, resolved if one assumes that some of the cesium appearing in the milk arises not from the soil, but by direct fallout on the plant.

The cesium fallout particle, being small, may be physically bound to plant surfaces and not removed even by rainfall. Soil potassium would have no effect on the concentration of this cesium on or in the plant.

Whether the radionuclide is retained on the leaf surface or absorbed, is immaterial, because the animal eats the entire plant. We do know that plants differ in extent of foliar absorption. Plant breeders might "build" a plant which will "shed" fallout and at the same time absorb a minimum amount through the leaf cuticle.

The application of potassium to soil through fertilizers may be beneficial in reducing the cesium-potassium ratio in the plant. However, a high soil ion exchange capacity is also important. A high potassium content may mobilize cesium in a low ion exchange capacity soil. It has been shown that the rate of leaching of cesium-137 increases in these four soils as the ion exchange capacity decreases.

Methods for the control of plant uptake of cesium-137 and strontium-90 are being sought. Past experience with these two radionuclides leads the authors to believe that the problem is extremely complex. Each soil and each plant must be considered as a separate problem.

As previously pointed out (*3*), a determination of the over-all ratio from soil to man is extremely difficult. In the case of cesium the difficulties are increased manyfold, probably because it is bound to the soil in such a way as to be unavailable to plants. The range of availability is soil-dependent and extensive.

Acknowledgment

The authors express their thanks to Lyle T. Alexander, Chief of Soil Survey, U. S. Department of Agriculture, for collecting and supplying the soils used in this study. Appreciation is also expressed to Howard Adams, George Johnson, Elgin Rex, and Richard Thomas, Group H-7, Los Alamos Scientific Laboratory, for assistance.

Literature Cited

(1) Auerbauch, S. I., Oak Ridge Natl. Laboratory, Health Physics Advisory Board Meeting, Oak Ridge, Tenn., November 1957.

(2) Christenson, C. W., Fowler, E. B., Johnson, G. L., Rex, E. H., Vigil, F. A., *Sewage and Ind. Wastes* **30,** 1478–89 (1958).

(3) Fowler, E. B., Christenson, C. W., *Science*, in press.

(4) Frysinger, Galen, Thomas, H. C., "Clays and Minerals," Natl. Acad. Sci.-Natl. Research Council Publ. **395,** 239–245 (1955).

(5) Klechkovsky, V. M., "On the Behavior of Fission Products in Soil," U. S. Atomic Energy Comm., **TR-2867** (1957).

(6) Langham, Wright, Anderson, E. C., "Entry of Radioactive Fallout into the Biosphere and Man," Swiss Acad. Medical Sci. Symposium on Noxious Effects of Low Level Radiation, Lausanne, Switzerland, March 1958.

(7) Menzel, R. G., *Soil Sci.* **77,** 419–25 (1954).

Received for review April 24, 1959. Accepted August 21, 1959. Division of Agricultural and Food Chemistry, 135th Meeting, ACS, Boston, Mass., April 1959.