Effect of Soil Nutrients on Plant Uptake of Fallout

Soil calcium and potassium decrease strontium-90/-calcium and cesium-137/potassium ratios in plants.

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The role of strontium-90 in the biosphere has reached marked prominence within the past decade. It is one of the products of nuclear fission (*1*); it has been shown to be a possible carcinogenic agent (*2*); and it has an appreciable half-life. Further, it is a close periodic relative of calcium; its relation to calcium metabolism has been reviewed (*2*).

Libby (*3*) and others have investigated the problem of strontium-90 fallout and have indicated the importance of the plant in the biological chain of transfer from soil to animal, to milk, and hence to man.

Collander (*4*) in 1937, using milligram amounts of calcium and strontium, showed that lettuce took up 500 times as much calcium as strontium. However, the ratio of calcium to strontium in the nutrient was of the order of 400 to 1. He places calcium and strontium in the same group relative to availability and relates availability to electrical mobility of the ions. If the work of Collander employed nutrient solutions, as is indicated, the results are probably not applicable to soils [see also (*5*)].

It was the purpose of the investigation discussed here to study the uptake of strontium-90 by plants growing in the presence of known amounts of the radionuclide and controlled concentrations of calcium.

Methods

Field tanks were chosen for this study, rather than greenhouse pots, to more nearly duplicate field conditions. Thirty tanks, each 5 by 5 feet and 2 feet deep, were filled to a depth of 18 inches with a soil composited from local Los Alamos soil, clay, and barnyard manure in the ratio of 4 to 1 to 1. The parent material of local soil is tuff, high in silicious matter and low in organic matter, with a low ion-exchange capacity. To assure uniformity, the separate fractions were passed through a shredder and then combined in the proper proportions and repassed.

A final mixing of the soil, radionuclides, and calcium was accomplished in a ½-yard concrete mixer; the radionuclides and calcium, as the chloride, were brought into solution and pumped into the mixer as tumbling was taking place. The use of calcium chloride was indicated because preliminary laboratory experiments had shown that calcium carbonate would not be available to the plant in this soil. Uniformity of distribution was determined by an analysis for leachable chloride, on the assumption that even distribution of calcium chloride indicated even distribution of the added radionuclides. The average variation among 90 random samples was less than 1 percent.

The concentration of strontium-90 and cesium-137 was such that 50 counts per minute per gram of soil were obtained. Preliminary calculations indicated that this level of activity would yield crop samples with satisfactory counting statistics. The amount of calcium chloride was varied from 1.5 to 18.1 milligrams of calcium per gram of soil in five increments. The intrinsic available calcium concentration of the mixture was 1.5 milligrams per gram of soil, and this level served as the control.

In addition, one tank received 1.5 milligrams of calcium per gram of soil, applied as calcium carbonate, and each of three tanks received 1.6 milligrams of strontium, applied as strontium nitrate, per gram of soil in lieu of calcium. Two tanks received spiked solutions applied at the surface with a sprinkler.

The average exchangeable potassium in 25 random samples of soil was 1.14 milligrams per gram of soil; the maximum was 1.25, and the minimum was 1.00. The soils were considered to have nearly uniform exchangeable potassium content.

The *p*H of the soils in the plots varied from 7.2 to 8.0. The *p*H of soil in two of the three control plots was 8.0; that of soil in the other plots varied in a random manner from 7.2 to 7.8.

Seed planted in soils containing more than 8 milligrams of calcium per gram of soil did not germinate uniformly; hence, to obtain uniform germination, untreated soil was overlayed to a depth of 2 inches on each tank, and this layer was reseeded to the respective crops.

The three crops grown in the 1957 season were lettuce, alfalfa, and mixed grass (*6*). These were randomly planted in replicate, except that only one control tank was used for each crop. Three crops were harvested; dry and ash weights were determined on samples of 100 grams cut weight per tank. All determinations were based on the ash weight of the sample. Plant ash was digested with 6*N* hydrochloric acid and made to volume in preparation for analyses.

Analyses were performed for calcium, strontium-90, and potassium by conventional methods. Available calcium in the soil samples was extracted with 1*N* ammonium acetate (*7*), and the respective concentrations were determined by flame photometry. Except for the plots to which strontium nitrate was added, all strontium data are given in counts per minute. The limit of detection of stable strontium by the method used is of the order of 2.5×10^{-3} milligram per gram. Strontium above this limit was not found in any of the soils. The amount of strontium added as strontium-90 was of the order of 10^{-13} gram per gram of soil.

Results

Ratios of strontium-90 per milligram of calcium in the plant were calculated and plotted against the concentration of calcium per gram of soil. Typical results are shown for two cuttings of lettuce in Fig. 1. It should be noted that in

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each case the concentration of strontium-90 based on plant calcium decreased as soil calcium increased. The first increments of soil calcium effect the most marked reduction in the strontium-90/calcium ratio in the plant; the curves break at about 4 to 6 milligrams of calcium per gram of soil and tend to level off after 9 milligrams of soil calcium has been reached.

The data in the middle section of Fig. 1 depict the results obtained from two cuttings of grass. The slopes of the curves are similar to those observed in the case of lettuce. It will be noted that grass accumulated a greater amount of strontium-90 per unit of calcium than did lettuce or alfalfa. This was especially true at the lower concentrations of soil calcium, where the strontium-90/calcium ratio in grass was 1.7 times that found in lettuce. The anomalous value at 18 milligrams of calcium per gram of soil is an average of four determinations and is believed to be real; it is possible that this high concentration of soil calcium may release strontium-90 from the soil colloid (this is discussed below).

The data relative to the adsorption of strontium-90 by alfalfa are presented in the right-hand section of Fig. 1. It is of interest that a lower strontium-90/calcium ratio is found in this plant than is found in lettuce or grass. The ratio is lower than that found in lettuce by a factor of 1.5 and lower than that found in grass by a factor of 2.5. The slopes of the curves are not as great as those found in lettuce and grass; this indicates that soil calcium has less effect in alfalfa upon adsorption of the radionuclide.

The calcium concentration in the crop ash also varied with the type of plant. It was higher in alfalfa than in grass by a factor of 10, and higher than in lettuce by a factor of 4.

The foregoing data indicate that increasing the concentration of soil calcium from 1.5 to 9.0 milligrams per gram of soil has a marked depressing effect on the concentration of strontium-90 per milligram of calcium in the plant.

Romney (*8*) has since indicated an inverse relationship between strontium-90 concentrations in the plant and available soil calcium.

Data were obtained from a comparable set of experiments (*9*) in which the soils were mixed with cesium-137, and the effect of potassium on this radionuclide was investigated. An increase, by a factor of about 2, in exchangeable soil potassium decreased the

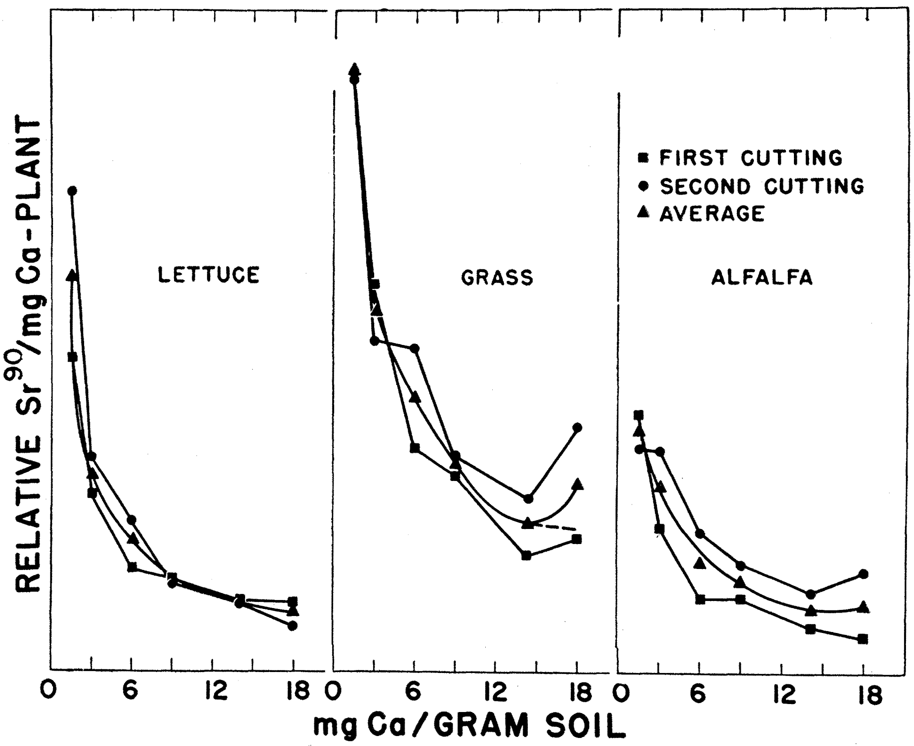

Fig. 1. Effect of increasing increments of soil calcium on strontium-90 uptake in plants.

cesium-137/potassium ratio in alfalfa by a factor of about 9.0. The reduction factors for lettuce and grass were about 5.

Libby (*3*) has reported a discrimination on the part of the plant against strontium-90. The curves in Fig. 1 suggest that discrimination is related to the type of plant and to the concentration of soil calcium. To determine the presence and extent of such a factor, the ratio strontium-90/milligram of calcium (soil) was divided by the ratio strontium-90/milligram of calcium (plant) for the three crops; the data were treated statistically. The results fell into three groups—roughly, high calcium, low calcium, and an intermediate range; these are presented in Fig. 2.

Discrimination against strontium-90 per unit of calcium added was most marked with the three crops when calcium concentrations were low. Lettuce and alfalfa demonstrated discrimination against strontium-90 at soil calcium levels up to 13 milligrams of calcium per gram of soil. However, at concentrations of 18 milligrams of calcium per gram of soil, the plants appeared to discriminate against calcium; this is especially true of alfalfa and grass. It is possible that the high concentration of soil calcium may release some of the strontium-90 which is bound to the soil colloid and thus make it more readily available to the plant. Other data indicate a possible disturbance of the plant's physiology, lettuce being more tolerant to calcium than is grass. It should be stressed that these data show trends and are not meant to imply absolute values. However, it is indicated that the type of plant as well as the calcium level is associated with discrimination against strontium.

Stable strontium nitrate was substituted for calcium in three plots at a level of 1.6 milligrams of strontium per gram of soil. The calcium level in the strontium-treated soils was comparable to that in the low-calcium soils. It was believed that the addition of stable strontium would decrease the strontium-90 uptake by physical dilution, and hence apparently increase the discrimination against strontium-90 by the plant. However, in the case of lettuce the discrimination against strontium in the stable-strontium plots was very similar to that obtained for the low-calcium soils (Fig. 2). When discrimination factors were calculated for alfalfa and grass grown in stable-strontium tanks, the factors were lower than similar factors obtained on low-calcium plots. Romney (*10*) has since observed a similar phenomenon with Ladino clover and attributes this to replacement of the strontium-90 on the soil colloid by stable strontium. As suggested above, a similar reaction may take place when the soil colloid is flooded with calcium, and thus in the soils where a large amount of calcium was added, the strontium-90 may have been released from the colloid and become available to the plant. Such a phenomenon could account for the decrease in discrimination factors with increase in soil calcium, as seen in Fig. 2, and for the apparent lack of a dilution of strontium-90 by stable strontium.

The highest discrimination (about 2.9) against strontium-90 was shown by alfalfa (Fig. 2), and the least discrimination (about 1.6) was shown by lettuce. It is interesting to contrast these values with those obtained for cesium-137 in a separate study (*9*). Alfalfa again discriminated to the greatest extent, but in this case the value is about 1150; grass discrimination is least, about 750. These marked differences are attributed to the greater binding of cesium to the soil, which makes it less available to the plant. Nishita *et al.* (*11*) also have since attributed the low uptake of cesium-137 by plants from soils to the nonavailability of the radionuclide.

In one case, calcium chloride was replaced by calcium carbonate to the extent of 1.5 milligrams of calcium per gram of soil, and the plot was planted to lettuce. This calcium did not have an effect comparable to that produced by calcium chloride. It is believed that the salt was not solubilized in this alkaline soil and thus that the calcium was not available to the plant. It is probable that calcium carbonate applied to an acid soil would be solubilized and would produce an effect comparable to that demonstrated with calcium chloride.

The results presented thus far have been concerned with the effect of calcium concentration on uptake of strontium. The soils were also spiked with cesium-137. The tanks which received a surface spike of cesium-137 yielded interesting preliminary data with regard to the zone of feeding of the plant, since cesium does not move easily through the soil.

In the tanks where cesium-137 and strontium-90 were applied as surface spikes, the average count of these radionuclides per gram of alfalfa ash from the first and second cuttings was only one-seventh the average for plants grown in similar tanks which were uniformly spiked. It would appear that the zone of feeding is associated with the area of root growth; this area moves progressively downward with the development of the plant. The implication with regard to fallout is important, since it has been suggested that fallout is presently concentrated within 2 to 4 inches of the surface (*12*). It was pointed out above that seeds were planted in a layer of untreated soil 2 inches deep. Further evidence supporting the importance of depth of feeding is offered by the observed concentration of radionuclide in the first and second cuttings. Since these concentrations were essentially the same after 5 to 6 weeks of growth as after 12 weeks of growth, it is apparent that the plants were feeding below the 2-inch untreated soil level at an early period in their development. The 2½-inch depth figure commonly occurs in the literature. It is our opinion that when strontium-90 concentrations in this depth are used to calculate discrimination factors, the results may be misleading. From our results we believe that absolute discrimination factors may actually be less than those reported. Menzel (*13*) has reported a Ksr equal to 0.38, which we believe translates to a discrimination factor, as calculated by us, of about 2.8. However, his results were obtained with bomb debris, part of which might be unavailable to the plant, and this would influence the calculation.

At the time of the third cutting, there was some evidence of leaching of calcium from the high-calcium plots, due to heavy rains. An analysis for available calcium at depths of 4, 8, 12, and 18 inches indicated that there was in fact some movement. It is of interest that some strontium-90 did move through the soil under these conditions, as was shown by depthwise assay of soil cores

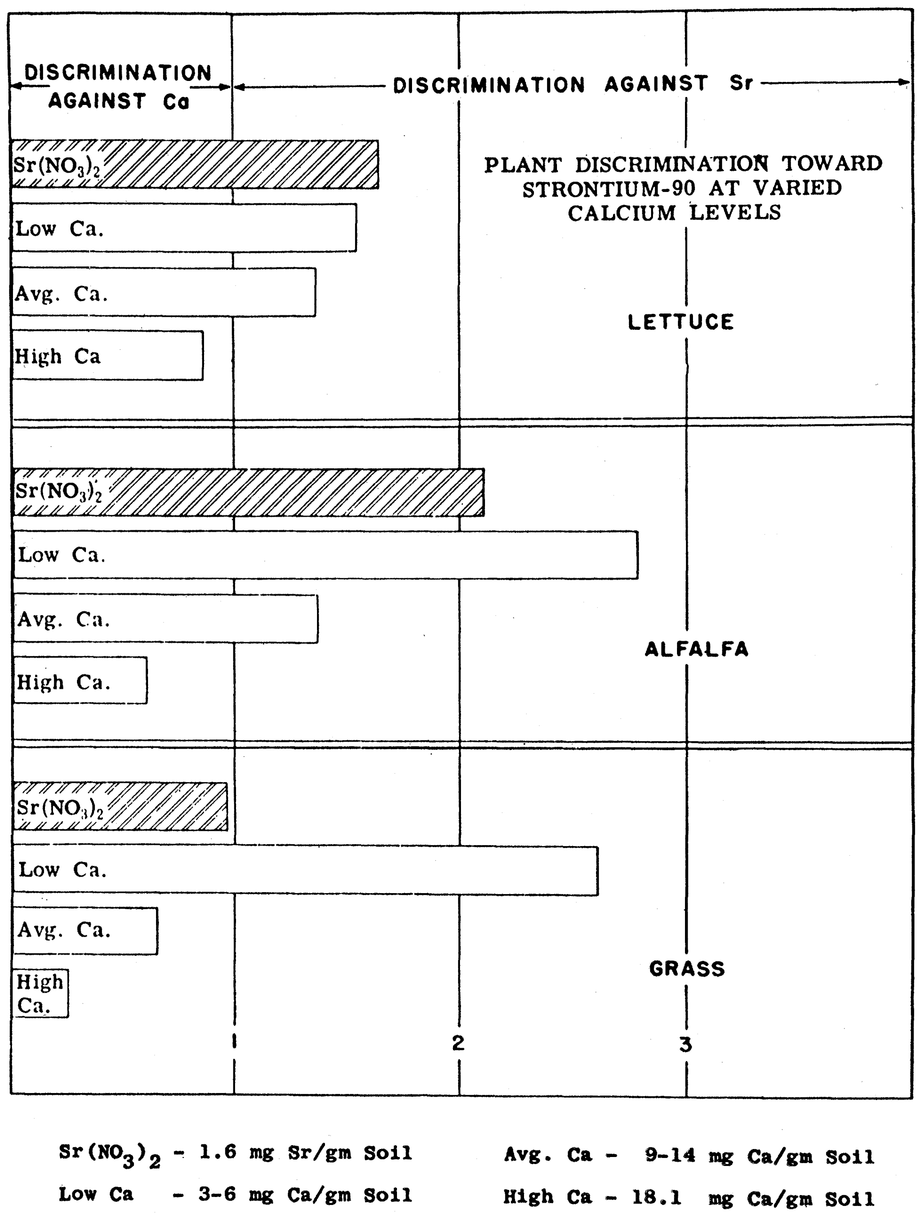

Fig. 2. Discrimination against strontium-90 in lettuce, alfalfa, and grass at various concentrations of calcium.

from plots to which a surface spike had been applied. However, attempts to repeat these results in the laboratory on 1-inch soil columns of varied length did not show a pattern of movement similar to that obtained in the field. Data indicate that high calcium in the soil increases the rate of leaching of strontium.

The ratio of strontium-90 to calcium in the plants from the third cutting plotted against the soil calcium found gave curves having the same general slope as those in Fig. 1, with the first increments of calcium again having the most marked depressing effect on the uptake of strontium-90. The observed movement of calcium at the time of the third cutting and the effect of high soil calcium on the movement of strontium-90 are of interest from the standpoint of fallout movement in soils. Because the movement of both calcium and strontium through the soils *after* the second cutting made calculations difficult, data obtained from the third cutting analyses are not included in the results or conclusions regarding the depressing effect of soil calcium on the uptake of strontium-90 by the three crops.

A greater reduction of soil calcium by leaching shortly before the third cutting was observed under lettuce than under grass. Such a relationship to the rate of water penetration would be expected. Leaching of calcium under the three types of cover follows the pattern: under grass < under alfalfa < under lettuce. Hence, both the concentration of soil calcium and the type of cover may affect the depth of penetration of the strontium-90. Other factors of importance in the movement of strontium-90 downward through the soil would include the exchange capacity of the soil for strontium-90, the soil *p*H, and the type of soil colloid. An attempt was made in this work to hold these factors constant through the use of a single soil type.

Libby (*3*) states that a lack of detailed correlation between strontium-90 in the soil and in alfalfa from samples collected on 11 midwestern farms is probably due to direct fallout on the plant. Libby's data do not indicate the calcium content of the soil upon which alfalfa was grown or the age of the plant. It is possible that alfalfa containing low concentrations of strontium-90 was harvested from old plants which were deep-feeding and, hence, contained a low concentration of strontium-90. Foliar absorption indeed plays an important role in concentration of strontium-90 in plants; however, the erratic concentration of strontium-90 observed by Libby might be accounted for in part by soil calcium concentration or depth of feeding and not entirely by foliar absorption.

Conclusions

1) The adsorption of strontium-90 by lettuce, grass, and alfalfa is progessively inhibited by increasing concentrations of soil calcium.

2) The inhibition is more marked in the case of lettuce and grass than in the case of alfalfa.

3) Lettuce and alfalfa apparently discriminate against strontium to a greater extent than does grass.

4) The discrimination noted against strontium (up to 2.8) is dependent upon the concentration of soil calcium.

5) Cesium-137 is removed by plants from soil to a lesser degree than is strontium-90. Apparent discrimination against cesium may be as great as 1100 and is dependent upon soil potassium and soil type. This high value may be due, in part, to the binding of cesium by the soil and to the consequent nonavailability of cesium to the plant (*9*).

6) The depth of feeding of the plant is an important factor in the natural adsorption from the soil of radionuclides arising from fallout.

Determination of the Over-all Ratio in Man

The determinations for a discrimination factor for or against strontium-90 as described in this article and reported by Libby (*3*) and Menzel (*13*) are quite close (1.0 to 1.4 against strontium-90), if one assumes that the strontium-90 in the bomb debris used by Menzel is only 50 percent available to the plant. However, we feel that these figures are of little or no value for determining the over-all ratio of strontium-90/calcium in soil to strontium-90/calcium in bone.

The reasons for this statement are fourfold. First, the root-feeding zone is, in all probability, much deeper than the strontium-90, which has been reported to be almost entirely in the top 2½ inches of soil. This factor is further complicated by the fact that root depth varies from 1 foot to more than 20 feet, according to the type and age of the plant in question. For example, the discrimination against strontium-90 was shown to be 1.5 in the case of alfalfa grown in a tank in which the strontium-90 was uniformly mixed with the soil. For alfalfa grown in a comparable tank with the strontium-90 added as a surface spike, the figure, calculated on the basis of the concentration of strontium-90 in the top 4 inches of soil, becomes 8.2. Second, it has been demonstrated that the soil calcium markedly affects the uptake of strontium-90, and this factor also varies over an extremely wide range. Third, foliar absorption perhaps plays an important though unknown role. This would be particularly true of deep-rooted plants, such as alfalfa, which feed largely in an area free of strontium-90. Much work remains to be done on this phase of the study. Fourth, specific land areas necessarily have a marked effect on the amount of strontium-90 taken up by a plant. Libby points out that arid regions will have little fallout and hence, very little foliar absorption, and that if the plants are irrigated, the irrigation water will contain little strontium-90, since much of the nuclide will have been previously adsorbed on soils contacted at the time of runoff.

It is quite apparent from the foregoing discussion that any determination of the over-all ratio from soils is going to be fraught with difficulties. It is entirely possible that average plants might better be used than soils as a base for determination of the over-all ratio, and it is becoming increasingly apparent that no matter how the over-all ratio is determined, one will be extremely fortunate to determine it within an order of magnitude (*14*).

Added Comments

A report of the 1957 hearings before the Joint Committee on Atomic Energy (*15*) has been published since completion of the work described above. The four factors presented here were discussed at some length by the committee and the attending scientists.

Since this work was first reported several other reports have appeared in the literature. They are briefly reviewed here.

Middleton (*16*) has pointed out that foliar absorption of cesium-137 and strontium-90 takes place to varying degrees according to the stage of maturity of the plant and the plant type. In the case of wheat, rainfall may remove as much as 85 percent of the applied isotope. In all cases, the higher percentage of applied strontium-90 was found to be associated with the leaves and stems of the plant.

Prout (*17*) has indicated the effect of *p*H and concentration on the ion-exchange capacity of a particular soil for strontium. Absorption reached a maximum at *p*H 7.0 and at lower concentrations of strontium. He has also indicated a decrease in absorptive capacity with increase in salt concentration. We had pointed out previously (*18*) the inverse of this reaction—that is, the desorption of strontium from tuff and soils by various salt solutions.

Nishita *et al.* (*11*), in an excellent study, have shown a utilization of nonexchangeable potassium by Ladino clover. They also point out that, as exchangeable potassium decreases in the soil, the amount of cesium-137 fixed by the soil increases. Evidence is presented for increased cesium-137 uptake as exchangeable soil potassium decreases due to continued cropping.

Romney *et al.* (*10*) have shown that stable strontium releases strontium-90 to the soil solution so that strontium-90 becomes more available to the plant up to applications of about 5 tons of strontium per acre. No practical dilution effect of strontium-90 by stable strontium was shown.

Romney *et al.* (*8*) have studied the uptake of strontium-90 and four other radioisotopes by five different plants from seven different soils. Strontium-90 was very significantly taken up by all plants. Both plant and soil effects were noted. These authors also point out an inverse relationship between strontium-90 in the plant and available calcium in the soil.

Klechkovsky (*5*) has reviewed extensively the work on behavior of fission products in soil in the U.S.S.R. He also points out the lack of cesium-137 uptake from soil by plants and notes an inverse relationship between concentrations of calcium in soil and of strontium-90 in plants. Of interest is his report of marked increase in the uptake of cesium-137 when plants are grown hydroponically. These results further indicate nonavailability of soil cesium and are attributed to the strong bond between cesium and the soil colloid.

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Gilbert Morgan Smith, Botanist

Unlike many scientists, Gilbert Morgan Smith, who was born in Beloit, Wisconsin, 6 January 1885, gave no indication of his future occupation in his boyhood hobbies and activities. Indeed, despite the fact that he came from an academic family—his father, Erastus Gilbert Smith, was professor of chemistry at Beloit College—Gilbert's career as a scholar began inauspiciously. He had difficulty with his studies (particularly the classics), and his pranks at Beloit High School were far more memorable than his academic record. Many of the present generation of botanists who were brought up on the two volumes of the scholarly *Cryptogamic Botany* will be shocked to learn that its author was almost expelled from high school for putting asafetida in the heating ducts. Through the influence of his father, he was accepted at Williston Academy, where the perceptive headmaster soon discovered his aptitude for science. From that time on, his progress was so rapid that his undergraduate record qualified him for election to Phi Beta Kappa when a chapter was established at Beloit.

After graduating from Beloit College in 1907, he taught science in high school in Stoughton, Wis., for a year before he began graduate study at the University of Wisconsin. He had intended to become a chemist, but while preparing for language examinations he became so fascinated with the algae that he decided to work in botany under C. E. Allen. He obtained his Ph.D. in 1913 and was married that same year to Helen Pfuderer, who shared his career from that time on. He remained on the staff at Wisconsin and moved steadily upward in academic rank. In 1923 he was invited to Stanford University for the autumn quarter, and two years later he succeeded Douglas Houghton Campbell as professor of botany at Stanford. He became emeritus professor of biology (botany) in 1950 but remained active scientifically until he died, on 11 July 1959.

Smith was accorded many honors. He was elected to the National Academy, the American Academy, Phi Beta Kappa, Beta Theta Pi, and Sigma Xi. Beloit College recognized his ability