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THE EXTENT AND CONSEQUENCES OF THE UPTAKE BY PLANTS OF RADIOACTIVE NUCLIDES¹

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INTRODUCTION

The title of this review was chosen by the Editors, who indicated that it should be interpreted broadly; discussion should not be limited to problems of plant physiology. There are obvious reasons for this directive. The wide attention which has lately been given to the entry of radioactive nuclides into plants is due neither to their role in metabolism nor to their effects on plant growth; the predominant source of interest is the internal radiation dose which they may deliver to man. The words "extent" and "consequences" in the present title have little meaning outside this context. Because the biological effects of radiation from all sources are additive, the significance of the radiation dose which man receives from radioactive materials which pass through food chains into his diet can be judged in perspective only when compared with doses from other sources to which he is simultaneously exposed. The first part of this review is therefore devoted to a brief summary of current knowledge on the sources and extent of radiation to which the population lately has been exposed. This will enable problems of plant physiology and soil science, which are of major practical importance, to be selected for subsequent discussion.

This approach should not be regarded as suggesting that the behaviour of individual radioactive substances in plants is not of considerable plant physiological interest. Rather it takes account of the fact that, in the terminology of a recent review of the organisation of research, investigations in this field at the present time can usually be described as "objective basic research" (1). This term is used in contra-distinction to "pure basic research." The difference between these two categories lies in that whereas the latter is pursued solely because of its intrinsic scientific interest, the former though frequently of equal interest, and equally demanding, in addition provides information which serves a specific practical end, and its pursuit is planned with this end in view.

SOURCES AND MAGNITUDE OF ENVIRONMENTAL RADIOACTIVITY

Natural sources.—Our knowledge of the natural sources of radiation to which man is exposed has increased considerably in recent years. The sec-

¹The survey of literature pertaining to this review was concluded in October 1962.

ond Report² of the *United Nations Scientific Committee on the Effects of Atomic Radiation* (2) contains the most comprehensive estimate which has yet been made of the average dose experienced by the world population. Their findings are summarised in Table I, column 1.

External radiation is received both from cosmic rays and from terrestrial sources, which consist of both the decay products of the uranium and thorium series and potassium 40 in the earth's crust. It is of interest that the dose from cosmic rays estimated in the U.N. Report (2) is appreciably higher than that which was assumed until recently; this is because the significance of the neutron component of cosmic rays has but lately become apparent. Whereas all tissues are exposed to a relatively constant radiation dose from external sources, that from ingested materials may vary widely dependent on their distribution between organs. Thus, the major dose from the uranium and thorium series (due mainly to radium and its daughter products) is received by the bone.

The average dose from all natural sources to different tissues is estimated as between 120 and 130 mrems per year.³ Considerable variations in exposure, however, occur between different localities on the earth's surface. The dose from cosmic rays, for example, rises both with increasing altitude and, at the same altitude, with distance from the equator. The dose from radioactive substances in the earth's crust is subject to even greater variations. In granitic areas, exposure may be several times above the average and appreciable differences can be caused by the choice of building materials (3). In exceptional areas of extremely high background, values of several 1000 mrems per year are known. The internal dose from the radium and thorium series varies also.

Man made sources.—The sources of extra radiation which man has created are, for present purposes, conveniently divided into those resulting from the contamination of the environment with the radioactive substances and those due to other causes. The latter group will be considered first; diagnostic radiology is the predominant source. Estimates of the mean dose to the populations in different countries where modern medical procedures are employed range from 10 mrems per year in Norway to 58 in France (2). Much smaller exposure comes from such sources as luminous watches, shoe-fitting equipment and television sets.

²This Report is an exhaustive source of information on all aspects of man's exposure to radiation. It contains extensive annexes and bibliographies covering not only medical questions but also natural radioactivity and the behaviour of radioactive substances in soils, plants and animals. The existence of this document makes it unnecessary here to give a comprehensive coverage of the literature, except on topics which have been selected for special consideration.

³The relative biological effective dose of radiation is usually measured in *rems*. Doses in *rems* are calculated by multiplying the absorbed dose in *rads* (1 rad is 100 ergs absorbed per gm) by the RBE (relative biological effectiveness) for the type of radiation in question. The RBE for beta and gamma radiation is 1, that for alpha and neutron radiation is taken as 10 (2).

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om cosmic rays and from terrestrial decay products of the uranium and earth's crust. It is of interest that the U.N. Report (2) is appreciably different recently; this is because the significance of cosmic rays has but lately become related to a relatively constant radiation level from ingested materials may vary widely in different organs. Thus, the major dose from radium is mainly to radium and its daughter

sources to different tissues is estimated per year.³ Considerable variations in dose exist in different localities on the earth's surface. The dose from cosmic rays rises both with increasing altitude and with distance from the equator. The dose from radium is subject to even greater variations. It may be several times above the average dose received by the choice of building materials. In areas of extremely high background, values of the internal dose from the radium

of extra radiation which man has received is conveniently divided into those resulting from natural sources and those from man-made sources. The former will be considered first; the latter later. Estimates of the mean dose from natural sources where modern medical procedures are used range from 100 per year in Norway to 58 in France (2). Other sources as luminous watches, shoe-

For information on all aspects of man's exposure to radiation, annexes and bibliographies covering not only the activity and the behaviour of radioactive substances but also the existence of this document makes it unnecessary to refer to the literature, except on topics not covered here.

The unit of radiation is usually measured in *rems*. The equivalent of the absorbed dose in *rads* (1 rad is 100 *rems* for the type of radiation) for the type of radiation is 1, that for alpha and

The extent of environmental contamination with world-wide fallout varies both with latitude (since the majority of weapons trials have been in the Northern Hemisphere) and with climate, since fallout is deposited mainly, though not entirely, in rain. The internal dose from ingested radioactive materials is also influenced by agricultural factors and by the choice of food. The evaluation of man's exposure to radiation from fallout is further complicated by the fact that whereas some components of the dose are received for a short period only, others will be experienced for many years. Accordingly it is appropriate to consider not only the annual exposures which have been received hitherto, but also the continuing level to which the population has been committed. Annual dose rates in the United Kingdom in the middle of 1959 are shown in Table I, column 2. That time is selected because the highest exposures from the early series of weapons trials were then experienced; the choice of the United Kingdom as an example rests partly on the availability of data and partly on the fact that it lies in a latitudinal band of relatively high deposition. Table I, column 3, based on the U.N. Report (2), shows the estimated exposure of the world population from weapons tests up to 1961, averaged over the years 1954 to 2000 AD; in column 4 of the table the fraction of the dose from these tests which will not be received until after 2000 AD is shown.

Iodine 131 is not considered in Table I, since the average exposure of the population gives little indication of its significance. This nuclide is of concern primarily as a source of exposure of infants who consume appreciable quantities of fresh milk, partly because of the very small size of their thyroid glands in which it is concentrated, and partly because milk is usually the most highly contaminated food. Doses to infants from iodine 131 have on occasions been considerably higher than those from any other component of fallout; for example, towards the end of 1961 it was estimated from the analysis of milk (5) that the thyroid glands of infants fed on fresh milk in the United Kingdom would have received about 170 mrem (2).

Table I shows that the doses from fallout have hitherto been small relative to those from natural sources; they are less even than that from diagnostic radiology. However, since the present evidence indicates that all exposure to radiation may be deleterious, these relatively small additions to man's exposure cannot be ignored. Furthermore the possibility of significantly higher levels of exposure in the future cannot be excluded. Nonetheless, the realization that past doses from fallout have been smaller than those accepted unknowingly by persons who move from areas of low to higher natural background, can aid a rational approach to new problems.

Ingested materials are the main sources of exposure from fallout and, of these, strontium 90, caesium 137 and carbon 14 are the most important. Carbon 14, though currently delivering relatively small doses, is, because of its long half life (5570 years), a continuing source of exposure. The total dose commitment from this nuclide released by past weapons trials should be approximately equal that from all other sources of fallout (2).

TABLE I

COMPARISON OF PRINCIPAL DOSE RATES FROM NATURAL BACKGROUND AND
WORLD-WIDE FALLOUT DOSE RATES IN MREM/YEAR¹

	Natural back- ground	World-wide fallout from nuclear weapons tests up to 1961		
		dose rate July 1959 UK only	average annual dose rate 1954-2000 AD	fraction of total dose not delivered until after 2000 AD
<i>External radiation to whole body:</i>				
Cosmic rays	50	—	—	—
From radioactive substances	50	4.2	0.65	3%
<i>Internal sources:</i>				
To whole body (range for differ- ent tissues):				
Potassium 40	15-20	—	—	—
Carbon 14	1.2	0.2	0.16-0.26	90%
Caesium 137	—	1.5	0.24-0.42	Nil
To new bone:				
Uranium & thorium series	13	—	—	—
Strontium 90	—	8.1	1.6	9%
To bone marrow (blood forming organs):				
Uranium & thorium series	5	—	—	—
Strontium 90	—	2.7	0.81	9%
Total dose (range for different tissues)	120-130	6-14	1.3	—

Notes: Column 1: world average from UN Report (2), p. 21.

Column 2: based on Loutit *et al.* (4).

Columns 3 & 4: world average based on UN Report (2), p. 27.

Fallout from nuclear weapons is, however, not the only source of environmental contamination; releases from nuclear reactors may also occur. In the normal operation of such establishments, iodine 131 is likely to be the most significant component released into the atmosphere. Dilute radioactive effluent containing both fission products and induced radioactive substances is usually discharged, but at controlled levels which make it an insignificant source of exposure to the population. Accidents at nuclear reactors may give rise to considerably higher levels of local contamination. Both past experience and theoretical studies indicate that internal and not external radiation will be the main risk to the population; iodine 131 is

again likely to be the dominant source of concern, followed probably by strontium 90 (6, 7).

Characteristics of radioactive substances which cause them to be important sources of internal radiation.—The relative importance of individual radioactive nuclides as sources of internal radiation depends on many factors apart from the quantities in which they are released into the environment. Of these the major are: (a) the extent to which they are transferred through food chains into diet; (b) the extent to which they are absorbed from the gastrointestinal tract into the body; (c) the extent to which they are accumulated and retained in tissues; (d) their half-lives; and (e) the type and energy of the radiation they emit.

In practice the nuclides which are readily absorbed from the gastrointestinal tracts of animals are either isotopes of elements important in metabolism or closely similar to them. Potassium 40 and caesium 137, both alkali metals, are absorbed and circulate freely throughout the body and, because of the gamma radiation they emit, irradiate all tissues; they are potential sources of genetic injury. Iodine 131 accompanies stable iodine to the thyroid gland. Strontium 90 and radium, both alkaline earths like calcium, pass with it to the bone. By virtue of their long biological retention and long half-lives (strontium 90, 27 years; radium 226, 1620 years; radium 228, 6.7 years) the bone and the bone marrow are irradiated for long periods. Carbon 14 becomes distributed throughout all living tissues. Because the basic nature of metabolic processes in plants and animals is similar, nuclides which are readily accumulated in animal tissues are usually those which pass most freely through food chains.

Of the nuclides mentioned in the previous paragraphs, iodine 131 and potassium 40 can be excluded from the present discussion. The short half life of iodine 131 (8 days) makes its absorption by plants of little importance; it enters food chains mainly through the direct contamination of plants, lodging on them in a manner similar to strontium 90 (8). Although potassium 40 is the main source of internal radiation to which man has yet been exposed, the considerable literature on the absorption of this element makes unnecessary its inclusion in this review. Attention will therefore be given mainly to strontium 90, caesium 137, radium and other members of the uranium and thorium series, and carbon 14. Other nuclides, however, cannot be always ignored; they are considered briefly in the final section.

STRONTIUM 90

This nuclide has received considerably more attention than any other in food chain studies. Before its behaviour had been studied experimentally it was natural to speculate on the basis of its chemical similarity to calcium. From some respects this analogy was misleading. It undoubtedly encouraged the belief, now disproved, that strontium 90 would always enter man's diet mainly, like calcium, from the soil. The comparison of the two elements, however, led also to the view, since amply vindicated, that the major sources

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FROM NATURAL BACKGROUND AND RATES IN MREM/YEAR²

World-wide fallout from nuclear weapons tests up to 1961		
dose rate July 1959 UK only	average annual dose rate 1954-2000 AD	fraction of total dose not delivered until after 2000 AD
—	—	—
4.2	0.65	3%
—	—	—
0.2	0.16-0.26	90%
1.5	0.24-0.42	Nil
—	—	—
8.1	1.6	9%
—	—	—
2.7	0.81	9%
6-14	1.3	—

Report (2), p. 21.

on UN Report (2), p. 27.

however, not the only source of en- from nuclear reactors may also occur. blishments, iodine 131 is likely to be ed into the atmosphere. Dilute radio- products and induced radioactive sub- controlled levels which make it an in- population. Accidents at nuclear re- higher levels of local contamination. studies indicate that internal and not risk to the population; iodine 131 is

of calcium in man's diet would in general also be the main sources of strontium 90, water being relatively unimportant; thus, for example, in European countries where milk is the main dietary source of calcium, it is the main source of strontium 90 also (2). The importance of studying the soil-plant-animal food chain was thus recognised. The amount of attention which has been given to strontium 90 from this viewpoint cannot, however, be explained solely in terms of the magnitude of the radiation dose it delivers, relative to that from other sources. The food chains whereby it enters man's diet merit greater study than those for many other nuclides because they are more complex and variable. There are two main reasons for this. Firstly, strontium is considerably more readily absorbed from the soil so that its effects can be assessed adequately only on the basis of its behaviour over very long periods. Secondly, like calcium it moves only basipetally throughout plants (9, 10, 11); thus strontium 90 which is absorbed into an individual leaf or other above ground tissue is not appreciably transferred to other organs. Accordingly, the levels of strontium 90 in tissues which man consumes cannot be inferred from the total quantity which the plant absorbs; very different situations may occur, depending on whether entry occurs into roots or through the aerial tissues. This question is of much less practical importance with nuclides such as caesium 137, which are freely redistributed.

Relationships between strontium and calcium.—The currently accepted terminology for defining relationships between strontium and calcium was originated by Comar *et al.* (12) who introduced the term "Observed Ratio" of strontium to calcium (OR) as defined by the equation:

$$\text{OR (sample/precursor)} = \frac{\text{Ratio: Sr/Ca in sample}}{\text{Ratio: Sr/Ca in precursor}}$$

In practice the transfer of the two ions through any biological process which can be studied experimentally, i.e., from diet to bone, or from the soil to the ear of a cereal plant, involves a number of successive steps; the term "Discrimination Factor" was introduced to define the relative behaviour of the two ions in each such step, the "OR" of the entire process being the product of the relevant discrimination factors.

Although widely and profitably used, difficulties have sometimes arisen in the use of this terminology. One cause has been disregard of the precise definition of the OR. Comar *et al.* (12) laid emphasis on the fact that the OR could be validly estimated only if the ratio in which the two ions were available for entry into the biological system was known, as well as the ratio within it. It is difficult to make precise estimates of the relative availability of ions in the soil and misleading conclusions as to the magnitude of the OR "plant/external medium" have on occasions been reached through the assumption that the ratio of added tracer strontium to "exchangeable" calcium in the soil validly reflects the ratio in which the two ions are

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available to plants (13). A second cause of difficulty in the application of the OR concept, which may legitimately be ascribed to over-enthusiasm in its use, has been the assumption that the OR for a given process should be numerically constant over all circumstances. The inadequacy of this view has been demonstrated particularly in animal studies which show that changes in discrimination may occur with age or from grossly abnormal physiological conditions such as can readily be arranged in the laboratory. These variations do not, however, significantly reduce the value of the concept for assessing relationships in normal circumstances. In particular the consideration of the two ions conjointly is appropriate for assessing the effects of the dietary intake of strontium 90 by man or animals. The level of radiation to which the body is exposed depends on the concentration of strontium 90 in bone; in practice this is determined by its ratio to calcium. Since the OR (bone/diet) is relatively constant (*ca.* 0.25) it follows that the radiation dose varies with the ratio of strontium 90 to calcium in the total daily intake (2).

The OR (plant shoot/rooting medium) is close to one (2, 14 to 18). Numerous investigations show that variations in the extent to which different species absorb strontium from uniformly contaminated soil are closely paralleled by variations in their absorption of calcium (19 to 22) though some small divergences have been observed (2, 22). More detailed studies of the relative behaviour of the two ions in plants, however, reveal an appreciable differential movement. When both ions enter through roots their ratio in the aerial organs most remote from the root is in general lower than the average in the plant. Varying relationships occur depending possibly on growth rate but the ratio of strontium to calcium in vegetative tissues is frequently about half that in underground storage organs and twice that in grain and seeds (21). Stems may show considerably higher ratios than other above ground tissues (13). The most detailed comparisons have been made between different tissues of wheat grain; the ratio in the endosperm has been found to be 0.7 of that in whole grain (23).

It appears, therefore, that the two ions interact in a series of exchange reactions in their upward passage through plants, strontium being more firmly retained. The possible nature of sites involved has been discussed by Biddulph *et al.* (24). The fact that despite this discrimination, the ratio of strontium to calcium in shoots is close to that in the outer medium appears to reflect the fact that the major part of both the calcium and the strontium in plant shoots is usually located in the foliage. The discriminatory mechanism in the stem may thus be likened to an ion exchange column which has "overrun." Although differential retention occurs in the column, the ratio of ions in the effluent becomes similar to that in the added solution after saturation.

These discriminatory processes have little effect on the ratio of strontium 90 to calcium in man's diet. The majority of dietary calcium comes directly,

or indirectly via milk, from leaf tissues in which the ratio of the two ions is similar to that in the rooting medium.

The extent to which stable strontium influences the relationship between strontium 90 and calcium has been considered. It has been shown both in water culture and in soil studies that variations in the concentration of stable strontium do not affect the absorption of strontium 90 provided that the ratio of stable strontium to calcium is low (10, 25, 26). Entry appears to be controlled by the total concentration of strontium plus calcium. Since in nature the ratio of stable strontium to calcium is usually 1 to 100 or less, and high concentrations of stable strontium are toxic, the carrier ion can be ignored in studies of plant/soil relations.

Direct contamination of plants.—The term "direct contamination" is used to describe all material which enters plants as a result of lodging on their above ground tissues, in contradistinction to absorption from the soil. Translocation within plants may lead to the direct contamination of tissues which are remote from the site of initial lodgement.

One of the most important and interesting aspects of food chain studies has been the investigation of the relative extent to which strontium 90 can enter plants by direct contamination under conditions of continuous deposition such as occur with world-wide fallout. The amount which enters in this way is determined by the deposit only in the recent past while absorption from the soil will be related to the cumulative total therein. Without knowledge of the relative magnitude of these two processes it was impossible to form any valid view on the long term consequences of world-wide fallout. The prediction of the manner in which dietary contamination would change after a massive accidental release, of short duration, also depends on the same information.

It has been found necessary to distinguish three types of direct contamination (27, 28) namely: (a) foliar contamination: retention by leaves, (b) floral contamination: retention by inflorescences, (c) plant base contamination: retention and absorption by the basal parts of plants, or surface roots, of material which has not been incorporated into the soil.

Strontium 90 which reaches plants by the first two routes will not be accompanied by calcium; some calcium may enter simultaneously with strontium 90 from the plant-base but to a much smaller extent than when strontium 90 is absorbed from the soil. The relative importance of these alternative routes of entry depends on the growth form of plants. Whereas foliar absorption can occur in all species, floral absorption is of practical significance with grain crops only; plant-base absorption is particularly important in perennial pastures though not necessarily confined to them. It is often difficult to define sharp boundaries between foliar absorption and either floral or plant-base entry. During rain the downward washing of contamination over the surfaces on the plant will occur concurrently with absorption; thus foliar and floral absorption merge into plant-base entry. Likewise,

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The term "direct contamination" is used to describe plants as a result of lodging on the ground. Distinction is made between direct contamination of tissues and external lodgement.

Interesting aspects of food chain studies are the relative extent to which strontium 90 can enter under conditions of continuous deposition and fallout. The amount which enters in this way in the recent past while absorption is still low is a cumulative total therein. Without knowledge of these two processes it was impossible to estimate the consequences of world-wide fallout. The extent of dietary contamination would change with the duration of short duration, also depends on the

to distinguish three types of direct contamination: (a) retention by leaves, (b) by inflorescences, (c) plant base contamination by the basal parts of plants, or surface contamination incorporated into the soil.

Contamination by the first two routes will not be as important as calcium may enter simultaneously with strontium to a much smaller extent than when strontium enters from the soil. The relative importance of these routes depends on the growth form of plants. Whereas in some species, floral absorption is of practical importance, plant-base absorption is particularly important and not necessarily confined to them. It is the boundary between foliar absorption and plant-base absorption and during rain the downward washing of contaminants will occur concurrently with absorption. Both routes merge into plant-base entry. Likewise,

as materials are leached from the plant-base zone into the soil, plant-base absorption gives place to indirect entry through roots.

Whereas the extent of foliar or floral absorption will depend on the quantity of fission products deposited during the growth of the individual leaf or inflorescence, material may be absorbed from the plant-base reservoir after it has been retained there for an appreciable period of time. This occurs particularly with perennial plants which are the main components of many pastures.

The extent of direct contamination is also influenced by the physical form of the deposit, especially its particle size and its solubility. Large particles usually rebound from leaf surfaces, being retained only if they lodge in re-entrant angles; it has been found that, in general, particles in excess of about 40μ in diameter are little retained (28). Particles considerably in excess of these dimensions will occur in the near-in fallout from nuclear weapons but not at considerable distances. Insoluble materials are a source of superficial contamination only. The greatest contamination of plants will therefore be caused by finely divided soluble deposits; world-wide fallout is in this form. It is important to realise that the findings on its behaviour are not applicable to large, sparingly soluble particles, which may be the major constituent of near-in fallout.

Perennial pastures—The mechanisms responsible for the contamination of pastures have received particular attention because of the importance of milk as a source of strontium 90 in diet. Information has been obtained both from experiments and from widespread surveys of strontium 90 in milk; since the OR (milk/diet) is about 0.11 (2, 19) changing levels in grass can be reasonably inferred from those in milk.

Observations at nuclear weapons trials and experiments both in the laboratory and in the field suggest that 20 to 30 per cent of finely divided deposits or solutions may be initially retained on the edible tissues of pasture 4 to 6 inches high (29, 30). Bartlett *et al.* (31) have analysed the results of a series of experiments in which strontium 89 was applied as a fine spray to pastures which were exposed to the normal summer climate of the United Kingdom after the spray had dried. Losses were variable, but the quantity remaining on the grass could be expressed by a simple exponential relationship of the form $y = e^{-kx}$, x being the number of days after spraying and y the fraction of the initial deposit which remained on the herbage. The value of the constant k was computed as 0.05 with the standard error of ± 0.02 ; thus some 50 per cent was on average lost within 14 days and about 90 per cent after 50 days. It appears that strontium is removed from leaves to about the same extent as other carrier-free nuclides (8) and the extent of loss computed by Bartlett *et al.* (31) is close to that estimated after the accidental release of iodine 131 from Windscale Works in 1957. In two weeks after that accident the levels of iodine 131 in herbage appeared to decrease by about twice the amount attributable to the decay

of radioactivity (6). Even after several weeks the major part of the deposit may remain superficial so that it can be removed by washing with water. Leaching in rain has been shown to be the major cause of loss under normal circumstances (32), but not the only one. Moorby & Squire (33) have shown that measurable, though small, quantities of strontium 89 which have been deposited as fine sprays on leaves can become air-borne if the plants are kept in dry conditions. The radioactivity is contained in fine particles (ca. 4μ) and the maximum loss may not take place until 2 to 3 weeks after contamination. It has been suggested that losses under these conditions are due to the dehiscence of fragments of cuticle.

The assessment of the relative importance of foliar and plant-base entry in permanent pastures first received attention in the United Kingdom. The suggestion that plant-base entry (or stem-base entry as it was first called) might be important was made in 1957 to account for the fact that in hill areas, strontium 90 was transferred considerably more readily to the bones of animals, and to milk, from slowly growing hill pastures than in the lowlands (34). The difference could not be explained in terms of enhanced absorption from the soil due to its low calcium status; the entrapment of strontium 90 at the plant-base appeared to be the probable explanation because of the considerable development of "mat," consisting of prostrate stems and surface roots, which is characteristic of the hill pastures. The hypothesis was readily compatible with observations that the major part of the cumulative deposit was in the mat zone and not in the underlying soil. It appeared that strontium 90 might remain readily available for plant-base absorption for a considerable period since, in the early years of world-wide fallout, the levels of strontium 90 in biological materials from some hill areas rose in a similar manner to the cumulative deposit (34). Experimental studies have since provided considerably stronger evidence for the importance of plant-base absorption under these circumstances. Middleton & Squire (35) applied strontium 90 as a spray to turves from various types of pasture which had been transplanted to boxes. Within a relatively short period, the levels of strontium 90 on herbage from lowlands swards fell to those expected as a result of absorption from the soil. In contrast, turves from heavily matted upland pastures showed for many years a markedly greater transfer to herbage and this could not be inhibited entirely by the application of lime. Ellis & Newbould (36) compared relationships between the levels of strontium 90 in soil and the contamination of pastures in a number of areas with the extent to which strontium 89 was absorbed from the underlying soils by grass grown under laboratory conditions. Their results indicate that the extent of direct contamination is markedly greater, presumably due to plant-base uptake, in hill areas. Other studies point to the same conclusions (37).

The interpretation of the mechanism whereby strontium 90 enters by direct contamination into rapidly growing lowland pastures has proved more difficult. There is strong evidence that, when the rate of fallout was relatively high, direct contamination was the major route of entry and the

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1 weeks the major part of the deposit be removed by washing with water. The major cause of loss under normal conditions is one. Moorby & Squire (33) have shown that quantities of strontium 89 which have been deposited on plants can become air-borne if the plants are cut. The activity is contained in fine particles which do not take place until 2 to 3 weeks after deposition. That losses under these conditions are considerable.

The importance of foliar and plant-base entry has attracted attention in the United Kingdom. The mechanism of stem-base entry as it was first called (37) is thought to account for the fact that in hill pastures strontium is absorbed considerably more readily to the bones of sheep growing on hill pastures than in the lowland pastures. It can be explained in terms of enhanced calcium status; the entrapment of strontium is considered to be the probable explanation. The development of "mat," consisting of prostrate vegetation characteristic of the hill pastures. The observations with strontium 89 show that the major part of the activity remains readily available for plant-base uptake since, in the early years of world-wide fallout, the cumulative deposit (34). Experimental evidence for the uptake of strontium 90 under these circumstances. Middleton (30) showed that strontium 90 as a spray to turves from various sources was readily absorbed and transplanted to boxes. Within a relatively short time strontium 90 on herbage from lowlands swards was readily absorbed from the soil. In contrast, lowland pastures showed for many years a low uptake of strontium 90 and this could not be inhibited entirely. Newbould (36) compared relationships between strontium in soil and the contamination of pastures to the extent to which strontium 89 was taken up by grass grown under laboratory conditions. The extent of direct contamination is thought to be due to plant-base uptake, in hill areas. Other mechanisms (37).

The mechanism whereby strontium 90 enters by direct uptake from growing lowland pastures has proved evidence that, when the rate of fallout was high, direct uptake was the major route of entry and the

mean delay period in the transfer of strontium 90 from rain to milk was sometimes only 1 to 2 months (see p. 286) and that permanent pastures were subject to considerably greater direct contamination than annual crops. Despite the large discrimination factor against strontium relative to calcium in its passage to milk the ratio of strontium 90 to calcium in milk has generally differed little from that in the vegetable component of diet (2). The same conclusion was indicated by the sampling of grass in a number of areas. Such observations do not, however, answer the question of whether foliar or plant-base absorption is the operative mechanism. The difficulty of distinguishing between them lies in the fact that in lowland, as opposed to upland, pastures strontium 90 does not remain readily accessible for absorption in the plant-base zone for long periods; thus the expected time course of the two mechanisms is relatively similar. Some evidence suggestive of plant-base uptake in lowland pastures was obtained in 1958 and 1959; for example the degree to which some lowland pastures were directly contaminated from world-wide fallout appears to increase with age and the consequent development of a surface mat (37). However, until relatively recently it remained possible that foliar uptake might be the major mechanism of direct contamination. The statistical studies of Bartlett & Mercer (38) now show that this is most unlikely. They examined the correlation between the ratios of strontium 90 to calcium observed in milk produced throughout England and Wales during the summers of 1958 to 1960, and the extent of fallout in the month preceding the mean date of sampling (f_1), one month earlier (f_2) and two months earlier (f_3). The correlation coefficients between the observed levels in milk and the deposits in months f_1 , f_2 , f_3 were respectively 0.83, 0.94 and 0.86. It was found also that the best linear relationship between the level in milk (m) and these quantities was given by the equation:

$$m = 4.7 + 6.61 f_2$$

No significant advantage was gained by including other terms. A "lag" of at least a month between the deposition of strontium 90 and its maximum effect on the contamination of milk was thus established. This delay could not be attributed to the metabolic process of the cow since the delay between the ingestion of strontium 90 and the occurrence of the highest levels in milk is about one week (39). The significance of the lag period is apparent when the rate at which direct contamination is lost from leaves is remembered; after 7 to 8 weeks some 90 per cent of the original deposit is likely to have been removed (31). Thus, if the pastures had been contaminated mainly by simple foliar retention the lag demonstrated by Bartlett & Mercer (38) could not have occurred; it is, however, readily compatible with plant-base absorption. The analysis of results for some local milk sources in the United Kingdom again demonstrated this lag phenomenon (38, 40) and it is, therefore, probable that plant-base contamination is the dominant mechanism whereby recently deposited fallout enters pastures of all types in that country. Comparable data are not available for other areas and quantita-

tively different relationships must be expected under contrasting conditions of climate and husbandry. However, as broad similarities between the pattern of contamination of pastures in North America and Europe are suggested by many results of surveys (2, 27), the situation in the United Kingdom is unlikely to be unique.

Cereals—Enquiry into the mechanism whereby strontium 90 enters cereal grains from world-wide fallout was first encouraged by the observation that the ratio of strontium 90 to calcium in flour might be two or more times higher than that in leaf vegetables or the majority of other foods, while that in whole grain was larger still, often about three times that in flour (2). Clear evidence that strontium 90 entered grain largely by direct floral contamination was provided by a comparison of the ratios of strontium 90 from world-wide fallout to stable strontium in the different tissues of cereal ears (41). As the soil is the sole source of stable strontium, the ratio of strontium 90 to stable strontium in all tissues formed at the same time would be constant if strontium 90 entered only from the soil. Variations in this ratio thus indicate direct contamination with strontium 90 unaccompanied by stable strontium. The analysis of the different tissues of grain harvested in the United Kingdom and in other countries in 1957 showed that the ratio of strontium 90 to stable strontium in flour was always considerably lower than that in other tissues, sometimes by a factor exceeding 10 (41). A minimum estimate of the extent to which strontium 90 had entered by direct contamination could be made by assuming that all strontium 90 in the flour came from the soil. On this basis it appeared that at least two-thirds of the strontium in grain was due to direct contamination; when results for later years became available the corresponding estimate was about 50 per cent (5). Comparable calculations for different parts of the United States gave values of 20 and 90 per cent in 1959 (42, 43).

The importance of floral contamination has also been demonstrated in laboratory experiments. If strontium 89 is applied as a spray after ear emergence the contamination of the grain at harvest may be some 15 times greater than if the same application is made shortly before the ears emerge and the protection of emerged ears with small caps reduces contamination to the level found when the spray is applied before ear emergence (32). Floral contamination has been shown to be important also with rice (44).

Other crops.—Laboratory experiments have been carried out in which strontium 89 has been applied in a fine spray to cabbages, potatoes, and sugar beet at different stages of their growth (32, 45). Since the edible heart tissues of cabbages are protected by outer leaves and strontium 90 does not move basipetally within plants, the manner in which they are "directly" contaminated is not altogether obvious. The occurrence of direct contamination of the heart tissues, though to a much smaller extent than in pasture grass, is however well established both by experimental studies and surveys of world-wide fallout (41). It has been suggested that this may be due to strontium 90 being carried down in rain and lodging in the saucer-

pected under contrasting conditions as broad similarities between the in North America and Europe are (2, 27), the situation in the United States whereby strontium 90 enters cereal is encouraged by the observation that in flour might be two or more times that in the majority of other foods, while often about three times that in flour entered grain largely by direct floral comparison of the ratios of strontium 90 in the different tissues of cereal source of stable strontium, the ratio of tissues formed at the same time would only from the soil. Variations in this ratio with strontium 90 unaccompanied the different tissues of grain harvested in countries in 1957 showed that the ratio in flour was always considerably lower than in other tissues by a factor exceeding 10 (41). A comparison of the ratios of strontium 90 which strontium 90 had entered by assuming that all strontium 90 in the grain is due to direct contamination; when results are compared with the estimate was about 50 per cent in 1959 (42, 43). Contamination has also been demonstrated in which strontium 89 is applied as a spray after ear emergence. The ratio of strontium 89 in grain at harvest may be some 15 times that in the soil. This is made shortly before the ears emerge and the use of small caps reduces contamination. The use of caps is applied before ear emergence and is shown to be important also with rice (44). Experiments have been carried out in which strontium 90 is applied as a fine spray to cabbages, potatoes, and other vegetables. Since the edible parts are protected by outer leaves and strontium 90 enters through the leaves, the manner in which they are contaminated is together obvious. The occurrence of direct contamination, though to a much smaller extent than in the case of cereals, is established both by experimental studies and by observations. It has been suggested that this may be due to rain and lodging in the saucer-

shaped cavities at leaf axils whence it is absorbed into the stem. Only a very small fraction of the strontium 90 which is deposited on the foliage of potato plants reaches their tubers; in experiments when sprayed plants were exposed to rain 0.05 per cent or less was found in them (45). However, if sprayed plants were protected from rain negligible levels were observed in the tubers (11); thus under normal conditions downward leaching by rain over the surfaces of stems appears to be the main mechanism of transfer.

The absorption of strontium 90 from soil.—Laboratory studies of the absorption of strontium 90 from soil have usually been directed to one or more of the following objectives: the identification of soil characteristics which influence the extent to which it is absorbed by plants; the development of methods of estimating absorption from soil; the study of the extent to which its absorption will change with time. Field experiments have been carried out both to test the conclusions reached in the laboratory and also to determine quantitative relationships.

Variations in the extent to which strontium 90 enters plants from different soils depend mainly on their calcium content (2, 16, 21, 22, 46 to 50); absorption is greatest from soils low in that element. However, when the "exchangeable" calcium content of soils, as measured by conventional extraction procedures, exceeds a certain limit, often 15 to 20 meq per 100 g soil further increase in soil calcium has little effect on the absorption of strontium 90, and the ratio in which strontium 90 and calcium are extracted from soil by such procedures is frequently higher than that found in plants (21, 51). These observations are in apparent conflict with the finding that the OR (plant shoot/outer medium) is relatively constant and close to one. The reason for this was identified by Schofield (52). The ratio in which the two ions are absorbed depends on their ratio in the soil solution and this may be very different from the ratio in the total "exchangeable" fraction which is extracted by conventional procedures, for example with 1 *N* ammonium acetate. In a number of soils, the ratio of the two ions in solution was 0.5 to 0.8 of that in the readily exchangeable ions on surfaces. No simple procedure is available to measure the absolute concentrations of ions in the soil solution under equilibrium conditions. However, it has been shown that the relative magnitude of the ratio of strontium 90 to calcium in the solution phases of different soils can be inferred from the extent to which strontium 90 is removed by equilibration with 0.01 *M* calcium chloride (53, 54, 55). This procedure depends on the fact that only small changes in the concentration of calcium usually occur when this solution is shaken with soil; even when the readily exchangeable calcium in soil ranges from less than 1 to over 30 meq per 100 g, the concentration of the solution may change by not more than 15 per cent and usually considerably less. Thus the quantities of strontium 90 removed from different soils by 0.01 *M* calcium chloride indicate the relative magnitudes of the ratios of the two ions in the soil solutions, and hence the ratios in which they enter

plants. Closely linear relationships between the percentages of the strontium 90 removed from such soils and the ratio of strontium 90 to calcium in plants demonstrate the practical utility of this procedure; in contrast, markedly curvilinear relationships can occur between the ratios of strontium 90 to calcium in plants and those in soil extracts made with 1 *N* ammonium acetate (54, 55). Fredriksson *et al.* (46) found that extraction with ammonium lactate provides an adequate basis for comparison in an extensive survey of Swedish soils; the view that this extractant removed appreciable quantities of both strontium 90 and calcium from surfaces as well from the solution phase is, however, possible as the ratio of strontium 90 to calcium in plant shoots was usually about 80 per cent of that in the soil extract (21).

The effect of the addition of lime on the absorption of strontium 90 from soil has been considered in many investigations (2). Appreciable reductions, though seldom by a factor exceeding 3, occur in soils low in calcium. However, if soils contain moderate or high levels of calcium the addition of lime does not increase the concentrations of calcium in the soil solution and the absorption of strontium 90 is therefore not depressed. Fredriksson *et al.* (56, 57) have shown that liming was of little importance when 50 per cent or more of the exchange capacity of the soil is saturated with calcium.

Characteristics of soils other than their calcium content usually have comparable effects on the absorption of both strontium and calcium; thus the ratio of the two ions in plants is little changed. There are, however, indications that some preferential absorption of strontium can occur from soils low in colloidal minerals or when much organic matter has been added (2). This may reflect the contrasting distribution of the two ions between the soil and solution phases. Under field conditions where strontium 90 is usually largely present in the upper soil layers, manurial treatments which alter distribution may markedly affect uptake.

The extent to which the availability of strontium 90 to plants may be reduced by fixation processes in the soil has also received much attention. Since fixation is irreversible, or nearly so, its extent will increase with time until the sites which retain ions are saturated. Experiments in which the state of strontium 90 has been studied for several years thus provide the most convincing evidence. Squire (58) compared the extent to which 0.01 *M* calcium chloride displaced strontium 89 and strontium 90 from a series of soils in which the former had been present for one week and the latter for about 3.5 years. The extent to which strontium could be displaced by calcium chloride decreased in different soils by 3 to 10 per cent during this interval. These figures would overestimate the extent of fixation if the recently added tracer had not attained equilibrium throughout the labile phases. Evidence of a small degree of fixation was, however, provided by the fact that about 1.5 per cent of the strontium 90 which had been present for 3.5 years could not be extracted by prolonged treatment with concentrated nitric acid. Schulz & Riedel (59) used similar extraction procedures

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on parallel batches of soil in which strontium 90 had been present either for about 3.5 years or for a short period; the quantities of strontium 90 displaced from different soils decreased with time, by 1.2 to 3.4 per cent. Morgan (60) concluded that strontium 90 which had been accumulated in soil from world-wide fallout over several years did not differ appreciably in availability from that of recently added strontium 89.

These results provide convincing evidence that the conversion of strontium 90 into sparingly soluble forms is unlikely to reduce its availability to plants to a significant extent over a period of several years. Some small "fixation" may occur but its magnitude is unlikely to be sufficiently great to modify appreciably the rate at which strontium 90 passes through food chains. Suggestions that strontium 90 may, to a large extent, be held in forms which are inaccessible to plants appear to rest on inadequate data. Thus, for example, it has been suggested that the strontium 90 displaced from soil by relatively mild extraction procedures was alone available to plants because the amount of strontium 90 absorbed by plants from different soils was more closely correlated to this fraction than to the total strontium 90 in the soil (61). The findings of Schulz *et al.* (62) show the danger of characterizing the strontium 90 in soils into available and unavailable fractions on the basis of such treatments. These workers investigated 26 California soils to which radioactive strontium had been added. More than 30 per cent frequently remained in the soils after a single extraction with 1 N ammonium acetate but, with repeated extraction, progressively more was removed and the results were in close agreement with a theoretical extraction curve based on Vanslow's exchange equation; thus it appeared that essentially all the radioactive strontium was retained in exchangeable or water soluble forms. The assumption that the availability of strontium 90 has been reduced solely because less may be absorbed as the growing season advances (63) rests on a still more tenuous basis.

In practice the most important factor which is likely to cause change with time in the entry of strontium 90 into plants is its penetration down the soil profile. This is a slow process. In undisturbed soil 50 or more per cent of the added strontium may be in the upper 5 cm several years after its deposition on the soil surface. Cultivation treatment which causes it to be distributed through 15 to 25 cm of soil may lower the absorption of shallow rooted crops such as ryegrass by a factor approaching 3 (50).

In the United Kingdom and in Sweden, field experiments have been carried out to provide quantitative information on the contamination of crops (49, 50, 64). In the United Kingdom strontium 89 was sprayed onto the surface of the soil which was then cultivated to varying depths. In this way the distribution in the soil expected shortly after deposition could be simulated as well as that which would occur after cultivation for many years. The results of experiments carried out at sites in different parts of England and Wales, which span the range of calcium content commonly found in agricultural land, led to the conclusion that the presence of 1 mc

Sr^{90} per km^2 would initially lead on average to about 1.6 pc $\text{Sr}^{90}/\text{gCa}$ in plants; with the passage of time the deeper penetration of strontium 90 in soil would cause this value for shallow rooted crops to decrease by about one-third (65). The analysis of the results of surveys of world-wide fallout have lead to broadly similar conclusions; so also did the field experiments in Sweden (2).

Relative importance of direct contamination and absorption from the soil in the contamination of plants with world-wide fallout.—The first convincing evidence, on a wide scale, of the importance of direct contamination was provided by the comparison of the ratios of strontium 89 to 90 in rain with that in milk during the years 1957 and 1958. Because the half life of strontium 89 is only 7 weeks, its ratio to strontium 90 in any sample of fission products decreases by a factor of 2 in this period. Information on the rapidity with which strontium 90 is transferred to diet could therefore be obtained. In North America and in Britain it was found that the ratio in milk was half or more of that in fallout during the summer (27). The average delay in the transfer of strontium 90 to milk was thus between one and two months. Later, when the suspension of weapons trials caused the rate of fallout to decrease throughout the Northern Hemisphere, though the cumulative total still rose, marked decreases in the levels of strontium 90 in foodstuffs demonstrated the importance of direct contamination in the earlier period (2). It is estimated that during 1958 and 1959 the contamination of milk in the United Kingdom was mainly due to the direct contamination of pastures with the recent deposit (66) and the situation appears to have been similar in the Netherlands (67). In the United States about half the strontium 90 which entered milk in the summer of these years has been attributed to direct contamination (68).

Attempts have been made to derive proportionality factors which relate the levels of strontium 90 in agricultural products to the rate of fallout and to the cumulative deposit in the soil (2, 65 to 71). Any such calculation is subject to obvious limitations; changes in weather and seasonal factors influence the entry of strontium 90 into plants. With milk, which has been particularly considered from this viewpoint, additional complications are introduced by the seasonal grazing pattern of cattle and the consumption of stored food in winter. Further uncertainties result from the imprecision of the survey data on which calculations are based. The effects of these limitations are likely to be considerably reduced when estimates are made for the average situation over an entire year or longer in a large area where extensive surveys have been carried out. Calculations which satisfy these criteria have been shown to be of considerable practical value for predicting the probable long-term consequences of environmental contamination (2).

CAESIUM 137

Caesium 137 in plants.—Caesium is readily absorbed by plants and like potassium it is freely redistributed within them. However, it is well established that the absorption of caesium is not related to that of potassium in

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the close manner shown by calcium and strontium (2, 64). The study of the interaction of caesium and potassium has been hindered by the fact that toxic symptoms may readily be induced by relatively high ratios of caesium to potassium. Cline & Hungate (72) found that the growth of plants was considerably reduced by 0.2 mM caesium in the presence of 0.07 mM potassium after 16 days. Uhler (73) showed that within a few hours respiration may be reduced by 7.5 mM caesium. The suggestion that two mechanisms are operative for the absorption of caesium, one at relatively low concentrations, the other at higher levels (74), may therefore be due to the occurrence of toxic effects under the latter circumstance. Cline (75) has, however, established that over extended periods the absorption of carrier-free caesium 137 (which will give rise to no toxic symptoms) bears no constant relationship to that of potassium; whereas the ratio of caesium 137 to potassium in tissues equalled that in the external solution when 10 mM potassium was applied, caesium 137 reached leaves to less than one-fiftieth of the extent of potassium when the concentration of the latter ion was 0.02 mM. Comparable results have also been obtained in short experiments, in which, however, interpretation is less certain because of the occurrence of exchange reactions (76). The dissimilar behaviour of the two ions does not, however, justify the conclusion that they enter plants by separate mechanisms. The ionic radii of the two ions contrast markedly; this could cause them to be retained to differing extents on sites of the same type. Uncertainties as to the basic nature of metabolic processes whereby ions are actively accumulated make it unwise to suggest any final conclusion.

Caesium 137 which was deposited on foliage of plants appears to be retained relatively similarly to strontium 90, and like strontium it is readily removed from foliage by rain (32). The concentration of caesium 137 within different tissues which results from direct contamination, however, can contrast very markedly with that caused by strontium 90. This is due to the mobility of caesium 137 within tissues; thus nearly 30 per cent of the caesium 137 which has been deposited on the foliage of potatoes may reach the tubers, as compared with less than 1 per cent of strontium 89 (45). It appears possible that in permanent pastures caesium 137 may remain in the plant-base region, readily accessible for absorption for an appreciably longer time than strontium 90 (77). More detailed investigations must, however, be carried out before any general conclusions are warranted.

Caesium 137 in the soil.—Caesium 137 moves downwards in undisturbed soil to a considerably smaller extent than strontium 90 (2); after 3 years more than 90 per cent of the caesium 137 which has been added to the soil surface may remain in the upper 5 cm (78). The most important contrast in the behaviour of the two ions in the soil, is however, that caesium 137 is usually bound in forms inaccessible to plants (2). Fredriksson *et al.* (79) have estimated that 95 to 99 per cent of the caesium 137 in typical Swedish soils is fixed in this way; the operative process appears to be entrapment of the caesium ion in the lattice structures of clay minerals, possibly illite.

The extent of fixation can be reduced by the addition of stable caesium (56, 80) and sometimes by potassium (79). The latter observation suggests that the process of fixation may be similar to that for potassium, though some results suggest the contrary view (81). There is evidence that the fixation process is not instantaneous and that the availability of caesium 137 may decrease progressively over two or more years (82). Thus, whereas the results of short-term experiments suggest that caesium 137 usually enters plants from typical temperate soil to about one-tenth the extent of strontium 90, the corresponding value after three or more years may be about one-twentyfifth (2). Until relatively recently it was assumed that caesium 137 was fixed to a considerable degree in all soils. This, however, has now been disproved. Fredriksson conducted an extensive series of experiments on soils from tropical areas in South America; many of these soils contained very small quantities of clay minerals and no evidence of fixation was obtained (64).

Entry of caesium 137 into plants from world-wide fallout.—The relative extent to which caesium 137 enters plants as a result of the direct contamination of vegetation as opposed to absorption from the soil, has been studied in considerably less detail than that of strontium 90. Because the latter ion is considerably more freely absorbed from the soil, and the extent of direct contamination of both is relatively similar, it would be expected that the content of caesium 137 in plants would be much more closely related to the rate of fallout in the immediate past. While this is, in general, borne out by surveys of dietary contamination the levels of caesium 137 in milk from some regions did not decrease in the manner expected, relative to those of strontium 90, when the rate of fallout declined in 1960 (2). A possible explanation is suggested by the two "delaying" mechanisms to which reference has already been made, namely the retention of caesium 137 in the plant-base zone for a longer period than strontium 90 and the slow rate at which fixation may proceed in the soil. Considerably more detailed investigations must be undertaken before relationships between the levels of caesium 137 in plants and both the rate and the cumulative total of fallout can be elucidated, even to the extent which has been achieved for strontium 90.

THE URANIUM AND THORIUM SERIES

The most important naturally occurring alpha-emitting nuclides belong to the uranium and thorium series; the actinium series is much less abundant. Because of the complex decay schemes of these series, measurements have frequently been reported as total alpha activity. Uncertainties as to the extent of isotopic equilibrium can cause difficulties in the interpretation of such observations, but it is apparent that radium is the element principally absorbed by plants (2, 83, 84). Thorium 232, the parent member of the thorium series, has not been detected in living tissues; thorium 228 has been observed but its presence can be attributed to the decay of the parent

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by the addition of stable caesium (79). The latter observation suggests similar to that for potassium, though (81). There is evidence that the and that the availability of caesium o or more years (82). Thus, whereas s suggest that caesium 137 usually e soil to about one-tenth the extent of e after three or more years may be tively recently it was assumed that e degree in all soils. This, however, n conducted an extensive series of eas in South America; many of these of clay minerals and no evidence of

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radium 228. Little experimental work on the absorption of thorium by plants has been carried out. However, in water culture experiments lasting 24 hr Mercer & Morrison (85) found no detectable quantities of thorium in shoots, but more prolonged studies must be carried out before it can be concluded that this element cannot move upwards in plants to any appreciable extent.

Radium.—Two isotopes of radium have sufficiently long half lives to deserve consideration from the viewpoint of absorption by plants, namely radium 228 (thorium series; half life 6.7 years) and radium 226 (uranium series; half life 1620 years). Numerous measurements of radium have been made in plant tissue and very widely ranging values have been found (2). Turner *et al.* (86) found that the content of different edible plant tissues ranged from less than 1 to 17,000 pc alpha activity/kg. The highest values were for Brazil nuts, cereals were intermediate (up to 580 pc/kg) while other fruits and leaf vegetables contained low quantities. The existence of an unusual accumulation mechanism in the Brazil nuts was suggested both by this observation and by the fact that the same species also accumulate barium to an unusual extent (87). However, as soil samples were not available from the areas where the nuts were collected for alpha assay, the possibility could not be excluded that the results were due to abnormal levels of activity in the soil. More recently a preliminary report has been made on different tissues of Brazil nuts and the underlying soil in British Guiana (88). The soil contained 22,000 to 24,000 pc alpha activity/kg which is within the normal range for many areas (83). The endosperm and pericarp of the nuts, however, sometimes contained more than 30,000 pc/kg, twice the previously reported figure; the values for the epicarp were lower than those for other tissues. Although the full spectrum of the alpha activities in these tissues has not yet been reported, radium 226 was the predominant nuclide; its accumulation to an unusual degree in the fruits of this plant is therefore established. The ratios of alpha activity to calcium were also examined in the Brazil nut samples from British Guiana. The values for the endosperm and epicarp appreciably exceeded those for leaves or such stem tissues as were examined. These findings do not, however, prove that radium is transferred preferentially to calcium in any step of the upward transfer mechanism. The higher ratio in the endosperm than other aerial tissues could be due to the preferential retention of radium in some stem tissues during the process of upward transfer to leaves and to its subsequent release at the time of fruit development. This suggestion is encouraged by the observation that strontium may be retained to a considerably greater extent than calcium in plant stems and that under some experimental conditions a higher ratio of strontium to calcium in grain than in other tissues can be induced, though the reverse usually occurs (13). The study of these relationships in the Brazil nut is rendered difficult both by the laborious procedures necessary to identify specific alpha-emitting substances in the low levels in which they occur in nature and by the

obvious problems of growing this tree under experimental conditions. No results are as yet available of physiological studies of the factors which control the absorption of radium in other species. The comparative study of radium and the other alkaline earths, magnesium, calcium, strontium, and barium, would appear to hold promise of elucidating some aspects of the mechanisms whereby cations are absorbed and transferred in plants.

Lead 210 and polonium 210.—Lead 210 (half life 19 years) and polonium 210 (half life 138 days) are the only two radioactive substances of appreciable half life which arise from radon 222, the gaseous emanation of radium. The polonium isotope, unlike lead 210, emits alpha radiation, and has been shown to be the major component of the alpha activity in grass grown under normal field conditions (89). It has been suggested that this is mainly due to the deposition in rain of the decay products of radon which escape from the earth's crust into the atmosphere. Other investigations provide evidence of the absorption of the lead isotope from the soil (2).

CARBON 14

Carbon 14, both natural and man-made, is formed in the atmosphere from nitrogen. Its natural occurrence is due to the action of cosmic rays; man-made carbon 14 results from the release of neutrons, formed mainly in nuclear weapons tests. The relative amounts in which carbon 14 and fission products arise from nuclear weapons depends on the ratio of total yield (fission plus fusion) to fission yield. Thus, carbon 14 will become proportionately more significant if larger or "cleaner" weapons are exploded; the adjective "clean" refers to the extent of fission only. Although carbon 14 enters into biological systems through photosynthesis, the evaluation of its effects presents few problems for the plant physiologist since the specific activity of organic carbon will reflect that of the atmospheric carbon dioxide from which it is derived.

The changes in the specific activity of atmospheric carbon dioxide because of nuclear weapons trials may, however, provide an important tool for certain types of biological research. The work of Libby (90) has caused the general principles of "carbon-dating" to become widely known. Until considerable releases of carbon dioxide from fossil sources occurred in the late 19th century the specific activity of atmospheric carbon dioxide is believed to have been constant for a considerable period; thus the date at which organic carbon was synthesised could be calculated from its specific activity on the basis of the half life of carbon 14 (5570 years). In the recent past, however, the specific activity of the atmospheric carbon dioxide has increased. In 1955 it was estimated that the mean value in the troposphere of the Northern Hemisphere was a few per cent above the natural level, but in 1959 it was enhanced to nearly 30 per cent (2). Further increases are to be expected. Accordingly, the specific activity of carbon in organic compounds which have been formed by photosynthesis in the last decade will vary considerably depending on their date of formation. Thus, whereas "carbon-

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under experimental conditions. No logical studies of the factors which affect other species. The comparative study of strontium, magnesium, calcium, strontium, and potassium promise of elucidating some aspects of the elements absorbed and transferred in plants.

Polonium 210 (half life 19 years) and polonium 214 (half life 163 days) are two radioactive substances of appreciable activity. Radium 222, the gaseous emanation of radium 226, emits alpha radiation, and has been shown to be the alpha activity in grass grown in the field. It has been suggested that this is mainly due to the decay products of radon which escape from the atmosphere. Other investigations provide evidence for the presence of a long lived isotope from the soil (2).

ION 14

Carbon-14, man-made, is formed in the atmosphere and its presence is due to the action of cosmic rays; the release of neutrons, formed mainly from nuclear weapons, depends on the ratio of total fission yield. Thus, carbon 14 will become more abundant in larger or "cleaner" weapons are exposed to the extent of fission only. Although carbon is assimilated through photosynthesis, the evaluation of the plant physiologist since the will reflect that of the atmospheric carbon

activity of atmospheric carbon dioxide. The work of Libby, however, provide an important tool for "carbon dating". The work of Libby (90) has caused "carbon dating" to become widely known. Until recently, the activity of atmospheric carbon dioxide from fossil sources occurred in a considerable period; thus the date at which a sample could be calculated from its specific activity of carbon 14 (5570 years). In the troposphere, the activity of the atmospheric carbon dioxide has increased that the mean value in the troposphere is now a few per cent above the natural level, but in some areas it is as high as 10 per cent (2). Further increases are to be expected as the activity of carbon in organic compounds increases. The synthesis of carbon in organic compounds in the last decade will vary considerably from that of formation. Thus, whereas "carbon-

dating" was previously applicable only to samples of considerable antiquity it is now possible to study the time-course of those carbon cycles which turn over rapidly in nature, for example the formation of organic matter in soil. The laborious techniques involved will, however, limit the exploitation of this opportunity.

OTHER RADIOACTIVE NUCLIDES

Fission products.—Apart from strontium 90, caesium 137 and iodine 131, mixed fission products contain about 180 other radioactive nuclides. However, if the mixture is deposited in fission yield, strontium 89 (half life 51 days) is the only product which is likely, at any time, to be a more important source of internal radiation than strontium 90, caesium 137 or iodine 131. Up till about 6 weeks after fission it may deliver a larger dose than strontium 90 but it soon becomes of little importance and it contributes little to the exposure of the population from world-wide fallout (2). In fresh fallout, barium 140 (half life 12 days) and short lived isotopes of iodine, for example, iodine 133 (half life 21 hours) may enhance the exposure of the bone or thyroid, though not being the major sources of exposure.

Fission products are not, however, always released in fission yield. Their ratio in the effluent from nuclear establishments may be much altered by chemical separation. This may cause nuclides which are relatively immobile in biological systems to assume particular interest. Thus, for example, the limits to the release of dilute effluent into the sea at Windscale Works are largely determined by the extent to which ruthenium 106 (half life 1 year) is absorbed onto the marine alga *Porphyra* which is consumed by limited population groups in Wales (91).

The absorption by plants of nuclides of relatively long half life, other than strontium 89 and 90 and caesium 137 has been examined, especially ruthenium 106, zirconium 95 (half life 63 days) and cerium 144 (half life 290 days) (92). All these substances enter plants from the soil considerably less readily than strontium 89 or 90 though they can be readily adsorbed onto the surfaces of roots from solutions. Between soil types the absorption of ruthenium varies widely but it is often of comparable order to that of caesium 137; except from acid soils, the absorption of cerium is extremely small. These relationships are to be expected on account both of the valency of these ions and their other chemical characteristics. Not only is absorption small, but the major fraction of that which enters plants is retained in roots or stem tissues, little reaching the leaves.

Induced activities.—Radioactive isotopes of many elements can be induced by neutron capture. This occurs both in nuclear weapons tests and in nuclear reactors. The latter is likely to be of particular significance when large amounts of cooling water are drawn from natural sources and returned to them. Thus at Hanford Works the induced activities of phosphorus 32 (half life 14 days) and zinc 65 (half life 245 days) rank next

to iodine 131 as the most significant releases of radioactivity into the environment (93). Both phosphorus 32 and zinc 65 have been detected in grass and milk in pastures irrigated by the Columbia River into which the Hanford cooling water is returned. Zinc 65, together with the induced activities, cobalt 59 and 60 and iron 55, has also been found to be the main source of radioactivity in fish and sea water soon after nuclear explosions (94). The low concentration of the carrier isotopes in water can cause these nuclides to be absorbed and concentrated to a spectacular extent in plants and animals.

Plutonium.—Because of its very long half life and high toxicity to animals consideration has been given to the entry into plants of the fissile element plutonium. A very slow rate of absorption is to be expected because it forms high valency (usually 4 or 6) ions; this has been confirmed in several studies and, over 1.5 years, grass grown in pot culture may absorb less than 10^{-4} per cent of that added to the soil (95, 96).

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