1 320

9

57

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

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

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.

[2] The 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).

:s where modern medical procedures
year in Norway to 58 in France (2).
:h sources as luminous watches, shoe-

: information on all aspects of man's ex-
annexes and bibliographies covering not
oactivity and the behaviour of radioactive
: existence of this document makes it un-
verage of the literature, except on topics
ration.
of radiation is usually measured in *rems*.
g the absorbed dose in *rads* (1 rad is 100
/e biological effectiveness) for the type of
d gamma radiation is 1, that for alpha and

nostic 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 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/YEAR[3]

	Natural background	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
External radiation to whole body:				
Cosmic rays	50	—	—	—
From radioactive substances	50	4.2	0.65	3%
Internal sources:				
To whole body (range for different 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

LL

I

FROM NATURAL BACKGROUND AND RATES IN MREM/YEAR[a]

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%
60 6–14	1.3	—

Report (2), p. 21.
).
d on UN Report (2), p. 27.

however, not the only source of en-
rom 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

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

European countries where milk is the main dietary source of calcium, 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

Ratio: Sr/Ca in precursor

ıs through any biological process which
om diet to bone, or from the soil to the
ber of successive steps; the term "Dis-
to define the relative behaviour of the
of the entire process being the product

ısed, difficulties have sometimes arisen
cause has been disregard of the precise
12) laid emphasis on the fact that the
if the ratio in which the two ions were
l system was known, as well as the ratio
ise estimates of the relative availability
conclusions as to the magnitude of the
on occasions been reached through the
d tracer strontium to "exchangeable"
the ratio in which the two ions are

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,

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,

iar contamination : retention by leaves, by inflorescences, (*c*) plant base con- by the basal parts of plants, or surface incorporated into the soil.

ıts by the first two routes will not be lcium may enter simultaneously with ıt to a much smaller extent than when soil. The relative importance of these on the growth form of plants. Whereas pecies, floral absorption is of practical plant-base absorption is particularly im- ı not necessarily confined to them. It is ndaries between foliar absorption and ring rain the downward washing of con- ılant will occur concurrently with absorp- ın merge into plant-base entry. Likewise,

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

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

oil was the major route [illegible]

[illegible] 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-

xpected under contrasting conditions
, as broad similarities between the
in North America and Europe are
(2, 27), the situation in the United

ism whereby strontium 90 enters cereal
st encouraged by the observation that
in flour might be two or more times
r the majority of other foods, while
often about three times that in flour
entered grain largely by direct floral
parison of the ratios of strontium 90
ntium in the different tissues of cereal
urce of stable strontium, the ratio of
tissues formed at the same time would
only from the soil. Variations in this
ion with strontium 90 unaccompanied
the different tissues of grain harvested
countries in 1957 showed that the ratio
in flour was always considerably lower
es by a factor exceeding 10 (41). A
which strontium 90 had entered by
by assuming that all strontium 90 in the
asis it appeared that at least two-thirds
to direct contamination; when results
e corresponding estimate was about 50
ions for different parts of the United
r cent in 1959 (42, 43).
nination has also been demonstrated in
um 89 is applied as a spray after ear
grain at harvest may be some 15 times
is made shortly before the ears emerge
s with small caps reduces contamination
is applied before ear emergence (32).
wn to be important also with rice (44).
riments have been carried out in which
a fine spray to cabbages, potatoes, and
their growth (32, 45). Since the edible
tected by outer leaves and strontium 90
plants, the manner in which they are
gether obvious. The occurrence of direct
, though to a much smaller extent than in
ablished both by experimental studies and
). It has been suggested that this may be
down in 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

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

the strontium 90 which

ed by prolonged treatment with concen-
(59) used similar extraction procedures

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

average delay in the transfer of strontium 90 to milk was [illegible] 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

of the importance of direct

ı of the ratios of strontium 89 to 90 in
ırs 1957 and 1958. Because the half life
ratio to strontium 90 in any sample of
or of 2 in this period. Information on
0 is transferred to diet could therefore
in Britain it was found that the ratio in
fallout during the summer (27). The
ontium 90 to milk was thus between one
uspension of weapons trials caused the
iout the Northern Hemisphere, though
:d decreases in the levels of strontium 90
ortance of direct contamination in the
ıat during 1958 and 1959 the contamina-
was mainly due to the direct contamina-
posit (66) and the situation appears to
ls (67). In the United States about half
k in the summer of these years has been
58).

rive proportionality factors which relate
ltural products to the rate of fallout and
il (2, 65 to 71). Any such calculation is
anges in weather and seasonal factors
) into plants. With milk, which has been
viewpoint, additional complications are
g pattern of cattle and the consumption
uncertainties result from the imprecision
ulations are based. The effects of these
erably reduced when estimates are made
ı entire year or longer in a large area
n carried out. Calculations which satisfy
be of considerable practical value for pre-
nsequences of environmental contamina-

ESIUM 137

ım is readily absorbed by plants and like
d within them. However, it is well estab-
ium is not related to that of potassium in

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 m*M* 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 m*M*. 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.

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

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

rom world-wide fallout.—The relative
plants as a result of the direct con-
to absorption from the soil, has been
han that of strontium 90. Because the
absorbed from the soil, and the extent
elatively similar, it would be expected
nts would be much more closely related
te past. While this is, in general, borne
ation the levels of caesium 137 in milk
se in the manner expected, relative to
ate of fallout declined in 1960 (2). A
by the two "delaying" mechanisms to
made, namely the retention of caesium
nger period than strontium 90 and the
proceed in the soil. Considerably more
dertaken before relationships between
s and both the rate and the cumulative
l, even to the extent which has been

AND THORIUM SERIES

occurring alpha-emitting nuclides belong
; the actinium series is much less abun-
y schemes of these series, measurements
total alpha activity. Uncertainties as to
can cause difficulties in the interpretation
parent that radium is the element prin-
84). Thorium 232, the parent member of
etected in living tissues; thorium 228 has
n be attributed to the decay of the parent

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

DOE ARCHIVES

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-

DOE ARCHIVES

few per cent above the natural level, but
) per cent (2). Further increases are to be
activity of carbon in organic compounds
synthesis in the last decade will vary con-
of formation. Thus, whereas "carbon-

duced 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).

LITERATURE CITED

1. Zuckerman, S., *Rept. Comm. Management Control Res. Develop.* (Her Majesty's Stationery Office, London, 1961)
2. United Nations Scientific Committee on the Effects of Atomic Radiation. *Rept. Gen. Assembly* (United Nations, New York, 1962)
3. Spiers, F. W., *Hazards Man Nucl. Allied Radiations, Cmnd. 1225, Annex D* (H.M. Stationery Office, London, 1960)
4. Loutit, J. F., Marley, W. G., Mayneord, W. V., and Russell, R. S., *Hazards Man Nucl. Allied Radiations, Cmnd. 1225, Annex F* (H.M. Stationery Office, London, 1960)
5. Agricultural Research Council Radiobiological Laboratory, *Ann. Rept. 1961-1962, ARCRL 8* (H.M. Stationery Office, London, 1962)
6. Loutit, J. F., Marley, W. G., and Russell, R. S., *Hazards Man Nucl. Allied Radiations, Cmnd. 1225, Annex H* (H.M. Stationery Office, London, 1960)
7. Farmer, F. R., and Fletcher, P. T., *Proc. Intern. Elec. Nucl. Conf. 6th,* **1,** pt. 1, 413–37 (1959)
8. Middleton, L. J., and Squire, H. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 5,* 50–51 (H.M. Stationery Office, HMSO, London, 1961)
9. Rediske, J. H., and Selders, A. A., *Plant Physiol.,* **28,** 594–605 (1953)
10. Russell, R. S., and Squire, H. M., *J. Exptl. Botany,* **9,** 262–76 (1958)
11. Moorby, J., and Squire, H. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 8,* 63–64 (H.M. Stationery Office, London, 1962)
12. Comar, C. L., Wasserman, R. H., and Nold, M. M., *Proc. Soc. Exptl. Biol. Med.,* **92,** 859–63 (1956)
13. Martin, R. P., Newbould, P., and Russell, R. S., *Radioisotopes Sci. Res., Proc. Intern. Conf., Paris, Sept., 1957,* **4,** 173–90, (1958)
14. Menzel, R. G., and Heald, W. R., *Soil Sci.,* **80,** 287–93 (1955)
15. Bowen, H. J. M., and Dymond, J. A., *J. Exptl. Botany,* **7,** 264–72 (1956)
16. Gulyakin, I. V., and Yudintseva, E. V., *Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva,* **18,** 476–85 (1958)
17. Asada, K., and Kasai, Z., *Mem. Res. Inst. Food Sci., Kyoto Univ.,* **20,** 22–30 (1960)
18. Comar, C. L., Russell, R. S., and Wasserman, R. H., *Science,* **126,** 485–92 (1957)
19. Mouat, M. C. H., *Nature,* **188,** 513–14 (1960)
20. Vose, P. B., and Koontz, H. V., *Hilgardia,* **29,** 575–85 (1960)
21. Fredriksson, L., and Eriksson, B., *Ann. Kgl. Landwirtschaftlichen Hochschule Schwedens* (In press 1962)

1957, **4**, 173–90, (1958)
14. Menzel, R. G., and Heald, W. R., *Soil Sci.*, **80**, 287–93 (1955)
15. Bowen, H. J. M., and Dymond, J. A., *J. Exptl. Botany*, **7**, 264–72 (1956)
16. Gulyakin, I. V., and Yudintseva, E. V., *Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva*, **18**, 476–85 (1958)
17. Asada, K., and Kasai, Z., *Mem. Res. Inst. Food Sci., Kyoto Univ.*, **20**, 22–30 (1960)
18. Comar, C. L., Russell, R. S., and Wasserman, R. H., *Science*, **126**, 485–92 (1957)
19. Mouat, M. C. H., *Nature*, **188**, 513–14 (1960)
20. Vose, P. B., and Koontz, H. V., *Hilgardia*, **29**, 575–85 (1960)
21. Fredriksson, L., and Eriksson, B., *Ann. Kgl. Landwirtschaftlichen Hochschule Schwedens* (In press 1962)
33. Moorby, J., and Squire, H. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 8*, 62–63 (H.M. Stationery Office, London, 1962)
34. Bryant, F. J., Morgan, A., and Spicer, G. S., *U.K. At. Energy Authority, Rept. AERE HP/R-2730* (1958)
35. Squire, H. M., and Middleton, L. J., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 2*, 40–41 (H.M. Stationery Office, London, 1960)
36. Ellis, F. B., and Newbould, P., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 5*, 58–62 (1961)
37. Ellis, F. B., and Mercer, E. R., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 4*, 48–63 (H.M. Stationery Office, London, 1961)
38. Bartlett, B. O., and Mercer, E. R., *Agr. Res. Council Radiobiological* (1954)
52. Russell, R. S., Schofield, R. K., and Newbould, P., *Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva*, **27**, 146–48 (1958)
53. Squire, H. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 4*, 35–37 (H.M. Stationery Office, London, 1961)
54. Milbourn, G. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 5*, 54–57 (H.M. Stationery Office, London, 1961)
55. Newbould P., and Squire, H. M. (In preparation)
56. Fredriksson, L., Eriksson, B., Rasmuson, B., Gahne, B., Edvarson, K., and Löw, K., *Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva*, **18**, 449–70 (1958)
57. Fredriksson, L., Eriksson, A., and

Haak, E., *Ann. Kgl. Landwirtschaftlichen Hochschule Schwedens* (In press 1962)
58. Squire, H. M., *Nature*, **188**, 518–19 (1960)
59. Schulz, R. K., and Riedel, H. H., *Soil Sci.*, **91**, 262–64 (1961)
60. Morgan, A., *J. Nucl. Energy, Pt. A*, **11**, 8–13 (1959)
61. Roberts, H., Jr., and Menzel, R. G., *J. Agr. Food Chem.*, **9**, 95–98 (1961)
62. Schulz, R. K., Overstreet, R., and Babcock, K. L., *Hilgardia*, **27**, 333–42 (1958)
63. Lee, C. C., *Science*, **138**, 41–42 (1962)
64. Fredriksson, L., *Ann. Kgl. Landwirtschaftlichen Hochschule Schwedens* (In press 1962)
65. Burton, J. D., Milbourn, G. M., and Russell, R. S., *Nature*, **185**, 498–500 (1960)
66. Bartlett, B. O., Burton, J. D., and Russell, R. S., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 5*, 62–63 (H.M. Stationery Office, London, 1961)
67. Mattern, F. C. M., and Strackee, L., *Nature*, **193**, 647–49 (1962)
68. Kulp, J. L., and Schulert, A. R., *Science*, **136**, 619–32 (1962)
69. Mercer, E. R., Bartlett, B. O., and Burton, J. D., (In preparation)
70. Knapp, H. A., *U.S. At. Energy Comm., Rept. TID-13945* (1961)
71. Rivera, J., *U.S. At. Energy Comm., Rept. TID-7632, Pt. 2*, 405–11 (1962)
72. Cline, J. F., and Hungate, F. P., *Plant Physiol.*, **35**, 826–29 (1960)
73. Uhler, R. L. (Personal communication, 1962)
74. Bange, G. G. J., and Overstreet, R., *Plant Physiol.*, **35**, 605–8 (1960)
75. Cline, J. F., *Nature*, **193**, 1302–3 (1962)
76. Middleton, L. J., Handley, R., and Overstreet, R., *Plant Physiol.*, **35**, 913–18 (1960)
77. Squire, H. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 8*, 64–66 (H.M. Stationery Office, London, 1962)
78. Squire, H. M., and Middleton, L. J., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 5*, 47–50 (H.M. Stationery Office, London, 1961)
79. Fredriksson, L., Eriksson, B., Eriksson, A., and Haak, E., *Ann. Kgl. Landwirtschaftlichen Hochschule Schwedens* (In press 1962)
80. Tensho, K., Yeh, K., and Mitsui, S., *Soil Plant Food (Tokyo)*, **6**, 176–83 (1961)
81. Schulz, R. K., Overstreet, R., and Barshad, I., *Soil Sci.*, **89**, 16–27 (1960)
82. Middleton, L. J., and Squire, H. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 4*, 63–64 (H.M. Stationery Office, London, 1961)
83. Mayneord, W. V., Turner, R. C., and Radley, J. M., *Nature*, **187**, 208–11 (1960)
84. Mayneord, W. V., and Hill, C. R., *Nature*, **184**, 667–69 (1959)
85. Mercer, E. R., and Morrison, T. M., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 8*, 83 (H.M. Stationery Office, London, 1962)
86. Turner, R. C., Radley, J. M., and Mayneord, W. V., *Health Phys.*, **1**, 268–75 (1958)
87. Robinson, W. O., Whetstone, R. R., and Edginton, G., *U.S. Dept. Agr., Tech. Bull., 1013* (1950)
88. Mercer, E. R., Mitchell, W. A., and Smith, K. A., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 8*, 82–83 (H.M. Stationery Office, London, 1962)
89. Hill, C. R., *Nature*, **187**, 211–12 (1960)
90. Libby, W. F., *Radiocarbon Dating*, 2nd ed. (University of Chicago Press, Chicago, Ill., 1955)
91. Dunster, H. J., *Proc. Intern. Conf. Peaceful Uses At. Energy, 2nd, Geneva*, **18**, 390–99 (1958)
92. Nishita, H., Romney, E. M., and Larson, K. H., *J. Agr. Food Chem.*, **9**, 101–6 (1961)
93. Bustad, L., *Radioisotopes in the Biosphere*, 243–54 (University of Minnesota Press, Minneapolis, Minn., 1960)
94. Welander, A. D., *U.S. At. Energy Comm., Rept. UWFL-55* (1958)
95. Jacobson, L., and Overstreet, R., *Soil Sci.*, **65**, 129–34 (1948)
96. Newbould, P., and Mercer, E. R., *Agr. Res. Council Radiobiological Lab., Rept. ARCRL 8*, 81–82 (H.M. Stationery Office, London, 1962)