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FALLOUT AND RADIOSTRONTIUM.

by

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A Thesis Submitted to the University of Glasgow

for the

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"Then we saw flashes of fire bright as the sun itself, rising into the sky. They rose about ten degrees from the horizon and the sky around them glowed fiery red and yellow. Someone yelled to the men below. 'The sun is rising in a strange fashion. Hurry up and see it.' Then I realised that what we were watching could not be the sun, for the light was coming from the west. It was at this moment that I first felt fear and first thought of the atomic bomb."

Sanjiro Masuda,

Crewman of the 'Fukuryu Maru'.

1st March 1954.

C O N T E N T S.

LIST OF ILLUSTRATIONS.

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INTRODUCTION.

This thesis is the description of an investigation carried out to study the fallout of Strontium-90, produced from nuclear weapons testing. Since strontium is chemically similar to calcium, it will become fixed in bone when taken in the diet, and the long half-life of Sr-90 means that it is a long-term hazard. Isolated samples of grass taken from the Perthshire hills in 1957 had shown a high level of Sr-90 activity compared with pasture grass from cultivated fields in the south of England. This difference was due partly to the much higher rainfall in Perthshire, and to the agricultural conditions. There was therefore a need to study the levels of Sr-90 contamination in high rainfall areas, since the Sr-90 taken up by grass finds its way into the human body, with cows' milk as the intermediate.

Although the first atomic bomb was exploded in July 1945, it was not until the first thermonuclear explosion in November 1952 that fallout began to assume its characteristic world-wide deposition. This is because the much greater release of energy from the 'hydrogen' bomb lifts the fission products into the long-range air currents of the upper atmosphere. The bombs tested before November 1952 were fission bombs of sufficiently low yield to prevent much distant fallout, and the radioactive debris was deposited near the test site, and over suitably uninhabited areas.

It was recognised from the beginning of weapons testing that fallout could be a serious hazard. The testing of the first Soviet atomic bomb, and the realisation that any nation possessing a monopoly of thermonuclear weapons would have complete military supremacy, started the present arms

race towards thermonuclear stockpiling. It is against this political background that the picture of Sr-90 fallout must be viewed. With the universal public concern over the dangers from fallout it is now expedient for the three nations at present in possession of thermonuclear weapons to self-impose a ban on tests.

Due to this concern, in 1956 the Food Science Department of the Royal College of Science and Technology, Glasgow, was invited to carry out a survey of Sr-90 in foodstuffs in the Glasgow area. The initial results of this survey led to the instigation of the more detailed investigation which comprises this thesis.

Part I.

PREAMBLE.

PRODUCTION AND DISTRIBUTION OF SR-90.

When a large enough mass of certain isotopes of heavy metals is assembled near to a source of neutrons a self-sustaining chain reaction occurs, in which the heavy nuclei undergo fission on neutron capture producing further neutrons to continue the process. The minimum mass which has to be assembled in any particular arrangement before this phenomenon can occur is called the "critical mass". Three nuclides exhibit this property - U-235, U-233, and Pu-239; the first of which occurs naturally in ordinary uranium (to the extent of 0.7%), the other two are manufactured in an atomic reactor. This happens when U-238 or Th-232 capture a low-energy neutron and, on the subsequent emission of two β particles, are transformed into Pu-239 and U-233 respectively. Any of these three fissile materials can form the basis of an atomic bomb or a nuclear reactor. The average energy of the neutrons released by the fission of U-235 is 1 Mev (1) which is not energetic enough to cause the fission of U-238, but the capture of a neutron of any energy by the three fissile isotopes will cause fission.

Since slow or "thermal" neutrons are more easily captured by the nucleus than fast neutrons, most nuclear reactors use a "moderator" to slow down the fast neutrons produced by the fission process in order to enable the chain reaction to continue. The moderator is usually deuterium (in the form of "heavy" water), or the less expensive graphite.

Very fast neutrons with an energy of several Mev will bring about the fission of U-238 (21), or Th-232. When any fission process takes place a great number of radioactive

fission products are formed, no matter whether the fission is in the form of the uncontrolled atomic bomb, or in a nuclear reactor. If the mass number of each fission product is plotted against its fission yield there results a curve having two peaks. For the slow neutron fission of U-235 these peaks occur at about mass numbers 95 and 139, and detectable amounts of elements having mass numbers between 72 and 161 are also produced.(2) The distribution curves for the slow neutron fission products of the fissionable isotopes are very similar, but the peaks of the curves shift slightly for each one. When fission is carried out with 14 Mev neutrons the nuclei split up into two almost equal halves. In the case of U-235, 14 Mev neutrons produce 100 times more medium sized fission products than do thermal neutrons, although the two peaks of greatest abundance still predominate. (2,3)

The Sr-90 yield from U-235 (thermal) is about 5.6% (4) but this is almost entirely produced by radioactive decay. For the elements of mass number 90 there are independent fission yields only for Kr-90 and its very short lived precursor Br-90, with the independent yields of Rb-90 and Sr-90 contributing only about 15% and 0.1% respectively of the total mass 90 yield. Most of the mass 90 radioisotopes have quite short half-lives, and quickly decay away until the long-lived Sr-90 is reached. Since the fission products generally have a neutron surplus most decay by β emission along a chain of constant mass number until a stable end product is reached. Such is the decay system for the mass 90 chain, which is as follows. All the isotopes are β active with the respective half-lives given in brackets.

Kr-90 -(33 seconds) - Rb-90 -(2.7 minutes) -
Sr-90 -(28 years) - Y-90 -(64.4 hours) -
Zr-90 -(stable).

The 28 year half-life of Sr-90 (5,6) and the 64.4 hours half-life of its daughter Y-90 (7,8) mean that a Sr-90 source will reach complete equilibrium with Y-90 after about 18 days. At equilibrium the disintegration rates of both isotopes are equal, and this important fact will be referred to many times.

The quantity and distribution of fission products from a nuclear explosion depends upon the power and type of the bomb, as well as the position from which it is fired.

The fission temperature of a bomb is high enough to bring about the fusion of the heavier isotopes of hydrogen; deuterium (mass 2), and tritium (mass 3). A cheaper bomb would result if deuterium could be used in place of the radioactive tritium, which has to be made by the interaction of Li-6 with neutrons in a nuclear reactor. The temperature of the fission explosion is not maintained for a sufficient length of time to enable the fusion of deuterium alone to take place. It is necessary to use a mixture of deuterium and tritium to initiate the fusion reaction, which, once started, enables the fusion of deuterium to take place. If a fission "trigger" is used to set off the fusion of a large mass of deuterium/tritium an explosion of great magnitude will result. This is the thermonuclear, or "hydrogen" bomb. The first fusion explosion used liquefied hydrogen isotopes. Subsequent thermonuclear devices were designed to produce a dropable "dry" bomb, and these probably use Li-6 as a source of tritium, and combined with deuterium, and perhaps tritium, in the form of lithium hydrides. The fission products from this type of device can naturally only come from the "trigger" which can be quite small, and so this is termed the "clean" bomb, as most of the explosive power comes from the fusion reaction.

As it was noted earlier, U-238 will undergo fission if the neutrons are energetic ones, and the fusion reaction produces vast quantities of surplus fast (14 Mev) neutrons. If a relatively small fusion bomb is surrounded by a large mass of natural uranium (mainly U-238), then a fission - fusion - fission reaction will take place, in which most of the explosive power will come from the U-238 "blanket". This device will release great quantities of fission products compared with a fission - fusion bomb (of equal magnitude) described in the last paragraph, and is therefore known as the "dirty" bomb. It should be realised that the terms "clean" and "dirty" are relative terms, and while they are widely used to denote the two main types of H-bomb their implications are misleading.

The explosive power of a nuclear bomb is expressed in the weight of conventional chemical explosive (as tons of T.N.T.) which would give the same release of energy. The "nominal" bomb, such as the Hiroshima one, has an equivalent yield of 20,000 tons of T.N.T., or 20 kilotons. Bombs with yields greater than 1,000,000 tons are expressed in megatons, but in the case of H-bombs only part of this will be from fission.

The rate of radioactive disintegration of a particular nuclide is measured in curies: one curie equals 3.7×10^{10} disintegrations per second. The approximate quantity of Sr-90 produced by fission is 146 curies (or 1.14 gm.) per kiloton of U-235 fission, and is lower for higher neutron energies, and for masses above 235. (4)

The mechanism of fallout whereby the fission products from a nuclear explosion are deposited on the ground is of three main types.

(i). Close-in fallout. When a bomb is exploded on the

ground, or near enough so that the fireball sucks up material off the ground, the heavier particles are deposited near the test site, and for several hundred miles down-wind. Ground material will become impregnated with fission products, and the surplus neutrons in and around the fireball will induce radioactivity in the soil and other debris sucked up by the rising cloud of hot gases from the explosion. Induced radioactivity will be a lesser problem for explosions over water, and close-in fallout will be almost negligible for an air burst which is high enough to prevent material being sucked up off the ground.

(ii). Tropospheric fallout. The cloud of hot gases from the explosion, containing the fission products, will continue its upward movement until cooling prevents further rising, and it then starts to follow the air currents which may carry it several times around the world, at about monthly intervals. This world-wide circulation generally takes place in the same latitude as the test site, and so the kiloton weapons do not contribute a great deal to the global deposition. Nearly all the debris is washed down by rain after a month or two. It has been reported (9) that on one occasion the bomb cloud from a Nevada test was involved in a thunderstorm 36 hours after the test, and 2,300 miles away, which caused the local gamma background radiation to be more than doubled over a period of ten weeks. This kind of localised fallout should not be neglected when calculating average world-wide fallout, since this could not have been termed close-in fallout.

(iii). Stratospheric fallout. The upper and lower atmospheric regions (stratosphere and troposphere) have quite a distinct boundary layer (tropopause) between them, and little circulation of air takes place in a vertical direction across this boundary. A hydrogen bomb in the megaton range

produces a sufficiently large fireball to raise the fission products through the tropopause and into the stratosphere. The radioactive cloud may rise to a height of 30 kilometres, or about 100,000 feet.

At its highest point at equatorial latitudes the tropopause is at about 20 kilometres (65,000 feet), and it decreases until, over the poles, it is only at about 10 kilometres (33,000 feet). The turbulent winds of the stratosphere appear to distribute the fission products over the hemisphere in which the explosion occurred. British H-bomb tests were carried out at latitude 2° N, the American tests at 11° N, and the Russian tests at about 45° N, so that most of the radioactivity has remained in the Northern Hemisphere.

Based on measurements of the seasonal variations in the total quantity of ozone in the atmosphere, Dobson (10) suggested that the mass of cold air which forms over the poles during the winter sinks during the spring, sweeping ozone-rich air downwards into the troposphere. Ozone is produced at above 30 kilometres, and some 8% of the total ozone is in the troposphere. By using high-flying jet planes for sampling, Dobson found extremely little water vapour at 15 Km. and concluded that this could only happen by the air passing through the very cold regions of the upper troposphere and lower stratosphere near the equator, with subsequent condensation and precipitation.

In 1949 Brewer (11) used measurements of helium and water vapour distribution to conclude that air enters the stratosphere at the equator where it is dried by condensation, travels in the stratosphere to temperate and polar regions and sinks into the troposphere. This theory supports the fact that there is little movement of fission products in either North/South direction, and maximum fallout occurs in

the middle latitudes of both hemispheres, but predominately in the Northern Hemisphere. The spring rise in ozone concentration is accompanied by a spring rise in fallout (12,13,14), but Martell (15) submits that these increases are mainly due to weapon testing the preceding autumn and winter, and that this indicates a short residence time in the stratosphere. Libby (16) has presented evidence to support Martell's theory of a short residence time, especially in the case of the Russian tests, although on earlier evidence he had believed that the mean stratospheric residence time to have been about 10 years. (17) Feely (18) gives a stratospheric residence half-time of 4 - 9 months for Russian tests, and 9 - 15 months for the American and British tests.

Another method of measuring the retention time is to use long-lived natural radioactivity as an atmospheric tracer, and such a study has been carried out by Burton and Stewart (19). They measured the ratio of radium-F (Po-210, an α emitter with a half-life of 138.4 days) to radium-D (Pb-210, a β emitter with a half-life of 19.4 years); both of which are decay products of the gas radon (Rn-222, an α emitter, half-life 3.8 days) one of the decay products of uranium. Results give a value of 22 days for the mean residence time of those atoms the whole airborne life of which is spent in the troposphere. By comparing the ratio in equatorial air with that in air in the lower stratosphere above Britain, the period of circulation is of the order of 6 months.

Lal and Peters (20) have suggested using cosmic ray produced radioisotopes as tracers for studying large-scale atmospheric circulation. These are Be-7 (half-life 53 days), P-32 (14.3 days), P-33 (25 days), and S-35 (87 days).

Deposition is almost entirely by "rainout" and not "fallout", although the latter term is always used to describe the overall process. Measurements in Britain of Sr-90 in rain have shown that fallout is proportional to rainfall. (12) The mechanism of rain scavenging is discussed by Greenfield (46), and the variation of the specific activity of rainwater with the size of the shower is given by Stewart et al. (12)

The preceding discussion on the production of Sr-90 on fission and the various meteorological systems and fallout patterns has only briefly touched upon these topics. All the general information has been mentioned in many papers by several authors, but the most complete and authoritative descriptions are given by Libby (22,23,24), Martell (15), and Machta (25,26,27). *

The most reliable published figures for the total fission yields from all bombs exploded up to the end of 1958 are those presented to the American Congressional Hearings on Fallout (29). These state that between 3rd October 1952 and 23rd September 1958 Britain exploded 21 nuclear devices. It is estimated that the total yield of all bombs exploded up to the end of 1958 was 69.2 megatons from air bursts, and 104.6 megatons from surface bursts. The total FISSION yields from air, ground surface, and water surface bursts, are respectively 37.8, 21.5, and 32.6 megatons.

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* Information regarding the political and scientific background to the production of the first atomic bomb, and the subsequent development of the hydrogen bomb, their manufacture and testing, were obtained from the books listed under Reference 28.

Data from American and British tests are more accurately known than those from Russian tests, and the total yield from the megaton weapons only, amounts to 58 megatons by these two nations. Based on a world-wide soil sampling programme, Holland (30) has estimated the total deposition of Sr-90, excluding local fallout, as 2.5 - 3 megacuries, as of mid-1958; with 1 megacurie of Sr-90 representing 10 megatons of fission. Alexander (31) has estimated the total ground deposition as 3.2 megacuries of Sr-90 by 1st October 1958. Later data of Walton's quoted by Kulp et al (32) show that at the end of 1959 the total Sr-90 ground deposition was 4.5 megacuries. At this period Feely (18) estimated a stratosphere reservoir of 0.6 megacuries of Sr-90.

Although this thesis is concerned with Sr-90 production and fallout, there are several other long-lived fission products formed in greater amounts than Sr-90. None of these other has the bone-seeking properties of Sr-90, and are not able to enter the food cycle with the facility of that isotope. The relative quantity of the isotopes produced depends upon the bomb material, and neutron energy. The following isotopes are the most important ones.

Caesium-137/Barium-137^{m}.
Cerium-144/Praseodymium-144.
Zirconium-95/Niobium-95.
Promethium-147.

Although Sr-89 has a half-life of only 50.4 days (33-36) its activity on fission is so much greater than the Sr-90 activity that even 1 year after fission the Sr-89/Sr-90 activity ratio is more than one.

Data tables are available (2,3) which show the decay characteristics of the fission products, and graphs (1) showing the relative activity of the more abundant isotopes after a short, medium, and long decay periods from a few

hours to 100 years, calculated for U-235 (thermal).

Radioactive isotopes which are not present as fission products may be formed by neutron activation. For example, Co-60 may be formed in significant amounts due to the presence of nickel and/or cobalt in the structural material of the bomb. It was found after one of the 1956 tests in the Marshall Islands that the fallout ratio of Co-60/Sr-90 was 0.27 (37). Weiss and Shipman (38) did not find any Co-60 in fallout exposed materials 1 year after a 1954 Marshall Island test, but two clams recovered 2 years after detonation showed 210,000 d.p.m. of Co-60 in 1,800 gm. wet weight; and 705,000 d.p.m. of Co-60 in 882 gm. wet weight. This points to an enormous concentration of this isotope.

The most hazardous isotope produced by neutron activation is C-14, which is a low-energy β emitter with a long half-life of 5,600 years. This isotope, of course, is produced by either fission or fusion bombs from atmospheric nitrogen, and in amounts proportional to the energy released. Libby (23) has calculated that to produce in the atmosphere an amount of C-14 equal to that naturally present by cosmic neutron activation would take 1,000 megatons of fission. Fission produces less surplus neutrons than fusion. Pauling (39) on the basis of Libby's information concluded that the somatic effects of bomb-produced C-14 are expected to be about equal to those of fission products, including Sr-90, with respect to leukaemia and bone cancer. The somatic effects will be greater than those of fission products with respect to diseases resulting from radiation damage of tissues other than bone tissue and bone marrow. Pauling does state that the estimated numbers of defects are subject to great uncertainty.

Totter et al (41) state that Libby's calculations are based on incorrect assumptions, and a much smaller quantity than 1,000 megatons of fission will produce an amount of C-14 equal to that which is naturally present in the atmosphere. It is also pointed out that the transmutation of C-14 to nitrogen, in genetic material, will produce roughly the same number of mutations as the radiation effect produces. The rate of entry of C-14 into human beings has been discussed by Broecker et al (42,43).

C-14 data has been used by Hagemann, Gray, Machta, and Turkevich (40) to give an indicated stratospheric residence time of 5 years for fallout debris. Since the C-14 is in the form of carbon dioxide, it is an ideal atmospheric tracer. This residence time agrees with the value obtained by Machta and List (27), which was based on Sr-90 data.

In order to obtain the whole picture of the dangers arising from nuclear explosions, consideration must be given to isotopes other than Sr-90. Although this isotope has become synonymous with fallout in the public mind, other fission products must also be investigated. The long-term hazard of C-14 might be greater than the Sr-90 hazard, for there has not been the extensive work done on C-14 that has been done on Sr-90 fallout.

DEPOSITION IN THE SEA.

The amount of Sr-90 deposited in the sea would be difficult to estimate, but this fallout could never be as hazardous as that falling on to the land. Measurements of the quantity of natural strontium in sea water show a calcium/strontium ratio of about 100/1, with a strontium concentration of about 8 mgm./litre (55-58). It appears that the natural strontium content is a constant fraction of the total salinity, since although the salinity increases with depth the Sr/Cl ratio remains the same. Due to the great dilution involved the measurement of Sr-90 in sea water is rather difficult; 100 litres may have to be analysed (59,60,61).

Bowen and Sugihara (62) found appreciable concentrations of Sr-90 at 1,000 - 1,200 metres in the Atlantic; and water columns had 3 - 4 times as much Sr-90 below 100 metres as above. While some marine algae (63), and plankton (55), have been found to take up strontium preferentially, many marine invertebrates discriminate against strontium (64). Boroughs et al (65) kept several species of sea fish in sea water to which Sr-89 had been added. When equilibrium had been reached the quantity of Sr-89 in the fish was about 0.3 of the Sr-89 in an equal weight of sea water. Using rainbow trout, Shiffman (66) found that the trout concentrated strontium from the water by a factor of 1.5 on an equal weight basis. It is probable that the uptake depends on the available calcium in the environment. Bidwell and Foreman (170) have carried out measurements on the Sr-90 uptake of weeds and fish growing in an enclosed pond spiked with Sr-90. They found that the accumulation factor for the weed was 270

compared with an equal weight of water; and that the fish bones contained 70 times the amount of Sr-90 in an equal weight of water.

ADSORPTION BY THE SOIL.

It is not generally realised that Sr-90 does exist "naturally", if only in trace amounts. In the same way that radium exists "naturally" by being one of the radioactive decay products of uranium, so Sr-90 is produced by the natural or spontaneous fission of uranium. Kuroda and Edwards (44) found by analysing pitchblende (in which the Sr-90 and U-238 decay schemes were in equilibrium) that one kilogram of U-238 was associated with about 13 micro-microcuries of Sr-90, and approximately the same amount of Sr-89 activity. This "natural" Sr-90 is hardly available to the biosphere, and so can be ignored.

E.A.Bryant et al (45) have made a study of the biological availability of Sr-90 from various types of atomic bomb tests. They measured the fraction of 1N HCl soluble debris collected from the bomb clouds a few hours after detonations from towers and balloons at Nevada, and from megaton ground bursts at the Pacific Proving Grounds. If the sucked up debris condenses after all the Kr-90 has decayed to solid Rb-90 or Sr-90, then the Sr-90 radioactivity may be trapped inside the particles, and will not be available to plants. The Pacific shots produced debris consisting of fluffy conglomerates of calcium carbonate from coral, and crystalline particles of sodium chloride from sea water. After contact with 1N HCl for 7 days this type of debris was completely soluble. Debris from the Nevada tests was rather less soluble, but only small kiloton weapons were tested there.

In all reported cases when rain containing dissolved fission products falls on soil the radioactivity is adsorbed in the top inch or two of soil, and soil carried away by

runoff was found by Menzel (47) to contain 10 times the concentration of Sr-90 compared with that in the remaining topsoil. Deposition and leaching tests carried out in the laboratory show that these microquantities of radiostrontium are strongly adsorbed by soils, but the adsorbed strontium is comparatively weakly held in the soil and may be displaced almost completely back into the solution by leaching with neutral salts such as calcium chloride or sodium nitrate (48). Both the mineral (clay) portion of the soil and any organic matter present are the binding agents. In cases where the soil contains carbonates the adsorption will be stronger due to chemical binding.

To obtain a mathematical model of the downward movement of radiostrontium (49,50) many factors have to be taken into consideration, including the cation-exchange capacity of the soil, strontium concentration in the soil, and the chemical composition of the rain and the soil solution. This water composition appears to be very important. Leaching experiments on columns of material spiked with Sr-90/Y-90 showed that tap water removed the adsorbed radioactivity much more quickly than distilled water (51), and 90% of this activity was due to Sr-90 alone. When Sr-90 solution was added to the core to the "breakthrough" point, and then the core washed with distilled water, the activity of the effluent falls away rapidly. If carbon dioxide saturated water is run through there is a large release of radioactivity of which only 5% is due to Y-90.

This preferential desorption leads to difficulty in estimating the amount of "available" Sr-90 when dealing with uptake by plants. Schultz et al (52) found that the major fraction of the Sr-85 added to several soils was extractable with ammonium acetate or calcium chloride solutions, no

matter whether the soils had been dried at 50°C, or at 110°C prior to leaching. Also, Hungate et al (53) found that Sr-85 newly applied to a field plot was absorbed into barley to the same extent as Sr-90 which had been applied to the same ground 3 years earlier. Cultivation seems to be the most important factor, since the same Laboratory has published reports showing that barley growing on field plots for three successive years removes more Sr-90 from the soil as the time between contamination and cropping increases (202,203). Nishita et al (54,174) found that of all the medium- and long-lived fission products Sr-90 is about the most water soluble and exchangeable isotope in several soils and clays.

To sum up, all fission products in rain are adsorbed strongly in the top layer of soil, but radiostrontium is the most easily removed by leaching, especially where the leaching solution contains dissolved salts.

UPTAKE BY PLANTS.

Plants are able to take up fission products either through the roots or directly through the aerial parts. In the absence of current bomb tests uptake by the roots will be the main process of accumulation, and a great deal of work has been carried out to determine the extent to which plants take up radiostrontium in competition with calcium.

By varying the concentrations of strontium and calcium in nutrient solutions, plant uptake of strontium was found to be proportional to the total Sr + Ca concentration, that is, the plants do not discriminate between the two (67-72,171). In solutions of strontium only, the uptake was proportional to the strontium concentration (71,73). Rediske and Hungate (73) showed that the uptake of Sr-90 from solutions was increased by decreasing the solution pH from 7 - 4.

There are two modes of uptake by roots. In the first instance the roots act as a cation exchanger, and then the absorption proper is by active transport of the ions by the plants' metabolic processes (166).

This simple relationship can not be applied to the uptake of Sr-90 from calcium-bearing soils, if only the total soil calcium and Sr-90 are considered. Misleading results may be obtained even if the ratio of Sr-90 to the "extractable" calcium is employed, due to the difficulty in measuring these components (69,70,74,193). "Exchangeable" ions are those which can be displaced from the soil by solutions of neutral salts, such as ammonium acetate. Where the "exchangeable" soil calcium has been measured, the ratio of Sr-90/Ca in the plant is equal to the ratio in the

soil extract (75-82,174,201), and Sr-90 uptake is inversely proportional to the calcium concentration. What is required to be measured is the ratio in the "Equilibrium Soil Solution", defined by Russell et al (83) as the composition of the solution with which a soil would be in equilibrium under field conditions, when its ionic concentrations would remain constant irrespective of the ratio of soil to solution. Schulz et al (52) state that when the free electrolytic concentration of the soil is low the Sr/Ca ratio in a water extract of the soil is to a good approximation a measure of this activity ratio of strontium and calcium in the soil. Graham (84) has found that the electrodialysis of soil will remove the exchangeable cations, and 96% of the adsorbed Sr-90.

For acidic, and calcium deficient soils, the Sr-90 uptake by plants is greatly reduced by adding calcium, but this effect is small when the exchangeable calcium reaches the value of about 7 milliequivalents of calcium per 100 gm. of soil (80,85). Schmehl et al (86) have suggested that the low calcium content of plants grown on acidic, calcium deficient soils may be due more to the acid-induced restricted root growth, than to the non-availability of calcium in the soil. In this case the addition of lime has the dual effect of neutralisation and increasing the calcium supply. Prout (49) has shown that maximum absorption of calcium occurred at pH 7.0. Adding too much calcium or stable strontium to a soil in an endeavour to reduce further the Sr-90 uptake by plants may have exactly the opposite effect (78,87,88). This is due to the Sr-90 adsorbed on the soil exchange complex being displaced into the soil solution where it is more readily available to plants. Bowen and Dymond (169) analysed the natural calcium and strontium content of soils and plants, and found some plants which

appeared to be strontium accumulators, but these were growing on soils with a very high calcium and strontium content, which may be the reason for the high strontium uptake.

Experiments have been reported where the addition of potassium salts to soils or nutrient solutions reduced Sr-90 uptake (48,81,89).

The chemical composition of the Sr-90 in soils has a bearing on the uptake by plants (90); in general, the soluble forms have much greater availability than insoluble forms. For acid soils the insoluble strontium compounds are equally available, but for alkaline soils $SrHPO_4$ was the least available, so that the addition of phosphate may reduce Sr-90 uptake. Similar experiments on the availability of calcium salts, by Ririe and Toth (173), showed that calcium from $CaCO_3$ was more available to alfalfa than calcium from $Ca(H_2PO_4)_2.H_2O$ or from $CaSO_4.2H_2O$.

Russell and Milbourn (168) have carried out experiments which indicate that contamination on herbage which is afterwards ploughed into the soil, is more readily available to plants than contamination directly incorporated into the soil.

When a layer of Sr-90 contaminated soil was placed at different depths in non-contaminated ground, the uptake by barley was reduced ten-fold when the spiked layer was changed from 2 inches down to 2 feet down (91). This is due to the root growth, and the difference was less between the two layers when the ground was not irrigated, suggesting a deeper root growth in this case. With surface contamination there was no difference in the Sr-90 uptake of barley when the ground was surface or sub-surface irrigated (92).

Where contamination is mainly on the surface soil the

depth of feeding of plants becomes an important factor in the uptake of Sr-90 (81). Nishita et al (175) have shown that on prolonged cropping up to a quarter of the Sr-90 added to soil was removed by 9 crops of clover, over a period of 17 months.

Sr-90 which has been absorbed through the roots collects in that part of the plant where current growth is most vigorous, and there is little or no translocation of the Sr-90 once it is laid down in the plant (48,71,93,171). This deposition is mainly in the stems and leaves (80,82,190, 191). With cereals only a small fraction of the total Sr-90 absorbed is deposited in the grains, and most of the activity in the grain is in the outer bran part, and will therefore be removed on milling (48,191,192,95,96). The direct uptake of Sr-90 by grain from rain will be expected to remain on the outside parts (94), so that this extra activity will add to the difference between the bran, and the endosperm and germ parts (192).

Wiebe and Kramer (172) showed that most translocation of minerals to the shoots occurs from a region of the roots several centimetres behind the root tips, at the region of maximum water absorption.

Plants are able to take up fission products dissolved in rain directly through the leaves. Morgan (97,132) has shown that 20% of the Sr-90 activity in rain is directly taken up by the aerial parts of ryegrass. By using simulated rainfall conditions Middleton (98,99) showed that the Sr-90 taken up by the leaves of a wide variety of plants is fixed on the part of the plant where the drops fell; there is no translocation. Similarly it was found that any translocation of activity from the leaves of directly deposited Sr-90 is due almost entirely to the daughter Y-90 (48,167).

There is a special case of direct absorption by the aerial parts of grass where this is in the form of permanent pasture having a thick root mat. Here, the Sr-90 activity in rain is not able to penetrate this mat and become equilibrated with the soil calcium. Russell (74) has called this "stem-base" absorption; and for some pastures 80% of the Sr-90 content of grass has not passed through the soil. The absorption effect is greater for plants growing in soil submerged in water, which can be looked upon as an extreme case of stem-base absorption. Measurements of mosses by Gorham (100,101) have shown high activity due to this effect. Activity was the same for mosses growing under wet and dry conditions which indicates that rainfall activity is fixed at the spot where it falls, due to the strong absorption by the root mat and soil.

UPTAKE BY ANIMALS.

The main source of dietary calcium for people in this country is cows' milk, and related dairy products (102,236). It is necessary, therefore, to be able to predict the level of Sr-90 contamination of milk which would result from a known Sr-90 level in the cows' diet. For convenience, the effects of tracer amounts of radiostrontium, and radiocalcium, on animals are studied under laboratory conditions, rather than by relying on fallout isotopes to provide the radioactive tracers. Many species of laboratory animals have been used in these experiments to find out the metabolic movement of Sr-90 after ingestion. The radiological effect of Sr-90 on animals has been studied in an attempt to assess, by extrapolation, the probable effects of large doses of Sr-90 on the human body.

In order to describe the overall discrimination which occurs in the movement of radiostrontium and radiocalcium from one place to another in a biological system, Comar, Wasserman, and Nold (103) proposed the term "Observed Ratio" (O.R.) which is defined as :

$$\text{O.R.(sample-precursor)} = \frac{\text{Sr/Ca of sample}}{\text{Sr/Ca of precursor}}$$

This term will be referred to often.

The need to use many replicates for statistical certainty means that rats are generally used in tracer feeding experiments where large numbers have to be sacrificed. An analysis of the radiostrontium content of the leg bones is usually sufficient in calculating the body retention of the isotope. Uptake of radiostrontium by rats has been shown to be dependent on the type of diet. Some amino acids and lactose, administered with the radiostrontium and radio-

calcium will double the retention of these isotopes (104, 105); while absorption of calcium in milk fed to rats is 1½ times the absorption from radiocalcium chloride solution, or from calcium chloride plus a grain diet (106). Deposition of strontium and calcium also depends on the amounts of calcium, phosphate, carbonate, and lactate in the diet (107,108,176,177).

Comar, Russell, and Wasserman (70) found that when rats were fed on a milk diet the O.R.(bone-diet) was 0.57. For an ordinary non-milk diet the O.R. was 0.27, a figure which is in agreement with other published results of similar experiments (109,110,176), using both double tracer experiments, and stable strontium measurements. These figures are from experiments where radioisotopes had been fed to the rats over a long enough period to ensure equilibrium. Where a single dose of radiostrontium and radiocalcium is fed or injected into animals it is laid down in that part of the bone undergoing active growth at that time. In experiments with adult rabbits (111) these sites of high activity persist, but with young rabbits they are almost entirely lost, and the active material becomes diffuse throughout the bones due to secondary deposition. After a single injection of radiostrontium to rabbits the activity in the teeth was followed (112). The concentration increases for about 30 days then falls off abruptly, due to the initial localised deposition wearing away. Analysis of the femurs showed a steady decrease in activity after the initial deposition.

Experiments carried out on pregnant rats (109,148) showed that the O.R.(fetal bone-maternal diet) was 0.20 for the fetus. For the growing rat the O.R.(bone-diet) was 0.28, so that just under 30% of the fetal calcium was derived from the maternal skeleton, the remainder from the

maternal diet. Bronner (113) has experimentally calculated for rats that 10 - 15% of the offspring's bone calcium, and the same amount of the calcium in the mother's milk, comes from the skeletal calcium of the mother. Radiocalcium studies by Comar (114) have shown that there is a rapid transfer across the placental barrier. Finkel (115) showed that when fatal doses of Sr-89 were given to mice, pregnant animals survived longer than the non-pregnant controls due to the transference of Sr-89 by the placenta to the offspring, and then through the milk.

The laboratory animal whose metabolic processes most closely resemble those of human beings is the monkey, and radiostrontium toxicity studies have been done by Ward; and Edington, Judd, and Ward (196,197,198). They found a straight line relationship when the Sr-90 body burden, and the survival time were plotted on logarithmic scales. It was estimated that the dose to give a survival time of 3 years was approximately 18,000 μμc Sr-90/gm. of bone calcium. It will be mentioned in the next section that the maximum permissible level of Sr-90 in bone for certain human workers is 1,000 μμc/gm. calcium.

Amounts of Sr-90 secreted in sheep's milk (116) were the same whether the Sr-90 was orally administered as the nitrate, or in plant material grown on contaminated ground. When Sr-90 as the nitrate was fed to sheep for 14 days apparent equilibrium was reached by the 5th day, and at that time about 4% of the Sr-90 ingested appeared in the milk. Lambs fed on a varying calcium diet, containing Sr-89, for over 20 days took up less Sr-89 as the calcium content of the diet increased (117). These experiments were continued and it was found that, compared with ewes, lambs showed a greater discrimination against strontium in favour of calcium in bone deposition (118). The transfer of maternal

skeletal calcium to the offspring, and to the mother's milk, was determined by Benzie et al (178,189). When the daily calcium intake was only 2 grams the maternal skeletal loss of calcium was 18.2% by mid-lactation, which was not replaced until 2 months after the end of lactation. The skeletal distribution of fallout Sr-90 in yearling sheep was investigated by Morgan and Wilkins (179) who found the lowest Sr-90/calcium ratio in the teeth; which may be due to accretion, and a diet of varying activity.

Wasserman, Lengemann, and Comar (119,194) carried out an extensive study on the comparative metabolism of calcium and strontium in lactating and non-lactating goats, using double tracer techniques. At steady state the O.R.(milk-diet) was 0.09. With data from the non-lactating animals it was found that the largest discrimination occurred in the gastrointestinal absorption of the two elements, with calcium absorption being 3 - 4 times the strontium absorption. The relative Sr/Ca ratios were:- diet - 1; plasma - 0.23; skeleton - 0.23; milk - 0.09; urine - 4.0. This means that there is no differential movement between blood and bone, and that strontium is preferentially excreted in the urinary process. The calcium in blood and extracellular fluid undergoes very rapid exchange with calcium in the bone (120).

The same facts appear from tracer studies of cows. Many research teams have been and are working on the evaluation of the O.R.(milk-diet) for lactating dairy cows, and results lie within the range 0.08 - 0.16 (17,70,121,124, 125,126). Cox, Morgan, and Tayler (126) determined the O.R.(milk-diet) of a herd of between 69 and 78 cows by measuring both the Sr-90 and stable strontium content of diet and milk. The study lasted about 15 months and took into account the varied forms of grazing over that time;

there being 5 distinct grazing periods with a total of 19 sample groups. From the Sr-90 measurements the mean O.R. for the 5 periods, and the standard deviation, was 0.10 ± 0.03. The summer values were lower than the winter ones probably due to the difficulty in obtaining a representative sample of the pasture. For 2 of the 19 sample groups stable strontium determinations were carried out, and both gave an O.R. of 0.11, which is in excellent agreement with the Sr-90 method. Cragle and Demott (125) found that for cows (as for goats) strontium was preferentially excreted in urine, with an O.R.(urine-diet) of 1.82, from a single dose of Sr-89 and Ca-45. The movement of radiocalcium in cows following oral and intravenous administration has been described by Visek et al (180), Swanson et al (181), and Squire et al (195).

In experiments to determine whether radiostrontium in milk could be replaced by calcium, Easterly, Demott, and Cragle (127,128) used cation exchange resins in the calcium form. They showed that complete or near complete exchange needed large quantities of resin compared with the quantity of milk used. They also found that ordinary milk spiked with tracer solutions gave slightly higher absorption effects than milk from cows fed with the same tracers. Even after standing for 16 hours equilibrium was not quite reached between the milk and the tracer solution. This difference in the availability of radiostrontium from milk and ordinary solutions might explain the difference in absorption of radiostrontium by animals on milk and non-milk diets, which was mentioned earlier.

Using milk containing Sr-89 and Ca-45 to make Cheddar Cheese, Demott and Cragle (129) found a slightly higher proportion of Sr-89 in the cheese than was in the milk.

The average O.R.(cheese-milk) for 14 vats was 1.23 ± 0.28. The significance of Sr-90 in milk has been the subject of reviews by Larson (130,131).

Comparisons of the Sr-89/Sr-90 ratios in milk and rain for the same periods show that much of the radiostrontium in milk, and therefore in grass, must be of recent origin, so that milk analyses will give an average value for the current level of pasture contamination (70,102,126,133,134,135). To obtain an indication of the Sr-90 levels of pasture Hawthorn and Duckworth (136) proposed using the Sr-90/calcium ratio of deer antlers, since these are grown rapidly and may be expected to contain proportionately more Sr-90 than the skeleton of the animal.

In the section dealing with the translocation of radioisotopes within plants it was noted that parent/daughter isotopes were translocated by different amounts (p. 24). This also happens in animals with some tissues. The large accumulation of iodine by the thyroid gland has long been known, but there may be other accumulative organs as yet unsuspected. The differential distribution of Sr-90/Y-90 in the eye of the rabbit showed large departures from the equilibrium ratios when the separate sections of the eye were analysed (137). Rabbit eye sections also accumulate more Ba-140 than the daughter La-140 (138). These differences can be expected in human eyes, and this accumulation of radioisotopes by particular organs may necessitate a revision of the maximum permissible concentrations of these isotopes, especially in the case of sensitive organs such as the eye.

UPTAKE BY HUMAN BEINGS.

For obvious reasons few tracer experiments have been carried out on human beings to determine the fate of radiostrontium. The addition of the short-lived Sr-85 isotope to two volunteers showed an initial rapid loss, finally leaving the relatively non-exchangeable part deposited in the bone (139). Comar et al (70) found an O.R.(bone-diet) in man having milk in the diet of 0.54; and for rats 0.57. Spencer et al (140) injected Sr-89 into four patients and found that the main excretion is via the kidney, and the rate of Sr-89 excretion appears to depend upon the state of bone metabolism. Further experiments by Spencer et al (141) showed that ammonium chloride increased the rate of excretion of Sr-89. 15 days after injecting nine patients with Sr-89 9 grams of ammonium chloride were given orally each day for 5 days. The amount of Sr-89 removed was less for those patients on a high-calcium diet and greater for those on a low-calcium diet.

Comar et al (142) studied strontium and calcium metabolism using radioisotopes with fatally ill patients, and found that the behaviour of radiostrontium in man is similar to that in animals. When given orally as a solution, 74% of the radiostrontium was excreted via the faeces, and 6% via the urine. When radiostrontium and radiocalcium were fed in milk for several days there was a discrimination against strontium in favour of calcium by a factor of about two. Due to the probable variation in the metabolism of these patients used for the experiments the results might not be typical of the average human being. Schulert et al (182) showed that for a single intravenous

dose the net retention of radiocalcium was 60%, whereas for radiostrontium it was about 25% since it was preferentially excreted by the kidneys.

To overcome this difficulty of using radioisotopes, measurements of the stable strontium content of human bone and diet will give the overall discrimination against strontium. A report by Bailey, Bryant, and Loutit (123) gives a simplified account of the role of calcium and strontium in the human body. Reports by Bryant et al (143-146) give the stable strontium content of human bones in this country for all age groups, and similar stable strontium measurements have been made to give an average value for the diet in Britain (102,147,236). A comparison of the two shows an overall discrimination factor of about four against strontium relative to calcium, in their passage from diet to bone.

It is not the object of this thesis to attempt to assess the hazards of Sr-90 to man when absorbed into the body. Nevertheless it is relevant to note that recommendations have been given by the United Nations Scientific Committee on the effects of Atomic Radiation (149), a summary of which has appeared in the journals 'Nature' (150), and 'Science' (151). A similar report was published earlier by the Medical Research Council (152), and in a Supplement of the British Journal of Radiology (153). A summary of the report of the Committee on Genetic Effects of Atomic Radiation is given in 'Science' (184).

The Maximum Permissible Concentration (M.P.C.) of Sr-90 in the human skeleton is put at 1,000 μμc of Sr-90/gm. of calcium for occupationally exposed workers; 100 μμc/gm of calcium for the population average; and 10 μμc/gm. of calcium for the population average as the "level which would indicate

the need for immediate consideration of the problem." (152, p. 128) A further report by the Medical Research Council (237) is to be published shortly. The 3rd report by the Agricultural Research Council Radiobiological Laboratory (236) suggests that the M.R.C. now accepts that the maximum permissible concentration of Sr-90 in human bone, for the average person in the population at large, should be 67 μμc/gm. of calcium, and that the maximum for individual members of the population should be reduced to 200 μμc/gm. of calcium. Any reassessment of the 10 μμc/gm. of calcium "warning level" must await the Medical Research Council's recommendations. Caster has criticised the M.P.C. value of 100 μμc/gm. of calcium for the population mean by calculating that certain minority groups will greatly exceed this value.

The Medical Research Council has recommended maximum permissible levels of dietary contamination after the accidental release of radioactive material from a nuclear reactor, assuming that only a very small proportion of the population is exposed to the hazard (122). The daily intake should not exceed 2,000 μμc of Sr-90/gm. of calcium, so that the concentration at sites of new bone growth will not exceed 500 μμc/gm. of calcium. For Sr-89 the corresponding maximum intake is 200,000 μμc of Sr-89/gm. of calcium. It is assumed that immediate steps will be taken to reduce the intake of radiostrontium as soon as possible after any accident which releases dangerous quantities of fission products.

A general treatment of the principles for dose-rate calculations for bone-seeking isotopes, and an extensive bibliography is given by Engstrom, Bjornerstedt, Clemendson, and Nelson (154). Bjornerstedt and Engstrom (155) indicate that Sr-90 taken into the body over a long period is less of a hazard than a single dose of the same magnitude,

due to the diffuse nature of the bone deposit of the first process. Bjornerstedt and Haggroth (183) have described a method to determine the distribution of low levels of Sr-90 in bone using long exposure autoradiographs. Bone doses and the biological hazard from Sr-90 have been discussed by Newcombe (186), Norris et al (187), and Libby (188).

A great deal of work has been done to forecast the future levels of Sr-90 which would result from the bombs exploded up to the present time; i.e., up to the end of 1958 when the testing of megaton weapons stopped. Earlier papers by Kulp, Schulert, Hodges, and Eckelmann (156,157,158) arrived at predictions which were too high in estimating future levels of Sr-90 in human bones since the stratospheric reservoir of Sr-90 was believed to be greater than in fact it was. The fourth article by these authors (32) uses the latest available data, which is that by the end of 1959 the stratosphere contained 0.6 megacuries of Sr-90, and that about 4.5 megacuries had been deposited on the ground. The Sr-90 in the human diet in Britain depends to a great extent on the rate of fallout rather than upon the total deposition in the soil (159). Since the total surface deposit will reach its maximum in 1961 and the rate of fallout is diminishing quickly, the maximum bone Sr-90 concentrations will be reached in the next year or two, in the early 1960's.

It is extremely difficult, if not impossible, to determine the minimum human bone level of Sr-90 which will cause bone tumors or leukaemia. Any estimates have been made by extrapolation from animal experiments, or from the study of the damage caused by the accidental human ingestion of radium (160). Finkel (161) concluded that the minimum effective dose in man is from 6 - 15 microcuries based on

the radium method, and 5 - 10 microcuries by extrapolating from experiments on mice, dogs, and cats. A threshold dose was proposed between 5 - 15 microcuries of Sr-90 in man. These conclusions have been the subject of criticism (162). Archer and Carroll (163) disagree with Finkel's assumption that the dose/effect relationship is non-linear (indicating a threshold) and point out that another interpretation of the data is possible which implies the opposite. Kamb and Pauling (164) state that there is no justification for Finkel's conclusion that there is a threshold of 5 - 15 microcuries, but that the data ~~is~~ are compatible with a zero threshold for Sr-90 in man.

The report of the Ad Hoc Committee of the National Committee on Radiation Protection and Measurements, 6th May 1959, (165) concluded that the present data are still insufficient to establish the character of the dose/response curve for somatic effects.

Part II.

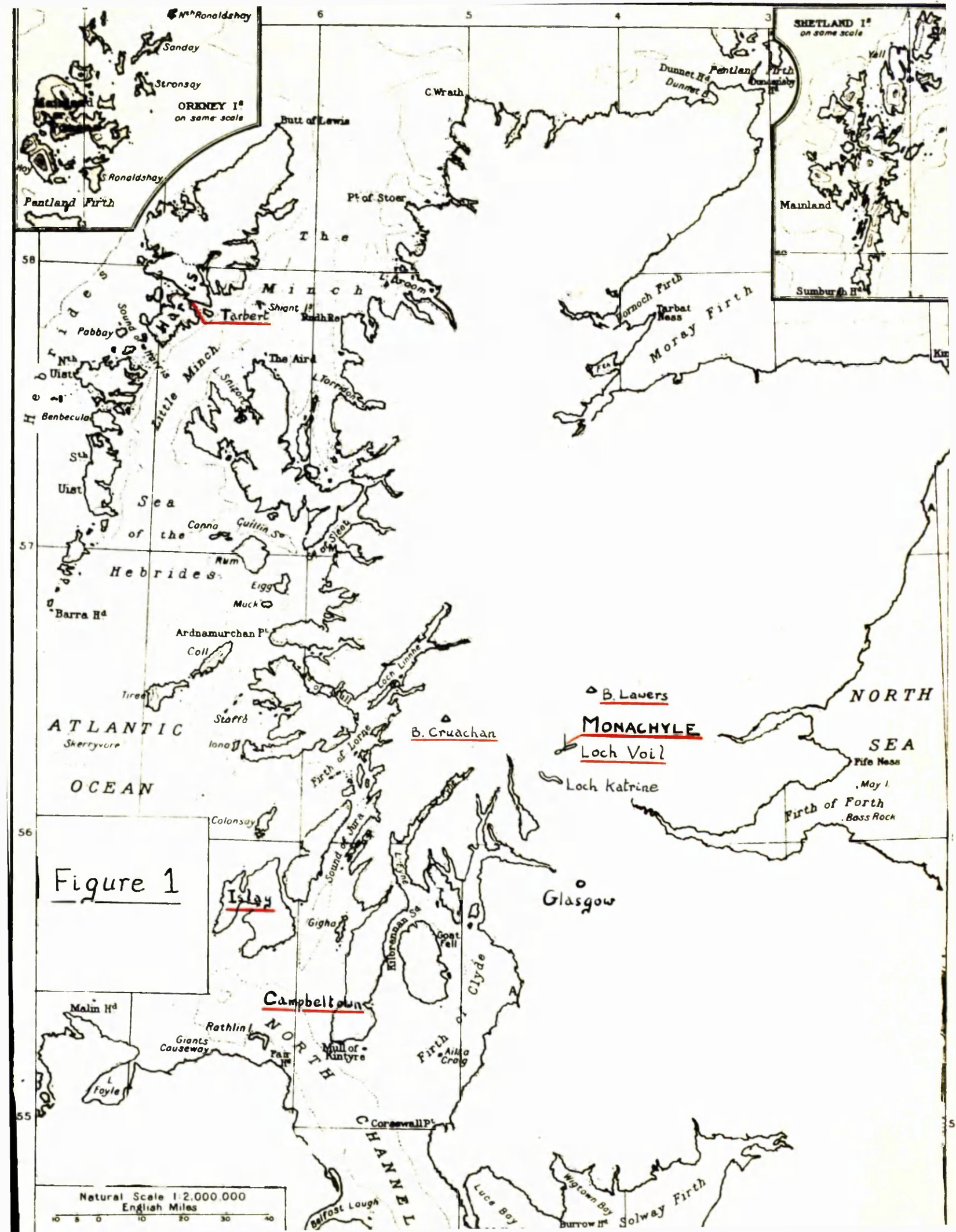
EXPERIMENTAL.

SAMPLING.

The areas from which samples were collected for analysis needed careful selection. They must satisfy several conditions. A knowledge of the agricultural history of the land is necessary to evaluate results from plants growing upon it. The samples chosen from a particular area or field must be representative and not isolated samples; so that personnel responsible for the sampling must be aware of the need to combine many samples in order to achieve an average selection. The radiochemical analysis is made easier where the sample contains sufficient radiostrontium to give good count rates from small quantities of the material sampled. For example, a grass sample of 100 gm. dry weight is sufficient for duplicate Sr-90 determinations where the activity of the grass is 3,000 μμc of Sr-90 per kilo dry weight. Where possible the samples should be from an area having a high rainfall, which will give high levels of fallout activity.

The choice of samples for this study was in keeping with the following aims. To study the magnitude of current fallout, and seasonal trends. To determine the extent of fallout deposited before the present study. To measure the uptake of radiostrontium by plants with a view to correlating these data with the levels in animal bones and in milk. From the above to seek any trends which might point to an estimate of the future levels of radiostrontium in the environment studied.

One area has been thoroughly investigated (Monachyle - Balquhidder region), and from two others (Campbeltown and Harris) a few samples have been obtained. In addition,


ORKNEY Is on same scale
SHETLAND Is on same scale
Mainland
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Dunnet Hd
C Wrath
Butt of Lewis
Pt of Stoer
The Minch
Tarbert
Shiant Is
Raasay
Pabbay
Sound of Harris
Little Minch
The Aird
L Shiel
L Torridon
L Broom
Tarbat Ness
Moray Firth
Benbecula
Uist
Sea of the Hebrides
Canna
Rum
Eigg
Muck
Barra Hd
Ardnamurchan Pt
Coll
Tiree
Staffa
Iona
ATLANTIC OCEAN
Skerryvore
B. Cruachan
Firth of Lorne
Loch Linnhe
Sound of Mull
B. Lawers
MONACHYLE
Loch Voil
Loch Katrine
NORTH SEA
Fife Ness
May I
Firth of Forth
Bass Rock
Colonsay
Sound of Jura
Islay
Gigha
Kilbrannan Sd
Goat Fell
Glasgow
Firth of Clyde
Campbeltown
Malin Hd
Rathlin I
Giants Causeway
Fair Hd
Mull of Kintyre
Ailsa Craig
L Foyle
NORTH CHANNEL
Coreswall Pt
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Natural Scale 1:2,000,000
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10 5 0 10 20 30 40
Figure 1

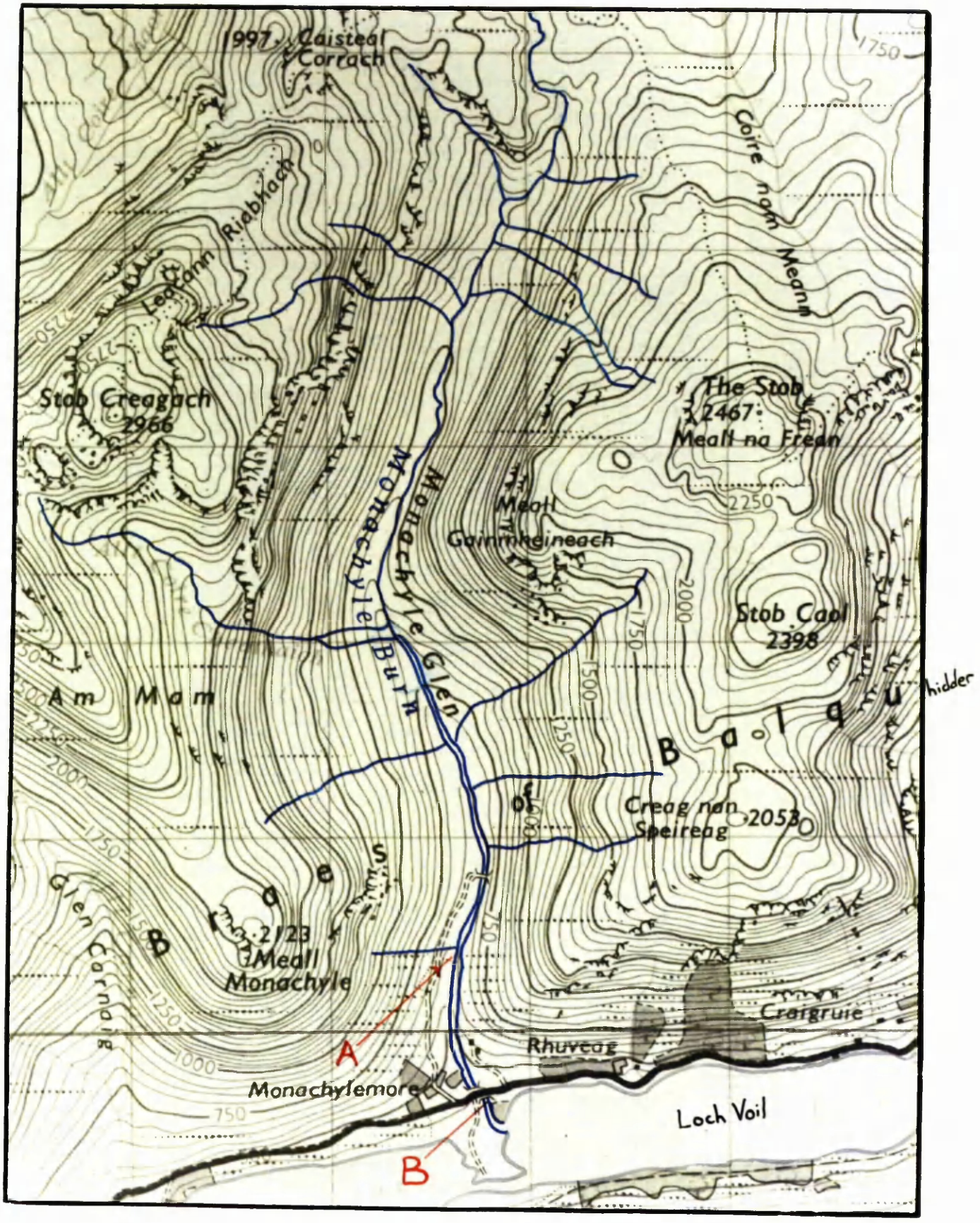

Figure 2. Monachyle Glen .

samples of animal bones have been collected from Perthshire and surrounding regions similar to Monachyle; and some deer antlers were obtained from Islay.

Monachyle.

At the beginning of this investigation the need was felt to study the fallout in a location which would be typical of the catchment area which supplies the domestic water to Glasgow and West of Scotland. Permission was granted by the landowner to set up a semi-permanent rain collection station on private land in the next valley to the north of Loch Katrine (the main reservoir of the Glasgow water board). This valley is in the Braes of Balquhidder area, and is known as the Monachyle Glen. Monachyle is indicated on the map of Scotland. (Figure 1.) The detail of Monachyle Glen is enlarged from the latest (1957) Ordnance Survey map (Sheet 54); Figure 2. It is readily seen from the contours that the Glen is surrounded by steep hills so that runoff empties into the central Monachyle Burn, which flows south into Loch Voil. The surface of Loch Voil is 414 feet above sea level. The local farm is at Monachylemore, which is reached by the road shown as a thick black line along the north shore of the loch. A small herd of Ayrshire cows is kept, and sheep are grazed on the hills. The cows graze on pasture which is limed (2 tons/acre) every four years; this was last done 2 years ago (1958).

On 27.1.59 a rain collector was erected by the side of the burn, and its position is shown on the map as point A. Figure 3 is a photograph of the rain collector tower, and Figure 4 shows a general view of the country, looking south. The collector is a square funnel made from 'Perspex',

Figure 3. Monachyle Rain Collector.

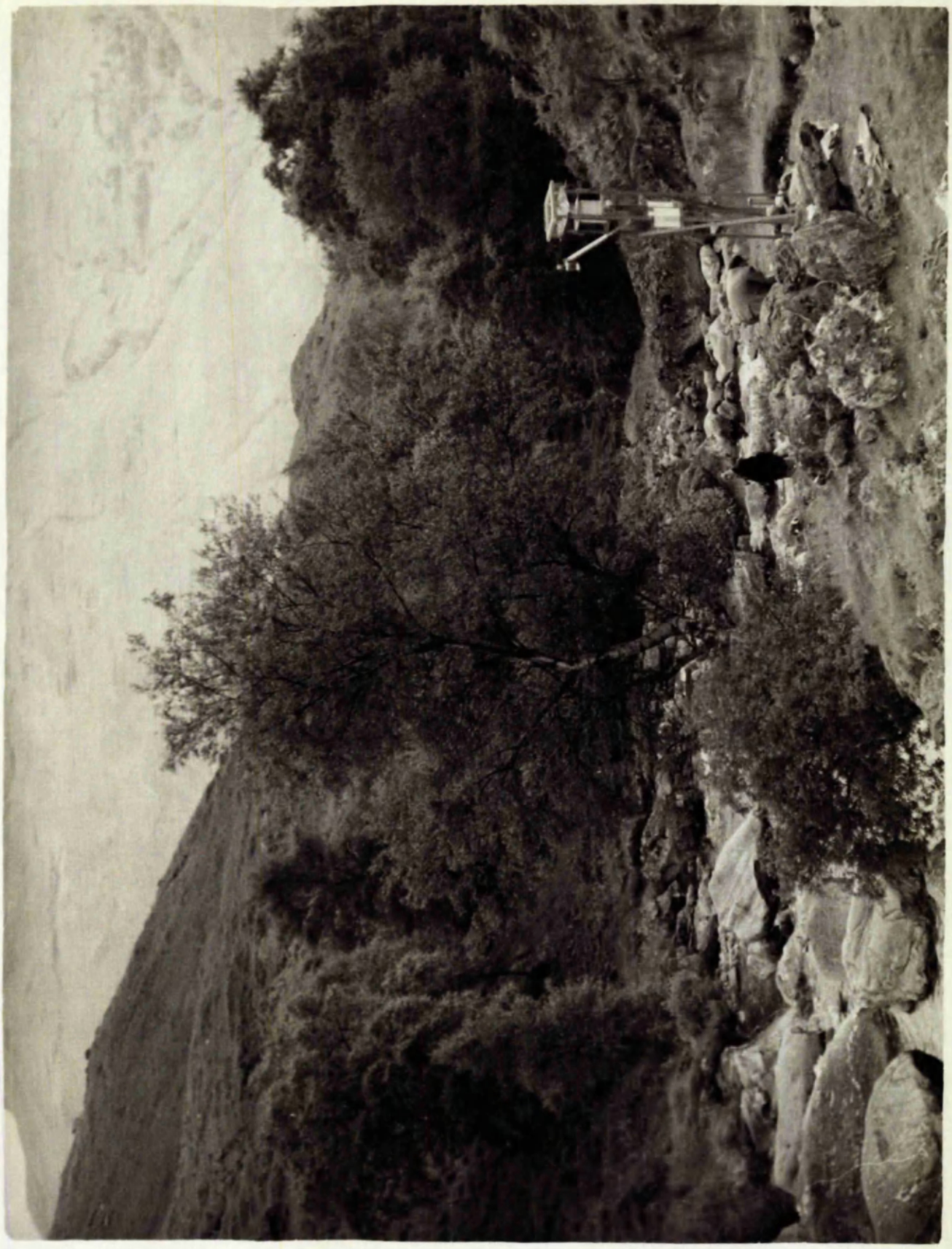

Figure 4. Monachyle Burn.

draining into a 10 litre polythene bottle. Originally the aperture of the funnel was 16 inches square, but due to the high rainfall this was covered by a 'Perspex' lid having an aperture of 11 inches square. On collection trips the rain collected is run off into four $2\frac{1}{2}$ litre Winchester bottles, which can be seen in figure 3. Strontium carrier and a little nitric acid are added to each Winchester previously to prevent adsorption of radio-strontium on to the glass. The volume of additives is subtracted to obtain the volume of rain water.

The rain gauge shown was added later (15.7.59) and it consists of a 4 inches high funnel with a 500 ml. collection bottle. The funnel was turned from 2 inches diameter aluminium rod, and it has an internal diameter of 1.875 inches (i.e.,a cross-sectional area of 17.81 sq. cm.) with a fine chamfer at the top giving a sharp edge. The outside was coated with several layers of matt black to increase heat absorption in an attempt to prevent icing-up in winter. To cut down losses by evaporation approximately 50 ml. of n-heptane (b.pt. 98^{o}C; s.g. 0.68) is poured into the 500 ml. collection bottle. The Air Ministry Meteorological Office have a rain collection station at Balquhidder (Tulloch Lodge) which is also situated on the northern side of the loch, at a point four kilometres along the road to the east of Monachylemore farm. Daily readings from this station have been obtained from the Air Ministry, and the following table gives a comparison between the Monachyle rain gauge and the Tulloch Lodge rain gauge totals over the same period. The rain is measured in, or converted into, centimetres. The numbers 7 - 15 refer to the collection periods; the dates for each period are given in Table 1 of the results.

PERIOD	MONACHYLE	TULLOCH LODGE
7	16.1	14.0
8	8.5	7.2
9	34.8	35.0
10	25.5	26.2
11	28.8	27.0
12	27.4	27.4
13	17.9	15.9
14	16.6	16.8
15	12.4	11.2

The totals, (15.7.59 - 3.6.60) are:- Monachyle 188.0 cm., Tulloch Lodge 180.5 cm. The agreement is quite good considering the simple rain gauge at Monachyle, so that in all subsequent calculations the Air Ministry figures are used.

On every collection trip a 25 litre polythene bottle containing strontium carrier and a little nitric acid is filled to the top with water from the Monachyle Burn. To reduce the distance the full bottle has to be carried to the car, the water is sampled where the road crosses the burn, and the spot is indicated by the point B on the map.

The samples of grass, moss (Sphagnum spp.), rushes, sheep droppings, and soil from Monachyle were all taken between the rain collector and the farm, unless otherwise stated. Vegetation was cropped with shears and did not include any root or soil. The exception to this was in the case of moss growing on soft ground where some brown stems were included with the green tops. Only recent sheep droppings were taken.

Most soil samples were taken with a corer made from a 2 inches diameter iron pipe, graduated in inches on the outside. On one occasion a small pit was dug with a shovel. The places where the soil cores were cut had a

root mat 1 - 1½ inches thick, which was analysed separately on some occasions. Where average soil samples were required 12 cores were taken between the rain collector and the farm, the soil being of a sandy-clay nature.

Several sheep carcases were found in the Monachyle Glen and along the shore of Loch Voil, and these may be considered to have grazed the same hill pastures of the Glen. Both the Monachyle and Perthshire sets of deer antlers came from animals which must have grazed over a wide area in Perthshire and beyond.

Sheep bone samples have been collected from the hills in Perthshire and Argyllshire. Two of these places, Ben Lawers (Perth) and Ben Cruachan (Argyll), are indicated on the map of Scotland; and show the approximate region from where these bones were obtained.

Campbeltown.

Some preliminary samples were sent from Carradale, Campbeltown, Argyll, between September 1958 and April 1959 with a view towards carrying out a more detailed survey should results prove to be interesting. Several samples of kale, which is used for animal feeding, were taken for analysis. It was thought that when the kale began growing again in the spring, after standing in the field over winter, the roots would quickly take up Sr-90 from the soil. Kale is a typical leaf crop which is grown on prepared fields. Due to the large leaf area it was thought that exceptionally high Sr-90 activity levels might be found after long exposure to the rain, and this activity would find its way into human bone by way of cows' milk. Since the land was well cultivated and limed, the Sr-90

activity of the kale was quite low (see Results) so that no more samples were collected from Campbeltown, and attention was focussed on the more interesting and accessible Monachyle. A sample of three brown trout and one sample of oats were also sent for analysis.

Harris.

Three samples of trout and a grass sample were sent from the Isle of Harris, near Tarbert, in September 1958. Transportation of samples, and the lack of supervision of sampling, made this location unsuitable for further study.

Islay.

Several samples of deer antlers were obtained from this island. These are of interest due to the enclosed area of grazing for the deer, so that a study of the annual variation in Sr-90 activity of the antlers over several years would be extremely useful. Since the antlers are shed and regrown each year Sr-90 analyses would serve to give an approximate average Sr-90 content of the island's vegetation for the period of growth of the new antlers.

Greenland.

A single sample of grass was brought back from southern Greenland by a member of this College during the late summer of 1958, and is included for general interest.

METHODS OF ANALYSIS.

The problem of Sr-90 analysis can be divided broadly into three parts:- (1) the extraction from the material under examination of the radioactive and non-radioactive strontium present; (2) the separation of this strontium from all other elements present, resulting finally in chemically pure strontium carbonate; (3) the counting and calculation of the amounts of Sr-90 and Sr-89 which may be present. This section deals with the basis of the chemical manipulations required to produce the radioactive strontium component of the materials to be analysed in a form which is suitable for counting.

One great handicap to the analyst is due to the minute quantities of natural strontium usually associated with the radioactive strontium isotopes. This quantity is not sufficient to be separated directly, and the unavoidable losses in the chemical separations would result in the loss of at least an unknown portion of this strontium, if not all of it. The second handicap is the abundance of the chemically similar element calcium, found associated with strontium in all the materials, with the sole exception of rain water, and to a lesser extent, burnwater. In high calcium samples such as bone and milk this becomes a serious obstacle. The problem of little natural strontium is overcome by adding a known amount of stable strontium carrier to the weighed portion of sample before the start of the analysis, and provided that the quantity added is many times greater than the natural strontium present a yield correction can be made at the end of the analysis to allow for what has been lost.

Although analysis of natural strontium is beyond the scope of this thesis, published data on the amounts present in biological materials and water in the U.K. show that in human bones the natural strontium concentration is between 200 - 350 p.p.m. of calcium (143-146,199,200). In Glasgow drinking water the strontium concentration is 4 - 8 mgm./gm. of calcium, and the calcium concentration is about 2 mgm./l. The amounts of natural strontium in these substances is very low compared with the amount of added strontium carrier.

(a). Rain water:

When the rain collector is full its total capacity can be divided into four $2\frac{1}{2}$ litre bottles. Before leaving for the sampling site 6 mgm. of strontium carrier (1 mgm. Sr/ml.) are run into each bottle from a burette, followed by 9 ml. of 5N nitric acid. Approximately the same quantity of rain water is run into each bottle until the rain collector is empty. The Winchester glass stoppers are then sealed with 'Sellotape'or firmly screwed tight if of the plastic screw-on type. The added carrier and acid prevents adsorption of radioactive strontium from the rain on to the glass surfaces. On return to the laboratory the water is evaporated to about 20 ml. Very little atmospheric pollution is found in the rain water from Monachyle so the water is not filtered at any stage.

(b). Burn water.

To give a convenient level of radiostrontium activity for counting purposes a much larger sample of burn water is taken, compared with the rain water. 5 ml. (25 mgm.) of strontium carrier are pipetted into a 25 litre

polythene bottle, 35 ml. of 5N nitric acid are added, and distilled water (60 ml.) to make the total volume of added solutions 100 ml. Burn water is taken from as far out into the middle of the fast flowing part as is possible, a stainless steel scoop being used to fill up the polythene bottle to a mark on the inside of the neck, giving a known volume of water - 26.45 litres, not counting the additives. The cap is screwed on tightly and the bottle rolled over once or twice to mix the contents. On return the water is evaporated to about 20 ml. as was done for the rain water. The burn water was always clear and required no filtering, but due to the peaty nature of the surrounding hills the water became brownish on evaporation. This did not interfere with the analysis as determined by the final chemical yield of strontium.

(c). Bones and antlers.

These require little treatment and, after first removing any flesh on the bones, they are simply weighed and ashed. The most convenient ashing temperature is 750 - 800°C in an electric muffle furnace, and the bones are left in until a white carbon-free ash is obtained, generally after 4 - 5 hours. On cooling, the friable pieces of ash are broken up with a mortar and pestle, and finally reduced to a fine powder in an electrically driven rotary grinder. Samples are weighed and bottled.

(d). Milk.

Murthy et al (215) have described a method for the direct and rapid ashing of milk without preliminary evaporation; and also a method to eliminate ashing by adding

strontium carrier to the milk followed by trichloroacetic acid (216). After filtration the strontium and calcium are precipitated from the filtrate by adding sodium hydroxide and sodium carbonate; but the quantities of trichloroacetic acid needed would be a serious drawback where many samples have to be analysed. Owers (219) has used an electro-dialysis technique to separate the calcium and strontium from the organic constituents of diluted milk.

The method used for the Monachyle samples is to evaporate one Winchester bottle of milk in a large porcelain dish on a hot-plate until a hard charred mass is obtained which can easily be chipped out in large flakes. These are placed loosely in silica trays, and ignited in the muffle furnace. Too high an ashing temperature gives a hard semi-fused ash which is difficult to work with, so that the sample is left overnight in the furnace at 650°C, with the furnace door ajar to admit air. This results in a soft purewhite ash, which is easily ground to powder.

(e). Vegetation.

Grass, rushes, and moss were generally cut just above ground level. Kale leaves were separated from any thick stalks. Samples were first dried in a cabinet tray drier through which hot air at about 70°C was blown, then, after weighing, were ashed at 650 - 700°C, and the ash weighed. Sheep droppings were dried, weighed, ashed, and re-weighed as for the vegetation samples.

(f). Soil.

Samples were dried and weighed. The top layers of soil which contained much vegetation, or included the

root mat, were ashed where only the total strontium activity was to be determined. If it was intended to determine the available calcium and Sr-90 in the soil, then the soil was not ashed. After ashing the soil was sieved through a 30 mesh wire gauze to break up the lumps, and any stones over about 3 mm. size were removed, the smaller ones being mixed with the sieved soil, or soil ash. The stones were weighed and the weight subtracted from the dried weight of soil. Ashing was also omitted where a sample contained little organic material, and where the size of the sample made ashing impracticable, in which case the dried soil was sieved and any stones removed.

(g). Fish.

It was considered neither necessary nor practicable to analyse only the bones, and since most of the calcium (and therefore Sr-90) is contained in the skeleton, ashing the entire fish results in what may be taken as a "bone ash".

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Only the broad outlines of the analytical methods are dealt with in this section; the details of the chemical separations are to be found in the Appendices at the end.

The chemical manipulations are divided into five sections which will be considered separately.

(1). Initial treatment, resulting in the solution of all the radioactive strontium present.

(2). Preliminary separation of calcium and strontium together from the bulk of the other interfering elements.

(3). Removal of calcium from the strontium.

(4). Removal of radioactive contaminating trace elements.

(5). Radioactive strontium counting and Y-90 separation and counting.

The analytical methods are based on those published in various U.K. Atomic Energy Authority Research Group Reports (121,204-208), the U.S. Atomic Energy Commission Health and Safety Laboratory Manual of Standard Procedures (209), and other sources (4,210-213).

(1). Initial treatment.

This is necessary where an ash is not at all soluble in nitric acid, and is obviously not used where the ash is completely soluble as with bone and milk ashes, and water. An alkali fusion will result in the complete solution of an ash but the additional alkali salts make the subsequent treatment difficult, and a simpler method is preferred. The one most commonly used for soil and vegetable ashes is to evaporate to dryness with strong nitric and perchloric acids. With 5 gm. of vegetable ash, strontium carrier is added, 50 ml. of distilled water, 35 ml. of fuming nitric acid, and 38 ml. of 72% perchloric acid. The mixture is evaporated to dense white fumes in a beaker on a hot plate. This is of necessity done in a fume cupboard. With soil ash large quantities are involved due to the low activity, and the liquid volumes are increased accordingly so that the acids that are evaporated are best condensed and collected to avoid damage to the fume cupboard extraction system. A 2 litre round-bottomed "Quickfit" flask is fitted with a water condenser and collector; traces of acid fumes are absorbed in a side arm at the cold end containing sodium hydroxide pellets, so that a closed system is avoided.

The residue is extracted by boiling with 10% hydrochloric acid, and filtering. After washing with water, the residue and filter paper are emptied into a beaker of suitable size and again boiled with dilute HCl and filtered. This process is continued until it is estimated that all the soluble material has been extracted from the residue. A useful indication of this is the colour of the filtrate due to iron leached from the soil, a colourless filtrate coming from the last extraction. All the filtrates are combined and evaporated to a suitable volume.

One sample of 1.5 kgm. of dry soil was extracted with 6N HCl, by leaving the mixture standing one working day with occasional stirring, and then left overnight. After centrifuging the liquid the residue was washed with water and again centrifuged. The residue was once more left overnight in 6N HCl and the extraction repeated, after which all the supernates were combined and evaporated to a suitable volume.

It was shown by Volchok et al (211) that if a soil sample was extracted three times with 6N HCl, 99% of the leachable activity was removed by the first extraction. If the residue from the final HCl extraction was then fused with sodium carbonate, the quantity of Sr-90 removed was 40% of the total from all the extractions. On another sample of ashed soil, therefore, several methods of extraction were carried out to find the differences in the degree of extraction possible. The methods were:- (i) evaporation with nitric and perchloric acids; (ii) evaporation with concentrated hydrochloric acid; (iii) low temperature sodium hydroxide/sodium carbonate fusion; (iv) high temperature sodium carbonate fusion. The residues from (iii) and (iv) were extracted with perchloric acid then evaporated to

dryness, and extracted with boiling 10% HCl. The residues from (i) and (ii) were leached with boiling 10% HCl.

All these methods of soil extraction result in a clear solution containing, in the case of the fusion extractions, large amounts of sodium salts. In addition, the solution has a strong orange colour from the ferric chloride present (with associated aluminium) from the clay part of the soil. The next stage removes these unwanted salts. The few grams of vegetable ashes analysed give little trouble, and do not involve the manipulation of large quantities of solution as is the case with soil analyses.

(2). Preliminary separation.

With soil extracts this takes the form of a calcium and strontium oxalate precipitation. Sufficient solid oxalic acid is added to the solution to complex all the iron present; using 100 - 200 gm. of soil ash 100 gm. of oxalic acid and 15 gm. of ammonium acetate are used. The solution is heated to 70 - 80°C, then ammonia is added to give a pH of 4, as determined with narrow range pH paper. A precipitate of ferric hydroxide before pH 4 is reached means that there is insufficient oxalic acid present to complex all the iron. The minimum amount of dilute HCl to dissolve the ferric hydroxide is added, and more oxalic acid, so that on raising the pH again to 4 only the white oxalate precipitate is seen. After the hot solution has been standing for a few hours the calcium and strontium oxalate precipitate is filtered, ignited to the carbonates, and taken up in dilute nitric acid. Any carbon is filtered off and rejected. Traces of iron and aluminium are removed by precipitating them as hydroxides with ammonia. The remaining calcium and strontium are

precipitated as carbonates and filtered into a tared sintered glass filter crucible containing a glass fibre filter paper. This weight, after it has been corrected for the quantity of strontium carbonate present, gives the percentage calcium in the ash. Dilute nitric acid is used to dissolve the mixed carbonates, and the solution is evaporated almost to dryness.

With the smaller quantities of extract from vegetable ashes a simpler method is used, consisting of precipitating all the insoluble phosphates present by the addition of phosphoric acid, followed by excess ammonia. The precipitation is done in a centrifuge bottle to eliminate the transfer of precipitate. After centrifuging, the supernate is discarded, the residue washed, and the mixture is centrifuged again. Fuming nitric acid is added to the residue in the centrifuge bottle which dissolves the phosphates. The solution is transfered to a beaker and either evaporated to dryness, or more fuming nitric acid added to precipitate the strontium nitrate. This method can be usefully employed when handling large amounts (over 10 gm.) of milk ash.

(3). Calcium separation.

At the end of stage (2) the solution contains strontium and calcium, with the latter in excess; sometimes in large excess. Also present are trace quantities of other elements which are dealt with in stage (4). The method of calcium separation used throughout is the one involving fractional crystallisation of strontium nitrate in strong nitric acid as described by Willard and Goodspeed (214). With an acid concentration of over 80% w/w, calcium nitrate begins to precipitate, so that successive recrystallisations

at 74% w/v strength will result in calcium-free strontium nitrate. For complete precipitation of strontium nitrate an acid concentration of about 79% w/v is necessary, but gives rise to calcium contamination, especially where large quantities of calcium are present. Some strontium is lost at the lower concentration used in practice, so this means that the total volume of acid solution used for a crystallisation should be kept to a minimum. The best method is to start with a higher acid concentration (76 - 78% w/w) when working with larger volumes of solution, and decreasing the acid concentration, and the total volume, in subsequent recrystallisations. The point at which no more calcium is present can be judged visually with experience.

It would obviously be a great advantage if an easy, clean, one-step method were available for separating calcium from strontium in an aqueous solution; and such methods have been devised, or proposed. Some have been put forward for the rapid analysis of milk as part of a Sr-90 monitoring programme. One method is to use raw liquid milk which is run through a cationic resin column under conditions where both calcium and strontium are absorbed, using the sodium form of the resin (217), or the strontium exchanged with the calcium form of the resin (218). Since no strontium carrier is added to the milk the exact yield is not accurately known, although an average figure may be experimentally derived which would be sufficiently accurate for a quick monitoring service. Other methods involve ashing the milk and extracting the calcium and strontium in a soluble form after adding strontium carrier. In this case both calcium and strontium can be absorbed on to a cationic resin and eluted separately (220), or the calcium can be complexed with ethylenediamine tetraacetic acid (EDTA) under very

carefully controlled conditions where the large calcium component of the milk ash passes through the resin column, and the non-complexed strontium ions are absorbed, and later eluted (221). Most methods suffer from the disadvantage that they are unable to handle large quantities of starting material, due to either column loading, or the large quantities of EDTA needed to complex all the calcium. The method by Milton and Grummitt (222) uses up to 15 gm. of milk ash, but here the resin column is thermostatically maintained at 80°C, and a nitric acid separation of traces of calcium from the strontium is necessary after column fractionation. A method separating calcium, strontium, and barium from a mixture using a cellulose chromatography column (223) proved unsatisfactory due to the overlapping of the fractions on elution, and the large volumes of organic eluate needed. The method of fractional crystallisation in nitric acid is the most flexible simple one of separating calcium from strontium.

(4). Trace contaminants.

Since the nitric acid concentration is adjusted to precipitate strontium nitrate, the remaining elements of Group II of the Periodic Table, namely barium and radium, are also precipitated as nitrates. Where materials, especially rain, are contaminated with fallout from recent nuclear tests a considerable quantity of Ba-140 may be present. Soil will naturally contain sufficient radium and thorium to give an abnormally high "Sr-90" count if these elements are not removed; an average value of one curie per square mile of radium in the top foot of soil has been quoted (224). Turner et al (225) have shown that the diet of an adult can contain more than 100 μμc of α-activity per day,

with the ratio of thorium to radium of about 0.75. Only about one tenth of the intake appears to be absorbed, but there seems to be little difference in the total α-activity concentration in human bones from all age groups.

These impurities are removed by "scavenging". "Scavenging", in a chemical sense, is the process of co-precipitating trace quantities of contaminants (in this case radioactive ones) using chemically related compounds. Due to the fact that radium salts are isomorphous with those of barium, a barium chromate scavenge will remove radiobarium and radium, and a ferric hydroxide scavenge is used to remove thorium, radium daughters, and any radioactive rare earths present. With soil samples two barium chromate scavenges are carried out to ensure complete radium elimination.

For the barium scavenge the pH of the solution is adjusted to about pH5 with acetic acid and ammonium acetate buffer to prevent precipitation of strontium chromate. The volume of the solution must not be too small or the strontium loss may be appreciable: a small percentage of the strontium is lost nevertheless (226). Murthy et al (227) showed that there was little effect on barium chromate precipitation by varying the pH between 6.5 and 3.5.

(5). Radiostrontium and Y-90 counting.

Where measureable quantities of Sr-89 are present a method is needed to differentiate between the two radiostrontium isotopes, Sr-89 and Sr-90. The half-life of Sr-89 (50.4 days) is too long to be followed conveniently, although this method could be used since the Sr-90/Y-90 equilibrium activity would not change significantly over a period of two months, due to the

long half-life of Sr-90 (28 years). The Sr-90 activity would actually fall only 0.4% over two months. A method which gives the result in a shorter time is required.

The method universally adopted is to measure the activity of the quantity of Y-90 associated with an equilibrium mixture of Sr-90/Y-90. At equilibrium the activity of each isotope is the same, so that measuring the Y-90 activity at equilibrium gives the Sr-90 activity directly. 97% of the equilibrium activity of Y-90 has grown in after 14 days from a Sr-90 source, and virtually 100% after 18 days. When the Sr-90 activity is known the Sr-89 activity is calculated as the difference between the Sr-90 activity and the mixed Sr-89 + Sr-90 activity.

The method used here to separate Y-90 from the mixed strontium isotopes is to precipitate the Y-90 (to which stable yttrium carrier has been added) with ammonia. The yttrium hydroxide precipitate is separated by centrifugation; dissolved in dilute nitric acid, and re-precipitated with ammonia to eliminate any occluded radiostrontium which may have been carried down with the first precipitate. After dissolving the hydroxide precipitate a second time in a little dilute nitric acid, oxalic acid solution is added, and the yttrium oxalate precipitate filtered. The filter paper is cemented on to an aluminium planchet, and the Y-90 activity measured.

A method of separating Y-90 into an immiscible organic liquid layer, from the aqueous solution, has been reported (227).

The Y-90 activity is measured every day for at least a week. The time of the first yttrium hydroxide precipitation is noted; this is the "zero time" for extrapolating the Y-90 decay curve, which is drawn from the daily observations.

Finally, the oxalate precipitate is removed from the planchet, and ashed to yttrium oxide (Y_2O_3), which is weighed to give the yttrium yield. The extrapolated Y-90 "zero time" count is corrected for both strontium and yttrium recovery yields, and the Sr-90 activity calculated.

Determining the mixed strontium activity can be done in either of two ways, which differ mainly in the time taken for each method. The first method is to precipitate the strontium from the two combined supernates left over after the yttrium has been precipitated and centrifuged. After weighing the strontium carbonate precipitate to determine the strontium recovery yield, the sample is left for at least 18 days for the Sr-90 present to reach equilibrium with its Y-90 daughter again, and the three nuclides (Sr-89, Sr-90, and Y-90) are counted in one sample. The calculated Sr-90 + Y-90 activity is subtracted from this total activity. Since the sample has to be left another 18 days to reach equilibrium prior to the Y-90 separation, this means that the sample has been set aside for a total of about six weeks while twice reaching equilibrium.

It would cut down the analysis time if the mixed radiostrontium activities were measured before any Y-90 had grown in; and this is the method used here. The ferric hydroxide scavenge described earlier removes all Y-90 present at that moment. If the strontium is then precipitated as carbonate, dried, and counted immediately, little Y-90 will have had time to grow in. The small amount which has grown in is calculated from the time of the iron scavenge to a point midway through the time of counting of the mixed radiostrontium: this amount is easily read off a time/activity graph for the growth of Y-90 from Sr-90, and subtracted from the mixed radiostrontium activity. When the strontium carbonate precipitate has been counted and weighed it is completely

dissolved in dilute nitric acid and stored in a small polythene bottle, with added yttrium carrier, until Sr-90/Y-90 equilibrium is reached, at which point the Y-90 is milked off and counted as described above.

It should be pointed out that taking the time of the iron scavenge as the point from which Y-90 begins to grow in again is a little arbitrary, since the strontium carbonate precipitation might not precipitate any Y-90 then present in solution. In practice it does not really matter whether the time at which the ferric hydroxide precipitation is made, or the time at which the strontium carbonate precipitation is made, is taken as the starting point for new Y-90 to grow in. The time between the two precipitations is only about 10 minutes so the amount of Y-90 which can grow in during this time is negligible; for example, after two hours about 2% of the equilibrium activity of Y-90 grows in from a pure Sr-90 sample.

A disadvantage of measuring the mixed radiostrontium activity after the yttrium hydroxide precipitations is that some strontium losses may occur in the process of precipitating strontium carbonate from the combined filtrates, and filtering the precipitate. Since the strontium yield therefore appears to be lower than it really was before the Y-90 was milked, the final value for the Sr-90 activity of the sample analysed will be higher than it should be. In the other method where the strontium carbonate precipitate is counted first, the corresponding losses (if any) will occur in the step where the precipitate is dissolved in dilute nitric acid and stored. During this operation the glass fibre filter paper used to hold the strontium carbonate precipitate is pulped in the acid, and the mixture filtered into a polythene bottle. Due to repeated washings with dilute nitric acid the losses of strontium in this

transfer is considered to be negligible. Chemical manipulations should be done where either strontium or yttrium carrier is present, otherwise losses can not be allowed for.

To sum up, the counting procedure briefly is as follows. Count the Sr-89 + Sr-90 carbonate immediately after the iron scavenge. Weigh the precipitate, dissolve it in dilute nitric acid, add yttrium carrier and store for 18 days. Separate the yttrium from the strontium by means of two yttrium hydroxide precipitations, and finally precipitate as yttrium oxalate. Count the Y-90 activity over a period of one week, then determine the yttrium yield by ashing to yttrium oxide and weighing. From the zero time Y-90 activity calculate the Sr-90 activity, and then the Sr-89 activity by difference.

Determination of calcium.

In samples where Sr-90 is associated with calcium, and the results are expressed in micromicrocuries per gram of calcium (Strontium Units) an accurate method for the determination of calcium is required. Gravimetric, volumetric, and physical methods are available.

Gravimetric: For samples with a high calcium content, and where 1 - 2 gm. of ash are available for the calcium determination, a simple gravimetric method is used (121,208). Bone, milk, and antler ashes are analysed in this way. 0.5 - 1.0 gm. of ash is dissolved in dilute HCl, oxalic acid is added, and the pH of the solution is increased to 4 with dilute ammonia. The calcium oxalate precipitate is left for a few hours to aggregate, then

filtered into a tared sintered glass crucible. The dry precipitate is weighed as calcium oxalate monohydrate.

Strontium and barium are also precipitated under these conditions, and if the concentration of these elements was thought to be high enough to give appreciable errors, a method has been described (226) for the removal of strontium and barium by ion exchange chromatography. The calcium is the first fraction eluted, and is a well-defined peak. This calcium is then precipitated and weighed.

In the case of soils the calcium is measured at the end of the first stage of the radiostrontium analysis. Here, the percentage calcium in the soil is required under the identical extraction procedure used for the radiostrontium analysis so that the two analyses are combined in one sample. When calcium and strontium oxalates are precipitated from the soil extract they are converted into the carbonates and weighed, the method being more fully described in the details for radiostrontium analysis of soils. The weight of strontium carbonate is subtracted after the strontium yield has been determined, and the percentage calcium in the soil is calculated.

Volumetric: In the standard volumetric method the oxalate precipitate (obtained as described above) is dissolved in dilute sulphuric acid , and the oxalic acid liberated is titrated against standardised potassium permanganate.

Physical: Methods have been described using emission spectrophotometry (228), atomic-absorption spectrophotometry (229), and flame photometry (238). The method used here is the one by Hemingway (230) using the "EEL" flame photometer; and is used for all vegetable ashes, and other ashes of low calcium content, and where there is not a sufficient quantity of ash to use the gravimetric method. Phosphate greatly interferes with the flame intensity and must be removed prior to measurement. One method (231) uses an oxalate precipitation to purify the calcium, and exchange phosphate for nitrate: the oxalate being dissolved in nitric acid. Hemingway uses a cation exchange column to absorb all cations, which are then eluted with 5N nitric acid and measured directly. This is a simple method which requires little supervision, and many samples can be handled at once.

Ashes which are insoluble in dilute acid are evaporated to dryness repeatedly with concentrated hydrochloric acid, and the residue extracted with boiling water.

Precise details of all the analytical methods used for radiostrontium and calcium determinations are given in the Appendices at the end.

COUNTING EQUIPMENT.

The three nuclides under study all emit β- particles of sufficient energy to make the use of end-window Geiger-Muller tubes ideally suited for the counting of them. Two types of G-M. tubes have been used in this investigation. The first type was an organic quenched GM4LB tube made by G.E.C., and having an aluminium window of thickness 7 mgm./sq. cm., and operated at about 1,200 volts. The second type was the halogen quenched EW3H tube made by 20th Century Electronics having a mica window of thickness 1.5 - 2.5 mgm./sq. cm., and operated at about 500 volts.

Two general purpose commercial counting assemblies are used to count the majority of samples which gave a count rate of over 50 c.p.m. These are manufactured by 'Panax', one being the model 100C with two hard valve decade units, and a four figure mechanical register to record hundreds. Samples are timed with a stop watch. The other is the Panax Automatic Counting Equipment type A.C. 300/6 which incorporates an automatic timer running at 100 pulses per second, and operating 6 Dekatron tubes so that a total counting time of 10^4 seconds (i.e. $2\frac{3}{4}$ hours) is obtained, after which counting stops. A pre-set time control can stop counting after .1, 4, 10, 40, 100, 400, 1000, 4000, or 10000 seconds; the 4000 second setting being particularly useful. The G-M pulses are fed into a similar row of Dekatron tubes, and it is also possible to time a pre-set number of counts from 100 to 1 million.

Each counting assembly is connected to a 1 inch thick lead castle holding the G-M tube; the scaler incorporates the necessary stabilised voltage for the G-M tube, and a

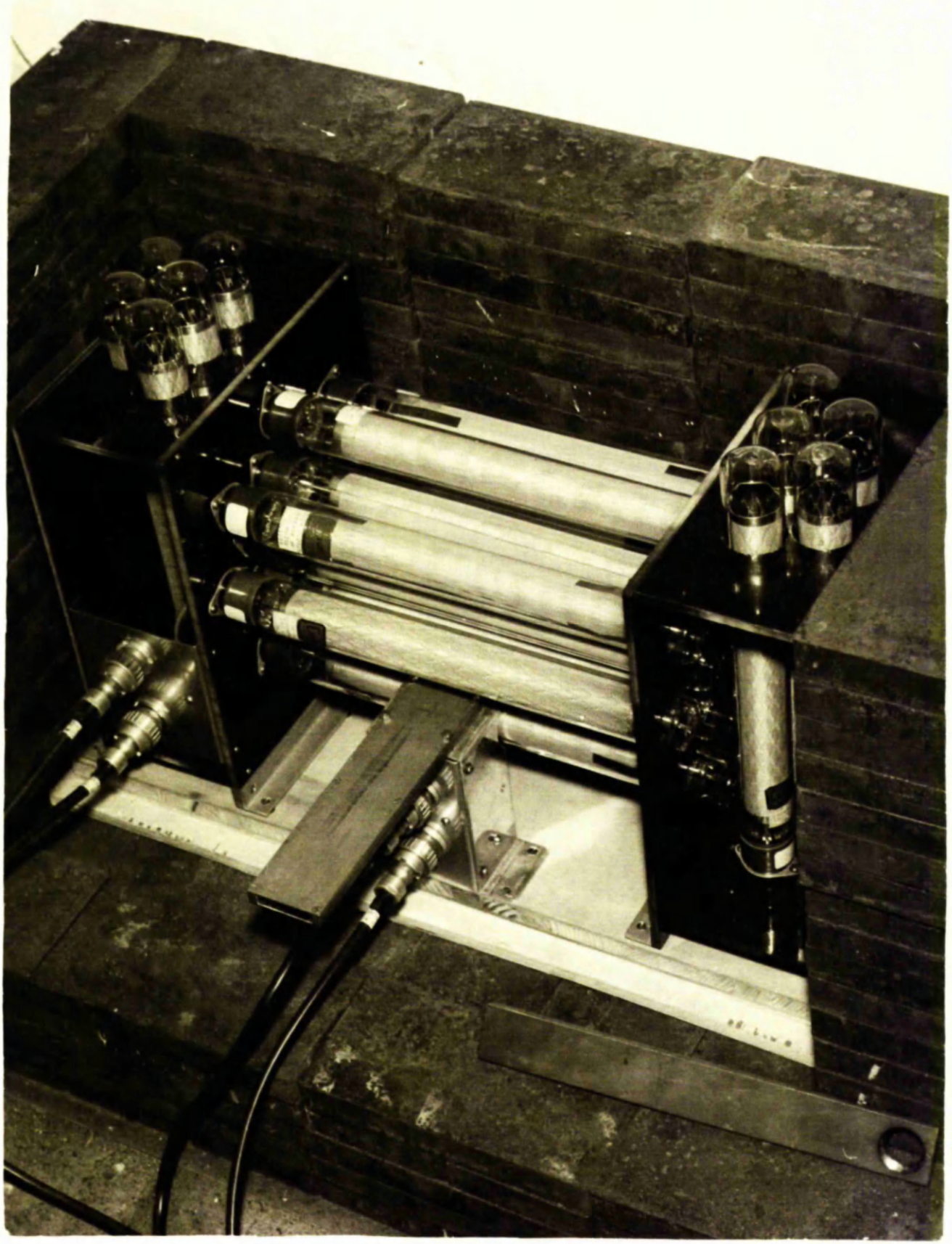


Figure 5. Anticoincidence low background counter.

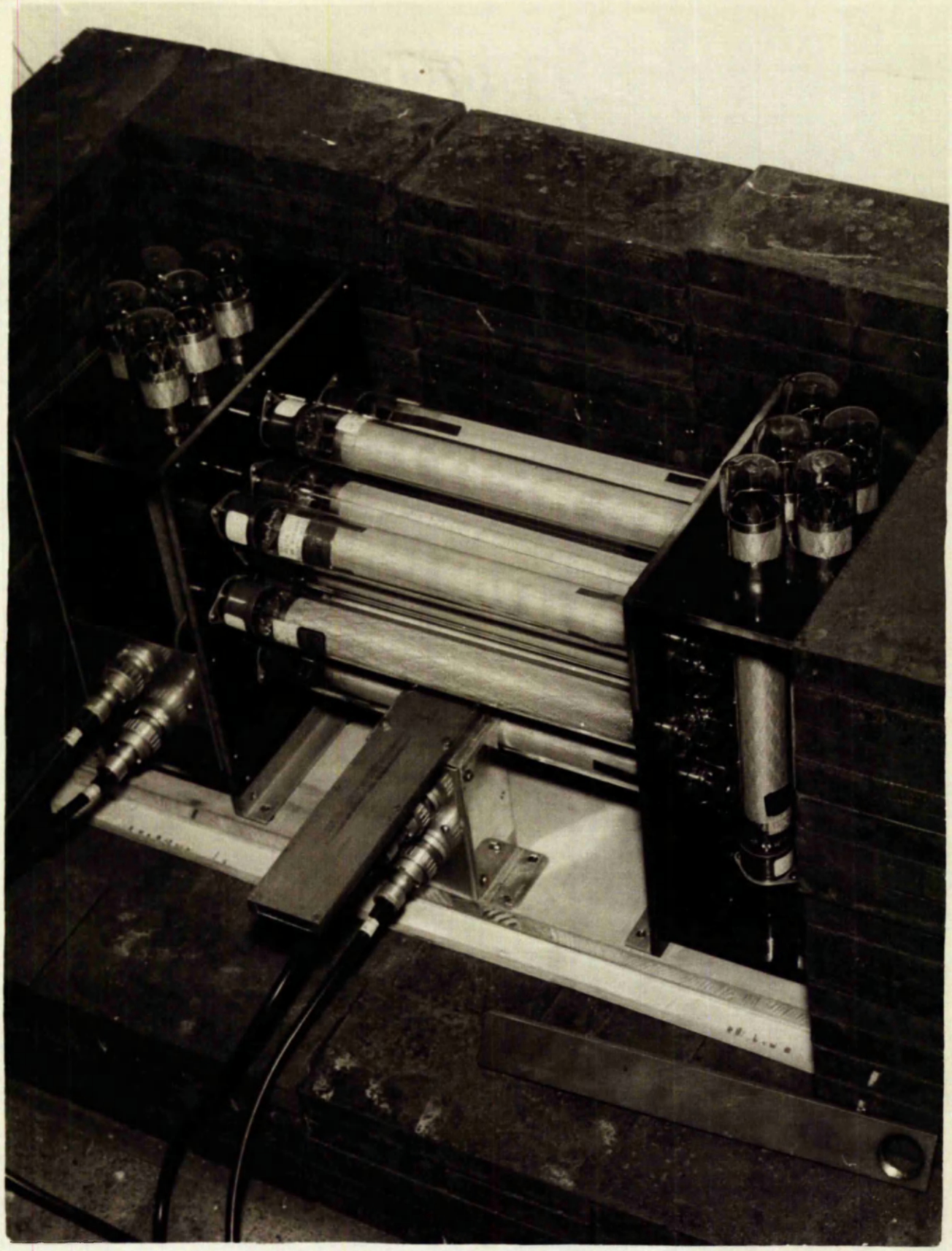


Figure 5. Anticoincidence low background counter.

3-position paralysis time selector of 50, 300, and 800 microseconds. The background count with this lead castle is about 7 c.p.m. using the GM4LB tubes, and about 14 c.p.m. using the EW3H tubes.

To count samples of low activity it is desirable to reduce the background count to a value such that the sample count rate is about 2 or 3 times the background count rate. The background count of the GM4LB tubes can be reduced to about 3 - 4 c.p.m. using 6 inches of steel shielding in place of 1 inch of lead. Doubling the thickness of shielding does not halve the background, and one must resort to electronic shielding methods. In this case the procedure is to surround the GM4LB tube (the counting tube) with a ring of gamma tubes. These tubes are arranged with an anticoincidence circuit so that a cosmic ray ~~which~~ causes the pulse from one or more of the screening tubes, to eliminate the pulse from the counting tube. In the accompanying photograph the 26 screening tubes are shown in position. These are high efficiency gamma tubes made by 20th Century Electronics; type G.26Pb. They have a lead cathode, an effective length of 20 cm., and are organic quenched. The counting tube is inside the centre of the screen, and samples are mounted in the circular recess of the brass slide seen in the right foreground. This is introduced into the slot projecting into the middle of the screening tubes. When the slide is pushed in to its furthest extent the sample is exactly underneath the window of the counting tube.

A black cloth is placed over the screening tubes to exclude any light which may come through the cracks between the steel bricks. After the steel walls have been built up a $\frac{1}{4}$ inch thick steel plate of the required area is placed exactly on top of the walls. The roof plate is finally covered to a depth of 6 inches with steel bricks. The walls

and the floor are four inches thick. The whole assembly is built on top of a one inch thick wooden board, raised off the ground on wooden battens to allow air to circulate underneath. A $\frac{1}{4}$ inch thick 'Perspex' sheet of the same size is placed on top of the wooden board as a moisture seal, and a thin aluminium sheet is laid over the Perspex to prevent scratching by the steel bricks.

The total weight of the steel shielding is about $1\frac{3}{4}$ tons so that care has to be taken to avoid overloading the floor. A basement was chosen for the assembly, which was placed near an outside wall. Due to the slope of the ground outside, the floor at this point is about five feet underground.

Throughout 1959 the background of this counter was about 0.8 - 0.9 c.p.m. From about February 1960 to July 1960 the equipment was running continuously at a background of 0.63 - 0.69 c.p.m. The counting assembly and its electronics were devised and built by the technicians of the Physics Department of the Western Regional Hospitals Board, Glasgow. A similar counting assembly there incorporates three GM4LB tubes inside the same ring of screening tubes, and all three channels have shown this reduction in background over this period. This is partly due to the greatly reduced amount of gamma active fission products washed out of the atmosphere by rain. A full description of this three channel low background unit has been published by the designers D. Rowan and W. Stevenson (232).

Paralysis time.

After a pulse has been recorded in a counter tube the pulse height falls to zero due to the time taken for the positive ions of the filling gas to move sufficiently

far away from the central anode: this is the 'dead time'. Another interval elapses before the counter is capable of registering a further pulse: this is the 'recovery time'. The 'paralysis time' is the total interval which elapses after one pulse before another can be recorded: i.e., paralysis time equals dead time plus recovery time. The listed paralysis time of the EW3H tubes is about 700 microseconds, so the scaler is set for a paralysis time of 800 microseconds and the following correction is made where the count rate is sufficiently high to make the paralysis time error significant.

$$R_t = \frac{Ro}{1 - R_o.T},$$

where:- R_t and R_o are the true and observed count rates, respectively, in c.p. second; and T is the paralysis time in seconds.

In the case of the anticoincidence assembly all pulses are scored on a mechanical register, and the paralysis time of the counting tube is insignificant compared with the resolution time of this mechanism. Although the register can except pulses, at a regular rate, at 1,200 pulses per minute, when dealing with the random pulses from the G-M tube samples were chosen having count rates of less than 100 c.p.m., with the majority of samples counting less than 20 c.p.m. It is possible to ascertain the necessary correction for resolution time by counting several standard radioactive sources, the activities of which increase in multiples of the first sample, chosen with a low count rate so that no resolution time correction need be made.

Self-absorption.

All radioactive samples are counted in the form of solid sources: Sr-89 and Sr-90 as carbonates, and Y-90

as the oxalate. When the source becomes too thick β particles from the bottom layers will not be able to penetrate through the source material and into the counter tube. The magnitude of this absorption effect will depend upon the maximum β energy of the particles, as well as on the weight of the sample.

Geometry.

The dimensions of the counting tube assembly have a bearing on the counting tube efficiency since the count rate of a sample will be reduced if the sample to tube distance is increased. This means that the efficiency of a particular tube might appear to change if it is set up in a different lead castle; this being due to the change in geometry.

Statistics.

Radioactive decay is a random process, so that there is no 'absolute' count rate, but only a 'mean' count rate can be described. Statistics can be used in the estimation of errors due to this randomness. A theoretical discussion of the statistics of counting can be obtained from standard text books, such as that by Faires and Parks (233); only the conclusions are given here.

The 'standard deviation' of an observed count is equal to the square root of the mean count, and is such that 32.7% of a large number of observations will have deviations greater than the standard deviation. The 'true' count is therefore $x \pm x^{\frac{1}{2}}$, for a total count of $\underline{x}$. If x counts are obtained in $\underline{a}$ minutes, then the standard deviation of the count rate x/a is $x^{\frac{1}{2}}/a$. Where the background count is

y counts in b minutes the standard deviation of the net sample count rate is

$$(x/a^2 + y/b^2)^{\frac{1}{2}}.$$

The sample count plus the background count which are recorded together can be described as the 'gross' count. The sample count, from which the background count has been subtracted, is termed the 'net' count.

From this formula it will be seen that in order to reduce the standard deviation as much as possible the ratio of the sample counting time to the background counting time should be equal to the ratio of the square roots of the respective count rates. In practice this is judged only roughly.

CALIBRATION OF COUNTERS.

When a radioactive sample is counted in one of the assemblies described in the last section the resulting measurement is calculated in terms of a count rate; generally counts per minute (c.p.m.). For a given sample the magnitude of this count rate is dependent upon the type of G-M tube used, and the geometry of the counting assembly. In addition, G-M tubes of any one kind may differ in counting characteristics from each other. To know the amount of radioactivity present in absolute units (e.g. in micromicrocuries) it is necessary to know the counting efficiency of the tube for that particular nuclide. Since the efficiency of a G-M tube depends upon the dimensions of the counting assembly ('geometry'); the radionuclide being counted (β energy); and the weight of the precipitate containing the nuclide ('self-absorption'); all these variables must be specified with the efficiency of the tube.

Calibrated Sr-90/Y-90, and Sr-89 solutions are obtained from the Radiochemical Centre, Amersham. The solutions are packed in sealed glass ampoules or in polythene screw-top bottles. With glass ampoules, the radioactive content is stated for the total contents so that the solution is transferred to a 100 ml. volumetric flask containing strontium and yttrium carriers, and made up to the mark. This stock solution is further diluted in the same way to give a convenient level of activity; generally of the order of about 1,000 d.p.m. of Sr-90 per 5 ml. aliquot. When the calibrated solutions are delivered in polythene bottles, the activity is quoted per millilitre; and a suitable aliquot is removed as required.

Using the diluted reference solutions the efficiency calibrations are carried out in a similar manner to the radiochemical procedures already described in the section on methods of analysis (p. 58), and in the Appendices.

Y-90 calibration.

Aliquots of the Sr-90/Y-90 calibrated solution are added to centrifuge tubes containing yttrium carrier, and the volumes diluted to about 40 ml. Two yttrium hydroxide precipitations are carried out to remove the Sr-90; finally the yttrium is precipitated as oxalate and counted immediately. The procedure is described in Appendix 1, beginning from paragraph (6) of the DETERMINATION. From the decay graph the zero time count rate is calculated and then corrected for yttrium recovery, taking into account the small quantity of yttrium carrier in the reference solution itself. The recovery of yttrium carrier is invariably greater than 80% so that there is no variation in Y-90 efficiency due to self-absorption over this narrow range of sample weight.

Sr-90 calibration.

Aliquots are added to 100 ml. beakers containing varying amounts of strontium carrier from 10 to 50 mgm., and each containing about 5 mgm. of yttrium and iron carriers. A hydroxide scavenge is carried out, the Sr-90 precipitated as the carbonate and counted immediately. The method is described in detail in Appendix 1, beginning from the middle of paragraph (3) of the DETERMINATION. The amounts of strontium carrier used cover the range of expected strontium recovery from samples where 50 mgm. of

strontium carrier have been added at the beginning of the chemical separation. A graph of strontium carbonate precipitate weight against strontium-90 efficiency is drawn for each G-M tube used, so that the efficiency for a given precipitate weight is easily read off.

Sr-89 calibrations.

Aliquots of the Sr-89 reference solution are added to varying amounts of strontium carrier, and the strontium precipitated as the carbonate and counted. The activity of the reference solution is corrected for radio-active decay. With a precipitate weight of less than about 70 mgm. of strontium carbonate there was no loss in efficiency due to self-absorption. In practice, samples have less than half this precipitate weight so that the Sr-89 efficiency is taken to be independent of the weight of precipitate.

Paralysis time corrections.

The low background anti-coincidence counter with its mechanical register is not suitable (nor intended) for counting samples giving several hundred c.p.m. To find the resolution error and to correct high counting samples for the losses in the register a correction graph was constructed.

Using 25 mgm. of strontium carrier for all samples, a series of Sr-89 standard samples were prepared, the activity of the first sample being 30 - 40 c.p.m. Other activities were multiples of this value. A count rate of 200 c.p.m. did not show any appreciable loss due to the recovery time of the register. An observed count rate of 800 c.p.m. was obtained from a sample with a 'true' count rate of 1460 c.p.m.

This counter was generally used for samples with a count rate of less than 100 c.p.m., when no paralysis time correction was necessary.

Samples with a count rate of over 100 c.p.m. were counted on the Panax counter with the automatic timer, and using an EW3H counting tube. The paralysis setting of 800 microseconds was used. Few samples gave count rates greater than 300 c.p.m., when no dead time correction was made.

A value for the inherent paralysis time of one particular tube was found by counting a 10,000 c.p.m. sample with the paralysis time setting at 50 then 800 microseconds. Based on a paralysis time of 800 microseconds the true count rate is calculated. Set at 50 microseconds it is assumed that only the paralysis time of the tube is operative. This was repeated using a sample giving 1,700 c.p.m. For the tube studied the two values obtained were 580 and 590 microseconds.

Part III.

RESULTS and CONCLUSIONS.

TABLE 1.

ANALYSIS OF RAINWATER COLLECTED AT MONACHYLE.

Sample number	Date	Collection time (days)	Rainfall (cms.) *	Sr-90 activity (μμc/l.)	Sr-89/ Sr-90 ratio **	Total Sr-90 fallout (mc/km^2)
START	27.1.59					
1.	8.3.59	40	12.0	13.6	24.3	1.63
2.	25.3.59	17	6.9	17.1	14.9	1.18
3.	17.4.59	23	12.7	18.9	16.1	2.40
4.	27.5.59	40	9.8	20.3	9.5	1.99
5.	16.6.59	20	7.7	13.5	6.1	1.04
6.	15.7.59	29	7.7	8.2	5.2	0.63
7.	18.8.59	34	14.0	4.1	2.5	0.57
8.	8.10.59	51	7.2	2.8	1.0	0.20
9.	18.11.59	41	35.0	1.5	0.7	0.53
10.	14.12.59	26	26.2	1.2	-	0.31
11.	18.1.60	35	27.0	1.5	-	0.40
12.	29.2.60	42	27.4	2.0	2.8†	0.55
13.	4.4.60	35	15.9	1.8	0.9	0.29
14.	29.4.60	25	16.8	3.3	-	0.56
15.	3.6.60	35	11.2	3.8	-	0.42

* Air Ministry Metoerological Office rainfall figures for the Balquhidder (Tulloch Lodge) station.

** The ratio is calculated to mid-way through the collection period, except for † which is calculated to the date of collection.

RAIN.

All the data from the rain analyses are given opposite in Table 1. The total rainfall (in cm.) is plotted with the Sr-90 activity of the rain (in μμc/l.) in histogram form, and shown in Figure 6. The width of each column represents the time of each sampling period superimposed on a monthly scale. It is apparent from Figure 6 that there was no direct correlation between total rainfall and the Sr-90 concentration in the rain.

When the average rate of rainfall (in mm./day) is plotted with the average rate of Sr-90 fallout (in μc/sq. km./day) for each sampling period, some correlation is seen: Figure 7. In this case a much more pronounced peak in fallout occurs in April 1959, and a much smaller peak in 1960, both of which are accompanied by increased rates of rainfall compared with the periods before and after. This seasonal variation in Sr-90 fallout has been found in rain collected at Milford Haven over the past several years, and Figure 8 is taken from a published graph (14) to show the spring peaks in the Sr-90 concentration in rain.

Although the Sr-90 concentration in rain at Monachyle during April and May 1960 was about three times the concentration during December 1959, this is partly due to the heavy rainfall during the winter months, because the rate of fallout was nearly constant from the middle of October 1959 until sampling stopped at the beginning of June 1960. This effect is seen in Figure 7. The fact that there is no direct correlation between the Sr-90 concentration in rain, and rainfall, is not surprising, and one would expect any relationship to be an inverse one. At a place with a high

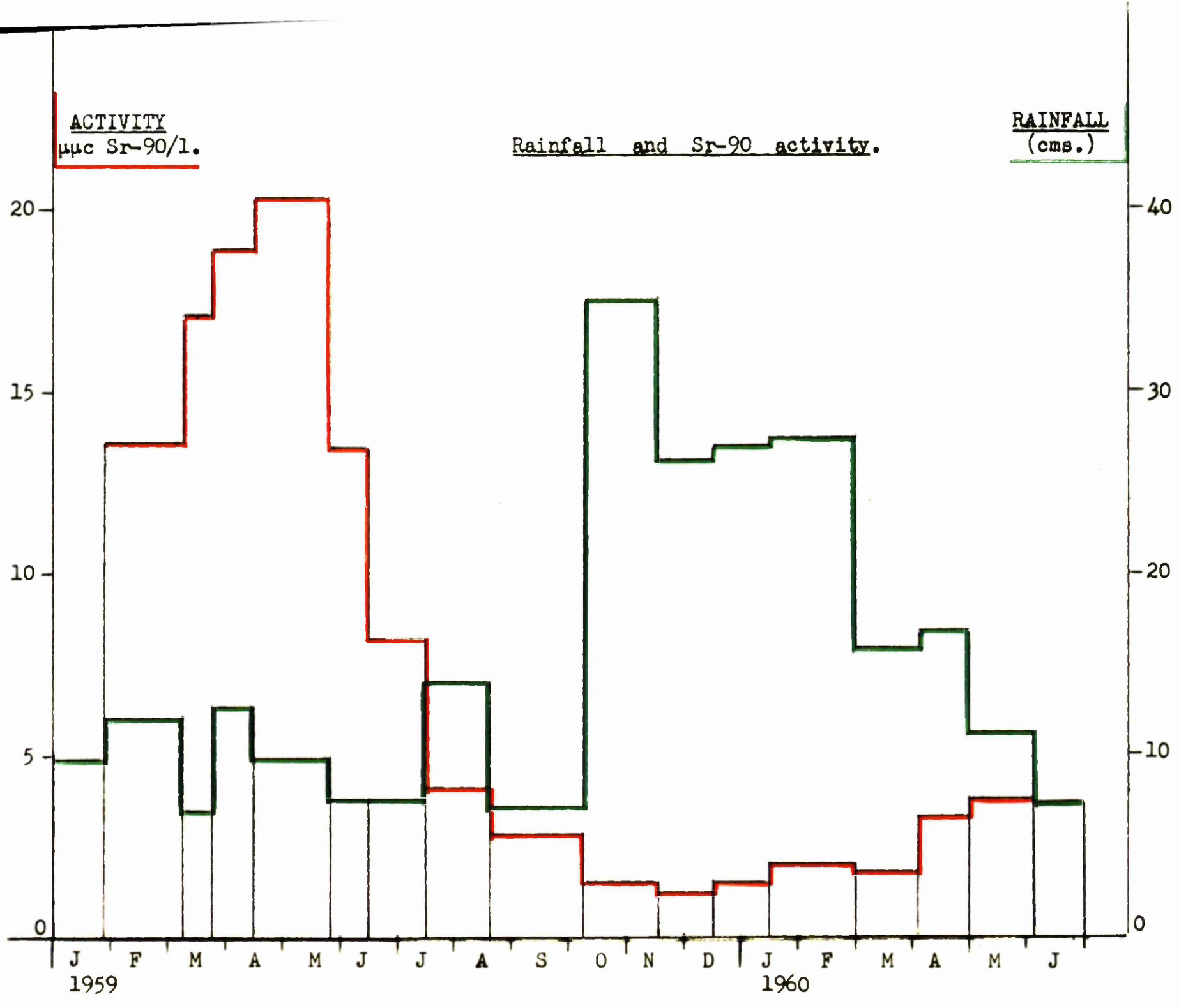
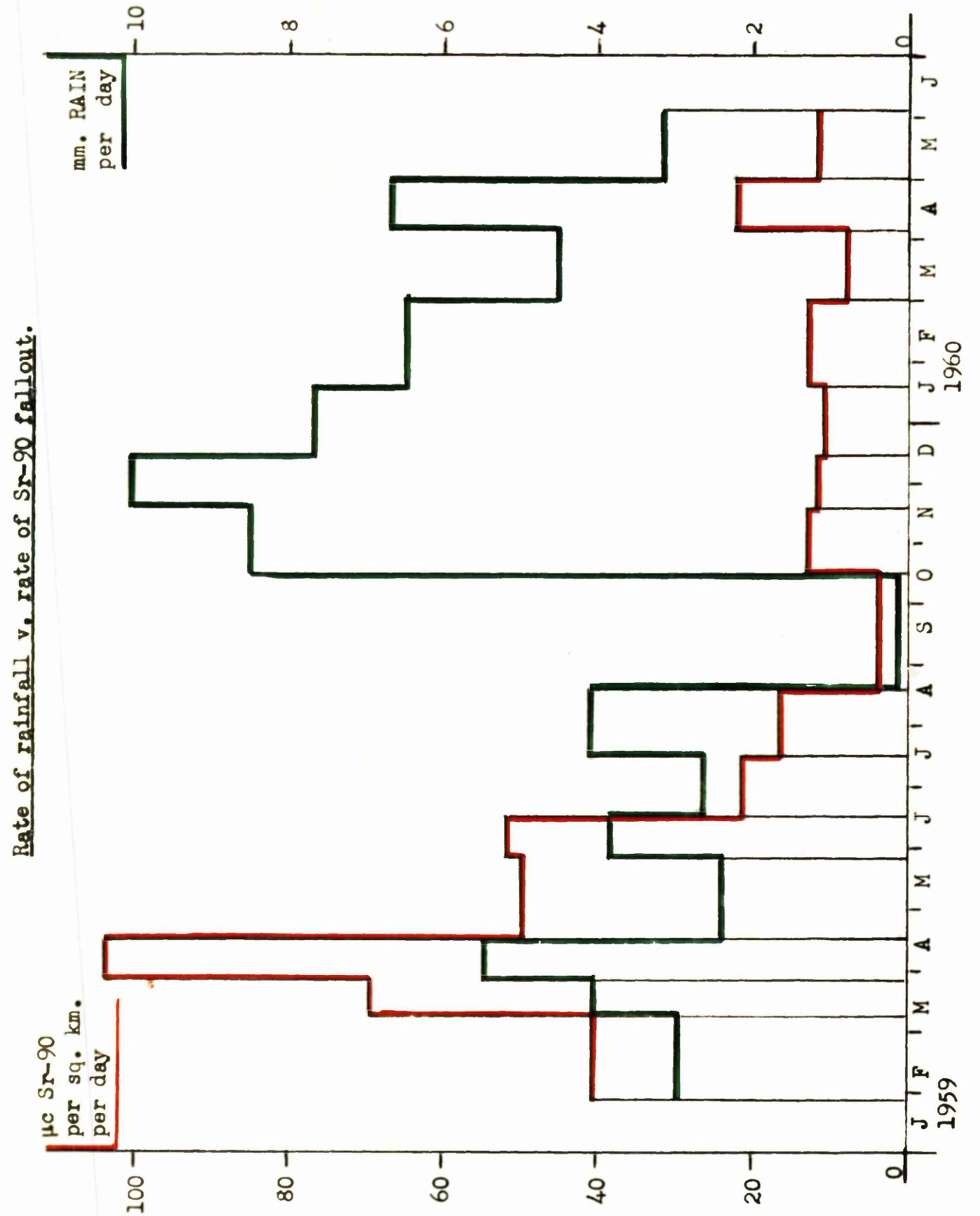

ACTIVITY
µµc Sr-90/l.
Rainfall and Sr-90 activity.
RAINFALL
(cms.)
20
15
10
5
0
40
30
20
10
0
J F M A M J J A S O N D J F M A M J
1959
1960

Figure 6.

Figure 7.


Rate of rainfall v. rate of Sr-90 fallout.
μc Sr-90 per sq. km. per day
mm. RAIN per day
100
80
60
40
20
0
10
8
6
4
2
0
J F M A M J J A S O N D J F M A M J
1959
1960

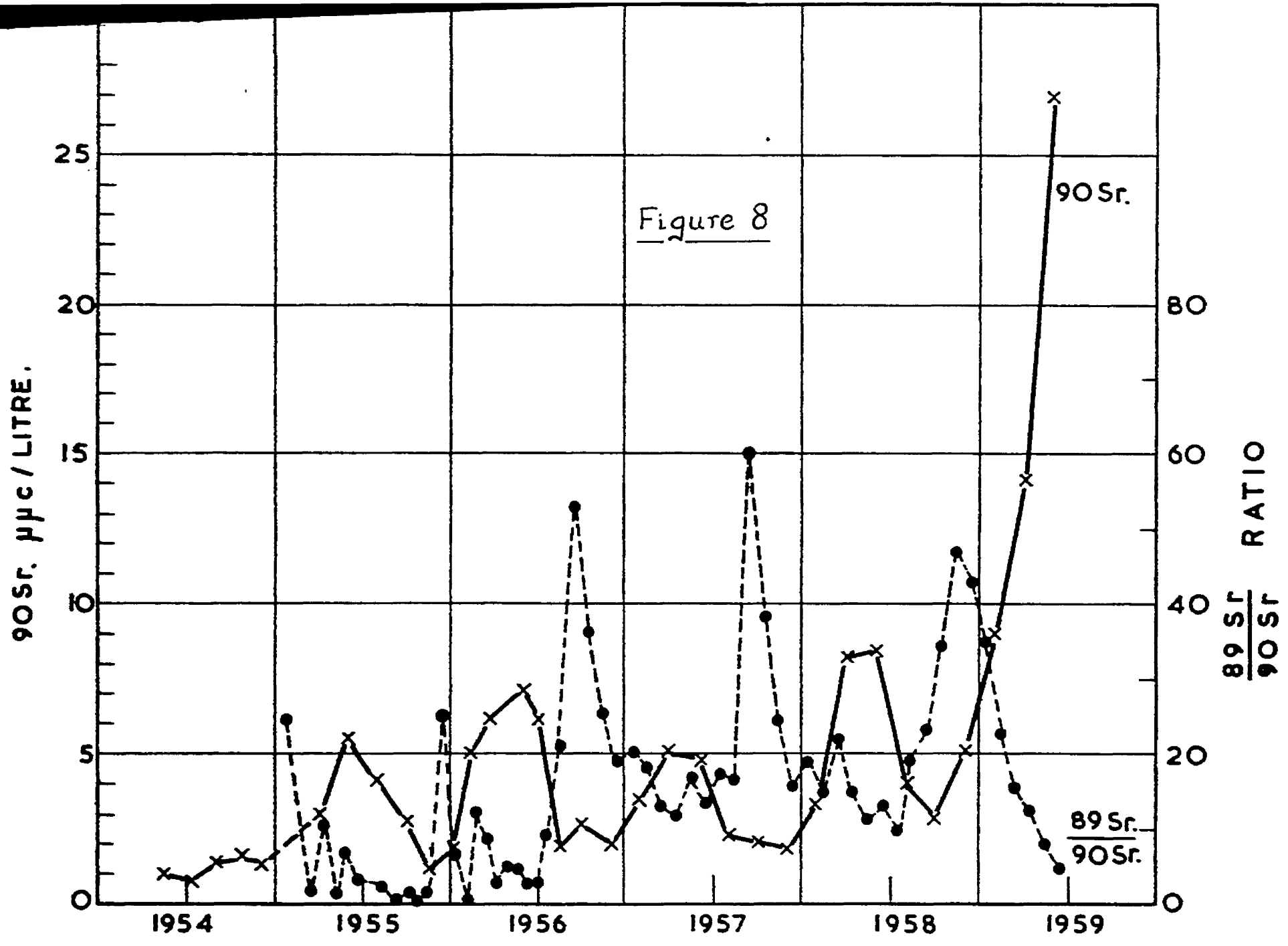


FIG. 2. 89 Sr AND 90 Sr CONTENT OF RAINWATER AT MILFORD HAVEN
90Sr PLOTTED AS 2 MONTHLY MEANS, 89 Sr / 90Sr AS MONTHLY FIGURES.

rainfall the first shower will wash out all the fine particulate debris present in that part of the atmosphere, and further rain will only dilute the activity thus washed out (12). Since fallout seems to be almost entirely due to the scavenging action of rain, the more often it rains the more fallout will be deposited. This effect can also be seen to some extent in Figure 7.

From Figures 6 and 7 the following conclusions can be drawn. (1). The Sr-90 concentration in rain reached a peak in April/May 1959, and this concentration was 5 - 6 times the concentration during the corresponding period in 1960. (2). The rate of Sr-90 fallout was to some extent directly proportional to the rate of rainfall. (3). The peak rate of fallout in April 1959 was 4 - 5 times the fallout rate in 1960.

Figure 9 shows, in histogram form, the total fallout (in mc./sq. km.) during each sampling period, and these periodic figures are successively added together giving (the red line) the cumulative deposition since 27.1.59. The initial steep increase in deposition diminishes about July 1959, and from then on the slope is roughly linear with a gradient of 0.34 mc./sq. km./month.

The decrease with time of the Sr-89/Sr-90 ratio in rain is shown in Figure 10. The slight increase of the 3rd point over the 2nd may be due to experimental error. If it is a real increase the reason could be incomplete mixing of old and new fission products in the stratosphere, but the theoretical Sr-89 decay curve from the first point closely follows the line of points shown on the graph. On 13.2.60 the first French atomic bomb was tested in the Sahara, and this date is shown by the vertical arrow in Figure 10. The following sample of rain collected on

Figure 9.

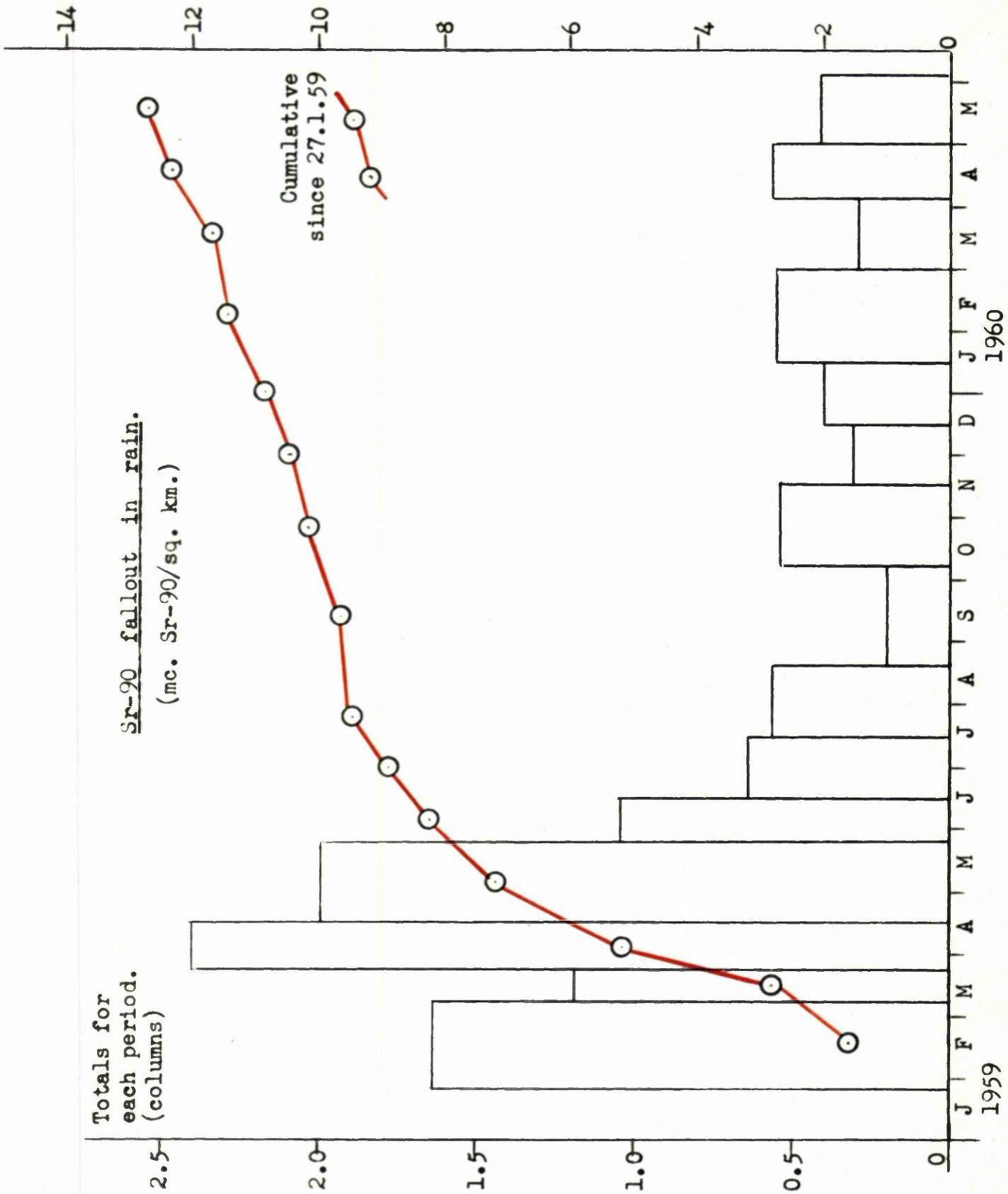
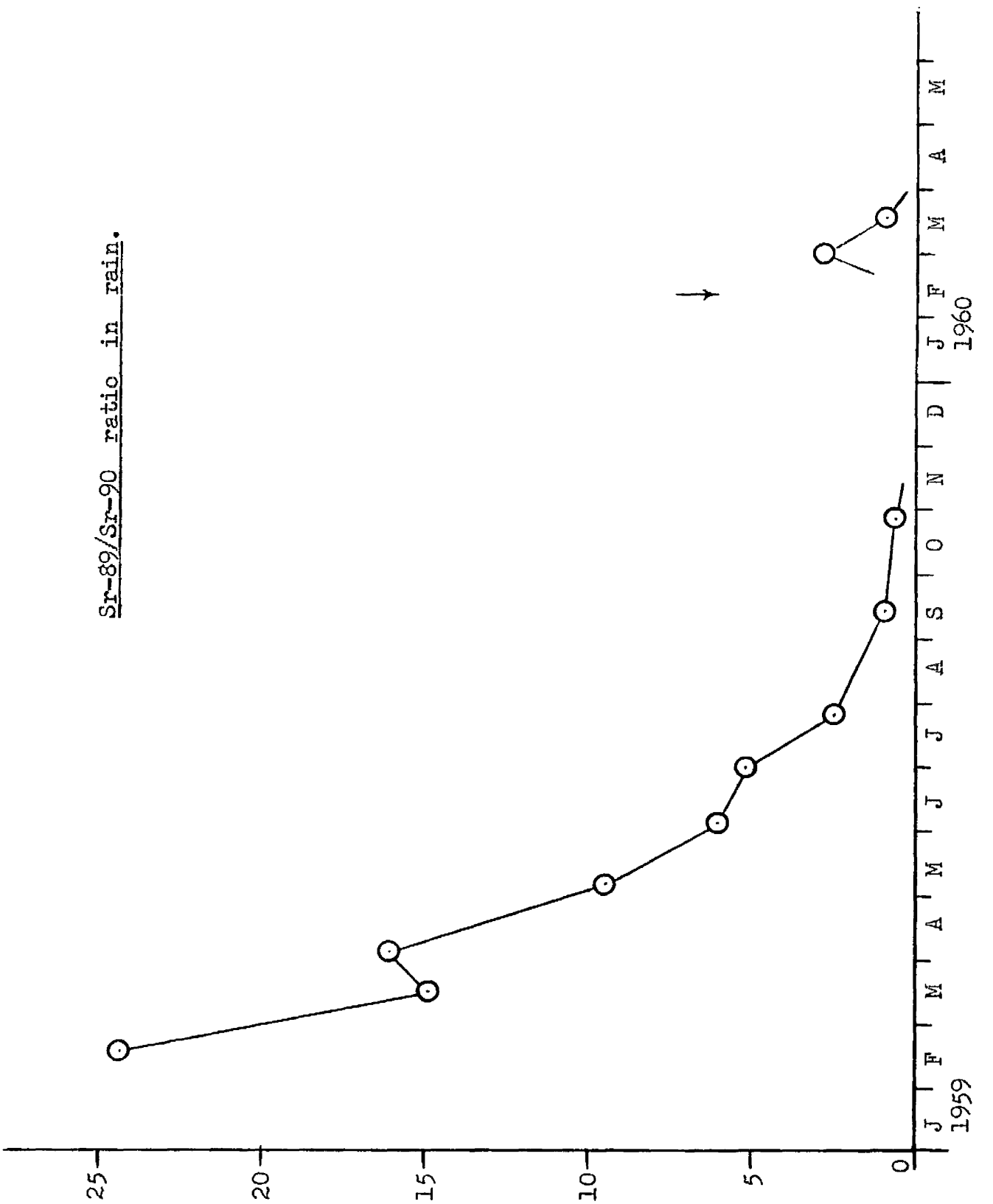

Sr-90 fallout in rain.
(mc. Sr-90/sq. km.)
Totals for each period. (columns)
Cumulative since 27.1.59
2.5
2.0
1.5
1.0
0.5
0
14
12
10
8
6
4
2
0
J F M A M J J A S O N D J F M A M
1959
1960

Figure 10.


Sr-89/Sr-90 ratio in rain.
25
20
15
10
5
0
J
F
M
A
M
J
J
A
S
O
N
D
J
F
M
A
M
1959
1960

29.2.60 showed a measureable amount of Sr-89, the activity of which is calculated to the date of collection and not to the middle of the collection period as in the case of the other samples. Measurements of airborne fission products (239) first showed a pronounced increase in recently produced fission products between February 28th and March 1st, so that the rain containing the Sr-89 must have fallen only a short time before the rain sample was collected. A trace of Sr-89 was found in the rain sample collected a month later. *

It is clear from the rainfall activity graphs that the storage time in the stratosphere is quite short, with a retention half-time of less than one year. Feely (18) has stated that more than half the debris from the Soviet tests in the autumn of 1958 fell out within 6 months, and at least half of the debris injected into the stratosphere by the American and British tests during 1958 fell out within 12 months. This means that the remaining debris in the stratosphere will not increase the existing soil reservoir of Sr-90 by a great deal. It will be shown later that over 50% of the Sr-90 activity in the grass samples was by direct foliar absorption, so that in the future it can be expected that the Sr-90 activity of grass and other vegetation will level off, if no land cultivation is carried out, or testing be resumed.

\- - -

* Unfortunately at the time of writing figures for the deposition of Sr-89 and Sr-90 in rain at Milford Haven are available only to mid-1959, Figure 8 (14). The Monachyle results to June 1960 are in general agreement with the Milford Haven results of Crooks for that period (234).

TABLE 2.

ANALYSIS OF BURN WATER COLLECTED AT MONACHYLE.

Sample number	Date	Sr-90 activity (μμc/l.)	Sr-89/ Sr-90 ratio
1.	8.3.59	0.84	4.8
2.	25.3.59	0.85	5.8
3.	17.4.59	0.67	5.6
4.	27.5.59	0.90	3.4
5.	16.6.59	sample lost	
6.	15.7.59	0.73	2.0
7.	18.8.59	0.94	0.9
8.	8.10.59	0.26	0.9
9.	18.11.59	0.59	0.2
10.	14.12.59	0.68	-
11.	18.1.60	0.63	-
12.	29.2.60	0.61	0.8
13.	4.4.60	0.94	-
14.	29.4.60	0.67	-
15.	3.6.60	0.69	-

Sample 15 contained 3.0 parts per million of calcium, so that the Sr-90 activity of that water sample was equivalent to 230 μμc of Sr-90/gm. of calcium.

BURN WATER.

The complete results are given opposite in Table 2. The variation in activity through 1959 to June 1960 is shown in Figure 11, and the decrease in the Sr-89/Sr-90 ratio in Figure 12. There was no large or seasonal change in the Sr-90 concentration, which remained practically constant throughout. It is interesting to note that the one low value occurred in a sample taken in a very dry spell of weather; in fact only 4.3 cm. of rain had fallen during the preceding 44 days. The Sr-89/Sr-90 ratio falls off in a similar manner to the ratio in rain, and also shows the appearance of a slight trace of Sr-89 from the Sahara test in the sample collected on29.2.60. A comparison of the Sr-89/Sr-90 ratios in rain water and burn water shows that for the first 6 or 7 samples collected the ratio in rain was about 2 or 3 times the ratio in the burn water.

There are three facts about the results which require an explanation. (1). The almost constant Sr-90 concentration of the water when the rain activity was fluctuating rapidly. (2). The one abnormally low concentration after a long period of dry weather. (3). The difference in the Sr-89/Sr-90 ratios in rain and burn water.

Radiostrontium can appear in the burn water by any of three routes. (i). By falling in the rain directly into the burn; this will be of little significance. (ii). By direct runoff before the radiostrontium in the rain has had time to be adsorbed or exchanged by the root mat or soil. (iii). By leaching from the root mat and soil by successive rainfalls. Routes (i) and (ii) are direct ones, whereas (iii) can be a long-term effect.

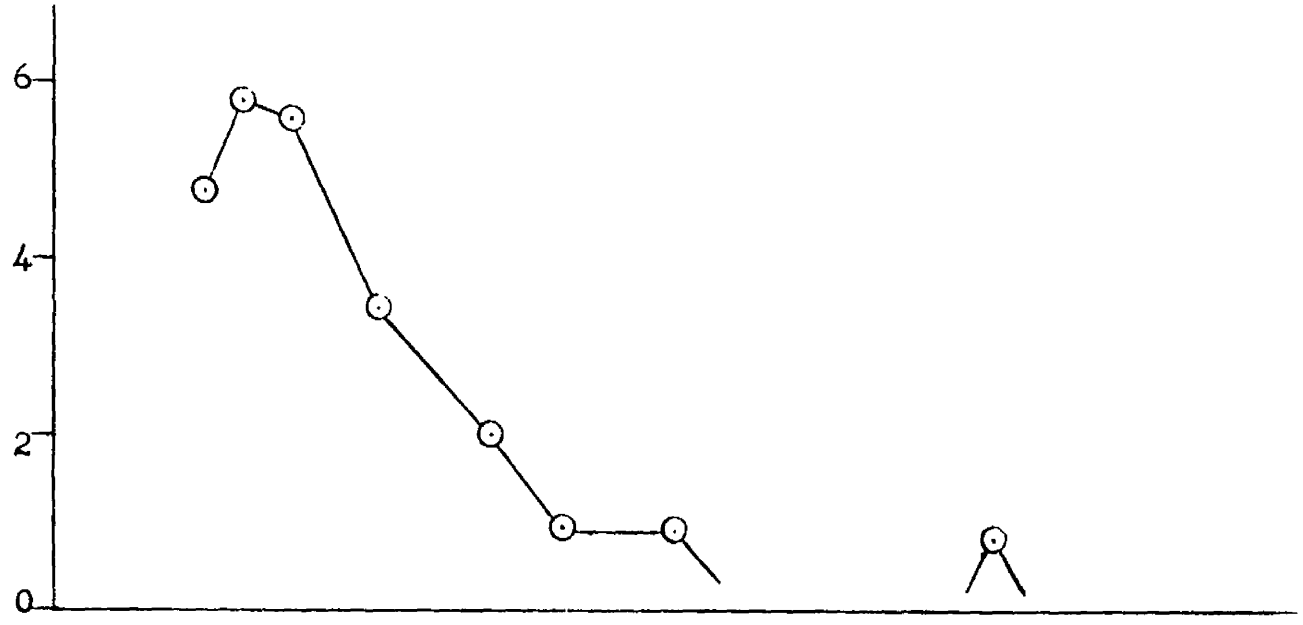
Figure 11.

Sr-90 activity of burn water.

(μμc Sr-90/l.)

Figure 12.

Sr-89/Sr-90 ratio in burn water.



The soil of the hills at Monachyle retains enough water to ensure that the Monachyle Burn does not dry up after a long spell of dry weather, but this water will reach equilibrium with the radiostrontium adsorbed by the soil as it slowly percolates down the hillside. Since none of the burn water after a dry spell will be from direct runoff the activity will be low due to this adsorption effect, and the longer contact time with the soil. The Sr-89/Sr-90 ratio for the low activity sample (No. 8 - 8.10.59) was 0.9, and since in the previous 13 days only 1.5 mm. of rain had fallen it must be assumed that this ratio of 0.9 represents the ratio leached from the soil alone. This burn water ratio will be higher than the ratio of the total Sr-89 deposition to the total Sr-90 deposition, as it is to be expected that earlier fallout (which will be predominately Sr-90 by October 1959) will have penetrated further into the ground than the recent fallout containing the Sr-89 produced by the late 1958 tests. The published figures for the total deposition of Sr-89 and Sr-90 at Milford Haven (14) show that at mid-May 1959 the Sr-89 deposition was 23 mc./sq. km., and the Sr-90 deposition was 17.3 mc./sq. km. The rate of Sr-89 fallout since the beginning of 1959 was far less than the rate of decay of the Sr-89 on the ground, so that the Sr-89 concentration in the soil will rapidly decline.

The almost constant Sr-90 concentration in the burn water is therefore perhaps due to a constant low level of leaching from the soil and root mat, with a variable contribution from direct runoff. It is impossible to estimate the Sr-89/Sr-90 leached from the soil to give a reliable picture of the retention time of fallout in the soil.

TABLE 3.

ANALYSIS OF MILK FROM MONACHYLE.

Sample number	Date	Volume in litres	Ash weight (gms.)	Percent calcium in ash	Sr-90 activity	
					µµc/l.	S.U.
1.	18.8.59	2.80	20.1	13.13	29.4	34.7
2.	8.10.59	2.48	17.5	13.34	20.2	25.7
3.	18.11.59	2.46	17.7	14.64	20.0	23.2
4.	14.12.59	2.46	18.7	14.82	16.1	14.3
5.	18.1.60	2.46	16.0	16.56	24.7	22.9
6.	29.2.60	2.70	17.5	16.86	40.0	36.7
7.	4.4.60	2.66	16.2	17.62	31.4	29.3
8.	29.4.60	2.72	19.2	14.97	28.2	26.6
9.	3.6.60	2.50	16.5	16.20	21.5	20.1

Sr-89/Sr-90 ratios:- Sample 1, 1.5

Sample 2, 0.7 approx.

MONACHYLE MILK.

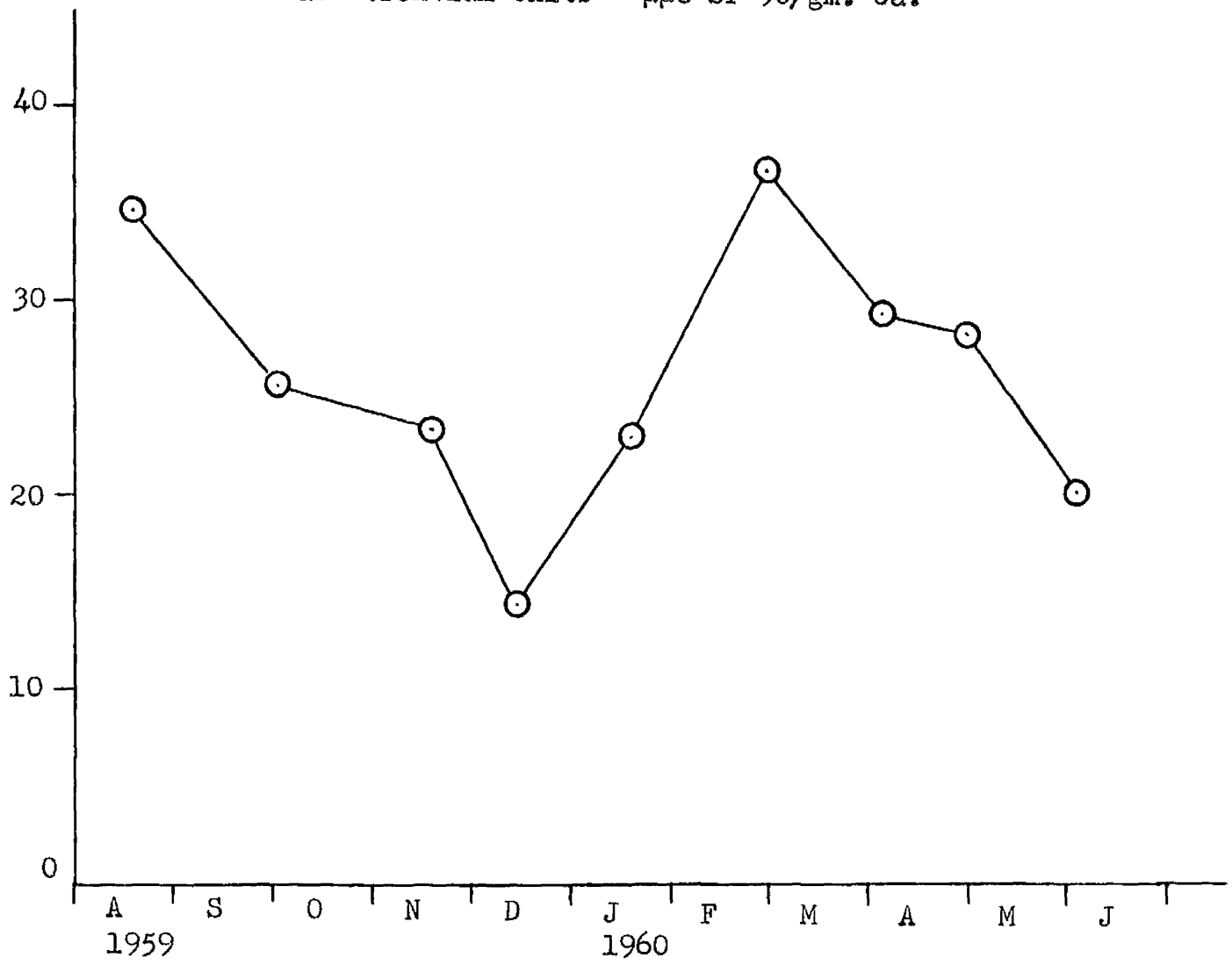
The results are given in Table 3 opposite, and the seasonal variation in Sr-90 content is plotted in Figure 13. The fall in activity to the minimum value in December 1959 corresponds to the winter foodstuffs, which due to the mineral calcium content will have much less Sr-90 activity than grass, when expressed on a calcium basis. The highest value obtained (36.7 S.U.) was on 29th February, and the activity became progressively less. Since the highest value in 1959 (34.7 S.U.) was the first sample collected (18th August), it is possible that before milk sampling was begun the activity had been greater than 40 S.U.

Figures are available in the ARCRL Reports Nos. 2 and 3 (135,236) for the Sr-90 content of milk sampled in Dunbartonshire and Argyllshire. These would be expected to show the same seasonal trends as the results of the Monachyle milk samples. The Sr-90 level in Dunbartonshire milk increased steadily during 1959, reaching a maximum value of 42.0 S.U. in July. The activity for the period 15th August - 15th September was only 15.2 S.U. The Report ARCRL 3 gives results of analyses of milk collected in Argyllshire. These results also show a peak level in Sr-90 activity in July 1959 of 37.2 S.U. Between 15th August and 15th September the activity had fallen to 23.0 S.U. The first Monachyle milk sample was collected on 18th August, and it contained 34.7 S.U. A sample of pasture grass taken on the same day at Monachyle contained 690 S.U. (MG3,Table 4), and a similar grass sample collected earlier on 27th May contained 1,040 S.U. (MG1) All this evidence points to the conclusion that the activity of

Figure 13.

Sr-90 in milk.

as Strontium Units - μμc Sr-90/gm. Ca.



milk at Monachyle, during July 1959, was greater than the activity of the first sample (34.7 S.U.) taken on 18th August.

It has been mentioned (p. 29) that a cow's diet contains between 7 and 12 times the ratio of Sr-90/calcium that appears in the milk. The activity of the diet giving 34.7 S.U. milk would be about 350 S.U., which is about half the value (690 S.U.) in the corresponding grass sample. This indicates that the samples of pasture grass collected were not representative of the cow's feed. If the cows were feeding on this pasture grass, having an activity of over 700 S.U. in 1960, then the milk activity would be about 70 S.U. It has also been pointed out (p. 33) that animals (including human beings) will have a Sr-90/calcium ratio in bone equal to about one quarter of the ratio in the diet. This fraction will be nearer one half for young people on a mainly milk diet. The significance of this is that if infants are fed mainly on milk, from cows grazing on pasture having an activity of 700 S.U., the Sr-90 level in these children's bones will approach 30 S.U. It will be remembered (p. 33) that 10 S.U. is the level indicating the need for immediate consideration of the problem.

TABLE 4.

ANALYSIS OF GRASS FROM MONACHYLE.

Sample number	Date	Type	Percent calcium in ash	Sr-89/ Sr-90 ratio	Sr-90 activity	
					µµc/kg.	S.U.
BG1.	25.3.59	A	0.96	11.5	6,200	15,600
BG2.	17.4.59	C	5.09	8.3	14,200	2,900
MG1.	27.5.59	D	5.2	4.3	4,190	1,040
MG2.	27.5.59	B	6.1	5.7	17,800	5,320
BG2/A.	15.7.59	B	6.16	2.9	11,400	3,490
MG4.	18.8.59	B	6.5	-	4,950	1,430
MG3.	18.8.59	D	5.03	-	2,940	690
MG5.	29.4.60	C	4.82	-	3,070	724

GRASS FROM MONACHYLE.

The results of the Monachyle grass samples are given in Table 4, opposite. The 'Types' of grass (column 3) are as follows.

"A" was taken at a point about two kilometres upstream from the site of the rain collector. As the grass was taken at the end of March it comprised only old growth. The quality of the grass was rather poor as the ash contained only 1% of calcium, equivalent to 0.43 gm. of calcium per kilo of dry weight.

The type "B" samples were all cut from the same 4 feet square patch situated near the rain collector. This spot is shown in the photograph (Figure 4), in the bottom right-hand corner.

The two "C" samples were taken exactly one year apart, towards the end of April 1959 and 1960, so that in both cases they were the first growth of the new season. They are representative samples, being the combined croppings of grass cut at many points between the rain collector and Monachylemore farm.

Samples "D" were cut in the field at the loch side, the actual position on the detailed map (Figure 3) being roughly where the dotted path shown cuts the lower (red) arrow. The first sample from this spot (MG1) was new growth at the end of May 1959, and the second sample (MG3) from there was the secondary growth after the first crop of hay had been cut from this field.

Samples of types "B", "C", and "D" all had similar calcium levels in the ash, with an average value of about 5.6%: the average calcium content of the grass samples when

expressed on a dry weight basis was about 4 gm. per kilo. The sample BG1 (type "A") had by far the highest level of Sr-90 when expressed on a calcium basis (15,600 S.U.), but only an average level taken on a dry weight basis (6,200 μμc/kg. dry grass).

As it was to be expected, later croppings of grass taken late in 1959 and early 1960 contained less Sr-90 than samples taken during the first half of 1959. This is due to the reduced amount of direct foliar uptake from rain, the activity of which was falling off rapidly during August/September 1959. In the three months between first and last samples of type "B" grass the activity fell four-fold. This decrease is also shown with the "C" samples. After a lapse of one year the Sr-90 activity had been reduced to one quarter of its 1959 value. The activity of the pasture grass "D" fell during the 1959 summer.

A comparison of the Sr-89/Sr-90 ratios in grass and rain for the same periods will indicate that proportion of the radiostrontium in the grass which comes from foliar uptake. If it is first assumed that the Sr-89/Sr-90 ratio in the root mat and soil is very low, it is calculated for sample BG1 which, being dead grass, was exposed to many months of direct fallout without taking up any from the roots, that only about 10% of the Sr-90 in the top parts of the grass that were cut came from the root system. For the next four samples in Table 4 the root contribution of Sr-90 is between 20% and 40% of the total Sr-90 present. At the beginning of 1959 the amount of Sr-89 in the soil and root mat was probably greater than the Sr-90 content, due to the recent fallout from the 1958 test series. Any Sr-89 taken up by the roots will mean that more than this value of 20 - 40% of the activity was by root uptake, and hence these percentages should be

taken as minimum ones.

It is very interesting to compare the activities of two samples of grass collected on the same day (27th May 1959). These are a sample of pasture grass (MG1), and a grass sample from ground which has never been ploughed (MG2). This "untreated" grass had an activity of 4 - 5 times that of the pasture grass, both on a calcium and a dry weight basis. Also, the Sr-89/Sr-90 ratio in the pasture grass was lower (4.3) than the ratio in the untreated grass (5.7). These facts indicate that the fallout on the pasture land has been to a large extent incorporated into the soil, both by ploughing and by the absence of the root mat which is characteristic of the untreated grass. The activity on the root mat of the untreated land is much more available to grass than activity in the soil, and the root mat activity will contain a higher ratio of Sr-89/Sr-90 from recent fallout than the soil, which will contain Sr-90 from past tests.

Samples from these two places were again collected on the same day, later in the year (18th August, 1959), but at this time there was not sufficient Sr-89 remaining to be determined accurately. The difference in Sr-90 activity is still apparent, but is not so great as in the case of the previous two samples. This is due to the reduced Sr-90 activity in rain, and the fact that grass had been cropped throughout the year.

It does appear from the 1960 grass sample that the Sr-90 activity in grass would fall below the 700 S.U. level on account of the falling Sr-90 content of rain. The residual activity of the grass will come from the reservoir of Sr-90 in the root mat and the top soil layer, and since only a fraction of this activity is taken up by the grass each year, an almost constant Sr-90 level in grass will

result.

If one could be sure that bomb testing (especially the testing of megaton weapons) would never be resumed this would now be an ideal time to plough under the top layer of pasture land, if this has not been done for several years. If the calcium deficient land is limed at the same time there would be expected to result a large decrease in the Sr-90 content of the grass subsequently grown on such land. Since present levels of Sr-90 in grass resulting from root uptake only are not considered to be hazardous, this procedure is unlikely to be carried out on the hill pastures on the grounds of the expense involved.

RUSHES FROM MONACHYLE.

The results are given below in Table 5.

TABLE 5. ANALYSIS OF RUSHES FROM MONACHYLE.

Sample number	Date	Type	Percent calcium in ash	Sr-89/ Sr-90 ratio	Sr-90 activity µµc/kg.	Sr-90 activity S.U.
R1.	25.3.59	Dead	9.1	9.8	11,600	7,560
R2.	27.5.59	Dead	5.5	6.5	14,800	14,900
		New	4.0	4.2	1,660	1,300
R3.	18.8.59	Dead	9.0	-	8,020	5,300
		New	6.6	-	1,660	730

Sr-90 activity levels are within the same range as the grass samples, but the most striking feature is the difference between the Sr-90 levels in the old and new growth. The rushes were collected near the site of the rain collector; the first sample being taken so early in the year that no new growth had occurred, and high levels were found. By the time the next sample was taken new growth had taken place, so that the sample collected was hand-picked in the laboratory to separate the old from the new. On a calcium basis the old growth of sample R2 had 11 times the Sr-90 activity of the new growth, and 9 times the activity on a dry weight basis. Similar differences were found in the next sample taken, R3.

On a calculation based on the Sr-89/Sr-90 ratios in the rushes and rain, at least 24% of the Sr-90 activity in both dead samples R1 and R2 came through the roots; and 42% in the new growth of R2.

The large difference between the activities of the old and new growth is principally due to the difference in the times of exposure to direct fallout and foliar uptake. The difference in Sr-89/Sr-90 ratios between old and new would indicate that when new growth started there would be a substantial amount of old Sr-90 taken up by the roots compared with the amount taken up by direct contact from the rain. The sample of new growth cut on 27.5.59 had accumulated half its activity through root uptake, therefore it would seem that the sample cut on 18.8.59 (R3) would have far more than half its activity taken up through the roots, since the Sr-90 concentration in the rain was falling off.

When the rate of Sr-90 fallout in rain becomes negligible the Sr-90 to calcium ratio in rushes will level off to an almost constant value from root uptake alone. This value may be approximately the same as for grass growing under the same conditions. Judging by the last available sample of new rush (R3) this constant Sr-90 level might be less than 500 S.U.

MOSS FROM MONACHYLE.

TABLE 6. ANALYSIS OF MOSS.

Sample number	Date	Sr-90 activity µµc/kg.	S.U.
BM1.	25.3.59	11,100	1,700
BM2.	27.5.59	11,600	21,000
BM2/A.	15.7.59	8,780	21,900
BM3.	18.8.59	11,900	12,400

All samples were cut near the site of the rain collector. Sample BM1 comprised the whole of the moss plant, including the root system. This gave a much greater proportion of ash than the other three samples, which were tops only, although BM1 had a similar calcium content in the ash as the others (about 1%). It is this large ash content which is responsible for the much lower Sr-90 activity of BM1, when expressed as Strontium Units. The low calcium content of the moss gives very high levels of Sr-90 activity in terms of S.U. Should animals feed to any large extent on moss tips then high Sr-90 bone levels would result. The Sr-89/ Sr-90 ratio in the sample BM2/A (2.8) is the same as in the grass sample BG2/A, which was collected at the same spot, and at the same time, indicating that at least 30% of the Sr-90 in these two samples was taken up via the roots.

The four samples analysed had consistent levels of activity, expressed on a dry weight basis. This activity was the same as in the grass samples cut nearby ("B" samples), and also in the samples of dead rushes. This

similarity on a weight basis supposes that uptake was mainly foliar, since the moss values on a calcium basis were much higher. The high activity was also due partly to the position of the place where the moss was growing, which was at the foot of a slope. At this point the ground is always very soft due to water trickling down from the hills above, so that under these circumstances the moss would be in contact with a slightly higher concentration of Sr-90 than the grass or rushes growing in the open.

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SHEEP DROPPINGS FROM MONACHYLE.

TABLE 7. ANALYSIS OF SHEEP DROPPINGS.

Sample number	Date	Sr-90 activity µµc/kg.	S.U.
BLSD1.	11.4.58		3,190
MSD1.	27.5.59	14,000	2,840
MSD2.	29.2.60	6,880	1,820
MSD3.	29.4.60	10,800	1,670
MSD4.	3.6.60	11,800	1,880

The first sample, BLSD1, came from Ben Lawers, all the others from Monachyle. Sheep droppings were collected because it would be extremely difficult to obtain samples

of vegetation corresponding to the average diet of sheep grazing over a wide area. In this way, Sr-90 analyses of the droppings give a direct estimate of the Sr-90 level of the diet, provided that allowance is made for the differential absorption of strontium and calcium by the sheep. The correlation between the Sr-90/calcium ratio in the grass (or droppings) and in sheep bones is discussed in the next section. Main interest is focussed on the Sr-90 levels taken on a calcium basis, and the activity per kilo dry weight has less importance.

The three samples collected during 1960 have similar activity levels (about 1,800 S.U.) which are lower than the two other samples each collected at previous intervals of one year. The Sr-89/Sr-90 ratio for sample MSD1 is 6.2, which is slightly higher than the ratio of 5.7 found in hill pasture grass collected at the same time.

All samples were collected between the end of February and the beginning of June. If the droppings collected were from ewes which had lambed that spring there would be a calcium demand on the ewes to produce milk for their offspring. This would result in a slightly greater absorption of calcium than strontium from the intestine. In double tracer experiments on lactating goats (119) the average Observed Ratio (p. 26) for faeces-diet was1.12. Since the droppings collected were representative of the flock of ewes the Sr-90/calcium ratio in the diet must have been slightly less than the ratios quoted for the droppings. It is assumed that the droppings collected had not been lying for more than a day or two, so that any leaching by or absorption from rain is neglected.

SHEEP BONES.

TABLE 8. ANALYSIS OF SHEEP BONES.

Sample number	Location	Approximate date of death	Type of bone	S.U.
SB1.	Argyll	Winter 1957-8	Long bone	233
SB2.	Perth	"	"	63
SB3.	Argyll	Spring 1958	"	114
SB4.	Monachyle	"	Mandible	167
SB5.	Monachyle	Winter 1958-9	Long bone	132
			Teeth	94
			Mandible	183
SB6.	Monachyle	"	Long bone	131
SB7.	Perth	"	Long bone	108
			Teeth	92
			Mandible	130
SB8.	Argyll	"	Long bone	423
SB9.	Harris	"	"	177
SB10.	Argyll	Spring 1959	"	220
SB11.	Monachyle	"	Long bone	242
			Teeth	188
			Mandible	193
SB12.	Monachyle	Spring 1960	Long bone	111

The bones from Perth and Argyll were obtained from the area roughly between Ben Cruachan and Ben Lawers, both of which are shown on the map of Scotland. Sample SB9 was from the Isle of Harris. Long bones refer to femurs or tibias, or both together. Sample SB11 was a young sheep

of perhaps 9 months; the bones were small, and the skull bones partly separated. All the other samples were from sheep over one year old. There is no apparent annual trend in the Sr-90 activity of the bones analysed.

In the case of three samples (numbers 5, 7, and 11) several bones were taken from the same skeleton; namely long bones, and both the lower jaw bones from which the teeth were extracted and analysed separately. In all cases the teeth had the lowest Sr-90 activity per gram of calcium. For SB5 and SB7 the long bones had relatively less Sr-90 than the mandibles, but for SB11 this order was reversed.

The area from which the sample SB8 was taken would bear further investigation, since the Sr-90 activity (423 S.U.) is about twice the mean of the other bones.

The bone levels should be about one quarter of the Sr-90/calcium ratio in the diet, or droppings, of the animal. In the young there is a more rapid uptake, but with an older animal there is little exchange with the calcium in the bone, and new calcium in the diet. For the bones of sample SB11 from the Monachyle lamb, the activity of the long bones was 242 S.U., which is representative of the Sr-90/calcium level in the whole skeleton. This would result from a diet containing about 1,000 S.U. during 1958, for which figures are not available for the Sr-90 activity of the grass at Monachyle.

Grass samples for Dunbartonshire and Argyllshire taken in 1958, and reported in ARCRL Report No. 2 (135) showed a Sr-90 activity level of between 1,500 and 2,000 S.U. These were obtained from unimproved hill pasture similar to the Monachyle pasture. If during the winter months the sheep's diet is supplemented with low activity fodder such

as turnips, then the estimated average feed level of 1,000 S.U. may be approached.

Sheep droppings collected mid-1959 had an activity of 2,840 S.U., while early in 1960 this activity had fallen to 1,800 S.U. These figures correspond to approximate bone levels of 700 and 400 S.U., respectively, if this dietary activity level is maintained over a long period. This also shows, therefore, that the sheep's diet must be supplemented with low activity fodder.

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DEER ANTLERS AND DEER BONES.

TABLE 9.

Sample number	Location	Date of death	S.U.
DA1.	Islay	3.11.57	80.2
DA4.	"	27.10.58	76.5
DA5.	"	17.12.58	158
DB2.			139
DA6.	Monachyle	Oct. 1958	118
DA7.	Perthshire	Winter 1959-60	147
DB3.			58

The deer bone sample DB2 was the femur of the same animal from which the antlers DA5 were taken. Similarly

for the samples DA7 and DB3. The antler samples DA2 and DA3, are not included in the table of results. They represent deer which were shot before 1952, and no detectable quantities of Sr-90 were found.

The difference in activity between the leg bone and antler of DA5/DB2 is due to the age of the animal, which was only 2 or 3 years old. During its lifetime there was an almost uniform rate of increase of fallout, and due to the redistribution of bone calcium and Sr-90 in bone, found in young animals, there is not a great deal of difference between antler and bone activity. The difference is no more marked than the differences between types of sheep bone from the same animal.

The antler and bone from Perthshire were taken from a skeleton of a fully grown animal, probably over 8 years old. When the deer had reached its full stature, and the rate of turnover of bone calcium was low, the level of contamination of hill pasture increased considerably, after, say, 1957. Results show a wide variation between the activity of the skeleton (as typified by the leg bone) formed when the grass activity was low, and the antler, which would perhaps be grown within a year of its death, formed with calcium from highly contaminated pasture. Only the activity found in a sheep bone (SB2, Table 8, p. 87) from an animal which had died in the winter of 1957-8 showed the same low level as that deer bone.

SOIL FROM MONACHYLE.

TABLE 10.

Sample number & date	Depth range (in.)	Sr-90 activity mc/sq.km	Calcium extracted gm/sq.m.	Method of analysis	Appendix number
S1. 8.19.59	0 - 1"	26.6	39	$HClO_4/HNO_3$	4.
	1"- 3"	8.8	23	"	4.
S2. 18.11.59	0 - 1"	25.4	14.8	"	4.
	1"- 3"	3.9	14.8	"	4.
	3"-6"	2.6	31	"	4.
	6"-11"	6	76	"	4.
S3. 14.12.59	0 - 1"	37.8	6.8	"	4.
	1"- 3"	5.7	6.3	"	4.
	3"- 6"	4.8	79	"	4.
	3"- 6"	4.7	14	6N HCl	5.
S4. 29.2.60	0 - 4"	32.9	103	$HClO_4/HNO_3$	4.
		33.1	92	NaOH fusion	6.
		41	120	Na_2CO_3 fusion	6.
		36	83	21% HCl	4.
S5. 29.4.60	1"- 3"	3.3	4.7	NH_4.Acetate	7.

Samples 1, 2, 4, and 5 were taken with a 2 inches diameter soil corer. 1, and 2 were taken from a small area of ground, several cores being combined. For samples 4, and 5, twelve cores were taken between the site of the rain collector and the farm; the complete cores down to a depth of 4 inches were combined for sample S4, while for sample S5 as much of the root mat as possible was removed

and discarded, leaving the soil. Sample S3 was taken with a small shovel, only one pit, 22 cm. square was dug. The thick layer of turf and root mat was torn off in one piece, and this is the 0 - 1" layer of S3. Where the same soil sample was analysed for total Sr-90 by four different methods the highest value was obtained using the vigorous sodium carbonate fusion method; which also extracted the greatest amount of calcium.

Analysis of different soil horizons (samples 2, and 3) show that in the undisturbed ground 79% of the total activity in the top 6 inches lies in the top 1 inch of grass and root mat.

The ammonium acetate leach of sample 5 extracted about half the Sr-90 from the 1" - 3" level that was obtained from the same soil horizon of samples 1, 2, and 3. The amount of 'exchangeable' calcium found by this method (4.7 gm. per square metre) is equivalent to 0.65 milli-equivalents of calcium per 100 gm. of dry soil. The pH of a slurry of one part of soil to 2 parts of water was 4.2.

The soil samples 2 and 3 were carefully taken to the exact soil horizons listed. Although the total activity (to a depth of 6 inches) per unit area for sample 2 was only two thirds that of sample 3, the vertical distribution was proportionally the same in both cases. The relative ratios of the amounts of Sr-90 activity per unit area in the layers 0 - 1 inch (root mat and grass); 1 - 3 inches; and 3 - 6 inches; were respectively 100; 15; 11½.

It is of interest that the vertical distribution below the root mat was about constant for sample 2. In the five inches between 1 and 6 inches the activity was 6.5 mμc/sq. m., and in the next five inches downwards the activity was 6 mμc.

For sample 3 two different methods of analysis were used for the 3" - 6" layer, and both extracted the same amount of Sr-90. The $HClO_4/HNO_3$ method extracted far more calcium from the soil than did the 6N HCl leaching method.

Sample 4 was taken to compare the efficacy of the four different extraction processes, with the most common $HClO_4/HNO_3$ method being used for comparison purposes. The quantities of calcium extracted show more variation than do the Sr-90 figures, since the Sr-90 absorption will be mainly a surface effect. When the samples of soil are active enough to be able to use under 100 gm. of soil for each determination, the sodium carbonate fusion is the best method of extraction. Preparation and fusion are easily carried out and the complete solution of the soil ash in the molten alkali will extract all the Sr-90 present. Since the root mat contains most of the Sr-90 activity this fusion method cannot be used where the low-activity soil beneath is to be analysed separately. In this case soil samples of 500 gm. may have to be analysed in one determination, and either refluxing with $HClO_4/HNO_3$, or constant boiling HCl is convenient.

The amount of Sr-90 in sample 5 leached out using N ammonium acetate (3.3 mμc/sq. m.) is comparable with the two other accurately known values for the same depth: 3.9 and 5.7 mμc/sq. m. in samples 2 and 3, respectively. The amount of 'exchangeable' calcium in the area of Monachyle covered by sample 5 is extremely low.

KALE FROM ARGYLL.

TABLE 11.

Sample number	Date	Percent calcium in ash	Sr-89/ Sr-90 ratio	Sr-90 activity	
				µµc/kg.	S.U.
K1.	17.9.58	18.2	2.1	760	33
K2.	30.9.58	21.6	5.6	413	15
K3.	17.12.58	19.5	13.4	927	41.7
K4.	12.2.59	18.4	14	1,490	50.8
K5.	2.4.59	18.1	9	2,530	100

All the samples listed in Table 11 were obtained from Argyll; K2 was from Lochgilphead, which is about 40 miles to the north of Campbeltown (see map), where the other four samples were grown. K5 was spring growth from the same field as K4, which had been cut at the beginning of February 1959 before growth had started.

Since the Sr-89/Sr-90 ratio in the rain collected at Milford Haven (14), and at Leatherhead, Surrey (235) is the same as the ratio in the rain collected at Monachyle, it can be assumed that these ratios hold for the rain falling over Campbeltown. Based on the difference in the ratios in kale and rain it is calculated that at least 44% of the Sr-90 in sample 4 was taken by the roots, and at least 36% for sample 5.

For the Campbeltown kale samples the expected spring rise in activity in 1959 did occur, with a three-fold increase between September 1958 and April 1959. Foliar and root uptake appear to have contributed equally to this increase.

BROWN TROUT AND SEA TROUT.

TABLE 12.

Sample number	Date	Type of trout, & number	Location	Percent calcium in ash	S.U.
T1.	5.9.58	3 brown -	Harris	21.3	95
T2.	24.9.58	3 brown -	Argyll	20.7	81
T3.	30.9.58	1 brown -	Harris	21.6	152
T4.	30.9.58	1 sea -	Harris	18.7	147

The trout were ashed whole. The Argyll sample was from Campbeltown. The Sr-90 levels in the Harris samples are of the same order of magnitude as the sheep bone sample SB9 from Harris. (Table 8, p. 87) Although the Sr-90 content of the water in S.U. may be as high as the activity of the fish in S.U., it is doubtful whether uptake from the water is anything but a minor source of the total Sr-90 body burden. The diet must provide most of the Sr-90 ingested.

MISCELLANEOUS SAMPLES.

TABLE 13.

Sample number	Date	Type	Location	Percent calcium in ash	Sr-90 activity	
					µµc/kg.	S.U.
HG1.	11.9.58	Grass	Harris	2.75	2,270	1,190
GG1.	July 1958	Grass	Greenland	4.08	626	105
01.	Sept 1958	Oats	Campbeltown	3.85	50.5	63.2

The Harris grass sample had an expected level of activity, since the rainfall in Harris is similar to the Monachyle rainfall. The sample from southern Greenland is appreciably lower. No rainfall data are known for this Greenland area, so that no inference can be placed upon it.

The sample of oats from Campbeltown comprised the whole grain, which had not been milled.

Part IV.

APPENDICES.

APPENDIX 1.

Reagents used:

(a). Fuming nitric acid - 96% w/w, S.G. 1.50.

(b). Dilute nitric acid - 5N, made by diluting 320 ml. of concentrated (S.G. 1.420) nitric acid to 1 litre.

(c). Strontium carrier - 5 mgm./ml. of strontium, made by dissolving 6.038 gm. of dry strontium nitrate in water, adding some dilute nitric acid, and making up to 500 ml. with water. The acid prevents mould formation.

(d). Yttrium carrier - 10 mgm./ml. of yttrium, made by dissolving 6.349 gm. of yttrium oxide in dilute nitric acid and making up to 250 ml. with water.

(e). Barium carrier - Approximately 10 mgm./ml., made by dissolving about 4.445 gm. of $BaCl_2.2H_20$ in water and making up to 250 ml.

(f). Iron carrier - Approximately 5 mgm./ml., made by dissolving about 5.76 gm. of $FeCl_3.6H_20$ in water, adding some dilute nitric acid, and making up to 250 ml. with water. The acid prevents hydroxide precipitation.

(g). Ammonia - Analytical Reagent grade concentrated ammonia solution (S.G. 0.880) is assumed to be free from carbonate before a new bottle is opened.

(h). Perchloric acid - 72% w/w, S.G. 1.70.

(i). Phosphoric acid (syrupy) - 88% w/w, S.G. 1.75.

(j). Water - This is 'ELGA' de-ionised water, unless otherwise stated.

The concentrations of other reagents are specified in the text.

In the determination of radiostrontium in a variety of biological and inorganic materials there are two distinct parts. The first is peculiar to each material (the "PRELIMINARY"), and the second part is common to all the materials (the "DETERMINATION"). The DETERMINATION is described only once, for the case of bone ash, and all other procedures are referred back to this point after each PRELIMINARY has been dealt with. Unless otherwise stated all samples are done in duplicate. Notes on technique are given at the end of the complete procedure for bone ash.

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THE DETERMINATION OF RADIOSTRONTIUM IN BONE ASH.

PRELIMINARY.

(i). Accurately weigh 5 gm. of bone ash into a 250 ml. beaker, pipette in 5 ml. of strontium, and 1 ml. of yttrium carriers. Pour in 34 ml. of water from a measuring cylinder, followed by 50 ml. of fuming nitric acid. When all the ash has dissolved, a further 50 ml. of fuming nitric acid are added to precipitate strontium nitrate. The beaker is covered with a watch glass, and placed in a refrigerator until cold. (NOTE 1).

(ii). Filter the solution through a 3.7 cm. Whatman GF/A glass fibre filter paper, supported on a coarse (porosity 1) sintered glass filter funnel, with applied suction. The residue is dissolved in 25 ml. of hot water, and the filtrate is collected in a 100 ml. beaker. (NOTE 2).

(iii). 57 ml. of fuming nitric acid are added, and the

beaker refrigerated. The precipitate is filtered off and dissolved in 25 ml. of hot water, the filtrate being collected in the same beaker. 57 ml. of fuming nitric acid are added, the solution cooled, and filtered as before. (NOTE 3).

Continue the analysis as described under DETERMINATION.

When the specific activity of the bone ash is low (for example, in the case of human bone ash) much larger samples must be used to give a convenient count rate at the end of the separation. 10 gm. samples of ash can be conveniently handled using 5 ml. of strontium carrier solution, giving reasonably high recovery of strontium at the end. Where possible, larger quantities than 10 gm. of ash should be analysed in 10 gm. batches and the yields combined when all, or most, of the calcium present has been eliminated. A single sample of up to 40 gm. of ash can be handled, but 10 ml. of strontium carrier should be used due to the greater losses of strontium from the increased number of nitric acid separations required, and the larger volumes of solution per separation.

The PRELIMINARY then becomes:-

(i). Weigh out up to 40 gm. of bone ash into a 400 ml. beaker. To it add 10 ml. of strontium carrier, 1 ml. of barium carrier, 44 ml. of water, and 60 ml. of fuming nitric acid. When the ash has dissolved add a further 60 ml. of fuming nitric acid. Cool in a refrigerator. (NOTE 1).

(ii). Filter through a glass fibre filter paper, and dissolve the residue by pouring through 50 ml. of hot water, collecting the filtrate in the same beaker. Add 113 ml. of fuming nitric acid and cool.

(iii). Repeat the last paragraph.

(iv). When the quantity of precipitate becomes

appreciably smaller reduce the quantities of water and acid to 40 ml. and 90 ml. respectively, and when it appears that all calcium has been removed dissolve the residue from the last nitric acid separation in 25 ml. of water and collect in a 100 ml. beaker. Add 57 ml. acid, and cool.

Continue the analysis as described under DETERMINATION.

DETERMINATION.

(1). Dissolve the strontium nitrate precipitate from the last stage of the PRELIMINARY in 15 - 20 ml. of hot water, add 1 ml. of barium carrier and 1-2 drops of methyl red indicator. Dilute ammonia (1:1) is added from a dropper until the indicator just turns yellow. The pH of the solution is adjusted to about 5 by adding 1 ml. of 6M acetic acid and 2 ml. of 3M ammonium acetate solution. After diluting the solution to 30 - 40 ml. (NOTE 4.) and warming to just below the boiling point, 1 ml. of 1.5M sodium chromate is pipetted in to precipitate barium chromate. The solution is kept hot for 5 - 10 minutes, then filtered through a 3.7 cm. glass fibre filter paper, under suction, into a 100 ml. pressure flask. The beaker is washed out with a little water which is used to wash the filter paper. After pouring the filtrate into a clean 100 ml. beaker, the pressure flask is washed twice with water, and the washings combined with the filtrate. The residue is rejected.

(2). Sufficient ammonia is added to make the solution alkaline (as seen by the chromate colour change from orange

to yellow-green) followed by 5 - 10 ml. of saturated ammonium carbonate solution, which precipitates strontium carbonate. (NOTE 5). Heat the solution for 5 - 10 minutes until the strontium carbonate has coagulated, and settled to the bottom of the beaker. The solution is filtered through a 3.7 cm. glass fibre filter paper under suction, and the filtrate rejected.

(3). Dissolve the carbonate precipitate in 10 - 15 ml. of dilute nitric acid and collect in a 100 ml. beaker. 2 - 3 drops of '50 volume' hydrogen peroxide are added (NOTE 6), and 1 ml. of iron carrier. Heat the solution to just on the boiling point, and stir vigorously with a glass rod to remove all carbon dioxide from the solution. (NOTE 7.). Dilute to about 30 ml. and make alkaline with carbonate-free ammonia. Heat 2 - 3 minutes to coagulate the ferric hydroxide precipitate. The beaker is left to stand for a few minutes in cold water until the precipitate settles to the bottom, so that filtering through a 3.7 cm. glass fibre filter paper under suction is made more rapid. The beaker and residue are washed with a little water, the filtrate poured into a 100 ml. beaker, and the pressure flask rinsed out with water. The time at which the ferric hydroxide precipitation is made is noted, and all subsequent manipulations are carried out with the minimum delay, compatible with care and accuracy.

(4). Saturated ammonium carbonate solution is added to the ammoniacal filtrate to precipitate strontium carbonate, the solution being heated for a few minutes to coagulate the precipitate, which is then filtered on to a tared 2.5 cm. glass fibre filter paper, under suction. The beaker is carefully washed out on to the filter paper, and the precipitate washed several times with water, and finally with a little acetone. The filter paper is lifted carefully

from the sintered glass filter disc and placed on a 1 inch aluminium planchet, which has had the sample serial number scratched on the reverse side for identification. After drying the filter paper under an infra-red lamp, the sample is placed immediately in a suitable counting assembly and counted. The time at which counting commenced is noted. After an hour or two when counting is completed the filter paper is reweighed to obtain the weight of strontium carbonate, and hence the strontium recovery factor. (NOTE 8).

(5). The filter paper is placed in a 50 ml. beaker and 10 ml. of dilute nitric acid added to dissolve the carbonate precipitate, and a glass rod is used to pulp the filter paper. The contents of the beaker are filtered through a 1 inch diameter folded Whatman No. 4 filter paper in a 2 inch filter funnel (NOTE 9.) and the filtrate collected in a 60 ml. polythene bottle having a screw top. The beaker is washed out 4 or 5 times with 5 - 10 ml. of dilute nitric acid, and the washings poured through the filter paper, until about 40 ml. of filtrate have been collected. Repeated washings ensure that the strontium carbonate precipitate is quantitatively transferred to the polythene bottle. 1 ml. of yttrium carrier is added to the filtrate, and the sample number and date of preparation written on the polythene bottle in crayon. The solution is left to stand for a minimum of 14 days (but generally at least 18 days) for Sr-90 to reach equilibrium with Y-90. Since all Y-90 samples are prepared for counting on the first Monday after equilibrium is reached, the lying time is often longer than 18 days.

(6). Transfer the solution into a 100 ml. straight-sided centrifuge tube. Add ammonia to precipitate yttrium hydroxide, and note the time of precipitation. This is the 'zero time' which is mentioned later. Centrifuge, and

reject the supernate. (NOTE 10.). Dissolve the hydroxide precipitate in dilute nitric acid, increase the volume to 30 - 40 ml. with water, re-precipitate with ammonia, centrifuge and reject the supernate.

(7). Dissolve the precipitate in a little dilute nitric acid, and dilute to about 10 - 15 ml. Add dilute (1:1) ammonia from a dropper, until a slight permanent yttrium hydroxide precipitate is formed, then add dilute nitric acid dropwise just to dissolve the precipitate. 15 - 20 ml. of saturated oxalic acid are added to the solution, and the centrifuge tube is heated in a boiling water bath to coagulate the yttrium oxalate precipitate. After 10 - 15 minutes the solution is cooled and filtered on to a Whatman No. 5 filter paper cut just under 1 inch diameter. The precipitate is washed several times with water, and finally with a little acetone. Mount the sample on a 1 inch aluminium planchet using an acetone-soluble polystyrene cement, and count immediately, noting the time at which counting begins.

(8). Count the sample each day for a week, or longer if thought necessary. Remove the sample from the planchet using a little acetone, and place it into a tared silica crucible. Gently burn off the acetone, cement, and filter paper, then ash the yttrium oxalate to the oxide (Y_2O_3) at 800 - 900^{o}C for about 20 minutes. Cool, and weigh the crucible. Calculate the yttrium recovery factor.

(9). Construct a decay graph for the Y-90 activity, and calculate the activity at 'zero time'. Hence calculate the Sr-90 and the Sr-89 content of the original material.

NOTES.

(1). The addition of fuming nitric acid makes the solution very hot so that it may take two hours or more to cool in the refrigerator. With several experiments generally being carried out simultaneously the time taken to cool is not important. The beaker may be rapidly cooled in cold running water for 20 - 30 minutes, after which the solution may be filtered.

(2). A measuring cylinder is filled to the 25 ml. mark with hot water. About half of this is poured into the 250 ml. beaker to dissolve the residue remaining there, and then this solution is poured through the filter, dissolving any strontium nitrate on the filter paper. The filtrate is collected in a 100 ml. beaker, and this is repeated with the remaining hot water to dissolve completely all strontium nitrate.

(3). Using 5 gm. of bone ash the three nitric acid separations described should remove all calcium present. Whether this in fact is the case is judged by experience. Further separations using 25 ml. of water and 57 ml. of fuming nitric acid are carried out until the analyst is satisfied that a complete separation has been achieved.

(4). It is easier to judge the methyl red colour change using the smaller volume of 15 - 20 ml. If barium chromate be precipitated from this volume of solution some strontium may be lost by occlusion and adsorption, so the solution is diluted to prevent this.

(5). With 5 ml. of strontium carrier solution, the strontium carbonate precipitate does not form until 5 - 10 seconds after the addition of the ammonium carbonate precipitant.

If an immediate, voluminous precipitate forms this means that the nitric acid separations have not removed all the calcium. The mixed carbonate precipitate is therefore dissolved in 25 ml. of water containing sufficient dilute nitric acid to dissolve the carbonates, and 57 ml. of fuming nitric acid added, as in the last paragraph of the PRELIMINARY. The DETERMINATION is started from the beginning again.
When larger quantities than 5 ml. of strontium carrier are used the strontium concentration in the chromate filtrate will be high enough to give immediate precipitation on adding ammonium carbonate solution, so this simple visual check on purity cannot easily be applied.

(6). The strontium carbonate precipitation from the chromate solution may contain occluded chromate, which is reduced by the peroxide to the trivalent Cr^{+++} state, in which form it is precipitated with the ferric hydroxide scavenge.

(7). If carbon dioxide remains in solution when ammonia is added for the iron scavenge, strontium carbonate will be precipitated and lost. For the same reason carbonate-free ammonia must be used for this stage.

(8). No increase in the weight of the filter paper and strontium carbonate precipitate due to moisture absorption was found after leaving a sample in a covered box for several days. Since the glass fibre filter papers used are limp, and readily assume the shape of the planchet no adhesive is necessary. These papers cannot be used, of course, for the yttrium oxalate precipitate, which has to be ashed after counting, and here an ordinary cellulose filter paper is used.

(9). The small piece of filter paper is used to prevent absorption and loss of strontium.

(10). The supernate, which contains the strontium, is retained until the yttrium has been successfully mounted, and counted for the first time, and only then is the supernate rejected. If the yttrium is accidentally lost this supernate is used to repeat the DETERMINATION from the iron scavenge.

APPENDIX 2.

THE DETERMINATION OF RADIOSTRONTIUM IN MILK ASH.

PRELIMINARY.

(i). Weigh out 20 gm. of ash into a 500 ml. polythene centrifuge bottle. Add 10 ml. of strontium carrier, 1 ml. of barium carrier, and about 200 ml. of water. Dissolve the ash by the addition of 25 ml. of fuming nitric acid. If appreciable quantities of carbon and/or silica (from the ashing dish) remain, filter the solution. Add 1 ml. of syrupy phosphoric acid, and excess ammonia. Centrifuge and reject the supernate after testing for excess phosphate by adding a drop of barium carrier solution. Wash the residue by stirring with 200 ml. of water containing about 0.5 ml. of phosphoric acid, and 2 ml. of ammonia. Centrifuge and reject the supernate.

(ii). Add just sufficient fuming nitric acid (10 - 15 ml.) to dissolve completely the mixed phosphate residue in the centrifuge bottle, and pour the solution into a 400 ml. beaker. Rinse out the centrifuge bottle, and add the washings to the beaker. Evaporate the solution to the point of dryness, cool, and add 40 ml. of water to the beaker. Measure out 90 ml. of fuming nitric acid and pour about half of it into the beaker. When the residue

has dissolved, add the remainder of the acid. Cool the beaker in a refrigerator, filter through a glass fibre filter paper, and reject the filtrate.

(iii). Dissolve the residue in 25 ml. of water, collecting the filtrate in the same beaker. Add 57 ml. of fuming nitric acid, cool, filter, and reject the filtrate.

(iv). Dissolve the residue in 25 ml. of water, collecting the filtrate in a 100 ml. beaker. (NOTE 2, p. 104) Add 57 ml. of fuming nitric acid, cool, and filter, and repeat this step until it is judged that all the calcium has been removed. Three 25/57 separations should be sufficient.

Continue as described under DETERMINATION, page 100.

APPENDIX 3.

THE DETERMINATION OF RADIOSTRONTIUM IN VEGETABLE ASH.

NOTE.

The method described uses 5 gm. of ash for each analysis, but in many cases the ashing of a sample of vegetation may result in a few grams of ash. In this case the calcium determination is done first and the remaining ash divided into two, giving duplicates for the radiostrontium determination. Also, the specific activity of an ash may be so high that 1 - 2 grams of ash per analysis gives a suitably high count rate at the end, and the smaller the sample the easier the manipulations. With low activity ashes 10 grams are used, and the acid volumes in step (i) are increased as becomes necessary.

PRELIMINARY.

(i). Weigh out 5 gm. of ash into a 400 ml. beaker. Add 5 ml. of strontium carrier, 1 ml. of barium carrier, about 50 ml. of water, then 35 ml. of fuming nitric acid. Stir, and add 40 ml. of perchloric acid. Evaporate almost to dryness on a hot-plate in an efficient fume

cupboard. Add about 5 ml. of concentrated hydrochloric acid and about 100 ml. of water; boil for several minutes, then filter under suction through a Whatman No. 42 filter paper. Wash the residue with a little water, then transfer the residue and filter paper to the beaker, add about 100 ml. of water and again boil for a few minutes. Filter, and combine the filtrates in a 500 ml. polythene centrifuge bottle.

(ii). Add 1 ml. of syrupy phosphoric acid and excess ammonia. Centrifuge and reject the supernate, after testing for excess phosphate by adding a drop of barium carrier solution. Wash the residue by stirring vigorously with 200 ml. of water containing 0.5 ml. of phosphoric acid and 2 ml. of ammonia. Centrifuge and reject the supernate.

Continue from paragraph (ii), Appendix 2, page 107.

APPENDIX 4.

THE DETERMINATION OF RADIOSTRONTIUM IN SOIL:

EXTRACTION BY NITRIC AND PERCHLORIC ACIDS.

PRELIMINARY.

(i). Weigh out 300 gm. of ashed soil into a 2 litre round-bottomed flask. Add 0.966 gm. of strontium nitrate (400 mgm. of Sr), about 240 ml. of water, and 150 ml. of fuming nitric acid. After mixing, pour in 80 ml. of perchloric acid. Distil over all the liquid, the operation taking 6 - 7 hours.

(ii). When the flask is cool, add 500 ml. of dilute (1 : 9) hydrochloric acid. Boil vigorously under reflux for 1 - 2 hours, until all the lumps in the flask have been broken down. Filter hot through a Whatman No. 54 paper under suction, and wash the residue with a little water.

(iii). Transfer the entire residue, including the filter paper, into a 2 litre beaker, add 400 ml. of boiling water, and 30 - 40 ml. of concentrated HCl. Boil with constant stirring for 5 - 10 minutes. Filter and wash as before.

(iv). Repeat the above paragraph until a filtrate is obtained which does not have the orange colour of dissolved ferric chloride from the soil. It is then assumed that

all calcium and strontium has been extracted. Reject the residue, and evaporate the combined filtrates to 1 - 1½ litres, in a 2 litre beaker.

(v). To the hot solution add 150 gm. of oxalic acid, and 20 gm. of ammonium acetate as buffer. Slowly add (1 : 1) ammonia until pH 4 is reached, as indicated by narrow range pH papers. Cover the beaker with a watch glass, and stand in a warm place for 3 - 4 hours. Filter under suction then transfer the residue and filter paper to a small silica dish. Ash at about 800°C for 20 - 30 minutes. Transfer the ash to a 400 ml. beaker, and wash out the silica dish with dilute nitric acid. Add sufficient dilute nitric acid to dissolve the entire carbonate residue, and boil to remove carbon dioxide.

(vi). Dilute the solution to about 200 ml. and add excess ammonia to precipitate any iron and aluminium present. Heat for a few minutes, then filter off the hydroxides, and any carbon and silica present. Add excess ammonium carbonate solution to the filtrate, and heat for a few minutes to coagulate the precipitate. Filter through a tared sintered filter crucible, containing a glass fibre filter paper. Wash, dry, and reweigh to obtain the weight of calcium carbonate from the soil, plus recovered strontium carrier.

(vii). Completely dissolve the mixed carbonates in dilute nitric acid, and evaporate the solution to the point of dryness. Add 40 ml. of water to dissolve the mixed nitrates, and then 90 ml. of fuming nitric acid. Cool in a refrigerator, and filter through a glass fibre filter paper.

(viii). Dissolve the residue in 25 ml. of hot water and transfer to a 100 ml. beaker. Add 57 ml. of fuming

nitric acid; cool, and filter as before.
Repeat this step.

(ix). Dissolve the residue in 15 - 20 ml. of water, add 1 ml. of barium carrier, and 1 - 2 drops of methyl red indicator. From a dropper add dilute (1 : 1) ammonia until the indicator just turns yellow. Add 1 ml. of 6M acetic acid, and 2 ml. of 3M ammonium acetate. Dilute the solution to 30 - 40 ml., warm to just below the boiling point, and add 1 ml. of 1.5M sodium chromate. Continue heating for 5 - 10 minutes, then filter. Wash, then reject the residue.

(x). Transfer the filtrate to a 100 ml. beaker, add ammonia until alkaline, then excess saturated ammonium carbonate solution. Heat for 5 - 10 minutes, filter, and reject the filtrate. Wash the residue to remove absorbed chromate.

(xi). Pour 25 ml. of water through the filter paper in several portions: the first two portions containing enough dilute nitric acid to dissolve the carbonate precipitate. Collect in a 100 ml. beaker, and add 57 ml. of fuming nitric acid. Cool in a refrigerator, and filter as before.

Continue as described under DETERMINATION, page 100.

NOTES:

In paragraph (v), if less than 150 gm. of oxalic acid are used a brown precipitate of ferric hydroxide may come down on adding ammonia, before pH 4 is reached. If this should happen, a little hydrochloric acid is added to dissolve the ferric hydroxide , and enough extra oxalic acid to complex all the iron, and prevent a recurrence of the trouble on raising the pH again to 4.

To eliminate all the radium present in soil two barium chromate scavenges are carried out; one is described above in the PRELIMINARY, and the second in the DETERMINATION.

In the case of one soil sample constant boiling hydrochloric acid was used in place of nitric and perchloric acids. 150 gm. of soil ash were weighed into the 2 litre flask, and 300 ml. of constant boiling (21%) hydrochloric acid added. The mixture was refluxed for about six hours, then the acid distilled off, until the flask was nearly dry. The extraction is continued as before, from paragraph (ii) of this Appendix.

APPENDIX 5.

THE DETERMINATION OF RADIOSTRONTIUM IN SOIL:

EXTRACTION BY 6 N HYDROCHLORIC ACID.

PRELIMINARY.

(i). Weigh out 1.5 kgm. of dry, sieved soil into a 5 litre beaker. Add 0.966 gm. of strontium nitrate (400 mgm. Sr), and 1.5 litres of 6 N HCl. Mix thoroughly with a glass rod, and stir occasionally during one day, and leave standing overnight.

(ii). The fine suspension of clay particles may make filtration impossible, therefore centrifuge the mixture in 500 ml. bottles, and filter the supernate through a Whatman No. 54 paper, under suction to remove floating organic material. Mix the residue with about 1 litre of water, centrifuge and filter. Combine the filtrates.

(iii). Empty the residue into the 5 l. beaker and add 1.5 l. of 6 N HCl. Again stir occasionally and leave overnight. Repeat paragraph (ii). Reject the residue, and combine all filtrates.

(iv). The filtrate contains too much HCl to be neutralised with ammonia, and is therefore evaporated to 150 - 200 ml. The solution is transferred to a 2 litre beaker, and diluted to about 1 litre.

Continue from paragraph (v) of Appendix 4, p.112.

APPENDIX 6.

THE DETERMINATION OF RADIOSTRONTIUM IN SOIL:

EXTRACTION BY ALKALI FUSION.

This method is used to give complete extraction, due to solution of the ash in the fused melt. The resulting large quantity of sodium salts makes the method suitable only for soils with a high specific activity. A maximum of 100 gm. of soil ash is recommended which is handled in two or more batches.

Two types of fusion have been carried out; the first using only sodium carbonate, while the other is a low-temperature fusion with sodium hydroxide and a little sodium carbonate. The NaOH fusion has the advantage of being carried out over a bunsen flame. Unfortunately some sputtering on adding soil ash to the melt is unavoidable, and at this stage the strontium carrier has not been homogeneously mixed with the soil ash, which will lead to inaccuracy. This difficulty is overcome if the fusion is done with sodium carbonate alone. Here it is easy to premix the ash and the alkali before fusion, and little or no sputtering takes place, since the mixture is gradually heated to the melting point. The disadvantage of this method is the high (900°C) fusion point. Where a suitable muffle furnace is available the sodium carbonate fusion method is recommended.

SODIUM CARBONATE FUSION.

PRELIMINARY.

(i). Weigh out 100 gm. of soil ash on a large sheet of paper, and mix in 0.966 gm. of strontium nitrate (400 mgm. of Sr), and 400 gm. of anhydrous sodium carbonate. Place about one quarter of this into a 270 ml. (80 X 80 mm.) nickel crucible. Fuse at 900 - 950°C in an electric muffle furnace, and leave in for 10 minutes after a clear melt has been obtained. Allow to cool, and knock out the fused mass. Repeat the fusion process with the three remaining quarters of the carbonate/ash mixture.

(ii). Break down the larger lumps in a mortar and pestle, and then reduce to a fine powder in an electric grinder. Heat 500 ml. of water in a 5 litre beaker, and add the ground material to it.

(iii). Taking great care add 400 ml. of 72% perchloric acid. Stand the beaker on a hot-plate at low heat in a fume cupboard. Stir the hot solution with a glass rod until all the alkali has been neutralised, and insoluble material remains as fine particles. Increase the temperature of the hot-plate to evaporate the solution to dryness. The gelatinous mass of silica which is produced is broken up with the stirring rod, and finally remains as a white powder.

(iv). When cool, add about 2 litres of water to the beaker, and stir until the silica is in fine suspension; then allow the solid material to settle to the bottom. Filter under suction through a Whatman No. 5 paper. Wash the residue in the beaker with 300 ml. of hot dilute (1 : 9) HCl, and pour through the filter; then repeat this step

using hot water. Reject the residue , and combine the filtrates in a 5 litre beaker.

(v). Pour in strong sodium hydroxide solution to raise the pH to 7. Warm the solution and, while stirring, add 50 gm. of sodium carbonate. When the carbonate has dissolved leave standing for 1 - 2 hours, or overnight. Filter through a Whatman No. 5 paper under suction. Dissolve the residue with dilute HCl, and collect in a 2 l. beaker. Adjust the volume of solution to about 1 litre with water, and heat to 70 - 80°C. Add 100 gm. of oxalic acid and 15 gm. of ammonium acetate. Stir until dissolved.

Continue from paragraph (v), Appendix 4, page 112.

SODIUM HYDROXIDE FUSION.

PRELIMINARY.

(i). Weigh out 50 gm. of soil ash on a sheet of paper and mix in 0.483 gm. of strontium nitrate (200 mgm. Sr). Fill a 270 ml. nickel crucible with 175 gm. of sodium hydroxide pellets, and heat the crucible over a low bunsen until the alkali has melted. Add the soil ash to the crucible in small portions, taking care that excessive frothing is avoided. Stir the mixture with a stout iron rod (2 - 3 mm.) after each addition. When all the ash has been added, empty 10 gm. of sodium carbonate into the crucible, heat vigorously, and stir until a homogeneous melt is obtained. Maintain heating for 5 - 10 minutes, then allow to cool with the iron rod in the melt.

(ii). Knock out the solid mass from the crucible into a 5 litre beaker, and wash the stirrer free from adhering material. Fill the crucible half full of water, boil and pour the solution into the beaker, when the residue in the crucible is in the form of a suspension.

Repeat paragraphs (i) and (ii) with a further 50 gm. of soil ash, combining the solid residues and all washings in the 5 litre beaker, giving a total volume of about 500 ml.

Continue from paragraph (iii) of this Appendix, page 117.

APPENDIX 7.

THE DETERMINATION OF RADIOSTRONTIUM IN SOIL:

EXTRACTION BY AMMONIUM ACETATE.

PRELIMINARY.

(i). Duplicates. Weigh 400 gm. of dry, sieved soil into a 5 litre beaker. Pour in 3.5 litres of N ammonium acetate, which has been adjusted to pH 7 by adding either acetic acid or ammonia if necessary. Leave standing for a week, but stir the mixture briskly at intervals during the day.

(ii). To sediment the suspension of clay particles, centrifuge the supernate in 500 ml. bottles, then filter through a Whatman No. 54 paper under suction. Finally transfer the sludge to the filter paper and draw off as much liquid as possible. Wash the residue with a little water, and return all the filtrates to a 5 litre beaker. Reject the residue.

(iii). Weigh out 0.483 gm. of strontium nitrate (200 mgm. of Sr) and transfer to the beaker. Warm the solution to about 70°C and stir to dissolve the strontium nitrate. Weigh out roughly 200 gm. of anhydrous sodium carbonate and add this slowly to the solution, while stirring constantly. After the carbonate has dissolved

cover the beaker, and leaving standing overnight.

(iv). Filter the calcium and strontium carbonates under suction, and reject the filtrate. Transfer the filter papers and precipitates from the duplicates into a silica dish, dry over a low bunsen, then ash at 800 - 900°C for half an hour. When cool transfer the residue into a 400 ml. beaker; wash the silica dish with water, then with dilute nitric acid, and add the washings to the beaker. Add sufficient dilute nitric acid to dissolve all the carbonates, and boil the solution for a few minutes to expell all carbon dioxide.

Continue from paragraph (vi), Appendix 4, page 112.

APPENDIX 8.

THE DETERMINATION OF RADIOSTRONTIUM IN WATERS.

NOTES:

Strontium carrier and nitric acid should be added to the collection vessel before the water is poured in. If this has not been done first, add the carrier and acid and leave overnight. The volume of water is measured.

This has been found to be a satisfactory method for the analysis of the rain and burn waters mentioned in this thesis. Should the water be very hard, or contain large quantities of dissolved salts, the residue, on evaporation to dryness, should be taken up in dilute hydrochloric acid, and the calcium and strontium precipitated as phosphates. This method is similar to that for the determination of radiostrontium in milk ash, Appendix 2, page107.

PRELIMINARY.

(i). Evaporate the water to 100 - 200 ml. in a 5 litre beaker; rinsing out the water containers with dilute nitirc acid and water. Transfer to a 400 ml. beaker, and wash out the 5 litre beaker with acid and water. Evaporate to dryness.

(ii). Add 40 ml. of water to the beaker, and measure out 90 ml. of fuming nitric acid. Pour about half the acid into beaker and stir to dissolve the residue, then add the remainder of the acid. Cool in a refrigerator.

(iii). Filter the solution through a glass fibre filter paper under suction, and reject the filtrate. Dissolve the residue in 25 ml. of hot water, and collect the solution in a 100 ml. beaker. Add 57 ml. of fuming nitric acid, and cool.

Continue as described under DETERMINATION, page 100.

APPENDIX 9.

DETERMINATION OF CALCIUM.

NOTE:

"Water" refers to 'ELGA' de-ionised water, unless otherwise stated.

Bone and milk ash.

Weigh out exactly 0.5 gm. of ash into a 250 ml. beaker, and dissolve in 5 ml. of concentrated hydrochloric acid in about 40 - 50 ml. of water. If a slight residue of silica or carbon remains, use only about 20 ml. of water to dissolve the ash, filter through a small paper, and wash repeatedly with water bringing the total volume of filtrate to 40 - 50 ml. Add about 35 ml. of saturated oxalic acid solution, and warm to about 80°C.

Using 2 drops of bromo-phenol blue as indicator, add dilute (1 : 2) ammonia from a burette to pH 4 (purple-green colour). The end point is also checked with narrow range pH paper. Stand the beaker on an asbestos pad over a low bunsen for 3 - 4 hours, then filter through a tared sintered glass filter crucible of No. 4 porosity. Wash the beaker several times with water and pour the washings through the crucible. Finally, wash twice with a little acetone, and dry in an oven at 110 - 115°C for 30 minutes. Cool in a desiccator and reweigh. The calcium is in the form of calcium oxalate monohydrate; the gravimetric factor being 0.2743. Test runs are carried out using pure calcium carbonate.

Vegetable ash.

This includes all the ashes which do not dissolve completely in dilute HCl, other than soil which is dealt with later. Two different methods can be used. Where the quantity of ash available is small, method (i) is used; for larger amounts of ash method (ii) may be used.

(i). EEL Flame Photometer Method.

Weigh out accurately 0.1 gm. of ash into a 100 ml. beaker. Add 10 ml. of concentrated HCl and evaporate to dryness on a hot-plate. Repeat with another 10 ml. of HCl. Boil the residue with about 30 ml. of water for 5 minutes, and filter into a 100 ml. volumetric flask. Boil several portions of water in the beaker, and wash the residue and filter paper with them until almost up to the mark. Cool the flask to its calibration temperature ($20^{\circ}C$), and make up to the mark with water.

Prepare a three inch resin column in a 25 ml. burette using Amberlite IR-120 (H) cationic exchange resin, which has been ground between 60 and 120 mesh and previously washed with 5N nitric acid, and water. Pipette a 25 ml. aliquot from the volumetric flask into the burette, and allow the solution to percolate through the column at a rate of about 1 ml. per minute. Fill the burette to the top with water and run it through the column at the same rate. Add 24 ml. of 5N nitric acid to the column from a measuring cylinder, and run the acid through the column at a rate of about 0.5 ml. per minute, and collect the eluate in a 25 ml. volumetric flask. After elution

make up to the mark with acid, and wash out the column with water to prepare it for the next estimation. Leave the columns standing filled with water, to prevent the resin from drying out. All the phosphate in the original ash has been removed and replaced by nitrate ions.

The calcium content of the acid solution is determined using the EEL Flame Photometer, with water as the "zero", and 50 parts per million of calcium in water giving the "100" mark on the galvanometer scale. A calibration chart converts the scale reading of the sample into p.p.m. of calcium, and from this the calcium content of the original ash is calculated.

If it is known in advance that 25 ml. of the sample solution will contain more than 50 p.p.m. of calcium (giving an off-the-scale deflection), a suitably smaller aliquot is taken, and the burette filled nearly to the top with water as the solution is being run in, to ensure proper draining of the pipette.

The light spot on the galvanometer scale can be read to $\pm \frac{1}{2}$ of one scale division, which is equivalent to $\pm$ 1% of the full scale deflection. The aliquot chosen should therefore give as high a scale deflection as possible to minimise scale reading errors. The efficiency of the resin column procedure is determined by passing 10 or 20 ml. aliquots of the 50 p.p.m. calcium standard solution through the column, and determining the calcium content of the eluate. Low results are generally due to fast flow rates through the column, and high results to residual calcium from the previous determination remaining on the column.

(ii). Gravimetric method.

Weigh out accurately 1 gm. of ash into a 100 ml. beaker; add 15 ml. of water, 15 ml. of fuming nitric acid, and 15 ml. of perchloric acid. Evaporate to dryness on a hot-plate. Cool, and rinse the sides of the beaker with a little water to wash down any adhering solid material. Add 15 ml. of perchloric acid, and evaporate to dryness again. Boil the residue with 20 ml. of water, and filter into a 250 ml. beaker. Wash the residue several times with boiling water until the combined filtrate has a volume of about 50 ml. Add about 35 ml. of saturated oxalic acid solution and warm to about 80°C.

Continue from the second paragraph of the bone ash procedure on page 124.

Soil.

Due to the different extraction techniques used for the radiostrontium analysis, it is convenient to determine calcium and radiostrontium on the same sample. With soils of low calcium content this has the advantage that the large samples used give measurable quantities of calcium. The calcium carbonate precipitate obtained in paragraph (vi) of Appendix 4, page 112, has to be corrected for the weight of strontium carbonate recovered at the end of the separation.

Part V.

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REFERENCES.

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Certain Aspects of Polymucle tide Biosynthesis

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Considerable interest has been focussed, in recent years, on enzyme systems, from various sources, which are capable of the *in vitro* synthesis of deoxyribonuclei acid. Deoxyribonucleic acid which is a linear copolyme of the 5'-monophosphates of deoxyadenosine, deoxyguanosi deoxycytidine and thymidine, in 3'-5'-diester linkages is formed from the four deoxyribonucleoside-5'-tri-phosphates, with the elimination of inorganic pyro-phosphate by an enzyme termed "deoxyribonucleic acid polymerase". The action of this enzyme has been shown to be reversed at certain concentrations of inorganic pyrophosphate and this back reaction has been termed pyrophosphorolysis. Parts I and II of this thesis describe methods for the preparation of the deoxyribo-nucleoside triphosphates, and Part III gives details of attempts to demonstrate the pyrophosphorolysis of deoxy-ribonucleic acid in various tissue extracts.

Part I. The ability of extracts of Ehrlich ascite carcinoma to phosphorylate the deoxyribonucleoside ~~tri-~~ mon phosphates was studied with a view to using this system for the preparation of deoxyribonucleoside triphosphates

on a large scale. The extracts were found to be capable of converting deoxyadenosine monophosphate to the triphosphate in yields of up to 80%, deoxyguanosine monophosphate to the triphosphate in yields of up to 60%, deoxycytidine monophosphate to the triphosphate in yields of around 15% and thymidine monophosphate to the triphosphate in about 1% yield. This system was obviously only suited to the large scale preparation of deoxyadenos and deoxyguanosine triphosphates and preparative methods for these compounds are described.

Part II. The method described by Smith and Khorana for the preparation of adenosine triphosphate from adenosine monophosphate was used to prepare thymidine triphosphate from thymidine monophosphate in 18% yield. An investigation of the purification procedure led to several improvements and thymidine triphosphate labelled with tritium was obtained from tritiated thymidine monophosphate in 36% yield.

The effect of various changes in the composition of the reaction mixture on the yield of deoxyribonucleoside triphosphate was studied and it was found that by decreasing the ratio of the deoxyribonucleoside monophosphate to the other constituents of the reaction mixture, to one third of that used by Smith and Khorana

and by reacting the deoxyribonucleoside triphosphate in the form of its tri-n-butylammonium salt, greatly improved yields of the deoxyribonucleoside triphosphates could be obtained.

The method of isolating the deoxyribonucleoside triphosphates was further improved by the introduction of a charcoal column procedure for the simultaneous removal of contaminating lithium chloride and inorganic polyphosphates.

By means of the altered reaction conditions and improved isolation procedure thymidine, deoxyadenosine and deoxyguanosine 5'-triphosphates were prepared from the respective monophosphates in yields of around 70%. Deoxycytidine triphosphate was prepared from deoxycytidine monophosphate in 50% yield.

^{32}P-labelled thymidine monophosphate was synthesized from thymidine by the methods of Hurwitz and Tener, converted to thymidine triphosphate and isolated in 70% yield. The specific activity of the product was 1.0 - 1.5 x 10^6 cpm/µmole. A slight modification of the Tener method for the preparation of ^{32}P-labelled thymidine monophosphate led to the preparation of ^{32}P-labelled thymidine triphosphate with a specific activity of 3.5 x 10^6 cpm/µmole.

Part III. Attempts were made to demonstrate the pyrophosphorolysis of deoxyribonucleic acid in extracts of Ehrlich ascites carcinoma by the use of ^{32}P-labelled inorganic pyrophosphate and an analytical procedure involving the separation of inorganic pyrophosphate from deoxyribonucleoside triphosphates on charcoal columns The results obtained were inconclusive because of the variability of the analytical procedure. In a different procedure the pyrophosphorolysis of tritium labelled deoxyribonucleic acid was studied by analysis of the reaction products on paper chromatograms. This procedure was used to assay crude and partially purified extracts of Ehrlich ascites carcinoma but no evidence for the occurrence of pyrophosphorolysis was obtained.

A new method of separating inorganic pyrophosphate from deoxyribonucleoside triphosphates was developed and used in conjuction with ^{32}P-labelled inorganic pyrophosph in attempts to demonstrate the pyrophosphorolysis of deoxyribonucleic acid in extracts of Ehrlich ascites carcinoma, rabbit thymus glands and calf thymus glands. No evidence for the occurrence of pyrophosphorolysis was obtained and it was concluded that pyrophosphorolysis, which has only been demonstrated in highly purified syste could not be demonstrated in crude extracts.