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STRONTIUM METABOLISM STUDIES

by

Janet M. Warren

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INTRODUCTION

Strontium was first recognised in 1791 by Thomas Hope, Professor of Medicine at Glasgow, when he was examining minerals from a lead mine in Strontian, a village in Argyllshire.(1) Later, this mineral was identified as the carbonate of a new alkaline earth and was given the name Strontia. It was not until 1808 that the element was isolated by Davy who began work on the alkaline earths in 1807 after isolating sodium and potassium.(2) Following the method described to him in a letter by Berzelius, he separated bases from barytes, strontia, lime, and magnesia, and proposed names for these new elements including strontium.

Strontium occurs in nature, chiefly as the carbonate (strontianite) and sulphate (celestine) and also associated with other alkaline earth minerals but it is in much lesser abundance than calcium and barium which it closely resembles. Until recent years, strontium was regarded to be of little importance for its commercial uses were few - the main ones being in sugar refining and the nitrate providing the red colouration in fireworks. Its uses in the medical field were also few.

With the advent of high powered nuclear weapon testing, strontium suddenly assumed a new importance with the creation

of a new isotope strontium 90. Until this time no radioactive isotopes of the element were known in nature. In the Spring of 1954 it was realised that isotopes of many elements (including those of strontium) were being deposited over the earth's surface. At this time scientists and medical authorities were becoming very conscious of the hazards to mankind from ionising radiations. It was thought advisable to keep a close check on the radioactive material which was 'falling out' of the atmosphere following nuclear tests. Several laboratories, notably in the U.S.A. and U.K. were set up solely for the measurement of such radiations. One of these was established in Glasgow.

This laboratory was sponsored (initially in 1958) by the Medical Research Council. Glasgow was thought to be a suitable location for such a project, firstly because it has a high rainfall and 'fall-out' was thought to be partly dependent on rainfall, and secondly because it has a hospital solely for children who for reasons to be given later would be the first to be affected by fall-out. It is also appropriate that work on strontium should be housed in the Child Health Department of the Faculty and University in which Hope, the discoverer of the element, was Professor.

The early work consisted almost entirely of post mortem bone analyses for strontium 90, as the bone, being the main calcium containing organ in the human body, is the most suitable

sample for radio-strontium estimations. This work is discussed and summarised later. More recently experimental work has been carried out on food materials and excreta in an attempt to assess radiostrontium content in the living child. These experiments are described in some detail.

SECTION I

CONTAMINATION BY FALL-OUT

CONTAMINATION BY FALL-OUT

(a) Mechanisms of fall-out

In nuclear explosion, energy may be released by one of two processes or by a combination of both 'fission' and 'fusion'. In the first, which is the breakdown into two new nuclei of Uranium 235 (or Uranium 233 or Plutonium 239) by the bombardment of neutrons, a large number (in the region of 200) of radioisotopes may be produced. These fall mainly into two categories - those whose atomic weight is approximately 90, and those with atomic weight in the region of 140. This rules out most of the elements of biological importance, such as carbon, oxygen, hydrogen, calcium, nitrogen, and iron, although there are several isotopes which are of biological significance. In the second case, nuclear energy results from a fusion process. This involves a binding together of lighter nuclei and the thermonuclear reaction results in a very high neutron flux which can cause the activation of materials in the vicinity of the explosion so that large quantities of carbon 14 are produced from atmospheric nitrogen. The second mechanism is that of Thermonuclear bombs while the first is the source of energy of Atomic bombs or, if controlled, of Atomic piles. It is thought, however, that Thermonuclear or H-bombs usually involve both fusion and fission and so fission products as well as the

low-weight radioisotopes are dispersed. Fall-out radiations can be divided into three categories. (a) Radiation in areas adjacent to the test or explosion site, (b) fall-out from the radioactive cloud in the troposphere, (c) that resulting from slow fall-out of fission products from the stratosphere.

Adjacent to the site

The fall-out in this region consists in the main of the larger particles of debris, and the rate of its deposition depends largely on the conditions of wind and weather prevailing at the time. In the case of thermonuclear weapons, dust, and other matter in the vicinity can be caught up, vaporized and made intensely radioactive by the high neutron flux, so that fall-out in the test site is very abundant. It was such circumstances which on March 1st 1954 affected the Japanese fishing vessel 'Fukuryu Maru' which was 94 miles away from a test site in the Marshall Islands. This type of activated material is, however, fairly rapidly deposited - within a few hours of the explosion.

Tropospheric fall-out

This finer grained material, although not distributed over the whole of the earth's surface, has a much wider distribution than that of the first category.

Material blown by the explosion into the troposphere, or lower atmosphere, may circle the earth several times during a period of a few months before it is completely returned to

the surface by rain precipitation, and sedimentation.

The most important radioisotopes, biologically, in this fall-out are those with half-lives in the region of the debris' duration in the atmosphere. For example, detectable amounts of iodine 131, with half-life of eight days, have been found in the thyroid following a bomb test. Also, quite significant amounts of strontium 89 (half-life 64 days) have been found. It has been suggested that the irradiation to bone as a result of selective concentration of such intermediate and short lived isotopes in active growing points may be more intense than if the isotopes were distributed throughout the skeleton.(3)

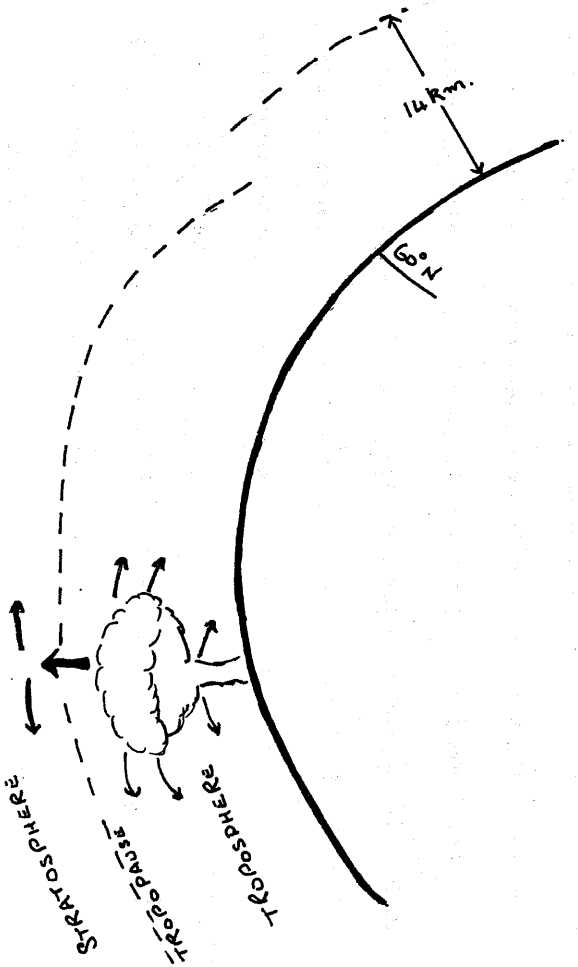
Stratospheric fall-out

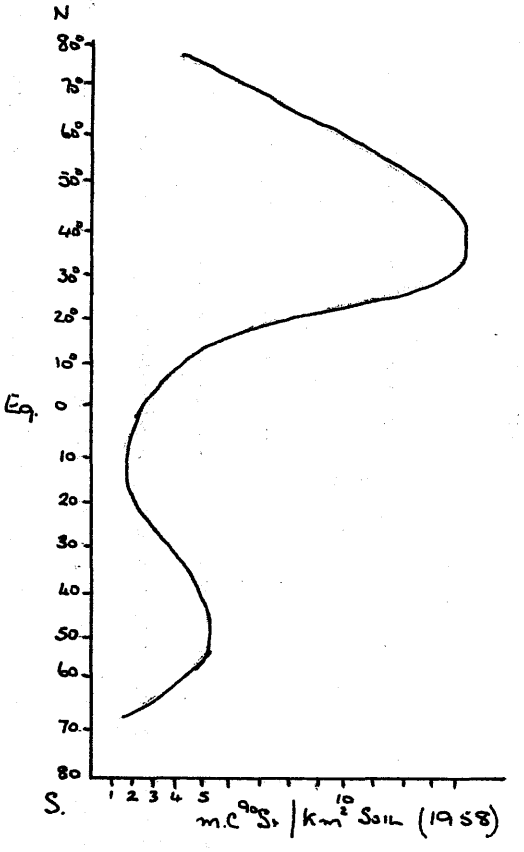
Stratospheric debris consists of the finer grained particles created in the explosion. Such material is carried by the force of the detonation above the region of atmosphere where weather processes are in progress, and so it may circle the earth in this region for several years.

Slowly the fission material filters back to the troposphere - either by the sinking of air in the polar regions, or via a break in the level of the tropopause in the middle latitudes.(4) See figure 1.

As much of the stratospheric debris remains in the upper atmosphere for some years, the fission elements of most significance in this group are those of long half-life, namely

FIGURE 1





VARIATION of STRONTIUM-90 DEPOSITION WITH LATITUDE. (61)

caesium 137 and strontium 90.(5)(6)

These isotopes have decay chains of a similar nature :-

FISSION PRODUCTS of MASS 90 :-

$$^{90}Kr \xrightarrow[33\,Secs.]{} {}^{90}Rb \xrightarrow[2.7m.]{} {}^{90}Sr \xrightarrow[27.7y.]{} {}^{90}Y \xrightarrow[64h.]{} {}^{90}Zr \text{ STABLE}$$

FISSION PRODUCTS of MASS 137 :-

$$^{137}I \xrightarrow[19\,Secs.]{} {}^{137}Xe \xrightarrow[3.4m.]{} {}^{137}Cs \xrightarrow[27y.]{} {}^{137m}Ba \xrightarrow[2.6m.]{} {}^{137}Ba \text{ STABLE.}$$

This similarity would suggest that the distribution of the nuclides will be similar in the bomb debris,(7) as would the fact that both isotopes have gaseous precursors and are formed relatively long after the bomb detonation. This accounts for their not being found in large proportions in the large particles which fall out locally, but rather being in the stratospheric store.

Martell(8) states that strontium reaches the stratosphere, in the main, as its precursor rubidium 90, which in common with other alkaline metals has relatively greater volatility and so condenses late. This is borne out by the evidence that strontium 90/total fission product ratio increases with decreasing particle size, and that the strontium 90 associated with large particles is concentrated in a thin surface layer.

Table 1 shows fission abundance of hazardous nuclides and it can be seen that caesium 137 and strontium 90 are those which combine relatively high fission abundance with long radiological half-life and high absorption on ingestion, and

TABLE 1

Potentially hazardous radionuclides in distant fallout from nuclear detonations (6)

RADIOELEMENT	TYPE of RADIATION	FISSION ABUNDANCE %	RADIOLOGICAL HALF-LIFE	ABSORPTION ON INGESTION %	M.P.L μC
Pu.-239	α	-	24,000 y.	3×10^{-3}	0.037
Cs-137	β γ	6.2	26.6 y	100	54
Sr-90, Y-90	β	5.1	27.7 y	30	1
Pm-147	β	2.6	2.6 y	1×10^{-2}	60
Ru, Rh-106	β γ	0.5	1.0 y	3	3
Ce, Pr-144	β γ	5.3	285 d.	1×10^{-2}	5
Zr-95	β γ	6.4	65 d.	1×10^{-2}	26
Y-91	β	5.9	58 d.	1×10^{-2}	5
Sr-89	β	4.6	51 d.	30	4
Nb-95	β γ	6.4	35 d.	1×10^{-2}	76
Ba-La-140	β γ	6.0	12.8 d.	5	4
I-131	β γ	2.8	8 d.	100	0.7

these two can be considered to be biologically most important.

Caesium, however, although it emits both β and γ radiation can be regarded as contributing to Man's general background radiation because chemically it is similar to potassium and so does not lodge in any particular organ of the body. Strontium, on the other hand, is an alkaline earth and so tends to follow the metabolic pattern of calcium. Therefore when strontium is ingested it tends to enter the skeleton replacing calcium ions in the hydroxyapatite crystal system. This means that strontium 90 is the only fall-out isotope which combines high fission yield, long half-life (27.7 years),[5] high absorption rate and low maximum permissible level.[6]

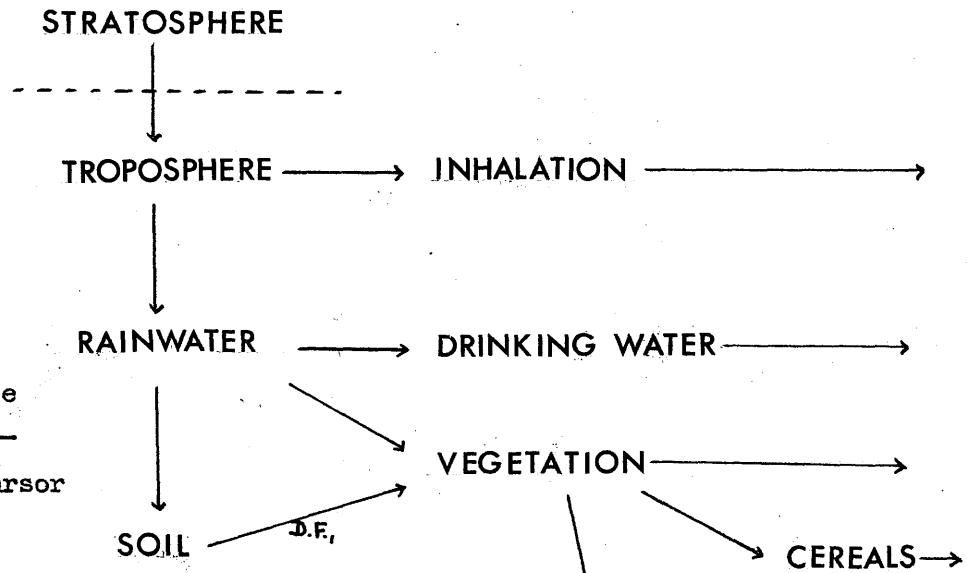
All these factors, therefore, make strontium the most important fall-out element biologically.

(b) The pathway of strontium to the body

Fall-out materials, whether tropospheric or stratospheric, eventually reach the lower atmosphere where they are washed by rainwater on to the earth's surface. There they may fall on plant leaves and be absorbed directly (strontium 90 unlike the other common fall-out radioisotope caesium 137 is mostly in a soluble form, 60 - 70% soluble, and so is readily absorbed by plants[9]), or else go into the soil and eventually reach the

FIGURE 2

STRONTIUM 90 PATHWAY TO THE BODY



$$D.F. = \frac{\text{Sr/Ca sample}}{\text{Sr/Ca precursor}}$$

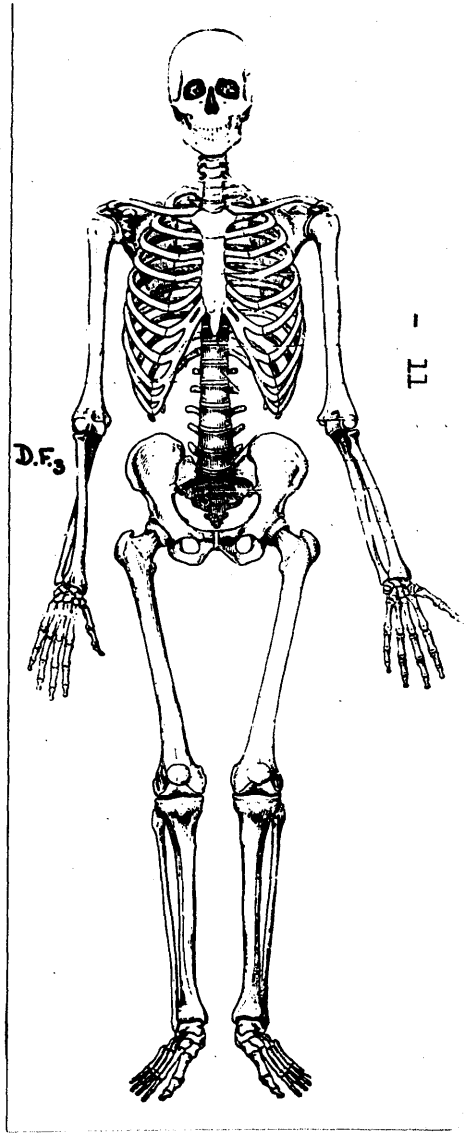
$D.F._1 = 0.7$

$D.F._2 = 0.125$

$D.F._3 = 0.25$

Total D.F. (bone/soil) $= 0.6 \times 0.7 \times 0.125 \times 0.25 + 0.4 \times 0.7 \times 0.25$

$= 0.08$ (where British diet is 60% milk derived Ca and Sr)



plant via root absorption. It has been shown that radiostrontium only penetrates the top few inches of soil, 80% being in the top two inches. Therefore, almost all fall-out radiostrontium is available for plant uptake. In the case of direct leaf absorption, the amount absorbed depends only on the level of activity at the time concerned, but in the case of radiostrontium absorbed via the roots from the soil, the depth of roots (the shallower the roots, the greater the activity available) and the calcium content of the soil (in the case of low calcium-containing soils, more strontium can be taken up by the plant) are factors which also govern the strontium activity in the plants. Therefore, strontium taken up by foliar absorption is undiluted by calcium whereas that by root absorption is diluted to a variable degree.

Vegetation may contribute directly to the human dietary intake of strontium 90 and also indirectly. Domestic animals live largely on vegetable material and so meat and eggs are contaminated by fall-out materials, as is milk. Milk and milk products contribute 56%[10] of the average persons dietary calcium in the U.K. and, as would be expected, they contribute almost 60% of the intake of strontium 90.[11] Fortunately, the cow discriminates in favour of calcium and against strontium in its secretion of milk so that the activity derived from milk and milk products is much less than would at first be

expected. The discrimination factor seems to favour calcium by 7 - 10.[10][12][13][14][15]

This factor is of particular importance to the diet of babies and young children whose calcium (and therefore strontium) is derived almost entirely from milk.

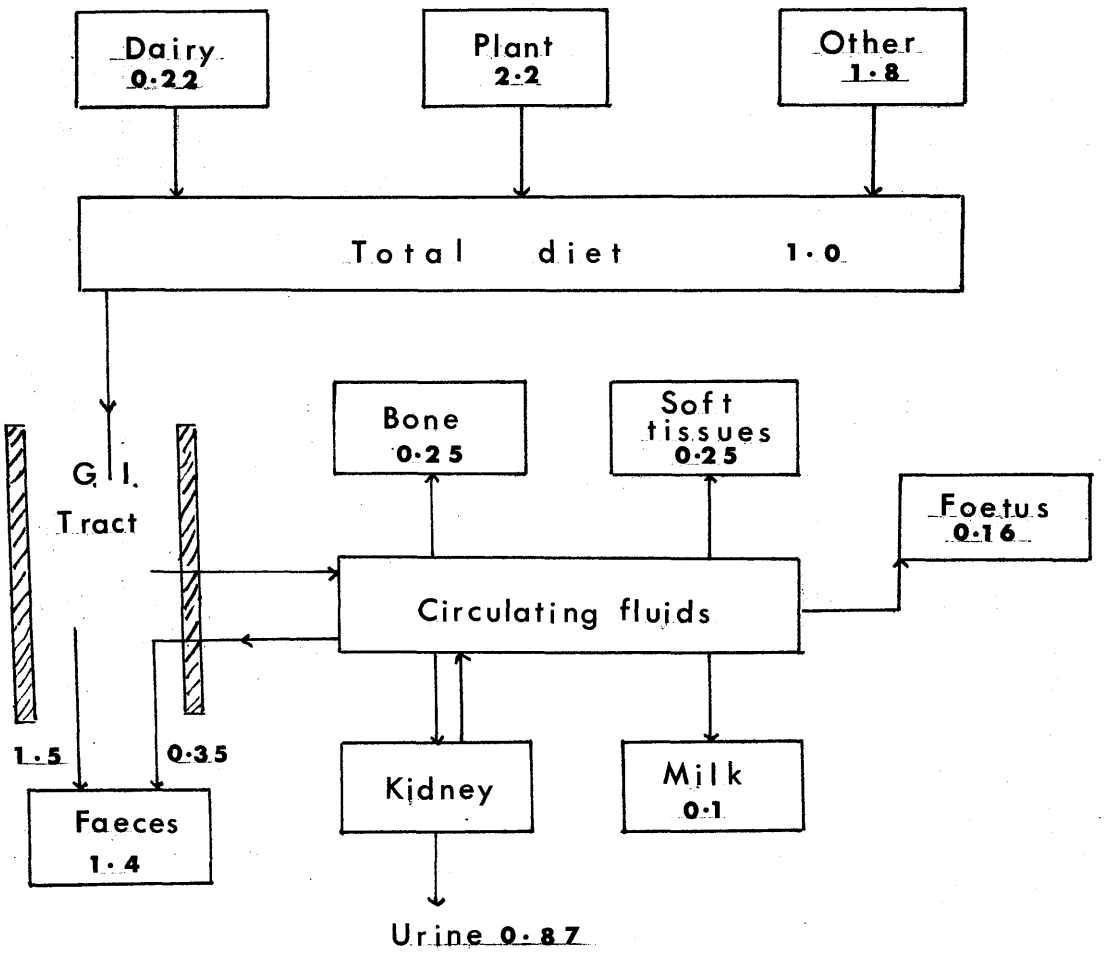
It should be noted that breast fed babies have a further protection as there is a similar discrimination in favour of calcium in human milk.[16]

In Eastern 'rice-eating' countries this safeguard will not exist as almost all the dietary calcium is derived from vegetables. Therefore it would be expected that strontium 90/ calcium ratios would be higher in human tissues in such countries.

A similar situation was reported in Canada in 1963. Eskimos in sub-Arctic regions were said to have higher strontium 90 levels due to the large proportion of caribou meat in their diet. Caribou in turn feed on lichens and as these have large flat leaves they are said to be ideal fall-out collectors. The bones of Eskimos are stated to have 0.5 μμc/g. Ca on average compared with 0.3 μμc/g. Ca in the rest of North America.[17]

Although calcium and strontium are both readily absorbed from the gastrointestinal tract, it seems that calcium is preferentially absorbed by a further factor of 4[18][19] and as newly deposited bone appears to be in equilibrium with the body

FIGURE 3



Typical ratios of stable Sr/Ca in diet, tissues, and excretions of Adult Man[51]
(Values normalised to a Sr/Ca value of 1 in total diet based on English diet with mineral Ca fortified bread)

fluids the ratio of strontium 90 to calcium in new bone and plasma is about one quarter of that in diet. In the case of new-born children there is an additional protection against strontium 90 as calcium seems to pass along the placenta more readily than strontium; for it is known that the strontium to calcium ratio in the foetus is about half of the ratio in the maternal blood level.

All these factors mean that at birth an infant will have a strontium 90/calcium ratio in its bone tissue which is about one eighth of the bone ratio in its mother's diet. The child's strontium 90/calcium ratio will rapidly increase as new bone is laid down and the existing bone is exchanged for new which has a strontium 90/calcium ratio about one quarter of the child's dietary ratio - the dietary ratio is highly variable depending on whether it is fed on mother's milk or cow's or dried milk. Bryant and Loutit report that an infant's prenatal bone is almost completely exchanged for new bone salt in the first year of its life, and then the turnover rate drops to an average of 10% per year between the third and eighth years and to 2.5% per year in the adult.(20)(21)

(c) The hazards of fall-out

Man has always been submitted to radiations for although this has only relatively recently been realised, radioactive elements have always been part of the earth and cosmic radiations are an intrinsic part of the Universe. Man has, however, by his own invention added to his radiation exposure - by the use of radiation in medicine and industry and by the production of radioactive isotopes in nuclear reactors and in the explosion of nuclear weapons.

When radiation is absorbed by body tissues it can cause either the death of the tissue cells and if this exceeds the rate of replacement of the cells then permanent damage occurs. Alternatively, it may cause genetic change. When the altered cells divide then the effect may be transferred to later generations of cells, or to descendants of the individual.

Radiation can be divided into four main types :-

(i) Alpha radiation which consists of high energy nuclei having low powers of penetration, < 0.1 mm in human tissue.

(ii) Beta radiation or electrons of varying energies and penetrating up to a few millimetres in soft tissue.

(iii) Gamma rays and x-rays, also of various energies and usually far-penetrating, therefore irradiating the whole body.

(iv) Neutrons which are uncharged and of wide range in energy and penetration power.

As radiation associated with a nuclear explosion is varied depending on the power and the distance of irradiated material from the explosion, then the dangers of fall-out are also various. In addition, biological tissues have differing sensitivities to radiation depending on the type of tissue, and on the age of the cells making up the tissue - immature cells being more sensitive than mature cells. Fall-out at the present time is below the level known to cause any measurable damage, and so that the degree of damage caused by low levels of radioactivity can only be based on inference. This leads to doubt as to whether such low levels do cause any damage; one train of thought is that a 'threshold' exists, below which radiation has no effect. But it is perhaps safer to assume that all radiation has damage potential and so should be avoided if possible. The damage caused by low levels is however difficult to assess and can only be inferred by work with experimental animals at higher levels. It is obviously not permissible to do experimental work on human guinea pigs.

As was previously noted, the radioisotopes of most biological significance in fall-out are iodine 131, calcium 137, strontium 89 and strontium 90.

Iodine 131 is only important immediately after an atomic burst or nuclear reactor accident (it should be remembered that although the term 'fall-out' is being generally used and is

usually associated with bomb explosions, most of the discussion is equally applicable to reactor accidents, so that any information accumulated from nuclear weapon testing might be of value if such cases arise) for it has a half-life of eight days and in the main, decays before it reaches ground level. It is mainly concentrated in the thyroid and several measurements have been made on the human thyroid during periods of fresh fall-out.(22) These give values of about 6 m. rads in the four week period December 1958 - January 1959, with other workers reporting up to ten times this figure. This is well within the permissible dose laid down by the International Commission on Radiological Protection (I.C.R.P.) of 1,000 m. rads/year.

Caesium 137, although having a physical half-life of 27 years behaves in the body as does potassium, therefore it has fairly rapid turnover giving it a biological life of about four months. Its chances of disintegrating within the body are therefore slight. But when it does disintegrate, it produces a radioactive daughter, barium 137 which almost immediately decays with the emission of γ rays. Because of its uniform distribution throughout the body, and therefore general irradiation, caesium 137 can be thought of as slightly increasing the 'natural background' radiation by about 1 - 2 m. rads per year (i.e. by about 1% of the gonad dose rate).

Strontium, however, we have already stated, behaves biologically in a fashion similar to calcium, i.e. the strontium absorbed from the gastrointestinal tract localises almost entirely in the skeleton. Strontium 89 has a relatively short half-life (51 days) and so although almost as abundant in fall-out as strontium 90 it is not considered so dangerous. Strontium 90 emits β radiation of low penetration power (0.8 mm in soft tissue and 1.4 mm in bone and ranges of 4.3 mm in soft tissue and 7.8 mm in bone tissue for the β radiation from the daughter nuclide yttrium 90),[23] so its genetic hazard is considered to be non-existent or slight. The main danger therefore is to the bone marrow, where blood is formed and irradiation might cause a higher incidence of leukaemia, or to the bone itself - for bone irradiation might result in bone sarcoma formation (this has been shown in experiments where animals have been given high dosage of radiostrontium but there is not any direct evidence of damage due to fall-out).

Maximum permissible levels of strontium 90 in bone were specified in 1960 by the Medical Research Council in their report 'Hazards to Man of Nuclear and Allied Radiations' as 200 pc. Sr 90/g. Ca for individual members of the population and 67 pc. Sr 90/g. Ca for the average over the whole population. These concentrations would give radiation dose rates of 540 m. rem and 180 m. rem/year respectively (the average bone dose rate

from natural background radiation is estimated at about 130 m. rem/year). As will be seen, levels to date do not even approach these concentrations.

SECTION II

ASPECTS OF STRONTIUM METABOLISM UNDER CONSIDERATION

ASPECTS OF STRONTIUM METABOLISM UNDER CONSIDERATION

(a) Strontium 90 in bone - linked with the activity in diet

In the study of strontium 90 and of its metabolism by children, the most logical starting point seems to be the establishment of levels of the isotope in bone tissue. This makes a good basis both from the point of view of elucidating the metabolic pattern and from that of devising methods of analysis. For these reasons methods for analysing bone for strontium 90 and summaries of results have been included. These results have been used in an attempt to link up the strontium 90 content in infants' bone with the levels of the isotope in diet. A simple relationship between diet and bone would be useful in estimating general trends quickly and without the laborious assay of post mortem bone specimens. Such a relationship would, however, be of no use in determining any particular child's bone strontium 90 concentration.

Although the Agricultural Research Council have published the results of measurements of strontium 90 in foodstuffs in the United Kingdom[24] and have estimated the radioactivity in the average mixed diet, no such information is available for the 'average mixed diet' in Scotland. As the newborn child derives its skeletal strontium and calcium from the diet of its mother during pregnancy some approximation to the strontium 90

activity in Scottish diet was required. This has been calculated from the Agricultural Research Council's figures for average mixed diet and U.K. and Scottish milks. As the ratio of mixed diet/U.K. milk is fairly constant the calculated figures seem to be reasonably good approximations. The calculation for the first half of 1964 is supported by results from several estimations on mixed diet.

In addition to these calculations on strontium 90 content in diet, and diet/bone ratios based on averages, an attempt has been made to study the effect of variation in diet of mothers during pregnancy on their children (where the children have died within two weeks of birth and post mortem investigations were carried out). A survey has been carried out of the diet of these mothers, and the relative amounts of those foods contributing most calcium to the total intake were noted. Milk and milk products, meat, vegetables and eggs were noted as being relatively high sources of strontium 90 together with calcium, while bread and other flour products, together with calcium tablets, contain mineral calcium which is uncontaminated with radiostrontium and so would be expected to reduce strontium 90 levels. A survey such as this could be extended to older children and adults but would be more difficult and perhaps less profitable than with 'stillbirths' whose bone tissue is of most uniform derivation and is laid down during the narrowest time

interval therefore most likely to be at some equilibrium with, in this case maternal, diet.

(b) Strontium 90 excretion compared with intake

So far, most of the analyses have been related to post mortem investigations. These give no direct evidence of the living child's handling of strontium and calcium (although much can be inferred from such studies[20]). In order to examine this, some methods must be available for the determination of strontium in food material and in excreta. Adaptations of the bone analysis procedure have been devised for the analysis of diet, faeces and urine. These adaptations are included in the appendices. Milk has been analysed with satisfactory results, by a method of Spicer.[25]

(c) Methods of attempting to increase strontium output relative to intake

When methods have been established for the measurement of intake and output of strontium 90, experiments can be undertaken in an attempt to reduce bone deposition of the isotope. Although such measures need not be taken at the moment, strontium 90 activity being well below the maximum permissible level, it would be useful to have the information at hand before it is required. There are two main methods of approach to this

problem. One is to reduce uptake of the isotope in the gastrointestinal tract, the second is to increase the renal clearance[26] of the absorbed strontium.

The first is, on the face of it, easiest to tackle. The simplest way of reducing bodily uptake is to remove the isotope from the intake altogether. This is highly feasible if the food product is in liquid form, as has been described by several authors. Drinking water and milk can be passed through an ion-exchange column which will remove the alkaline earths.[27] This means, of course, that calcium as well as strontium will be removed by this process, and as milk is the main source of dietary calcium, it must be replaced by adding mineral calcium to the treated milk. Another interesting means of removing strontium from milk has been described by Singer and Armstrong.[28] This method consists of passing milk through a column of bovine bone which has been modified by a preparatory treatment. This is reported to be 75% successful in removing strontium with no loss of calcium, but the flow rate of 10 ml./minute would hardly make the process practical on a commercial scale.

With solid foodstuffs, however, the problem is not so easily surmounted. Strontium cannot be removed without destruction of the food. Therefore some mechanism must be found to reduce strontium absorption in the gastrointestinal tract. Several authors have reported a dilution technique to be

effective. Wasserman, Comar, and Papadopoulou[29] found that dilution of the strontium with a calcium enriched diet reduced strontium uptake in growing rats. Palmer and Thomson[30] in their in vivo experiments on rats, decreased the amounts of radiostrontium and radiocalcium absorbed, although the radiostrontium/radiocalcium ratio was increased. In general, however, large increases in calcium are required for relatively small decreases in strontium uptake.[31]

An experiment of this kind has been attempted on several hospital bound children between the ages of 4 and 13 years. During the period of the experiment - approximately four weeks - the children were given a diet which was fairly varied although of reasonably uniform (approximately 1 gram per day) calcium content. After an equilibration period of about one week, complete collections of all urine and faeces are made for at least six days. During this time an exact duplicate of the child's food is also kept. The procedure is then repeated giving the child, in addition to its diet as before, an extra gram of calcium daily. With the first two 'balance' subjects this calcium was as B.P. calcium carbonate suspended in a syrup to make it pleasant to take. The other 'volunteers' were given calcium monohydrogen phosphate B.P.C., as the phosphate is thought to be more readily absorbed in the gastrointestinal tract than the carbonate which tends to neutralise the gastric juices and

so reduce their powers of digestion. The treatment of the samples collected during the two periods is described later.

Although this seems, in theory, to be a relatively simple exercise to measure strontium and calcium intake and output during two periods of six days, in practice, it did not prove to be. Most of the difficulties were linked to the choice of sick children as 'guinea pigs'. Their metabolic pattern is upset during the experiment and it is difficult to ensure that faecal specimens correspond exactly to the metabolic period under consideration. A carmine marker was used for several of the collections but it was not thought to be very accurate (the practice was, in general, to start and end a balance period at the midnight following the passage of a stool).

In addition to increasing calcium intake in order to increase its uptake at the expense of strontium, Widdowson, McCance, Harrison and Sutton have found that phosphate supplements give similar effect - calcium metabolising more readily when phosphate levels are high.(32)(33) The alternative to reducing gastrointestinal uptake of strontium in order to prevent strontium 90 being laid down in bone, is to increase renal clearance of the isotope. Spencer et al.(34) have reported the effectiveness of ammonium chloride (in conjunction with intravenously infused calcium) in the removal of radiostrontium from man, and Clark and Smith have used magnesium ions when

working on rats.(35) The individual's calcium metabolism comes into play in all these experiments so that reduction of strontium uptake will be of variable effectiveness.

In addition to measurements of strontium 90 content in bone, diet and excreta, some measurements have been made on teeth. Although no individual is likely to shed enough teeth to enable samples to be analysed on an individual basis, deciduous teeth of people of similar ages can be pooled. Such samples should be a good indication of average levels of strontium 90 in teeth.

SECTION III

REVIEW OF ANALYTICAL TECHNIQUES

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Before the effect of present day fall-out can be determined some estimation of its content in foodstuffs and in biological material must be made. Although in this case strontium 90 has been chosen as the isotope for investigation, much of the chemistry is equally applicable to strontium 89.

In expressing levels of radioactive strontium two criteria are available. One is to express the radioactive material relative to the stable isotopes, i.e. specific activity which is the usual method of reporting radioactivity. In the case of strontium however, the chemistry and metabolism are so close to those of calcium that most investigators of strontium 90 have expressed their results relative to calcium. Whatever way is chosen accurate methods of determining stable strontium and calcium must be found, as well as techniques for the separation of the radioactive isotope.

(a) Radiochemical techniques

As strontium is a trace element in most materials, the main object in its determination is to separate it from the elements making up the bulk of the sample. This is particularly important with the isotope strontium 90, for, although it is generally possible to estimate radioisotopes in the presence of

the predominating 'impurities', strontium 90 is a weak β -ray emitter - its radiation having a maximum penetration of a few millimetres. This means that strontium 90 must be concentrated into small bulk so that its radiation is not absorbed before it reaches the detecting unit or counter.

Three main methods of separation of radiostrontium from foodstuffs and biological materials are available. These are :-

i. an ion exchange technique.(36)(37)

ii. partition between solvents.(38)

iii. a chemical separation procedure.(39)

The ion exchange technique is particularly useful in the case of liquid samples such as milk and rain water. The sample is mixed with the resin, a cation exchange resin, and the strontium eluted with suitable agents.(36) The method can also be applied to solid samples, if these are ashed first and the ash taken up in a suitable solvent.(40)

Extraction by one solvent from another has also been reported.(38)(41) This involves preferential extraction of equilibrium yttrium 90 by tri- or di-butyl phosphate from nitric acid solution of bone ash. Although the method is speedier than others, it requires fairly active samples as yttrium 90 is short lived and so should be counted quickly (which is impossible at one or two counts per minute levels).

The method which has been used here is the chemical

separation technique. This depends for its success on the differential solubilities of the alkaline earths in nitric acid.(39) The main advantage of this separation procedure is that with some variation of initial treatment, it can be applied to almost any type of sample with any level of activity. As most of the work has been on very low levels of activity, this is important. The main disadvantage is the length of each estimation which has many steps.

The procedure for the estimation of strontium 90 is based on a method for the separation of strontium and barium from calcium by their precipitation as nitrates. Willard and Goodspeed report that the variation of nitric acid concentration has marked effect on the solubilities of these salts. At 80% acid concentration barium and strontium nitrates are completely insoluble whereas calcium is soluble. Bryant, Morgan, and Spicer(42) have used this property in their method for the 'Determination of Radiostrontium in Biological Materials'. In addition to this process which combines the concentration of strontium into small bulk with the elimination of calcium, phosphate and other impurities, additional scavenges are carried out for barium and iron. Barium is removed by chromate precipitation in an acetate buffered solution, and iron is precipitated as hydroxide. Finally, the strontium is precipitated as carbonate and mounted on an aluminium planchet.

As this method has been used for all samples in which fall-out strontium 90 was to be determined, it has been laid out in full in the appendices. Modifications, usually in the form of pretreatment, for samples other than bone ash are also detailed.

Assay of the radioactive content of the samples has been made with G.M.4 LB. counters in anti-coincidence shield units.(43)(44) Further details of the counting is given in Appendix IV.

(b) Estimation of stable strontium and calcium

As strontium is exceeded by calcium by a factor of $10^3 - 5 \times 10^3$ in most of the samples to be analysed, some very sensitive method of analysis must be used. Of the two techniques available, neutron activation analysis and flame photometry, the first has not been of use here because of the lack (until recently) of an atomic reactor. This method involves the inducing of radioactivity in the element to be determined by submitting it to a suitable flux of neutrons. After a suitable activation period the sample is removed and with a minimum of chemical treatment the radioactivity measured. By comparison with known standards treated in a similar way, the concentration of the element in the sample can be calculated. Sowden and Stitch(45) have used activation analysis successfully

for the estimation of strontium in biological materials but it has been rejected here in favour of flame photometry.

This method of analysis which has been used for both stable strontium and calcium determination, is based on emission spectrometry. When salt solutions are sprayed into a flame, the molecules of the salt are dissociated and the resultant atoms may absorb energy from the flame to put them into an excited state, i.e. to displace one of the atom's electrons into a higher energy state. This energy may then be released as light which is of wave lengths characteristic to the element in question. Therefore by choosing a suitable instrument to measure the emitted light this property is a useful means of the estimation of elements.

Early workers on flame photometry were Talbot[46] who, in the early part of the 19th century, studied lithium and strontium, and Kirchoff and Bunsen who discovered caesium and rubidium in 1860 and 1861 respectively. Later workers realised the possibility of using flame spectra quantitatively, but it was not until Lundegårdh[47] devised a suitable means of introducing solutions into the flame, that this became a practical proposition. In more recent years great advances have been made in this direction. Details of the instrumentation and methods of analysis are in Appendix II

SECTION IV

RESULTS AND CONCLUSIONS

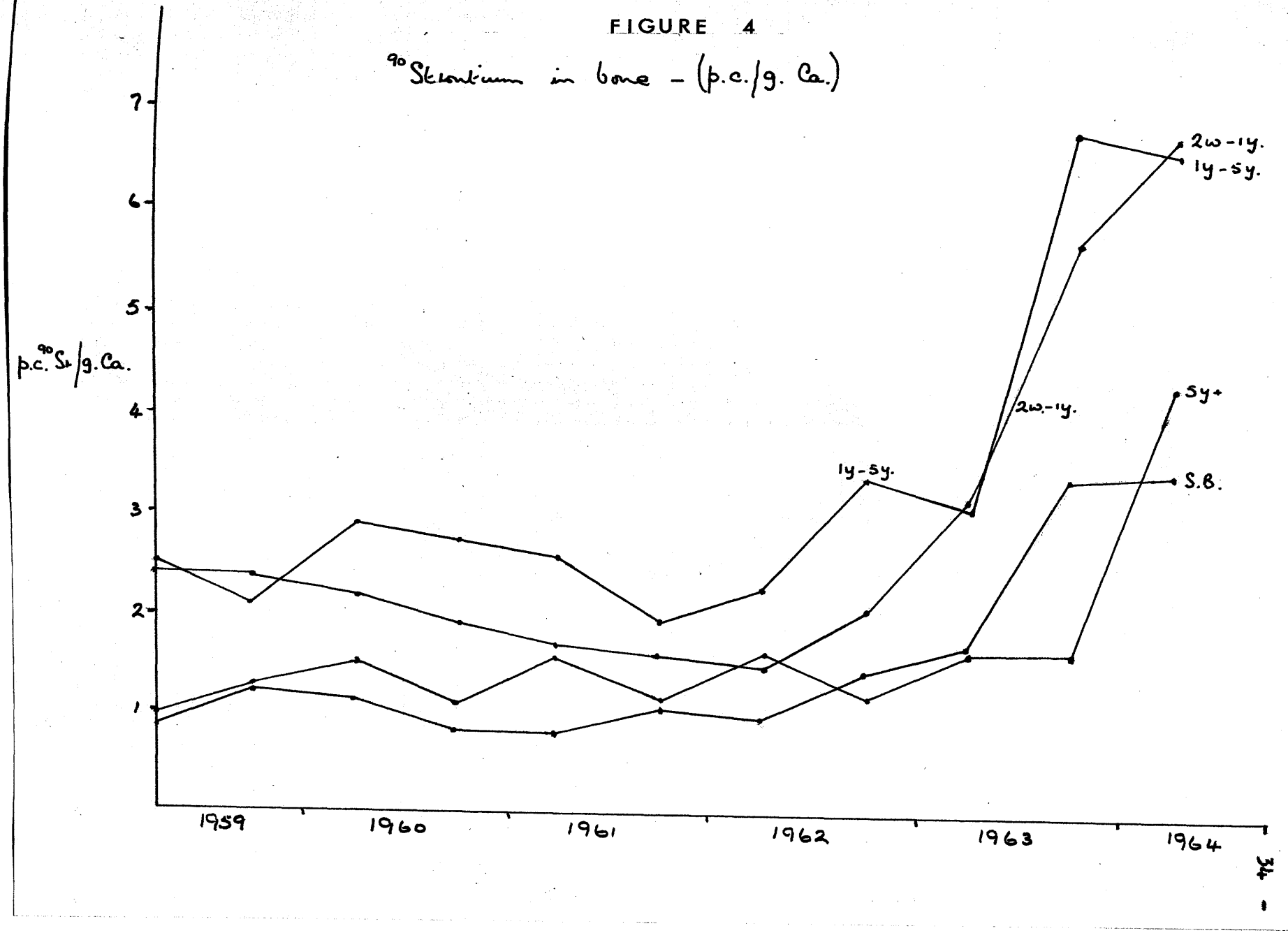
TABLE 2

^{90}Strontium in human bone (Glasgow)

Age group	1959		1960		1961		1962		1963		1964
	JAN. - JUNE	JULY - DEC.	JAN. - JUNE	JULY - DEC.	JAN. - JUNE	JULY - DEC.	JAN. - JUNE	JULY - DEC.	JAN. - JUNE	JULY - DEC	JAN. - JUNE
Stillbirths	0·82	1·2	1·11	0·8	0·79	1·04	0·89	1·4	1·65	3·37	3·42
2w. - 1y.	2·4	2·36	2·16	1·85	1·65	1·55	1·43	2·02	3·09	5·68	6·74
1y - 5y	2·45	2·1	2·9	2·73	2·53	1·8	2·23	3·35	3·02	6·78	6·56
5y +	0·97	1·25	1·5	1·03	1·5	1·07	1·57	1·15	1·6	1·6	4·28

UNITS - pc. | g. Ca

FIGURE 4


90Strontium in bone - (p.c./g. Ca.)
p.c. 90Sr/g.Ca.
7
6
5
4
3
2
1
2w-1y.
1y-5y.
5y+
S.B.
1959
1960
1961
1962
1963
1964
Yrs

RESULTS AND CONCLUSIONS

(a) i. Strontium 90 in human bone[48] (Figure 4, Table 2)

Since the second half of 1962 there has been a rise in the mean strontium 90 concentrations. This results from the effects of nuclear weapon tests in 1961 and 1962. The rise follows a period of fluctuating levels with a slight net decrease from 1959-62 (although this report does not cover 1959-62 these bone averages have been included for comparison). The increases are most marked in the groups 2 weeks - 1 year and 1 year - 5 years. The age group, >5 years, shows a rather delayed rise - it does not become obvious until the first half of 1964. This delay is however to be expected as the rate of turnover of calcium and strontium decreases with increasing age. The 'stillbirth' group show a slight increase in 1962 with the steepest rise in July - December 1963. By 1964 this group are showing signs of levelling off again so the other groups would be expected to level off although the > 5 years group should take longest to do so. The stillbirths, although showing a steep rise in strontium 90 content in 1963, are the 'age group' with the lowest increase over the whole period mid-1962 - mid-1964. This reflects the additional protection the foetus has from fall-out in the 'discrimination factor' across the placenta.

TABLE 3

CALCIUM AND STRONTIUM CONTENT OF 10 BONE-ASH SAMPLES.

P.M No.	AGE of SUBJECT	Ca % of ash	Sr. µg/g.Ca.	^{90}Sr µc./g Ca	Standard error of COUNTING
10491	1 7/12 y.	38.63	151	7.4	0.07
10403	1 10/12 y.	38.01	284	6.6	0.3
10506	3y.	38.1	171	-	-
10394	4 2/12 y.	38.31	176	2.3	0.1
10284	4 1/2 y.	38.08	273	1.9	0.07
10451	4 10/12 y.	37.7	250	3.0	0.1
10467	5y.	38.24	207	3.5	0.1
10482	5 5/12 y.	38.03	267	5.4	0.08
10315	7y.	37.51	241	2.1	0.15
10281	9 8/12 y.	38.38	208	1.8	0.05

Au. 38.1 Au. 223.

ii. Calcium and stable strontium in bone

In general, bone samples have been too small to allow routine analyses for calcium and stable strontium. Table 3 shows results obtained for ten samples where sufficient material was available after the assay of strontium 90.

Calcium in bone ash is relatively uniform, regardless of age, although there is a slight increase with age. The ten samples analysed give an average of 38.1% calcium in bone ash. This is not very different from the value of 38% which has been used in the calculation of strontium 90/g. calcium in bone, where no actual analysis of calcium has been made.

Stable strontium however has a wider range - from 151 - 284 ug/g. calcium. Although the tendency is for strontium content to increase with age[49] there is a wide scatter in each age group. The average for the ten samples analysed is 223 ug/g. calcium.

(b) Strontium 90 in Scottish diet

This was done by simple proportion, using the Agricultural Research Council's figures for National diet, National milk and Scottish milk. The ratio of Sr 90/Ca in average diet/Sr 90 in milk was fairly constant and so average Scottish diet was expected to be in a similar ratio to Scottish milk.

TABLE 4

	1962	1963	1964 (year ending June)
U.K. Diet	9.9 pc/g. Ca	22.8	29.3*
U.K. Milk (average)	11.7	25.6	33.7
Scottish Milk	16.8	33.4	43.3
Scottish Diet (calculated)	14.2*	29.7*	37.6*

* estimated figure

The figures calculated have been tabulated (table 4), and are used in the comparisons with bone and with urine in an attempt to discover something of the body's handling of strontium with respect to calcium.

(c) i. Comparison of strontium 90 in diet with that in bone

The ratio of strontium 90 per gram of calcium diet/bone has been calculated using the Agricultural Research Council's figures for diet[50] and both the Atomic Energy Research Establishment's and our Scottish results for bone.[48] Table 5 shows this diet/bone ratio for three age groups during the period 1959 - June 1964.

During 1959-62 the ratios are fairly stable within each group. This reflects a period of little fresh fall-out and the ratios should be a reasonable indication of the body's discrimination in favour of calcium and against strontium. In the older age groups, the figures are higher than in the 0-5 years group due to lower bone turnover rates. This disparity increases with increasing 'fresh' fall-out although the ratios in all age groups increase with fresh fall-out as there is a certain time lag between strontium 90 reaching food and attaining an equilibrium with bone tissue.

As the Agricultural Research Council's figures for strontium 90 in an average mixed diet were not thought to be representative

TABLE 5

RATIOS — $\frac{^{90}Sr.\ p.c./g.\ Ca.\ Diet}{^{90}Sr.\ p.c./g.\ Ca.\ one}$

		1959	1960	1961	1962	1963	1964
A.R.C. diet A.E.R.E. bone	S.B.	7.8	7.45	8.6	9.9	11.7	-
	0-5y.	2.95	2.2	3.2	4.45	5.05	-
	5-20y.	10.0	5.8	5.9	8.8	14.9	-
A.R.C. diet Glasgow bone	S.B.	8.2	6.4	6.95	9.4	8.6	-
	0-5y.	3.6	3.05	3.55	5.5	5.35	-
	5-20y.	11.25	5.3	4.8	5.8	14.3	-
Scottish diet Glasgow bone	S.B.	-	-	-	13.5	11.4	11.0
	0-5y.	-	-	-	7.9	7.0	5.6
	5-20y.	-	-	-	8.3	18.6	8.8

of a Scottish diet, an estimation has been made (table 4) which is thought to be a better approximation of local activities in food. These figures have been used in the last three lines of table 5 but in 1962-64 there is no equilibrium between diet and bone due to the period of 'fresh' fall-out.

These ratios support the discrimination factor of 4 between diet and bone (the 0-5 years age group show a ratio of between 3 and 4 in the years 1959-62. This group are all post 1953 and so are more likely than older groups to be in a state of equilibrium with diet.) The ratio is approximately 8 in the stillbirth group. As the bone mineral of these children is laid down 'in utero', this confirms an additional factor of 2 across the placenta.

ii. Strontium 90 levels in the bones of neonates with respect to maternal dietary habits

As the neonates were the group with least variables - only the mother's diet need be considered as the strontium 90 levels do not increase much in the first few days of life - this group was first to be considered and is tabulated and summarised.

TABLE 6

Average values of Sr 90/g. Ca in bones of neonates whose mothers' diet fell into the following categories (during 1963)

	High	Average	Low	Average S.U. for stillbirths
Bread	2.3	-	2.2	
Brassica	2.5	2.4	1.85	
Milk Products	2.6	-	1.95	2.24
High Milk Prods. + High Brassica	2.4	-	1.95	

It was thought that bread containing mineral calcium (with no fall-out strontium) would, if eaten in large amounts, have a depressing effect on the strontium 90 levels. There is, however, no significant difference between the children of 'large-small bread-eating' mothers. Apart from this, the differences are very much as would be expected, on average, with higher levels of strontium 90 in the bones of children of high consumers of milk products and green vegetables. The levels have been averaged for 1963 only as there are insufficient numbers in 1962 and 1964.

(d) Strontium 90 in teeth

As an individual is unlikely to provide sufficient material for estimation at any one time, samples must be composite ones drawn from people of the same age at approximately the same time. Results therefore are likely to resemble average bone levels.

TABLE 7

Sample No.	Habitat of Donors	Age	Time of Sampling	Sr. pc/g. Ca.	Stable Sr.
T1	Norway - within Arctic circle	5-6 y.	Late 1962	2.35 ± 0.04	-
T2	Glasgow	2-3 y.	Sept. 1964	2.86 ± 0.04	237 ug/g. Ca.

Although T1 compares with British bone samples of the same age group, T2 is rather lower than would be expected. Bone averages for this age group in 1964 are nearer 4-5 pc/g. Ca. The stable strontium content of 237 ug/g. Ca is similar to the Atomic Energy Research Establishment's average value of 274 ug Sr/g. Ca for the bones of children in the 2-3 years age group.

The rather lower than expected strontium 90 activity in sample T2 is accounted for by the fact that teeth are not in a state of continual mineral exchange with plasma as is bone. This means that when dietary strontium 90 activity is rising, as it was when T2 were collected, the activity in teeth will lag behind that in bone.

(e) Stable strontium and strontium 90 in samples of children's diet

Aliquots of food samples prepared for stable strontium and calcium determination in the 'balance' experiments were assayed for strontium 90 in addition. Results for these estimations are tabulated (table 8) and average for six samples examined during the period October 1963 to March 1964 is 36.9 pc Sr 90/g. Ca. This figure approximates well with that calculated for Scottish diet from figures given for the National diet and Scottish milk averages. The calculated figure for the year ending June 1964

TABLE 8

STABLE - Sr AND ^{90}Sr IN DIET.

PATIENT	DATE of SAMPLING	AGE of PATIENT	^{90}Sr. p.c./g Ca	Stable Sr. µg/g Ca	^{90}Sr / Sr p.c./mg.
B.M.	10/63	11y.	44.8	1000	44.8
E.M^{c}I.	11/63	12y.	36.8	1010	36.4
B.C.	11/63	9y.	32.1	-	-
J.M^{c}D.	12/63	13y.	37.0	995	37.2
K.M^{c}G.	1/64	6½y.	38.1	715	53.1
L.C.	3/64	9½y.	32.8	780	42.1

Av. 900

is 37.6 pc/g. Ca.

Stable strontium in the diet samples averages 900 ug/g. Ca. This is approximately four times the stable strontium per gram of calcium in the average of ten bone samples analysed (see table 3). This would seem to confirm a factor of 4 which favours the deposition of calcium rather than strontium in the bone, i.e.

$$D.F. \quad \frac{\text{Sr/Ca Diet}}{\text{Sr/Ca Bone}} = 4$$

(f) Strontium 90 in urine

The results are tabulated (table 9).

The average for 1962 is 15.4 pc/g. Ca (four samples) which is somewhat higher than the average National dietary level for 1962 (9.9 pc/g. Ca) or the calculated level for Scotland (14.2 pc/g. Ca), but as these samples are all from the latter half of the year when levels were rising generally, this is perhaps not so surprising.

In 1963 the average level of strontium 90 in children's urine is 22.5 pc/g. Ca (30 samples). This figure, when compared with the calculated Scottish diet Sr 90/Ca figure of 29.7 pc/g. Ca, gives a ratio urine/diet of 0.76 which compares with Comar's ratio of 0.87[51] but is rather higher than the ratio reported

TABLE 9

Sample No.	Date of Sampling	Age of Subject	Volume in litres	Calcium mg/l.	Sr 90 pc/l.	Sr 90 pc/g. Ca
GU. 11	7/62	7 y.	1.5	40.0	1.12	28.1 ± 5.7
12	8/62	12 y.	1.5	105.0	1.75	16.6 ± 2.0
13	8/62	pooled	1.5	95.0	0.37	3.9 ± 0.5
14	8/62	12 y.	1.5	87.3	1.13	12.9 ± 1.5
15	3/63	pooled	1.5	70.0	1.45	20.7 ± 2.0
16	3/63	4 y.	1.5	83.0	1.64	19.7 ± 1.5
17	3/63	pooled	1.5	95.0	0.84	8.8 ± 1.0
18	3/63	"	1.5	80.0	1.31	16.4 ± 2.8
19	4/63	11 y.	1.4	202.0	1.76	8.7 ± 0.7
20	4/63	pooled	1.5	64.0	1.50	23.3 ± 2.5
21	4/63	10 y.	1.5	38.0	0.45	12.0 ± 3.2
22	4/63	12½ y.	1.5	12.7	0.375	29.5 ± 1.5
23	4/63	pooled	1.5	20.0	0.47	23.4 ± 8.0
24	4/63	"	1.5	95.0	1.64	17.3 ± 1.2
25	4/63	"	1.5	110.5	1.58	14.3 ± 1.0
26	5/63	"	1.5	66.0	0.72	11.0 ± 2.7
27	5/63	12 y.	1.5	38.0	0.057	1.5 ± 1.5
28	5/63	pooled	1.2	75.0	1.13	15.0 ± 15.0
29	5/63	"	1.5	130.0	0.95	7.3 ± 1.2
30	9/63	7 y.	1.5	27.4	1.08	39.5 ± 5.4

Sample No.	Date of Sampling	Age of Subject	Volume in litres	Calcium mg/l.	Sr 90 pc/l.	Sr 90 pc/g. Ca
GU 32	9/63	7 y.	1.5	149.5	4.25	28.5 ± 1.1
33	9/63	7 y.	1.5	149.5	1.21	8.1 ± 1.2
34	9/63	13 y.	1.5	96.0	2.2	23.5 ± 1.0
35	9/63	13 y.	1.5	96.0	1.9	20.0 ± 2.8
36	9/63	7 y.	1.5	34.8	1.73	49.6 ± 5.6
37	9/63	7 y.	1.5	34.8	1.67	48.0 ± 4.6
38	9/63	6½ y.	1.5	294.0	2.8	9.5 ± 0.6
39	9/63	6½ y.	1.5	294.0	2.1	7.2 ± 0.5
40	9/63	8½ y.	1.5	155.0	1.4	9.1 ± 0.9
41	9/63	8½ y.	1.5	155.0	1.63	10.5 ± 0.65
42	10/63	9½ y.	1.5	160.0	5.7	35.5 ± 1.0
43	10/63	6½ y.	1.5	57.5	4.4	76.4 ± 2.7
44	10/63	10 y.	1.5	149.0	3.6	24.2 ± 1.2
45	10/63	pooled	1.5	94.5	5.9	62.3 ± 2.2
46	11/63	9 y.	1.5	261.0	4.13	15.8 ± 0.7
47	11/63	12 y.	1.5	468.0	7.35	15.7 ± 0.4
48	11/63	9 y.	1.5	165.0	5.15	31.2 ± 1.1
49	11/63	12 y.	1.5	242.0	2.55	10.5 ± 0.6
50	1/64	1 m.	0.17	212.0	8.8	41.5 ± 10.0
51	2/64	3 w.	0.195	58.0	1.9	32.8 ± 2.6
52	2/64	2 m.	0.4	76.0	4.4	57.8 ± 9.4
53	2/64	8½ y.	1.5	33.7	0.98	29.1 ± 3.4

Sample No.	Date of Sampling	Age of Subject	Volume in litres	Calcium mg/l.	Sr 90 pc/l.	Sr 90 pc/g. Ca
GU 54	3/64	$10\frac{1}{2}$ y.	1.5	586.0	5.3	9.1 ± 0.35
55	1/64	6 y.	1.5	19.0	0.4	21.4 ± 8.0
56	1/64	9 y.	1.5	135.0	4.85	36.1 ± 1.7
57	4/64	$10\frac{1}{2}$ y.	1.5	307.0	4.2	13.7 ± 0.5
58	4/64	$4\frac{3}{4}$ y.	1.5	46.2	1.015	22.0 ± 2.4
59	3/64	$9\frac{1}{2}$ y.	1.5	277.0	4.05	14.6 ± 0.6
60	5/64	7 y.	1.5	104.5	0.23	2.2 ± 0.5

by Harrison et al.[52] and Schulert in his 'Assessment of Dietary Strontium 90 through Urine Assays.'[53] These authors report a urine/diet ratio of 0.5.

The average of eleven assays in 1964 is 25.5 pc Sr 90/g. Ca. Unfortunately no National diet figures are yet available so the calculated Scottish average of 37.6 pc/g. Ca is arrived at with less certainty than that for 1963 although experimental evidence is fairly corroborative (two diet samples assayed in 1964 give an average of 35.5 pc/g. Ca). These figures give a urine/diet ratio of 0.70 which is lower than the 1963 ratio but still higher than the Harrison and Schulert value.

TABLE 10

Patient	Age in years	Date of Sampling	S.U.* in Diet	S.U.* in Urine	O.R. Urine/ Diet
BM	11	10/63	44.8	31.2	0.7
EMcI	12	11/63	36.8	15.7	0.43
BC	9	11/63	32.1	15.8	0.49
JMcD	13	12/63	37.0	10.6	0.29
KMcG	$6\frac{5}{12}$	1/64	38.1	21.4	0.56
LC	$9\frac{1}{2}$	3/64	32.8	14.6	0.44
U51	3 w.	2/64	34.5	32.8	
U52	2 m.	2/64	56.5	57.8	

* S.U. = pc. Sr 90/g. Ca

In a few cases urine strontium 90 has been measured in patients whose dietary intake has also been measured. These results are tabulated (table 10), and as can be seen patients on mixed diet in the 6-13 years of age group have urine levels much below the dietary level. The average dietary intake is 36.9 pc/g. Ca compared with a urine level of 18.2 pc/g. Ca on average. This gives a diet/urine ratio of approximately 2. This therefore seems to agree with the Harrison figure but numbers of samples are not sufficient for conclusive evidence.

Two estimations have been made on the milk intake and urinary output of infants of 3 weeks and 2 months (table 10). In both cases the urinary strontium 90 level approximates to the milk strontium 90/calcium ratio. This seems to suggest that in infants, the discrimination between calcium and strontium is not so marked. (It was suggested possible that the discrimination against strontium might not be so marked in the very young in their Report to the Medical Research Council by the Committee on Protection against Ionising Radiations.[54])

(g) Strontium 90 excreted in the faeces compared with the dietary intake

Results are given for strontium 90 per gram of calcium in the diet and faeces of five patients, during periods of normal and high calcium intake.

TABLE 11

Patient	Date of Sampling	Age	High or Normal Calcium	Diet Sr 90 pc/g. Ca	Faeces Sr 90 pc/g. Ca	Faeces/ Diet
BM	10/63	11 y.	N	44.8	35.2	0.785
			H	25.6	20.0	0.78
JMcD	12/63	13 y.	N	37.0	32.8	0.89
			H	14.8	15.8	1.07
KMcG	1/64	$6^{5}/12$ y.	N	38.1	34.9	0.92
			H	18.4	21.1	1.15
AD	4/64	$10\frac{1}{2}$ y.	N	33.3	22.8	0.68
			H	17.6	12.3	0.71
SC	4/64	$4\frac{3}{4}$ y.	N	44.0	26.2	0.59
			H	19.0	12.1	0.64

The additional calcium is given in the form of mineral calcium orthophosphate and is effective in diluting the radiostrontium. It does not however, reduce the total radiostrontium in diet as the calcium is given as a supplement and does not replace calcium from a 'natural' dietary source.

The ratio Sr 90 pc/g. Ca in faeces/Sr 90 pc/g. Ca in diet for each period has been included in the table and in each case, except the first where the ratio does not alter, there is an increase in faecal output relative to intake, from the 'normal'

to 'high calcium' periods. This would seem to indicate that 'high calcium' intake does increase excretion of radiostrontium although the increase is small compared with the approximate doubling of the calcium intake.

Table 12 shows the same results expressed as activity of strontium 90 per day. These absolute figures permit the calculation of the 'absorption'. In all cases except the first, this is reduced by the increase of dietary calcium. The negative

TABLE 12

Patient	High or Normal Calcium	Diet pc Sr 90 /day	Faeces pc Sr 90 /day	% Absorption $\frac{D-F}{D} \times 100$	Faeces/ Diet
BM	N	41.6	28.2	32.2	0.68
	H	50.5	30.3	40.0	0.60
JMcD	N	32.4	27.0	16.7	0.83
	H	25.7	28.6	-11.3	1.11
KMcG	N	44.7	23.8	46.7	0.53
	H	39.3	27.3	30.5	0.70
AD	N	24.3	21.6	11.1	0.89
	H	31.3	32.6	-4.15	1.04
SC	N	33.8	21.9	35.3	0.65
	H	33.3	23.4	29.7	0.72

nature of the 'absorption' in two cases (especially JMcD) is due to the inaccuracies of the metabolic experiment. It is hardly conceivable that growing children would be in negative balance. For this reason the concentration of strontium 90 relative to calcium is of more reliability than the absolute activity, although the second set of figures confirms the first to some degree.

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APPENDICES

Appendix I

ANALYTICAL PROCEDURES FOR DETERMINATION OF RADIOSTRONTIUM

Preliminary treatment of samples

Bone is dried in an air oven at approximately 110°C for several days before ashing in a muffle furnace at 700-750°C. Large femora are sectioned and as much fat is removed as is possible before ashing. The ash is ground well in a mortar to ensure a homogeneous powder from which aliquots may be taken for strontium 90, stable strontium and calcium assays.

Teeth require no previous treatment unless they have any amalgam filling. It is desirable that this is removed.

Food specimens analysed are, in the main, composite diet samples (the only exceptions are a few milk samples which require no preliminary treatment). These samples should be homogenised, dried at 110° and ashed at 500-550°C. The ashing temperature should be lower than that for bone samples as diet contains silicon which would fuse at the higher temperature and so make the sample more difficult to deal with at later stages. The lower temperature means that the ashing may take several days, whereas a bone specimen takes only a few hours for a small one

up to a maximum of about 24 hours for that from an adult. The ashed diet is thoroughly ground in a mortar to ensure a homogeneous ash.

Faecal specimens are dried in a porcelain crucible on a hot-plate before grinding in a coffee mill and ashing under the same conditions as food samples. Again the ash is well ground with a mortar and pestle.

Urine is collected in a bottle containing a few crystals of thymol as preservative. Unless the sample is to be analysed immediately it is acidified to ensure the calcium remains in solution. Urine, on standing, becomes progressively more alkaline with time due to the breakdown of some of its constituents. As it becomes more alkaline, calcium tends to precipitate out and so erroneous results will be obtained with urine which is not processed when fresh, unless it is well acidified. Hydrochloric acid is used.

Chemical treatment for strontium 90 determination

(a) Bone samples(42)

In addition to the usual reagents (of analytical grade always) the following special reagents were used :-

Fuming nitric acid 95%

Dilute acetic acid 6 M.

Ammonium acetate 3 M

Sodium chromate 1.5 M

Strontium carrier solution ($Sr(NO_3)_2$)
5 mg Sr/ml. standardised

Yttrium carrier solution ($Y(NO_3)_3$). 6 H_2O
10 mg Y/ml. standardised

Barium carrier solution ($Ba(NO_3)_2$) 10 mg Ba/ml.

Ferric iron carrier solution ($Fe(NO_3)_3$). 9 H_2O
5 mg Fe/ml.

Elga deionised water is used throughout.

Procedure: As much bone ash as is available (up to a maximum of about 50 grams) is weighed out into a 250 ml. centrifuge bottle. About one gram should be left, if possible, for calcium and stable strontium determinations but most of the samples were less than 3 grams or so, and in these cases all the ash was used for radiostrontium assay and an average calcium value (38%) was used.

To the bottle is added 5 ml. of strontium carrier solution, 1 ml. of barium carrier solution and (a) for sample weights less than 5 grams 34 ml. of water and 50 ml. of fuming nitric acid to dissolve the bone ash. When solution is complete a further 50 ml. of acid is added. (b) For sample weights greater than 5 grams 45 ml. of water and 60 ml. of acid is used to dissolve the bone and a further 60 ml. of acid.

In both cases the bottles are cooled in running water for 30 minutes with occasional stirring to precipitate the barium

and strontium. (The acid concentration at this stage is approximately 80% at which concentration strontium is insoluble but calcium soluble. Barium is insoluble in 76% nitric acid and calcium solubility decreases rapidly after 80%.)[39][55] By allowing the sample to stand for 30 minutes, cooling is effected (the mixing of acid and water involves a considerable rise in temperature) so precipitation is increased as well as allowing some of the precipitated calcium to redissolve.[39]

After this, the supernate is removed by centrifuging and decanting off the acid. The acid separation is repeated on the residue using (a) 40 ml. water to dissolve the residue and 90 ml. 95% nitric acid to reprecipitate the strontium (b) 50 ml. water and 113 ml. acid. (In the majority of assays these acid separations will be sufficient to remove the calcium, but with larger samples the last stage should be repeated until the residue is equal in bulk to that of the 'blank' sample which is assayed with each batch.)

The residue is transferred to a 40 ml. centrifuge tube using about 30 ml. of water, the solution made alkaline with 0.88 ammonia solution, solid ammonium carbonate added, and the tube heated in a boiling water bath to coagulate the strontium carbonate precipitate. After centrifuging, the supernate is rejected and two further acid separations carried out on the residue. Each uses 10 ml. of water and 22.5 ml. fuming nitric acid.

The nitrate residue is dissolved in 10 - 15 ml. water, 1 ml. barium carrier added and the solution neutralised with 6 M ammonium hydroxide using methyl red indicator. The neutral solution is buffered with 1 ml. 6 M acetic acid and 2 ml. 3 M ammonium acetate, diluted to about 30 ml. (to reduce loss of strontium by occlusion with barium chromate) and heated on a boiling water bath. 1 ml. of 1.5 M sodium chromate is added and heating continued for 5 minutes to precipitate the barium chromate.(55) The tube is centrifuged and the supernate transferred to a second tube. This stage, as well as removing barium, is effective in precipitating radium and its radioactive daughters which may contaminate the sample.

The solution is made alkaline with 0.88 NH_3 solution and solid ammonium carbonate added to precipitate the strontium as carbonate. (These carbonate precipitations serve to reduce the bulk of the sample.) The precipitate is coagulated by heating in a boiling water bath then the tube is centrifuged and the supernate rejected.

The residue is dissolved in 6 M nitric acid and one drop of 100 volumes hydrogen peroxide added to reduce the chromium to the trivalent state so that it can be precipitated as hydroxide in the next stage. 1 ml. of iron carrier is added and the solution heated in a boiling water bath to drive off the carbon dioxide. It is diluted to 15 - 20 ml. and made alkaline

with 'carbonate free' ammonium hydroxide (in practice this is 'AnalaR' 0.88 NH_3 freshly drawn from the Winchester bottle so that it has not been in contact with the atmosphere). Heating is continued to complete the precipitation of ferric hydroxide which is then removed by centrifuging - it is usually necessary to filter the supernate at this stage to remove the last traces of precipitate.

Ammonium carbonate is added to the supernate to precipitate the strontium as carbonate and after heating on a water bath for a few minutes to complete the precipitation, the solution is filtered through a Whatman No. 42 filter paper of known weight. The precipitate is washed twice with water and three times with methanol before being allowed to dry for two hours, and weighed.

Finally the paper with precipitate is transferred to an aluminium planchet and a few drops of 3% Gelva (polyvinylacetate) solution added to keep the precipitate intact.

The source is counted after storing for at least 14 days to allow the strontium 90 to come to equilibrium with its daughter yttrium 90. When there is a lapse of more than one year between the detonation of a nuclear weapon and analyses of samples, one assay of about 3000 counts is sufficient for the determination of strontium 90 but if an explosion is more recent than this then at least two counts are necessary with a time interval of 6 - 8 weeks between. By doing this a correction

for any strontium 89 can be made.

A more conclusive way of determining only strontium 90, free from any other radioisotopes, is to precipitate the yttrium daughter. This can be done by adding yttrium carrier to the sample and allowing the supernate from the ferric hydroxide scavenge to stand for 14 days to allow the strontium 90/ yttrium 90 to come to equilibrium before milking off the daughter isotope as hydroxide, filtering, and mounting for counting. If yttrium is to be counted, counters must be readily available, for yttrium 90 has a half-life of only 64 hours and so must be counted immediately. Also the time of precipitation must be noted so that a correction for decay can be applied. Because of the large number of samples to be determined it has been found more practicable to count the strontium 90 in equilibrium with its daughter because any delay in counting is not then important.

(b) Teeth samples

An attempt was made to assay teeth samples in the same way as bone, but this was not successful. The main difficulty was in obtaining a homogeneous ash, as the hardness of the enamel made the ashed teeth impossible to be ground down. A similar difficulty was reported by Bryant, Henderson and Holgate.(56)

The sample was weighed into a volumetric flask

and after strontium and barium carriers had been added it was dissolved in dilute nitric acid and made up to a known volume. An aliquot was removed for calcium and stable strontium determinations, then the remainder was transferred to centrifuge bottles where the nitric acid concentration was increased to precipitate the strontium and, for the first separation, most of the calcium. If the sample is in more than one bottle the residues are combined using 50 ml. of water, and 113 ml. of fuming nitric acid is added to reprecipitate the strontium. This is repeated as with the bone samples until the bulk is reduced to that of the 'blank' when the procedure is continued in the same way as for bone - removing barium, etc. as chromate and iron as hydroxide before finally precipitating the strontium as carbonate.

(c) i. Composite diet ash

As the ash is of much more varied composition than bone ash it requires rather more chemical treatment, in particular to remove silicates.

10 - 20 grams of the ash are weighed into a 600 ml. beaker and the carrier solutions added as before. 200 ml. water is added together with 40 ml. 95% nitric acid to dissolve the sample. If insoluble material remains, it should be removed by filtration at this stage. The calcium and strontium are

coprecipitated as phosphate by the addition of 1 ml. of 60% orthophosphoric acid and sufficient 0.88 ammonia solution to make the sample alkaline. The phosphate precipitate is removed by centrifuging and then dissolved in 20 ml. 95% nitric acid. The volume(V) of the resultant solution is measured and a further (2.5 V - 40) ml. of nitric acid added to precipitate the strontium. To cool the sample and complete precipitation the bottle is allowed to stand for 30 minutes in running water and the contents stirred frequently. After centrifuging, the supernate is removed and the procedure continued as for bone samples.

ii. Milk samples

A method for the determination of strontium 90 in liquid milk has been devised by Spicer.(25) By using this procedure ashing could be eliminated.

After an aliquot has been removed for calcium determination the sample is measured into a 2 litre beaker and carriers added as before. The beaker is warmed on a hotplate with continuous stirring. 100 ml. of concentrated hydrochloric acid is added and stirring continued for about 5 minutes. This acidification ensures that the calcium and strontium are ionised and so remain in solution and at the same time the protein is precipitated. This, with fat, is removed by adding 50 grams of kieselguhr to the beaker, stirring for 30 seconds, and filtering while warm

through an 18.5 cm. Whatman No. 54 filter paper in a Buchner funnel. The filtrate is transferred to a clean beaker and 20 g. of oxalic acid added. The sample is warmed to dissolve the oxalic acid, before neutralising with ammonium hydroxide solution using methyl red as indicator. The beaker is allowed to stand for 15 - 20 minutes and the supernatant liquid siphoned off. The residual liquid is removed by centrifuging.

30 ml. of water are added to the precipitate, followed by 80 ml. of 95% nitric acid to dissolve the oxalates. When solution is complete a further 40 ml. of acid are added and the bottle cooled in running water or ice-water.

After centrifuging the residue is dissolved in 40 ml. of water and 90 ml. acid added to reprecipitate the nitrates. Thereafter the procedure is continued as bone ash.

(d) Faecal ash

The Bryant et al. procedure for the determination of radiostrontium in vegetable ash was successfully applied to specimens of faecal ash, although the preliminary fusion with sodium carbonate as employed by Spencer-Laszlo, Samachson, Hardy and Rivera in their strontium 90 balances[57] has also been tried. This was not pursued as no better results were obtained.

About 10 grams of ash are weighed out into a platinum

crucible and 5 ml. of strontium carrier solution and 1 ml. of barium carrier solution added, together with 10 ml. of water, 40 ml. of 40% hydrofluoric acid and 30 ml. 60% perchloric acid. The mixture is evaporated, in a fume cupboard, until almost dry. The crucible is cooled and a further 30 ml. of the perchloric acid added and the evaporation repeated - this time until all the white fumes are driven off. The residue, after cooling, is dissolved in water with the addition of about 5 ml. of 11 M, [redacted] hydrochloric acid. The solution is transferred to a 600 ml. beaker, the volume made up to about 250 ml. and boiled. After cooling and filtering, strontium is precipitated as phosphate by the addition of about 5 ml. of orthophosphoric acid and sufficient 0.88 ammonia to make the filtrate alkaline.

The sample is transferred to a centrifuge bottle and the precipitate washed with 100 ml. of a solution containing 3 ml. of phosphoric acid and 20 ml. of 0.88 ammonia to every 2 litres of solution. After centrifuging the precipitate is dissolved in 20 ml. of 95% nitric acid and the volume(V) of solution measured. A further (2.5 V - 40) ml. of nitric acid are added and the centrifuge bottle cooled in running water for 30 minutes. The method is continued as for bone samples.

(e) Urine samples

Additional special reagents :-

Oxalate solution (made by dissolving 25 g. oxalic acid, 25 g. ammonium oxalate and 50 ml. glacial acetic acid in 600 ml. of water at 40-50°C, and after cooling the volume is made up to 750 ml.)[58]

Calcium chloride solution - 0.1 g $CaCl_2$/ml.
(This is necessary to act as carrier in the initial precipitation as the calcium content in urine is very low.)

Procedure: 1.5 litres (or more if available) of the sample are measured into a 2 litre beaker and 5 ml. strontium carrier, 5 ml. barium carrier, and 10 ml. of calcium carrier solutions added. Slowly, with stirring is added 150 ml. (or $^1/_{10}$th of the volume if a larger sample of urine is assayed) oxalate solution. The beaker is stirred well then covered with a clock glass before allowing to stand overnight. If the sample is acidic then it may be necessary to add 0.88 ammonia to precipitate the oxalates.

The supernate is removed partly by siphoning and more completely by centrifuging before the precipitate is dissolved in a mixture of 40 ml. water and 50 ml. 95% nitric acid. When solution is complete the strontium is precipitated by adding a further 50 ml. of acid. The procedure is then continued as for bone samples.

Appendix II

DETERMINATION OF CALCIUM

(i) Bone samples by gravimetric method

About 0.5 g. of the ash is weighed accurately into a 250 ml. beaker and 40 ml. water and 5 ml. concentrated hydrochloric acid added. When solution is complete (if there is any insoluble material it may be removed at this stage by filtration) 40 ml. 8% oxalic acid is added and the sample neutralised to pH 4 with ammonia, using bromocresol green as indicator. The beaker is heated on a boiling water bath for 3 - 4 hours and the contents filtered through a sintered glass crucible. The precipitate is washed with distilled water and methanol before drying in a hot air oven for about 30 minutes. The crucible is reweighed with the dried calcium oxalate monohydrate and the percentage calcium content of the bone ash calculated.

As bone ash is, if well ashed, fairly pure calcium phosphate this gravimetric method is feasible and accurate but for other specimens the impurities and interfering materials are too great so the flame spectrophotometric method has been used.

This is of course also applicable to bone ash.

(ii) Flame photometric method(59)

The flame photometer consists of five main parts - atomiser, burner, an optical system to pick out the required wavelengths, a photomultiplier or other photosensitive detector, and a recording instrument such as a galvanometer.

The instrument used here is a Zeiss PMQ II spectrophotometer with a flame attachment. This has an integral type of burner, that is the atomiser and burner are such that the sample is sprayed directly into the flame - which is oxy-acetylene.

Calcium presents several problems in flame photometry. Firstly, a very hot flame is necessary to activate the calcium atoms. This must be at least the temperature of an air/acetylene flame and preferably that of oxy-acetylene. Secondly, various interference effects are encountered. Although calcium emits light of several wavelengths, there is an overlapping of the bands with emissions from other elements. The most satisfactory wavelength for the measurement of calcium is 422.7 mμ. Calcium emission is depressed by the presence of phosphate and sulphate - the depression due to phosphate is important in biological work and is most marked at the high temperatures required for calcium estimation. Two methods of eliminating this interference effect are available, however. One is to use an internal standard technique, the other is to increase the interference to a maximum by the addition of phosphate solutions and compare

the unknown with standards in a similar 'high phosphate' medium.

A. Reagents and solutions :-

Calcium stock solution: 25 mM calcium chloride is prepared by dissolving 'Spec-pure' calcium carbonate in a minimum of hydrochloric acid and diluting to the appropriate volume with water.

Mixed salt solution A contains 30 mM KCl, 5 mM K_2SO_4, 1.4 M NaCl, 50 mM KH_2PO_4.

Magnesium solution: 8 mM $MgCl_2$.

Phosphate solution: 44.4 mM KH_2PO_4.

Sodium solution: 200 mM NaCl.

Calcium standards ranging from 0.0 - 20.0 parts per million (ppm) are made up by adding 10 ml. of mixed salt solution A, and 10 ml. magnesium solution to an appropriate volume of the calcium stock solution. About 700 ml. of water is added before 50 ml. of perchloric acid (60%) and the volume is made up to one litre.

Diluting and deproteinising solution is prepared by diluting 10 ml. of phosphate solution and 25 ml. of sodium solution to approximately 700 ml. before adding 55.5 ml. of perchloric acid (60%) and finally making up the volume to a litre.

All reagents and stock solutions are stored in polythene bottles.

Preparation of samples

(a) Bone

0.2 grams of ash are weighed accurately and dissolved in a minimum of concentrated HCl. The volume is made up to 500 ml. or 1 litre and the solution shaken. 1 ml. of the resultant solution is diluted 10 or 20 fold with diluting fluid.

(b) Teeth

The aliquot of teeth in acid solution is diluted to give a solution of approximately 100 ppm. This solution is then further diluted 10 or 20 fold with diluting fluid.

(c) i. Diet ash

0.2 grams of ash are dissolved in a minimum of hydrochloric acid and the solution made up to 250 or 500 ml. As this solution is not always absolutely free from insoluble matter it is well shaken and allowed to settle before 1 ml. is withdrawn from the top and diluted 10 or 20 fold with diluting fluid (any solid material might block the burner and this method of avoidance is preferable to filtration).

ii. Milk

As the strontium 90 is determined on the liquid milk to avoid ashing, the calcium aliquot must be evaporated in a silica basin and ashed before diluting for calcium assay.

Milk on heating forms a skin of milk solids and readily

boils over - this is prevented by evaporating with a bunsen burner from above the sample instead of heating the bottom of the basin.

When dry the milk is ashed at 700°C in a muffle furnace. The ash is dissolved in a minimum of concentrated HCl and made up to a suitable volume so that when further diluted with diluting fluid the resultant solution is 5 - 10 ppm of calcium. Filtration of the acid digested ash may be necessary as some siliceous material may be present. If this is so, the apparatus should be well washed out to avoid any losses.

(d) Faecal ash

0.15 - 0.2 g. of ash are made up in a similar manner to diet ash so that the final solution is 5 - 10 ppm of calcium after a 10 or 20 fold dilution with diluting fluid.

(e) Urine

This is diluted directly with diluting fluid - again 10 or 20 fold.

Alternatively the urine, 5 or 10 ml., may be treated with saturated ammonium oxalate solution and a few drops of ammonia to precipitate the calcium as oxalate. The sample is centrifuged and the residue dissolved in hydrochloric acid and made up to the original volume. This solution is diluted as before.

Analytical procedure

The apparatus is set up with the acetylene pressure adjusted to 150 mm. The slit width used is 0.02 mm. and the wavelength is set at 422.7 mμ. The sensitivity is adjusted and using one of the standards the wavelength and position of burner are both adjusted to give maximum readings on the galvanometer. When all these settings have been made, the standards are measured in turn and then the samples are sprayed, each one being bracketed with the standard above and below the unknown in strength. The concentration of each sample is then calculated by interpolation.

B. Another way of avoiding erroneous results due to interference effects is to introduce into the unknown samples an internal standard. In theory, this means that the interference is identical in sample and standard.

The samples are made up as before but in this case about 25 ml. of solution are required. A small volume (0.1 ml.) of the standard (1000 ppm strength) pipetted into a 10 ml. flask and the volume made up with the unknown solution. The emission of the solution with and without the internal standard is measured and off peak (about ± 5 mμ) readings are also noted (these are subtracted from the results to correct for interfering ions). The concentration of the unknown is calculated by

simple proportion

$$X = \frac{(D_1 - D_M) \times 10}{D_2 - D_1} \text{ ppm calcium}$$

where X is the unknown concentration.

D_1 = deflection for unknown solution (X ppm Ca)

D_2 = " " added standard solution (X + 10 ppm Ca)

D_M = mean reading for off-peak measurement

This method is only applicable where there is a linear relationship between galvanometer readings and concentration. If the relationship is not linear then the McIntyre method gives a better approximation. As the instrument used does give a linearity when the standard concentration is plotted against galvanometer reading, either method is suitable.

(iii) Eel titrator method

A third method was attempted for urine samples but was later given up when the Zeiss instrument became available.

Acidified urine is passed through an Amberlite Resin IR-4B (OH) ion exchange column to remove phosphate ions. The urine is then titrated with E.D.T.A. in the presence of ammonium purpurate solution. Ammonium purpurate gives a red colour with calcium ions and turns blue when all the calcium is complexed with the E.D.T.A. The end-point is detected on the galvanometer of the Eel titrator.

Appendix III

DETERMINATION OF STABLE STRONTIUM[60]

Aliquots of bone, diet or faecal ash (in the case of bone or faeces 0.5 g. and diet 2 - 3 g.) are weighed out and put into centrifuge tubes. Strontium 85 of activity about 1500 cpm is added to the samples. The ash is dissolved in about 5 ml. concentrated hydrochloric acid with the addition of 5 - 10 ml. water. When solution is complete (any insoluble material is removed by centrifuging and pouring the supernate into a clean tube) a few ml. of saturated ammonium oxalate solution are added, and the solution made alkaline to pH 8 - 9 with 0.88 NH_3 solution (B.D.H. Universal indicator is used). The tube is centrifuged and the supernate discarded. The precipitate is dissolved in a few ml. of fuming nitric acid, and when solution is complete more nitric acid is added until the nitrates of calcium and strontium begin to precipitate. The tube is cooled in ice-water before centrifuging and pouring off the supernate. The nitrate precipitate is dissolved in a minimum of water and the 95% nitric acid added to reprecipitate the nitrates. The acid supernate is removed as before, and the precipitate dissolved in water and transferred to a 10 ml. graduated tube which is made up to the mark. The percentage recovery is estimated by counting

3 ml. of the solution on a scintillation counter and comparing the count rate with that of a standard made up from the same aliquot of strontium 85 as was added to the samples - diluted to 10 ml. and 3 ml. taken.

Urine is treated in the same way as solid samples with an additional phosphate precipitation to concentrate the calcium and strontium at the beginning. About 1 ml. of syrupy phosphoric acid is added to the urine sample in a beaker and the sample made alkaline with 0.88 ammonia solution. The beaker is warmed gently then after stirring well allowed to stand overnight. The supernatant liquid is siphoned off and the residual liquid removed by centrifuging. The precipitate is transferred to a silica crucible and ashed in a muffle furnace at 500°C. The ash is then taken up as before.

After counting, the 3 ml. aliquot is returned to the 10 ml. tube. The solution is diluted in a 25 ml. volumetric flask to give a concentration of 1 - 5 ppm. of strontium. An aliquot (0.1 ml.) of standard strontium solution (200 ppm) is pipetted into a 10 ml. volumetric flask and the volume made up with the sample solution.

After a suitable 'warming up' period the Zeiss flame spectrophotometer is set at wavelength 460.7 mμ (checked for maximum deflection with a standard solution of 2 ppm. strontium), slit width 0.02 mm. and acetylene pressure 100 mm.

The emission due to the internal standard solution D_2 and the sample solution D_1 are measured in turn and the deflection for these solutions at about 5 mμ off the strontium wavelength is also noted. The mean of these 'off-peak' readings is a measure of the calcium interference effects.

The concentration of strontium in the unknown is then calculated using the formula

$$X = \frac{(D_1 - D_m) \times 2}{D_2 - D_1} \text{ ppm}$$

where X is the unknown concentration of strontium.

D_1 is the deflection of the unknown solution

D_2 " " " " " " "
+ 2 ppm internal standard

D_m is the mean deflection of the solutions measured 'off-peak'

Appendix IV

LOW LEVEL COUNTING

One of the main difficulties in the determination of fission materials is that of obtaining counting devices sufficiently sensitive to discriminate between their low activities, and the comparatively high background radiation. As strontium 90 is a β emitter a Geiger Muller counter must be used. It is desirable that samples with activity of 1 or 2 disintegrations per minute (dpm) be estimated, but natural background may give a count rate of 16 - 20 counts per minute (cpm) on an unshielded G.M. counter and even with heavy shielding it is difficult to reduce this to below 7 cpm.(43)

In order to reduce the background count rate to below 1 cpm, which is necessary for low level measurements of strontium 90, Rowan and Stevenson have devised a counting set-up.(44) This consists of three 'low background' Geiger Muller tubes of type G.M. 4 LB mounted in a cylinder of screening Geiger tubes, G. 26 Pb, with more of these tubes at either end of the cylinder. The whole assembly is enclosed in a 4" thick shield of steel bricks and the samples fed into position in long brass holders. The theory of the anticoincidence set-up is that only radiation from the sample is measured because it is only detected on the

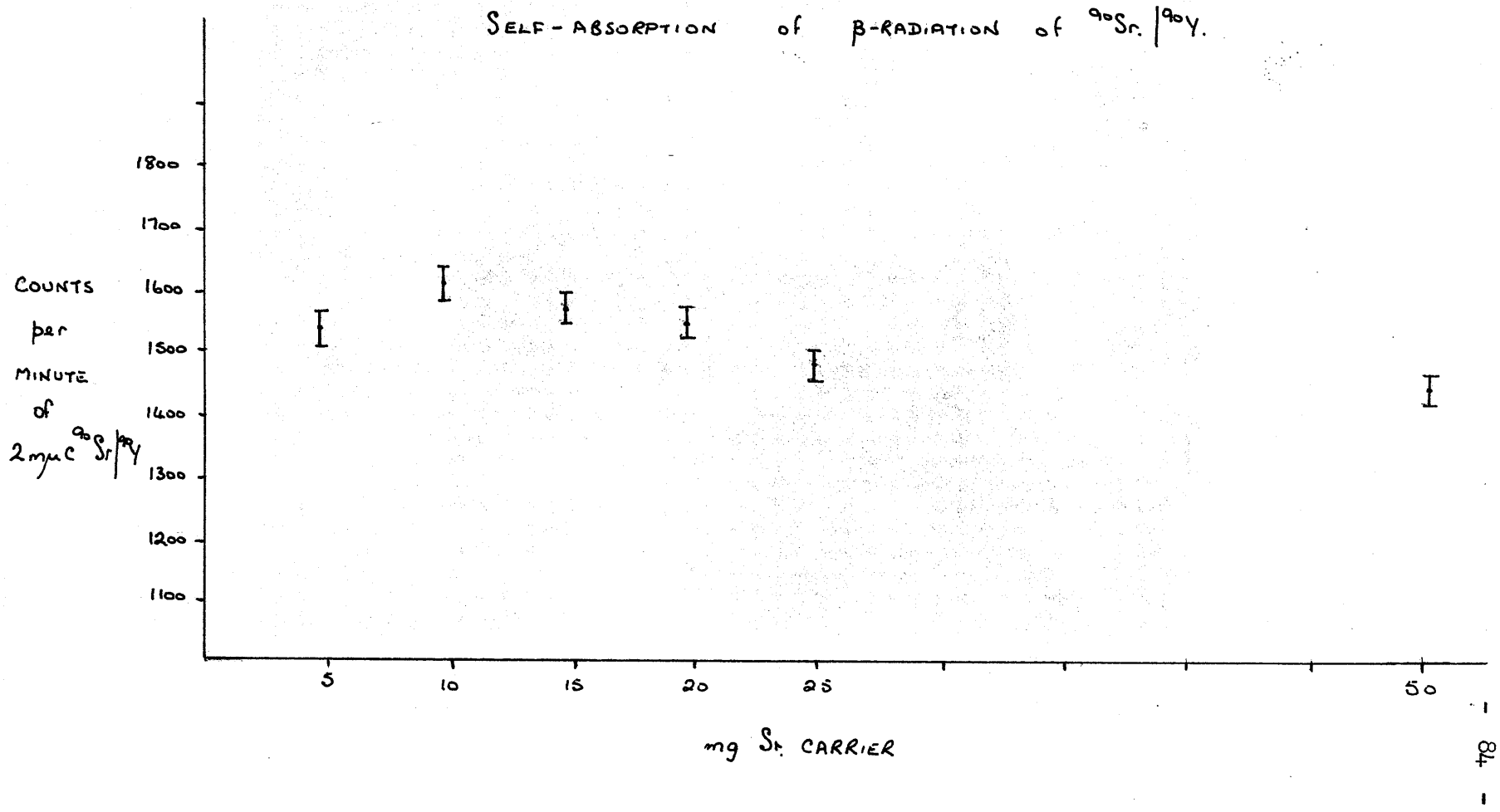
G.M. 4 LB tubes. Radiation from gamma or cosmic rays will trigger off a screening tube as well as a G.M. 4 LB. tube and any such pulses are not registered in the sample count. Although there is no screening between the channels, the sources are pure β emitters so no interaction is possible.

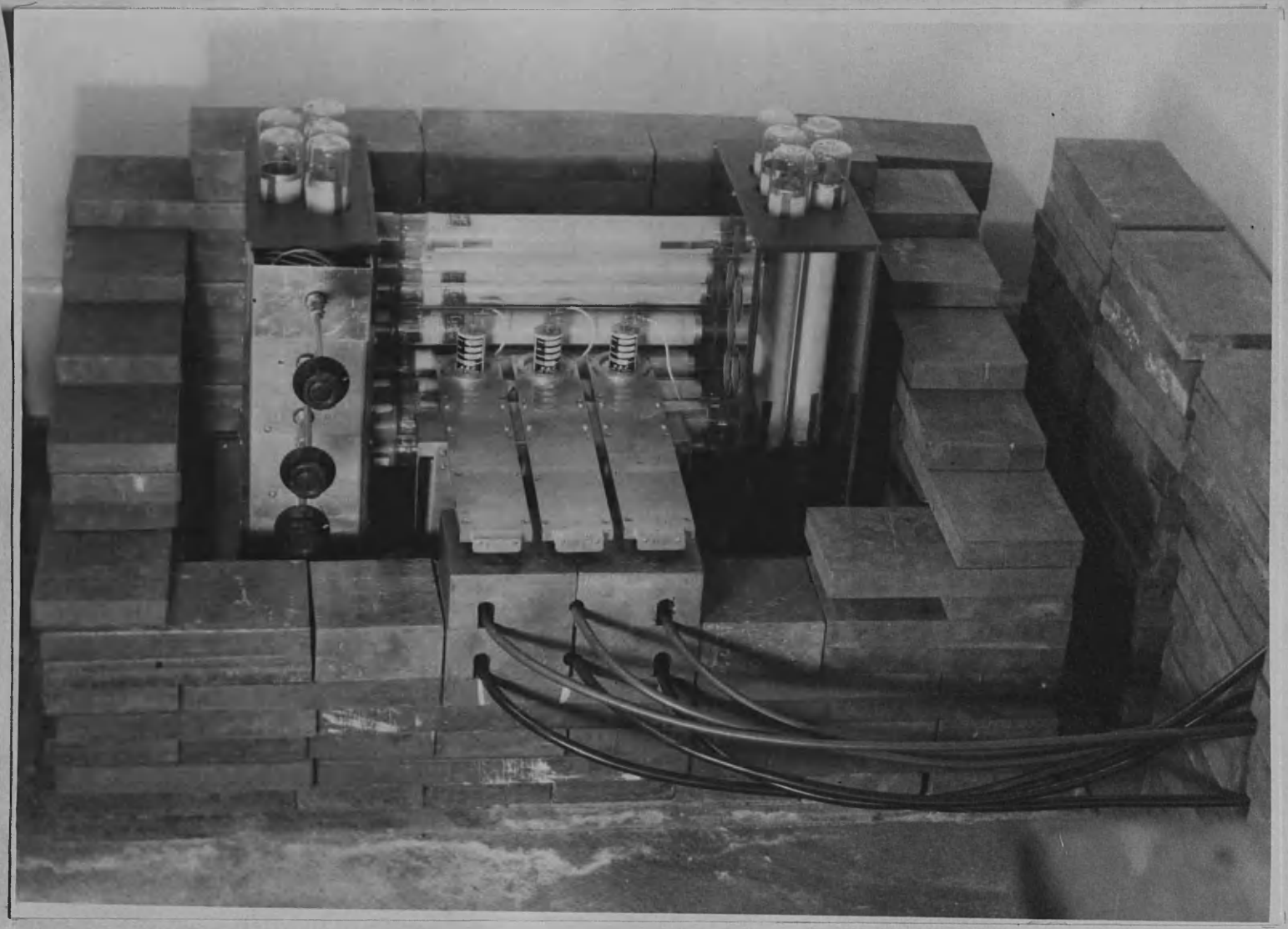
The efficiency of these counting channels is estimated by adding 0.01 mμc. of strontium 90 in equilibrium with its daughter yttrium 90 to a solution containing 25 milligrams of strontium carrier and precipitating it as carbonate for filtering and mounting on an aluminium planchet. After standing for 14 days to ensure equilibrium of the strontium with yttrium, the standard sample is counted on each channel in turn. In general the efficiency is 30-40%. This is checked for each channel every month or so.

As strontium 90 has a low energy β emission it is possible that the varying thicknesses of precipitate in samples, due to the variation in recovery of stable strontium carrier (from one sample to another), introduce an error due to self absorption of these β rays.

This was checked by precipitating 0.01 mμc strontium 90 with a varying amount of strontium carrier and plotting the count rate against thickness of sample. It was found that self absorption is not significant up to about 25 mg. of strontium carrier. Any inaccuracies are within the statistical deviation of the experiment. This is in the region of 10% on average.

FIGURE 5


SELF-ABSORPTION of β-RADIATION of ^{90}Sr | ^{90}Y.
COUNTS per MINUTE of 2 mμc ^{90}Sr | ^{90}Y
1800
1700
1600
1500
1400
1300
1200
1100
5
10
15
20
25
50
mg Sr CARRIER



The partly assembled steel castle and 'guard' ring showing the three counters in position