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Atomic Energy Research Establishment, Harwell

## **A Survey of British Work on Radioactive Fallout**

(covering results up to mid-1957)

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### *Introduction*

Considerable public concern has been expressed over the possible biological effects of the radioactivity deposited upon the earth as a result of the injection into the atmosphere of the fission products from nuclear explosions. Evaluation of hazards from this cause can only be made from a knowledge of the radiation doses being incurred by various tissues of the human body and from an estimation of the future levels of radiation exposure. For this reason, the measurement of the present fission product radioactivity in tissues of the human body and determinations of the levels of external radiation and estimates of the levels to be expected in the future are a pre-requisite of any evaluation.

Measurements of the concentration of fission products in the air over the U.K. were commenced about 1948 and continuous measurements of the radioactivity level in rainfall were instituted at two stations in the U.K. in 1951. The early measurements were mostly concerned with the fission products liberated into the troposphere from nuclear explosions in the "kiloton" range (corresponding in energy release to the explosion of a few thousand tons of T.N.T.). Study of the fission product activity in the lower atmosphere from such explosions shows that, when allowance is made for the decay of the radioactivity the debris disappears from the atmosphere at a rate such that only one-tenth of it is left after approximately 11 weeks (*Stewart et al., 1956*), the material at the greater distances being deposited primarily by wash-out by rain at the surface of the earth.

An entirely different picture emerges in regard to debris from explosions in the "megaton" range. The dust cloud from such explosions

penetrates the tropopause and reaches a considerable height in the stratosphere. Diffusion in the stratosphere is a very slow process and the material returns to the lower layers of the atmosphere at a very slow rate. The mean residence time of debris in the stratospheric reservoir appears to be of the order of 5 to 10 years. The vertical concentration of activity was measured in the U.K. at heights up to 15000 metres in 1954 and 1955 (Stewart et al., 1956) and some further measurements are now being undertaken. The measurements show a marked gradient of increasing fission product activity with height above the tropopause, but the gradient was small below the tropopause, consistent with the much greater turbulence in the lower atmosphere.

With the early measurements it was possible to identify the date of origin of the sample and to calculate the amounts of the long-lived isotopes present. This is no longer a feasible procedure and, accordingly, since 1954, the measurements have been made by direct radiochemical analysis for the particular nuclide in question.

#### *Measurements of the Deposition of $Sr^{90}$ , $Cs^{137}$ and $Sr^{89}$*

The predominant mode of deposition of the fission product activity in the U.K. is by wash-out in rain and, accordingly, the monthly depo-

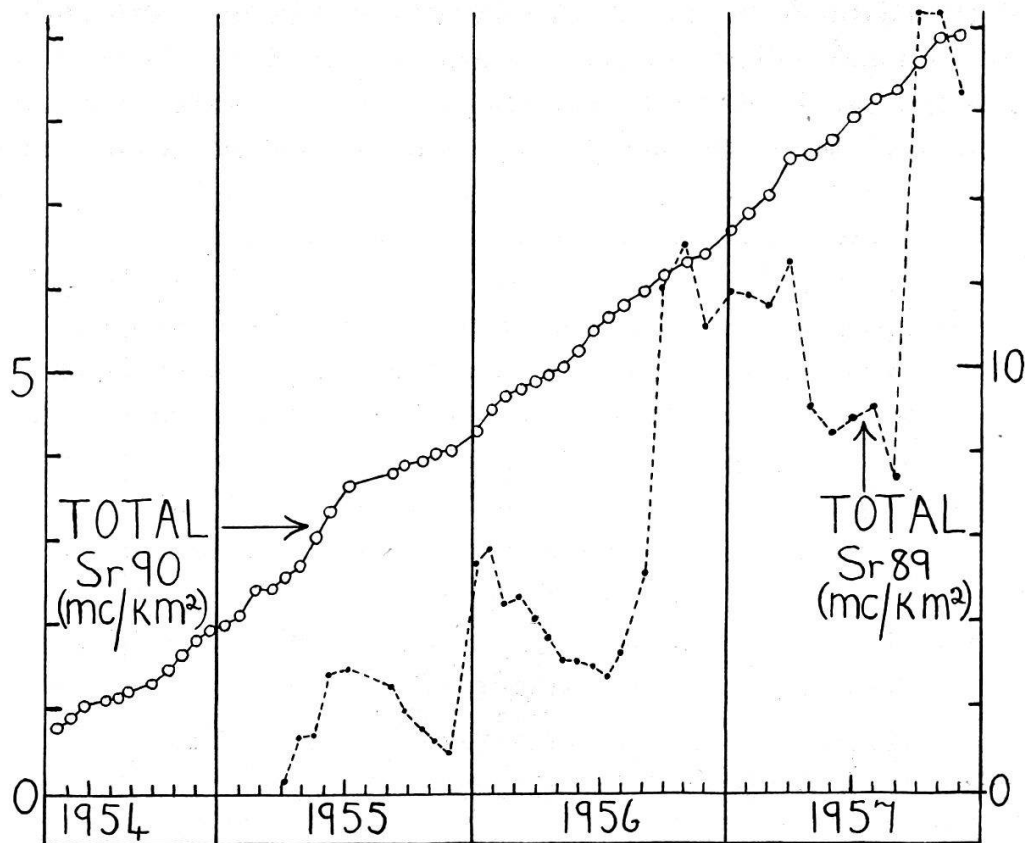


Fig. 1. Cumulative deposition of  $Sr^{89}$  and  $Sr^{90}$  at Milford Haven (Stewart et al., 1957).

sition of the nuclides of interest has been determined by radiochemical analysis of the monthly rainfall at two stations in the U.K. since 1954. The cumulative level for  $\text{Sr}^{90}$  for the station at Milford Haven in Wales is shown in fig. 1 (Stewart et al., 1957). The small accumulated deposit prior to March, 1954, was calculated from the earlier measurements of total fallout and the estimated age of the debris. Since 1954, the rate of fallout of  $\text{Sr}^{90}$  has been remarkably constant with a mean value of  $2.3 \text{ mc/km}^2/\text{year}$ . The cumulative total deposition at Milford Haven of  $\text{Sr}^{90}$  up to October, 1957, is  $8.6 \text{ mc/km}^2$ . Measurements of the specific activity of rainwater at six stations in the U.K. having widely different rainfall rates, showed that the specific activity of the rain is substantially independent of the amount of rainfall and accordingly, for the U.K. area, the total deposition of long-lived activity is approximately proportional to the local rainfall. Confirmation of this is seen from radiochemical measurements of the  $\text{Sr}^{90}$  present in the top 10 cm of soil in various parts of the U.K. These measurements are shown in fig. 3 plotted against the rainfall for the locality over the years 1955 to 1957.

The rainfall measurements have been extended to thirteen overseas stations at various latitudes including some in the southern hemisphere. A marked dependance of total fallout with latitude has been found and is shown in fig. 2 (Stewart et al., 1957). The fallout is a maximum between the latitudes  $30^\circ$  and  $50^\circ \text{ N}$  and the levels in the southern hemisphere, and particularly in the tropical belt, have been found to be much below this level. Some indication of seasonal variation of fallout has also been obtained although this has been, to some extent, masked

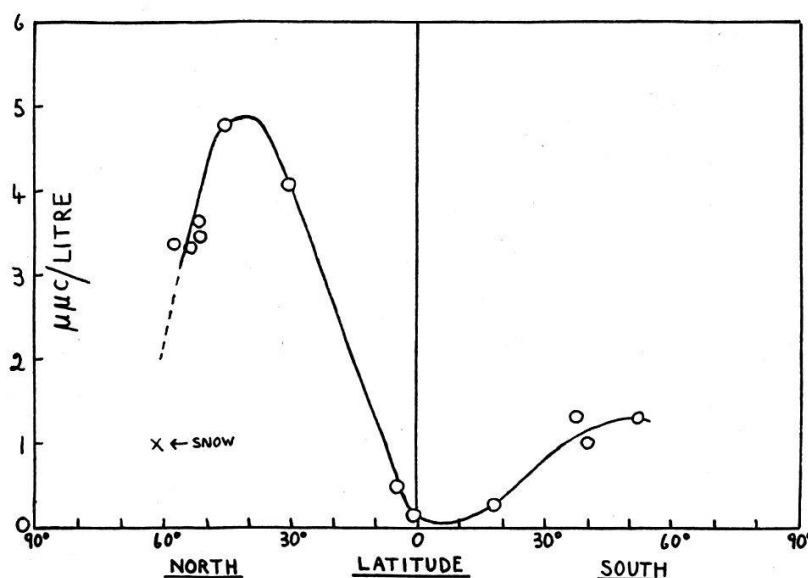


Fig. 2. Mean  $\text{Sr}^{90}$  content of rainwater at various latitudes 1955–1957 (Stewart et al., 1957).



by the seasonal variation in the conduct of nuclear explosions. The world-wide distribution of the fission products diffusing from the stratosphere is being compared with other stratospheric physics data and appears to be in accord with existing data on stratospheric circulation.

Calculations of the external radiation dose from deposited fission products on the ground show that this arises primarily from the long-lived  $\text{Cs}^{137}$  isotope. Measurements of the level of  $\text{Cs}^{137}$  in the human body show that the dose to the gonads from the internal source is much greater than that from the external radiation. The internal dose measurements are reported below.

### *Measurement of $\text{Sr}^{90}$ in the Human Food Chain*

Apart from the genetic effects of external irradiation, the biological effect of the radioactive fallout depends upon the extent to which the individual fission products are taken up in the human body through the human food chain. The fission product of greatest biological hazard is  $\text{Sr}^{90}$  (Medical Research Council, 1956, paragraph 236) and radiochemical analysis for  $\text{Sr}^{90}$  in biological material was commenced in the U.K. in 1954. The two most important factors influencing the levels in biological materials were originally thought to be the amount of rainfall, as affecting the total fallout, and the soil conditions, as affecting the uptake from the soil to plants. However, it now appears that foliar retention and root mat absorption of  $\text{Sr}^{90}$  are the dominant factors, at least on some soils, and this has proved to be the case over much of the area of the U.K. up to the present. The earlier measurements were concentrated on the uptake of radioactivity in sheep bones, which were used as a monitoring agency. An extensive human bone sampling programme was commenced in October, 1955, and the work has been successively expanded to include elements of the human food chain. Measurements have now been made on all the components of representative diets in several parts of the country.

The methods developed for the satisfactory determination of radiostrontium in various biological materials have been described by *Bryant et al.* (1956) and for rainwater by *Osmond et al.* (1957). For the determination of the very low levels of  $\text{Sr}^{90}$  in human bone anti-coincidence counting methods are necessary, the backgrounds of the assemblies used being of the order of 0.5 counts/minute.

During the development of the work, samples have been exchanged between various laboratories engaged in this type of work and excellent agreement by two British laboratories and three American laboratories

has been obtained (*J. H. Harley et al.*, 1956). It cannot be too strongly emphasized that for work of this kind reliability in measurement is of paramount importance and sufficient laboratory time must be allowed for check determinations to be made, for determinations of the  $Y^{90}$  daughter of the  $Sr^{90}$  where necessary, and to allow the development of new methods or improvements of existing methods as the work proceeds.

In view of the similarity in the chemistry and metabolism of strontium and calcium, it is frequently desirable to express results as micromicrocuries per gram of calcium in the specimen,  $\mu\mu c$   $Sr^{90}/g$  Ca, or Strontium Units (formerly known as Sunshine Units in the United States). In order to provide a wider basis for estimation of future trends, the measurements now include the stable strontium content per gram of calcium in the sample.

The  $Sr^{90}/Ca$  ratio in the roots of plants growing in the soil is more nearly represented by the exchangeable than by the total strontium and calcium content of the soil but it is very difficult to reproduce the extraction effected by the plant by any simple chemical procedure (*Bowen and Dymond*, 1955, 1956). Since the exchangeable calcium in the soil is an imprecise concept, it was decided to make extraction with 6 M hydrochloric acid the standard procedure. It has been shown that this method extracts as much  $Sr^{90}$  from the soil as the more heroic method of fusion with sodium carbonate, and the results agree well with the estimated cumulative fallout in rain as shown in fig. 3.

#### *$Sr^{90}$ in Soil, Grass and Sheep Bone*

The Government Ministry of Agriculture has been responsible for organizing the sampling at various sampling sites arranged to cover a range of different soil types and situations. The results for  $Sr^{90}$  measurements in soil, grass and sheep bone in 1956 are summarized in table 1 (*Bryant, Chamberlain, Morgan and Spicer*, 1957). Evidence from fig. 3 shows that  $Sr^{90}$  remains substantially in the top soil and available to HCl extraction for periods of the order of years. Measurements from depths 4 to 8 inches and 8 to 12 inches support the view that  $Sr^{90}$  remains substantially in the top soil (*Booker et al.*, 1956). The subsequent behaviour of the  $Sr^{90}$  depends on the exact mode in which it is retained in the soil. There may, in fact, be considerable non-uniformity over the top 4 inches. On acid soils there is often a layer of root matt between the vegetation and the soil, and in circumstances where a separate sample could be taken, the matt was found to contain about half the total activity in  $\mu\mu c/m^2$  and the S.U. ratio in it was  $4\frac{1}{2}$  times that of the soil beneath.

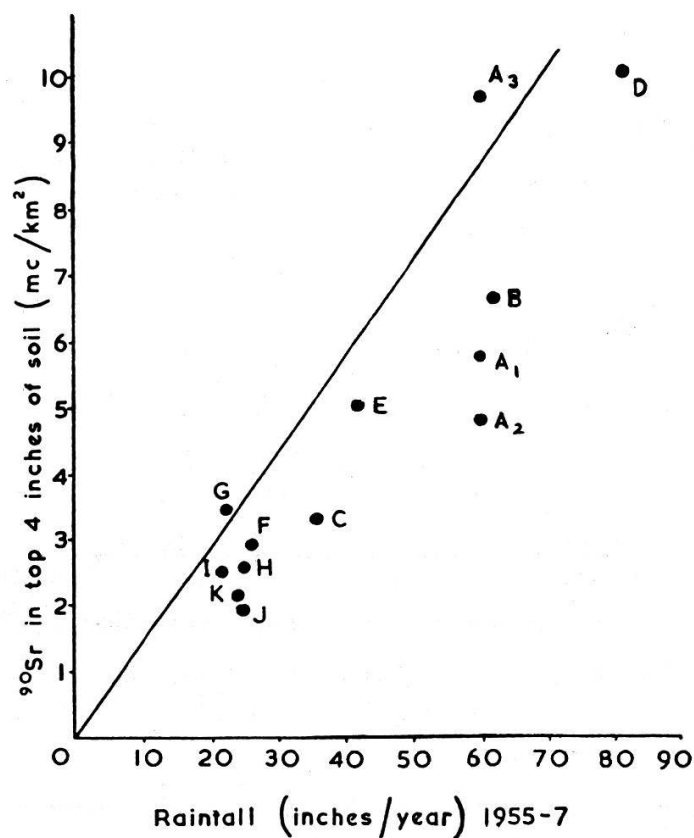


Fig. 3. Correlation of soil  $\text{Sr}^{90}$  and rainfall. Points show measured soil values plotted against local rainfall: line shows fallout computed for various rainfall amounts from fallout measurements in rain at Milford Haven (Bryant et al., 1957).

Table 1  
 $\text{Sr}^{90}$  in Soil, Grass and Sheep Bone in 1956

Station	pH	Soil (HCl extraction)			Grass			Sheep Bone $\mu\mu\text{c/g Ca}$
		$\text{g Ca/kg}$	$\mu\mu\text{c/m}^2$	$\mu\mu\text{c/g Ca}$	$\mu\mu\text{c/m}^2$	$\mu\mu\text{c/kg}$	$\mu\mu\text{c/g Ca}$	
A1	4.3	0.14	4900	800	60	2550	2100	160
A2	4.5	0.17	5700	760	90	2500	1400	
A3	4.9	1.1	9600	130	15	230	116	
B	5.4	1.8	6600	59	145	1000	123	
C	6.2	2.7	3300	14	—	—	—	
D	5.6	5.8	10000	28	50	2100	125	53
E	3.6	0.37	5000	220	64	790	625	71
F	7.5	4.7	2900	5.2	20	450	64	13
G	6.8	14.6	3400	2.6	15	250	41	8.7
H	6.0	1.6	2600	19	a) 24	150	68	
					b) 1.3	68	48	
I	6.6	3.0	2500	8.0	a) 24	100	33	
					b) 2.3	89	26	
J	7.1	39	1900	0.66	a) 18	100	32	28
					b) 2.5	72	26	
K	8.0	156	2200	0.15	a) 27	190	33	
					b) 4.8	210	28	

a) Accumulated growth. b) Fresh growth.

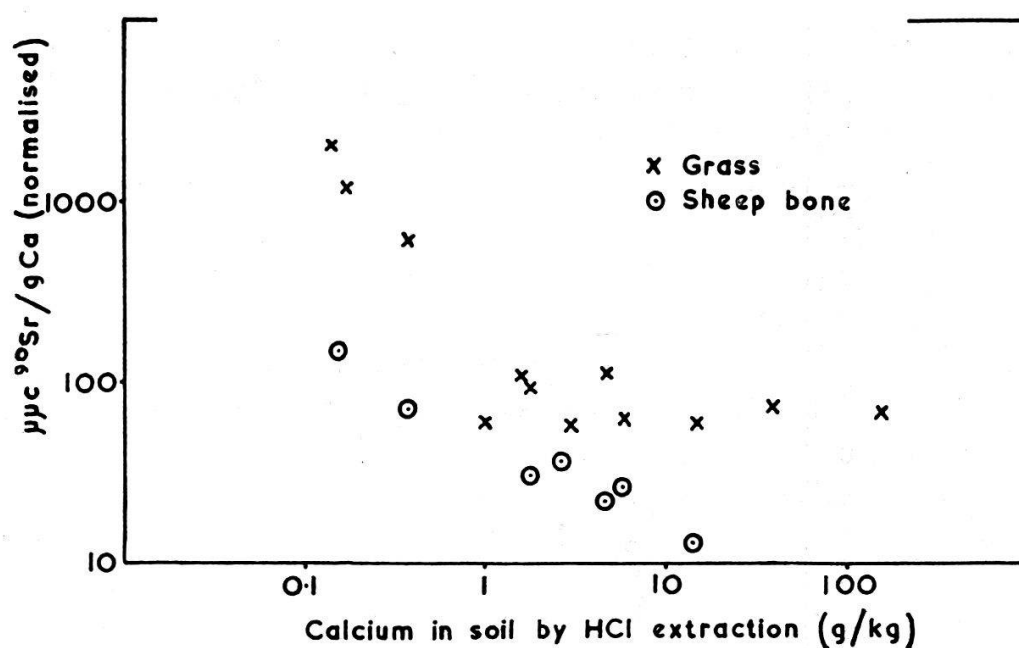


Fig. 4. Correlation of  $\text{Sr}^{90}$  in grass and sheep bone with soil calcium (normalized to  $5000 \mu\mu\text{c}/\text{m}^3$  in soil).

The level in grass in mid-1956 for each of the stations is plotted in fig. 4 against the calcium content of the top 4 inches of soil. In making the comparison the variation in total fallout of  $\text{Sr}^{90}$  ( $\mu\mu\text{c}/\text{m}^2$  in soil) has been allowed for by normalizing the results of the various stations to a nominal fallout of  $5000 \mu\mu\text{c}/\text{m}^2$ . The normalized S.U. ratio in grass does not show any correlation with soil calcium when the latter is 1 g/kg dry weight or over, as determined by HCl extraction. Thus the normalized S.U. in grass from samples A3 and K are about the same, though the soil at the former station has 1 g Ca/kg and pH 4.9 whereas the latter has 156 g Ca/kg and pH 8.0. The normalized S.U. values in vegetation on the uncultivated, acid and very low calcium soils A1, A2 and E are higher by a factor of 10 to 60 than the values for normal soils. These results are in accord with those obtained by Romney et al. (1957) from laboratory experiments on the uptake of  $\text{Sr}^{90}$  from soils of various types. The relatively high root uptake in soils of very low calcium content, which may in part be due to the presence of root matt, is being closely investigated by the Agricultural Research Council especially in view of its importance in determining the future levels to be expected in herbage and crops on such soils.

An extensive programme of research into the uptake of radioactive strontium from soils of various types is also being conducted by the Agricultural Research Council in the U.K. The results of preliminary experiments on one type of soil using  $\text{Sr}^{89}$  have been reported by Russell and Milbourn (1957) and the work was extended last year to five different

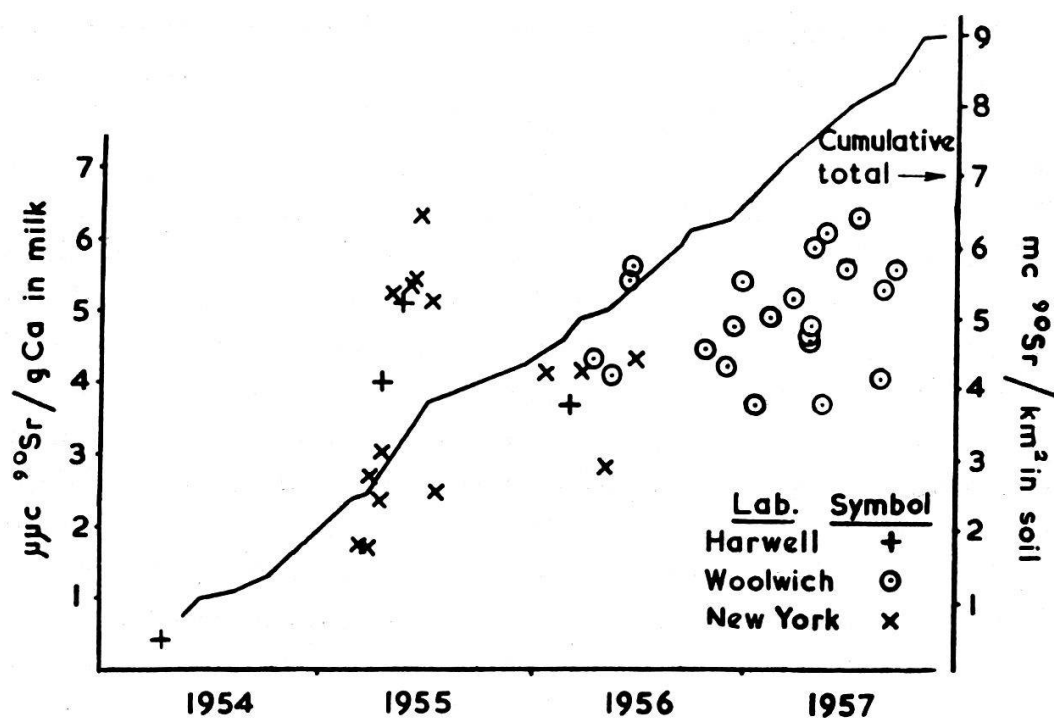


Fig. 5.  $\text{Sr}^{90}$  in Somerset milk.

representative areas of other soil types in different parts of the country. The results for the single (calcareous) soil so far published show that, for the specific types of soil cultivation used, less than 1 per cent of the  $\text{Sr}^{89}$  was recovered by crops in 1 year. It is pointed out by *Russell* and *Milbourn* that physico-chemico equilibrium in the soil may cause the absorption of  $\text{Sr}^{90}$  to decrease with time to a greater extent than would be predicted solely on the basis of its decay rate (*C. L. Comar, R. H. Wasserman and R. S. Russell, 1957*).

The measurement of the ratio of  $\text{Sr}^{90}$  and of stable strontium to calcium in sheep bone at stations A to G in 1956 show that for both  $\text{Sr}^{90}$  and stable strontium the ratio is about 0.24 of the ratio in the grass grazed although there is considerable scatter in the  $\text{Sr}^{90}$  results (*F. J. Bryant et al., 1957*). This factor is in excellent agreement with that given by *Comar et al. (loc. cit.)* for various animal species.

### *Radio-Strontium in Milk*

Systematic measurements of the  $\text{Sr}^{90}$ /calcium level in spray-dried skimmed milk from a factory at Frome, Somerset, have been made continuously since the Spring of 1955 and the results are shown in fig. 5 compared with the cumulative fallout level. It is known from Canadian work that the  $\text{Sr}^{90}$ /calcium ratio is the same in the cream as in the skimmed milk and the same also in dried as in fluid milk. The levels in milk are complicated by the use of stored animal feed in Winter, the



animals thus being exposed to foodstuffs in which the  $\text{Sr}^{90}$  levels correspond to those of the previous summer: this is particularly apparent where the  $\text{Sr}^{89}/\text{Sr}^{90}$  ratio is determined, the decay of the  $\text{Sr}^{89}$  thus reducing the ratio by a factor of up to 8 during the storage period. The  $\text{Sr}^{90}$  measurements in milk show average levels of 4.1 S.U. in 1955, 4.4 S.U. in 1956, and 5.1 S.U. for the first 9 months of 1957 for the Frome district. The increase over these years is not proportional to the cumulative deposit, which increased by a factor of 2.3 between mid-1955 and mid-1957. The milk level is apparently determined not only by the root uptake of the herbage (dependent upon cumulative deposit) but also by direct foliar contamination (dependent upon the rate of deposition): the rate of deposition of  $\text{Sr}^{90}$  was substantially constant over the years 1955 to 1957. Further evidence of this is provided by the  $\text{Sr}^{89}/\text{Sr}^{90}$  ratio which is determined by the age of the contamination: the ratio for herbage is intermediate between that for soil and that in rainwater (*Bryant et al.*, 1957).

Milk sampling was extended in 1956 to cover a representative range of districts in the U.K. and the regional results showed a minimum level of  $2.9 \mu\mu\text{c/g Ca}$  and a maximum  $10.3 \mu\mu\text{c/g Ca}$ . The median levels for Somerset were 4.4 and for the other areas 6.7. The regional results for 1957 are not yet complete but show a similar pattern. The median levels for milk are some 8 to 10 times lower than corresponding grass levels: this factor, arising from interposition of the cow in the human food chain, would not be applicable in countries where dietary calcium is derived directly from vegetation (*Hiyama*, 1957).

#### *Radio-Strontium and Stable Strontium in Human Bone*

Of the 48 measurements made on human bone samples during 1956 (*Bryant et al.*, 1957) there were 34 femora and 3 tibiae, and the results from these are shown in fig. 6. About half the samples came from children under 5 years of age. The specimens were mostly from south-east England and the Midlands, but there were a few from the west and north-west including the two showing the highest radio-strontium activity, namely, 1.55 S.U. in a 2-year-old Plymouth child and 1.3 S.U. in a 3-month-old child from Carlisle. The results quoted for human bone are those for total radio-strontium and not for specifically identified  $\text{Sr}^{90}$ . On specimens for which  $\text{Y}^{90}$  was measured independently, the  $\text{Sr}^{90}$  activity deduced was within 15 per cent of that estimated for total strontium. There is considerable scatter in the results on bones from infants under 1 year but the median value for children under 5 years was 0.7 S.U. and

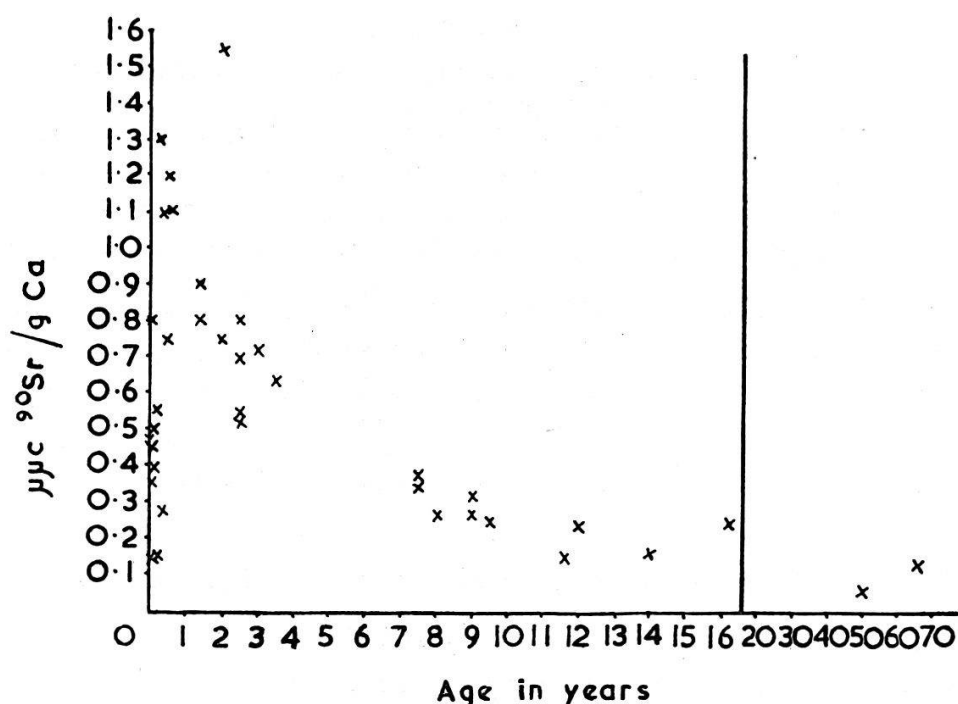


Fig. 6.  $\text{Sr}^{90}$  in human bone in 1956.

thereafter there is a steady fall off in the  $\text{Sr}^{90}$ /calcium ratio with age. The level for stillborns tends to be about half that for children in the age range 1 month to 5 years.

Human bone samples for 1957 have been mainly from the high rainfall areas in the north and west of the country which were not specifically sampled in previous measurements. Amongst the sixteen results for children's bones obtained up to mid-1957 (*Bryant et al.*, 1957) are values of 2.3 S.U. in a 1-year-old child from Keswick, Cumberland, and 2.4 S.U. from a 6-month-old child from Liverpool. Apart from one further result of 1.3 S.U. all other results were less than 1 S.U. Fifty further measurements on later 1957 samples have now been made but none of the results exceed the maximum value of 2.4 quoted. The 1957 results are thus not out of line with the pattern of results reported for 1956 when allowance is made for the somewhat enhanced contribution in the high-rainfall areas.

A series of measurements of the stable strontium content of human bones as a function of age has been made by *Sowden and Stitch* (1956) and a further series by *Bryant et al.* (1958). These measurements are in excellent agreement and show that at any one age the stable strontium content per gram of calcium in the bone shows a scatter of about a factor 2 and the average value rises from rather less than 200  $\mu\text{g Sr}$  per gram of calcium at birth to nearly 300 in adolescence, and thereafter tends to remain substantially constant.



Table 2  
Average Daily Intake of Strontium and  $\text{Sr}^{90}$  in Various Items of Diet  
(Cumberland and Dumfries area) 1957  
(F. J. Bryant, A. C. Chamberlain, G. S. Spicer and M. S. W. Webb, 1958.)

Food	Daily intake		
	Calcium mg	Strontium $\mu\text{g}$	$\text{Sr}^{90}$ $\mu\mu\text{c}$
Milk and milk products . . . . .	508	213	2.69
Flour, bread, etc. . . . .	255	484	0.36
Rest of diet . . . . .	114	368	1.74
Totals . . . . .	877	1,065	4.79

*Contribution of Various Elements of Diet to the  $\text{Sr}^{90}$  Level of Human Bone*

During 1957 the sampling and analysis was extended to an investigation of the stable and radio-strontium contents of the principal items of diet in addition to milk. Most of the samples were taken in the west and north of Britain, since these are the regions of highest rainfall and therefore highest fallout. This also had the advantage that the proportion of milk and vegetables imported from a distance in the sampling areas was less than would be the case in the Midlands and south-east England. Stable strontium as well as  $\text{Sr}^{90}$  was measured so that the passage of the isotopes in parallel through food chains could be studied. Measurements were made on various foodstuffs in four regions of the U.K. The daily intake of these materials was then computed for representative diets for families of four or more children. A summary of the results for the Cumberland and Dumfries areas is shown in table 2. The results show that the average  $\text{Sr}^{90}/\text{Ca}$  level in the diet is about the same as that in the milk portion of it, namely, 5 to 6  $\mu\mu\text{c/g}$  Ca. This is because the low ratio in the calcium-fortified flour balances the higher ratio in vegetables and meat. A more primitive diet would show a higher average ratio. It will also be seen that the average stable strontium/calcium ratio is five times that in the milk portion. The regional variation in average diet does not appear large. It should be recalled that the contribution to the  $\text{Sr}^{90}$  content of the diet is changing rather slowly with time in view of its considerable dependance on rate of fallout. The strontium/calcium ratios in respective daily diet averaged for the four areas sampled show the following ratios:

Stable strontium/calcium 1170  $\mu\text{g/g}$ :  $\text{Sr}^{90}/\text{calcium}$  5.6  $\mu\mu\text{c/g}$ :  $\text{Sr}^{90}/\text{Strontium}$  4.8  $m\mu\text{c/g}$ .

These results give some indication of the extent to which the bones of children are in equilibrium with the environment. The stable strontium/

calcium ratio in children's bones in the age range 0 to 5 years is 208  $\mu\text{g}$  per gram, whereas the level in the diet averaged for the four regions of the U.K. is 1170  $\mu\text{g/g}$ , a ratio of about 1/5.6. Since the level of  $\text{Sr}^{90}$  in the diet expressed per gram of calcium is 5.6  $\mu\mu\text{c/g}$ , one would expect children's bones in equilibrium to have a level of about 1  $\mu\mu\text{c/g}$  Ca, which is approximately the level observed. The strontium metabolism of adults in the U.K. is being studied directly and also by the use of the tracer isotope  $\text{Sr}^{85}$ . From the results obtained (Harrison et al., 1957) it appears that the metabolism in the child is not very different from that in the adult and that the children's bones are approximately in equilibrium with the environment.

A further aspect of dietary  $\text{Sr}^{90}$  is the level in drinking water. Measurements in the U.K. show that the level in the drinking water of towns is from 5 to 100 times less than the level of 3 to 5  $\mu\mu\text{c/litre}$  observed for rainwater, and that the contribution of water supplies to the dietary  $\text{Sr}^{90}$  intake is relatively small.

#### *Summary of $\text{Sr}^{90}$ Results for Biological Materials in 1956*

Table 3 summarizes the results for the various biological materials in 1956. Where results for 1957 are available these have been reported in the text.

Table 3  
 $\text{Sr}^{90}$  in Biological Materials in 1956

Material	No. of samples	$\text{Sr}^{90}$ activity ( $\mu\mu\text{c/g}$ Ca)		
		Max.	Min.	Median
Grass (acid hill soils) . . . . .	9	2,100	91	130
Grass (normal soils) . . . . .	61	77	11	37
Sheep bones (hills) . . . . .	6	170	24	57
Sheep bones (lowland) . . . . .	7	15.6	7.8	13.7
Milk (Somerset) . . . . .	13	5.7	2.9	4.4
Milk (other areas) . . . . .	10	10.3	3.9	6.7
Human bones (femora and tibiae)				
0 to 5 years . . . . .	25	1.55	0.15	0.70
5 to 20 years . . . . .	10	0.38	0.15	0.26
Over 20 years . . . . .	2	0.13	0.06	—

#### *$\text{Cs}^{137}$ in Foodstuffs and in the Human Body*

A gamma-spectrometer method has been used (D. V. Booker, 1957) for the rapid determination of  $\text{Cs}^{137}$  levels in milk. The samples comprising about 2 kg of dried milk are placed in an aluminium pan with

Table 4  
Cs<sup>137</sup> Levels in Dried Milk Samples from Five Districts in the U.K. in 1956

Sample number	Source	Date	$\mu\mu\text{c Cs}^{137}$ per kg	g K/kg	$\mu\mu\text{c Cs}^{137}$ per g K	$\mu\mu\text{c Sr}^{90}$ per g Ca
C53	Carmarthen	17. 10. 56	640	11.8	54	8.0
C68	Wales	29. 12. 56	620	12.2	51	—
C54	Driffield	16. 10. 56	245	11.7	25	4.3
C72	Yorks	27. 12. 56	320	11.3	28	—
C55	Carlisle	19. 10. 56	360	11.4	31	6.5
C70	Cumberland	25. 12. 56	335	11.6	29	—
C56	Ballymoney	19. 10. 56	800	11.1	72	6.9
C69	N. Ireland	28. 12. 56	720	11.4	63	—
C57	Coleraine	17. 10. 56	800	11.4	70	10.3
C71	N. Ireland	27. 12. 56	700	11.3	62	—
Errors		—	$\pm 35$	$\pm 0.3$	$\pm 3$	—

a recessed base which fits over the 75-mm sodium-iodide crystal, the whole being placed inside a 10 cm thick lead castle and used in conjunction with a 5-channel pulse-height analyser which enabled the pulses due to Cs<sup>137</sup> and K<sup>40</sup> to be separately determined. The results obtained on samples from five districts in the U.K. mainly in the higher fallout area of the north and west are shown in table 4 above. The table also shows the corresponding Sr<sup>90</sup> levels.

Measurements have been made of the Cs<sup>137</sup> content of the human body using a total body gamma spectrometer. This equipment is being extensively used at A.E.R.E. in studying the instances of internal contamination of the human body with radioactive materials arising in atomic energy work. For the present purpose, however, care has been taken to

Table 5  
Dose Rate from Sr<sup>90</sup> and Natural Radioactivity to Bone

Source of radiation	Dose rate (mrem/year)
Natural radiation	
external sources . . . . .	82
radium in bone* . . . . .	39
Total . . . . .	121
Sr <sup>90</sup> in children under 5	
median level for 1956 (0.70 S.U.) . . . . .	2
maximum level up to mid-1957 (2.4 S.U.) . . . . .	6

\* R.B.E. of  $\alpha$  rays taken as 10 (I.C.R.P., 1955).

select from the records those persons who are known not to have had any previous contact with  $\text{Cs}^{137}$  in the course of their work. On this basis, the level in 16 men has been determined in the period June, 1956, to July, 1957 (*Rundo*, 1958). The average  $\text{Cs}^{137}$  content was found to be 5.4  $\mu\text{mc}$  in the body. The potassium level was measured at the same time and it was concluded that the caesium content could be expressed per gram of potassium as 35  $\mu\text{mc/g K}$ . The potassium content was found to have a mean value of  $0.212 \pm 0.023$  per cent of body weight. Some repeat measurements were made on some of the individuals at different times and variations both up and down in the period of observation were observed. This is not unexpected in view of the moderately short biological life of  $\text{Cs}^{137}$  in the body which has been estimated as 140 days (*Marinelli et al.*, 1956). In view of the suspected low uptake of this isotope from the soil by plants, there is reason to expect the level in the human body to reflect the level of direct foliage contamination and therefore to be dependent upon the rate of  $\text{Cs}^{137}$  fallout which has been substantially constant since early 1954. The radiation dose level from  $\text{Cs}^{137}$  in the body, and in particular in gonad tissue, from the level of contamination quoted above, is computed to be 0.6 millirem per year which compares with the background level in the gonads of about 100 millirem per year. If the rate of fallout continues to remain constant, the dose-rate would thus likewise be expected to remain constant.

#### *Dose Levels in Tissue and Future Predictions*

The dose levels in human bone due to the  $\text{Sr}^{90}$  levels observed in 1956 are compared in table 5 below with those from natural radioactivity (*Spiers*, 1956).

The maximum permissible body burden for occupational workers is 1  $\mu\text{c}$  (I.C.R.P., 1955) which is approximately equivalent to 1000 S.U. The Medical Research Council Committee (1956) have proposed a limit of one-tenth of the occupational body burden for the general population and state also that "immediate consideration would be required if the concentration in human bones showed signs of rising greatly beyond one-hundredth of that corresponding to the maximum permissible occupational level". The highest  $\text{Sr}^{90}$  activity in human bone found in the U.K. was 2.4 S.U. which is one-fortieth of the maximum permissible for the general population, and one-fourth of the level above which "immediate consideration would be required".

It has been suggested that a dose rate 10 times the human occupational level would be acceptable for animals (*Chamberlain, Loutit, Russell* and

*Martin*, 1956). The  $\text{Sr}^{90}$  maximum permissible level for sheep would then be 10000 S.U. The highest levels recorded in Britain are at Cwmystwyth, when 183 S.U. was found in October, 1955 (*Bryant et al.*, 1956), and 160 S.U. in July, 1956 (table 1).

The levels of gonad exposure and of  $\text{Sr}^{90}$  in bone to be expected in the future may be deduced from the calculation of future levels of fallout made by *Stewart et al.* (1957). It appears that the gonad dose will remain at less than 1 mrem/year in the U.K. if the present constant rate of fallout is maintained.

Estimates of the future levels in bone will depend on the fraction of the present level which is due to cumulative deposit. The trend in the  $\text{Sr}^{90}$  levels in milk over the past 3 years suggests that the present level in the U.K. is to a considerable extent due to direct foliar contamination, which is dependent on the rate of fallout, rather than to root uptake, which is dependent on the cumulative deposit. Effort is being directed at an elucidation of this partition with a view to enabling the future levels in bone to be evaluated. The future levels in a particular area will, however, depend upon many critical factors, such as the nature and calcium status of the soil, dietary considerations, and climatic and geographical factors which affect the mode of agriculture as well as the local level of fallout. The levels depend, of course, above all upon the future injection of fission products into the stratosphere.

### *Summary*

The report reviews various aspects of the extensive research carried out in the United Kingdom during recent years on radioactive fallout from nuclear test explosions. Continuous measurements of the gross fission-product radioactivity in rainwater were commenced in 1951, and, more recently, radiochemical determinations of the fallout rates of the isotopes  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  have been made at several stations over the world. The cumulative total of  $\text{Sr}^{90}$  in the fallout in the United Kingdom up to Oct., 1957 was found to be  $8.6 \text{ mc/km}^2$  with a substantially constant annual fallout rate of  $2.3 \text{ mc/km}^2 \text{ year}$ . A marked variation of the rate of fallout has been found with season and with latitude.

A detailed study has been made of the fate of the long-lived isotopes  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  in human food chains involving measurements on soil, grass, animal bones, milk and foodstuffs, in relation to the amount of stable calcium present in the foodstuff and also in the whole diet and, in some instances, to the stable strontium content. The importance of direct foliar contamination and the influence of very low calcium content



of the soil in some areas in promoting high uptake through the roots has been studied. The work carried out directly on fallout activities in agricultural materials has been supported by extensive soil and agriculture studies on the behaviour of  $\text{Sr}^{89}$  in soil and plants, carried out by the Agricultural Research Council.

The  $\text{Sr}^{90}$  activity in human bones expressed in terms of the stable calcium content is shown to be dependent on age, the activity of 25 children's bones in the age range 1 month to 5 years having a median value of 0.7 and a maximum value of  $1.55 \mu\mu\text{c}$   $\text{Sr}^{90}$  per gramme of calcium in 1956. Human bone samples for 1957 have been mainly from the high rainfall areas in the north and west of the country and the values obtained are somewhat higher, the highest being  $2.4 \mu\mu\text{c}$   $\text{Sr}^{90}/\text{g Ca}$ . Measurements of the level of the  $\text{Cs}^{137}$  activity in the human body have been made by total-body  $\gamma$ -ray spectrometry and showed an average level of  $5.4 \mu\mu\text{c}$  in the body (16 men) in the period mid-1956 to mid-1957, corresponding to a radiation dose-rate of less than 1 millirad per year. Corresponding measurements have been obtained for the  $\text{Cs}^{137}$  content of dried milk and other foodstuffs.

An evaluation of the effects on man of the irradiation from fallout material, and from other sources of ionizing radiations was made by the British Medical Research Council in 1956 and reference is made to the conclusions of this study.

### *Zusammenfassung*

Der Bericht befaßt sich mit verschiedenen Aspekten der ausgedehnten Forschungen, die im Vereinigten Königreich während der letzten Jahre über den radioaktiven Niederschlag von Atomtestexplosionen unternommen wurden. Seit 1951 wurde dauernd die totale Radioaktivität von Zerfallsprodukten im Regenwasser gemessen, und seit kurzem wurden auf verschiedenen Stationen, verteilt über die ganze Erde, radiochemische Bestimmungen der im Niederschlag enthaltenen Isotope  $\text{Sr}^{90}$  und  $\text{Cs}^{137}$  vorgenommen. Die totale niedergeschlagene Aktivität von  $\text{Sr}^{90}$  im Vereinigten Königreich bis Okt. 1957 betrug  $8,6 \text{ mc}/\text{km}^2$  bei einer im wesentlichen konstanten Niederschlagsrate von  $2,3 \text{ mc}/\text{km}^2$  im Jahr. Eine starke Abhängigkeit der Ausfallsrate von der Jahreszeit und der geographischen Breite wurde gefunden.

Eine detaillierte Untersuchung des Weges der langlebigen Isotope  $\text{Sr}^{90}$  und  $\text{Cs}^{137}$  in menschlichen Ernährungszyklen wurde unternommen. Dabei erfolgten Messungen im Boden, im Gras, in Tierknochen, in der Milch und in Lebensmitteln im Vergleich zum stabilen Calciumgehalt der Lebensmittel und der gesamten Ernährung. In einigen Fällen wurde

auch mit dem Gehalt an stabilem Strontium verglichen. Die Auswirkung einer direkten Verseuchung der Blätter und der Einfluß eines niedrigen Calciumgehalts des Bodens in einigen Regionen, der eine verstärkte Aufnahme durch die Wurzeln bewirkt, wurde studiert. Die Arbeiten, die sich direkt mit Aktivitäten von landwirtschaftlich wichtigen Pflanzen befaßten, wurden durch ausgedehnte Boden- und agrikulturtechnische Untersuchungen (Agricultural Research Council) über das Verhalten von  $\text{Sr}^{89}$  in Boden und Pflanzen unterstützt. Es wurde gezeigt, daß die  $\text{Sr}^{90}$ -Aktivität in menschlichen Knochen in Abhängigkeit des Gehaltes an stabilem Calcium vom Alter bestimmt wird. In Knochen von 25 Kindern im Alter zwischen 1 Monat und 5 Jahren wurde 1956 ein mittlerer Gehalt von 0,7 und ein Höchstwert von 1,55  $\mu\mu\text{c}$  pro g Calcium festgestellt. Die menschlichen Knochenproben des Jahres 1957 stammen hauptsächlich aus regenreichen Gebieten im Norden und Osten des Landes; es wurden etwas höhere Werte erhalten; die höchsten betragen 2,4  $\mu\mu\text{c}$   $\text{Sr}^{90}$ /g Ca.

Messungen des  $\text{Cs}^{137}$ -Aktivität des menschlichen Körpers erfolgten mit Hilfe der  $\gamma$ -Strahlen-Spektrometrie. Sie zeigten pro g natürliches Kalium in der Zeit von Mitte 1956 bis Mitte 1957 bei 16 Männern einen Mittelwert von 5,4  $\mu\mu\text{c}$ . Dies entspricht einer Dosisleistung von weniger als 1 millirad pro Jahr. Entsprechende Meßergebnisse wurden für den  $\text{Cs}^{137}$ -Gehalt getrockneter Milch und anderer Nahrungsmittel gefunden.

Eine Auswertung der Effekte auf den Menschen infolge Bestrahlungen durch den radioaktiven Niederschlag und anderer Quellen ionisierender Strahlen wurde 1956 vom British Medical Research Council durchgeführt, auf dessen Folgerungen Bezug genommen wird.

### *Résumé*

Ce travail passe en revue les différentes données obtenues par l'étude sur une grande échelle des retombées radioactives au cours de ces dernières années dans le Royaume Uni, à la suite des essais d'explosion atomique. En 1951, on a commencé à faire des déterminations continues de la radioactivité des produits de fission présents dans l'eau de pluie, et récemment on s'est mis à déterminer la teneur en  $\text{Sr}^{90}$  et  $\text{Cs}^{137}$  dans les retombées radioactives dans plusieurs stations placées en de nombreux points du globe. La somme totale de  $\text{Sr}^{90}$  dans les précipitations radioactives en Grande Bretagne, jusqu'en oct. 1957, a été de 8,6 mc/km<sup>2</sup>, avec un taux constant annuel des précipitations de 2,3 mc/km<sup>2</sup>. On a constaté une grande différence d'intensité des retombées radioactives selon les saisons et les latitudes. On a étudié en détail l'évolution des isotopes à longue durée de vie tels que le  $\text{Sr}^{90}$  et  $\text{Cs}^{137}$  dans l'alimentation



humaine, en partant de leur présence dans le sol, l'herbe, le squelette des animaux, le lait et les aliments, et en proportion avec la teneur en calcium stable contenu dans les aliments en général, dans certains cas aussi en proportion avec la teneur en strontium stable. Cette étude souligne le rôle de la contamination directe des feuilles et l'influence exercée par une teneur très faible du sol en calcium sur l'absorption augmentée d'éléments radioactifs par les racines. Ces travaux sur les radioactivités des plantes importantes pour l'agriculture sont appuyés par des recherches étendues, en particulier, sur le comportement du  $\text{Sr}^{89}$  dans le sol et dans les plantes, et accomplies sous les auspices du Conseil de Recherche en Agriculture.

L'activité du  $\text{Sr}^{90}$  dans le squelette humain, par rapport à la teneur en calcium stable, s'est montrée être en relation directe avec l'âge des personnes étudiées; l'activité du  $\text{Sr}^{90}$  dans les os de 25 enfants âgés de 1 mois à 5 ans, a donné une valeur moyenne de 0,7 ainsi qu'une valeur maximum de  $1,55 \mu\mu\text{c}$  de  $\text{Sr}^{90}$  par g de calcium en 1956. Les épreuves d'os humains de l'année 1957 proviennent surtout des contrées riches en pluie du nord et de l'ouest du pays; les valeurs obtenues sont un peu plus grandes, le maximum étant de  $2,4 \mu\mu\text{c Sr}^{90}/\text{g Ca}$ .

Les mesures de l'activité du  $\text{Cs}^{137}$  dans le corps humain ont été faites par la spectrométrie du rayonnement  $\gamma$  du corps entier et ont donné une valeur moyenne de  $5,4 \mu\mu\text{c}$  (16 hommes), durant la période allant du milieu de 1956 au milieu de l'année 1957, ce qui correspond à une dose d'irradiation de moins de 1 millirad par année. Des valeurs correspondantes ont été obtenues pour la teneur de  $\text{Cs}^{137}$  du lait en poudre et d'autres aliments. Sous les auspices du Conseil Médical de la Recherche Britannique, on a cherché à évaluer les effets chez l'homme des irradiations par de produits de fission et ceux dus à d'autres sources de radiations ionisantes au cours de l'année 1956; on fait mention des résultats de ces études dans le présent travail.

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