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# PREDICTION OF FUTURE LEVELS OF LONG-LIVED FISSION PRODUCTS IN MILK

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CINCE 1958, when the United Nations Scientific Committee on the Effects of Atomic Radiation<sup>1</sup> made its first comprehensive review of the consequences of world-wide fall-out, many attempts have been made to derive formulae for calculating the level of radioactivity in foodstuffs which is likely to be caused by different patterns of environmental contamination. Information has been obtained mainly by analysing relationships between measurements of fall-out and of dietary contamination made in environmental surveys. Controlled experiments on food-chain mechanisms and information on agricultural practices are, however, important supplementary sources of guidance; besides indicating whether relationships suggested by survey results are causal or fortuitous they can reveal factors which, though unimportant under conditions so far experienced, may require greater consideration in the future. Improved information can be expected as the period during which observations have been made becomes longer because both climatic factors and variations in the seasonal distribution of fall-out influence the transfer of radioactivity to diet. Conclusions previously reached in the United Kingdom<sup>2-4</sup> have, therefore, been reviewed in the light of the more extensive data which are now available.

Consideration has been limited to two nuclides, strontium-90 and caesium-137, in a single foodstuff, milk. These two nuclides are mainly responsible for the internal radiation doses to which long-lived fission products give rise and it is well established that the contamination in milk has provided a good guide not only to the exposure of children but also to that of adults who consume a mixed diet of the type normal in Western countries<sup>5</sup>. Moreover, the data on the contamination of milk are considerably fuller and more reliable than those for other foodstuffs.

The annual average levels of contamination in milk throughout the United Kingdom have alone been considered; predictions for smaller areas or shorter periods would be subject to much greater uncertainty. The country-wide average deposition of strontium-90 used in the present calculations has been derived, from the

results of surveys conducted by the United Kingdom Atomic Energy Authority<sup>6-12</sup>, by multiplying the mean concentration in rain from all sites for which quarterly determinations were made by the mean rainfall throughout the United Kingdom. The average ratio of caesium-137 : strontium-90 in fall-out is relatively constant, and the deposition of caesium-137 has therefore been calculated from that of strontium-90 taking the ratio of caesium-137 : strontium-90 as 1.7 up to 1961 and 1.5 since then<sup>13</sup>. The country-wide mean levels of radioactivity in milk were taken from the reports of the Agricultural Research Council Radiobiological Laboratory<sup>14-29</sup>.

#### Strontium-90

It has been recognized for many years that strontium-90 may enter milk as a result of both the 'direct' contamination of pastures and other forage crops with airborne debris and the absorption of strontium-90 through plant roots from the soil. The magnitude of these two components is related, respectively, to the recent rate of fall-out and the cumulative deposit in the soil. Equations of the following type have, therefore, been widely used to relate the ratio of strontium-90 to calcium in milk to the rate of fall-out and cumulative deposit of strontium-90:

 $C = p_r F_r + p_d F_d \tag{1}$ 

where C is the 12-month mean ratio of strontium-90 to calcium in milk (pc./g),  $F_r$  is the annual deposit of strontium-90 (mc./km<sup>i</sup>/year),  $F_d$  is the cumulative deposit of strontium-90 at the middle of the year (mc./km<sup>2</sup>), and  $p_r$ ,  $p_d$  are described as the 'rate' and 'soil' proportionality factors. Such equations have been shown to provide a useful basis for predicting both the average levels of contamination in milk in a number of countries and also the world-wide situation<sup>4,5,21</sup>. None the less, the relationship between the pattern of fall-out and the contamination of milk is considerably more complex than the equation suggests. The assumption that the soil factor is constant ignores the progressive penetration of strontium-90 deeper into the soil which will reduce its availability to pasture plants<sup>3</sup>. A further defect is that the equation takes no account of the fact that an appreciable, though variable, fraction of the diet of cattle consists of stored food grown in the previous year; thus the rate of fall-out at that time will influence the extent to which the current diet of cattle has been directly contaminated by airborne debris. In some areas this 'lag' effect of the rate of fall-out may be enhanced due to the absorption by plants of strontium-90 which has lodged in the basal tissues and been retained for several months without being incorporated in the soil.

The inadequacy of equation (1) from this point of view is apparent when the mean annual ratios of strontium-90

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to calcium observed in milk throughout the United Kingdom between 1958 and 1964 are compared with those which would have been expected from the measured deposit of fall-out on the basis of the proportionality factors for equation (1), shown in Table 1, which were derived by a least squares analysis of the results of surveys between 1958 and 1961. The average agreement between calculated and observed values was good (Table 2) but for individual years there were appreciable divergences. The calculated values were too low for years when the lag effect was expected to be greatest because of high fall-out in the previous year (that is, 1960, 1963 and 1964); conversely, the calculated values were too high when the rate of deposition had previously been relatively low. An approximately linear relationship was found between the magnitude of these discrepancies and the extent of fall-out in the second half of the previous year. Only a small improvement was obtained when the rate factor in equation (1) was related to the deposition of fall-out in the twelve months ending in June or September instead of to that in the current year. The following equation was, however, found to give a significantly better fit to the data (P < 0.01), the residual standard error being reduced from 16 to 6 per cent:

$$C = p_r F_r + p_i F_i + p_d F_d \tag{2}$$

where  $F_i$  is the deposit of strontium-90 in the second half of the previous year (mc./km<sup>2</sup>) and  $p_i$  is the 'lag-rate' factor, the other symbols being defined as in equation (1). When the lag-rate factor was related to the deposit in the whole of the previous year, or in the summer months only, a poorer fit was obtained. Proportionality factors for equation (2), derived by least squares analysis, are shown in Table 1. The levels of strontium-90 in milk calculated on this basis agree closely with those observed in each year (Table 2) and it is evident that equation (2) describes the situation in past years more satisfactorily than equation (1).

The introduction of the lag-rate factor leads to a lower value for the soil factor, and a smaller fraction of strontium-90 in milk is thus attributed to absorption from cumulative deposit; it appears from equation (2) that strontium-90 from this source was responsible for from 20 to 50 per cent of the contamination of milk in past years (Table

Table 1. ESTIMATES OF PROPORTIONALITY FACTORS FOR THE TRANSFER OF STRONTIUM-90 TO MILK IN THE UNITED KINGDOM

	Equation 1	Equation 2	
Rate factor $(p_r)$ (pc. ***Sr/g Ca per mc./km*/year)	0.76*	<b>0</b> ·70	
(pc, *°Sr/g Ca per mc./km <sup>2</sup> in second half of previous year)		1.13	
Soil factor $(p_d)$ (pc. **Sr/g Ca per mc./km*)	0.19*	0-11	
Calculated from survey data for the	period 1958 to	1961 (ref. 28	١.



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Year		Deposition of strontium-90 (mc./km²)			Strontium-90 in milk			Benearte as of strenthum 90	
					Observed	Calculated values as		in milk attributable to	
		January to June	July to December	Total	(pc.**Sr/g Ca)	Equation 1	Equation 2	Equation 1	Equation 2
	1957 1958 1959 1960 1961 1962 1963 1963	1.6 3.2 6.3 1.0 0.7 5.3 9.5 9.6	1-5 2-1 1-8 0-9 1-4 5-4 9-6 5-8	3·1 5·3 8·1 1·9 2·1 10·7 19·1 14·9	7-2 9-8 6-4 5-9 11-7 25-6 28-0	91 104 92 107 119 90 82	97 107 94 90 108 96 101	38 40 76 74 42 37 51	22 23 44 53 28 21 25

# Table 2. COMPARISON OF CALCULATED AND OBSERVED LEVELS OF STRONTIUM-90 IN MILK IN THE UNITED KINGDOM





2). The use of the lower soil factor derived by equation (2) receives some independent support from the fact that a value of 0.14 has been derived from the results of tracer experiments<sup>3</sup> in the United Kingdom in which strontium-89 was distributed in the soil in a manner which is broadly comparable with that observed in surveys of fall-out in permanent pasture in the recent past<sup>19,29</sup>.

The adoption of equation (2) causes lower levels of strontium-90 in milk to be predicted when the cumulative deposit is an important source of contamination, the reverse being true when the contribution of the recent deposit, especially that in the second half of the previous year, is large. To illustrate the extent of the difference between predictions based on the two equations, calculations have been made for two hypothetical models of future fall-out, namely: Model A, the rate of fall-out will decrease by a factor of 2 annually. Model B, the rate of fall-out will remain constant. In both models it has been assumed that the annual deposit in the first year is 45 per cent of the cumulative deposit measured at mid-year (that was, the approximate situation in 1963, the year in which the highest levels of fall-out were observed) and that the rate of fall-out was similar in the second half of the previous year. The decrease in the rate of fall-out in Model A is greater than that anticipated in the absence of further weapon trials<sup>13</sup> while the amount of fall-out assumed in Model B would over a few years much exceed that caused by all weapon trials hitherto. Future situ-ations resulting from weapons testing may thus be expected to lie between the two models.

Predictions made by the two equations are compared in Table 3. In considering the situation over long periods it is necessary to take account of loss of strontium-90 from the soil which arises mainly from leaching or from absorption by plants; following the United Nations Scientific Committee on the Effects of Atomic Radiation<sup>5</sup> an average loss of 2 per cent per annum has been assumed.

For the first year the level of contamination in milk predicted by equation (1) is about 20 per cent lower.

Table 3. COMPARISON OF PREDICTED LEVELS OF CONTAMINATION OF MILK, BASED ON DIFFERENT EQUATIONS, FOR TWO HYPOTHETICAL MODELS OF FALL-OUT

	First year		Years 1-10 Integrated value		Years 1–50 Integrated value	
Model	Bo	th models	A	В	A	В
Ratio of strontium-9 (relative to year )	0 <i>to ce</i> 1 calc	<i>ilcium in mi</i> ulated by eo	lk Juation 2	)		
Based on equation	1	0.8	4.6	12	10	110
-	2	1.0	4.1	12	7	90
Caesium-137 per l. o	f mill	5				
(relative to year 1	l calci	ulated by eq	uation 4	1		
Based on equation	3	0.8	1.9	8	1.9	40
-	4	1.0	2.6	10	3.0	54

Modèl A, fall-out rate halved each year. Model B, fall-out rate remains constant. The rate and cumulative total of fall-out in year 1 are taken to be the same in both models, the annual deposit being 45 per cent of the cumulative deposit at mid-year (that is, the relationship in 1963); it is further assumed that the rate was similar in the previous year.

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 $\sum_{i=1}^{n}$ 

However, under conditions of decreasing fall-out (Model A) the relative magnitude of predictions based on the two equations changes progressively, the integrated values over 10 and 50 years being about 10 and 40 per cent higher respectively when derived by equation (1). For Model B the two bases of prediction differ to a smaller extent and the difference would be still smaller if an increasing rate of fall-out were assumed.

#### Caesium-137

Unlike strontium-90, caesium-137 is usually subject to a considerable degree of 'fixation' in the soil on account of its entrapment in the lattice structure of clay minerals and it usually enters plant roots much less readily. It has been suggested that current fall-out is the only significant source of caesium-137 in foodstuffs, but this was disproved in 1960 and 1961. In many countries the decreasing rate of fall-out then caused smaller reductions in the levels of caesium-137 in milk than would have been expected if they were determined solely by the current fall-out<sup>13</sup>. An explanation of the situation seemed to be provided by the fact that caesium-137 can enter plants relatively freely from soils which contain large quantities of organic matter. The surface soil underlying permanent pastures usually shows this characteristic, and experimental. investigations have indicated that caesium-137 continues to enter the edible tissues of pastures relatively freely during the 2 years after its deposition<sup>22</sup>. This led to the suggestion that an equation similar to (1) might be applicable except that the soil factor related only to the deposit in the previous two years<sup>23</sup>, namely:

$$C = p_r F_r + p_d F_d \tag{3}$$

 $A^{(1)}$ 

where C is the 12-month mean concentration of caesium-137 in milk (mc./km<sup>2</sup>/year),  $F_r$  is the annual deposit of caesium-137 (mc./km<sup>2</sup>/year), and  $F_d$  is the deposit of caesium-137 accumulated over the preceding two years (mc./km<sup>2</sup>). The values of the proportionality factors (Table 4) were considerably higher than those for strontium-90 because caesium-137 is transferred from the diet of cattle to milk ten or more times as readily as strontium-90 (ref. 24). Levels of caesium-137 in milk between 1960 and 1963 calculated by equation (3) agreed reasonably with those which were observed (Table 5) and it was found that the body-burden of caesium-137 in man could be related to the pattern of fall-out in a similar manner<sup>13,23</sup>. When, however, the proportionality factors were applied to fall-out data for 1964 the calculated level in milk differed appreciably from that which was observed (Table 5). As the manner in which caesium-137 is initially retained on edible herbage and removed from it by rain differs little from that of strontium-90 (ref. 25), it appeared possible that a lag-rate factor similar to that

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of decreasing fall-out (Model A) predictions based on the two sively, the integrated values about 10 and 40 per cent higher by equation (1). For Model Bon differ to a smaller extent be still smaller if an increasing ued.

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used for strontium-90 in equation (2) might be applicable. The following equation, which differs from equation (2) only in units used, was therefore applied to the survey data for caesium-137 from 1960 until 1964:

$$C = p_r F_r + p_l F_l + p_d F_d \tag{4}$$

where C is the 12-month mean concentration of caesium-137 in milk (pc./l.),  $F_r$  is the annual deposit of caesium-137 (mc./km<sup>2</sup>/year),  $F_i$  is the deposit of caesium-137 (mc./km<sup>2</sup>) in the last half of the previous year, and  $F_d$  is the cumulative deposit of caesium-137 in the soil (mc./km<sup>2</sup>). Values for the rate and lag-rate factors derived from survey data by least squares analysis are shown in Table 4; the soil factor appeared to be zero. Calculated values for the mean level of caesium-137 in milk for each year based on these rate and lag-rate factors agreed more closely with those observed than when equation (3) was used (Table 5).

Although the analysis of the survey data provides no evidence that the cumulative deposit of caesium-137 has contributed hitherto to the contamination of milk, experiments with tracer levels of .caesium-137 indicate that this nuclide continues to enter the roots of plants, though to a small extent, for many years after its entry into the soil. Thus if equation (4) is to be used for predicting levels of caesium-137 in milk in future situations when the cumulative deposit may exceed the annual rate by much larger factors than has occurred hitherto, it is necessary to consider the possible magnitude of the soil factor. Some basis for so doing is provided by the results of experiments on several British soils; 3-5 years after they had been contaminated the ratio in which caesium-137 and strontium.90 were absorbed by plants was on average only one-fortieth of that in the soil<sup>26</sup>. Using

#### Table 4. ESTIMATES OF PROPORTIONALITY FACTORS FOR THE TRANSFER OF CAESIUM-137 TO MILK IN THE UNITED KINGDOM

	Equation 3	Equation 4
Rate factor $(p_r)$ (pc. <sup>137</sup> Cs/l. per mc./km <sup>2</sup> /year)	3.57	3.00
Lag-rate factor $(p_l)$ (pc. <sup>137</sup> Cs/l. per mc./km <sup>2</sup> in	_	6-00
Soil factor $(p_d)$ (pc. <sup>137</sup> Cs/l, per mc./km <sup>2</sup> )	0.65 (relative to deposit	0-03* (relative to cumula-
	in previous 2 years	tive deposit)

• Unlike the other factors, which are based on the analysis of survey data, the soil factor in equation (4) has been deduced from experimental results.

Table 5. COMPARISON OF CALCULATED AND OBSERVED LEVELS OF CAESIUM-137 IN MILK IN THE UNITED KINGDOM

		TH WITTE TH .	THE OWN	UPD RIMOD	om	
Year	Deposi January	tion of caesiu (mc./km <sup>2</sup> ) July to December	m-137 Total	Caesium-137 in milk Observed Calculated values as (pc./l.) percentage of observed Equation 2 Equation		
1959 1960 1961 1962 1963	10.7 1.7 1.2 8.0 14.3	3·1 1·5 2·4 8·0 14·4	13.83.23.616.028.729.3	26 21 62 135 153	98 116 100 86 71	106 96 101 100

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this figure and assuming (a) that caesium-137 is transferred from the diet of cattle to milk about ten times as readily as strontium-90 (ref. 24), and (b) that the average calcium content in milk is about  $1\cdot 1$  g/l., a soil factor for caesium-137 of about  $0\cdot 03$  can be derived from the value for strontium-90 calculated by equation (2) (Table 1). On this basis, the cumulative deposit of caesium-137 would on average have been expected to account for less than 1 per cent of the caesium-137 which has hitherto entered milk from world-wide fall-out. The analysis of survey results for past years could not be expected to reveal this component and at the present time the soil factor can thus be estimated only from laboratory experiments.

The concentrations of caesium-137 in milk which would be predicted on the basis of equations (3) and (4) are compared in Table 3 for the two models of fall-out which have already been outlined; no allowance has been made for losses of caesium-137 from the soil by processes other than the decay of radioactivity because its movement in soil and absorption by crops are much smaller than those of strontium-90. Integrated over the first 10 years, equation (4) leads to predicted values 20-30 per cent higher than those given by equation (3) for both models; over 50 years under conditions of decreasing fall-out the integrated value given by equation (4) is 50 per cent higher. Over this period the soil would contribute about 20 per cent of the total.

## Conclusions

Evidence reviewed in this article indicates that the annual average level of strontium-90 in milk in the United Kingdom is influenced to an appreciable extent by the rate of fall-out in the last half of the previous year as well as by the current rate of fall-out and cumulative deposit. An improved basis for predicting future levels of strontium-90 in milk is therefore provided by an equation which includes a proportionality factor for this 'lag-rate' effect in addition to the rate and soil factors used in former assessments, equation (2). The same type of equation is applicable to caesium-137 (equation 4) though with that nuclide the contribution from the cumulative deposit has been so small that it can be ignored under conditions of fall-out experienced hitherto.

The levels of both nuclides in milk which would be predicted under conditions of fall-out which may be expected during the next decade as a result of weapon testing are not greatly altered when allowance is made for the lag-rate effect. In any year the greatest difference is unlikely to exceed about 20 per cent and averaged over 10 years predictions for strontium-90 would be altered by a smaller amount. This degree of agreement between different methods for prediction encourages confidence

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that caesium-137 is transto milk about ten times as 4), and (b) that the average out 1·1 g/l., a soil factor for n be derived from the value by equation (2) (Table 1). ive deposit of caesium-137 expected to account for less ium-137 which has hitherto le fall-out. The analysis of s could not be expected to it the present time the soil only from laboratory experi-

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des in milk which would be of fall-out which may be lecade as a result of weapon of when allowance is made for year the greatest difference is ) per cent and averaged over rontium-90 would be altered degree of agreement between iction encourages confidence in their use to forecast situations over relatively short periods of years; much greater uncertainty besets many other phases of the assessment of the effects of environmental contamination.

If, however, the integrated levels of dietary contamination are considered over long periods, especially when the rate of fall-out is declining steadily, predictions on the new basis diverge substantially from those which were made formerly. Thus, if the rate of fall-out were to decrease by a factor of two annually, the integrated levels of strontium-90 and caesium-137 in milk over 50 years would be expected on the new basis to be respectively 30 per cent lower and rather more than 50 per cent higher than those formerly predicted; this reflects the reduced soil factor for strontium-90 and the small soil factor now deduced for caesium-137. In consequence the ratio of the integrated level of caesium-137 in milk to that of strontium-90 over this period would be twice that previously calculated.

Long-term predictions of this type are subject to particular uncertainties owing to the lack of direct information on the behaviour of nuclides in soil over long periods. None the less, they deserve consideration as they form the basis for estimating the integrated radiation dose, or dose commitment, which will be received by the population; this is regarded as the best available guide to the total impact of radiation from weapon testing<sup>5,13</sup>. The revised proportionality factors suggest that an appreciably larger fraction of the total dose commitment is due to caesium-137 and a smaller fraction to strontium-90 than has hitherto been assumed.

In conclusion it is to be emphasized, first, that although the method of prediction here outlined appears to give a reasonable description of the average situation, appreciable divergences in individual years must be expected because of climatic variation, and secondly, that the proportionality factors here proposed have been derived for the United Kingdom only. Moreover, variations are to be expected between countries which contrast in climate, soil type and agricultural practices. Thus a recent analysis of relationships for caesium-137 in milk in Sweden<sup>27</sup> gives a rate factor more than three times that calculated for the United Kingdom and a lag-rate factor (which was related to the deposit in the entire previous year) similar to the factor which relates to the deposit during 6 months only in the United Kingdom. Differences, though of lesser magnitude, are apparent between proportionality factors calculated for milk in other countries<sup>5</sup>.

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