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[illegible] 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[2-4] 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[5]. 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[6–12], 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[13]. The country-wide mean levels of radioactivity in milk were taken from the reports of the Agricultural Research Council Radiobiological Laboratory[14–20].

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:

$$C = p_r F_r + p_d F_d \qquad (1)$$

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./km²/year), F_d is the cumulative deposit of strontium-90 at the middle of the year (mc./km²), 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[4,5,21]. 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[3]. 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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tive 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. ^{90}Sr/g Ca per mc./km^2/year)	0·76*	0·70
Lag-rate factor (p_l) (pc. ^{90}Sr/g Ca per mc./km^2 in second half of previous year)	—	1·13
Soil factor (p_d) (pc. ^{90}Sr/g Ca per mc./km^2)	0·19*	0·11

* Calculated from survey data for the period 1958 to 1961 (ref. 28).

Table 2. COMPARISON OF CALCULAT

Year	Deposition of strontium-90 (mc./km²)		
	January to June	July to December	To
1957	1·6	1·5	3
1958	3·2	2·1	5
1959	6·3	1·8	8
1960	1·0	0·9	1
1961	0·7	1·4	2
1962	5·3	5·4	10
1963	9·5	9·6	19
1964	9·6	5·3	14

	to June	December
1957	1·6	1·5
1958	3·2	2·1
1959	6·3	1·8
1960	1·0	0·9
1961	0·7	1·4
1962	5·3	5·4
1963	9·5	9·6
1964	9·6	5·3

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

Model		First year	Years 1–10 Integrated value		Years 1–50 Integrated value	
		Both models	*A*	*B*	*A*	*B*
Ratio of strontium-90 to calcium in milk (relative to year 1 calculated by equation 2)						
Based on equation	1	0·8	4·6	12	10	110
	2	1·0	4·1	12	7	90
Caesium-137 per l. of milk (relative to year 1 calculated by equation 4)						
Based on equation	3	0·8	1·9	8	1·9	40
	4	1·0	2·6	10	3·0	54

Model *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.

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[13]. 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[22]. 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[23], namely:

$$C = p_r F_r + p_d F_d \qquad (3)$$

where C is the 12-month mean concentration of caesium-137 in milk (mc./km^2/year), F_r is the annual deposit of caesium-137 (mc./km^2/year), and F_d is the deposit of caesium-137 accumulated over the preceding two years (mc./km^2). 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[13,23]. 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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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[26]. 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. ^{137}Cs/l. per mc./km^2/year)	3·57	3·00
Lag-rate factor (p_l) (pc. ^{137}Cs/l. per mc./km^2 in second half of previous year)	—	6·00
Soil factor (p_d) (pc. ^{137}Cs/l. per mc./km^2)	0·65 (relative to deposit in previous 2 years only)	0·03* (relative to cumulative 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

Year	Deposition of caesium-137 (mc./km^2)			Caesium-137 in milk		
	January to June	July to December	Total	Observed (pc./l.)	Calculated values as percentage of observed: Equation 3	Equation 4
1959	10·7	3·1	13·8	—	—	—
1960	1·7	1·5	3·2	26	98	106
1961	1·2	2·4	3·6	21	116	96
1962	8·0	8·0	16·0	62	100	101
1963	14·3	14·4	28·7	135	86	100
1964	14·3	8·0	22·3	153	71	100

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 1·1 g/l., a soil factor for caesium-137 of about 0·03 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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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[5,13]. 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[27] 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[5].

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