UPTAKE OF RADIONUCLIDES BY FARM ANIMALS CLOSE TO A MAJOR NUCLEAR INSTALLATION

T J Sumerling, N Green and N J Dodd
National Radiological Protection Board
Chilton, Didcot, Oxfordshire

INTRODUCTION

A field investigation of the transfer of artificially produced radionuclides in the pasture-cow-milk pathway has been made at a farm close to the nuclear fuel reprocessing installation at Sellafield on the north-west coast of England. The routine discharges from the plant have resulted in enhanced levels of several artificial radionuclides in the local environment. For example, during the period of this investigation the annual depositions of ^{90}Sr and ^{137}Cs at the farm were a factor of about five higher than the average deposition of these radionuclides in the United Kingdom (1). Even if extremely cautious assumptions concerning local eating habits are made, the consumption of meat and dairy products from this farm would give rise to an annual activity intake of less than one percent of the limit for adult members of the public (2). Nevertheless, the presence of enhanced levels of certain artificial radionuclides provides a rare opportunity to study their transfer in the environment.

METHODS

The methods of sample collection and analysis have been described in detail elsewhere (1, 3). Briefly, samples of airborne particulates, rainwater, herbage, supplementary feed and milk were collected at monthly or two-weekly intervals over a period of two years. Samples of soil and cattle faeces were collected at less frequent intervals. At the end of each year, a single cast dairy cow was sacrificed to provide tissue samples. In order to estimate and minimise the uncertainties due to variations between individual samples, multiplicate samples were taken of all media except airborne particulates and rainwater.

After appropriate pretreatment, e.g. concentration by evaporation or drying and milling, the content of ^{137}Cs and other gamma-ray emitting nuclides of all samples was measured with germanium or sodium iodide detectors. The ^{90}Sr content of bulked feed samples, representing intake in each month, and individual samples of rainwater, milk, soil and tissues was determined by an yttrium separation method followed by β particle counting. The stable calcium content of aliquots from the bulked feed, milk and tissue samples was determined by a colorimetric method. Samples of rainwater, feed and milk were each combined to form representative quarterly samples. These and individual samples of soil and tissues were analysed by a radiochemical separation technique followed by α particle spectrometry to determine the content of plutonium isotopes and ^{241}Am. The daily fodder intake of the herd was estimated from a consideration of the theoretical dry matter and metabolisable energy requirements of the cows (1).

RESULTS

Coefficients of transfer from feed to milk, F_m, were calculated by the formula

$$F_m = \frac{\text{concentration of radionuclide in milk (Bq l}^{-1}\text{)}}{\text{daily intake of radionuclide (Bq d}^{-1}\text{)}}$$

For ^{90}Sr and ^{137}Cs, this was calculated for each month over a two year period; the mean of the monthly coefficients for each major feeding period are shown in Table 1. The concentrations of plutonium isotopes and ^{241}Am in milk were very low and usually did not exceed the limit of detection. Hence, only maximum values of F_m could be calculated for these radionuclides.

Similarly coefficients of transfer from feed to flesh (meat or liver) F_f, were calculated by the formula

$$F_f = \frac{\text{specific activity of radionuclide in flesh (Bq kg}^{-1}\text{)}}{\text{daily intake of radionuclide (Bq d}^{-1}\text{)}}$$

In this case the activities in flesh are those determined in tissues from the two slaughtered animals and the daily intake was taken to be the mean daily intake during the year preceding slaughter. Table 2 shows best estimates of the coefficients of transfer to meat, liver and milk for each of ^{90}Sr, ^{137}Cs, ^{239}Pu and ^{241}Am; for ^{90}Sr and ^{137}Cs these are equilibrium coefficients, for ^{239}Pu and ^{241}Am these refer to activity at the time of slaughter.

Table 3 shows mean specific activities of ^{239}Pu and ^{241}Am in tissues from two cast dairy cows, sacrificed at six years of age. Estimates of the total activities present in lung, liver, meat and bone are also given. During the two years of the study, the average annual intake by ingestion of each cow of the dairy herd was estimated to be 10 kBq a^{-1} of ^{239}Pu and 5 kBq a^{-1} of ^{241}Am, 90% of which was derived from the consumption of contaminated herbage and soil during the grazing season. The average annual intake by inhalation of each cow was estimated to be 0.16 Bq a^{-1} of ^{239}Pu and 0.07 Bq a^{-1} of ^{241}Am. This estimate is based on measurements of the concentration of these nuclides in air and assumes an inhalation rate of 1.5×10^{-3} m^3 s^{-1}. Measurements of titanium (a soil trace element) in lung, muscle and soil samples indicated that inhalation of soil did not account for the measured activities of ^{239}Pu and ^{241}Am in lung tissues.

DISCUSSION

Strontium-90

The calculated daily intake of ^{90}Sr by the herd varied markedly during each year and was, on average, a factor of three higher during the grazing season than during the winter periods due to the differences in specific activities of pasture herbage and the stored feeds that were supplied. However, the concentration of ^{90}Sr in milk was relatively constant and varied only by a factor of about 50%. The daily intake of stable calcium and concentration of stable calcium in milk were both found to be relatively constant and could not account for the changes in ^{90}Sr transfer coefficient. The observed ratio, OR, of $^{90}Sr/Ca$ in milk divided by $^{90}Sr/Ca$ in daily intake was found to be unusually high during the winter months (OR = 0.4) but lower during the summer months (OR = 0.1, that is within the range found by other investigators). It was concluded that the concentration of ^{90}Sr in milk was being supported both by recent intakes and by activity remobilised from bone; during the summer months a dynamic equilibrium was reached but during the winter months the activity in milk was supported mainly by remobilised activity.

Caesium-137

On a monthly basis, the concentration of ^{137}Cs in milk followed the calculated daily intake very closely with no evidence for a long term retention component. However, as shown in Table 1, coefficients of transfer obtained for the summer months were a factor of about two lower than those obtained for the winter months. The specific activity of ^{137}Cs in surface soil at the farm is relatively high (200 to 300 Bq kg^{-1} dry weight) so that small traces of soil adhering to the surface of herbage can make a significant contribution to the total measured activity of the herbage. Measurements of herbage sample weight after ashing and measurements of titanium in herbage and soil samples were used to quantify the level of soil contamination on herbage and other feed samples. It was found that the presence of soil on feed samples accounted for between 30 and 60% of the total measured ^{137}Cs activity of herbage but less than 5% of the total measured ^{137}Cs of stored feeds. Measurements ofthe physical and chemical disposition of ^{137}Cs in soil from the farm showed that 85% of the ^{137}Cs was permanently bound to silicaceous mineral particles and would be totally unavailable for biological uptake. It was concluded that the inclusion in daily intake of ^{137}Cs associated with soil on herbage surface could account for the lower coefficients of transfer observed during the summer months. In order to calculate the coefficients of transfer for ^{137}Cs shown in Table 2 daily intake was recalculated excluding the fraction of ^{137}Cs that was estimated to be bound to mineral particulates.

Plutonium-239 and Americium-241

Even if a relatively low gut uptake factor and a relatively high fractional uptake from the respiratory tract are assumed it would appear that intake by ingestion was responsible for the bulk of the systemic activity of these radionuclides in cattle on the farm. The distribution of ^{239}Pu between liver and bone was very similar to that found after experiments in which dairy cows were orally dosed with ^{238}Pu in oxide and citrate forms but the proportion of activity present in meat is approximately twice that found following oral dosing with ^{238}Pu citrate and almost on order of magnitude greater than that found after oral dosing with ^{238}Pu oxide (4). The ratios of ^{239}Pu and ^{241}Am activities in meat to activities in liver and bone were approximately an order of magnitude higher than found in animals grazing on more heavily contaminated pastures in the area (5). It is possible that at these very low levels of intake a greater proportion of systemic intake does indeed become deposited in meat but it should be noted that the absolute levels of activity are very low and subject to analytical and sampling uncertainties. If it is assumed that there has been negligible systemic clearance of plutonium and americium from these animals and also that their intake in previous years was similar to that calculated for the two years of the study, then it is possible to estimate fractional gut uptake values for these animals of 8×10^{-5} for ^{239}Pu and 2×10^{-4} for ^{241}Am. These values are lower than found in oral dosing experiments (4) but higher than usually associated with these radionuclides.

REFERENCES

1. Sumerling T. J., Dodd, N. J. and Green N., The transfer of ^{90}Sr and ^{137}Cs to milk in a dairy herd grazing near a major nuclear installation. To be published in Sci. Tot. Environment.
2. Sumerling, T. J. and Crick M. J., A preliminary evaluation of a dynamic model for the transfer of radionuclides in the pasture-cow-milk pathway with data from a field investigation, in Proc. CEC Seminar. The transfer of radioactive materials in the terrestrial environment subsequent to an accidental release to atmosphere, Dublin 1983.
3. Green N., Radiochemical analysis of samples from an environmental pathway study, Laboratory of the Government Chemist 4th Symposium on the Determination of Radionuclides in Environmental and Biological Materials, 1983.
4. Stanley R. E., Bretthauer, E. W and Sutton, W. W., Absorption, distribution and excretion of plutonium by dairy cattle, in Radioecology of Plutonium and Other Transuranics in Desert Environments. Eds. White M. G. and Dunaway P. B., NVO 153 1975.
5. Popplewell, D. S. P., Personal Communication, 1983.

Table 1: Mean of monthly coefficients of transfer from feed to milk, F_m, for ^{90}Sr and ^{137}Cs.

Season	Feed	F_m, d l^{-1}	
		^{90}Sr	^{137}Cs
Winter 1980/81	hay and concentrates	3.8×10^{-3}*	6.4×10^{-3}
Summer 1981	grazed pasture	1.2×10^{-3}	3.8×10^{-3}
Winter 1981/82	silage and concentrates	2.9×10^{-3}*	7.2×10^{-3}
Summer 1982	grazed pasture	1.0×10^{-3}	3.4×10^{-3}
Winter 1982/83	silage and concentrates	---	7.4×10^{-3}

* not in equilibrium, see discussion.

Table 2: Best estimates of coefficients of transfer from feed to meat, liver and milk.

Product	Coefficient of transfer, d kg^{-1} or d l^{-1}			
	^{90}Sr	^{137}Cs*	^{239}Pu	^{241}Am
Meat	5×10^{-4}	1.4×10^{-2}	3×10^{-4}	5×10^{-4}
Liver	--	9×10^{-3}	1.6×10^{-3}	2.1×10^{-3}
Milk	1×10^{-3}	7×10^{-3}	$< 7 \times 10^{-6}$	$< 1 \times 10^{-4}$

* excludes intake of ^{137}Cs bound to mineral particulates, see discussion.

Table 3: Mean specific activities and total activities of ^{239}Pu and ^{241}Am in tissues from two slaughtered cows.

Tissue	Mean Weight kg	Specific activities, mBq kg^{-1}		Total activities, Bq	
		^{239}Pu	^{241}Am	^{239}Pu	^{241}Am
Lungs (2)	4.1	12	7	0.05	0.03
Liver	8.4	54	29	0.45	0.24
Meat	140*	6	4	0.8	0.6
Bone	70*	51	78	3.6	5.5

*estimated from dressed carcase weight, excludes head and hooves.