A STUDY OF RADIOCESIUM CONTAMINATION AND DECONTAMINATION OF SHEEP'S MILK

P.A. ASSIMAKOPOULOS¹, K.G. IOANNIDES¹, A.A. PAKOU¹ and A.S. MANTZIOS²
¹Nuclear Physics Laboratory, The University of Ioannina, 451 10 Ioannina,Greece.
²Agricultural Research Station of Ioannina, P.O. Box 1103, 451 10 Ioannina, Greece

SUMMARY
The radiocesium contamination and decontamination of sheep's milk were studied under a constant level of activity concentration in the sheep's diet. Two sets of experiments were performed: one at the end of the animal's lactating period and one during the main lactating period. The data were in satisfactory agreement with the predictions of a simple two-compartment model. At the stage of equilibrium the data yielded the transfer coefficient $f_m$ with an average value of $f_m = 0.063 \pm 0.005 \text{ d}^{-1}$. In the second experiment a detailed study of the decontamination phase revealed a two-component decay with amplitudes 53% and 43% and half-lives 1.5 d and 6.9 d, respectively. A small 4% long-lived ($T_{1/2} = 170 \text{ d}$) third component could not be distinguished from an overall background decay, measured in control animals.

INTRODUCTION
The transport of radionuclides from a lactating animal's diet to its milk has been the subject of several investigations in the past (refs. 1-6). Such studies indicate that, to a first approximation, a two-compartment model can adequately describe the experimental results if the animal's daily diet contains a constant level of radioactive contamination.

In the research presented here this simple model was investigated in a realistic situation following the nuclear accident at Chernobyl. A secondary purpose of this research was the determination of the transfer coefficient from a sheep's diet to its milk in order to assess milk contamination levels during the winter lactation period expected from sheep feeding on stockpiled food of known radioactivity concentration.

The first experiment was conducted during the month of August 1986. Since during that period the animals were near the end of the lactating period the decontamination process was not adequately sampled. For this reason the experiment was repeated in the Spring of 1987, with emphasis on the decontamination process.

THEORY
According to the two-compartment model, when radioactive contamination is introduced into the diet of a lactating animal at a constant rate $P(\text{Bq d}^{-1})$, the
ensuing milk contamination $C(t)$ (Bq l$^{-1}$) is described by the differential equation (see, e.g. ref. 4)

$$\frac{dC}{dt} = \gamma P - \lambda C,$$

in which $\gamma$ (L$^{-1}$) is the transfer rate of contaminant from the animal's diet to its milk and

$$\lambda = \lambda_M + \lambda_R.$$ 

In the last expression, $\lambda_R$ (d$^{-1}$) is the radioactive decay rate of the isotope and $\lambda_M$ the rate of transfer to the udder. For long-lived radionuclides, such as $^{134}$Cs and $^{137}$Cs, $\lambda_R$ does not contribute significantly to the total decay rate and may be omitted in eqns (1) and (2).

The solution of eqn (1) for constant daily contamination intake $P$ may be written as

$$C(t) = \frac{\gamma P}{\lambda} (1 - e^{-\lambda t}) + S,$$ 

in which $S$ is a constant contamination level arising from sources other than the known contaminating agent in the animal's diet. This is an increasing function which soon (within a time interval of a few $\lambda^{-1}$) reaches asymptotically a constant value $\gamma P/\lambda + S$. Thus, in the state of equilibrium, the constant transfer of contamination from the animal's diet to its milk is governed by the relation

$$C_{eq} = f_m P,$$ 

where the transfer coefficient $f_m$ is given by

$$f_m = \frac{\gamma}{\lambda}.$$ 

If at a later time $t_0$ the contaminating agent is removed from the animal's diet, the contamination level of the milk produced will correspondingly decrease. In this case ($P = 0$) eqn (1) yields a decreasing exponential behavior for $C(t)$. We may thus write the complete solution of eqn (1) during the two time intervals as

$$C(t) = \frac{\gamma P}{\lambda} (1 - e^{-\lambda t}) + S, \quad t < t_0,$$

$$C(t) = [C(t_0) - S] e^{-\lambda (t - t_0)} + S, \quad t > t_0.$$ 

In this solution again a constant background contamination level $S$ is assumed in the milk which arises from all sources other than $P$.

It should be noted that although a simple decay is predicted by eqn 6 during the decontamination process, several authors have in the past reported decontamination of the milk according to a multi-exponential function of the form

$$C(t) = \sum_{i=1}^{N} A_i \exp(-\lambda_i t).$$
METHOD AND MATERIALS

Two experiments were conducted, details of which are shown in Table 1. The ewes were segregated from a herd at the Ioannina Agricultural Research Station and held in isolated pens for the duration of the investigation. The animals in experiment I were maintained on the contaminated diet for 20 days and in experiment II for 50 days. They were subsequently returned to a contamination free diet. Uncontaminated food was given to the control group in experiment II throughout the duration of the investigation.

The milk obtained daily was measured for radioisotope contamination at the Nuclear Physics Laboratory of the University of Ioannina. Samples were measured in a standard geometry of 400 mL with a 1.9 keV resolution (for the 661.65 keV line of $^{137}$Cs), 18% efficiency, intrinsic Ge detector. The detector was shielded with 5 cm of lead against background radiation. Standard electronics were used, and the spectra were accumulated in 1024 channels. The analysis was performed on the Nuclear Physics Laboratory's computer, with a modified version of code ANNA (ref. 7). The detector was calibrated for efficiency versus energy with a standard 129.5 kBq L$^{-1}$ $^{152}$Eu source. The time required for the accumulation of a spectrum with adequate statistics ranged from 1000 s to 5000 s.

Several spectra of grass samples were also accumulated and analyzed to obtain estimates of radiocesium concentration.

RESULTS

The radiocesium concentrations measured in the animals' milk are presented in Fig. 1 and Fig. 2.

Fig. 1 contains data of experiment I from both the contamination and decontamination process. These values were fitted to eqn 6 by treating the

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Experimental details.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experiment</td>
<td>I</td>
</tr>
<tr>
<td>Number of animals in each experiment</td>
<td>10</td>
</tr>
<tr>
<td>Number of animals in the control group</td>
<td>—</td>
</tr>
<tr>
<td>Weight of animals</td>
<td>36</td>
</tr>
<tr>
<td>Yield of milk/day/animal (mL)</td>
<td>110</td>
</tr>
<tr>
<td>Radiocesium intake (Bq d$^{-1}$)</td>
<td>$832 \pm 50$</td>
</tr>
</tbody>
</table>
Fig. 1. Total radiocesium activity concentration in sheep's milk during the contamination and decontamination phase of experiment I. The solid curve represents the best fit of the expression in eqns (6),(7).

Fig. 2. Total radiocesium activity concentration in sheep's milk during the decontamination phase of experiment II. The solid curve represents the best fit of the expression in eqn (8). The three solid straight lines are the contribution of each term in eqn (8). The dashed straight line is a background component, obtained from the monitoring group.
quantities $v, \lambda$ and $S$ as free parameters. The best fit ($\chi^2 = 1.6$ confidence level 95\%) is plotted as a continuous curve in Fig. 1 and corresponds to the values of the free parameters

\begin{align*}
v &= 0.018 \pm 0.003 \, \text{L}^{-1} \\
\lambda &= 0.31 \pm 0.05 \, \text{d}^{-1} \\
S &= 18 \pm 5 \, \text{BqL}^{-1}.
\end{align*}

The transfer coefficient obtained from these values and eqn (6) is

$$f_m = 0.058 \pm 0.007 \, \text{dL}^{-1}$$

while the decay half life during decontamination is $(2.24 \pm 0.36)\,\text{d}$.

Fig. 2 shows the data from experiment II during the decontamination period. These values suggest that the excretion of cesium $C(t)$ into milk may be described by a sum of decaying exponential functions. The number of terms involved in the sum (see eqn (7)) was determined by successive fits to the data for functions with $N = 1, 2, 3, \ldots$. It was established in this manner that for $N \geq 3$ the best fit of eqn (7) yielded only three distinct parameters $\lambda_i$. The data in Fig. 2 were therefore fitted to a three-component decay function. The best fit ($\chi^2 = 1.35$, confidence level 95\%) is plotted as a continuous curve through the data and corresponds to a three-component decay with amplitudes $(53 \pm 21)\%$, $(43 \pm 8)\%$, $(4 \pm 1)\%$ and half-lives of $(1.5 \pm 0.4)\,\text{d}$, $(6.9 \pm 0.7)\,\text{d}$, $(173 \pm 87)\,\text{d}$, respectively. The small third component, could not be distinguished from effects of background decay observed in the control group. The best fit on these control data are shown as a dashed line in Fig. 2 and might be interpreted as a long-term residual decay from the summer of 1986 when the entire herd was grazing freely on pastures affected by the Chernobyl fallout.

The transfer coefficient obtained in experiment II was

$$f_m = 0.071 \pm 0.009 \, \text{dL}^{-1}$$

in very good agreement with the result of experiment I (eqn (11)). Thus an average value was calculated as

$$f_m = 0.063 \pm 0.005 \, \text{dL}^{-1}$$

**DISCUSSION AND CONCLUSIONS**

The experimental results contained in Fig. 1 are seen to confirm the applicability of the two-compartment model for the prediction of milk contamination when a constant daily amount of contaminating agent is introduced into the animal's diet. It is noted that in this model the build-up of radioactivity concentration following the introduction of contamination in the animal's diet and the decay following the removal of contamination are both governed by the same rate constant. This is confirmed within the accuracy represented
by the data in Fig. 1.

Transfer coefficients measured in our two experiments are in very good consistency. These values are also in very good agreement with other results in sheep milk (ref. 8).

The decay constants obtained in this research can be compared with literature values for Cs excretion measured in sheep, although reported results vary considerably. A three-component excretion pattern has been observed by Twardock and Crackel (ref. 9) with average half-lives 1.09, 5.27 and 25 d, accounting for the fractional excretion of the initial dose of 0.26, 0.34 and 0.41 respectively. On the other hand, Buldakov (refs. 9-10) has reported an excretion rate which could be described as the sum of only two exponential components with half-lives of 24.4 and 49.5 d for the excretion of 36% and 64% of the initial burden respectively.

The two-component exponential decay observed in the data of Fig. 2, suggests excretion of cesium from two distinct compartments. The nature of these compartments is, however, uncertain. Multiple-component exponential decay might, for instance, represent release of cesium, trapped in different organs of the animal, which exhibit different retentive properties. A more plausible explanation is that the two-component decay represents release from the intracellular and extracellular volumes. If this is the case, then multi-compartment behaviour should appear upon closer scrutiny of the contamination phase. Further, the relative amplitude of the two components in the decontamination phase should depend on the duration of the equilibrium phase. These factors will be investigated in a series of forthcoming experiments.

REFERENCES

2 F.O. Hoffman, A review of measured values of the milk transfer coefficient ($f_m$) for iodine, Health Phys., 35 (1978) 413-416.
