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Countermeasures for animal products: a review of effectiveness and potential usefulness after an accident

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Abstract

Over the last decade, there has been considerable progress in the development of countermeasures for preventing or reducing contamination of animal products by radio-isotopes of iodine, caesium and strontium. In this paper, recent significant technical improvements are summarised and the current availability of countermeasures and their usefulness in the event of a nuclear accident reviewed. An improved understanding of factors controlling the metabolism of radioiodine and radiostrontium has enabled previously suggested countermeasures to be either optimised or dismissed. For radiocaesium in particular, experience since the Chernobyl accident has enabled effective and feasible countermeasures to be identified and successfully implemented in different situations. It has also been more widely understood that countermeasure effectiveness, although important, is not the only criterion which needs to be determined. In addition, cost and practical considerations such as availability, technical feasibility, acceptability and side-effects need to be taken into account. Evaluation of these factors has shown that some previously recommended countermeasures are unlikely to be feasible. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Countermeasures; Radioiodine; Radiocaesium; Radiostrontium; Review; Ruminants

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1. Introduction

Animal products generally contribute substantially to the ingested dose after radioactive contamination of the environment. The need, and resources available, to develop and improve countermeasures to reduce internal dose are inevitably connected to the occurrence and severity of accidental releases. Thus, many studies were conducted on radioiodine and radiostrontium, in particular, in the 1950–1970s in response to the Windscale and Kyshtym accidents and to the development and testing of nuclear weapons. Probably the greatest impetus to the development of countermeasures occurred in response to the Chernobyl accident. Much of the focus in the first few years after the Chernobyl accident was, by necessity, on development and application of effective countermeasures for radiocaesium in agricultural and forest ecosystems. The development and implementation of these countermeasures was described within Howard and Desmet (1993); this compilation included some of the first peer-reviewed literature of results from the former Soviet Union (FSU). Subsequently, the International Atomic Energy Agency (IAEA) Technical Report on Guidelines for Agricultural Countermeasures (IAEA, 1994) was published which provided an overview of strategies for the introduction of agricultural countermeasures. In addition, the results of a large European Commission (EC) funded series of projects (Karaoglou, Desmet, Kelly, & Menzel, 1996) were reported which covered countermeasure applications in the FSU countries after the Chernobyl accident. There has been significant further progress with respect to countermeasure techniques not only for radiocaesium, but also for other mobile radionuclides. These studies have focused not only on effectiveness, but also on other issues such as cost, acceptability and practical considerations (e.g. availability, feasibility and side-effects) (Voigt et al., 2000). Furthermore, improved mechanistic understanding of the behaviour of radionuclides in animals has allowed the effect of some countermeasures to be incorporated into predictive models (Crout, Beresford, Howard, Mayes, & Hansen, 1998; Crout et al., 2000).

The aim of this paper is to provide a review of recent developments concerning countermeasures for reducing radioactive contamination of terrestrial animal products for the three most important mobile radionuclides, namely radiocaesium, radioiodine and radiostrontium. Recommendations are made on the most appropriate, currently available, countermeasures and requirements for further development. Information reviewed excludes a detailed consideration of treatment of soil to reduce contamination of animal feed, although the effectiveness of these measures are compared to animal-based techniques in the text and considered in the conclusions and recommendations. The focus here is on ruminants because contamination of their products is important for all three radionuclides; countermeasures for contamination of non-ruminants are also considered for radiocaesium.

2. Countermeasure categories

Countermeasures for reducing internal dose from animal products (excluding those aimed at reducing the contamination of animal feed) principally fall into three major categories (Skuterud, Strand, & Howard, 1997)

(a) Restrictions on use of contaminated animal products by humans

1. Ban on entry of contaminated products into the foodchain
2. Monitoring of animal-derived products (monitoring of live animals and/or of milk and tissues after slaughter)
3. Dietary advice for consumers (to change consumption habits or use recommended food preparation techniques)

(b) Animal management

1. Provision of uncontaminated or less contaminated feedstuff, preferably to housed animals, especially shortly before slaughter
2. Remove animals from contaminated to less contaminated land
3. Selective grazing of available pasture
4. Change slaughter times especially for wild ranging animals such as deer
5. Change from milk to meat production
6. Change animal species to one in which the transfer of radionuclides is lower (e.g. from goats to cows)

(c) Use of additives to reduce or prevent contamination in the animal

1. Administration of binders which prevents or reduces gut uptake
2. Administration of a stable element or analogue

Most recent information relates to the use of additives, but there have also been developments under the other categories.

3. Non-radionuclide-specific countermeasures

The provision of uncontaminated feed or pasture to previously contaminated animals for an appropriate period before slaughter (often termed "clean feeding") has been one of the most frequently used countermeasures for agricultural animals in both the FSU and Western Europe after the Chernobyl accident to derive products with radionuclide activity concentrations below internationally or nationally agreed intervention levels (Firsakova, Zhuchenko, Fesenko, Kuchma, & Dvornik, 1998; Tveten, Brynildsen, Amundsen, & Bergan, 1998; Åhman, 1999; Nisbet & Woodman, 2000). Clean feeding has two benefits: it reduces uptake of the contaminating radionuclides and allows the reduction of those radionuclides already incorporated into the body.

The EC has previously published recommended Maximum Permitted Levels (MPLs) of radiocaesium in marketed animal feedingstuffs (CEC, 1990) intended to prevent activity concentrations in animal products exceeding intervention levels (CEC, 1989). Because these MPLs take little account of national or regional variation in relevant factors affecting transfer, and only consider commercial feedstuffs, a recent study has assessed their applicability in the UK (Woodman & Nisbet, 1999). The study demonstrated that detailed information on the major contributors to radionuclide intake within the diet of agricultural animals would be required for effective management in the event of an accident in many countries. Results of the study for radiocaesium and radiostrontium are discussed below.

Pasture grass, hay or ensilaged crops are often important contributors to radionuclide intake. Therefore, the roughage ration could be replaced with grain-based feedstuffs which would be less contaminated than forage crops growing in the same contaminated soil (Salt et al., 1999a). However, there is a limit to the percentage contribution these could make to the diet. For instance, the animals health may be detrimentally affected (such as parakeratosis and acidosis) if the percentage contribution of concentrates in the diet of dairy cattle is increased above 60–70% (Wattiaux & Howard, 1999). Such feeding practices can also lead to impairment in the quality of the animal-based food products (e.g. reduced fat content of cow's milk; "soft" fat in lamb carcasses). In the United Kingdom (UK), the advised percentage of concentrates in the diet of dairy cattle ranges from about 20% (summer) to 33% (winter) (Nix, 1994) and hence could potentially be doubled . Many of the feeding problems associated with feeding high-grain diets relate to the period of changeover from a high-roughage to a high-concentrate diet; few problems are evident in animals fed high-cereal diets over a longer period.

Although conceptually dietary manipulation may appear simple, implementation of this countermeasure requires consideration of a large range of factors. For example, when considering the cost-effectiveness, direct costs of the "clean" feed itself, transportation, enclosures or housing if required, duration of special feeding (which will depend on the contamination level and biological half-life), regulatory management, liaison with farmers and compensation all need to be evaluated. Differences in the costs of the feedingstuffs and transport affect the relative merits of bringing in uncontaminated grain or roughage to contaminated areas compared to using on-site feedingstuffs. Because the nutrient density of grain is higher than that of roughage, the transport cost of the former is lower than that of roughage; however, roughage is a cheaper feed than grain. The combined costs of these factors may make this approach less cost-effective than other countermeasures and its application is highly dependent on availability of suitable feedstuffs, roughage or pasture land. Where uncontaminated feedstuffs are not nationally available in sufficient quantities, they may need to be imported. In the FSU, importation of uncontaminated feedstuffs after the Chernobyl accident was precluded because of the high additional costs involved (Nisbet, 1995).

Various potential side effects of dietary manipulation and the associated potential housing requirement have been identified (Salt et al., 1999a; Salt, Solheim Hansen, Kirchner, Lettner, & Rekolainen, 1999b; Nisbet & Woodman, 2000). Countermeasures which involve more intensive livestock feeding regimes and longer periods of housing could be perceived as detrimental to animal welfare. For instance, housing of animals will increase the risk of diseases such as footrot and Pasteurella. Similarly, Tveten et al. (1998) reported problems associated with the clean feeding of semi-domesticated reindeer which are not accustomed to being enclosed and hence have to be gradually adjusted to the management regime and feedstuffs to avoid sickness. Occasionally, individual animals are "mobbed" by other animals and need to be separated. If the proportion of cereals fed to animals was increased a number of possible environmental side effects could occur including enhanced soil erosion, increased inputs of phosphorus and nitrogen and reduced biodiversity (Salt et al., 1999a).

The extent of such side-effects will be highly dependent on the severity and scale of the contamination event and the extent to which farming systems are adapted. For an increased use of cereals, many side-effects could be avoided if cereals were imported from uncontaminated areas with excess production.

4. Review for selected radionuclides

4.1. Radiocaesium

About 80% of ingested plant-associated radiocaesium is absorbed from the ruminant gut and is subsequently transported to all soft tissues, milk, urine and faeces (Beresford et al., 1995; Mayes, Beresford, Howard, Vandecasteele, & Stakelum, 1996). The muscle and milk represent the most important sources of radiocaesium from animal products entering the human foodchain.

In the early phase after an accident, radiocaesium is intercepted directly on vegetation surfaces, during both dry or wet deposition, and consumed by animals. As radiocaesium is transferred into the soil, the importance of soil-to-plant uptake increases, with the highest uptake occurring in areas with highly organic or sandy soils; hence long-term problems may not only occur in the areas of highest contamination. The persistent uptake of radiocaesium in some contaminated areas after the Chernobyl accident (Smith et al., 2000) has prompted the development and refinement of a wide range of different countermeasures for ^{137}Cs contamination of animals.

Using information on typical animal diets, relative activity concentrations in different feedstuffs and feed-to-animal product transfer, Woodman and Nisbet (1999) found that in the UK only one or two feedstuffs typically contribute 5% or more to intakes of radiocaesium, and that grass and silage were particularly important sources of intake for animals. Their study concluded that specific guidance was needed for the UK since the "working levels" (similar to the EC MPL values) derived for radiocaesium in important feedstuffs ranged from 22-fold lower to 20-fold higher than the EC MPL. In most cases, the MPL was too cautious, implying that feedstuffs in excess of the MPL could be fed to animals and their resultant products would be below intervention levels for the human foodchain. Clearly, the dietary composition of agricultural animals varies between countries, but feed to animal transfer has also been shown to vary (Beresford et al., 2000a). Thus, due to the spatial variation in transfer rates and local influences such as soil type, animal breed, management system and production level, specific working levels may need to be derived for different countries (or even regions).

During clean feeding of previously contaminated animals, studies of growing lambs have demonstrated that an increased dry matter intake results in a more rapid decrease in radiocaesium activity concentration in muscle (Howard et al., 1995a).

This observation was probably the consequence of a higher live-weight gain of those lambs receiving greater quantities of feed.

There have been many studies on different binders for radiocaesium which reduce absorption in the gut of both ingested and recirculated radiocaesium (Hove, 1993; Voigt, 1993; Åhman, 1996). Two main classes of compounds have been considered, clay minerals and hexacyanoferrates.

There has been relatively little recent work on the use of clay minerals as radiocaesium binders, with the exception of work with reindeer, where Åhman (1996) reported that administration of $25\,g\,d^{-1}$ of bentonite to reindeer reduced radiocaesium transfer by three-fold. Åhman noted that, although higher doses may give greater reductions, this may cause problems of increased water requirements in reindeer. Bentonite has the advantage of being relatively cheap and readily available and was used in Sweden in the first few years after the Chernobyl accident to decrease the biological half-life of radiocaesium in reindeer by ca. 20% during clean feeding (Åhman, 1996), but the cost was considered to be high relative to the "extra" effect so the practice was discontinued. Since then, clean feeding and alteration of slaughter times have been the main countermeasures for reindeer in Sweden (Åhman, 1999).

The most effective binders to reduce radiocaesium absorption in the gut are the hexacyanoferrate compounds commonly referred to as "Prussian Blue". Of these, ammonium-ferric(III)-cyano-ferrate (II) (AFCF) has been commonly tested in Western Europe, although the commercially available compound is relatively expensive. In 1996, the EC passed regulations allowing the use of AFCF as an animal feed additive to reduce radiocaesium absorption in the European Union (CEC, 1996); these regulations are effective until 2001. Many different formulations of hexacyanoferrates have been developed in different countries, partially to identify the most effective compound and partly to produce a cheaper, locally available product. Most such studies have confirmed that hexacyanoferrate compounds are highly effective binders (e.g. Ioannides et al., 1996; Ratnikov et al., 1998).

In Western Europe, much of the long-term problem after the Chernobyl accident has been connected with free-ranging or wild animals. To cope with this, delivery systems which do not need daily access to animals were developed in which the hexacyanoferrate binder is incorporated into different matrixes, including rumen-dwelling boli and salt licks (Hove, 1993). The original boli, developed in Norway (typically 16–20 mm in diameter and 50–65 mm length) consisted of a compressed mixture of 15% AFCF, 10% beeswax and 75% barite (Hove & Hansen, 1993). The design has recently been modified, by adding a protective surface coating of wax, to delay the onset of AFCF release so that their effectiveness is increased at the time when animals are collected for slaughter (Hansen, Hove, & Barvik, 1996). By doing this, the duration of effect in free-ranging sheep was enhanced from 43–75% over 4–8 weeks for an unmodified boli to 48–65% over 9–11 weeks for the wax-coated boli (based on three boli per animal). Despite the high relative cost of AFCF, costs of boli production and animal handling costs, Brynildsen, Selnaes, Strand, and Hove (1996) estimated that the use of boli as a countermeasure for sheep was 2.5 times as cost-effective as feeding with uncontaminated feed. In the UK, the boli used in Norway were too large to administer to upland lambs. Therefore, smaller boli (14 mm in diameter and 50 mm length) with a 20% AFCF (the maximum possible to retain bolus cohesion) and a wax coating have been developed (Beresford et al., 1999a) and successfully used on an upland farm (Beresford et al., 1999b). A reduction of $>30\%$ in the muscle radiocaesium activity concentrations of lambs treated with three boli was achieved, 51 d after administration at the time when the lambs would be sold from the farm. In the FSU, the high cost of imported AFCF was prohibitive. Therefore, a locally manufactured hexacyanoferrate called ferrocyn (a mixture of 5% $KFe[Fe(CN)_6]$ and 95% $Fe_4[Fe(CN)_6]$) was developed and is now commercially produced in Russia and Belarus. It has been administered as 98% pure powder, in boli (15% ferrocyn), salt licks (10% ferrocyn) and as sawdust with 10% adsorbed ferrocyn (called bifege) (Ratnikov et al., 1998). The administration of 3–5 g of bifege was most effective (possibly due to a more uniform mixing in the feedstuff), but each delivery method produced good reductions under experimental conditions. However, when applied under unsupervised agricultural conditions, the effectiveness was lower, demonstrating the importance of ensuring that recommended procedures are followed by farm workers. An important aspect of the chosen delivery method is the acceptability of boli to the farming community. In Norway, Tveten et al. (1998) reported that the use of boli for reindeer is not as high as originally anticipated. This is because of additional work required in administration connected with reindeer and the need to ensure that the boli are administered properly without hurting the animal (using a special delivery device, Hove, Staaland, Pedersen, Ensby, & Sæthre, 1991). There can be uncertainty concerning the effectiveness of boli administration for individual animals due to; poor dosing techniques (so that animals do not swallow the bolus); regurgitation (usually soon after dosing) and the possibility of too rapid breakdown of boli in the rumen. In Wales, Nisbet and Woodman (2000) suggested that farmers felt that the use of AFCF may adversely affect the image of Welsh lamb as an "organic" product. A further potential problem with the use of AFCF boli in the event of a future nuclear accident is that there is currently no commercial production facility outside the FSU. However, salt-licks containing AFCF are commercially produced in Germany for use in Norway (Tveten et al., 1999) and boli, made on an ad-hoc basis by a university, have been an important and effective part of the restoration strategy for sheep production after the Chernobyl accident in Norway.

When AFCF was sprayed onto grass before ensiling, subsequent administration at a rate equivalent to 21 mg AFCF d^{-1} to lambs (38 kg liveweight) reduced ^{134}Cs transfer by 45% (Paasikallio, Sormunen-Cristian, Jaakola, & Kaikkonen, 2000). This is consistent with reductions achieved by other delivery mechanisms for ruminants (Hove, 1993) of 60% reduction in absorption at a daily administration rate of 1 mg kg^{-1} live weight. Increasing the intake of AFCF in silage to 50, 100 and 150 mg AFCF d^{-1} reduced ^{134}Cs transfer to sheep muscle by 75, 82 and 86%, respectively. Compared with incorporation of AFCF into concentrate or salt licks, where AFCF intake may be somewhat unpredictable since animals may choose not to ingest these supplements, this method may offer some benefits in that, if silage was the sole component of the diet, intake of AFCF would be directly proportional to the intake of the silage. This would assume that complete mixing was ensured and that AFCF was not subsequently lost from the silage. Appropriate techniques and equipment would need to be further tested and developed before it could be used on a large scale.

There are no known effects of AFCF on animal welfare or production (Giese, 1988, 1989; Pearce, 1994). When hexacyanoferrates are administered to animals, the compound is excreted in faeces, which will subsequently be deposited onto soil, either directly or as manure. Uptake of ^{137}Cs by legumes and grasses was reduced by 37–51% when the soil was fertilised with manure from cows receiving AFCF supplements (Hove et al., 1995). In response to concern about the possible negative effects of the continual deposition of AFCF in faeces onto soil, experiments were carried out where AFCF and radiocaesium-contaminated faeces was introduced onto sandy soil. There was no effect on plant growth and no detectable amounts of free cyanide released (Vandenhove, VanHees, DeBrouwer, & Vandecasteele, 1997; Vandenhove, VanHees, & Vandecasteele, 2000). Instead of a detrimental effect, a net benefit resulted since, as reported by Hove et al. (1995), a reduction in radiocaesium transfer to grass was observed.

In previous reviews of the effectiveness of hexacyanoferrate, the data were largely for ruminants and pigs, but there was a lack of available data for poultry (Voigt, 1993) although administration of AFCF achieved a high reduction in transfer to chicken meat by a factor of up to 100 (Voigt, Müller, Paretzke, Bauer, & Röhrmoser, 1991). In recent years, two further studies have been published demonstrating that the administration of AFCF reduces transfer of radiocaesium to chickens, although the effectiveness seems to be highly variable (Vitorovic et al., 1997; Pöschl & Balas, 2000) and dependent on several factors. In the studies performed by Pöschl and Balas (2000), RADEKONT, a natural clinoptilolite modified with FeHCF, mixed and fed with the daily feed each day decreased the uptake of radiocaesium in chicken meat by 1.3–2.4. A higher transfer to chicken meat after administration of radiocaesium incorporated into wheat was observed compared with other radiocaesium sources, combined with a higher reduction factor of 3.0–3.7; the reason for this observation is not clear.

In the FSU, the remaining radiation exposure of certain population groups above intervention limits due to Chernobyl accident and the resulting necessity of countermeasures (Jacob et al., 2000) combined with an increasing lack of centrally financed resources, have prompted a consideration of appropriate community-based strategies (Hériard Dubreuil et al., 1999), which might be used by people living in affected areas and who generally produce most of their own food. People need to understand how various countermeasures for animals work, so that they can manage their own resources to minimise the animal's radionuclide intake at the appropriate times (Frank et al., 1998; Beresford et al., 2001). For instance

- monitoring of feedstuffs (e.g. hay) and milk can allow informed decisions on how best to use the available resources. It can also ensure that time and effort is not wasted in taking animals which exceed accepted limits to market;

- the most contaminated milk or hay should be fed to young meat animals or non-lactating breeding stock rather than to lactating animals or to animals within 1 month of their going to market;
- available ferrocyn should be used for lactating animals and not for dairy animals when they are not lactating
- providing clear information regarding the biological half-lives of contaminant radionuclides to allow people to assess the timing and impact of "clean" feeding

4.1.1. Comparison with soil-based techniques

Some of the most frequently used countermeasures in the FSU after the Chernobyl accident were soil-based agricultural countermeasures such as disking, ploughing and fertilising (liming and NPK) which effectively reduced plant uptake of radiocaesium and its subsequent transfer via the foodchain. These measures are, however, strongly dependent on the soil type, nutrient status of the soils and plant community present. For K fertiliser rates of 90–240 t ha^{-1} in agricultural lands in the FSU, the reduction factor for radiocaesium transfer to ryegrass varied from 0.7 (i.e. an increase) to 3.5, with the highest reduction in peat and podzolic soils and less pronounced for chernozem (Colgan et al., 1996). In unimproved natural meadows, reductions of a factor of more than five occurred depending on soil type, with highest reductions of up to 33 in flooded soddy gley soils (Prister, Perepelyatnikov, & Perepelynatikova, 1993; Firsakova, Zhuchenko, & Voigt, 2000). In recent experiments, disking and ploughing and reseeding achieved an eight-fold reduction in plant uptake of radiocaesium in natural meadows (Rauret, Vidal, Rigol, & Camps, 1999). The decrease in transfer was often accompanied by a two-fold increase in biomass production.

Radical (disking and ploughing, application of mineral fertilisers, liming, reseeding with grasses) and surface (as above without ploughing) improvement is effective several years after application. However, the decline of the radionuclide content in grass varies with time. Usually, a maximum effect is achieved in the 2nd year after application and effectiveness thereafter declines (Table 1). Therefore, it is conventional practice to repeat radical improvement every 4–5 years.

Substances other than mineral fertilisers have also been tested for their effectiveness in reducing radiocaesium uptake; these include manure, zeolites, bentonites, vermiculites and liming, the application of Sapropel, Polygorskite, Phosphorite (Russian/Ukrainian compounds containing mineral clays (details about composition of these compounds are given in Rauret, Vidal, Rigol, & Camps, 1999) and sand or clay soils (Colgan et al., 1996; Rauret et al., 1999)). A maximum reduction factor of 2.0 fold (for peat) was achieved with average values for different soil types of between 0.9 and 1.8.

The applicability and effectiveness of soil-based measures are highly dependent on the characteristics of the soil in the contaminated site. They are generally restricted to agriculturally managed systems and need resources other than the direct costs of the fertiliser (e.g. transport, machinery, personnel, although many of these resources would be used in normal practice already). Though reductions have been achieved by enhanced K fertiliser application or soil additives for both agricultural and natural

Table 1
Long-term effectiveness for ^{137}Cs content in grass stands after radical and surface improvement (Jacob et al., 2000)

Year after treatment	Reduction factor	
	Radical improvement	Surface improvement
1	3.5	1.4
2	4.2	1.2
3	2.8	1.2
4	2.7	1.2
5	2.1	1.1

meadows in the FSU, it is questionable whether similar reductions could be achieved in highly fertilised agricultural systems such as those prevailing in Western Europe.

4.1.2. Conclusion

Condemnation of meat is an immediately available and effective countermeasure to reduce ingested dose from animal products. This was a widely used countermeasure after the Chernobyl accident, but retrospective evaluations have concluded that this was very expensive and resulted in large quantities of contaminated waste. It was therefore not considered to have been a good solution and clean feeding would have been a better alternative.

The most useful, and in the aftermath of Chernobyl, widely used countermeasures for radiocaesium contamination of animals are clean feeding, the administration of hexacyanoferrates, changing slaughter times and use of live-monitoring. Hexacyanoferrate compounds are highly effective with reported reduction factors of up to 10 in the FSU (Prister, Perepelyatnikov, & Perepelyatnikova, 1993), have low toxicity (Giese, 1988, 1989) and AFCF has been temporarily authorised in the EU (to 2001) and some other countries. They can now be incorporated into many different delivery systems for use in different farming systems and for different species. Cost-effectiveness has been found to be good in certain situations, and for free-ranging animals, boli are more cost-effective than clean feeding, although they are somewhat less practical for reindeer than other ruminants. The public acceptability of using AFCF in the farming community is a factor which needs consideration; some delivery methods are less intrusive than others. Furthermore, the lack of a commercial manufacturer of AFCF boli would currently restrict their use in the event of a nuclear accident.

To summarise, animal-based countermeasures for radiocaesium might often be more generally applicable, practical and cost-effective than soil-based measures, especially for animal products from the private sector and from unimproved, semi-natural environments. In practice, the optimum solution is often likely to be the development of strategies combining soil-based measures for both pasture and production of pasture with the use of a series of animal-based measures, to produce an additive effect.

4.2. Radioiodine

Absorption of radioiodine in the gut is complete, regardless of source of dietary iodine intake rates, and there is a subsequent rapid transfer to the thyroid and milk (Howard, Voigt, Segal, & Ward, 1996; Vandecasteele, VanHees, Hardeman, Voigt, & Howard, 2000; Beresford et al., 2000a). Despite the short physical half-life of many radioiodine isotopes, ingestion of contaminated milk represents a potentially important source of internal dose after an accident, and has been identified as a major source of ingested dose for children who developed thyroid cancer after the Chernobyl accident (Kazakov, Demidchick, & Astachova, 1992; Likhtarev et al., 1994). Iodine is an important trace element required by the thyroid for hormone synthesis, and the metabolism and excretion of radioiodine is controlled by the individual's stable iodine status. Transfer to important edible issues is low. Variation in stable iodine intake may occur since contents in soil, and hence vegetation, vary spatially, being generally higher in coastal areas and sometimes endemically low in certain continental areas. However, iodine supplementation in concentrates would prevent low iodine intake in many areas.

Howard, Voigt, Segal, and Ward (1996) have previously identified oral administration of stable iodine as a potentially useful countermeasure to reduce radioiodine to milk with reported reductions of up to a factor of three. The reduction occurs because the transfer of iodide to the mammary gland is an active process (Falconer, 1963) which is saturable at high iodide plasma concentrations. Radioiodine activity concentrations in milk are reduced by isotopic dilution. The relationship between stable iodine administration rate and effect is complex, but various experiments in dairy cows suggested that at least 1 g I d^{-1} would be needed to reduce the ^{131}I activity concentration in milk by a factor of two to three (Kiefer & Voigt, 1999; Vandecasteele, Van Hees, Hardeman, Voigt, & Howard, 2000).

Recently, two experimental studies with dairy cows have confirmed that the administration of stable iodine to cow milk will reduce transfer of radioiodine. Reduction values ranging from 1.1 to 2.2 have been determined (Kiefer & Voigt, 1999; Vandecasteele et al., 2000) dependent on the stable iodine intake of the animals, with the highest value achieved from the administration of 1 g d^{-1} stable iodine. The reduction is not due to effects on absorption (Vandecasteele et al., 2000) and must therefore be related to the differential affinities and saturation levels of the thyroid and milk pathways for iodide circulating in the plasma (Crout et al., 2000).

Most models describing radioiodine transfer are empirically based and do not allow for the interaction between radioiodine and stable iodine. A model developed by Coughtrey, Jackson, and Thorne (1983) considered the effect of stable iodine, to a limited extent, by incorporating the interaction of stable iodine and thyroid metabolism. Recently, models have been developed which more fully incorporate the effect of stable iodine on the metabolism of radioiodine in cows (Crout & Voigt, 1996) and goats (Crout et al., 2000). In the later study, where the model was developed using new experimental data for goats, the transfer of iodide from the plasma to milk was represented by using Michaelis–Menten kinetics. The model predicts that there will not be a linear relationship between stable iodine intake and the radioiodine activity concentration in milk. Over the range of 1–10 mg I d^{-1}, the radioiodine activity concentration in milk increases (with the highest proportional increase at the lowest stable iodine intakes). Between 10 and 20 mg I d^{-1}, the increase in radioiodine activity concentration ceases, and above 20 mg I d^{-1} the radioiodine activity concentration decreases. Some experimental evidence confirms that administration of small quantities of stable iodine may lead to increased radioiodine activity concentrations in cow milk (Howard et al., 1996; Vandecasteele et al., 2000). The effect can be explained by considering the relative transfer of iodide to the thyroid and mammary gland. As the stable dietary iodine increases, the proportion of the daily radioiodine intake which is transferred to the thyroid declines because of the constant rate of uptake of iodide by the thyroid. This is implemented accordingly in the iodine model of Crout et al. (2000). Thus, a smaller fraction of the daily iodide intake (stable iodine and radioiodine isotopes) is transferred to the thyroid and the proportion available for transfer to the mammary gland increases. At high stable iodine daily intakes, this effect is offset by the saturation of the transfer from plasma to milk represented by Michaelis–Menten kinetics. Therefore, increasing stable iodine intakes above a certain threshold would be expected to have little additional effect in reducing radioiodine transfer to milk.

The relative timing of stable iodine administration compared to that of the ingestion of radioiodine has been investigated by Beresford et al. (1997) in goats. They showed that administration of 1 g KI 12 h before a pulse administration of radioiodine reduced transfer by 50%, whereas the reduction was 40% if the stable iodine was administered 12 h after radioiodine. When only half of quantity of KI (i.e. 0.5 g) was administered 6 h before radioiodine rather than 12 h, the same effect was achieved (Crout et al., 2000). Another temporal effect discussed by Crout et al. (2000) was that the time of maximum radioiodine activity concentration in milk was predicted to vary with the expected range in stable I intakes by 2 days. The time at which the predicted peak ^{131}I activity concentration in milk occurs increased with increasing stable iodine intake.

Recently, Beresford et al. (1997) showed that a single administration of 1 g of KI increased stable iodine concentrations in the milk of goats to 20 mg I kg^{-1}, which was 40-fold higher than maximum permissible levels of iodine in milk of 0.5 mg I kg^{-1} recommended by World Health Organisation (WHO) and applied in some countries. The same effect has also been reported by Kiefer and Voigt (1999) in dairy cows, who found that stable iodine intakes exceeding 50 mg I d^{-1} greatly exceeded the WHO recommended value. Such observations were also predicted in the model described above for goats (Crout et al., 2000).

Contrary to previous recommendations, model predictions by Crout et al. (2000) and experimental data of Kiefer and Voigt (1999) and Voigt, Schotola, Probstmeier, & Röhrmoser (1994) suggest that reductions in stable iodine intake from normal levels might be an effective countermeasure because a higher proportion of the dietary radioiodine would be accumulated in the thyroid and less in the milk. However, for large scale application after accidental releases of radioiodine, it is unlikely that such a response (which would only need to replace the farmers' own feedingstuffs for a short period) could be organised in the timescale needed given the short physical half-lives of most radioiodine isotopes and the need to know current, site-specific, stable iodine intake rates.

The practicality of storing or disposing of contaminated milk has been discussed recently in various fora, including the International Union of Radioecologists (Voigt et al., 2000) and a UK working group (Mondon & Nisbet, 1999). If emergency preparedness plans envisage processing and storage of contaminated products, then prior agreement between regulatory authorities and potential operators regarding required procedures would be highly beneficial. If disposal is considered, then it will be necessary to ensure that the envisaged route of disposal is feasible and environmentally acceptable and that any legal constraints can be overcome at short notice. In a review of disposal options for milk, which was focused on radioiodine, but is also relevant for radiocaesium and radiostrontium, Woodman, Nisbet, and Penfold (1997) identified five possible options, in order of recommended implementation, as

- feeding contaminated milk to animals,
- spreading contaminated milk on agricultural land,
- anaerobic digestion,
- discharge to sea,
- dilution/processing of contaminated milk.

4.2.1. Conclusion

Contamination of milk by short-lived radioiodine isotopes is a major source of potential ingested exposure after many types of nuclear accident. It is therefore important that emergency preparedness plans for this eventuality are well informed and appropriately focused.

Previous recommendations in the literature regarding the use of stable iodine are probably unjustified: the reduction achieved is only two- to three-fold and there are significant side-effects in that the stable iodine in milk would probably exceed acceptable limits. Furthermore, there may be problems in obtaining, distributing and administering suitable iodine sources within the time-scale required. Ensuring that animals can eat uncontaminated fodder is probably the best solution, especially for short-lived radioiodine isotopes. However, moving animals to uncontaminated sites to achieve this provision of clean fodder may be difficult to arrange within the time-scale required; the feasibility of this option will depend on the scale of the contamination and the number of animals affected. The most effective and practical solution remains the provision of uncontaminated feed to the animals, but the availability of suitable stocks of feedstuffs will vary seasonally.

Storage of contaminated milk or conversion to other storable products would be another effective option, but may not be commercially or socially acceptable and would need to take account of other radionuclides also present. Prior arrangement for this option with relevant organisations and commercial concerns would be beneficial.

4.3. Radiostrontium

Radiostrontium absorption from the gut depends upon the calcium requirement and intake of an animal, with mean reported values for the degree of true absorption varying between 12% and 72% (Beresford et al., 2000a). Absorbed radiostrontium is subsequently largely deposited in bone and transferred to milk; the latter forms the main exposure route to humans.

Woodman and Nisbet (1999) have recently recommended "working levels" for feedstuffs (i.e. that would result in the production of animal products with activity concentrations approaching the relevant Council Food Intervention Level) for radiostrontium; MPLs for this isotope are not currently given by the EC. The recommended values for ^{90}Sr in feedstuffs were generally much higher than those for radiocaesium, with the exception of those for dairy cattle and laying hens. This is because, with the exception of milk and eggs, transfer coefficients of ^{90}Sr to animal products are lower than those of radiocaesium.

The behaviour of radiostrontium in animals is governed by that of its homeostatically controlled analogue, calcium. Recent studies have shown that there is a relationship between calcium intake and the transfer coefficient (F_m) of radiostrontium to milk. The relationship was initially derived for dairy cattle (Howard et al., 1997) and has since been extended to derive a generic relationship (Fig. 1) for ruminants (Beresford et al., 1998a) whereby

$$F_m\mathrm{Sr} = \frac{0.11[\mathrm{Ca}]_{\mathrm{milk}}}{I_{\mathrm{Ca}}}, \tag{1}$$

where $[\mathrm{Ca}]_{\mathrm{milk}}$ ($\mathrm{g\,kg^{-1}}$) is the concentration of calcium in milk, and I_{Ca} the daily intake of Ca ($\mathrm{g\,d^{-1}}$) and 0.11 is the reported value of the observed ratio between strontium and calcium concentrations in milk divided by those in the feed.

The application of this relationship allows (i) values of the transfer coefficient for radiostrontium to be derived for differing daily calcium intake rates and (ii) the reduction in transfer of radiostrontium in milk achieved by different additional administrations of calcium to be predicted. Because of the relationship between calcium and radiostrontium, published reduction factors (reviewed in Voigt, 1993), which predict the reduction achieved by giving additional calcium, are not generally applicable, because the reduction will depend on the animal's basal calcium intake and status. In general terms, if the dietary intake of calcium is doubled then the radiostrontium activity concentration in milk would be halved. Larger reductions would be achievable in those animals with low dietary calcium intakes prior to calcium supplementation. Typically, in countries with farming systems similar to the UK, the calcium intake for dairy goats is in the range 15–20 $\mathrm{g\,d^{-1}}$ (Beresford et al., 1998b), whilst that of dairy cows is 70–150 $\mathrm{g\,d^{-1}}$ (Beresford et al., 2000b). Application of the relationship suggests that a reduction in F_m of 40–60% would be expected if dairy cattle were supplemented with 100–200 $\mathrm{g\,d^{-1}}$. The upper value is probably the maximum supplement rate that can be used to avoid exceeding the upper limit for calcium intake for ruminants of 1–2% of their daily dry matter intake, because of the need to prevent any reduction in absorption of other essential

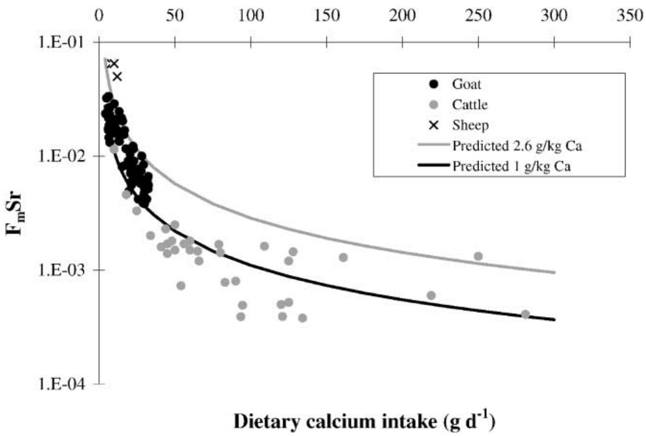

Fig. 1. The relationship between dietary calcium intake and the transfer of radiostrontium to milk of different farm ruminants as presented by Beresford et al. (1998a); figure also includes more recent data for cattle taken from Beresford et al. (2000b). The lines represent predicted values from Eq. (1) based upon calcium contents in milk of 1 and 2.6 g kg^{-1}, respectively.

nutrients (NRC, 1980, 1989). To achieve a greater effect, dietary intakes of dairy animals would need to be assessed on a farm-by-farm basis—some farmers would already have this information. Recent experiments with cows by Beresford et al. (2000b) validated the relationship and this data are included in Fig. 1. In addition to its efficiency, the cost of dietary calcium supplementation is low.

A model of radiostrontium transfer to dairy goats has been developed which is based on calcium metabolism (Crout, Beresford, Howard, Mayes, & Hansen, 1998). In addition to incorporating the effect of dietary calcium intake, the model also accounts for the effect of an animal's calcium requirement. The model is therefore capable of simulating the effect of additional dietary calcium intake under different physiological conditions.

Previously, there has been contradictory evidence of the ability of stable strontium to reduce the deposition of radiostrontium to bone or its transfer to milk (Howard, Beresford, Kennedy, & Barnett, 1995b). Recent experiments where stable strontium was administered to dairy goats have shown a lack of effect (Hove et al. in Howard et al., 1995a). This may be expected given the relationship between strontium and calcium, and that calcium is present in the diet in much larger quantities.

Over many years, a wide range of different binders have been tested with the objective of reducing transfer of radiostrontium in the gut (Voigt, 1993). This is difficult because the strontium cation binds exchangeably to many anions and has a wide range of potential competitors (in addition to calcium) in an animal's gut. In addition, it is important to ensure that administration of a binder does not cause deficiencies in essential elements by also preventing their absorption in the gut.

Recently reported studies of possible strontium binders consider clays and alginates. Some clay minerals such as aluminium pillar-layered clays (Karamanis et al., 1997) and modified zeolites (Marinov & Zlatev, 1995) had good sorption properties in vitro. However, good binding in vivo has not been achieved, probably because the high pH in parts of the animal gut causes dissociation of the binding (Howard, Beresford, Kennedy, & Barnett, 1995b). Hansen, Sæther, Asper, and Hove (1995) tested a range of different clay minerals in dairy goats, only sodium-aluminiumsilicate (Zeolite A(Na)), administered at a rate of $0.5\,g\,kg^{-1}$ live weight d^{-1} was effective, reducing the radiostrontium activity concentration in milk by ca. 40%. This compound is widely used in the chemical industry, is readily available and cheap. However, this compound influences the absorption of a number of essential elements, and the potential implications have not been adequately considered.

Past studies on sodium alginate with dairy cows achieved a 70% reduction when fed as 5–7% of the diet, but the cattle found the diet unpalatable, probably because of the highly viscous solutions given, and it was therefore thought to be impractical (Voigt, 1993). To overcome this problem, Beresford, Mayes, MacEachern, Dodd, and Lamb (1999c) and Beresford et al. (2000b) incorporated sodium alginate and calcium alginate into pelleted feed at 4–5% dry weight. This achieved a reduction in radiostrontium activity concentrations in goat milk of 50% (calcium alginate) and in cow milk of 30–40% (both alginates) without affecting the palatability of the feed. The effectiveness of the alginates was much lower than that previously reported for monogastrics, and the authors suggested that this may have been due to the observed fermentation of the alginate in the reticulorumen. The cost of using commercially available alginates of ca. 13 Euro per cow per day (1998) to achieve a ca.50% reduction is ca. >2000 fold higher than that of doubling Ca intake, and would preclude its use. Alginates are derived from seaweeds, and although not yet tested, seaweed meal may be a viable, cost-effective, alternative as alginic acids in seaweeds undergo the same base exchange reactions as isolated alginates (Percival & McDowell, 1967).

4.3.1. Comparison with soil-based techniques

Radiostrontium is generally highly available for uptake by plants from many soil types and is not immobilised by clay minerals. The similarity of calcium and strontium would imply that calcium is a potentially effective countermeasure for reducing radiostrontium uptake by plants. Thus, the application of lime or other calcium-containing minerals will increase exchangeable Ca levels in soils and thus the Ca : Sr concentration ratio in soil solutions. Factors which affect reductions in soil-plant uptake achieved by calcium application include: the base saturation of the contaminated soil, Sr/Ca selectivity of contaminated soils, and the prevailing Cs : Sr ratio in the soil solution for root uptake. Reduction factors of up to 10 (but more often two to three) have been obtained by liming of mineral soils with a low percentage base saturation. Organic matter achieved a five-fold reduction, and phosphates, sulphates and silicates a reduction of up to ten-fold (Nisbet et al., 1993). In recent experiments, performed in natural meadows in the FSU, radical improvement of meadows (including disking, ploughing, liming and reseeding) achieved reductions of two- to four-fold for a range of different soil types. However, the effect was much lower in the 2nd year after treatment (Rauret et al., 1999).

Additional application of fertilisers had only a minor influence on soil-plant transfer, as did the application of turf, Sapropel, mineral, sand and clay soil to meadows with a decreasing effectiveness in the second and following years.

4.3.2. Conclusion

There is a limit to the potential effectiveness of calcium supplementation to the diet of dairy animals because of interference with availability of other essential nutrients. For animals receiving basal dietary calcium levels typical for Western Europe, a doubling of calcium intake is recommended with an anticipated reduction of 40–60% in the transfer of radiostrontium to milk. However, it may be possible to increase its effectiveness on the basis of farm-specific assessments.

Currently, the use of binders cannot be recommended. There are three different reasons for this conclusion: (i) many are ineffective, (ii) alginates are effective but are currently too expensive and (iii) some potentially effective binders have possible side-effects which have been inadequately tested.

Soil treatment or soil amendments provide a viable and practical alternative to animal-based countermeasures for radiostrontium.

5. Discussion

For each of the three most important, mobile radionuclides, our understanding of the effectiveness of possible animal-based countermeasures and their potential usefulness has significantly improved since the early 1990s. For radiocaesium, hexacyanoferrates are routinely used for post-Chernobyl management in some countries; cheaper effective sources of these compounds are now produced and AFCF can be legally used in the EC and elsewhere. The cost-effectiveness of boli has been demonstrated and they have been modified to improve effectiveness and widen their applicability. Overall, there is now a wide range of practical and cost-effective countermeasures available for radiocaesium, which should cover most situations. For radioiodine, the situation is different. The use of stable iodine cannot now be recommended and the only obviously effective countermeasure is clean feeding or storage to allow for radioactive decay (depending on the presence of other radionuclides). For radiostrontium, there is now better defined advice available on calcium intake manipulation and its effect. Palatable and effective sources of alginates are available, but they are currently too expensive to contemplate their use.

The usefulness of various countermeasures and assessment of whether they are realistically likely to be useful and effective for contamination of animals will vary with many different factors, both within and between countries, in addition to those already discussed above. These factors will include the

- Severity of contamination—the extent of the affected area and the inventory of contamination will be major criteria in deciding which countermeasures can be used.

- Importance of affected areas to national economies—the proportion of animal production affected and whether it can be readily replaced by importation.
- Type of ecosystem affected and existing management systems—the response to contamination of intensive agricultural systems (for instance of cow milk) will differ greatly to that of extensive systems.
- Priority given to maintaining existing ways of life—the numbers of people in critical groups and their views may influence the selection of countermeasures and the severity and potential disruption of the response. It may be considered appropriate to raise the intervention limit (combined with dietary and other advice) for some animal products to safeguard certain communities, as was implemented in Scandinavia after the Chernobyl accident.
- Animal species and products contaminated—whether the contaminated products form an important component of the national diet. Rates of transfer of radiocaesium are particularly high to some animal products such as reindeer and goat milk-derived whey cheese, and the transfer of radioiodine to goat and sheep milk is greater than to cow milk.
- Way of life—some countermeasures would only be appropriate for certain types of community. For example, self-help measures suggested for private cows in rural communities in the FSU may not be transferable to Western European communities.
- National economy—the acceptable cost per averted ManSv may differ between countries.
- Response of stakeholders such as the food industry and consumers—the extent and duration of any retail and consumer reaction to contamination of animal products.

Given the above, it is clear that suitable countermeasures are likely to vary considerably between different areas and communities, and national authorities need to evaluate their own relevant criteria. Such assessments will need to be frequently reviewed to cope with changes in factors such as feeding regimes of agricultural animals.

The legal and administrative framework for the use of countermeasures is also important. For instance, regulations for the application of restrictions for contaminated free-ranging animals after the Chernobyl accident differ—in the UK farms are restricted if a single sheep is measured over the intervention limit whereas in Norway restrictions are based on the measured average values for a flock.

Contamination of animals and their products is likely to give rise to considerable public concern in the event of an accident. Some problems and potential solutions with regard to contamination of animals would benefit from prior consideration and agreement with relevant stakeholders, such as that ongoing within a consultation group in the UK (Mondon & Nisbet, 1999). Such bodies would include organisations which do not necessarily have a direct regulatory role regarding nuclear issues, but which would or might be involved in dealing with the consequences of accident, such as farmers unions, the dairy industry and consumer bodies.

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