Review of the Transfer of Technetium to Terrestrial Crops and Animal Products

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ABSTRACT

Technetium-99 (^{99}Tc) has been dispersed in the environment from many sources such as nuclear weapons testing, releases from medical or industrial processes, nuclear power plants and nuclear fuel processing facilities. For ^{99}Tc, interest in environmental transfer has increased because of its relative importance in radiological assessments for nuclear waste repositories. An adequate knowledge of any radionuclide's behaviour in terrestrial foodchains is important for assessing the radiological impact on people following the release of radioactive material into the environment.

The aim of this study was to carry out a critical review of published data on the transfer of technetium into terrestrial crops and animal products and to recommend transfer factor (TF) values for use in UK based radiological assessments.

This review has identified that the chemical form of technetium in soil is the main factor that determines the degree to which it is available for uptake to crops. TF values have been compiled for a range of crop types and for technetium in a chemically reduced and non-reduced form, where data are available.

The compiled TF values have been compared with default parameters currently used in HPA's foodchain model, FARMLAND. Recommended values of transfer factors for specific applications of FARMLAND for radiological assessments in the UK are proposed.

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Chilton, Didcot
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Approval: August 2011
Publication: August 2011
£21.00
ISBN 978-0-85951-703-4

This work was undertaken under the Environmental Assessment Department’s Quality Management System, which has been approved by Lloyd's Register Quality Assurance to the Quality Management Standards ISO 9001:2008 and Tick IT Guide Issue 5.5, Certificate No: LRQ 0956546.

Report version 1.0

CONTENTS

1 INTRODUCTION

There are twenty two known isotopes of technetium. While all isotopes of this element are radioactive, attention is focused on technetium-99 (^{99}Tc), a beta emitting radionuclide with a radiological half-life of 2.1 10^{5} years. Technetium-99, produced by the fission of uranium-235 (^{235}U) and plutonium-239 (^{239}Pu) has been dispersed in the environment from many sources such as nuclear weapons testing, releases from medical or industrial processes, nuclear power plants and nuclear fuel processing facilities.

Environmental impact assessments associated with the licensing, operation and decommissioning of nuclear facilities usually include the evaluation of radiation exposure pathways that involve the transport of radionuclides through agricultural soils, their uptake by plants and animals and their subsequent ingestion by man. For ^{99}Tc, interest in predictions of soil to plant transfer is increasing because of its importance to radiological assessments for permanent terrestrial nuclear waste repositories (Echevaria et al, 1998). In addition, as part of decontamination and decommissioning assessments, there is current interest in the potential of plants to extract toxic elemental pollutants, including radionuclides such as ^{99}Tc, from contaminated effluents and soils (Meagher, 2000; Willey et al, 2002).

An adequate knowledge of any radionuclide's behaviour in terrestrial ecosystems is important for assessing the radiological impact on people following the release of radioactive material into the environment. The accuracy of the predictions will depend on the extent to which the models of environmental transfer replicate the actual situation, and on the values chosen for the parameters in these models. In terms of environmental transfer, technetium has not been studied as extensively as other radioisotopes, for example plutonium, americium, strontium and caesium. Extensive reviews carried out by Desmet and Myttteneare (1984) and by Wildung et al (1989) in the 1980s indicated that a better understanding of the behaviour of technetium in the environment would be useful as technetium is relatively mobile in the environment and high transfer from soil to plants had been observed. The mobility of technetium in the terrestrial foodchain makes it of particular interest when assessing possible exposures to people from the ingestion of food.

Since the 1980s, technetium, notably ^{99}Tc, has received more research attention and more information is now available on which to base radiological assessments. However, there are still considerable uncertainties associated with the uptake of technetium from soil to plants and from feed to animals and a wide range of values for transfer factors have been derived from laboratory, field and animal studies.

This report describes a review of data on the transfer of technetium into terrestrial crops and animal products, focusing on environmental conditions such as soil type and climate that are similar to those encountered in the UK. The transfer of technetium from feed to animal products formed part of an earlier review conducted by Green and Woodman (2003). However, further data have since been located and for completeness all available data on transfer to animal products have been reviewed here.

Recommended values for use in radiological assessments are given. These have been chosen based on criteria designed to eliminate those studies that are not applicable to the UK. Values are recommended for use in the foodchain model, FARMLAND (Brown and Simmonds, 1995) for radiological assessments in the UK. Comparisons are made with current parameter values used in the model and implications discussed in this review.

2 OBJECTIVES

The objectives of this review were as follows:

a compile published information on the transfer of technetium along the soil to plant and feed to animal pathway;

b carry out a critical review of the data in terms of their applicability to the UK;

c identify factors (eg, study type, chemical form and age of technetium deposit, soil characteristics) that affect the transfer of technetium to crops and animal products;

d compare the results with values currently used in the FARMLAND model;

e compare results derived from the review with those given in the International Atomic Energy Agency's (IAEA) Technical Document 1616[1] (IAEA, 2009) for crops and IAEA Technical Report 364 (IAEA,1994) for animals and highlight any areas of difference;

f recommend transfer values for specific applications of the FARMLAND model.

3 RADIONUCLIDE TRANSFER IN THE FOODCHAIN

3.1 Transfer of radionuclides to crops

The processes by which radionuclides can become incorporated into edible parts of crops are as follows:

a uptake of activity from the soil via the root system;

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[1] Based on a review of available data, IAEA in their latest publication (IAEA-1616, 2009) do not recommend transfer parameters to animal products for technetium.

b interception of activity by external parts, either directly from the atmosphere or from re-suspended material, followed by subsequent translocation from the external surfaces of the plant to the edible plant parts.

Measurements of activity concentrations in the edible parts of the crop and the corresponding soil are often expressed in terms of an observed concentration ratio (CR):

$$\mathrm{CR} = \frac{\text{Activity concentration in edible crop (Bq kg}^{-1}\text{ fresh or dry mass)}}{\text{Activity concentration in soil (Bq kg}^{-1}\text{ dry mass)}}$$

In situations where deposition of radioactivity, either directly or from resuspension makes a significant contribution to the overall activity concentration in the plant, the CR value represents a situation at a given time and can combine a number of processes affecting the activity concentration in the crop. Such values are not then of direct use in assessment models. However, if direct deposition can be excluded and the CR value is high, then any contribution from resuspension onto the edible crop will be negligible.

In the absence of direct deposition or resuspension, the uptake from the soil via roots is the only process that controls activity concentration in the crops. This value can then be regarded as the soil to plant transfer factor (TF):

$$\mathrm{TF} = \frac{\text{Activity concentration in edible crop from root uptake (Bq kg}^{-1}\text{ fresh or dry mass)}}{\text{Activity concentration in soil (Bq kg}^{-1}\text{ dry mass)}}$$

As is the case for CR values, any possible contamination from resuspension of contamination from the soil onto the edible crop can be ignored if the observed TF value is high.

This review has found several factors that affect the transfer of technetium into crops which are discussed in Section 5.1.

As this review was concerned with providing information on TFs for use in predictive food chain models that are used to estimate doses to people in the UK, published data for crops were carefully evaluated against a set of criteria designed to eliminate those studies in which transfer parameters were not true soil to plant transfer factors and which were not applicable for use in the UK. These criteria are described in Section 6.

3.2 Transfer of radionuclides to animals

The principal routes by which animals can intake radionuclides are:

a ingestion of contaminated feed, soil and drinking water;

b inhalation, including gaseous compounds, aerosols and particles.

Intake via drinking water and inhalation are generally small contributors to the total radionuclide intake and are not considered further in this review. These intakes may however need to be considered in specific situations following accidental releases.

To quantify the transfer of radionuclides to animals, the transfer coefficient has been widely adopted. It is defined as the amount of an animal's daily intake that is transferred to one litre of milk (F_m, d L^{-1}), to one kilogram of animal tissue (F_f, d kg^{-1}) or to eggs (F_f).

Very few data were found by this review on the transfer of technetium into animal products and so criteria were difficult to establish. Subsequently, each publication was critically reviewed on a case by case basis.

4 IAEA REVIEWS OF RADIONUCLIDE TRANSFER

For more than thirty years IAEA has published a set of documents aimed at the limitation of the radiation exposure of the population from various nuclear activities. In 1994, IAEA published Technical Report Series 364 (Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments, (IAEA, 1994). The handbook which is based on available data up to 1992, provides recommended TFs for specific soil to plant and plant to animal combinations. In recent years, several publications have been produced which led IAEA to revise the transfer parameters in the handbook. In 2009, IAEA published Technical Document 1616 (IAEA, 2009) which comprises revised transfer parameter values as well as new data. In both publications, the amount of data on the transfer of technetium to crops is limited with only minor differences noted between the two reviews.

For the purposes of this review, criteria for accepting experimental data were designed that were specific to the UK (Section 6). When the compiled data are compared to values recommended by IAEA for wider soil types some differences in transfer parameters can be expected. For crops and pasture, where recommended transfer values for technetium are given in IAEA-1616, they are discussed.

In their latest publication (IAEA-1616), IAEA do not recommend transfer parameters to animal products for technetium (IAEA, 2009). IAEA found that limited data exist on the transfer of technetium to animals and for those studies that have been published many use short life gamma-emitting radioisotopes such as ^{95m}Tc and ^{99m}Tc which have been reported as showing noticeable isotope-specific variation in the transfer parameters (Ennis et al, 1988a; Johnson et al, 1988).

5 BEHAVIOUR OF TECHNETIUM IN THE ENVIRONMENT

The behaviour of technetium in the environment has been studied since the element was discovered by Perrier and Segre in 1937. A great deal of research was carried out on technetium in the late 1970s and early 1980s (Wildung et al, 1974; Landa et al, 1977; Routson et al, 1978; Gast et al, 1978; Sisson et al, 1979; Balogh and Grigal, 1980; Eriksson 1982; Sheppard et al, 1983). Overall, the results showed that the characteristics of sorption of technetium in soil were:

a low but measurable under aerated conditions as TcO_4^- (Pertechnetate);

b increased substantially under reducing conditions, presumably as TcO_2;

c decreased over the course of time, presumably via the reduction of TcO_4^- to TcO_2;

d increased in the presence of higher organic matter content especially under anaerobic (ie, reducing) conditions;

e decreased as the pH of the soil increased.

Since then, advances in soil science have provided new information to assess technetium transfer to crops and those parameters considered important for root uptake.

This review found very little data on the transfer of technetium from feed into animal products and found only two studies suggesting that the chemical form of technetium could have an affect on uptake into animal products (Ennis et al, 1988a; Jones, 1989). It is therefore not possible to draw any firm conclusions on the impact of chemical form on the transfer of technetium along the plant to animal pathway. However, from other radionuclides which have been studied more extensively, some general statements can be drawn and these factors are discussed in Section 5.2.

5.1 Factors affecting the transfer of technetium to crops

5.1.1 Chemical form of radionuclide and age of contamination

The chemical form of technetium in soil is one factor that determines the degree to which it is available to crops. The pertechnetate ion (TcO_4^-) is the form produced during the nuclear fuel cycle and the most likely to be released into the environment (Till, 1984; Harms et al, 1999).

From a review of relevant literature, Bennett and Willey (2003) concluded that in aerobic soils, TcO_4^- was the most stable form of technetium. Many other authors have reported that this form of technetium is very soluble in water over a wide pH range and is readily taken up by plants (Echevarria, et al, 1994; Vandecasteele et al, 1989 and Sheppard et al, 1983). Similar conclusions can be drawn from a review conducted by Bergstrom and Wilkens (1983) and from information gathered by Turcotte (1982).

Several studies report a decrease over time in the rate of soil to plant transfer of technetium when the activity was originally applied as $^{99}TcO_4^-$ (Mousny and Myttenaeare, 1982; Garten et al, 1984; Stalmans et al, 1986; Vandecasteele et al, 1986; Vandecasteele et al, 1989; Echevarria et al, 1994; Bennett and Willey, 2003). Results from some of these studies that investigated uptake of technetium into pasture are illustrated in Figures 1 and 2.

Soil to plant transfer factor data obtained from some of the studies cited in this section can only be regarded as observed soil to plant CR values as the radionuclide was not applied homogenously to the soil. They are however of use, since they demonstrate the reduction in uptake of ^{99}Tc over time. For ease of discussion, all data relating to CR values and TFs (as defined in Section 3.1) are referred to as soil to plant transfer values in this section.

The individual studies shown in Figures 1 and 2 have different delay periods between the contamination of the soil and the first harvest of pasture. In all cases, soil to plant transfers for the first harvest are given as the 100% value on the y-axis of the graphs. The numerical values for the changes are not therefore directly comparable. However, the results do show a similar trend in decrease of the soil to plant transfer over time for the different studies (field, pot and lysimeter).

Figure 1 shows results of studies investigating the relationship between soil to plant transfer of ^{99}Tc in pasture and soil type for soils contaminated either on the surface or homogeneously mixed (Mousny and Myttenaeare, 1982; Vandecasteele et al, 1989; Garten et al, 1984). Mousny and Myttenaeare (1982) investigated pasture grown in pots containing clay, loam and peat based soils that were either homogeneously contaminated with $^{99}TcO_4^-$ or had radioactivity applied to the soil surface. All the soils were maintained at field moisture capacity for a period of 21 days, seeds were then sown and the pasture was harvested after a further 30, 60 and 90 days. The results of this experiment show a decrease in the observed transfer values from one harvest to the next. For those soils that were homogeneously contaminated, the decrease in the transfer factor values for the harvest at 90 days were more pronounced with around an 80% to 90% reduction in soil to plant transfer for all soil types except clay. In comparison, for soils that received surface contamination, the reduction in soil to plant transfer over time was generally less pronounced and more variable, with reductions varying from 10% to 80%.

Garten et al (1984) carried out field studies investigating soil to plant transfer values for pasture grown in silt-loam soils where the soil surface had originally been surface contaminated with $^{99}TcO_4^-$. The pasture was harvested at various times up to 189 days after contamination. The study demonstrated a rapid decrease in the observed soil to plant transfer over time with a reduction at 90 days after contamination of over 90%. After this period of time there was little change in soil to plant transfer up to the last harvest carried out 189 days after contamination, as shown in Figure 1. Comparable results were also obtained from lysimeter studies conducted by Vandecasteele et al (1989) who grew pasture in loam based soils that had $^{99}TcO_4^-$ applied to the soil surface. Harvests were carried out at various times up to 1200 days after contamination. The results given in Figure 1 show a reduction in the observed transfer value of around 30% at the second harvest, carried out 76 days after contamination. At 140 days this reduction was around 95%, with little further change in the transfer to pasture observed up to 1200 days.

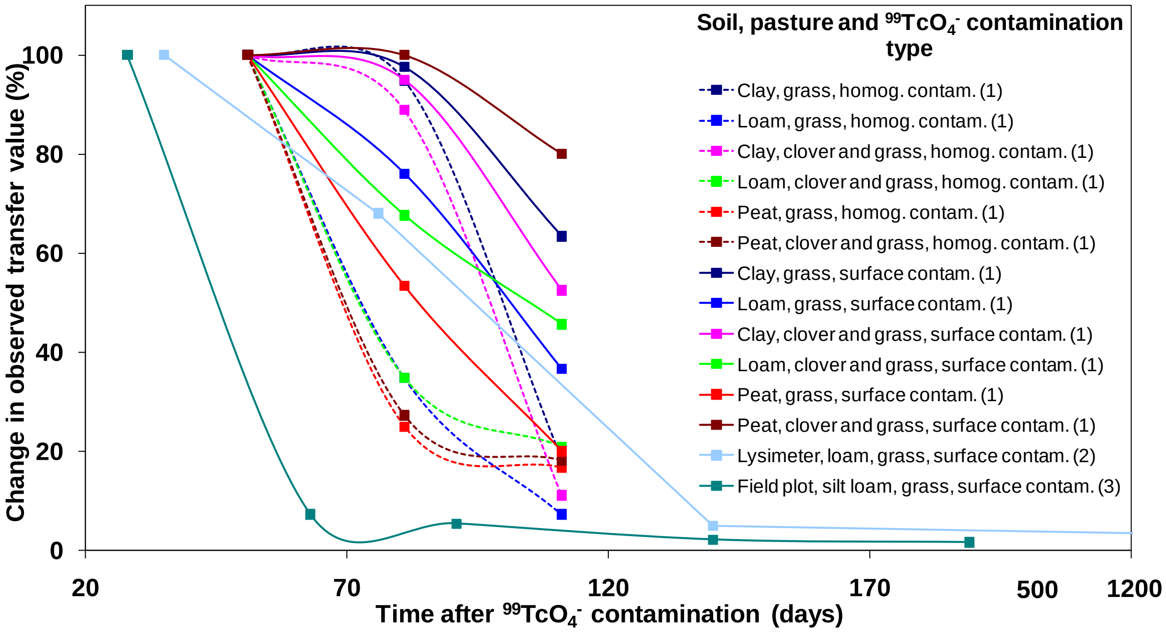

Figure 1 Relationship between soil to plant transfer of 99Tc in pasture and the age and type of contamination. Data extracted from Mousny and Myttenaere, 1982 (Pot study) (1); Vandecasteele et al, 1989 (Lysimeter study) (2); Garten et al, 1984 (Field plot study) (3).

Figure 2 shows that similar trends were observed from pot experiments investigating uptake into rye grass using two soil types homogeneously contaminated with different levels of activity of $^{99}TcO_4^-$ (Echevarria et al, 1994). Rye grass was sown straight after contamination and the results showed that for both soil types around a 70-85% reduction in the soil to plant transfer value was observed 90 days after contamination. The results from this study are therefore reasonably consistent with those summarised in Figure 1.

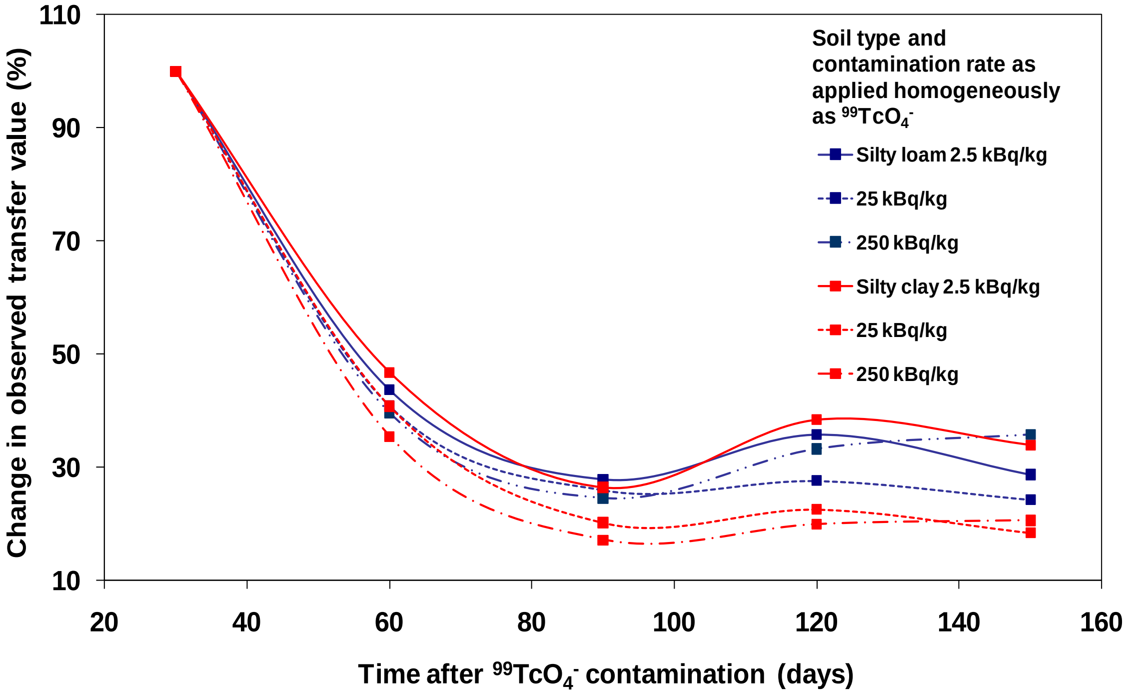

Figure 2 Relationship between soil to plant transfer of ^{99}Tc in pasture, soil type and age of contamination (Echevarria et al, 1994).

The variation of transfer factors with time can be rationalised in terms of speciation changes from the highly soluble and readily available TcO_4^- to less available and highly insoluble forms of technetium, such as TcO_2, sulphides and high molecular weight organic complexes (Sheppard et al, 1990; Tagami and Uchida, 1996). These effects have been ascribed to anaerobic conditions leading to the reduction of technetium to lower oxidation states (Stalmans et al, 1986). It may seem surprising that TcO_4^- can be reduced in soils that are aerobic. However, it is well known that soils cannot be considered homogeneous and that anaerobic centres can develop in apparently well drained aerobic soils. These are caused by depletion of oxygen by microbial growth in small water-filled pores, resulting in a sluggish, diffusion-controlled replenishment of oxygen (Currie, 1962; Greenwood and Goodman, 1964; Rowell, 1981).

Green et al (1995a, 1995b) performed root uptake experiments under field conditions using contaminated land reclaimed from the sea, where water logging had occurred. The observed soil to crop transfers were significantly lower than those often obtained following contamination of soil in pots, lysimeters and field plot studies under controlled conditions. The lower transfer values obtained in these field studies may have resulted from prolonged contact between ^{99}Tc and soil in the field, perhaps including periods of anaerobic conditions where the ^{99}Tc would be likely to reduce to a far less soluble form such as $^{99}TcO_2$. Alternatively, the ^{99}Tc could have been in a reduced form when adsorbed on to the marine sediment from which this soil was derived. The maintenance of a reduced form could have been further aided by the presence of organic matter, which under anaerobic conditions has been reported to strongly retard the movement of ^{99}Tc (Sheppard et al, 1990; Tagami and Uchida, 1996, 1997).

Some reports suggest that once ^{99}Tc is in an insoluble form such as $^{99}TcO_2$, it is oxidised very slowly limiting the release of TcO_4^- back into the soil solution (Sheppard and Evenden, 1991; Echevarria et al, 1997). Tagami and Uchida (1999) investigated the chemical change of ^{99}Tc in soil and noted that a dry aerobic period after water logging did not remobilise $^{99}TcO_2$. Sheppard and Evenden (1991) showed that aquatic macrophytes known for their ability to oxidise the rhizosphere (the zone that surrounds the roots of plants), around plants such as rice, were unable to oxidise technetium in flooded anaerobic soils sufficiently quickly to enhance its availability. The results of these studies support the fact that once technetium is oxidised to insoluble forms such TcO_2 it is unlikely to become readily available for plant uptake.

In many temperate areas, soils become waterlogged periodically, while those used for agriculture generally contain at least some organic matter. When taken together with the slow rate of oxidation of species such as TcO_2, these factors may account for a sustained reduction in the proportion of available technetium over time.

5.1.2 Complexation and soil type

Under aerobic conditions in soil, TcO_4^- does not form complexes with humic acid, but remains as a free ion in the soil solution (Van Loon and Lembrechts, 1984; Takahashi et al, 1999). Complexation of TcO_4^- with humic acid and other dissolved organic matter is only possible under more reducing conditions (Rößler et al, 2000). This is principally possible only under certain anaerobic conditions in the presence of pyrite containing rocks and the reducing effect of ferrous ion (Fe^{2+}) (Lieser and Bauscher, 1987). There are therefore only limited, infrequent conditions under which humic acid can form complexes with TcO_4^- and thereby influence its mobility.

In anaerobic soils, especially those high in organic matter, the movement of ^{99}Tc is strongly retarded, probably as an insoluble form such as TcO_2 (Sheppard et al, 1990; Tagami and Uchida, 1996, 1997). Bergstrom and Wilkens (1983) reviewed several studies that showed that the uptake of ^{99}Tc tended to be lower from organic compared to mineral soils. This conclusion is consistent with investigations by Sheppard et al (1990) who examined the variation in ^{99}Tc sorption among 34 different soil types. Among the 7 mineral soils studied, results implied that there would be very little sorption and that ^{99}Tc from these soil types would be readily taken up by crops under aerated conditions. In contrast, there was substantial sorption of ^{99}Tc in anaerobic conditions, especially in the presence of organic matter.

The uptake of $^{99}TcO_4^-$ into Swiss chard using mineral sand and peat soils has been investigated using a lysimeter scale study (Sheppard et al, 1983). The soils were homogeneously contaminated with different activity concentrations of $^{99}TcO_4^-$ and then left for a period of 70 days under moist conditions. Mature Swiss chard plants were then planted into the lysimeters and these were subsequently harvested after a further 40 days. The results showed that the observed soil to plant TFs for peat were significantly lower than those obtained for sand by around 2 orders of magnitude (Table 1).

TABLE 1 Observed soil to plant TFs for technetium in different soil types

Author	No of samples	Soil type	Contamination rate	Observed TF_{dry}[a]
Sheppard et al (1983)	15	Mineral sand	0.5 10^{-4} ug g^{-1}, ^{99}Tc	2100, Swiss chard
	15		5.0	2000
	15		50	3000
	15		500	2600
			Geometric mean	**2400**
	15	Peat	1.0 10^{-4} ug g^{-1}, ^{99}Tc	12, Swiss chard
	15		10	11
	15		100	32
	15		1000	40
			Geometric mean	**20**
Echevarria et al (1994)	1	Silty loam	2.5, kBq kg^{-1}, ^{99}Tc	100, rye grass
	1		25	118
	1		250	156
			Geometric mean	**123**
	1	Silty clay	2.5, kBq kg^{-1}, ^{99}Tc	123, rye grass
	1		25	153
	1		250	157
			Geometric mean	**143**

Notes

a. Expressed in terms of Bq kg^{-1} dry mass of crop and soil.

The study concluded that this reduction was due to the immobilisation of ^{99}Tc in the peat, most probably caused by $^{99}TcO_4^-$ reduction to a far less soluble form such as $^{99}TcO_2$. This conclusion is consistent with the statement that in peat soils, anaerobic conditions may have occurred and these, with the presence of organic matter, would have resulted in the reduction of the ^{99}Tc (Sheppard et al, 1990; Tagami and Uchida, 1996, 1997). It is also important to note that the plants were harvested 110 days after the soil had been contaminated, which is comparable to the period over which other work indicates that uptake into pasture decreased markedly (see Section 5.1.1). The mineral sand used in the study by Sheppard et al (1983) is not representative of agricultural soils found in the UK, where sandy soils would generally be augmented with organic matter to improve moisture retention and fertility. In such soils, which would still be well drained, largely aerobic and contain relatively small amounts of organic matter, it is conceivable that ^{99}Tc may, over a longer time period, undergo reduction to a less available form leading to lower uptake than observed in Sheppard et al (1983).

Echevarria et al, 1994 investigated uptake into rye grass using two soil types, silty loam and silty clay, homogeneously contaminated with different levels of activity of $^{99}TcO_4^-$. Results taken from a harvest 60 days after contamination showed little difference between the two soil types (Figure 2). It might be expected that water retention in the clay soil could cause anaerobic centres to develop leading to more rapid reduction of ^{99}Tc to a less plant available form compared to that of a well drained loam soil. However, due to the silty nature of both soil types they could be expected to be well drained and largely anaerobic in nature. This is demonstrated from further results of this study which shows that over a period of 90 days a reduction in plant transfer around a 70-85% was observed for both soil types (Figure 2).

The results from two studies (Sheppard et al, 1983 and Echevarria et al, 1994) shown in Table 1 indicate that for the soil types shown, the observed soil to plant TFs obtained are sensitive to the type of soil used but not sensitive to the concentration of ^{99}Tc in the soils. This is fundamental to using the TF approach and is in agreement with the work of others (Van Loon and Lembrechts, 1984; Murphy and Johnson, 1993; Yanagisawa and Muramatsu, 1993).

5.2 Factors affecting the transfer of technetium to animals

As previously mentioned in Section 3.2 the most important transfer pathway to animals is the ingestion of contaminated feed. Few publications have been found on the transfer of technetium into animal products and limited information could be located to support factors affecting its transfer. The evidence summarised in this Section is general to the transfer of radionuclides to animals and where specific information relating to technetium was found, this is discussed.

5.2.1 Factors influencing the estimation of transfer coefficients to animal products

A number of authors (Ennis et al, 1988a; Howard et al, 1989, 1996, 1997, 2007; Hansen and Hove, 1991, 1993; Belli et al, 1993; Assimakopoulos et al, 1994; Beresford et al, 1998, 2000) have reported variations in transfer coefficients due to factors including the age and body mass of the animals, physical and chemical form of the radionuclide and problems with estimating radionuclide intake and establishing if equilibrium in the animal has been reached. Two of these studies used technetium (Ennis et al, 1988a, Johnson et al, 1988). Some of these factors are discussed below.

5.2.1.1 Effects of age and body mass

Nalezinski et al (1996) reported that transfer coefficients of radionuclides are generally higher for animals with a lower body mass. For example, transfer coefficients to lambs will generally be larger than those to ewes.

Experiments carried out by Johnson et al (1988) obtained transfer coefficients for cows and goats that had been given ^{99m}Tc and ^{95m}Tc orally. A single oral dosage of $^{99m}TcO_4^-$ was given to 2 cows and 26 goats and $^{95m}TcO_4^-$ was given to 2 cows and 6 goats. Although differences between the two isotopes were noted, ratios of about 6 and 6.5 for the goat to cow milk transfer can be derived for ^{95m}Tc and ^{99m}Tc, respectively. These ratios are comparable and support the general assumption that the transfer coefficient for cows is about one-tenth of that for goats. However, the study also used $^{99}TcO_4^-$ and obtained transfer coefficients to goat's milk that were between 10 to 70 times greater than those obtained from using ^{95m}Tc and ^{99m}Tc, respectively. This high value for the transfer to goat's milk using ^{99}Tc was not expected and no clear explanation was offered by the authors. In addition, no value was given for ^{99}Tc transfer to cow's milk and so comparisons could not easily be made.

5.2.1.2 Physical and chemical form

The physical and chemical form of a radionuclide can affect its transfer to animals. Jones (1989) compared results obtained from goats using ^{95m}Tc where one group (control group) was given 5 MBq via a stomach tube and the second group of goats (abomasum group) received the same activity concentration injected directly into their abomasums (fourth-stomach). Samples of milk, urine and faecal material were collected twice daily for 200 hours after the administration of ^{95m}Tc. The percentage of the ^{95m}Tc dose given was measured in the samples and the results are shown in Table 2. The abosamal group secreted on average 9 times more ^{95m}Tc in milk than the control group. For the urine and faecal excretion there was no significant difference between the two groups. Although there is no further investigation, Jones (1983 and 1989) suggests that the microbial and other processes in the forestomachs of the goats can change the chemical form of the TcO_4^-, which could affect absorption in the intestines.

TABLE 2 Milk production and total secretion of ^{95m}Tc in the milk, urine and faeces of goats

Author	Group	No of goats	Milk (l^{-1}) Production	(% given dose)		
				Milk excretion	Urine excretion	Faecal excretion
Jones (1989)	Control	9	6.6 (1.8-10.0)[a]	0.1 (0.002-0.3)		
		11			1.2(0.4-2.5)	90.1 (71.4-100.4)
	Abosamal (fourth stomach)	2	7.8 (7.7-7.8)	0.9 (0.6-1.3)	3.4 (2.1-4.7)	83.7 (78.6-88.8)

Notes

a. Data ranges given in brackets, along with the mean from the study.

5.2.1.3 Equilibrium assumption

By definition, for a transfer coefficient to be valid the radionuclide activity concentration in the animal products must be at equilibrium with the dietary intake of the radionuclide. For milk, an approximate equilibrium is reached rapidly for many radionuclides. However, experimentss from which transfer coefficients are derived, are often not conducted for long enough for equilibrium to be reached in animal products. The requirement of equilibrium is often not reached for those radionuclides with a short physical half life. Equally, care must be taken when using radionuclides with long radioactive and biological half life's since the activity concentrations in tissues will not have equilibrated with diet by the time of slaughter. Hence, transfer coefficients derived from comparatively short-term experiments could potentially underestimate equilibrium transfer coefficients.

For technetium, many of the studies reviewed (Weichen et al, 1983; Bondietti and Garten et al, 1984; Thomas et al, 1984 and Ennis et al, 1988a) have tended to use the short half life gamma emitting radioisotopes, ^{99m}Tc and ^{95m}Tc with half life's of 6.0 hours and 61 days, respectively. As discussed above, due to the potential of equilibrium not being reached when using short half life isotopes, care is needed when interpreting such data (Howard et al, 2009). For example, Ennis et al (1988a) who studied the transfer to the milk of lactating goats using three pertechnetate (TcO_4^-) isotopes of technetium (^{99m}Tc, ^{95m}Tc, ^{99}Tc) demonstrated that the milk transfer coefficients increased with decreasing specific activity and increasing half-life.

5.3 Implications for the selection of parameter values for transfer of technetium to crops and animals

The uptake of technetium into crops depends critically on whether technetium is in an oxidised form as TcO_4^- or a reduced form such as TcO_2. The evidence summarised in Section 5.1 suggests that when considering the types of agricultural soil generally encountered in the UK, the age of the contamination is the most important factor in determining uptake of technetium into crops. Soil to crop TFs have been compiled for non-reduced and reduced forms of technetium but deciding on appropriate TF values for the non-reduced form are complicated by the rapid changes in the observed values over time (Figures 1 and 2). For the purposes of this review, it has been assumed that the technetium will be in a chemically reduced form in soils if the contamination has been present for more than about 90 to 120 days. The specific values used when considering data for each crop type are given in Section 7.

This review found few data for transfer coefficients for technetium in animal products. In addition, studies on the effects of chemical form on transfer to animal products were very limited with only two studies suggesting that this may have an effect (Ennis et al, 1988a and Jones, 1989). However, no further evidence could be found to support these claims. From the evidence found for crops, it might be expected that the chemical form of technetium could affect transfer to animal products. Due to the limited data found for animals, care was taken when interpreting such data with each study critically reviewed on a case by case basis.

6 REVIEW METHODOLOGY

Each source of data for the transfer of technetium from soil to crops was evaluated in relation to a set of criteria designed to eliminate those studies in which transfer parameters were not applicable for use in food chain transfer models or in the UK.

For crops, these criteria were as follows:

a individual results must be from matched crop and soil combinations; summary data from reviews were excluded;
b data on crops must relate to the edible parts grown to full maturity;
c minimum mass of 10kg of soil used for pot experiments (standard International Union of Radioecologists protocol), (IUR, 1989);
d clear and concise experimental design, eg, crop preparation prior to analysis, details on chemical form of technetium (if applied directly to soil), harvest periods, age of deposit, if applicable;
e plant species must be similar to those typically found in the UK;
f agricultural soils should be comparable with those found in the UK.

Data were also only considered to be admissible if it was evident that direct deposition or resuspension were not contributing significantly to the observed activity concentrations in the crop. Care was also taken to ensure that data were not duplicated. The application of the above criteria for crops meant that in some cases only subsets of published data were used.

For animal products rigid criteria were difficult to establish due to the lack of available data found for technetium. Subsequently, publications were reviewed on a case by case basis with consideration given to the factors that can effect the estimation of transfer coefficients.

For both crops and animals, some data that strictly were considered inadmissible have been included in the review because they support the interpretation and evaluation of the available data. Where such data have been used, they are clearly identified and discussed in the main body of the text. Data from inadmissible studies are presented in tables in Appendix A and are discussed in Appendix B.

6.1 Data recording

All information was taken into account when reviewing source papers against criteria and a recording sheet was designed so that data identified from publications could be recorded in a standard format.

6.2 Analysis and presentation of data in the review

Due to the inherent sampling uncertainties in individual field studies and/or low activity concentrations of technetium typically found in some crop types and animal products, there is considerable variability associated with individual measured data and the mean values calculated. Differences of an order of magnitude between different samples of the same crop from the same site were common. In addition to the variability associated with individual values, most datasets were small and wide ranging. Consequently, confidence intervals about the datasets were too large to be useful and have not been calculated.

The data were analysed with the objective of deriving representative transfer values for technetium for a given crop type or animal product which can be used in radiological assessments. All data taken from individual publications are expressed to the level of accuracy given by the authors. For the purposes of calculating representative transfer values, the data have been rounded to reflect the variability in the published values and the use of these data within radiological assessments. Typically, these representative values are presented to 1 significant figure.

The values obtained from the review have been compared with the default values currently used in the FARMLAND model (Brown and Simmonds, 1995) and with those published for crops in IAEA-1616 (IAEA, 2009) and for animal products in IAEA-364 (IAEA,1994).

Predictive models such as FARMLAND often use broad categories of crop. The FARMLAND model groups crops into six broad categories:

a pasture;
b green vegetables (includes salad vegetables, brassicas and legumes);
c root vegetables;
d potatoes;
e cereals (grain);
f soft fruit.

It is important to note that the crop categories considered in the literature can differ from those adopted in the model. Since one aim was to determine whether any sub-divisions of the broad categories used in the model were warranted, this review has divided crops into the following seven categories:

a pasture;
b salad vegetables and brassicas ;
c root vegetables;
d tubers such as potatoes;
e cereals;
f other crop types such as legumes and the onion family;
g soft fruit.

Swedes and turnips are strictly categorised as brassicas, however due to the fact that only the root is eaten, they were allocated to the root vegetable group.

Data on transfer from feed to animal products for technetium were limited. Transfer coefficients were grouped for different animal products such as meat, milk and eggs.

The default transfer factor values in the FARMLAND model are expressed in terms of the fresh mass of the crop and dry mass of the soil, since this is the form that is most appropriate for radiological assessments. However, comparisons of transfer values are best based on the dry mass of the crop because the effects of differences in moisture content in the individual crops are removed. Where comparisons are made between FARMLAND and the reviewed data in the main part of this report, the quoted TF values in FARMLAND are expressed in dry mass of the crop and do not correspond numerically with those published by Brown and Simmonds (1995). In Section 8 of this report any recommendations for default values used in FARMLAND have been given in terms of fresh and dry mass of crop. The percent of dry matter content used to convert FARMLAND values to dry mass which are given in Appendix C, Table C2 are:

a pasture, 26%;
b green vegetables (including brassicas and legumes), 13%;
c root vegetables, 10%;
d potatoes, 21%;
e cereals (grain), 90%;
f soft fruit 6%.

Where necessary, published values presented in this review were also converted to a dry mass basis either by using specific data given in the publication itself or by using existing data for specific crop types as detailed in Appendix C.

Data extracted from publications containing admissible soil to plant TF values for crop groups are presented in tables and are denoted TF_{dry}. They are presented as calculated geometric means from individual studies together with corresponding data ranges, if available. Inadmissible data, which support discussion on factors influencing transfer are discussed within the text. Inadmissible TF values are presented in tables denoted as either TF_{dry} or as CR_{dry} in Appendix A. An additional study providing inadmissible data is discussed in Appendix B. Both admissible and inadmissible data were further divided based on whether the technetium was assumed to be in a chemically reduced or non-reduced form. Finally, if sufficient data existed (from two or more studies) overall geometric means and ranges of the admissible values are then given.

For animal products, data were evaluated and are presented in a similar way to the crops. Transfer coefficients are discussed within the text and are presented in tables denoted as F_m for milk and F_f for other animal products and eggs in the tables. It should be noted that none of the transfer coefficient data reviewed were considered strictly admissible. This is discussed further in Section 7.2 and Section 8.2.

7 RESULTS AND DISCUSSION

7.1.1 Technetium uptake into pasture

A significant amount of data was found on the uptake of ^{99}Tc into pasture and forage, including one study that was carried out in the UK. Admissible soil to plant TFs on a dry mass basis are given in Table 3, where ^{99}Tc is assumed to be in non-reduced form and in Table 4, where it is assumed to be chemically reduced. A value of 90 days has been used as the cut-off for assuming the ^{99}Tc is in the non-reduced form. Inadmissible data that have been used to support the discussions in the main text are given in Appendix A, Table A1.

For technetium present in the non-reduced form, a geometric mean value of a TF of 140 with a data range of 30 to 440 has been derived from admissible data for pasture (Table 3). Data in Table 3 show that for forage crops such as pasture, the TF values can vary considerably. IAEA-1616 (IAEA, 2009) gives a geometric mean TF value for pasture of 76 with a range of values of 7.9 to 470. The value recommended in this review is around 2 times higher than that recommended by the IAEA. The range of values taken to derive a TF of 140 in this review was within the IAEA range of values.

The TF for pasture when the technetium is assumed to be in a reduced form (elapsed time from initial contamination greater than 90 days) are shown in Table 4 and are drawn from 3 studies. In one of these studies, the TF values at 112 and 140 days after contamination were slightly greater than those after 90 days (Echevarria et al, 1994). However, given the likely measurement uncertainties and the assumption that the technetium will reduce over a period of around 90 to 120 days, these differences are not significant.

A geometric mean TF from Table 4 of 20 has been derived. This is within the data range of 7.9 to 470 specified in IAEA-1616 (IAEA, 2009). The lower end of the range of data found by this review includes results from field studies carried out by Green et al (1995a) in the UK who obtained TF values of around 2 for root uptake into pasture. These relate to aged technetium deposits of marine origin and are very much lower than the other values in Table 4 and are outside the data range published in IAEA-1616 (IAEA, 2009). IAEA did not consider the Green et al (1995a) study in their publication.

TABLE 3 Admissible soil to plant TFs for technetium uptake into pasture where it was assumed to be in a non-reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} Pasture[a]	Age of contamination (days)[b]
Mousny and Myttenaere (1982)	3	Pot experiment, grass, 10-15kg clay soil,	$^{99}TcO_4^-$, homogeneous contamination	190	51
				180	81
	3	Pot experiment, grass, 10-15kg loam soil	$^{99}TcO_4^-$, homogeneous contamination	345	51
				120	81
	3	Pot experiment, grass, 10-15kg peat soil	$^{99}TcO_4^-$, homogeneous contamination	120	51
				30	81
	3	Pot experiment, clover and grass, 10-15kg clay soil,	$^{99}TcO_4^-$, homogeneous contamination	180	51
				160	81
	3	Pot experiment, clover and grass, 10-15kg loam soil,	$^{99}TcO_4^-$, homogeneous contamination	345	51
				120	81
	3	Pot experiment, clover and grass, 10-15kg peat soil	$^{99}TcO_4^-$, homogeneous contamination	110	51
				30	81
Echevarria et al (1994)	30	Pot experiment, rye grass, 10kg silty loam soil	$^{99}TcO_4^-$, 2.5 kBq kg^{-1}, homogeneous contamination	230	28
				100	56
				60	84
	30	Pot experiment, rye grass, 10kg silty loam soil	$^{99}TcO_4^-$, 25 kBq kg^{-1}, homogeneous contamination	290	28
				120	56
				75	84
	30	Pot experiment, rye grass , 10kg silty loam soil	$^{99}TcO_4^-$, 250 kBq kg^{-1}, homogeneous contamination	400	28
				160	56
				100	84
	30	Pot experiment, rye grass, 10kg silty clay loam soil	$^{99}TcO_4^-$, 2.5 kBq kg^{-1}, homogeneous contamination	260	28
				120	56
				70	84
	30	Pot experiment, rye grass, 10kg silty clay loam soil	$^{99}TcO_4^-$, 25 kBq kg^{-1}, homogeneous contamination	375	28
				150	56
				80	84
	30	Pot, rye grass , 10kg silty clay loam soil	$^{99}TcO_4^-$, 250 kBq kg^{-1}, homogeneous contamination	440	28
				160	56
				75	84
Geometric mean and data range of studies				140 (30-440)	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to the nearest decade where given to a higher level of precision by the authors or where values have been converted from values based on fresh mass of crop.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times up to 90 days were used to represent technetium assumed to be in a non-reduced form.

Long term field studies have been carried out where ^{99}Tc activity has been applied directly to the surface of the soil (Garten et al, 1984), and similar studies have been undertaken using lysimeters (Vandecasteele et al, 1989). The results from these studies are given in Appendix A, Table A1. Due to the selection criteria adopted, the way in which the activity was applied strictly renders these data inadmissible. However, the observed values in the longer period of time after contamination obtained from these two studies show that the TF values are as low as those derived by Green et al (1995a), and are at the lower end of the range of admissible data in Table 4.

Results from field measurements made in the vicinity of operational gaseous diffusion plants are summarised in Appendix A, Table A1 (taken from Hoffman et al, 1982; Hoffman Jr, 1982). These data were considered inadmissible because of the possible effects of direct deposition on to the foliage from ongoing discharges, together with the likelihood of non-uniform distribution of activity with depth in the soil. The observed CR values with a range of 1-44 are also at the lower end of the range of admissible data in Table 4 supporting the evidence that technetium in soils reduces over time. Although the sampling protocol adopted meant that the data were inadmissible for the purposes of this review, monitoring data for sea-washed pasture in northwest England (MAFF, 1997) inferred CR values in the range 0.03 - 1 for technetium which are also very low. These very low values could be the result of the generally high moisture content of sea-washed soils which would result in anaerobic conditions causing the reduction of technetium to forms that are far less available to plants (see Section 5.1.1)

TABLE 4 Admissible soil to plant TFs for technetium uptake into pasture where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF$_{dry}$ Pasture[a]	Age of contamination (days)[b]
Mousny and Myttenaere (1982)	3	Pot, grass, 10-15kg clay soil,	^{99}TcO$_4^-$, homogeneous contamination	35	111
	3	Pot, grass, 10-15kg loam soil	^{99}TcO$_4^-$, homogeneous contamination	25	111
	3	Pot, grass, 10-15kg peat soil	^{99}TcO$_4^-$, homogeneous contamination	20	111
	3	Pot, clover and grass, 10-15kg clay soil,	^{99}TcO$_4^-$, homogeneous contamination	20	111
	3	Pot, clover and grass, 10-15kg loam soil,	^{99}TcO$_4^-$, homogeneous contamination	25	111
	3	Pot, clover and grass, 10-15kg peat soil	^{99}TcO$_4^-$, homogeneous contamination	20	111
Echevarria et al (1994)	5	Pot, grass, 10kg silty loam soil	^{99}TcO$_4^-$, 2.5kBq kg^{-1}, homogeneous contamination	100	112
				70	140
	5	Pot, grass, 10kg silty loam soil	^{99}TcO$_4^-$, 25kBq kg^{-1}, homogeneous contamination	80	112
				70	140
	5	Pot, grass, 10kg silty loam soil	^{99}TcO$_4^-$, 250kBq kg^{-1}, homogeneous contamination	130	112
				140	140
	5	Pot, grass, 10kg silty clay loam soil	^{99}TcO$_4^-$, 2.5kBq kg^{-1}, homogeneous contamination	100	112
				90	140
	5	Pot, grass, 10kg silty clay loam soil	^{99}TcO$_4^-$, 25kBq kg^{-1}, homogeneous contamination	90	112
				70	140
	5	Pot, grass, 10kg silty clay loam soil	^{99}TcO$_4^-$, 250kBq kg^{-1}, homogeneous contamination	90	112
				90	140
Green et al (1995a)	Not known	Field study, grass, UK based	Aged of marine origin	2 (1 – 4)[c]	Aged
Geometric mean and data range of studies				20 (1–140)	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to the nearest decade where given to a higher level of precision by the authors or where values have been converted from values based on fresh mass of crop.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

7.1.2 Technetium uptake into salad vegetables and brassicas

All of the data considered admissible for salad vegetables and brassicas crops are for soil that had been contaminated for a period of time long enough for it to be assumed that the technetium is chemically reduced. These data are summarised in Table 5. Inadmissible data that have been used to assist the discussion are given in Appendix A (Tables A2, A3 and A4) and Appendix B.

Table 5 shows that only two studies reported data that are considered admissible. One was a UK based field study carried out with aged deposits of marine origin where TFs for both salads and brassicas were reported (Green et al,1995b). The other admissible study, Sheppard et al (1983), provided data on the uptake of technetium into brassicas (Swiss chard) grown in lysimeters containing contaminated peat. The values for Swiss chard in Sheppard et al (1983) are at least 10 times higher than those reported by Green et al (1995b). These higher values are comparable with results for Swiss chard from an inadmissible study shown in Appendix A (Table A4) (Garten et al, 1986). These two studies provide some evidence that chard may have higher root uptake than other brassicas. The results from the inadmissible study on transfer to crops from soil amended with seaweed described in Appendix B, showed that Swiss chard, broccoli, kale and spinach are crops with the highest CR values compared with of the rest of the crops studied (Figures B1 and B3). These data provide evidence that supports the fact that root uptake for leafy green vegetables such as chard and spinach is higher.

Sheppard et al (1983) also investigated the uptake of $^{99}TcO_4^-$ into Swiss chard grown in mineral sand (Appendix A, Tables A2) and obtained TFs around two orders of magnitude higher when compared with those obtained from peat soil (Table 5). In both cases, the crops were harvested 110 days after the soils had been contaminated, which is comparable with the period over which the uptake of technetium can be assumed to be reduced in typical soils used to grow crops in the UK. Transfer values obtained from the mineral sand, although regarded as soil to plant TFs, are not considered admissible for the purposes of this review due to the fact that mineral sand is not normally used to grow crops in the UK. Data from Appendix A, Table A2 show that the Swiss chard was harvested 110 days after contamination, slightly greater than the suggested 90 day cut off period when the technetium can be assumed to be reduced. However, the values are much higher than would be expected if the technetium was reduced. In this case the technetium may not be in a reduced form because of the low organic matter content in mineral sand.

TABLE 5 Admissible soil to plant TFs for technetium for salad vegetables and brassicas where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry}[a]	Age of contam. (days)[b]
Green et al (1995b)	4	Field, UK	Aged of marine origin	4 (lettuce)[c] 1 (0.9-2)[d] (cabbage) 0.9 (cauliflower) 1.5 (sprouts)	Aged
Sheppard et al (1983)	60	Lysimeter, peat soil	homogeneous contamination, 1.0 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	12 (Swiss chard)	110
			Homogeneous contamination, 10 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	11 (Swiss chard)	110
			Homogeneous contamination, 100 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	32 (Swiss chard)	110
			Homogeneous contamination, 1000 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	40 (Swiss chard)	110
Geometric mean and data range of studies			Brassicas	4 (1 – 40)	
			Salad vegetables	Insufficient data[e]	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

c. Crop types are given in brackets.

d. Data ranges given in brackets, along with the geometric mean from the study.

e. Only one study (Green et al, 1995b) provided admissible data for one crop type (lettuce) which was considered insufficient for the derivation of an overall geometric mean, see text.

Studies conducted in Japan by Yanagisava and Muramatsu (1993) provided data on the root uptake of technetium into a variety of crops grown in pots in a growth chamber containing an organic soil that had been contaminated homogeneously with $^{95m}TcO_4^-$. The data are summarised in Appendix A, Table A3, but were not considered admissible because the amount of soil used for each pot was only 3 kg. The age of the contamination in these experiments was 85 days, around the time when it is thought that technetium becomes reduced in the soil and root uptake becomes significantly lower. However, it is worth noting that since the crops were grown in an organic soil the technetium may become reduced in a shorter period of time. The values are comparable to those from soils where it is believed that the technetium was in a reduced form such as the values for cabbage from field experiments conducted by Green et al (1995b), given in Table 5.

Results from a pot study carried by Garten et al (1986) were not considered admissible because the amount of soil used for each pot was estimated to be around 8 kg, slightly less than criteria considered acceptable for this review. The observed values (given in Appendix A, Table A4) are, however, comparable with those from a controlled lysimeter study for the same crop type (Table 5).

A geometric mean TF for brassicas of 4 for technetium in a reduced form can be calculated. Admissible values found by this review for brassicas are at the lower end of those given for different soils in IAEA-1616 (IAEA, 2009). TF values in the range of 4.5 to 3400 with a geometric mean of 180 for leafy vegetables were published by IAEA for a mixture of soils (sand and loam). The reported data range in IAEA-1616 is large and as discussed previously, the criteria used to evaluate suitability of data for inclusion in this review may be different to that used by IAEA, since they are specific to the UK. Consequently, it might be expected to observe a difference in reported data ranges.

For salad crops, only one study gave admissible data for reduced technetium and so a geometric mean could not be derived. Limited evidence from this review suggests that, for soil that has been contaminated for many years, uptake is likely to be similar to that of brassicas. IAEA-1616 (IAEA, 2009) does not include a separate crop category for salads and so comparisons cannot be made.

For salad vegetables and brassicas, if the soil has only been contaminated for a few weeks or months prior to harvest, then a higher uptake might be expected. No direct experimental evidence to support this has been found but for pasture, TF values for soil that had been contaminated for a few months were around an order of magnitude greater than those observed for aged deposits (Tables 3 and 4).

7.1.3 Technetium uptake into root crops

Very few data have been published on the uptake of technetium into root crops with only one UK based study reporting admissible TF values (Green et al, 1995b). Data from this study are summarised in Table 6 and are for a field investigation on land that had been reclaimed from the sea. Values of TF of 1 – 3 were reported for 3 different types of root crops.

TABLE 6 Admissible soil to crop TFs for technetium uptake into root crops where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} root crops[a]	Age of contamination (days)[b]
Green et al (1995b)	4	Field, UK	Aged of marine origin	3(2-3)[c] (beetroot)[d] 2 (carrots) 1 (swede)	Aged
Geometric mean and data range of studies				Insufficient data[e]	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Crop types are given in brackets

e. Only one study provided admissible data which was considered insufficient for the derivation of an overall geometric mean, see text.

For carrots, TF values of approximately 2 were also obtained from pot experiments carried out over a period of around 90 days using an organic soil (Yanagisava and Muranmatsu, 1993). These data are in good agreement with the field data in Table 6, but were considered inadmissible since they were obtained from crops grown in only 3 kg of soil. The data from these pot experiments are given in Appendix A, Table A5.

IAEA Technical Document 1616 (IAEA, 2009) gives a geometric mean TF for technetium in root crops of 46 with a range covering 14 – 79. The Green et al (1995b) study has not been considered by IAEA and in addition, IAEA reported values that were obtained from two studies which include a mixture of soils which did not meet the criteria used in this review. It should be noted that the low TFs found in Green et al (1995b) can be supported with data presented in Appendix B also for technetium of marine origin. However, since these data are considered to be CR values they were not considered admissible for the purposes of this review.

7.1.4 Technetium uptake into potato tubers

Similarly to root vegetables, very few data have been published on the uptake of technetium into potatoes, with only one UK field study reporting admissible TF values (Green et al, 1995b). Data from this study are given in Table 7 and are for an aged technetium deposit of marine origin. Values of TF of around 0.1 - 0.7 were observed which are comparable with values from a study summarised in Appendix A (Table A6) in which sweet potatoes were grown in pots containing organic soils homogenously contaminated with $^{95m}TcO_4^-$ (Yanagisava and Muramatsu, 1993). The potatoes in this study were harvested 90 days after contamination around the same time period that it can be assumed that the technetium would be in a chemically reduced form. Values of TF from this study were not strictly admissible since the crops were grown in only 3 kg of soil.

Admissible values found by this review for potato tubers are within the range of those given for different soil types in IAEA-1616 (IAEA, 2009). TF values in the range of 0.013 - 0.65 with a geometric mean of 0.23 were published by IAEA for a mixture of soils (sand and loam).

TABLE 7 Admissible soil to plant TFs for technetium uptake into potato tubers where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} potato tubers[a]	Age of contamination (days)[b]
Green et al (1995b)	3	Field, UK	Aged of marine origin	0.3 (0.1 – 0.7)[c]	Aged
Geometric mean and data range of studies				Insufficient data[d]	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Only one study provided admissible data which was considered insufficient for the derivation of an overall geometric mean, see text.

7.1.5 Technetium uptake into cereal crops

Only one lysimeter study reported data considered admissible in this review (Eriksson, 1982), which are summarised in Table 8. The soils from the lysimeter study were homogenously contaminated with $^{99}TcO_4^-$ and left for a period of about 2 years before wheat was grown. The investigation observed that TF values continued to decrease in the grain from the first harvest to the next. The study concluded that, over time, the TcO_4^- was being converted to an insoluble form which was less available for root uptake; a conclusion reached by many studies found in this review. However, further reductions (although small) in root uptake were still being observed in the period from 2 to 3 years after the initial contamination, much later than the period of around 90 days at which most of the technetium can be expected to be in a chemically reduced form. This continued long-term reduction in uptake is consistent with the lower TF values observed using aged deposits in field studies compared with those found in other studies (Green et al, 1995b).

TABLE 8 Admissible soil to plant TFs for technetium uptake into cereals where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} Cereals[a]	Age of contamination (days)[b]
Eriksson (1982)	24	Lysimeter study, various soil types	Contaminated with 370 kBq, $^{99}TcO_4^-$	0.11(0.02-0.2)[c] (wheat grain)[d]	730
				0.05(0.02-0.1) (wheat grain)	1095
Geometric mean and data range of studies				Insufficient data[e]	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Crop types are given in brackets.

e. Only one study provided admissible data which was considered insufficient for the derivation of an overall geometric mean, see text.

The 2009 IAEA review (IAEA, 2009) gives a range of TF values for cereal grain of 0.18 – 2.4 and a wide range for maize grain of 0.50 – 52 with a geometric mean of 3.8. The values observed in this review were either outside or at the lower end of the range given in IAEA-1616. As previously discussed, the criteria used to evaluate suitability of data for inclusion in this review may be different to IAEA since they are specific to the UK. Consequently, it might be expected to observe a difference in reported data ranges.

Other studies on cereal crops were located but the data were considered inadmissible due to the mass of soil used for the growing experiments. However, they have been used to support discussion and are summarised in Appendix A (Tables A7 and A8). A comparison of the root uptake of technetium by rice and wheat has been made using small-scale pot experiments (Yanagiswa et al, 1992). Further studies were subsequently carried out on uptake to rice only, one involving a comparison between flooded and non-flooded soils (Yanagiswa and Muramatsu, 1995a) and the other a comparison between two different soil types, both under flooded conditions (Yanagiswa and Muramatsu, 1995b). The CR values for rice grown in flooded conditions were lower than those obtained for wheat grain. However, the CR values for rice, which was grown under non-flooded conditions, were considerably higher than those obtained using flooded soil and were similar to those obtained in the earlier study for wheat. There was a difference between the two different soil types, but both were well below the value obtained for a non-flooded soil. Although the age of contamination was not given, these studies help support the evidence that under anaerobic soil conditions, which would exist in a flooded environment, the technetium would be expected to be far less available for plant uptake (Appendix A, Table A7).

The remainder of the results given in Appendix A, Table A8 involved growing wheat to maturity in small pots over the period when the technetium is expected to be non-reduced.

7.1.6 Technetium uptake into other crop types

Limited data were found on the root uptake of technetium into other crop types. Of the two studies found, only one provided TF values considered admissible for the purposes of this review (Green et al, 1995b). The field study giving admissible data involved aged deposits of marine origin and the results are summarised in Table 9. The TF values were in the range 0.1 - 1.2 for legumes, 0.25 - 2.6 for onions and around 2 for tomatoes.

Data from a pot scale study on uptake to onions and tomatoes were considered inadmissible because of the mass of soil that was used (Yanagisava and Muramatsu, 1993). The data are summarised in Appendix A, Table A9. The CR values for onions are similar to those obtained in the field study set out in Table 9, whereas for tomatoes they were slightly lower. Although inadmissible, the study described in Appendix B provides valuable information on the difference in transfer between crop species and provides CR values broadly similarly to those found by this review. For tomatoes, a CR value on a dry mass basis of $9\ 10^{-3}$ was found which is significantly lower than the value for tomatoes found in this review. For legumes, CR values in the range of $8\ 10^{-3}$ – $4\ 10^{-1}$ were found which are also lower than those found in the review.

IAEA Technical Document 1616 (IAEA, 2009) reports values for legumes of 1.1 - 30.0 with a geometric mean of 4.3 for a mixture of soils (sand and loam). The values from this review were either outside or at the lower range of values published by IAEA. As previously discussed, the criteria used to evaluate suitability of data for inclusion in this review may be different to IAEA since they are specific to the UK. Consequently, it might be expected to observe a difference in reported data ranges.

TABLE 9 Admissible soil to plant TFs for technetium uptake into other crop types where it is assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} legumes[a]	TF_{dry} onions[a]	TF_{dry} soft fruit[a]	Age of contam (days)[b]
Green et al (1995b)	9	Field, UK	Aged of marine origin	0.6 (mange tout)[c] 0.1 (peas) 1.2 (French beans) 0.1 (broad beans)	0.3 (0.25-0.3)[d] (onion from seed) 0.9 (0.3-2.6) (onion from sets) 0.4 (leeks)	2 (tomato)	Aged for all crops
Geometric mean and data range of studies				Insufficient data[e]	Insufficient data[e]	Insufficient data[e]	

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

c. Crop types are given in brackets.

d. Data ranges given in brackets, along with the geometric mean from the study.

e. Only one study provided admissible data which was considered insufficient for the derivation of an overall geometric mean, see text.

7.2 Technetium transfer from feed to animal products

7.2.1 Technetium transfer from feed to milk

Few published studies on transfer coefficients for technetium from feed to milk, F_m are available, with none of them from the UK. The published data on transfer coefficients for technetium from feed to milk, F_m for cows and goats are given in Table 10.

Ennis et al (1988a) which is an accompanying paper to experiments conducted by Johnson et al (1988) reported the transfer of technetium to the milk of goats to be $1.5\ 10^{-4}$ d l^{-1}, $8.5\ 10^{-4}$ d l^{-1} and $1.1\ 10^{-2}$ d l^{-1} for ^{99m}Tc, ^{95m}Tc and ^{99}Tc, respectively. The authors found that the milk transfer coefficients increased with decreasing specific activity and increasing half-life.

Goats are often used experimentally to derive transfer coefficients for cattle. Investigations by Johnson et al (1988) compared transfer coefficients for both milk and meat from dairy cows and goats using different isotopes of technetium; the results are shown in Table 10. Literature surveys carried out by Ng and Hoffman (1983), Till et al (1985) and Hoffman and Baes (1979) suggest that transfer coefficients are around a factor of 10 higher in goats than cows and that goat's milk could therefore provide a reliable estimate of the transfer of ^{99}Tc to cow's milk. Results from studies conducted by Johnson et al (1988) showed this to be the case only for ^{99m}Tc (Table 10). However, care must be taken when interpreting data from this study since the authors used the short lived radioisotopes, ^{99m}Tc and ^{95m}Tc and obtained F_m values that were reported by Ennis et al (1988a) to be inversely proportional to the specific activity of the isotopes. Since it was not clear whether these differences could be due to the fact that equilibrium had not been reached in the animal products, the data were considered inadmissible for the purposes of this review. However, a value for transfer to goat milk of $1.1\ 10^{-2}$ using

^{99}Tc was also derived from this study. Although there is no corresponding value for cow's milk for comparison, this value should be considered as a potential recommended value.

Field studies using 12 dairy goats grazing on pasture that had been spray contaminated with ^{95m}Tc were conducted by Bondietti and Garten Jr (1984). The pasture to milk F_m value derived was around 1.4 10^{-4} d l^{-1} (Table 10). The authors considered that equilibrium had been reached around 5 days after contamination. The values from this study are in reasonable agreement with some of those published by Johnson et al (1988).

Wiechen et al (1983) derived F_m values for cow's milk using ^{99m}Tc administered using a fistula and obtained values significantly lower than most of the studies found by this review. However, this study was considered inadmissible for the purpose of this review since experimental details were not clear.

Technetium in the form of a single dose of $^{95m}TcO_4$ was administered to a three year old lactating cow in studies conducted by Voigt (1988). However, due to the maximum activity available (37 kBq) the cow could only be milked once at around 10.5 hours after application before the concentration in the milk was measured at below the minimum detectable activity (MDA). The authors reported an F_m value in milk from the first milking of 5 10^{-6} d l^{-1} with an estimated upper limit of 1.7 10^{-4} d $l^{-1.}$ The upper limit was derived on the assumption of an activity concentration in the milk at the level of the MDA after day 1 decreasing with the physical half-life. Due to the uncertainties and the fact that a state of equilibrium in the animal product could not be demonstrated, the values are not considered admissible by this review.

TABLE 10 Transfer coefficients from feed to milk of cows and goats

Author	No of samples	Study type	$F_m{}^a$(x 10^{-4})		Comments
			Cow	Goat	
Weichen et al (1983)	Not known	^{99m}Tc administered using a rumen fistula	0.03 (0.001-0.1)[b]		Inadmissible
Johnson et al (1988)	4	Orally administered capsule of $^{99}TcO_4^-$		110	For consideration
	28	Orally administered capsule of $^{99m}TcO_4^-$	0.23	1.5	Inadmissible
	8	Orally administered capsule of $^{95m}TcO_4^-$	1.4	8.5	Inadmissible
Voigt (1988)	Not known	Orally administered dose of $^{95m}TcO_4^-$	0.05, 1.7		Inadmissible
Bondietti and Garten Jr (1984)	Not known	Field study, $^{95m}TcO_4^-$ spray contaminated pasture		1.4 (0.8 - 1.8)	Inadmissible
Geometric mean and data range of studies			Insufficient data[c]		

Notes

a. F_m denotes the fraction of the daily intake by ingestion transferred to a litre of milk.

b. Data ranges given in brackets, along with the geometric mean from the study.

c. Data considered insufficient for the derivation of an overall geometric mean, see text.

The same sources of data were used in IAEA-364 for ^{99m}Tc and ^{95m}Tc in cow's milk (Johnson et al, 1988). Similarly, for goat's milk IAEA used the same sources of data for ^{99}Tc, ^{99m}Tc and ^{95m}Tc and so comparisons cannot be made with data included in this review.

Insufficient data are available to recommend a value of F_m in either cow or goat's milk from any of the studies found by this review. The values for cow's milk for studies using ^{99m}Tc and ^{95}Tc are not significantly different to the current recommended default value of 3 10^{-4} d l^{-1} in the FARMLAND model.

7.2.2 Technetium transfer from feed to meat products

Only one publication could be found reporting F_f values in cow and goat meat (Table 11). The literature surveys described earlier (Ng and Hoffman, 1983; Till et al, 1985 and Hoffman and Baes, 1979) suggested that F_f values are around a factor of 10 higher in goats than in cows. More research is required, but limited experimental results from the Johnson study showed that this is not the case. The transfer coefficient derived for goat meat by Johnson et al (1988) using ^{99m}Tc obtained an F_f value of 2.2 10^{-4}, over 300 times greater than the corresponding value derived for cow meat. In addition, care must be taken when interpreting data from this study since the authors used short lived ^{99m}Tc. Given the half life of ^{99m}Tc is 6.0 hours and there are no details of whether equilibrium was reached in the animal products, the data are considered inadmissible for the purposes of this review.

Experimental values obtained by this review cannot be compared to those recommended in IAEA-364 since they were derived from the same source (Johnson et al, 1988). No recommended values were calculated in this review for F_f in beef or goat meat.

TABLE 11 Transfer coefficients from feed to animal products

Author	No of samples	Study type	F_f meat[a] ($\times 10^{-4}$)	
			Cow	Goat
Johnson et al (1988)[b]	29	Orally administered capsule of $^{99m}TcO_4^-$	0.007	2.2

Notes

a. F_f meat denotes the fraction of the daily intake by ingestion transferred to a kilogram of muscle

b. Data from this experiment is deemed inadmissible, see text.

7.2.3 Technetium transfer from feed to poultry products

Japanese quail are often considered a suitable substitute for commercial laying hens when studying transfer to meat and eggs (Wilson et al, 1961 and Woodward et al, 1973) and these birds were used to obtain transfer coefficients in investigations conducted by Thomas et al (1984). The quails were fed alfalfa grown on solutions containing ^{95m}Tc and results were obtained for quail meat, offal and eggs. The investigation indicated that the biological half life in quail meat was approximately 1-2 days, since each quail reached equilibrium (about 5 biological half-lives) after 5-10 days of chronic ingestion. The study concluded that equilibrium in the eggs was reached at around 7 days. Table 12 shows the data that were published on the transfer of technetium from feed to poultry products.

Values for transfer to chicken meat and hens' eggs of 0.2 and 2, respectively, have been published by Ng (1982). These values were derived from a literature review and it was not possible to obtain the original material; consequently they were not considered admissible by this review.

The F_f value recommended in IAEA-364 (IAEA, 1994) for ^{95m}Tc in chicken meat and eggs cannot be compared to experimental results since it was obtained from the same source (Ennis et al, 1988b) and no default values are given for the uptake of technetium from feed to poultry products in the FARMLAND model.

The experimental values for F_f in chicken and quail meat were less than those quoted by Ng (1982) and, for hens' eggs the experimental values were in reasonable agreement, with those for quails eggs being slightly greater.

TABLE 12 Transfer coefficients from feed to quail and chicken products

Author	No of samples	Study type	F_f meat[a]		F_f other[b]			F_f eggs[c]	
			Chicken	Quail	Quail			Hen	Quail
					Gizzard	Liver	Heart		
Ennis et al (1988b)	6	Orally administered capsule of $^{95m}TcO_4^-$	0.03					3	
Thomas et al (1984)		$^{95m}TcO_4^-$ contaminated feed		0.09 (0.07-0.12)[d]	3.8 (3.7-4)	0.8 (0.6-1.2)	0.3 (0.26-0.34)		8.7 (6.1-11.4)
Ng (1982)		Literature survey	0.2					2	
Geometric mean and data range of studies			Insufficient data[e]		Insufficient data[e]			Insufficient data[e]	

Notes

a. F_f meat denotes the fraction of the daily intake by ingestion transferred to a kilogram of muscle.

b. F_f other denotes the fraction of the daily intake by ingestion transferred to a kilogram of offal.

c. F_f eggs denotes the fraction of the daily intake by ingestion transferred to eggs.

d. Data ranges given in brackets, along with the geometric mean.

e. Data considered insufficient for derivation of an overall geometric mean, see text.

8 RECOMMENDED VALUES AND IMPLICATIONS FOR THE FARMLAND MODEL

8.1 Recommended soil to plant transfer factors for technetium

The chemical form of technetium in the soil determines the degree to which it is available for uptake to crops. Due to the differences in the behaviour of the reduced and non-reduced forms of technetium in the environment, two geometric means were calculated from admissible data, one for each chemical form.

Table 13 gives recommended soil to crop transfer factors on a fresh mass basis for pasture and brassicas which are the only two crops for which geometric means can be calculated. The recommended value for pasture is 5 for the reduced form and 40 for the non-reduced form of ^{99}Tc. For brassicas, a value of 0.5 is recommended for technetium in a reduced form. Data are not available to recommend a value for the non-reduced form.

Table 14 gives values where geometric means could not be derived because, in most cases, only one admissible study was found. It is difficult to draw conclusions and recommend TF values for each category of crop when the values are only based on one admissible study. Therefore, a generic soil to crop TF has been considered for the recommended value, which comprises all categories of crop except pasture.

TABLE 13 Recommended soil to crop transfer factors for technetium on a fresh and dry mass basis by this review

Crop	Recommended by this review				FARMLAND (Brown and Simmonds, 1995)	
	reduced technetium		non-reduced technetium			
	Dry[b]	Fresh[c]	Dry[b]	Fresh[c]	Dry[b]	Fresh[c]
Brassicas[a]	4	0.5	NA	NA[e]	40	5
Pasture	20	5	140	40	20	5

Notes

a. FARMLAND considers salads, legumes and brassicas to be part of its green vegetable crop category (see Section 6.2).

b. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

c. Expressed in terms of Bq kg^{-1} fresh mass crop and Bq kg^{-1} dry mass soil.

d. NA, no appropriate data available.

For all the crop types considered in Table 14 the overall geometric mean TF (on a fresh mass basis) for technetium in a reduced form is 0.11 with a range of values between 0.04 and 0.2. From the CR data shown in Appendix B (Figures B1 and B2) a generic value of 0.1 could be considered acceptable for most of the crops except salad vegetables and brassicas; high values of soil to crop TFs for salad vegetables and brassicas are discussed in Section 7.1.2. For example, Sheppard et al 1983, reported high soil to crop TFs for Swiss chard grown in a peat soil, with a range of values of 1.0 - 5.0 and data from Brown et al (2009) show several CR values for salads and vegetables around 1. For this reason, it was decided that a generic TF value of 0.5 would be appropriate and although this is a conservative value for most of the other crops, it encompasses salads and brassicas.

There are no values reported for the TF for non-reduced technetium for edible crops. From Table 13, the ratio for pasture of the TF values for non-reduced:reduced technetium is 8. A ratio of 10 is therefore suggested as a robust value that can be applied for other crop categories to take account of the difference that could be expected in values of TF for non-reduced technetium compared to the recommended value for the reduced form.

The recommended values for TF to be used in radiological assessments in the UK and within the FARMLAND model are shown in Table 15.

TABLE 14 Soil to crop transfer factors for technetium on a fresh and dry mass basis

Crop	Transfer factors found from individual studies				FARMLAND (Brown and Simmonds, 1995)	
	reduced technetium		non-reduced technetium			
	Dry[b]	Fresh[c]	Dry[b]	Fresh[c]	Dry[b]	Fresh[c]
Salad vegetables[a]	4	0.2	NA	NA[e]	40	5
Legumes[a]	0.3	0.07	NA	NA	40	5
Cereals	0.07	0.06	NA	NA	6	5
Root vegetables	2	0.2	NA	NA	50	5
Potato tubers	0.3	0.06	NA	NA	25	5
Onion[d]	0.5	0.04	NA	NA	NA	NA
Soft fruit	2[f]	0.1[f]	NA	NA	80	5

Notes

a. FARMLAND considers salads, legumes and brassicas to be part of its green vegetable crop category (see Section 6.2).

b. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

c. Expressed in terms of Bq kg^{-1} fresh mass crop and Bq kg^{-1} dry mass soil.

d. FARMLAND does not have a crop category for onions, see text.

e. NA, no appropriate data available.

f. Value represents only tomato fruit, see Section 7.1.6

TABLE 15 Recommended default soil to crop transfer factors for technetium in the UK and within the FARMLAND model

Crop	reduced technetium[a]	non-reduced technetium[a]
Pasture	5	40
Any other crop[b]	0.5	5

Notes

a Expressed in terms of Bq kg^{-1} fresh mass crop and Bq kg^{-1} dry mass soil.

b Includes, brassicas, salad vegetables, legumes, cereals, root vegetables, potato tubers, onions and soft fruit.

8.2 Recommended transfer coefficients from feed to animal products for technetium

8.2.1 Technetium transfer from feed to milk

Johnson et al (1988) derived an F_m value for goat milk of 1.1 10^{-2} d l^{-1}. However, since $^{99}TcO_4^-$ was orally administered in this experiment the value does not represent a true feed to animal transfer coefficient. In addition, this transfer coefficient is around two orders of magnitude higher than the maximum transfer coefficient of 1.4 10^{-4} d l^{-1} estimated by Bondietti and Garten Jr (1984) who derived values using ^{95m}Tc sprayed onto pasture.

Insufficient data are available to recommend a value of F_m in either cow or goat's milk from any of the studies found by this review. For these reasons it is recommended that the current default value of 3 10^{-4} d l^{-1} for cow's milk in FARMLAND should be retained.

For situations where assessments are needed for goats milk, it is suggested that a value of 1 10^{-2} d l^{-1} is used, based on the information in Section 7.2.1.

8.2.2 Technetium transfer from feed to meat products

Transfer coefficients from feed to meat product for cattle and goats were only reported in Johnson et al (1988) but the study was carried out using ^{99m}Tc, which could lead to underestimates due to equilibrium not being reached. Nevertheless, the ratio between values for goats and cows was over 200, which is not consistent with the general view that it could be 10 times higher for goats than for cows. For these reasons no recommended values of F_f are given in this report and the current default values in FARMLAND should be retained. Values in the model for cow meat and liver are 1 10^{-2} and 4 10^{-2} d $kg^{-1,}$ respectively and for sheep meat and liver the values are 1 10^{-1} and 3 10^{-1} d kg^{-1}, respectively.

8.2.3 Technetium transfer from feed to poultry and chicken products

This review found that only two studies have published transfer coefficients from feed to poultry and chicken products (Ennis et al, 1988b and Thomas et al, 1984), both of which used ^{95m}Tc to estimate transfer for ^{99}Tc. As previously mentioned, using such short half life isotopes to predict behaviour for ^{99}Tc transfer can lead to an underestimation in transfer and for this reason no values are recommended for use by this review. No default values are currently used in FARMLAND. However, for situations where assessments are needed it is suggested that values of 3 d kg^{-1} and 1 d kg^{-1} for transfer to eggs and chicken meat, respectively are used based on the limited data presented in Table 12.

8.3 Technetium soil to plant transfer factors and implications for their use in the FARMLAND model

Currently, the FARMLAND model only uses a single value for soil-plant transfer and does not take into account the chemical form of technetium in the soil. The review of soil-plant transfer factors for technetium has shown that the chemical form makes a significant difference to its availability for root uptake. This section looks at the implications of this for the FARMLAND model and the choice of default values to be used for different applications of the model in the UK.

8.3.1 Routine releases to atmosphere

The default value for soil to crop transfer used in FARMLAND for technetium is 5 for all crop types on a fresh mass basis. Evidence gathered by this review suggests that two values should ideally be used dependent on the chemical form of technetium. Values of 0.5 for reduced and 5 for non-reduced chemical forms of technetium have been recommended for crops and 5 and 40 for pasture for the reduced and non-reduced forms of technetium.

The impact of choosing values of TF of 0.5, 5 and 40 has been explored and the results are shown in Appendix D. For routine use of the FARMLAND model it is recommended that a single default TF value of 5 continues to be used for both pasture and edible crops within FARMLAND. Appendix D describes the basis for this decision. Not taking account of the higher transfer to pasture grass of the non-reduced form of technetium in the first 120 days does not have any significant impact on the predicted activity concentrations in animal products for continuous deposition as in routine releases to atmosphere. However, the use of a TF value of 5 is a cautious assumption for edible crops and will lead to an overestimation of activity concentrations at times greater than a year. Given the sparsity of data, the low contribution of technetium when assessing doses from routine discharges to atmosphere and to be consistent with other regulatory models used in the UK, this approach is considered appropriate. If a more realistic study is needed for technetium, then a value of 0.5 could be used for technetium that has been in the soil for more than 3 – 4 months.

8.3.2 Accidental releases to atmosphere

For accidental releases, a conservative approach is recommended for the choice of TF values. It is very unlikely that an accidental release of technetium to atmosphere would occur or be a significant contributor to doses received. However, for all accidental releases it is more important to accurately estimate activity concentrations in food in the first year which is the period when activity concentrations vary more markedly with time. On this basis, it is recommended that for routine use of the model, single TF values of 40 and 5 are used for pasture and edible crops, respectively. This approach will overestimate the activity concentrations after the first year. Further details of the differences between the activity concentrations predicted by FARMLAND when the model is run using TF values for reduced and non-reduced forms of technetium are given in Appendix D.

8.3.3 Other applications where FARMLAND may be used

For specific applications of the FARMLAND model where it is known that technetium is newly deposited, the TF value for the non-reduced chemical form should be used, ie, 40 for pasture and 5 for edible crops. For applications where the technetium has aged in the environment or is of marine origin, eg, on sea-washed pastures, application of sewage sludge to land or from seaweed application to land, a TF value for the reduced from of technetium should be used, ie, a value of 5 for pasture and 0.5 for other crops. For very specific situations different TF values can be used at different times following the introduction of technetium into the terrestrial environment; however, this is not necessary in normal circumstances as discussed above and in more detail in Appendix D.

9 CONCLUSIONS

A critical review of published data on the transfer of technetium into terrestrial crops and animal products has been conducted and recommended transfer factor values for use in UK based radiological assessments are proposed.

This review has found that the uptake of technetium into crops depends on whether the technetium is in a chemically non-reduced form such as TcO_4^- or a reduced form such as TcO_2. The evidence found by this review suggests that when considering the types of agricultural soil generally encountered in the UK, the age of contamination is the most important factor in determining uptake of technetium into crops. Deciding on appropriate TF values for the chemical forms of technetium is complicated by the rapid changes in the observed values over time.

Based on a set of criteria designed to eliminate those studies in which transfer parameters were not applicable for use in radiological assessments in the UK and taking into consideration the differences in the behaviour of technetium in the environment, soil to crop TFs have been estimated. Since one aim of the review was to compare estimated TF values with those currently used in radiological assessment models such as FARMLAND, the crops were divided into seven broad categories.

Due to a lack of admissible data, values for soil to crop TFs for UK based radiological assessments have only been recommended for pasture (both chemical forms) and brassicas (the reduced chemical form). For the other crop categories, it is not possible to recommend TF values for each category due to the values only being based on one admissible study. Therefore, the use of a generic soil to crop TF value is recommended. For all crop types except pasture, generic values of 0.5 for the reduced chemical form and 5 for the non-reduced chemical form are recommended and for pasture, values of 5 and 40 are recommended for reduced and non-reduced forms, respectively. For assessments where more site specific data are required this review has compiled further data which may be more appropriate but no recommendations are made on their use.

For the transfer of technetium to animal products this review has identified the most important transfer pathway to be the ingestion of contaminated feed. Few publications on the transfer of technetium into animal products have been found and limited information could be located to determine the factors affecting its transfer. From the evidence summarised by this review, it is recommended that current default values for transfer to milk and meat products in the FARMLAND model should continue to be used. No values are currently given in the model for transfer from feed to poultry and chicken products. However, based on limited data, values have been suggested for use in assessments.

Values of transfer factors for the routine use of FARMLAND for different types of radiological assessments in the UK have been proposed. For routine releases, it is recommended that a single TF value of 5 continues to be used for both pasture and edible crops within FARMLAND. Not taking account of the higher transfer to pasture grass of the non-reduced form of technetium in the first 120 days does not have any significant impact on the predicted activity concentrations in animal products. However, the use of a TF value of 5 is a cautious assumption for edible crops and will lead to an overestimation of activity concentrations at times greater than a year. For accidental releases, it is recommended that single TF values of 40 and 5 are used for pasture and edible crops, respectively.

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APPENDIX A Inadmissible data used to aid discussion on recommended soil-to-plant transfer factors

This review has evaluated each source of data found for transfer of technetium from soil to crops and from crops to animals using criteria that were designed to eliminate those studies in which transfer parameters might not be applicable for use in food chain models used for radiological assessments in the UK (Section 6).

Data derived from studies that do not meet the criteria but have supported discussion in the main text of this review are given in Tables in the following pages. Details of these tables are given below.

Table A1	Inadmissible soil to plant CR values for technetium uptake into pasture
Table A2	Inadmissible soil to plant TFs for technetium for salad vegetables and brassicas where it was assumed to be in a non reduced form
Table A3	Inadmissible soil to plant TFs for technetium for salad vegetables and brassicas where it was assumed to be in a reduced form
Table A4	Inadmissible soil to plant CR values for technetium uptake into brassicas (Swiss Chard)
Table A5	Inadmissible soil to crop TFs for technetium uptake into root crops assumed to be in a reduced form
Table A6	Inadmissible soil to plant TFs for technetium uptake into potato tubers that have been used to aid discussion where it was assumed to be in a reduced form
Table A7	Inadmissible soil to plant TFs for technetium uptake into cereals where it was assumed to be in a reduced form
Table A8	Inadmissible soil to plant TFs for technetium uptake into cereals where it was assumed to be in a non-reduced form
Table A9	Inadmissible soil to plant TFs for technetium uptake into other crop types where it was assumed to be in a reduced form

TABLE A1 Inadmissible soil to plant CR values for technetium uptake into pasture

Author	No of samples	Study type	Form of technetium originally applied or discharged	CR_{dry}[a] pasture	Age of contamination (days)[b]
Hoffman et al (1982)	Not known	Field, near operational gaseous diffusion plant, mixed grass, silty loam soil	Likely to be discharged as $^{99}TcO_4^-$	7 (3 – 18)[c]	Potential continuous deposition from plant
	Not known	Field, near operational gaseous diffusion plant, mixed grass, silty loam soil	Likely to be discharged as $^{99}TcO_4^-$	16 (9 – 29)	Potential continuous deposition from plant
	Not known	Field, near operational gaseous diffusion plant, mixed grass, henry silt loam soil	Likely to be discharged as $^{99}TcO_4^-$	7 (5 – 11)	Potential continuous deposition from plant
Hoffman Jr (1982)	64	Field, Tennessee near operational gaseous diffusion plant	Likely to be discharged as $^{99}TcO_4^-$	7 (3 –16)	Potential continuous deposition from plant
	64	Field, Paduch, Kentucky near operational gaseous diffusion plant	Likely to be discharged as $^{99}TcO_4^-$	16 (5 – 44)	Potential continuous deposition from plant
	64	Field, Portsmouth, Ohio, near operational gaseous diffusion plant	Likely to be discharged as $^{99}TcO_4^-$	7 (1 – 26)	Potential continuous deposition from plant
Mousny and Myttenaere (1982)	3	Pot, grass, 10-15 kg clay soil	$^{99}TcO_4$,surface contamination	200	51
				200	81
				130	111
	3	Pot, grass, 10-15 kg loam soil	$^{99}TcO_4^-$, surface contamination	360	51
				270	81
				130	111
	3	Pot, grass, 10-15 kg peat soil	$^{99}TcO_4^-$,surface contamination	150	51
				80	81
				30	111
	3	Pot, clover and grass, 10-15 kg clay soil	$^{99}TcO_4^-$, surface contamination	200	51
				190	81
				100	111
	3	Pot, clover and grass, 10-15 kg loam soil	$^{99}TcO_4^-$, surface contamination	340	51
				230	81
				100	111
	3	Pot, clover and grass, 10-15 kg peat soil	$^{99}TcO_4^-$, surface contamination	150	51
				50	81
				120	111

TABLE A1 (cont) Inadmissible soil to plant CR values for technetium uptake into pasture

Author	No of samples	Study type	Form of technetium originally applied or discharged	CR_{dry}[a] pasture	Age of contamination (days)[b]
Garten et al (1984)	5	Field plot, grass, 10-15 kg silt loam	TcO_4^- surface contaminated	40	28
				3	63
				2	91
				1	140
				0.6	189
Vandecasteele et al (1989)	60	Lysimeter, grass, soil with 6.9% organic and 33% clay content	First surface contamination with 18.5 kBq $^{99}TcO_4^-$	400	30
				100	63
				100	100
			Second surface contamination with ^{99}18.5 kBq TcO_4^-	500	35
				340	76
				30	140
				10	304
				40	313
				40	389
				20	448
				5	698
				6	789
				3	958
				3	1098
				2	1198

Notes

a Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b Elapsed time from initial contamination to crop harvest, where applicable.

c Data ranges given in brackets, along with the geometric mean from the study.

TABLE A2 Inadmissible soil to plant TFs for technetium for salad vegetables and brassicas where it was assumed to be in a non reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} Brassicas[a]	Age of contamination (days)[b]
Sheppard et al (1983)	60	Lysimeter, mineral sand	Homogeneous contamination, 0.5 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	2100 (Swiss Chard)[c]	110
			Homogeneous contamination, 5.0 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	2000 (Swiss Chard)	110
			Homogeneous contamination, 50 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	3000 (Swiss Chard)	110
			Homogeneous contamination, 500 10^{-4} ug g^{-1}, $^{99}TcO_4^-$	2600 (Swiss Chard)	110

Notes

a Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b Elapsed time from initial contamination to crop harvest, where applicable. The values are much higher than would be expected if the technetium was reduced. In this case the technetium may not be in a reduced form because of the low organic matter content in mineral sand (see main text).

c Crop types given in brackets.

TABLE A3 Inadmissible soil to plant TFs for technetium for salad vegetables and brassicas where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} brassicas[a]	Age of contamination (days)[b]
Yanagisava and Muramatsu (1993)	2	Pot, 3.0 kg organic soil, growth chamber	Homogeneous contamination, $^{95m}TcO_4^-$	0.9(0.3-5)[c] (cabbage)[d]	Approx 85
				0.5(0.1-4) (Chinese cabbage)	Approx 85

Notes

Expressed in terms of Bq kg^{-1} dry mass crop and soil.

Elapsed time from initial contamination to crop harvest, where applicable. Times greater than 90 days were used to represent technetium assumed to be in a reduced form.

Data ranges given in brackets, along with the geometric mean from the study.

Crop types given in brackets.

TABLE A4 Inadmissible soil to plant CR values for technetium uptake into brassicas (Swiss Chard)

Author	No of samples	Study type	Form of technetium originally applied	CR_{dry} Brassicas[a]	Age of contamination (days)[b]
Garten et al (1986)	80	Pot experiment in field, silty clay loam	Likely to be aged ^{99}Tc	4(3 - 5)[c]	Greater than 10 years
			Homogeneously contaminated , $^{99}TcO_4^-$	14(12 – 17)	Freshly contaminated

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable.

c. Data ranges given in brackets, along with the geometric mean from the study.

TABLE A5 Inadmissible soil to crop TFs for technetium uptake into root crops assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} root crops[a]	Age of contamination (days)[b]
Yanagisava and Muramatsu (1993)	2	Pot, 3.0 kg organic soil, growth chamber	Homogeneous contamination, $^{95m}TcO_4^-$	2 (1.7-2.1)[c] (peeled carrot)[d] (2-4) (carrot peel)	90

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than about 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Crop types given in brackets.

TABLE A6 Inadmissible soil to plant TFs for technetium uptake into potato tubers that have been used to aid discussion where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} potato tubers[a]	Age of contamination (days)[b]
Yanagisava and Muramatsu (1993)	4	Pot, 3.0 kg organic soil, growth chamber	Homogeneous contamination, $^{95m}TcO_4^-$	0.08(0.07-0.09)[c] (Sweet potato, whole)[d] 0.37(0.37-0.38) (Sweet potato, peel)	90

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than about 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Crop types given in brackets.

TABLE A7 Inadmissible soil to plant TFs for technetium uptake into cereals where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} cereals[a]	Age of contam (days)[b]
Yanagisawa et al (1995a)	10	Pot, 2.8 kg soil under flooded conditions, growth chamber	Homogeneous contamination, $^{95m}TcO_4^-$	<0.0002 (rice grain)[c]	Not given
Yanagisawa and Muramatsu (1995b)	3-6	Pot, 2.8 kg Andosol soil, under flooded conditions, growth chamber,	Homogeneous contamination $^{95m}TcO_4^-$, 740kBq kg^{-1-}	0.00005 (rice grain)	Not given
	3-6	Pot, 2.8 kg Gray lowland soil, under flooded conditions, growth chamber,	Homogeneous contamination $^{95m}TcO_4^-$, 740kBq kg^{-1}	0.0006 (rice grain)	Not given
Yanagisawa et al (1992)	3	Pot, 2.8 kg soil under flooded conditions, growth chamber	Homogeneous contamination, $^{95m}TcO_4^-$	<0.005 (rice grain)	Not given

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Under flooded conditions it is assumed that the technetium will be in a reduced form. See Section 7.1.5.

c. Crop types given in brackets.

TABLE A8 Inadmissible soil to plant TFs for technetium uptake into cereals where it was assumed to be in a non-reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} cereals[a]	Age of contam (days)[b]
Yanagisawa et al (1995a)	10	Pot, 2.8 kg soil under non- flooded conditions, growth chamber	Homogeneous contamination, $^{95m}TcO_4^-$	0.021(0.016 - 0.026)[c] (rice grain)[d]	Not given
Yanagisawa et al (1992)	3	Pot, 2.8 kg soil, non-flooded conditions, growth chamber	Homogeneous contamination $^{95m}TcO_4^-$	0.027 (wheat grain)	Not given
Echevarria et al (1995)	Not known	Pot, 1.5 kg silty loam soil, growth chamber	Homogeneous contamination $^{99}TcO_4^-$, 2.5kBq kg^{-1}	5(3 – 7) (wheat grain)	Not given
			Homogeneous contamination $^{99}TcO_4^-$, 25kBq kg^{-1}	0.3(0.2 – 0.4) (wheat grain)	Not given

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Under non-flooded conditions it is assumed that the technetium will be in a non-reduced form. See Section 7.1.5.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Crop types given in brackets.

TABLE A9 Inadmissible soil to plant TFs for technetium uptake into other crop types where it was assumed to be in a reduced form

Author	No of samples	Study type	Form of technetium originally applied	TF_{dry} onions[a]	Age of contam. (days)[b]	TF_{dry} soft fruit[a]	Age of contam (days)[b]
Yanagisava and Muramatsu (1993)	4	Pot, 3.0 kg organic soil, growth chamber	Homogeneous. Contamination, $^{95m}TcO_4^-$	0.3(0.1-2.1)[c]	115	0.3(0.2-0.4) (tomato)[d]	120

Notes

a. Expressed in terms of Bq kg^{-1} dry mass crop and soil.

b. Elapsed time from initial contamination to crop harvest, where applicable. Times greater than about 90 days were used to represent technetium assumed to be in a reduced form.

c. Data ranges given in brackets, along with the geometric mean from the study.

d. Crop types given in brackets.

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APPENDIX B Inadmissible data used to aid discussion: Concentration ratios for the uptake of Technetium-99 in terrestrial food following seaweed application to agricultural land

This Appendix provides extra information that can be used to support the choice of recommended transfer factor values for use in the FARMLAND Model. The results presented are a summary of data extracted from a study of the transfer of radioactivity from seaweed to terrestrial foods (Brown et al 2009).

The presence of technetium-99 (^{99}Tc) in soil originates from the application of seaweed used as fertiliser or a soil conditioner. It is important to note that seaweed has been applied to the soils for several months prior to the crops being grown and, in many cases, will have been regularly applied to the land over many years. Therefore, it is reasonable to assume that the technetium will be in a reduced form. This study provides information for a wide range of crops that have been planted under normal agricultural conditions and not under controlled experimental conditions. It is therefore possible that the activity concentration of ^{99}Tc in the crop might include contributions from different uptake pathways such as resuspension or direct deposition from sea spray. Since this cannot be ruled out, the data presented can only be regarded as soil to crop concentration ratios. These data were therefore not considered admissible in this review but have been used to support discussion.

B1 BACKGROUND

The main source of ^{99}Tc in the UK comes from the authorised liquid discharges into the environment from the nuclear industry. Technetium-99 is concentrated in seaweed as indicated by routine surveillance monitoring and in some parts of Scotland and England ^{99}Tc has been incorporated into the soil by the historic use of seaweed as a fertilizer and soil conditioner.

The data that are presented in this appendix do not meet the criteria to be included in the main body of the review. The soil sampling protocol does not meet the standard soil depths set by the IUR, 10 cm for grass and 20 cm for all other crops. These soil and crop samples were mainly taken to identify the extent to which ^{99}Tc is incorporated into the soil and crops due to the use of seaweed as a fertiliser. However, calculated concentration ratios using measurements of ^{99}Tc in crops and soils from the same location can provide very useful information to support the selection of appropriate soil to plant transfer factors. The seaweed study looked at a wide variety of fruit, cereals and vegetables (Brown et al, 2009). A list of all the crops for which measurements were made are given in Table B1. The variety of crops studied can be used to investigate differences in CR values between the different crop categories considered in the review (see Section 6).

B2 RESULTS AND DISCUSSION

It is important to be aware that the results shown in this appendix are concentration ratios of ^{99}Tc from soil to crop and the source of contamination of the radionuclide comes from seaweed. The main part of this review has derived transfer factors where other potential contributions to the uptake of ^{99}Tc due to different pathways have been removed. Also the method of soil contamination in most of the literature published and reviewed has ensured a homogeneous contamination of soil with a chemical solution that contains $^{99}TcO_4^-$.

The main review suggests that over a period of 90-120 days it is expected that the TcO_4^- has been reduced to TcO_2 and, as a consequence, the root uptake will decrease. In this study there is no certainty in which oxidation state the ^{99}Tc is likely to be present although it is reasonable to assume that it is TcO_2.

The crops have been classified into different groups according to Section 6.3 of the main report: salad vegetable and brassicas, root vegetables, tubers, cereal crops, fruit and other crops.

The calculated concentration ratios (CR) values for the wide variety of crops studied across 15 locations are listed in Table B1. Situations where the ^{99}Tc concentration in soil was below the limit of detection have been discounted. However, where the activity concentration in the soil was measurable but the activity concentration of the crop was below the limit of detection, the data can be used to indicate a maximum CR for the crop. Maximum CR values are indicated in Table B1.

TABLE B1 Technetium-99 concentration ratios (activity concentration fresh crop / activity concentration in dry soil)

Location and sample description	Group	Concentration Ratio
Location 1		
Broad beans	Other crops	$1\ 10^{-2}$
Cabbage	Salad vegetable + brassicas	$4\ 10^{-2}$
Carrots	Root crops	$3\ 10^{-2}$
Potatoes (early variety)	Tubers	$4\ 10^{-1}$
Potatoes	Tubers	$1\ 10^{-2}$
Location 2		
Potatoes,	Tubers	$8\ 10^{-2}$ [a]
Location 3		
Artichokes	Root crops	$4\ 10^{-3}$
Kale from plot 1	Salad vegetable + brassicas	$1\ 10^{-1}$
Kale from plot 2	Salad vegetable + brassicas	$9\ 10^{-2}$
Location 4		
Potatoes	Tubers	$7\ 10^{-2}$
Potatoes (Desiree)	Tubers	$3\ 10^{-2}$
Potatoes (Sharps Express)	Tubers	$5\ 10^{-2}$
Location 6		
Potatoes	Tubers	$1\ 10^{-2}$
Location 8		
Potatoes ,	Tubers	$7\ 10^{-2}$ [a]
Location 9		
Cabbage	Salad vegetable + brassicas	$4\ 10^{-2}$
Carrots	Root crops	$4\ 10^{-2}$
Lettuce plot	Salad vegetable + brassicas	$1\ 10^{-2}$
Mixed legumes (broad beans, peas, beans)	Other crops	$2\ 10^{-2}$
Strawberries	Fruit	$2\ 10^{-2}$ [a]
Location 10		
Blackcurrants	Fruit	$5\ 10^{-2}$ [a]
Broad beans	Other crops	$8\ 10^{-2}$ [a]
Kale	Salad vegetable + brassicas	$9\ 10^{-2}$
Red cabbage	Salad vegetable + brassicas	$9\ 10^{-3}$
Location 11		
Broccoli	Salad vegetable + brassicas	$3\ 10^{-1}$
Cabbages	Salad vegetable + brassicas	1
Lettuce (mixed varieties)	Salad vegetable + brassicas	$4\ 10^{-2}$
Swiss chard (red)	Salad vegetable + brassicas	1
Location 12		
Lettuce	Salad vegetable + brassicas	$6\ 10^{-1}$
Potatoes,	Tubers	$1\ 10^{-2}$
Location 13		
Cabbage and cauliflower	Salad vegetable + brassicas	$3\ 10^{-2}$
Peas and sugar snap peas	Other crops	$2\ 10^{-3}$ [a]

TABLE B1 Technetium-99 concentration ratios (activity concentration fresh crop / activity concentration in dry soil)

Location and sample description	Group	Concentration Ratio
Potatoes (early variety)	Tubers	$2\ 10^{-2}$
Location 14		
Celery	Salad vegetable + brassicas	$8\ 10^{-3}$ [a]
Currants and raspberries	Fruit	$3\ 10^{-3}$ [a]
Location 15		
Apples	Fruit	$2\ 10^{-3}$ [a]
Broad beans	Other crops	$1\ 10^{-2}$ [a]
Kale	Salad vegetable + brassicas	$7\ 10^{-3}$
Mixed berries	Fruit	$6\ 10^{-3}$ [a]
Location 16		
Blueberries	Fruit	$2\ 10^{-2}$ [a]
Raspberries	Fruit	$1\ 10^{-2}$ [a]
White currants	Fruit	$8\ 10^{-3}$ [a]
Location 17		
Calibrese / kale	Salad vegetable + brassicas	1
Potatoes	Tubers	$2\ 10^{-3}$ [a]
Location 19		
Chard	Salad vegetable + brassicas	$6\ 10^{-2}$
Potatoes	Tubers	$7\ 10^{-3}$
Tomatoes	Fruit	$5\ 10^{-4}$

a. Maximum value for the concentration ratio estimated as ^{99}Tc concentration in fresh crop is below of limit of detection.

The CR values for all crops were plotted within their corresponding category. Figure B1 shows a graph of the CR values for each crop group. The CR values for all crops are within the range of $5\ 10^{-4}$ to 1 as shown in Table B2. Figure B1 and Table B2 show that there is a wide distribution in the CR values within the same crop group, approximately 2 orders of magnitude; most of the values are less than $1\ 10^{-1}$ with a few values above this and 3 CR values which are approximately 1. In general, it is expected that the technetium present in the soil will be in a reduced form because of its marine origin; the low CR values support this and they broadly agree with the experimental data for technetium in a reduced form considered in the review. On this basis they can be used to show possible differences between crop categories.

Salad vegetables and brassicas is the group where there is the highest CR values ranging from $1\ 10^{-1}$ to 1. This crop group contains 16 measured CR values and these have been considered in more detail to determine if there are differences between individual crops in this category. The CR values are shown in more detail in Figure B2. It can be seen in Figure B2 that almost 40% of the CR values are higher than $1\ 10^{-1}$, which correspond to the maximum CR values for the rest of the crop groups. The three highest CR values are for kale, cabbage and Swiss chard; however CR values for kale and cabbage can be found within the lower part of the range. For Swiss chard there is only one measurement and no conclusion can be therefore be made on a range of values for this crop. For lettuces, the 3 measurements cover the whole CR value range. It is therefore reasonable to suggest that there is no significant difference in root uptake between individual species of salad vegetables and brassicas. However, it can be concluded that the leafy green vegetables may have higher root uptake than other vegetables.

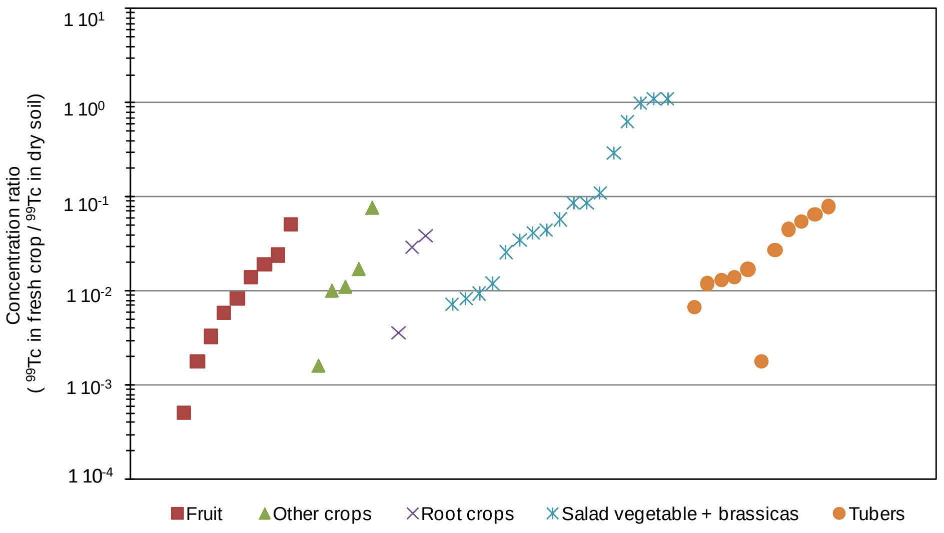

Figure B1 Concentration ratios of ^{99}Tc for different crop categories

Table B2 Concentration ratio range values for different crop groups

Crop Group	Number of values	Range Min - Max
Fruit	9	$5\ 10^{-4}$ – $5\ 10^{-2}$
	8	($2\ 10^{-3}$ - $5\ 10^{-2}$)[a]
Other crops[b]	5	$2\ 10^{-3}$ – $8\ 10^{-2}$
Root crops	3	$4\ 10^{-3}$ – $4\ 10^{-2}$
Salad vegetables and brassicas	16	$7\ 10^{-3}$ - 1
Tubers	11	$2\ 10^{-3}$ - $8\ 10^{-2}$

a. Excluding tomato.

b. All crops in this category are legumes.

The concentration factors for the root, tuber and “others” crops are not significantly different.

Fruit is the crop group with the lowest CR values with a range from $5\ 10^{-4}$ to $5\ 10^{-2}$. Most of the measurements in the fruit category are for berries, which cover the majority of the range of fruit CR values. There are two other types of fruit in this category, apples and tomatoes with CR values of $2\ 10^{-3}$ and $5\ 10^{-4}$, respectively, which are at the lower end of the range of fruit CR values. The CR value for tomatoes is the lowest value found in this category; although tomatoes are part of the fruit family, they would not be considered routinely in radiological assessments and have not been considered in the development of the fruit model within FARMLAND. The literature review (Section 7.1.6) shows that there is only one transfer factor for tomatoes. If the tomato CR value is excluded, the geometric mean of the CR values for fruit (apples and berries) is 1 10^{-1}. Given the similarity in the ranges of CR values in this study with the admissible TF values discussed in Sections 7 and 8, it is reasonable to propose that a TF value of 1 10^{-1} would be appropriate to use for soft fruit if a specific assessment was required.

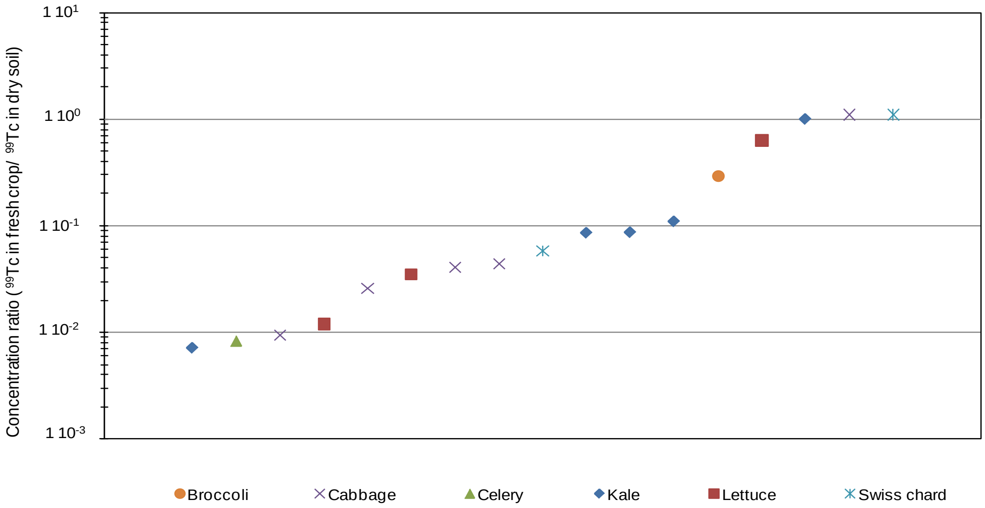

Figure B2 Concentration ratio of ^{99}Tc in salad vegetables and brassicas

B3 REFERENCES

Brown J, Hammond D, Wilding D, Wilkins BT and Gow C (2009). Transfer of Radioactivity from Seaweed to Terrestrial Foods and Potential Radiation Exposures to Members of the Public. Chilton, HPA-RPD-059.

APPENDIX C Dry matter content of crops

Where necessary, published values presented in this review were converted to a dry mass basis either by using specific data given in the publication itself or by using existing data for specific crop types as given in Table C1.

In this review, FARMLAND default values have been given in terms of fresh and dry mass of the crop. The percentage dry matter content used to convert the broad FARMLAND crop categories to dry mass were averaged from available data given in Table C1. These data are presented in Table C2.

TABLE C1 Percentage dry matter content for various crop types

	Dry matter content (%)	
Crop	Green et al (1995)[a]	Others
Salads		
Endive	–	6.3[b]
Lettuce	2.7	4.8[b]
Root vegetables		
Carrot	12	10.2[b]
Beetroot	11.2	12.9[b]
Radish	3.9	6.7[b]
Swedes	11.7	9.6[b]
Parsnip	–	17.5[b]
Turnips	–	6.7[b]
Tubers		
Potatoes	21.3	21.2[b]
Brassicas		
Brussel sprouts	17.6	15.7[b]
Cauliflower	8.5	8.9[b]
Cabbage	7.1	10.9[b]
Broccoli	12.3	–
Legumes		
Broad beans	31.3	–
French beans	8	–
Runner beans		8.4[b]
Peas	39.1	21.5[b]
Mange Tout	12.7	
Cereals		

TABLE C1 Percentage dry matter content for various crop types

	Dry matter content (%)	
Crop	Green et al (1995)[a]	Others
Cereal grains	–	90[b]
Barley	90	89[d]
Oats	89.3	–
Onions		
Onion	9.9	7.2[b]
Spring onion	6	13.2[b]
Leek	9.7	10[b]
Soft fruit		
Tomatoes	4.8	–
Strawberries	8	–
Cucumber	4	6[d]
Hard fruit		
Apples (variety Fiesta)	10.4	–
Other crops		
Celery	–	6[d]
Marrow	4.2	–
Pasture		26[e]

Notes

a. Data reproduced from Green et al (1995).
b. Data reproduced from McCance and Widdowson (1960).
c. Data reproduced from Orr, (1982).
d. Date reproduced from Frissel and Heisterkamp (1989).
e. Unpublished data from studies conducted by NRPB (1999).

TABLE C2 Percentage dry matter content for crops used to convert FARMLAND values

FARMLAND Crop category	Dry matter content (%)[a]
Pasture	26
Green vegetables[b]	13
Root vegetables	10
Potatoes	21
Cereals (grain)	90
Soft fruit	6

Notes

a. Average derived from available crop data given in Table C1.
b. The green vegetable category in FARMLAND includes salads, legumes and brassicas.

C1 REFERENCES

Frissel MJ and Heisterkamp S (1989). Geometric mean TF-values calculated with multilinear regression analysis, 6th report of IUR working group on soil to plant transfer factors. RIVM, Bilthoven, Netherlands.

Green N, Wilkins BT and Hammond DJ and Davidson MF (1995). Transfer of radionuclides to vegetables and other crops in an area of land reclaimed from the sea: a compilation of data. Chilton, NRPB-M538.

McCance RA and Widdowson EM (1960). The composition of foods. Medical Research Council special report series No 297. London, HMSO.

Orr RM (1982). Animal feedstuffs. IN Primrose McConnell's, The Agricultural Notebook. (Ed: RJ Halley), 17th edition. Butterworths, London, 341-350.

APPENDIX D Impact of choice of transfer factors on activity concentrations in different foods estimated by the FARMLAND model

The main report has recommended two values of transfer factors for crops and pasture, the transfer factor value used for the calculation of ^{99}Tc in food being dependent on the chemical form of the technetium. Based on the recommendations of the literature review, it can be assumed that the ^{99}Tc released is in a non-reduced form and would mostly become chemically reduced in the soil after a period of approximately 90-120 days. To represent this situation within the FARMLAND model (Brown and Simmonds 1995), the use of two soil to crop TF values would be most appropriate; these are values of 5 for the first 120 days followed by a value of 0.5 for crops and a value of 40 for the first 120 days followed by a value of 5 for pasture. However, for ease of use in radiological assessments it is desirable to use a single TF value.

This appendix justifies the continued use of single TF values within radiological assessments following the release of ^{99}Tc to the atmosphere as a routine or accidental release. Green vegetables, cereals, root crops and cattle grazing pasture (animal products) are used to look at the impact of the choice of TF value on predicted radionuclide concentrations in food using the FARMLAND model for ^{99}Tc released to the atmosphere as a routine and accidental release. The results have been used to support the choice of the most appropriate TF value for a particular scenario.

D1 ROUTINE RELEASE TO ATMOSPHERE

The time integrated concentrations of ^{99}Tc in crops and in milk and meat from cattle grazing pasture assuming a continuous deposition of 1 Bq m^{-2} s^{-1} were calculated using three different sets of soil to crop transfer factors. The TF values chosen were:

a 5, the value recommended for technetium in the non-reduced form. This is the current TF value used in FARMLAND;

b 0.5, the value recommended for technetium in the reduced form

c a combination of 5 for the first 120 days followed by 0.5 for crops (the recommendation for the literature review). The TF combination is represented in the remainder of this section as (TF=5,0.5);

d a combination of 40 for the first 120 days followed by 5 for pasture (the recommendation for the literature review). The TF combination is represented in the remainder of this section as (TF=40,5).

The calculated time integrated activity concentrations in green vegetables are shown in Figure D1 and Table D1. Compared with using the optimum combination of TF = 5,0.5, the results show that the choice of a single TF value of 5 gives good agreement between the estimated time integrated activity concentrations over the first year but may lead to an overestimation of the concentration after the first year which increases with time up to a factor of about 6 after 50 years. If a single TF value of 0.5 is used, then in the first few years the time integrated activity concentrations may be underestimated by up to a factor of about 2 but at longer times there is no difference compared to using the combination of TF values (TF=5,0.5).

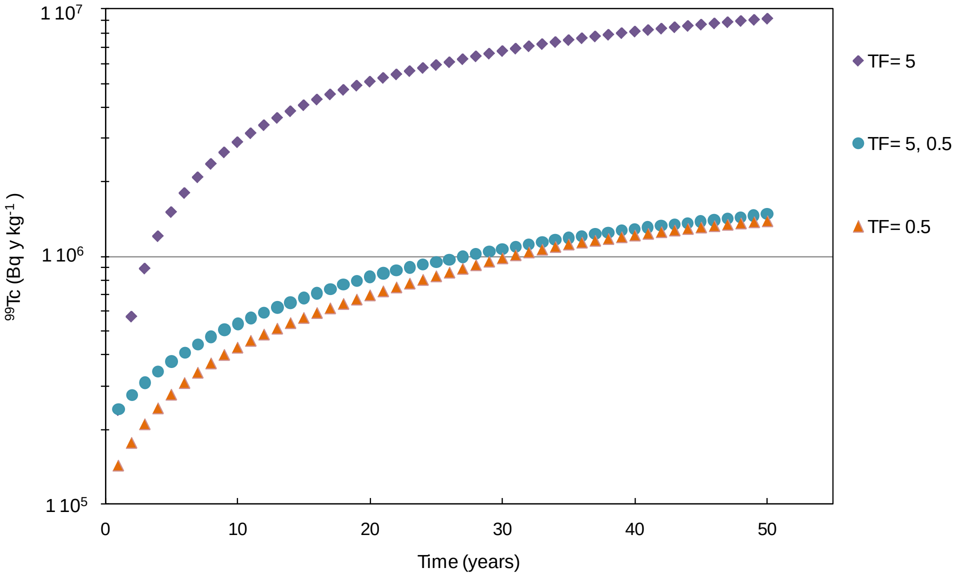

Figure D1 Time integrated activity concentration for green vegetables using different soil to crop TF values of 0.5 and 5 compared to the optimum combination of TF= 5,0.5

Table D1 Time integrated activity concentration in different crops for routine release

	Green vegetables (Bq y kg^{-1})			Cereal (Bq y kg^{-1})			Root vegetables (Bq y kg^{-1})		
Time (year)	TF = 0.5	TF =5,0.5	TF = 5	TF = 0.5	TF =5,0.5	TF = 5	TF = 0.5	TF =5,0.5	TF = 5
1	1.4 10^5	2.4 10^5	2.4 10^5	5.1 10^5	6.1 10^5	6.1 10^5	1.3 10^5	2.2 10^5	2.2 10^5
5	2.8 10^5	3.8 10^5	1.5 10^6	6.5 10^5	7.5 10^5	2.0 10^6	2.7 10^5	3.6 10^5	1.5 10^6
10	4.3 10^5	5.3 10^5	2.9 10^6	8.1 10^5	9.1 10^5	3.6 10^6	4.3 10^5	5.2 10^5	2.7 10^6
20	7.0 10^5	8.3 10^5	5.1 10^6	1.1 10^6	1.2 10^6	6.5 10^6	7.3 10^5	8.1 10^5	4.7 10^6
40	1.2 10^6	1.3 10^6	8.1 10^6	1.7 10^6	1.8 10^6	1.2 10^7	1.2 10^6	1.3 10^6	6.9 10^6
50	1.4 10^6	1.5 10^6	9.2 10^6	1.9 10^6	2.0 10^6	1.4 10^7	1.5 10^6	1.5 10^6	7.5 10^6

Table D1, Figures D2 and D3 also give results data for time integrated activity concentrations for cereals and root crops, respectively. The same differences are seen for the other crops when the use of single TF values of 5 and 0.5 are compared to the optimum combination of TF values (TF=5,0.5).

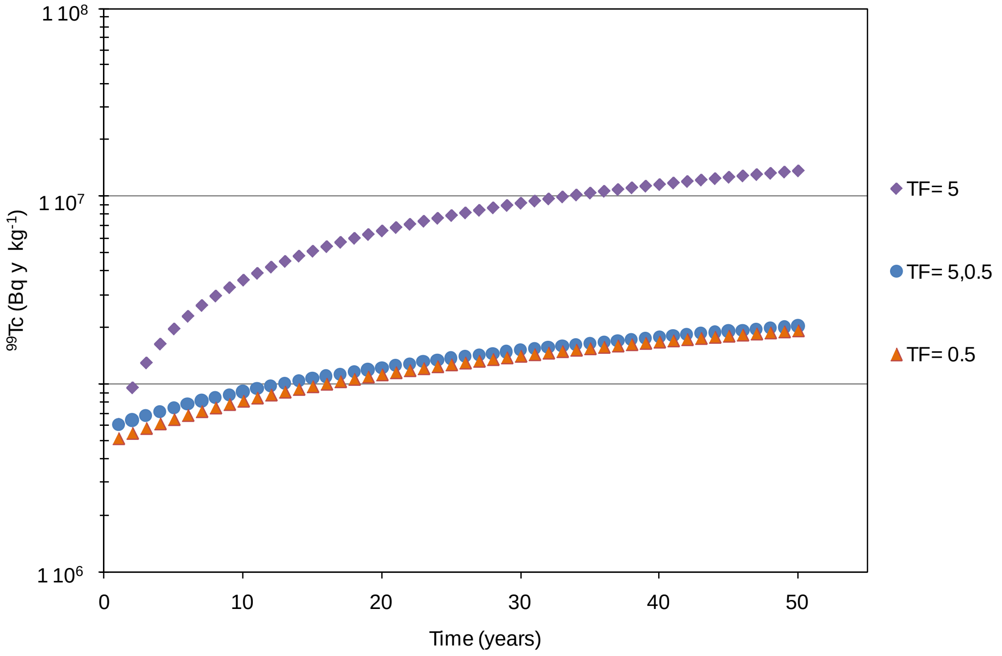

Figure D2 Time integrated activity concentration for cereals using different soil to crop TF values of 0.5 and 5 compared to the optimum combination of TF= 5,0.5

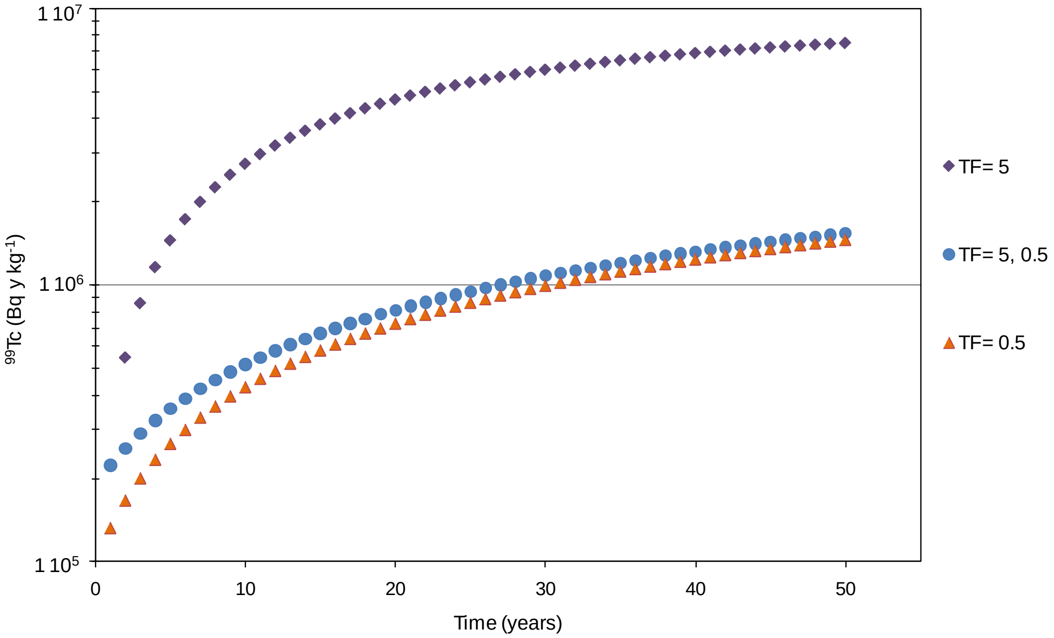

Figure D3 Time integrated activity concentrations for root vegetables using different soil to crop TF values of 0.5 and 5 compared to the optimum combination of TF= 5,0.5

The calculated time integrated activity concentrations in milk and meat from cattle grazing pasture are shown in Table D2. Compared with using the optimum combination of TF = 40,5, the results show that the choice of a single TF value of 5, ie, the value for the reduced form of technetium gives very similar predictions of time integrated activity concentrations in milk and meat.

Table D2 Time integrated activity concentration for animal products using different soil to crop transfer factors for pasture.

Time (years)	Milk (Bq y l^{-1})			Meat (Bq y kg^{-1})		
	TF= 5	TF= 40,5	TF= 40	TF= 5	TF= 40,5	TF= 40
1	1.7 10^5	1.7 10^5	6.3 10^5	5.5 10^6	5.5 10^6	2.1 10^7
5	7.4 10^5	7.5 10^5	2.9 10^6	2.5 10^7	2.5 10^7	9.7 10^7
10	1.4 10^6	1.4 10^6	5.3 10^6	4.6 10^7	4.7 10^7	1.8 10^8
20	2.6 10^6	2.6 10^6	8.8 10^6	8.6 10^7	8.8 10^7	2.9 10^8
40	4.6 10^6	4.7 10^6	1.3 10^7	1.5 10^8	1.6 10^8	4.2 10^8
50	5.5 10^6	5.6 10^6	1.4 10^7	1.8 10^8	1.9 10^8	4.5 10^8

D1.1 Recommended TF values for use in the FARMLAND model for routine release assessments

It is recommended that the use of a single TF value of 5 is appropriate for both pasture and edible crops within radiological assessments using FARMLAND. As described above, not taking account of the higher transfer factor of the non-reduced form of technetium in the first 120 days does not have any impact on the predicted activity concentrations in animal products. However the use of a transfer factor of 5 is a cautious assumption for crops and will lead to an overestimation of activity concentrations at times greater than one year.

This recommendation is based on the assumption that ^{99}Tc will not be the main contributor in a radioactive release and therefore the dose exposure to man will be dominated by other radionuclides. If this assumption is not valid, then the combination of TF values for the non-reduced and reduced forms of technetium (TF = 5,0.5 for crops and TF= 40,5 for pasture) should be used.

D2 ACCIDENTAL RELEASES TO ATMOSPHERE

For accidental releases it is important to be aware of the impact of the choice of TF values on the activity concentrations as a function of time because if an incident occurs a ban on the sale of crops may need to be implemented.

The activity concentrations as a function of time and time integrated activity concentration per year have been calculated using FARMLAND for a single total deposition per unit area of 1 Bq m^{-2} (assumed to be instantaneous). The predicted activity concentrations as a function of time are presented in Table D3 for green vegetables, cereals and root vegetables.

Compared to the optimum combination of TF values (TF=5,0.5), it can be seen that the activity concentration in green vegetables after the first year is the same after 1 year and the activity concentrations are subsequently overestimated by a factor of about 4 at longer times. In contrast, if a single TF value of 0.5 is used, ie, for technetium in a reduced form, the activity concentrations are approximately 10 times lower after one year and after a few years the predicted activity concentrations are similar. For cereals and root vegetables, a similar trend is seen, although the differences between using the optimum combination of TF values and the use of a single value is less than a factor of 3.

Table D3 Activity concentrations for an accidental release scenario using different soil to crop TFs

	Green vegetables (Bq kg^{-1})			Cereal (Bq kg^{-1})			Root vegetables (Bq kg^{-1})		
Time (year)	TF = 0.5	TF =5,0.5	TF = 5	TF = 0.5	TF =5,0.5	TF = 5	TF = 0.5	TF =5,0.5	TF = 5
1	$1.1\ 10^{-3}$	$1.0\ 10^{-2}$	$1.0\ 10^{-2}$	$1.9\ 10^{-2}$	$2.9\ 10^{-2}$	$2.9\ 10^{-2}$	$5.4\ 10^{-3}$	1.410^{-2}	$1.5\ 10^{-2}$
10	$9.8\ 10^{-4}$	$9.6\ 10^{-4}$	$8.0\ 10^{-3}$	$1.0\ 10^{-3}$	$1.0\ 10^{-3}$	$9.8\ 10^{-3}$	$1.0\ 10^{-3}$	9.610^{-4}	$7.3\ 10^{-3}$
20	$9.0\ 10^{-4}$	$8.8\ 10^{-4}$	$6.0\ 10^{-3}$	$9.5\ 10^{-4}$	$9.5\ 10^{-4}$	$8.8\ 10^{-3}$	$9.0\ 10^{-4}$	8.710^{-4}	$4.9\ 10^{-3}$
50	$6.8\ 10^{-4}$	$6.7\ 10^{-4}$	$2.5\ 10^{-3}$	$7.6\ 10^{-4}$	$7.6\ 10^{-4}$	$6.2\ 10^{-3}$	$6.6\ 10^{-4}$	6.410^{-4}	$1.5\ 10^{-3}$

Additional details on how the activity concentrations vary over the first 3 years following deposition for these crops using the different soil-to-plant transfer factors are shown in Figures D4 and D5.

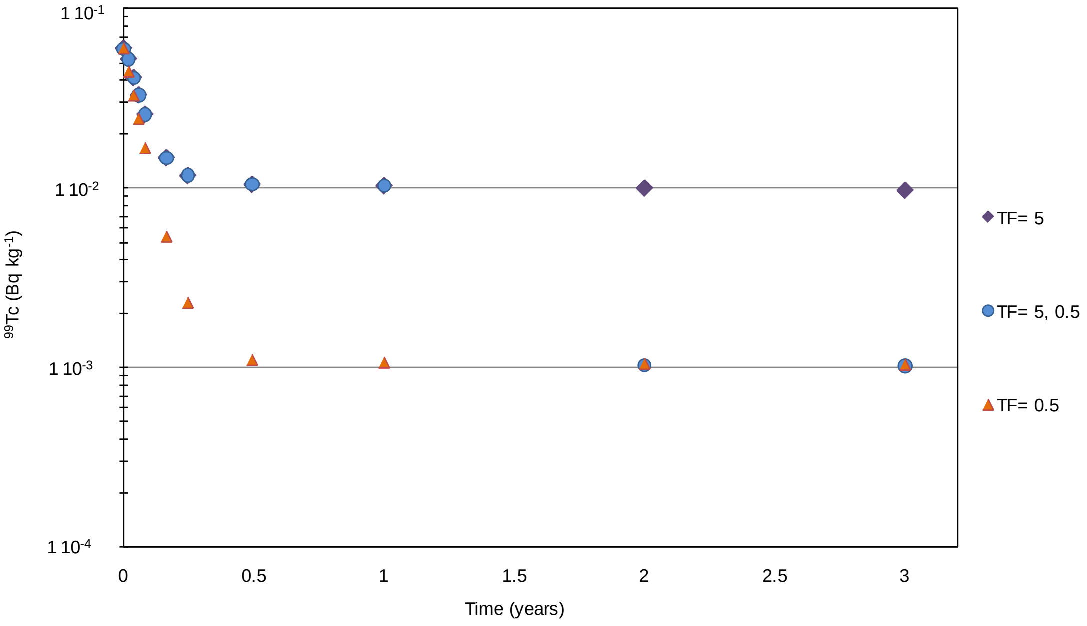

Figure D4 Activity concentrations for green vegetables for an accidental release scenario using different soil to crop TF values of 0.5 and 5 compared to the optimum combination of TF= 5,0.5

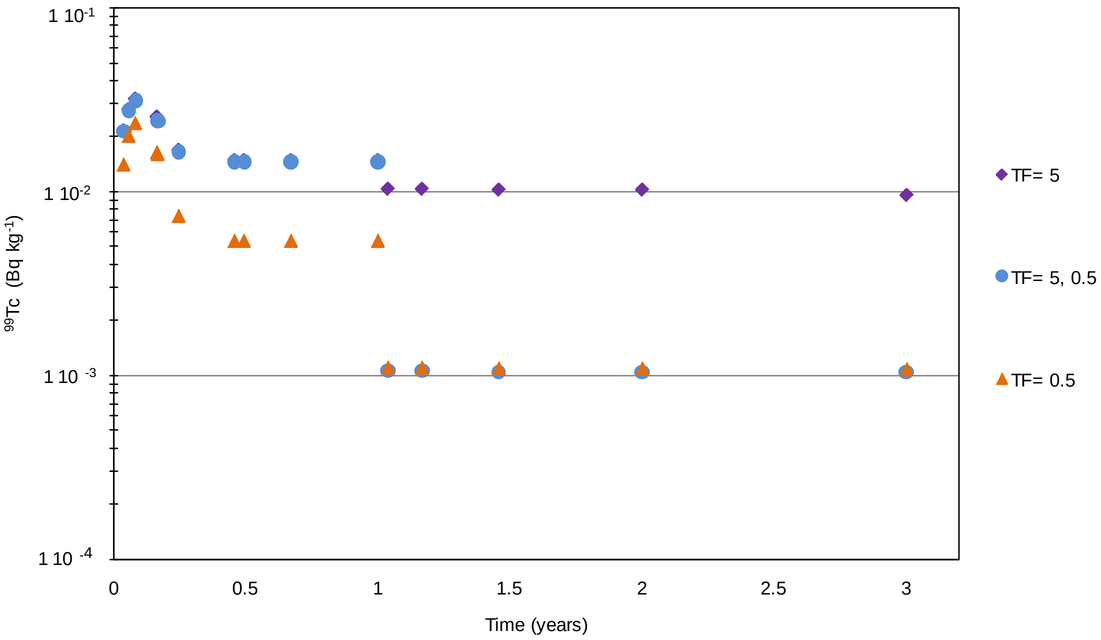

Figure D5 Activity concentrations for root crops for an accidental release scenario using different soil to crop TF values of 0.5 and 5 compared to the optimum combination of TF= 5,0.5

Although for accidental releases the activity concentrations during the first year are the most important for short term decisions, it is still necessary to estimate the impact on the choice of TF value on estimated radiation doses from consumption of contaminated food in the long term. Table D4 shows the predicted time integrated activity concentrations for green vegetables, cereals and root crops using different values of TF. Similar results are seen as those for the activity concentrations as a function of time. The use of a single TF value of 5 overestimates the time integrated activity concentrations over long times compared to the optimum choice of TF=5,0.5. However, if a single TF value of 0.5 is used instead of the optimum values (TF=5,0.5), the time integrated concentrations are underestimated over the first 10 years but there is no significant difference after 50 years, as shown in Figure D6.

Table D4 Time integrated activity concentrations for an accidental release scenario for different soil to crop TFs

	Green vegetables (Bq y kg^{-1})			Cereal (Bq y kg^{-1})			Root vegetables (Bq y kg^{-1})		
Time (year)	TF = 0.5	TF =5,0.5	TF = 5	TF = 0.5	TF =5,0.5	TF = 5	TF = 0.5	TF =5,0.5	TF = 5
1	$4.8\ 10^{-3}$	$1.4\ 10^{-2}$	$1.4\ 10^{-2}$	$1.4\ 10^{-2}$	$2.2\ 10^{-2}$	$2.2\ 10^{-2}$	$7.8\ 10^{-3}$	$1.6\ 10^{-2}$	$1.7\ 10^{-2}$
10	$1.4\ 10^{-2}$	$2.3\ 10^{-2}$	$9.6\ 10^{-2}$	$2.9\ 10^{-2}$	$3.8\ 10^{-2}$	$1.2\ 10^{-1}$	$1.8\ 10^{-2}$	$2.6\ 10^{-2}$	$1.0\ 10^{-1}$
20	$2.3\ 10^{-2}$	$3.2\ 10^{-2}$	$1.7\ 10^{-1}$	$3.8\ 10^{-2}$	$4.8\ 10^{-2}$	$2.1\ 10^{-1}$	$2.7\ 10^{-2}$	$3.5\ 10^{-2}$	$1.6\ 10^{-1}$
50	$4.7\ 10^{-2}$	$5.5\ 10^{-2}$	$2.9\ 10^{-1}$	$6.4\ 10^{-2}$	$7.4\ 10^{-2}$	$4.4\ 10^{-1}$	$5.1\ 10^{-2}$	$5.8\ 10^{-2}$	$2.5\ 10^{-1}$

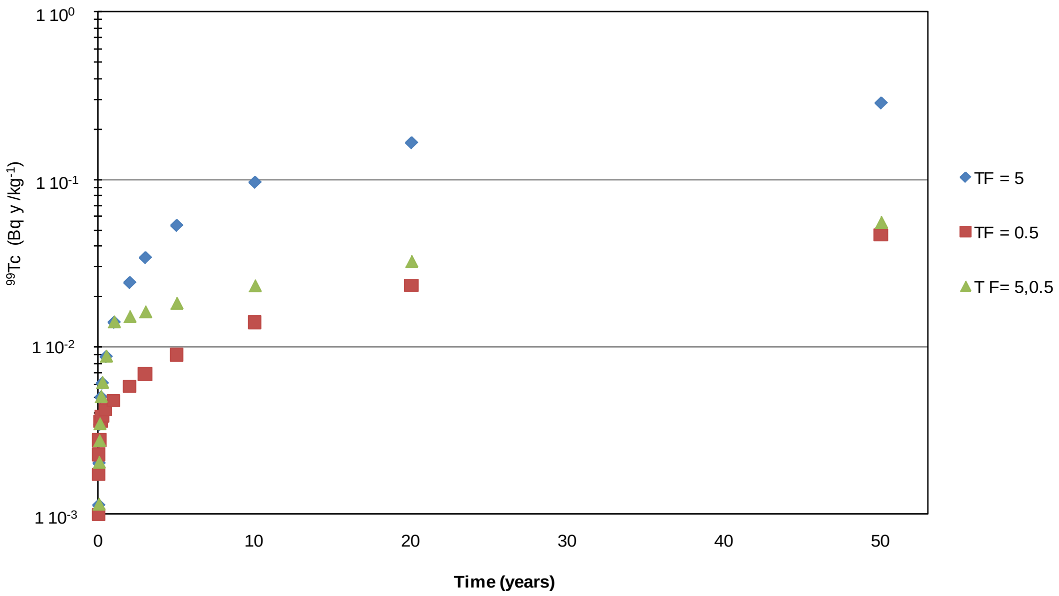

Figure D6 Time integrated activity concentration for green vegetables for an accidental release using different soil to crop TF values of 0.5 and 5 compared to the optimum combination of TF= 5,0.5

The calculated time integrated activity concentrations in milk and meat from cattle grazing pasture are shown in Table D5. Compared with using the optimum combination of TF values (TF= 40,5), the table shows that the choice of a single TF value of 5, ie, the value for the reduced form of technetium, underestimates the time integrated activity concentrations in milk and meat by about a factor of 2 after 1 year but at subsequent times the concentrations are very similar. The choice of a single TF value of 40 overestimates the time integrated activity concentration by factors of between 2 and 3 at all times following deposition.

Table D5 Time integrated activity concentrations for milk and meat products for an accidental release using different soil to crop TF values of 40 and 5 compared to the optimum combination of TF= 40,5

	Milk (Bq y l^{-1})			Meat (Bq y kg^{-1})		
Time (years)	TF=5	TF=40,5	TF=40	TF=5	TF=40,5	TF=40
0	0	0	0	0	0	0
1	$5.3\ 10^{-3}$	$1.0\ 10^{-2}$	$2.0\ 10^{-2}$	$1.8\ 10^{-1}$	$3.4\ 10^{-1}$	$6.6\ 10^{-1}$
10	$4.4\ 10^{-2}$	$4.9\ 10^{-2}$	$1.7\ 10^{-1}$	$1.5\ 10^{0}$	$1.6\ 10^{0}$	$5.5\ 10^{0}$
20	$8.3\ 10^{-2}$	$8.7\ 10^{-2}$	$2.7\ 10^{-1}$	$2.8\ 10^{0}$	$2.9\ 10^{0}$	$9.1\ 10^{0}$
50	$1.8\ 10^{-1}$	$1.8\ 10^{-1}$	$4.2\ 10^{-1}$	$5.9\ 10^{0}$	$6.0\ 10^{0}$	$1.4\ 10^{1}$

D2.1 Recommended TF values for use in the FARMLAND model for accidental release assessments

It is recommended that the use of a single TF value of 5 is appropriate for both pasture and edible crops within radiological assessments for accidental releases using FARMLAND. The use of a single value is important because other computer models might not have the flexibility to change the TF value for the different periods of time used in modelling to represent changes in agricultural practices over the year. This is a conservative approach and activity concentrations will be overestimated after the first year, as discussed above. However, similarly to routine releases, it is expected that ^{99}Tc will be a minor contributor when assessing doses and, in the case of an accidental release, it is more important to accurately estimate activity concentrations in the first year which is the period when they vary more markedly with time. However, if an overestimation after a year is not acceptable when calculating doses, then the combination of soil to crop TF values of 5 and 0.5 for edible crops and 40 and 5 for pasture should be used in the FARMLAND model. If it is only possible to use a single value and it is important to reflect the reduction of technetium in the terrestrial environment over time, two model runs could be undertaken using values of 5 (40 for pasture) and 0.5 (5 for pasture), which are then combined.

D3 REFERENCES

Brown J and Simmonds JR (1995). FARMLAND: a dynamic model for the transfer of radionuclides through terrestrial foodchains. Chilton, NRPB-R273.