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Experimental Studies on the Uptake of Technetium-99 to Terrestrial Crops

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Experimental Studies on the Uptake of Technetium-99 to Terrestrial Crops

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ABSTRACT

Technetium-99 is dispersed in the environment from a number of sources, the main ones being nuclear weapons testing, nuclear power plants, nuclear fuel processing facilities and facilities that treat or store radioactive waste. The pertechnetate ion, $^{99}TcO_4^-$, is the form produced during the nuclear fuel cycle and the most likely to be released into the environment.

A review published in 2011 by the Health Protection Agency (whose functions transferred to Public Health England in 2013) found that the availability for the root uptake of technetium into crops depends on whether the technetium is in a chemically non-reduced, more plant available form, such as TcO_4^-, or a chemically reduced, less plant available form, such as TcO_2. Based on the review, generic soil to crop transfer factor (TF) values for use in non-site-specific UK based radiological assessments were proposed, with the TF value for the reduced form of technetium in crops around a factor of 10 lower than that for the non-reduced form.

The aim of this small-scale experimental study was to provide further evidence that the generic assumption made on the difference between soil to crop TF values for non-reduced and reduced forms of technetium is valid. The study was also designed to establish likely time periods over which the chemical reduction of technetium takes place and to provide additional soil to crop TF values for use in UK based radiological assessments.

Soil to crop TFs for crops harvested from loam and peat based soils up to four months after contamination are about a factor of 10 higher than those seen in soil contaminated more than a year previously, indicating that the technetium has become reduced over this period. The values presented in this report are in good agreement with those proposed in the earlier review and also support the use of a generic soil to crop TF. For crops grown in peat based soil, TFs were typically a factor of three lower than those observed in the crops grown in loam based soil over the same period. Values obtained for peat soils also suggest a much faster chemical reduction of the technetium in the soil. Additional soil to crop TFs have also been measured for a range of crops, which can be used where more site-specific data for UK based radiological assessments are required.

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CONTENTS

1 INTRODUCTION

Technetium-99 (^{99}Tc) is a beta emitting radionuclide with a radiological half-life of 2.5 10^5 years produced by the fission of uranium-235 (^{235}U) and plutonium-239 (^{239}Pu). It is dispersed in the environment from a number of sources, the main ones being nuclear weapons testing, nuclear power plants, nuclear fuel processing facilities and facilities that treat or store radioactive waste. The pertechnetate ion ($^{99}TcO_4^-$) 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).

In 2011 the Health Protection Agency (whose functions transferred to Public Health England in 2013) published a critical review of data on the transfer of technetium into terrestrial crops and animal products and recommended soil to crop transfer factor (TF) values for use in UK based radiological assessments (Ewers et al, 2011). The data reviewed showed that the availability for the root uptake of technetium into crops depends on whether the technetium is in a chemically non-reduced, more plant available form, such as TcO_4^-, or a chemically reduced, less plant available form, such as TcO_2. Evidence from the review suggested that for the types of agricultural soil generally encountered in the UK, the time since contamination is the most important factor in determining the magnitude of uptake of technetium into crops.

Taking into consideration the differences in the chemical behaviour of technetium in the environment, Ewers et al (2011) presented soil to crop TFs for use in UK based radiological assessments for seven broad categories of crop. The technetium was assumed to be in a chemically reduced form for uptake into crops if it had been present in the soil for more than about four months. Based on this assumption, soil to crop TF values were categorised according to whether the technetium was assumed to be in a chemically reduced or non-reduced form. Based on limited data, Ewers et al (2011) proposed the use of a generic soil to crop TF value. The generic soil to crop TF value for the reduced form of technetium was assumed to be around a factor of 10 lower than that of the non-reduced form.

A small-scale experimental study has been carried out to provide further evidence that the generic assumption made on the difference between soil to crop TF values for non-reduced and reduced forms of technetium is valid. The study was also designed to establish likely time periods over which the chemical reduction of technetium takes place and to provide additional soil to crop TF values for use in UK based radiological assessments.

2 TRANSFER OF RADIONUCLIDES TO CROPS

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 be regarded as the soil to crop transfer factor (TF):

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

Any possible contamination from resuspension of contamination from the soil on to the edible crop can be ignored if the observed TF value is high (Brown and Simmonds, 1995).

Comparisons of transfer values are most reliable when based on the dry mass of the crop because the effects of differences in moisture content in the individual crops are removed. Calculated soil to crop TF values for crops from this experimental study are presented on a dry mass basis and will be denoted as TF_{dry} in this report. Transfer factors for crops used in radiological assessments are often expressed in terms of the fresh mass of the crop and, where such values are required, soil to crop TF values will be reported and denoted as TF_{fresh} in this report.

There are several factors that can affect the transfer of technetium into crops and the main ones are briefly discussed in Section 3. Further information can be found in the literature review produced by Ewers et al (2011).

3 FACTORS AFFECTING THE TRANSFER OF TECHNETIUM TO CROPS

The findings of the literature review conducted by Ewers et al (2011) showed that the magnitude of the uptake of technetium to crops depends critically on whether the technetium is in a chemically non-reduced form, such as TcO_4^-, or a reduced form, such as TcO_2. Several authors have reported that in aerobic soils TcO_4^-, which is the most stable form of technetium (Bennett and Willey, 2003), is very soluble in water over a wide pH range and is readily taken up by plants (Sheppard et al, 1983; Vandecasteele et al, 1989; Echevarria et al, 1994). Several studies investigating the uptake into pasture have reported a decrease over time in the rate of soil to plant transfer of technetium-99 when applied as TcO_4^- (Mousny and Myttenaere, 1982; Garten et al, 1984; Vandecasteele et al, 1989; Echevarria et al, 1994). This variation of transfer factors with time can be explained in terms of speciation changes from the highly soluble and readily available TcO_4^- to less plant 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). Ewers et al (2011) suggested that when considering the types of agricultural soil found in the UK, the time since contamination is the most important factor determining the magnitude of the uptake of technetium into crops and that after more than about four months following soil contamination uptake can decrease markedly.

Uptake of TcO_4^- can be lower from organic compared to mineral soils (Bergstrom and Wilkens, 1983). The uptake of technetium-99 applied as TcO_4^- into Swiss chard using mineral sand and peat soils was investigated using a lysimeter-scale study (Sheppard et al, 1983). The values showed that the observed soil to plant TF_{dry} values for Swiss chard grown in peat were around two orders of magnitude lower than those obtained for sand. Further information on the effects of soil type on the uptake of technetium into crops can be found in Ewers et al (2011).

4 EXPERIMENTAL DESIGN

4.1 Soil preparation

Two soils were considered in this project: a loam and a peat based soil. These soils were selected to represent two of the common types of agricultural soil found in the UK. Twelve batches of loam and four batches of peat soil with a mass of at least 10 kg each

(Standard International Union of Radioecologists protocol – IUR, 1989) were homogeneously contaminated with $^{99}TcO_4^-$. The soil was contaminated to give an activity concentration of approximately 5 kBq kg^{-1}, which ensured that sufficient activity could be measured in the crops grown (Ewers et al, 2011). In order to achieve homogeneous contamination, the soils were thoroughly mixed with a metal paddle attached to a power drill while an aqueous solution of $^{99}TcO_4^-$ was slowly added in aliquots.

To enable a range of different crops to be grown, a series of small containers was used. Water cisterns of 20-litre capacity were used with dimensions of 0.45 m x 0.3 m x 0.27 m. Drainage holes were drilled in each container and a second container was used to collect the drainage water.

Contaminated soils were then placed into the tubs, tilled, gently watered and left to settle for a minimum of seven days before seeding or planting.

4.2 Growing and harvesting of crops

Initially, in late summer 2010, contaminated loam based soil was placed in four containers and left for about nine months so that they could be used to investigate the longer term uptake of technetium in crops. Crops were grown in these containers at 293, 416 and 702 days following contamination, as detailed in Table 1.

TABLE 1 Soil type and crop assignment

Soil type	Days following soil contamination before crops seeded or planted	Crop type* and variety grown
Loam based	7	Swiss chard (Fordhook Giant)
	21	Dwarf French beans (The Prince)
	42	
	56	
	293	Swiss chard (Fordhook Giant)
		Dwarf French beans (The Prince)
		Tomatoes, (Bellstar)
		Potatoes (early variety
	416	Carrot (Autumn King 2)
		Spring onion (White Lisbon)
		Lettuce (Butterhead)
		Cabbage (Savoy King F1)
		Strawberries (Domanil)
	702	Swiss chard (Fordhook Giant)
Peat based	7	Swiss chard (Fordhook Giant)
	21	
	42	
	56	
	416	

* Swiss chard, carrot, spring onion, lettuce, savoy cabbage and dwarf French bean plants were cultivated from seeds. Potatoes were grown from seed tubers; tomato and strawberry plants were grown under greenhouse conditions to a height of around 15 cm before transplanting into containers.

In spring 2011, a further 12 containers were prepared. Eight containers were filled with loam and four with contaminated peat based soils. Crops were then grown at 7, 21, 43 and 56 days following contamination in both soils. Crops were also grown in the peat based soil in 2012, 416 days following contamination (see Table 1).

Prior to growing crops, samples of approximately 70–100 g of soil were collected from each tub to confirm the activity concentrations of technetium-99 and soil homogeneity. The crops grown were chosen to represent some of the broad categories used for UK based radiological assessments and to provide additional data where information is sparse or not available (Ewers et al, 2011); details are given in Table 1. Swiss chard, which is not a common crop, was specifically chosen due to the expected high transfer of technetium from soil which had been identified during the earlier review.

During the growing season crops were watered as required, regularly fed with a proprietary liquid feed and treated as necessary with plant insecticides and pesticides. The containers and experimental set-up are shown in Figure 1.

FIGURE 1 Experimental set up for growing crops in soils contaminated with $^{99}TcO_4^-$

Crops with an associated soil sample were collected at crop maturity. For dwarf French beans, tomatoes and strawberries that mature over several weeks, edible samples were collected from successive harvests and combined as one sample. Soils associated with these crops were collected at the first and final harvests and combined as one sample. After the initial harvest of Swiss chard the crops were allowed to re-grow from their root base and a second set of samples was collected for analysis together with the associated soils; this was approximately 25–60 days after the end of the first harvesting period.

4.3 Sample preparation and analyses

All crops were thoroughly washed in tap water to avoid the possibility of soil contamination and edible parts were separated. For both the potato and carrots, peel and flesh were prepared separately to determine the distribution of activity between the two parts. Similarly, for the cabbage the inner and outer leaves were removed and analysed separately. To stabilise and prepare samples for analysis, crops were roughly chopped or shredded, freeze-dried and then homogenised to a fine powder. The soil samples were oven dried at 105–110 ºC and processed to produce a finely milled, homogeneous product. Moisture content was determined in both crop and soil samples.

The determination of technetium-99 in the samples was based on an established method by Holm and Riosecco (1984), which has been further developed and validated as an ISO 17025:2005 accredited method for use in the PHE laboratory by Hammond et al (2011).

Based on the expected activity concentrations, the dried samples used for the analysis were about 3 g for the soils and between 2 g and 20 g for the edible crops. All activity concentrations were calculated as Bq g^{-1} with respect to dry mass for crops and soils.

5 EXPERIMENTAL STUDIES: VALUES AND DISCUSSION

5.1 Data analysis and presentation of data

Activity concentrations in edible crops and soils were measured with the objective of estimating representative transfer factor values for technetium-99 in a given crop.

For the purposes of calculating soil to crop TFs, the data has been rounded to two significant figures to reflect the use of this data within radiological assessments. In cases where duplicate samples were analysed, the reported TFs represent a weighted mean of the values for the individual crops and associated soil samples.

Data was first reviewed and categorised based on whether the technetium was assumed to be in a chemically reduced form (contamination present in the soil at harvest for more than about four months) or a non-reduced form (present in the soil at harvest for less than about four months) (Ewers et al, 2011). Geometric means were then calculated from the individual TFs and presented with corresponding data ranges. Calculated soil to crop TF values for crops from this experimental study are presented on either a dry or fresh mass basis, and are denoted as TF_{dry} or TF_{fresh}, respectively, in this report.

5.2 Effect of chemical form and the time since contamination on the uptake of technetium into crops

Transfer factors for the uptake of technetium-99 into Swiss chard and dwarf French beans grown in loam based soils are presented in Tables 2 and 3, respectively. As described previously, after the first Swiss chard crop was harvested it was allowed to re-grow from the base of the plant and a second crop and associated soils were then collected and processed; this data is also given. Although samples collected from the second growth do not strictly relate to crops grown from seed, the consistent data obtained suggests that they are valid and can be compared to the other values given (see Table 2).

TABLE 2 Soil to crop TFs* for technetium uptake into Swiss chard grown in loam based soil

Days following soil contamination before crops sown	Days following soil contamination at harvest	TF_{dry}
7	61	38†
Regrowth‡	125	150
21	76	150
Regrowth	125	110
42	83	97
Regrowth	125	120
56	97	100
Regrowth	138	150
293	383	7.4
702	791	30

* Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to two significant figures.
† Result excluded from further evaluation, see Section 5.2.
‡ Data represents re-growth of crop from root base following original harvest, see Section 5.2.

TABLE 3 Soil to crop TFs* for technetium uptake into dwarf French beans grown in loam based soil

Days following soil contamination before crop seeded	Days following soil contamination at midpoint of harvest	TF_{dry}
7	98	0.9
21	108	1.2
42	125	1.7
56	131	2.0
293	384	1.2

* Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to two significant figures.

Data for Swiss chard harvested 61 days after soil contamination is not consistent with the other values given in Table 2. The measurements showed a variation in activity concentrations in the associated soil of around 50% which was not observed in other crop and soil combinations that were analysed, where good agreement between duplicate measurements was found. This difference could be as a result of a non-uniform distribution of the technetium in the soil. The TF_{dry} value obtained from the re-growth of the same chard crop harvested 125 days after soil contamination was much more consistent with other observed values (see Table 2). This could be due to the technetium in the soil becoming more homogeneously distributed over time as a result of natural weathering, for example.

Based on the review by Ewers et al (2011) it was assumed that technetium-99 would be in a chemically reduced form for uptake into crops in an agricultural based soil such as loam if the contamination had been present for more than about four months at the time of harvest. Results from this study for the uptake of technetium-99 into Swiss chard grown in loam soil showed no significant decrease in the range of TF_{dry} values obtained at harvest around four months after soil contamination, with values of 97–150 (see Table 2). The results therefore provide further evidence that it is reasonable to assume that technetium does not become significantly reduced in loam soil at times less than about four months following contamination.

Lower TF_{dry} values of 7.4 and 30 were observed for Swiss chard harvested around 380 and 790 days, respectively, following soil contamination (see Table 2). These results strongly suggest that the technetium present in the soil became chemically reduced to a far less available form such as $^{99}TcO_2$ for uptake at some time after four months following soil contamination. Evidence suggests that the TF_{dry} value observed at harvest at about 800 days following soil contamination should be lower than that obtained at about 380 days (Ewers et al, 2011) but this was not observed in this study (see Table 2). However, it is reasonable to assume that the technetium is in a reduced form because the value is still considerably lower than that seen for harvests within a few months of contamination.

Overall, the change in observed values over time given in Table 2 is in agreement with other studies (Mousny and Myttenaeare, 1982; Sheppard et al, 1983; Garten et al, 1984; Stalmans et al, 1986; Vandecasteele et al, 1989; Echevarria et al, 1994; Bennett and Willey, 2003).

Observed soil to crop TF_{dry} values for dwarf French beans (at the mid-point of harvest following soil contamination) were significantly lower than those obtained for Swiss chard. The range of values obtained of 0.9–2.0 showed no variation across all time periods after soil contamination, including the sample harvested at about 380 days (see Table 3). It is expected that the technetium in the soil should have become chemically reduced and less available for plant uptake by this time and the results for Swiss chard (see Table 2) support this view. However, despite an investigation no reason could be found as to why no significant difference in root uptake over time was observed in the bean samples. Dwarf French beans were grown again in 2012 but unfortunately due to weather conditions the crops failed to grow and no measurements could be made. Based on the other information available, the TF_{dry} value of 1.2 observed for samples harvested about 380 days after soil contamination has been assumed to be representative of technetium in a chemically reduced form.

5.3 Effect of soil type on the uptake of technetium into crops

Observed soil to crop TF_{dry} values for uptake of technetium-99 into Swiss chard grown in peat and loam based soils are presented in Table 4 and Figure 2. For comparative purposes data observed for crops grown in the loam based soils is reproduced from Table 2. As described previously, after harvesting the first crop the chard was allowed to re-grow and the second harvests, together with the associated soils, were subsequently collected and processed (see Table 4 and Figure 2).

Table 4 shows that the observed soil to crop TF_{dry} values for Swiss chard harvested up to three to four months following soil contamination are higher for loam based soils than for peat based soils by up to a factor of five. Based on the results for loam based soils discussed in Section 5.2 and other experimental studies (Ewers et al, 2011), it is reasonable to assume that

TABLE 4 Soil to crop TFs* for uptake of technetium into Swiss chard grown in loam and peat based soils

Days following soil contamination before crops seeded	Days following soil contamination at harvest	TF_{dry}	
		Loam	Peat
7	61	38†	55
Regrowth‡	125	150	27
21	76	150	46
Regrowth	125	110	30
42	83	97	63
Regrowth	125	120	14
56	97	100	48
Regrowth	138	150	–§

* Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to two significant figures.
† Result excluded from further evaluation, see Section 5.2.
‡ Data represents re-growth of crop from root base following original harvest, see Section 5.2.
§ No data available; crop did not grow.

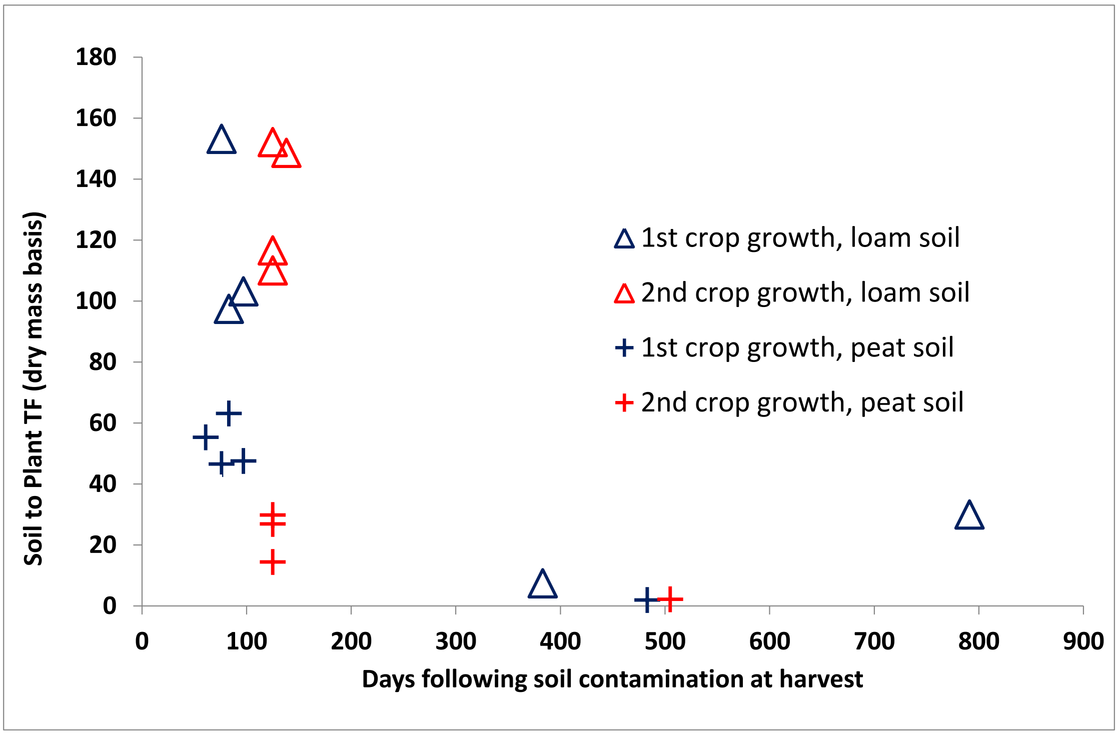

FIGURE 2 Soil to crop TFs for Swiss chard grown in loam and peat based soils

the technetium should be in the non-reduced, more plant available form over this period. TF values for Swiss chard harvested from both soil types over longer time periods of between one and two years following contamination were also measured and are shown in Figure 2. The soil to crop TF_{dry} values are broadly similar in both soil types, when taking into account the uncertainties associated with the measurements, and can be assumed to be representative of uptake from soils where technetium is in the reduced chemical form. However, the results suggest that the technetium is becoming reduced much faster in the peat based soil due to the characteristics of peat soils, leading to much lower TF values within three to four months of contamination (see Table 4 and Figure 2).

Over longer periods of one to two years, the observed uptake values for both soil types become similar (see Figure 2).

Soil to crop TF_{dry} values from the re-growth of the chard following the first harvest were obtained for both soil types (see Table 4 and Figure 2). In contrast to chard grown in the loam based soil, values for crops grown in peat gave TF values that were about a factor of two lower when compared to those obtained from the first crop samples (see Figure 2). It is difficult to draw firm conclusions from these values due to the fact that these TF_{dry} values represent crops that were grown from the root base. However, they do support the continuing faster reduction of technetium in peat based soils within a three to four month period following contamination compared to the loam based soil. The TF_{dry} measured at about 500 days in chard re-growth in peat based soils is very similar to that in the first harvest at that time, adding weight to the expectation that the technetium has reduced.

5.4 Soil to crop transfer factors for other crop types

Potatoes, tomatoes, carrots, spring onions, lettuce, cabbage and strawberries were identified as crops where soil to crop transfer data for the uptake of technetium was considered sparse (Ewers et al, 2011). Table 5 shows the observed soil to crop TF_{dry} values obtained from these crop types grown in a loam based soil contaminated more than a year earlier. Owing to the length of time that the contamination had been present in the soil, the observed soil to crop TF_{dry} values can be assumed to be representative of those in the reduced, less plant available form.

Several studies have shown activity concentrations of certain radionuclides in both the peel and flesh of the edible crop. For example, isotopes of plutonium and americium are considerably higher in the peel than in the flesh of the potato tuber, but for caesium this distribution has been found to be more uniform (Popplewell et al, 1984; Green et al, 1995). In this experimental study, in both potatoes and carrots no significant difference in the distribution of technetium-99 activity between the flesh and the peel was observed (Table 5). This result is supported by results from a study conducted by Yanagisawa and Muramatsu (1993). For cabbage, a small difference in the distribution of technetium-99 activity between the inner and outer leaves was found (Table 5).

TABLE 5 Soil to crop TFs* for technetium uptake into other crop types grown in loam based soil

	Days following soil contamination	
Crop type	**At harvest**	**TF_{dry}**
Tomato	397[b]	0.3
Strawberry	437[b]	0.3
Potato tuber	389	0.1 (flesh) 0.2 (peel)
Carrot	483	2.0 (flesh) 1.7 (peel)
Spring onion	505	3.2
Lettuce	483	4.1
Cabbage	559	1.4 (outer leaf) 5.5 (inner leaf)

* Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to two significant figures.
† Represents soil contamination at the midpoint of the harvest.

6 SUMMARY AND DATA COMPARISON

6.1 Summary of experimental data

As discussed previously, the form of technetium determines the degree to which it is available for uptake into crops. A summary of the soil to crop TF_{dry} values obtained from this experimental study is provided in Table 6. Based on the discussions in Section 5, soil to crop TF_{dry} values have been categorised according to whether they are representative of technetium uptake in the reduced form at crop harvest (contamination present in the soil for more than about four months) or in a non-reduced, more plant available form (present in the soil for less than about four months).

6.2 Comparison of experimental data with published values

A range of soil to crop TF_{dry} values extracted from the review published by Ewers et al (2011) is given in Table 7, together with values observed in this study.

No admissible data representative of TF_{dry} values in a non-reduced form was found in the review and so comparisons can only be made with those assumed to be in a chemically reduced form. With the exception of tomatoes and strawberries, the majority of observed TF_{dry} values from this experimental study were within the range of admissible crop values produced by Ewers et al (2011).

TABLE 6 Summary of experimental soil to crop TFs* obtained for technetium

Crop	Soil type	Days following soil contamination at harvest	TF_{dry}	Assumed chemical form of $^{99}Tc^{\dagger}$
Swiss chard	Loam	76–138 383–791	120 (97–150)‡ 15 (7.4–30)	Non-reduced Reduced
Swiss chard	Peat	61–125 483–505	37 (14–63) 2.0 (1.9–2.2)	Non-reduced Reduced
Dwarf French beans	Loam	98–131 384	1.5 (0.9–2.0) 1.2	Non-reduced Reduced
Whole potato tubers	Loam	389	0.16[d]	Reduced
Tomato fruit	Loam	397	0.32	Reduced
Strawberry	Loam	437	0.34	Reduced
Whole carrot	Loam	483	1.8[d]	Reduced
Spring onion	Loam	505	3.2	Reduced
Lettuce	Loam	483	4.1	Reduced
Whole cabbage	Loam	559	2.7§	Reduced

* Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to two significant figures.

† Data assumed to be representative of TF_{dry} values in either the reduced or non-reduced chemical form.

‡ Geometric mean and data range in brackets.

§ Geometric mean of TF_{dry} values for potato flesh and peel, carrot flesh and peel, and inner and outer cabbage leaves.

Further comparisons can be made for individual crops. For dwarf French beans, a TF_{dry} value of 1.2 was calculated from this experimental study, which is the same as the value for French beans in the earlier review. The measured TF_{dry} value of 0.16 for whole potato tubers is within the range of admissible data found in the review where technetium is assumed to be in a chemically reduced form (Ewers et al, 2011). For tomatoes, only one admissible study was found in the review, which relates to a field study involving long-term deposits of technetium of marine origin (Green et al, 1995). The value of 2 is significantly greater than the value of around 0.3 observed for both tomato and strawberry fruits in this study. Other admissible TF_{dry} values were also published by Green et al (1995), including lettuce, carrots and onions, all of which are in reasonable agreement with those observed in this experimental study (Table 7).

A study investigating the transfer of radioactivity from seaweed to terrestrial plants (Brown et al, 2009) is a further source of soil to crop data. However, since contributions other than root uptake could not be ruled out, results from this study could only be regarded as soil to crop concentration ratios (CR_{dry}). Values for uptake of technetium of around 0.01 in tomato were observed by Brown et al (2009), significantly lower than the value found in this study and others (Ewers et al, 2011). The study also reported CR_{dry} values for other crop types. For potato tubers, the majority of values were around 0.1, which were within the range of values published in the review and obtained experimentally (see Table 7). For Swiss chard, two wide-ranging CR_{dry} values of 0.6 and 10 were observed by Brown et al (2009), with the highest value similar to the range of values given in the table where technetium is assumed to be in a chemically reduced form. For carrots, values were within the range of 0.25 to 0.33, both lower than those found in the review and obtained experimentally. Values from 0.13 to 0.56 were found by Brown et al (2009), which are lower that the values presented in Table 7. Two CR_{dry} values of 0.37 and 22 for lettuce were found by Brown et al (2009). These values cover a wide range and span the values published in the review and obtained experimentally (see Table 7).

TABLE 7 Comparison of published soil to crop TFs* for technetium assumed to be in the reduced chemical form

Experimental (this study)		Ewers et al (2011)	
Crop types†	**TF_{dry}**	**Crop types**	**TF_{dry}**
Legumes (dwarf French beans)	1.2	Legumes (French beans, broad beans, peas, mange tout)	0.3 (0.1–1.2)‡
Leafy vegetables/brassicas (Swiss chard§, whole cabbage)	4.8 (1.9–30)	Leafy vegetables/brassicas (Swiss chard§, cabbage, cauliflower, sprouts)	4 (0.9–40)
Potatoes	0.16	Potatoes	0.3 (0.1–0.7)
Tomato, strawberry	0.3	Tomato	2
Spring onion	3.2	Onion from sets and seeds, leeks	0.5 (0.25–2.6)
Lettuce	4.1	Lettuce	4
Carrot	2	Carrot	–

* Expressed in terms of Bq kg^{-1} dry mass crop and soil and rounded to two significant figures. Data assumed to be representative of TF_{dry} values in the reduced chemical form, see Section 5.

† Where data exists crop types were grouped into the same broad category such as brassicas, eg Swiss chard and cabbage.

‡ Geometric mean and data range in brackets.

§ Includes results from Swiss chard grown in both loam and peat soil types.

6.3 Comparison of experimental data with those currently recommended for use in UK based radiological assessments

For radiological assessments, soil to crop TF values are often expressed on a fresh mass basis for use in predictive models such as the PHE dynamic foodchain model FARMLAND (Brown and Simmonds, 1995). Currently, the FARMLAND model does not take into account the variation over time in the transfer of technetium from soil due to changes in its chemical form. The review by Ewers et al (2011) and data from this experimental study have demonstrated that there is a significant difference in the availability of technetium for root uptake into crops depending on its chemical form.

Owing to limited data found for the uptake of technetium into crops, the use of a generic soil to crop TF value for all crop types was proposed in the earlier review for non-site-specific radiological assessments, one for each assumed chemical form. A factor of 10 difference between the reduced and non-reduced forms was recommended, with generic TF_{fresh} values of 0.5 and 5 for the reduced and non-reduced chemical forms, respectively, for all crops

(Ewers et al, 2011). Guidance on the choice of value for different types of radiological assessment was also provided. Experimental TF_{fresh} values obtained from this study for Swiss chard grown in both loam and peat based soils demonstrate that this is a reasonable assumption, with values of around 0.5 and 7 for reduced and non-reduced forms, respectively, being observed (see Table 8). For French beans no significant difference was found with time following contamination. As stated previously (see Section 5.2), this was investigated and no reason could be given as to why no significant difference in root uptake over time was observed in the bean samples. The TF_{fresh} values obtained from this study for French beans where technetium was assumed to be in a non-reduced form were around a factor of 30 lower than the recommended generic value of five; values for the reduced form were in reasonable agreement with that recommended by the review (Table 8).

TABLE 8 Comparison of experimental soil to crop TFs* (fresh mass basis) for technetium to those currently recommended for use in UK based radiological assessments, with technetium assumed to be in: (a) non-reduced form and (b) reduced form

(a) Technetium assumed to be in a non-reduced chemical form

Experimental (this study)		Recommended default values†	
Crop type	**TF_{fresh}**	**Crop type**	**TF_{fresh}**
Swiss chard‡	6.7 (1.2–15)§	All crop types¶	5
Dwarf French beans	0.18 (0.11–0.22)	All crop types	5

(b) Technetium assumed to be in a reduced chemical form

Experimental (this study)		Recommended default values†	
Crop type	**TF_{fresh}**	**Crop type**	**TF_{fresh}**
Swiss chard‡, whole cabbage	0.48 (0.22–2.50)	All crop types¶	0.5
Dwarf French beans	0.15	All crop types	0.5
Whole potato tubers	0.03	All crop types	0.5
Tomato, strawberry	0.04	All crop types	0.5
Spring onion	0.27	All crop types	0.5
Lettuce	0.24	All crop types	0.5
Whole carrot	0.21	All crop types	0.5

* Expressed in terms of Bq kg^{-1} fresh mass crop and dry mass soil and rounded to two significant figures. Data assumed to be representative of TF_{fresh} values in either the reduced or non-reduced chemical form.

† Recommended soil to crop default values for technetium for non-site specific radiological assessments within the FARMLAND model in the UK (Ewers et al, 2011).

‡ Includes results from Swiss chard grown in both loam and peat soil types.

§ Geometric mean and data range in brackets.

¶ Recommended default values proposed are for all crop types in the FARMLAND model (Ewers et al, 2011).

Where technetium was assumed to be reduced, TF_{fresh} values of around 0.2 for dwarf French beans, spring onions, lettuce and carrots were obtained, which are in reasonable agreement with the recommended value of 0.5. Values about an order of magnitude lower were obtained for potatoes, tomatoes and strawberries (Table 8).

In summary, the generic TF_{fresh} values proposed in the review are conservative for most crops but do encompass brassicas, such as Swiss chard, which have been found to give higher soil to crop TF values. The results from this study support the recommendations for generic soil to crop TF values proposed in the review for non-site-specific radiological assessments within the UK. For assessments where more site-specific data are required, additional data published in the review and from this study should be considered.

7 CONCLUSIONS

A small-scale experimental study on the transfer of technetium into terrestrial crops has been conducted. The purpose was to provide further evidence that the generic assumption made on the difference between soil to crop TF values for non-reduced and reduced forms of technetium is valid. The study was also designed to establish likely time periods over which the chemical reduction of technetium could take place and provide additional soil to crop TF values for use in UK based radiological assessments.

This study has found that TF values for brassicas, such as Swiss chard, harvested from peat and loam based soils up to four months after soil contamination are approximately a factor of 10 higher than those seen in the same crop grown in a soil that had been contaminated with technetium over a year previously. This finding supports the recommendations made in an earlier review that the soil to crop TF value for the chemically reduced form of technetium is around a factor of 10 lower than that for the non-reduced form (Ewers et al, 2011). The results suggest that at some point after four months the technetium had become chemically reduced and therefore less plant available for uptake, a finding identified in the review.

For Swiss chard grown in peat based soil, TFs were typically around a factor of three lower than when compared to those from the loam based soil over the same time periods. The values obtained from peat also support the evidence that the uptake of technetium is around a factor of 10 higher between the reduced and non-reduced forms. The lower values obtained are likely to be due to a much faster chemical reduction of technetium in the soil, which would lead to it being less available for root uptake. This supports the findings of the earlier review by Ewers et al (2011). For peat based soil, data from this study could be used for more site-specific radiological assessments within the UK.

Over the same growing period and under the same experimental conditions, the factor of 10 difference between the soil to crop TF values for the reduced and non-reduced forms of technetium was not observed in French beans. Although no reason could be identified why this was the case, the TF values observed experimentally were found to be in good agreement with those found in the open literature (Ewers et al, 2011).

This experimental study has provided additional soil to crop TFs where data for the UK was considered sparse. With the exception of tomatoes, experimental values for potatoes, spring onion, lettuce and carrot were found to be lower, significantly in some cases, than the generic value recommended for use in radiological assessments, but were in reasonable agreement with the limited data published in the open literature. They can therefore be used to provide additional data to support site-specific radiological assessments within the UK.

Overall, the generic TF_{fresh} values proposed by Ewers et al (2011) are conservative for most crops but do encompass brassicas, such as Swiss chard, which have been found to give higher soil to crop TF values. The experimental results from this study support the recommendations for the generic soil to crop TF values proposed in the review. For assessments where more site-specific data is required, additional data published in the review and from this study should be considered.

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