A Long-term Study of the Transfer of Radionuclides from Soil to Fruit

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ABSTRACT

This report brings together the results of long term studies of the uptake of selected radionuclides by perennial fruit crops. Over a nine-year period, apples, blackcurrants, gooseberries and strawberries have been grown in an area of land that has been reclaimed from the sea. This land is in north-west England and provided an opportunity for field research because of the authorised discharges of radionuclides from Sellafield. Apples have also been grown over periods of up to six years in an established lysimeter facility where three diverse soil types have been artificially contaminated.

The results indicated that, when radionuclide uptake occurs only via transfer from soil, then any storage of activity in plant parts such as branches does not result in increased transfer to fruit in later years. In these circumstances, soil:crop transfer parameters derived from short term experiments would also be applicable in the longer term, at least for radiological assessment purposes. In terms of soil:crop transfer factors, changes should be considered for the default values for isotopes of caesium, plutonium and americium currently assumed in the dynamic foodchain model FARMLAND. The current value for strontium should be retained.

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EXECUTIVE SUMMARY

Fruit is an increasingly important dietary component, but data on transfer from soil to crop are scarce. This report brings together results from two parallel studies carried out over several years. The first study made use of an area of land in north-west England that had been reclaimed from the sea in the late 1970s. The soil contains a selection of radionuclides as a result of the authorised discharges into the Irish Sea from the Sellafield nuclear fuel reprocessing plant. Activity concentrations in the soil are sufficient to provide opportunities for field research, although the resultant concentrations in foodstuffs are well below those that would give rise to radiological concern. The second study involved an established lysimeter facility containing three diverse soil types that had been artificially contaminated in 1983.

In this project, the aim was to investigate long term transfer to different crops at the field site, and to elicit differences in transfer between soil types using a single crop at the lysimeter facility. The species grown at the reclaimed land site were blackcurrants, gooseberries, strawberries and two varieties of apple, Egremont Russet and Fiesta. The same varieties of apple were grown in the lysimeter facility. The radionuclides of interest were ^{137}Cs, ^{90}Sr, ^{239,240}Pu and ^{241}Am.

For perennial fruiting species such as trees and bushes, there is the possibility that radionuclides may be taken up from the soil but stored in plant parts such as branches. There is then the potential for translocation of activity to fruits in a later season. A comprehensive evaluation of the distribution of radionuclides between plant parts and the potential for translocation to fruit has not been undertaken here. Instead, the approach adopted was to use the observed temporal changes in activity concentrations in fruit together with the derived soil:crop transfer factors (TFs) to determine whether storage and translocation were important from the radiological assessment point of view.

The results from both parts of the investigation indicated that there was no systematic increase in the activity concentrations in fruit over time. Variations in soil:crop TF values were comparable with those observed in earlier studies, and again no systematic increases were observed over time. Overall, the results indicated that, when root uptake is the only process by which activity is entering the plant, then long term storage in plant parts and subsequent translocation is not an important contributor to activity concentrations in fruit. In such cases, uptake in the longer term could be reasonably predicted on the basis of short-term experiments, at least for radiological protection purposes. In contrast, information published elsewhere indicates that if the plant was originally contaminated via direct deposition, then storage and translocation could be important.

The implications for the soil:crop TFs currently assumed in the HPA-RPD dynamic foodchain model FARMLAND are that the value presently assumed for ^{90}Sr is reasonable but that the values observed for the other radionuclides of interest

were much lower than the defaults. Consideration should therefore be given to the adoption of lower values for isotopes of caesium, plutonium and americium. The TF values observed for peat soils differed from those for sand and loam, which was consistent with previous results for other types of crop. However, fruit crops are not generally grown in peat soils and so these results do not have implications for the general use of the FARMLAND model.

CONTENTS

1 INTRODUCTION

Soil-crop transfer factors are important components of radiological assessments dealing with accidental and routine releases of radionuclides to the environment. Fruit is an increasingly important dietary component, but data on transfer from soil to crop are scarce (Venter et al, 2001). In consequence, some assessment models use soil:crop transfer factors (TFs) for other crops such as green vegetables as a basis for the prediction of transfer to fruit. The significance of the transfer of radionuclides to fruit was recognised by the International Union of Radioecologists (IUR) and the International Atomic Energy Agency (IAEA). In September 1997, the two organisations set up a working group (usually referred to as the Fruits Working Group) under the Biosphere Modelling and Assessment (BIOMASS) Co-ordinated Research Programme. The work of this group included model intercomparisons and model validations using a scenario based on direct deposition from atmosphere. Although the models included compartments using soil:crop TFs, initial deposition onto fruit and leaf surfaces were the most important pathways (IAEA, 2003).

In the late 1980s and early 1990s, the National Radiological Protection Board (NRPB)[*] carried out a comprehensive field study of radionuclide transfer along terrestrial foodchains, making use of a plot of land that had been reclaimed from the sea. The study included the uptake of radionuclides from soil to pasture and thence to milk and meat, as well as radionuclide transfer from soil into a wide range of vegetables, fruits and grain. The results have been issued in a series of publications (Green et al, 1994; Green et al, 1995a and 1995b; Green and Wilkins, 1995a and 1995b; Green et al, 1997).

One of these publications dealt with the transfer of radionuclides to a range of fruit species grown on reclaimed land during the period 1993-1995 (Green et al, 1997). The paper also contained data on limited species grown during the same period in a lysimeter facility where different soils had been artificially contaminated. In both cases the radionuclides of interest were ^{137}Cs, ^{90}Sr, ^{239,240}Pu and ^{241}Am. The results of these two sets of experiments were combined to elicit the implications for the parameter values that had been assumed in the NRPB's dynamic foodchain model FARMLAND (Mayall, 1995).

Studies at both the reclaimed land site and in the lysimeters continued after 1995, in particular to ascertain whether there were any longer term temporal changes in the transfer parameters. This report presents the results of the measurements made from 1996 to 2002 together with those from the earlier work in order to derive the implications for radiological assessments.

[*] NRPB became part of the Health Protection Agency (HPA) with effect from 1 April 2005.

2 EXPERIMENTAL

From the botanical point of view, vegetables such as peas or beans, cereals such as oats and other crops such as tomatoes are strictly defined as fruits, but such crops were not considered within this project. Desai and Salunkhe (1991) have defined fruit as "plant parts that have fragrant, aromatic flavours and are either naturally sweet or normally sweetened with sugar". The species selected for study within this project conformed with this more practical, culinary definition.

2.1 The reclaimed land site

The field study was based at an area of land on the north west coast of England that was reclaimed from the sea in the late 1970s. The site is about 70 km from the Sellafield nuclear fuel reprocessing facility. It receives negligible deposition from radionuclides discharged to atmosphere from either the Sellafield facility or the nearby Heysham nuclear power station (Green et al, 1995a). The soil in the reclaimed land contains radionuclides originally discharged into the Irish sea from Sellafield, and the concentrations are sufficient for studies of soil:plant transfer to be feasible. However, the activity concentrations measured in the fresh crops are only small fractions of the relevant Generalised Derived Limits (GDLs) (NRPB, 1998), and so are not of radiological significance. As in the case of the earlier work on uptake by fruit, the radionuclides of interest in this longer term study were ^{137}Cs, ^{90}Sr, ^{239,240}Pu and ^{241}Am.

The varieties of fruit grown were chosen to represent those that could reasonably be cultivated in the UK. For the long-term study the fruits grown were apples, blackcurrants, gooseberries and strawberries. These broadly represent a tree fruit, bush fruits and herbaceous fruit, which correspond well with those chosen for the BIOMASS programme (IAEA, 2003). As part of the earlier study, melons and rhubarb had also been grown (Green et al, 1997), but these have not been considered further in this report.

This coastal site is susceptible to strong onshore winds and deposits of salt spray. For this reason, the apple trees were grown inside a polythene tunnel. Two varieties were chosen in order to provide cross-pollination. These were Egremont Russet, hereafter referred to as Egremont, and Fiesta. The height of the tunnel was limited, and so both of these varieties had been grafted on to an M.26 dwarfing rootstock and were also controlled by pruning. Blackcurrants, gooseberries and strawberries could be grown successfully outdoors. Raspberries are a popular soft fruit in the UK but could not be grown successfully at this site.

The site was managed by an experienced local gardener. Farmyard manure had been applied regularly to the land during the previous study on vegetables (Green et al, 1995a). Once the fruit plants were established, a surface application of a commercial NPK fertiliser (Growmore) was made at the rate recommended by the manufacturer (68 g m^{-2} monthly).

2.2 The lysimeter facility

This part of the study made use of an established facility at the NRPB's laboratories at Chilton in central southern England. The facility contains three diverse soil types, Hamble loam, Fyfield sand and Adventurers peat. Each soil had been artificially contaminated with ^{90}Sr, ^{137}Cs, $^{239,240}Pu$ and ^{241}Am during the winter of 1983/4. The facility contains three pairs of lysimeters, with one pair being allocated to each of the three soil types.

The fruit crops grown in the lysimeters were originally confined to apples and strawberries. The varieties chosen were the same as those used at the reclaimed land site. Plants and trees for both parts of the study were purchased from the same supplier at the same time. For each of the three pairs of lysimeters, one contained two trees of each variety of apple and the other was planted with strawberries. The long term study described in this report was confined to apples.

The soil in all of the lysimeters had been well-mixed during cultivation of other crops in earlier years, and had been dug over again prior to the planting of the fruit crops. The soils were not disturbed after planting. A surface application of a commercial NPK fertiliser (Growmore) was made on a monthly basis during the growing season. The application rate was that recommended by the manufacturer (68 g m^{-2}).

2.3 Sampling

For both parts of the study, all crops were sampled at maturity. Unpalatable parts normally removed from strawberries and gooseberries were separated immediately after sampling and the fruit was washed and frozen; blackcurrants were washed and frozen as picked. Samples of strawberries, gooseberries and blackcurrants from the field study were transported to the Chilton laboratory in the frozen state and kept so until required for analysis. Apples from the field site were transported to the laboratory within two days of sampling and did not therefore require freezing. Apples from both the field site and the lysimeters were washed and then kept in a refrigerator until required for analysis.

Previous studies at the reclaimed land site had indicated that the concentrations of radionuclides in the soil across the plot was non-uniform even after extensive cultivation (Green et al, 1995a). Consequently, in order to provide the most rigorous estimates of soil:crop TFs, samples of soil were collected at various locations within the growing area for each crop. The cultivated soil was not amenable to the use of a soil corer, and so for each sample an area of 100 mm diameter was marked out and all of the soil to a depth of 100 mm was removed using standard gardening tools. Typically three such samples were collected from a row of about 14 m in length, fewer samples being collected from shorter rows on a *pro rata* basis. Each sample was kept and treated separately.

In both parts of the study, samples of soil were not always collected each time that a crop was harvested. The reasons for this are discussed in Section 3.1.

2.4 Sample preparation

Prior to beginning radiochemical analysis, any fruit that had been frozen was allowed to defrost and any that had not been already cleaned was washed thoroughly. The cores were removed from all apples using a cork-boring tool of an appropriate diameter. All samples of fruit were dried at 110°C, milled and thoroughly mixed. An aliquot was packed into a suitable container, usually a 1 l re-entrant container and the ^{137}Cs content determined using high-resolution γ-ray spectrometry. These aliquots were then ashed and taken into solution. The total mass of solution was determined. Weighed aliquots corresponding to about 80% of the total mass were then taken for the sequential determination of $^{239,240}Pu$, ^{241}Am and ^{90}Sr.

Each sample of soil was dried at a temperature not exceeding 80°C, roughly ground and mixed well. An aliquot was packed into a standard Petri dish of 90 mm diameter for the determination of ^{137}Cs by high resolution γ-ray spectrometry. This aliquot was then ashed at 450 °C, leached with boiling aqua regia and filtered. The filtrate was weighed and stored in a sealed plastic bottle. Separate weighed aliquots were taken for the determination of ^{90}Sr (equivalent to about 80% of the sample), and actinides (about 5%).

High-resolution γ-ray spectrometry was carried out using hyperpure Ge detectors, housed in a purpose-built facility and appropriately calibrated. The analytical methods for ^{90}Sr and the actinides used for all samples are in regular operation at the Board's laboratories and are carried out within a formal agreement with the United Kingdom Accreditation Service (accreditation number 1269).

3 RESULTS AND DISCUSSION

Studies of radionuclide transfer along terrestrial foodchain pathways may provide results in terms of either plant:soil concentration ratios or soil:plant transfer factors (TFs). The former is an operational quantity that is defined by the circumstances prevailing at the time of the experiment. The observed value may be affected by processes such as direct deposition from atmosphere, translocation of directly deposited activity from other plant parts, uptake from soil via roots or the resuspension of soil on to foliage. Soil:plant TFs relate specifically to the uptake of activity from soil into the plant. As noted earlier, direct deposition at the field site has been shown to be largely absent (Green et al, 1995a) and would not occur at the lysimeter facility. In addition, all edible plant parts were thoroughly washed prior to analysis. Any adhering soil should then have been removed. This is important because soil contamination can have a very marked effect on the concentration measured in vegetation, especially for isotopes of plutonium and americium that are not readily taken up via roots (Zach and Mayo, 1984; Nisbet and Shaw, 1994).

Under the conditions used in this study, the derived concentration ratios may therefore be regarded as soil:plant transfer factors. However, for perennial plants such as trees and bushes, there is the possibility that radionuclides may be taken up from the soil but stored in plant parts such as branches. There is then the potential for translocation of activity to fruits in a later season. Published data relating to the uptake of radionuclides from soil into fruit and the distribution of radionuclides between the different parts of perennial fruit crops have been reviewed (Carini, 2001). The results indicated that translocation within species such as trees was important when the original route of contamination was via a single episode of direct deposition from atmosphere. Field measurements made after the Chernobyl accident illustrated this point. However, no long-term studies were reported where uptake from soil via roots was the only route of transfer of radionuclides into the plants. A comprehensive evaluation of the distribution of radionuclides between plant parts and the potential for translocation to fruit has not been undertaken as part of this study. Instead, the approach adopted was to use the observed temporal changes in activity concentrations in fruit together with the derived soil:crop TF values to determine whether storage and translocation were important from the radiological assessment point of view.

Plant:soil concentration ratios and soil:plant transfer factors can be expressed in terms of either the fresh mass or the dry mass of plant, together with the dry mass of soil. Values in terms of fresh mass are used in radiological assessment models because the objective is to estimate doses due to ingestion and consumption rates for foodstuffs are expressed on a fresh mass basis. Values in terms of dry mass are commonly used in comparative studies such as that operated by the International Union of Radioecologists (IUR) (IUR, 1989). Such an approach is considered useful since variability between samples of the same crop due to differences in moisture content are removed and the effects of varying local conditions at the time of collection or during the growth period are reduced. In this paper, TFs are given in terms of both the fresh and dry mass of fruit and both forms have been used in the interpretation and use of data.

IUR recommends that concentration ratios for crops should be based on a uniform distribution of activity in soil to a depth of 150 mm (IUR, 1989). Although soil samples within the field site used in this study were taken to only 100 mm, at any given point the soil was assumed to be well-mixed to a depth of 300 mm. This approach was considered reasonable because prior to the cultivation of any crops the plot had been ploughed. In addition, since that time the soil had frequently been dug over manually to a depth of 300 mm using a spade of that depth. As a result of continual cultivation, the soil was not sufficiently consolidated to permit any determination of the distribution of activity with depth.

3.1 Changes in transfer with time

3.1.1 Fruit grown on reclaimed land

The observed concentrations in apples grown on reclaimed land, together with the associated data for soils, are shown in Table 1. The corresponding data for blackcurrants, gooseberries and strawberries are shown in Tables 2, 3 and 4 respectively. Soil:crop TFs have been derived from these data and these are shown in Tables 5, 6, 7 and 8 for apples, blackcurrants, gooseberries and strawberries respectively.

All of the data available from the field site for the period 1993-2002 are given in these tables. Data for all crops are not however available for each year, largely because of occasional crop failures but also because of losses of samples during analysis. Samples of soil were not collected each time that fruit was harvested. In some cases, notably blackcurrants in 1994, more than one harvest was made in a single year. In such cases, the data for each harvest have been taken with a single set of samples of soil for that year. In other cases, the determination of ^{137}Cs in soil gave values similar to previous years, and so the more labour intensive analyses for the other radionuclides of interest were not carried out. Instead, TF values were derived using the data for the most recent similar year. In the case of apples, a single set of soil samples was collected from within the polythene tunnel. The resultant analytical data were applied to both of the varieties grown.

There were some years for which no samples of soil were collected, generally in the case of apples. In such cases TF values were derived using typical data from an earlier year. This approximation was considered reasonable given that, apart from irrigation and surface applications of fertiliser, the soil around perennial plants remained largely undisturbed.

For apples, the mean concentrations of all of the radionuclides of interest in the underlying soil varied by no more than a factor of 4 across the 9-year period (Table 1). As might be expected, there were no consistent trends with time, and the variability reflects the continued non-uniformity across the site even after frequent cultivation. To elicit any temporal changes, radionuclide concentrations in fruit and the corresponding TF values are best considered on a dry mass basis so as to avoid any effects of differences in moisture content. Concentrations in fruit were in most cases within a factor of 5 across the 9-year period. For TF values, the variability between years was generally within a factor of around 5. For comparison, the variation between years for the same variety of annual crops at this site was typically 2-3 (Green et al, 1995a).

There were no indications of systematic increases in TF values over the 9-year period. There were a few exceptional values, notably that for the transfer of ^{241}Am to Egremont apples in 1997. The corresponding data for ^{239,240}Pu were within the range of values for other years, and so it is unlikely that the unusual value for ^{241}Am was due to small amounts of soil in the sample of fruit. This result was not considered reliable and has not been considered further.

Similar trends were observed for blackcurrants (Tables 2 and 6) and gooseberries (Tables 3 and 7). In 1994, it was possible to make three harvests of blackcurrants between July and September, and it is worth noting that the variations in concentrations and TF values were up to an order of magnitude when expressed on a dry mass basis. Two harvests of gooseberries were made over the same period, and for the actinides the TF values differed considerably. This could be due to small amounts of soil remaining with one sample of crops. This would have a noticeable effect on the very low TF values observed for these radionuclides. Overall however, for both types of fruit and all of the radionuclides studied there was no evidence of a systematic increase in the apparent TF value with time.

The results for strawberries also gave no indication of systematic increases in transfer with time (Tables 4 and 8). However, over the timescales of this study some of the data derived from plants propagated via runners from the originals established in 1993. For this reason, strawberries have not therefore been considered as a perennial crop in the context of the present study.

Some storage of radionuclides in parts of perennial plants such as branches would be expected, but overall the results for fruit trees and bushes suggest that this process does not give rise to increased concentrations in fruit in later years. Thus for radiological assessment purposes, it would be reasonable to assume that when root uptake is the only process affecting transfer to fruit, TF values derived on the basis of relatively short - term experiments would not underestimate transfer in the longer term.

3.1.2 Fruit grown in lysimeters

The available data on activity concentrations in soils and fruit from the lysimeter study are given in Table 9, and the derived soil:crop TF values are given in Table 10. As noted earlier, the long term study was confined to apples. All of the data available for strawberries have been reported previously (Green et al, 1997) and are not reproduced here. There were frequent crop failures for the apples grown in the lysimeter facility. In addition, crop yields were extremely variable and often differed between varieties in a given year. Consequently, useful data were produced for only a few years. The soils in the lysimeters had been well-mixed several times since they had first been contaminated in 1983. As part of this study, activity concentrations in soil were determined only in 1995. However, in preparation for a separate study some further analyses were carried out in 2004. The results were in reasonable general agreement with the values for 1995. The TF values given in Table 10 have therefore been based on the activity concentrations in soils as determined in 1995.

The data for any given soil-variety combination are limited. In terms of concentrations in fruit, there were generally no systematic trends over time

(Table 9). There were some occasions, notably with Fiesta apples grown in sand and loam soils in 1995, where concentrations of actinides in the fruit were much higher than in other years. Despite the steps taken to wash fruit prior to analysis, it is likely that these are due to small amounts of soil adhering to the fruit, since higher values were observed for both $^{239,240}Pu$ and ^{241}Am. No systematic temporal changes were observed in TF values for any of the radionuclides studied (Table 10). The longest sequence of data, a 6-year period, was for Fiesta apples grown in peat soil. When expressed on a dry mass basis, the TF values varied by around a factor of 4 or less, which is comparable with the results from the field study (Section 3.1.1). Overall therefore, there was no evidence of systematic increases in transfer over time, in agreement with the findings from the field study.

3.2 Parameter values for radiological assessments

Tables 5,6,7,8 and 10 give the default soil:crop TF values currently used in FARMLAND. These are intended to be applied to all domestically-produced fruit, but are based on data for apples (Mayall, 1995). This is because apples are considered to be the dominant contributor to intakes of domestically produced fruit in the UK.

Table 10 indicates that, for all of the radionuclides considered, TF values for the peat soil differed from those for sand and loam, which were reasonably consistent with each other. For ^{137}Cs, values were higher for peat soil, whereas for the other radionuclides they were generally lower. These trends were observed in the earlier study (Green et al*,* 1997) and were expected on the basis of results for other crops grown in the lysimeter facility (Nisbet and Shaw, 1994). Fruit is not generally cultivated on this type of soil in the UK, and so for the purposes of this evaluation values for peat have not been taken into account. Thus the mean TF values given in Table 10 relate to sand and loam soils only. In addition, some of the data for actinide uptake from sand and loam soils were considered unreliable because of the possible presence of soil in the samples of crops (Section 3.1.2). These data were not therefore considered in the derivation of mean values in Table 10.

For ^{137}Cs, the mean soil:plant TF value for sand and loam soils, expressed in terms of the fresh mass of the crop, was $2.9\ 10^{-3}$. This was a factor of almost 3 higher than the mean value obtained in the field study but much less than the current default of $20\ 10^{-3}$ assumed in the dynamic foodchain model FARMLAND. On the basis of the earlier results from this study, a default value of $5\ 10^{-3}$ was suggested (Green et al*,* 1997). Given the range of values in Tables 5 and 10 this would seem a suitably cautious value for general assessments. The mean TF value for ^{90}Sr, again based on the fresh mass of crop, was $1.8\ 10^{-2}$, close to the default model value of $4\ 10^{-2}$. On the basis of this and the range of values in Tables 5 and 10 it would seem reasonable to retain the current default value.

The TF values for $^{239,240}Pu$ and ^{241}Am observed in sand and loam soils were very much less than those obtained in the field study, which in turn were about an order of magnitude lower than the default values assumed in FARMLAND. Given the large range involved, it would be imprudent to base predictive models purely on the results from the lysimeter facility. In the earlier study, a value of 5 10^{-5} was put forward as a suitably cautious default for predicting the transfer of Pu and Am to fruit. The larger amount of data now available supports this view.

For the reclaimed land site, the mean value derived for apples was in some cases much lower than those derived for other fruit crops (Tables 5,6,7 and 8). The implications for radiological assessments have been set out in an earlier publication, making use of data for a wider range of crops grown between 1993 and 1995 (Green et al, 1997). Briefly however, for general assessments, data based on apples should not underestimate radiological impact unduly because of the major contribution from apples to the consumption of domestically produced fruit in the UK. For more rigorous assessments however it may be necessary to use more specific data for particular species of fruit. There was however no evidence of systematic differences in transfer between the two varieties of apple in either the field or the lysimeter studies.

4 CONCLUSIONS

Long term studies of the transfer of ^{137}Cs, ^{90}Sr, $^{239,240}Pu$ and ^{241}Am from soil to fruit crops have been carried out using a field site that had been reclaimed from the sea together with a lysimeter facility containing a diverse range of soil types. A small range of perennial crops was grown at the field site, including two varieties of apple. The same two varieties were grown in the lysimeter facility. No systematic increases in the soil:plant TF values were observed. On this basis, it is reasonable to assume that, for radiological assessment purposes, long term storage in plant parts and subsequent translocation to fruit is not an important contributor to activity concentrations in fruit if root uptake is the only mechanism by which the plant is contaminated. In such cases, uptake in the longer term could be reasonably predicted on the basis of short term experiments. In contrast, information published elsewhere indicates that if the plant is originally contaminated via direct deposition, then storage and translocation are likely to be important in terms of transfer to the crop in later years (Carini 2001).

The data now available support the suggestions from an earlier study regarding the values for soil:crop TFs currently assumed in the dynamic foodchain model FARMLAND. It would be reasonable to retain the default value presently assumed for ^{90}Sr. However, consideration should be given to the adoption of lower values for ^{137}Cs and isotopes of plutonium and americium.

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7 TABLES

TABLE 1 Activity concentrations of radionuclides in apples and associated soil from the reclaimed land site, Bq kg^{-1}[(a)]

Variety	Date	^{137}Cs			^{90}Sr			^{239,240}Pu			^{241}Am		
		Fresh	Dry	Soil[c]	Fresh	Dry	Soil[c]	Fresh	Dry	Soil[c]	Fresh	Dry	Soil[c]
Egremont	Sep 1993	0.25	1.29	259	0.05	0.26	5.44	0.0012	0.0060	26.4	0.0015	0.0078	52.3
Egremont	Sep 1994	0.42	2.52	470	0.15	0.88	9.25	0.0010	0.0057	61.3	0.0022	0.0133	113
Egremont	Sep 1995[d]	0.28	1.99	259	0.04	0.28	5.44	0.0009	0.0064	26.4	0.0011	0.0076	52.3
Egremont	Oct 1997[e]	0.26	1.58	240	0.14	0.83	5.44	0.0019	0.0114	26.4	0.0191	0.117	52.3
Egremont	Aug 1998[d]	0.23	1.68	259	SL	SL	5.44	0.0003	0.0024	26.4	0.0007	0.0048	52.3
Egremont	Sep 1999	0.15	1.04	205	0.33	2.36	3.65	0.0007	0.0052	29.7	0.0008	0.0059	23.9
Egremont	Sep 2000	0.16	1.23	192	0.10	0.76	4.23	0.0004	0.0033	31.1	0.0008	0.0059	48.4
Fiesta	Sep 1993	0.11	0.90	259	0.04	0.28	5.44	0.0022	0.0180	26.4	0.0026	0.0207	52.3
Fiesta	Sep 1994	0.11	1.03	228	0.07	0.64	3.78	0.0007	0.0061	27.1	0.0010	0.0089	49.1
Fiesta	Sep 1995[d]	0.50	6.43	259	0.08	1.03	5.44	0.0004	0.0046	26.4	0.0008	0.0103	52.3
Fiesta	Oct 1997[d]	0.41	3.54	240	0.09	0.76	5.44	0.0005	0.0044	26.4	0.0021	0.0181	55.5
Fiesta	Aug 1998[d]	0.17	1.66	259	0.06	0.62	5.44	0.0007	0.0064	26.4	0.0015	0.0143	52.3
Fiesta	Sep 1999	0.18	1.66	205	0.08	0.75	3.65	0.0003	0.0026	29.7	0.0003	0.0031	23.9
Fiesta	Sep 2000	0.28	2.66	192	0.11	1.01	4.23	0.0003	0.0029	31.1	0.0005	0.0044	48.4
Apples[b]	Aug 2001	0.36	2.52	121	0.19	1.32	9.79	0.0004	0.0028	35.1	0.0009	0.0064	55.3
Apples[b]	Oct 2002	0.33	2.37	181	0.15	1.08	3.78	0.0005	0.0038	26.3	0.0006	0.0044	45.7

[a] uncertainties in the activity concentrations in dry vegetation were estimated from the bounds of the 95% confidence interval and were typically 10% for ^{137}Cs, 16% for ^{90}Sr and 20% for ^{239,240}Pu and ^{241}Am. Values in terms of fresh mass were derived using the measured values in terms of dry mass and the observed fresh:dry mass ratios. Values for soils represent the mean of several individual analyses (see text).

[b] the different varieties could not be distinguished

[c] expressed in terms of dry mass

d no soil samples collected, data relate to samples taken in 1993

[e] ^{137}Cs values in soil close to those for 1993; values for other radionuclides based on those for 1993

SL sample lost

TABLE 2 Activity concentrations of radionuclides in blackcurrants and associated soils at the reclaimed land site, Bq kg^{-1} [(a)]

Date	^{137}Cs			^{90}Sr			$^{239,240}Pu$			^{241}Am		
	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]
Jul 1993	0.87	7.00	229	0.12	0.98	1.57	0.010	0.083	23.5	0.015	0.121	38.2
Jul 1994[c]	0.48	3.12	178	0.19	1.20	2.65	0.003	0.018	20.1	0.004	0.024	33.6
Sep 1994[c]	0.84	6.04	178	0.68	4.87	2.65	0.005	0.034	20.1	0.006	0.043	33.6
Sep 1994[c]	0.39	2.73	178	0.07	0.51	2.65	0.003	0.023	20.1	0.005	0.037	33.6
Jul 1997	0.47	3.11	91	0.24	1.60	3.04	0.003	0.017	19.4	0.005	0.032	15.9
Jul 1998	0.37	2.60	140	SL	SL	3.04	0.001	0.010	19.4	0.002	0.014	29.7
Aug 2001	0.29	1.80	138	0.37	2.28	5.22	0.001	0.007	18.0	0.002	0.013	26.9
Aug 2002	0.40	1.95	122	0.69	3.32	2.59	0.004	0.018	14.7	0.003	0.014	23.6

[a] uncertainties in the activity concentrations in dry vegetation were estimated from the bounds of the 95% confidence interval and were typically 10% for ^{137}Cs, 16% for ^{90}Sr and 20% for $^{239,240}Pu$ and ^{241}Am. Values in terms of fresh mass were derived using the measured values in terms of dry mass and the observed fresh:dry mass ratios. Values for soils represent the mean of several individual analyses (see text).

[b] expressed in terms of dry mass

[c] a single set of soil samples was taken for use with all harvests in 1994

SL sample lost

TABLE 3 Activity concentrations of radionuclides in gooseberries and associated soils at the reclaimed land site, Bq kg^{-1}[(a)]

Date	^{137}Cs			^{90}Sr			$^{239,240}Pu$			^{241}Am		
	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]
Jun 1993	0.13	1.44	231	0.21	2.29	2.48	0.0019	0.021	38.1	0.0021	0.023	46.1
Jul 1994[c]	0.16	1.97	250	0.06	0.81	3.15	0.0009	0.012	30.3	0.0018	0.022	47.2
Sep 1994[c]	0.21	3.57	250	0.08	1.28	3.15	0.0038	0.064	30.3	0.0053	0.089	47.2
Jul 1997	0.13	1.33	125	0.14	1.39	3.14	0.0010	0.009	25.3	0.0039	0.039	32.0
Jul 1998	0.17	1.64	157	SL	SL	3.14	0.0003	0.003	25.3	0.0005	0.005	32.0
Jun 1999	0.18	2.18	149	SL	SL	1.64	SL	SL	36.6	SL	SL	27.3
Aug 2000	<0.19	<1.4	132	0.39	2.85	2.81	0.0004	0.003	16.8	0.0008	0.006	24.7
Aug 2001	0.16	1.52	159	0.13	1.23	5.47	0.0008	0.008	22.2	0.0006	0.006	33.1
Aug 2002	0.14	1.21	107	0.19	1.65	3.12	0.0005	0.004	13.9	0.0006	0.005	23.1

[a] uncertainties in the activity concentrations in dry vegetation were estimated from the bounds of the 95% confidence interval and were typically 10% for ^{137}Cs, 16% for ^{90}Sr and 20% for $^{239,240}Pu$ and ^{241}Am. Values in terms of fresh mass were derived using the measured values in terms of dry mass and the observed fresh:dry mass ratios. Values for soils represent the mean of several individual analyses (see text).

[b] expressed in terms of dry mass

[c] a single set of soil samples was taken for use with all harvests in 1994

SL sample lost

TABLE 4 Activity concentrations of radionuclides in strawberries and associated soils at the reclaimed land site, Bq kg^{-1}[(a)]

Date	^{137}Cs			^{90}Sr			$^{239,240}Pu$			^{241}Am		
	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]
Jul 1993	0.45	5.59	197	0.08	1.02	2.79	0.0027	0.034	23.5	0.0032	0.040	36.7
Jul 1994[c]	0.37	5.04	596	0.10	1.40	7.3	0.0060	0.081	69.2	0.0095	0.129	153
Sep 1994[c]	0.31	5.79	596	0.15	2.69	7.3	0.0008	0.012	69.2	0.0016	0.029	153
Aug 1998	0.86	10.1	253	SL	SL	4.2	0.020	0.233	35.3	0.036	0.418	65.0
Jun 1999	0.63	8.53	285	0.13	1.75	3.6	0.027	0.367	22.4	0.043	0.582	53.6
Aug 2000	0.48	8.01	251	0.16	1.73	4.3	0.0047	0.049	38.4	0.008	0.080	62.6
Aug 2002	0.39	3.70	200	0.26	2.43	2.1	0.025	0.233	32.7	0.044	0.419	49.0

[a] uncertainties in the activity concentrations in dry vegetation were estimated from the bounds of the 95% confidence interval and were typically 10% for ^{137}Cs, 16% for ^{90}Sr and 20% for $^{239,240}Pu$ and ^{241}Am. Values in terms of fresh mass were derived using the measured values in terms of dry mass and the observed fresh:dry mass ratios. Values for soils represent the mean of several individual analyses (see text).

[b] expressed in terms of dry mass

[c] a single set of soil samples was taken for use with all harvests in 1994

SL sample lost

TABLE 5 TFs for apples from the reclaimed land site in terms of fresh and dry mass of crop and dry mass of soil

Variety	Date	^{137}Cs		^{90}Sr		$^{239,240}Pu$		^{241}Am	
		Fresh X 10^{-3}	Dry x 10^{-2}	Fresh x 10^{-2}	Dry x 10^{-1}	Fresh X 10^{-5}	Dry x 10^{-4}	Fresh x 10^{-5}	Dry x 10^{-4}
Egremont	Sep 1993	1.0	0.5	0.9	0.5	4.4	2.3	2.9	1.5
Egremont	Sep 1994	0.9	0.5	1.6	1.0	1.6	0.9	2.0	1.2
Egremont	Sep 1995	1.1	0.8	0.7	0.5	3.4	2.4	2.0	1.5
Egremont	Oct 1997	1.1	0.7	2.5	1.5	7.0	4.3	36.4	22.4
Egremont	Aug 1998	0.9	0.7	SL	SL	1.3	0.9	1.3	0.9
Egremont	Sep 1999	0.7	0.5	9.0	6.5	2.5	1.8	3.5	2.5
Egremont	Sep 2000	0.8	0.6	2.3	1.8	1.3	1.0	1.6	1.2
Fiesta	Sep 1993	0.4	0.4	0.6	0.5	8.5	6.8	4.9	4.0
Fiesta	Sep 1994	0.5	0.5	1.8	1.7	2.4	2.3	2.0	1.8
Fiesta	Sep 1995	2.0	2.5	1.5	1.9	1.4	1.7	1.5	2.0
Fiesta	Oct 1997	1.7	1.5	1.6	1.4	1.9	1.6	3.80	3.3
Fiesta	Aug 1998	0.7	0.6	1.2	1.1	2.5	2.4	2.8	2.7
Fiesta	Sep 1999	0.9	0.8	2.2	2.1	0.9	0.9	1.4	1.3
Fiesta	Sep 2000	1.5	1.4	2.5	2.4	1.0	0.9	1.0	0.9
Apples[a]	Aug 2001	3.0	2.1	1.9	1.3	1.1	0.8	1.7	1.2
Apples[a]	Oct 2002	1.8	1.3	4.0	2.9	2.0	1.4	1.3	1.0
Mean (rounded)		1.2	1.0	2.3	1.8	2.7	2.0	4.4 (2.2[b])	3.1 (1.8[b])
FARMLAND default value[c]		20		4		30		80	

[a] the different varieties could not be distinguished.

[b] excluding the value for the Egremont apples from October 1997 (see text).

[c] taken from Mayall, 1995.

SL sample lost.

TABLE 6 TFs for blackcurrants from the reclaimed land site in terms of fresh and dry mass of crop and dry mass of soil

Date	^{137}Cs		^{90}Sr		$^{239,240}Pu$		^{241}Am	
	Fresh x 10^{-3}	Dry X 10^{-2}	Fresh x 10^{-2}	Dry x 10^{-1}	Fresh x 10^{-4}	Dry X 10^{-3}	Fresh x 10^{-4}	Dry x 10^{-3}
Jul 1993	3.8	3.1	7.8	6.3	4.4	3.5	4.0	3.2
Jul 1994	2.7	1.8	7.0	4.5	1.4	0.9	1.1	0.7
Sep 1994	4.7	3.4	25.6	18.4	2.4	1.7	1.8	1.3
Sep 1994	2.2	1.5	2.8	1.9	1.7	1.1	1.6	1.1
Jul 1997	5.2	3.4	8.0	5.3	1.3	0.9	3.1	2.0
Jul 1998	2.7	1.9	SL	SL	0.7	0.5	0.7	0.5
Aug 2001	2.1	1.3	7.1	4.4	0.7	0.4	0.8	0.5
Aug 2002	3.3	1.6	26.5	12.8	2.5	1.2	1.2	0.6
Mean (rounded)	3.3	2.2	12.1	7.7	1.9	1.3	1.8	1.2
FARMLAND default value[a]	20		4		3		8	

SL sample lost

[a] taken from Mayall, 1995.

TABLE 7 TFs for gooseberries from the reclaimed land site in terms of fresh and dry mass of crop and dry mass of soil

Date	^{137}Cs		^{90}Sr		$^{239,240}Pu$		^{241}Am	
	Fresh X 10^{-3}	Dry x 10^{-2}	Fresh x 10^{-2}	Dry x 10^{-1}	Fresh x 10^{-5}	Dry X 10^{-4}	Fresh X 10^{-5}	Dry x 10^{-4}
Jun 1993	0.6	0.6	8.3	9.3	5.0	5.5	4.6	5.1
Jul 1994	0.6	0.8	2.1	2.7	3.0	3.8	3.7	4.7
Sep 1994	0.9	1.4	2.5	4.2	12.7	21.1	11.3	18.9
Jul 1997	1.1	1.1	4.4	4.4	3.7	3.72	12.2	12.1
Jul 1998	1.1	1.0	SL	SL	1.2	1.2	1.6	1.6
Jun 1999	1.2	1.5	SL	SL	SL	SL	SL	SL
Aug 2000	1.4	1.0	14.0	10.1	2.3	1.7	3.2	2.3
Aug 2001	1.0	1.0	2.3	2.2	3.7	3.6	1.7	1.7
Aug 2002	1.3	1.1	6.1	5.3	3.3	2.9	2.7	2.3
Mean	1.0	1.1	5.7	5.5	4.4	5.4	5.1	6.1
FARMLAND default value[a]	20		4		30		80	

SL sample lost

[a] taken from Mayall, 1995.

TABLE 8 TFs for strawberries from the reclaimed land site in terms of fresh and dry mass of crop and dry mass of soil

Date	^{137}Cs		^{90}Sr		$^{239,240}Pu$		^{241}Am	
	Fresh x 10^{-3}	Dry x 10^{-2}	Fresh x 10^{-2}	Dry x 10^{-1}	Fresh x 10^{-4}	Dry x 10^{-3}	Fresh x 10^{-4}	Dry x 10^{-3}
Jul 1993	2.3	2.8	2.9	3.7	1.2	1.4	0.9	1.1
Jul 1994	0.6	0.9	1.4	1.9	0.9	1.2	0.6	0.8
Sep 1994	0.5	1.0	2.0	3.7	0.1	0.2	0.1	0.2
Aug 1998	3.4	4.0	SL	SL	5.6	6.6	5.5	6.4
Jun 1999	2.2	3.0	3.7	4.9	12.2	16.4	8.1	10.9
Aug 2000	1.9	2.0	3.9	4.1	1.2	1.3	1.2	1.3
Aug 2002	2.0	1.8	12.4	11.7	7.5	7.1	9.0	8.6
Mean (rounded)	1.8	2.2	4.4	5.0	4.1	4.9	3.6	4.2
FARMLAND default value[a]	20		4		3		8	

SL sample lost

[a] taken from Mayall, 1995.

TABLE 9 Activity concentrations of radionuclides in apples and associated soils from the lysimeter facility, Bq kg^{-1}

Variety - soil combination	Year	^{137}Cs			^{90}Sr			$^{239,240}Pu$			^{241}Am		
		Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]	Fresh	Dry	Soil[b]
Egremont -sand	1994	2.3	15.2		17	112		0.007	0.048		0.012	0.08	
Egremont - sand	1997	5.03	29.9		13.2	78.2		0.003	0.016		0.009	0.056	
Egremont - loam	1995	0.78	5.1	1320	6.9	44.8	680	0.004	0.029	1280	0.004	0.024	1440
Egremont - peat	1995	55.3	334	3450	3.5	21.2	4800	0.003	0.021	3300	0.003	0.017	4100
Egremont - peat	1997	59.4	286		9.1	43.6		0.004	0.019		0.024	0.116	
Egremont - peat	1998	35.9	198		12.5	68.9		0.003	0.015		0.004	0.021	
Fiesta - sand	1995	1.57	11.7	1050	7.2	53.3	480	0.030	0.19	1100	0.025	0.19	1300
Fiesta - sand	1997	6.6	45.2		7.3	49.9		0.002	0.012		0.004	0.025	
Fiesta - sand	1998	5.4	39.3		7.1	51.7		0.001	0.010		0.002	0.013	
Fiesta - loam	1995	1.7	12.2	1320	9.4	67.8	680	0.02	0.110	1280	0.02	0.14	1440
Fiesta - loam	1998	1.8	14.7		7.7	62.0		0.005	0.037		0.004	0.034	
Fiesta - peat	1993	273	1665		6.9	42.0		0.008	0.049		0.010	0.064	
Fiesta - peat	1995	63.3	547	3450	8.1	70.3	4800	0.002	0.014	3300	0.002	0.021	4100
Fiesta - peat	1997	136	775		9.3	53.0		0.002	0.012		0.004	0.022	
Fiesta - peat	1998	73.7	492		7.7	51.4		0.002	0.011		0.002	0.016	

[a] uncertainties in the measured activity concentrations in dry vegetation were in all cases typically about 5%, based on the bounds of the 95% confidence interval. Values in terms of fresh mass were derived using these data together with the observed fresh:dry mass ratio.

[b] expressed in terms of dry mass, mean of 3 determinations

TABLE 10 TFs for apples from the lysimeter facility in terms of fresh and dry mass of crop and dry mass of soil[a]

Variety - soil combination	Year	^{137}Cs		^{90}Sr		$^{239,240}Pu$		^{241}Am	
		Fresh x 10^{-3}	Dry x 10^{-2}	Fresh x 10^{-2}	Dry x 10^{-1}	Fresh X 10^{-6}	Dry x 10^{-5}	Fresh x 10^{-6}	Dry x 10^{-5}
Egremont - sand	1994	2.2	1.4	3.5	2.3	6.5	4.3	9.5	6.3
Egremont - sand	1997	4.8	2.8	2.7	1.6	2.5	1.5	7.4	4.4
Egremont - loam	1995	0.6	0.4	1.0	0.7	3.4	2.2	2.5	1.7
Egremont - peat	1995	16.0	9.7	0.07	0.04	1.0	0.6	0.7	0.4
Egremont - peat	1997	17.2	8.3	0.19	0.09	1.2	0.6	5.8	2.8
Egremont - peat	1998	10.4	5.7	0.25	0.14	0.8	0.4	0.9	0.5
Fiesta - sand	1995	1.5	1.1	1.5	1.1	23.1	17.3	20.1	15.0
Fiesta - sand	1997	6.3	4.3	1.5	1.0	1.6	1.1	2.9	2.0
Fiesta - sand	1998	5.1	3.7	1.5	1.1	1.2	0.9	1.4	1.0
Fiesta - loam	1995	1.3	0.9	1.4	1.0	12.0	8.6	13.5	9.7
Fiesta - loam	1998	1.4	1.1	1.1	0.9	3.6	2.9	3.0	2.4
Fiesta - peat	1993	79.1	48.2	0.14	0.09	2.4	1.5	2.6	1.6
Fiesta - peat	1995	18.3	15.9	0.17	0.15	0.5	0.4	0.6	0.5
Fiesta - peat	1997	39.4	22.4	0.19	0.11	0.6	0.4	0.9	0.5
Fiesta - peat	1998	21.3	14.3	0.16	0.11	0.5	0.3	0.6	0.4
Mean[b]		2.9	2.0	1.8	1.2	3.1[c]	2.2[c]	4.5[c]	3.0[c]
FARMLAND default value[d]		20		4		300		800	

[a] TF values were based on measured values in soil for 1995 (see text). Both varieties of apple were grown in the same lysimeter.

[b] excluding values for peat soil (see text).

[c] excluding values for Fiesta apples grown in sand and loam for 1995 (see text).

[d] taken from Mayall, 1995.