Abstract. The transfer was investigated of $^{134}$Cs and $^{85}$Sr to a bush plant, blackberry, at short term after an acute release. Results presented of the first experimental year have been produced for validation of SPADE suite of codes. The objective was to improve the assessment of risk from ingestion of fruit contaminated with radionuclides. Blackberry plants were grown in pots filled with a mixture of peat, pumice and compost, and placed under a ventilated tunnel in field. An acute wet deposition was simulated of $^{134}$Cs and $^{85}$Sr in soluble form, onto the above-ground part of the plant or onto the soil surface, at three phenological stages: predormancy, anthesis, and ripening. Leaf to fruit translocation coefficients are one order of magnitude higher for $^{134}$Cs than for $^{85}$Sr. Direct deposition affects fruit activity, accounting for 55% of $^{134}$Cs and 75% of $^{85}$Sr detectable at harvest. Soil to fruit transfer coefficients are approximately one order of magnitude higher for $^{85}$Sr than for $^{134}$Cs.

1. INTRODUCTION

This paper presents results from an experimental study carried out by the Università Cattolica del Sacro Cuore (Italy) for Mouchel Consulting Ltd (UK) and funded by the Food Standards Agency (FSA) (formerly Ministry of Agriculture, Fisheries and Food), Radiological Safety Division (Project No. RP0157). Data have been produced for validation of SPADE suite of codes, to improve the assessment of risk from ingestion of fruit contaminated with radionuclides. The overall objective of the project is to improve understanding of the processes determining the level of radioactivity in fruit plant components, as affected by the time between contamination and harvest.

The radiocontamination of fruits by airborne pathway can result from direct deposition of radionuclides onto the fruit surface, from deposition onto other above ground parts of the plant, or from deposition onto soil. The relative contribution of these processes depends on many variables, such as the kind of radionuclide, the plant species and the plant phenological stage at time of deposition.

Scarce information is available on bushes bearing fruits. Blackberry is a bush plant, widespread in Northern Europe, with a perennial root apparatus and a biannual aerial part. The consumption of blackberry fruit is common particularly in northern countries, and can play a role in the diet of particular groups of population.

The objective of the present experimental work was to assess blackberry fruit contamination following an acute release of $^{134}$Cs and $^{85}$Sr. The main processes investigated were: leaf to fruit translocation and soil to fruit transfer at different phenological stages.

2. MATERIALS AND METHODS

2.1 Agricultural practices

Domesticated blackberry, cultivar Chester, thornless, has been utilized in the present research. 40 two-year-old plants were transplanted between 10$^{th}$ and 13$^{th}$ September 1999 in 20 L pots. The growth medium was a mixture of peat (55% of the total), pumice and compost, used for growing horticultural and vegetable products in greenhouse. Its main physical-chemical characteristics are reported in section 3.2.1.
The pots were placed under two tunnels, covered with PVC from the top down, leaving a margin of 1m from the ground. This minimised loss of radioactivity by rain whilst allowing natural ventilation, thus permitting insect and wind pollination. The plants were arranged in 4 rows under the tunnels. Distances between plants and between rows were 1m and 2m respectively. Plants, that have a semiprostrate growth habit, were supported by strong espaliers, giving to them the shape of a double T.

Plants were irrigated as required by an automatic irrigation system and regularly fertilized and treated with pesticides for disease control.

### 2.2 Experimental design

The experimental design considered blackberry contamination with $^{134}$CsCl and $^{85}$SrCl$_2$ in aqueous solution through two pathways: via soil or via the epigeous part of the plant (Table 1). The scenario of a wet deposition after an acute release was simulated. The application of radionuclides to the soil was identified as “Soil contamination” and that to the above-ground part of the plant as “Foliar contamination”.

Plants were contaminated at three different phenological stages of their growing cycle: pre-dormancy, identified as “Autumn contamination”, anthesis, identified as “First Spring contamination”, beginning of ripening, identified as “Second Spring contamination”. Each kind of contamination was carried out on three replicates.

#### Table 1. Experimental design

<table>
<thead>
<tr>
<th>Contamination</th>
<th>Stage at time of contamination</th>
<th>Contamination pathway</th>
</tr>
</thead>
<tbody>
<tr>
<td>Autumn 13/10/1999</td>
<td>Pre-dormancy</td>
<td>soil foliar</td>
</tr>
<tr>
<td>First Spring 11-15/5/2000</td>
<td>Anthesis</td>
<td>soil foliar</td>
</tr>
<tr>
<td>Second Spring 30/6/2000</td>
<td>Beginning of ripening</td>
<td>foliar</td>
</tr>
</tbody>
</table>

### 2.3 Contamination treatments

#### 2.3.1. Foliar contamination

The aboveground part of each plant was uniformly sprinkled with an aqueous solution containing $^{134}$CsCl and $^{85}$SrCl$_2$. In order to avoid soil direct contamination during sprinkling, the soil surface of each pot was covered with a plastic sheet before treatment.

Intercepted activity was worked out by cutting the above-ground part of 3 plants when dry, and separating it into leaves, canes and fruits, where present, to measure direct contamination.

#### 2.3.2. Soil contamination

The whole soil surface of each pot was moistened with an aqueous solution containing $^{134}$CsCl and $^{85}$SrCl$_2$. After contamination the soil surface was covered with a layer of expanded clay to separate the leaves from the soil and prevent their direct contamination.

### 2.4 Harvests

Ripening of fruits was scalar and lasted approximately one month, from 2 to 24 July. Fruits were picked regularly every two days and kept separately according to plant. The whole above-ground part of the plant was harvested at the end of the fruit season, between 25 and 29 July 2000 and was divided into leaves, canes and roots.

All the samples were weighted as collected and dried at 60°C. Only fruits were frozen. Each sample was minced and homogenized before being analysed by direct gamma spectrometry.
2.5 Gamma analyses

The concentration of $^{134}\text{Cs}$ and $^{85}\text{Sr}$ in each sample was measured by direct gamma spectrometry. The detector employed is a HpGe with an efficiency of 38% and a resolution FWHM of 1.76 keV at 1.33 MeV of the $^{60}\text{Co}$.

Each sample was analysed for a time sufficient to collect at least 4000 net counts in the peaks of interest to reach an error of maximum 3-4% at 95% of confidence. Different counting geometries were employed, depending on the size of the sample analysed.

3. RESULTS AND DISCUSSION

3.1 Foliar contamination

3.1.1 Fruit activity

The processes leading to the contamination of fruit at short term after foliar deposition may be: the translocation from the above-ground part of the plant to fruit and the direct contamination of fruit, when present. In the present work fruit activity is called Translocation Coefficient, even when the contribution of direct deposition to fruit is likely.

Translocation Coefficients ($TC$) were calculated to represent the share of $^{134}\text{Cs}$ or of $^{85}\text{Sr}$ activity ascertained in fruits at ripening, after contamination of the aboveground part of the plant. The unitary $TC$ of the $i$th sample ($TC_i$) is expressed by the ratio:

$$TC_{fr} = \frac{(\text{Bq} \cdot \text{g}^{-1} \text{ f.w. fruit}),}{\text{Bq intercepted} \cdot \text{plant}^{-1}}$$

where $i = 1^{\text{st}}, 2^{\text{nd}}, 3^{\text{rd}}$ replicate.

$TC_{fr}$ are expressed on a fresh weight basis, considering the juicy feature of fruits and their way of consumption. Leaf to fruit $TC$s for $^{134}\text{Cs}$ and $^{85}\text{Sr}$ are reported in Figures 1 and 2.

![Figure 1. Soil and leaf to fruit TCs of $^{134}\text{Cs}$ after deposition at different growing stages](image)

The unitary fruit activity, expressed as $TC_{fr}$, is lowest after deposition in Autumn, both for $^{134}\text{Cs}$, $(3.0 \pm 0.4) \times 10^{-5}$, and for $^{85}\text{Sr}$, $(1.6 \pm 0.2) \times 10^{-6}$. Considering that fifty percent of contaminated leaves are lost from the plant over Winter, the activity in fruit in the following Spring will be the result of the processes of translocation from the remaining originally contaminated leaves and canes to fruits, and
retranslocation from the storage organs, such as canes and roots, where a share of radionuclides had been allocated before leaf abscission.

Fruit activity after deposition at anthesis, 1st Spring, is the result of the process of translocation from leaves, canes and flowers to fruit. It amounts to $(4.2 \pm 0.2)\times 10^{-5}$ for $^{134}$Cs and $(2.8 \pm 0.2)\times 10^{-6}$ for $^{85}$Sr.

The activity of fruit is the highest when deposition occurs at ripening (2nd Spring), being affected by the process of direct deposition to fruit that overlaps that of leaf to fruit translocation. This is particularly manifest for $^{85}$Sr, radionuclide characterized by a very low mobility in the phloematic pathway. $^{85}$Sr activity in fruit after deposition at ripening is $(6.7 \pm 0.7)\times 10^{-5}$, approximately 24 times higher than after deposition at anthesis, while $^{134}$Cs activity, $(1.0 \pm 0.1)\times 10^{-4}$, is only 2.4 times higher than after deposition at anthesis.

![Figure 2. Soil and leaf to fruit TCs of $^{85}$Sr after deposition at different growing stages](image)

Generally speaking, leaf to fruit TCs are one order of magnitude higher for $^{134}$Cs than for $^{85}$Sr, independently of the plant stage at time of foliar deposition. This result was to be expected on the basis of the different mobility of the two radionuclides in the phloem. While Cs is mobile, Sr is transported only to a very small extent in the phloem [1] and tends to remain at the site of application.

### 3.1.2 Activity in the components of the plant

$^{134}$Cs and $^{85}$Sr distribution in the plant components at harvest, after foliar contamination at three phenological stages, is reported in Figure 3. It is expressed as a percentage of intercepted activity.

![Figure 3. Distribution of $^{134}$Cs and $^{85}$Sr in the plant components after foliar deposition](image)

After deposition of radionuclides to the epigeous part of the plant, $^{134}$Cs is translocated from leaves to canes, fruits and roots. $^{85}$Sr translocation also occurs, even if to a very minor extent. Fruit activity of
$^{134}$Cs and $^{85}$Sr after 2nd Spring contamination is mainly ascribable to the direct contamination of this component.

The ability of $^{134}$Cs to translocate from leaves to the other components of the plant results in a $^{134}$Cs activity lower than $^{85}$Sr in leaves, and higher than $^{85}$Sr in fruits, canes and roots. Fruit activity after foliar contamination is higher when deposition occurs at phenological stages closer to ripening, behaviour more evident for $^{134}$Cs than for $^{85}$Sr.

### 3.2 Soil contamination

#### 3.2.1 Fruit activity

Soil to fruit $T_{C_{fw}}$ were calculated as:

$$T_{C_{fw}} = \frac{(\text{Bq} \cdot \text{g}^{-1} \text{ f.w. fruit})}{\text{Bq administered} \cdot \text{plant}^{-1}}$$

and are shown in Figures 1 and 2.

They are significantly higher for $^{85}$Sr than for $^{134}$Cs. More in particular, $^{134}$Cs $T_C$ is $(8.3 \pm 0.9) \times 10^{-7}$ after Autumn contamination and $(6.9 \pm 0.5) \times 10^{-7}$ after Spring contamination, while $^{85}$Sr $T_C$ is approximately one order of magnitude higher both after Autumn contamination, $(1.0 \pm 0.3) \times 10^{-5}$, and after Spring contamination, $(8.2 \pm 1.4) \times 10^{-5}$. $^{85}$Sr:$^{134}$Cs ratio in fruit is 12:1.

The growth substrate used for blackberries is a mixture of peat (55% of the total substrate), pumice and compost. It is a subacid medium (pH in H$_2$O 6.6), rich in organic matter (39.1%) and with a high CEC (33.2 meq/100 g). Its total K and Ca content, 187.5 and 235 ppm respectively, is low. These elements were regularly added to the substrate through fertilizers. The transfer of $^{134}$Cs and $^{85}$Sr from the medium used in the present work to blackberry plants reflects their behaviour in mineral, rather than in organic soils [2].

In order to compare the results from this experimental work with data in the literature, $T_C$ in fruits have been expressed as Bq kg$^{-1}$ f.w. fruit/ Bq kg$^{-1}$ d.w. soil, considering the weight of the growing substrate around 10 kg d.w. pot$^{-1}$. Average $T_C$s of $7.6 \times 10^{-5}$ for $^{134}$Cs and of $9.1 \times 10^{-2}$ for $^{85}$Sr are consistent with data reported for fruits borne by shrubs and herbaceous plants [3]. More in particular they fall in the upper range of shrub values, and are similar to TFs for blackcurrant [4, 5] and raspberry [6] grown on loam or clay loam soil.

$^{134}$Cs and $^{85}$Sr are transferred from soil to fruit to a greater extent when contamination occurs in Autumn than in Spring. More in particular both $^{134}$Cs and $^{85}$Sr transfer is 1.2 times higher in plants contaminated in Autumn, approximately 9 months before, than in Spring. The longer the time the radionuclide remains in soil, the higher root absorption.

#### 3.2.2 Activity in the components of the plant

$^{134}$Cs and $^{85}$Sr distribution in the plant components at harvest, after soil contamination at two phenological stages, is reported in Figure 4, expressed as a percentage of administered activity.

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Figure 4. Distribution of $^{134}$Cs and $^{85}$Sr in the plant components after soil deposition
$^{134}$Cs is translocated from soil to plant to a lower extent than $^{85}$Sr. The difference is more evident for plants contaminated in Autumn, than for those contaminated in Spring.

The activity transferred from soil to plant after Autumn contamination is higher than that transferred after Spring contamination. This trend particularly holds good for $^{85}$Sr, whose activity ascertainable in the plant is 1.7 times higher in those plants contaminated in Autumn than in Spring. The difference is not so striking for $^{134}$Cs.

4. CONCLUSIONS

4.1 Foliar contamination

- After foliar deposition, fruit activity is one order of magnitude higher for $^{134}$Cs than for $^{85}$Sr, independently of the phenological stage at time of deposition. This result was to be expected on the basis of the higher mobility of $^{134}$Cs than $^{85}$Sr in the phloem.
- Fruit interception capability is low as compared with that of leaves, which are the main receptors of radioactivity. However, the natural plant habit and the human management of blackberries favour direct deposition and interception of pollutants by fruit. Direct interception is rather important for those radionuclides, like $^{85}$Sr, which do not easily translocate from leaf to fruit.
- The total activity of the whole plant is similar for $^{134}$Cs and $^{85}$Sr after contamination at each phenological stage.

4.2 Soil contamination

- Fruit activity after soil contamination at both phenological stages is one order of magnitude higher for $^{85}$Sr than for $^{134}$Cs. This is ascribable to the absorption of $^{134}$Cs on clay minerals of soil, that reduces its availability to biological systems.
- Fruit activity increases as time from soil contamination increases.
- Soil to plant transfer increases with time, trend that particularly holds good for $^{85}$Sr.

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References