

Chapter 14

Radionuclides Behavior in Fruit Plants

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Abstract This paper summarizes research carried out on fruits by the Università Cattolica del Sacro Cuore (UCSC) in Piacenza, Italy. Among the fruit crops studied, strawberry, blackberry, grapevine, apple, pear, and olive, research on strawberry and blackberry was funded by the Food Standard Agency (UK). Fruit plants were grown in pots, kept under tunnels or in open field, and contaminated with ^{134}Cs and ^{85}Sr via leaves or via soil. Interception in strawberry plants ranges 39–17 % for ^{134}Cs , from anthesis (April) to predormancy (November). Leaf-to-fruit translocation occurs to a greater extent for ^{134}Cs than for ^{85}Sr . The distribution of contamination in fruit crops is an element-specific process: ^{134}Cs is preferentially allocated to fruits and ^{85}Sr to leaves. However, the activity in leaves is also species-specific: fruit species show different leaf-to-fruit translocation. Results on apple, pear, and grape crops indicate that the highest transfer from leaf to fruit occurs in apple crops. Olive plants also show ^{134}Cs translocation from leaves to trunks. Grapevines grown on mineral soil show a root uptake higher for ^{85}Sr than for ^{134}Cs , while strawberries grown on a peat substrate show a root uptake higher for ^{134}Cs than for ^{85}Sr . Rinsing directly contaminated fruits removes ^{85}Sr (36 %) to a greater degree than ^{134}Cs (24 %). Transfer to olive oil is low. A 57 % of ^{134}Cs is transferred from grapes to white wine.

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Keywords ^{134}Cs • ^{85}Sr • Fruits • Interception • Leaf-to-fruit translocation • Soil-to-fruit transfer • Wine-making • Food processing

14.1 Introduction

Radioactive contamination of the agricultural environment and the transfer of radionuclides through the food chain, as a consequence of a nuclear release, may require measures to protect human health and the environment. Understanding the behaviour of radionuclides in these situations is crucial if these measures are to be effective.

There are relatively few radioecological studies of fruit crops, which is surprising given that they contribute about 8 % of world food production and have a high economic value [1]. The fate of radionuclides in fruit systems is affected by a combination of biological, chemical, and physical processes and is sensitive to the timing of contamination relative to plant growth.

Contamination of fruits following an airborne release can occur from direct deposition of radionuclides onto the fruit surface, from deposition onto other above-ground parts of the plant, or after deposition onto soil. The relative contribution of these processes depends on many variables, such as the radionuclide involved, the plant species, the plant phenological stage at time of deposition, and the soil type. This topic was discussed under the IAEA BIOMASS program [2] and further updated in IAEA TRS 472 [3] and the accompanying TECDOC-1616 [4].

Various projects on fruit production systems have been carried out by the Università Cattolica del Sacro Cuore (UCSC) in Piacenza, Italy. Among the fruit crops studied were: strawberry, blackberry, grapevine, apple, pear, and olive. These studies considered plants grown in pots, kept under tunnels or in open field, and contaminated with ^{134}Cs and ^{85}Sr via aboveground plant parts or via soil. Research on strawberry and blackberry was funded by the Food Standard Agency (UK) and carried out as a collaborative project between Mouchel (UK) and the UCSC [5]. The objective of these projects was to understand the main processes that lead to fruit contamination in the short term after deposition. Processes of interception, leaf-to-fruit translocation and soil-to-fruit transfer are discussed in the following, as well as processing activities associated with fruits after harvest, such as pressing olives to produce oil and grapes for wine-making. Selected results from this research are compared and summarized in this paper.

14.2 Interception

Results are presented on interception by strawberry plants, *Fragaria x ananassa* Duchesne. Strawberry is a herbaceous perennial member of the rose family (Rosaceae). The study used June bearer plants, cultivar Miss, which are

representative of commercial production in Italy. Young strawberry plants are transplanted at the end of July, develop flower buds in the autumn, lose all old leaves at the resumption of growth (in March), flower in the spring, and produce a single crop from early May to June. After harvest, plants are removed and replaced by new plants.

The aboveground part of different sets of strawberry plants was contaminated with ^{134}Cs and ^{85}Sr at various phenological stages: predormancy (November), preanthesis (early April), anthesis (end of April), and beginning of ripening (May). The fresh weight activity (Bq per kg fresh weight), biomass (g dry weight per plant), and leaf area (cm^2 per plant) were measured for each of the four replicates. Intercepted activity (%) was calculated from the ratio of activity deposited on the plant per total activity applied:

$$I(\%) = \frac{\text{Bq in plant component}}{\text{Bq applied} \cdot \text{plant}^{-1}} \times 100$$

Results for ^{134}Cs are reported in Fig. 14.1.

The results show that interception for ^{134}Cs is highest at anthesis (the period of flower development that starts with the opening of the flower buds), 39 %, and

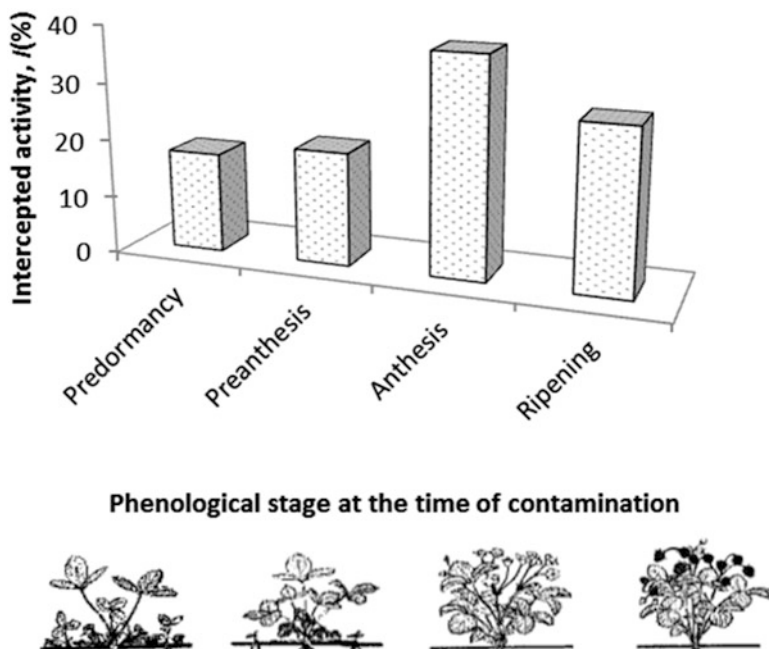


Fig. 14.1 ^{137}Cs intercepted by strawberry plants after deposition at different phenological stages: predormancy, preanthesis, anthesis, and ripening. $I(\%)$ is expressed as $(\text{Bq in plant component per Bq sprinkled} \cdot \text{plant}^{-1}) \cdot 100$

lowest at predormancy, 17 % [6]. Fruits, when present, have a lower interception capability than leaves, even if crop management, aimed at increasing the exposure of fruits to light, favors direct deposition and interception by fruit.

In a number of experimental studies, the interception of wet deposited radioactivity was found to be positively correlated with the leaf area and/or the dry biomass of the crop [7]. However, in the study described here there was no correlation between the interception capacity of strawberry plants and leaf area or biomass. Interception, at the growth stages considered, seems rather to be affected by variables such as the leaf senescence and the posture and physical orientation of leaves. A similar result was obtained contaminating tomato plants at two growing stages, demonstrating that an increase in the leaf surface area at later growing stages does not necessarily imply an increase in the level of interception of wet deposition [8].

Renaud and Gonze [9], studying the ^{134}Cs contamination of orchard fruits in Japan in 2011, observed that some correlation exists between cesium concentration in fruits, at first harvest, and ground surface deposit. They calculated the aggregated transfer factors, T_{ag} , expressed in $\text{Bq} \cdot \text{kg}^{-1}$ of fresh fruit per $\text{Bq} \cdot \text{m}^{-2}$ deposited on the ground surface. They observed that T_{ag} values of apricot samples were apparently higher when collected in low-elevation coastal areas (i.e., in Minamisoma-shi and Soma-shi municipalities, located to the North of the nuclear site, and Mito-shi to the South in Ibaraki Prefecture) than those collected from sites in mountainous areas. They suggested these differences were due to the stage of the vegetative cycle, as flowering would have occurred earlier in coastal regions than in inland elevated areas, resulting in greater transfer [9]. Similarly, the radiocesium activities of the foliar parts of woody species, 5 months after the Fukushima accident, were higher in evergreen species than in deciduous species, because the foliar parts of evergreen species (leaves from previous year) were present at the time of fallout but those of the deciduous species were not [10].

14.3 Leaf-to-Fruit Translocation

Fruits can receive radionuclides via translocation from contaminated aerial parts of plants, the most receptive of which, when present, is foliage. Leaf-to-fruit translocation has been studied in apple, pear, grapes, olives, blackberry, and strawberry plants, after contamination of leaves by wet deposition of ^{134}Cs and ^{85}Sr in open field conditions [6, 11–14]. The activity in fruits (Bq per kg fresh weight) was measured at ripening. Leaf-to-fruit translocation coefficients were calculated to represent the share of ^{134}Cs or ^{85}Sr activity found in fruits at ripening. Translocation coefficients, $\text{TC}(\%)$, are expressed as:

$$\text{TC}(\%) = \frac{\text{Bq} \cdot \text{kg}^{-1} \text{ fresh weight fruit}}{\text{Bq intercepted} \cdot \text{plant}^{-1}} \times 100$$

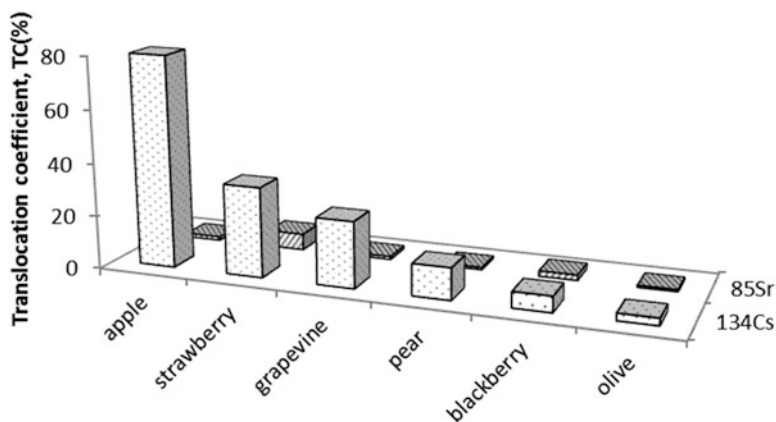


Fig. 14.2 Leaf-to-fruit translocation of ^{134}Cs and ^{85}Sr in various fruit crops. Translocation coefficients, TC(%), are expressed as $(\text{Bq kg}^{-1} \text{ fresh weight fruit per Bq intercepted plant}^{-1}) \cdot 100$

Results, reported in Fig. 14.2, indicate that the leaf-to-fruit TCs are on average one order of magnitude higher for ^{134}Cs than for ^{85}Sr in all fruit crops studied. This result is expected on the basis of greater foliar absorption of Cs^+ than Sr^{2+} [15] and greater mobility, after absorption, in the phloem of monovalent ions than divalent ones [16, 17]. This makes the fruits sinks for cesium.

Differences in the process of foliar absorption and leaf-to-fruit translocation also exist between fruit species. Our results showed TCs for ^{134}Cs declined in the following order: apple trees, strawberry, grapevine, pear, blackberry, and olive plants (Fig. 14.2). A different trend was observed after the Chernobyl accident, in 1986, when Baldini et al. [18] found greater TCs for grapevine and peach than for apple and pear, and ascribed the differences to the more active metabolism of grapevine and peach.

However leaf-to-fruit translocation is also highly dependent on the time at which contamination occurs during the growth period of crops [19] and this can explain the results under different experimental conditions. In this regard, plants of strawberry and blackberry were contaminated with ^{134}Cs and ^{85}Sr via leaves at different growing stages: predormancy (November for strawberry and October for blackberry), anthesis (April for strawberry and May for blackberry), and beginning of ripening (May for strawberry and June for blackberry) [5, 6, 14, 20, 21]. Ripe fruits were picked and analyzed.

The growth cycle of strawberry plants was described earlier and a short description of blackberry plants is given in the following.

Blackberry is a bush plant, widespread in Northern Europe, with a perennial root apparatus and a biannual aerial part: the canes dry after having borne fruits. Blossoming is scalar, lasting up to 5–6 weeks. As a consequence, ripening is also very prolonged, from mid-July onward. The consumption of blackberry fruit is common particularly in northern countries, and can play a role in the diet of particular population groups.

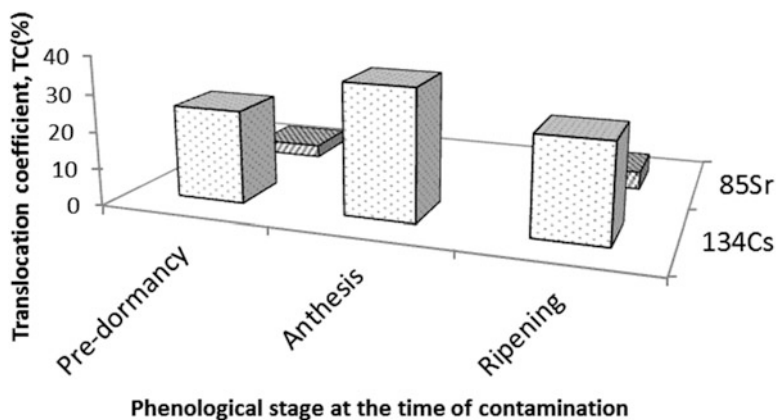


Fig. 14.3 Leaf-to-fruit translocation in strawberry: ¹³⁴Cs and ⁸⁵Sr activity translocated in fruit after leaf deposition at different phenological stages: predormancy, anthesis, and ripening. Translocation coefficients, TC(%), are expressed as (Bq kg⁻¹ fresh weight fruit per Bq intercepted plant⁻¹) · 100

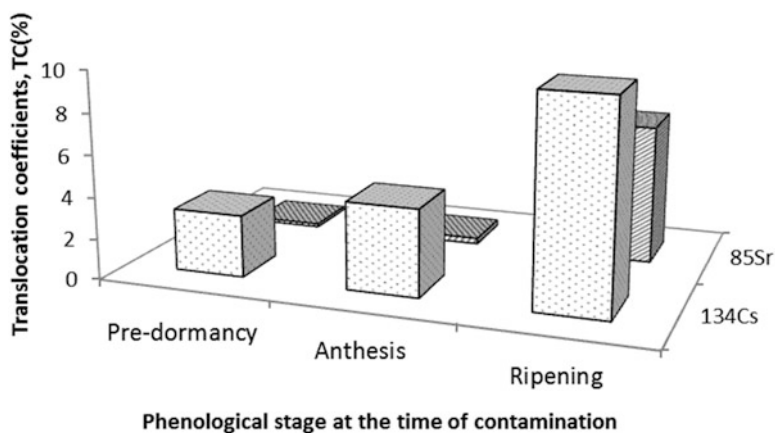


Fig. 14.4 Leaf-to-fruit translocation in blackberry: ¹³⁴Cs and ⁸⁵Sr activity translocated in fruit after leaf deposition at different phenological stages: predormancy, anthesis, and ripening. Translocation coefficients, TC(%), are expressed as (Bq kg⁻¹ fresh weight fruit per Bq intercepted plant⁻¹) · 100

The activity (Bq per kg fresh weight) of fruits was determined and TCs(%) calculated; results are shown in Figs. 14.3 and 14.4 for strawberry and blackberry, respectively. The objective of this study was to show the dependence of plant contamination on the time of the year when the deposition occurs, referred to as seasonality by Aarkrog [22]. In strawberry, leaf-to-fruit translocation is greatest at

anthesis, following the pattern observed for interception (Figs. 14.1 and 14.3). This result supports the hypothesis that interception is also affected by leaf age and by the metabolic activity of the plant. Increasing leaf age seems to reduce the absorption of radionuclides [23, 24].

When deposition occurs at predormancy, strawberry plants present well-developed leaves to aerial deposition, even if some are aging at this stage of the growing cycle. The plants have also developed flower buds for the following spring. A proportion of the radionuclides intercepted by, and absorbed into the leaves, is remobilized and translocated to the storage organs (roots and crowns) before leaf drop. Remobilization of ^{134}Cs is presumably higher than that of ^{85}Sr , given its higher mobility in the phloem. A few months later, the activity in ripe fruits is one order of magnitude greater for ^{134}Cs than for ^{85}Sr , an indication of the greater translocation of ^{134}Cs than ^{85}Sr from roots and crowns to fruits (Fig. 14.3).

In blackberry plants, the leaf-to-fruit translocation is greatest when foliar contamination occurs at ripening, and is one order of magnitude higher for ^{134}Cs than for ^{85}Sr (Fig. 14.4). When blackberry plant contamination occurs in Autumn, at predormancy, the plant system loses 60 % of the intercepted activity through dead leaves during Winter. A remaining 20 % is released into the soil and the environment. Fruit activity at harvest will be the result of retranslocation from roots and shoots (the storage organs) toward the fruits.

Recent observations after the Fukushima accident provide evidence that translocation to fruit does occur from leaf and bark, the amount of translocation differs between plant types, and that translocation is very sensitive to plant growth stage at the time of deposition [9, 25–27]. Another process that can contribute to fruit activity, ascertained by various authors after the Fukushima accident, is the secondary contamination, resulting from weathering, by the action of wind and rain [10, 27–30 cited by 28].

14.4 Soil-to-Fruit Transfer

Soil-to-fruit transfer was studied for grapevines, blackberry, and strawberry plants grown in pots [5, 11, 14, 21]. The crops were grown under different conditions using soils most suited to their cultivation. Data are not therefore comparable and are discussed separately as follows.

Root uptake for blackberry and strawberry plants was studied at two phenological stages: predormancy and anthesis. Results are presented as transfer factors, expressed as in IAEA [4]:

$$F_v = \frac{\text{Bq} \cdot \text{kg}^{-1} \text{ fresh weight fruit}}{\text{Bq} \cdot \text{kg}^{-1} \text{ dry soil}}$$

14.4.1 Grapevines

Two-year-old grapevines, variety Pinot Blanc, were grown in pots of 10 L capacity and kept in open field conditions. Pots were filled with a substrate of mineral soil (70 %) and sand (30 %). The mineral soil is moderately acidic (pH in H₂O 5.7), with low organic matter content (OM = 1.6 %) and low cation exchange capacity (CEC = 13.2 cmol₍₊₎/kg). The substrate of each pot was contaminated by moistening the surface with 250 mL of an aqueous solution containing 5305 kBq of ¹³⁴Cs and 2063 kBq of ⁸⁵Sr per pot after the fruit setting, at the beginning of July.

The objective of this study was to assess the transfer of ¹³⁴Cs and ⁸⁵Sr to fruit and other parts of the vine. At ripening, 60 days after soil contamination, F_v from soil to the whole plant was greater for ⁸⁵Sr than for ¹³⁴Cs, but while ⁸⁵Sr concentrates mainly in leaves and shoots, ¹³⁴Cs is redistributed throughout the plant. As a result, ¹³⁴Cs and ⁸⁵Sr F_v (mean \pm standard error) in grapes are similar: $(8.0 \pm 0.9) \cdot 10^{-2}$ and $(6.6 \pm 1.2) \cdot 10^{-2}$, respectively [12].

14.4.2 Blackberry Plants

Two-year-old blackberry plants, cultivar Chester Thornless, were grown in 20 L capacity pots, filled with a mixture of peat (55 % of the total substrate), pumice, and compost. This is a medium with pH in H₂O 6.6, rich in organic matter (39.1 %), and with a high CEC (33.2 cmol₍₊₎/kg). Different sets of plants were contaminated in Autumn, at predormancy (October), and in Spring, at the beginning of anthesis (May). The soil surface was moistened with 800 mL of an aqueous solution containing 298 kBq of ¹³⁴Cs and 1921 kBq of ⁸⁵Sr per pot. The experiment lasted for 2 years following the growth cycle of this crop. The radionuclide content of fruit was determined at ripening, 276 or 74 days, respectively, after soil contamination.

The F_v are reported in Fig. 14.5. The F_v for ⁸⁵Sr is one order of magnitude greater than for ¹³⁴Cs and reflects the behavior of these radionuclides in mineral rather than organic soils [31]. The ratio of ⁸⁵Sr to ¹³⁴Cs in fruit was 12:1.

As for the phenological stages, a significant difference is shown ($p < 0.05$) for ⁸⁵Sr F_v mean and standard error: $(7.9 \pm 0.2) \cdot 10^{-1}$ at predormancy and $(5.9 \pm 0.2) \cdot 10^{-1}$ at anthesis; no significant difference is shown for ¹³⁴Cs: $(7.7 \pm 0.2) \cdot 10^{-2}$, at both plant stages.

14.4.3 Strawberry

Strawberry plants, cultivar MISS, were grown in 4.5 L capacity pots filled with peat, the normal substrate under horticultural growing conditions. Peat characteristics

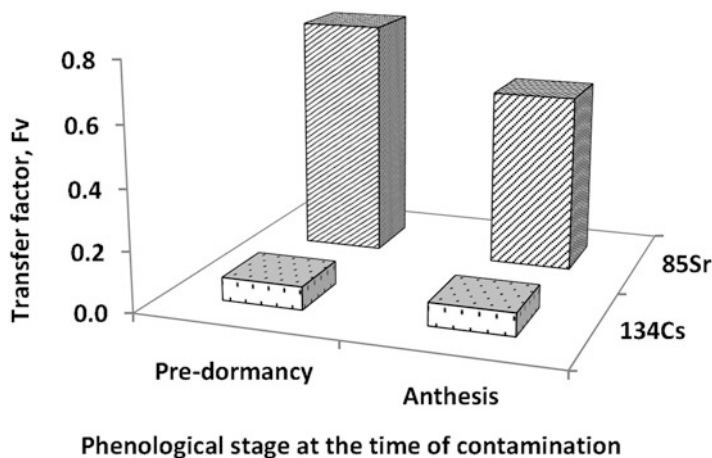


Fig. 14.5 Soil-to-fruit transfer factors F_v of ^{134}Cs and ^{85}Sr to blackberry following soil contamination at two phenological stages: predormancy and anthesis. F_v are expressed as (Bq kg^{-1} fresh weight fruit per Bq kg^{-1} dry soil)

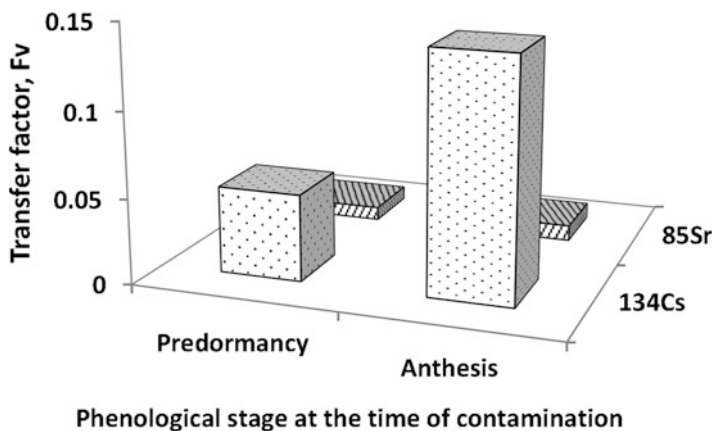


Fig. 14.6 Soil-to-fruit transfer factors F_v of ^{134}Cs and ^{85}Sr to strawberry following soil contamination at two phenological stages: predormancy and anthesis. F_v are expressed as (Bq kg^{-1} fresh weight fruit per Bq kg^{-1} dry soil)

were: a moderate acid pH (5.7), high OM (48.1 %), and high CEC ($111.5 \text{ cmol}_{(+)}/\text{kg}$). The soil surface of each pot was moistened with 250 mL of an aqueous solution containing 74.8 kBq of ^{134}Cs and 91.6 kBq of ^{85}Sr at two phenological stages: predormancy (November) and anthesis (April). The experiment lasted for 3 years.

The radionuclide content of fruit was determined at ripening, 200 and 70 days respectively, after soil contamination. F_v are reported in Fig. 14.6 and show greater root uptake for ^{134}Cs , $(5.1 \pm 0.3) \cdot 10^{-2}$ at predormancy and $(1.4 \pm 0.1) \cdot 10^{-1}$ at

anthesis, than for ^{85}Sr , $(7.8 \pm 0.9) \cdot 10^{-3}$ and $(9.2 \pm 1.6) \cdot 10^{-3}$, respectively. The ratio of ^{134}Cs to ^{85}Sr in fruit was 15:1.

The high organic matter content of the peat substrate, responsible for a large part of the CEC, reduces ^{134}Cs fixation on clay minerals, leaving it more available for root uptake, as highlighted by Nisbet and Shaw [31]. In contrast, it has been demonstrated that the organic matter of peat may form complex compounds with ^{85}Sr , reducing its availability to plants.

Differences in transfer to fruit after soil contamination at different phenological stages is apparent only for ^{134}Cs : F_v , are three times higher after contamination at anthesis than at predormancy (Fig. 14.6), the opposite was observed for blackberry fruits (Fig. 14.5).

Results on soil-to-plant transfer for blackberry and strawberry plants highlight time-dependent changes of root uptake following acute soil deposition during plant growth, as discussed by Choi [32]. These changes are more significant for an annual plant like strawberry, than for a perennial plant (with a perennial root apparatus) like blackberry and are more apparent for those radionuclides with the greatest transfer: ^{134}Cs on peat and ^{85}Sr on mineral soil.

14.5 Food Processing

Food processing losses can vary considerably depending on the type of radionuclide and between processes at the industrial and domestic scales. Rinsing directly contaminated apples (Golden Delicious) and grapes (Chardonnay) with tap water, simulating common domestic practice before consumption, removes ^{85}Sr to a greater degree than ^{134}Cs [13]. The processing factor P_f [3], called food processing retention factor, f_{fp} , in ICRU 65 [33], is the ratio of the radionuclide activity concentrations in the food after and before processing:

$$P_f = \frac{\text{Bq} \cdot \text{kg}^{-1} \text{ processed food}}{\text{Bq} \cdot \text{kg}^{-1} \text{ raw food}}$$

In our observations, it corresponds to 0.7 for ^{85}Sr and 0.8 for ^{134}Cs .

14.5.1 Wine-Making

A study of Pinot Blanc grapes, contaminated following an application of ^{134}Cs to leaves at the beginning of ripening, replicated the wine-making process on a small scale under laboratory conditions. Grapes were picked at the ripening stage, 35 days after contamination, analyzed for ^{134}Cs and squeezed to separate skin, pulp, and seeds from must (the juice extracted from grapes). The processing factor, P_f , for must was 0.94.

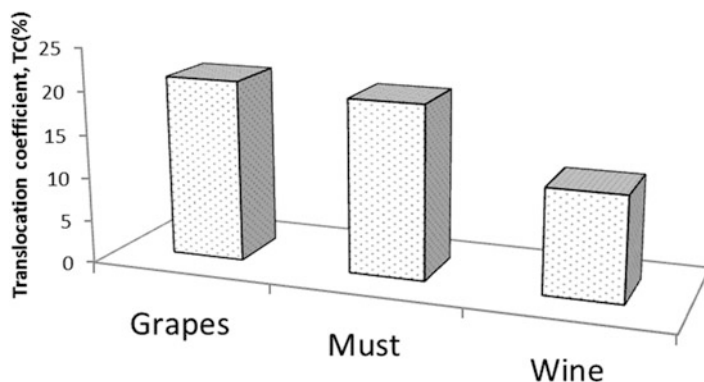
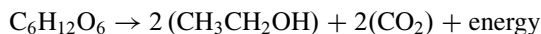


Fig. 14.7 Translocation coefficients, TC(%), from leaf to grapes, must, and wine for ^{134}Cs , expressed as $(\text{Bq kg}^{-1} \text{ fresh weight per Bq intercepted plant}^{-1}) \cdot 100$

Must was then inoculated (under controlled conditions) with $2.8 \cdot 10^5$ cells of the yeast *Saccaromyces cerevisiae* $\cdot \text{mL}^{-1}$ to begin the fermentation process. The must was fermented at 25°C until constant weight was achieved. During the fermentation process, sugar changes into alcohol releasing CO_2 with a loss of weight:



At the end of the process, the fermented must was filtered to remove any sediment. The ^{134}Cs content of wine corresponded to 61 % of the must activity (Fig. 14.7). Several different wine-making processes are practiced in industry, but all entail filtration, which is also carried out for home-made wine.

In practical situations the activity in wine can be quantified by the food processing factor P_f , if the activity of grapes is known, as follows: $P_f = (\text{Bq} \cdot \text{kg}^{-1} \text{ wine})$ per $(\text{Bq} \cdot \text{kg}^{-1} \text{ grapes})$. In this experimental study P_f for ^{134}Cs from grapes to wine was of 0.57. From the recent literature, the transfer of stable ^{133}Cs from rice to Japanese sake has been reported by Okuda et al. [34]. The authors calculated a P_f value of 0.04 from brown rice grains to sake.

14.5.2 Olive-Oil-Making

A second study was carried out using olive plants, *Gentile di Chieti*, contaminated via leaves with ^{134}Cs and ^{85}Sr at the beginning of ripening (at the end of September) [35]. At ripening, 10 days after contamination, olives were picked, analyzed for ^{134}Cs and ^{85}Sr , crushed by a mill, and pressed using a hydraulic jack, producing a fluid comprising oil, wastewater, and solid impurities. The oil was then separated from wastewater and solids by centrifugation. TCs were calculated for leaf-to-olives, leaf-to-wastewater, and leaf-to-oil. Results are shown in Fig. 14.8.

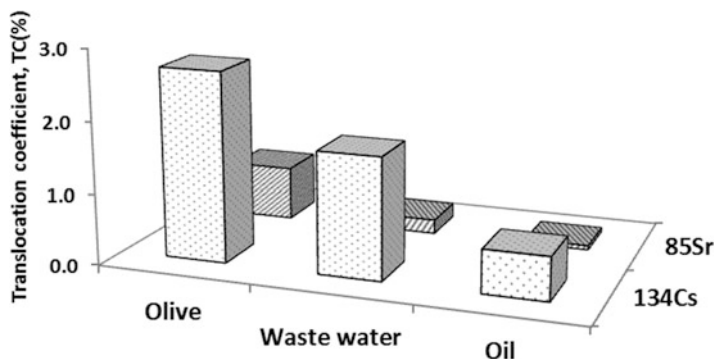


Fig. 14.8 Translocation coefficients, TC(%), from leaf to olive, wastewater, and oil, expressed as $(\text{Bq kg}^{-1} \text{ fresh weight per Bq intercepted plant}^{-1}) \cdot 100$

Only a small fraction of ^{134}Cs in fruit is transferred to oil. ^{85}Sr transfer to oil is not significant. P_f in this research, expressed as $(\text{Bq} \cdot \text{kg}^{-1} \text{ oil})$ per $(\text{Bq} \cdot \text{kg}^{-1} \text{ olives})$, gives values of 0.23 for ^{134}Cs and of 0.08 for ^{85}Sr , indicating that 77 % of ^{134}Cs and 92 % for ^{85}Sr are removed in the process of oil-making. Other studies on olive plants contaminated via soil report that a significant soil-to-fruit transfer of ^{134}Cs may occur, but no transfer to oil is detected [36]. Results from Cancio et al. [37] show that processing into olive oil removes ~ 90 % of the ^{134}Cs contamination initially contained in olive fruit. The P_f for olive oil obtained from data reported in the IAEA TRS 472 gives a value of 0.65 for ^{134}Cs [3].

14.6 Conclusions

From research at UCSC carried out on various fruit plants concerning the behavior of ^{134}Cs and ^{85}Sr after aerial or soil contamination, the following conclusions can be drawn:

- Interception seems to be affected by variables such as leaf senescence and the posture and physical orientation of leaves, rather than by leaf area or biomass.
- Leaf-to-fruit translocation is always higher for ^{134}Cs than for ^{85}Sr . It is also affected by the metabolic activity of the plant and by the phenological stage at time of contamination. The reproductive stages, like anthesis, lead to greater contamination of fruit.
- Soil-to-fruit transfer depends on the growing substrate. It is greater for ^{85}Sr on mineral soils, and greater for ^{134}Cs on peaty soils. Time-dependent changes of soil-to-fruit transfer following acute deposition are not insignificant in annual plants.
- The process of wine-making removes more than 40 % of ^{134}Cs present in grapes; that of oil-making removes about 75 % of ^{134}Cs and 90 % of ^{85}Sr in olives.

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