# Radionuclide Uptake by Plants: Soil-to-Plant Transfer Factors, Kinetics of Absorption, and Internal Radionuclide Distribution of  $137Cs$  and  $40K$  in South American Species

#### Hugo Velasco, Roberto Meigikos dos Anjos and Jimena Juri Ayub

Abstract Soil-to-plant transfer factors  $(F_v)$  are commonly used as a key link to estimate the transference of radionuclides along the human food chain. The use of  $F<sub>v</sub>$  assumes that the radionuclide concentration in plant is linearly related with the radionuclide concentration in soil. However, for different ecosystems and even considering similar soil type and plant species, a large variability in  $F<sub>v</sub>$  values has been reported, showing that other variables in addition to radionuclide soil concentration have influence on the radionuclide uptake by plants. Another possible source of uncertainty in  $F_v$  could come from the heterogeneous distribution of the radionuclide in the plant. In this chapter, we analyze two situations: (a) the root uptake of  $Cs<sup>+</sup>$  and  $K<sup>+</sup>$  using electrophysiological techniques, for three grass species that grow in seminatural grassland of central Argentina and (b) the incorporation of  $137Cs$  and  $40K$  by tropical plants and their translocation to fruits. In both cases, we have considered components that are incorporated into the human food chain and, consequently, there is a need of adjusting their dosimetric contribution on the population.

**Keywords** Soil-to-plant transfer factor  $\cdot^{137}$ Cs  $\cdot^{40}$ K  $\cdot$  Michaelis–Menten kinetics  $\cdot$ Plant uptake

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### 1 Variability of Soil-to-Plant Transfer Factor

The consumption of fruits and vegetables containing radionuclides represents the main way of radionuclide intake by humans and concomitant internal exposure (Shaw and Bell [2001](#page-12-0)). Primarily, the plants incorporate radionuclides through root absorption; the radionuclide transference from soil to vegetation is usually modeled using the soil-to-plant transfer factor  $(F_v)$ . For a particular radionuclide,  $F_v$  is defined as (IAEA [2010\)](#page-12-0):

$$
F_{\nu} = \frac{\text{Activity concentration in plant (Bq kg}^{-1} \text{ dry mass})}{\text{Activity concentration in soil (Bq kg}^{-1} \text{ dry mass in upper 10 or 20 cm)}}\tag{1}
$$

 $F<sub>v</sub>$  values have been collected in databases developed by the US Nuclear Regulatory Commission (Ng et al. [1979](#page-12-0)) and the International Union of Radioecology (Frissel and van Bergeijk [1989](#page-12-0); Frissel et al. [2002\)](#page-12-0). These data were gathered mainly for temperate regions belong to Northern Hemisphere as a consequence of Chernobyl incident. The International Atomic Energy Agency recently published the largest database available today of radionuclide transfer coefficients in terrestrial and freshwater environments, including information of tropical and subtropical environments and regions of the Southern Hemisphere (IAEA [2010](#page-12-0)). The large number of observations that have been accumulated demonstrates that, for most radionuclide, particularly for those of long-lived, soil-to-plant transfer factors show variations, which may exceed three orders of magnitude (Frissel [2002;](#page-12-0) Velasco et al. [2008a](#page-12-0)). In fact, Velasco et al. ([2008b\)](#page-12-0) analyze more than 2,700  $F_v$ values from tropical and subtropical environment with the purpose of exploring the influence of crop types and soil properties on radionuclide uptake by plants. They conclude that  $F_v$  values for  $^{137}Cs$  present differences of five orders of magnitude. Even when the same group of plants and soil type are evaluated, the  $F_v$  variability is wide. In temperate woody trees, for instance,  $F<sub>v</sub>$  values for radiocesium range from 8.6  $\times$  10<sup>-4</sup> to 8.0  $\times$  10<sup>-2</sup> (Carini [2009](#page-11-0); IAEA [2010](#page-12-0)). In contrast, they range from  $5.0 \times 10^{-2}$  to 8.7 in tropical and subtropical environments (IAEA [2010;](#page-12-0) Velasco et al. [2008b\)](#page-12-0). While this variability has been recognized by many authors, different hypotheses have been proposed to understand and reduce the uncertainty <span id="page-2-0"></span>(Frissel and van Bergeijk [1989;](#page-12-0) Ehlken and Kirchner [1996](#page-12-0), [2002;](#page-12-0) Ciuffo et al. [2002;](#page-12-0) Velasco et al. [2004;](#page-12-0) Carini et al. [2005](#page-12-0)). This wide variability suggests that other variables, besides soil concentration, would influence the amount of radionuclide to be uptaken by the plant. The large number of parameters involved and the interdependence between them, make it difficult to assess the effect of each single factor on the entire transfer process.

Considering these constraints in the use of  $F_v$ , in this study, we incorporate two situations that can give some answers to reduce the  $F<sub>v</sub>$  uncertainty. The first is to explore the cellular mechanisms that influence the uptake by plants of monovalent ions, such as  $Cs^+$  and its chemical analogue  $K^+$ . Three grass species (*Distichlis* spicata, Eragrostis curvula, and Cynodon sp) growing in seminatural grassland of central Argentina were selected to investigate their capability to incorporate these cations using electrophysiological techniques (Juri Ayub et al. [2008](#page-11-0)). Kinetic experiments indicated the presence of high-affinity transport systems for the uptake of potassium and cesium in the roots of the analyzed species. Both potassium and cesium uptake data were fitted to Michaelis–Menten model, and the parameters obtained in both cases are comparatively analyzed. Also, for one species, Cynodon sp, the competitive effect of  $K^+$  addition on  $Cs^+$  uptake by roots was evaluated. Both ions compete for the union sites of the same carriers, but do not behave as strict analogues. That could explain the differences observed in  $F_v$ for K<sup>+</sup> and Cs<sup>+</sup>, considered a priori with similar environmental behavior from a radioecological viewpoint.

In the second situation, we evaluate the soil uptake of  $137Cs$  and  $40K$  by tropical plants and their consequent translocation to fruits. The changes in  ${}^{40}$ K and  ${}^{137}$ Cs activity concentrations during the fruit growth of lemon tree were followed from gestation to maturity. A mathematical model was calibrated from the experimental data allowing simulating the radionuclide incorporation by fruits. Although the fruit incorporates much more potassium than cesium, both radionuclides present similar absorption patterns during the entire growth period. Considering the manner in which plants absorb elements present in the soil, and the complex internal distribution of these elements in different parts of plants, it would be possible to reduce the  $F_v$  variability and diminish the uncertainty when food chain models are used to approach the radionuclide content in each compartment.

## 2  $Cs<sup>+</sup>$  and  $K<sup>+</sup>$  Kinetics of Absorption

Three grass species grown in seminatural grassland of central Argentina: Eragrostis curvula, Cynodon sp, and Distichlis spicata were selected to explore the capability to incorporate the monovalent cations  $Cs<sup>+</sup>$  and  $K<sup>+</sup>$ . These plants grow in soils in which  $137$ Cs fallout inventory range from 328 to 730 Bq m<sup>-2</sup>, and  $^{40}$ K specific activity range from 740 to 770 Bq kg<sup>-1</sup>. Individuals of *Cynodon* sp and D. spicata were extracted from the field and cultivated in hydroponic culture.

Parameters <i>D. spicata</i>			Cynodon sp		E. curvula	
	$K^+$	$\mathsf{C}^{\mathsf{c}^+}$	$K^+$	$\mathsf{C}^{\mathsf{c}^+}$	$K^+$	$\mathsf{C}^{\mathsf{c}^+}$
		$D_{MAX}$ (mV) 18.4 $\pm$ 0.9 13.7 $\pm$ 1.5 21 $\pm$ 3		$12 \pm 2$	$56.6 \pm 4.4$ 37.0 $\pm$ 4.1	
$K_M$ (µM)				$97.4 \pm 16.2$ $163.3 \pm 51.9$ $73.3 \pm 34.1$ $120.2 \pm 52.7$ $142 \pm 48$ $607 \pm 268$		
$R^2$	0.98	0.95	0.81	0.91	0.98	0.99

Table 1 Michaelis–Menten kinetics parameters for potassium and cesium uptake by root cells

Commercial seeds of E. curvula were germinated in distilled water and later transferred to hydroponic culture (Juri Ayub et al. [2008\)](#page-11-0). Previously to electrophysiological experiments, all plants were submitted to  $K^+$  starvation for three days. Membrane potentials  $(E_m)$  were determined by electrophysiology techniques (Heredia et al. [2002](#page-12-0)), and variations of this parameter were recorded at different external potassium or cesium concentrations. Changes of the membrane potential  $(E_m)$  can be considered an estimate of ion uptake, and these data could be fitted to the Michaelis–Menten equation:

$$
V = \frac{V_{\text{MAX}} S}{K_M + S} \tag{2}
$$

where S ( $\mu$ M) is the substrate concentration in assay medium; V (estimated by  $\Delta E_m$ , in mV) is the uptake by cell root,  $V_{MAX}$  (estimated by  $D_{MAX}$ , maximum depolarization, in mV) is the maximal rate of uptake by cell root, and  $K_M(\mu M)$  is the Michaelis–Menten constant. High-affinity transport systems (System 1) were detected for the uptake of potassium and cesium in the roots of the three analyzed species. Both potassium and cesium uptake data were fitted to Michaelis–Menten model, showing  $R^2$  values higher than 0.8 (Table 1). In all plants, the affinity constant,  $K_M$ , was smaller for  $K^+$  than for  $Cs^+$ , indicating the higher affinity of the carriers for  $K^+$  than for  $Cs^+$ . Both ions compete for the union sites of the same carriers, but do not behave as strict analogues, as indicated also by the values of  $D_{\text{max}}$  that were always lower for  $Cs^+$  than for  $K^+$ .

Direct effect of increasing  $K^+$  concentrations on  $Cs^+$  uptake was evaluated in Cynodon sp (Fig. [1](#page-4-0)). In culture medium without  $K^+$ , the addition of a saturating concentration of  $Cs^+$  (0.5 mM) induced a membrane depolarization of 14 mV (Fig. [1](#page-4-0), left side). This depolarization value was similar to maximum depolarization obtained in  $Cs^+$  uptake kinetic experiments (Table 1). However, in the presence of 0.5 and 1 mM of  $K^+$  in the assay medium, the addition of  $Cs^+$ (0.5 mM), after membrane potential stabilization, caused a depolarization 3- and 7-fold lower, respectively, than in the absence of  $K^+$  (Fig. [1](#page-4-0), middle). The addition of 0.5 mM  $Cs<sup>+</sup>$  at 5 mM  $K<sup>+</sup>$  did not induce a clear depolarization. An increase in external  $K^+$  concentration causes then a decrease in the absorption of  $Cs^+$ . This effect is due to the competition of both ions for union sites of high-affinity potassium carriers, as it has been shown for these transporters (Heredia et al. [2002\)](#page-12-0).

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The relative permeability of the cellular membrane to these two ions was analyzed for D. spicata and Cynodon sp. The relative permeability was estimated to be 0.74 and 0.71, respectively. These findings indicate that the membranes of the root cells of the species analyzed are highly permeable to  $Cs<sup>+</sup>$ , being around 70 % of  $K^+$  permeability.

### 3 Radionuclides Internal Distribution in Plants

As was previously emphasized, when the transfer factors are assessed, the procedure presumes that the concentration in the plant compartment and in the soil remains constant over time. That implies that if food chain model is applied, the steady state in radionuclide concentration in each compartment is assumed (Ng et al. [1979\)](#page-12-0). This condition is acceptable for the radionuclide soil concentration, but it could be a severe limitation for inaccurate analysis about the radionuclide content in part of the plants. A typical deviation to this behavior occurs particularly when the incorporation of radionuclides by fruits is evaluated. They can be consumed at various stages of their growth and the specify activity usually changes strongly during this period.

In this section, we analyze the changes in  ${}^{40}$ K and  ${}^{137}$ Cs activity concentrations during the growth of tropical lemon fruits (Citrus limon) from gestation until maturity. The experiments were performed at the Laboratório de Radioecologia e Alterações Ambientais (LARA) of the Instituto de Fisica of Universidade Federal Fluminense using lemon trees grown in Goiânia region, in central Brazil, where in 1997, there was a radiological accident involving a source of  $137Cs$  (IAEA [1988;](#page-12-0) Anjos et al. [2002,](#page-11-0) [2007,](#page-11-0) [2009a](#page-11-0), [b](#page-11-0); Carvalho et al. [2006](#page-12-0); Mosquera et al. [2006\)](#page-12-0). The soil profile was analyzed from the surface to 50 cm deep, in 5-cm-thick layers. Soil samples were dried, submitted to a homogenization process, and sieved in order to separate the soil particles from the stones, grass, roots, and other organic materials.

<span id="page-5-0"></span>The fruit samples were washed with distilled water, dried in stove at 105  $\degree$ C, and finally were ground to powder. Afterward, all samples were packed into cylindrical plastic containers, dry-weighed, and sealed. The amounts of  $^{137}Cs$  and  $^{40}K$  in the soil samples were determined by standard gamma spectrometry. The presence of potassium in fruit samples was quantified by neutron activation analysis (NAA) performed at the IEA-R1 research reactor of the Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP). Samples were bombarded with neutrons, resulting in the production of radioactive isotopes of the elements of interest. Gamma rays emitted by the radioactive isotopes were then analyzed by gamma-ray spectrometry.

The activity concentration of  $^{137}Cs$  and  $^{40}K$  in lemon fruit was evaluated at different times (Table [2](#page-6-0)). The fruit dry mass for each time and the radionuclide activity concentration in the 20 cm of top soil are also indicated.

Data obtained were used to calibrate a mathematical model for describing the growth dynamics of fruits and the temporal evolution of the soil-to-fruit transfer factor (Velasco et al. [2012](#page-13-0); Cid et al. [2013](#page-12-0)). The model assumes that fruit dry mass grows according to a logistic model taking into account two main assumptions:

• The growth curve of the fruit dry matter follows the differential equation:

$$
\frac{dM(t)}{dt} = \alpha M(t)[M_m - M(t)] \tag{3}
$$

where *M* is the fruit dry mass (g), *t* the time (d),  $\alpha$  the maximum growth rate  $(d^{-1})$ , and  $M_m$  the maximum value of  $M(t)$ . The solution to Eq. (3) is

$$
M(t) = \frac{M M_m}{(M_m - M_0) \exp(-\alpha t) + M_0}
$$
\n(4)

where  $M_0 = M(0)$ .

• The radionuclide activity concentration in fruits decreases exponentially following the differential equation:

$$
\frac{\mathrm{d}C}{\mathrm{d}t} = -\beta (C - C_{\min})\tag{5}
$$

where C is the radionuclide activity concentration in fruit (Bq kg<sup>-1</sup>),  $\beta$  is the exponential decreasing parameter  $(d^{-1})$ , and  $C_{\text{min}}$  is the minimum radionuclide activity concentration value (Bq  $kg^{-1}$ ). Its solution is



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$$
C(t) = C_r \exp(-\beta t) + C_{\min} \tag{6}
$$

where  $C_r = C(0)-C_{\text{min}}$ .

From the multiplication of Eq. [13.5](http://dx.doi.org/10.1007/978-3-319-07665-2_13) and [13.6](http://dx.doi.org/10.1007/978-3-319-07665-2_13), the radionuclide activity (Bq) in the entire fruit as a function of time can be obtained as

$$
A(t) = M(t) \times C(t) \tag{7}
$$

For this case, the soil-to-fruit transfer factor  $(F_v)$  is (Eq. [1](#page-1-0))

$$
F_{\nu} = \frac{\text{Concentration of radionuclide in fruit (Bq kg-1 dry mass)}}{\text{Concentration of radionuclide in soil (Bq kg-1 soil dry mass in upper 20 cm)}} \tag{8}
$$

Assuming that the radionuclide activity concentration in soil does not change during the fruit growth period, then the soil-to-fruit transfer factor is

$$
F_v(t) = \frac{C_F(t)}{C_S} = \frac{C_{Fr}}{C_S} \exp(-\beta t) + \frac{C_{Fvmin}}{C_S}
$$
  
=  $F_{vr} \exp(-\beta t) + F_{vmin}$  (9)

where  $F_{\text{vmin}}$  is the minimum value of  $F_v$  and the sum  $F_{vr} + F_{vmin}$  is the maximum value of the soil-to-fruit transfer factor.

The theoretical approach for describing the growth of the fruit dry mass (Eq. [3](#page-5-0)) has been represented in Fig. 2. The parameter values obtained from the model calibration are showed in the same figure. Fruit development shows a sigmoidal growth with a maximum growth rate around  $t = 100$  d. After the time  $t = 160$  d,



the growth rate decreases, reaching the fruit the final stage, with a dry mass of 18–19 g.

Figures 3 and [4](#page-9-0) show  ${}^{40}$ K and  ${}^{137}$ Cs activity concentration in fruit, respectively, for the entire development period, and the curves represent the theoretical approach. The values of  $\beta$  is the same for both radionuclides suggesting a similar behavior in which the fruit incorporates both cations (Delvaux et al. [2000](#page-12-0); White and Broadley [2000](#page-13-0); Zhu and Smolders [2000\)](#page-13-0). Robison et al. ([2009\)](#page-12-0), studying the changes in  $40K$  and  $137Cs$  concentrations in the development of coconut trees fronds, find a similar decrease in radionuclides concentrations, for plant grown in low potassium content in soil. These plants show a retrieval of  $K^+$  and  $Cs^+$  from the fronds to the plant when leaves reach maturity and senescence. Although fruits and leaves are not physiologically identical, our results show that in the development of lemon fruits, some potassium and cesium are recovered by the plant when they reach maturity, similar to what occurs in coconut leaves.

The rate of  ${}^{40}$ K and  ${}^{137}$ Cs incorporation by lemon fruits is represented in Fig. [5](#page-9-0) (Eq. [7](#page-7-0)), the derivative function of  $A(t)$ . This figure shows the greater preference for the fruits to absorb  ${}^{40}$ K in relation to  ${}^{137}$ Cs throughout the growth period. Although the same transport mechanisms are implicated in the transfer of ions to fruit, differences in the uptake rates has been reported for several plant species between both potassium and cesium, showing the plant capability for discriminating between potassium and cesium, preferably by the first ion (Shaw and Bell [1989;](#page-12-0) Juri Ayub et al. [2008](#page-11-0)).

Finally, in Fig. [6](#page-10-0), we represent  $F_v$  as a function of time. The theoretical curves have been obtained from Eq. [9.](#page-7-0) The characteristic factor of the exponential decreasing function ( $\beta = 0.5$ ) is the same for both radionuclides. At the beginning

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of fruit development period,  $F_v$  is about 17 times greater for <sup>40</sup>K than for <sup>137</sup>Cs. In the maturing period, this difference seems to increase, being the soil-to-fruit transfer factor for  ${}^{40}$ K about 25 times higher than for  ${}^{137}$ Cs.

<span id="page-10-0"></span>

Fig. 6 Temporal dependence of  $^{137}Cs$  and  $^{40}K$  soil-to-fruit transfer factor for tropical lemon trees (Velasco et al. [2012\)](#page-13-0)

### 4 Conclusions

Soil-to-plant transfer factors are commonly used as a key link to estimate the transference of radionuclides along the human food chain. The use of  $F_v$  in the calculations of this transfer involves the assumption that the radionuclide concentrations in plant and in the rooting zone of the soil are linearly related. However, this condition usually involves an oversimplification. Physical, chemical, and biological mechanisms occur both in the absorption process of radionuclides by root and in the process of redistribution of radionuclides in different parts of the plant. That makes this linear relationship is usually not satisfied or that the dispersion in the  $F_v$  measured values is extremely wide.

In this study, we explore two situations that can help us to partially explain this uncertainty and thus reduce it. In the first, grass species grown in seminatural grassland of central Argentina were selected to follow their capability to incorporate the monovalent cations  $Cs<sup>+</sup>$  and  $K<sup>+</sup>$ . The results obtained show that the permeability of the membrane of the root cells to  $Cs<sup>+</sup>$  is almost 70 % of  $K<sup>+</sup>$ permeability, indicating that potentially cesium can be incorporated in plant when is available in soil.

On the other hand, plants have a high-affinity system to uptake potassium and cesium. The transport of both ions are carried out by the same potassium carriers, but showing a lower affinity for  $Cs^+$  and also a lower maximum uptake rate. Electrophysiological experiment allows detecting interspecific differences in  $V_{MAX}$ and  $K_M$  for K<sup>+</sup> and Cs<sup>+</sup> uptake, as it has been described for Cs<sup>+</sup> uptake kinetics in another grass species.

<span id="page-11-0"></span>In addition, we have shown that addition of  $K^+$  to assay culture decreases strongly  $Cs<sup>+</sup>$  uptake in *Cynodon* sp. The external concentration of  $K<sup>+</sup>$  has then a direct influence on  $Cs^+$  incorporation by plants. Kinetic studies of  $Cs^+/K^+$  interaction can help us to understand the wide variability detected in transfer factors and could also provide a screening procedure to select plants with different cesium affinities, for example with low cesium uptake capacity.

In the second situation, we explore the  $^{137}Cs$  and  $^{40}K$  soil-to-fruit transfer factor for tropical lemon trees. We conclude that  $F_v$  for <sup>40</sup>K, at the initial stage of fruit development, is more than twice that in the fruit maturation stage, while  $F_v$  for  $137Cs$  is three times higher. In spite of the chemical analogies between potassium and cesium,  $F_v$  is at least one order of the magnitude higher for <sup>40</sup>K during the entire fruit development period. However, the temporal pattern for radionuclides incorporation by fruits was very similar for both radionuclides. The logistic model was applied to describe the dry mass growth for fruits. The corresponding parameters were obtained when compared with experimental data. The fruit growth is a dynamic process and determines the degree of contamination by radionuclides. This is of fundamental importance because it involves the edible part of the plant, and any radionuclide content is transferred to the consumer.

A theoretical approach was introduced describing the evolution of  ${}^{40}$ K and  $137$ Cs activity concentrations in fruits as a function of time. The agreement with the experimental data is satisfactory. Then, the radionuclide input rate was calculated for both radionuclides. Finally, the transfer factor was determined for the entire growth period, demonstrating its temporal variability.

In the explored cases, the agreement between the experimental data and the theoretical approach is very good. The results could be a significant advance in the modeling of nutrients accumulation by fruits and the explanation of the variability of the radionuclide soil-to-fruit transference.

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