

Variability of ^{137}Cs and ^{40}K soil-to-fruit transfer factor in tropical lemon trees during the fruit development period

H. Velasco^{a,*}, A.S. Cid^b, R.M. Anjos^b, C.B. Zamboni^c, M. Rizzotto^a, D.L. Valladares^a, J. Juri Ayub^{a,b}

^aGEA - Instituto de Matemática Aplicada San Luis (IMASL), Universidad Nacional de San Luis, CCT-San Luis CONICET, Ej. de los Andes 950, D5700HHW San Luis, Argentina

^bLARA - Laboratório de Radioecologia, Instituto de Física, Universidade Federal Fluminense, Av. Gal Milton Tavares de Souza, s/no Gragoatá, 24210 340 Niterói, RJ, Brazil

^cInstituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN), Av. Lineu Prestes 2242 - Cidade Universitária, 05508 000 São Paulo, SP, Brazil

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ABSTRACT

In this investigation we evaluate the soil uptake of ^{137}Cs and ^{40}K by tropical plants and their consequent translocation to fruits, by calculating the soil-to-fruit transfer factors defined as $F_v = [\text{concentration of radionuclide in fruit (Bq kg}^{-1}\text{ dry mass)}/\text{concentration of radionuclide in soil (Bq kg}^{-1}\text{ dry mass in upper 20 cm)}]$. In order to obtain F_v values, the accumulation of these radionuclides in fruits of lemon trees (*Citrus limon B.*) during the fruit growth was measured. A mathematical model was calibrated from the experimental data allowing simulating the incorporation process of these radionuclides by fruits. Although the fruit incorporates a lot more potassium than cesium, both radionuclides present similar absorption patterns during the entire growth period. F_v ranged from 0.54 to 1.02 for ^{40}K and from 0.02 to 0.06 for ^{137}Cs . Maximum F_v values are reached at the initial time of fruit growth and decrease as the fruit develops, being lowest at the maturation period. As a result of applying the model a decreasing exponential function is derived for F_v as time increases. The agreement between the theoretical approach and the experimental values is satisfactory.

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1. Introduction

More than half of the elements of the periodic table have been found in plant tissues, but not all are essential for plant growth and reproduction. Potassium is an essential or building element present in ionic form in vegetal tissues, and directly involved in the plant physiology (Barker and Pilbean, 2007; Mengel, 2007; Pallardy, 2008). Potassium is highly mobile in plants, is involved in enzyme activation, protein synthesis, osmoregulation, stomatal opening and closing, photosynthesis, and cell expansion. Under potassium starvation conditions plants are unable to complete their life cycle. Radioecological studies have shown that others univalent cationic species with a similar size and water mantle can have comparable behavior to K^+ in the biogeochemical pathway (IAEA, 2010). In the environment, cesium is mainly present in soil solution as a free hydrated cation (Cs^+) with little or no tendency to form soluble complexes. Similarly to K^+ , $^{137}\text{Cs}^+$ can be absorbed by plant roots from soil solution and translocated to the above-ground plant parts (Delvaux et al., 2000; White and Broadley, 2000; Zhu and Smolders, 2000). Additionally, radiocaesium uptake can be

influenced by the roots distribution within the soil profile, soil type, soil water status and characteristic rate uptake of fruit species as well as by specific activity of the roots (Atkinson and Webster, 2001).

Several experimental studies regarding the radionuclide uptake by fruit plants have been carried out in the last decades. These studies were mainly focused on the behavior of fallout radionuclides, mainly ^{137}Cs , but rarely addressed on K. Additionally, in most cases, the information was not sufficient to understand the ^{137}Cs incorporation by plants (Carini, 1999). Recently, the International Atomic Energy Agency published *The Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments* (IAEA, 2010), which contains a full collection of reviewed transfer factor data and the methods used to obtain the tabulated data values.

Studies regarding the behavior of univalent inorganic cations in tropical fruit species have been performed by our groups since 1999 (Carvalho et al., 2006; Mosquera et al., 2006; Sanches et al., 2008; Anjos, 2006; Anjos et al., 2009a, 2009b; Velasco et al., 2010). Previous results obtained analyzing plant tissues of lemon, orange, guava, mango, avocado, banana, papaya and chili peppers, have shown that nutrients and radioactive elements uptake by roots largely depends on the kind of plant. The concentrations of K^+ , NH_4^+ , $^{137}\text{Cs}^+$ in roots, stems, leaves and fruits are influenced by both

* Corresponding author. Tel./fax: +54 2652 422803.

E-mail address: hvelasco@unsl.edu.ar (H. Velasco).

direct soil uptake and movement of these cations among plant compartments. The extent of this movement, or translocation, differs greatly, depending upon the type of radionuclide and the growth activity of the tree. Tissues rich in water show high K^+ and $^{137}Cs^+$ concentrations. Plant organs rich in these radionuclides are young leaves, young roots, and fleshy fruits.

Radionuclides uptake by plants is usually assessed using soil to plant transfer factors (F_v). This F_v is calculated as the ratio between the activity concentration in the considered plant compartment (stem, leaf, fruit, whole plant, etc) and the activity concentration in the topsoil; both concentrations are assumed to be constant over time. F_v values have been collected in databases developed by the US Nuclear Regulatory Commission (Ng et al., 1979), the International Union of Radioecology (Frissel and van Bergeijk, 1989; Frissel et al., 2002) and the International Atomic Energy Agency (IAEA, 2010). However, these values have shown a wide variability, particularly for fruit plants. Usually, transfer factors evaluated for the same plant species and the same plant compartment can have significantly different values, ranging by several orders of magnitude.

Aiming to deepen the knowledge about the incorporation of monovalent inorganic cations during the process of growth and maturation of the edible parts in tropical and subtropical plants (Anjos et al., 2009), fruit samples at various stages of development were taken from lemon trees. Furthermore, with the purpose to understand the F_v variability for fruit of tropical lemon trees [*Citrus limon* (L.) Burm. f.], we calculate soil-to-fruit transfer factor for ^{40}K and ^{137}Cs , from the time of gestation of the fruit until its maturity date. A mathematical model was also developed for describing the growth dynamics of fruits and the temporal evolution of the soil-to-fruit transfer factor for ^{40}K and ^{137}Cs in tropical fruits.

2. Material and methods

This work involved the participation of three research Institutions. The sample preparation and the gamma-ray analysis were carried out at the Laboratório de Radioecologia (LARA), Universidade Federal Fluminense, Brazil. The neutron activation analysis was performed at the IEA-R1 research reactor, Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN), Brazil. The data analysis and modeling were developed by the Grupo de Estudios Ambientales (GEA), UNSL-Conicet, Argentina.

2.1. Sampling

The lemon trees were grown in Goiânia region, where a radiological accident involving a source of ^{137}Cs occurred in 1987 (IAEA, 1988; Anjos et al., 2001, 2002; Facure et al., 2001).

The soils from the surrounding of each tree were sampled. The soil samples were taken using a post-hole digger. The rooting depth was defined as 20 cm for agricultural crops, following the protocol for the experimental determination of soil to plant transfer factors (concentration ratios) to be used in radiological assessment models (IUR, 1992; Frissel, 1997).

The soil samples were dried, submitted to a homogenization process, and sieved through a 1-mm screen in order to separate the soil particles from the stones, grass, roots and other organic materials. Afterward, all samples were packed into cylindrical plastic containers, dry-weighed and sealed.

According to EMBRAPA (1999), the soils in Goiânia are classified as dark red latosol. In general, this kind of soil tends to be acid, but by presenting construction remains, it shows an alkaline soil. Among its main features, the total potassium content was $2.4 \pm 0.2\%$, the activity concentrations of ^{137}Cs was $3.6 \pm 0.3 \text{ kBq kg}^{-1}$, the sand content ranged from 47% to 58%, the silt

content from 35% to 43%, and the clay content from 7.3% to 10%. The soil pH was measured in 1 M KCl in a solid–liquid ratio of 1:2.5 after 1 h equilibrium. The pH value ranged from 6.2 to 7.8. The organic C content was measured with a wet combustion technique using $K_2Cr_2O_7$ and H_2SO_4 followed by CO_2 analysis. The organic C content ranged from 1.5% to 1.7%. The cation exchange capacity (CEC) was measured using silver thiourea as the index cation in a 0.1-M NH_4OAc buffer. CEC ranged from 5.0 to 5.5 $cmol \text{ kg}^{-1}$ soil.

Lemon trees, a member of the family Rutaceae, can be grown in both dry and humid atmospheres, preferring areas with abundant sunlight and annual rainfall from 25 to 125 cm (Morton, 1987). Lemon trees reached a height of 3–4 m. The leaves are light-green when young and became dark-green when old. They are elliptic, 6–11 cm long, with slender wings on the petioles. The mildly fragrant flowers are solitary or there are two or more clustered in the leaf axils. The fruits are oval with a nipple-like protuberance at the apex, 5–7 cm long. Since the lemon tree bears fruit twice a year, there were two collecting sets, the first going from March to May 2009 and the second from September to December 2009. The first collect was divided in six sampling sets and second was divided in ten sets. Three individual of lemon trees of 6–8 years old, were used in this work. From each lemon plant, 5 lemon fruits were randomly collected in each sampling time. The fruit samples were washed with distilled water, dried in stove at 105 °C, and finally were ground to powder. The lemon trees were cultivated in the same place, in which there are no significant variations of soil properties. Similarly to soil samples, fruit samples were packed into cylindrical plastic containers, dry-weighed and sealed.

2.2. Measurements

There are a few chemical and physical analytical procedures for extraction and quantification of potassium and radiocaesium in plants. Since ^{137}Cs is a radioactive element, its amount in a sample can be determined using nuclear analytical methods, such as gamma spectrometry. Measure amount of K can also be accomplished by such methods, taking into account the basic underlying assumption that is the isotopic composition of potassium in nature is essentially constant, with variations not exceeding 1% in terms of the $^{40}K/K$ ratio (McDougall and Mark Harrison, 1999). Additionally, all isotopes of potassium (and cesium) show the same behavior in plants (Mengel, 2007). Therefore, ^{40}K can be used to trace the potassium behavior in plants.

The amounts of ^{137}Cs and ^{40}K in soil and fruit samples were initially determined by gamma spectrometry. The dry weights of each soil sample were about 200 g. About 2.0–3.0 g dry weight of fruit sample was used for measurements of ^{137}Cs activity concentration. However, about 30–50 g dry weight of fruit sample are necessary to determine the activity concentration of ^{40}K . This material amount was required to compensate the decrease of count rate due to the lower detection efficiency of this radionuclide compared to ^{137}Cs . In effect, this problem is also reflected in determining the MDA (Minimum Detectable Activity) of ^{40}K , which is about ten times higher than that for ^{137}Cs (see item 2.2.1). Thus the gamma spectrometry technique became unfeasible to determine the ^{40}K activity concentration in lemon samples due to the large amount of fruit weight necessary to obtain a single sample. This caused that for the first sampling set (from March to May 2009) limited data of ^{40}K activity concentration were obtained (with large error bar).

Aiming to solve this problem, we conducted a second sampling (from September to December 2009) and we chose to use the method of Neutron Activation Analysis (NAA) to measure K concentration in fruit samples. The results were consistent with the first sampling and enabled to obtain a potassium distribution with

low statistical error. Therefore, measurements of ^{137}Cs and ^{40}K in soil, and ^{137}Cs in lemons were performed by gamma spectrometry (first collect divided in six sampling sets). Measurements of K in lemons were performed by NAA (second collect divided in ten sampling sets).

2.2.1. Determination of ^{137}Cs in soil and fruit and of ^{40}K in soil using gamma spectrometry

Gamma-ray analysis was performed using a 55% efficiency high-purity Germanium detector (HPGe), in order to obtain the concentrations of ^{137}Cs and ^{40}K in soil and ^{137}Cs in fruits (Anjos et al., 2009a). Energy spectra measurements from soil and fruit samples were accumulated for 4 h and 24 h, for ^{137}Cs and ^{40}K , respectively. Radionuclide activities in soil and fruit samples were calculated from the net full energy peaks (661.7 keV gamma-ray line for ^{137}Cs and 1460.8 keV for ^{40}K) and the measured efficiency of the detector. The ^{40}K calibration was performed using a reference material (RGK-1) obtained from the International Atomic Energy Agency. Standards of radiocaesium were made by mixing uncontaminated sawdust with the liquid ^{137}Cs solution prepared by the Comissão Nacional de Energia Nuclear of Brazil (CNEN). Their results were inter-compared with two laboratories in Brazil. Technical details of sample preparation and analysis can be obtained from Mosquera et al. (2006) and Carvalho et al. (2006). Hence, using the dry weight of the samples, their respective activity concentrations of ^{137}Cs and ^{40}K could be expressed in activity per unit weight (Bq kg^{-1}). The uncertainties of the specific ^{137}Cs activity from each individual measurement of fruit and soil samples ranged from 3 to 7%. The uncertainty of ^{40}K from soil samples was estimated to be around 10%. The MDA (Minimum Detectable Activity) was estimated to be 0.7 Bq kg^{-1} d.w. for ^{137}Cs and 6.3 Bq kg^{-1} d.w. for ^{40}K .

2.2.2. Determination of K in fruit using NAA

The potassium content in fruit samples was quantified by Neutron Activation Analysis – NAA (Vasconcellos et al., 2004). In this technique, fruit samples and certified standard reference material (NIST 1573a Tomato Leaves) were irradiated together, with thermal neutrons, allowing the simultaneous activation of these materials under the exact same irradiation conditions. Gamma rays emitted by the radioactive isotopes were then analyzed by gamma spectrometry.

Several aliquots of each fruit (50–600 mg) and standard (100–250 mg) were accurately weighed and sealed into individual pre-cleaned polyethylene bag and irradiated in a pneumatic station in the IEA-R1 nuclear reactor (2–4 MW, pool type) using irradiation times of 120 s, decay time of 600 s and counting time of 300 s (for samples and standard). Each fruit sample was analyzed in triplicate and the reported results are the mean value. Gamma spectrometry was performed using a 60% efficiency high-purity Germanium detector (HPGe) and the concentration was obtained using in-house software. The uncertainty associated was about 5% for potassium content. The inter-comparison between INAA and conventional gamma spectrometry results for K content in fruit samples were very satisfactory.

The total potassium weight concentration, T_K (g kg^{-1} d.w.), in the fruit sample may be converted to the activity concentration of ^{40}K , C_K , (Bq kg^{-1} d.w.), by the following equation:

$$T_K (\text{g kg}^{-1}) = 7.56 \times 10^{-17} \cdot A_w \cdot T_{1/2} \cdot C_K (\text{Bq kg}^{-1}) \quad (1)$$

where A_w is the radionuclide atomic weight in g mol^{-1} , and $T_{1/2}$ is the radionuclide half-life in years.

2.3. Theoretical approach

In radioecological studies, the assessments of radionuclide contaminations in edible parts of a plant following soil contamination is an important subject to estimate doses rates associated with potential releases of radionuclides to the environment and, consequently, to man. The input of radionuclides into food chains may require the application of measurements intended to limit or reduce the internal dose to man resulting from consumption of contaminated food (Carini, 2001). Fruits represent an important component of the total diet of the population. Since there are radioactive elements and mineral nutrients that are transported to the aerial parts of the plant in a similar way, it is interesting to study how they behave during the growth of fruits.

The time required for fruit growth varies widely among species and genotypes. The period from anthesis to fruit ripening varies from about three weeks in chili pepper to 40 weeks in coconut. Various types of fruits grow at different rates and reach different sizes at maturity. The fruit growth involves cell division and cell expansion. During anthesis there is little cell division. After a fruit is set, however, it becomes an active carbohydrate sink, and many of its tissues become meristematic (Pallardy, 2008).

According to Pallardy (2008) and Carini (2001), growth curves of fruits are of two general types. The first is a simple sigmoid type in which there is initially an exponential increase in size followed by slowing down of growth in a sigmoid fashion. This type of growth is characteristic of some tropical and subtropical species, such as orange, banana, avocado, mango, coconut and lemon. The precise shape of the growth curve differs somewhat with the plant variety. The second type is a more complex growth curve, involving two periods of fast growth with a period of slow or suspended growth among them. This is common in stone fruits (such as peach, apricot plum and coffee), as well as some nonstone fruits (such as grape and currant).

Based on these evidences and experimental results, we have developed a theoretical model aiming to describe the temporal evolution of ^{137}Cs and ^{40}K incorporation during the growth of tropical fruits (Anjos et al., 2009b). The model assumes that fruit dry weight grows according to a logistic model, taking into account two main assumptions:

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

$$\frac{dM(t)}{dt} = \alpha M(t)[M_m - M(t)] \quad (2)$$

where M is the fruit dry weight (g d.w.), t the time (d), α the maximum growth rate (d^{-1}), and M_m the maximum value of $M(t)$. The solution to Eq. (2) is:

$$M(t) = \frac{M_0 M_m}{(M_m - M_0) \exp(-\alpha t) + M_0} \quad (3)$$

where $M_0 = M(0)$.

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

$$\frac{dC}{dt} = -\beta(C - C_{\min}) \quad (4)$$

where C is the radionuclide activity concentration in fruit (Bq kg^{-1} d.w.), β is the exponential decreasing parameter (d^{-1}), and

Table 1

Fruit dry weight (g) and mean values of ^{137}Cs and ^{40}K activity concentrations in fruit (Bq kg^{-1} d.w.) measured at various times during the fruit development. Mean values of activity concentrations of ^{137}Cs and ^{40}K in soil measured at one point in time are also given.

Time (d)		20 (7)	60 (7)	90 (7)	120 (7)	140 (7)	160 (7)				
^{137}Cs activity concentration (Bq kg^{-1} d.w.)	Fruit	209 (25)	114 (15)	93 (13)	83 (13)	76 (12)	83 (13)				
	Soil	3659 (292)									
Time (d)		16 (7)	31 (7)	49 (7)	55 (7)	71 (7)	87 (7)	113 (7)	129 (7)	158 (7)	165 (7)
^{40}K activity concentration (Bq kg^{-1} d.w.)	Fruit	805 (105)	637 (48)	482 (57)	422 (38)	440 (25)	422 (51)	407 (44)	365 (35)	437 (48)	431 (47)
	Soil	792 (32)									
Fruit dry weight (g)		0.117 (0.05)	0.523 (0.16)	0.847 (0.16)	3.08 (0.5)	3.92 (0.83)	5.17 (0.65)	7.39 (1.0)	10.28 (1.3)	18.90 (2.0)	17.22 (2.0)

The values given in parentheses indicate the standard deviations of the mean. The error associated with time (7 days) aims to take into account the uncertainty in time between the period of flowering and the beginning of fructification.

C_{\min} is the minimum radionuclide activity concentration value (Bq kg^{-1} d.w.). Its solution is:

$$C(t) = C_r \exp(-\beta t) + C_{\min} \quad (5)$$

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

From the multiplication of Equations (3) and (5), the radionuclide activity (Bq) in the entire fruit as a function of time can be obtained:

$$A(t) = M(t) \times C(t) \quad (6)$$

The concept of *Transfer Factor* has been developed to predict the radionuclide concentration in any particular organism (or compartment of this), taking into account the radionuclide soil concentration as reference. This concept is based on the assumption that if equilibrium has been achieved then the degree of radionuclide bioaccumulation in the organism is proportional to the concentration of this radionuclide in the supporting environment (Twining, 2011). The transfer factor (F_v) for the plant uptake of any radionuclide from soil is a dimensionless quantity (concentration ratio), defined as the ratio between the radionuclide concentration in the plant compartment (dry weight) and the radionuclide concentration in soil (dry weight), in a defined soil layer (IAEA, 2010):

$$F_v = \frac{\text{Concentration of radionuclide in fruit} (\text{Bq kg}^{-1} \text{ dry weight})}{\text{Concentration of radionuclide in soil} (\text{Bq kg}^{-1} \text{ soil dry weight in upper 20 cm})} \quad (7)$$

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

$$F_v(t) = \frac{C_F(t)}{C_S} = \frac{C_{Fr} \exp(-\beta t) + C_{F\min}}{C_S} \quad (8)$$

$$= F_{vr} \exp(-\beta t) + F_{v\min}$$

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

3. Results and discussion

Table 1 shows the mean values of ^{137}Cs and ^{40}K activity concentrations in fruit (on dry matter weight basis) measured during ripening process of lemon fruits, grown on soils contaminated by ^{137}Cs . Measurements of ^{137}Cs activity concentrations in lemons were performed from first collecting (divided in six sampling sets) by gamma spectrometry. Measurements of ^{40}K activity concentrations in lemons were performed from second collecting (divided in ten sampling sets) by NAA. The fruit dry

weight for each of the ten sampling times and the radionuclide activity concentration in the 20 cm of topsoil are also indicated. During ripening process of lemons, its fresh weight increased from 0.56 to 159 g, corresponding to a variation in dry weight bases ranged from 0.11 to 17.22 g, indicating that the average percentage of water content of lemons fruits ranged from 79 to 89%. Due to the fact that K-stable and ^{40}K have the same physical-chemical properties and that ^{40}K activity concentration in lemons can be converted into total potassium (K) content using Eq. (1), the behavior of K content in lemons can also be investigated. Potassium was consumed in large quantities, so that ^{40}K activity concentration ranged from 431 to 805 Bq kg^{-1} . On the other hand, the ^{137}Cs activity concentrations ranged from 83 to 209 Bq kg^{-1} . These results indicate that the incorporation of K^+ and $^{137}\text{Cs}^+$ in fruit is age-dependent, more in particular their concentration decreases when fruit age increases. However, considering that fruit weight increases with age, the total fruit content of K and Cs increases from anthesis to ripening.

Fig. 1 shows the growth curve of lemons. The data were fitted by Eq. (3), confirming that the 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 weight of 18–19 g.

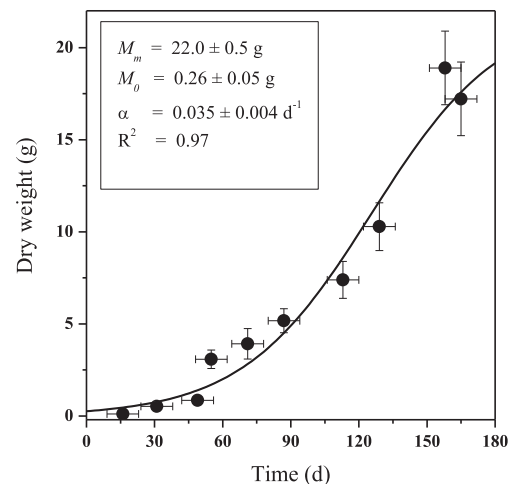


Fig. 1. Fruit dry weight as a function of time. Measurements and theoretical approach.

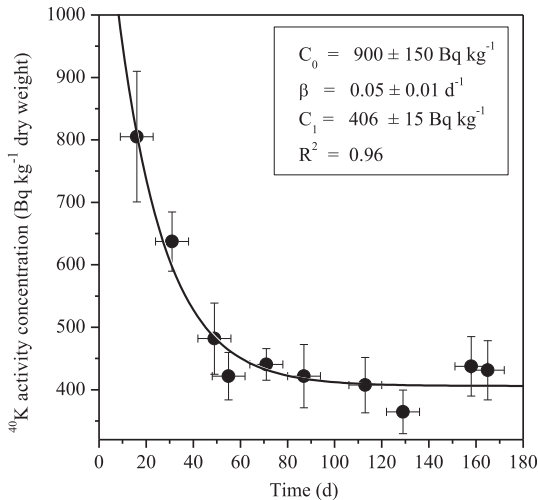


Fig. 2. ^{40}K activity concentration in fruit as a function of time. Measurements and theoretical approach.

Fig. 2 and Fig. 3 show the temporal evolution of ^{40}K and ^{137}Cs activity concentration in lemons, respectively. Like K^+ , $^{137}\text{Cs}^+$ distribution shows an exponential decreasing when fruit age increases. The theoretical curves, given by Eq. (5), have been also represented in these figures. In both cases, the fit of the model is very satisfactory. The parameter β , characteristic of the exponential decreasing function, takes the same value in both cases ($\beta = 0.05 \pm 0.01 \text{ d}^{-1}$), confirming the similar behavior in which the fruit incorporates both cations (Delvaux et al., 2000; White and Broadley, 2000; Zhu and Smolders, 2000). This significant correlation between the activity concentrations of ^{137}Cs and ^{40}K in fruits has been observed in previous investigation (Mosquera et al., 2006; Sanches et al., 2008; Anjos et al., 2009a, 2009b).

The temporal derivative of the function $A_T(t)$, given in Eq.(6), gives us the rate of radionuclide incorporation by fruit. The rate of ^{40}K and ^{137}Cs incorporation by lemon is represented in Fig. 4. This figure suggests the greater preference for the fruits to absorb ^{40}K in relation to ^{137}Cs during its growth period. Shaw and Bell (1989) has reported that, although the same transport mechanisms are implicated in the transfer of K^+ and Cs^+ , differences in the uptake

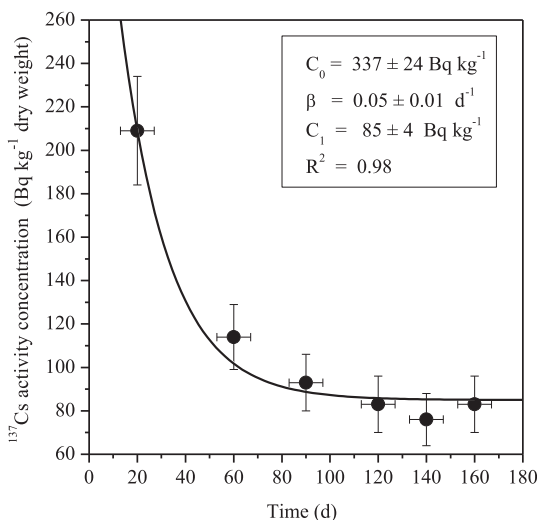


Fig. 3. ^{137}Cs activity concentration in fruit as a function of time. Measurements and theoretical approach.

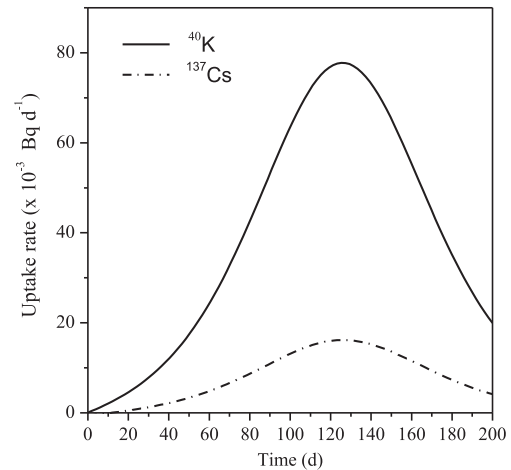


Fig. 4. ^{40}K and ^{137}Cs uptake rate by fruit as function of time. These theoretical represent the temporal derivative of the function $A_T(t)$ given by Eq. (7).

rates would indicate that plants have an ability for discriminating between these two ions, giving preference to potassium. However, if we analyze the amount of radiocaesium and potassium in the soil and inside the plant (in units of g kg^{-1} and not in Bq kg^{-1}), we note that the amount of potassium in soil is markedly higher than the amount of cesium. The same result is observed when lemon fruits are analyzed. When we say that the activity concentration of ^{40}K in the fruits ranged from 431 to 805 Bq kg^{-1} , while the ^{137}Cs activity concentrations ranged from 83 to 209 Bq kg^{-1} , this means a range from 11 g kg^{-1} to 25 g kg^{-1} for K and 2.6×10^{-11} to $6.5 \times 10^{-11} \text{ g kg}^{-1}$ for ^{137}Cs . ^{137}Cs can be considered as trace element (in fruit). Nevertheless, the similar distribution or behavior of these two ions indicates that the same transport mechanisms are implicated in the transfer of K^+ and $^{137}\text{Cs}^+$ ions to fruits.

Contamination of the edible parts of a plant following soil contamination; is the result of a series of closely co-ordinated steps from root uptake, transport across the root and transport to the aerial parts of the plant. Numerous factors can affect the transfer of radioactivity from soil to fruits, such as chemical-physical characteristics of the soil (soil moisture, pH, Cation Exchange Capacity - CEC, redox potential, quantity of organic matter, microbial activity and fertilizer application), root interception with chemical elements, ion uptake by roots, ion transport across root membranes and its transport in the vascular system of the xylem and phloem, etc (Carini, 2001).

Several experimental studies regarding the element or radionuclide uptake by fruit plants have been carried out in the last decades. Soil to plant transfer factor values for temperate and tropical environments and for a wide combination of soil types, plant groups, and plant compartments, have been collected in databases developed by the US Nuclear Regulatory Commission (Ng et al., 1979), the International Union of Radioecology (Frissel and van Bergeijk, 1989; Frissel et al., 2002) and the International Atomic Energy Agency (IAEA, 2010); with the aim of identifying the main variables and processes affecting the behavior of radionuclides in fruit plants. In spite of the soil-to-fruit transfer is nuclide specific, it is important to notice that when the transfer factors are calculated, the procedure presumes that the concentration in the soil remains constant over time. This means that soil-to-fruit transfer factor available in literature is defined at the equilibrium. Furthermore, when food-chain models for describing key processes in radionuclides transfer are applied, a steady state in the radionuclide concentration in each compartment is assumed (Ng et al.,

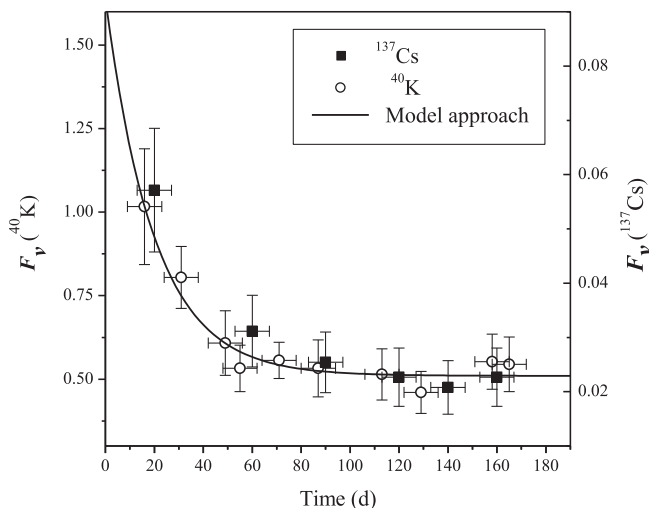


Fig. 5. Temporal dependence of ^{137}Cs and ^{40}K soil-to-fruit transfer factor for tropical lemon trees.

1979). A wide variability of F_v values was found for all radionuclide or element studied, showing variations of up to 5 orders of magnitude for different combinations of soil and plants. For temperate woody trees, for instance, F_v values for radiocaesium range from 8.6×10^{-4} to 8.0×10^{-2} (Carini, 2009; IAEA, 2010). In contrast, they range from 5.0×10^{-2} to 8.7 in tropical and subtropical environments (IAEA, 2010; Velasco et al., 2009). While this variability has been recognized by many authors and assigned to the various factors cited above, different hypotheses have been proposed in order to reduce the uncertainty (Frissel and van Bergeijk, 1989; Ehlken and Kirchner, 1996, 2002; Ciuffo et al., 2002; Velasco et al., 2004; Carini et al., 2005), since it is certainly propagated to the estimation of radionuclide accumulation in the subsequent steps of the food chain and, consequently, to the calculation of human exposure to radionuclide via the food chain (Carini, 2001, 2010; Shaw and Bell, 2001). In the situation of our study, all parameters related to soils properties are fixed, since it is not expected significant changes in these parameters during the fruit growth period. The F_v values as a function of time were presented in Fig. 5. Curves were obtained using Eq. (8). The experimental and theoretical values indicate that $F_v(t)$ for ^{40}K and ^{137}Cs is age-dependent. Moreover, $F_v(t)$ behavior of ^{137}Cs is proportional to ^{40}K , decreasing with increasing the age of fruits. On the other hand, while the variability of F_v values in literature for ^{137}Cs in tropical and subtropical environments is around 2–3 orders of magnitude and even larger, the variability of F_v calculated in our research is lower: twice, three times. This indicates that although the variability of F_v during the fruit growing-ripening is lower than the variability caused by soil properties, this fact must be taken into account in Radioecological studies.

According to IUR recommendation (Frissel and van Bergeijk, 1989; Frissel et al., 2002), F_v must be measured when the product is ready to be consumed. In spite of Fig. 5 shows clearly that close to the final mature stage the variation of F_v is minimum, we believe that the fact of fruit can be harvested and consumed at different stages of maturation should be considered. While in radioecology it is considered that the fruits are harvested and eaten when they reach maturity, this is not strictly true. Many fruits are collected before the complete maturation stage. This stage is usually achieved during the fruit transport, the storage period or when they are exposed for sales. Moreover, there are others examples of fruits that are consumed at different stages of

development, such red and green peppers (used intensively in foods), green and red chili peppers (researches have shown that the concentration of green chili pepper is significantly higher than red chili pepper, see Carvalho et al., 2006), olives (green and black) and those used to make olive oil, fruits (citrus) collected for juice, not necessarily with the same maturation stage and canned fruits and vegetables.

Additionally, Fig. 5 shows that in the beginning of fruit development period, F_v is about 17 times greater for ^{40}K than for ^{137}Cs . In the maturation period this difference seems to increase, being the soil-to-fruit transfer factor for ^{40}K about 25 times higher than for ^{137}Cs . Since $F_v(t)$ is a dimensionless parameter, the comparison between F_v values for ^{137}Cs and ^{40}K does not depend on concentration units of the elements involved, indicating that although the plants absorb and distribute ^{137}Cs and ^{40}K [or total potassium, according to Eq. (2)] in fruits similarly, they show a greater affinity for K than ^{137}Cs . Just now confirming the behavior observed by Shaw and Bell (1989), where the lemon tree has a capability for discriminating between K^+ and $^{137}\text{Cs}^+$, giving preference to the first ion. This evidence is also in agreement with Zhu et al. (2002), according to whom potassium, used in soils in relatively large quantities as fertilizer, is believed to be effective in inhibiting the uptake of radiocaesium, due to the ability of their ions to block radiocaesium uptake by plant roots.

4. Conclusions

In this study we examine the temporal dependence of ^{137}Cs and ^{40}K soil-to-fruit transfer factor for tropical lemon trees. It aims to partially explain the wide variability observed in this key parameter from the experiments. We conclude that F_v for ^{40}K , at the initial stage of fruit development, is more than twice that in the fruit maturation stage, while F_v for ^{137}Cs is three times higher. In spite of the chemical analogies between potassium and cesium, F_v is at least one order of magnitude higher for ^{40}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 weight growth for fruits. The corresponding parameters were obtained in comparing 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 model was proposed to describe the evolution of ^{40}K and ^{137}Cs activity concentrations in fruits and then the activity in the whole fruit as a function of time. The agreement with the experimental data is satisfactory. Then the radionuclide uptake rate was calculated for both radionuclides. Finally, the transfer factor was calculated 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. Further studies should be performed with other plant species in order to confirm the trends obtained in the present investigation.

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