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Temporal trends of ^{137}Cs and ^{40}K activity flux from soil to plant in grassland ecosystems

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Abstract

A data set of ^{137}Cs and ^{40}K activity values in plant and soil samples, obtained under identical conditions during a four-year period, was analysed in order to determine the temporal trend of the soil-to-plant transfer process. The aggregated transfer factor (*Tag*) was determined in each sampling site, showing the typical seasonal variations and the well-known uncertainties. The activity flux density (*FD*) and the activity flux density per soil activity superficial concentration (*J*) were used as more suitable variables to quantify the radionuclide soil-to-plant transfer process. A negative correlation between $\log J(^{137}\text{Cs})$ and *t* was obtained ($R = -0.99$). In contrast, $\log J(^{40}\text{K})$ remains approximately constant with time ($R = -0.67$). The influence of the ^{40}K content in soil on ^{137}Cs soil-to-plant transfer is discussed. Temporal mean value of *FD* was 50 times larger for ^{40}K than for ^{137}Cs . However, comparing temporal mean values, $J(^{40}\text{K})$ was 26 times $J(^{137}\text{Cs})$. The soil concentration level of ^{137}Cs and ^{40}K explains only 50% of the higher uptake of K than Cs in plants. The $J(^{40}\text{K}):J(^{137}\text{Cs})$ relationship was five for the first year and 32 for the last year, a behaviour that could be attributed to the radiocaesium fixation to soil components.

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Keywords: Flux density; Soil-to-plant transfer; Transfer factor; ^{137}Cs ; ^{40}K

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1. Introduction

Radionuclide soil-to-plant transfer constitutes a crucial process in the assessment of human exposure to radionuclides via the food chain. The incorporation of radionuclides into plant tissues from contaminated soils is usually evaluated through the calculation of soil-to-plant transfer factors. Two parameters are most frequently used to assess this transfer process: the aggregated transfer factor (*Tag*) and the concentration ratio (*CR*). These parameters are calculated for the uppermost layer using a standardised thickness (Steiner et al., 2002).

Transfer factors constitute useful tools to obtain basic information on the instantaneous uptake of radionuclides by plants, mainly in the first years after deposition. Values obtained of these empirical quantities are part of the two principal databases established by the US Nuclear Regulatory Commission (Ng et al., 1979) and by the International Union of Radioecology (Frissel and van Bergeijk, 1989; Frissel, 1992). Due to the large quantity of *Tag* (or *CR*) values determined (more than 7000 in the IUR database) and the wide range of values obtained in similar environments, the use of these parameters to predict the soil-to-plant transfer through mathematical models is limited. Unfortunately, the uncertainty in *Tag* values propagates the same error in the calculation of radionuclide accumulation in the following steps of the food chain (Shaw and Bell, 2001).

One of the main causes of this variability can be attributed to the time variable. First, the transfer process shows an appreciable seasonal variability (Strebl et al., 2002). On the other hand, the changes in the radionuclide bioavailability, caused by both their downward mobility and their fixation to soil constituents, lead to a diminution with time of the radionuclide plant uptake (Frissel, 1992; Ehlen and Kirchner, 1996, 2002).

With the aim of eliminating seasonal fluctuations, we introduce the annual activity flux density, *FD* ($\text{Bq m}^{-2} \text{y}^{-1}$), and *J* (y^{-1}), the activity flux density per soil activity concentration. Soil remediation or impact on a large scale after an accidental radionuclide release should be better quantified with *FD* and *J* if these quantities show, as in our case, a lesser variability than *Tag* or *CR*.

In the present study, we have analysed an important data set of radiocaesium (^{137}Cs and ^{134}Cs) and radiopotassium (^{40}K) activity concentrations in soil and vegetation samples obtained during a four-year period. Sampling stations were situated in the northeastern part of Italy. More than 800 activity values make up the database. Samples were taken by the same people, using identical methodology, and radionuclide activities were determined in the same laboratory.

2. Materials and methods

The data set used in this study was collected during the period 1989–1992, in upland meadows situated in the mountains of the Friuli Venezia Giulia region in the northeast of Italy (Fig. 1). The study area is located in semi-natural grasslands, where the dominant vegetation consists of herbaceous leguminous (*Medicago sativa*, *Trifolium repens*, *T. pratensis*) and gramineous species used for hay pro-

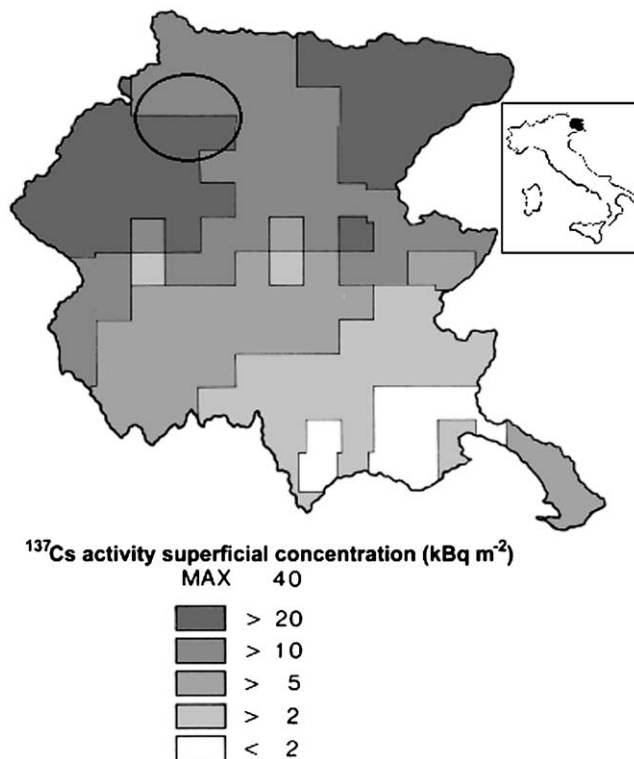


Fig. 1. Location of the study area and deposition of ^{137}Cs (kBq m^{-2}) from the Chernobyl accident (ENEA, 1989).

duction. The sampling area was divided into nine sites (identified with the first nine letters). All the stations were located between 400 and 500 m above sea level. The soils were of the Sand or Sand–Loamy Sand type, derived from calcareous rocks. Table 1 shows the main soil characteristics, including organic matter content, exchangeable cations and total nitrogen. The similarity of these characteristics permits one to assume that soil properties were homogenous in the nine analysed sites.

Soil and plant samples were taken while the crop was growing, two or three times a year. In each meadow, an area of $10 \text{ m} \times 10 \text{ m}$ was defined. The soil sample consists in a monolith of $30 \text{ cm} \times 30 \text{ cm}$ surface area. Each one was located in the four corners and in the centre of the selected area. Then, they were divided into three layers of different depth (L_1 : 0–2.5 cm, L_2 : 2.5–7.5 cm and L_3 : 7.5–12.5 cm). We considered that root activity levels are confined to the upper 10 cm zone (Lembrechts et al., 1991). The five layers of the same depth obtained in this way were merged into one sample, air-dried and sieved in a 2-mm mesh, using standard procedures. Soil bulk densities were calculated by determining the mass of dried soil sample per fresh volume of soil (McGee et al., 1995).

Table 1
Soil properties for each sampling site

Sampling site	Grain size distribution (%)			Soil type	Organic matter (%)	Cations (ppm)			Total N (mg/100g soil)
	Sand	Silt	Clay			Exchangeable K	Exchangeable P	Ca	
A	93.1	6.0	0.9	Sand	7.1	98	13.6	856	140
B	87.5	8.7	3.8	Loamy Sand	5.6	102	17.5	773	112
C	93.3	5.5	1.2	Sand	6.8	110	18.2	884	129
D	90.5	7.7	1.8	Sand	7.6	120	21.4	840	143
E	91.5	7.3	1.2	Sand	6.8	117	15.7	864	121
F	90.6	7.7	1.7	Sand	6.3	119	18.7	788	126
G	87.8	9.7	2.5	Sand–Loamy Sand	7.3	100	18.0	826	143
H	87.0	11.9	1.1	Sand–Loamy Sand	6.9	–	–	787	–
I	82.2	12.8	5.0	Loamy Sand	6.6	119	17.0	811	127

Depending on the meadow productivity, vegetation samples were collected from an area of 1 or 4 m². The samples were dried at 105 °C for 24 h and ground. Likewise, the annual biomass density, $BD(\text{kg (d.w.) m}^{-2} \text{ y}^{-1})$, was determined yearly for each site. All samples were analysed by high-resolution gamma spectrometry, using HPGe detectors. The counting time was selected in order to achieve counting errors lower than 5% on ¹³⁷Cs. All activities of Cs isotopes were decay-corrected to 1 May 1986. The total ¹³⁷Cs soil activity was separated into the weapons fallout and Chernobyl contributions using the ¹³⁷Cs:¹³⁴Cs ratio of 2, measured in air and rainfall samples gathered in May 1986.

The Rabes I model (Toso and Velasco, 2001) was used to simulate the radio-caesium vertical migration process. This model assumes that, for every time, the radionuclide vertical distribution pattern can be approximated by a decreasing exponential function. The relaxation length L (cm) of the distribution depends on time t as:

$$L(t) = L_R[1 - \exp(-k_R t)] \quad (1)$$

where L_R (cm) is the relaxation depth in the equilibrium and $k_R(\text{d}^{-1})$ is the rate of change of the vertical profile. The dependence of relaxation depth with the ¹³⁷Cs soil-to-plant transfer process has been noted in previous studies (Schuller et al., 2002).

The dynamic process of root uptake was determined using the Tag , which is usually defined as (Steiner et al., 2002):

$$Tag = \frac{\text{Activity concentration in plant (Bq kg}^{-1}) \text{ (dry weight)}}{\text{Activity superficial concentration on ground (Bq m}^{-2})} \quad (2)$$

At the same time, the activity flux density from soil to plant, $FD(\text{Bq m}^{-2} \text{ y}^{-1})$, was introduced to quantify this transfer process. For a particular radionuclide, FD represents the activity that passes from soil to plant per square meter and per year. If n is the number of plant samples collected per year, and $Cv_i(\text{Bq kg}^{-1})$ and $BD_i(\text{kg m}^{-2} \text{ y}^{-1})$ are, respectively, the plant radionuclide activity concentration and the biomass density of the plant i -sample, then:

$$FD = \sum_{i=1}^n Cv_i BD_i \quad (3)$$

The activity flux density per soil activity superficial concentration $J(\text{y}^{-1})$ is defined as:

$$J = \frac{FD}{D} \quad (4)$$

$D(\text{Bq m}^{-2})$ is the radionuclide activity superficial concentration in soil. J is a more adequate variable to compare the soil-to-plant transfer in different scenarios of radionuclide contamination.

Table 2
Radionuclide activity in soil and plant, ^{137}Cs relaxation depth in soil (L_R) and aggregated soil-to-plant transfer factors (Tag)

Quantities	Soil layer	Distribution type	Mean	SD	CV % ^a	Range		N^b
						Maximum	Minimum	
$^{137}\text{Cs}_{\text{SCHER}}$ soil activity concentration (Bq kg^{-1})	L ₁	Log normal	699	419	60	2806	229	74
	L ₂	Log normal	179	87	49	461	46	72
	L ₃	Log normal	77	65	84	223	8	73
^{40}K soil activity concentration (Bq kg^{-1})	L ₁	Normal	502	269	54	1070	40	74
	L ₂	Normal	518	283	55	1090	40	72
	L ₃	Normal	526	294	56	1090	8	73
$^{137}\text{Cs}_{\text{SCHER}}$ soil activity superficial concentration (Bq m^{-2})		Log normal	2.7E+04	9.6E+03	36	9.1E+04	1.3E+04	74
^{40}K plant activity concentration (Bq kg^{-1})		Normal	5.6E+04	3.8E+04	68	1.9E+05	3.2E+03	74
^{137}Cs plant activity concentration (Bq kg^{-1})		Log normal	21	31	148	194	0.80	59
^{40}K plant activity concentration (Bq kg^{-1})		Normal	578	174	30	997	210	63
L_R (cm)		Log normal	3.0	1.8	60	6.7	0.8	74
$Tag^{137}\text{Cs}$ ($\text{m}^2 \text{ kg}^{-1}$)		Log normal	9.27E-04	1.47E-03	159	9.53E-03	4.45E-05	59
$Tag^{40}\text{K}$ ($\text{m}^2 \text{ kg}^{-1}$)		Normal	1.55E-02	1.12E-02	72	9.19E-02	4.19E-03	63

^a Coefficient of variation (= SD/mean $\times$ 100).

^b Number of samples.

3. Results and discussion

Table 2 shows the obtained results for the distribution type and statistical quantities associated with ^{137}Cs and ^{40}K activity values in each soil layer, soil activity superficial concentration, concentration in plants, and T_{ag} and L_{R} values for ^{137}Cs .

3.1. Soil and plant activity

In soil samples, only the component of ^{137}Cs attributable to the Chernobyl accident ($^{137}\text{Cs}_{\text{CHER}}$) was considered to assess the transfer of radiocaesium from soil to plant and its temporal variation (Colgan et al., 1990; Belli et al., 1994). A log normal distribution for $^{137}\text{Cs}_{\text{CHER}}$ activity concentration data was obtained. Instead, the soil concentration of ^{40}K showed a normal distribution for the three layers. In general, the data set available indicates that radiocaesium is distributed within the first centimetres of soil and that its vertical distribution with depth can be acceptably fitted by an exponential decrease function (Velasco et al., 1993, 1997; Toso and Velasco, 2001). As was expected, the $^{137}\text{Cs}_{\text{CHER}}:^{137}\text{Cs}_{\text{TOTAL}}$ relationship decreases with soil depth. Fig. 2(a) shows $^{137}\text{Cs}_{\text{CHER}}$ vs. $^{137}\text{Cs}_{\text{TOTAL}}$ for the first soil layer: 0–2.5 cm (L_1). The points are optimally fitted with a simple linear regression ($R = 0.99$). The slope value obtained is $B_1 = 0.93$, indicating that 93% of the ^{137}Cs present in L_1 corresponds to the $^{137}\text{Cs}_{\text{CHER}}$ fraction. One of the sampling sites (site D) presents, in the superficial soil layer, radiocaesium activity concentration values substantially greater than all other sites ($>2500 \text{ Bq kg}^{-1}$) (points situated on the upper-right sector in Fig. 2(a)). Fig. 2(b) shows the same variables for L_2 (second soil layer: 2.5–7.5 cm), where $B_2 = 0.80$ ($R = 0.94$); therefore, 80% ^{137}Cs activity corresponds to the $^{137}\text{Cs}_{\text{CHER}}$ fraction. For L_3 (third soil layer: 7.5–12.5 cm) (Fig. 2(c)) a larger dispersion of the points was found, and the straight slope of the linear regression was $B_3 = 0.66$, with a lower correlation coefficient ($R = 0.88$). In L_3 , 66% of the ^{137}Cs activity measured corresponded to the $^{137}\text{Cs}_{\text{CHER}}$ fraction. The linear relation between both variables in each soil layer

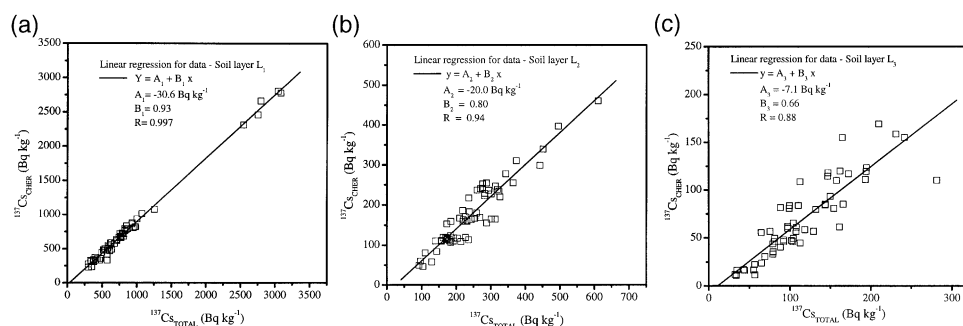


Fig. 2. ^{137}Cs in soil attributable to Chernobyl deposition vs. ^{137}Cs total content in soil. (a) Soil layer L_1 : 0–2.5 cm; (b) soil layer L_2 : 2.5–7.5 cm; (c) soil layer L_3 : 7.5–12.5 cm.

was used to determine the $^{137}\text{Cs}_{\text{CHER}}$ fraction in samples where ^{134}Cs was not detected.

Both $^{137}\text{Cs}_{\text{CHER}}$ soil activity concentration (Bq kg^{-1}) and $^{137}\text{Cs}_{\text{CHER}}$ soil activity superficial concentration (Bq m^{-2}) showed considerable variability (Table 2). This variability is marked both within one site and among different sites. Large variations in activity concentration levels of radiocaesium derived from Chernobyl within local areas have been reported (Clark and Smith, 1988; Block and Pimpl, 1990; Colgan et al., 1990; McGee et al., 1991, 1995; Monte, 1991).

The Rabes I model (Toso and Velasco, 2001) was used to simulate the temporal change of the $^{137}\text{Cs}_{\text{CHER}}$ soil vertical profile. In all sampling sites, it was found that the equilibrium state was reached. L_R remains approximately constant with a weak tendency to decline with time. Fig. 3(a) shows the frequency histogram for L_R considering all sampling sites and Fig. 3(b) shows the temporal trend of L_R . The tendency of relaxation depth to decline with time could be attributed to two factors. First, the radionuclide plant uptake takes place immediately below the superficial layer. In the present study we found that, on average, about $60 \text{ Bq m}^{-2} \text{ y}^{-1}$ passed from soil to plants in the first sampling time. Second, the radionuclide physical decay leads to a progressive impossibility to detect ^{137}Cs activity in the deeper layers, where the activity values are close to the equipment detection limit.

The ^{40}K soil distribution shows a more homogeneous distribution among layers in the same station (Nimis et al., 1994). Compared with ^{137}Cs soil distribution, ^{40}K is more uniform in a horizontal microscale. The different patterns observed for ^{137}Cs and ^{40}K in the soil cannot be explained by the different soil properties. All analysed soils were of the Sand or Sand-Loamy Sand type and their physico-chemical properties are similar (Table 1). The variability of ^{40}K among stations could be due to differences in land management, for example, fertilization and plowing practices. ^{40}K activity concentration shows a weak tendency to increase with depth. The general pattern of this radionuclide is consistent with the fact that

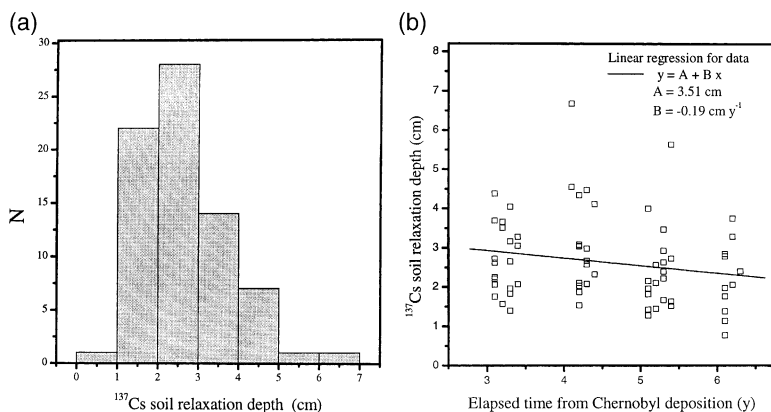


Fig. 3. (a) ^{137}Cs soil relaxation depth: frequency histogram; (b) ^{137}Cs soil relaxation depth vs. time from Chernobyl deposition.

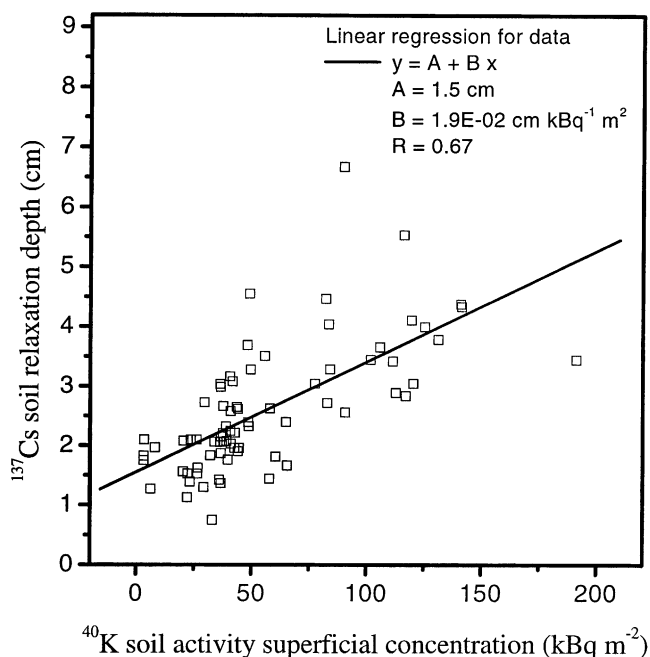


Fig. 4. ^{137}Cs soil relaxation depth vs. ^{40}K soil activity superficial concentration.

being a natural element in the soil, it has reached the equilibrium between the solid and the liquid phase in the soil layers.

A positive correlation was obtained between the ^{40}K soil activity superficial concentration and the ^{137}Cs relaxation depth ($R = 0.67$) (Fig. 4). This phenomenon has been extensively recognised and it is attributable to the competitive effect between these two ions (Coughtrey and Thorne, 1983; Cawse, 1990; Heaton et al., 1990; McGee et al., 1995). Cs^+ competes with K^+ for the exchange sites, in the complex of interchange of the soil matrix. Assuming a constant relationship between K-radioactive and K-stable, it can be concluded that high contents of ^{40}K in soil lead to greater leaching of ^{137}Cs to deeper layers.

Table 2 shows the statistical values associated with radiocaesium and radiopotassium plant activity concentrations. The ^{137}Cs plant concentration shows a log normal distribution, while the ^{40}K plant concentration was better fitted by a normal distribution. The plant activity concentration was almost 27 times greater for ^{40}K than for ^{137}Cs ; however the coefficient of variation was one order of magnitude less for ^{40}K . The wide difference among the variability of plant activities cannot be explained from the difference observed in the variability associated with the soil activity concentration for both radionuclides.

Despite the high variability observed in plant concentration, it is possible to infer that ^{137}Cs plant activities decrease with the time elapsed since the soil deposition event. This behaviour has been previously reported. Cremers et al. (1990), Heaton

et al. (1990), Henrich et al. (1990), Antonopoulos-Domis et al. (1990), Sanzharova et al. (1994), Antonopoulos-Domis et al. (1997) and Askbrant and Sandalls (1998) attributed this phenomenon to the decrease in availability of this radionuclide for plants with time. Other authors have argued that the ^{137}Cs temporal trends in plants can be explained by the distribution change of this radionuclide within the rooting zone and their evolution with time. In some situations, previous studies indicate that ^{137}Cs activity concentration in plants decreases with time by the migration to deeper soil layer rather than by fixation to mineral clay (Ehlken and Kirchner, 1996, 2002).

3.2. Soil-to-plant transfer

Tag for ^{137}Cs and ^{40}K were determined in each sampling site. The distribution of $\text{Tag}(^{137}\text{Cs})$ was log normal, while that of $\text{Tag}(^{40}\text{K})$ was normal (Table 2). $\text{Tag}(^{40}\text{K})$ values were in all cases greater than for $\text{Tag}(^{137}\text{Cs})$. Mean of $\text{Tag}(^{40}\text{K})$ was more than 17 times higher than mean of $\text{Tag}(^{137}\text{Cs})$. This difference confirms the existence of a selection phenomenon between the two ions, in favour of K, by plant roots (Nimis et al., 1994).

Tag values, both for ^{137}Cs and ^{40}K , showed marked variability among samplings. Likewise, $\text{Tag}(^{137}\text{Cs})$ values present high annual variability in the same sampling sites. Considering all the sampling stations, we obtained a difference of two orders of magnitude between maximum and minimum $\text{Tag}(^{137}\text{Cs})$ with a coefficient of variation of 159. In the case of $\text{Tag}(^{40}\text{K})$, the relationship between maximum and minimum values was about 22-fold and the coefficient of variation was 72. As expected, *Tag* seasonal variability was observed for both radionuclides. *Tag* was lower in late spring, and increased in summer and late summer–early autumn. This increase of *Tag* corresponded to the increment in the activity level and of the requirement of nutrients by vegetal tissue. A similar seasonal variability of radio-caesium transfer factors has been reported in previous studies (Strebl et al., 2002). The *Tag* seasonal variability has usually been attributed to two factors: the physiology and the development of plants. Plants present different and distinctive activity patterns. Activity is low or null in winter and autumn, the seasons with adverse conditions for growth and reproduction. In spring and summer, the activity level gradually increases, reaching its highest point in summer (Salisbury and Ross, 1994). A positive correlation has been found between plant nutrients uptake and increment of plant activity (Colgan et al., 1990; van Bergeijk et al., 1992).

Several authors have recognised that ^{137}Cs soil-to-plant transfer is time dependent (Rosén et al., 1999; Ehlken and Kirchner, 2002; Frissel et al., 2002; Strebl et al., 2002). In the sampling sites analysed in this study, the dependence of $\text{Tag}(^{137}\text{Cs})$ with time from deposition is not clear.

Table 3 shows the *FD* and *J* values (mean and SD), calculated yearly, for ^{137}Cs and ^{40}K considering all sampling sites. *FD* and *J* showed a normal distribution for both radionuclides. For all time and for all sampling sites, except site D, $\text{FD}(^{40}\text{K})$ was greater than $\text{FD}(^{137}\text{Cs})$. Comparing the soil-to-plant activity flux density for

Table 3
Activity flux density and activity flux density per soil activity superficial concentration for ¹³⁷Cs and ⁴⁰K. Mean values and standard deviations for each sampling time. Initial time corresponds to the Chernobyl deposition time (May, 1986)

Sampling time, y	$FD(^{137}\text{Cs}), \text{Bq m}^{-2} \text{y}^{-1}$		$FD(^{40}\text{K}), \text{Bq m}^{-2} \text{y}^{-1}$		$J(^{137}\text{Cs}), \times 10^{-4} \text{y}^{-1}$		$J(^{40}\text{K}), \times 10^{-4} \text{y}^{-1}$	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1	59	136	294	194	15.0	26.7	78.8	50.9
2	22	46	207	100	5.6	8.1	54.1	22.0
3	14	22	289	147	3.2	2.6	62.7	27.2
4	4	2	358	261	1.6	0.7	51.6	16.8

both radionuclides, we obtained that the ratio $FD_t(^{40}\text{K}):FD_t(^{137}\text{Cs})$ was 50, when FD_t stands for the temporal mean value of FD considering all sampling sites.

Likewise, for all sampling sites $J(^{40}\text{K})$ was larger than $J(^{137}\text{Cs})$. The ratio $J_t(^{40}\text{K}):J_t(^{137}\text{Cs})$ was 26. These results make it clear that, in the analysed situations, only 50% of the observed difference between the soil-to-plant activity flux density for ⁴⁰K and for ¹³⁷Cs could be attributed to the larger ⁴⁰K soil activity superficial concentration.

Potassium content in soil limits the radiocaesium soil-to-plant transfer process. Fig. 5 shows $J(^{137}\text{Cs})$ as a function of ⁴⁰K soil activity superficial concentration.

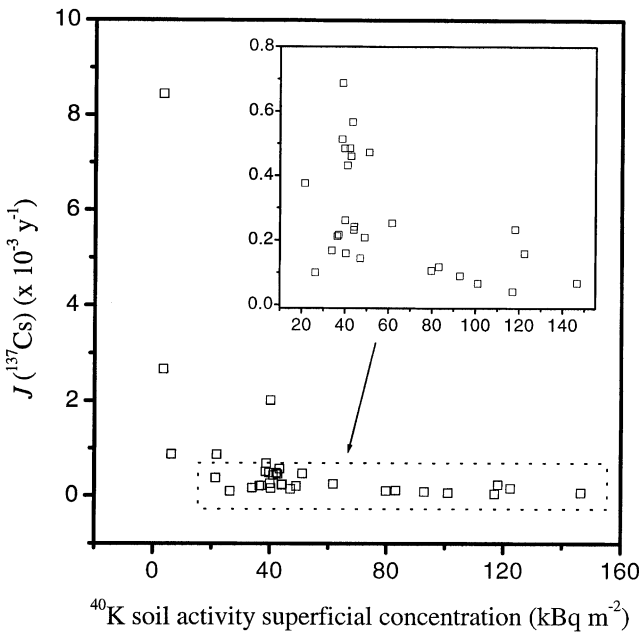


Fig. 5. Plot of $J(^{137}\text{Cs})$ vs. ⁴⁰K soil activity superficial concentration.

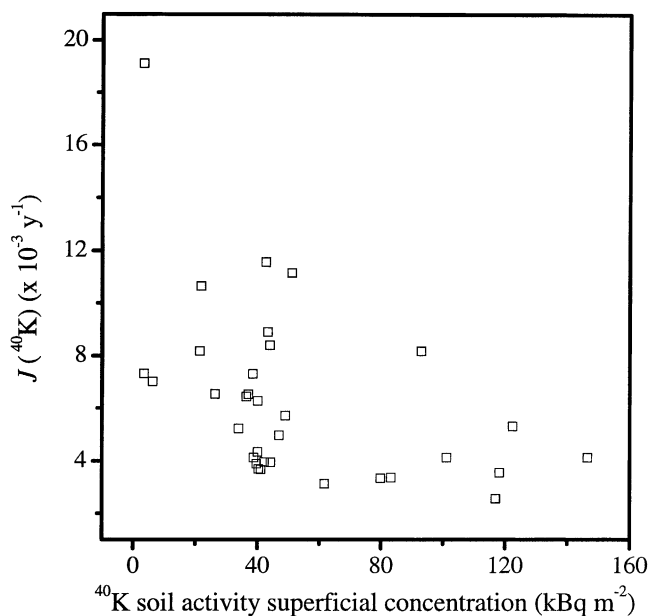


Fig. 6. Plot of $J(^{40}\text{K})$ vs. ^{40}K soil activity superficial concentration.

This transference falls abruptly for ^{40}K soil activity superficial concentration values up to 50 kBq m^{-2} . For values greater than 50 kBq m^{-2} , $J(^{137}\text{Cs})$ exhibits a smooth tendency to the minimum value.

For the analysed data set, $J(^{40}\text{K})$ was not found to depend on ^{137}Cs soil content. In contrast, $J(^{40}\text{K})$ shows a tendency to decrease with the increment of ^{40}K soil content (Fig. 6). It is known that uptake by plants is a very complex mechanism which cannot be explained only by concentration of nutrients in soil. Many intrinsic factors regulate this process. In our case, a saturation phenomenon in the uptake of potassium is observed (Salisbury and Ross, 1994).

In order to determine the possible influence of ^{137}Cs soil vertical distribution on the transfer of this radionuclide from soil to plant, $J(^{137}\text{Cs})$ was plotted vs. the ^{137}Cs relaxation depth in soil (Fig. 7). An inverse relationship between these variables was obtained. This result does not agree with the relation found between the ^{137}Cs concentration ratio plant:soil and the ^{137}Cs relaxation depth in Chilean soils (Schuller et al., 2002). This difference could be explained by the fact that in the present study we considered the component of ^{137}Cs derived only from the Chernobyl accident (this component was not detected in South American soils). Consequently, at the sampling time, radiocaesium remained available for plants in the superficial soil layer, where the root density in grassland environments is greater. An increment of the leaching process leads to an increase of the radiocaesium

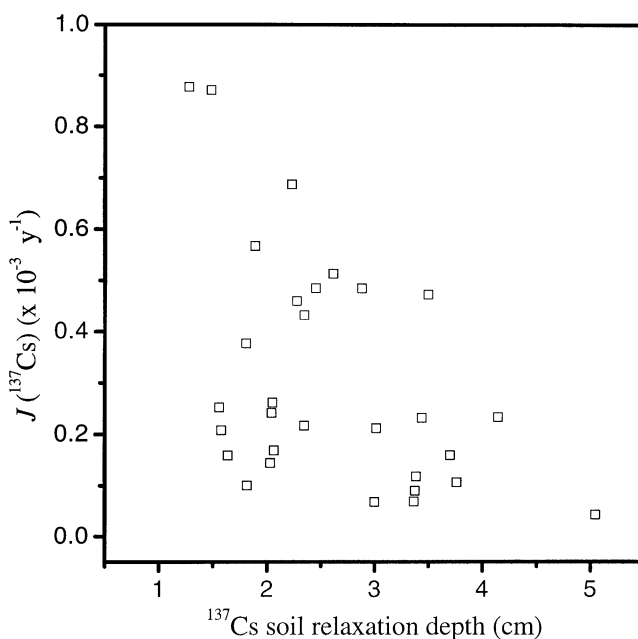


Fig. 7. Plot of $J(^{137}\text{Cs})$ vs. ^{137}Cs soil relaxation depth.

relaxation depth and, consequently, a reduction of the soil-to-plant transfer can be expected.

3.3. Temporal trend of FD and J

Table 3 shows the values obtained yearly for FD and J , for both radionuclides. The $FD(^{40}\text{K}):FD(^{137}\text{Cs})$ ratio was 5 in the first sampling years and it increased with time reaching a value close to 90 in the last sampling year (Fig. 8). The $J(^{40}\text{K}):J(^{137}\text{Cs})$ ratio showed a similar behaviour but with a more attenuated increase with time due to the continuous temporal decrease in the ^{137}Cs soil activity superficial concentration. $J(^{40}\text{K}):J(^{137}\text{Cs})$ was 5.2 for the first sampling year and reached about 32 for the last sampling year (Fig. 8). The temporal change in these relationships could be explained taking into consideration the gradual fixation process of radiocaesium to soil components and its gradual unavailability for plants.

Fig. 9 shows the temporal variation of $\log J(^{40}\text{K})$ and $\log J(^{137}\text{Cs})$. In the first case, no substantial variation with time was found. A straight slope of -0.05 ($R = -0.67$) was obtained from linear regression of the data. Potassium remains equally available to plants with time. Meaningfully, a very good linear fit was found when $\log J(^{137}\text{Cs})$ vs. t was considered. In this case, a negative straight slope of -0.33 with a correlation coefficient of $R = -0.99$ was obtained.

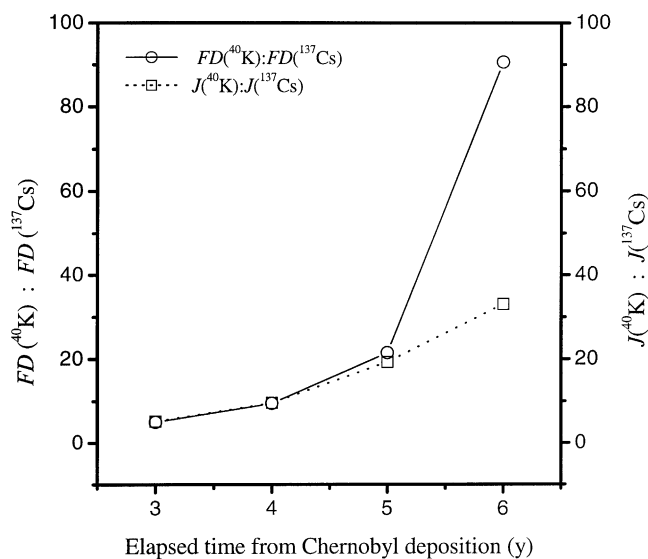


Fig. 8. Temporal trend for the $FD(^{40}\text{K}):FD(^{137}\text{Cs})$ (○), and $J(^{40}\text{K}):J(^{137}\text{Cs})$ (□) relationships for the sampling period.

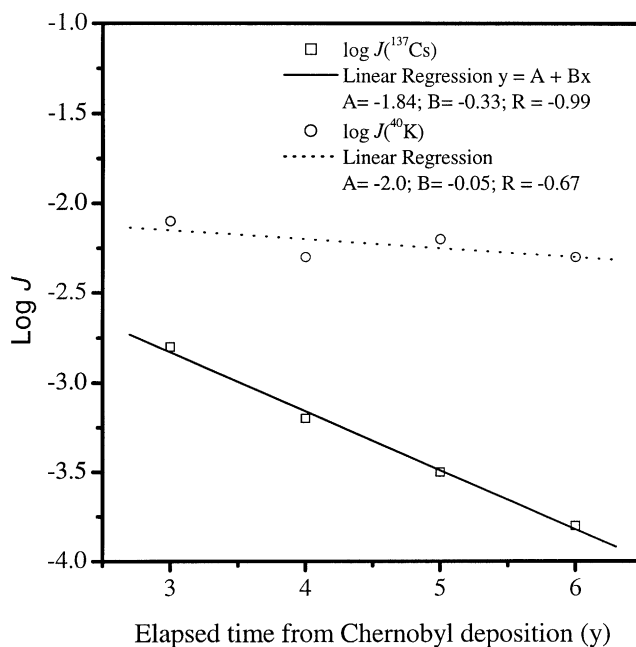


Fig. 9. Temporal trend of $\log J(^{40}\text{K})$ and $\log J(^{137}\text{Cs})$ for the sampling period.

4. Conclusions

A large set of empirical data of ^{40}K and ^{137}Cs activity concentration in plants and soils sampling has been reviewed in order to assess the radionuclide soil-to-plant transfer process in semi-natural environments and its temporal dependence.

The main results obtained are:

- Different distribution patterns were obtained for ^{40}K and ^{137}Cs in soil. While radiopotassium presents an approximately uniform distribution with depth, radiocaesium vertical profile was fitted with a decreasing exponential function and relaxation depth was determined in each case.
- *Tag* values were calculated in all sampling sites. This coefficient shows a wide variability with pronounced seasonal changes. On average, *Tag* was 17 times larger for ^{40}K than ^{137}Cs .
- Activity flux density was introduced to evaluate the annual activity that passes from soil to plants. This variable presents lower variability than *Tag* and it permits to better assess the influence of time on this transfer process. *FD* values for ^{40}K and for ^{137}Cs were obtained and a comparative analysis was carried out.
- A linear relationship with negative slope was found between $\log J(^{137}\text{Cs})$ and the elapsed time since radiocaesium deposition. On the other hand, $\log J(^{40}\text{K})$ remained approximately constant with time.

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