

## Radioactive trace in semi natural grassland. Effect of $^{40}\text{K}$ in soil and potential remediation

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**Keywords:**  $^{137}\text{Cs}$  plant uptake, radioactive traces, radiocaesium, radiopotassium.

**Abbreviations:** CV: variation coefficient  
GM: geometric mean  
TF: transfer factor

The uptake of radionuclides by plant roots constitutes the main pathway for the migration of radiocaesium from soil to humans, via food chain. In this study we assessed radiocaesium uptake by plant in order to piece together information on factors affecting uptake processes, particularly K supply and differential uptake among different plant species. Soil contaminated by the Chernobyl accident and forage from a semi-natural alpine grassland, situated in Tarvisio, Italy, were sampled during 1999. Under field conditions,  $^{137}\text{Cs}$  uptake for Gramineae and *Taraxacum officinale* seem to behave in a comparable way. Higher  $^{40}\text{K}$  soil activity concentration leads to a lower  $^{137}\text{Cs}$  plant uptake, suggesting an inhibitory pattern of potassium on radiocaesium plants uptake. For forage samples, a similar tendency was observed. We analyzed the influence of the ratio of  $^{137}\text{Cs}/^{40}\text{K}$  in soil on  $^{137}\text{Cs}$  plant uptake. Under field conditions, the ratio observed varied in a range of 0.5 to 1.3. For most of the species, at higher  $^{40}\text{K}$  soil concentration a lower  $^{137}\text{Cs}$  uptake was observed, a fact that reflects the resulting effect of the complexity of factors controlling ion absorption from soil.

The understanding of mechanisms that affect the radiocaesium uptake by plant species which growth under field conditions, become a subject of increasing interest. Radiocaesium bioavailability in soil is strongly influenced by soil properties such as K status, clay content, pH and soil organic matter (Absalom et al. 2001). On the other hand, the radiocaesium availability in soil is one of factors controlling ion uptake by plant roots. It is generally accepted that, following the Chernobyl accident, radiocaesium has been retained in soil surface mainly due to reaction with clay and humic components and soil microflora (Thiry and Myttenaere, 1993).

Radionuclides are present in the environment either naturally or artificially.  $^{137}\text{Cs}$  is an artificial radioelement, generated in the past by nuclear weapon testing and the Chernobyl accident. The radioactive isotopes of anthropic origin are introduced into the environment, principally in soil, and generate great concern by the impact of contamination in the environment and the man.  $^{40}\text{K}$  is a natural isotope present in soil and used as a tracer of K, an essential plant's nutrient. Under field conditions, plants can suffer from potassium starvation (or potassium deficiency) periodically or constantly throughout the growing season, due to spatial and temporal variations in the potassium status of agricultural soil (Zhu et al. 2000).

Radionuclides in soil are taken up by plants, thereby becoming available for further redistribution within food

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K enters to the plant roots via ion channels or specific transporters, while Cs<sup>+</sup> uptake takes place by means of K<sup>+</sup> channels. On the other hand, <sup>137</sup>Cs, an artificial radioelement and a non-strict analogue of K, is a competitor at the K<sup>+</sup> uptake plant mechanisms (Smolders et al. 1996a; Smolders et al. 1996b; Marscher, 1998; Zhu and Smolders, 2000).

K<sup>+</sup> and Cs<sup>+</sup> ionic radius are 0.33 nm and 0.31 nm (hydrated ion) respectively, and the absorption rate are 26 μmol g<sup>-1</sup> fresh wt per 3 hrs for potassium and 12 μmol g<sup>-1</sup> fresh wt per 3 hrs for caesium. Despite its smaller diameter, Cs<sup>+</sup> is taken up at a much lower rate than K<sup>+</sup>. Obviously, factors other than ionic diameter are involved in the uptake regulation, one of them being the affinity for membrane-bound carriers, or channels (Marscher, 1998). Consequently, high K concentration in soil acts as inhibitor and prevents Cs uptake. Radionuclide transfer from soil-to-plant is affected by a number of factors, summarized in **Figure 1**. It is well accepted that K<sup>+</sup> uptake by plant roots is a consequence of two different, but parallel, mechanisms present on plasma membranes: a) high-affinity energized transporter system and b) low-affinity mechanism, usually associated with ion-channels (Smolders et al. 1996a; Smolders et al. 1996b; Zhu and Smolders, 2000). Thus, it becomes of great importance, to study the relationship between both elements in soil.

A number of field studies in contaminated areas have been conducted since the Chernobyl accident in order to better understand the behaviour of <sup>137</sup>Cs in soil-plant systems (McGee et al. 1996; Nisbet and Woodman, 2000). In a previous study we performed an analysis of the soil-to-plant relationship in alpine pastures, and compared our results with a similar area in Germany. We observed a very strong negative correlation between <sup>40</sup>K soil activity and <sup>137</sup>Cs

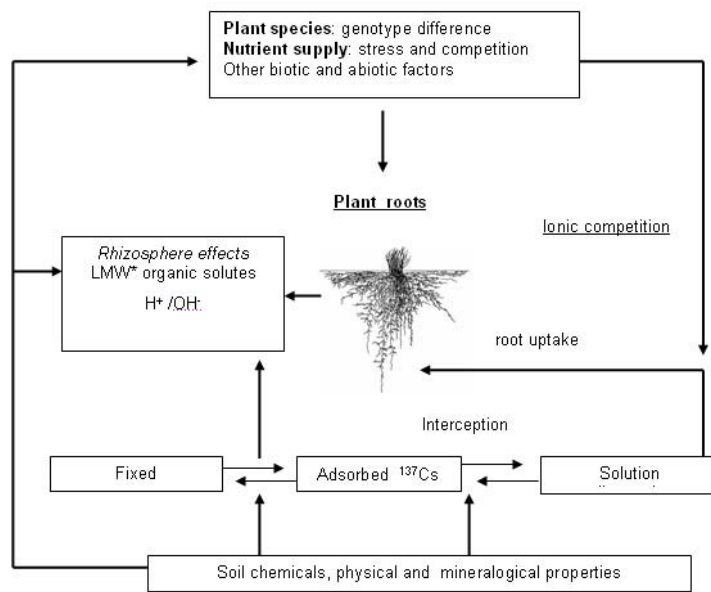
**Table 1. Geometric mean (GM), range of values for <sup>137</sup>Cs and <sup>40</sup>K in soil samples and variation coefficient (CV) (n = 24).**

Radioelement	GM	Min.-Max. values	CV(%)
<sup>137</sup> Cs (Bq m <sup>-2</sup> )	2.4E + 04	1.7E + 04 – 3.4E + 04	16
<sup>40</sup> K (Bq m <sup>-2</sup> )	3.1E + 04	2.0E + 04 – 4.4E + 04	21

plant activity for both pastures (Bunzl et al. 2000). This observation indicates that when a large variety of different plant species is considered, radiocaesium and radiopotassium do not necessarily behave in an analogous way (Bunzl et al. 2000).

Our observations in semi-natural grassland suggest that the Transfer Factor (TF) is a highly variable parameter that appears to be independent of the radionuclide activity in soil (Ciuffo et al. 2002). <sup>137</sup>Cs uptake was assessed for different plant species from two sites in a semi-natural grassland, at the Giulia Alps, Italy, and we found that TF values are within the range of commonly reported values, suggesting that these pastures were apt for forage use. The soil-to-plant relationship for radiocaesium was analyzed because of its direct importance regarding human exposure to radiocaesium through the food chain (Ciuffo et al. 2003).

The present study assess the radiocaesium uptake by plants in order to piece together information on factors affecting the uptake processes, particularly K supply and differences among different plant species. The study was conducted in a semi-natural grassland contaminated by the Chernobyl accident.



**Figure 1. Factors affecting radiocaesium uptake by plant roots. LMW\* =Low molecular weight.**

## MATERIALS AND METHODS

The area of study was situated in the Tarvisio Woodlands, in Friuli -Venezia Giulia region, Italy. Sampling of natural soils and plants was conducted in a semi-natural grassland contaminated by the fallout of Chernobyl accident (mean deposition of approximately  $40 \text{ kBq m}^{-2}$  (Belli et al. 1989; European Communities, 2001).

Soil and grass were sampled simultaneously –within an area of one hectare along three equidistant transect– on the same day (June 1999). Twenty-four parallel samples of soil and grass were taken. Grass samples were collected by cutting the total herbage growing at each  $1 \times 1\text{-m}$  plot, 2 cm above ground, while avoiding contamination with soil. A fraction of each vegetation sample was taken and processed separately. The aboveground plant material was weighed, dried at  $105^\circ\text{C}$  until constant weight and milled before radioactivity quantification.

Soil samples: monoliths of  $15 \times 15 \text{ cm}$  were collected to a

depth of 10 cm. Soil samples were air-dried, ground to pass through a 2-mm sieve, homogenized and weighed.

Dried soil and grass samples were analyzed for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  by gamma spectrometry with a high purity Germanium detector (HPGe). The  $^{137}\text{Cs}$  activity was decay-corrected to the date of the Chernobyl accident, May 1986.

## Statistics

Data were analyzed by using Statgraphic package.

## RESULTS AND DISCUSSION

### Soil characteristic

We performed an analysis of the natural  $^{40}\text{K}$  and artificial  $^{137}\text{Cs}$  radioelements present in soil from an area contaminated by the Chernobyl accident. In a previous study we showed that soil activity deposition expressed in  $\text{Bq m}^{-2}$  provides a more homogeneous value than soil

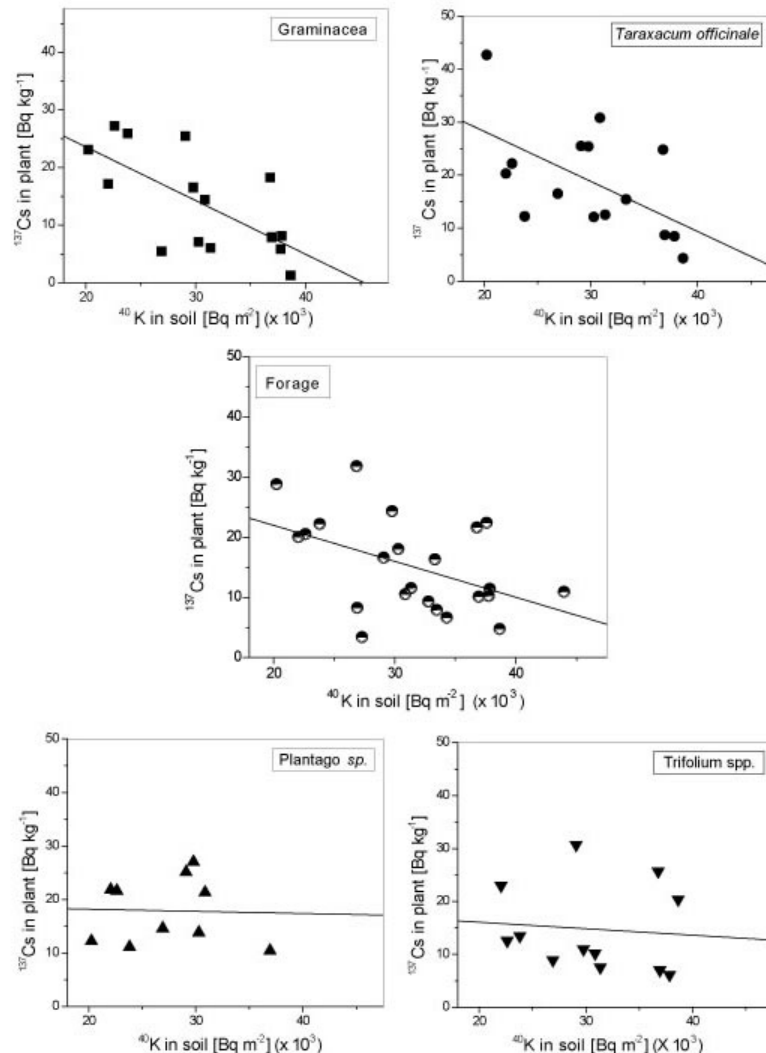


Figure 2: Scattergram of  $^{137}\text{Cs}$  plant activity concentration for forage and different plant and  $^{40}\text{K}$  activity concentration superficial in soil.

activity concentration values (Ciuffo et al. 2002) expressed in Bq Kg<sup>-1</sup> for <sup>137</sup>Cs. Thus, Table 1 shows the variability of <sup>137</sup>Cs activity deposition and <sup>40</sup>K superficial activity concentration in soil from grassland [Bq m<sup>-2</sup>], where the geometric mean (GM), range of values and variation coefficient (CV) are included (n = 24).

### Differential activity by plant species

To understand the effect of <sup>40</sup>K soil content in <sup>137</sup>Cs sorption by plant roots, we performed an analysis for the different plant species present in the sampled area. We are reporting data of the predominant plant species present: *Taraxacum officinale*, *Trifolium* spp., Graminaceae, *Plantago* sp.

We observed that different plant species behave in a different way with respect to <sup>137</sup>Cs sorption against <sup>40</sup>K soil content. The ANOVA evaluation of the <sup>137</sup>Cs activity concentration in plants [Bq kg<sup>-1</sup>] with respect to the <sup>40</sup>K activity present in superficial soil [Bq m<sup>-2</sup>], exhibited a statistically significant relationship between variables for Graminaceae and *Taraxacum officinale* (R= -0.67 and -0.56, respectively; P < 0.05; 95% confidence level). On the other hand, the ANOVA analysis indicates a relatively poor relationship between variables for *Plantago* sp. and *Trifolium* spp. (R = 0.43 and -0.09, respectively). Figure 2 shows the scattergram of <sup>137</sup>Cs plant activity concentration for forage and different plants against <sup>40</sup>K superficial activity concentration in soil.

Graminaceae and *T. officinale*, behave in a comparable way under field conditions, with increased K concentration in soil: higher K soil concentration causes low <sup>137</sup>Cs uptake by plant roots, in agreement with previous results from other authors (Zhu and Shaw, 2000; Zhu and Smolders, 2000; Zhu et al. 2000). However, for forage samples, which comprise mixed species, a statically less significant relationship was observed than for individual species in particular (R= -0.47; P < 0.05; 95% confidence level). Evidently, when forage samples were considered, the individual effect of each species was damped.

### Cs /K ratio in soil

Several authors made a number of contributions, mainly by laboratory experimental studies (Smolders et al. 1996a; Smolders et al. 1996b; Zhu and Shaw, 2000), and demonstrate the inhibitory effect of potassium on the uptake of radiocaesium by plants. The effect is better expressed when <sup>137</sup>Cs activity concentration in plant was related to the ratio of <sup>137</sup>Cs/ <sup>40</sup>K in soil.

To evaluate the effect of the ratio of <sup>137</sup>Cs/<sup>40</sup>K in soil on plant uptake, we analyzed the effect of <sup>40</sup>K concentration increment in soil on <sup>137</sup>Cs absorption by plant, using field experimental values. Under field conditions, the ratio observed varied in a range of 0.5 to 1.3 (Figure 3). We should point out that total <sup>137</sup>Cs and <sup>40</sup>K activities were

considered, the cationic fraction in the solution not was discriminated.

Figure 3 shows a comparison of the mobile means of <sup>137</sup>Cs activity concentration in plant, for different plant species under study and forage samples against ratio of <sup>137</sup>Cs/ <sup>40</sup>K in soil. At higher <sup>40</sup>K concentrations a lower absorption of <sup>137</sup>Cs by plant was observed, an observation valid at low ratio values of <sup>137</sup>Cs/ <sup>40</sup>K in soil. However, for *Plantago* sp. and <sup>137</sup>Cs/ <sup>40</sup>K ratio values in soil higher than 0.8, we observed a sensible decrease in <sup>137</sup>Cs absorption. This observation could be interpreted as a singular characteristic of *Plantago* sp.

The inhibitory pattern of K on Cs uptake by plant roots has been previously described by Sacchi et al. (1997). Several authors (Maathuis and Sanders, 1996; Ichida et al. 1999; Marscher, 1998; Zhu et al. 2000) provide evidence for the presence of two distinct pathways for root K<sup>+</sup> uptake. One system of low selectivity for Cs<sup>+</sup>, Rb<sup>+</sup> and K<sup>+</sup>, a carrier-mediated transporter, which operates at low external concentration of K<sup>+</sup>, probably driven by cation symport with H<sup>+</sup>. A second system highly selective for K<sup>+</sup>, channel mediated, which operates at high external K<sup>+</sup> concentration (Maathuis and Sanders, 1996).

An inhibitory effect in the kinetics of Cs uptake, at low external K<sup>+</sup> concentrations was described in the briophyte *Riccia fluitans* L. after preculture at different K concentrations. This observation and our present results agree with an inhibitory effect of K in Cs uptake at high affinity K channels (Casadesus et al. 2001).

Our results suggest that <sup>137</sup>Cs plant uptake is better described when ratio of <sup>137</sup>Cs/ <sup>40</sup>K in soil is considered, because an increment of K concentration in soil inhibits

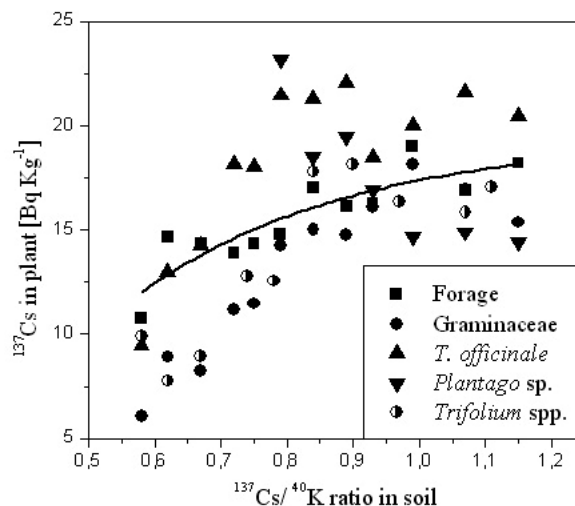


Figure 3. Mobile mean of <sup>137</sup>Cs activity concentration in plant for different plant and forage samples, against ratio of <sup>137</sup>Cs / <sup>40</sup>K in soil (layer 0 to 10 cm). The solid line shows the tendency for forage samples.

$^{137}\text{Cs}$  uptake by the plant. This observation suggests an inhibitory effect of potassium on radiocaesium uptake by plant roots.

Considering different plant species and uptake nutrients from soil, the analysis performed provides evidence that, under field conditions,  $\text{K}^+$  is more efficiently absorbed than  $\text{Cs}^+$ .

We analyzed the influence of the ratio of  $^{137}\text{Cs}/^{40}\text{K}$  in soil on  $^{137}\text{Cs}$  plant uptake. Under field conditions, the ratio  $^{137}\text{Cs}/^{40}\text{K}$  varied in a range of 0.5 to 1.3. For most of the species, at higher  $^{40}\text{K}$  soil concentration a lower  $^{137}\text{Cs}$  uptake was observed, a fact that reflects the resulting effect of the complexity of factors controlling ion absorption from soil.

The present results suggest that K fertilization could prevent radiocaesium uptake under field conditions. Bioremediation is emerging as an alternative approach to reduce the contamination level of  $^{137}\text{Cs}$ , such as application of minerals or chemical fertilizers in agricultural production systems with low levels of contamination. However, further investigation is still needed to optimize these countermeasures.

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