Comparison of spatial patterns of ^{137}Cs and ^{40}K in natural grassland soil and soil-to-plant relationship

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Abstract. ^{137}Cs and ^{40}K activity in natural grassland was studied at two sampling locations in the North-East of Italy, during 1997 and 1999. The present spatial patterns of ^{137}Cs and ^{40}K , their mobility and availability for uptake and transfer to grass. To know the spatial and temporal variation of these factors is essential for prediction the radioelement transfer; GIS tools (ILWIS) were applied. The spatial pattern of distribution evidence element redistribution in soil as consequence of the following parts: 1.- ^{40}K migration allows ^{137}Cs adsorption by clay soil components, although clays have low retention capacity. 2.- Lateral migration is predominant over vertical migration for both radioelements as consequence of: clay content of low retention capacity, higher porosity and strong acidic pH in site 1; low O.M. content, clay content of low retention capacity and moderate acidic pH in site 2. Multitemporal analysis evidences high increment of ^{137}Cs soil inventory which varies between one and six times. The spatial and temporal variation in ^{137}Cs soil inventory might be attributed to radioelement redistribution in soil, as shown by the spatial and temporal pattern. Radiocaesium redistribution determines changes in spatial patterns of soil contamination in time. Multitemporal analysis evidences loss of ^{40}K soil inventory.

1. INTRODUCTION

^{137}Cs is by far the most widely used fallout radionuclide in soil erosion and sedimentation research. The knowledge of its behavior and distribution in soils is vital for understanding its movement within the environment. The redistribution of ^{137}Cs between and within landscape elements provides information on spatial patterns of distribution. It is well known that radiocaesium tends to be readily adsorbed by clay minerals in soil, which leads to a decline in its uptake by plants in time. However, for soils with very low clay content, such as sandy podzolic and peat bog-soils, radiocaesium can still be available for uptake and so be transferred into food chains.

The spatial distribution of ^{137}Cs radionuclide in soils on a field scale level in undisturbed grassland environments is not well documented. The data generated by ^{137}Cs measurements are ideally suited to coupling with GIS (Geographical Information System), which can be used to predict spatial and temporal variation of radionuclides. The spatial and temporal radionuclide behavior is important in order to predict contamination levels of relevant agricultural products over different time scales.

The present study was based on sampling of natural soils and plants within an alpine pasture contaminated by the fallout of Chernobyl accident, an area which received a radiocaesium mean deposition of approximately 40 kBq m^{-2} [1-2]. Assuming that the fallout input of radionuclides was spatially uniform and was strongly and rapidly adsorbed by surface soil, the spatial variation in the inventory values can be attributed to soil redistribution. Post-depositional redistribution of radiocaesium within the soil occurs in response to a range of physical, physico-chemical and biological processes operating in the soil system, which in turn results in temporal changes in the radionuclide distribution in the soil profile.

The purpose of this research is to evaluate the present spatial distribution of ^{137}Cs and ^{40}K, their vertical and lateral migration, and to determine their changes by means of a multitemporal study in undisturbed soil. The spatial and temporal distribution and radioelement migration provide basic tools to progress in the categorization of soils with regard to trace Cs mobility and phytoavailability.

2. MATERIALS AND METHODS

The study was based on sampling of natural soils and plants from two sites with alpine pasture, during 1997 and 1999. The area of study is situated in the Tarvisio Woodlands, North-East of Italy. The geologic substratum in this area is moraine detritus, with numerous marl elements, covered with fine material probably from aeolian origin. The area is characterized by deep brown earth (podzolics).

Field sampling: soil and vegetation were sampled simultaneously, within an area of one hectare, 24 and 17 plots in site 1 and 24 plots in site 2 taken in three equidistant transects at the same daytime.

Soil samples: July 1997, site 1, monoliths of 30 x 30 cm were collected every 10 m over three transects separated by 50 m. The samples were divided into layers of 0 – 2.5 cm and 2.5 – 7.5 cm and processed. One sample per transect was taken to a depth of 40 cm (profile sample) and subsequently divided in layers of 0 – 2.5 cm ; 2.5 – 7.5 cm; 7.5 –10 cm; 10-15 cm; 15 –20 cm, 20- 40 cm. For year 1999, (28/June and 13/July), at both sites, monoliths of 15 x 15 cm were collected to a depth 10 cm. The distance between transects was 50 m in site 1, 40 m in site 2. One sample per transect was taken to a depth of 12.5 cm and subsequently divided into layers of 0-2.5 cm; 2.5-7.5 cm; 7.5-12.5 cm. Soil samples were air-dried and then weighed, ground to pass through a 2-mm sieve, and well homogenized.

Grass samples: were collected by cutting the total herbage growing at each 1 m x 1 m plot 2 cm above ground, avoiding contamination with soil. Grass material was weighed, dried at 105°C until constant weight and milled.

Measurements of activity were carried out with a high purity Germanium detector (HPGe). The activity of ^{137}Cs was decay-corrected to the date of the Chernobyl accident, May 1986.

Data Process by GIS tools (ILWIS ITC 98, [3]): A Digital Elevation Model (DEM) of field was created from topographic map by interpolation method. A DEM of ^{137}Cs and ^{40}K soil inventory was obtained by application of a linear interpolation function to the raster map of isoactivity curves of each radioelements. The spatial patterns of ^{137}Cs and ^{40}K soil inventory and Multitemporal Analysis were obtained by mean cross function.

3. RESULTS and DISCUSSION

3.1 Spatial and temporal soil distribution patterns of ^{137}Cs and ^{40}K

The behavior of ^{137}Cs and ^{40}K on seminatural grasslands was evaluated using GIS tools [3] in order to: 1) study the influence of slope and soil physical-chemical characteristic on its migration, 2) determine the temporal pattern of distribution by means of a multitemporal analysis of site 1 (years 1997-1999).

3.1.1 Slope influence and soil physical-chemical characteristics

The activity in the superficial soil layer is crucial for the soil-to-plant transfer process regarding herbaceous vegetation. For this reason, in the present study we considered the upper soil layer (7.5 cm or 10 cm depth). Soil characteristic, pH, organic C, total N, organic matter content (O.M.), cation exchange capacity (C.E.C.) and K, Mg, Ca exchange are summarized in Table 1 for both sites.

Site 1, sampling 1997: For ^{137}Cs soil inventory (Bq m^{-2}), although the Geometric Coefficient of Variation (GCV) was 40 % for the first level and 28 % for the second level, the pattern of distribution on the whole area was more homogeneous for the first level (0-2.5 cm) than for the second level (2.5-7.5 cm) less homogeneous pattern was observed (Fig. 1A).

^{40}K soil inventory (Bq kg^{-1}) at the first level on the whole area showed a non homogeneous distribution pattern as a consequence of ^{40}K eluviation, in vertical and lateral directions, according to the slope direction. At the second level an homogeneous distribution pattern was observed (Fig. 1B). ^{40}K soil inventory showed low variability among samples, GCV 9 % and 5 %, first and second level respectively.

Profiles until a depth of 40 cm, showed a predominant lateral migration at all levels.

Table 1. Soil characteristic.

Site	Depth (cm)	PH (H2O 1:2,5)	C org. (%)	O.M. (%)	N tot. (%)	K (mg/kg)	Mg (mg/kg)	Ca (mg/kg)	C.E.C. [meg 100g^{-1}]
1	0 – 2.5	5.0	9.8	16.9	1.00	381	465	5020	48.3
1	2.5 – 7.5	4.8	4.3	7.41	0.55	35	132	2250	29.8
1	7.5 – 12.5	5.7	2.7	4.65	0.30	6	78	2320	25.3
2	0 – 2.5	6.0	7.3	12.6	0.80	130	710	5174	39.1
2	2.5 – 7.5	5.6	4.2	7.24	0.52	44	443	3680	32.2
2	7.5 – 12.5	5.5	3.3	5.69	0.40	24	299	3190	29.6

Sites 1 and 2, 1999: significant spatial variability for both ^{137}Cs and ^{40}K inventory values provides non homogeneous patterns of distribution for both study areas.

The spatial pattern of ^{137}Cs soil inventory (Bq m^{-2}) in site 1 (Fig. 1C) shows high activity in the South East and East sectors, suggesting a low lateral mobility of the element.

The slope shows an inclination of about 18 %, between 900 m to 915 m altitude. The spatial pattern of ^{137}Cs soil inventory showed a higher variation at the upper part with respect to the lower part, evidencing changes of the spatial pattern due to migration of radioelements compared with 1997 sampling. The GCV values are shown in Table 2.

Table 2. Geometric Coefficient of Variation (GCV) for ^{137}Cs and ^{40}K soil inventory for different altitude.

	Site 1-1997		Site 1-1999		Site 2-1999	
	^{137}Cs (Bq m^{-2}) GCV (%)	^{40}K (Bq kg^{-1}) GCV (%)	^{137}Cs (Bq m^{-2}) GCV (%)	^{40}K (Bq kg^{-1}) GCV (%)	^{137}Cs (Bq m^{-2}) GCV (%)	^{40}K (Bq kg^{-1}) GCV (%)
Lower part	18	38	17	11	19	7
Middle part	24	21	15	9	12	2
Upper part	40	39	61	7	15	13

The ^{137}Cs soil inventory (Bq m^{-2}) on site 2 (Fig. E) shows a high lateral mobility. Within this area, the low O.M. content and clay content of low retention capacity were the determinant factors for the high lateral migration in spite of the smooth slope (7%) and moderate acidic pH.

In site 1, ^{40}K soil inventory (Bq kg^{-1}) shows mainly lateral migration, in the slope direction. At different levels of the soil profile an equilibrium was observed for lateral and vertical migration. An irregular migration behavior was observed for radiopotassium in soil profile, with prevalence of lateral mobility (Fig. D), probably because of the particular physical-chemical characteristics of the soil, particularly because of porosity variability.

In site 2, an important lateral mobility in the direction of the slope was observed for ^{40}K soil inventory (Bq kg^{-1}). However, on the soil profile, a vertical homogeneous migration was observed (Fig. F), an effect probably due to soil porosity. The topography shows an inclination of about 7%. For this area, the GCV values of ^{137}Cs and ^{40}K soil inventory exhibit low variation at different altitude (see Table 2).

The behavior of ^{137}Cs could be affected mainly by ^{40}K soil activity, as others authors have shown [4-5]. ^{40}K migration allows ^{137}Cs adsorption by clay soil components, although clays have low retention capacity. Our results evidence opposing spatial pattern of distribution for ^{137}Cs and ^{40}K soil inventories, as described earlier [4-5].

As reported in studies carried out by other authors [5-6], our experimental data for both sites clearly show that the mean activity of ^{40}K is lower for the organic part and greater for the mineral part of soil components. On the other hand, an inverse relationship was observed for ^{137}Cs, whose the activity decreased with greater soil depth. We found that organic material exhibited the highest ^{137}Cs activity, while the activity decreased rapidly in the mineral part of the soil. Our results confirm the tendency of an exponential decrease with depth of ^{137}Cs in soil profiles.

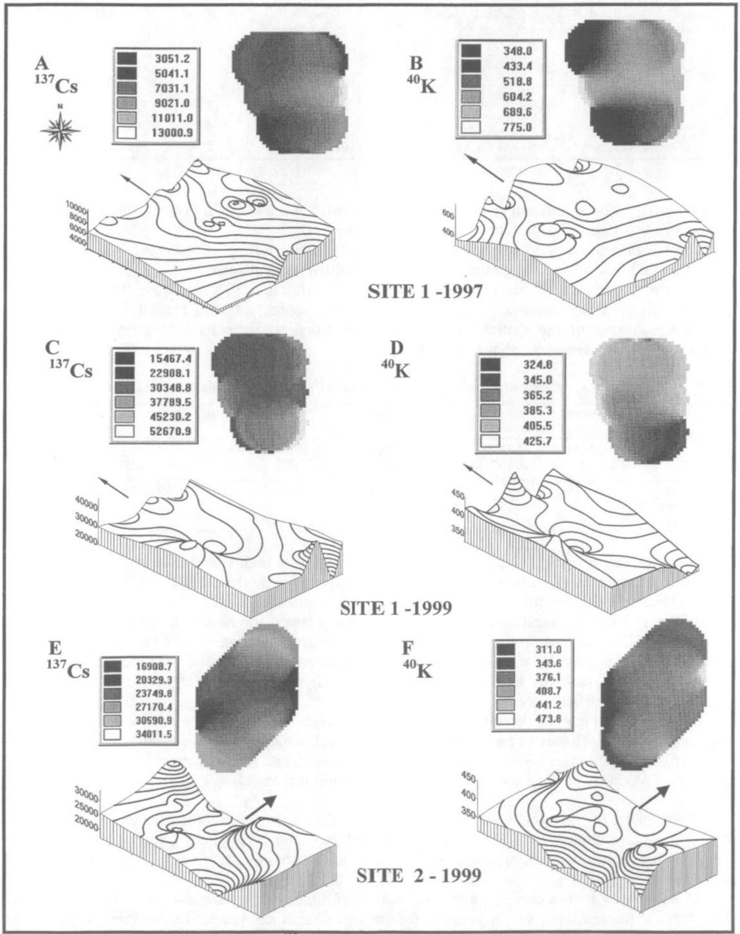

Figure 1. Interpolated map of isoactivities for ^{137}Cs and ^{40}K soil inventory. Slope direction: ↗

3.1.2 *Multitemporal analysis of soil inventory*

The data derived from the measures obtained for both ^{137}Cs and ^{40}K inventories of site 1, years 1997-1999, were analyzed with ILWIS by cross function to generate the maps shown in Fig. 2 (A-B).

For ^{137}Cs soil inventory (Bq m^{-2}), the multitemporal analysis shows comparatively higher values for the year 1999 than for 1997. The percentage of variation was calculated according to: $[^{137}Cs_{(97)} - {}^{137}Cs_{(99)}] / {}^{137}Cs_{(97)} * 100$. The increment varies between one and six times compared to the previous values (1997) (see Fig.2A). The spatial variation of ^{137}Cs soil inventory might be attributed to radioelement redistribution in soil, as shown by the spatial and temporal patterns. ^{40}K migration is another coadjuvant variable to ^{137}Cs retention, since ^{40}K migration leaves empty spaces on the soil components which are occupied by ^{137}Cs.

In the multitemporal analysis performed for ^{40}K soil inventory (Bq kg^{-1}), the spatial and temporal pattern obtained shows lower values for 1999 than for 1997 for most of the sample points. The percentage of variation was calculated according to: $[^{40}K_{(97)} - {}^{40}K_{(99)}] / {}^{40}K_{(97)} * 100$. Figure 2B shows the percentage of loss obtained for ^{40}K, only a minimum sector of the studied area with higher values in 1999 (index –4.2).

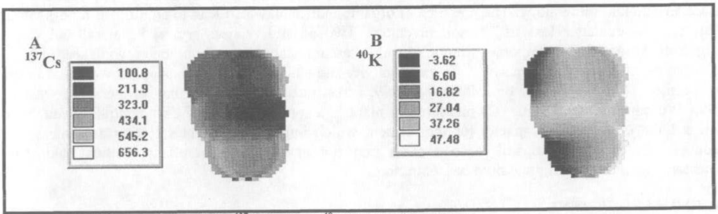

Figure 2. Multitemporal analysis for ^{137}Cs (left) and ^{40}K (right) soil inventory.

3.2 Soil-to-plant relationship of ^{137}Cs and ^{40}K

Soil-to-plant transfer factors were determined by the usual definition: *TF = activity per kg dry grass mass (Bq kg^{-1}) / activity in dry soil (Bq m^{-2}).*

The multitemporal analysis (site 1, 1997–1999) of ^{137}Cs grass activity and soil-to-grass relationship showed higher uptake during 1999 on the East sector. In spite of the higher Cs availability on soil during this year, grass transference was lower than in 1997 at the same season. Figure 3 shows the differences obtained for year 1999 as compared to year 1997.

The multitemporal analysis for ^{40}K grass activity and soil-to-grass relationship showed higher activity for 1997 for the same season in 1999 (Figure 3), in agreement with bioavailability of the element for the same period. Figure 3 shows the differences obtained between year 1999 and 1997.

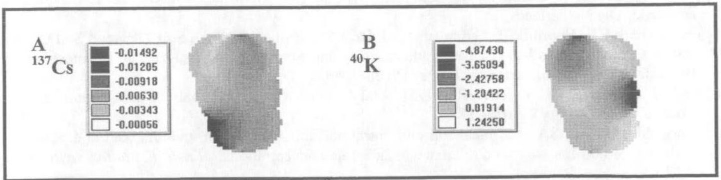

Figure 3. Multitemporal analysis for ^{137}Cs (left) and ^{40}K (right) soil-to-plant relationship.

4. CONCLUSIONS

The spatial pattern of distribution analyzed evidenced the element redistribution in soil as a consequence of following facts: 1.- ^{40}K migration allows ^{137}Cs adsorption by clay soil components, although clays have low retention capacity. 2.- Lateral migration is predominant over vertical migration for both radioelements as a consequence of: clay content of low retention capacity, higher porosity and strong acidic pH in site 1; low O.M. content, clay content of low retention capacity and moderate acidic pH in site 2.

Besides the physical-chemical soil characteristics, ^{40}K could also influence ^{137}Cs behavior. The migration of K^+ ions induces a sink effect which can provoke transformation of clay minerals in soil and therefore allow ^{137}Cs adsorption by clay soil components. In summary, the K^+ migration in soil due either to the eluviation effect or K^+ depletion because plant uptake might strongly influence the Cs redistribution in soil. ^{40}K soil activity shows mainly a lateral migration in slope direction.

Multitemporal analysis evidenced high increment of ^{137}Cs soil inventory, which varied between one and six times. The spatial and temporal variation in ^{137}Cs soil inventory might be attributed to radioelement redistribution in soil, as shown by the spatial and temporal patterns. Although Caesium soil adsorption, and hence its mobility and availability, are determined by clay content, physical-chemical characteristic and mineralogy. The presence of organic matter may also lead to significant modifications. This analysis evidence loss of ^{40}K soil inventory. Two possible causes are: a) K lateral and vertical migration, b) spatial and temporal variation of soil Potassium status throughout the growing season.

The multitemporal analysis of ^{137}Cs grass activity and soil-to-grass relationship showed, that in spite of the high ^{137}Cs availability on soil during 1999, grass transfer was lower than in the same season of 1997. We could assume that ^{137}Cs measured in plant is a combination of ^{137}Cs potentially available on soil and the plant uptake capacity for the element which implies that plant selection as a measure to reduce ^{137}Cs transfer from soil to food chain may not have significant effects without taking into consideration the external potassium concentration.

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