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REGULAR ARTICLE

Disparity in ^{90}Sr and ^{137}Cs uptake in Alpine plants: phylogenetic effect and Ca and K availability

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Abstract
Background and aims Uptake of ^{90}Sr and ^{137}Cs in plants varies widely between soil types and between plant species. It is now recognized that the radionuclide uptake in plants is more influenced by site-specific and plant-specific parameters rather than the bulk radionuclide concentration in soil. We hypothesized that the stress which Alpine plants experience because of the short growing season may enhance the phylogenetic effect on the ^{137}Cs and ^{90}Sr transfer factors as well as the dependency of the uptake by plant to the concentrations of exchangeable Ca and K of soils.
Methods We carried out a field study on the ^{90}Sr and ^{137}Cs uptake by 11 species of Alpine plants growing on 6 undisturbed and geochemically different soils in the Alpine valley of Piora, Switzerland.
Results Results show that a strong correlation exists between the log TF and the log of exchangeable Ca or K of the soils.
Conclusions Cs uptake by *Phleum rhaeticum* (Poales) and *Alchemilla xanthochlora* (Rosales) is more sensitive to the amount of exchangeable K in the soil than the corresponding uptake by other orders. Moreover, the ^{90}Sr results indicate a phylogenetic effect between Non-Eudicot and Eudicots: the order Poales (*Phleum rhaeticum*) concentrating much less ^{90}Sr than Eudicots do.

Keywords ^{90}Sr · ^{137}Cs · Plant · Phylogenetic effect · Uptake · Soil

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Introduction

The radionuclides ^{137}Cs and ^{90}Sr have been widely dispersed in the environment as a consequence of the atmospheric nuclear bomb tests (NBT) of the 1950's and 1960's. Moreover, the accident of the Chernobyl nuclear power plant increased the ^{137}Cs soil inventory in most European countries. Because ^{90}Sr is easily transferred from soil to plant and along the food chain, it can still be determined above the detection limit in milk teeth of children and in human vertebrae today (Froidevaux et al. 2006; Froidevaux et al. 2010). A commonly used parameter to express the extent of the transfer of radionuclides from soil to plants is the transfer factor (TF), which is the ratio of the mass activity concentration of the plant to that of the soil. One prerequisite for the use of the soil-to-plant transfer factor concept is the presence of a statistically significant relationship between the content of a given radionuclide in the soil and plant (Bunzl et al. 2000). However, TFs found in the literature are highly variable, even for the same plant species, which introduces considerable uncertainty in modeling (Frissel 1992; Staunton et al. 2003; Vandenhove and Van Hees 2007). It is now recognized that the radionuclide uptake in plants is more influenced by site-specific and plant-specific parameters rather than by the bulk radionuclide concentration in soil (Bunzl et al. 2000; Ciuffo et al. 2002). These specific parameters are: 1) the radionuclide bioavailability in the soil solution (Sanchez et al. 2002), 2) the capacity of plant species to absorb bioavailable radionuclides; 3) the availability of competing ions in the soil solution (Ehlken and Kirchner 2002), e. g. K as a chemical analogue to ^{137}Cs and Ca as an analogue to ^{90}Sr (Absalom et al. 2001; Camps et al. 2004; Delvaux et al. 2000; Roca-Jove and Vallejo-Calzada 2000; Sanchez et al. 2002; Smolders et al. 1997). For instance, Smolders et al. (1997) in a laboratory (pot trial) study described a negative correlation between the log- CR (plant to soil solution concentration ratio) of ^{137}Cs and the log of the K concentration in the soil solution in 30 different soils covering a wide range of textures. These authors concluded that 94% of the TF variability can be explained by the K concentration in the soil solution and by the ^{137}Cs distribution coefficient in soil, K_D. Furthermore, several soil parameters have been reported to play an important role in ^{90}Sr and ^{137}Cs transfer to plants: cation exchange capacity (CEC) of soil and associated pH, soil organic matter content (SOM), radiocesium interception parameter (RIP).In addition, heterogeneous distribution of radionuclide in soil affects the plant uptake (Bundt et al. 2000; Centofanti and Frossard 2006).

Broadley et al. (1999) proposed a method to assess taxonomic variation in plant uptake of cesium, with Cariophyllidae having the highest Cs concentration in shoot among superorders. Furthermore, Willey et al. (2005) used a database of 273 plant taxa to show that there exists a phylogenetic effect in the ^{137}Cs concentration in plants, with Caryophyllales and Poales presenting the highest and the lowest uptake, respectively. In addition, Willey and Fawcett (2006) showed that plant species take up Sr differently and that the uptake is linked through plant phylogeny. Combining both the phylogenetic approach and the K concentration in the soil solution, Waegeneers et al. (2001) found that the genotypic effects in the phytoavailability of ^{137}Cs are increased at a low K content in the soil.

There exist few studies conducted in Alpine ecosystems, regardless of the fact that radionuclide contamination of the soil is persistent in this environment and that the plant uptake is higher than in lowlands (Bunzl et al. 2000; Ciuffo et al. 2003; Ciuffo et al. 2002; Pourcelot et al. 2003; Pourcelot et al. 2007). The scarcity of field studies may lead to a bias in the data found in the literature because equilibrium between the soil, as well as the rhizosphere, and the deposited radionuclide is time-dependant and is often not reached in laboratory experiments (Waegeneers et al. 2009). Additionally, ^{137}Cs and ^{90}Sr concentration in soil solution are generally not measurable due to their very low concentrations. While TF is a questionable parameter in soil to plant transfer, it is a readily available quantity that is often tabulated in studies dealing with soil to plant transfer of radionuclide. On the other hand, the effect of exchangeable Ca and K on TFs is also questionable, particularly for ^{137}Cs for which RIP seems to be a better parameter. Nevertheless, CEC is probably the simplest parameter that accompanies the soil classification in the general literature while RIP necessitates a complex determination that has been carried out only on a few soils samples (Sanchez et al. 2002). Thus there is an interest to link the exchangeable K and Ca in the soil to the soil to plant ^{137}Cs and ^{90}Sr TF. This relationship should involve various plant species with different uptake potential. This may help to classify plant species and soils according to their capability to transfer radionuclide to the food chain.

The aim of this study was to investigate the relationship between the ^{90}Sr and the ^{137}Cs TFs for different species of plants and the exchangeable Ca and K in the soils. In addition to the effect of competing ions, we hoped to show any possible phylogenetic effects on the TFs. We assumed that Alpine plants may show enhanced phylogenetic effects on bioaccumulation due to the stress created by the short growth period at an altitude of 2000 m a.s.l. Because a short growth season means a fast biomass increase rate, it results in a higher requirement in minerals. This might influence the uptake of ^{137}Cs and ^{90}Sr. To reach these objectives we chose an Alpine area with different soil types. We collected plants from different taxa that were present on the same soil and similar plant species present on different types of soils.

Materials and methods

Sampling sites

The study was conducted in an Alpine valley in Switzerland (Val Piora, Ticino; ca 2000 m a.s.l.). This 23 km^2 valley is used in the summer for pasturing and is covered with snow from October to May. Six sites were selected: three of them, referred hereafter as sites 1, 4 and 6, are alpine pastures with increasing cattle pressure; site 2 is a fen, site 3 is a non-pastured wet meadow and site 5 is a resting place for cattle. The vegetation is described according to the Braun-Blanquet phytosociological classical approach and for each site, soils horizons have been described according to AFES (Association Française pour l'Etude du Sol; Baize and Girard 2009) and soils have been classified according to WRB (World Reference Base for soil resources, FAO 2006). A full description of each soil profile, along with ^{137}Cs and ^{90}Sr soil profiles and a full description of the overlaying vegetation are available in Supporting Information.

Sampling

Soils were sampled in August 2007, described and analyzed for CEC, pH and exchangeable K and Ca as displayed in the supplementary material (Table SM-1). Water content was estimated to be at least 50% of the soil capacity during the growing season. Soil 2 was permanently saturated. Plant samples were collected between July and August 2007 and again in July 2008, on the same surfaces. 500 to 2000 g of wet aerial material were carefully picked for each species. We collected 4 species (*Achillea millefolium*, *Alchemilla xanthochlora*, *Phleum rhaeticum* and *Ranunculus acris*), all present together on sites 1, 3 and 4. These species and *R. alpinus* collected on sites 4, 5 and 6 form the dataset for site-specific modeling. In addition, we collected other plant species on sites 1 to 6 as described in Table 1. The depth of the rooting layer of each species was estimated through field observation, in the soil profile. Plant samples were carefully washed several times with tap water to remove adhering soil particles; afterwards they were air-dried and crushed. Plant samples were ashed at 600°C for 48 h. Ashes were dissolved in 8 M HNO_3 in a pressurized microwave apparatus (MLS ultraCLAVE; Milestone Inc., Monroe, CT) and filtered before measurement. Ca, as well as stable Cs and Sr, were measured on an aliquot using the same techniques as used for the soil samples.

Radionuclide analysis

^{137}Cs activity was measured on air-dried samples of soil and plants in a volume of 250 or 500 ml using γ-spectrometry as described in Saidou et al. (2008). The total concentration of K in the soil and plant sample was calculated from the activity of ^{40}K.

^{90}Sr activity was determined indirectly from the measurement of its daughter product, ^{90}Y, using a slightly modified version of the method described in Froidevaux et al. (2002). The modification consists, for a soil sample, in introducing an additional purification step of ^{90}Y through an extraction of ^{210}Bi (as a potential β-emitter interference) on an anionic exchanger (AG1x8) in 9 M HCl media. The detection limit for ^{90}Sr analysis was 0.5 Bq per kg of dry mass for soil and 0.1 Bq per kg of dry mass for plant.

Calculation

The concentrations of elements in the soil horizons were reported to the volume of a soil layer of one centimeter thick for one square meter. This particular calculation allows for the comparison of different soils with different densities. Transfer factors were calculated for each horizon following Eq. (1):

$$TF = \frac{[A_{plant}](\mathrm{Bq\ kg^{-1}})}{(A_{soil})(\mathrm{Bq\ m^{-2}\ cm^{-1}})} \qquad (1)$$

Results

Soils

The main characteristics of each soil site are presented in Table SM-1 (Supplementary Material). The majority

Table 1 ^{137}Cs and ^{90}Sr activities (Bq kg^{-1} of dry mass) and K and Ca content (mg g^{-1} of dry mass) of plants on different sites

				Bq kg^{-1} dry mass				mg g^{-1} dry mass			
species	Class a)	site	abbreviation	^{137}Cs	u95	^{90}Sr	u95	K	u95	Ca	u95
Achillea millefolium	E	1	1 am	182.5	6.9	92.0	1.6	20.2	2.1	22.5	0.4
Alchemila xanthochlora	E		1ax	71.9	5.3	74.5	1.2	12.5	3.5	19.3	0.3
m2 b)			1 m2	62.4	1.3	91.9	2.1	9.3	0.4	18.9	0.4
Phleum rhaeticum	NE		1pr	119.3	6.9	16.7	0.3	13.8	2.4	4.6	0.1
Ranunculus acris	E		1rac	44.1	3.3	83.1	1.2	11.7	1.5	20.2	0.3
Rhododendron ferrugineum, wood	E		1rfb	5.7	0.2	28.2	0.4	n.m	n.m	5.0	0.1
Rhododendron ferrugineum, leaves	E		1rff	11.5	0.7	51.5	1.0	5.5	0.3	13.1	0.3
Allium schoenoprasum	NE	2	2as	11.1	0.5	4.2	0.3	13.3	0.4	24.4	1.7
m2 b)			2 m2	61.3	1.2	2.6	0.2	12.9	0.4	22.1	2.0
Poaceae c)	NE		2poa	16.5	0.5	1.0	0.1	9.3	0.3	2.8	0.3
Trifolium badium	E		2tb	36.3	0.9	4.5	0.3	14.7	0.5	20.7	1.3
Achillea millefolium	E	3	3 am	25.5	2.1	62.6	1.5	26.1	1.7	15.2	0.4
Alchemia xanthochlora	E		3ax	209	6.5	52.4	0.7	12.3	1.5	17.2	0.2
Gentiana purpurea	E		3gp	129.1	3.1	24.7	0.6	7.7	0.7	7.9	0.2
Peucedanum ostruthium	E		3po	26.7	0.9	112.2	1.3	15.2	0.5	16.8	0.2
Phleum rhaeticum	NE		3pr	107.4	3.1	12.8	0.3	7.9	0.8	1.3	0.0
Phleum rhaeticum	NE		3bpr	149.1	6.2	9.7	0.3	16.3	2.0	2.3	0.1
Ranunculus acris	E		3rac	80.5	3.8	79.7	0.8	19.8	1.8	15.3	0.2
Veratrum album	NE		3va	47.2	1.2	184.4	2.4	17.2	0.5	18.8	0.2
Achillea millefolium	E	4	4 am	31.1	1.8	30.0	1.1	50.9	1.7	26.7	0.9
Alchemila xanthochlora	E		4ax	13.8	1.4	42.3	0.6	17.4	1.0	24.7	0.3
Geranium sylvaticum	E		4gs	9.3	1.6	35.6	1.0	15.2	1.4	25.3	0.7
Phleum rhaeticum	NE		4pr	9.4	1.2	6.0	0.2	14.0	0.9	1.7	0.1
Rrumex alpinus	E		4ra	19.3	0.7	17.4	1.1	31.9	0.7	7.4	0.5
Ranunculus acris	E		4rac	33.5	4.4	39.5	0.8	30.7	3.8	24.6	0.5
Alchemilla xanthochlora	E	5	5ax	5	0.8	47.5	0.7	20.6	0.9	8.7	0.1
Rumex alpinus	E		5ra	2.3	0.5	12.5	0.5	35.8	0.9	3.2	0.2
Alchemila xanthochlora	E	6	6ax	21.7	0.9	33.9	0.7	19.9	0.6	12.9	0.3
Rumex alpinus	E		6ra	19.7	1.3	11.3	0.5	38.2	1.4	4.0	0.2

a. E: Eudicot; NE: Non-Eudicot

b. One square meter is sampled, all species together

c. Mix of all Poaceae present on this site

of the soils are classified as Cambisols. Nevertheless, they differ in their pH, the amount of exchangeable cations and the number of saturated exchangeable sites. Soil 1 has the lowest amount of exchangeable Ca and Mg of all the studied Cambisols, and is thus classified as Cambisol (Dystric). Soil 2 is a Calcic Histosol with a nearly basic pH caused by lateral input from upslope runoff rich in Ca. Soil 3 is a sandy acidic soil with the lowest amount of exchangeable cations of all the sites. It is classified as a Gleysol (Dystric). Soil 4 and soil 5 have the highest amount of exchangeable K. They are classified as Fluvic Cambisol (Eutric) and Cambisol (Eutric), respectively. Despite a low amount of exchangeable K, site 6 is saturated with Ca in the whole profile and is thus also classified as a Cambisol (Eutric). The high amount of Ca in this site is due to the underlying dolomite. Figure SM-1 displays the CEC of the six soils as a function of depth.

Results of the surface deposition of ^{137}Cs and ^{90}Sr for each soil are given in Table SM-2. On average, ^{137}Cs inventory is about 5000 Bq m^{-2} except for soil 2 where depletion is observed due to runoff, and for soils 4 and 6, which are situated in a small depression, where Chernobyl-originating ^{137}Cs probably accumulated after snow melting (Pourcelot et al. 2003). On average, ^{90}Sr inventory is 1200 Bq m^{-2}, apart from soil 2 which is depleted in surface, due to enhanced mobility (Chawla et al. 2010), and soil 4, enriched because of its situation in a small depression. The depth distribution of the ^{137}Cs activity is typical of alpine soil with accumulation in the top soil layers. Depth distribution of ^{90}Sr and ^{137}Cs in site 2 shows enhanced mobility for both radionuclides due to the organic-rich nature and strong runoff features of this soil (Chawla et al. 2010). In addition, the depth distribution of ^{90}Sr shows that inventories for soil 2, soil 3 and soil 6 are most likely underestimated because of the high mobility of ^{90}Sr in these soils (see ^{90}Sr profiles in Supplementary Material).

Radioisotope activities in plants and TFs

Table 1 displays the ^{137}Cs and ^{90}Sr activity and K and Ca content measured in the different plants species. Plant activities as well as K content are highly variable among species and between the different sites. Two orders of magnitude were found for ^{137}Cs (2.3–209 Bq kg^{-1}) and ^{90}Sr (0.97–184 Bq kg^{-1}) between plant samples, with activities log-normal distributed. In the following, uptake is expressed as the TF calculated using the inventory of the first horizon for ^{137}Cs and of the second horizon for ^{90}Sr, if not otherwise stated. This choice resulted from the depth distribution of ^{137}Cs and ^{90}Sr in the soils and the observed rooting zone of species that we selected as representative of the rooting (absorption) zone. The observed rooting zone typically includes the two first horizons, while the activity of ^{90}Sr is often at its maximum in the second horizon. Figure 1a shows the ^{137}Cs uptake by 4 species of Alpine plants that co-existed in three different types of soils. The results demonstrate that the uptake by plant species is dependent on the

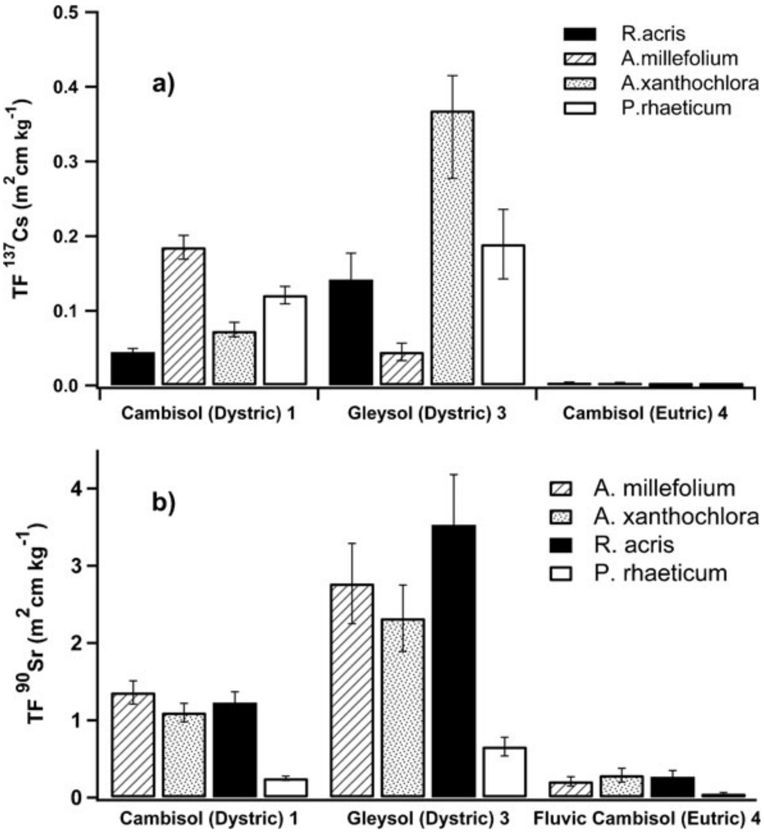

Fig. 1 TFs of 4 species collected on 3 different soil types: (**a**) ^{137}Cs; TFs are calculated with the radionuclide inventory of the first horizon. (**b**) ^{90}Sr; TFs are calculated with the radionuclide inventory of the second horizon. Uncertainty is given for *k=1*

underlying soil. All four species have a minimal uptake when growing on soil 4, despite a ^{137}Cs inventory, which is much higher in this soil than in the other soils. However, soil 4 has the highest exchangeable K content of all soils. Figure 1b displays the results of ^{90}Sr uptake in the same 4 plant species growing on each of the three soils. The results demonstrate that *A. millefolium*, *A. xanthochlora* and *R. acris* (Dicot) have similar uptake in a given soil and their maximum uptake is on the same soil (Gleysol Distric 3). On the other hand, *P. rhaeticum* (Monocot) has a distinctly lower uptake on all three soils.

Plotting the activity of ^{137}Cs in the plant against the content of its chemical analogue potassium in the plant, a pattern appears as displayed in Figure 2 (a): either the plants have a low K content and may have a wide range of ^{137}Cs activity; or they have a high K content, in which case they have not taken up a large quantity of ^{137}Cs. Non-Eudicots have a low K content in every case but ^{137}Cs covers a large range of activity. Results are very different for ^{90}Sr and its chemical analogue calcium: there exists a strong correlation ($r=0.83$; p-value$<2\cdot10^{-6}$, $n=20$) between the ^{90}Sr content and the Ca content of the plant grown on soil 1, 3 and 5 (Fig. 2b). These soils have a ratio of ^{90}Sr to exchangeable Ca in the rooting zone above 13 Bq gCa^{-1}. For soils with a ratio of ^{90}Sr to exchangeable Ca in the rooting zone below 6 Bq gCa^{-1} (soil 2, 4 and 6), no statistical relationship was observed between the ^{90}Sr and Ca content of plant ($r=0.25$, $n=9$). This is because soil 2 receives Ca-rich runoff and soils 4 and 6 have very high CEC and exchangeable Ca content. Consequently, Ca is highly available in these soils, which results in a comparatively very low ^{90}Sr uptake. In addition, Figure 2b shows that non-Eudicots (Poales and Liliales) contain very little ^{90}Sr and Ca.

These results show that the soil to plant TFs ^{137}Cs and ^{90}Sr are strongly influenced by soil properties (exchangeable K and Ca) as well as by a phylogenetic

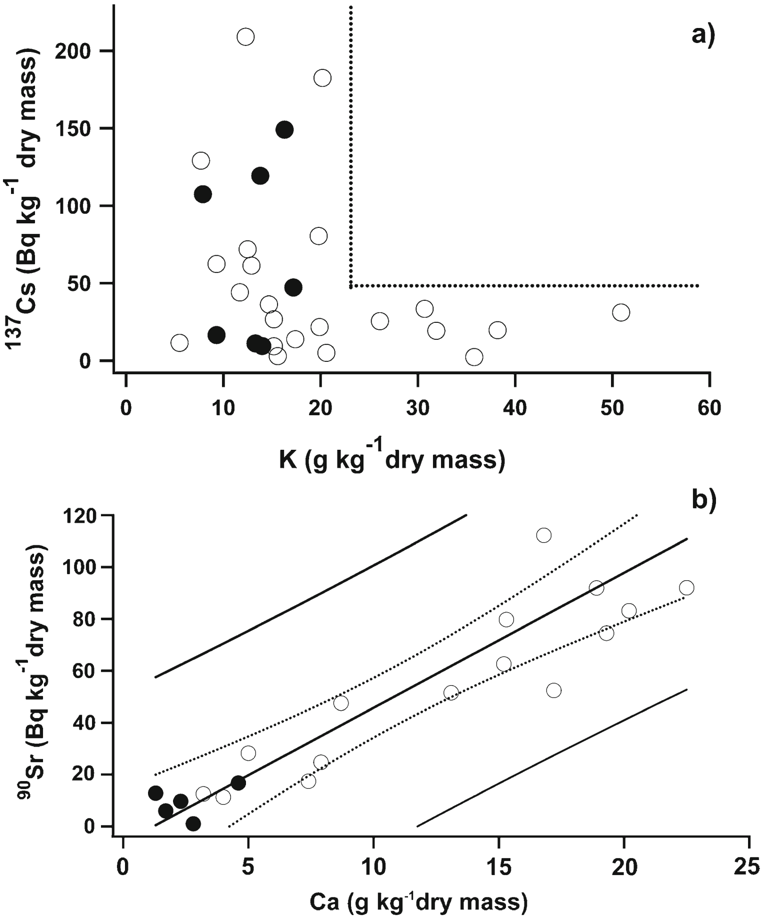

Fig. 2 ^{137}Cs activity in the plant as a function of the K content in the plant (**a**) and ^{90}Sr activity in the plant as a function of the Ca content in the plant. Dotted lines limit a region where no values are found (**b**). Black dots: Non-Eudicots. Open dots: Eudicot. Solid external lines represent 95% confidence interval of the regression data, dotted lines represent 95% confidence interval on the regression line

effect. For every site, we calculated a linear regression between the log-transformation of the average TFs (on all plant species) and the exchangeable K or Ca, respectively. A significant correlation exists between log TFs and log exchangeable K or Ca content. For ^{137}Cs and K, the Pearson coefficient was 0.91 (p-value$<$ 0.012, $n=26$ plant samples collected on 6 different soils, Fig. 3a). The corresponding calculation for ^{90}Sr and Ca yielded a Pearson coefficient of 0.82 (p-value$<$ 0.027, $n=26$; Fig. 3b).

In addition, for 5 plant species (*A. millefolium, A. xanthochlora, P. rhaeticum, R. acris, R. alpinus*), we calculated the linear regression of the log-transformation of the TFs versus K or Ca for the three first horizons separately, using the radionuclide inventory of each horizon and the exchangeable amount of K or Ca of the corresponding horizon. This calculation was meant to show, for each species found on at least three different soils, a specific effect of the exchangeable K and Ca. Results are presented in Table 2. The calculation for ^{137}Cs shows that for all species, apart from *R. alpinus*, the best fit is obtained using the data of the first horizon. This is in agreement with the observed rooting zone of *P. rhaeticum* and *R. acris* in the first horizon (0–5 cm) while *R. alpinus* has a deeper rooting zone. The calculation for ^{90}Sr shows that the best fit is observed using data from the first horizon for *A. millefolium*, *P. rhaeticum* and *R. acris* and data from the second horizon for *A. xanthochlora* and *R. alpinus* (for specific rooting zone and ^{137}Cs and ^{90}Sr distribution in soils, see supplementary material, Figs. SI4 to SI9). However, the statistical significance of all the fits is weak because each selected species is found on only 3 to 5 different soils at most. Figure 4 displays a graphical representation of the best fit of the linear regression of the log-transformation of the TFs versus the exchangeable K or Ca. For ^{137}Cs uptake, the presence of two different slopes (Fig. 4a) for five different plant species can be interpreted as a

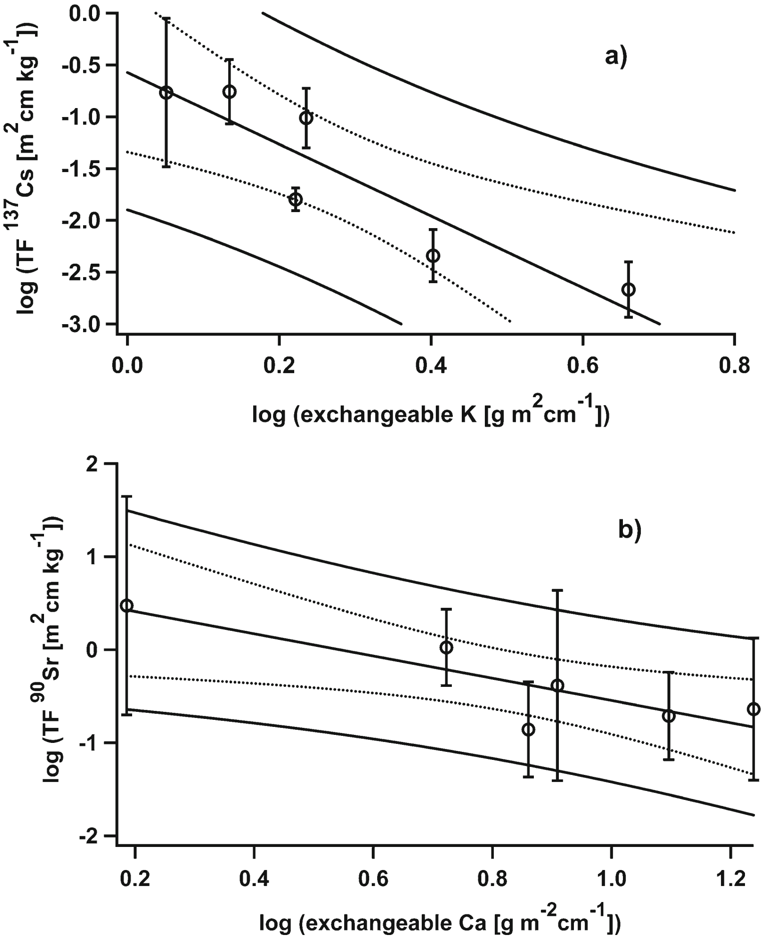

Fig. 3 Regression on log-transformed average TFs data of ^{137}Cs (**a**) or ^{90}Sr (**b**) as a function of log-transformed exchangeable K or Ca in 6 soils. Solid external lines represent 95% confidence interval of the regression data, dotted lines represent 95% confidence interval on the regression line

Table 2 Parameters of the log TF versus log exchangeable K or Ca linear regression using values of ^{137}Cs or ^{90}Sr deposition in horizon 1, horizon 2 and horizon 3

	^{137}Cs							
	horizon 1		horizon 2		horizon 3		parameter of regression	
species	R^2	p-value	R^2	p-value	R^2	p-value	Slope a	Intercept b
A. millefolium	0.47	0.34	0.42	0.36	0.99	0.05	-2.70^{i}	-0.59^{i}
A.xanthochlora	0.76	0.03	0.45	0.13	0.10	0.31	-4.03^{i}	-0.34^{i}
P.rhaeticum	0.98	0.07	0.93	0.12	0.80	0.21	-4.46^{i}	-0.01^{i}
R.acris	0.96	0.09	0.94	0.11	0.11	0.47	-2.87^{i}	-0.56^{i}
R.alpinus	0.57	0.31	-0.98	0.93	0.61	0.29	-1.84^{i}	-1.55^{i}
	^{90}Sr							
	horizon 1		horizon 2		horizon 3		parameter of regression	
species	R^2	p-value	R^2	p-value	R^2	p-value	Slope a	Intercept b
A. millefolium	0.85	0.18	0.75	0.23	0.21	0.43	-0.81^{i}	0.26^{i}
A.xanthochlora	0.47	0.12	0.91	0.01	0.75	0.04	-0.87^{ii}	0.57^{ii}
P.rhaeticum	0.91	0.14	0.81	0.20	0.14	0.46	-0.83^{i}	-0.43^{i}
R.acris	0.99	0.06	0.92	0.13	0.00	0.50	-0.85^{i}	0.35^{i}
R.alpinus	0.69	0.74	0.74	0.24	0.72	0.25	-0.54^{ii}	-0.30^{ii}

i: parameter of regression of horizon 1; ii: parameter of regression of horizon 2.

different response of the plants to the exchangeable K content depending on the species. *A. millefolium*, *R. acris* and *R. alpinus* accumulate more ^{137}Cs at a high exchangeable K content but are less sensitive to the decrease in the exchangeable K content (slope in the range of −2.87 to −1.84), while *A. xanthochlora* and *P. rhaeticum* accumulate somewhat less ^{137}Cs at a high K content but are more sensitive to its variation (slope in the range −4.03 to −4.46). Despite the low significance of the statistics, *t*-test reveals two different group of values (bilateral, heteroscedastic, p-value=0.019) that confirms that *A. xanthochlora* and *P. rhaeticum* behave differently than *A. millefolium*, *R. acris* and *R. alpinus*. Conversely, ^{90}Sr uptake by all the selected species gives the same response to any variation in the exchangeable Ca content of the soils (slope in the range −0.87 to −0.81).

From this observation it follows that plant activity can be calculated using the empirical relation described in Eq. (2) using the concentration of exchangeable K or Ca in the soil and the slope of the linear regression determined on the log-transformation of TFs (Eq. 2):

$$A_{plant} = A_{soil} \cdot C_{soil}^{a} \cdot 10^{b} \qquad (2)$$

where A_{plant} is the activity of ^{137}Cs or ^{90}Sr in the plant (Bq kg^{-1}), A_{soil} is the inventory of ^{137}Cs or ^{90}Sr in the soil (Bq m^{-2} cm^{-1}), C_{soil} is the concentration of exchangeable K or Ca in the corresponding rooting zone of the soil (g m^{-2} cm^{-1}) and *a* and *b* are parameters as given in Table 2. *a* is an empirical parameter that can be understood as the sensitivity of the plant species to the presence of competing K or Ca in the soil solution.

Discussion

The results of our study show that the relationship between the plant uptake of ^{137}Cs and ^{90}Sr and the concentration of exchangeable K or Ca in the soils, as previously reported by others (Delvaux et al. 2000; Sanchez et al. 2002; Smolders et al. 1997; Waegeneers et al. 2001), can also be worked out from our field data acquired at an undisturbed Alpine environment. This means that ^{137}Cs and ^{90}Sr uptake by plants is strongly dependent on the presence of available (exchangeable) chemical analogues in the soil. In particular, we found a significant linear correlation between the log-transformation of the TFs and the exchangeable K or Ca content of the soil. Apparently, simple soil properties such as exchangeable K and Ca can be used to predict a soil's ability to transfer ^{90}Sr or ^{137}Cs to plant

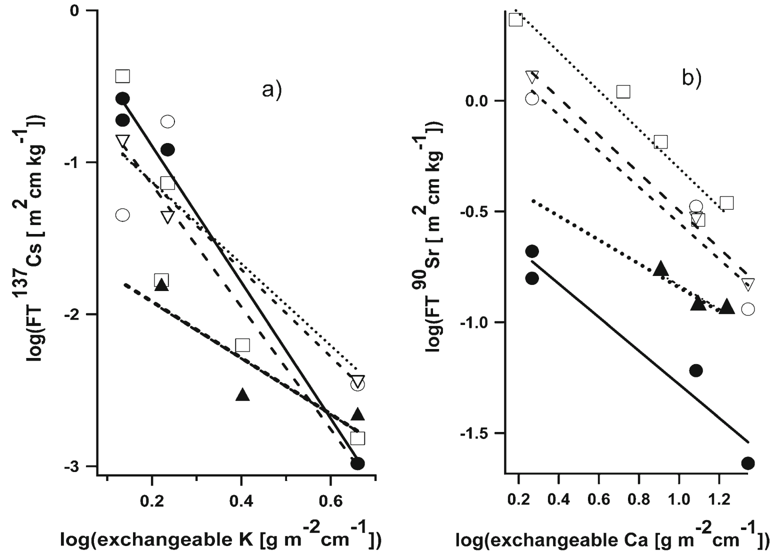

Fig. 4 Regression on log TFs data of ^{137}Cs (**a**) or ^{90}Sr (**b**) of 5 species as a function of log exchangeable K or Ca. Symbol: ○: *A. millefolium*; □: *A. xanthochlora*; ●: *P. rhaeticum*; ▲: *R. alpinus*; ▿: *R. acris*. Calculations are made using ^{137}Cs inventory of the first horizon for all plants species and ^{90}Sr inventory of the first horizon for *A. millefolium*, *P. rhaeticum* and *R. acris*, and of the second horizon for *A. xanthochlora* and *R. alpinus*

when the radionuclides are at chemical equilibrium due to a long time period of presence in soil. This has been found in our study of the silty-sandy mineral, partly organic-rich, soils of the Val Piora. It possibly applies also to similar sites, even if there are limitations to the use of the exchangeable cations rather than the competitive cations concentration of the soil solution in the interpretation of TFs (Gil-Garcia et al. 2009). As a result, the link between the ^{90}Sr and ^{137}Cs uptake by plant and the exchangeable K and Ca of soil, a very basic characteristic of soil that can be found almost for every soil in the literature, seems established, at least for this specific alpine environment.

Furthermore, we found species-dependent differences for the selected plants with respect to the response of the ^{137}Cs uptake to the amount of exchangeable K in the soil. More specifically, we found that *A. millefolium*, *R. acris* and *R. alpinus* are less sensitive to variations in the amount of exchangeable K in the soil than *A. xanthochlora* and *P. rhaeticum*. Since the "sensitive species" include an Dicot and a Monocot the observed phylogenetic effect seems to be a property of individual species rather than larger group of plants.

In contrast, we found that the ^{90}Sr uptake of all investigated Alpine species has a similar sensitivity to changes in the exchangeable Ca content of soils. There is no evidence for a phylogenetic effect with this respect. Consequently, the results of our study on ^{90}Sr uptake could be used to predict the ^{90}Sr uptake over a wide range of soil exchangeable Ca content for plant for which only one measurement of ^{90}Sr TF and one measurement of the corresponding exchangeable Ca content exist. While the above described Ca-sensitivity of TFs for ^{90}Sr does not depend on the species, the level of ^{90}Sr accumulation does. There are genuine differences in TFs among species for one given site that are maintained across the different sites. We observed a strong difference in the ability of plant species to accumulate ^{90}Sr; *V. album* (Liliaceae), *P. ostruthium* (Apiaceae) and *G. sylvaticum* (Geraniaceae) in addition to *A. xanthochlora* (Rosaceae), *A. millefolium* (Asteraceae) and *R. acris* (Ranunculaceae) are species that accumulate large amounts of ^{90}Sr. On the other hand, *P. rhaeticum* (Poaceae) has a very low uptake of ^{90}Sr in all types of soils studied here. This confirms results of others (Fuhrmann et al. 2002; Willey and Fawcett 2006) that demonstrate that plants in the Poales order do not accumulate ^{90}Sr and Ca to a large extent.

White and Broadley (2000) reviewed the mechanisms of caesium uptake by plants. The general observations are that K$^+$ permeable channels are also permeable to Cs$^+$, with VIC (voltage-independent channel) channels explaining most (30%–90%) of the Cs$^+$ influx under physiological conditions. Nevertheless, higher-affinity K$^+$ channels also play a significant role in Cs influx, possibly at low external $[K^+]_{ext}$ concentration. While our study was not designed as a plant physiology study, our results conform to these observations. For instance, we observed high K content together with low ^{137}Cs content for plants growing on soils with a CEC higher than 1000 cmol$^+$ m^2 cm (soils 4,5 and 6). Thus non-selective channels (e.g. VIC channels) might be responsible to the high K uptake together with low ^{137}Cs uptake (competition), while at low external $[K^+]_{ext}$ concentration (low CEC) much higher ^{137}Cs influx has been measured.

Similarly, Ca enters plant cells through Ca^{2+}-permeable channels (White and Broadley 2003). These authors state that there seems to be no competition between Ca^{2+}, Ba^{2+} and Sr^{2+} in transport from xylem to the shoot. Moreover their accumulation in the shoot is often linearly correlated to their concentration in the rizosphere solution. Here also, our results on ^{90}Sr accumulation in the plants are in accordance with a non-selective influx of Ca or Sr (Fig. 2b). To the opposite to K$^+$ uptake, higher Ca^{2+} uptake in plant is observed on soils with CEC lower than 500 cmol$^+$ m^2 cm (soil 1 and 3). This higher Ca^{2+} uptake is accompanied by higher ^{90}Sr uptake.

Conclusions

We studied radionuclide uptake in undisturbed alpine soils. In these soils ^{90}Sr has been deposited in the early sixties as a result of fallout of the NBT. ^{137}Cs has been deposited during two events: the fallout of NBT and the fallout of the Chernobyl accident. Both events contribute to about 50% of the current ^{137}Cs activity in surface soils of the sampling sites (Chawla et al. 2010). The long-time presence of the radionuclides in the soil allowed for the establishment of a chemical equilibrium between ^{90}Sr and ^{137}Cs and soil components (e.g. clay minerals). Thus our experiment differs strongly from pot experiments that form basis for many literature data on radionuclide uptake by plants. Here we demonstrate that exchangeable K and Ca in soil can be used as a proxy for the determination of the extent of ^{137}Cs and ^{90}Sr transfer from soil to plant, at least for (organic-rich) silty-sandy soils and for plant species subjected to a short growing period. In addition we found that the dependency of ^{90}Sr TF in function of exchangeable Ca was similar for 5 different Alpine plant species. This result may be used to predict the ^{90}Sr uptake by plant for which only one determination of TF and exchangeable Ca per type of soil are known.

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