Interception and Retention of Wet-deposited Radiocaesium and Radiostrontium on a Ley Mixture of Grass and Clover

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Abstract

The aims of this study were to assess the potential radioactive contamination of fodder in the case of accidental radionuclide fallout, and to analyse the relationship between interception and retention of radionuclides as a function of biomass and Leaf Area Index (LAI). The interception and the retention of wet deposited ^{134}Cs and ^{85}Sr in ley (a mixture of grass and clover) were measured after artificial wet deposition in a field train in Uppsala (eastern central Sweden). The field trial had a randomised block design with three replicates. ^{134}Cs and ^{85}Sr were deposited at six different growth stages during two growing seasons (20101 and 2011) using a rainfall simulator. The biomass was sampled in the centre of each parcel 2 to 3 h after deposition and at later growth stages (1 to 5) during the growing season. The above ground biomass and LAI were measured as well.

The interception of radionuclides by the ley was largest at the late growth stages; the spike and tassel/flowering (code 5:6) in the 1st year, and at flowering/initial flowering (code 6:5) in the 2nd year. There was a correlation between radionuclide interception and above ground biomass, as well with LAI, for both radionuclides in both years. The highest activity concentrations of both radionuclides were measured after deposition at the late growth stages and were found to be higher in the 2nd year. There weathering half-lives were shorter at the earlier growth stages than at the later growth stages for both radionuclides. For the magnitude of deposition chosen in our experiment, it can be concluded that the above ground biomass is a good predictor and the LAI a more uncertain predictor of the interception of radiocaesium and radiostrontium by ley grass and clover.

Keywords: fodder crops, growth stages, radioactive deposition, weathering half-life, aggregated transfer factor

1. Introduction

The release of radionuclides into the atmosphere can result in their deposition on crops and soils. Several studies have reported that wet deposited radionuclides, *e.g.* radiocaesium and radiostrontium, can be intercepted and taken up directly the vegetation, for example through leaves (Bengtsson *et al*. 2013; Middleton, 1958; Middleton, 1959; Scotti and Carini, 2000). One of the most important pathway for the transfer of radionuclides, ^{90}Sr and ^{137}Cs, to humans is through milk and meat consumption of animals that feed on contaminated fodder from ley and pasture (Fasenko *et al*., 2009; Gastberger *et al*., 2001; UNSCEAR, 2008). Milk obtained from contaminated agricultural fodder is a main source of the total collective dose to humans (Lepicard and Dubreuil, 2001; Madoz-Escande *et al*., 2004; Middleton, 1958). The levels of milk contamination by radionuclides depends on, among others, the foliar absorption by plant leaves and shoots, the root uptake of radionuclides from soil to crops, the consumption of contaminated soil by animals and the transfer of radionuclides from forage vegetation to milk (Gastberger *et al*., 2001).

The magnitude of radionuclide interception by crops depends on the concentration of radionuclides from the fallout, the intensity of the precipitation, time of year, plant morphology, *i.e.* the Leaf Area Index (LAI), the angle of leaves, above ground plant biomass, growth stage of the crop, the total amount of rain and wind (Eriksson *et al*., 1998; Hoffman *et al*., 1992; IAEA, 2010; Kinnersley *et al*., 1997; Pröhl, 2009). Other factors that can influence the level of radionuclide interception are the physical and chemical forms, *i.e.* the molecular mass and the valence (Bréchignac *et al*., 2000). Post-deposition retention of radionuclides is related to weathering processes such as wash-off of intercepted radionuclides by rain or irrigation, plant surface abrasion, re-suspension, age of plant tissues, litter fall, growth of new plant tissues, volatilisation, and evaporation (IAEA, 2009; Milbourne and Taylor, 1965). The proportion of the weathering loss of

radionuclides depends on a number of factors, e.g. solubility, adsorption to the plant's surface and shedding of epicuticular wax (IAEA, 2009; Milbourne and Taylor, 1965). The cuticle layer of the epidermis of leaves is rather impermeable, but it contains cracks and defects through which radionuclides can enter the plant (Handley and Babcock, 1972; Hossain and Ryu, 2009; Tukey *et al*., 1961). The rate at which radionuclides can enter plants through their cuticle layer depends among others on temperature, light, pH, the carrier of the radionuclides in the solution, the valence of the radionuclides and the type of crop (Marschner, 1986: Tukey *et al*., 1961). The length of the time period between interception of the radionuclides and the harvest of the crop also has an effect on the redistribution of the radionuclides within the crop (Coughtrey and Thorne, 1983; Kirchmann *et al*., 1967; Tukey *et al*., 1961). For grass crops, the concentration of radiocaesium in the biomass is likely to be highest in the first year after fallout. However, the transfer to the second cut can also be high, due to contributions from the previous seasons (Rosén *et al*., 1996; Rosén and Vinichuk, 2014).

The proportion of radioactivity directly intercepted by standing crop can be estimated as the interception fraction (*f*), which is defined as the ratio between the activity concentration in the above ground plant biomass directly after deposition (Bq m^{-2}) and the total amount of radionuclides deposited per unit area (Bq m^{-2}) (IAEA, 2010; Pröhl, 2009). Interception of radionuclides depends strongly on the biomass, and to account for this dependency the mass interception fraction (f_B, m^2 kg^{-1}) is introduced. The mass interception fraction is defined as the normalisation of the interception fraction of the above ground plant biomass (*B*, kg m^{-2}, dry weight) (IAEA, 2010; Pröhl, 2009). The weathering effects of intercepted radionuclides can be estimated as the weathering half-life (T_w, days), which is defined as when 50% of the initial intercepted activity of radionuclides on the crops is obtained (IAEA, 2009). The transfer of radionuclides from the environment to crops in a given situation can be described by an aggregated transfer factor (T_{ag}, m^2 kg^{-1}); which is defined as the ratio between the post-deposited retained activity concentration of radionuclides in the crops (Bq kg^{-1}) and the amount of radionuclides deposited per unit area (Bq m^{-2}) (Dahlgaard, 1994; Howard et al., 1996; IAEA, 2010) T_{ag}-values calculated in connection to a previous deposition, or experimentally determined, can be used to evaluate the situation after a new radioactive deposition occasion occurs to facilitate decisions on countermeasures for prevention or at least reducing the potential transfer of radionuclides to animal fodder (Bengtsson *et al*., 2013; Howard *et al*., 1996; Kostiainen *et al*., 2002; Rosén *et al*., 1996; Salbu, 2000).

The aims of this study were to measure the interception and activity concentrations of wet deposited ^{134}Cs and ^{85}Sr in ley at different growth stages; to examine the relationships between interception and activity concentrations and biomass in LAI and growth stage; to calculate the transfer of ^{134}Cs and ^{85}Sr in the above ground biomass through T_{ag}-values. The hypothesis was ^{134}Cs and ^{85}Sr interception and activity are related to the type of radionuclide, the biomass, the LAI and the growth stage of the plant at the time of deposition.

2. Materials and methods

2.1 Study area

The study was conducted at the Ultuna meteorological and agricultural field station in Uppsala, Sweden (59°48′45″N and 17°38′45″E). The soil's texture was: clay (60% clay, 20% silt and 20% sand), pH 6.5, plant available (ammonium lactate-acetate) phosphorus (P) 57 mg kg^{-1}, potassium (K) 202 mg kg^{-1}, and calcium (Ca) 3692 mg kg^{-1}, total (HCl-soluble) P 638 mg kg^{-1}, K 5720 mg kg^{-1} and Ca 6236 mg kg^{-1} (Bengtsson *et al*., 2012; Sandsborg and Wilert, 1975/1976).

The long-term (30 years, 1961-1990) annual mean air temperature was 5.6°C and the annual mean precipitation sum was 588 mm (SMHI, 2012). The growing season lasted from the 1st May to the 30th September, in both 2010 (1st year) and 2011 (2nd year). The mean annual air temperature of the growing season (1st May to 30th September) was 15°C in both the 1st and 2nd years, and the precipitation sums were 293 mm and 287 mm in the 1st and 2nd years, respectively (Bengtsson *et al*., 2013; Department of Crop Production Ecology, 2013). Air temperature at the deposition and sampling occasions varied between 10 and 21°C, and there was very little or no precipitation in connection with deposition and sampling on any occasion during the study years. Wind speed at the deposition and sampling occasions was low and varied between 1.3 and 3.6 m s^{-1} in the 1st year, and, 1.3 and 2.7 m s^{-1} in the 2nd year (Department of Crop Production Ecology, 2013).

2.2 Design of the experiment

A trial with a randomised block design (each parcel was given a unique number and these numbers were randomised by using the computer program Excel 2010 v. 14.0.7128.5000 (© Microsoft Corporation, (2010)), with 81 parcels (27 parcels (including 6 control parcels) × 3 replicates) of 1 × 1 m^2 parcels was laid out in 2010 (Table 1). In order to cover seasonal variations, a new trial was laid out, with another 81

parcels, in the adjacent field in 2011 (with already developed ley). The experimental ley was a mixture of clover and grass, which was composed of: 6% red clover (*Trifólium praténse* L.), 4% white clover (*Trifólium repens* L.), 60% timothy (*Phleum praténse* L.), and 30% meadow fescue (*Festúca praténsis* L.). Ley was sown in the middle of May in 2010 on bare soil. Seedling rate was 25 kg ha^{-1}, and fertiliser rates were equivalent to 104 kg N ha^{-1} and 19 kg P ha^{-1}. As clay soil has a relatively high natural capacity for delivering K through weathering (202 mg kg^{-1}) corresponds to a K-AL class of IV-V (of V), no K-fertiliser was needed according to Swedish standards (Yara International ASA, 2013).

The treatments in the experiment consisted of deposition of the ^{134}Cs (half-life of 2.07 years) and ^{85}Sr (half-life of 64.9 days) at six different growth stages, each treatment was carried out on three replicates. The growth stages were a combination of the growth stages of grass and clover (grass/clover). The growth stages, according to Halling (2005), were at the time of deposition in the 1st year, rising, code 0:0; leaves, code 1:1 (leaves, leaves and petiole); tillering/growth of internode, code 2:2 (one node visible, most plants have visible internodes); beginning stem extension/initial budding, code 3:3 (part of spike and panicle visible:major stalk buddings visible); stem extension/initial flowering, code 4:5 (flag leaf visible:flowers visible on major stalk); and, spike and panicle/flowering, code 5:6 (spike and panicle fully visible:flowers visible on major stalk and side stalks) (Table 2).

The growth stages in the 2nd year at the time of deposition were somewhat more advanced compared to the 1st year; the growth stages at the time of deposition in the 2nd year were spike and panicle/initial budding, code 4:3 (spike and panicle fully visible:separated buds in bud cluster); flowering/initial flowering, code 6:5 (full flowering:flowers visible on major stalk); flowering/flowering, code 6:6 (full flowering:flowers visible on major stalk and side stalks); post-flowering/post-flowering, code 7:7; beginning stem extension/flowering, code 3:6 (part of spike and panicle visible:flowers visible on major stalk and side stalks); flowering/post-flowering, code 6:7 (full flowering:post-flowering) (Table 2).

A sampling of the above ground biomass was done directly after deposition and at the subsequent growth stages each time a new treatment started; thus, the number of samplings depended on how early in the season the treatment started, the first one had six samplings in total, the second one had five samplings in total etc. (Table 1). In addition three control plots were sampled every event.

The normal harvest of ley in the 1st year corresponded to the growth stage code 5:6 (2nd of September). In the 2nd year, the first normal harvest corresponded to the growth stage code 4:3 (26th of May); sub-sequent regrowth after code 7:7 rendered a second normal harvest at the growth stage with code 3:6 (24th of August).

2.3 Deposition of artificial radioactive rain

In both years, the artificial rainwater was prepared from stock solutions and applied with a rainfall simulator, as described by Bengtsson *et al*. (2012). The stock solutions contained 5 MBq L^{-1} ^{134}Cs and 15 MBq L^{-1} ^{85}Sr in the 1st year, and 40 MBq L^{-1} ^{134}Cs and 37 MBq L^{-1} ^{85}Sr in the 2nd year. In the 1st year, ^{134}Cs was in the form of caesium chloride (CsCl) in a 0.1 M HCl solution (expanded uncertainty of ± 0.68%) (GE™ Healthcare Limited, Amersham, UK), and ^{85}Sr was in the form of strontium chloride ($SrCl_2$) in a 0.5 M HCl solution (no expanded uncertainty provided) (Eckert & Ziegler™, Santa Clarita, CA, USA). In the 2nd year, ^{134}Cs was in the form of CsCl in a 0.1 M HCl solution (expanded uncertainty of ± 2.5%) (Areva Cerca Lea, Pierrelatte Cedex, France). In the 1st year, the amount of ^{134}Cs applied at different growth stages ranged from 24.5 to 30.9 kBq m^{-2} and ^{85}Sr ranged from 28.5 to 49.8 kBq m^{-2} (details are given in Table 2). In the 2nd year, the amounts of ^{134}Cs ranged from 40.5 to 41.0 kBq m^{-2} and ^{85}Sr ranged from 39.5 to 41.0 kBq m^{-2} (details are given in Table 2).

Due to stock solutions in the 1st year being prepared with ultra-purified water in glass bottles, there was an adsorption of the radionuclides on the glass surface-Si-OH group of the bottles through ion exchange adsorption (Lehto and Hou, 2011), so the amount of ^{134}Cs and ^{85}Sr that were applied differed between the two years. To minimise adsorption of the radionuclides in the 2nd year, stable isotopes of Cs and Sr in the form as CsCl (150 g L^{-1}) in a 0.1 M HCl solution and $SrCl_2$ (150 g L^{-1}) in a 0.1 M HCl solution were added to the stock solutions, and they were stored in hydrophobic plastic bottles.

The amount of precipitation applied at each treatment was 1.00 ± 0.01 mm with an intensity of 1 mm 30 s^{-1} on a 60 × 60 cm^2 area in the middle of each parcel. A windshield was used to prevent wind disturbance during deposition in the early growth stages.

2.4 Samplings and analysis

The plants in three replicates were cut 5 cm above the soil surface (normal for harvest of a ley crop, and as well to avoid soil contamination) within a frame (25 × 25 cm^2 square) in the middle of each parcel; two-three hours after deposition, additional replicates of each treatment were sampled with the following deposition occasions of all ongoing treatments (Table 1). In addition three replicates of control were sampled for each sampling occasion. The plant material was

weighed fresh, and then air dried (at maximum of 40°C for about 14 days) before being re-weighed for determination of the dry weight (d.w.). Thereafter, the plant material was milled through a 2 mm sieve and placed in 35 mL or 60 mL plastic jars with a suitable geometry for measuring activity concentration.

Table 1. The schedule for the deposition and samplings, with three replicates for each combination of growth stage at deposition and sampling, where Y = both deposition and sampling, X = only sampling (time of deposition indicated by Y in the same row).

Year	Growth stage at deposition	Growth stage at sampling					
		0:0	1:1	2:2	**3:3**	**4:5**	**5:6**
1st	0:0	Y	X	X	X	X	X
	1:1		Y	X	X	X	X
	2:2			Y	X	X	X
	3:3				Y	X	X
	4:5					Y	X
	5:6						Y
		4:3	**6:5**	6:6	7:7	**3:6***	6:7*
2nd	4:3	Y	X	X	X	X	X
	6:5		Y	X	X	X	X
	6:6			Y	X	X	X
	7:7				Y	X	X
	3:6*					Y	X
	6:7*						Y

The activity concentration of the radionuclides were expressed as Bq kg^{-1} d.w. and corrected for the decay. The samples obtained of the early growth stages with small amounts of biomass were corrected for the degree of filling of the 35 mL jars, and a correction factor for each detector was determined as described Bengtsson *et al*. (2012).

Measurements of ^{134}Cs and ^{85}Sr activity concentration were performed by using High Purity Germanium (HPGe)-detectors (GMX-13200, GMX-33210, GMX-20200), and results were analysed and presented with the computer software Genie™ 2000 (© Canberra, Meriden, Connecticut, USA (2009)). The measured activity concentration included uncertainties of the efficiency calibration of the HPGe-detectors, which were assumed to be one of the main sources of the total uncertainty (Bronson *et al*., 2008; Bengtsson *et al*., 2012). Uncertainty of the total amount of radionuclides deposited was estimated according to the method described in the *Guide to the Expression of Uncertainty in Measurements* (GUM) (Ellison et al., 2000; ISO, 1993), and reported as the combined standard uncertainty $u_c(y)$ for the total amount of radionuclides deposited. For the interception fraction (f), the mass interception fraction (f_B) and the aggregated transfer factor (T_{ag}), the standard deviation was reported.

2.5 Calculations of interception and transfer

The interception fraction (f) was calculated as the ratio between the activity in the above ground biomass directly after deposition (A_i, Bq m^{-2}, d.w.), and the total amount of activity deposited (A_t, Bq m^{-2}, d.w.) (Eq. (1): (IAEA, 2010; Pröhl, 2009)).

$$f = A_i/A_t \qquad [-] \qquad (1)$$

The mass interception fraction (f_B, m^2 kg^{-1}) was calculated by normalising the interception fraction to the above ground plant biomass (B, kg m^{-2} d.w.) (Eq. 2): (IAEA, 2010; Pröhl, 2009)).

$$f_B = f/B \qquad [m^2\,kg^{-1}] \qquad (2)$$

The aggregated transfer factors (T_{ag}, m^2 kg^{-1}) were calculated as the activity concentration of the specific radionuclide in the plants at harvest (sampling) (A_c, Bq kg^{-1}, d.w.), divided by the amount of deposited activity (A_t, Bq m^{-2}) (Eq (3): (Dahlgaard, 1994; IAEA, 2010)).

$$T_{ag} = A_c/A_t \qquad [m^2\,kg^{-1}] \qquad (3)$$

2.6 Statistics

Analyses of variance (ANOVA) were applied for ^{134}Cs and ^{85}Sr interception (only in the 2nd year due to the different amounts in the 1st year) to determine whether the interception was affected by the type of radionuclide. ANOVA were applied for the Tag-values of ^{134}Cs and ^{85}Sr to determine whether the Tag-values were affected by the growth stage, the year of deposition, or the type of radionuclide. To determine whether different types of radionuclides affected the total concentration of the two radionuclides in the ley at different sampling occasions, the ^{85}Sr interception from the linear regression was plotted against the ^{134}Cs interception, with one plot for each year. All statistical analyses were computed with the program R version 2.15.2 (© The R Foundation for Statistical Computing, Vienna, Austria, 2012).

Table 2. Shows the above ground plant biomass (kg m^{-2}) and LAI (m^2 m^{-2}), the means with standard deviation, the total amount (A_t) of ^{134}Cs and ^{85}Sr (kBq m^{-2}) deposited with the total uncertainty (u_c); the intercepted amount (A_i) of ^{134}Cs and ^{85}Sr (kBq m^{-2}) with standard deviation; the interception fraction (f) of ^{134}Cs and ^{85}Sr with standard deviation; the mass interception fraction (f_B) of ^{134}Cs and ^{85}Sr (m^2 kg^{-1} × 10^{-3}) with standard deviation. The means are of three replicates (n = 3).

Year	Growth stage at deposition	Biomass	LAI	Total amount deposited (A_t)		Intercepted amount (A_i)	
				^{134}Cs	^{85}Sr	^{134}Cs	^{85}Sr
1st	0:0	0.002±0.001	-	24.5±0.2	41.9±1.3	0.0±0.0	0.0±0.0
	1:1	0.008±0.03	0.6±0.2	26.8±0.3	28.5±0.9	0.2±0.2	0.27±0.2
	2:2	0.04±0.02	1.5±0.4	25.9±1.9	41.2±1.2	0.6±0.3	1.42±0.5
	3:3	0.09±0.05	2.3±0.7	26.8±1.7	38.5±2.7	1.0±0.7	2.13±1.4
	4:5	0.17±0.03	4.4±0.4	30.9±2.0	49.8±1.8	2.7±1.1	4.85±1.7
	5:6	0.35±0.15	5.7±0.6	26.6±1.7	49.4±3.4	7.3±1.8	15.8±4.5
2nd	4:3	0.06±0.06	1.6±0.8	40.6±0.34	39.5±0.9	1.0±1.0	1.0±1.1
	6:5	0.93±0.56	1.8±0.6	40.5±0.34	40.6±0.9	13.9±15.3	15.0±15.7
	6:6	0.67±0.14	1.1±1.8	41.0±0.4	40.6±0.9	12.6±1.9	13.3±1.4
	7:7	0.33±0.09	0.9±0.9	41.0±0.4	41.0±0.9	6.5±2.8	6.9±3.0
	3:6*	0.20±0.09	0.5±0.5	40.6±0.4	40.6±0.9	8.5±2.9	8.6±2.9
	6:7*	0.17±0.08	2.2±2.2	41.0±0.4	40.4±0.9	3.9±2.0	5.1±2.5

3. Results and Discussion

3.1 Interception of ^{134}Cs and ^{85}Sr

For both radionuclides, the fraction intercepted by ley increased with the increase in above ground biomass and LAI in both years (Fig. 1). The relationship between the intercepted fraction (*f*) of radionuclides and above ground biomass (d.w.) was significant in both years (Figs. 1A and B). A similar relationship between *f* and above ground biomass was found by Vandecasteele *et al*. (2001) for wheat, although their values for this crop were higher (0.84 for ^{134}Cs and 0.88 for ^{85}Sr). The relationship between the intercepted fraction of each radionuclide and the LAI in both years was also significant (Figs. 1C and D), although a weaker relationship was found for the 2nd year.

The intercepted fraction (*f*) and mass intercepted fraction (f_B) of both radionuclides were highest after deposition just before the last normal harvest (growth stage 5:6) in the 1st year (Fig. 2 and Table 2) and the control plots did not intercepted any activity of the radionuclides as they did not receive any radioactive deposition. In the 2nd year, *f* was low at the first deposition occasion, and then increased drastically at the 2nd deposition occasion, followed by a gradual decrease at later depositions occasions. For the f_B, the lowest values for both radionuclides were at the first deposition occasion, and then the values increased at the 2nd deposition occasion, followed by more or less the same magnitude at later deposition occasions, followed by a drastic increase at the deposition at growth stage 3:6. Variations in *f* can be explained by the variation from the biomass (from 0.35 ± 0.15 kg m^{-2} at last sampling occasion (growth stage 5:6) in the 1st year, to 0.06 ± 0.06 kg m^{-2} at the first sampling occasion (growth stage 4:3) in the 2nd year. In the 2nd year, the ley had a quick regrowth in the beginning of the growing season, which decreased its (living) biomass later on as a fold of the ageing and dry spells (Table 2), and the harvest further reduced the biomass. Intercepted *f*- and f_B-values were higher for ^{85}Sr than for ^{134}Cs at several growth stages. Higher interception values for radiostrontium than for radiocaesium were also observed for spring wheat by Bengtsson *et al*. (2012) and Vandecasteele *et al*. (2001). Divalent ions, like ^{85}Sr, are expected to be more efficiently adsorbed on plant surfaces than monovalent ions, like ^{134}Cs (Aarkrog, 1969; Bréchignac *et al*., 2000; Vandecasteele *et al*., 2001).

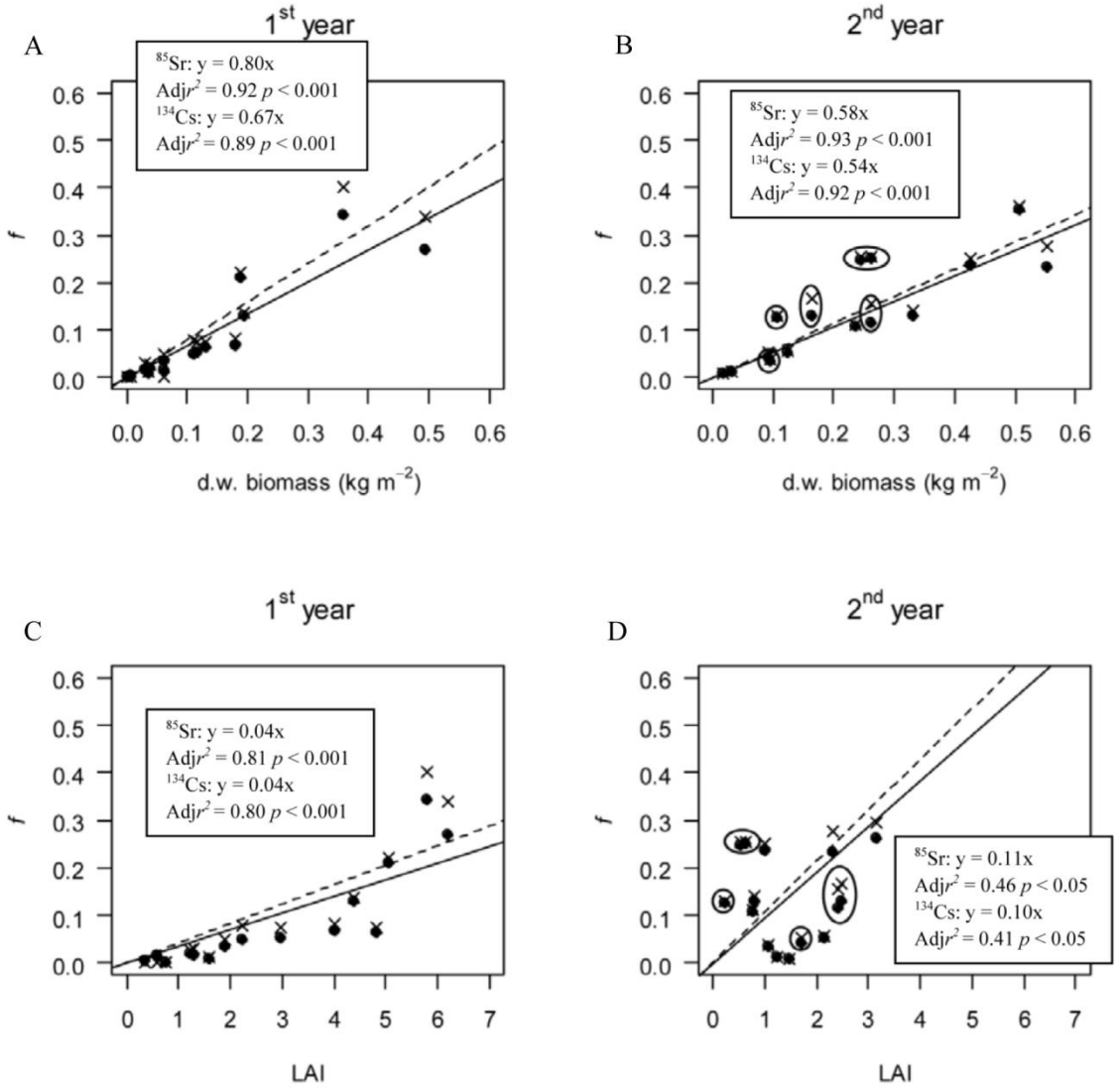

Figure 1A-D. The relationship between interception fraction (*f*) for ^{134}Cs (●, —) and ^{85}Sr (×, - -) deposited on a ley and above ground plant biomass d.w. (1A and 1B) and LAI (1C and 1D). The encircled points in 2nd year are values measured in normal sampling times for the second harvest of a regrowth.

This effect might be more pronounced when sampling occurs shortly after deposition; as in this short time-lag, negligible amounts of radionuclides were washed-off. However, the fraction of radionuclides intercepted by ley was not only related to the valence of radionuclides, but it was also related to the water storage capacity and accumulation of the radionuclides on crop surfaces (Kinnersley *et al*., 1997). Our results might have been influenced by the high rain intensity during the 30 second application (1 mm 30 s^{-1}). The bigger drops connected to higher rain intensities tend to splash off the plants to a higher degree than the smaller ones at lower rain intensities (Keim *et al*., 2006; Wang *et al*., 2005). Moreover, the application of more than 1 mm of rain could have resulted in a lower interception of radionuclides, because as the maximum water storage capacity on the crops surfaces is reached more quickly, further water will run off from the crops surfaces (Kinnersley *et al*., 1997).

As an average, ley intercepted (mean values of three replicates) up to 27% (7 kBq m^{-2}) of the deposited amount of ^{134}Cs in the 1st year, and up to 34% (14 kBq m^{-2}) (mean values of three replicates) in the 2nd year. This is quite similar to the data reported by Eriksson *et al*. (1998), where up to 39% of ^{134}Cs was intercepted by ley. The percentage of intercepted ^{85}Sr (mean values of three replicates) were up to 32% (16 kBq m^{-2}) in the 1st year and up to 37% (15 kBq m^{-2}) (mean values of three replicates) in the 2nd year; which is in agreement with data obtained by Milbourne and Taylor (1965), where up to 30% of the deposited amount of ^{85}Sr was

intercepted on a permanent pasture. However, it is possible that interception of radionuclides could have been underestimated rather than overestimated, since some of the amount of radioactivity was likely to be intercepted by the lower parts of the plant.

In a comparable experiment, Chadwick and Chamberlain (1970), found the interception of grass to be in a similar range (^{85}Sr: 0.20-0.82), as in the presented study (^{134}Cs: >0.01-0.76; ^{85}Sr: >0.01-0.80). Similarly, Eriksson *et al*. (1998) and Madoz-Escande *et al*. (2004) found in a comparable experiment on leys and common beans that the highest *f* for ^{134}Cs (0.71, ours: 0.76) and for ^{85}Sr (1.11, ours: 0.80) is 24 h after deposition, with *f* increasing with the biomass.

Based on ANOVA analyses, the *f*-values for the 2nd year were more or less the same for ^{85}Sr and ^{134}Cs (p = 0.80 when testing the hypothesis that they were different) when compared over different deposition occasions. ANOVA analyses were done only for the 2nd year, as higher amount of ^{85}Sr was deposited in the 1st year due to experimental error (Table 2).

An intercepted fraction that has been determined in one situation needs to be used with caution for prediction of crop contamination in a new situation, since its magnitude can vary with plant type, type of radionuclide, magnitude and intensity of rainfall, the concentration of radionuclides in the rain, weather conditions, and biomass (Bengtsson *et al*., 2012; Hoffman *et al*., 1992; Kinnersley *et al*., 1997).

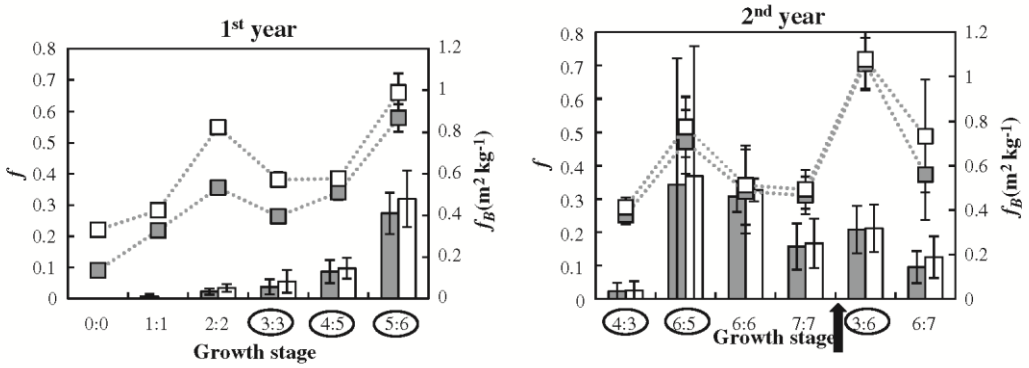

Figure 2. The interception fraction (*f*) and the mass interception fraction (f_B) after deposition of ^{134}Cs (■) and ^{85}Sr (□) at different growth stages in a ley (n = 3 at all growth stages). The error bars indicate standard deviation. The arrow in the 2nd year diagram indicates when regrowth started after the first harvest.

The relationship between radionuclide intercepted fraction and LAI in both years was also significant (Figure 1, ^{134}Cs interception / ^{85}Sr interception vs. LAI, and by year), showing that the interception increases with increasing LAI.

An intercepted fraction determined in one situation need to be used with caution for prediction of crop contamination in a new situation since its magnitude varies with plant type, type of radionuclide, level of rainfall and the concentrations of radionuclides in the rain (Hoffman *et al*., 1992; Kinnersley *et al*., 1997).

The fraction of radionuclides intercepted by ley is not directly related to the intensity of rainfall but rather to the water storage capacity and accumulation of the radionuclides on the crop surfaces (Kinnersley *et al*., 1997). Still, our results might have been influenced by the high rain intensity during the 30 second application (1 mm 30 s^{-1}). Higher rain intensity could have resulted in lower values of interception, as bigger droplets tend to splash off the plants to a higher degree than at lower rain intensities with smaller drops (Keim *et al*., 2006; Wang *et al*., 2005). The risks that water interception capacity was exceeded at prevailing LAI was low as only 1 mm was added (Kinnersley *et al*., 1997).

3.2 Activity concentration of ^{134}Cs and ^{85}Sr

The weathering half-life (T_w) of the retained intercepted radionuclides was in general much shorter at the earlier growth stages when the ley had a quick growth of its biomass (10 days for ^{134}Cs and 8 days for ^{85}Sr) (Table 3). At later growth stages the weathering half-life was in general longer for both radionuclides (24 days for ^{134}Cs and 22 days for ^{85}Sr).

Table 3. Shows the weathering half-life (T_w) of ^{134}Cs and ^{85}Sr (days), including growth dilution.

Year	Growth stage at deposition	Weathering half-life (T_w)	
		^{134}Cs	^{85}Sr
1st	0:0	8†	8 (8–8.5)
	1:1	8.5 (8–9)	8 (7.5–9)
	2:2	6 (5.5–6)	5 (5–5.5)
	3:3	15.5 (15.5–16)	15 (15–15.5)
2nd	4:3	10 (7–13.5)	7.5 (6.5–9)
	6:5	14 (13.5–14.5)	7 (6.5–7)
	6:6	26 (15.5–44)	24 (14.5–41)
	7:7	21 (13–31.5)	20 (13–30.5)

The means and ranges are of three replicates (n = 3), except where †n = 1. The two last deposition occasions were not included due to lack of data.

The activity concentration of both radionuclides was in general lower in the 1st year than in the 2nd year, and this could be explained by the fact that the ley was established in the 1st year on bare soil and/or also by yearly variations in the season (Fig. 3). It should be borne in mind that in the 1st year, normal harvest (growth stage 5:6) was made very late in the season; i.e. in the beginning of September, since the ley grass was sown in the spring and needed some time for establish itself. In the 2nd year, the 1st normal harvest (growth stage 4:3) occurred in the beginning of June. The controls did not have any activity concentrations of the radionuclides, as they did not receive any radioactive deposition.

Generally, the closer the sampling after deposition and the more developed the crop, the higher the activity concentration of both radionuclides (Fig. 3). This is in agreement with data obtained by Mårtensson (2012) for ley. The dynamics of the activity concentrations showed that quite some radioactivity was lost between sampling occasions, for instance between sampling at growth stage 3:3 and 4:5 for the 2nd, 3rd and 4th deposition in the 1st year, and sampling at growth stage 6:5 in the 2nd year; the explanation for this could be related to precipitation.

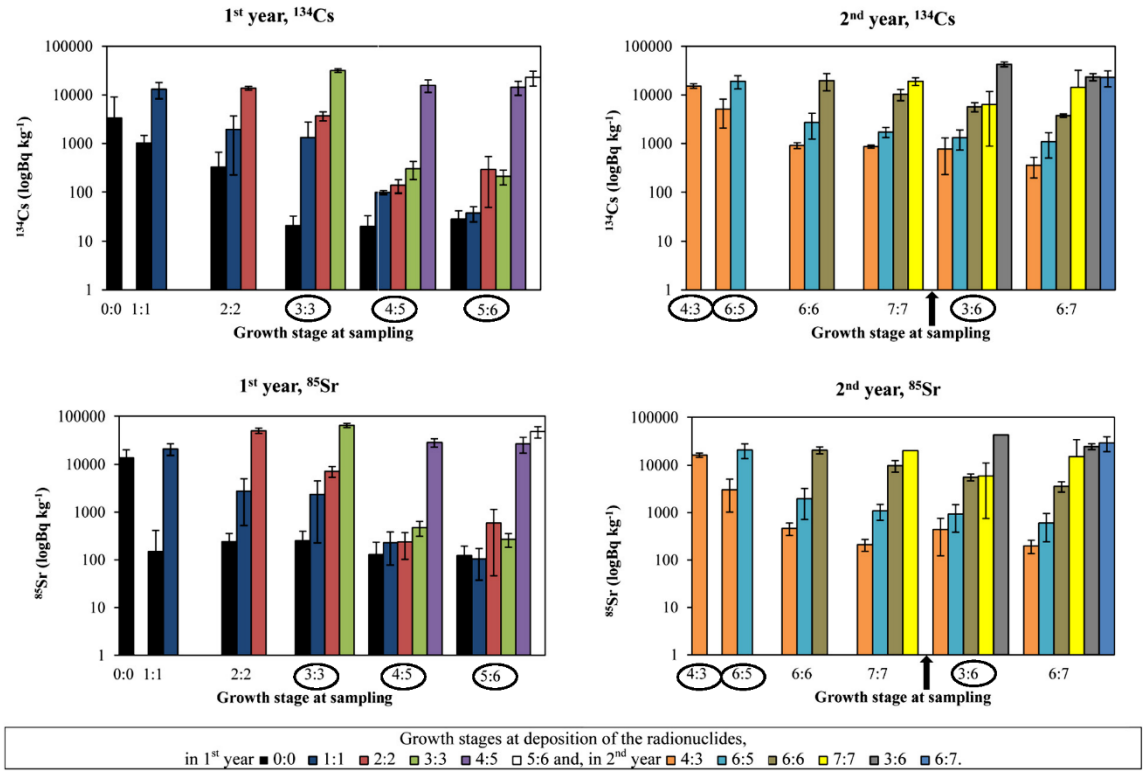

Figure 3. Logged activity concentrations of ^{134}Cs and ^{85}Sr at different sampling occasions after deposition at different growth stages (n = 3 at all growth stages except at deposition/sampling in the 2nd year at deposition/sampling (6:5/6:6) where n = 2). Error bars indicate standard deviation. Encircled are the growth stages at which harvest normally occurs. The arrow indicates when regrowth started after the first harvest in the 2nd year.

In general, in the 1st year the activity concentrations of radiostrontium in the ley were found to be higher than that of radiocaesium; however, this could be explained by the higher amount of ^{85}Sr deposited due to experimental error (Table 2). In the 2nd year, the activity concentrations of radiocaesium in the ley were found to be higher than that of radiostrontium, and this was in agreement with Eriksson *et al*. (1998), who found that the activity concentration of radiocaesium in ley was generally higher than that of radiostrontium. All comparisons between radionuclide accumulation in the ley were significant (AdjR^2 = 0.99, p < 0.05 for both years and dy/dx was 1.16 1st year and 1.07 2nd year, with intercept of 0.00 in both years) and plots of the data can be found in the Appendix in Supplementary Data 2. This indicates that both radionuclides were similarly absorbed by the plant tissue. The smaller amount of radionuclides absorbed by the ley at deposition in early growth stages could be due to more wash-off after fall-off in early growth stages, and the dilution effects because more rapid growth of the plant in the early stages took place (Colle *et al*., 2009; Eriksson *et al*., 1998).

The T_{ag}-values (Table 4) varied between treatments in a similar way to the activity concentrations of the two radionuclides, since a more or less equal amount of radioactivity was applied per square meter in the 2nd year in all treatments and on all occasions (*i.e.* the denominator in Eq. (2) is constant).

Table 4. Shows aggregated transfer factors (T_{ag}) (m^2 kg^{-1} × 10^{-3}) of ^{134}Cs and ^{85}Sr for the different combinations of wet-depositions and samplings of ley at different growth stage, with the mean and standard deviation of three replicates (n = 3).

Year	Growth stage at deposition	^{134}Cs				
		Growth stage at sampling				
		1:1	2:2	**3:3**	**4:5**	**5:6**
1st	0:0	28±27	6.2±1.8	**0.29±0.5**	**0.55±0.61**	**1.2±0.58**
	1:1		73±65	**50±53**	**2.5±1.8**	**1.4±0.49**
	2:2			**143±31**	**5.3±1.6**	**11±9.4**
	3:3				**11±4.6**	**7.9±2.7**
	4:5					**468±177**
		6:5	6:6	7:7	**3:6***	6:7*
2nd	4:3	**126±75**	23±3.1	21±2.9	**19±13**	8.8±4.0
	6:5		67±37	43±8.4	**32±15**	18±19
	6:6			252±62	**140±29**	92±7.1
	7:7				**155±134**	352±430
	3:6*					581±97
		^{85}Sr				
		Growth stage at sampling				
		1:1	2:2	**3:3**	**4:5**	**5:6**
1st	0:0	3.6±6.2	3.9±3.9	**2.0±3.5**	**2.1±2.5**	**3.0±1.7**
	1:1		97±79	**83±75**	**4.1±5.5**	**3.7±2.4**
	2:2			**175±45**	**5.8±3.3**	**14±13**
	3:3				**12±4.2**	**7.0±2.3**
	4:5					**544±198**
		6:5	6:6	7:7	**3:6***	6:7*
2nd	4:3	**77±51**	12±3.5	5.4±1.5	**11±8**	5.0±1.6
	6:5		48±31	27±9.9	**23±13**	10±11
	6:6			241±67	**138±23**	88±22
	7:7				**150±127**	371±473
	3:6*					609±87

* indicates regrowth with a second harvest in the 2nd year (2011) after growth stage 7:7. The growth stages relevant for a normal harvest of ley are bolded.

Since the aggregated transfer factors have a tendency to get higher the closer the deposition occasions were to the sampling occasion, the interception alone does not explain the final activity concentration in the crop. In the 2nd year, the T_{ag}-values increased even when the interception decreased, and this can be explained by the fact that a lower biomass will contribute to higher T_{ag}-values, even with lower interception (see Table 2 for values on the biomass). Other contributing factors could be the dilution of radionuclide concentration due to the growth of the biomass (Coughtrey and Thorne, 1983), the leaves falling-off during the time between the deposition and sampling occasion (Colle *et al*., 2009; Eriksson *et al*., 1998) and the decay rate of the radionuclides (Choi *et al*., 2002).

According to the ANOVA test, T_{ag}-values for both radionuclides were dependent on the growth stage of the crops ($p < 0.05$), and on the year of the experiment ($p < 0.001$). However, the T_{ag}-values did not differ significantly between the radionuclides ($p = 0.93$), as they ranged between <0.00 and 0.849 for ^{134}Cs and <0.00 to 0.917 for ^{85}Sr. A similar range in T_{ag}-values (^{134}Cs: 0.005-1.594; ^{85}Sr: 0.000-1.140) was reported by Eriksson *et al*. (1998). Even if the transfer factors were in agreement with those in other studies, the variation of the T_{ag}-values stresses the uncertainty of Tag-values, and the need for more experiments and analyses to be done with emphasis placed on weather. For this reason, preliminary assessment of activity concentration in fodder crops needs to be done by continuous sampling and monitoring in the case of a radioactive deposition.

4. Conclusions

The interception of radionuclides was highest when deposition occurred at the well-developed ley during the later or latest growth stages before harvest. For the magnitude of deposition chosen in our experiment, it can be concluded that the above ground biomass is a good predictor and the LAI a more uncertain predictor of the interception of radiocaesium and radiostrontium by ley grass and clover. The T_w-values of retained intercepted radionuclides were much higher when deposition occurred at the well-developed ley during the later or latest growth stages before harvest.

The variation in magnitude of aggregated transfer factor values between different deposition occasions fits well with those found in other studies. However, it should be noted that there are few investigations where transfer factors are related to deposition rates and to the deposition of radionuclides in growing ley. The variation in Tag-values, which could then be used as trustworthy prediction measures of radionuclides in crops after possible future fallout

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Supplementary data 1

The activity concentration (Bq kg^{-1}) of ^{134}Cs and ^{85}Sr at different harvest occasions after deposition at different growth stages. The means and standard deviation of three replicates (n = 3). * indicates regrowth within the second harvest in the 2nd year (2011) after growth stage 7:7.

Year	Growth stage at deposition	^{134}Cs Growth stage at sampling					
		0:0	1:1	2:2	3:3	4:5	5:6
1st	0:0	3333±5774	1025±442	327±336	21±12	20±13	29±14
	1:1		4850±13196	1955±1731	1331±1421	99±8	38±13
	2:2			13822±1199	3709±796	138±43	293±243
	3:3				31845±2752	304±122	211±71
	4:5					15800±4613	14453±5458
	5:6						23133±7849
		4:3	6:5	6:6	7:7	3:6*	6:7*
2nd	4:3	15333±1662	5133±3054	913±126	866±116	768±538	357±162
	6:5		19100±5821	2720±1485	1733±339	1321±588	1086±586
	6:6			19867±7736	10333±2538	5723±1196	3763±290
	7:7				19100±3477	6357±5475	14433±17645
	3:6*					42867±4800	23567±3951
	6:7*						22967±8344
		^{85}Sr Growth stage at sampling					
		0:0	1:1	2:2	3:3	4:5	5:6
1st	0:0	13880±6317	151±262	116±82	252±146	130±104	124±70
	1:1		21274±5836	2243±1295	2366±2139	232±153	106±68
	2:2			50973±6389	7191±1834	238±135	594±316
	3:3				66101±6872	477±163	270±87
	4:5					287667±5771	27100±9875
	5:6						48900±13066
		4:3	6:5	6:6	7:7	3:6*	6:7*
2nd	4:3	16300±1735	3044±2021	465±136	212±59	440±317	199±62
	6:5		20967±5490	1960±1247	1090±401	927±540	602±359
	6:6			20800±7227	9787±2707	5593±941	3580±875
	7:7				20233±3564	5941±5189	15213±19408
	3:6*					43700±5333	24733±3513
	6:7*						29600±10227

Supplementary data 2

Selected linear regression models for estimating activity concentration in the two crops (^{134}Cs interception fraction vs. ^{85}Sr interception fraction), see table 3. In the 1st year, a much higher amount of ^{85}Sr was deposited than that of ^{134}Cs. In the 2nd year, the amounts of both radionuclides were more or less the same (see table 1). (--) represents the 1:1 line.