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Influence of development stage of spring oilseed rape and spring wheat on interception of wet-deposited radiocaesium and radiostrontium.

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Abstract

The dry and wet deposition of radionuclides released into the atmosphere can be intercepted by vegetation in terrestrial ecosystems. The aim of this study was to quantify the interception of wet deposited $^{134}$Cs and $^{85}$Sr by spring oilseed rape ($Brassica napus$ L.) and spring wheat ($Triticum aestivum$ L.). The dependency of the intercepted fraction ($f$) on total above ground plant biomass, growing stage and the Leaf Area Index (LAI) was quantified. A trial was established in Uppsala (east central Sweden), with land management in accordance to common agricultural practices. The field trial was a randomised block design of $1 \times 1$ m$^2$ parcels with three replicates. During the growing season of 2010, a rainfall simulator deposited $^{134}$Cs and $^{85}$Sr during six different growth stages. Two to three hours after deposition, the biomass of the centre $25 \times 25$ cm$^2$ area of each parcel was sampled and above ground biomass and LAI were measured. The radioactivity concentration and radioactivity of samples were measured by High Purity Germanium (HPGe)-detectors.

For $^{134}$Cs, there was a correlation between $f$ and LAI ($r^2 = 0.55$, $p < 0.05$) for spring wheat, but not for spring oilseed rape ($r^2 = 0.28$, $p > 0.05$). For $^{85}$Sr, there was a correlation between $f$ and LAI for both crops ($r^2 = 0.41$, $p < 0.05$ for spring oilseed rape and $r^2 = 0.48$, $p <$
0.05 for spring wheat). There was no correlation between $f$ and above ground plant biomass in spring oilseed rape for either $^{134}$Cs ($r^2 = 0.01, p > 0.05$) or for $^{85}$Sr ($r^2 = 0.11, p > 0.05$). For spring wheat, there was a correlation for both $^{134}$Cs ($r^2 = 0.36, p < 0.05$) and $^{85}$Sr ($r^2 = 0.32, p < 0.05$). For spring oilseed rape, $f$ was highest at growth stage ‘stem elongation’ for $^{134}$Cs (0.32±0.22) and $^{85}$Sr (0.41±0.29). For spring wheat, $f$ was highest at growth stage ‘ripening’ for both radionuclides ($^{134}$Cs was 0.36±0.14 and $^{85}$Sr was 0.48±0.18). Thus, LAI can be used to quantify interception of both radionuclides for both crops, whereas, above ground plant biomass is a weak measure of interception of wet deposited radiocaesium and radiostrontium.

1. Introduction

The release of radionuclides into the atmosphere from different sources, for instance nuclear power accidents or test firing of nuclear weapons, can cause both the dry and wet deposition of radionuclides onto vegetation (Hoffman et al., 1995; Kinnersley et al., 1997). Some wet deposited radionuclides, e.g. radiocaesium, can be directly taken up by the vegetation through leaves (Middleton, 1958, 1959; Scotti and Carini, 2000). The strongest contamination of the food chain may occur during deposition onto standing crops during the growing season (Anspaugh et al., 2002), as when deposition occurs during the growing season, the uptake of radioactive substances through leaves is assumed larger than uptake through roots (Johnson et al., 1966; Russell, 1965). In the event of a nuclear power accident or an atom bomb explosion, the release of radionuclides comprises a large part of the collective dose to humans through intake of contaminated agricultural foodstuffs (Madoz-Escande et al., 2004; Middleton, 1958).

The level of radionuclide interception by different parts of important agricultural plants, e.g. grass, broad bean and wheat, may be dependent on plant morphology i.e. Leaf Area Index (LAI), the angle of leaves, above ground plant biomass, and the maximum water storage.
capacity of the plant canopy (IAEA, 2010; Kinnersley et al., 1997). Other factors affecting the level of interception include the physical and chemical form of the radionuclides, such as molecular mass and the valence (Salbu et al., 2004): divalent radiostrontium ions fix more easily to the surfaces of leaves than monovalent radiocaesium ions do (Mueller and Proehl, 1993; Vandecasteele et al., 2001). The size of the radioactive particles and fragments and the weather conditions i.e. precipitation and wind speed also affect interception (Aarkrog, 1975; Kinnersley et al., 1997). According to Hoffman et al. (1992), the interception of radionuclides is more dependent on the above ground plant biomass than on the amount of rainfall, and the time between deposition and harvest will affect the concentration of radionuclides in plants at harvest and depend on ‘field losses’, for example wash-off and volatilisation (Chadwick and Chamberlain, 1970).

Therefore, after wet deposition onto a growing crop, the potential risk of transfer to plant parts used for food production needs to be known in order to reduce transfer to humans. Information on the level of interception of radionuclides in different situations is essential for the risk assessment of transfer through the food chain and for planning effective countermeasures for reducing human exposure.

Radiocaesium and radiostrontium are the main harmful, long-living radionuclides released during a nuclear power plant accident and test firing of nuclear weapons. Radiocaesium spreads evenly in human bodies, somewhat more in muscles than in bones, and is the cause of different kinds of cancer. Radiostrontium accumulates in the human skeleton and presents an additional risk of cancer in young people.

The aim of this study was to quantify interception of wet deposited $^{134}\text{Cs}$ and $^{85}\text{Sr}$ by spring oilseed rape ($\text{Brássica napus}$ L.) and spring wheat ($\text{Triticum aestívum}$ L.) at different growth stages. The hypothesis was interception of radiocaesium and radiostrontium was related to above ground plant biomass, LAI, type of radionuclide, and type of crop.
2. Materials and methods

2.1 Study area

The study was conducted at the Ultuna meteorological and agricultural field station, Uppsala, Sweden (59°48’45”N and 17°38’45”E). The meteorological station monitors air temperature, precipitation, and wind speed (Table 1) (Karlsson and Fagerberg, 1995). The 30-year (1961-1990) annual mean temperature is 5.7°C and the annual mean precipitation sum is 545 mm (Geovetenskaper, 2012). During the growing season in 2010 (1st of May to 30th of September), the site had a mean temperature of 14.8°C and precipitation sum of 58.6 mm, according to data from the nearby Ultuna meteorological station. The temperature at deposition and sampling varied between 10 and 21°C and there was no precipitation in connection with deposition and sampling on any occasion. Wind speed at deposition and sampling varied between 1.3 and 3.6 m s⁻¹.

The texture of the soil at the site was loamy clay (Table 2): texture was determined through a combination of wet sieving for large particle size fractions and sedimentation analysis with the pipette method for finer fractions, with a modified method as described by Ljung (1987). Soil pH was measured in water with a soil:water ratio of 1:5. Soluble phosphorus (P), potassium (K) and calcium (Ca) were extracted with a solution of 0.10 M ammonium lactate and 0.40 M acetic acid at a pH of 3.75 (AL-solution), as described by Egnér et al. (1960). The amount of P, K, and Ca in the AL-extracts was determined by Inductively Coupled Plasma (ICP) analysis.

2.2 Design of the experiment

The trial was a randomised block design with 3 blocks including 1 × 1 m² parcels, with three replicates. The experimental crops, spring oilseed rape (*Brássica napus* L.) variety
‘Larissa’ and spring wheat (Triticum aestivum L.) variety ‘Triso’, were sown and managed according to common agricultural practices, except for covering the sowing beds with a non-woven fabric for three weeks.

The Swedish Radiation Safety Authority gave permission for this type of open field experiments with the requirement that isotopes with short half-life should preferentially be used. Thus, the isotopes selected for the field experiment were $^{134}$Cs (half-life of 2.07 years) and $^{85}$Sr (half-life of 64.9 days): it was assumed these isotopes behaved in the same manner as $^{137}$Cs and $^{90}$Sr. An artificial rain simulator was used to deposit $^{134}$Cs and $^{85}$Sr at six plant growth stages to each crop.

The experimental crops were sown in the middle of May. The seeding rates were 8 kg ha$^{-1}$ for spring oilseed rape and 230 kg ha$^{-1}$ for spring wheat. After sowing, the beds were covered with a non-woven polypropylene fabric for three weeks to promote quicker growth.

For both crops, fertiliser rates were equivalent to 104 kg N ha$^{-1}$ and 19 kg P ha$^{-1}$, no potassium (K) was added as illitic clay has high natural capacity for delivering K through weathering (ammonium lactate-acetate soluble K is 202 mg kg$^{-1}$ (Table 1), which according to Swedish standards indicates no demand for potassium fertiliser).

Radionuclides were deposited on spring oilseed rape at six growth stages, according to the BBCH scale by Hack et al. (1992) (Figure 1, sketch by Nigrinis, 2010 after Bayer Crop Science (2011a), Lancashire et al.(1991), and Weber and Bleiholder (1990)). These stages were: leaf development, code 13 (three leaves unfold); stem elongation, code 32 (two visible extended internodes); 10% of flowers on main raceme open, code 61; full flowering, code 65; beginning of ripening, code 80; and, fully ripe, code 89.

For the spring wheat, the corresponding growth stages, according to the BBCH scale were: tillering, code 21 (headshot and one side shot); stem extension, code 37 (flag leaf visible); flowering, code 65 (on-going flowering); ripening, code 70 (dough ripeness);
ripening, fully ripe, code 89; and, senescence, over-ripe, code 92 (Figure 2, after Bayer Crop Science (2011b), Lancashire et al.(1991), Witzenberger et al. (1989) and Zadoks et al. (1974)).

2.3 Preparation and deposition of artificial radioactive rain

The artificial rainwater was prepared from stock solutions containing 5 MBq L⁻¹ for ¹³⁴Cs and 15 MBq L⁻¹ for ⁸⁵Sr: ¹³⁴Cs was in the form of caesium chloride (CsCl) in 0.1 M HCl (expanded uncertainty of ±0.68%) (GE™ Healthcare Limited, Amersham, UK), and ⁸⁵Sr was in the form of strontium chloride (SrCl₂) in 0.5 M HCl (no expanded uncertainty provided) (Eckert & Ziegler™, Santa Clarita, CA, USA). The two radionuclides were mixed and diluted to the desired concentration in ultra-purified water (purity to 18.2 MΩ·cm (8 S cm⁻¹). The amount of ¹³⁴Cs applied was in the range 24.5±0.23 to 30.9±1.97 kBq m⁻² and the amount of ⁸⁵Sr applied was in the range 28.5±0.86 to 49.8±1.75 kBq m⁻².

The radionuclides were applied with a rainfall simulator that was a modified version of the drip infiltrometer described by Joel and Messing (2001). The amount of precipitation applied in each treatment was 1.00±0.01 mm at an intensity of 1 mm 30 s⁻¹. When deposition was in the early growth stages, a windshield was used to prevent wind disturbance.

2.4 Sampling and measurements analyses

Two-three hours after deposition, a sampling frame was placed in the central 25 × 25 cm² square of each parcel and the plants were sampled. The plant materials were weighed fresh, then air dried (at 30°C for a minimum of 14 days) before being re-weighed for dry weight (d.w.): the plants were then milled. After milling, samples were placed in 35 mL or 60 mL plastic jars with a suitable geometry for measuring radioactive concentration. The activity concentrations of the radionuclides were expressed as Bq kg⁻¹ d.w. and decay was corrected
for the sampling date. Due to a small amount of plant material, samples from early growth stages measured in 35mL jars were corrected for the degree of filling.

2.4.1 Measurement of Leaf Area Index

Leaf Area Index (LAI) is an indicator of the morphology of plant canopies and corresponds to layers of leaf biomass projected on the soil surface. LAI is determined by measuring the intensity of sunlight below the canopy (Anderssen et al., 1985; Lang and Yueqin, 1986; Lang et al., 1985). On the day of sampling, LAI was measured with a LAI-2000 device (© LI-COR Biosciences Inc., Nebraska, USA); the standard error was given by the device.

2.5 Analyses

The actual concentration of the radionuclides in the artificial rainwater and in the plant materials were measured by High Purity Germanium (HPGe)-detectors (GMX-13200), and the measured concentration of the radionuclides was analysed and presented with the computer software Genie™ 2000 (© Canberra, Meriden, Connecticut, USA, (2009)).

The correction factor of each HPGe-detector (GMX-13200, GMX-33210 and GMX-20200) was determined through measurements with a dilution trial. In the dilution trial, 4 mL of the stock solution (representing a filling degree of 10%) was added to a 35 mL plastic jar, and the activity was measured. Then 3 mL of CsCl was added and the activity was remeasured: this step was repeated until the jar was 100% filled. The measured activity values for each step were divided by the activity measured at 100% filled to obtain a correction factor (CF) that was plotted against the percentage filled and adapted to the second order polynomial model in Equation 1:
where: $y$ is the filling degree, $a$ and $b$ stand for unknown parameters, $x$ is the scalar variable (in this case CF), and $c$ is the random error. From the curves, the CF was calculated for four different filling degrees: 10, 25, 50 and 75%. The corrected values of radioactivity concentration ($A_c$) were calculated for different filling degrees with Equation 2.

\[ A_c = A \times CF \]  

2.5.1 Calibration of the HPGe-detectors

The measured activity concentrations included uncertainties of the efficiency calibration of the HPGe-detector, which is one of the dominant components of the total measured uncertainty (Bronson et al., 2008). The HPGe-detectors were calibrated with a “calibration standard” containing a number of specific radioisotopes dissolved in water. The composition of the calibration standard used for this study is described in Table 3 and was made according to principles presented in Bronson and Young (1997) and ANSI (1978).

2.6 Calculations of interception fraction and statistical analyses

The interception of radionuclides by crops was expressed as the interception fraction, $f$, according to Equation 3, after Pröhl (2009). The interception fraction was the ratio between the activity in the d.w. above ground plant biomass directly after deposition ($A_i$, Bq m$^{-2}$) and the total amount of activity deposited ($A_t$, Bq m$^{-2}$):
Statistical analyses were through a balanced analysis of variance (ANOVA) with Minitab 16® (© Minitab Inc., Pennsylvania, USA, (2010)) and regression analysis with Microsoft Excel 2010 (© Microsoft Inc., Washington, USA, (2010)).

2.7 Uncertainties in measurement

Uncertainty was estimated according to the method described by the Guide to the Expression of Uncertainty in Measurement (GUM) (Ellison et al., 2000; ISO, 1993). The uncertainties are reported as the combined standard uncertainty $u_c(y)$ for measurement of above ground plant biomass and for the concentrations of radionuclides. The combined standard uncertainty was the combined standard uncertainty of the output estimate $y$, and was calculated according to Equation 4.

\[
u_c(y) = y \times \left( \sqrt{\sum_{i=1}^{\infty} \left( \frac{u(x_N)}{x_N} \right)^2} \right)
\]

where: $y$ is the output estimate and $x_N$ is the input estimates.

The uncertainties considered in this study were the purity of the radionuclides, difficulty in obtaining plant samples from a well-defined area (estimated), variation in the d.w. of samples, error in measuring the exact activity concentration in the deposition liquid, the uncertainty of the volume prepared for the deposition event, and, error in the liquid volume deposited by the rainfall simulator. The absorption of radionuclides on the surfaces of the rainfall simulator was not considered among the uncertainties. The radionuclide concentration in the rainwater...
was measured before and after passing through the rainfall simulator: there was no reduction in the concentration of radionuclides after passing through the rainfall simulator.

For LAI values, the standard deviation $S$ was reported. For $f$, the expanded uncertainty $U$ was reported as a 95% confidence interval and was equal to a coverage factor $k$ times the combined standard uncertainty $u_c(y)$ of $y$: $U = k \times u_c(y)$ (Ellison et al., 2000; ISO, 1993).

3. Results

3.1 The development of above ground plant biomass and LAI

Above ground plant biomass of spring oilseed rape reached a maximum at the fruit stage (code 80) (1.37±0.23 d.w. kg m$^{-2}$), and then declined until senescence (code 89). The maximum above ground plant biomass of spring wheat was measured at the start of ripening (code 89) (1.76±0.29 d.w. kg m$^{-2}$), which then declined until the end of ripening (code 92). The maximum LAI was measured at flowering (growth stages (code 61) for spring oilseed rape and (code 65) for spring wheat), and declined until harvest in both crops. Spring oilseed rape had a maximum LAI of 3.7±0.23 m$^2$ m$^{-2}$ and spring wheat had a maximum LAI of 4.69±0.20 m$^2$ m$^{-2}$. There was a correlation between the above ground plant biomass and LAI for both crops ($r^2 = 0.43$, $p < 0.05$ for spring oilseed rape and $r^2 = 0.58$, $p < 0.05$ for spring wheat) (Figure 3).

3.2 Interception fractions of $^{134}$Cs and $^{85}$Sr

For spring oilseed rape the maximum $f$ were 0.32±0.22 for $^{134}$Cs and 0.41±0.29 for $^{85}$Sr, and for spring wheat, the maximum $f$ were 0.36±0.14 for $^{134}$Cs and 0.48±0.18 for $^{85}$Sr. The maxima $f$ for both $^{134}$Cs and $^{85}$Sr were measured at stem elongation stage (code 32) for spring oilseed rape and at ripening stage (code 70) for spring wheat. There was a significant, but
weak correlation between the $f$ and LAI for $^{85}\text{Sr}$ ($r^2 = 0.48$, $p < 0.05$) in spring oilseed rape and for both $^{134}\text{Cs}$ ($r^2 = 0.55$, $p < 0.05$) and $^{85}\text{Sr}$ ($r^2 = 0.41$, $p < 0.05$) in spring wheat (Figure 4). There was no correlation between the $f$ and above ground plant biomass for $^{134}\text{Cs}$ in spring oilseed rape ($r^2 = 0.01$, $p > 0.05$), but there was a weak correlation for spring wheat ($r^2 = 0.36$, $p < 0.05$); a similar result was found for $^{85}\text{Sr}$ ($r^2 = 0.01$, $p > 0.05$ for spring oilseed rape and $r^2 = 0.32$, $p < 0.05$ for spring wheat) (Figure 5). In spring oilseed rape, $f$ for both radionuclides reached a maximum around growth stage 32, and tended to be more or less constant thereafter (Figure 6). In spring wheat, $f$ continuously increased up to growth stage 70 and then decreased in the later stages. According to the ANOVA test, the $f$ for $^{85}\text{Sr}$ was higher than for $^{134}\text{Cs}$ for spring oilseed rape ($p = 0.06$), but there was no difference between the two radionuclides for spring wheat ($p = 0.58$).

4. Discussion

Despite large uncertainties, significant statistical relationships were identified. For the interception fraction, the uncertainty in the later growth stages appeared related to the sampling area for both crops. This was due to difficulties in placing the sampling frame at later growth stages.

4.1 Above ground plant biomass and LAI

The highest values for the biomass for spring wheat were in the growth stage of ripening. Although this agreed with the results of Vandecasteele et al. (2001) and Eriksson et al. (1998), the real maximum could have been between growth stages (code 70) and (code 89).

For the LAI, the highest values (4.7±0.2 m$^2$ m$^{-2}$) were in the growth stage of flowering (growth stage (code 65) in this study) for spring wheat. The highest LAI values found by Vandecasteele et al. (2001) were 7.5 m$^2$ m$^{-2}$ in wheat at a slightly earlier growth stage; stem
extension 4 nodes detectable (growth stage (code 37) in this study). These differences could be explained by the use of different techniques for measuring LAI (e.g. type of device) and different weather conditions during the growth period. Although LAI can be both overestimated and/or underestimated if there are gaps between the plants, LAI is a suitable measure of crop development (Lang et al., 1985). There was a relation between LAI and the above ground plant biomass for both crops (Figure 3); however, at increasing values of above ground plant biomass, the values for LAI declined. This is because above ground plant biomass continues to increase until seeds and grains are fully developed, whereas, LAI decreases at later growth stages due to decline and drop of leaves during the ripening process.

4.2 Interception fraction of $^{134}$Cs and $^{85}$Sr

The highest values for $f$ were at the growth stage ripening for spring wheat: this was in agreement with Vandecasteele et al. (2001) (dry deposition) and Eriksson et al. (1998) (wet deposition).

The $f$ was related to LAI for both crops but only related to above ground plant biomass for spring wheat. A similar relationship between $f$ and above ground plant biomass is presented by Vandecasteele et al. (2001) for spring wheat, although the values are higher (0.84 for $^{137}$Cs and 0.88 for $^{90}$Sr) than determined in this study (0.36 for $^{134}$Cs and 0.48 for $^{85}$Sr): Vandecasteele et al. (2001) measured a few hours after deposition. The interception fraction tended to follow the growth of LAI; in spring wheat, higher values of LAI had higher $f$ values (Figure 4). At several growth stages for spring oilseed rape, but not for spring wheat, $f$ values were higher for $^{85}$Sr than for $^{134}$Cs. One explanation for the weaker relation between above ground plant biomass and interception fraction for spring oilseed rape was that the plants drop leaves, thereby interception capacity, at later growth stages, whereas, total biomass still increases. Higher values for radiostrontium than for radiocaesium are observed
by Madoz-Escande et al. (2004) (dry deposition on bean) and Vandecasteele et al. (2001) (wet deposition on cereals). Carini et al. (2003) (wet deposition on strawberry) found interception is higher for $^{85}$Sr (0.47) than for $^{134}$Cs (0.37) on strawberry, and is probably explained by a difference in valence between $^{85}$Sr (divalent) and $^{134}$Cs (monovalent) ions. Divalent ions are assumed more strongly absorbed by plants than monovalent ions are (Bréchignac et al., 2000; Vandecasteele et al., 2001).

Vandecasteele et al. (2001) found a correlation between the $f$ for $^{137}$Cs and $^{90}$Sr and LAI ($r = 0.98$) in spring wheat. The differences between the results from this study and those of Vandecasteele et al. (2001) and Eriksson et al. (1998) might be due to differences in methodology: the results of Vandecasteele et al. (2001) and Eriksson et al. (1998) are from wet deposition. On a pasture crop sampled two hours after the deposition event (Chadwick and Chamberlain, 1970), the interception of $^{85}$Sr is in the range 0.20 to 0.82, and the highest $f$ for $^{134}$Cs (0.71) and $^{85}$Sr (1.11) is 24 hours after deposition (Eriksson et al., 1998), with $f$ increasing with plant growth (Eriksson et al., 1998; Madoz-Escande et al., 2004). However, although a dip in the $f$ was found at growth stage (61) for spring oilseed rape, this difference could be explained by errors in the deposition or sampling of the crops. A reduction in $f$ was found at growth stage (89) for both crops, but could be explained by the reduction in the area of above ground plant biomass that could intercept deposited radionuclides (Figure 5).

The results of this study could have been influenced by the high intensity rain application during a 30-second burst. A lower intensity of rain application results in higher values of interception, as the droplets tend to splash off the plants to a lesser degree (Keim et al., 2006; Wang et al., 2005). An alternative measure for $f$ is the mass interception fraction, which is $f$ normalised for its biomass (Hoffman et al., 1992; Hoffman et al., 1995; Pröhl, 2009), however, as a better relationship was found with LAI than with biomass, $f$ was more suitable for this study.
5. Conclusions

LAI can be used to quantify interception of both $^{134}\text{Cs}$ and $^{85}\text{Sr}$ in spring oilseed rape and spring wheat, whereas, above ground plant biomass can only be used to quantify interception on spring wheat. The levels of interception are highest at the ripening stage, whereas, in later growth stages (senescence) there is a decline in the level of interception. However, the results in this study could have been influenced by the amount and intensity of the rain that was applied.

The urgency of further research is emphasised by the Fukushima Dai-ichi nuclear power plant accident in 2011, the limited number of field studies, and the abundance and age of nuclear power plants. We suggest field experiments with more food and fodder crops and a wider range of radionuclides than studied here, including iodine, are warranted for developing suitable countermeasures for reducing human exposure to radioactivity.

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