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Accumulation of wet-deposited radiocaesium and radiostrontium by spring oilseed rape (*Brássica napus* L.) and spring wheat (*Tríticum aestívum* L.)

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ABSTRACT

The accumulation of ^{134}Cs and ^{85}Sr within different parts of spring oilseed rape and spring wheat plants was investigated, with a particular focus on transfer to seeds after artificial wet deposition at different growth stages during a two-year field trial. In general, the accumulation of radionuclides in plant parts increased when deposition was closer to harvest. The seed of spring oilseed rape had lower concentrations of ^{85}Sr than spring wheat grain. The plants accumulated more ^{134}Cs than ^{85}Sr. We conclude that radionuclides can be transferred into human food chain at all growing stages, especially at the later stages. The variation in transfer factors during the investigation, and in comparison to previous results, implies the estimation of the risk for possible transfer of radionuclides to seeds in the event of future fallout during a growing season is still subject to considerable uncertainty.

1. Introduction

Radionuclides intercepted on plants can be taken up and redistributed to edible plant parts, for example seeds. The rate of uptake and redistribution of radionuclides depends on the growth stage of the crop, weather conditions and the type of radionuclide (IAEA, 2010; Kinnersley et al., 1997; Pröhl, 2009), and radionuclide uptake is through either foliage (foliar uptake) or roots (root uptake). Foliar uptake is assumed the dominant pathway when deposition occurs during the growing season (Pröhl, 2009); as a well-developed crop with its large leaf area intercepts a majority of the deposited radionuclides (Bengtsson et al., 2012; Vandecasteele et al., 2001). The cuticle layer of the leaf epidermis is assumed to be impermeable; however, it contains cracks and defects where radionuclides can enter (Handley and Babcock, 1972; Hossain and Ryu, 2009; Tukey et al., 1961). The rate of radionuclide entrance through the cuticle layer depends on different physical and chemical factors, such as temperature, light, pH, the carrier of the radionuclide in the solution, the valence of the radionuclides, and the type of crop (Tukey et al., 1961). Radionuclides can also enter the plant system through the stomata, but this pathway is assumed to contribute a small fraction of the total amount of radionuclides entering the leaf (Eichert and Burkhardt, 2001; Eichert et al., 2002; Tukey et al., 1961). Similarly, the time elapsed since interception of the radioactive deposition and the time of the crops harvest affected the redistribution of the radionuclides within the crop (Coughtrey and Thorne, 1983; Kirchmann et al., 1967; Tukey et al., 1961). When radionuclides enter through the cuticle layer, they are actively transported inside the plant cells through both the symplastic pathway and an exchange mechanism between the phloem and the xylem i.e. vascular bundle system (Thiessen et al., 1999). The redistribution of radionuclides is regulated mainly by their physiological behaviour within the crop and the time during the growing season when the deposition occurred (Thiessen et al., 1999).

The monovalent radiocaesium ion is redistributed within crops to a higher degree than the divalent radiostrontium ion (Aarkrog, 1969, 1975, 1983; Eriksson et al., 1998; Müller and Pröhl, 1993). The redistribution of radiocaesium through the vascular bundle system takes place at different rates depending on the plant part targeted: 5–30% of the intercepted radiocaesium is taken up through the epidermis, and of this, about one-third is redistributed to the seeds (Coughtrey and Thorne, 1983). However, if the entrance of radiocaesium into crops occurs at an early growth stage or just

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before harvest, the degree of redistribution to the seeds is lower than if entrance of radiocaesium occurs at the time of seed development (Coughtrey and Thorne, 1983). The decrease in radiocaesium concentration in plants with time can be explained through plant growth rather than being due to activity loss (Coughtrey and Thorne, 1983).

Radiostrontium is retained by the cuticle layer of the epidermis to a higher degree than radiocaesium (Vandecasteele et al., 2001), therefore, there is less redistribution to other plant parts in the vascular bundle system (Aarkrog, 1969, 1975, 1983; Bréchignac et al., 2000; Müller and Pröhl, 1993). A maximum of 25% of the total radiostrontium taken up by cereals is redistributed to other plant parts within the vascular bundle system of which 5–10% of the radiostrontium being redistributed to the grain, and up to 50% to the roots (Coughtrey and Thorne, 1983).

The transfer of radionuclides from the environment to edible plant parts in a given situation can be described by a transfer factor (TF, $m^2\ kg^{-1}$). This is defined as a ratio between the activity concentration of radionuclides in plant parts (Bq kg^{-1}), for example seeds, and the amount of radionuclides deposited per unit area (Bq m^{-2}) (Ehlken and Kirchner, 2002; Howard et al., 1996; Rosén et al., 1996). TF values calculated in connection to a previous deposition, or experimentally determined scenario, can be used to estimate the transfer to edible plant parts after a new radioactive deposition event to facilitate the making of decisions on countermeasures for reducing the distribution of radionuclides to foodstuffs (Howard et al., 1996; Kostiainen et al., 2002; Rosén, 1996). Transfer factors have only been determined for a limited number of possible scenarios and have strong seasonal variation (IAEA, 2010). If a TF value relevant for a specific situation, i.e. determined under similar circumstances, is unavailable, the assessment of the situation after deposition of radioactivity can be challenging and wrong decisions, either too cautious or too careless, about suitable measures for preventing the transfer of radionuclides to foodstuffs may be made (Salbu, 2000). The transfer of the intercepted radionuclides to edible plant parts can be described by a translocation factor (TLF, $m^2\ kg^{-1}$). This is defined as the ratio between the activity concentration of radionuclides in plant parts (Bq kg^{-1}), for example seeds, and the amount of intercepted radionuclides by the plant canopy per unit area (Bq m^{-2}) (Thiessen et al., 1999; Vandecasteele et al., 2001).

The aims of this study were to investigate ^{134}Cs and ^{85}Sr accumulation in spring oilseed rape (*Brássica napus* L.) and spring wheat (*Tríticum aestívum* L.) after wet deposition at different growth stages; to calculate the distribution of ^{134}Cs and ^{85}Sr between plant parts; and, to describe transfer of ^{134}Cs and ^{85}Sr to seeds through TF and TLF values. The hypothesis was ^{134}Cs and ^{85}Sr accumulation in seeds depended on the growth stage of the plant, the type of radionuclide, and the plant type.

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 texture of the soil at the site was clay (60% clay, 20% silt and 20% sand). Soil texture, pH (6.5), plant available phosphorus (57 mg kg^{-1}), potassium (202 mg kg^{-1}) and calcium (3692 mg kg^{-1}) were determined in 2010 and described by Bengtsson et al. (2012).

The meteorological station, described by Karlsson and Fagerberg (1995), monitors daily air temperature, precipitation, and wind speed. 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 from 1st May to 30th of September 2010 had a mean air temperature of 15 °C and precipitation sum of 293 mm (Department of Crop Production Ecology, 2013). During the second growing season in 2011, mean air temperature was 15 °C and precipitation sum 287 mm (Department of Crop Production Ecology, 2013). The temperature at the deposition and sampling occasions varied between 10 and 21 °C and there was no precipitation in connection with deposition and sampling on any occasion during the years of study (2010 and 2011), except for the last deposition and sampling occasion for spring wheat in 2011 (0.1 mm) (Department of Crop Production Ecology, 2013). Wind speed at deposition and sampling occasions was low; it varied between 1.3 and 3.6 m s^{-1} in 2010, and, 1.3 and 2.7 m s^{-1} in 2011 (Department of Crop Production Ecology, 2013).

2.2. Design of the trial

A trial with a randomised block design, with 1 × 1 m^2 parcels in three replicates (in total 60 parcels), was laid out in 2010. In order to cover seasonal variations, a new trial with the same design was laid on a nearby site in 2011. The experimental crops, spring oilseed rape (*Brássica napus* L.) variety 'Larissa' and spring wheat (*Tríticum aestívum* L.) variety 'Triso', were sown 12 May 2010 and 27 April 2011 as described in Bengtsson et al. (2012) and managed according to common agricultural practises for the region. The radionuclides 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 radionuclides behaved in the same manner as ^{137}Cs and ^{90}Sr. The selected radionuclides were applied through artificial rain.

The radionuclides were deposited on plants of spring oilseed rape at five different growth stages, which were according to the Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH)-scale, (Hack et al., 1992). In 2010 these stages were leaf development, code 13 (three leafs unfold); stem elongation, code 32 (two visible extended internodes); 10% of flowers on main raceme open, code 61; full flowering, code 65; and, beginning of ripening, code 80. In 2011, the growth stages were leaf development, codes 15–19 (five to nine leafs unfold); full flowering, code 65; end of flowering, code 69; development of fruit, code 76 (60% of pods have reached final size); and, ripening, code 82 (20% of pods ripe, seeds dark and hard) (Fig. 1a).

For the spring wheat in 2010, the 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); development of fruit, code 70 (medium milk); and, ripening, fully ripe, code 89. In 2011, the growth stages were stem extension, code 37 (flag leaf visible); flowering, code 65 (on-going flowering); ripening, code 85 (dough ripeness); ripening, fully ripe, code 89; and, senescence, over-ripe, code 92 (Fig. 1b).

2.3. Preparation and deposition of artificial radioactive rain

The artificial rainwater solution was prepared from stock solutions as described by Bengtsson et al. (2012). In 2010 the stock solutions contained 5 MBq L^{-1} ^{134}Cs and 15 MBq L^{-1} ^{85}Sr and in 2011, they contained 40 MBq L^{-1} ^{134}Cs and 37 MBq L^{-1} ^{85}Sr. ^{134}Cs was in the form of caesium chloride (CsCl) in 0.1 M HCl solution (expanded uncertainty of ±10%) (Institute of Atomic Energy POLATOM, Otwock-Świerk, Poland). ^{85}Sr was in the form of strontium chloride ($SrCl_2$) in 0.1 M HCl solution (expanded uncertainty of ±2.5%) (Areva Cerca Lea, Pierrelatte Cedex, France). In 2010 the amount of ^{134}Cs applied at the 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}. In 2011, the amount of ^{134}Cs ranged from 40.2 to 41.0 kBq m^{-2} and ^{85}Sr ranged from 39.4 to 41.0 kBq m^{-2}.

The radionuclides were applied with a rainfall simulator, as described in Bengtsson et al. (2012). The simulator was a modified version of the drip infiltrometer developed by Joel and Messing (2001). The amount of precipitation applied at each treatment was 1.00 ± 0.01 mm at an intensity of 1 mm 30 s^{-1} and the equipment used was a 520 series process pump manufactured by Watson–Marlow. A windshield was used to prevent wind disturbance during deposition in the early growth stages.

2.4. Sampling and analyses

In both years plants were cut 5 cm above ground within a frame (25 × 25 cm^2 square) placed in the middle of each parcel two-three hours after deposition in three replicates, and in another three at harvest time (when plants were fully ripe): the whole plants were sampled. Spring oilseed rape was separated into four plant compartments; stem (stem and attached dead leaves), siliques (except seeds), and remaining seed materials. Likewise spring wheat was separated into four plant compartments; stem (stem and attached dead leaves), ears (husks) except seeds, and the seeds.

The plant materials were weighed fresh, and then air-dried (at a maximum of 40 °C for a minimum of 14 days) before being re-weighed for dry weight (d.w.). Thereafter, the plant material was milled and placed in 35 mL or 60 mL plastic jars with a suitable geometry for measuring activity concentration. The activity concentrations of the radionuclides were expressed as Bq kg^{-1} d.w. and corrected for the decay between sampling and analysis. The results obtained for samples from early growth stages measured in 35 mL jars were corrected for the degree of filling due to small amount of plant material; the correction factor for each detector was determined according to Bengtsson et al. (2012).

The actual concentration of radionuclides in the artificial rainwater and in the plant materials were measured by High Purity Germanium (HPGe)-detectors (GMX-13200, GMX-33210, GMX-20200), and the measured activity concentrations were