Interception and Storage of Wet Deposited Radionuclides in Crops

Field Experiments and Modelling

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Cover: A flowering oilseed rape field with the nuclear power plant Barsebäck in the background.
(photo: B. Bengtsson)

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Interception and Storage of Wet Deposited Radionuclides in Crops: Field Experiments and Modelling

Abstract

The emission of radionuclides into the atmosphere from various sources, such as nuclear power plant accidents and nuclear bomb explosions, can result in the interception and uptake of radionuclides by crops in the agricultural ecosystem. These radionuclides *e.g.* radiocaesium (^{134,} ^{137}Cs) and radiostrontium (^{85,} ^{90}Sr), can be transferred to foodstuffs via seeds or animal feed.

Therefore, in this thesis, the goal was to study the amount of ^{134}Cs and ^{85}Sr that have been intercepted, taken-up and redistributed to different plant parts during wet deposition at different growth stages of spring oilseed rape, spring wheat and ley. For spring oilseed rape and spring wheat, the focus was on the transfer to the seeds after wet deposition of ^{134}Cs and ^{85}Sr. The dependence between the interception of radionuclides and the growth stage, *e.g.* the total standing plant biomass and the leaf area index (LAI) were also studied.

There was a positive correlation between the interception of ^{134}Cs and ^{85}Sr and LAI for all three crops. A positive correlation between the standing plant interception and the biomass of ^{134}Cs and ^{85}Sr was found for spring wheat and ley, but not for spring oilseed rape. The highest interceptions of ^{134}Cs and ^{85}Sr were at shooting for spring oilseed rape, and at maturity for spring wheat. For ley, the highest interception was at the well-developed stages.

Accumulation of ^{134}Cs and ^{85}Sr in the different plant parts increased when deposition was close to harvest and the crops accumulated more ^{134}Cs than ^{85}Sr. The concentration of ^{85}Sr was lower in spring oilseed rape than in wheat grains. There was an indication that the distribution of radionuclides between the above ground plant parts was independent of the way that they entered into the plant after deposition of ^{134}Cs and ^{85}Sr.

The variation in transfer factors found in this thesis in comparison with results from other studies suggest, that the estimate of the risk of possible uptake to crops in the event of future deposition during the growing season, is still subject to uncertainties.

Keywords: concentration, radionuclide, transfer factor, translocation factor, deposition, food crops, biomass, leaf area index, growth stage

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Dedication

To all the people that have been affected by the two, so far, most severe nuclear power plant disasters, in Chernobyl, Ukraine 1986 and in Fukushima Dai-ichi, Japan 2011.

Nothing in life is to be feared, it is only to be understood. Now is the time to understand more, so that we may fear less.

Marie Curie

Contents

List of Publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I Bengtsson, S.B., Eriksson, J., Gärdenäs, A.I., Rosén, K. (2012). Influence of development stage of spring oilseed rape and spring wheat on interception of wet-deposited radiocaesium and radiostrontium. *Atmospheric Environment* 60, 227-233.

II Bengtsson, S.B., Eriksson, J., Gärdenäs, A.I., Vinichuk, M., Rosén, K. (2013). Accumulation of wet-deposited radiocaesium and radiostrontium in spring oilseed rape (*Brássica napus* L.) and spring wheat (*Tríticum aestívum* L.). *Environmental Pollution* 182, 335-342.

III Bengtsson, S.B., Eriksson, J., Gärdenäs, A.I., Vinichuk, M., Rosén, K. Interception and absorption of wet-deposited radiocaesium and radiostrontium by a ley. (manuscript).

IV Gärdenäs, A.I., Berglund, L.S., Bengtsson, S.B., Rosén, K. The uptake and storage of caesium and strontium by spring wheat - a modelling study based on a field experiment. (manuscript).

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The contribution of Stefan B. Bengtsson to the papers included in this thesis was as follows:

I Planned the experimental work together with the co-authors. Performed the practical fieldwork, with some assistance. Performed data analyses and wrote the main part with some assistance from the co-authors.

II Planned the experimental work together with the co-authors. Performed the practical fieldwork, with some assistance. Performed data analyses and wrote the main part with some assistance from the co-authors.

III Planned the experimental work together with the co-authors. Performed the practical fieldwork, with some assistance. Performed data analyses and wrote the main part with some assistance from the co-authors.

IV Contributed with the input parameters to the model and wrote it with assistance from the co-authors.

Abbreviations

Cs	caesium
f	interception fraction
LAI	leaf area index
Sr	strontium
TE_{Seed}	simulated storage in grains
$TE_{IntSeed}$	simulated intercepted storage on grains
TF	transfer factor
TLF	translocation factor

1 Introduction

The release of radionuclides into the atmosphere from nuclear power plant accidents or test firing of nuclear weapons can result in both dry and wet deposition of radionuclides onto food crops (Kinnersley *et al.*, 1997; Hoffman *et al.*, 1995). The main harmful long-living radionuclides released from nuclear power plant accidents and test firing of nuclear weapons are radiocaesium ($^{134, 137}Cs$), isotopes of plutonium ($^{238, 239, 240, 241}Pu$) and radiostrontium ($^{89, 90}Sr$). The intake of radiocaesium via contaminated foodstuffs spreads evenly in human bodies, somewhat more in muscles than in bones (Burovina *et al.*, 1965; Yamagata, 1962), and is a possible cause of different types of cancers (Nikula *et al.*, 1995). Radiostrontium intake via contaminated foodstuffs mostly accumulates in the human skeleton and presents an additional risk of cancer during pregnancy and in young people (Valentin, 2004). Due to the harmful effects of radionuclides to humans, it is of significance to be able to reduce the transfer of radionuclides to human food.

The level of radionuclide interception by different plant parts is dependent on plant morphology, for example the leaf area index (LAI), the angle of the leaf, the standing plant biomass and the maximum external 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 forms of the radionuclides, such as the molecular mass and valence of ions (Salbu *et al.*, 2004): a divalent ion of radiostrontium can fix itself more easily to the surface of leaves than a monovalent ion of radiocaesium (Vandecasteele *et al.*, 2001; Müller & Pröhl, 1993). The size of the radioactive particles and weather conditions, such as precipitation and wind speed, also affect the level of radionuclide interception (Kinnersley *et al.*, 1997; Aarkrog, 1975). According to Hoffman *et al.* (1992), the interception of radionuclides is more dependent on the standing plant biomass than on the amount of precipitation. The time between the deposition and the harvest affects the concentration of radionuclides in the plant at harvest: this depends on "field losses", for example wash-off effects and volatilisation (Chadwick & Chamberlain, 1970).

Wet deposited and intercepted radionuclides, for example radiocaesium, can be taken up directly by vegetation through leaves (Scotti & Carini, 2000; Middleton, 1959; Middleton, 1958), and inside the plant tissues; the radionuclides can then be redistributed to edible plant parts, such as seeds. Wet deposition of radionuclides during the growing season increases the risk of crop contamination in the first year (Anspaugh *et al.*, 2002). The uptake of radioactive substances through the leaf is assumed to be higher than uptake through the roots (Johnson *et al.*, 1966; Russell, 1965), and the rate of uptake and redistribution of radionuclides will change depending on the growth stage of the crop and the type of radionuclide. In a well-developed crop, most of the deposited radionuclides will be intercepted directly by leaves (Bengtsson *et al.*, 2012; Vandecasteele *et al.*, 2001). Although, the impermeability of the cuticle layer of the leaf's epidermis makes it difficult for radionuclides to enter into the leaf; the cuticle layer contains cracks and defects where radionuclides can enter (Hossain & Ryu, 2009; Handley & Babcock, 1972; 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 of the solution, the radionuclide carrier in the solution, valence and the type of crop (Tukey *et al.*, 1961).

Information about the level of wet deposited radionuclide interception under different circumstances and its subsequent redistribution within plant parts used for food consumption is essential for the risk assessment of radionuclide transfer through the food chain, and for planning effective countermeasures to reduce human exposure to radionuclides. The interception, uptake and redistribution of deposited radionuclides vary, depending on the specific situation and seasonality.

2 Aim

The overall aim of this thesis was to determine the amount of radiocaesium (^{134}Cs) and radiostrontium (^{85}Sr) intercepted, taken up and redistributed to different plant parts during wet deposition at different growth stages of spring oilseed rape, spring wheat and ley. From the information obtained a model describing the expected contamination levels in spring wheat in relation to the time of fallout during the growing stages was developed. This knowledge is of importance after emissions of radionuclides from various sources, such as nuclear power plant accidents and nuclear bomb explosions, which can result in the interception and uptake of radionuclides by agricultural crops.

The specific aims were to:

- Measure the interception of wet deposited ^{134}Cs and ^{85}Sr by spring oilseed rape, spring wheat, and ley at different growth stages (Papers I and III).
- Clarify whether the interception of ^{134}Cs and ^{85}Sr was related to the standing plant biomass, the leaf area, the type of radionuclide and the type of crop (Papers I and III).
- Investigate the accumulation of wet deposited ^{134}Cs and ^{85}Sr in spring oilseed rape, spring wheat and ley at different growth stages (Papers II and III).
- Calculate the distribution of ^{134}Cs and ^{85}Sr between plant parts of spring oilseed rape and spring wheat (Paper II).
- Describe the transfer of wet deposited ^{134}Cs and ^{85}Sr to seeds and other plant parts through calculation of transfer factors (TF) (Papers II and III) and translocation factors (TLF) (Paper II).
- Calculate the transfer of ^{134}Cs and ^{85}Sr to beef and cow's milk from ley (Paper III).
- Extend a trace element cycling model by including wet deposited radionuclides with a description of interception and foliar uptake for spring wheat (Paper IV).

3 Background

The deposition of radioactive compounds from the Chernobyl accident in 1986 could have had important consequences for agricultural systems in Sweden. The deposition of ^{137}Cs in these regions ranged from 10 to >100 kBq m^{-2}. In the southern part of Sweden, where only dry deposition occurred, the range was 0.2 to 5 kBq m^{-2} of ^{137}Cs (Moberg, 2001). The total fallout of ^{137}Cs in Sweden was estimated to be 4.25 PBq, or about 5% of that released from Chernobyl, with a mean deposition in Sweden of 10 kBq m^{-2} (Moberg, 2001). However, the impact was limited, as the accident occurred relatively early (26th April) in the growing season (Moberg, 2001). If deposition had occurred later in the growing season, the consequences would have been more severe (Eriksson *et al.*, 1998a; Eriksson *et al.*, 1998b). Since the Chernobyl accident, the uptake of radioactive substances by crops in contaminated agricultural areas has been widely investigated (Fesenko *et al.*, 2007; Rosén *et al.*, 1998; Rosén, 1996; Aarkrog, 1988). In the same year, a radioactive accident occurred, direct deposition resulted in higher distribution of radioactivity in the crops than direct uptake from the soil (Madoz-Escande *et al.*, 2004).

The consumption of contaminated agricultural foodstuffs can expose humans to high collective doses of radiation (Hasegawa *et al.*, 2009; Madoz-Escande *et al.*, 2004). Therefore, risk assessment for predicting the content of radioactive substances in crops, especially the edible parts, is vital. If the concentrations of radionuclides in edible parts of crops are known, then appropriate countermeasures for preventing or reducing the transfer to humans can be taken. There is limited knowledge about the relationship between the level of radionuclide deposition onto growing crops and its activity concentration in harvested products. There is, however, considerable knowledge about the relationship between the concentrations in both soil and crops, and how these are controlled by different factors (IAEA, 2010). Data on the relationship between soil and crops is useful for predicting uptake into crops after deposition on bare soil, and in the years after an event has taken place; however, this data is not useful when radionuclides are deposited directly onto the crop and can be redistributed to harvested products.

The knowledge about how radiocaesium ($^{134, 137}Cs$) and radiostrontium ($^{85, 89, 90}Sr$) are captured and redistributed to harvested plant parts after being deposited onto a growing crop, and the size and time of deposition are important. Gathering data on the amount and time of deposition is essential for estimating the level of activity concentration in crops, and for suggesting appropriate countermeasures in different scenarios to prevent further spread of radionuclides to foodstuffs. With access to this data, it would be possible to estimate the levels of radionuclides in a crop occurring in different scenarios.

The degree of capture depends on the growing stage of the crop, the leaf area and the density of the canopy. The time that elapses between deposition and harvest affects the total concentration of radioactive substances in the crop, as plant growth has a dilution effect and a small fraction of the captured radioactive substance is lost through leaf drop, rain flushing or wind removal (Rosén & Eriksson, 2008; Eriksson *et al.*, 1998a; Eriksson *et al.*, 1998b). The growth and storage mechanisms of the crop and the variation of concentration in the crop may depend on the weather conditions; and the effects of countermeasures can be dynamically simulated through models such as ECOSYS-87 (Müller & Pröhl, 1993) and the Tracey model (Gärdenäs *et al.*, 2009).

3.1 Entrance of radionuclides into plants

There are two main types of deposition: *dry deposition* and *wet deposition*. In dry deposition, particles are deposited directly onto surfaces, and the transfer of radioactive particles to plants is through absorption, impaction and sedimentation in water (IAEA, 2010; Smith, 2001). In wet deposition, the radioactive particles are smaller and are deposited when it hails, rains or snows (IAEA, 2010).

Radioactive particles deposited by rain can result in two different processes: *wash-out*; the entrainment (a turbulent flow captures a non-turbulent flow) of radioactive particles by falling rain drops, and *rain-out*; the condensation of air vapour into rain drops forming around radioactive particles. With wet deposition, radioactive particles are transported from the atmosphere at a higher rate than during dry deposition. Plant uptake of radionuclides from both dry- and wet deposition are controlled by different factors, *e.g.* the physical and chemical properties of the radioactive particles, the growth stage of the crop at the time of contamination, the total amount of precipitation (Kinnersley *et al.*, 1997), the intensity of precipitation (IAEA, 2010) and the ability of the canopy to hold water.

The uptake of radionuclides differs depending on the growth stage of the crop. At a well-developed stage, the majority of deposited radionuclides are taken up directly by the leaves (Andersson *et al.*, 2002). The proportion of radionuclides in precipitation that can be held by a crop quickly reaches its contamination maximum when the external water storage capacity of the plant canopy reaches its maximum. However, if contamination continues to increase after the maximum external water storage capacity has been reached, then radioactive particles will be absorbed through the surface of the leaf. The degree of continuous uptake is related to the chemical form of the radionuclide deposited (Kinnersley *et al.*, 1997).

3.1.1 Interception of radionuclides by plants

The interception of radioactive particles is a complicated process related to molecular mass and valence (Pröhl, 2009; Salbu *et al.*, 2004; Oughton & Salbu, 1994). The negatively charged surfaces of plants act as a cation exchanger or exchange resin, causing a weaker retention of anions and monovalent ions on the plant surface than for divalent and polyvalent cations (Pröhl, 2009; Vandecasteele *et al.*, 2001; Kinnersley *et al.*, 1997).

Interception of radionuclides is related to plant morphology, such as leaf area, the angle of the leaves, the standing plant biomass, and the maximum external water storage capacity of the plant canopy (IAEA, 2010; Kinnersley *et al.*, 1997). The size of the radioactive particles and fragments and the weather conditions, such as precipitation and wind speed, can have an impact on radionuclide interception (Kinnersley *et al.*, 1997; Aarkrog, 1975). The interception of radionuclides is more dependent on the standing plant biomass than on the amount of precipitation (Hoffman *et al.*, 1992). The time between the deposition event and the harvest affects the concentration of radionuclides in the plants at harvest, and depends on "field losses"; for example wash-off and volatilisation (Chadwick & Chamberlain, 1970).

There are different ways of describing interception levels of deposited radionuclides. The interception can be described by an interception fraction (f, $\text{Bq m}^{-2}/\text{Bq m}^{-2}$), which is defined as the ratio between the activity concentration retained by the vegetation immediately after the deposition event, and the total activity deposited. Another way of describing interception is by a mass interception fraction (f_B, $m^2\ kg^{-1}$), which is dependent on the development stage of the plant, and is defined by normalising the interception fraction to the standing plant biomass (B, $kg\ m^{-2}$, dry mass) (Pröhl, 2009).

The level of interception increases at the same rate as the plant canopy expands. Canopy growth can be described by plant biomass per unit area and the leaf area per soil unit, leaf area index (LAI). In the early stages of plant growth, there is a strong correlation between biomass and leaf area, but this correlation decreases at the end of growth.

Interception of precipitation increases linearly with LAI (Pröhl, 2009) and is connected to the water storage capacity of the plant canopy, which in turn, is dependent on the LAI. However, interception decreases as the amount of precipitation increases (Figure 1), which means that the interception fraction is inversely proportional to the amount of precipitation (Pröhl, 2009).

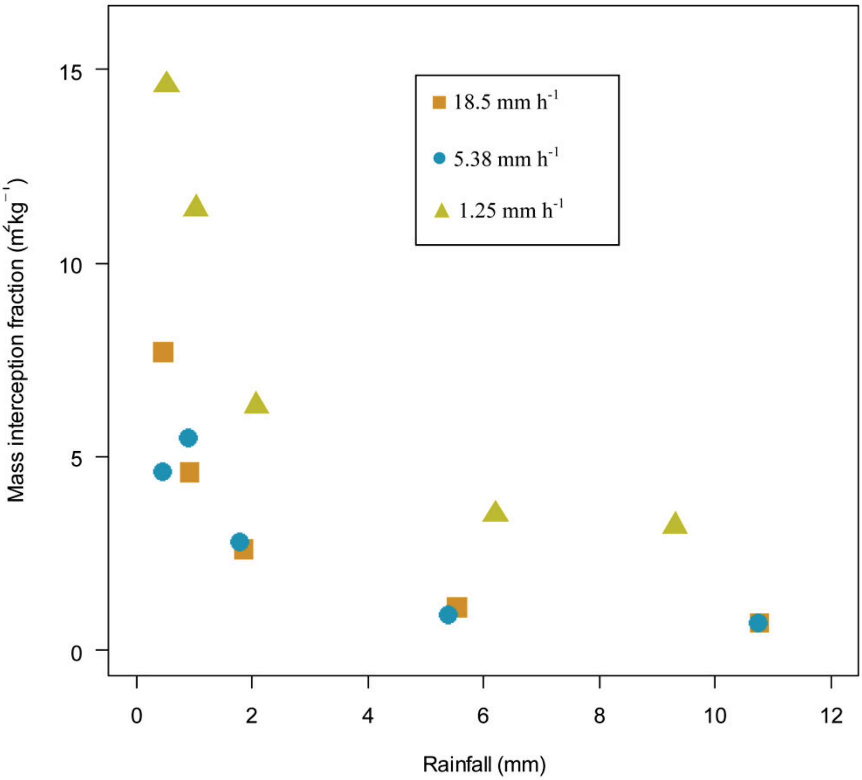

Figure 1. Interception of ^{137}Cs by grass as a function of the amount of precipitation and precipitation intensity, modelled after Kinnersley *et al.* (1997).

3.1.2 Uptake of radionuclides by plants

Radionuclides cannot easily enter through the cuticle layers of the leaf's epidermis; however, the cuticle contains cracks and defects where entrance can take place (Figure 2) (Hossain & Ryu, 2009; Handley & Babcock, 1972; Tukey *et al.*, 1961). Radionuclides can also enter the plant system through the stomata, but this pathway accounts for a smaller fraction of the total amount of radionuclides being absorbed (Eichert *et al.*, 2002; Eichert & Burkhardt, 2001; Tukey *et al.*, 1961).

In the cuticle layer, specialised epidermal cells (surface veins consisting of thin-walled parenchyma tissue) are easily penetrated by radionuclides. The rate of entrance through the cuticle layer depends on physical and chemical factors such as temperature, light, pH, the carrier of the radionuclides in the solution, the valence of the radionuclides and the plant species. The time between interception of the radionuclides by the standing plant and the crop harvest affects the distribution of the radionuclide within the plant (Coughtrey & Thorne, 1983; Kirchmann *et al.*, 1967; Tukey *et al.*, 1961). Radionuclides such as ^{45}Ca, ^{42}K and ^{32}P are lost through the leaves of squash, beans and corn, whereas in strawberries, up to 75% of radiostrontium uptake is lost through the berries (Tukey *et al.*, 1961).

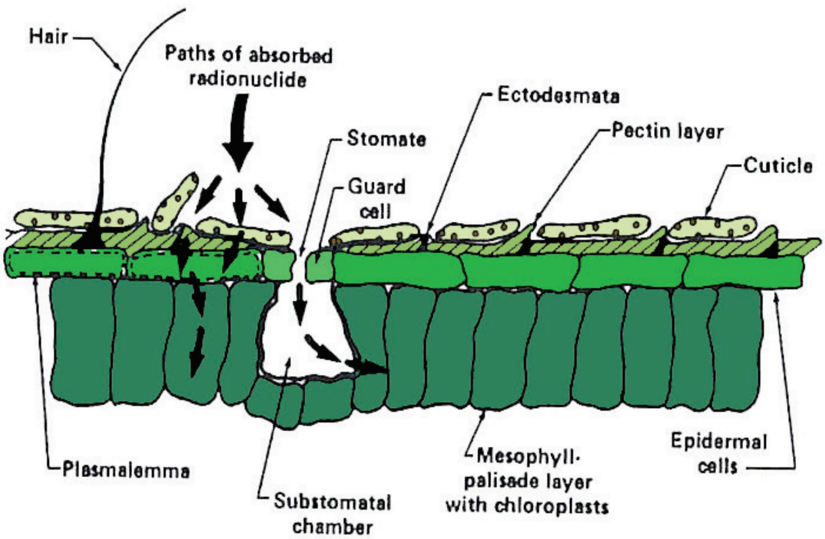

Figure 2. Cross sectional diagram of the leaf's surface showing where the entrance of radionuclides can take place. Illustration with permission from Koranda & Robison (1978).

Radionuclides that enter through the cuticle layer are actively transported inside the plant cells through the symplastic pathways (the inner side of the plasma membrane in which water can freely diffuse), and by an exchange mechanism between the phloem and the xylem (vascular bundle system). The redistribution of radionuclides is regulated by the physiological stage of the plants (growth stage) and the time when deposition took place during the growth season (Thiessen *et al.*, 1999).

Plants do not appear to distinguish between the transfer of the divalent cations calcium (Ca^{2+}) and strontium (Sr^{2+}), to different plant parts (White, 2001): the transportation of Ca^{2+} and Sr^{2+} is linear to the concentration of these two cations in a nutrient medium (Young & Rasmusso, 1966; Bowen & Dymond, 1956; Menzel & Heald, 1955; Collander, 1941). These divalent cations cannot be transferred through the different plasma membranes when they enter the xylem system of plants (White, 2001), but similarly to calcium, radiostrontium can be taken up via the roots (Veresoglou *et al.*, 1996). Radiocaesium has less availability to plants, as it is readily absorbed and fixed into clay interlayers (Absalom *et al.*, 2001; White, 2001; Zhu & Smolders, 2000; Absalom *et al.*, 1995). The transfer of caesium to different plant parts follows the pathways of potassium transport. Minor differences between the concentrations of caesium and potassium in different plant parts are found throughout vegetative growth (Menzel & Heald, 1955), which implies that plant parts with a lower potassium concentration, such as ears, will also have a low concentration of caesium (Bilo *et al.*, 1993). However, there may be differences in the transport of potassium and caesium (Gommers *et al.*, 2000), these differences are assumed to be related to the caesium:potassium discrimination in the membrane transport system (Buysse *et al.*, 1995). Moreover, the effective potassium transporter, which can transport caesium efficiently, has only been found in root cells and not in above ground parts (Zhu & Smolders, 2000).

3.1.3 Distribution of radionuclides in plants

The cuticle layer of the epidermis retains more radiostrontium, which is divalent, than radiocaesium (Vandecasteele *et al.*, 2001); therefore, radiostrontium has a lower redistribution to other plant parts in the vascular bundle system than radiocaesium (Bréchignac *et al.*, 2000; Müller & Pröhl, 1993; Aarkrog, 1983; Aarkrog, 1975; Aarkrog, 1969). At maximum, 25% of the total radiostrontium uptake by cereals is redistributed to other plant parts within the vascular bundle system, with between 5 to 10% being redistributed to the grains and up to 50% to the roots (Coughtrey & Thorne, 1983).

In later growth stages, radiostrontium is washed off, as the plant material contaminated with radiostrontium is lost to desquamation of the cuticle (Madoz-Escande *et al.*, 2004). If contamination of the plant occurs close to harvest, then the uptake of both radiocaesium and radiostrontium will be more effective (Baeza *et al.*, 1999). In turnips and broad beans the uptake of radiostrontium is lower than the uptake of radiocaesium, and the uptake by their roots is higher than the uptake by their leaves (Baeza *et al.*, 1999); which confirms that radiostrontium has lower mobility than radiocaesium. In the grains of cereal species, the concentration of radiostrontium is lower than radiocaesium, and it is transported from different plant parts to the grain (Aarkrog, 1969). In soybeans, 0.7% g^{-1} of radiostrontium is absorbed by the leaves and there are lower concentrations of radiostrontium in the seeds, indicating that radiostrontium accumulates in seeds through deposition from atmospheric fallout (Shinonaga & Ambe, 1998). As radiostrontium has low redistribution in the plant after it has been deposited on plant parts, the deposition of radiostrontium during an early growth stage leads to low translocation to newly grown plant parts (Tukey *et al.*, 1961).

The redistribution of radiocaesium in crops can differ by a factor of 100 over the growing season and is related to the physiological development (growth stage) of the crop. The contamination of cereals before anthesis (the period from flower opening to fruit set) results in less radiocaesium being redistributed from the leaves to the seeds. If contamination takes place at anthesis, then a large fraction of the radiocaesium will be redistributed from the ears to other plant parts (Gerdung *et al.*, 1999). If precipitation occurs within two days after deposition, then more radiocaesium will be washed-off of the leaves than if precipitation occurs at a later stage, due to the quick uptake of radiocaesium by the plant (Madoz-Escande *et al.*, 2004). Radiocaesium is redistributed in the plant to the grain in different cereal species over time (Aarkrog, 1969), and if deposition occurs in the early growth stages, then the radiocaesium will be redistributed to the developing plant parts (Tukey *et al.*, 1961).

3.2 Transfer of radionuclides

The transfer of radionuclides from the environment to foodstuffs is controlled by the rate of direct uptake and root uptake by plant parts. The concentration of a radionuclide in a plant or plant part is linearly related to the concentration in the root zone of the soil. This proportionality is defined by a transfer factor (Ehlken & Kirchner, 2002) that is used to describe the transfer of radionuclides from the environment to plants or edible plant parts in a specific situation (von Fircks *et al.*, 2002). The transfer factor (TF, m^2 kg^{-1}) is defined as the ratio between the activity concentration of radionuclides in a plant or plant parts (Bq kg^{-1}), and the amount of radionuclides deposited per unit area (Bq m^{-2}) (Rosén *et al.*, 2011; Ehlken & Kirchner, 2002; Howard B.J. *et al.*, 1996; Rosén *et al.*, 1996). The transfer of intercepted radionuclides to edible plant parts can be described by a translocation factor (TLF, m^2 kg^{-1}), defined as the ratio between the activity concentration of radionuclides in plant parts (Bq kg^{-1}), and the amount of intercepted radionuclides by the plant foliage per unit area (Bq m^{-2}) (Vandecasteele *et al.*, 2001; Thiessen *et al.*, 1999).

The estimation of radionuclides transfer is used in decision making for the implementation of agricultural countermeasures to reduce the content of radionuclides in foodstuffs. The transfer of radionuclides through root uptake can be calculated in the first year after deposition. Data for calculating the transfer factor is readily available for root uptake, but the data is limited for the redistribution of radionuclides from leaves to other plant parts in certain scenarios, which has a strong seasonal variation (IAEA, 2010; Kostiainen *et al.*, 2002). If transfer factors relevant to a specific situation are unavailable, then the assessment of the situation after radioactive deposition can be challenging and wrong decisions might be made for the measures used to prevent the transfer of radionuclides to foodstuffs (Salbu, 2000).

3.3 Models of radionuclide transportation in ecosystems

After an accidental release of radionuclides into the environment, there is an urgent need to predict the level of exposure to the human population. Computer models are used to provide data for decision making on the mitigation of potential consequences and for implementing countermeasures (Thiessen *et al.*, 1999). There are different types of models that relate to different aspects of radionuclide transport and each model has its own temporal and spatial scales and temporal resolution. The simplest models consist of transfer functions and mechanistically describe the addition and losses of radionuclides from the soil-plant-atmosphere system. However, these models do not describe the uptake by the plant, or the translocation and storage processes in the plant. Some examples of these types of models are Pathway (Whicker & Kirchner, 1987) and Comida (Abbott & Rood, 1993). The model ECOSYS-87 (Müller & Pröhl, 1993) describes human exposure to radionuclides through multiple pathways *e.g.* inhalation, edible plant parts and processed food products. This model is also one of the models that provides information to the European decision support systems ARGOS and RODOS, which are used for assessment and decision making in a nuclear or radiological emergency (Andersson *et al.*, 2011a; Andersson *et al.*, 2011b).

3.3.1 Tracey model

The Tracey model (Gärdenäs *et al.*, 2009) describes radionuclide cycling, as trace element cycling in a soil-plant system affected by weather, vegetation development and management (Gärdenäs *et al.*, 2009). The radionuclide fluxes are estimated in proportion to the carbon or water fluxes, which are simulated with the external biophysical ecosystem model CoupModel (Jansson & Karlberg, 2004), and the ratio in the respective pool between the radionuclides and the amount of water or carbon. The plant root uptake of radionuclides is described in two ways: *passive uptake*; controlled by water uptake, and *active uptake*; controlled by growth, redistribution, and accumulation of the radionuclides in the plant due to carbon reallocation. These are described for different plant parts such as seeds, leaves, stems and roots. The soil is divided into different layers and in each layer, the radionuclide fluxes between the different organic pools; litter and humus, and between the adsorbed and solved pools, is described. The model is linked to the sensitivity toolbox Eikos (Ekström, 2005) for Monte-Carlo simulations.

3.3.2 CoupModel

The CoupModel simulates the flows of water, heat, carbon and nitrogen in the soil-plant-atmosphere system for different time steps from 1 hour up to several days. The fluxes of water, heat, carbon and nitrogen are dynamically coupled in each time-step (Jansson, 2012; Jansson & Karlberg, 2004), and the different fluxes of water, heat, carbon and nitrogen affect each other equally for every time step. The CoupModel is a successful combination of the SOIL (Jansson, 1998; Jansson & Halldin, 1979) and SOILN (Eckersten *et al.*, 1998; Johnsson *et al.*, 1987) models, all three have been used for different ecosystems and climate regions. Examples from agricultural ecosystems in northern Europe by Eckersten & Jansson (1991) and Blombäck *et al.* (1995) were relevant for this thesis. The same plant parts that were used in the Tracey model were considered. The soil profile was divided into a maximum of 100 layers with specified properties, such as hydraulic conductivity, root density and carbon and nitrogen content in litter and humus (Jansson, 2012; Jansson & Karlberg, 2004).

4 Materials and Methods

4.1 Study area

The field trial was conducted at the Ultuna meteorological and agricultural field station in Uppsala, Sweden (59°48′45″N and 17°38′45″E) during two growing seasons in 2010 and 2011. A nearby meteorological station monitors daily air temperature, precipitation and wind speed (Karlsson & Fagerberg, 1995).

The soil at the experimental site had a clay texture, and the main physical and chemical characteristics of the topsoil (0-30 cm) are presented in Table 1.

Table 1. *Summary of physical and chemical characteristics of the topsoil (0-30 cm) at the experimental site (Bengtsson et al., 2012).*

Soil parameter	
Particle size distribution:	
% clay (< 0.002 mm)	60
% silt (0.02 − 0.002 mm)	20
% sand (2 − 0.02 mm)	20
pH (H_2O)	6.5
AL-extractable ions (mg kg^{-1} soil)	
Ca	3690
K	202
P	57
HCl-extractable ions (mg kg^{-1} soil)	
Ca	6240
K	5720
P	640

The long-term (30-year, 1961-1999) annual mean air temperature was 5.6°C and the annual mean precipitation sum was 588 mm (SMHI, 2012). The 1st year growing season (2010: 1st of May to 30th of September) had a mean air temperature of 15°C and a precipitation sum of 293 mm. During the 2nd year growing season (2011: 1st of May to 30th of September), the mean air temperature was 15°C with a precipitation sum of 287 mm. The temperature at the deposition and sampling occasion varied between 10°C and 21°C, and there was no precipitation in connection with deposition and sampling on any occasion during the years of study, except for the last deposition and sampling occasion for spring wheat in the 2nd year. The wind speed at the time of deposition and sampling 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.

4.2 Design of the trial

A field trial with a randomised block design of 1 × 1 m^2 parcels in three replicates (in total 180 parcels) was laid out in the 1st year. In order to cover seasonal variations, a new trial with the same design was laid out on a nearby site in the 2nd year. Experimental crops were sown and managed according to common agricultural practises, except for covering the sowing beds with a non-woven fabric for three weeks after sowing to promote quicker growth. The experimental crops were spring oilseed rape (*Brássica napus* L.) variety 'Larissa' (Papers I and II), spring wheat (*Tríticum aestívum* L.) variety 'Triso' (Papers I and II) and a ley consisting 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.) (Paper III). Sowing took place in the middle of May in each year (except for the ley, which was only sown in the 1st year on bare soil), with seeding rates of 8 kg ha^{-1} for spring oilseed rape, 230 kg ha^{-1} for spring wheat and 25 kg ha^{-1} for the ley. For all of the 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 a high natural capacity for delivering K through weathering: the ammonium lactate-acetate soluble K was 202 mg kg^{-1} (Table 1), which according to Swedish standards, indicated no demand for potassium fertiliser (Yara-International-ASA, 2013).

The radionuclides chosen (^{134}Cs and ^{85}Sr) were deposited on the plants of spring oilseed rape at six different growth stages (Papers I and II), according to the Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH)-scale (Hack *et al.*, 1992). In the 1st year, 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 69; and, the beginning of ripening, code 80. In the 2nd year (Paper II), the growth stages were leaf development, code 15−19 (five to nine leaves 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) (Figure 3).

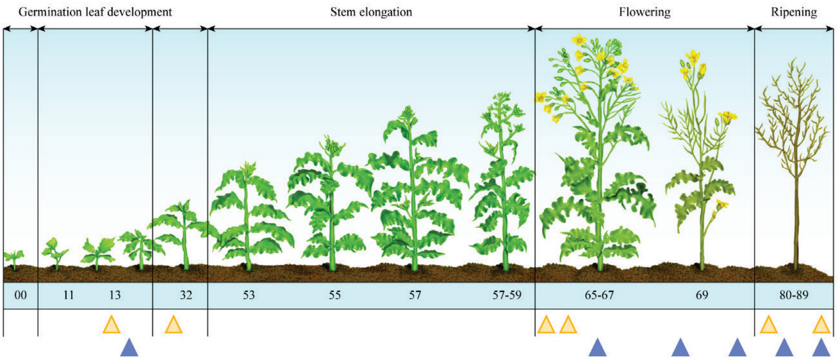

Figure 3. Growth stages in spring oilseed rape. Triangles indicate stages when deposition was carried out (△ 2010 and ▲ 2011) (Illustration by Elsevier B.V. Illustrator, 2012).

For spring wheat in the 1st year (Papers I and II), 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 the 2nd year (Paper II), 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 (Figure 4).

The reason why deposition took place at different growth stages in the 1st and 2nd years was the difficulties of being "on time" for exactly the same growth stages in both years.

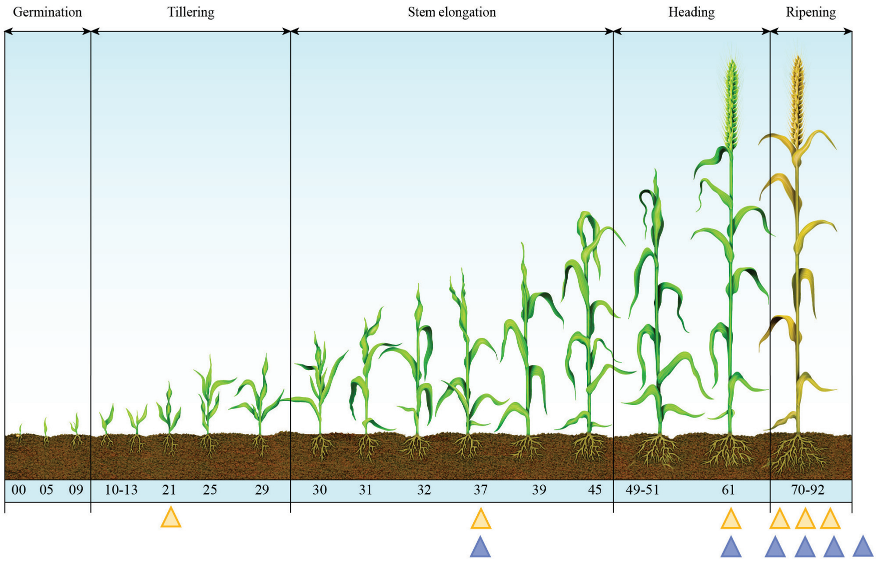

Figure 4. Growth stages in spring wheat adapted from Bayer Crop (2011). Triangles indicate stages when deposition was carried out (△ 2010 and ▲ 2011) (Illustration by Elsevier B.V. Illustrator, 2012).

The establishment and first harvest of ley in the 1st year corresponded to code 5:6 (Paper III), and the growth stages were a combination of the growth stages of grass and clover. The growth stages, according to Halling (2005), were 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 tassel visible:major stalk buddings visible); stem extension:initial flowering, code 4:5 (flag leaf visible:flowers visible on major stalk); and spike and tassel:flowering, code 5:6 (spike and tassel fully visible:flowers visible on major stalk and side stalk).

In the 2nd year, the first harvest of ley corresponded to code 4:3, and subsequent regrowth after code 7:7 rendered a second harvest at code 3:6 (Paper III). The growth stages were spike and tassel:initial budding, code 4:3 (spike and tassel 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 tassel visible:flowers visible on major stalk and side stalks); and, flowering:post-flowering, code 6:7 (full flowering:post-flowering) (Table 2).

Table 2. *Schedule for deposition and sampling of ley, three replicates for each combination of the growth stage at deposition and growth stage at sampling. Y = both the deposition and sampling, X = only sampling (time of deposition indicated by Y in the same row). *indicates regrowth after cutting at growth stage 7:7 in the 2nd year (2011). Growth stages relevant for a normal harvest of ley are emboldened.*

Year	Growth stage at deposition	Growth stage at sampling					
		0:0	1:1	2:2	**3:3**	**4:5**	**5:6**
1st year	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 year	**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 reason deposition took place at different growth stages in the 1st and 2nd years was due to the ley being established in the 1st year, which meant it started to grow much earlier and quicker in the 2nd year, as it was already established.

4.3 Preparation and deposition of artificial radioactive rain

An artificial rainwater solution was prepared from stock solutions. In the 1st year, the stock solutions contained 5 MBq L^{-1} ^{134}Cs and 15 MBq L^{-1} ^{85}Sr; and in the 2nd year, the solution contained 40 MBq L^{-1} ^{134}Cs and 37 MBq L^{-1} ^{85}Sr (Papers I, II and III). ^{134}Cs was in the form of caesium chloride (CsCl) in 0.1 M HCl solution, and ^{85}Sr was in the form of strontium chloride ($SrCl_2$) in 0.1 M HCl solution. Both radionuclides were mixed and diluted to the desired concentration in ultra-purified water (purity to 18.2 MΩ-cm (8 S cm^{-1})). In the 1st year, the amount of ^{134}Cs applied at different growth stages ranged from 24.5−30.9 kBq m^{-2}, and the amount of ^{85}Sr applied ranged from 28.5−49.8 kBq m^{-2}. In the 2nd year, the amount of ^{134}Cs ranged from 40.2−41.0 kBq m^{-2}, and the amount of ^{85}Sr ranged from 39.4−41.0 kBq m^{-2}.

The amounts of ^{134}Cs and ^{85}Sr differed between the two years because the stock solutions prepared with ultra-purified water in the 1st year resulted in absorption of the radionuclides on the glass surface−Si−OH groups of the bottles due to the ion exchange adsorption (Lehto & Hou, 2011). To avoid this problem in the 2nd year, stable isotopes of the same elements (Cs and Sr) in the form of CsCl and $SrCl_2$ were added to the radionuclide stock solutions, which were stored in hydrophobic plastic bottles to avoid ion exchange adsorption (Lehto & Hou, 2011).

The radionuclides were applied with a rainfall simulator (Figures 5 and 6) that was a modified version of the drip infiltrometer developed by Joel and Messing (2001). In both years, a Watson-Marlow 520 series process pump was used to apply the precipitation, and during each treatment, the amount of precipitation applied was 1.00±0.01 mm at an intensity of 1 mm 30 s^{-1}. During deposition in the early growth stages, a windshield was used to prevent wind disturbance.

Figure 5. Picture of the rainfall simulator used in the field trial. Photo taken by Stefan B. Bengtsson.

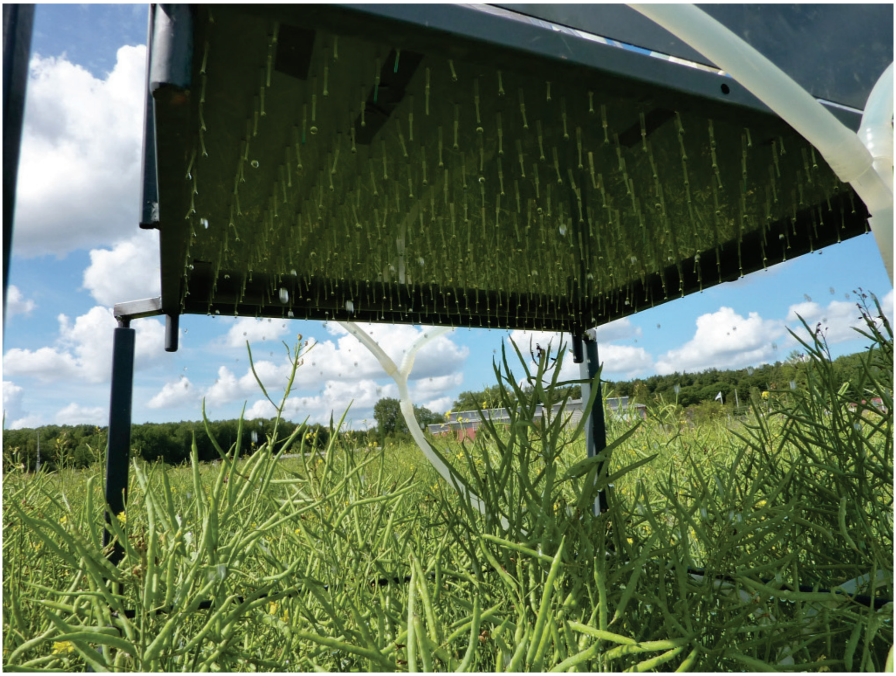

Figure 6. Close up of the rainfall simulator showing the bottom with the individual pipes where the rain drops disengaged. Photo taken by Anna-Lisa Mårtensson (Mårtensson, 2012).

4.4 Sampling and measurements

In both years, the plants were sampled within a frame (25 × 25 cm^2 square) placed in the middle of each parcel. Sampling was two-three hours after deposition in one set of replicates, and in another set at harvest (Papers I and II) or at later growth stages (Paper III). The whole plants were sampled (all crops) and the plant compartments were separated (only spring oilseed rape and spring wheat). Spring oilseed rape was separated into stems (stem and attached dead leaves), leaves, flowers, siliques (except seeds) and remaining seed materials (Paper II). Spring wheat was separated into stems (stem and attached dead leaves), leaves, flower spikes, ears (husk) except for grains and remaining grain material (Paper II).

The plant material was 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 (depending on the amount of plant material) with a suitable geometry for measuring activity concentration. The activity concentrations of the radionuclides were expressed as Bq kg^{-1} d.w. and a correction for the decay between sampling the date and the date of analysis was calculated. Samples from early growth stages were measured in 35 mL jars and the results obtained were corrected for the degree of jars filling due to the small amount of plant material. The determination of the correction factor for each detector has been described in Paper I.

4.5 Measurement and analyses

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

4.5.1 Calibration of the HPGe detectors

The measured activity concentrations included uncertainties of the efficiency calibration of the HPGe detector, which was assumed as one of the dominant components of the total measured uncertainty (Boson *et al.*, 2009; 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 been described in Paper I, and was according to principles presented in Bronson and Young (1997) and ANSI (1978).

4.6 Calculations

4.6.1 Calculation of the interception fraction (Papers I and III)

The interception of wet deposited radionuclides by the crops was expressed as the interception fraction, *f*, according to Equation (1) (Pröhl, 2009). The interception fraction was the ratio between the activity in the standing plant 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.):

$$f = A_i/A_t \qquad \text{Equation (1)}$$

4.6.2 Calculation of the transfer factors (Papers II and III) and translocation factors (Paper II)

The concentration of radionuclides in edible plant parts, *i.e.* seeds, to the amount of deposition was calculated as the transfer factor, TF (m^2 kg^{-1}), with Equation (2) (Ehlken & Kirchner, 2002; Howard B.J. *et al.*, 1996; Rosén *et al.*, 1996). The transfer factor is the ratio between the total activity in seeds at sampling (A_c, Bq kg^{-1}, d.w.), and the amount of activity deposited (A_t, Bq m^{-2}, d.w.).

$$\mathrm{TF} = A_c/A_t \qquad [\mathrm{m^2\ kg^{-1}}] \qquad \text{Equation (2)}$$

The concentration of radionuclides in edible plant parts in relation to the amount of interception was calculated as the translocation factor, TLF (m^2 kg^{-1}, d.w.), with Equation (3) (Vandecasteele *et al.*, 2001; Thiessen *et al.*, 1999). The translocation factor is the ratio between the total activity in seeds at sampling (A_c, Bq kg^{-1}, d.w.), and the amount of activity intercepted at deposition (A_i, Bq m^{-2}, d.w.).

$$\mathrm{TLF} = A_c/A_i \qquad [\mathrm{m^2\ kg^{-1}}] \qquad \text{Equation (3)}$$

4.6.3 Calculation of radionuclide transfer to beef and cow's milk (Paper III)

The transfer of radionuclides to beef was calculated by multiplying the daily intake (d) of d.w. ley by the cattle (estimated to 9 kg d.w. daily (Rosén & Eriksson, 2008)) and the transfer coefficient (*Ff*), which is 0.022 d kg^{-1} for Cs and 0.0013 d kg^{-1} for Sr (IAEA, 1994).

The transfer of radionuclides to cow's milk was calculated by multiplying the daily intake (d) of d.w. ley by the cows (estimated to 10 kg d.w. daily (Rosén & Eriksson, 2008)) and the transfer coefficient for cow's milk (*Fm*), which is 0.0046 d L^{-1} for Cs and 0.0013 d L^{-1} for Sr (IAEA, 2010).

4.7 Statistics (Papers I, II, III)

The relationship between the standing plant biomass, leaf area index (LAI), interception fraction and mass interception fraction were identified by

Student's t-test and correlation (Papers I and III). The relationship between ^{134}Cs and ^{85}Sr interception fractions (Papers I and III) were identified by analyses of variances (ANOVA). Relationships between ^{134}Cs, ^{85}Sr concentrations, growth stage and years were identified by paired t-test (Paper II) or Student's t-test (Paper III), correlation and ANOVA (Papers II and III). In Papers II and III, statistical analyses were made with the computer program R version 2.15.2 (© The R Foundation for Statistical Computing, Vienna, Austria (2012; 2011). In Paper I, Minitab 16® (© Minitab Inc. Pennsylvania, USA (2010)) was used for ANOVA and Microsoft Excel 2010 (© Microsoft Inc. Washington, USA (2010)) was used for regression analyses.

4.8 Uncertainties in the measurements (Papers I, II and III)

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 were reported as the combined standard uncertainty, $u_c(y)$, for measurement of standing plant biomass (Papers I and III) and for the concentration of radionuclides (Papers I, II and III). The combined standard uncertainty of the output estimate, y, was calculated according to Equation (4).

$$u_c(y) = y \times \left(\sqrt{\sum_{i=n}^{\infty}\left(\frac{u(x_N)}{x_N}\right)^2}\right) \qquad \text{Equation (4)}$$

Where: y is the output estimate and x_N is the input estimates.

The uncertainties considered were the purity of the radionuclides, the difficulty in obtaining plant samples from a well-defined area (estimated), the variation in the d.w. of samples, the error in measuring the exact activity concentration in the deposited liquid, the uncertainty of the volume prepared for the deposition event and the error in the liquid volume deposited by the rainfall simulator (Papers I and III). The absorption of radionuclides onto the surface of the rainwater simulator 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).

4.9 Model development (Paper IV)

4.9.1 Tracey extension (Paper IV)

The Tracey model was extended to include a description of the cycling of ^{134}Cs and ^{85}Sr in the agricultural ecosystem after contamination via wet deposition. A schematic description of the existing fluxes and the added fluxes is presented in Figure 7. Carbon fluxes were added to the model to describe both forest (existing flux) and agricultural ecosystems (added flux). In addition a number of water fluxes were added as driving variables; these fluxes were precipitation, interception, throughfall, infiltration and evaporation, which were simulated in the CoupModel.

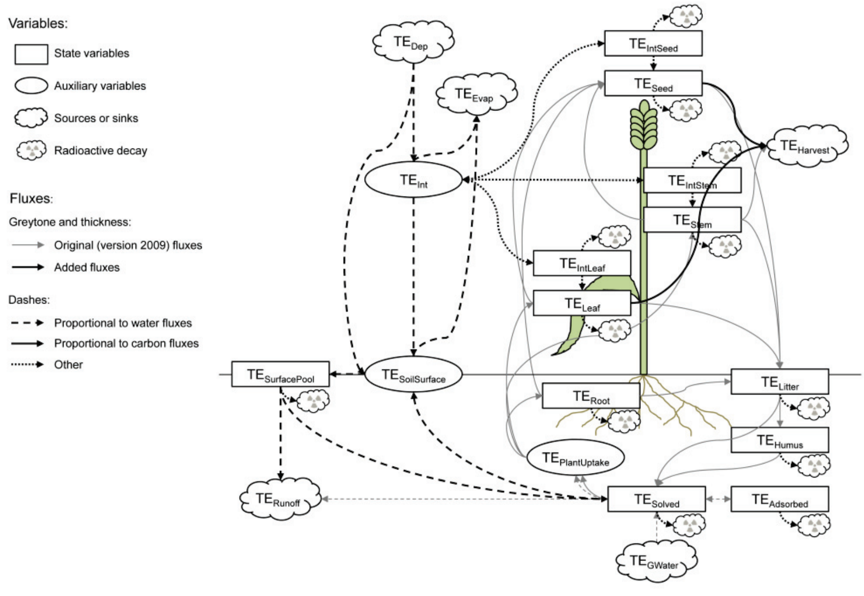

Figure 7. Pools and fluxes of trace elements (TE) in the model. The grey arrows represent fluxes of the original version from 2009, and the black arrows represent the added fluxes. The solid arrows represent TE fluxes that are proportional to carbon fluxes, the dashed arrows represent TE fluxes that are proportional to water fluxes and the dotted arrows are other proportionalities. The boxes represent state variables, the clouds represent sinks or sources, and the circles are auxiliary variables. The soil profile is divided into different layers, each of which includes all of the soil's TE pools.

Interception and foliar uptake

Interception of trace elements (TE) on the crop's surface at each time step was defined as a function of intercepted water flux multiplied with the concentration of radionuclides in the deposited water. Intercepted trace elements were divided into the plant parts leaf, stem and seed, in relation to the ratio of each plant part's biomass. Scaling factors were used to describe the negative and/or positive discrimination of trace element fluxes to the equivalent water or carbon fluxes, *e.g.* to describe a higher or lower interception capacity of leaves over stems for the same amount of biomass.

The fixation or foliar uptake of trace elements was defined as a function of the fixation rate of the plant parts. The definition of foliar uptake was extended to include uptake by all of the above ground plant parts, each having its own fixation rate.

Throughfall, infiltration and volatilisation

The sum of direct and indirect throughfall fluxes was classed as throughfall. Direct throughfall is the flux of trace elements that are not intercepted by the crop at deposition, and indirect throughfall is the flux of trace elements first intercepted but then washed-off at later precipitation when the storage capacity of water interception is exceeded. Thus, weathering (*e.g.* wind and rain) is described by a function of the corresponding water flux. Weathering due to other forces is indirectly summed in the scaling factor of indirect throughfall; similarly, the scaling factor for indirect throughfall is inversely proportional to the retention of the intercepted radionuclide.

In the soil, the infiltration of trace elements was denoted as a function of the trace element to the water ratio in the throughfall and the capacity of infiltration at the soil's surface. A pool of water with trace elements at the soil's surface is formed if the water throughfall exceeds the water infiltration capacity. The trace elements in the surface pool can infiltrate with a delay into the soil, or be lost as surface runoff.

The volatilisation of radionuclides was assumed to be negligible.

Radioactive decay

A description of the radionuclide decay of ^{134}Cs and ^{85}Sr was added to all the pools of trace elements.

4.9.2 Tracey application (Paper IV)

Contamination

The radioactive contamination was made by adding the same amount of radionuclides to the atmospheric deposition pool in the model as in the experiment for the different deposition occasions. One millimetre of rain was added to precipitation in the CoupModel simulations at these occasions.

Sensitivity analysis

The sensitivity analyses were made with the software package Eikos (Ekström, 2005), with added scripts. The parameters and distributions used in the application of Tracey are described in Table 3, where *s*-parameters are scaling factors, *k*-parameters are rate parameters, *f*-parameters describe fractions and *p*-parameters are other types of parameters. Atmospheric parameters are associated with fluxes such as deposition, interception, throughfall, and infiltration. Plant parameters are associated with fluxes such as foliar uptake, translocation. Litter fall, and harvest. Soil parameters are associated with fluxes in the soil such as adsorption, movement within water in soil, organic matter decomposition, and root uptake. Different parameters were used for the active and passive model approach for the root uptake. Discrimination of several TE fluxes was disregarded and their scaling factors were set to one. Thus, 18 and 17 parameters remained to be used in the sensitivity analyses with the active respectively passive root uptake approach.

A distribution on a logarithmic scale was chosen when most of the values were assumed to occur at the lower end of the range. A log-triangular distribution was chosen when minimum, maximum and a most likely nominal value were found in the literature. A uniform distribution was chosen when no information about the distribution was available.

Different parameters were used for ^{134}Cs and ^{85}Sr when such information was available. Several parameter values for ^{85}Sr were set to half the values of ^{134}Cs translocation within plants.

The parameters were set with the Latin Hypercube sampling technique included in Eikos (Ekström, 2005). Parameter distributions were divided into equal probability intervals, and each interval was sampled exactly once, ensuring that the entire range of the distribution was explored, and this with fewer samples than with simple random sampling. A total of 1, 000 parameters sets were drawn for ^{134}Cs and ^{85}Sr, for both active and passive root uptake for the six deposition treatments during both years, giving a total of 48, 000 simulations.

Spearman’s rank correlation coefficients were used as sensitivity measures and these calculations were carried out using Eikos.

Table 3. *Parameters and their distributions used in the application of Tracey were arranged by sphere; atmosphere, biosphere and lithosphere, where s-parameters are scaling factors, k-parameters are rate parameters, f-parameters describe fraction and p-parameters are other types of parameters. Some of the soil parameters are general, some concern plant water uptake and allocation and are specific for either active root uptake or passive root uptake. See the Appendix in paper 4, for equations and parameter definitions of added parameters, and Gärdenäs et al. (2009) for equations and definitions of parameters and the original model version.*

Name (unit)	Prior distribution (mean, sd, min, max, nominal)	Posterior distribution (mean, sd, min, max)
Atmosphere		
$s_{TEDep \rightarrow SoilSurface}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TEDep \rightarrow Int}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TEInt \rightarrow IntSeed}$ (-)	Uniform (-, -, 0, 1, -)[1]	Normal (0.21, 0.22, 0, 1)
$s_{TEInt \rightarrow IntLeaf}$ (-)	Uniform (-, -, 0, 1, -)[1]	-
$s_{TEInt \rightarrow IntStem}$ (-)	Uniform (-, -, 0, 1, -)[1]	Normal (0.37, 0.21, 0, 1)
$s_{TEInt \rightarrow SoilSurface}$ (-)	Cs: Log-normal (0.7, 0.4, 0, 1.5, -)[1, 7]	-
	Sr: Log-normal (0.35, 0.4, 0, 1.5, -)[1, 7]	Sr: Lognormal (0.39, 0.45, 0, 1.5)
$s_{TEInt \rightarrow Evap}$ (-)	- (-, -, -, -, 0)[1]	-
$s_{TESoilSurface \rightarrow Solved}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TESurfacePool \rightarrow Solved}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TESurfacePool \rightarrow Runoff}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TESolved \rightarrow Evap}$ (-)	- (-, -, -, -, 0)[1]	-
Plant		
$k_{TEFixSeed}$ (d^{-1})	Cs: Log-triangular (-, -, 0, 1, 0.0055)[14]	-
	Sr: Log-triangular (-, -, 0, 1, 0.001)[14]	-
$k_{TEFixLeaf}$ (d^{-1})	Cs: Log-triangular (-, -, 0, 1, 0.0055)[14]	Cs: Log-normal (0.04, 0.67, 0, 1)
	Sr: Log-triangular (-, -, 0, 1, 0.001)[14]	Sr: Log-normal (0.01, 0.10, 0, 1)

$k_{TEFixStem}$ (d^{-1})	Cs: Log-triangular (-, -, 0, 1, 0.0055)[14]	Cs: Log-normal (0.04, 0.47, 0, 1)
	Sr: Log-triangular (-, -, 0, 1, 0.001)[14]	Sr: Log-normal (0.01, 0.08, 0, 1)
$s_{TESeed \rightarrow Litter}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TELeaf \rightarrow Litter}$ (-)	Uniform (-, -, 0, 1, -)[1]	-
$s_{TEStem \rightarrow Litter}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TERoot \rightarrow Litter}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TELeaf \rightarrow Seed}$ (-)	Cs: Log-normal (0.7, 0.15, 0, 1,-)[1, 13]	-
	Sr: Log-normal (0.35, 0.15, 0, 1,-)[1, 13]	-
$s_{TEStem \rightarrow Seed}$ (-)	Cs: Log-normal (0.7, 0.15, 0, 1,-)[1, 13]	-
	Sr: Log-normal (0.35, 0.15, 0, 1,-)[1, 13]	-
$s_{TERoot \rightarrow Seed}$ (-)	Cs: Log-normal (0.7, 0.15, 0, 1,-)[1, 13]	-
	Sr: Log-normal (0.35, 0.15, 0, 1,-)[1, 13]	-
$s_{TEStem \rightarrow Harvest}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TELeaf \rightarrow Harvest}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TESeed \rightarrow Harvest}$ (-)	- (-, -, -, -, 1)[1]	-
Soil		
p_{Kd} (m^3 kg^{-1})	Cs: Log-triangular (-, -, 0.1, 10, 1)[3]	-
	Sr: Log-triangular (-, -, 0.001, 0.1, 0.01)[3]	Sr: Lognormal (0.02, 0.02, 0, 1)
$p_{BulkDensity}$ (kg m^{-3})	Normal (1418, 60, -, -, -)[2, 10]	
$s_{TEWaterFlow}$ (-)	Log-normal (0.35, 0.4, 0, 1.5, -)[1, 4, 12]	-
$s_{TELitter1 \rightarrow Solved}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TELitter1 \rightarrow Humus}$ (-)	- (-, -, -, -, 1)[1]	-
$s_{TEHumus \rightarrow Solved}$ (-)	- (-, -, -, -, 1)[1]	-
Active uptake		
$k_{TESolvedUptake}$ (d^{-1})	Cs: Log-normal (0.1, 0.3, 0, 1, -)[1, 5, 6, 11]	-

	Sr: Log-normal (0.05, 0.3, 0, 1, -)[1, 5, 6, 11]	-
$p_{MaxTECLeaf}$ (mg TE (g C)$^{-1}$)	Cs: Log-normal (65, 17, -, -, -)[1, 9]	-
	Sr: Log-normal (30, 12, -, -, -)[1, 9]	-
$p_{MaxTECStem}$ (mg TE (g C)$^{-1}$)	Cs: Log-normal (30, 8, -, -, -)[1, 2]	-
	Sr: Log-normal (10, 4, -, -, -)[1, 2]	-
$p_{MaxTECRoot}$ (mg TE (g C)$^{-1}$)	Cs: Log-normal (50, 28, -, -, -)[1]	-
	Sr: Log-normal (22, 20, -, -, -)[1]	-
Passive uptake		
$s_{TEWaterUptake}$ (-)	Cs: Log-normal (0.7, 0.15, 0, 1,-)[1, 6]	-
	Sr: Log-normal (0.35, 0.15, 0, 1,-)[1, 6]	-
f_{PULeaf} (-)	Normal (0.5, 0.1, 0, 1, -)[1, 8]	-
f_{PUStem} (-)	Normal (0.3, 0.1, 0, 1, -)[1, 8]	-
Radioactive decay		
$p_{HalfLife}$ (d)	Cs:- (-, -, -, -, 754)	-
	Sr: - (-, -, -, -, 65)	-

1. Assumed
2. Bengtsson *et al.* (2013)
3. Bergström *et al.* (1999)
4. Carrillo-Gonzalez *et al.* (2006)
5. Eckersten *et al.* (2007)
6. Marschner (2012)
7. Müller and Pröhl (1993)
8. Nabipour *et al.* (2007)
9. Rasmusson *et al.* (1971)
10. Sandsborg and Wiklert (1975/1976)
11. Shaw (1993)
12. Simunek *et al.* (2006)
13. Strebl *et al.* (2007)
14. Whicker and Kirchner (1987)

Criteria of acceptance

Simulated estimates of radionuclides in and adsorbed on grains were accepted if they were within the 95% confidence interval of the mean value of the replicates. The sum of simulated storage in grains (TE_{Seed}) and simulated intercepted storage on grains ($TE_{IntSeed}$) was used as it was not possible to distinguish between intercepted and stored radionuclide activity in the measured values. The sum of measured grains and husks was used as the model did not distinguish between grain and husk. Comparison was first made for the 6th sampling at harvest, and then for the 5th sampling and so on until there were no longer any parameter sets left that could accurately simulate the whole measured dynamics. In some cases, it was not possible to make comparison due to that all the replicates were zero.

Posterior distribution

The Kolmogorov-Smirnov test (Marsaglia & Tsang, 2003; Massey, 1951) was used to test if there were significant differences between the prior and the posterior parameter distributions. The posterior distribution parameters were estimated using maximum-likelihood, with truncated distributions when necessary.

4.9.3 CoupModel application (Paper IV)

The water and carbon fluxes were simulated with the CoupModel (Jansson & Karlberg, 2004). The daily values for air temperature, precipitation, wind speed, relative humidity and solar radiation were measured by the Ultuna meteorological station. The leaf area index (LAI) values measured at deposition and harvest were used to estimate the leaf biomass development.

The soil was divided into 10 layers according to the U12 profile for the same location by Sandsborg and Wiklert (1975/1976): the physical properties of the soil, water retention curves and hydraulic conductivity were from the U12 profile. The initial soil moisture was set to 60 cm water pressure, and the initial soil carbon (C) and nitrogen (N) content was adjusted to the measurements.

The root depth was 170 cm for wheat, in accordance to observations by Sandsborg and Wiklert (1975/1976), and the critical water pressure for root water uptake was 10 000 cm. The parameters for the Lohammar equation for transpiration, the half saturation light response R_o of 5106 MJm^{-2}day^{-1}, the maximum conductance g_{max} 0.012 ms^{-1} and the vapour pressure deficit at 50% closure of stomata 1300 Pa were from Heidmann *et al.* (2000). Water interception capacity per LAI was set to 0.2 mm LAI^{-1} (Kang *et al.*, 2005).

Carbon and nitrogen parameters were based on Eckersten and Jansson (1991) and Blombäck *et al.* (1995), *e.g.* minimum C:N ratio of different plant parts (leaves 7.5, roots 18 and straw 22.5) and maximum C:N ratio of leaf litter (50). The fluxes of carbon to grain from other plant parts were highest for leaves (0.06 d^{-1} in the 1st year and 0.04 d^{-1} in the 2nd year), stems (0.02 d^{-1}) and roots (0.01 d^{-1}).

In order to mimic the late, but exponential, growth in spring of the 1st year, the specific leaf area was set to 11.6 g C m^{-2} for the 1st year and 20 g C m^{-2} for the 2nd year.

Many parameters, *e.g.* solar radiation, use efficiency (3.5 g d.w. MJ^{-1}, with 2.2−8.2 g d.w. MJ^{-1} being reported to be the physical range of spring wheat (Loomis & Amthor, 1999)) and specific leaf area, were included to provide calibration with good agreement between the simulated and measured soil moisture dynamics at 30 and 70 cm in depth, and between the simulated and measured C content of the standing biomass, leaves, stems, flowers and/or grains (the C content of plant dry weight was assumed to be 45%).

5 Results and Discussion

5.1 Interception of ^{134}Cs and ^{85}Sr by crops (Papers I and III)

The amounts of wet deposited ^{134}Cs and ^{85}Sr intercepted by standing crops depend on both the plant's biomass and its leaf area. The interception fraction (*f*) for both radionuclides increased in relation to increasing values of standing plant biomass in spring wheat and ley (Figures 8 and 9): a similar relationship for spring wheat has also been found by Vandecasteele *et al.* (2001). However, in spring oilseed rape, the interception fraction (*f*) for both radionuclides decreased with higher values of standing plant biomass (Figure 8). The weaker relation between interception and standing plant biomass for spring oilseed rape could be explained by plants shedding leaves at later growth stages, whereas the total above ground biomass still increased due to the rapid growth of siliques (a fruit (seed capsule) of 2 fused carpels).

In all crops, increasing values of leaf area (LAI) were related to interception of both radionuclides (Figure 8 and 9); although this relation was weaker for spring oilseed rape (Figure 8). Vandecasteele *et al.* (2001) found a similar correlation between the interception for ^{137}Cs and ^{90}Sr on spring wheat.

The crops studied intercepted different fractions of ^{134}Cs and ^{85}Sr. The fraction of ^{134}Cs and ^{85}Sr intercepted by spring oilseed rape was lowest after the first deposition, and remained constant thereafter. The fraction of ^{134}Cs and ^{85}Sr intercepted by spring wheat was highest after the middle deposition, which was at fruit development (code 70): in similar experiments on spring wheat, Vandecasteele *et al*. (2001) and Eriksson *et al.* (1998b) found that the highest interception occurs at the same growth stages as presented in this thesis. Interception of both radionuclides by ley in the 1st year increased during later growth stages and peaked after the last deposition at spike and tassel:flowering (code 5:6), and could be explained by the ley being established in the 1st year. However, in the 2nd year, radionuclide interception was lowest after the first deposition, and increased drastically after the second deposition, but was constant at the later growth stages. This could be explained by the low amount of ley biomass, as the ley "died off" during the winter period.

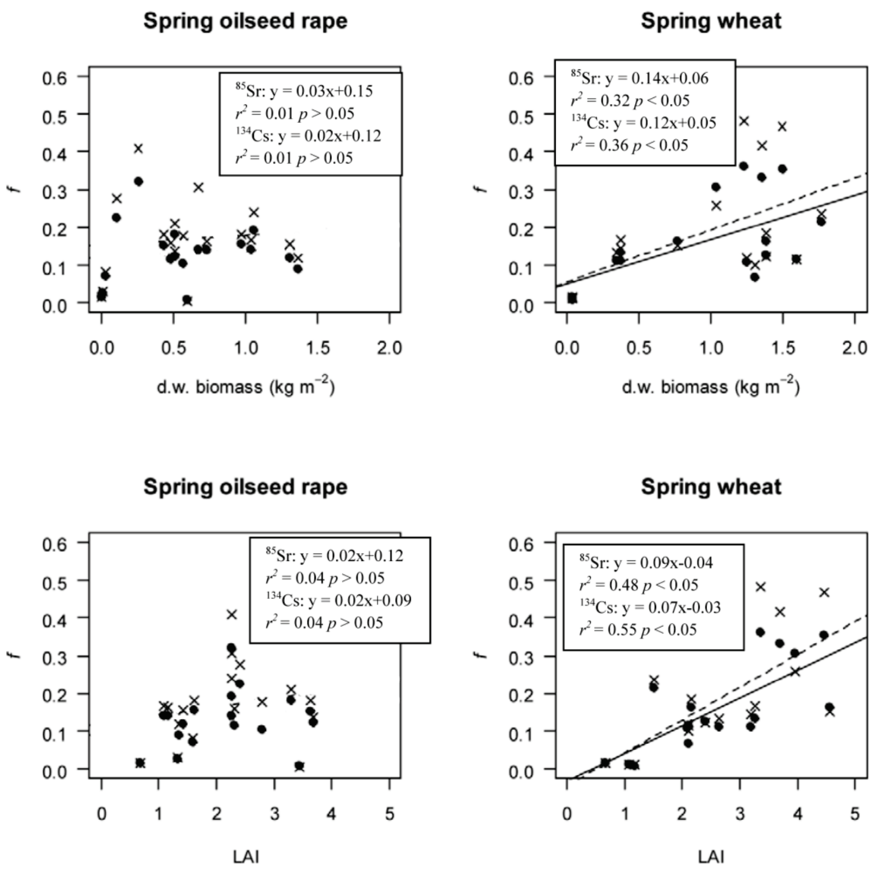

Figure 8. Relationship between intercepted fraction (*f*) of ^{134}Cs (●, −) and ^{85}Sr (×, - -) deposited on spring oilseed rape and spring wheat as a function of standing biomass dry weight (d.w.) and leaf area index (LAI).

Generally, interception was slightly higher for ^{85}Sr than for ^{134}Cs in all three crops (Figures 8 and 9). This could be explained by the difference in valence between ^{85}Sr (divalent) and ^{134}Cs (monovalent) ions. Divalent ions are assumed to bind more strongly to the surface of plants than monovalent ions (Vandecasteele *et al.*, 2001; Bréchignac *et al.*, 2000).

However, when estimating the degree of radionuclide contamination of crops, the interception fraction should be used with caution, as the estimation of interception requires a single value for each crop, radionuclide, level of precipitation and radionuclide concentration in precipitation (Kinnersley *et al.*, 1997; Hoffman *et al.*, 1992).

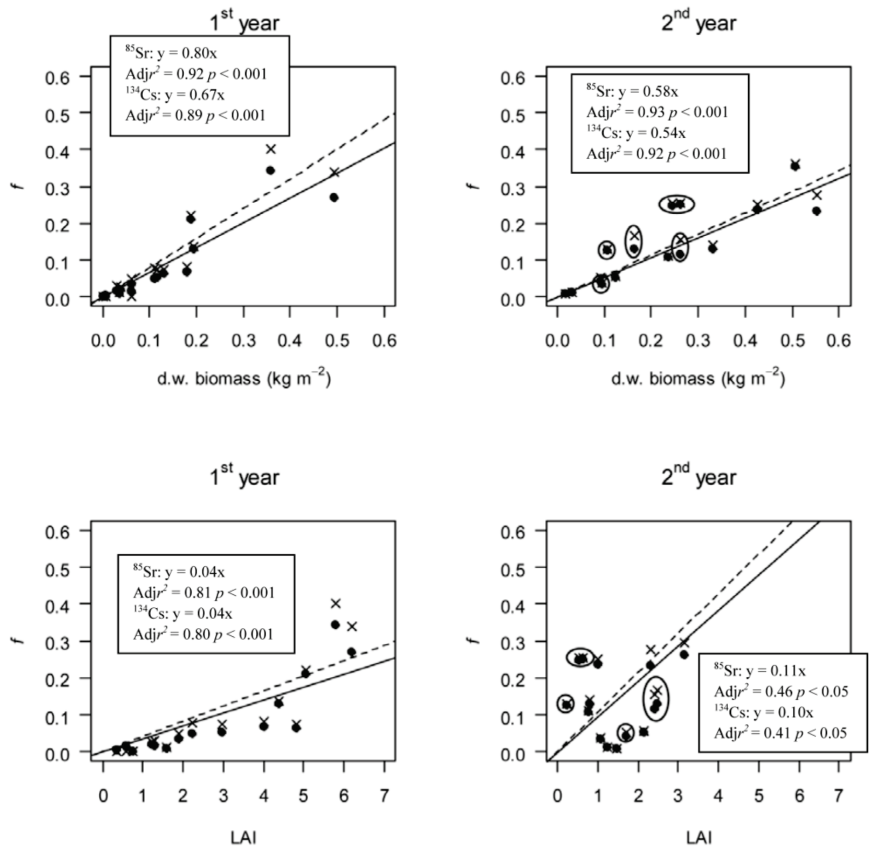

Figure 9. Relationship between intercepted fraction (*f*) of ^{134}Cs (●, −) and ^{85}Sr (×, - -) deposited on ley as a function of standing plant biomass dry weight (d.w.) and leaf area index (LAI). Encircled points in the 2nd year are values measured at the second harvest of a regrowth.

Furthermore, the level of radionuclide-interception is not directly related to the intensity of precipitation but rather to the external water storage capacity of the plant and the accumulation of radionuclides on the crop's surface (Kinnersley *et al.*, 1997).

There was a possibility that the intercepted fraction of the deposited radionuclides was affected by the high intensity of precipitation during the 30-s application. Therefore, the lower precipitation intensity might have rendered higher interception values, as there is less "splash off" from the crop at lower intensities than at higher precipitation intensities (Keim *et al.*, 2006; Wang *et al.*, 2005).

5.2 Activity concentration of ^{134}Cs and ^{85}Sr in crops (Papers II and III)

Wet deposition of radionuclides that occurred later in the growing season caused higher ^{134}Cs and ^{85}Sr activity concentrations in the different plant parts of both spring oilseed rape and spring wheat. The plant parts with the highest activity concentration and activity of both radionuclides were siliques (except seeds) of spring oilseed rape and the husk of spring wheat. The highest activity concentration in ley was found when deposition was closer to harvest (sampling) and the crop was more developed (Figure 10). After radiocaesium is released into the atmosphere, one of the important pathways for radiocaesium entry into plants is through direct contamination onto crops (Vandecasteele *et al.*, 2001). Radiocaesium is relatively mobile within the plant tissues, but lower availability for root uptake from soils, especially from soils high in clay content due to fixation of caesium to clay minerals (Absalom *et al.*, 2001; Absalom *et al.*, 1995). This can explain why direct contamination of crops by radiocaesium is an important pathway.

In the whole plants of spring wheat, the activity concentration for ^{85}Sr was higher than for ^{134}Cs. In the 1st year, the highest activity concentration in ley was for ^{85}Sr; but in the 2nd year, it was ^{134}Cs. In a similar experiment, the activity concentration of ^{134}Cs in ley was found to be higher than the activity concentration of ^{85}Sr (Eriksson *et al.*, 1998a). The lower activity concentration of ^{134}Cs and ^{85}Sr could be explained by lower interception, with radionuclide retention due to fall-off and wash-off during early growth stages (Colle *et al.*, 2009; Eriksson *et al.*, 1998b).

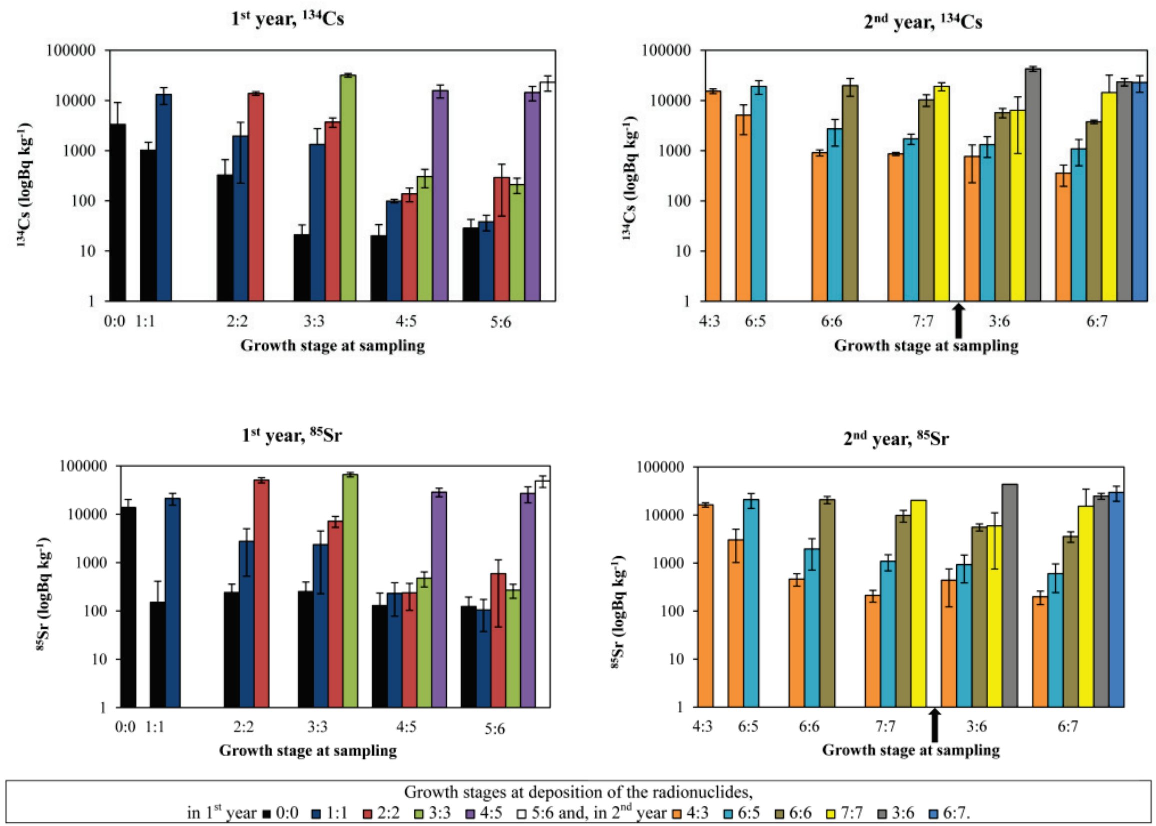

Figure 10. Logged activity concentration of ^{134}Cs and ^{85}Sr at different harvest occasions after deposition at different growth stages in ley (n = 3 at all growth stages, except at deposition/sampling in the 2nd year (6:5/6:6) where n = 2). Error bars indicate standard deviation. The arrow indicates where regrowth with the second harvest started in the 2nd year.

The activity concentrations of ^{134}Cs and ^{85}Sr in seeds of spring oilseed rape and spring wheat at harvest varied depending on the time of deposition. The lowest activity concentrations of ^{134}Cs and ^{85}Sr were when deposition took place during the early growth stages, and concentrations were higher when deposition took place during the growth of the crops and at flowering. In spring oilseed rape and spring wheat, the highest activity concentrations of ^{134}Cs and ^{85}Sr occurred when deposition occurred at the ripening phase. The increase in activity concentration of both radionuclides in seeds in relation to growth was lower than in other plant parts *e.g.* the straw and siliques (without seeds), and could be explained by a dilution effect; as seed biomass increases in later growth stages (Coughtrey & Thorne, 1983). After foliar contamination of spring wheat, the accumulation of ^{134}Cs and ^{85}Sr in seeds increased at later growth stages (Eriksson *et al.*, 1998b; Aarkrog, 1969): ^{85}Sr concentration in seeds is lower than in the straw, and the highest concentration occurs if contamination occurs before harvest. For spring oilseed rape, data on radionuclide interception and transfer to seeds after direct deposition onto a growing crop is lacking. However, data available for cereals (barley, wheat, rice, and rye) indicates that the highest transfer of radionuclides occurs after the emergence of the ears (Colle *et al.*, 2009; Vandecasteele *et al.*, 2001; Eriksson *et al.*, 1998b; Voigt *et al.*, 1991; Middleton, 1959; Middleton, 1958).

5.2.1 Distribution of wet deposited ^{134}Cs and ^{85}Sr between plant parts (Paper II)

The majority of wet deposited radionuclides taken up by spring oilseed rape and spring wheat was found in the straw, with a smaller fraction being in the seeds (Table 4). Radionuclide distribution between different parts of spring oilseed rape varied depending on the time of deposition, indicating ^{134}Cs increased in siliques (except seeds) and levelled-off or decreased in the straw after deposition occurred in later growth stages.

For spring wheat, the majority of ^{134}Cs was redistributed to the straw and to the grains. In the 2nd year, more ^{134}Cs was found in the grains than in the straw, and the ratio between the grain and the straw's biomass was higher than in the 1st year; whereas, the grain's biomass did not differ between the two years. For ^{85}Sr, the redistribution to grain was higher in the 2nd year, but the amount was still highest in the straw. The distribution of ^{85}Sr among different plant parts did not appear to be related to the deposition occasion. After root uptake by oilseed rape 12 years after the Chernobyl accident, 65% of ^{137}Cs, and 82% of ^{90}Sr were found in the straw, and only 3% of ^{137}Cs and 6% of ^{90}Sr were found in the seeds (Bogdevitch *et al.*, 2002). After root uptake by spring wheat growing on contaminated loamy sand soil, 85% of ^{137}Cs and 91% of ^{90}Sr were found in the straw, and 18% of ^{137}Cs and 8% of ^{90}Sr were found in the grain (Putyatin *et al.*, 2006). The pattern of radionuclide distribution between the above ground plant parts after wet deposition was comparable to other studies by *e.g.* Bogdevitch *et al.* (2002) and Putyatin *et al.* (2006), where the uptake of radionuclides was entirely from the soil. This suggests that the distribution of radionuclides among above ground plant parts appeared to be independent in the route of uptake of radionuclides (uptake by roots or foliar uptake).

However, radionuclides might have fallen directly onto the ground after being washed-off from the plant surface; thus, they were available for root uptake from the soil after deposition.

Table 4. *The percentages of ^{134}Cs and ^{85}Sr in different plant parts and stages in spring oilseed rape and spring wheat at harvest (September) after wet deposition occurred at different growth stages. Means are from three replicates (n = 3), except where ‡ indicates n = 2. The percentage was not estimated due to activity below minimum detectable limit is denoted by *.*

year / crop	Growth stage at deposition	^{134}Cs			^{85}Sr		
		Seeds	Siliques (except seeds)	Straw	Seeds	Siliques (except seeds)	Straw
2010 / Oilseed	13	20	8	72	14	14	72
	32	11	25	64	*	*	*
	61	14	34	52	3	57	40
	65	13	37	50	6	50	44
	80	8	39	53	4	51	45
2011 / Oilseed	15-19	21	15	64	13	31	56
	65	16‡	18‡	66‡	*	*	*
	69	23	32	45	9	79	12
	76	21	25	54	18	37	45
	82	17	44	39	19	49	32
		Grain	Husk	Straw	Grain	Husk	Straw
2010 / Wheat	21	49	18	33	*	*	*
	37	34	6	60	*	*	*
	65	34	4	62	7	6	87
	70	24	7	69	11	7	82
	89	11	16	73	10	15	75
2011 / Wheat	37	35‡	9‡	56‡	*	*	*
	65	68	16	16	36	28	36
	85	44	28	28	32	31	37
	89	35‡	35‡	30‡	31‡	33‡	36‡
	92	17	40	43	18	37	45

A part of this foliar and root uptake of radionuclides could not be distinguished, as this would require combining the data with a dynamic simulation model describing the dependencies of foliar and root uptake on weather, growth stage and radionuclide. The activity concentration at harvest varies depending on growth stage at deposition. Subsequent uptake of radionuclides from soil is generally lower in the first year of deposition (Rosén, 1996); however, there is little information on how much of the radionuclides are absorbed into to the straw and siliques or husks.

5.3 Foliar uptake of wet deposited ^{134}Cs and ^{85}Sr (Papers II and III)

The transfer factor (TF) values were used to estimate foliar uptake of wet-deposited ^{134}Cs and ^{85}Sr in the crops. As the amount of radioactivity applied per square meter in all treatments and on all occasions was approximately the same (*i.e.* the denominator in Equation 2 was constant), the TF values for seeds of spring oilseed rape, spring wheat (Figure 11) and ley had the same trends as the activity concentration of both radionuclides.

Conversely, the translocation factor (TLF) values for spring oilseed rape and spring wheat had a weaker correlation with the deposited activity concentrations; as TLF values are a function of the intercepted amount of radioactivity (Equation 3), which differed between the different deposition occasions. Even though the levels of each TLF value varied among the different deposition events, the pattern was similar to the TF values.

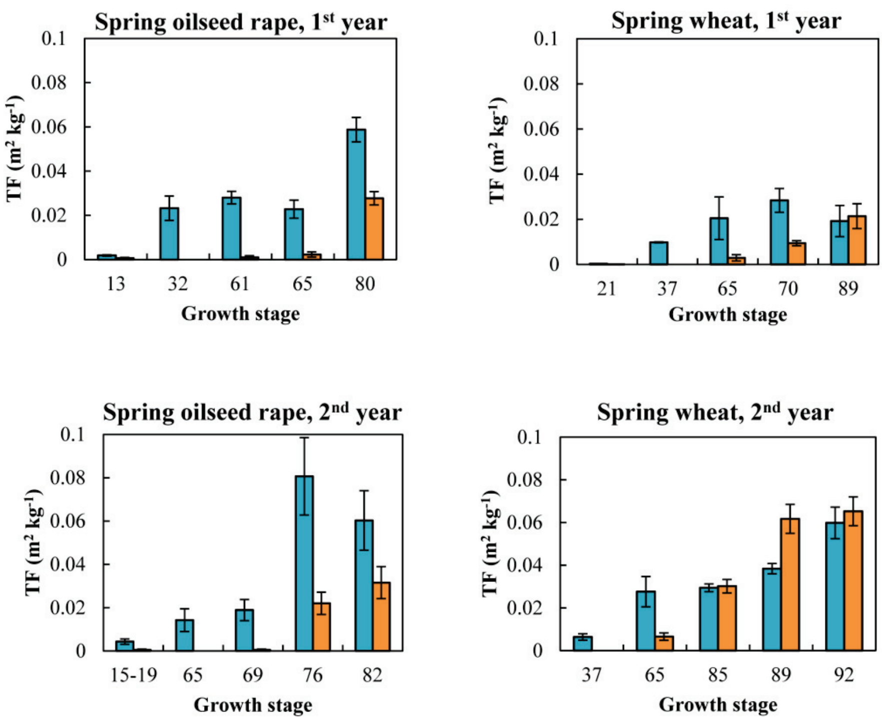

Figure 11. Average transfer factors (TF) (m^2 kg^{-1}) of ^{134}Cs ( ) and ^{85}Sr ( ) for seeds after wet deposition at five different growth stages in spring oilseed rape and spring wheat. For all growing stages, the average number of observations is $n = 3$; except spring oilseed rape (2nd year), $n = 2$ at growth stage 65 for ^{134}Cs. Error bars indicate the standard error of the mean.

The TF values tended to increase during the later deposition occasions, indicating that interception alone did not explain the activity concentrations in the seeds and ley. Other factors could have an effect, including the dilution of radionuclide concentration during biomass growth (Coughtrey & Thorne, 1983), fall-off during the time from deposition to harvest (Colle *et al.*, 2009; Eriksson *et al.*, 1998a; Eriksson *et al.*, 1998b) and/or the decay rate of the radionuclides (Choi *et al.*, 2002).

For both radionuclides, the TF and TLF values were dependent on the growth stage of the crop, the type of crop and the year, but not the type of radionuclide. The range of TF values for spring wheat and ley (information for spring oilseed rape is limited) were similar to the range found by Eriksson *et al.* (1998a; 1998b) for both radionuclides; and the TLF values for spring wheat were comparable with the findings of Vandecasteele *et al.* (2001).

Although the transfer factors for spring wheat and ley were in agreement with other studies, the variation in TF and TLF values for the two years means the use of these values for predicting possible contamination of food or fodder items in a real situation is unsuitable due to the high uncertainty. Therefore, preliminary assessments of activity concentrations in crops require continuous sampling and monitoring.

5.4 Calculated transfer of wet deposited ^{134}Cs and ^{85}Sr from ley to beef and cow's milk (Paper III)

The measured activity concentration in ley at the growth stages relevant for a normal harvest were used to calculate the transfer of ^{134}Cs and ^{85}Sr to beef and cow's milk. It was assumed that the transfer of both radionuclides from ley to beef and cow's milk would increase when deposition took place shortly before the ley harvest (Table 5). Generally, ^{134}Cs provided higher activity concentration in both beef and cow's milk than ^{85}Sr. This reversed trend to ley could be explained by caesium being more mobile than strontium in animal tissue, as strontium only bonds to bone marrow. On some occasions, the transfer of ^{134}Cs to beef exceeded the maximum permitted level of ^{134}Cs in beef inside the European Union (1250 Bq kg^{-1}) (The-Council-of-the-European-Communities, 1989; The-Council-of-the-European-Communities, 1987). As the allowed maximum permitted level of ^{134}Cs (1000 Bq kg^{-1}) and ^{85}Sr (750 Bq kg^{-1}) is lower for cow's milk than for beef (The-Council-of-the-European-Communities, 1989; The-Council-of-the-European-Communities, 1987), this meant that on some occasions, the maximum permitted levels for ^{85}Sr and ^{134}Cs in cow's milk represent the levels if no countermeasures are taken to reduce the intake of contaminated ley by livestock.

Table 5. *Estimated levels of ^{134}Cs and ^{85}Sr activity in beef (Bq kg^{-1}) and cow's milk (Bq L^{-1}) after wet deposition at different growth stages, which are relevant for normal harvests of ley, calculated with IAEA:s transfer coefficient. The mean and standard deviation of three replicates (calculation for beef and cow's milk were made for each single measurement of activity concentration of ley in the trials) (n = 3).*

year / foodstuff	Growth stage at deposition	^{134}Cs Growth stage at sampling			^{85}Sr Growth stage at sampling		
		3:3	4:5	5:6	3:3	4:5	5:6
2010 / beef	0:0	1±1	3±3	6±3	1±1	1±1	2±1
	1:1	264±281	20±2	8±3	28±25	3±2	1±1
	2:2	734±158	27±8	58±48	84±22	3±2	7±6
	3:3	2102±545†	60±24	42±14	258±80	6±2	3±1
	4:5		3128±913†	2862±1081†		337±68	317±116
	5:6			4580±1554†			572±153
		4:3	6:5	3:6	4:3	6:5	3:6
2011 / beef	4:3	3036±329†	1016±605	152±107	191±20	36±24	5±4
	6:5		3782±1153†	262±116		245±64	11±6
	6:6			1133±237			65±11
	7:7			1259±1084			70±61
	3:6			4547±1652†			346±120
		3:3	4:5	5:6	3:3	4:5	5:6
2010 / milk	0:0	0±0	1±1	1±1	1±1	1±1	2±1
	1:1	61±65	3±3	2±1	31±28	3±2	1±1
	2:2	171±37	6±2	14±11	94±24	3±2	8±7
	3:3	488±127	14±7	10±3	286±89†	6±2	4±1
	4:5		727±212	665±251		374±75†	352±128†
	5:6			1064±361†			636±170†
		4:3	6:5	3:6	4:3	6:5	3:6
2011 / milk	4:3	705±77	236±141	35±25	212±23†	40±26	6±4
	6:5		879±268	61±27		273±71†	12±7
	6:6			263±55			73±13
	7:7			292±252			77±68
	3:6			1057±384†			385±133†

† = value exceeding maximum permitted levels of ^{134}Cs (1250 Bq kg^{-1} for beef and 1000 Bq kg^{-1} for cow's milk) or ^{85}Sr (750 Bq kg^{-1} for beef and 125 Bq kg^{-1} for cow's milk) according to EU regulations (The-Council-of-the-European-Communities, 1989; The-Council-of-the-European-Communities, 1987).

5.5 Modelling the uptake and storage of ^{134}Cs and ^{85}Sr in spring wheat (Paper IV)

The linked Tracey CoupModel for radionuclide cycling was able to dynamical estimate the radionuclide balance as a function of weather conditions and growth during the two experimental years. It was possible to analyse the direct and indirect impact of weather, which is important for the correct estimation of interception and fixation (Pröhl, 2009).

5.5.1 Model performance of extended Tracey (Paper IV)

The comparison of measured and simulated Cs and Sr grains at harvest for the different deposition occasions is presented in Figure 12, with the 95% confidence interval of the measured activity of ^{134}Cs and ^{85}Sr of the grains (plus husks) and the vertical frequency distributions of the simulated TE_{Seed} plus $TE_{IntSeed,}$ at harvest.

The extended model simulated the dynamics of TE_{Seed} plus $TE_{IntSeed}$ from first to last sampling within the limits of acceptance for all scenarios (excluding those where the confidence interval could not be calculated). In total, 11% of all Cs and 10% of all Sr simulations were accepted, with 10.5% of the passive and 10.2% of the active root uptake simulations being accepted; however, more 2011-simulations (15%) were accepted than 2010-simulations (6%).

The model overestimated the storage in and on grains more than it underestimated the storage (Figure 12), which might be partly explained by the overestimation of the C content in above ground biomass, in particular that in grains. The overestimation of the grain C content might also explain the better modelling results for 2011 than for 2010. The highest number of simulations per scenario were accepted for D6 2011, 72% of Cs and 51% of Sr scenarios. This indicated the introduced module of interception functioned well; little or no foliar and root uptake took place in the two-three hours between the last deposition and harvest. The model also performed well at GS 37 (D2 in 2010 and D1 in 2011), when foliar uptake started to play a role.

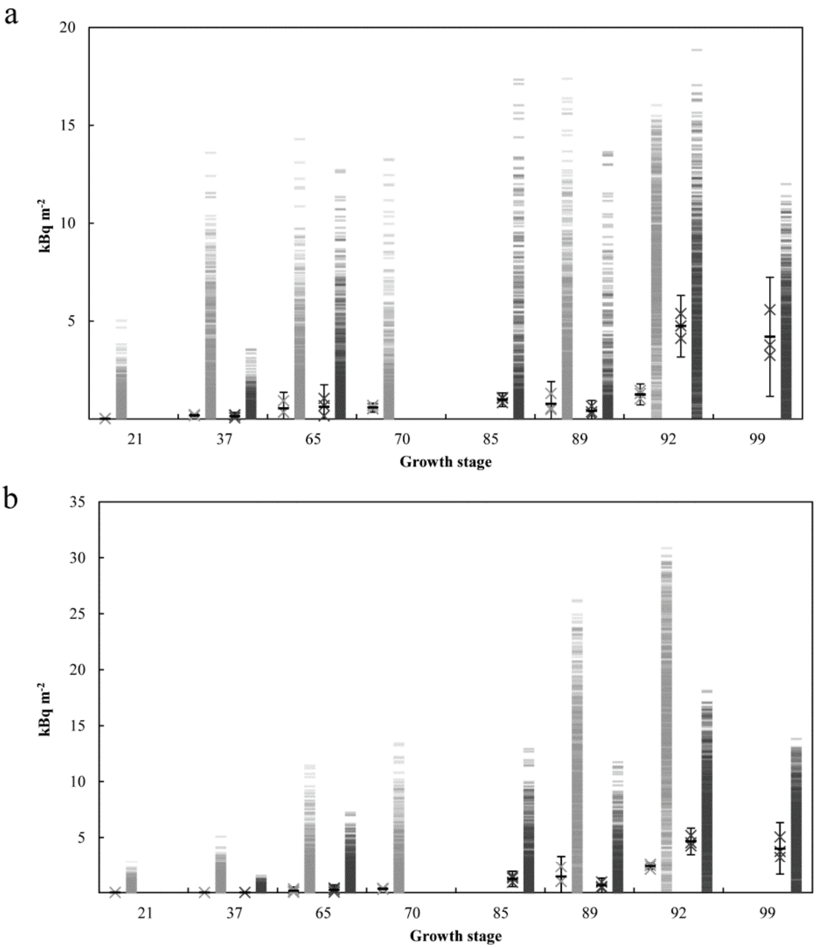

Figure 12. A comparison of measured and simulated Cs (a) and Sr (b) in and on the grains at harvest for the different deposition occasions and passive root uptake. The staples represent the simulated values of 2010 (light grey) and 2011 (dark grey); the higher the number of the simulations that is estimated for a certain activity, the darker that part of the staple is. For measured values: × denotes a replicate and the mean value with error bars of a 95% confidence interval. The growth stages (GS) at which the deposition took place is given along the x-axis to facilitate a comparison between 2010 and 2011. The deposition occasions in 2011 were somewhat delayed compared to those in 2010, for instance at GS 37, the second deposition took place in 2010 and the first deposition in 2011. For statistics of all the scenarios, see Table 3 in paper 4. The 2010 Cs' depositions contained roughly half the radioactivity of that of the Cs from 2011, and those of Sr in 2010 and 2011.

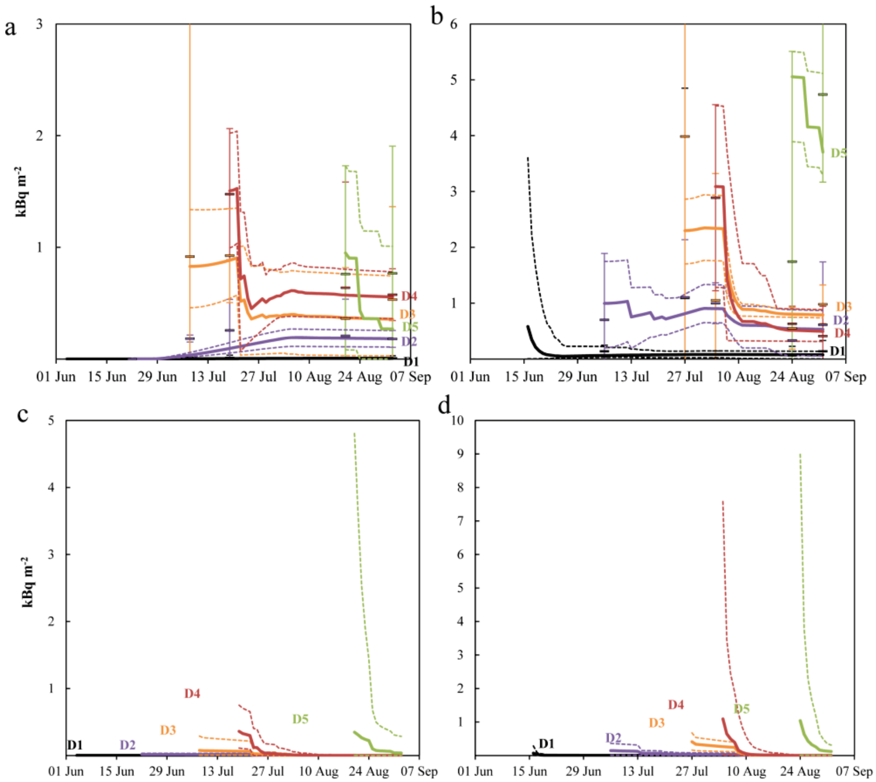

Figure 13. The means (-) of the measured and the accepted simulations (bold lines) of Cs in TE_{Seed}+$TE_{IntSeed}$ as well as the uncertainty bounds (dashed lines) (a: 2010 and b: 2011) and simulated foliar uptake of Cs (c: 2010 and d: 2011) using passive root uptake scenarios. The number of accepted simulations of grain Cs for the deposition treatments D1-D5 is given in brackets. No grains existed at the first sampling S1, and no confidence interval could be calculated at several samplings of the D1 and D2 treatment both years.

5.5.2 Simulated dynamics of the grains' storage, foliar and root uptake (Paper IV)

The temporal variation of the simulated grain storage of Cs (Figure 13ab) and Sr highlighted the increase of importance of interception after flowers are formed; from D3 in 2010 and D2 in 2011. In some later treatments, such as D3-D5 in 2010 and 2011, there was a rapid decline shortly after deposition; these declines were due to rainfall exceeding the water interception capacity, *i.e.*, the radionuclides were washed-off. Some Cs-scenarios, such as D4 in 2010 and D2 in 2011, recovered their losses, whereas, the Sr scenarios did not. The recoveries were due to reallocation of Cs with corresponding C fluxes through phloem, for instance from leaves to grains during ripening. Reallocation through the phloem was assumed to be much more pronounced for Cs than for

Sr in accordance with Strebl *et al.* (2007); Thiessen *et al.* (1999); Smolders and Merckx (1993).

The temporal variation in grains storage of Cs and Sr from the deposition treatments before flowering (D1 and D2 in 2010 and D1 in 2011) increased smoothly with increasing Cs (Figure 13ab) and Sr content in the grains. The elevated values of Sr and Cs at the beginning of D1 in 2011 were probably due to not being able to calculate the 95% confidence interval for the first sampling. The temporal variation of the simulated Cs and Sr foliar uptake was strongly related to interception of radionuclides (Figure 13cd for Cs) with high initial uncertainty. Foliar uptake totally dominated the uptake from GS 37 (stem extension) onwards (from flowering onwards for Sr in 2011), foliar uptake stood on average for 99 and 93% of total plant uptake of Cs and Sr respectively. As other plant parts are included, the sum of mean simulated accumulated foliar and root uptake can be larger than the mean simulated grain storage. According to the measurements, between 0.02-11.2% of the deposited Sr was found in (and on) the grains and husks and 0.02-20.5% in the total above ground biomass at harvest (Bengtsson *et al.*, 2013). Corresponding ranges of deposited Sr according to the model simulations were 0.03-10.1% (TE_{Seed}+$TE_{IntSeed}$) and 0.2-58.5% ($TE_{AbovegroundPlant}$+$TE_{IntAbovegroundPlant}$). Moreover, according to the model estimates, 0-0.05% of deposited Sr was stored in roots and 37.6-73.6% in the soil at harvest.

The increased knowledge from the modelling can be used for the estimation of potential doses from radionuclides in plant parts that have been used for food consumption from earlier nuclear power plant accidents depending on local weather and soil conditions, and also the plant type and its growth stage. This study can contribute to improving preparedness in the event of radioactive contamination through providing tools for retaining food security and food production.

This field experiment offers a unique data set that can be used for quantifying and analysing the importance of weather, growth stages and crop type, especially as this type of field experiment has been prohibited for a long-time.

6 Conclusions

- The highest interception of both radionuclides was at the ripening stage for both spring oilseed rape and spring wheat. For ley, the highest interception was when deposition occurred at high values of standing plant biomass and LAI. Therefore, LAI can be used for measuring the interception of both radionuclides in all three crops; whereas the standing plant biomass can only be used for measuring interception of both radionuclides in spring wheat and ley.

- The transfer of radionuclides to seeds was highest when deposition took place at growth stages close to harvest. The seeds of spring oilseed rape preferred ^{134}Cs, whereas spring wheat grain preferred ^{85}Sr. In ley, the highest transfer of the radionuclides was when the deposition took place close to the later growth stages. Therefore, the highest risk for transfer of radionuclides to humans via the food chain is when deposition occurs at the end of the growing season for spring oilseed rape and spring wheat, and on deposition at later growth stages for ley.

- The majority of radionuclide uptake by spring oilseed rape was distributed to the straw, with a smaller fraction found in the seeds. For spring wheat, a smaller fraction was directed to the husk. The amount of radionuclides did not vary systematically between the different plant parts at harvest and deposition occasions.

- The variation in magnitude of each transfer factor (TF and TLF) between different deposition occasions followed a similar pattern. A number of transfer factors for spring wheat and ley are already published; however, transfer factors relating to activity concentrations of radionuclides in seeds at harvest in growing oilseed crops are lacking. The variations between the two years (2010 and 2011) in this thesis and between earlier published transfer factors stress the need for further field and modelling experiments for increasing the understanding of the mechanisms that cause these variations.

- The calculations based on ^{134}Cs and ^{85}Sr contaminated ley transfer to beef and cow's milk exceeded the maximum permitted levels when deposition occurred at the latest growth stages, and higher activity concentrations of ^{134}Cs could be transferred to beef.

7 Future Perspectives

The focus in this thesis was on the radionuclides ^{137}Cs and ^{90}Sr, as they are important contaminants over an extended period after an accidental nuclear release to the agricultural environment. In a short-term perspective, other radionuclides, such as iodine (^{131}I) and caesium (^{134}Cs) may also be important, because they spread quickly from animal feed to milk for consumption by humans. This highlights the necessity for additional field studies on radionuclides of iodine, caesium and strontium. Extended time series are also necessary to improve understanding of the annual variations associated with crop development that affects the transfer of radionuclides to crops. Similarly, an understanding on how radioactivity concentrations in harvested parts of the crops are transferred in food processing, *e.g.* oil from rapeseed and flour from wheat grains, and through by-products from food processing, *e.g.* rape cake and husk used as animal fodder, are central for understanding the transfer of radionuclides to animal food products intended for human consumption.

Since nuclear power technology was introduced, a number of severe accidents have released radionuclides that threaten land areas used for food production. The Chernobyl nuclear power plant accident in 1986 had a great impact on food production systems in both countries in the former western part of the USSR (Smith & Beresford, 2005), and in the Nordic countries; particularly for the reindeer husbandry industry (Andersson *et al.*, 2007; Åhman & Åhman, 1994). The severe accident at the Fukushima Dai-ichi nuclear power plant in 2011 had a great impact on the areas close to the power plant and could have had a greater impact on the food production systems in Japan; as the main rice producing areas are located further north of the power plant (Statistical-Research-and-Training-Institute, 2012). These two examples strengthen the importance of obtaining a better understanding of the way radionuclide release influences food producing areas.

The increasing worldwide construction of different types of spallation sources, *e.g.* the European Spallation Source (ESS) outside Lund in the south of Sweden, has highlighted the importance of understanding other radionuclides produced in a spallation neutron source; such as radioberyllium (^{7}Be), tritium (^{3}H), radioiron (^{55}Fe) and radiosodium (^{22}Na) (Nordlinder *et al.*, 2012). The areas close to spallation sources are often used for intensive agricultural production, and there are concerns about whether a release of radionuclides from spallation sources could contaminate these areas. Therefore, understanding the colloidal transport and migration of different types of radionuclides and the behaviour of radionuclides in important agricultural crops (cereals, oilseeds, wheat and rice) is crucial.

8 Sammanfattning (Swedish Summary)

Utsläpp av radioaktiva ämnen till atmosfären från olika källor, såsom kärnkraftolyckor och nukleära explosioner, kan resultera i uppfångning och upptag av dessa radionuklider hos grödor i jordbrukets ekosystem. Dessa radionuklider t.ex. radiocesium och radiostrontium, kan överföras till livsmedel via spannmål eller djurfoder.

I doktorsavhandlingen var målet att uppskatta uppfångning och ackumulation av våt deponerat radiocesium (^{134}Cs) och radiostrontium (^{85}Sr) hos vårraps (*Brassíca napus* L.), vårvete (*Tríticum aestívum* L.) och klöver-gräsvall vid olika utvecklingsstadier i ett två årigt fältförsök. För vårraps och vårvete låg fokus på överföringen till fröna efter våtdeposition. Sambandet mellan radionukliduppfångningen och utvecklingsstadiet studerades också.

Fältförsöket etablerades i Uppsala, med markanvändning enligt normal lantbrukspraxis. Fältförsöket var upplagt som ett randomiserat blockförsök med 1×1 m^2 parceller med tre upprepningar. Under växtsäsongerna åren 2010 och 2011, deponerade en regnsimulator ^{134}Cs och ^{85}Sr vid sex olika utvecklingsstadier. Två till tre timmar efter deponering, prov togs biomassan i mitten från en yta på 25×25 cm^2 i varje parcell. Provtagen biomassa vägdes och LAI mättes. Radioaktivitetskoncentrationen och radioaktiviteten hos proven mättes med germanium (HPGe)-detektorer.

Det fanns en korrelation mellan uppfångningen av båda radionukliderna och LAI för samtliga tre grödor. Det fanns en korrelation mellan biomassan ovan jord och uppfångningen av båda radionukliderna för vårvete och för klöver-gräsvall, men inte för vårraps. Högsta uppfångningen av båda radionukliderna var vid stamskjutning för vårraps, och vid mognad för vårvete. För klöver-gräsvallen var den högsta uppfångningen vid de välutvecklade växtstadierna.

Ackumulationen av radionukliderna i de olika växtdelarna ökade när depositionen ägde rum nära skörd. Rapsfrön hade en lägre koncentration av ^{85}Sr än vetekärnor. Grödorna ackumulerade mer ^{134}Cs än ^{85}Sr. Det fanns också en indikation på att fördelningen av radionukliderna mellan växtdelarna var oberoende av hur de kom in i växten efter depositionen av radionuklider.

Variationen i överföringsfaktorerna var stor i jämförelse med tidigare studier. Detta tyder på att uppskattningen av risken för möjligt upptag av radionuklider i händelse av ett framtida nedfall under växtsäsongen är osäker.

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