Temporal and Spatial Variation of Radiocaesium in Moose (*Alces alces*) Following the Chernobyl Fallout in Sweden

Analyses of Data from Long-term Monitoring in Two Specific Areas

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Licentiate Thesis
Swedish University of Agricultural Sciences
Uppsala 2015

ISSN 1652-6880
ISBN (print version) 978-91-576-9341-9
ISBN (electronic version) 978-91-576-9342-6

Print: SLU Service/Repro, Uppsala 2015

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Abstract

In my thesis, I have analysed long-term monitoring data of ^{137}Cs activity concentrations in moose (*Alces alces*) from the two Swedish municipalities Heby and Gävle. These study areas are adjacent and dominated by managed coniferous forests. The percentage of farmland in Heby is 20% but only 4% in the Gävle area. Heby is located about 70 km inland from the Baltic coast while the study site Gävle is more close to the border of to the Baltic Sea.

The decline in the annual geometric mean ^{137}Cs concentrations in moose from 1986 to 2012 for Heby and 1986-2008 for Gävle corresponds to an effective ecological half-life of 16 years for both areas. Calves were found to have higher ^{137}Cs-concentrations than adults, on the average 14% (SD=18%) and 18% (SD=7%) in the Heby and Gävle areas, respectively. The aggregated transfer of ^{137}Cs, that describes the transfer rate from soil (activity per m^2) to moose muscle (activity per kg), seemed to be higher in Gävle (0.023 kg m^{-2}) than in Heby (0.016 kg m^{-2}).

The separate data sets were modelled by partial least squares regression (PLS) to examine the influence of environmental and physiological parameters on the caesium uptake by moose. Both data sets were supplemented with information on weather conditions prior to the hunting season, i.e. June-September. The Heby data set was more comprehensive and contained coordinates of most killing sites. That made it possible to add information of habitat types and estimated mean ground deposition around each killing site. The results of the PLS-analysis revealed at to which degree the examined parameters influenced the uptake of ^{137}Cs by moose. Apart from time since the accident and the amount of deposition, the most dominating parameters were the proportion of different habitat types around the killing site and the moose age. The model based on the more detailed Heby data set, explained the variation in ^{137}Cs concentrations in moose to a higher degree, than the model based on the Gävle data set. These results contribute to better understanding of the long-term consequences from accidental releases of ^{137}Cs on game, e.g. moose, residing in a boreal forest ecosystem.

Keywords: Radiocaesium, ^{137}Cs, Moose, *Alces alces*, Long-term Monitoring, Modelling, PLS, Chernobyl.

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Dedication

This work is dedicated to my loving family.

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 Robert N. Weimer, Synnöve Sundell-Bergman, Lars Sonesten, Camilla Wikenros & Klas Rosén. Long-term Trends of Radiocaesium Concentrations in Moose (*Alces alces*) Harvested in Sweden (manuscript).

II Robert N. Weimer, Synnöve Sundell-Bergman, Lars Sonesten, Camilla Wikenros & Klas Rosén. Modelling ^{137}Cs Concentrations in Moose (1986-2012) from Areas Highly Contaminated by the Chernobyl Fallout (manuscript).

The contribution of Robert N. Weimer (RNW) to the papers included in this thesis was as follows:

I RNW planned and designed large parts of the work, was involved in monitoring (2010-2012), performed data treatment and was the lead author for the manuscript.

II RNW planned and performed large parts of the modelling study, was involved in the design, and performed statistical analysis. RNW was the lead author for the manuscript.

Abbreviations

Bq	Becquerel, unit for nuclei disintegration per second
$^{137,134}Cs$	The radioactive isotopes of caesium
d.w.	Dry weight
f.w.	Fresh weight
GIS	Geographic Information System
PLS	Partial Least Squares Regression
Sv	Sievert, unit for equivalent radiation dose
T_{ag}	Aggregated transfer factor from soil to moose muscle
$T_{1/2}$	Physical half-life of a radionuclide
$T_{1/2\ eco}$	Ecological half-life of a radionuclide
$T_{1/2\ eff}$	Effective ecological half-life of a radionuclide

1 Introduction

Large accidental releases of radioactivity into the environment can potentially cause harmful effects on both man and biota for many decades. Considerable amounts of radioactivity can spread over long distances and cause large fallout, even thousands of kilometres away. This will eventually lead to the contamination of food products, both in agricultural ecosystems, e.g. different crops, milk, and meat, and in forest ecosystems, e.g. game species, berries and mushrooms. Lakes and freshwater ecosystems are in the short-term mostly affected by direct deposition on the surface water, but in the longer term inflow of radioactive material from the surrounding catchment areas becomes relevant (Dahlgaard, 1994). The more critical pathway in the long term for humans is via the ingestion of contaminated food products (Rantavaara, 1994).

Contaminated food products from the agricultural ecosystems are of greater concern in a shorter time span than the contamination of edible products from the forest ecosystems, but generally easier to countermeasure. This is partially because common managing actions, i.e. ploughing and fertilizing, may limit the availability for uptake by plants. Short-lived radionuclides decay within days or months, irrespective of the ecosystem but are of greater concern in parts of the agriculture, e.g. transfer of iodine-131(^{131}I) to milk (Stricht and Kirchmann, 2001). In forest ecosystems and in the long-term perspective the long-lived radionuclides, e.g. radiocaesium (^{137}Cs, half-life 30 years and ^{134}Cs, half-life 2 years) or radiostrontium (^{90}Sr, half-life 29 years) are of greatest concern (Howard et al., 2001).

The Chernobyl nuclear power plant accident on April 26, 1986, caused large radioactive fallout over many parts of Europe, e.g. Belarus, Austria, Germany and Scandinavia. The main fallout in Sweden occurred during a couple of weeks in April and May, and the deposition of ^{137}Cs locally reached levels up to 200 kBq m^{-2} (Edvarson, 1991). Other radionuclides were present in the fallout such as ^{131}I (half-life 8 days), zirconium-95 (^{95}Zr, half-life 64 days) and tellur-132 (^{132}Te, half-life 78 hours) (Edvarson, 1991). High levels of especially radiocaesium (^{134}Cs and ^{137}Cs) became a major concern for hunters, since moose (*Alces alces*) and roe deer (*Capreolus capreolus*), in the following hunting season showed high levels. Also reindeer (*Rangifer tarandus*) became highly contaminated due to the high deposition in some parts of northern Sweden.

The intervention limit for ^{137}Cs in all foodstuffs was set by the National Food Agency in Sweden (NFA), to 300 Bq kg^{-1} in 1986, but was in the following year increased to 1 500 Bq kg^{-1} for meat from game and reindeer, fish, berries and mushrooms (NFA, 1987). This new limit allowed for an annual consumption of 50 kg foodstuffs that would, in maximum, lead to an intake of 75 kBq. An annual intake of 75 kBq of ^{137}Cs corresponds to a dose of 1 mSv (ICRP, 1996). The annual background dose is in Sweden approximately 1.3 mSv per year, excluding doses from radon and medical diagnostics, (SSM, 2015).

In the east-central parts of Sweden, some 150 km north of Stockholm, i.e. in the Heby and Gävle municipalities, measurements of ^{137}Cs levels in several game species, e.g. moose, roe deer, mountain hare (*Lepus timidus*) and capercaille (*Tetrao urogallus*), started in 1986. These measurements of ^{137}Cs levels in moose meat have continued annually since 1986 in both the Heby and Gävle areas. Results from such continuous long-term monitoring of radiocaesium in large game species are unique in the scientific literature and offers good opportunity to understand the long-term behaviour of radiocaesium in the forest ecosystems.

Moose is the most important game species in Sweden and about one third (80 000-100 000) of the population is harvested annually (Lavsund et al., 2003). Adult moose weigh approximately 200-550 kg, with bulls generally being 20% larger than cows, and the average amount of meat produced per moose is around 100 kg (SJF, 2015). Radiocaesium accumulate (analogous to K) in all soft tissues and in particular muscles (Howard et al., 2001) with a biological half-life of around 30 days in moose (Nelin, 1994). However, the concentrations may fluctuate depending on food choice.

Moose exhibit seasonal variations in feeding preferences and during late summer and autumn they shift to a diet that includes plants with relatively higher caesium concentrations (Table 1), e.g. heather (*Calluna vulgaris*) and blueberries (*Vaccinium myrtillus*) (von Bothmer et al., 1990; Fawaris & Johanson, 1994; Rosén et al., 2011). Several investigations have suggested fungi as a main contributor to elevated ^{137}Cs concentrations in moose meat in autumn (Johanson et al., 1994; Palo et al., 2003; Bergman et al., 2005). This is due to higher ^{137}Cs concentrations in fungi than in plants (Karlén et al., 1991), frequently 10-100 times higher (Rosén et al., 2011) and to the abundance of mushrooms in some years, supposedly with favorable weather conditions. However, low content of fungi (< 2%) in moose rumen has been observed in some analyses (Cederlund et al., 1980; Johanson et al., 1994; Palo & Wallin 1996). Kostiainen (2007) was unable to link differences in ^{137}Cs activity concentrations in moose meat during different years to the variations in the mushroom abundance. These studies show that moose increases its intake of foodstuffs, containing relatively higher ^{137}Cs concentrations during late summer and autumn and that any correlation between intake of fungi in autumn and the concentrations of radiocaesium may be more complex than anticipated.

Table 1. *Relations between the ^{137}Cs concentrations, in Bq kg^{-1} d.w., in common plants in the moose diet in Sweden. All samples from the Harbo area.*

Species	[a,] von Bothmer et al. 1990	[b,] Fawaris & Johanson 1994	[c,] Unpublished data, ash-project 2012
Heather (*Calluna vulgaris*)	13 000	12 300	3 800
Blueberry (*Vaccinium myrtillus*)	4 100	4 700	1 200
Lingonberry (*Vaccinium vitis-idea*)	7 500	4 300	800
Pine (*Pinus sylvestris*)	2 500	3 500	1 100
Birch (*Betula* spp.)	1 200	3 200	250

a. Annual mean 1986-1988

b. July - September 1986-1989

c. Data from project with spreading of contaminated wood-ash in a forest in the Harbo area

2 Aim

The aims of this thesis were to:

- study the contamination of ^{137}Cs in meat from moose in forest and mixed ecosystems (Paper I).

- evaluate the long-term consequences of radiocaesium in species inhabiting boreal ecosystems (Paper I).

- investigate the influence of environmental (weather, habitat and ground deposition) variables and physiological characteristics (gender, age and weight) on the ^{137}Cs-concentrations in moose from two areas (Heby and Gävle) between 1986 and 2012 (Paper II).

3 Materials and Methods

3.1 Study areas

Monitoring of the concentration of radiocaesium in moose muscle has been conducted during the hunting season since 1986 in Heby and Gävle in east-central Sweden (60°28'N, 17°0'E) (Fig. 1). Although close, there are some differences between the areas and concomitantly the data sets. The Heby area where the samples have been taken is about half the size of the Gävle area, 1 000 km^2 and 1 800 km^2, respectively. Managed coniferous forests dominate both areas, but the Heby area also contains around 20% farmland while the Gävle area contains only around 4% farmland. The study areas have been without major changes during the whole sampling period, 1986-2012 (SCB, 2015). Heby is inland, located about 70 km from the Baltic Sea, while Gävle has a long coastline to the Baltic Sea. The fallout of ^{137}Cs in spring 1986 was on the average 35 kBq m^{-2} in the Heby area and on the average 50 kBq m^{-2} in the Gävle area, with an increasing gradient from south to north in Heby and a strong from west to east gradient in Gävle (Figs. 1 and 3).

The moose hunting season starts on the second Monday of October and last to the end of January the following year, in both areas. Most moose (80% in Gävle and 96% in Heby) were harvested already during the first 3-4 weeks of the hunting. At both areas hunters have collected the muscle samples from the harvested moose, normally from the upper part of the front or the rear leg. The sample, together with detailed information about each animal, has then been sent to the laboratory and analysed for the activity concentrations of ^{137}Cs. In the Heby samples ^{134}Cs was also analysed. The samples from the Heby area have been analysed at the Swedish University of Agricultural Sciences (SLU) while those from Gävle have been analysed by different municipal (Gävle) or commercial (Svelab 1996-1997 and Falma provtagning 1998-2013) laboratories. A measurement error of less than 10% has been achieved in 99% in the Heby data set and in 78% in the Gävle data set.

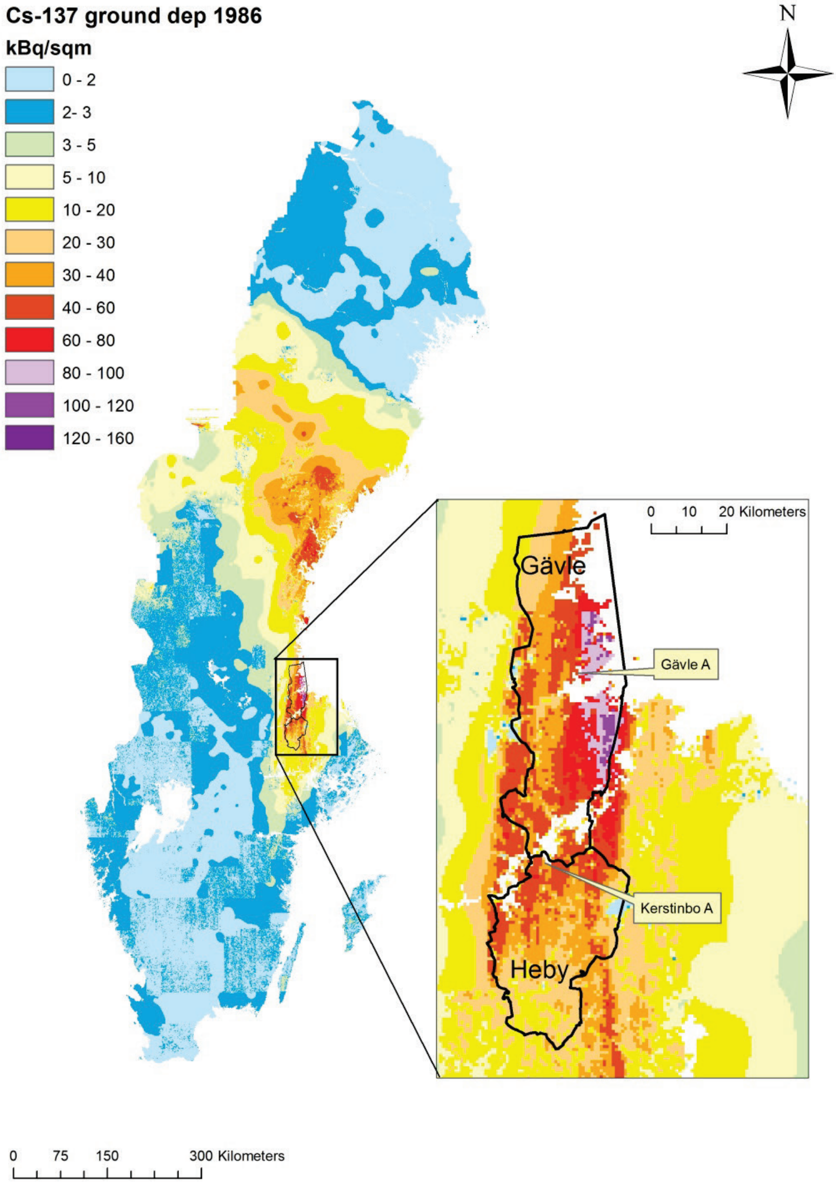

Figure 1. Ground deposition of ^{137}Cs in Sweden 1986. The two study areas of Heby and Gävle are displayed together with the two weather stations Kerstinbo A and Gävle A (adapted from fallout map supplied by the Swedish Radiation Safety Authority - SSM).

3.2 Long-term data on ^{137}Cs in moose

The monitoring for the two areas has resulted in two different data sets of ^{137}Cs concentrations for a large number of adult and juveniles from 1986-2012 for Heby and 1986-2008 for Gävle. The Heby data set contains fewer samples (3 652 specimens) than the Gävle data set (11 211 specimens). Information on hunter, hunting team, parish, animal gender, age class (calf/adult), age (for adults), weight (weighed or estimated total weight, carcass weight or weight without internal organs) and shooting date is included in the Heby data for most samples taken. In the Heby area the location of the killing site has been marked on a map, for 2 710 sites, and the coordinates have been retrieved by staff at SLU. The Gävle data set is less detailed and lacks information on gender for most of the calves, weight, and age of adults and coordinates of the killing site. The yearly number of samples from moose killed at other times than during normal hunting season, e.g. road accidents, was not used in the analyses. The number of samples coming from the Gävle area has been rather constant per year, while those from the Heby area have shown a steady decline.

3.3 Supplementary data

3.3.1 Weather conditions prior to hunting season

The influence of weather conditions during the summer, defined as from June to September, on the ^{137}Cs concentrations in moose was examined in Paper II. Weather data from two nearby weather stations (Kerstinbo A and Gävle A) were downloaded from the Swedish Meteorological and Hydrological Institute (SMHI, 2014). The data cover the years 1995-2012 for Heby and 1986-2008 for Gävle. These data were used to calculate monthly mean temperature and precipitation in the Gävle area and the Heby area (Table 2).

Including the data on temperature and precipitation in the modelling, see paragraph 3.5, was primarily aimed as an indirect indicator (proxy) of mushroom abundance in autumn, as there is no data available on actual mushroom abundance from year to year. It was expected that high precipitation in summer and the beginning of autumn would imply high amount of mushrooms in October. Many mushrooms species contain much higher concentrations of ^{137}Cs compared to plants from the same area (Rosén et al., 2011), and could, if included to some notable extent in the moose diet, contribute to the total ^{137}Cs intake by moose. A positive correlation between warm summers and mushroom production in autumn has been shown in some studies (Eveling et al., 1990; Straatsma et al., 2001) as well as a positive correlation between precipitation in summer and mushroom abundance in autumn (Eveling et al., 1990; Straatsma et al., 2001; Krebs et al., 2008).

3.3.2 ^{137}Cs ground deposition and habitat

The influence of ^{137}Cs ground deposition and habitat, around killing sites, on the ^{137}Cs concentrations in moose harvested in Heby was examined in Paper II. Data on land cover type (Fig. 2) and ^{137}Cs ground deposition (Fig. 3) in the Heby area was kindly supplied by the Swedish Geological Survey (SGU) and the Swedish Authority of Land Survey (Lantmäteriet), respectively. The data were in pixel format, 250x250 meters for land cover data and 200x200 meters for the ground deposition data. A circular theoretical home range for moose with a radius of 1.7 km was created around each moose killing site, which corresponds to an area of 9.1 km^2 that was used to calculate both the proportion of habitat types and the ^{137}Cs ground deposition within that area. A summer home range size (July – September) of 9.1 km^2 for adult females (Cederlund & Okarma, 1988) was used in a study by Nelin (1995). Mean ground deposition around individual moose killing sites was thus calculated from around 200 pixel-values. Creating circular ranges was done for sites with known geographic coordinates at a minimum distance of 1.7 km from lake and/or city (n=1 120). The proportion of land cover types around individual moose killing sites was determined by the proportion of pixel-values representing forests, wetlands, water surfaces and farmland (n=2 710).

The circular theoretical home ranges (called buffer zones in the GIS software ArcMAP) overlapped in most cases. ArcMAP (ESRI 10.2 for Desktop) cannot calculate data in overlapping so called buffer zones as default. This was solved by creating a basic model using the Model Builder-tool in ArcMAP, including the tool Zonal statistics that calculate the mean, maximum and minimum values within each home range. All killing sites in relation to land cover and ^{137}Cs ground deposition in the Heby area are displayed in Fig. 2 and Fig. 3 respectively.

3.4 Long-term trend of ^{137}Cs in moose (Paper I)

The two data sets were used to study the long-term behaviour of ^{137}Cs in moose from the two areas. Annual mean, maximum and minimum ^{137}Cs concentrations in moose were calculated. The effective ecological half-life of ^{137}Cs in moose muscle was calculated by the equation for the slope of the annual mean values. Since the number of samples varies between the years the variation in ^{137}Cs concentrations was expressed as the coefficient of variation (CV), which is the standard deviation through the mean (Box et al., 2005).

Activity concentrations of ^{134}Cs were available in the Heby data set and thus the effective half-life could also be determined for this radionuclide in the same manner. The activity concentrations of ^{137}Cs and ^{134}Cs in moose were non-normally distributed and hence geometric means were estimated.

The relation between the ground surface deposition of ^{137}Cs and the concentration in moose muscle can be described by the aggregated transfer factor (T_{ag}), which was calculated as:

$$T_{ag} = \frac{\text{activity concentration in meat, Bq kg}^{-1}\text{(f.w.)}}{^{137}Cs \text{ ground deposition, Bq m}^{-2}}$$

The T_{ag}-values for moose in the Heby were, for comparison, calculated both by using individual data, on ^{137}Cs concentration and ground deposition, and by using annual mean concentrations and mean ground deposition for the whole municipality. The T_{ag}-values for moose in the Gävle area were calculated using annual means of ^{137}Cs concentrations and the mean ground deposition for the whole municipality. In Heby the deposition around individual killing sites was determined by GIS, and thus could T_{ag}-values be calculated on an individual level.

The influence of age (i.e. to investigate if there was any difference between calves and adults) and gender (for adults) on the ^{137}Cs concentrations in moose was examined. The difference in T_{ag}-values between the two study areas was also examined. The groups were compared by using ANOVA GLM in Minitab (Minitab® ver. 17.2.1).

3.5 Influence of environmental factors on ^{137}Cs variation (Paper II)

The data sets were analysed with partial least squares regression (PLS) in order to examine the influence of all current, and additional, parameters in the data sets on the variation of ^{137}Cs in moose among and within years. Partial least square regression examines the correlation between a number of explanatory variables (X) and one or more response variables (Y), in this case ^{137}Cs concentration, and is especially suited to use on multi-collinear data. Both data sets were supplemented with data on weather conditions, i.e. precipitation and temperature, in the four months prior to the hunting season. The Heby data set was also supplemented with data on ground deposition (n=1 120) and on land cover types (n=2 710) around individual killing sites. Thus, the final data set for Heby contained more explanatory variables in the model (Table 2) than the Gävle model. The models were pruned by stepwise removal of non-significant explanatory variables (95% confidence interval). PLS-models also give, apart from defining the statistically significant environmental variables, values for predictability and explanatory power of the models (Paper II). The analyses were performed in SIMCA (ver. 13.0 by MKS Umetrics AB) that can work well with random missing data, ≤ 20%, in the data set (Eriksson et al., 2013).

The geographical distribution of the ^{137}Cs deposition does not change over time, and the proportions of different land cover types remained constant in the study areas (SCB, 2015). The models for the two areas are not completely inter-comparable since the underlying data sets are not equivalent.

Table 2. *Abbreviations and values of explanatory variables used to model the influence on ^{137}Cs concentrations in moose, during hunting season, from Heby municipality in east-central Sweden, 1986 – 2012. Data on precipitation and temperature in 1986-1994 and in June-July 1995 was not available. Percentage of available data within parenthesis.*

Variable	Unit	Mean	Min	Max	Description
Cat					Calf, born same year, or Adult (99%)
Age	years	1.3	0	15	Age, estimated by hunter. Calf = 0 years (93%)
Sex					Female or male (91%)
Weight	kg	117	24	270	Carcass weight, estimated or weighed by hunter (38%)
Season					Year of hunting[a] (100%)
Week					Week of hunting during hunting season (100%)
Month					Month of hunting during hunting season (100%)
Tot P. 6-9	mm	260	178	364	Total precipitation June through September
Tot P. 7-9	mm	197	133	244	Total precipitation July through September
Pres. 6	mm	62	7	137	Total precipitation in June
Pres. 7	mm	84	38	204	Total precipitation in July
Pres. 8	mm	65	15	163	Total precipitation in August
Pres. 9	mm	50	15	94	Total precipitation in September
Av T 6-9	°C	14.4	12.9	16.4	Average temperature June through September

Av T 7-9	°C	14.5	12.8	16.6	Average temperature July through September
Temp. 6	°C	14.2	12.2	16.3	Average temperature in June
Temp. 7	°C	16.7	14.4	19.4	Average temperature in July
Temp. 8	°C	15.7	13.2	18.6	Average temperature in August
Temp. 9	°C	11.0	8.7	13.5	Average temperature in September
Deposition	kBq/m^2	27.4	11.1	52.2	Mean ^{137}Cs ground deposition around individual moose, radius 1.7 km, at shooting date (31%)
% forest	%	77.7	13.4	100	Proportion of forest cover[b] (74%)
% open	%	15.5	0	84.2	Proportion of open area[b] (74%)
% mire	%	4.6	0	38.1	Proportion of wetlands [b] (74%)
% water	%	2.2	0	65.3	Proportion of water surface[b] (74%)

a. Including January the following year.

b. Proportion between forest, open area, mire and water surface. Areas representing cities/villages excluded.

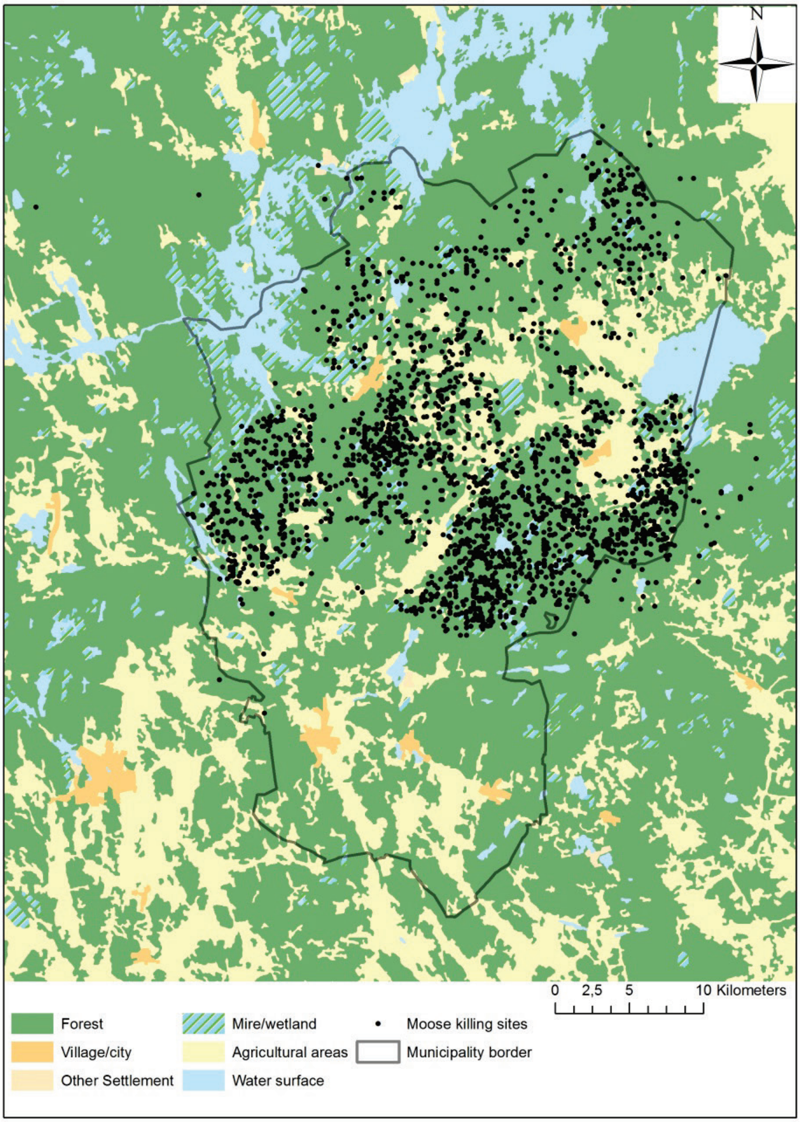

Figure 2. Land cover types in and around Heby municipality (black line) and killing sites of moose (n=2 710) harvested during 1986-2012. Land cover data supplied by the Swedish Authority of Land Survey (Lantmäteriet). Pixel size 250x250 m.

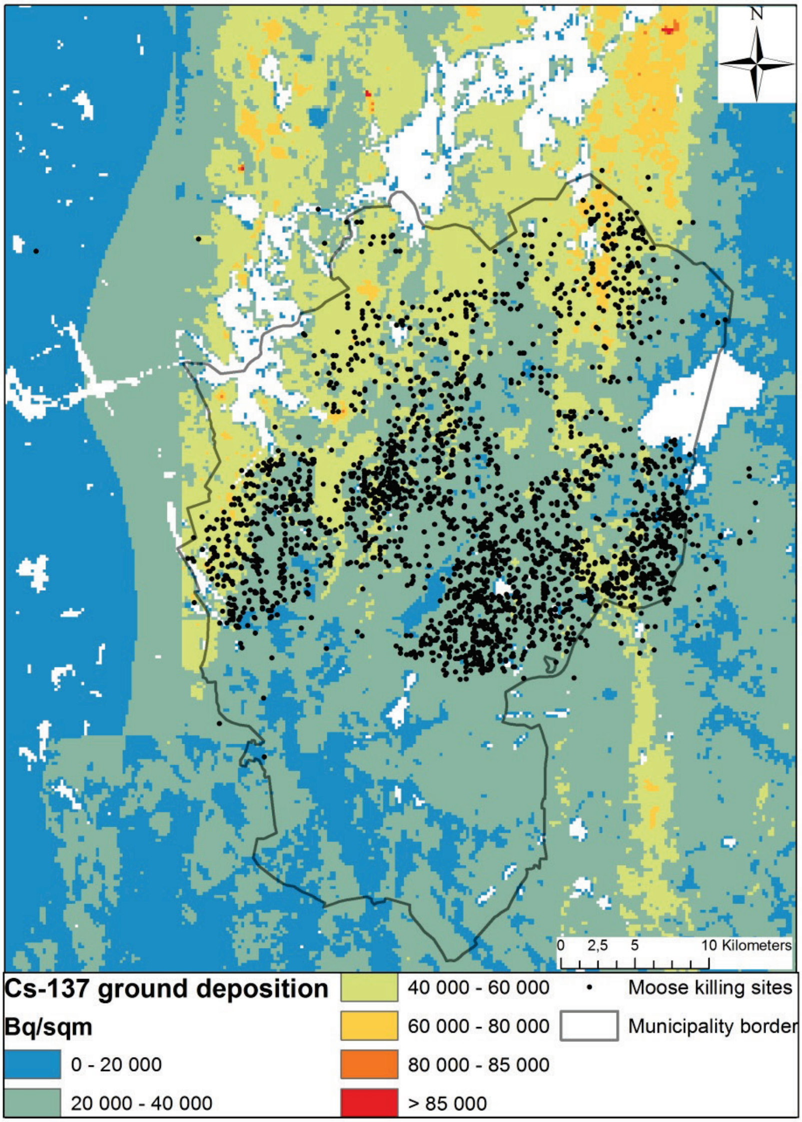

Figure 3. Ground deposition of ^{137}Cs in and around Heby municipality (black line) in 1986 and killing sites of moose (n=2 710) harvested during 1986-2012. The ground deposition decreases with time, but with the same relative distribution. Deposition data supplied by the Swedish Geological Survey (SGU). Pixel size 200x200 m.

4 Results

4.1 ^{137}Cs in moose in the long term perspective (Paper I, II)

During the sampling period from 1986 to 2012 for Heby respectively 1986 to 2008 for the Gävle area the $T_{1/2eff}$ was found to be 16 years. However if including data only up to 2008 in the Heby dataset the calculated half-life became longer or 19 years (Paper I). As expected the impact of time since the accident and the corresponding fallout was identified as important factors. Accordingly, in the PLS-models these parameters showed the strongest correlation to ^{137}Cs concentrations in moose (Fig. 5).

The $T_{1/2eff}$ of ^{134}Cs in moose from Heby was calculated to be 3 years. This is slightly longer than the $T_{1/2phys}$ of 2.1 years. In Fig. 4 the time-trends of the annual mean activity concentrations of ^{137}Cs and ^{134}Cs are shown.

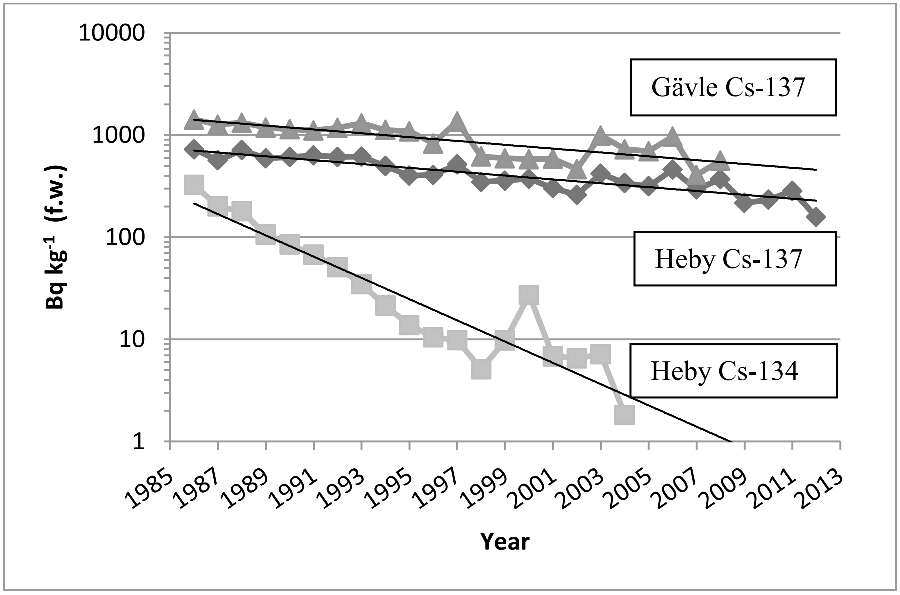

Figure 4. Annual geometric means of ^{137}Cs and ^{134}Cs in muscle from moose harvested in the Heby (1986-2012) and Gävle (1986-2008) areas during hunting season. Note that for the Gävle area there was no data on ^{134}Cs available.

4.2 ^{137}Cs concentrations in relation to age and sex of moose (Paper I,II)

A significant difference in the ^{137}Cs concentrations between adults and calves was found (see Paper I). Calves from Heby had 14% (SD=18%) higher concentrations compared to adults (DF=1, f=7.65, p=0.008). The corresponding value for Gävle was 18% (SD=7%) (DF=1, f=5.58, p=0.023). Age showed a significant negative correlation with the ^{137}Cs concentrations in the Heby model (see Paper II) (Fig. 5). In the Gävle model the age category also correlated with the ^{137}Cs concentrations (see Paper II). The negative correlation between ^{137}Cs concentration and the age of the animal seems therefore to be important for all age groups. No significant impact of gender on the long-term concentrations of ^{137}Cs in moose was found (see Paper I). The parameter for gender was not significant in either PLS-model, and thus excluded from the final models (see Paper II). The parameter weight was also excluded from the final model on the Heby data set since it was found to be insignificant.

4.3 ^{137}Cs concentration in relation to habitat and ground deposition (Paper I, II)

A difference in the aggregated transfer factor (T_{ag}) was found for the areas (see Paper I). Significantly lower T_{ag}-values were found for the Heby data set compared to the Gävle data set (DF=1, f=16.11, p=0.000). The average T_{ag} for moose from the Heby area was estimated to be 0.016 kg m^{-2} (range 0.010 – 0.036) and 0.023 kg m^{-2} (range 0.013 – 0.035) for moose from the Gävle area. A decreasing trend in T_{ag}-values of ^{137}Cs was found for both areas over time (DF=1, f=10.05, p=0.003)

In both models the regression coefficients for the variables describing deposition and the time since deposition showed the strongest correlation with ^{137}Cs concentrations in moose (see Paper II). In the Heby model the variables representing habitat, the percentage of open land (negatively) and forest (positively) showed the strongest correlation the ^{137}Cs concentrations in moose (Fig. 5). The proportions of water and wetland were also significant and both positively correlated to the ^{137}Cs concentrations. In the Gävle data set the variables describing the approximate location, northward and eastward, of the killing site showed a positive correlation to the ^{137}Cs in moose (see Paper II).

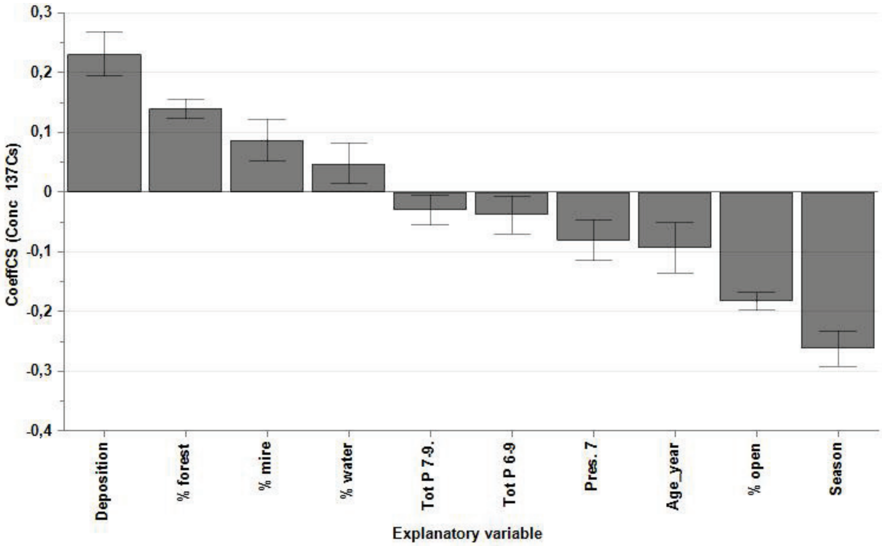

Figure 5. Strength and direction of the regression coefficients between auto-scaled explanatory variables and ^{137}Cs concentrations in muscle from moose harvested in Heby municipality in east-central Sweden during 1986-2012. PLS-analysis with two components, error bars indicate 95% confidence intervals. For abbreviations see Table 2.

4.4 ^{137}Cs concentration in relation to summer weather conditions (Paper II)

Several weather parameters were statistically significant and included in the final PLS-models. In the Heby model the only variables describing precipitation were significant (precipitation in July and total precipitation in June-September and July-September) (Fig. 5). All of the weather variables showed a negative correlation with ^{137}Cs concentrations in moose (Fig. 5). In the Gävle model both temperature and precipitation variables were significant. Temperature in July and precipitation in August showed a positive correlation to ^{137}Cs concentrations in moose, while temperature in June and September and average precipitation as well as precipitation in June and July showed a negative correlation (See Paper II).

4.5 PLS-models (Paper II)

Both PLS-models contained two significant PLS components, with both a predictive behavior and degree of explanation of the variation in the ^{137}Cs concentration in moose, at 38% for the Heby model (R^2-Y=0.38 and Q^2 = 0.375), and 22% for the Gävle model (R^2-Y=0.219 and Q^2 = 0.218). Permutation tests confirmed the validity of the models, i.e. low background correlation in the environmental data, with both R^2 and Q^2 clearly separated from permutated models.

5 Discussion

The initial deposition of ^{137}Cs sets the baseline for the mean ^{137}Cs activity concentrations in moose in an area over time. However, biological, ecological and meteorological factors might subsequently influence the bioavailability and thus contamination of ^{137}Cs in moose. All together this has implications for hunters that hunt moose, as they and their families probably will consume most of the meat.

5.1 Long-term trend of ^{137}Cs in moose (Paper I)

The observed high variability of ^{137}Cs concentrations in moose among years has a high inter- and intra-annual variation in both data sets, that even seem to increase at the end of the monitoring period in the Heby data set. The intra-annual variation might be due to a decreasing number of samples from the Heby area over time. The number of samples from the Gävle area has been rather constant over time with a stable variation (CV) of 53% (SD=4%). There was a strong correlation between the annual mean concentrations from the two areas, which indicates that common factor(s), e.g. summer precipitation and temperature, were influencing the among year variation (Palo et al., 2003).

Several studies have suggested that high inter-annual variation in ^{137}Cs concentrations in moose depends on the variability of the habitat (Johanson & Bergström, 1989; Nelin, 1995; Kostiainen, 2007) and increased consumption of mushrooms during years with relatively high mushroom abundance (Bergman et al., 2005; Johanson et al., 1994; Palo et al. 2003). However, weather conditions could very well affect the net intake of ^{137}Cs by moose through other mechanisms, beside the abundance of mushrooms in autumn, such as the proportion of different plants included in the moose diet (Bø & Hjeljford, 1991), although the information on this is scarce.

The effective ecological half-lives of ^{137}Cs in moose of 16 years for the two study areas are among the longest found in the deer family (*Cervidae* spp.) in Europe. Effective ecological half-lives ranging from eight to twelve years have been found for roe-deer in Germany and Poland (Pröhl et al., 2006; Strebl and Tataruch, 2007; Kapala et al., 2015) and for red deer (*Cervus elaphus*) in Poland (Kapala et al., 2015). From a dose assessment perspective, however, this suggests that mitigation strategies (e.g. time of hunting season) and recommendations on intake might have to be different, depending on game species, from a short, first 5-10 years, to a longer, > 10 years, perspective. Even in the long-term perspective the high variability in ^{137}Cs concentrations contributes to the uncertainties. This is illustrated by analysing the Heby data set only up to 2008 (instead of up to 2012), which modifies the estimated effective ecological half-life from 16 years to 19 years. No information on effective ecological half-lives of ^{134}Cs in moose has been found, but this study suggests that this is primarily governed by the physical half-life of ^{134}Cs. It is shown that already five years after the fallout the ^{134}Cs levels in moose were less than 20% of the levels of ^{137}Cs and rapidly declining (Fig. 4).

T_{ag}-values for moose, i.e. the relation between ^{137}Cs ground deposition per m^2 and the activity concentration per kg in moose meat, harvested in the Gävle area were higher, 44% on total means, than for those harvested in the Heby area during 1986 – 2008 (DF=1, f=16.11 $p<0.001$). Even in comparison with T_{ag}-values reporting values in the range of 0.010 – 0.016 for adults and 0.013 – 0.020 for calves (IAEA, 1996) the T_{ag}-values in the Gävle area were comparatively high. The Gävle municipality has a long coast-line along the Baltic Sea, and likely this proximity to the sea might influence the transfer of ^{137}Cs to moose. Higher T_{ag}-values in a coastal region have also been reported by Palo et al. (2003), who suggested that this was due to an earlier onset of winter and thus to winter diet that is lower in ^{137}Cs content than the autumn diet.

The T_{ag}-values for Heby-moose, based on deposition data around the individual killing sites, did not differ from the values based on a mean deposition for the whole study area. Considering that the variation in deposition within the Gävle area is larger than that for the Heby area it would have been of interest to examine if individual T_{ag}-values would differ from the means of the whole area. However, the Gävle data set did not contain locations for individual killing sites, and thus such an analysis was not possible to do.

The difference in activity concentrations between calves and adults (14%, (SD=18%) and 18% (SD=7%) higher in calves from Heby respectively Gävle), was seen for both study areas. Taking into account the large number of samples and the length of the data set, this difference between adults and calves is representative, for moose hunted in these types of ecosystems. Larger differences between concentrations in adults and calves, around 40%, have been found further north in Sweden (Danell et al., 1989; Palo et al., 1991). Probably, this is partly explained by ecosystem differences (e.g. plant species composition and soil types) and an earlier start of the hunting season. In the northern parts of Sweden the hunting season starts already in September. Consuming meat from calves, coming from the Heby and Gävle area, could thus pose a somewhat higher risk compared to consuming meat from adults.

The prevailing intervention limit for ^{137}Cs set by the Swedish National Food Agency is 1 500 Bq kg^{-1} for game meat up for sale on the market, and it is also advised to not regularly consume meat exceeding that level (NFA, 2015). Numerous samples (in total 25% and on average 8% during 2006-2008) of moose harvested in the Gävle area exceeded this limit, which mainly is attributed to the comparatively larger deposition in this area. However, also a higher transfer of ^{137}Cs from soil to muscle in the Gävle area than in the adjacent Heby area may have contributed to this observation.

5.2 Variation of ^{137}Cs in moose (Paper II)

The results for the Heby model showed that habitat type, especially the proportions of forest and farmland, as well as the deposition around the killing site and the number of years since fallout are key descriptors of the long-term spatial and temporal variation of ^{137}Cs activity concentrations in moose.

A weak correlation between ^{137}Cs ground deposition and measured activity concentration in moose meat was found by Palo et al. (2003), who suggested that this depended on variability in radiocaesium uptake due to habitat shifts by moose among years. Nevertheless our results show that the initial deposition still is among the most important variables for explaining the variation of ^{137}Cs in moose meat.

This study further stresses that the fallout magnitude can be of less importance if low contaminated fodder, i.e. different kinds of crops, is available to moose before the hunting season. Radiocaesium concentrations in agricultural plants have an ecological half-life of 1-2 years, and rapidly decline during the first years after fallout (IAEA, 2010). Rosén et al., (1996) also found low transfer rates to grass and grain on fertilized and ploughed soils. My study suggests that the extension of agricultural areas in the feeding habitat of moose to a large extent determines the uptake of radiocaesium in moose since it was the second most important parameter that negatively correlated with ^{137}Cs concentrations in moose from the Heby area. This is supported by studies that show lower ^{137}Cs levels in moose harvested close to agricultural areas than those in forests (Johanson & Bergström 1989; Nelin, 1995; Kostiainen 2007).

In the model based on the Heby data wetlands and water surface areas contributed to a smaller extent to the ^{137}Cs uptake by moose. Nevertheless wetlands and water had some importance in the model. The proportion of wetlands was expected to be important in the model as Nelin (1995) found highest ^{137}Cs levels in moose harvested in areas with a large proportion of mires, further north in Sweden. Likely the proportion of wetlands in the Heby area is too low (mean < 5%) to have a significant impact, especially in proportion to the amount of farmland (around 20%). The proportion of water surface areas, i.e. lakes and streams, could be of intermediate importance since moose preferentially feed on aquatic plants in summer to meet their need for sodium (Belovsky & Jordan, 1981), where for example water lily (*Nymphaea* spp.) and water horsetail (*Equisetum fluviatile*) can contain relatively high ^{137}Cs concentrations (Johanson et al., 1994; Nelin & Nylén, 1994). However, also the distribution of salt licks in spring and summer, which is a common practice by hunters, may contribute as salt licks in forests could decrease the need by moose to feed on aquatic plants to satisfy their need for sodium and hence reduce exposure to higher radiocaesium levels from consumption of aquatic plants.

The fraction of radiocaesium available to moose decreases with time due to physical and ecological decay, and consequently the variable describing years since deposition shows the strongest negative correlation to ^{137}Cs concentration in moose in both models. This confirms with effective ecological half-lives of ^{137}Cs at 16 years in moose from the Heby and the Gävle areas (Paper I).

PLS-models indicate that younger moose have a higher net uptake of radiocaesium than older moose, since the parameters representing the age of the moose were significant in both models. However, in the Gävle model moose age was only categorized as adult or calf, while in the Heby model the estimated age of the moose was used. Lower radiocaesium concentrations in meat from adults than in meat from calves have also been reported in other studies (Danell et al., 1989; Kostiainen, 2007; Palo et al., 1991). Interestingly, the results in the Heby model indicate that radiocaesium concentrations continue to decrease with age even during adulthood.

Weather variables such as temperature and precipitation were included in both models. These variables were used as an indirect link between mushroom abundance in autumn and ^{137}Cs concentrations in moose in autumn. In the Heby model three weather variables (total precipitation June-September and July-September as well as precipitation in July), all describing precipitation, remained significant and were solely having a negative correlation to ^{137}Cs

concentrations. In the Gävle model six weather variables were significant, with five (temperature June and September and precipitation June and July as well as average precipitation June-September) positively correlating and two (temperature in July and precipitation in August) negatively correlating to ^{137}Cs concentration in moose. It is unclear if the mechanism is through the effect of weather on mushroom abundance or via some other effect on ^{137}Cs levels in fodder plants, for example habitat choice by moose or by fodder quality within a habitat. The various weather variables in the respective month could correspond to either higher or lower fungal sporocarp production, respectively, in autumn. However, the influence of weather seems more profound in the Gävle area where more of the weather variables were significant in the PLS. A possible explanation for this could be the higher number of samples (about 4 times) in the Gävle model than in the Heby model, and that weather data was available for all years. With less access to uncontaminated food for moose in Gävle (4% farmland) than in Heby (20% farmland), access to mushrooms, even at small amounts, would relatively have a larger effect on the total ^{137}Cs uptake. This subsequently suggests that fungi may be of importance in particular to forest-residing moose, i.e. those residing far away from agricultural areas. It has been observed that the fungal production during one year is also dependent on the previous year's production irrespective weather conditions (Lange, 1978; Krebs et al., 2008). This suggest that simultaneous monitoring of mushroom abundance is also needed in order to elucidate to what extent fungi actually contribute to the variation in ^{137}Cs concentrations in moose.

Time-series of radionuclide concentrations in environmental samples have been of great value for determining the extent and spread of contamination following a nuclear accident in the long term. This study shows that empirical data from long time-series provide a valuable source of information for evaluation even if the sampling originally was not designed for scientific purposes. However, our study shows that precise geographical information of killing sites greatly improved the model, as this was used to determine both the deposition and the habitat type around the killing sites of individual moose. This lack of precise geographical information in the Gävle data set is apparent when comparing the explanatory power of the two models, with the Heby model explaining about 38% and the Gävle model explaining about 22% of the variation in ^{137}Cs concentrations. No site-specific variables, such as the actual coordinates that were used as variable in the Gävle model, were included in the model based on data from Heby, which means that the conclusions drawn from the Heby model, concerning the influence of environmental and physiological parameters on radiocaesium in moose, could be valid for other similar ecosystems in the boreal environment.

This investigation is unique through its long-term follow up of monitoring ^{137}Cs activity concentrations in moose. It should be recognized that the samples have been collected in the same area, by the almost same hunting team and same time every year. The extensive amount of data also provides an opportunity to evaluate the relevance of the current recommendations issued by the national authorities. My study also provides information on how long-term monitoring following accidental releases may be designed in order to provide valuable information for researchers as well as the management teams.

6 Conclusions

In a forest dominated ecosystem, intermixed with agricultural areas, the effective ecological half-life of ^{137}Cs in moose is close to two decades. Thus, the consequences will remain for generations for people that regularly consume game meat, and other forest food products, following large releases and depositions of long-lived radionuclides.

The difference in ^{137}Cs concentrations between adults and calves remained constant throughout the whole study period, which was 22 years for Gävle and 26 years for Heby. No sex-specific differences in ^{137}Cs concentrations in adults were found. In my study the transfer of ^{137}Cs from soil, though based on aerial measurements, to moose was higher in the Gävle area compared to in the Heby area.

Modelling of the behavior of radiocaesium, deposited after a single fallout event, in a large wild herbivore, e.g. moose, in forest habitats is a challenging task. However, the PLS-models identified several parameters as having an influence on the ^{137}Cs concentrations in moose. Of particular interest was the continuing influence on ^{137}Cs uptake by age even in adulthood and the positive correlation between water surfaces, around the killing site, and the ^{137}Cs concentrations in moose.

Additionally, it was found in the PLS-models that the weather conditions prior to the hunting season have an influence on the ^{137}Cs-uptake by moose. The mechanism behind the influence of weather conditions remains complex and should be further investigated.

7 Future perspectives

The knowledge of the behavior of radiocaesium in boreal forest ecosystem is still limited though thirty years have passed since the Chernobyl accident contaminated the Swedish environment. In order to gain better knowledge on the factors that affect the high variability of ^{137}Cs in moose, one solution could be to attach GPS collars to moose in the monitoring areas as a way towards better assessment of their habitat utilization. Further modelling work, to unravel the observed high variation of ^{137}Cs concentrations in moose, probably could benefit from including more variables such as forestry data on clear cuttings and actions (fertilization) as a part of habitat variability within forests.

The selection of food by moose in the months prior to the hunting season will have a high influence as to which degree the moose ingests the available ^{137}Cs from the surrounding environment. Subsequently, increasing the knowledge of the food choice by moose (plants, herbs and mushrooms), and the activity concentrations therein is crucial. The fractions of mushrooms in the moose diet as well as which factors that influences the variation in fodder quality and ^{137}Cs content from year to year need to be further investigated, as the diet is a key component to the ^{137}Cs uptake by moose.

Any other indicators of increased ^{137}Cs turnover in the forest ecosystems, such as outflow of ^{137}Cs from forests to streams and rivers related to variations in annual weather conditions, should also be investigated.

From a risk management perspective an examination of the influence of variation in game species populations over time would be needed. These variations could manifest themselves by different effective half-lives among different species and hence the consumption of meat from different animals by humans. More focus should also be put on research activities that study the possibilities to countermeasure the transfer of ^{137}Cs from game to humans. Some early experimental studies in Sweden indicated that by distributing saltlicks, containing Prussian blue which inhibits the gastrointestinal uptake of ^{137}Cs, in forests the ^{137}Cs concentrations in moose could be reduced.

The results of this study may be supportive in the long-term evaluation of the consequences of the fallout following the Fukushima nuclear power plant accident in Japan in 2011, where the deposition on land mainly occurred in the forest ecosystems.

8 Sammanfattning (Swedish Summary)

I denna licentiatavhandling har jag analyserat långa tidsserier med data innehållandes uppgifter om koncentrationer av radiocesium (^{137}Cs och ^{134}Cs) i älgkött. Datat i tidsserierna härrör från den miljöövervakning av radiocesium som påbörjades 1986 i Heby och Gävle kommun och täcker för Heby in data fram till 2012 och för Gävle fram till 2008. Studieområdena gränsar till varandra och består huvudsakligen av produktionsskog av barrträd, med cirka 20% jordbruksmark i Heby jämfört med cirka 4% i Gävle. Gävleområdet har en lång kust mot Östersjön, medan Heby ligger ca: 70 km från kusten.

Den långsiktigt nedåtgående trenden av ^{137}Cs i älgkött beräknades motsvara en effektiv ekologisk halveringstid, $T_{1/2eff}$, på motsvarande 16 år. Kalvkött från Hebyområdet innehöll i genomsnitt 14% (SD=18%) högre ^{137}Cs koncentrationer än kött från vuxna djur, medan kalvkött från Gävleområdet i genomsnitt innehöll 18% (SD=7%) högre koncentrationer än kött från vuxna djur. Den beräkande aggregerade överföringsfaktorn, T_{ag}, som beskriver ^{137}Cs koncentration i mark per m^2 jämfört med ^{137}Cs koncentration per kg älgkött var högre i Gävle (0.023 kg m^{-2}) än i Hebyområdet (0.016 kg m^{-2}).

Till de två dataseten med information från studieområdena lades ytterligare externa data till innan de analyserades separat med Partial Least Squares regression (PLS). Båda dataseten kompletterades med information om temperatur och nederbördsmängder under de fyra månaderna, juni-september, innan jaktsäsongen startar. I Hebydatat fanns desutom koordinater angivna för varje skottplats och med hjälp av det kunde även data för habitat och genomsnittlig markdeposition runt varje skottplatts räknas fram och läggas till. Resultaten av PLS-analysen indikerar att, förutom tid sedan nedfallet och nedfallets storlek, påverkades ^{137}Cs koncntrationerna i älg mest av i vilket habitat älgen sköts i samt vilken ålder älgen uppnått. Modellen baserat på Hebydatat innehöll fler variabler och hade en högre förklaringsgrad än modellen baserat på Gävledatat.

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Acknowledgements

My work was financed by the Swedish Authority for Radiation Safety (SSM) and the Swedish University of Agricultural Sciences (SLU) for which I'm very grateful.

I also wish to extend my gratitude to:

Willem Goedkoop who kindly assigned as main supervisor and for kindly admitting me as a Licentiate-student at the Department of Aquatic Sciences and Assessment.

My assistant supervisors and also co-authors, Synnöve, Camilla, Lars and especially Klas who got the funding from SSM.

Former colleagues at the Radioecology group: José Luis Guiterrez Villanueva, Yuri Chaiko, Juri Mandro (Ukraine) and Mykhailo Vinichuk.

Rolf Bergman for valuable input on the PLS-modelling.

Anders Dahlberg for valuable information on mushrooms and their relation to weather conditions and for kindly supplying with references.

All hunters that during all years faithfully collected the muscle samples from moose.

All "old" colleagues and the Department for Soil and Environment as well as all "new" colleagues at the Department of Aquatic Sciences and Assessment.

Everybody in the "Ultimate Frisbee" group for helping me to work hard on the field and to forget about writing and science for moment.

My parents, Margaretha and Christer, for being proud of me.

My wife Jenny and my two children, Elsa and Nicholas, for putting up with a very absent-minded husband and father.