The impact of climate change on radiocesium mobility

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Summary

We investigate variations in ^{137}Cs activities, temperature and rainfall with time and predict how climate change will interact with existing ^{137}Cs anomalies. We focus on several case-studies affected by distal fallout from Chernobyl; the Kymijoki watershed in Finland, Gävle in Sweden and Bavaria in Southeast Germany. In addition, we investigate ^{137}Cs activities in Japan as a contrasting location proximal to the Fukushima nuclear power plant that was damaged during the tsunami in 2011.

In Europe we find that ^{137}Cs anomalies show up in moose, mushrooms and ground radiation levels in specific years that differ from place to place. There is little indication of direct relationships between weather conditions and ^{137}Cs anomalies. However, environmental processes play critical roles in radionuclide behavior and may themselves be influenced by weather conditions and furthermore by changing climate. Erosion likely remobilizes subsurface ^{137}Cs and transports it downstream where it may be deposited. Erosion is driven by heavy rainfall, snowmelt, and daily variations in temperature that alternatively freeze and thaw the ground. These insights have helped us interpret signs of erosion and deposition of soil contaminated with ^{137}Cs in our case-study areas. Future variations in temperature, precipitation and extreme weather events will likely increase the occurrence of erosion and hence the redistribution of ^{137}Cs in the environment.

We have identified several areas sensitive to accumulation of and hence contamination by ^{137}Cs in Europe. In most places the levels of ^{137}Cs accumulation are rarely hazardous, however we recommend that local authorities assess the risks of new construction sites, agricultural practices and free-time activities such as hunting game and gathering mushrooms in areas that are sensitive to ^{137}Cs accumulation.

Sammanfattning

Vi undersöker variationer i ^{137}Cs aktiviteter, temperatur och nederbörd över tid och förutsäger hur klimatförändringarna kommer att påverka befintliga ^{137}Cs-anomalier. Vi fokuserar på flera fallstudier som påverkats av nedfall från Tjernobyl; Kymijoki vattendrag i Finland, Gävle i Sverige och Bayern i sydöstra Tyskland. Dessutom undersöker vi variationer i ^{137}Cs i Japan som en kontrasterande plats nära kärnkraftverket Fukushima Dai-ichi som skadades av tsunamin 2011.

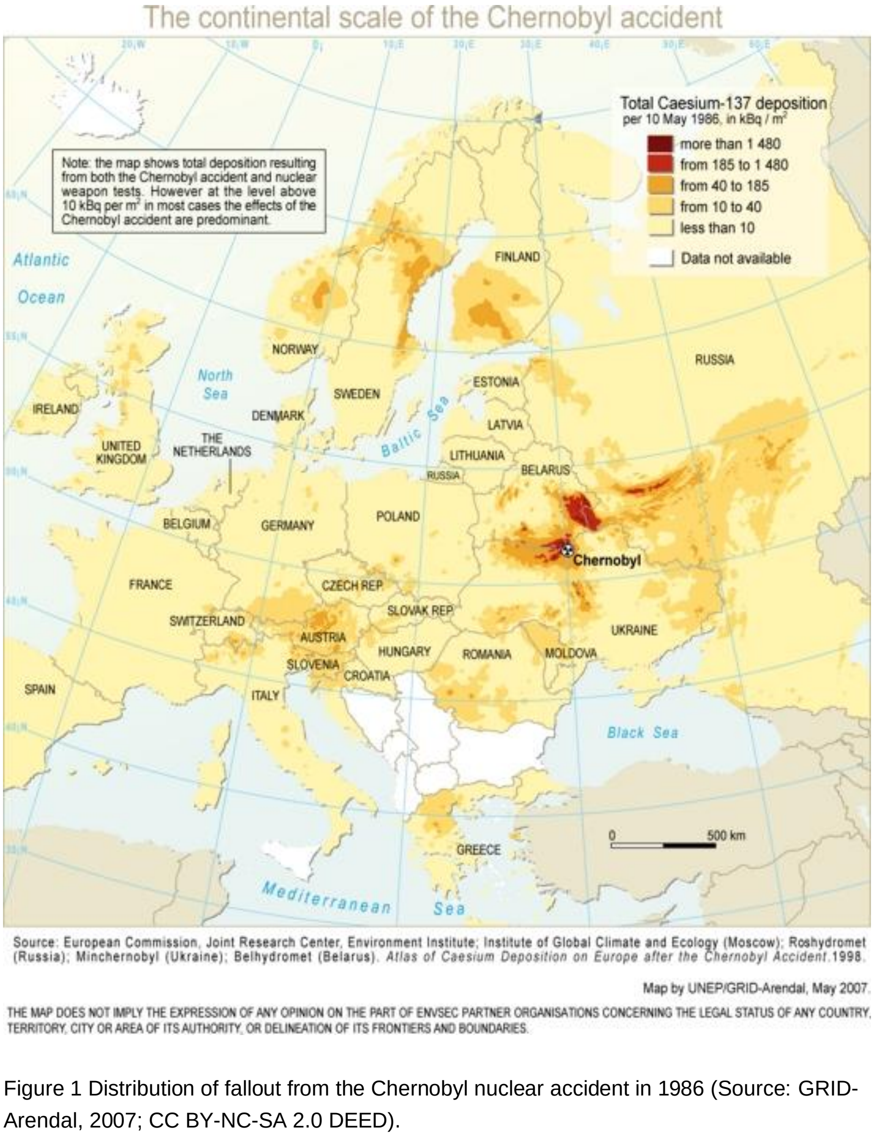
I Europa finner vi att ^{137}Cs-anomalier dyker upp i älgar, svampar och strålningsnivåer på marken under specifika år som skiljer sig från plats till plats. Det finns få indikationer på direkta samband mellan väderförhållanden och ^{137}Cs-anomalier. Miljöprocesser spelar dock en avgörande roll för radionukliders beteende och kan själva påverkas av väderförhållanden och dessutom av klimatförändringar. Erosion rör om ^{137}Cs i marken och transporterar det nedströms där det kan deponeras. Erosionen drivs på av nederbörd, snösmältning och dagliga temperaturvariationer som omväxlande fryser och tinar marken. Dessa insikter har hjälpt oss att tolka tecken på erosion och ackumulering av jord som förorenats med ^{137}Cs i våra fallstudieområden. Framtida variationer i temperatur, nederbörd och extrema väderförhållanden kommer sannolikt att öka förekomsten av erosion och därmed omfördelningen av ^{137}Cs i miljön.

Vi har identifierat flera områden i Europa som är känsliga för ackumulering av och därmed kontaminering med ^{137}Cs. På de flesta platser är nivåerna av ^{137}Cs ackumulering sällan farliga, men vi rekommenderar att lokala myndigheter undersöka potentiella risker förknippade med nya byggande, jordbruk och fritidsaktiviteter som jakt på vilt och insamling av svamp i områden som är känsliga för ^{137}Cs ackumulering.

Content

1. Introduction and objectives

Radionuclides including ^{137}Cs from the Chernobyl nuclear power plant accident in 1986 were deposited across large areas of northern and central Europe (Figure 1; Steinhauser et al., 2014; Nakajima et al., 2019). More recently the tsunami in 2011 caused damage to the Fukushima Dai-ichi nuclear power plant in Japan releasing radionuclides into the environment (Buesseler et al., 2011). Finland and Sweden each received ca. 5% of the fallout from Chernobyl and share similar terrains and proximity to marine basins with more than one half-life since the Chernobyl accident in 1986. Southeast Germany is also in the distal region of the Chernobyl fallout zone, which is a landlocked lowland landscape that is geomorphologically downslope from the high elevation Alpine mountains. Japan is in the early phase after contamination associated with the Fukushima tsunami in 2011. Additionally, Japan is proximal to the point source of the anomaly and with a coastal location for significant hydrosphere-lithosphere interaction.



Source: European Commission, Joint Research Center, Environment Institute; Institute of Global Climate and Ecology (Moscow); Roshydromet (Russia); Minchernobyl (Ukraine); Belhydromet (Belarus). *Atlas of Caesium Deposition on Europe after the Chernobyl Accident*.1998.

Map by UNEP/GRID-Arendal, May 2007.

THE MAP DOES NOT IMPLY THE EXPRESSION OF ANY OPINION ON THE PART OF ENVSEC PARTNER ORGANISATIONS CONCERNING THE LEGAL STATUS OF ANY COUNTRY, TERRITORY, CITY OR AREA OF ITS AUTHORITY, OR DELINEATION OF ITS FRONTIERS AND BOUNDARIES.

Figure 1 Distribution of fallout from the Chernobyl nuclear accident in 1986 (Source: GRID-Arendal, 2007; CC BY-NC-SA 2.0 DEED).

The cesium cycle in the environment begins with transfer from the atmosphere to the soil, where Cs shows affinity for clay or zeolite particles and becomes fixed to these particles (Absalom et al., 1996; Wauters & Cremers, 1996; Isaksson & Erlandsson, 1998; Isaksson et al., 2001; Sanchez-Valle et al., 2010; Andersson et al., 2014). The soil profile makes ^{137}Cs bioavailable to vegetation and fungi (Isaksson & Erlandsson, 1998; Dighton et al., 2008; Vinichuk et al., 2011; Snyder et al., 2012; Bahram et al., 2018). Grazing wild animals are therefore exposed to ^{137}Cs from the soil via vegetation and elevated activities have been recorded in Gävleborg, Heby and further South in Sweden as well as in Bavaria (Hysing, 2011; Hallqvist, 2013; SSM, 2018; Stäger et al., 2023).

Erosion occurs at the Earth's surface, where the lithosphere is in contact with the hydrosphere or atmosphere, i.e. temperature, precipitation, water and wind all cause erosion (Hilton & West, 2020). Weathering is the physical and chemical alteration of the soil horizon. Chemical weathering involves reactions with water that dissolve components such as K and Cs which are then transported by water (Land et al., 1999; Olsson & Melkerud, 2000). Physical weathering and erosion break down soil and till releasing solid material that is carried away by streams and rivers (sediment load). In this manner, weathering and erosion exposes formerly buried and stored radiocesium and associated ^{137}Cs anomalies can be remobilized and redistributed. Soil, wetlands, lakes and the Baltic Sea have acted as sinks for deposition and burial of ^{137}Cs (Knapińska-Skiba et al., 1994; Wauters & Cremers 1996; HELCOM MORS EG, 2014). We consider natural erosion from physical and chemical weathering associated with precipitation (rain and snow) and temperature changes, that are enhanced by climate change.

This project aims to investigate the impacts of climate change on radiocesium (^{137}Cs) anomalies. We investigate temporal variations in ^{137}Cs activities, temperature, rainfall, snow, freeze thaw cycles and predict how climate change will interact with existing ^{137}Cs anomalies. To assess what has happened to the ^{137}Cs anomalies associated with the Chernobyl accident in 1986 and the Fukushima tsunami 2011, we investigate time series data for four case studies; Finland, Sweden, Germany and Japan. Temporal variations in the ^{137}Cs anomalies for surface activities, moose and mushrooms will be presented. The processes we consider are radioactive decay, moose behavior, deposition, weathering and erosion.

2. Background

Accidents at nuclear power plants such as Chernobyl 1986 and the earthquake-tsunami related event at Fukushima 2011, as well as nuclear weapons testing in the 1950's to 1970's and nuclear warfare like Hiroshima and Nagasaki 1945, eject radioactive nuclides into the atmosphere (Almgren & Isaksson, 2006; Pittauernová et al., 2011; Snyder et al., 2012). Among the ejected radionuclides are noble gases which are distributed around the globe, while other elements, such as cesium, are deposited as aerosols or particles and subject to nuclear fallout (Masson et al., 2022). The aerosols and particles can be deposited dry but they are more efficiently extracted wet, therefore the highest anomalies will correlate with rainfall patterns (Du et al, 2010).

When radionuclides decay they emit radiation in the form of beta or alpha particles, often accompanied by gamma radiation. The radioactive decay time is a function of a nuclide's decay rate. Nuclear fallout introduces a variety of radionuclides into the environment such as ^{131}I, with a half-life of 8 days, which would decay quickly and not be detectable a few months after release. However, nuclear fallout also contains radionuclides such as ^{137}Cs with a half-life 30.2 years. Radiocesium may therefore remain in the environment for several hundred years before reaching negligible levels (Faure & Mensing, 2005). Environmental half-lives are often shorter than physical half-lives and therefore the effects of Chernobyl are often considered negligible after one physical half-life, due to burial removing the ^{137}Cs from the immediate surface environment (Hysing, 2011). In comparison only 12 years has passed since the Fukushima accident, therefore environmental processes are still actively in contact with the radionuclides (Nakajima et al., 2019).

Cesium enters the soil in water or as aerosol deposits on the surface, Cs fixation can occur by adsorption onto clay particles or as zeolites form (Absalom et al., 1996; Wauters & Cremers, 1996; Andersson, et al. 2014; Sanchez-Valle et al. 2010). Once the Cs is fixed, minimal vertical or lateral movement of the Cs occurs (Isaksson & Erlandsson, 1998; Isaksson et al., 2001; Buesseler et al., 2011). However, there is the potential for flood events to remobilize Cs, thus diluting the Cs rich horizon (Snyder et al., 2012). The soil profile is bioavailable to plant roots, down to about 60 cm, whereas the ^{137}Cs contaminated horizons are in the top 20 cm of the soil where most fungi occur (Snyder et al., 2012; Isaksson & Erlandsson, 1998; Dighton, et al., 2008; Vinichuk et al., 2011; Bahram et al., 2018).

Furthermore, Cs is fluid mobile and hence shows high solubility in rivers, lakes and oceans (Du et al., 2010). Dissolved salts in seawater help to bind ^{137}Cs and facilitate sedimentation (Knapińska-Skiba et al., 1994). River water moves rapidly and is therefore mostly a transport agent for ^{137}Cs. Sources of Cs to lakes and oceans are derived directly from wet or dry atmospheric deposition, as well as from river water, water sediment load and erosion (Kansanen et al. 1991; Frank, 2002; Du et al., 2010). Sediments that precipitate from water are likely to be fine grained and clay-rich which promotes the adsorption of Cs (Absalom et al., 1996; Wauters & Cremers, 1996; Andersson, et al., 2014).

Geochemistry provides useful proxies to trace environmental processes. Trace elements such as K, Rb and Ba show similar behavior to Cs in soil and they can be mobilized by chemical or physical weathering particularly in clay-poor and acidic soils (Land et al., 1999; Land & Öhlander, 2000; Casetou-Gustafson et a., 2020; Loba et al., 2022). Additionally, radionuclides like ^{137}Cs and ^{40}K are used to trace environmental processes, particularly erosion, deposition and burial (Quine et al., 1997; Djodjic & Spännar, 2012; Sac & İçhedef, 2015). Activities of ^{137}Cs and ^{40}K decrease in areas of erosion as soil containing radionuclides is removed from the site, burial also decreases ^{137}Cs and ^{40}K activities at the surface, burying it lower down in the soil profile (Quine et al., 1997; Fujiyoshi & Sawamura, 2004; Djodjic & Spännar, 2012; Jagercikova et al., 2015). For example, soil profiles in Skåne show subsurface ^{137}Cs anomalies associated with fallout from Chernobyl and nuclear weapons testing that have since been buried (Isaksson & Erlandsson, 1998; Isaksson et al., 2001). In contrast, sites where eroded sediment accumulate and deposit lead to a corresponding increase in radionuclides such as ^{137}Cs and ^{40}K. Bioturbation causes local mixing which can also expose previously buried radionuclides.

3. Results and Discussion

We present and discuss the findings of ^{137}Cs time series analysis and associated influence of weather and environmental processes for case studies from Finland, Sweden and Germany related to fallout from the Chernobyl accident in 1986. We follow up with a case study from Japan associated with contamination by the Fukushima tsunami and accompanying nuclear power plant accident in 2011.

3.1 Kymijoki watershed, Finland

The Kymijoki river in southern Finland was in the zone of Chernobyl fallout over Finland and has a catchment size of 37 200 km^2 that discharges into the Gulf of Finland (Figure 2). Lake Päijanne, the largest lake in the catchment, supplies drinking water to approximately one million people, mainly in the Helsinki metropolitan area.

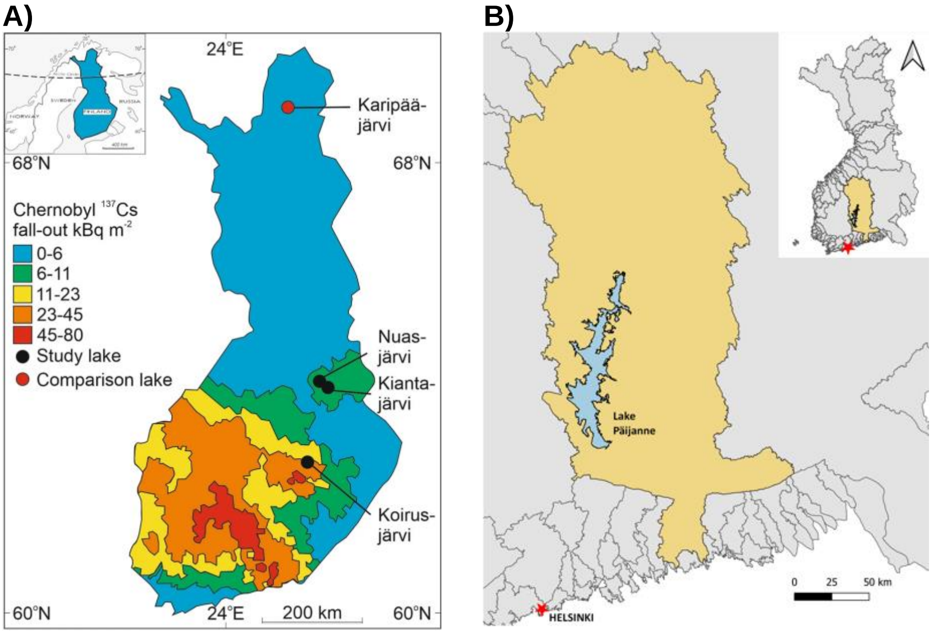


Figure 2 A) Map of Chernobyl fallout over Finland. (Source: Appleby et al., 2022; CC BY 4.0). B) Map of Kymijoki Watershed, Finland, showing Lake Päijanne that supplies drinking water to the Helsinki metropolitan area.

The Kymijoki river is approximately 200 km long and has a mean annual flow of 282 m^3 s^{-1} with minimum and maximum recorded flow rates of 65 m^3 s^{-1} and 816 m^3 s^{-1} respectively. Water discharge from the Southeast of the Kymijoki watershed varies between 150 and 490 m^3 s^{-1} and the suspended sediment load ranges from 1.5 to 6.6 mg L^{-1}. The variations in sediment load often reflect the discharge, however there are some occasions where suspended sediment is at odds with the discharge (Figure 3). Elevation in the watershed ranges from 0 to 266 m a.s.l. with the highest parts found in the Northwest (Figure 4a; Nilsson et al., 2023a). The northern part mainly consists of moraine soils whereas the southern part is dominated by rock covered with a thin soil layer. Clayey soil, although only constituting 6 % of the total soil, is the most common in the very southern tip of the watershed (Figure 4b; Nilsson et al., 2023a). Land use is predominantly forest, with 37% evergreen and 34% mixed evergreen and deciduous forest. Agricultural land accounts for 5%, coinciding with the southernmost part of the catchment where we find clayey soils. Finally, water covers 19% of the total catchment area (Figure 4c; Nilsson et al., 2023a).

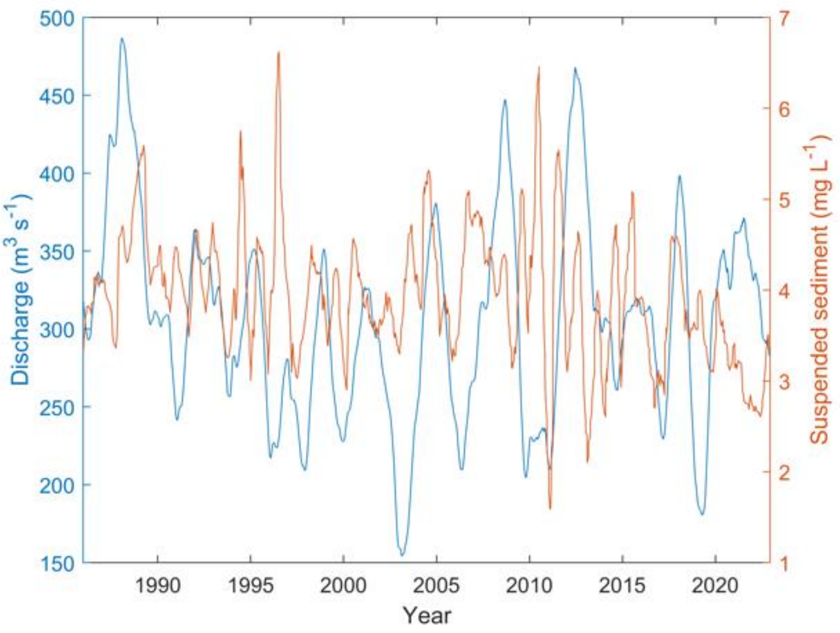
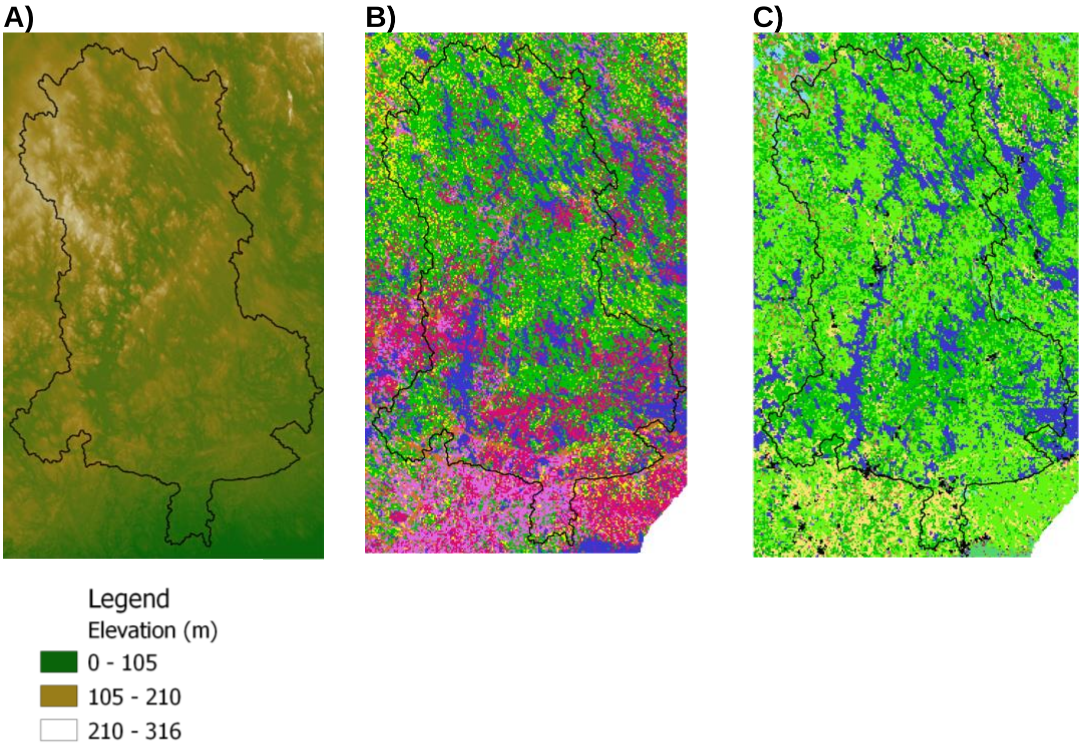


Figure 3 Discharge and suspended sediment in Kymijoki river on a one year moving average 1986 to 2022. Data from Finnish Environment Institute and Centers for Economic Development, Transport and the Environment (CC BY 4.0).



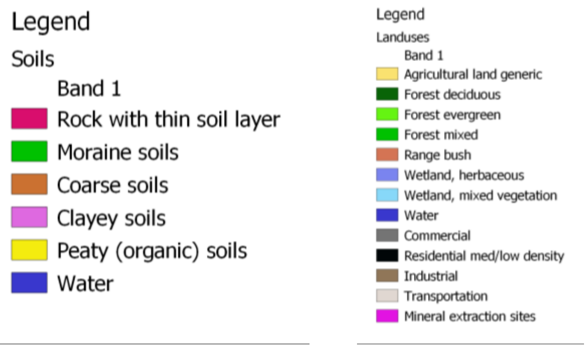


Figure 4 A) Topographic map B) Soil map, C) Landuse map of the Kymijoki Watershed, Finland.

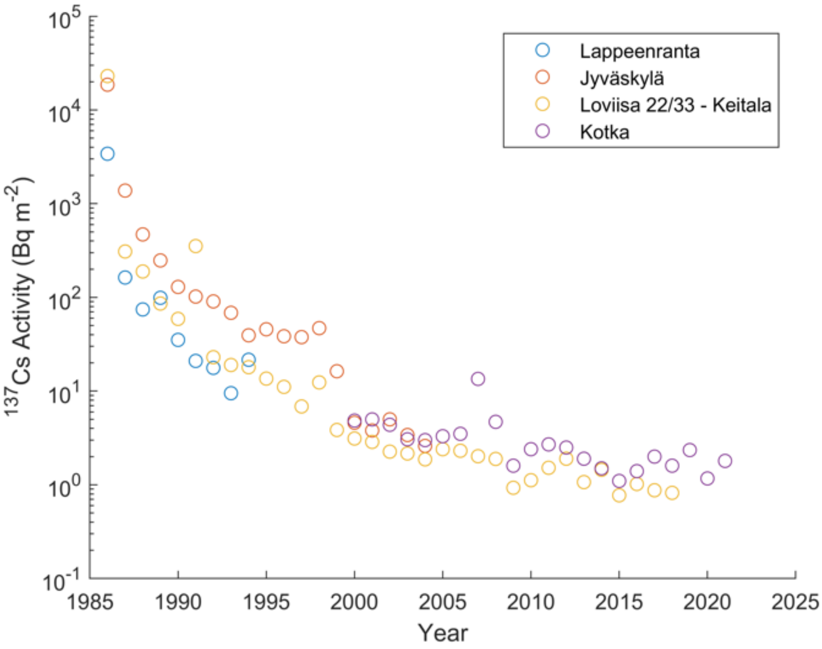


Figure 5 Ground deposition of ^{137}Cs measured annually, the high values observed from 1986 are associated with the Chernobyl nuclear power plant accident. The four stations are within or in proximity of the Kymijoki watershed. Data source: STUK and Loviisa nuclear power plant.

Atmospheric fallout of ^{137}Cs in the Kymijoki watershed ranged from 3500 to 25000 Bq/m^2 in 1986, since then the corresponding ^{137}Cs activities have decreased to ^{137}Cs of ca. <2 Bq/m^2 (Figure 5). The decrease with time is much more than expected from physical decay with a half-life of 30.2 years, indicating that environmental processes are involved. Although the ^{137}Cs activities could be depleted by weathering and erosion mobilizing and transporting it elsewhere, these curves are classic for transfer of ^{137}Cs to clay particles in the soil which then get buried as more soil is produced. However, there are perturbations to the overall decreases in ^{137}Cs activity, such as ^{137}Cs activities up to 15 Bq/m^2 in Kotka in 2007. Additionally, we observe uniform ^{137}Cs activities of ca. 40 Bq/m^2 in Jyväskylä between 1994 and 1998 that are elevated compared to activities of ca. 15 Bq/m^2 at other locations nearby (Nilsson et al., 2023a). These perturbations in ^{137}Cs activities are likely linked with mobilization and redeposition of ^{137}Cs bearing particles by erosion, potentially due to wind (Bärring et al., 2003; Chappell & Warren, 2003). Modelling of the weather conditions, geographical and hydrological parameters together with ^{137}Cs activities will help explain the causes of these anomalies as well as redistribution of ^{137}Cs within the Kymijoki watershed.

We construct a model of the dynamics of the Kymijoki watershed which focuses on the chain of precipitation, erosion and deposition with the aim of simulating water flow and erosion of particles (Nilsson et al., 2023b). Runoff is expected to transport ^{137}Cs as particles of suspended sediment in the hydrological system. Deposition of particles from water is a function of flow rate, which depends on topography, turbulence, water volume, and particle type, e.g. clay particles are suspended in water at much lower flow rates than larger grain sizes (Li et al., 2017). For this we use SWAT*Plus* to model erosion and runoff, which takes topography, soil, meteorology and landuse into consideration (Neitsch et al., 2005, 2011). Preliminary hydrological and erosion modelling in SWAT*Plus* has identified sub-basins within the Kymijoki watershed as well as constrained hydrological response units from the hydrological properties that are associated with common landuse, soil types and topography (Figure 6). These hydrological response units are equivalent to grid cells in other types of numerical models. In this case the cells are not square but are defined by geographical properties in a way that fits the landscape (Neitsch et al., 2005, 2011). Like grid cells, the hydrological response unit have inputs and outputs that are associated with neighbouring units to simulate detailed water flow and sediment transport within the entire Kymijoki watershed. The purpose of the model is to trace the soil and sediment erosion and deposition and the relationship to water flow. To calibrate the model, the water discharge and sediment load data from the hydrological response units will be compared to the discharge and suspended sediment from the stations in the South of the Kymijoki watershed (Fig. 3). This will allow us to explain differences in the behavior of water discharge and suspended sediment. In additional steps, the ^{137}Cs module we have developed for SWAT*Plus* will simulate the redistribution of ^{137}Cs by erosion and deposition. In turn the ^{137}Cs module output will be calibrated with the ^{137}Cs activities in soil, water and atmospheric deposition and facilitate explanation of the ^{137}Cs variations in terms of environmental processes and the influence of weather (Figure 5).

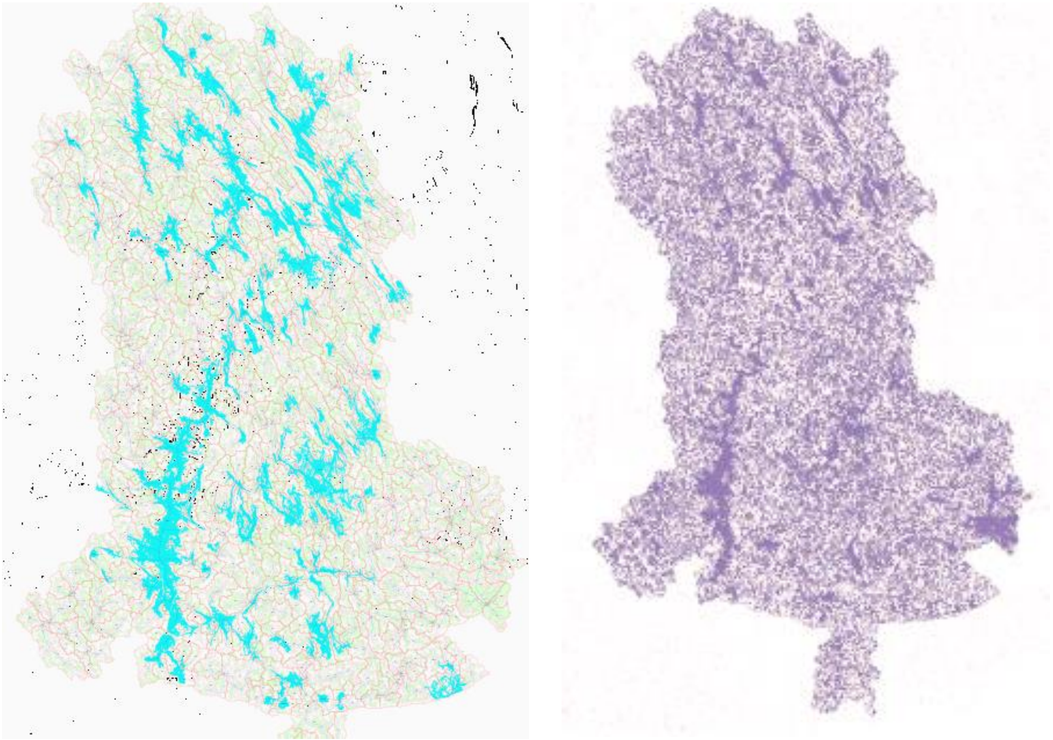


Figure 6 Sub-basins (left; red lines) and hydrological response units (right; purple) with the same landuse, soil type and topography for the Kymijoki watershed generated by modelling with SWAT*Plus.*

3.2 Gävle, Sweden

Sweden received approximately 5% of the fallout from the Chernobyl accident in 1986, when wind brought the radionuclides up over Sweden and rainfall deposited radionuclides over Gävle and Västernorrlands up into Västerbotten (Figure 7).

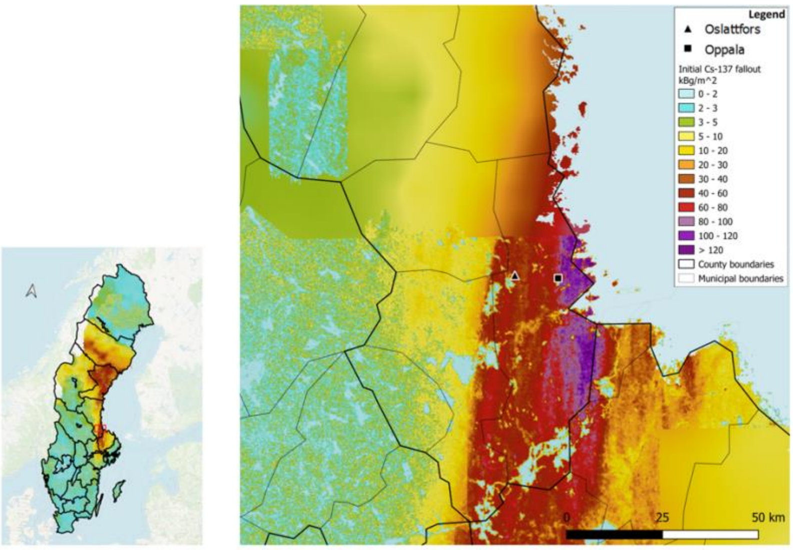


Figure 7 Map of ^{137}Cs fallout over Sweden from Chernobyl nuclear power plant accident in 1986 with detailed map of the Gävle area and study site.

We have focused on ^{137}Cs activities from moose in the Gävle area. We processed the data relative to local fallout, firstly by correcting the atmospheric deposition (Bq/m^2) for radioactive decay with time. Then normalized the measured ^{137}Cs activities (Bq/kg) to the decay corrected atmospheric deposition for the corresponding year to provide decay corrected ^{137}Cs values (m^2/kg). If the ^{137}Cs in the environment was only influenced by physical decay the data would form a horizontal line, deviations are due to environmental processes, such as burial that decreases the ^{137}Cs with time, whereas increases in ^{137}Cs with time would reflect addition likely from accumulation and deposition (Quine et al., 1997).

Compilations of ^{137}Cs activities from moose hunted annually in the Gävle area in late September to early October are expected to show a decrease in activities with time as seen overall between 1986 and 2020 (Figure 8). However, time series decay corrected ^{137}Cs activities show more complex patterns than the expected systematic decreases. For instance, we observe wider ranges and more elevated ^{137}Cs activities in 1997, 2003 and 2006 as well as low activities from 1998 to 2002, whereas the ^{137}Cs activities remain elevated between 2003 and 2006 (Figure 8; Jakabová et al., 2023a). To test whether these years show anomalous ^{137}Cs activities compared to preceding and subsequent years we have compared the cumulative frequency distributions (Figure 9; Jakabová et al., 2023a). The cumulative frequency distributions and Kolmogorov-Smirnov test confirm that decay corrected ^{137}Cs activities in 1997, 2003 and 2006 are anomalous with 99% significance. These anomalies in ^{137}Cs activities are independent of the age of the moose and are also confirmed by roe deer (Weimer, 2015).

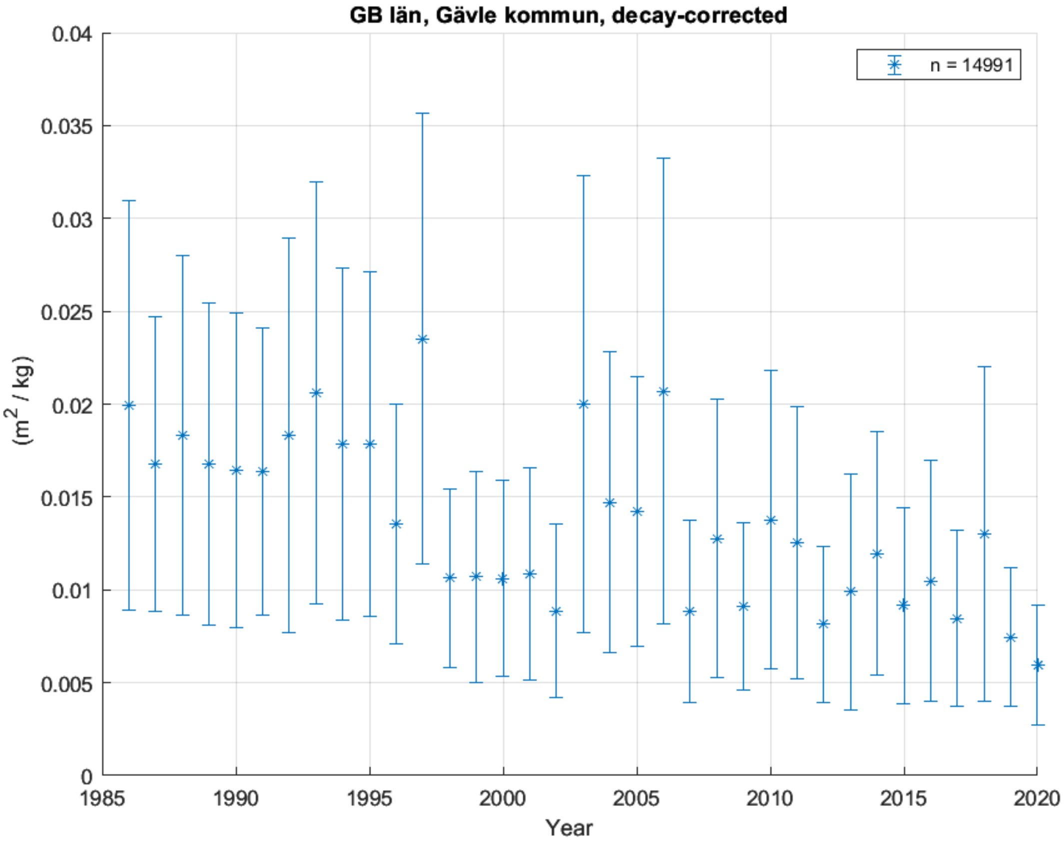


Figure 8 Decay corrected ^{137}Cs to account for atmospheric deposition and radioactive decay with time. Data from SSM's Cs database.

We consider the influence of moose behavior on their uptake of ^{137}Cs and the reliability of employing moose to trace environmental processes. Moose tend to have summer habitats and move to their winter grazing locations in September (Melin et al., 2014). On location in their summer residencies they likely graze within an area of approximately 1000 to 1700 ha (Allan et al., 2016). Additionally, ^{137}Cs resides in moose for two to three months, therefore their diets in the summer and early autumn will affect their ^{137}Cs activities during the hunting season. Moose mainly consume vegetation from trees and shrubs (von Bothmer et al., 1990). During the summer months moose eat vegetation from birch, willow, rowan, aspen and oak trees and shrubs like blueberry and lingonberry, they have a preference towards blueberry plants (pers comm, Jonas Malmsten; von Bothmer et al., 1990; Stålfelt 1993; Fawaris and Johanson, 1994; Rosen et al., 2011). Moose prefer cool wet summer weather suffering from heat stress above 15 to 20°C (Renecker & Hudson, 1986; Bubenik, 1998; Melin et al., 2014). They may hide in the forest during hot dry summer weather however this does not tend to affect their diet. Instead, their health suffers from the reduced nutritional value of drier woodier plants compared to cool, wet summers with lush vegetation (pers. comm. Jonas Malmsten). In addition, our analysis of ^{137}Cs activities and summer temperature and rainfall shows a lack of consistent trends, indicating that summer weather does not influence grazing behaviors. Instead, the ^{137}Cs of moose seems to reflect the ^{137}Cs levels of the plants that make up their diet, which in turn take up ^{137}Cs from the soil. Variations in ^{137}Cs in moose are therefore a reflection of the environmental processes affecting the soil composition. Activities of ^{137}Cs in moose provide annual records of ^{137}Cs in the environment and therefore changes are tracing processes that occur in the annual cycle of seasons.

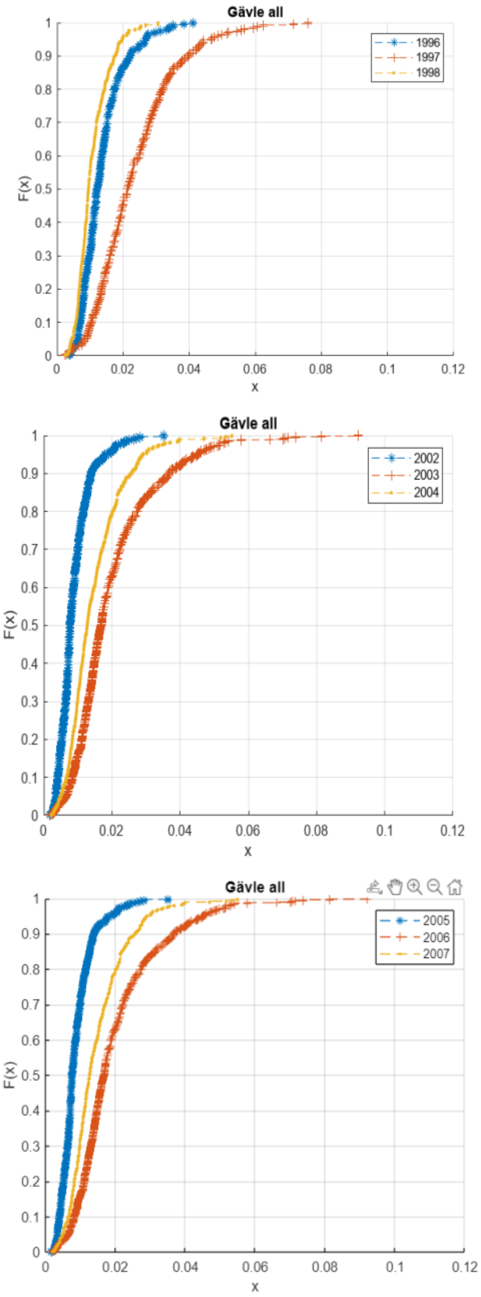


Figure 9 Cumulative frequency distributions of decay corrected ^{137}Cs activities for discrete years to test for statistically significant differences between years.

To investigate the behavior of Cs and spatial distribution of environmental processes, we have explored the relationship between Cs and Zr in soil from the Gävle area. To do so, we have assessed soil geochemistry data from SGU, they sample the C-horizon of soil profiles between 75 and 100 cm depth, taking a sample at least every 9 km^2 (www.sgu.se). We use these trace element compositions in soil to evaluate loss and gain of Cs. The premise is to compare total elemental Cs and K with Zr, which is immobile in soil (Casetou-Gustafson et a., 2020). If the total Cs and K have not changed then the relationship between elements will give a straight line, with a high correlation coefficient. Figure 10a and b show that total Cs and K form elongated trends that slightly increase with increasing Zr. There is significant scatter in Cs and K and both elements show outliers with very high Cs or K content. To constrain the enrichment and depletion of Cs with respect to Zr, we estimate the change from a regression line (Fig. 10a; Casetou-Gustafson et a., 2020). Samples lying above the regression line are interpreted to have been enriched in Cs concentration (positive mass change) and those falling below the line are depleted in total Cs concentration (negative mass change).

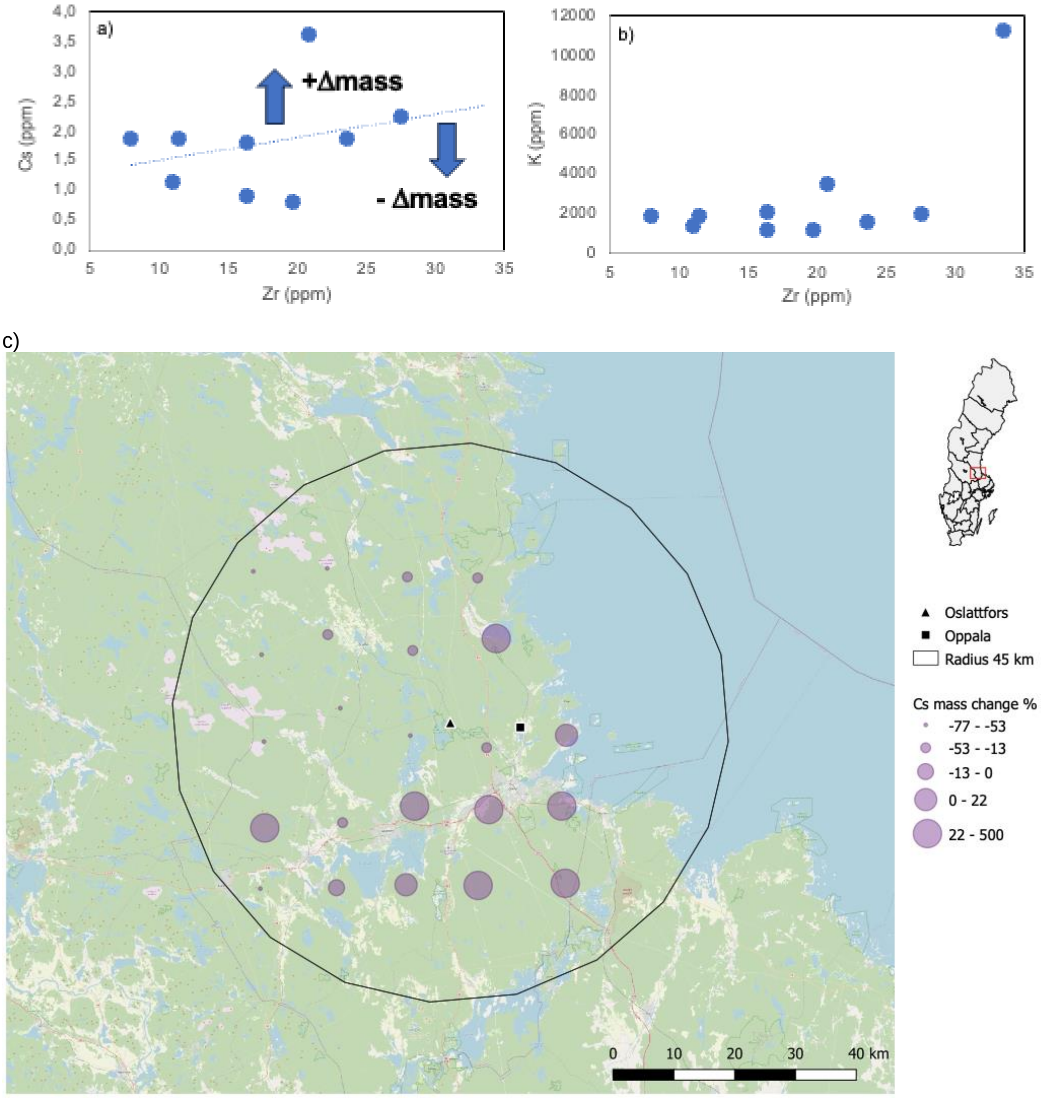


Figure 10 a) Cs vs Zr concentration in ppm in soil, showing that mass change is positive above the regression line and negative below the regression line b) K vs Zr concentration in ppm in soil, c) Mass change of Cs with respect to Zr in till for Gävle study area. Circle marks likely moose mobility within summer habitat. Data source: SGU.

A negative mass change suggests that Cs has been removed from the soil by weathering and erosion relative to the resistant and immobile Zr. In the Northwest of the study area, defined by likely moose migration within their summer grazing grounds, we observe Cs loss by weathering and erosion (Figure 10c; Jakabová et al., 2023b). However, in the South and East of the study area we observe Cs enrichment relative to Zr, indicating accumulation of Cs in this area (Figure 10c). This spatial distribution of Cs enrichment and depletion is also recorded by K, Ba and Rb, confirming that weathering and erosion is occurring in the Northwest and deposition and accumulation of eroded material occurs in the South and East (Jakabová et al., 2023b). The ^{137}Cs will behave the same way as the other naturally occurring isotopes of Cs in the environment, so we would expect it to be mobilized from the same places and deposited and accumulated in the same locations and by the same processes as total Cs. Furthermore, soil erosion has been linked to high suspended sediment and low ^{137}Cs activities, whereas areas of high ^{137}Cs activities correspond to zones of accumulation (Quine et al., 1997; Djodic & Spännar, 2012). Additional support for the accumulation of material in the area is the presence of high ^{137}Cs activities in wetlands at Hille located near Oppala (Stark et al., 2006). Hence the area of total Cs, K, Ba and Rb accumulation recorded by soil is likely to facilitate current and future accumulation of eroded material containing ^{137}Cs. In areas where total Cs including ^{137}Cs is enriched in the soil by deposition of mobilized Cs it will become bioavailable to moose via the plants that grow from the Cs enriched soil.

In order to explain the relationship between erosion and deposition of material in the study area and the years with uptake of anomalous ^{137}Cs in moose we explore the influence of weather on erosion. Erosion rates have been shown to change dramatically due to weather events, an example from Busssjösjön, southern Sweden records between 218 tons winter 1986/1987 compared with only 4 tons over the winter of 1987/1988, associated river bank and slope instability (Dearing et al., 1989). Rainstorms in 1998 in Abisko, northern Sweden caused strong erosion of steep slopes and river banks (Jonasson & Nyberg, 1999). Erosion is promoted by freeze-thaw cycles and seasonal variations in weather such as autumn rainfall, spring snowmelt and rain on partly thawed soils (Ulén et al., 2012). We have investigated river discharge in spring to trace snowmelt, temperatures and rainfall during the spring and summer months for the study area and no clear relationships with anomalous ^{137}Cs activities in 1997, 2003 and 2006 were found. Preliminary relationships between ^{137}Cs activities and freeze thaw cycles are apparent in 1997 when there was less snow cover and more freeze thaw cycles compared to 1996 (Jakabová et al., 2023b).

Sometimes the accumulation of ^{137}Cs activities show a single anomalous year, e.g. 1997, whereas other times such as 2003, the ^{137}Cs activities decrease in the following years but do not return to background values immediately, see 2004 and 2005 (Fig. 8, 9). When we look at Oslättfors in detail we find that the ^{137}Cs activities in 2004 are no lower than 2003 but remain high. The persistence of ^{137}Cs in the environment between 2003 and 2006 is likely connected to relatively slow burial rates and significant bioturbation of the top soil by wildlife, which spreads out and reconcentrates the ^{137}Cs. The low ^{137}Cs activity in 1998 directly after the elevated ^{137}Cs activity in 1997 was combined with low snow cover and frequent freeze-thaw cycles in both 1997 and 1998. This suggests further erosion occurred in 1998 from the site where ^{137}Cs was deposited in 1997, the erosion likely remobilized ^{137}Cs and transported it down slope i.e. potentially to lakes or wetlands nearby alternatively out to the Baltic Sea.

Climate change is expected to increase warming in Sweden, which would likely lead to more years with reduced snow cover and corresponding increases in freeze thaw cycles, which would further promote erosion. Enhanced erosion would serve to mobilize buried ^{137}Cs and promote transportation and accumulation of ^{137}Cs in areas like the South and East of Gävle, key localities are likely to be Oslättfors, Oppala and Hille. Hence the future will likely episodically increase the ^{137}Cs activities similarly to 1997, 2003 and

2006 in moose and deer grazing in this area, as well as mushrooms, plants and wetland environments. This may continue to occur in specific years that may be predictable by the winter weather conditions and could also become a more general pattern as warm winters become more common. Once snow, frost and frozen ground conditions are excluded by positive winter temperatures, erosion by freeze thaw will significantly decrease and hence also the ^{137}Cs mobility that is incurred. Additionally, the ^{137}Cs in the environment is finite, introduced by the Chernobyl accident and nuclear weapons testing, therefore erosion and accumulation of ^{137}Cs will decrease and stop as ^{137}Cs buried in the environment is removed and the remaining ^{137}Cs decays. Therefore, the environmental issues with ^{137}Cs will decrease with decreasing erosion, removal of ^{137}Cs from areas of erosion and burial of ^{137}Cs in the deposition and accumulation zones.

3.3 Bavaria, Germany

Bavaria in the Southeast of Germany was also within the zone of high fallout with 10,000 to 80,000 Bq/m^2 of ^{137}Cs deposited in 1986 following the Chernobyl accident (Figure 11). Locally the ^{137}Cs deposition was highly heterogeneous (Figure 11).

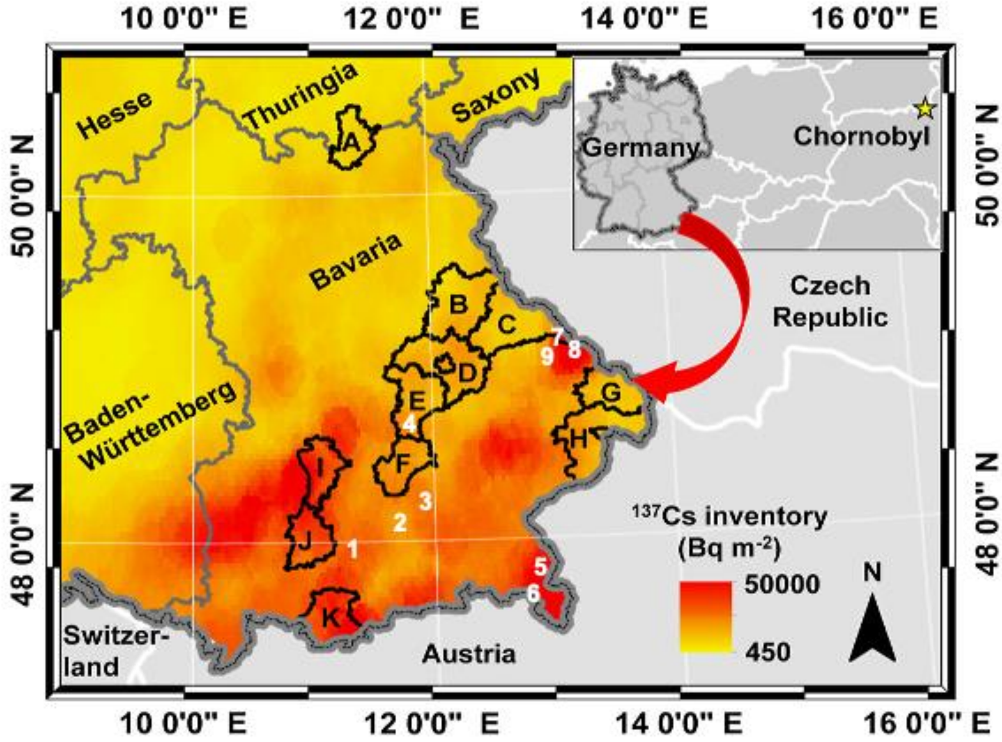


Figure 11 Map of ^{137}Cs fallout in Southeast Germany 1986 (source: Stäger et al., 2023; CC-BY 4.0). The ^{137}Cs data (Bq/m^2) is decay corrected to 1986 (source: Federal Office for Radiation Protection (BfS)). Numbers mark sampling sites for the data presented in this study (source: Federal Office for Radiation Protection (BfS)). Letters mark the following locations (A) Kronach; (B) Schwandorf; (C) Cham; (D) Regensburg; (E) Kelheim; (F) Freising; (G) Freyung-Grafenau; (H) Passau; (I) Aichach; (J) Landsberg; and (K) Garmisch-Partenkirchen.

The German Federal Office for Radiation Protection (BfS) have carried out a longterm monitoring campaign of ^{137}Cs and ^{40}K in mushroom samples from Bavaria, Southeast Germany. They have routinely sampled at ten sampling sites (Figure 11), and collected, analysed and reported between 1 and 4 analyses per species for the different sample localities on an annual basis (BfS reports). This data has been compiled and we present data for samples of six mushroom species that report a reasonable amount of data for the time period 2005 to 2021 (Figure 12). The ^{137}Cs activities have been corrected for initial atmospheric deposition and radioactive decay since 1986, using the same protocol as for data from Gävle, all figures show decay corrected data.

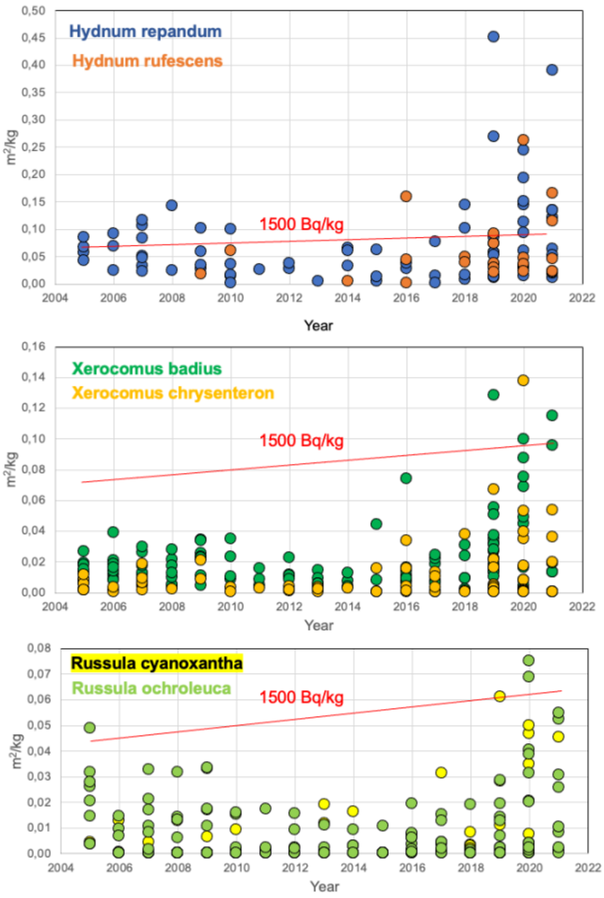


Figure 12 Decay corrected ^{137}Cs for different mushroom species collected in Bavaria from 2005 to 2021. Red line marks 1500 Bq/kg, which is the limit recommended for regular human consumption by livsmedelverket. Analysis of fresh, wet weight and each point is a single mushroom sample. Data from BFS.

The six mushroom species presented show an intermediate range in decay corrected ^{137}Cs activities from 2005 to 2009 (Figure 12). From 2011 to 2015 the ^{137}Cs activities are lower, the difference from 2005 to 2009 and 2011 to 2015 is more pronounced for the *Russula ochroleuca* samples than the other species. Subsequently from 2019 to 2021 much higher ^{137}Cs activities are observed in all mushroom species (Figure 12). The maximum activities for the different species are ^{137}Cs 1000 to 7400 Bq/kg (fresh weight). The cumulative frequency distributions for *Hydnum repandum* show consistently higher ^{137}Cs activities in 2020 and 2021 than in 2007, 2009 and 2014-2015, suggesting that the ^{137}Cs activities are statistically higher in 2020 and 2021 than earlier (Figure 13). Notably the 2019 data for *Hydnum repandum* has two outliers at ^{137}Cs 4400 and 7400 Bq/kg, however the other samples are within the expected distribution of the previous years, these outliers come from sites 8 and 6 respectively (Figure 11). The ^{137}Cs activities for

Hydnum repandum in 2020 and 2021 range from 220 to 3900 and 300 to 6100 Bq/kg respectively.

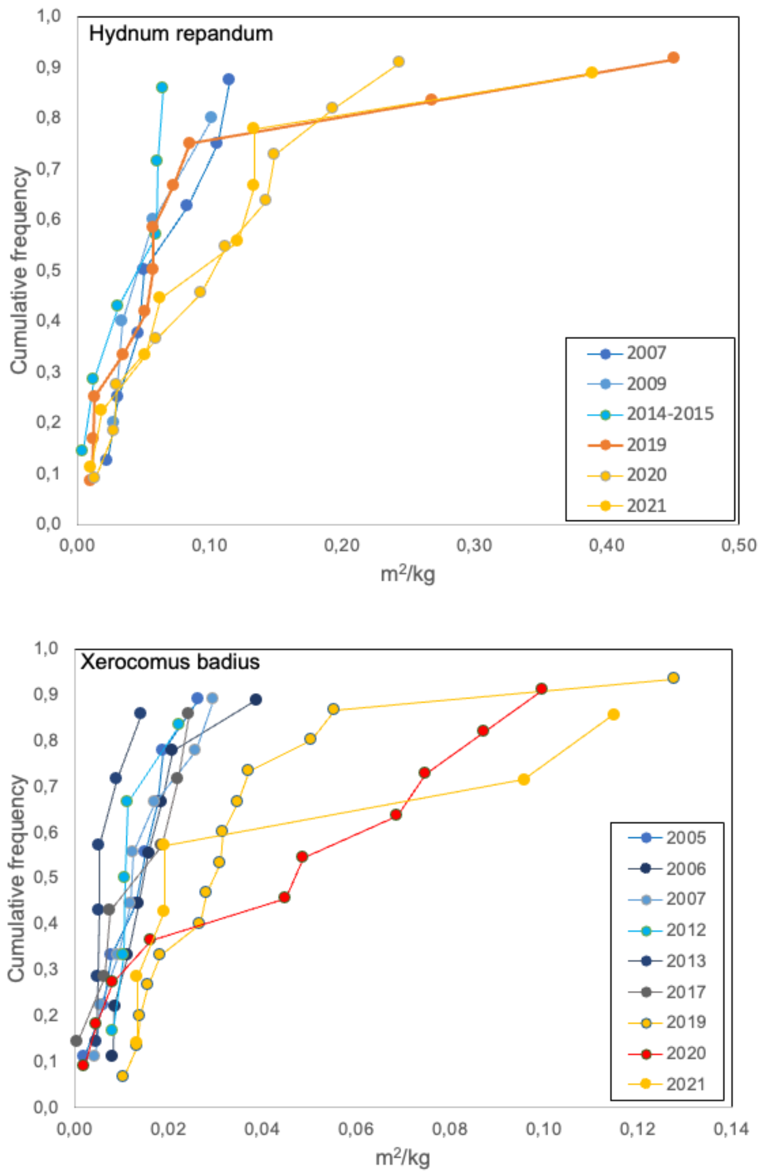


Figure 13 Cumulative frequency distribution of decay corrected ^{137}Cs data for Hydnum repandum and Xerocomus badius. Years selected mostly based on sample numbers.

For *Xerocumus badius* (*Imleria badia*) the ^{137}Cs activities for years 2005 to 2017 and 2012, 2013 and 2017 have similar cumulative frequency distributions, with a hint that 2012 and 2013 have the lowest ^{137}Cs activities (Figure 13). The ^{137}Cs activities increase systematically in 2019 and then more in 2020 in this case, with activities of 170 to 910 Bq/kg and a max of 2100 Bq/kg for 2019 and mostly in the range 720 to 1600 Bq/kg for 2020. In 2021 the ^{137}Cs activities in *Xerocumus badius* (*Imleria badia*) had returned to normal except two outliers at 1500 and 1800 Bq/kg from site 6 (Figure 11). The ^{40}K activities data backup the variations we have observed in ^{137}Cs activities, with the widest ranges in 2019 to 2021, and also *Hydnum repandum* tentatively displaying intermediate ranges between 2005 and 2007. The samples with highest ^{40}K activities are found in

Hydnum refuscens and *Xerocumus chrysenteron* measured at ^{40}K of 410 and 460 Bq/kg, were sampled from site 4 in 2019 (Figures 11, 14). *Russula ochroleuca* has an outlier in 2015 of ^{40}K at 610 Bq/kg however since it is the only sample of that species analysed in that year it is hard to assign any meaning to the analysis.

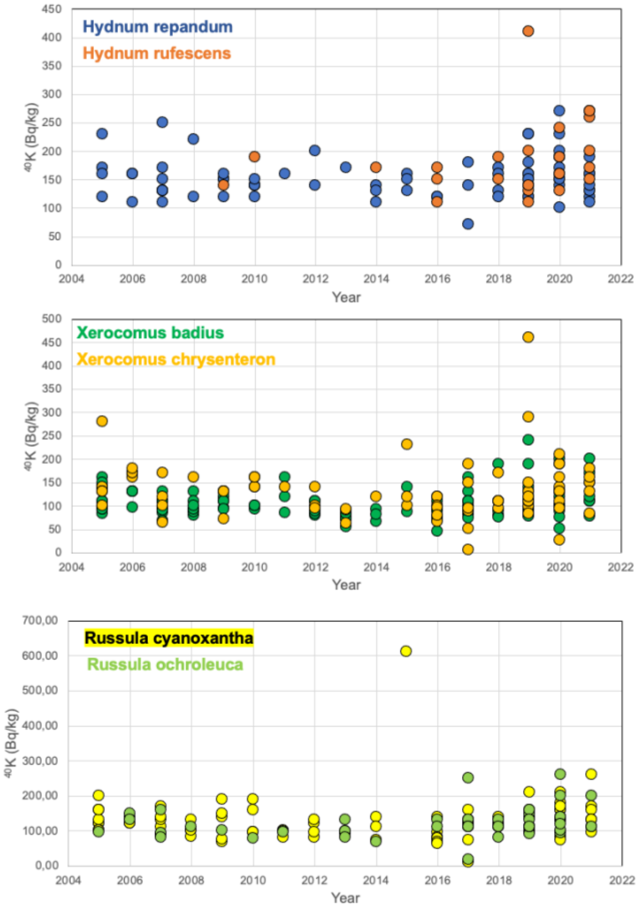


Figure 14 ^{40}K for different mushroom species collected in Bavaria from 2005 to 2021. Analysis of fresh, wet weight. Data from BFS.

The mushrooms species from Bavaria show elevated ^{137}Cs activities in 2019 to 2021 (Figures 12 & 13). Stäger et al. (2023) have also observed extremely high ^{137}Cs activities of up to 15,000 Bq/kg in wild boar during 2019 to 2021, they have shown that testing of nuclear weapons in the 1950's to 1970's contributes significantly to these elevated ^{137}Cs activities. The ^{137}Cs activities in mushrooms from Bavaria are confirmed by the higher activities of naturally occurring ^{40}K. Bioturbation by wildlife and in particular wild boar would create a mixing zone with similar ^{137}Cs not exceeding previous years, i.e. the levels should be roughly constant with time (Holby & Evans, 1996; Tyler et al., 2001; Fujiyoshi & Sawamura, 2004; Jagercikova et al., 2015; Jarvis et al., 2020). In contrast, significant increases in ^{137}Cs activities indicates input from external sources (Quine et al., 1997; Djodic & Spännar, 2012; Appleby et al., 2023). This is confirmed by the relationship between different sources, in Bavaria ^{137}Cs fallout from the Chernobyl power plant accident was much higher than the fallout from nuclear weapons testing, which would also reside deeper in the soil profile (Appleby et al., 2023). However, the boar data show that nuclear weapons testing contributes 10 to 70% of the ^{137}Cs in the area confirming that environmental processes are depositing ^{137}Cs and ^{40}K in the area (Stäger et al., 2023). Integrating the evidence above indicates that Bavaria and especially sites 6 and 8 are locations of deposition of eroded material that become enriched in ^{137}Cs and ^{40}K. Bavaria can be described as the foothills of the Alps, therefore it is natural that material would be eroded at higher elevations and transported to the lower elevations of Bavaria. The increases in ^{137}Cs and ^{40}K activities in 2019 to 2021 would imply an increase in erosion in recent years, this may be related to retreating glaciers, warmer winters with less snow and more freeze thaw action as well as other weather patterns that increase erosion (Ulén et al., 2012). Enhanced erosion due to climate change will likely increase the redistribution of ^{137}Cs in the future and this study shows that Bavaria and specific locations within the region are sensitive zones of ^{137}Cs accumulation.

3.4 Fukushima, Japan

The 2011 nuclear accident at Fukushima Dai-ichi nuclear power plant (FDNPP) released radionuclides to the surrounding areas. About 80 percent of the releases ended up in the Pacific Ocean, whereas the remaining part affected land areas primarily in Fukushima Prefecture.

A comprehensive decontamination effort in the affected areas was initiated by the Japanese government and local authorities. The purpose was to enable people to return to their homes and to secure the continuation of agriculture. The main radioactive nuclides of concern are ^{137}Cs, with 30 years half-life, and, to a lesser extent, ^{134}Cs with two years half-life. Here we mainly consider the behaviour of ^{137}Cs.

The Fukushima Prefecture has mountains reaching 2000 m in the western part, lower altitude mountains covered by forests in the eastern part, and lowland areas along parts of the coast along the Pacific Ocean. The areas affected by the radionuclides from the accident are primarily in the eastern part, where villages, forests and agricultural areas have been affected. About 70% of the total land area of Fukushima Prefecture is covered by forests, and about 10% is agricultural land. Figure 15 shows the distribution of ^{137}Cs and ^{134}Cs in eastern Fukushima as of April 2011 (Japan Atomic Energy Agency, 2013). The four cities and communities Fukushima City, Iitate, Kawauchi and Namie are indicated on the map as data from those locations will be discussed in Section 3.4.1 and 3.4.2.

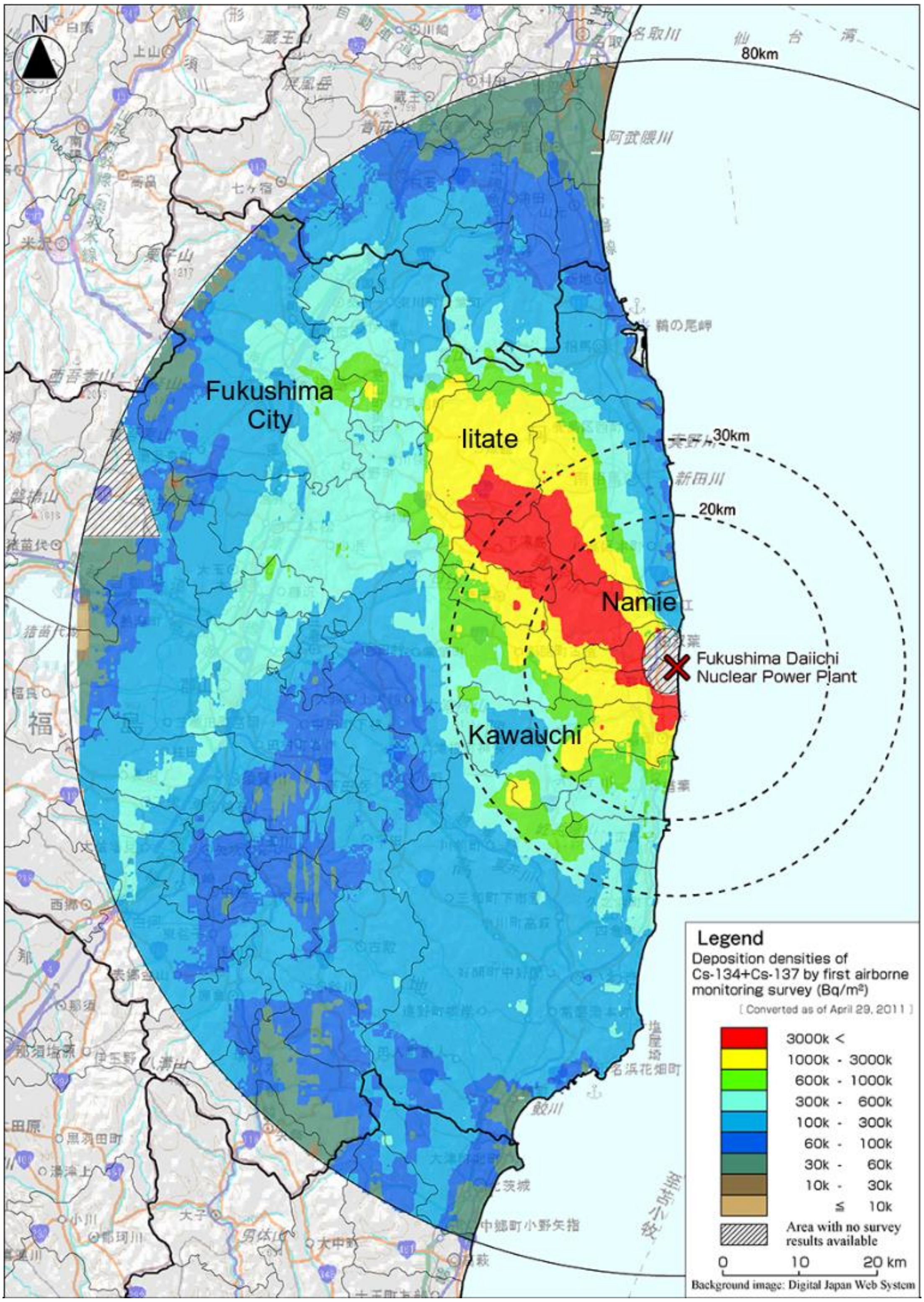


Figure 15 The distribution of ^{137}Cs and ^{134}Cs as of 23 April 2011 in the eastern half of Fukushima Prefecture. Image from Japan Atomic Energy Agency (2013).

Figure 16 shows the average monthly temperature, precipitation and snow depth in Fukushima Prefecture for the time period 2010-2022 (Japan Meteorological Agency, 2023). The average annual precipitation in Fukushima Prefecture is about 1200 mm, but the monthly averages may vary substantially, especially at the occurrence of typhoons during the summer season. During a typhoon or other severe weather there may be more than 100 mm of rain on an individual day. This results in drastic variations in local river runoff (Lepage, 2015) and due to the fact that most watersheds are in mountainous terrain the level of erosion may be substantial. Furthermore, the topography of the prefecture leads to substantial local variation. The long-term trend for weather data is that the average temperature has increased with about 2 °C over the last hundred years, while precipitation and snow depths have been rather constant, although individual years may vary (Japan Meteorological Agency, 2023).

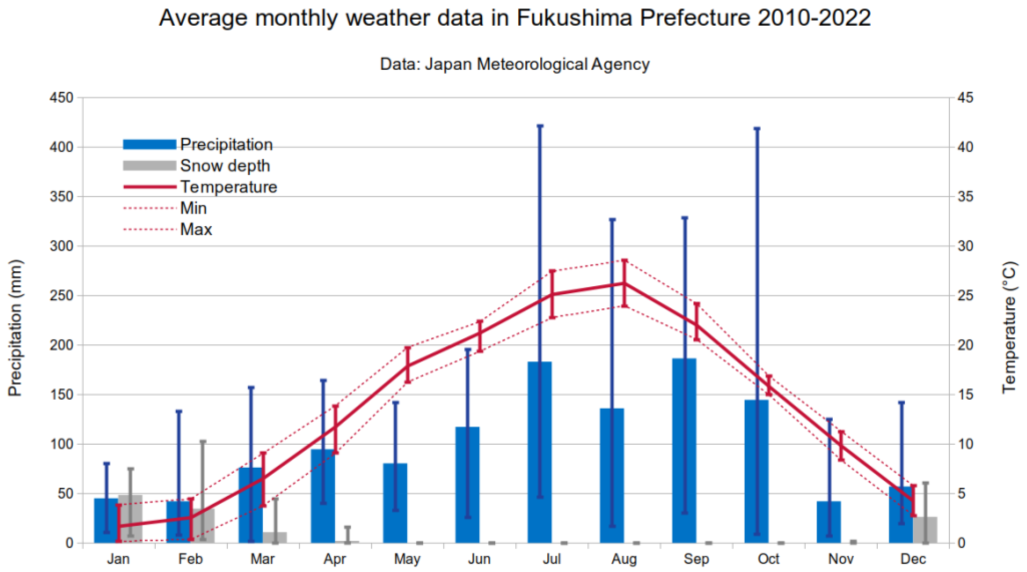


Figure 16 Average monthly weather data for Fukushima Prefecture for the time period 2010-2022. The error bars show the maximum and minimum values within the time period. Data from Japan Meteorological Agency.

There is a large amount of scientific literature on studying the distribution and movement of radionuclides after the FDNPP accident. The focus here is on ^{137}Cs as it is the radionuclide of relevance for longer time periods, and the one that is easiest to compare with data from other parts of the world.

Due to the relatively short time span since the accident (12 years) about 75 percent of the ^{137}Cs is still available in the ecosystem when considering the physical half-life. However, the ecological half-life may be substantially shorter due to precipitation, weathering and erosion.

In spite of the vast amount of data there are challenges to couple the data to weather patterns. A few reasons are given here.

- Some data have been gathered rather intensely for individual years, but without follow-up in the same area at later years. Therefore, it is difficult to establish time trends, though it varies depending on which kind of study that is performed.
- For mushroom there are certain general time trends that can be observed (Figure 17), but when looking at municipalities and for specific species, data may be

scarce. Furthermore, in cases where there are very high levels of ^{137}Cs it tends to come from single samples of some species that is not usually gathered.

- Soil data and water runoff within watersheds may be affected by the dedicated decontamination efforts that have been organized by the Japanese government and local municipalities. There are a number of studies looking into the effects of the decontamination and an important factor is the soil type, which determines how far down the radionuclides have migrated before decontamination (Takahashi et al., 2015, 2018). The decontamination efforts clearly disturb the signal for studies on weather related correlation with the release or reactivation of ^{137}Cs. However, the decontamination efforts focus on villages and agricultural land while forested areas tend not to be decontaminated, or limited to the edges of forests facing inhabited areas, although more extensive decontamination efforts of forested areas are considered (Evrard et al., 2023). Furthermore, as 70% of Fukushima Prefecture is forested area there are large incentives for performing such studies.
- The combined effect of scattered data, decontamination efforts, and the fact that not long enough time has passed since the FDNPP accident, makes it difficult to find clear patterns related to different weather factors such as precipitation and temperature. It is therefore difficult at this time to identify anomalous time trends similar to what has been seen in data from Sweden and Bavaria.

3.4.1 Mushroom

In 2012 the Japanese Government decreased the regulation limit for ^{137}Cs in food to 100 Bq/kg. In general, a very low fraction of agricultural products, home-grown or grown by farmers, have activities above that limit. For mushroom, which are picked in forests, the situation is very different, with a large fraction of the picked mushroom having higher values (Orita et al., 2016; Kunii et al., 2018). A general trend is that mycorrhizal mushroom have higher levels of ^{137}Cs than saprobiontic mushroom (Prand-Stritzko & Steinhauser, 2018) and that the levels tend to decrease slower than the levels in the corresponding litter and upper soil layers (Yamada, 2023). This can be understood as being due to that the mycorrhiza extends down into the ground and manages to extract ^{137}Cs at almost any depth in the surrounding soil, similar to what is observed in Bavaria (section 3.3).

In Figure 17 the ^{137}Cs activity for five different species of mushroom are shown for the time period 2011 to 2017 in Fukushima Prefecture (Hashimoto et al. 2020). It is tempting to interpret the downward trend as an effect of effective ecological half-life, i.e. the combination of physical half-life and decrease of cesium content due to transfer through different natural processes. But data is relatively scarce and in this case there is no detailed information about the location, so it cannot be correlated to the ground deposition.

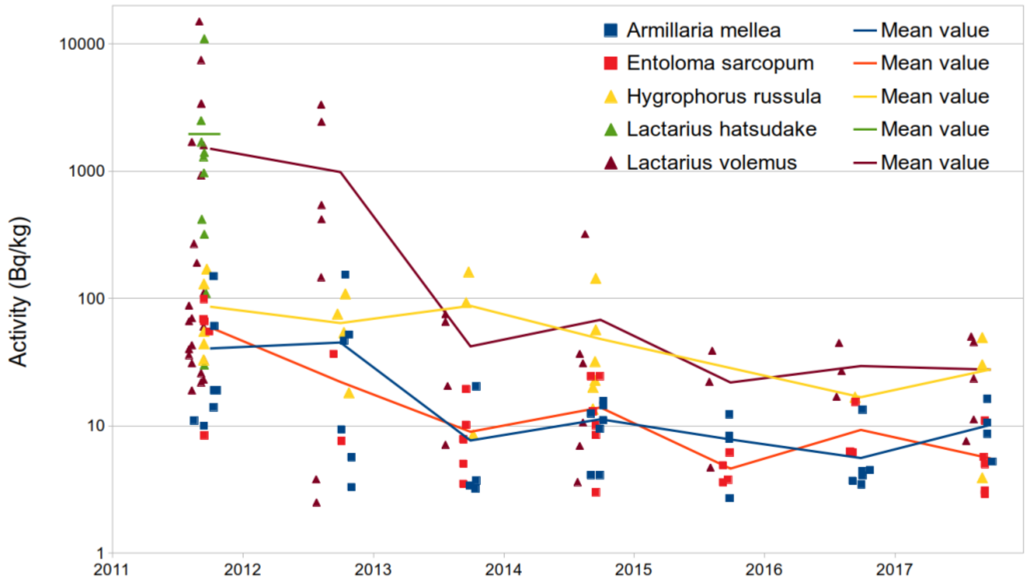


Figure 17 ^{137}Cs for four species in different parts of Fukushima Prefecture 2011-2017 (Hashimoto, et al., 2020). The solid lines indicate the arithmetic mean values for each species.

Figure 18 shows data from a series of studies performed in Kawauchi Village (Nakashima et al., 2015; Orita et al., 2017; Cui et al., 2020) where different species of mushroom were collected in the hilly forests over seven years. In Figure 15 Kawauchi Village is indicated on the map. Here it is difficult to see any trend at all, and as the number of samples vary and the spread in the activity has a very wide range, therefore caution is advised. The data has not been corrected with respect to activity, and as the local residents do not want to reveal where they find their mushroom it is not possible to derive transfer factors from this data set. However, under the assumption that the samples have been collected relatively uniformly, it is clear that the mushroom extracts ^{137}Cs from all depths, alternatively that the downward migration of ^{137}Cs is very slow in this region.

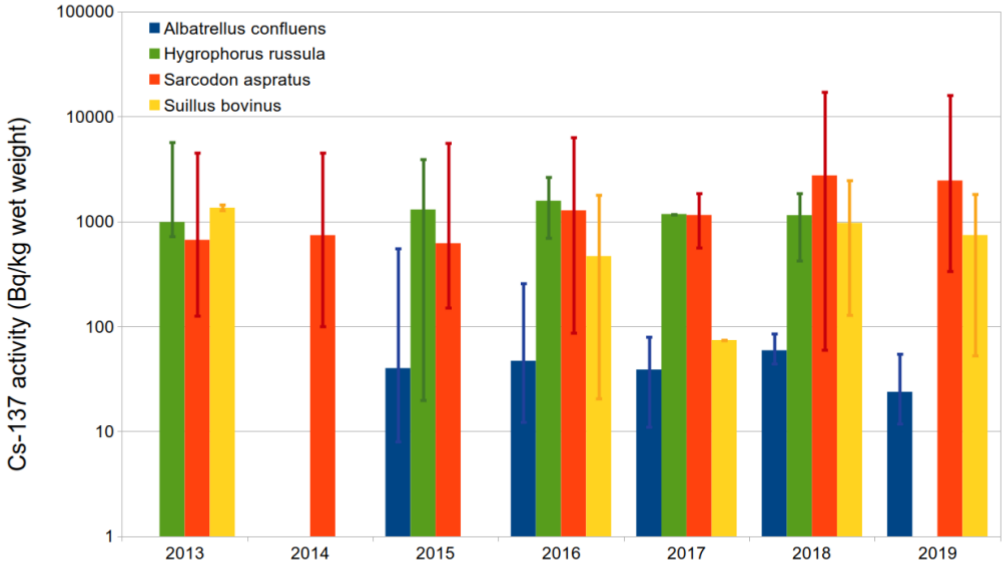


Figure 18 ^{137}Cs for four mushroom species collected 2013-2019 near Kawauchi Village, Fukushima Prefecture (Nakashima et al., 2015; Orita et al., 2017; Cui et al., 2020). The error bars indicate maximum and minimum values.

3.4.2 Litter and soil

There are a number of studies on soil samples where repeated sampling at the same locations have been performed. These studies with repeated sampling display patterns of downward migration of ^{137}Cs, the migration rate depends mainly on the soil type. Usually these studies are performed over too short time periods order to attempt correlating them to weather patterns (Takahashi, 2015, 2018), but may be valuable for such studies if repeated sampling is performed over the coming years. In one study, performed in 2011 before and after the first rainy season after the FDNPP accident, the effect of the rain on the ^{137}Cs downward migration turned out to be very low (Matsunaga et al. 2013). The reason was considered to be the physicochemical properties of the soils at the studied locations.

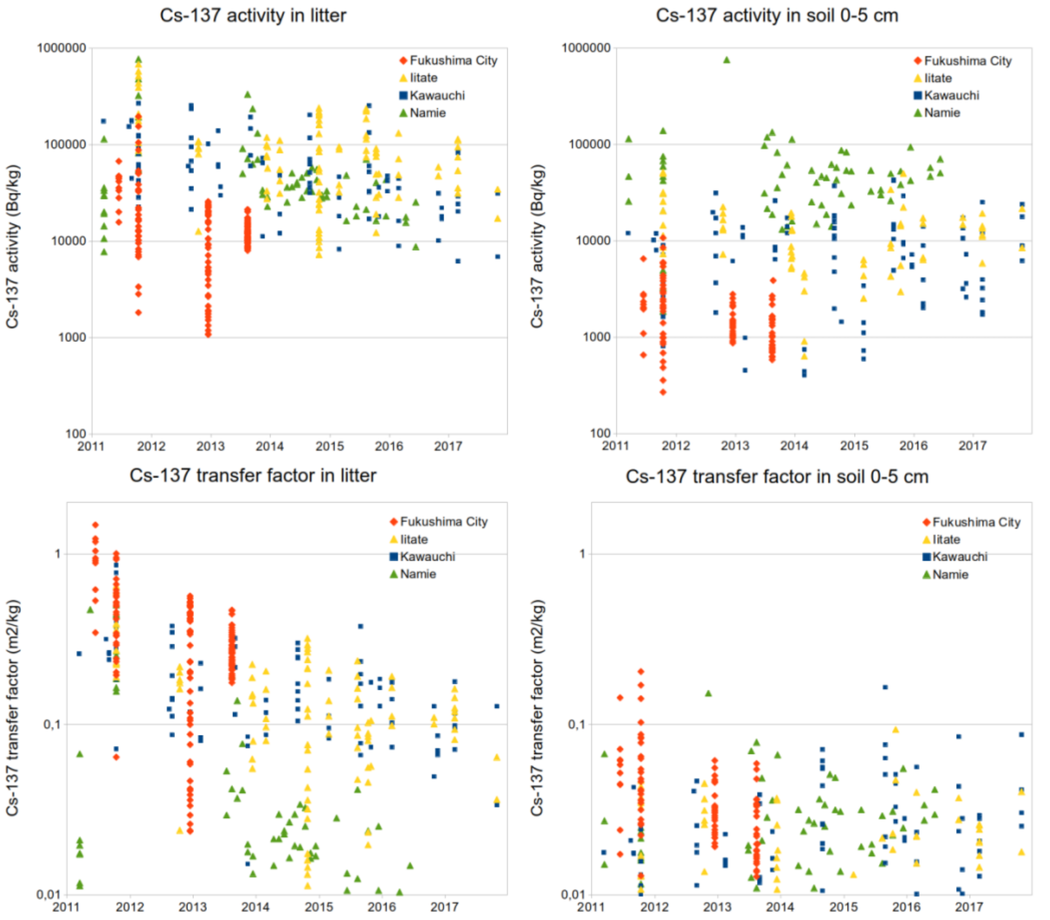


Figure 19 ^{137}Cs activity and transfer factors in litter and soil 0-5 cm in four different municipalities in Fukushima Prefecture over the time 2011-2017 (Hashimoto et al., 2020).

Litter and soil data display some general patterns, in Figure 19 the ^{137}Cs activity for litter and soil is displayed for four different municipalities in Fukushima Prefecture over the time period 2011-2017 (Hashimoto et al., 2020). The upper left panel shows the activity in litter, where there are downward trends for all four municipalities, while in upper soil layer 0-5 cm (upper right panel) there is no obvious trend in the activity. The lower panels show the activity in litter (left) and soil 0-5 cm (right) normalized to the ^{137}Cs ground deposition at each sample location (here somewhat incorrectly called transfer factor, as it is not a transfer from the soil to any living species). A reasonable interpretation is that the litter, originating from surrounding vegetation falling down on the ground, reflect the

^{137}Cs deposited on plants directly after the accident. This is an important contribution to ^{137}Cs activities initially and then decreases as there is no new deposition after 2011. The top soil layer displays lower ^{137}Cs activity than the litter, but it does not show any clear indication of being transported at the locations where the samples were taken, and the migration downwards is likely very slow.

3.4.3 Runoff in watersheds

There are a number of studies on how radionuclides have been transferred through different watersheds in Fukushima Prefecture, including how the transfer has been affected by different weather patterns (Chartin et al., 2013; Lepage, 2015; Evrard et al., 2015; Eyrolle-Boyer et al., 2016; Iwagami et al., 2017; Delmas et al., 2019; Hayashi et al., 2022; Feng et al., 2023; Ikenoue et al., 2023; Tatsuno et al., 2023; Tsuji et al., 2023; Yamasaki et al., 2023). Within the present project it has not been possible to explore this topic further in detail, but available data, which has been obtained or modeled with different approaches, are promising for further studies using the SWAT*Plus* model that has been used for the Kymijoki watershed (Nilsson et al., 2023b).

It is clear from data in the aforementioned studies that erosion related to intense precipitation, in particular during typhoons, plays an important role in the migration of ^{137}Cs in Fukushima Prefecture.

4. Conclusions & implications

For case studies from Finland, Sweden and Germany that were affected by fallout from Chernobyl, we have presented time series for ^{137}Cs activities in moose, mushrooms and surface deposition. These time series do not show systematic decreases expected from physical decay and environmental processes such as burial. Instead, the time series data reveal ^{137}Cs anomalies in specific years, 1994 to 1998 and 2007 locally in the Kymijoki catchment, 1997, 2003 and 2006 in Gävle, 2019 to 2021 in Bavaria. The 1997 ^{137}Cs anomaly in Gävle is also observed throughout Sweden. We have shown that ^{137}Cs activities in moose and mushrooms vary temporally and spatially tracing accumulation of ^{137}Cs in the environment. The accumulation of ^{137}Cs is associated with deposition of eroded material, and since erosion is influenced by rainfall, melt water and freeze thaw cycles, these weather needs to be considered. Anomalous ^{137}Cs activities are not associated with rainfall, temperature or river discharge that traces snow melt, however, there seems to be a connection between snow cover and freeze thaw cycles in Gävle in 1997 compared to 1996. Given that temperature and precipitation rates will vary in northern and central Europe during climate change, we suggest that climate change will lead to enhanced erosion, which may well increase accumulation of ^{137}Cs at specific locations that are sensitive accumulation zones in Gävle, Sweden and Bavaria, Germany.

Data on ^{137}Cs activity for the first decade after the nuclear accident in Japan show no clear patterns that indicate an influence of varying weather. Part of the reason is that plenty of data that was accumulated in the first years after the accident have been gathered in order to resolve immediate societal needs. Some of those research efforts have not been pursued after the acute phase, and this makes interpretations of time trends more difficult. However, more data are continuously gathered, which will enable more comprehensible studies in the coming years. One exception is that heavy precipitation in relation to typhoons has a large impact on erosion and thus also for the migration of radionuclides within watersheds. Data from such studies will be valuable for further use and development of the SWAT*Plus* model.

Prediction of erosion and accumulation of ^{137}Cs for a given season based on weather conditions is likely to remain challenging, because of complex environmental systems and pathways, uncertain source areas for erosion and the interplay of many factors that potentially cause erosion. However, this study contributes to identification of sensitive accumulation zones, allowing for recommendations of areas to avoid hunting game and gathering mushrooms and berries to be considered based on the extent of the hazard. For instance some localities in Bavaria have high probability of ^{137}Cs activities that exceed the 1500 Bq/kg recommended for regular consumption (livsmedelverket), whereas in Sweden and Finland the radiation in these sensitive areas of accumulation is typically not so hazardous. Considering these sensitive zones with respect to landuse and agricultural activities, areas of ^{137}Cs accumulation will also receive episodic inputs of remobilized ^{137}Cs, however like the moose habitat in Sweden that is likely not much of a concern. Construction activities also excavate the soil and till, in the case of the area around lake Hille, it may be worth assessing and taking into account radiation risks for new sites of development due to ^{137}Cs accumulation and subsurface burial. In Bavaria the authorities may also want to consider the risks posed by new agricultural and construction activities in the areas that are sensitive to ^{137}Cs accumulation.

5. Outlook and further research

We plan to follow up the preliminary SWAT*Plus* model for the Kymijoki watershed in southern Finland by calibrating and validating the hydrological outputs. Subsequently, we would pursue implementation of the ^{137}Cs module developed during this project and the associated calibration and validation. Potential future work with the modelling includes application to other areas of ^{137}Cs fallout, including some of the watersheds in Fukushima Prefecture. Further development of the code could include K as a trace element and ^{40}K as well as predictive modelling of hydrological catchments for future climate scenarios.

All the monitoring of ^{137}Cs activities since the Chernobyl and Fukushima accidents and related radioactive fallout has provided extensive datasets on diverse media. To make the most of this resource together with spatial variations in the landscape, availability of soil and till parameters and geochemical data, in addition to high temporal resolution weather records requires techniques that can handle and integrate big data from a variety of sources and on different spatial and temporal scales. That is the challenge that lies ahead.

6. Acknowledgements

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