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^{137}Cs activity in Sweden after the Chernobyl Nuclear Power Plant accident in relation to quaternary geology and land use

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ABSTRACT

After the Chernobyl Nuclear Power Plant accident ^{137}Cs was deposited in Mid-Sweden, with the highest activity in Gävle municipality. We compared aerial measurements of ^{137}Cs on the ground over time in Gävle municipality since 1986. Maps of soil types and land use were used to analyse the decrease in activity of ^{137}Cs. A spatial pattern was identified where the reduction of activity was greater in sand than in organic soil. For land use the reduction was greater in urban areas than in forests, with agricultural land in between. Long-term external radiation from ^{137}Cs depends not only on the physical decay of ^{137}Cs but also indirectly on geology, and this should be considered when calculating external radiation doses to individuals in epidemiological studies.

1. Introduction

On 26 April 1986 an accident occurred at the Chernobyl Nuclear Power Plant in Ukraine and a large amount of radioactive material was released. The highest ground deposition of ^{137}Cs was recorded in Belarus, Russia and Ukraine (De Court and Tsaturov, 1996). The most important radionuclides in the medium to long term after a nuclear power plant accident are ^{137}Cs and ^{90}Sr. The deposition of radionuclides may occur either as aerosols or as (fuel core) particles. Aerosols can more easily dissolve and migrate into the soil layers compared to particles after deposition on the ground. In total 85 PBq ^{137}Cs was released from the Chernobyl reactor core (United Nations Scientific Committee on the Effects of Atomic Radiation, 2000).

Five per cent (4.25 PBq) of the total released ^{137}Cs from the Chernobyl reactor was deposited in Sweden in the ensuing days, especially during the heavy rainfall on April 28–29. The ground deposition was unequally distributed, with the highest activity in the eastern coastal regions from Stockholm in the south to Umeå in the north (Mattsson and Moberg, 1991). The municipality of Gävle (87,431 inhabitants in 1986) consists of a city surrounded by rural areas dominated by forests, but also by lakes and agricultural land.

The municipality of Gävle has been mapped by the Swedish Geological Survey (Lundegårdh, 1967; Lundqvist, 1963). The area is a low lying coastal plain which was entirely flooded by the sea after the latest glaciation. The landscape has rather low relief without any dominant mountains or deep valleys. The underlying bedrock has eroded to a Precambrian peneplane leaving a tiny remnant of Ordovician limestone on the small island of Limön in the Bay of Gävle. The youngest Precambrian rocks are Jotnian sandstones, stretching in a narrow zone inland from Gävle for some 25 km. South of Gävle there are alternating zones of granites, grano-diorites and rhyolites. North of the Jotnian sandstone, granites and rhyolites occupy a narrow east-west zone which then, to the north, are replaced by a larger area of mica schists and metagraywackes. These metamorphic sediments become interspersed with metamorphic granitoids and rhyolites in the North. All over the area small intrusions of gabbro, andesite and basalt occur, with a similar age as the surrounding bedrock. Mineralisations are present, although on a small scale.

The dominant soil type in the municipality of Gävle is moraine, a large part of which is rich in boulders. The moraine is usually of a sandy type with fine sands and silt dominating at higher elevations. All moraines in this area have been inundated by the Baltic sea in historical time. This explains the absence of the finest fractions in most of the moraines. Another reason is the lack of soft host rocks north of the moraine deposits. As clayey moraines are absent, almost all clays are connected to the glaciofluvial deposits, of which there are several in the area. The largest is the Gävle Esker [Gävleåsen in Swedish], which enters the area in the southwest and then strikes northwards for some 25 km. The Gävle Esker then turns to the northeast and then again to the north, ending in a delta near the present seashore at Trödje, about

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halfway between Gävle city and Hamrånge. A smaller esker runs to the north-northwest from Hamrånge for some 20 km. The eskers are surrounded by glacial and postglacial deposits, of sand and clay. All these glacifluvial deposits, especially the eskers, are very water permeable and subject to vigorous flows of groundwater. Another major soiltype in Gävle municipality is organic soil, in this area often divided into fens, bogs and mires. The largest concentration of bogs is found in the forested plains south of Gävle city. This area is flat, hence these organic deposits are characterized by stagnant groundwater. Organic soil is also found in the northern part of the municipality, but more patchy and dispersed.

Knowledge of the dynamics of radio-caesium on the soil surface and its downward migration is important for predicting the external radiation dose and the degree of food-chain transfer to humans. Therefore, in this study we compared two datasets of aerial measurements of ^{137}Cs to investigate the relative importance of soil types and land use in predicting the fixation in the ground.

2. Materials and methods

2.1. Aerial measurements

At the request of the Swedish Radiation Safety Authority, the Geological Survey of Sweden did initial aerial measurements of ^{137}Cs, on 1–8 May 1986, in the Gävle area. The nominal flight altitude was 150 m above ground, and the airplane was equipped with four NaI (T1) crystals measuring the gamma-spectrum from the ground, flying with at a line distance of 50,000 m apart. At this altitude, 90% of the registered radiation originates from an area having a radius of approximately 300 m from a point directly below the plane. The measurement program was extended to cover Sweden, from 9 May to 19 June, with a line distance of 50,000 m and, in certain areas with large fallout, the line distance was 2000–10,000 m. In our study we produced a map of ^{137}Cs fallout over the Gävle municipality based on the aerial surveys from 14 May to 6 June 1986 (Lindén and Mellander, 1986).

Since 1986 the Geological Survey of Sweden has continued to perform annual aerial measurements of ^{137}Cs over different parts of Sweden at a lower altitude of 30 or 60 m above the ground. The dataset used in our analyses includes measurements until 2010. Considering all measurements over time the line spacing could be reduced down to 200–5000 m over Sweden, with the most narrow line spacing in areas with the highest deposition of ^{137}Cs. With the results from all these measurements, a database was created with 9.9 million measurements of ^{137}Cs in Bq/m^2 in a 200 × 200 m grid backdated to 1 May 1986 to account for the physical decay (Byström, 2000).

We have compared these two datasets, recognizing the first as an instant measurement of high deposition and the second as a time-integrated and more detailed measurement. The first dataset was kindly provided to us by Sören Byström, who was responsible for the measurements at the Geological Survey of Sweden, and the second dataset was provided by the Swedish Radiation Safety Authority. We restricted the study area to Gävle municipality (1600 km^2) as the municipality with the highest average of ^{137}Cs in Sweden, 51,830 Bq/m^2, calculated from the second dataset by the Swedish Radiation Safety Authority, after excluding water surfaces.

2.2. Quaternary geological map

A digital map on soil types in the Gävle municipality was provided to us from the Geological Survey of Sweden. In the geological map four categories of soil could be used for the analyses: clay, organic, sand and moraine. Hence, we omitted data points from water, rock and unclassified soils in all our statistical analyses.

2.3. Land use map

A map of land use for the Gävle municipality was provided to us by the Swedish National Land Survey. The land use map is based on satellite images over Sweden in the year 2000 and presented in 86 mutually exclusive categories. We subsumed these categories into four larger entities: urban and park, agricultural, mixed forest, and coniferous forest, respectively.

2.4. Statistical methods

The measurements of ^{137}Cs in the grid points were randomly sampled in order to obtain reasonably even coverage over the municipality. The reason for the random sampling was the huge number of observations, especially in the second data set, and the various distances between the flight paths. Due to scarcity of aerial measurements in the initial data set in the southern part of Gävle municipality, Hedesunda parish was excluded from the statistical analyses of soil and land use in relation to ^{137}Cs. The random sampling was repeated several times to control for any discrepancies in sampling. The next step was to calculate variograms for both datasets (Cressie, 1993; Cressie and Wikle, 2011). These were produced both as isotropic and anisotropic variograms. The rational for the anisotropic variogram was the west-to-east increasing ground deposition of ^{137}Cs.

The variograms were then used in the geostatistical models to interpolate the data onto regular grids. The routine for conventional kriging, “krige.conv” in the library “GeoR” (R Core Team., 2015), is a grid setting of 500 × 500 m. This might seem to be a very coarse grid, but choosing any smaller values would render too many grid points for the program, written in ‘R’, to handle. Instead, a second interpolation inside the GIS program was chosen. The two resulting grids were then fed into a GIS program (QGIS Development Team, 2015) and put through another round of interpolations. This time a grid setting of 100 × 100 m was used to produce the two digital maps used for the analyses, that is, one map created from the initial measurements and a second map from the time-integrated database. With a geometric join the two grid maps were interpolated, and the measurements in the second map were subtracted from the first map at each grid point. The differences obtained were expressed in per cent of the initial ^{137}Cs value. Only in a few cases were small negative values obtained, and these were set as *nil*.

The next step was to super-impose this map of differences onto the maps of quaternary geology and land use, in a three-layer analysis. The resulting dataset could be used to calculate basic statistics about soil types, land use and their interactions. The values of the percentage differences were submitted to rather more advanced methods due to the complicated distributions present. A Bayesian method was therefore used in the statistical analysis as superior in precision to predict medians and 2.5-percentiles, compared to classical non-parametric statistics (Lunn et al., 2013). Non-overlapping prediction intervals were regarded as statistically significant.

3. Results

The distribution of the fallout of ^{137}Cs in Sweden and Gävle municipality is presented in Fig. 1.

Comparison between the initial and the time-integrated dataset is given in Table 1.

The number of measurements in the initial dataset of 4717 represents about 95% of the total measurements. A random selection was made in the time-integrated data set, with 22,788 measurements (30%). A histogram of grid point values of ^{137}Cs from the two data sets clearly shows different distributions (Fig. 2).

Subtracting these two datasets resulted in a map expressed as percentage change of the initial value at each grid point (Fig. 3). A small difference between the two data sets indicates retention in the soil

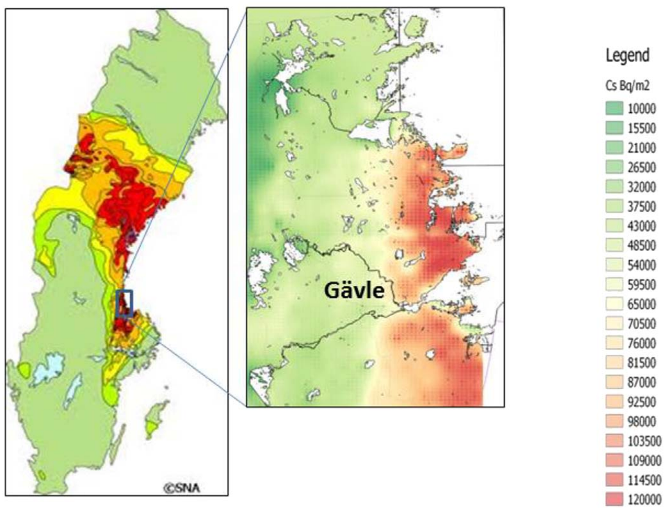

Fig. 1. Map on the fallout of ^{137}Cs (Bq/m^2) over Sweden with Gävle municality indicated.

Table 1
Comparison of the two datasets for Gävle municipality (excluding Hedesunda parish) used in the analyses.

Measurements	Line spacing (metres)	Data points (n)	^{137}Cs (Bq/m^2)		
			Average	SD	Min – max
Initial 14 May–6 June 1986	2000	4717	50,412	33,525	0 - 164,910
Time-integrated 1986–2010	200	22,788	46,772	27,556	0 - 156,490

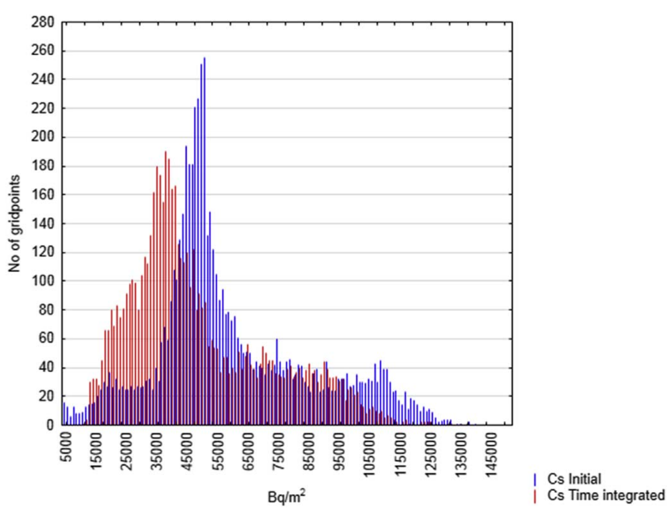

Fig. 2. Histograms of grid point values of ^{137}Cs (Bq/m^2) in Gävle municipality (excluding Hedesunda parish) from the initial (blue) and the time-integrated (red) measurements, respectively.

(white), and a large difference could be explained by wash-off from hard surfaces and/or deep migration into the soil (black).

In Table 2 the results from analysing the distribution of soil types and land use are presented in categories. Moraine (45%) and coniferous forest (62%) is totally dominating soil and land use, respectively. Coniferous forest prefers to grow on moraine, and mixed forest is more common on organic soil. A relatively large proportion of the soil types (16%) could not be classified into the major soil categories. The landuse category urban and park consists of virtually everything man-made, from built up city environments to parks, roads, industrial areas and villages. Agricultural areas, like fields and grasslands, cover a rather small portion, only a mere 4% of Gävle municipality.

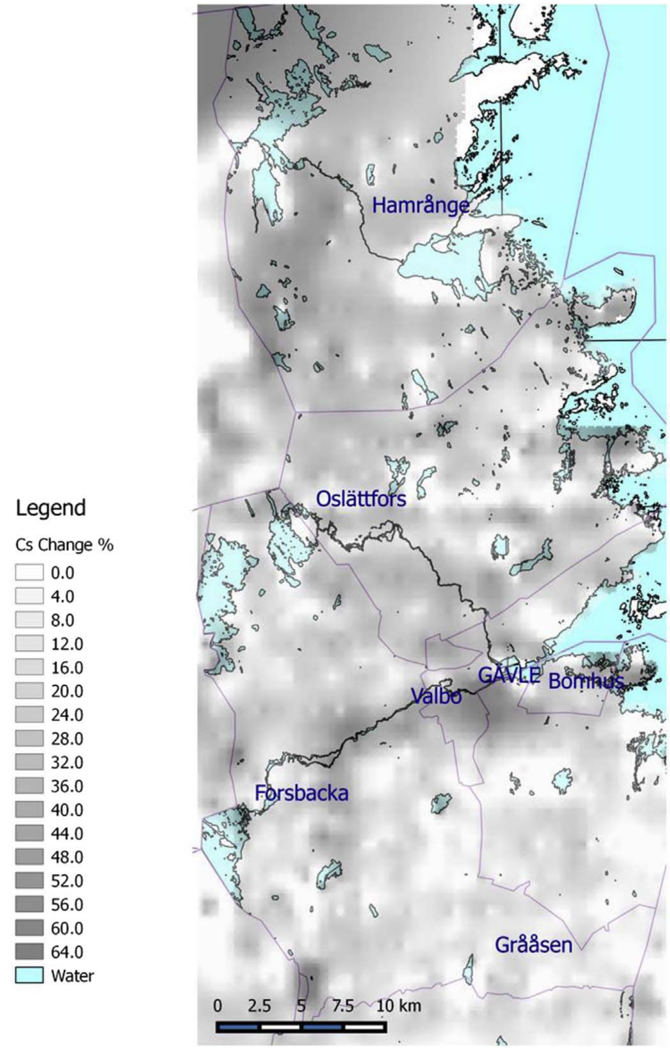

Fig. 3. Map of the difference between the initial and the time integrated measurements at each grid point expressed as percent change.

Table 3 is a cross-tabulation of soil type and land use with the reduction of ^{137}Cs expressed by the median value in each combination. Overall, organic soil has the lowest and sand the highest median value, indicating that the ^{137}Cs remained to a higher extent in the organic topsoil than in the sand. Also, human influence seemed to be rather important, as the lowest median value was seen in the forest compared to the highest median value in the urban and park, with agricultural land in between the two.

In Fig. 4 soil types stratified for land use are shown with median values and 2.5-percentiles. Urban and park have the highest median values regardless of soil type. Clay and sand showed similar patterns comparing land use, but organic soil suffered from low statistical power when stratified for land use, as some combinations became rare. Moraine, on the other hand, showed a relative narrow range of median values (20–25%), not so sensitive to stratification for land use.

4. Discussion

To the best of our knowledge this is the first time aerial measurements of ^{137}Cs after the Chernobyl Nuclear Power Plant accident have been used to analyse the weathering in relation to soil types and land use, respectively. According to our results, organic soil has the strongest and sandy soil the weakest retention and the land use urban and park

Table 2
Distribution of soil types and land use in Gävle municipality, respectively. Subcategories from the geological map (soil types) and land use map subsumed in broader category for this study.

	Categories	Area in km^2 (%)	Subcategories from map
Soil types	Moraine	720 (45)	Moraine (various), landfill
	Unclassified	256 (16)	
	Organic	224 (14)	Peat, mud
	Sand	176 (11)	Postglacial sand, washed sedimentary deposits, esker sediments.
	Water	96 (6)	
	Clay	80 (5)	Glacial clay, clay silt, postglacial clay
	Rock	48 (3)	Bedrock, igneous
	Total soil	1600 (100)	
Land use	Coniferous forest	992 (62)	Coniferous forests and clearings
	Mixed forest	208 (13)	Mixed and deciduous forests
	Water	144 (9)	Rivers, lakes, coastal waters
	Unclassified	32 (2)	
	Urban and park	80 (6)	Built up areas, parks, roads, railways etc.
	Agricultural	64 (4)	Fields, pastures
	Wetland	64 (4)	Sparsely forested mires and bogs
	Total land use	1584 (100)	

Table 3
Reduction of ^{137}Cs in per cent between the initial and the time-integrated measurements in relation to soil types stratified for land use. Bayesian estimation is used to calculate the medians.

		Soil type (median values)				
		Clay	Organic	Sand	Moraine	Total
Land use	Urban and park	37.38	22.25	38.18	26.91	31.11
	Agricultural	25.09	15.70	29.46	24.04	26.33
	Mixed forest	20.91	16.78	21.88	19.68	19.32
	Coniferous forest	17.15	18.99	18.89	20.56	20.23
	Total	24.61	18.48	26.64	20.85	21.20

has the highest and forest the lowest weathering.

The term weathering can be defined as the reduction of exposure rate due to natural removal phenomena such as run-off, resuspension and migration as well as the reduction of exposure rate due to human activities such as road sweeping and traffic (Karlberg, 1987). Migration of ^{137}Cs in the soil depends on several factors, such as precipitation (rain and snow), mineral content, porosity, and absorption property of the soil. High fixation rates are seen in clay minerals and in soils having high content of organic matter, which results in a slow vertical migration speed. The mobility of caesium has, as expected, generally been found to be considerably greater in peaty soils than in mineral soils. The vertical migration is affected by the land use, that is, ploughing of arable land will result in a more homogeneous distribution, compared to forest with a much slower mobility of ^{137}Cs. The activity on the surface will decrease with time by physical decay, plant uptake, accumulation of soil, and downward migration (Andersson, 2009).

The strong retention in organic soil, which has also been shown in previous studies, is explained by the higher ion binding properties in organic material (Forsberg and Strandmark, 2001). The soil type with the highest organic content in Gävle is mires. In Belarus it has been found that ^{137}Cs was removed from a peatbog system at a slow rate of 0.3% per year, 8 years after the Chernobyl Nuclear Power Plant accident (Kudelsky et al., 1996). In a raised open bog in Sweden with a ^{137}Cs ground deposition of 23,000 Bq/m^2, the migration rate was 0.57 cm per year. In this bog 50 percent of the total ^{137}Cs inventory remained in the 0–6 cm top layer in the year 2005 (Rosén et al., 2009). In Scotland the migration velocity in heather moors was 0.34–0.72 cm per year (Shand et al., 2013). Unfortunately, mires were too rare to be analysed separately in our study.

Mineral soils also have a high capacity to bind ^{137}Cs. In 33 sampling sites in Sweden the vertical migration was slow, 0–0.35 cm per year (Almgren and Isaksson, 2006). The migration velocity (cm per year) was calculated at 0.18–0.37 in mineral soils and 0.23–0.59 in organic soils (Rosén et al., 1999). Our results could not be directly compared to those of Rosén et al. because of different classification of soils, but it seems that we could not find support for our results in their migration rates. Somewhat surprisingly, we found a faster weathering rate in clay soil, which has been related to slow migration of ^{137}Cs (Forsberg and Strandmark, 2001).

Ordinary hydrological models predict very slow migration in soil because of strong retention. However, a surprisingly fast migration has

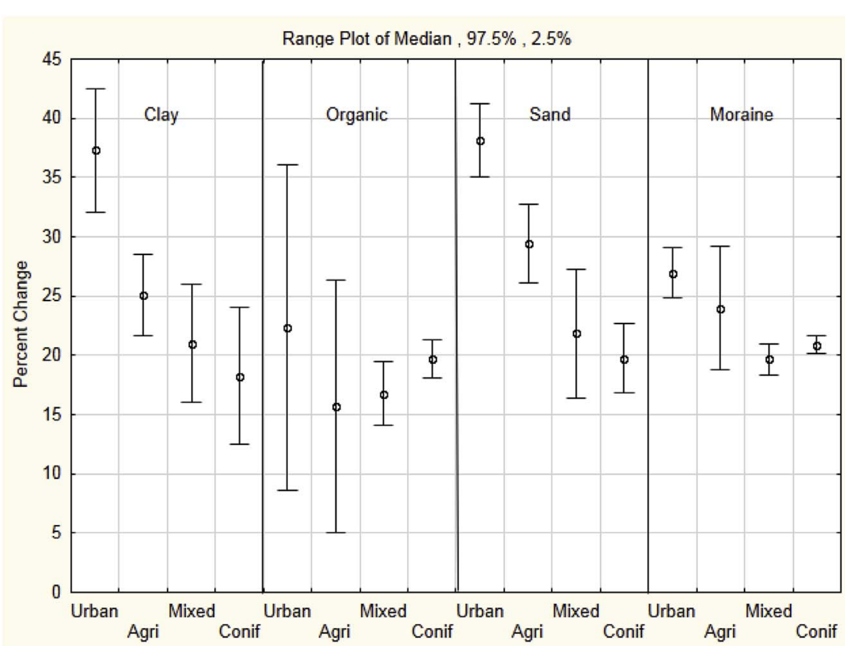

Fig. 4. Diagram of the reduction of ^{137}Cs in percent between the initial and the time-integrated measurements in relation to soil types and land use, respectively. Bayesian estimation is used to calculate the median values and the 2.5-percentiles.

been measured in field studies, suggesting mechanisms other than water solution, such as, colloidal transport (Bréchignac et al., 2000; Kirchner and Baumgartner, 1992). Since clay also consists of colloids, a faster transport might be at hand, as colloidal transport is mediated by mass-flow via flow paths such as cracks, root paths and worm holes. It can be expected that this colloidal transport would have the greatest influence initially (Bréchignac et al., 2000). This might explain why we observed a lower retention in clay soils. Migration can also be mediated via biological activity, root transportation, mycorhizal transport and soil migration through earth worms.

Because of fixation of ^{137}Cs in the soil, annual precipitation does not always alter this slow velocity. For example in western Norway, with an annual precipitation > 2000 mm per year, 20% of the activity still remains in the top soil in 2005 (Gjelsvik and Steinnes, 2013). Instead bioturbation through mixing of soil by earthworms or ploughing is generally a more important factor in terms of migration in soil, than precipitation. These are important factors contributing to the lower retention that we could relate to human activities of land use.

The migration velocity has been shown to be slow, about 0.26 cm per year, in undisturbed grassland (Schimmack and Feria Márquez, 2006; Schimmack and Schultz, 2006). In Sweden 8 sites of temporary and permanent grassland have been studied in relation to soil types with a ground deposition ranging from 14,000 to 184,000 Bq/m^2. Eight years after the Chernobyl Nuclear Power Plant accident most (50–92%) of the ^{137}Cs was still present in the upper 5 cm. In Sweden the reported effective half-lives in soil is 8–10 years in natural grassland (Isaksson et al., 2001) compared to 19 years in mountainous natural pastures with a dominating soil type of podsolized gravelly and sandy moraine, interfoliated by peat bogs (Andersson et al., 2001). Chemically, ^{137}Cs is found in its monovalent form Cs^{1+} and is very strongly bound, like other monovalent ions such as K^{1+} and NH^{4+}, to the so called frayed edge sites (FES) on illitic clay minerals and displacement only occurs via competitive ion exchange (Bolt et al., 1963; Cremers et al., 1988). Therefore, the use of fertilizers is a way of enhancing migration of caesium from the top layers of the soil, especially in combination with uptake in crowing crops. This is a possible explanation for our finding in this study of retention in agricultural land in between urban and forest land use.

In six samples from a Swedish forest it was shown that 85% of the total ^{137}Cs activity was in the upper 5 cm in 1991 (Fawaris and Johanson, 1994). Undisturbed forest is therefore one of the environments with the highest expected retentions, especially if low organic content in the soil, confirmed in our results with coniferous forest predominantly growing on moraine.

As expected, the land use urban and park had the overall largest reduction of ^{137}Cs as it entails hard surfaces, hence a more effective washout, in contrast to the strongest retention in the forest. In the urban setting the behaviour of radionuclides is somewhat different, compared to soil. A model of migration of ^{137}Cs has been developed based on the experience in Gävle including trees, paved areas, walls, roofs and non-paved areas in the model (Gallego, 2006). Even though streets are more easily washed off by rain, snow and cleaning, it took some 10 years of weathering for ^{137}Cs to be reduced to 2% of the initial activity (Andersson et al., 2002). Unfortunately, we could not differentiate between hard surfaces (streets, roofs, parking lots) in the city of Gävle and green areas with higher fixation (parks and lawns).

Radioactive nuclides deposited on a surface can be washed away or become airborne again in a process termed re-suspension. In the urban environment, vehicular activity has been regarded as possibly the most important mechanical action leading to re-suspension in many circumstances (Nicholson, 2009). In rural areas wind is important for re-suspension of deposited ^{137}Cs, until they are fixed in the ground.

In a recent study from Japan retention of ^{137}Cs has been studied in forests, agricultural lands and urban areas, as in our study (Adhiraga Pratama et al., 2015). They found that a significant amount of radiocaesium is transported both in liquid and solid states, from wash-off from the ground via river systems into the ocean. It would therefore be of interest to establish how much of ^{137}Cs in our case has been transported via the catchment area of the Gävle River, and how much has been transported via the Gävle esker, whose groundwater functions as a significant river. That might be established by analysing sediments from the Bay of Gävle and from the sea outside Trödje, respectively.

The strength of our study is the high resolution in 1) the ^{137}Cs exposure (200 × 200 m grid), 2) the geological map (scale 1:20,000–1:200,000) and 3) the land use map (resolution 25 m). This is rather unique and allows us to carry out very specific statistical analyses. However, due to a relatively small area (Gävle municipality) we had to aggregate both soil types and land use in broader categories, which limited the spatial resolution allowed by the primary data.

The most serious limitation is the time integrated fallout map, which did not allow us to study the fixation/weathering in more detail in relation to ground characteristics. Access to annual measurements would have made it possible to study the ecological decay of ^{137}Cs in relation to soil and land use. Due to the unexpected nature of the Chernobyl Nuclear Power Plant accident the preparedness for quick and accurate measurements in May was somewhat limited. Although there is an annual calibration programme for the instruments, they did not initially have maximum sensitivity, although they improved over time. As the first measurements were made, when the ^{137}Cs was still airborne, the outside of the aeroplane became contaminated, and it is not clear whether the adjustment could fully compensate for this unpredicted bias (Lindén and Mellander, 1986).

5. Conclusions

When backdating time series of ^{137}Cs measurements and from large areas it is important to take into account not only physical decay, but also soil types and land use. This is important when estimating the external radiation dose to man in areas with radioactive contamination after a nuclear accident. If this study could be regarded as a pilot study for a new cost effective risk assessment over time, it could be desirable to enlarge this study to the most contaminated counties, allowing for more specific soil and land use categories. This method could be considered as predicting spatial effective half times on the ground in counties with existing maps on geology and land use after a radionuclide incident.

Conflict of interest

The authors have no competing financial, personal or organizational interest.

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