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Ecological half-lives of ^{137}Cs and ^{90}Sr in dairy milk in regions with different soil types

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Abstract. Long-lived radionuclides ^{137}Cs and ^{90}Sr have been regularly monitored since the 1960's in dairy milk in Finland. The aim of this work was to study regional differences in long-term behaviour of ^{137}Cs and ^{90}Sr in dairy milk including the nuclear weapons tests period and the period after the Chernobyl fallout. In the milk production areas in Finland soil types vary from clayish to humus and peat soils. Although total ^{137}Cs deposition from nuclear weapon testings was approximately the same in all regions, ^{137}Cs concentrations in dairy milk during the 1960s ranged between 5 and 35 Bq/l being highest in regions with high proportion of peat soils. The uneven deposition after the Chernobyl accident before the start of the growing season gave rise to ^{137}Cs concentrations in dairy milk ranging from 3 Bq/l to 60 Bq/l. In the milk production areas dominated by organic soils the ecological half-lives for decrease of ^{137}Cs were higher in the regions with clayish soils. Half-lives of ^{90}Sr for longer time intervals were considerably longer than those for ^{137}Cs. The long-term data on behaviour of ^{137}Cs and ^{90}Sr in dairy milk can be utilized in testing food chain models.

1. INTRODUCTION

Milk is an important part of the diet in northern Europe and therefore it contributes significantly to the radiation dose received via ingestion after a radioactive fallout. Knowledge on the transfer of radionuclides to milk and on the trends in contamination levels in different environments for short and longer time periods after a radioactive fallout is required for estimation of ingestion doses in case of accidental releases of radionuclides to the environment. Milk is a sensitive indicator of radionuclide contamination because radionuclides are easily transferred to the grass used for feeding of cows. Milk sampling in dairies has significant advantages, resulting from the fact that the samples represent rather constant areas from which milk is delivered to the dairies. The radioactivity concentrations in dairy milk are independent of individual differences in the metabolism of a cow as well as slight differences of feeding of cows. The long-time constant surveillance programmes of dairy milk give information both for dose assessment and the time trends of radioactivity concentrations in milk in various environments.

STUK has regularly analysed long-lived radionuclides ^{137}Cs and ^{90}Sr since the 1960's in dairy milk in Finland. The sampling areas differ by soil types, nutrient contents of soils, climate, and also by the amount of ^{137}Cs deposition after the Chernobyl accident. The areas for this study were chosen to represent different soil types; sandy soils and peat and clay soils, which are known to be extreme in uptake of radiocesium.

Both the short-term decrease after the radionuclide deposition and long-term decline in contamination of milk are important for estimating the total exposure of fallout. In this study the ecological half-lives for decrease of ^{137}Cs and ^{90}Sr in milk after the period of nuclear weapon testings and after the Chernobyl accident were estimated.

2. MATERIAL AND METHODS

Data for this study was aggregated from the results of the dairy milk surveillance programme conducted by STUK [1-8]. The long-lived radionuclides ^{137}Cs and ^{90}Sr have been regularly analysed since the 1960's in dairy milk in Finland. The dairy milk sampling areas for this study were chosen to represent areas located in south-western, western, eastern and northern parts of Finland (Figure 1).

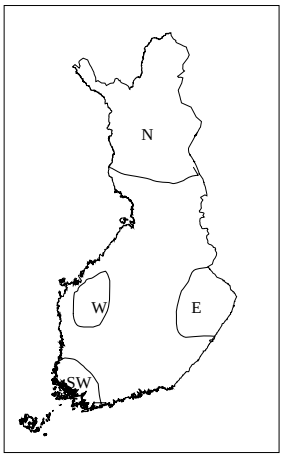

Figure 1. Sampling areas of dairy milk. N=northern, W= western, SW=south-western, E=eastern.

Milk sampling was started in south-western, northern and western areas in the beginning of the 1960's, and in the eastern area in 1983. The samples were taken monthly from milk powder factories in south-western area until 1986, in western area until 1992 and in eastern area in 1983-1984. Later on samples were taken weekly in dairies, and combined monthly for analysing. In the northern area weekly samples were taken in the dairy for the whole study period. The dairy milk samples represented the whole, rather constant, delivery areas of the dairies. The milk production areas where samples were taken, have remained nearly the same during the whole study period. The slight changes in production areas have not caused any significant variation in radionuclide concentrations of milk of the dairies.

The average annual precipitation in northern Finland is 500-550 mm per year as also is in western milk production area. Annual precipitation in eastern Finland ranges from 600 to 650 mm per year. In south-western area there is more variation in the precipitation rates, from 550 to 750 mm per year. In northern Finland the growing season is shorter, where the average annual temperature is from 0 to -3 ˚C, whereas in other areas it ranges from +2 to +5˚C.

The milk sampling areas differ in the types of cultivated soils [9]. The cultivated land in northern Finland is predominantly peat and humus soils; there are no clay soils in the area. The prevailing soil types are clay and sand in south-western area, and finesand and silt in western and eastern milk production areas. The western and eastern areas differ from each other by the proportion of humus soils, and the absence of clay soils in the eastern area (Table 1).

Table 1. Percentage proportions of different soil types of arable land in the milk production areas.

Soil type	SW	W	E	N
sand and fine sand moraine	5%	10%	19%	24%
finesand soils	2%	55%	35%	32%
silt soils	20%	2%	29%	-
clay soils (sandy and silty clay)	66%	7%	-	-
humus soils	6%	24%	12%	8%
Carex peat soils	-	-	-	33%

The pH of cultivated soils is lowest (5.3-5.6) in south-western area and highest in northern area (5.8-6.1). The calcium and potassium contents of cultivated land in the clayish south-western area are considerably higher than those in the other areas studied. Due to the changes in fertilization practises since 1960's, the potassium and calcium contents in the soils have increased gradually in all the studied areas. The contents of nutrients in cultivated land in northern Finland are still substantially lower than those in the other areas (Table 2).

Table 2. Characteristics of arable soils in the milk sampling areas in 1960-2000.

	SW	W	E	N
pH	5.3 - 5.6	5.5 - 5.7	5.5 - 5.8	5.8 - 6.1
Ca, mg/l				
in 1960-1990	1800 - 1900	1200 - 1300	1200 - 1300	800 - 1100
in 1990-2000	2200 - 2300	1400 - 1500	1400	1300 - 1400
K, mg/l	210 - 230	100 -130	100 - 110	80 - 100

^{137}Cs and ^{90}Sr depositions after the nuclear weapon testings period did not significantly differ between the areas studied; the depositions of ^{137}Cs and ^{90}Sr accumulated at the end of 1985 in Finland were 1.1 and 1.8 kBq m^{-2}, respectively. The highest deposition values were measured in 1963, decreasing rapidly by 1967, while during the 1970's they stayed low and rather constant [4].

^{137}Cs deposition after the Chernobyl accident in 1986 was most unevenly distributed all over the country (Figure 2) [10]. The average ^{137}Cs deposition (1.10.1987) was around 1.9 kBq m^{-2} in the northern, 2.5 kBq m^{-2} in the eastern, 19 kBq m^{-2} in the south-western and 26 kBq m^{-2} in the western milk production areas. The ^{90}Sr deposition after the Chernobyl accident was less than 0.5 kBq m^{-2} in all the areas, being highest in western and south-western areas. In the northern area almost no increase in the ^{90}Sr deposition was detected [11-12].

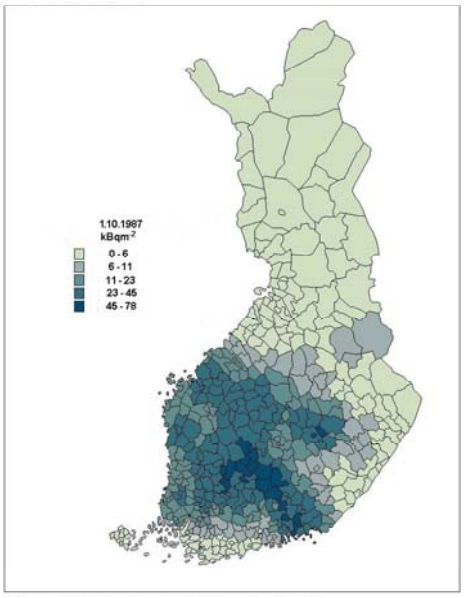

Figure 2. Chernobyl ^{137}Cs deposition in Finland (kBq m^{-2}) [10].

2.1 Data treatment

The concentrations of ^{137}Cs and ^{90}Sr measured from milk powder samples were converted to concentrations in liquid milk by using the average concentrations of calcium and potassium in Finnish milk (1.2 gCa l^{-1} and 1.58 gK l^{-1}). The ecological half-lives for the decrease of ^{137}Cs and ^{90}Sr concentrations in dairy milk were estimated using log-linear regression analysis performed with annual mean values. Time periods were chosen for short term to represent the years after the next growing seasons after the highest deposition, and for longer periods to correspond the years after the rapid decrease in concentrations.

3. RESULTS AND DISCUSSION

Although the total ^{137}Cs from nuclear weapon testings was approximately the same in all the areas, the ^{137}Cs concentrations in dairy milk during the 1960's and 1970's were significantly higher in the northern area and also to some extent higher in the eastern area than in the western areas (Figure 3.). Because the annual precipitation rates vary only between 500 – 700 mm per year there are other factors affecting the ^{137}Cs and ^{90}Sr concentrations in milk in different areas. The factors affecting the transfer of radionuclides from soil to plants are the amount of clay and humus, and contents of nutrients in soils among others. The transfer of ^{137}Cs into milk from northern soils was found to be more than 3-5 times higher than in intensively-cultivated clayish areas. This was mainly due to the high proportion of peat soils and nutrient deficiency of the pastures in northern Finland. The production of grass or hay per field area is less in nutrient deficient fields resulting that more area is needed for feeding cows, and also more contamination is thereby uptaken by cows.

The ^{137}Cs deposition after the Chernobyl accident was in northern Finland about the same as the deposition from nuclear weapon testings. However, the highest ^{137}Cs concentrations of milk were less than 4 Bq l^{-1} in northern Finland in 1986-1987, being considerably lower than the peak values 30 - 40 Bq l^{-1} in the 1960's [13]. The long-term pre-Chernobyl deposition with annual maximums in summer, resulted in higher contamination levels compared to the short-term Chernobyl deposition before the start of the growing season. The nutrient levels in the northern soils increased significantly since the 1960's, which also decreased the radionuclide intake of grass in later years. The increase in ^{137}Cs concentrations of dairy milk after the Chernobyl accident in 1986 in all the areas was nearly in proportion to the deposited ^{137}Cs. The ^{137}Cs concentrations of dairy milk in the growing season 1987 were in the south-western and eastern areas less than 60% and in western and northern areas 70-75% of the peak ^{137}Cs concentrations in 1986. The decline of ^{137}Cs contents in dairy milk was most rapid in the south-western clayish area. Although the deposition in south-western area was 7-10 times higher than in eastern and northern areas, the ^{137}Cs concentrations in milk decreased to the same level in all these areas by 1992, being since then lowest in the south-western area.

Figure 3. Monthly ^{137}Cs concentrations (Bq l^{-1}) in dairy milk in four sampling areas in Finland 1960-2003.

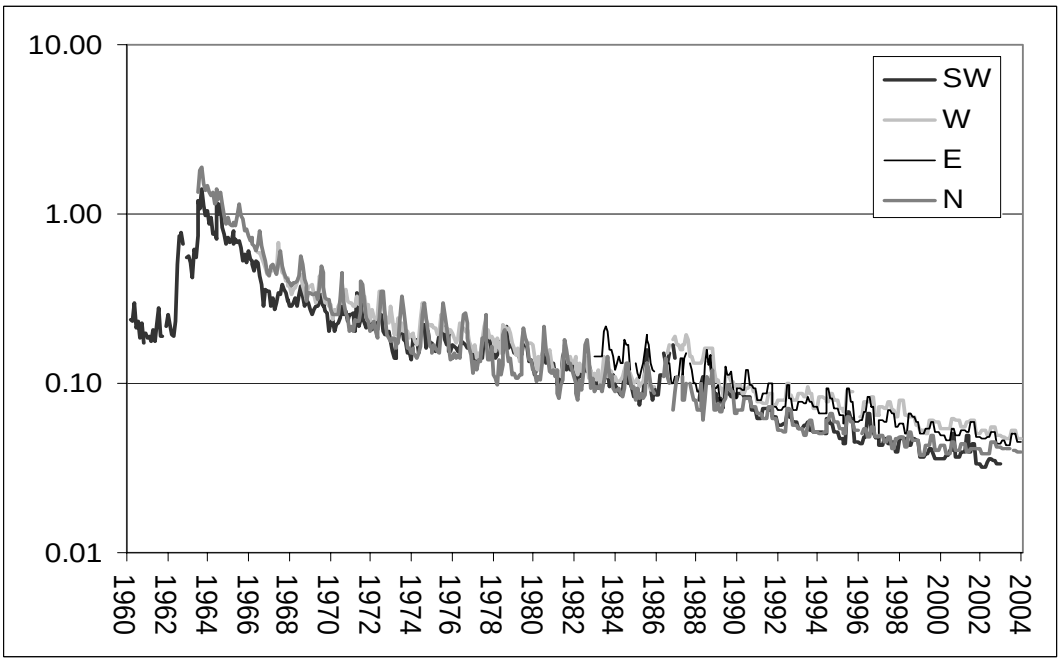

Figure 4. Monthly ^{90}Sr concentrations (Bq l^{-1}) in dairy milk in four sampling areas in Finland 1960-2003.

The ^{90}Sr in milk in Finland mainly originates from the fallout of the nuclear weapon testings. The ^{90}Sr deposition after the Chernobyl accident was low compared to the earlier fallout. The slight increase in contents of ^{90}Sr in milk was seen in the western and south-western areas with a higher ^{90}Sr deposition;

in the other areas the increase was negligible (Figure 4). The ^{90}Sr concentrations in dairy milk were highest in eastern and western areas, where the proportion of sandy soils is also higher. One reason to slightly higher concentrations of ^{90}Sr in milk in northern Finland after the nuclear weapons testing fallout may be the low calcium contents of pastures before the 1990's.

The clay in soils in south-western and western areas binds cesium more efficiently, which is seen in the proportion of ^{137}Cs and ^{90}Sr concentrations in milk after the nuclear testings period. The Cs/Sr proportion was 10-20 in the northern area during 1970-1980, versus 2-3 in the south-western area and 3-5 in western area. After the Chernobyl deposition, in the 1990's after the rapid decrease since 1986, the proportion of Cs/Sr was 6-12 in the south-western and eastern areas, slightly higher 8-15 in the northern area, and in the western area, which was exposed to highest deposition, 22-30. This proportion of Cs/Sr also suggests that the fixation of deposited ^{137}Cs to the soil is slower in areas with higher proportion of humus soils.

Ecological half-lives of ^{137}Cs were nearly at the same level in all the areas after the nuclear weapons testing period, for short periods 2-3 years and for longer periods 5-7 years (Table 3). During the first years after the Chernobyl deposition the ecological half-lives of ^{137}Cs were from two to three years, and since 1990 from 5 years in western and south-western areas to 7-9 years in northern and eastern areas. The ecological half-lives of ^{137}Cs for short periods after the Chernobyl deposition were in agreement with those given in earlier studies [14-16] The half-lives for ^{90}Sr were almost twice as high as those for ^{137}Cs, 10-15 years, in all the areas after the rapid decrease in the 1960's (Table 4).

Table 3. Ecological half-lives of ^{137}Cs in dairy milk in different areas.

Area	Half-lives (years), period			
	1965-1969	1970-1985	1987-1989	1990-2003
SW	2.0	6.8	1.8	4.7
W	2.8	5.0	1.9	4.6
E	-	4.1 (1982-85)	2.6	8.7
N	2.0	5.1	3.2	7.4

Table 4. Ecological half-lives of ^{90}Sr in dairy milk in different areas.

Area	Half-lives (years), period		
	1965-1969	1970-1985	1988-2003
SW	3.4	11	11
W	4.7	9.6	15
E	-	-	12
N	3.1	9.7	13

The data produced in the long-term regular surveillance programme of dairy milk shows the trends in behaviour of radionuclides after a radioactive fallout in various types of environments. The data can be utilized in testing food chain models, and in fitting the parameters in models for regions with different soil types.

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