

^{90}Sr and ^{137}Cs in deposition, grass and milk in Northern Finland

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Abstract. Soil-grass-milk is one of the main food chains leading radioactivity in man in Finland. From 1963 onwards dairy and farm milk and from 1972 deposition have been regularly collected at several sites in Northern Finland; some grass and AIV-silage samples have been taken simultaneously. ^{137}Cs and ^{90}Sr have been analysed by the Radiation and Nuclear Safety Authority (STUK) in the Regional Laboratory in Northern Finland. In this study the activity concentrations and decrease rates of ^{90}Sr and ^{137}Cs were studied in the years following the atmospheric nuclear weapon testings and after the Chernobyl accident in 1986 until 2008. Also the transfer factors were calculated and the ecological half-lives were estimated in the study. The concentrations of ^{90}Sr and ^{137}Cs were highest in the beginning of 1960 due to the nuclear weapon tests. The radioactive fallout in Finnish Lapland from the Chernobyl accident in April 1986 was low. The increase of the ^{137}Cs concentrations in milk was found out in July 1986 and it decreased rapidly after summer 1987. There was no increase of ^{90}Sr concentrations in milk and in deposition after Chernobyl fallout.

1. INTRODUCTION

^{90}Sr and ^{137}Cs are fission products with relatively long half-lives (about 30 years). As a result of the atmospheric weapon testing and Chernobyl nuclear power accident, they have entered into the environment and, consequently, into the food chain. Since ^{90}Sr has very similar chemical behaviour as calcium and ^{137}Cs as potassium, they follow the paths of these elements in the food chain and enter the human body. Grass is an efficient collector of atmospheric contaminants, and radionuclides are fast passed from grass to milk. In Finland milk is the main pathway of ^{90}Sr to human.

STUK has regularly analysed ^{90}Sr and ^{137}Cs in deposition, grass and milk samples from different sites in Northern Finland. The sampling sites of milk were chosen to provide representative information about radioactivity in milk produced in Northern Finland.

The aim of this study was to investigate the changes of ^{90}Sr and ^{137}Cs concentrations after the atmospheric nuclear weapon testings and after the Chernobyl accident and to estimate and compare the ecological half-lives in milk and deposition.

2. MATERIALS AND METHODS

Milk was sampled at different sites in Finnish Lapland and in the Province of Oulu. Dairy milk was sampled from Kursu dairy from 1963 to 1987. Milk powder was sampled from a dairy in Rovaniemi from 1966 to 1975 and dairy milk from 1986 onwards. Milk produced in individual farms was sampled from Apukka in years 1975–1977 and again in 1986–1991 and from Vikajärvi from 1991 onwards. Farm milk was also sampled from Hyrynsalmi and Kemijärvi. Also some grass and AIV-silage were taken from the farms. The sampling sites are shown in Fig. 1. Deposition was sampled in Apukka from October 1972 onwards. The sampling period was three months at the time. The sample collector is made from stainless steel with a surface area of 0.07m^2 . The wet and dry depositions were collected together.

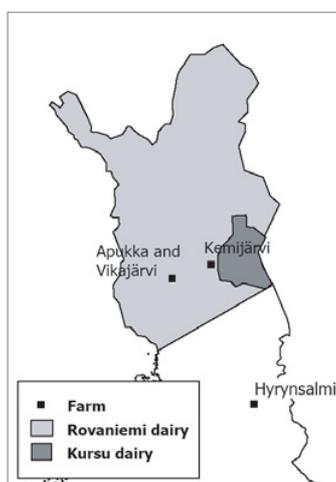


Figure 1. The sampling sites.

Sr was separated from samples using two different methods, traditional fuming nitric acid method and extraction chromatography using Sr-resin (Eichrom). Sr was measured through its yttrium daughter with proportional counter. ^{137}Cs was measured by gamma spectrometry.

3. RESULTS AND DISCUSSION

3.1 Milk and grass

The activity concentrations and the ecological half-lives of ^{90}Sr and ^{137}Cs in milk samples are shown in Fig. 2. ^{90}Sr and ^{137}Cs activity concentrations in present time are below 0,1 Bq/L and 1,8 Bq/L, respectively in all analysed sites. The concentrations of ^{90}Sr and ^{137}Cs were highest in the beginning of 1960 due to the nuclear weapon tests. Although the total deposition of ^{90}Sr and ^{137}Cs from the nuclear weapon tests was about the same in whole Finland, the highest ^{90}Sr and ^{137}Cs concentrations were found in Lapland [1, 2]. This was mainly due to the high proportion of peat soils and nutrient deficiency of the pasture in Lapland. The radioactive fallout in Finnish Lapland from the Chernobyl accident in April 1986 was low; ^{137}Cs was in average 1000 Bq/m² and the fallout occurred before the start of the growing season [3]. The increase of the ^{137}Cs concentrations was found out in July 1986 and it decreased rapidly after summer 1987. There was no increase of ^{90}Sr concentrations after Chernobyl fallout. ^{90}Sr activity concentrations from different areas were relatively similar. ^{137}Cs concentrations in Hyrynsalmi after Chernobyl were somewhat higher because of the higher fallout level in the area. A seasonal signal was shown in ^{90}Sr activity concentrations with higher values in summer time, when cows are put out to pasture or fed with fresh grass, same signal was also found in other studies [4]. However, similar signal was not shown for ^{137}Cs . The reason for this discrepancy may depend on difference in soil types, grass species and fertilisation. The ecological half-lives of ^{90}Sr and ^{137}Cs in 1963–1966 were nearly similar, about two years. The half-lives for ^{90}Sr in 1975–1985 were almost twice as high as those for ^{137}Cs and six years longer in 1993–2008.

^{137}Cs concentrations [Bq/kg f.w.] before Chernobyl varied in grass between 17–33 and in AIV-silage between 2.3–3.6 without one exceptional high value 13.7 measured in AIV-silage from Kemijärvi. After Chernobyl ^{137}Cs concentrations in AIV-silage varied between 0.25–0.35 and in grass between 0.8–10 in Kemijärvi. Single values during the same sampling period in Vikjärvi were 1.4 in AIV-silage and 4.6 in grass. ^{90}Sr concentrations [Bq/kg f.w.] were between 1.9–2.6 in grass and 0.3–1.8 in AIV-silage.

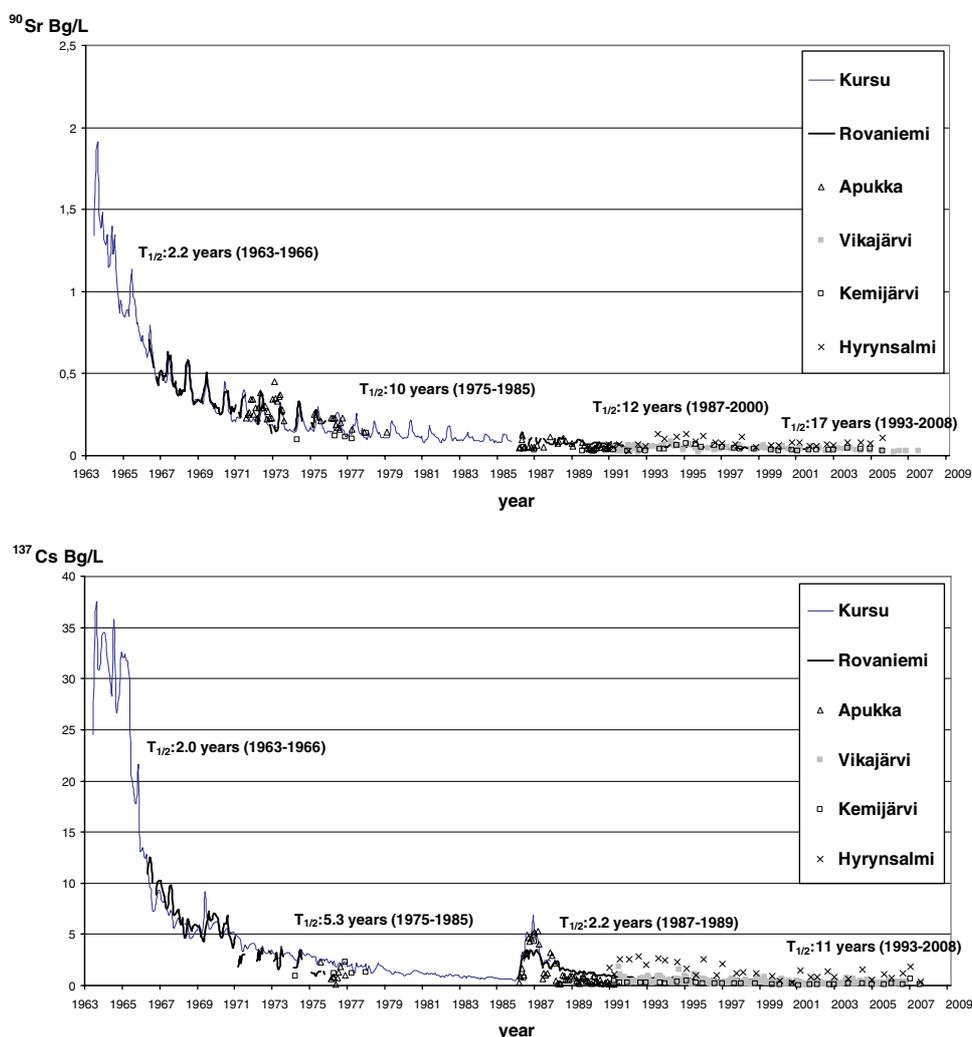


Figure 2. ⁹⁰Sr and ¹³⁷Cs concentrations and the ecological half-lives in milk in Lapland 1963–2008.

Table 1. The aggregated transfer coefficients T_{ag} (¹³⁷Cs) from fallout to grass and AIV-silage.

| Time Period | Sampling time | Sampling site | n | Grass | | | n | AIV-silage | | |
|----------------|---------------|---------------|---|--------|--------|--------|---|------------|--------|--------|
| | | | | Mean | Min | Max | | Mean | Min | Max |
| “nuclear era” | 1976 | Apukka | 3 | 0.0133 | 0.0092 | 0.0181 | 3 | 0.0015 | 0.0013 | 0.0019 |
| | 1977–1978 | Kemijärvi | 2 | 0.0188 | 0.0181 | 0.0195 | 3 | 0.0038 | 0.0018 | 0.0075 |
| post Chernobyl | 2000–2006 | Apukka | 1 | 0.0007 | | | 4 | 0.0013 | 0.0003 | 0.0024 |
| | 2000–2006 | Kemijärvi | 3 | 0.0022 | 0.0004 | 0.0051 | 3 | 0.0002 | 0.0001 | 0.0002 |

The aggregated transfer coefficients T_{ag} were calculated and the results are presented in Table 1. In calculations before Chernobyl, the year 1972 was used as a reference date and the fallout value used was 2000 Bq/m² (from Fig. 3). After Chernobyl, the equivalent values were year 1986 and 1000 Bq/m².

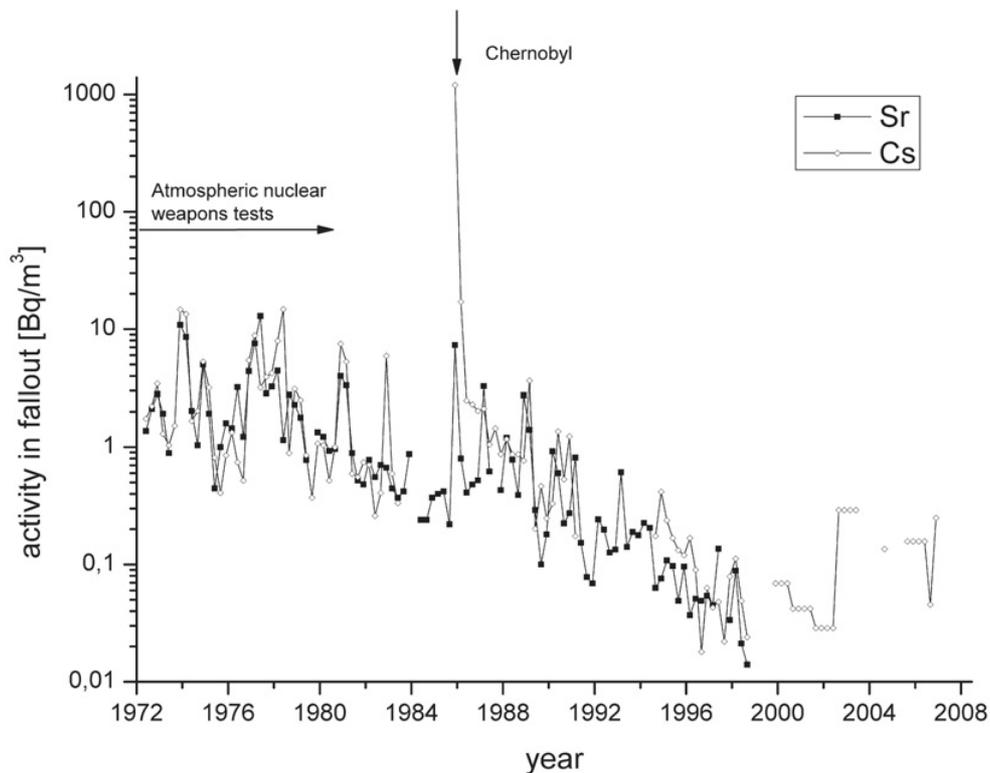


Figure 3. Measured ^{90}Sr and ^{137}Cs activity in deposition at Apukka, Rovaniemi.

3.2 Deposition

The sampling of deposition started in October 1972 when China was still conducting nuclear weapon tests in atmosphere at Lop Nur test site. Chinese tests were still releasing ^{90}Sr and ^{137}Cs into the atmosphere. This resulted in a global fallout and small amounts of ^{90}Sr and ^{137}Cs from these tests were deposited at Finnish Lapland. Fig. 3 shows the time series of ^{90}Sr and ^{137}Cs activity in deposition measured at Apukka, Rovaniemi from October 1972 to March 1999 for ^{90}Sr and from October 1972 to June 2007 for ^{137}Cs . The main source of ^{90}Sr and ^{137}Cs in Finnish Lapland is the atmospheric nuclear weapon tests. The Chernobyl accident caused an increase in the ^{137}Cs deposition.

The effect of the atmospheric nuclear tests can be seen if the time series is divided into three sections in Table 3.2 and 3.2. The First period is the “nuclear era” which covers the time period October 1972–June 1981 when China had made the last atmospheric nuclear weapon tests. The second period is the “middle years” from June 1981 to March 1986 which are the years after the nuclear weapons test period to the time of Chernobyl accident. The third period is the “post Chernobyl” years from August 1986 to March 1999 or June 2007. Half-lives of ^{90}Sr and ^{137}Cs in the deposition in different time periods can be extracted from the time series presented above.

The ^{90}Sr activity in the deposition is low throughout the whole sampling period. Also the ^{137}Cs activity is relatively low if the Chernobyl accident is disregarded. The half-lives of the ^{90}Sr and ^{137}Cs activity in the deposition during the middle years and in the post Chernobyl period agree well with the measured half-life of the total beta activity in ambient air [5]. The long half-life in the nuclear era can be attributed to atmospheric nuclear weapon tests. The tests were conducted far away from Finnish Lapland

Table 2. Summary of the observed ^{137}Cs data.

| Period | Time | Half-life [a] | ^{137}Cs fallout [Bq/m^2] |
|----------------|------------------------|---------------|------------------------------------------------------|
| nuclear era | October 1972–June 1981 | 11,4 | 0,4–15 |
| middle years | June 1981–March 1986 | 2,7 | 0,3–6,0 |
| post Chernobyl | August 1986–June 2007 | 2,4 | 0,01–3,3 |

Table 3. Summary of the observed ^{90}Sr data.

| Period | Time | Half-life [a] | ^{90}Sr fallout [Bq/m^2] |
|----------------|------------------------|---------------|-----------------------------------------------------|
| nuclear era | October 1972–June 1981 | 29,9 | 1–12 |
| middle years | June 1981–March 1986 | 2,7 | 0,2–4,0 |
| post Chernobyl | August 1986–March 1999 | 2,5 | 0,01–3,3 |

and low activity increases from these tests were enough to keep the activity level at relatively constant level.

4. CONCLUSIONS

The figures 2 and 3 as well as the tables 1 and 3.2 show half-lives of ^{90}Sr and ^{137}Cs in different samples. The half-lives of these nuclides in deposition represent the input to the food chain soil-grass-milk while the half-lives in the milk represent the output. The half-lives can be compared, and in milk longer half-lives of ^{90}Sr than for ^{137}Cs for longer time intervals, can be observed. Similar half-lives in deposition and in milk can be observed for ^{137}Cs in 1980's before the Chernobyl accident. Longer half-lives could be expected in milk at the present time since contaminants are still enriched in the food chain soil-grass-milk.

Finnish Lapland is very vulnerable to radioactive fallout due to high transfer rates in arctic and sub arctic environment. Finland has also high consumption of milk and milk products, about 15 kg/capita/year in Northern Finland [6]. Therefore, the monitoring of ^{90}Sr and ^{137}Cs in milk and deposition is very important for radiation protection and also for detecting all remarkable changes in activity in order to properly respond to radiological emergencies.

References

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