



## **EcoDoses. Improving radiological assessment of doses to man from terrestrial ecosystems**

**Nielsen, S.P.; Isaksson, M.; Saxén, R.; Kostianen, E.; Rissanen, K.; Joensen, H.P.; Palsson, S.E.; Bergan, T.D.; Skuterud, L.; Thørring, H.**

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Edited by Tone D. Bergan and Astrid Liland  
NRPA, Norway

May 2004

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## Abstract

The NKS B-programme EcoDoses project started in 2003 as a collaboration between all the Nordic countries. The aim of the project is to improve the radiological assessments of doses to man from terrestrial ecosystems.

The first part, conducted in 2003, has focussed on an extensive collation and review of both published and unpublished data from all the Nordic countries for the nuclear weapons fallout period and the post-Chernobyl period. This included data on radionuclides in air filters, precipitation, soil samples, milk and reindeer. Based on this, an improved model for estimating radioactive fallout based on precipitation data during the nuclear weapons fallout period has been developed. Effective ecological half-lives for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in milk have been calculated for the nuclear weapons fallout period. For reindeer the ecological half-lives for  $^{137}\text{Cs}$  have been calculated for both the nuclear weapons fallout period and the post-Chernobyl period. The data were also used to compare modelling results with observed concentrations. This was done at a workshop where the radioecological food-and-dose module in the ARGOS decision support system was used to predict transfer of deposited radionuclides to foodstuffs and subsequent radiation doses to man.

The work conducted the first year is presented in this report and gives interesting, new results relevant for terrestrial radioecology.

## Key words

Nuclear weapons fallout, deposition modelling, food chain modelling, ecological half-lives in reindeer and milk

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Improving radiological assessment of doses to man from terrestrial ecosystems

Edited by : Tone Bergan



# **EcoDoses**

## **Improving radiological assessment of doses to man from terrestrial ecosystems**

A status report for the NKS-B project 2003

**Edited by Tone D. Bergan and Astrid Liland**

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## 1. Introduction

The NKS B-programme EcoDoses project started in 2003 as a collaboration between all the Nordic countries. The aim of the project is to improve the radiological assessments of doses to man from terrestrial ecosystems.

The first part, conducted in 2003, has focussed on an extensive collation and review of both published and unpublished data from all the Nordic countries for the nuclear weapons fallout period and the post-Chernobyl period. This included data on radionuclides in air filters, precipitation, soil samples, milk and reindeer. Based on this, an improved model for estimating radioactive fallout based on precipitation data during the nuclear weapons fallout period has been developed. Effective ecological half-lives for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in milk have been calculated for the nuclear weapons fallout period. For reindeer the ecological half-lives for  $^{137}\text{Cs}$  have been calculated for both the nuclear weapons fallout period and the post-Chernobyl period. The data were also used to compare modelling results with observed concentrations. This was done at a workshop where the radioecological food-and-dose module in the ARGOS decision support system was used to predict transfer of deposited radionuclides to foodstuffs and subsequent radiation doses to man.

The work conducted the first year is presented in this report and gives interesting, new results relevant for terrestrial radioecology.

## 2. Atmospheric nuclear tests

As of January 1999, there have been 2,532 known nuclear tests worldwide (Carter, 1979; Norris & Arkin, 1996; UNSCEAR, 2000), see Table 1 and Figure 1 and 2. Different sources report different numbers, as the number of tests depends on the definition of a test<sup>1</sup>, as well as access to restricted information. According to UNSCEAR, almost 22 percent of the tests (543) were conducted in the atmosphere - most of them prior to 1963. This includes 39 safety tests with no nuclear yield, but with possible local plutonium contamination. The atmospheric nuclear weapons tests are considered to be the most significant source of radioactive fallout across the world.

The total yield of the atmospheric tests is estimated to have a size equivalent of 440 Mt, of which 189 Mt was fission yield. Deposited fission yield is estimated to have a size equivalent of 160 Mt. The most intensive test period was from September 1961 to December 1962, when 57% of the total explosive force was detonated. The largest test, conducted by the former Soviet Union in 1961, had a total explosion yield of 58 Mt and is estimated to have a fission yield of only 3% and a fusion yield of 97%. About 80% of the radioactivity from the atmospheric nuclear tests was later deposited as global fallout (Eisenbud & Gesell, 1997), the remaining 20% as either local or regional fallout. Figure 3 shows the approximate locations of test sites.

The atmospheric testing period lasted from 1945 to 1980, and can be divided into three phases. Early in the period, the explosions were comparatively small so that the nuclear debris was confined zonally, within the troposphere and roughly to the latitude of the explosion sites. The stratosphere was penetrated for the first time by a thermonuclear device detonated by the USA in 1952 (Peirson, 1971). The device, the 10.4 Mt surface bomb Mike in the Ivy series, detonated at the US test site at Eniwetok in the Pacific Ocean on 31 October. The partitioned fission yield was 2.85 Mt in the local and regional troposphere and 2.85 Mt in the stratosphere.

The first global fallout phase covers the period from the Ivy Mike test in October 1952 until November 1958, when the Soviet Union, the United Kingdom, and the United States complied with a temporary nuclear test moratorium. It consisted of moderate-sized explosions, and much of the debris was confined within the lower stratosphere. In this phase, there was a total fission yield of 80 Mt, producing approximately 310 PBq of <sup>90</sup>Sr (using a production estimate of 3.9 PBq/Mt, UNSCEAR, 2000). It is estimated that 35-40% of the yield was deposited in the vicinity of the explosion sites (Eisenbud & Gesell, 1997). The moratorium ended in September 1961, when there was an outbreak of tests. The second phase covers the period 1961-1962, and is dominated by larger explosions and the insertion of debris higher into the stratosphere. In this period, there was a total of 90 Mt fission yield, producing approximately 350 PBq of <sup>90</sup>Sr. Following this, came a peak in deposition rate of <sup>90</sup>Sr during 1963, with a steady fall until 1967. The third phase of nuclear weapons testing (1967-1980) started with a period of occasional explosions, producing a few Mt each year and in total 18 Mt resulting in approximately 70 PBq of <sup>90</sup>Sr. About 85% of the total fallout was deposited in the period 1950-1965. On 10 October 1963, a partial test ban treaty came into force, banning nuclear tests in the atmosphere, underwater and in space. Neither France

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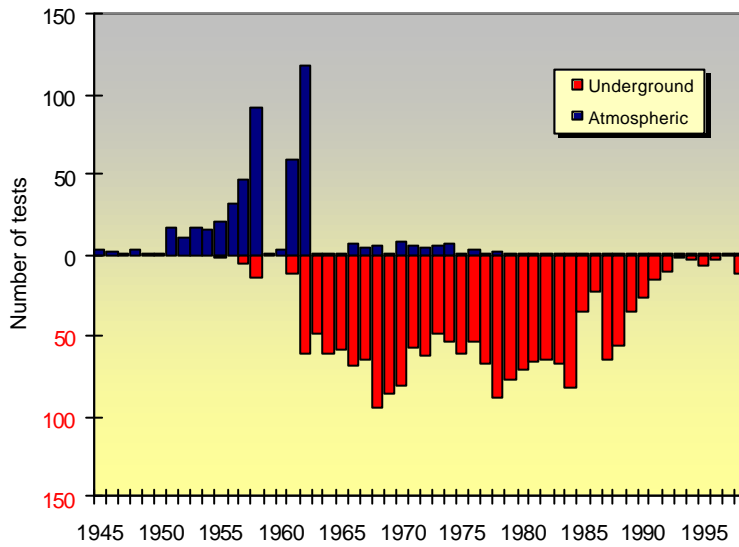
<sup>1</sup> For reference: The definition adapted by the United States and the Soviet Union/Russia states that a test is a single explosion, or two or more explosions fired within 0.1 seconds within a diameter of two kilometres.

nor China, both being nuclear weapon powers, signed the treaty. They have however honoured it since 1980.

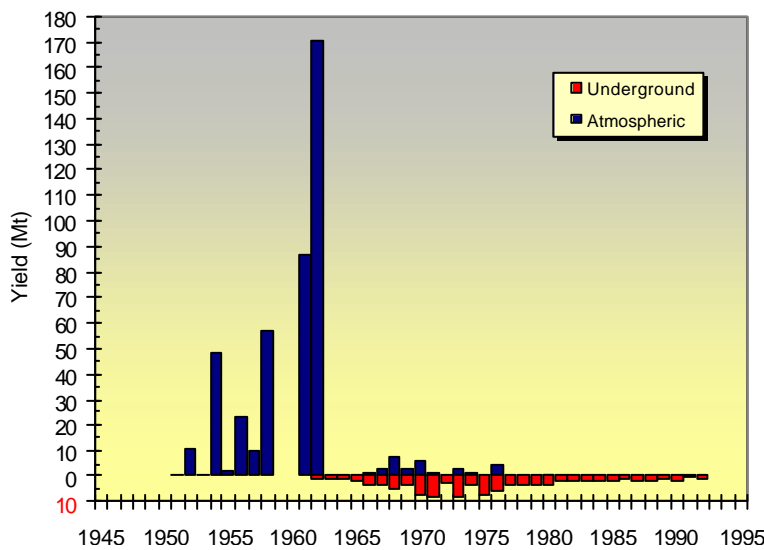
**Table 1: All known nuclear detonations up to 1999 (UNSCEAR, 2000; Norris & Arkin, 1996; Matuschchenko et. al. 1998). “Peaceful” detonations are detonations with a civilian purpose, such as mining, oil exploration, construction work, etc.**

Country	Time period	Atmospheric	Underwater	Underground	“Peaceful”	Total
France	1960-1974	45		160		205
India	1974-1998			6	1	7
Pakistan	1998			6		6
China	1964-1980	22		22		44
Soviet	1949-1962	216	3	750	124	1093
Great Britain	1952-1953	21		24		45
USA	1945-1962	192	5	908	27	1132
<b>Total</b>		<b>496<sup>a</sup></b>	<b>8</b>	<b>1876</b>	<b>152</b>	<b>2532</b>

<sup>a</sup> Including 39 safety tests; no nuclear yield, but possible local contamination of e.g. plutonium.



**Figure 1: Number of atmospheric and underground nuclear weapons tests.**



**Figure 2: Total yield of atmospheric and underground nuclear detonations.**

Of special interest to the Nordic countries, are the nuclear tests conducted on Novaya Zemlya. Out of the 219 atmospheric tests performed in the former Soviet Union, Matuschchenko *et al.* (1998) reports a total of 88 atmospheric, two surface water and three underground tests on Novaya Zemlya. Among these tests is the world's largest single detonation of 58 Mt, which occurred in October 1961. There has been some speculation as to whether Norway, with a close proximity to Novaya Zemlya, received local fallout from the tests, especially in connection with the surface or underwater tests in 1955, 1957 and 1961.

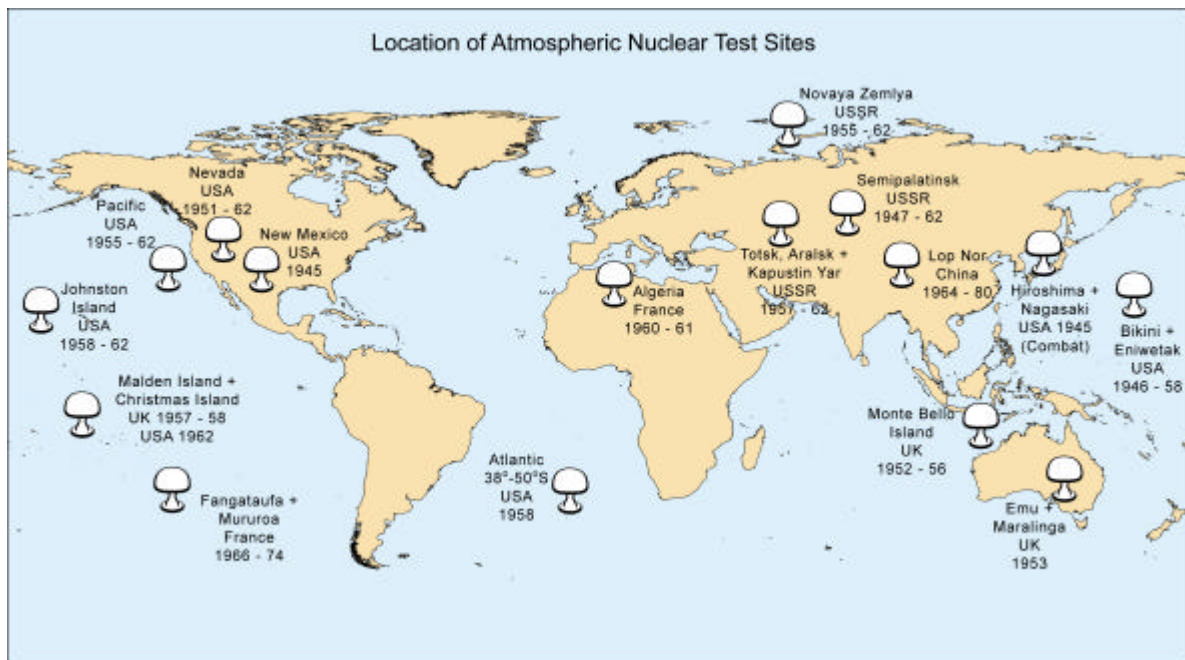


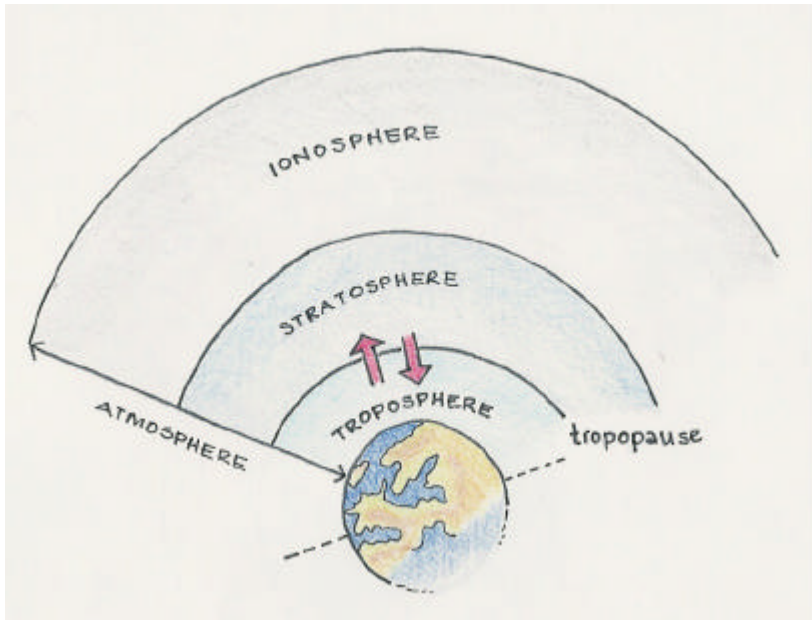
Figure 3: Approximate locations of atmospheric nuclear weapons test sites from 1945 to 1980.

### 2.1 Global deposition, the UNSCEAR model

A nuclear test produces about 150 fission products with half-lives, or progeny half-lives, long enough to contribute to radioactive fallout (Hicks, 1981), and spreads a considerable amount of unfissioned fissile material. Plutonium isotopes and tritium are produced as both a result of their use as a fuel in the lower yield devices, as well as from their production by activation of  $^{238}\text{U}$  and by fusion reactions in high-yield devices. A variety of other radionuclides is produced by neutron activation of bomb constituents and surrounding materials. Finally, particular compounds were included in a number of tests to produce radioactive tracers by neutron activation for use in studying the transportation of the fallout debris through the atmosphere. These tracer nuclides include  $^{185}\text{W}$ ,  $^{109}\text{Cd}$  and  $^{102}\text{Rh}$ .

Radioactive debris injected into the stratosphere will move slowly down into the troposphere, from where the debris is relatively rapidly removed and deposited on the ground, mainly by precipitation (Eisenbud & Gesell, 1997). There are seasonal and latitudinal variations in the concentration of debris in ground-level air and the rate of deposition (Davis *et al.*, 1960). These variations can mostly be explained by spatial and temporal differences in precipitation

by a simplified fallout model, where the removal of debris from the troposphere is proportional to precipitation and tropospheric concentration (Hvinden *et al.*, 1965). The correlations may appear only in average values over reasonable wide areas and long periods. Also, the precipitation values from a limited group of monitoring stations are not fully representative of the washout history of the measured air (Hvinden *et al.*, 1965).



**Figure 4: Distribution of radioactivity in the atmosphere**

A peak in late spring and a minimum in winter have characterised the temporal distribution of the fallout during a typical year. The spring peak can be explained by an enhanced transport from the stratosphere, into which the main component of the debris from atmospheric nuclear tests is injected, down through the tropopause (Figure 4). Dust injected into the lower polar stratosphere by Russian thermonuclear explosions had a mean residence time of less than six months, whereas in tropical latitudes the residence time was as long as 2-3 years in the middle stratosphere and 5-10 years if injected at 100 km or more above ground. In the troposphere, fallout had a mean residence time of 20-40 days (UNSCEAR, 1982). Average residence time of debris in the atmosphere is about 1.3 years. Mean residence time of aerosols in the lower stratosphere ranges from 3 to 12 months in the polar regions, and 8 to 24 months in the equatorial regions.

The global release of radionuclides and the annual deposition as predicted by UNSCEAR are given in Figure 5 and Table 2.

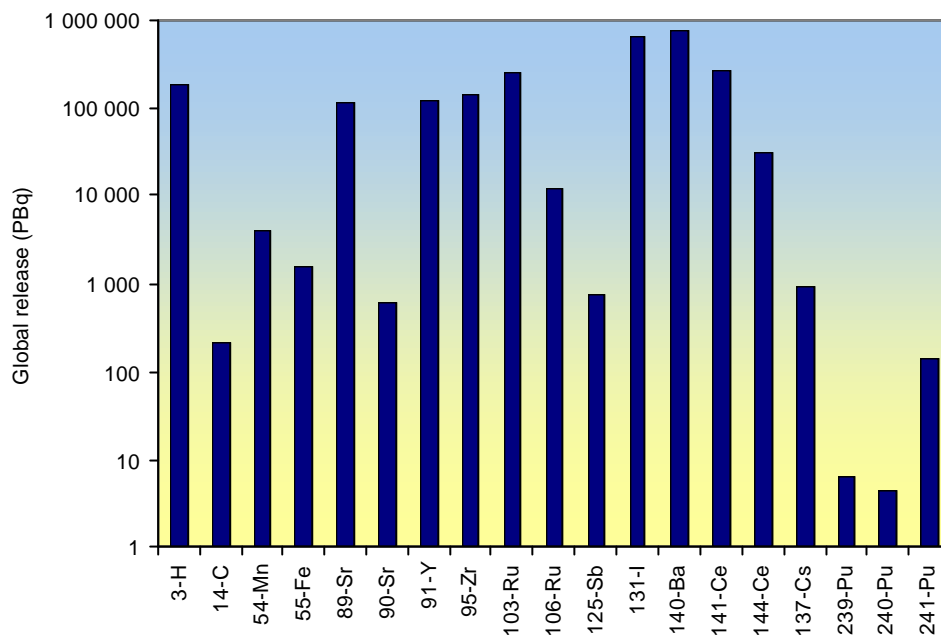


Figure 5: Global release of radionuclides (UNSCEAR, 2000)

Table 2: Annual deposition (PBq) (UNSCEAR 2000)

Nuclide	Time period						Total
	Before 1952	1952-1958	1959-1962	1963-1967	1968-1980	After 1980	
Northern hemisphere							
<sup>131</sup> I	139	1916	1666	120	159	0.023	4000
<sup>140</sup> Ba	246	3590	3084	268	316	0.52	7500
<sup>141</sup> Ce	127	2339	2639	551	348	6.9	6000
<sup>103</sup> Ru	180	3274	2871	753	419	10.4	7500
<sup>89</sup> Sr	91.1	1774	1595	571	261	8.2	4300
<sup>91</sup> Y	117	2383	2186	923	381	13.2	6000
<sup>95</sup> Zr	111	2453	2979	1409	538	19.9	7500
<sup>144</sup> Ce	54.3	1981	2757	3876	858	35.6	9560
<sup>54</sup> Mn	0.24	133	380	576	53.4	0.731	1144
<sup>106</sup> Ru	31.2	1240	1271	1925	408	16.39	4892
<sup>125</sup> Sb	1.99	101	102	201	38.9	1.47	446
<sup>55</sup> Fe	0.1	70.9	211	477	38	0.473	797
<sup>90</sup> Sr	1.72	97.4	115	205	48.2	2.81	474
<sup>137</sup> Cs	2.55	146	173	308	72.3	4.23	706

- Derived from estimated fission/fusion yields of tests with atmospheric model. Measured results used preferentially for Sr-90 and Cs-137 during 1958-1985. Model values for I-131, Ba-144, Ce-141, Ru-103, Sr-89, Y-91, and Zr-95 normalized to total hemispheric deposition estimated from available measurements.

Bursts that take place high in the atmosphere produce small particles that remain suspended for considerable periods of time. Airburst particles cannot grow to more than 0.3 μm by condensation (Lockhart *et al.*, 1965). In general, the fallout can be divided into three fractions; large particles that deposited from the atmosphere within hours, smaller particles that remained in the troposphere from which they were removed on a time scale of days, and the fraction injected into the stratosphere, from which they were removed on a time scale of months to years.

Prior to 1952, all the nuclear explosions were in the kiloton range, and the atmospheric radioactivity diminished at a rate corresponding to the residence half-life of dust in the lower atmosphere at about 20 days. Tropospheric fallout contains only a small fraction (not more than 5% of the total radioactive yield) of the long-lived radionuclides from bombs in the Mt range, but can be responsible for heavy exposure from short-lived tropospheric debris, notable  $^{131}\text{I}$ . Local fallout in this period accounted for an average of 80% for land surface explosions, 20% for explosions on the surface of the water and 10% for explosions in the air. The stratospheric inventory was not influenced significantly by the many detonations in the kiloton range since these did not penetrate appreciably into the stratosphere (Eisenbud & Gesell, 1997).

On a global scale, the deposition of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  shows a maximum at the temperate latitudes and a minimum at the poles and equator (Figure 6 and Figure 7). Most tests were performed in the northern hemisphere, and the mean integrated fallout in the  $60^{\circ}$ - $70^{\circ}$  N latitude band can be calculated from UNSCEAR (2000) to be  $2.7 \text{ kBq/m}^2$  for  $^{137}\text{Cs}$  and  $1.8 \text{ kBq/m}^2$  for  $^{90}\text{Sr}$ . Although describing the global pattern of deposition, the model does not describe regional differences in detail.

The British atomic energy research establishment (AERE) and the US Naval Research Laboratory started, systematically, monitoring of the atmosphere for fission products in 1955 and 1957, respectively (Monetti, 1996). Tromsø ( $69^{\circ} 40' \text{ N}$ ) and Bodø ( $67^{\circ} 17' \text{ N}$ ) were included as monitoring stations in the AERE-network from 1957 for rainwater, and Tromsø for air from 1977. Figure 8 illustrates the deposition of  $^{137}\text{Cs}$  calculated for the  $60^{\circ}$ - $70^{\circ}$  N latitude using the UNSCEAR global fallout model (UNSCEAR, 2000), compared with measurements of deposited  $^{137}\text{Cs}$  in Bodø and Tromsø. Estimated deposition of  $^{137}\text{Cs}$  in the years 1957-1963 is calculated from actual measurements of  $^{90}\text{Sr}$  deposition using a ratio of 1.5 for  $^{137}\text{Cs}/^{90}\text{Sr}$ . When the annual deposition of  $^{137}\text{Cs}$  calculated for the  $60^{\circ}$ - $70^{\circ}$  N latitude using the UNSCEAR global fallout model is compared with measurements of deposited  $^{137}\text{Cs}$  in Bodø and Tromsø, it becomes evident that the global model does not take into account the relatively rapid deposition of radionuclides in the northern hemisphere originating from the Soviet tests in 1958. The deposited  $^{137}\text{Cs}$  in 1958 is also accompanied by high levels of  $^{131}\text{I}$ . The UNSCEAR model also significantly underestimates the annual deposition in Norway.



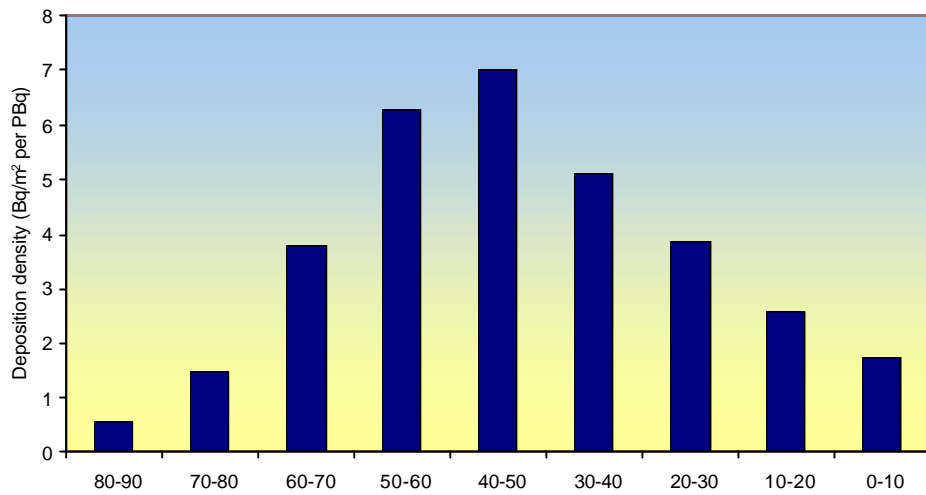


Figure 6: Deposition density of <sup>90</sup>Sr in latitude bands in the northern hemisphere (UNSCEAR 2000, global model)

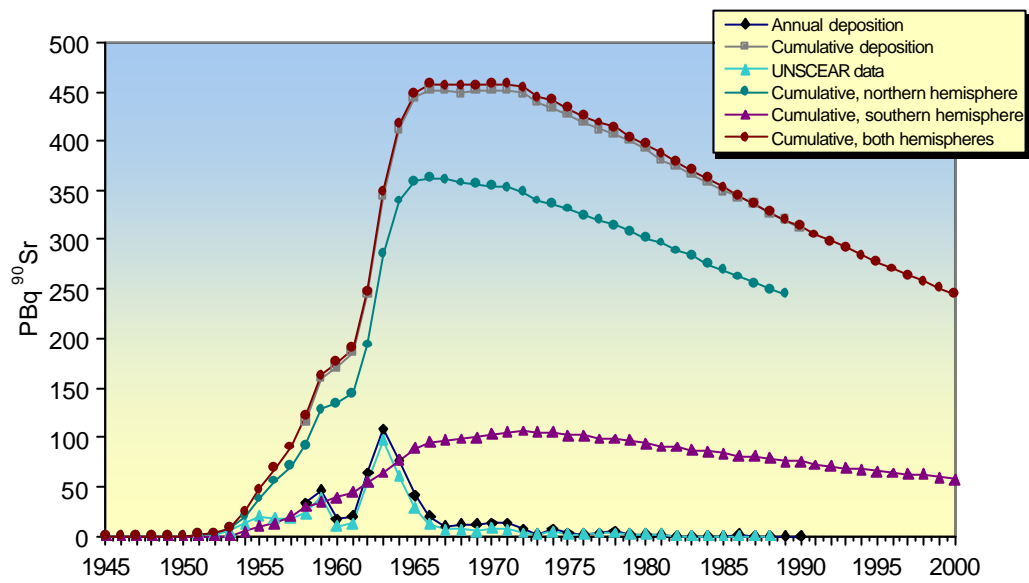


Figure 7: Worldwide deposition of <sup>90</sup>Sr (UNSCEAR, 2000)

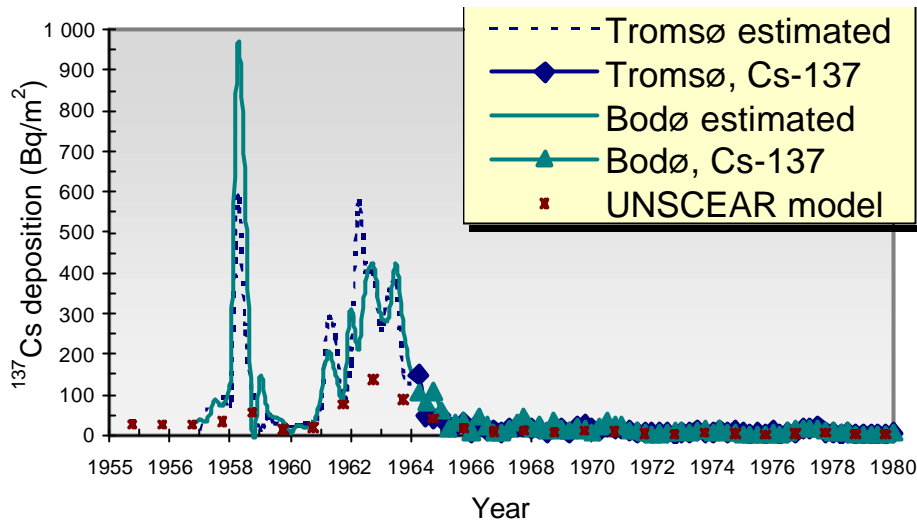


Figure 8: Deposition of  $^{137}\text{Cs}$  calculated for the  $60^{\circ}$ - $70^{\circ}$  N latitude (UNSCEAR, 2000), compared with measurements of  $^{137}\text{Cs}$  deposition in Bodø ( $67^{\circ}$   $17'$  N) and Tromsø ( $69^{\circ}$   $40'$  N). Deposition of  $^{137}\text{Cs}$  for the period 1957-1964 is calculated from measurements of  $^{90}\text{Sr}$  deposition using a ratio of 1.5 for  $^{137}\text{Cs}/^{90}\text{Sr}$ .

## 2.2 Measurements of fallout

Extensive monitoring programmes were initiated to measure the radioactive yield resulting from the atmospheric nuclear tests, the earliest in the 1950s. The most thorough monitoring arrangements were of radioactivity in air and precipitation, but a variety of other substances were measured as well, including soil samples and food products, etc.

### 2.2.1 Air filters

Radioisotopes in the air were collected in air filters, and measured for total beta activity after a decaying period of some days. This led to short-lived isotopes, like most of the daughter nuclides of radon, having decayed away prior to the measurements. Filters were additionally added up and measured for  $^{137}\text{Cs}$  with gamma spectroscopy.

### 2.2.2 Precipitation

Snow, rain and/or settling dust was collected in stainless steel or polyethylene containers, which were emptied regularly. If there had been no precipitation, the container would be flushed with distilled water. The precipitation or the flushing water was subsequently evaporated and measured for total beta activity. Some selected samples were measured with gamma spectroscopy as well. Also, irregular measurements of the contents of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were made.

### 2.2.3 Milk

Milk is the principal food through which  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  enter the human diet in Scandinavia, and it constitutes a major portion of the diet of infants. One considered therefore measurements of the activity in milk samples to be of great importance. Initially, the main concern was with  $^{90}\text{Sr}$ , which largely follows calcium in the metabolic chain, but it was also

well known that  $^{131}\text{I}$  and  $^{137}\text{Cs}$  are secreted into milk. The caesium isotope  $^{137}\text{Cs}$  follows the metabolism of potassium, while the iodide isotope  $^{131}\text{I}$  follows the natural iodine and is easily absorbed by the body, where it is mainly accumulated in the thyroid gland. Milk was analysed regularly for  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{131}\text{I}$ . On occasion, dry milk samples of equivalent size were used.

#### 2.2.4 Soil samples

The fallout activity in soil samples was measured and compared with the results from the precipitation measurements.

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### 3. Prediction of spatial variation in global fallout of $^{137}\text{Cs}$ using precipitation

#### 3.1 Introduction

Deposition from atmospheric nuclear weapons tests has in many cases been shown to be proportional to the rate of precipitation, although at sites with low precipitation rates this may not be true due to the dry deposition of radionuclides. Establishing the relationship between deposition and precipitation at a reference site allows radionuclide deposition at other sites to be estimated using precipitation data (e.g. Wright *et al.*, 1999; Pálsson *et al.*, 2002). However, care needs to be taken when the relationship between deposition and precipitation is calculated and described. The concentration of radionuclides in precipitation can be highly variable during extended periods of fallout. Such temporal variability should be taken into account when the relationship between precipitation and deposition is being derived and used for the estimation of radionuclide deposition at another site. This chapter describes methods which optimise the available precipitation and deposition information for a reference site for the estimation of radionuclide deposition following atmospheric nuclear weapons tests at other locations. The usefulness of the method is demonstrated for a test site in Iceland and the method is then applied to a few data sets from the other Nordic countries.

The relationship between precipitation and deposition is often inadequately characterised making it difficult to compare results between different studies. In some papers, the relationship is expressed as activity per unit area corresponding to a given rainfall (e.g. Bq/m<sup>2</sup> per 1000 mm rain). This type of expression is meaningful during a period of deposition or if the time period is explicitly stated. However, such an expression becomes meaningless after the deposition has ceased, because it fails to take into account temporal changes in this value. Likewise, comparing cumulative deposition with cumulative precipitation should be done with caution, because temporal variability in the relationship of deposition to precipitation may not be accounted for.

It is often better to relate radionuclide deposition to the *average annual precipitation rate* (e.g. mm rain per year) rather than *cumulative precipitation* (e.g. mm rain). The former value reflects the likelihood that one location receiving double the average precipitation rate compared to another location will have twice the radionuclide deposition. However, for the latter value, comparing cumulative precipitation between two sites is meaningless without referring to a given time interval. Furthermore, if a reference date during the time of deposition is used (e.g. in 1965), then it has to be clearly stated whether the estimate takes into account future deposition (e.g. until the present) with estimates time corrected back to the reference date or if the estimate only takes into account deposition for the specified year (which would normally be the case for data reported in that year).

#### 3.2 Methods of estimating fallout deposition

Calculating the *average annual precipitation rate* can be done in various ways, all providing differing estimates. The general formulation for calculating average annual precipitation rate can be expressed as:

$$\overline{P_X} = m \cdot \sum_i w_i \cdot P_{X_i} \quad (1)$$

where

$m$  is the number of time sub-periods in a year

$w_i$  are normalised weighting factors (their sum is 1)

$P_{Xi}$  is the precipitation amount (unit:  $y^{-1}$ ) at any site X during time period  $i$

If a simple average is to be calculated over a given time period, then each  $w_i = 1/n$ , where  $n$  is the total number of sub-periods. If monthly precipitation measurements are available then  $m = 12 y^{-1}$  is used to calculate the annual average precipitation rate.

Average precipitation rates are often compiled in terms of 30 year periods, e.g. 1931-60, 1961-90. Since the rate of precipitation can be temporally variable, better estimates of radionuclide deposition should be obtained by using a shorter time period window than 30 years, preferentially focusing on the years of maximum fallout. Before 1960, about 29% of the total global fallout in the northern hemisphere was deposited. Subsequently, in the period of highest test activity, 60% of the global fallout occurred during the years 1960-1967 and 11% thereafter (derived from UNSCEAR 2000).

It is possible, however, to circumvent the problem of selecting a suitable time window and instead have it (at least in theory) infinitely wide. This can be done by using variable weighting factors instead of constant ones. One such choice is:

$$w_i = \frac{C_{R_i}}{\sum_i C_{R_i}} \quad (2)$$

where  $C_{R_i}$  is the decay corrected  $^{137}\text{Cs}$  concentration in precipitation at the reference site R during the time period  $i$  ( $\text{Bq m}^{-3}$ ). Each time period has thus weight that is proportional to its potential relative contribution to the total deposition. Then precipitation during periods of high  $^{137}\text{Cs}$  concentrations is allocated greater weight than that during periods of lower concentrations. This estimate of the radionuclide concentration in precipitation needs not to be based on a direct measurement. It can be estimated from the decay corrected measured deposition during the relevant period by dividing by the amount of precipitation during the period:

$$C_{R_i} = \frac{D_{R_i}}{P_{R_i}} \quad (3)$$

where:

$D_R$  is the total accumulated decay corrected deposition at the reference site ( $\text{Bq m}^{-2}$ ),

$\overline{P}_R$  is the average annual precipitation at reference site R ( $\text{m y}^{-1}$ )

The radionuclide deposition can be estimated for the reference site directly from the decay corrected concentration of the nuclide in rain and the amount of precipitation.

$$D_R = \sum_i C_{R_i} \cdot P_{R_i} \quad (4)$$

The ratio of deposition to the average annual precipitation rate at the reference site R can then be shown to be, using the symbol  $C_\Sigma$  :

$$C_\Sigma = \frac{D_R}{\overline{P}_R} = \frac{1}{m} \sum_i C_{R_i} \quad (5)$$

When the above method of calculating average annual precipitation rate is used,  $C_{\Sigma}$  has a distinct physical meaning. The ratio of radionuclide deposition to the average annual precipitation rate is then simply the *time integrated decay corrected concentration of the radionuclide in precipitation*. Furthermore, this shows that the ratio  $C_{\Sigma}$  can be determined for a reference site in the absence of precipitation data, solely from values for the radionuclide concentration in precipitation. If the concentration in rain water is described with a continuous function, the time integrated decay corrected concentration in rain is given by:

$$C_{\Sigma} = \int_0^T C_R(t) dt \quad (6)$$

Then we can calculate the deposition at another site X,  $D_X$ , from the average precipitation rate at that site, if we assume that the ratio we calculated for the reference site holds for site X.

$$D_X = C_{\Sigma} \cdot \overline{P_X} \quad (7)$$

This equation can also be shown to be equivalent to:

$$D_X = \sum_i C_{Ri} \cdot P_{X_i} \quad (8)$$

The deposition estimate at the site X can thus be expressed as the product of a constant and the average precipitation rate at the site and also as the sum of the product of the concentration at a reference site during time period  $i$  and the precipitation at site X during the same period.

### 3.3 Case-study Iceland

Measurements of fallout from nuclear weapons tests in soil, vegetation and agricultural products started in Iceland over 40 years ago (Pálsson, 1996). Considerable variability was present in the results, even between adjacent sites, probably due to the mountainous terrain, changing strong winds and highly variable levels of precipitation. This variability has been especially noticeable in the case of soil samples but measurements of contamination in many areas are sparse, in part due to the difficult, remote and inaccessible terrain of much of the country. A recent study by Sigurgeirsson et al (submitted) has provided improved information on the spatial variation in  $^{137}\text{Cs}$  deposition at various sites across Iceland, especially in areas used for agriculture. The sampling sites were deliberately located close to meteorological measurement stations so that good representative data was available on precipitation for each sampling site, and the exact location of each site was recorded using a GPS instrument. The sites were selected so that they would span a range of precipitation rates as well as representing different areas and soil types. Soils were sampled with 17 and 19 mm diameter sampling probes to a depth of up to 30 cm. Twenty cores were collected at each site at 1 meter intervals along a 20 m long transect and bulked. The measured  $^{137}\text{Cs}$  content per unit area of soil varied from 900-4700 Bq m<sup>-2</sup>, with higher deposition in the south of Iceland which receives more precipitation.

### Precipitation and global fallout data

For the test a base station close to Reykjavík was chosen. Data for  $^{137}\text{Cs}$  concentration come from measurements conducted by the United Kingdom Atomic Energy Authority (UKAEA) in Harwell since April 1959 and the data are reported in annual reports. The values for  $^{137}\text{Cs}$  concentration in rain for 1959-1964 are estimates based on measurements of  $^{90}\text{Sr}$  and the values since 1965 are based on actual measurements. The precipitation data used in this study come from the Icelandic Meteorological Office and are regularly reported in their journal (Veðráttan, 1959-1991).

The data from this base station shows that 83% of the decay corrected deposition of  $^{137}\text{Cs}$  in the period 1959-1991 occurred during the eight years 1960-1967. The emphasis in this study was therefore placed on meteorological stations that have been in operation from 1959 to 1991; deposition for these years was estimated based on precipitation data for this period.

### Estimating annual precipitation

The average annual precipitation was calculated using the “optimized” method described in this chapter. For comparison an 8 year simple average (1960-1967) and a 30 year standard average period (1961-1990) were also tested.

The optimized method and the 8 year average gave similar estimates, but the 30 year average gave a considerably higher (ca. 30%) one.

### ***3.4 Correlation between calculated average annual precipitation and measured $^{137}\text{Cs}$ deposition***

The method used in the AMAP study for comparing deposition estimates and actual measurements was to force the regression line to go through the origin and calculate correlation coefficients on that basis. This approach will give a considerably higher value for the correlation coefficient than for an unbound regression line, but it can be justified on the basis that the assumption being tested is that deposition is directly proportional to precipitation. This same approach was used for each of the methods in this study.

The difference between the methods was not great, but the optimized method presented in this chapter gave highest correlation between estimates and actual measurements. Using the 8 year average gave very similar results and the 30 year average gave somewhat lower correlation.

A comparison of measured and predicted values for the first 14 sites in the study is shown in Figure 9.



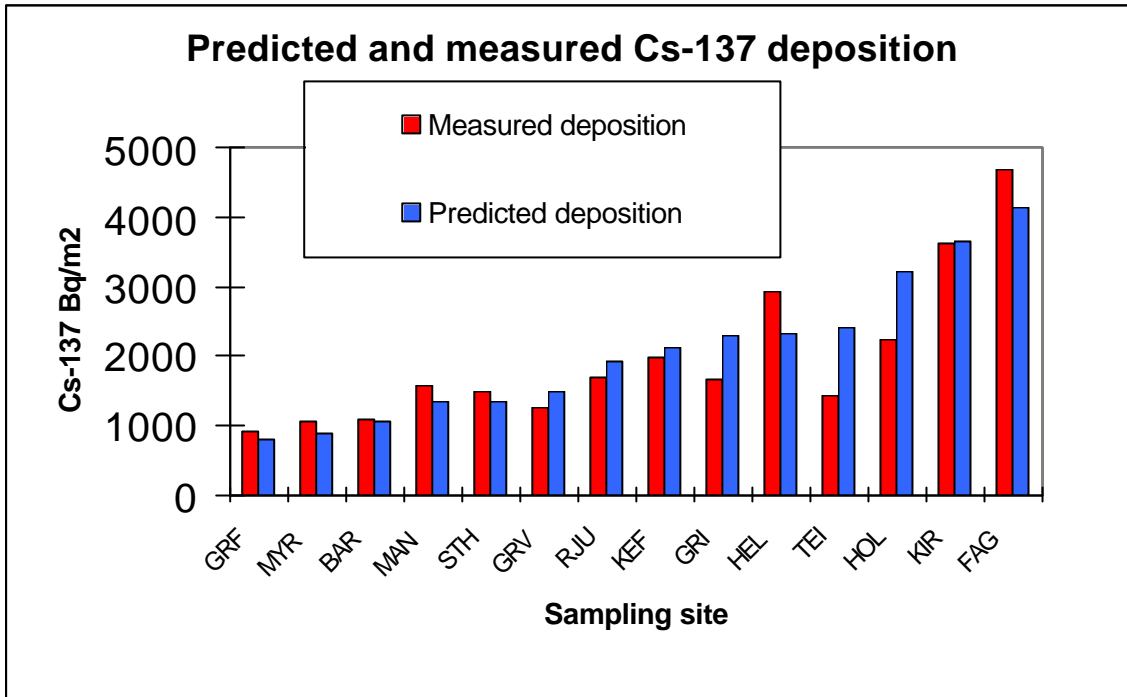


Figure 9: Measured and predicted (estimated) deposition for the first 14 sites in the study. Ratio of measured to estimated is 0.98 and  $r^2$  (for a line through origin) is 0.96. More sites have now been added, but the results are similar.

### 3.5 Discussion

The results show that the approaches outlined in this paper, of using precipitation data to predict spatial variation in global fallout  $^{137}\text{Cs}$  deposition, are valid and give reliable predictions. However, the strength of the correlation is surprising considering that dry deposition is not accounted for, although because of the high precipitation rate in much of Iceland dry deposition may not be expected to be an important contributor to total  $^{137}\text{Cs}$  deposition. In addition aeolian activity and lateral transport by erosion would be expected to occur in some Icelandic areas (Sigurgeirsson *et al*, submitted).

Wright *et al.* (1999) gave an estimate of the ratio of  $^{137}\text{Cs}$  deposition to precipitation of  $3.69 \pm 0.97 \text{ kBq m}^{-2}$  per 1000 mm precipitation for the end of 1985. This corresponds to  $2.6 \text{ kBq m}^{-2}$  per 1000 mm precipitation if the end of year 2000 is used as a reference. The Icelandic data gives corresponding values of 2.0 for a regression line and 2.4 for an average ratio. The value given for the normalised deposition density is equivalent to  $C_2$  for the same reference time.

The comparison of predicted and measured values for Iceland was much stronger than that originally reported by Wright *et al* (1999). In the AMAP study, data were analysed for 50 samples from Greenland, Norway and Russia from the period 1961-1985. A line through the origin was fitted to the data set using least squares regression giving an  $r^2$  value of 0.51 based on a coarse precipitation data set, disparate sources of measured  $^{137}\text{Cs}$  deposition using different sampling methodologies. In Iceland, the best comparison of predicted and measured values gives a corresponding  $r^2$  value of 0.95. This same correlation was obtained, both when the optimised method (using time varying weight factors) was used and when the prediction was based on average annual precipitation for the years 1960-1967. Some of the improved correlation compared with the AMAP study is probably due to the proximity of meteorological stations, where precipitation has been measured in a consistent manner.

Furthermore, the soil sampling was conducted by the same team, with a consistent, rigorous methodology over a short period of time. Consistent methodology of soil sampling and compiling precipitation data can be crucial for successful application of precipitation based deposition estimates.

### **Extending the time integrated decay corrected concentration coefficient to a wider area**

The preceding section shows that the time integrated decay corrected concentration can successfully be used as a tool for estimating radionuclide deposition from precipitation data over a country of the size of Iceland. The next step was to test over what range geographically the same coefficient can be used, if it could e.g. be used on a Nordic scale.

Time series for  $^{137}\text{Cs}$  concentration in rain water were obtained from the Nordic countries and the UK for the period 1960 – 1979. In some cases  $^{137}\text{Cs}$  data were not available for the whole period, but  $^{90}\text{Sr}$  data were. Gaps in the  $^{137}\text{Cs}$  time series in these cases were filled by  $^{90}\text{Sr}$  values multiplied by 1.5. This is standard ratio used by UNSCEAR in its latest report. It should be noted that experience shows that this ratio can be affected by various factors and that it should indeed not be regarded as a constant. In other cases the available data did not cover the whole of the period.

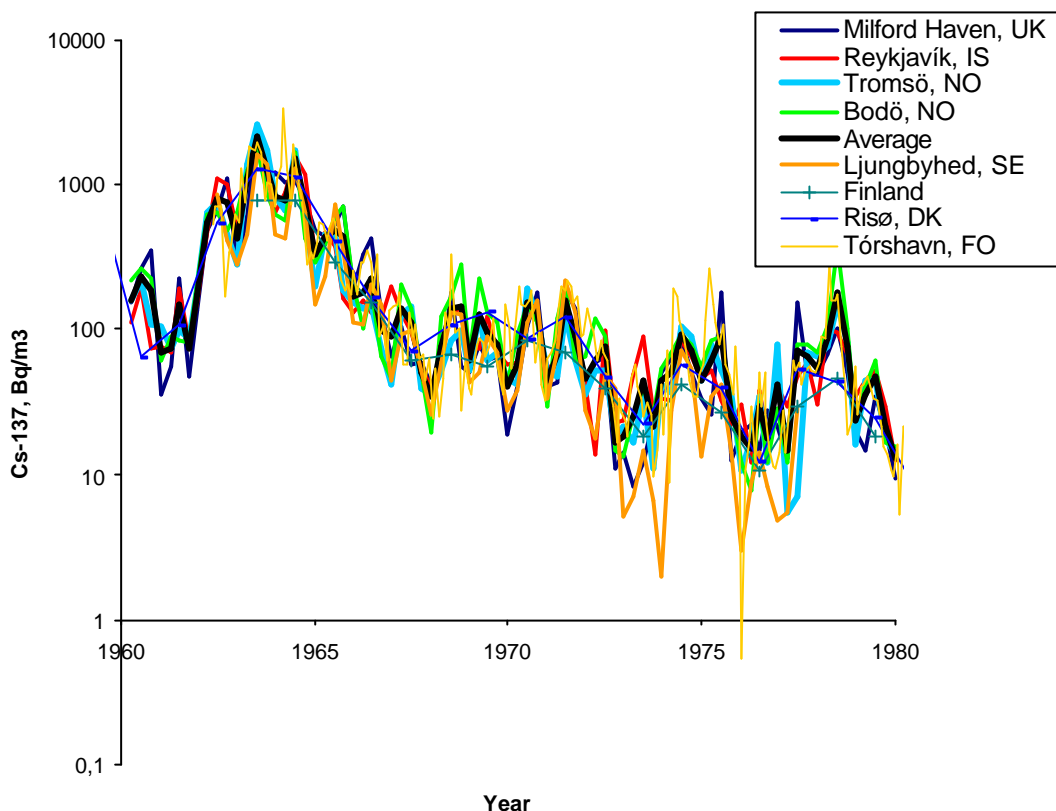
Data were obtained from the following sites

1. Reykjavík, IS
2. Tromsø, NO
3. Bodø, NO
4. Milford Haven, UK
5. Ljungbyhed, SE
6. Finland
7. Risø, DK
8. Tórshavn, FO

The first 4 sites were part of a network run by the Harwell Laboratory of the United Kingdom. The average for these 4 stations was used as a reference for comparing results. The time series for the concentration of  $^{137}\text{Cs}$  in rain water can be seen in Figure 10.

The time series are quite similar, even though some fluctuation between series is apparent. In some cases the fluctuations can be expected to even out, a higher value in one period can be compensated by a lower one in the next period. It should also be noted that the frequency of sampling has its effect.

### Cs-137 in rain water, decay corrected to 1.1.1965



**Figure 10: Time series for concentration of  $^{137}\text{Cs}$  in rain water at a few sites in the Nordic countries and one in the United Kingdom.**

The time integrated decay corrected concentration,  $C_T$ , can be used to test how well the assumption holds, that the  $^{137}\text{Cs}$  concentration in the rain water is similar over a wide area, at least with respect to a cumulative effect such as radionuclide deposition. The value  $C_T$  was calculated for each site and compared with the reference value calculated based on the average for the 4 stations in the Harwell network (Table 3). If a complete time series was not available for the period of interest, then a comparison was made with a corresponding part of the time series for the 4 station Harwell network average.

**Table 3: Deviation of estimated value of  $C_S$  for each station from the average for the four stations in the Harwell network (Milford Haven, Reykjavik, Tromsø and Bodø)**

Site:	Reykjavík	Tromsö	Bodö	Milford Haven	Ljungbyhed	Finland	Risø	Tórshavn
% deviation	0 %	- 2 %	- 1 %	2 %	- 23 %	- 5 %	1 %	8 %

All the sites, apart from one, show good agreement, especially bearing in mind that the stations are from different networks and in some cases the  $^{137}\text{Cs}$  values are derived from  $^{90}\text{Sr}$  values.

### 3.6 Conclusion

It is clear that the method presented can be used successfully as an aid to estimate  $^{137}\text{Cs}$  fallout deposition values from precipitation data, not only on a national level but also on a wider scale.

### 3.7 References

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## 4. Contamination of radionuclides in milk

### 4.1 Description of collated milk results

Data concerning milk contaminated with  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  has been collated from all Nordic countries for both the nuclear weapons fallout (NWF) period and the post-Chernobyl period. All results have been registered in an excel database. The data sometimes lack consistency (fresh vs dry milk; weekly, monthly, quarterly or yearly sampling; varying sampling place for different years), but some very good time series have also been collated. The emphasis has thus been on long time series with good consistency. Some results for  $^{131}\text{I}$  in milk are also gathered, but due to the short physical half-life of this isotope, the data are sparse.

#### 4.1.1 Cs-137 in cow`s milk

The data from Denmark consist of monthly samples from the Danish islands (Sjælland, Bornholm, Fyn) and from the peninsula Jylland, and form two continuous time-series from 1959 onwards. The samples are mainly dry milk but some samples of pasteurised milk and untreated whole milk are also included. All samples are from Danish dairies. The results are originally reported as Bq  $^{137}\text{Cs}$  per kg K, therefore a conversion factor of 1.66 g K per litre milk have been used to estimate content in fresh milk.

The same approach was used for reported data from the Faroe Islands. The monthly samples were locally produced milk from Klaksvig and Tvøroyri, and from a dairy in Torshavn which collects milk from most of the country.

The Icelandic data are monthly values from dairies in Egilsstaðir, Grafarnes, Hornafjörður and Reykjavik for the NWF period while samples were taken from dairies in Akureyri, Blonduos, Borgarnes and Selfoss both in the NWF and the post-Chernobyl period. The data from the NWF period are only from 1964 to 1967 and the post-Chernobyl data starts in 1990.

For Finland, milk data from two regions are available. In south-western Finland dry milk was sampled from 1960 to 1989; after this, samples have been dairy milk. In Lapland the samples were from dairies (Kursu and Rovaniemi) and from individual farms (Apukkajärvi and Vikajärvi). In Kursu, sampling lasted from 1963-88, whereas samples from Rovaniemi were gathered in two periods (milk powder in 1966-75 and dairy milk from 1986 onwards). Farm samples were taken in Apukkajärvi in 1975-77 and after the Chernobyl accident 1986-1991. After that farm specific sampling continued in Vikajärvi (1991 onwards). Data on  $^{137}\text{Cs}$  in Finnish milk is published in Castrén, 1966; Paakola and Castrén, 1967; Rantavaara et al., 1998; and the annual reports on Studies of environmental radioactivity from STUK, 1968-1988.

In Norway, samples have been collected from many dairies and farms from most parts of the country. However, some time series are rather short. The longest and most consistent series are from Vadsø, Kautokeino, Bardufoss/Målselv, Bodø, Værnes, Trondheim, Røros, Lillestrøm, Kristiansand, Bergen and Florø. For these sites data from 1960-81 are available in the database as yearly, monthly or quarterly values. During 1962-65 yearly averages from a large amount of dairies from all counties were taken. Corresponding investigations were done post-Chernobyl. Those data are not included at present, but will be included in the database shortly. In 1988 the summer monitoring program in Norway was started, with the aim of

collecting weekly milk samples from the most contaminated areas during the grazing period (May-September). These data are all farm specific.

No Swedish data are included in the database for the NWF period. Time-series for several dairies from the early 1960`s onwards are presented in Suomela and Melin (1992), and data from the two northernmost dairies (Vittangi and Tarnby) are included in the AMAP report (2004). At present these data are not included in the excel database, but they will be included shortly. For the post-Chernobyl period, the Swedish data comprised a weighted country average as well as dairy samples from Malmö, Tomelilla, Karlskrona, Kalmar, Vaxjö, Borgholm, Vastervik, Visby, Halmstad, Jönköping, Vetlanda, Dingle, Göteborg, Skövde, Örebro, Karlstad, Vasterås, Enskede, Kallhall, Uppsala, Grådö, Gävle, Bollnas, Sundsvall, Östersund, Örnsköldsvik, Umeå, Lycksele, Skellefteå, Piteå, Luleå and Hedenaset. They were all quarterly samples of fresh milk.

#### **4.1.2 Sr-90 in cow`s milk**

The NWF and post-Chernobyl data for  $^{90}\text{Sr}$  in Denmark are from the Danish islands (excluding Bornholm) and from the peninsula Jylland. Some samples are given for East Denmark including Bornholm as well. The samples are mainly dry milk but some samples of pasteurised milk and untreated whole milk are also included. All samples are from Danish dairies. The results are given in Bq  $^{90}\text{Sr}$  per kg Ca, and the conversion factor used to calculate content in fresh milk is 1.2 g Ca per litre milk.

Results are reported in the same manner for the Faroe Islands where samples are locally produced milk from Klaksvig and Tvøroyri, and from a dairy in Torshavn which collects milk from most of the country. Both the NWF and the post-Chernobyl period are covered.

In Finland the  $^{90}\text{Sr}$  results were from Lapland and south western Finland. In Kursu samples were fresh milk from the NWF period and 1986. In Apukka time series for fresh milk are from both the NWF and post-Chernobyl period, while for Rovaniemi the data from pre-Chernobyl was dry milk. In Vikajärvi the samples were all fresh milk from the post-Chernobyl period. The samples from south-western Finland were dry milk from the NWF period and up to 1987, after that fresh milk was measured. The data on  $^{90}\text{Sr}$  in milk is published in Paakola, 1966; Paakola and Castrén, 1967; and the annual reports on Studies of environmental radioactivity from STUK, 1968-1988.

The longest time series for  $^{90}\text{Sr}$  in Norway is from the Kjeller area near Oslo, including both the NWF and post-Chernobyl period (1964 onwards). Before 1980, weekly samples were combined to monthly samples before analysis. From 1980 the samples were combined to quarterly samples before analysis. Some farm and dairy results also exist from 1997 to 2001 (Liland et al., 2002).

In Sweden the  $^{90}\text{Sr}$  data are quarterly samples from dairies in Malmö, Visby, Kalmar, Göteborg, Stockholm, Uppsala, Gävle, Sundsvall og Lycksele, all post-Chernobyl.

There are no  $^{90}\text{Sr}$  data for milk from Iceland.

### 4.1.3 Iodine -131 in cow`s milk

Some data concerning  $^{131}\text{I}$  in milk have also been collated. For the NWF period, only data from Norway have been available, the most important being:

- Data from 10 (non-specified) dairies (1953-63)
- Data-series from Lillestrøm, Røros, Sandnes and Oslo (1957)
- Data-series from Bardufoss, Bergen, Bodø, Florø, Kautokeino, Kristiansand, Lillestrøm, Røros, Trondheim, Vadsø and Værnes (1961 and 1962).

For the post-Chernobyl period, data from BIOMOVs (1991) have been collated; the 3 series included are Risø, Denmark (May 1986); Tranvik, Sweden (April to June 1986); and Loviisa, Finland (April to July 1986).

## 4.2 Description of sampling locations

Information on soil properties and precipitation rates has been gathered for different sampling regions. The degree of details varies from country to country, depending on available information.

### 4.2.1 Finland

The cultivated land in northern Finland is predominantly peat and humus soils; there are no clay soils in the area. In south-western Finland prevailing soil types are clay and sand. The pH of cultivated soils in south-western Finland is lower (5.3-5.6) than in northern Finland (5.8-6.1). The calcium and potassium contents of cultivated land in northern Finland are considerably lower than those in south-western Finland. The fertilization practises have changed from the 1960's, but the contents of nutrients in cultivated land in northern Finland are still substantially lower than those in south-western Finland.

Percentage proportions of different soil types of arable land and soil characteristics are given in Table 4 and 5 for the milk production areas of northern and south-western Finland (Viljavuuspalvelu Oy, 2001).

**Table 4: Northern Finland – soil types and characteristics**

Carex peat soils	33%
humus soils	8%
fine and finer sand soils	32%
sand and fine sand moraine soils	24%
Ca	960 mg/l in the 1960's and gradually increasing up to 1320 mg/l in the 1990's
K	80 mg/l in the 1960's and gradually increasing up to 104 mg/l in the 1990's

**Table 5: South-western Finland – soil types and characteristics**

clay soils (sandy and silty clay)	66%
humus soils	6%
fine sand and silt soils	22%
moraine soils	5%
Ca	1850 mg/l in the 1960's and gradually increasing up to 2280 mg/l in the 1990's
K	210 – 230 mg/l

In northern Finland the annual precipitation is 500-550 mm while in south-western Finland the precipitation ranges from 550 to 750 mm per year (E. Kostainen, pers. com.).

#### **4.2.2 Denmark**

In the western part of Denmark (Jylland) the dominating soil classes are loamy sand, sandy clay loam and sand. The precipitation range is 654 to 840 mm/y. In the islands, the dominating soil classes are sandy clay loam and sandy loam. The precipitation is 517-685 mm/y. The pH in Danish soils is 6-7 (S.P. Nielsen, pers. com.).

#### **4.2.3 The Faroe Islands**

Soil descriptions do not exist for the milk sampling stations. However, data exist for sheep pasture areas, some of which are close to the milk sampling stations. The following descriptions are probably representative for the milk stations:

Torshavn: 30% organic material, pH 5.1, 60 mg K and 39.8 mg Na per 100 g soil.

Klaksvik: 67% organic material, pH 4.7, 64.3 mg K and 43.8 mg Na per 100 g soil.

Tvøroyri: 59.5% organic material, pH 5.0, 71.4 mg K and 83.1 mg Na per 100 g soil.

The Torshavn dairy collects milk from most of the country, so the results there would not be reflected by the soil properties in Torshavn alone.

The annual precipitation given as the norm for 1961-90 is 2710mm in Klaksvík and 1284mm in Tórshavn. No reference value is given for Tvøroyri (Cappelen, J. and Laursen, E. V., 1998).

#### **4.2.4 Iceland**

Data for soil types and precipitation for the milk sampling stations in Iceland are given in Table 6. The west Iceland area is dominated by histosols, a soil type with more than 20 % organic matter and low pH (4.0 – 5.5). The other areas are dominated by andosols varying from histic andosols (poorly drained areas, 13-18 % organic matter, pH 4.5 – 5.5) to brown andosols (freely drained areas, 2-7 % organic matter, pH 5.5 – 7.5).



**Table 6: Milk sampling stations in Iceland**

Area	Sampling station	Main soil type	Precipitation, mm/y
West Iceland	Borgarnes	Histosol	800-900
North west Iceland	Blöndus	Brown, gleyic and histic andosols	400-600
Eyjafjördur	Akureyri	Brown, gleyic and histic andosols	500-600
East and south east Iceland	Egilsstaðir Hornafjörður	Brown, gleyic and histic andosols	600-800 1200-1400
Central south Iceland	Selfoss	Brown, gleyic and histic andosols	1100-1200
South west Iceland	Reykjavik Grafarnes	Brown andosol	800-1000

#### 4.2.5 Sweden

The milk sampling stations in Sweden can be divided in three groups according to average annual precipitation, given in Table 7 (Gyldendal, 1995). Data on soil types are not yet available.

**Table 7: Annual precipitation for Swedish sampling stations**

250-500 mm/y	500-750 mm/y	750-1000 mm/y
Malmö	Hedenäset	Halmstad
Tomelilla	Luleå	Göteborg
Karlskrona	Umeå	
Kalmar	Örnsköldsvik	
Visby	Östersund	
Gävle	Skövde	
Bollnäs	Karlstad	
Sundsvall	Örebro	
Lycksele	Jönköping	
Stockholm	Grådö	
	Uppsala	

#### 4.2.6 Norway

The annual precipitation average for 1960 to 1974 and dominating soil types in the main sampling areas (NWF) are presented in Table 8. Podzols are acid soils with low cation exchange capacity consisting of sandy or coarse-loamy texture mixed with organic matter. Podzoluvisols have high clay content, mixed with other minerals and/or organic matter and a base saturation of >50 %. Leptosols are weakly developed shallow soils with a base saturation <50 %. Gleysols are formed from unconsolidated materials. They are water logged part of the year, and the low redox conditions results in mobile iron compounds (FAO, 1988).

**Table 8: Precipitation rates and dominating soil types for some Norwegian milk sampling areas**

<b>Location</b>	<b>Precipitation (mm/y)</b>	<b>Soil types</b>
Kristiansand	1367	Podzol
Lillestrøm	924	Podzol, Podzoluvisol
Bergen	2039	Leptosol, Podzol
Florø	1854	Leptosol
Røros	497	Podzol
Trondheim	844	Podzoluvisol, Podzol
Værnes	850	Podzol, Podzoluvisol
Bodø	1032	Podzol
Bardufoss	641	Podzol, Leptosol
Vadsø	513	Leptosol, Gleysol, Podzol
Kautokeino	336	Podzol

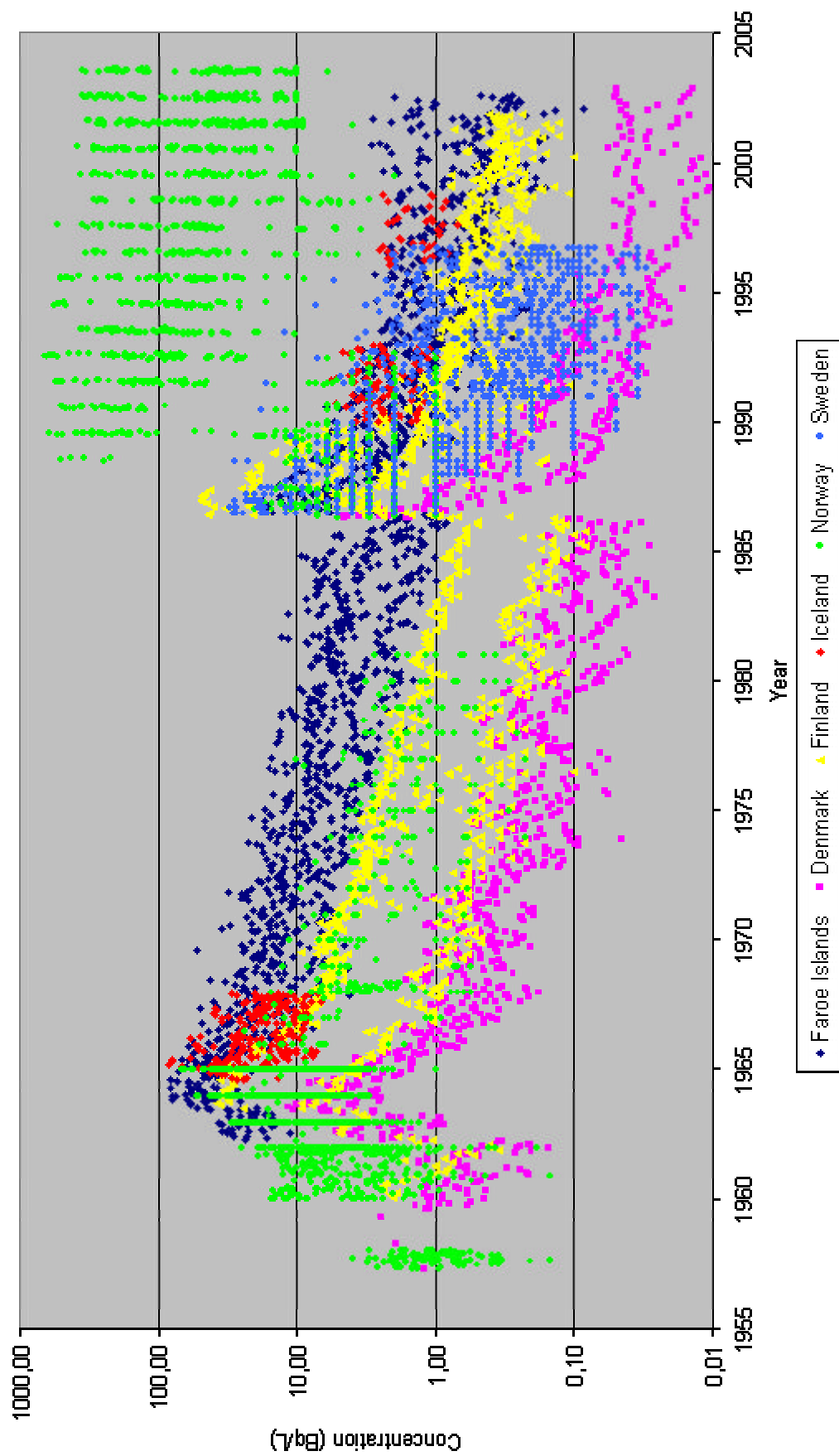


Figure 11: Cs-137 in cow's milk from the Nordic countries

### 4.3 Summary of data

A summary of the collated data for  $^{137}\text{Cs}$  in cow's milk from all the Nordic countries is shown in Figure 11. All the data follows approximately the same time trend. The contamination level varies considerably, though, dependent on factors such as deposition level and soil properties in different regions. This is particularly evident in Finland where there is a marked difference between Lapland (top yellow clustering) and the south west (bottom yellow clustering). During the NWF period the deposition level was similar in the two areas, but the contamination level in Lapland is more than twice as high as in the south west. This is most likely due to predominantly organic soils and nutrient deficiency which gives a higher transfer of Cs-137 in Lapland compared to the clayish soils in the south west.

In Denmark there is a difference in contamination level between Jylland (top pink clustering) and the islands (bottom pink clustering) after the Chernobyl accident. This is probably due to the difference in deposition since Jylland had areas with higher deposition than the islands.

The Faroe Islands show no distinct differences between regions. The milk contamination is the highest reported for the Nordic countries in the NWF period. The data seem to correspond with the Icelandic data, but since samples from Iceland are only for limited time periods (1964-67 and 1990-98) it is difficult to make a good comparison between Iceland and other countries.

The Swedish data are from 1986 until 1996. There is a regional difference in the reported data. This is due to the spotty character of the Chernobyl deposition. The higher level samples are from the mid-eastern part of Sweden which experienced the highest national deposition. Many reported values were below the detection limit. The value plotted was set at one half of the detection limit.

The data from Norway show a large variance. They take an intermediate stand in contamination level during the NWF period, while the post-Chernobyl data are clearly much higher in parts of Norway than in the other Nordic countries. This is mainly due to high deposition in some Norwegian areas. Many farms joined the summer monitoring program in 1988/89 which explains why the peak values are found around these years instead of in 1986/87. The data from individual farms are only summer values which may add to the explanation of the very high reported values in Norway post-Chernobyl.

The time series for  $^{90}\text{Sr}$  in cow's milk is shown in Figure 12. Again the Faroese data show the highest contamination level in the Nordic countries for the NWF period, while the Norwegian data generally are highest for the post-Chernobyl period. No results from Iceland are available.

There is a gap in contamination levels for the NWF data for Norway. The higher green band is data from Bergen and Florø, situated on the west coast of Norway. Due to the high precipitation rate in this coastal area the estimated deposition was higher here than in other parts of the country.

No large regional differences within other countries are apparent for  $^{90}\text{Sr}$ . This indicates that soil types are less important in determining the transfer factors to milk for  $^{90}\text{Sr}$  than for  $^{137}\text{Cs}$ . The time trends are not similar for all the different countries, indicating that the factors influencing the ecological half-life for  $^{90}\text{Sr}$  are not entirely the same as for  $^{137}\text{Cs}$  in the pasture – milk system.

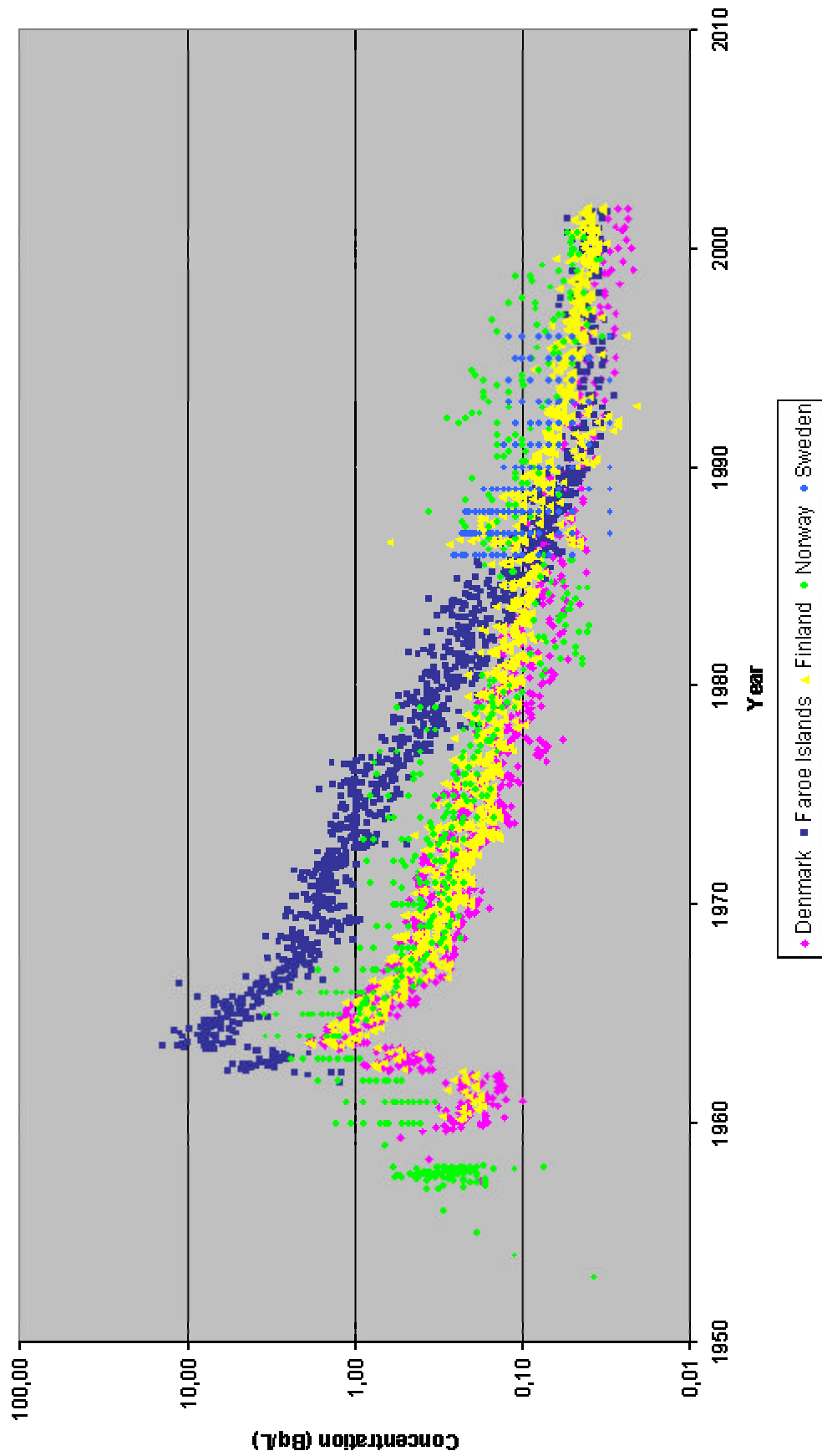
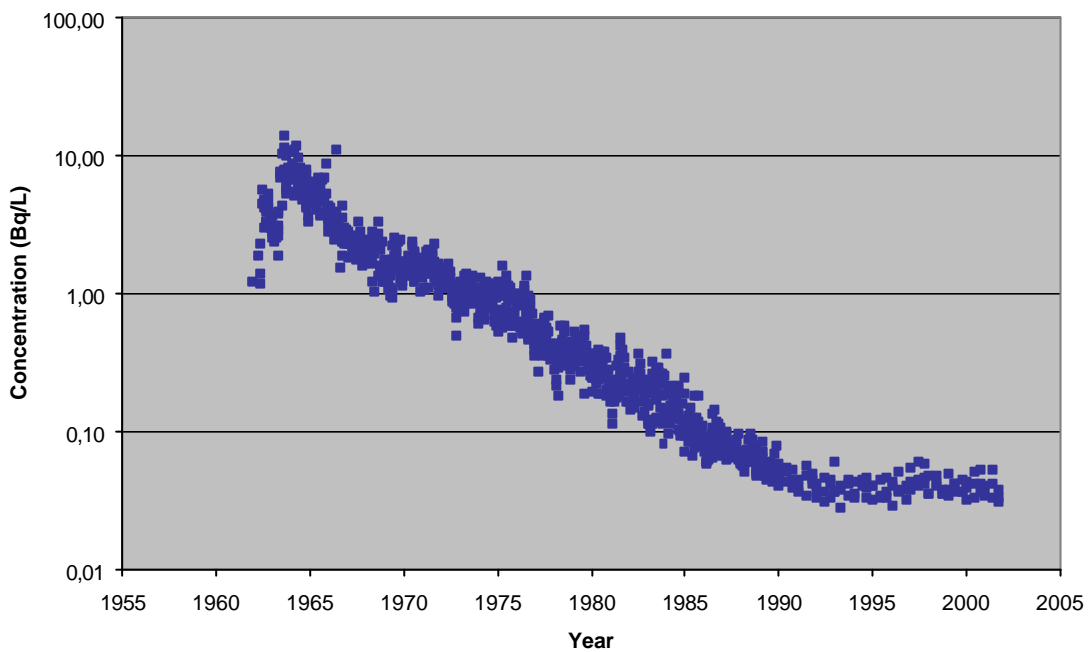


Figure 12: Sr-90 in cow's milk from the Nordic countries

The time trend for the three sampling sites at the Faroe Islands is extracted in Figure 13. The trend is differing from the other Nordic countries presented in Figure 12. The Chernobyl accident did not contribute significantly to the  $^{90}\text{Sr}$  milk contamination in the Faroe Islands. There is a decreasing trend in the contamination level until 1990 when the contamination seems to flatten out. There is no apparent reason for this. Actually, we would expect a continuing decrease also after 1990. The results are not close to the detection limit, so it cannot be explained by the measurement techniques. Measurements of  $^{90}\text{Sr}$  in precipitation in the Faroe Islands follow the same trend, but this still doesn't explain why the contamination in milk flattens out.



**Figure 13: Monthly values of  $^{90}\text{Sr}$  in milk in the Faroe Islands from 1961 to 2001**

Average levels of  $^{131}\text{I}$  in milk from 10 Norwegian dairies are given in Figure 14. Seasonal and geographical variations in  $^{131}\text{I}$  activity concentrations in 1957 and 1962 are shown in Figure 15 and Figure 16, respectively. For the latter series particularly high levels are observed for sampling stations in northernmost Norway (i.e. Kautokeino). This is most likely due to the nuclear tests at Novaja Zemlja in September-October 1962. Such a trend is also observed in Swedish data from certain farms in Norrland - where milk levels, during the actual time-span reached 75 Bq/L (Suomela and Melin, 1992).

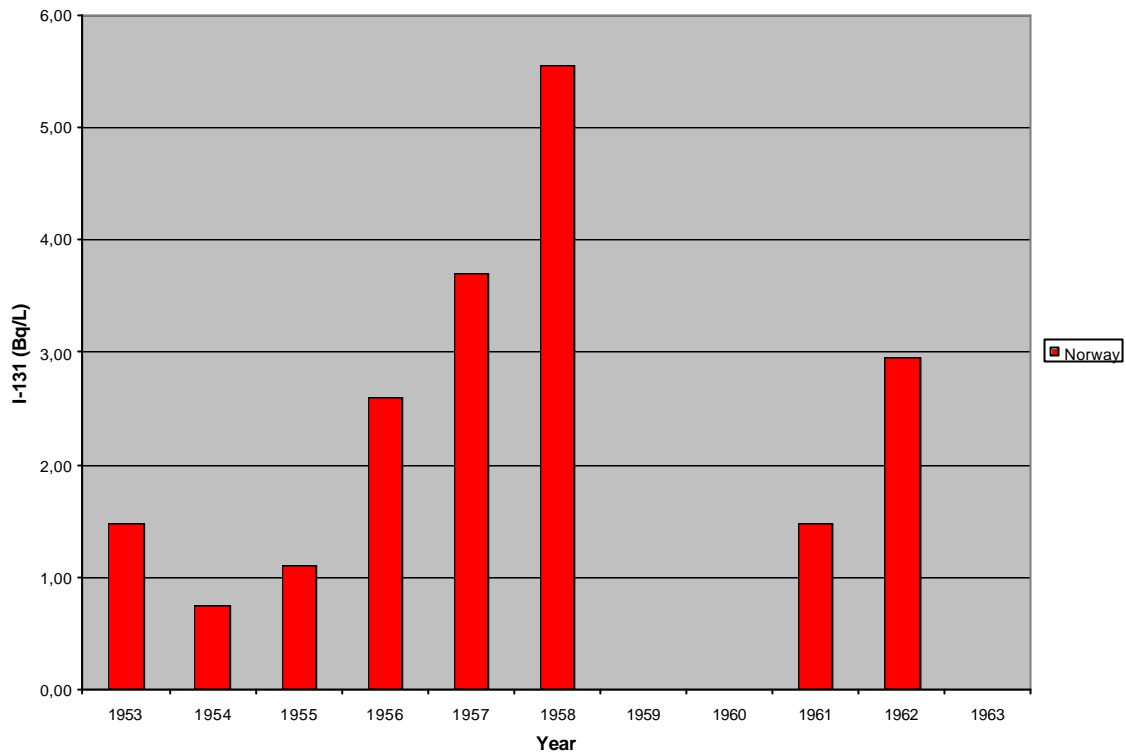


Figure 14: I-131 activity concentrations in milk (Bq/L) from 10 dairies in Norway

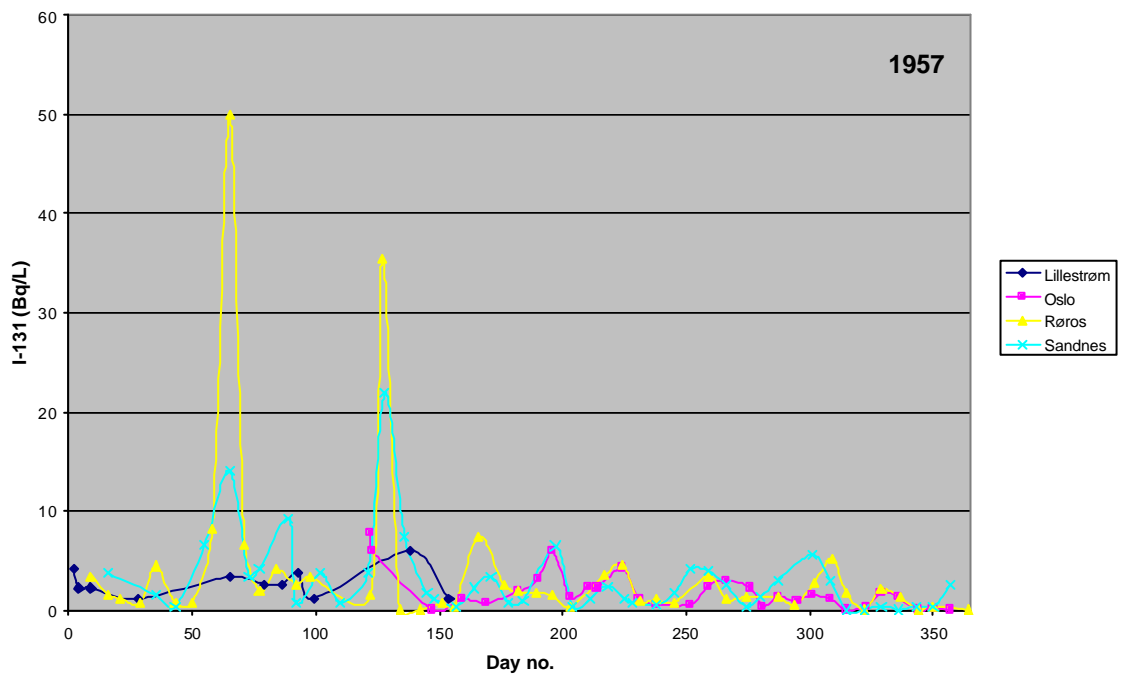


Figure 15: Temporal variability of  $^{131}\text{I}$  activity concentrations in milk from various Norwegian stations in 1957

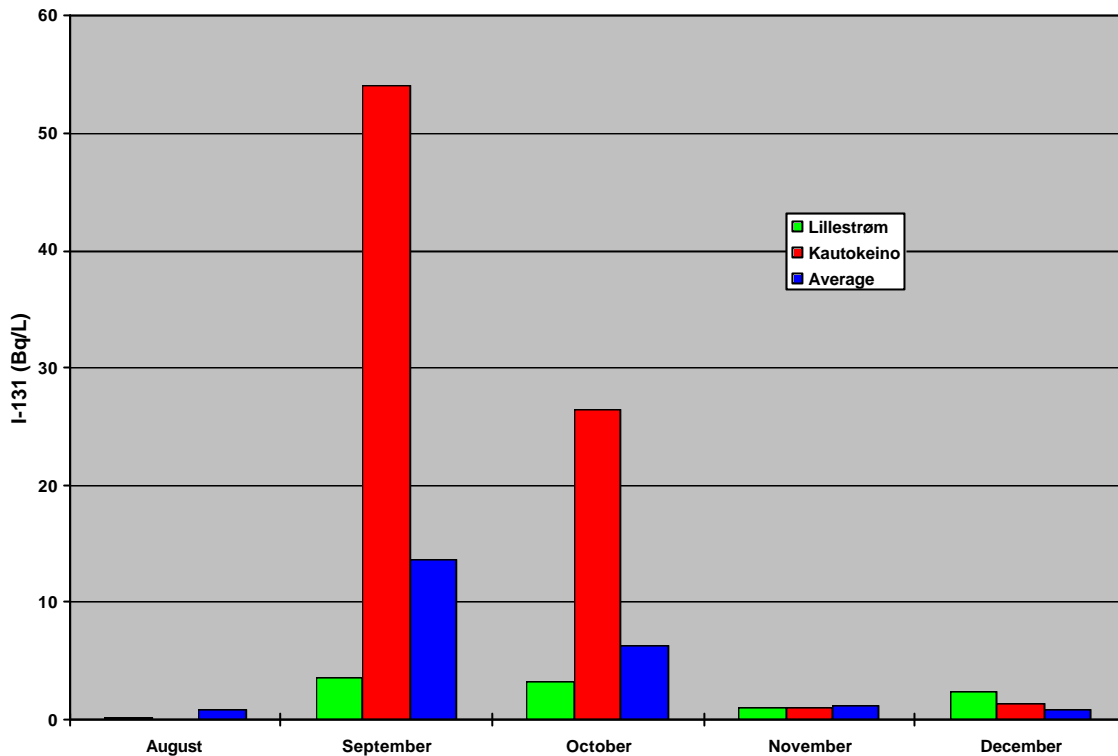


Figure 16: I-131 activity concentrations in milk sampled autumn/winter 1962 from 11 Norwegian sites

After the Chernobyl accident, the highest levels of  $^{131}\text{I}$  in Swedish milk were measured in Gotland during April-May 1986. Milk from some farms even contained as much as 600 Bq/L, whereas dairy milk activity concentrations peaked at about one third of that level. In contrast, dairy milk from Uppsala and Gävle did not exceed 60 Bq/L (SSI-report 12, 1986). The earlier mentioned time-series for Sweden, Finland and Denmark are given in Figure 17. Here, peak values correspond to the beginning of the pasture period for dairy cows.

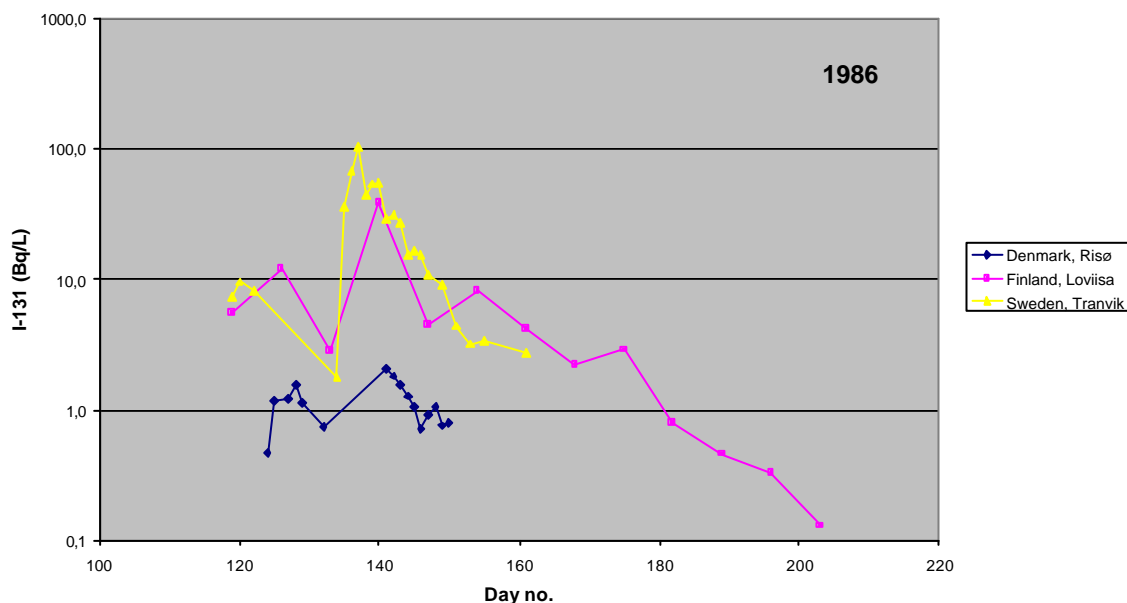


Figure 17: Time-series for  $^{131}\text{I}$  from various Nordic areas after the Chernobyl accident



#### 4.4 Effective ecological half-lives for the NWF period for some selected time-series

As a basis for a more thorough study, a dual exponential regression analysis on some of the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  time-series was performed. Earlier studies (e.g. H. S. Hansen and I. Andersson, 1994; E. Kostianen and K. Rissanen, 2003) have shown that when using single exponential decay, the effective ecological half-life increases with time. This suggests that there is actually a two-component decay with one fast half-life early on, followed by a longer component. A dual regression analysis has thus been used to explain the contamination development with time.

The most suitable time-series, in this respect, were the two Danish series, the three series from the Faroe Islands and the two from Finland. These series were chosen since they represent the whole concentration range (from Denmark to the Faroe Islands), they are long, rather detailed and consistent – monthly values are given from the early sixties to the Chernobyl accident. SPSS non-linear regression analysis was used. The data were ln-transformed to decrease the impact of inhomogeneous variance. Consequently, the model expression was as follows:

$$Y = \ln \left[ A1 \frac{\ln 2 \cdot t}{T1} + A2 \frac{\ln 2 \cdot t}{T2} \right] \quad (9)$$

where

A1: Start activity, component 1

T1: Effective ecological half-life for component 1 (fast component)

A2: Start activity, component 2

T2: Effective ecological half-life for component 2 (slow component)

t : Time elapsed since reference date

Since the milk data show considerable seasonal variation, the monthly values were combined to an annual average before the regression analysis was performed. Starting point of the regression was 1964, and the end point was 1985. Figure 18 shows the results for  $^{137}\text{Cs}$  for time-series from Thorshavn in the Faroe Islands, Kursu and south-western Finland, and the Danish islands. The results for  $^{90}\text{Sr}$  are shown in Figure 19. The regression lines seem to well represent the time development of contamination in milk. The results from the regression analyses are summarised in Table 9. In most cases the fast component, T1, is largely dependent on the starting point of the regression analysis (i.e. year and month). The starting point of the regression was set at 1964 since this corresponds to the year of peak values for most time-series. Looking at the results, we see that T1 is around 1 year for all sites for both radionuclides. The slow component, T2, is quite stable regardless of starting point. The long half-life for  $^{137}\text{Cs}$  is about 6 years for all localities except for south-western Finland that shows a slightly longer T2 half-life of about 7 years. For  $^{90}\text{Sr}$ , on the other hand, there is a considerable geographical variation in the T2 half-life. The shortest T2 half-life is for the Faroe Islands of about 4 years, while the Danish data gives a T2 half-life of about 7 years. The Finnish data give a T2 half-life of about 12 years, approximately 3 times longer than for the Faroe Islands.

The results for caesium indicates that soil types have an influence on the amount of deposited  $^{137}\text{Cs}$  that is transferred to the milk, but that this does not influence on the effective ecological half-lives in the pasture-milk system.

For strontium there are clearly factors that influence on the long term component since the T2 effective ecological half-lives varies from about 4 to about 12. There is no consistent trend

related to chemical soil parameters (pH, % clay, % organic matter), annual precipitation or intensive vs extensive farming practice. The reason for the differences is thus not known, but needs to be investigated further.

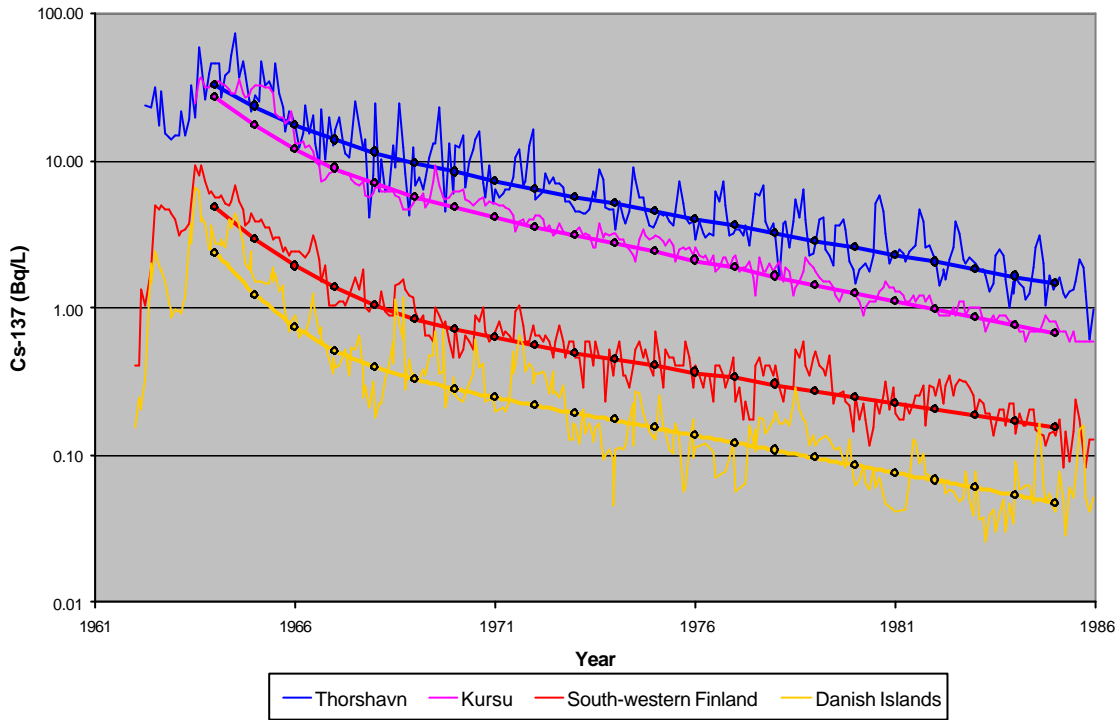


Figure 18: Cs-137 dual exponential regression analysis for four time-series from 1964 to 1985

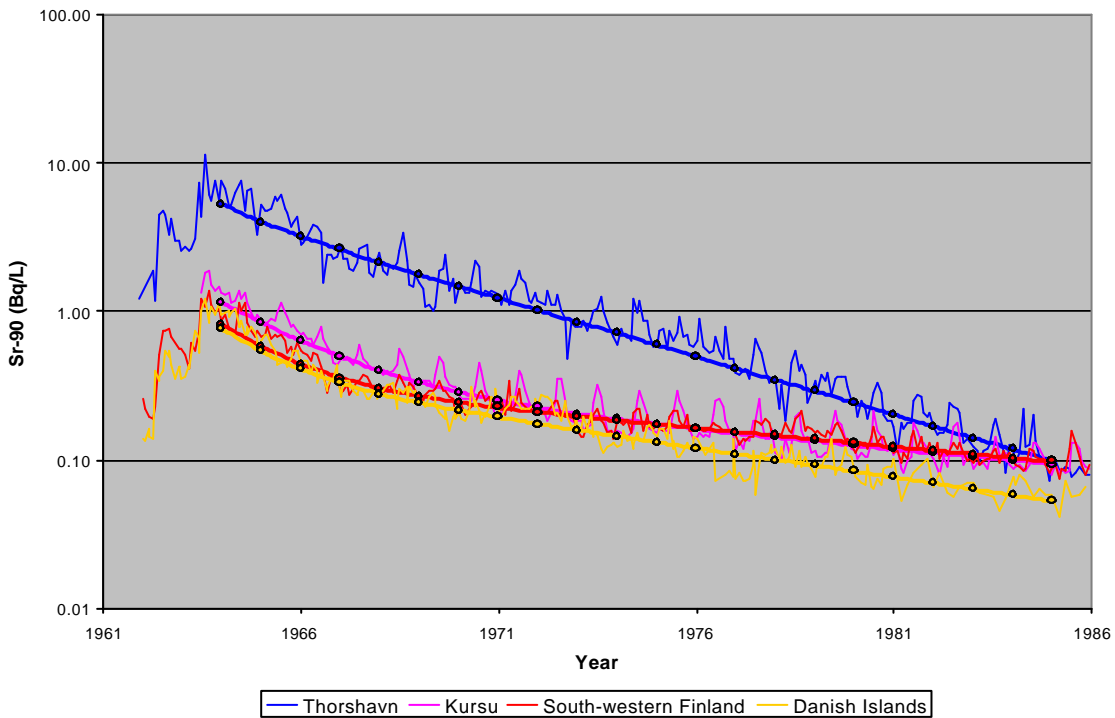


Figure 19: Sr-90 dual exponential regression analysis for four time-series from 1964 to 1985

**Table 9: Summary of regression analyses results for selected time-series**

	<b>Effective ecological half-life 1±SE<sup>2</sup> (years)</b>	<b>Effective ecological half-life 2±SE (years)</b>
<b>Cs-137</b>		
West-Denmark (Jutland)	0.7±0.1	5.8±0.2
East-Denmark (Islands)	0.8±0.1	6.0±0.3
Faroe Islands (Thorshavn)	1.1±0.2	6.2±0.4
Faroe Islands (Klaksvig) <sup>3</sup>	-	-
Faroe Islands (Tvøroyri)	1.0±0.3	6.5±0.3
North Finland (Kursu)	1.0±0.1	5.4±0.1
South-western Finland	1.0±0.1	7.2±0.4
<b>Sr-90</b>		
West-Denmark (Jutland)	0.9±0.1	7.0±0.2
East-Denmark (Islands)	1.1±0.1	7.6±0.3
Faroe Islands (Thorshavn)	0.8±0.4	3.8±0.1
Faroe Islands (Klaksvig)	0.8±0.2	4.2±0.1
Faroe Islands (Tvøroyri)	0.7±0.2	4.4±0.1
North Finland (Kursu)	1.6±0.1	11.9±1.2
South-western Finland	1.2±0.1	12.0±0.6

#### 4.5 Conclusions

A large amount of data has been collated from the Nordic countries and registered in an excel database. More data will be incorporated when available. The measurement results of <sup>137</sup>Cs and <sup>90</sup>Sr in milk show that the differences in contamination level can be substantial not only between countries but also between regions within one country. The transfer of <sup>137</sup>Cs to milk is dependent on the deposited amount and soil types in the pasture areas, while the transfer of <sup>90</sup>Sr to milk seems to be dominated by the deposited amount.

The traditional calculation of effective ecological half-life has been done using a single exponential decay regression. Most reported values for <sup>137</sup>Cs in milk from various studies lie in the range 1-3 years with some longer estimates for certain regions e.g. Lapland in Finland. The dual component regression used in this study fits the data well with a small appurtenant standard error. For <sup>137</sup>Cs the effective ecological half-life seems to be fairly equal for the different investigated regions - about 1 year for the fast component and 6 years for the long component. No difference in half-life is apparent between areas with different soil types, precipitation rates or intensive vs. extensive farming practices. The long component of about 6 years is considerably longer than most other reported effective ecological half-lives.

The effective ecological half-life for <sup>90</sup>Sr is about 1 year for the fast component in all investigated regions while the long component varies between 4 and 12 years. The reason for this is not apparent and needs further investigation.

<sup>2</sup> Asymptotic standard error

<sup>3</sup>Regression analysis gave anomalous results (compared to the other two Faroese series): the T2 was more than twice as long and had a large appurtenant standard error (about 30%). This suggests that the method used is not suitable for these data. Thus the results have not been included.

#### 4.6 Recommendations for further work

This study has revealed interesting similarities and differences in milk contamination features across the Nordic countries. Yet, some Nordic contamination data for milk has not been included in the database. This will be our first priority for the next project period. Secondly, further elaboration of the Nordic data is necessary to produce more time-series for the NWF period where dual component regression can be done. A third step will be to elaborate good time-series for the post-Chernobyl period and perform dual component regression analysis to determine effective ecological half-lives in the pasture-milk system. The results from the NWF period can then be compared to the post-Chernobyl ones. Finally, we would like to combine the deposition estimates from precipitation rates with the milk contamination data to determine transfer factors for the NWF period for both  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ .

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## 5. Regional differences in $^{137}\text{Cs}$ effective ecological half-lives in reindeer

With the contribution of B. Åhman, Reindeer Husbandry Unit, Dept. of Animal breeding and Genetics, Swedish University of Agricultural Sciences.

### 5.1 Introduction

Radiocaesium contamination of reindeer has been extensively studied in the Nordic countries and many Arctic areas since the early 1960s, following the finding of higher radiocaesium concentrations in humans who habitually consumed reindeer (Lidén 1961). The vulnerability of reindeer and reindeer meat consumers to radiocaesium contamination was again demonstrated by the consequences of the Chernobyl accident for reindeer herding in mid Sweden and mid and south Norway (e.g., Åhman and Åhman 1994, Ågren 1998, Strand et al. 1992, Mehli et al. 2000). Seventeen years after the Chernobyl accident, countermeasures are still required in some Norwegian (see Figure 20 and Figure 21) and Swedish reindeer herding districts before animals can be slaughtered for trade to comply with radiocaesium food intervention limits of  $3000 \text{ Bq kg}^{-1}$  in Norway and  $1500 \text{ Bq kg}^{-1}$  in Sweden. Radiocaesium contamination levels in the southern Saami population are today comparable to, or higher than, those in people close to Chernobyl. A thorough understanding of reindeer radioecology is therefore an inevitable prerequisite in Nordic emergency preparedness.

Early studies of the transfer of radiocaesium in the lichen - reindeer – human foodchain focused primarily on the northern areas of Fennoscandia. The consequences of the Chernobyl accident were more significant further south. As long term Chernobyl data series are becoming available, attention has been given to emerging differences in effective ecological half-lives between different regions (e.g. Åhman et al. 2001, Rissanen et al. 2003). This review will attempt to summarize the present knowledge.

Although historically most emphasis has been given to caesium isotopes, other radionuclides are also important in the lichen – reindeer – human foodchain. The natural nuclide  $^{210}\text{Po}$  will, in many areas, contribute more to the radiation dose of reindeer meat consumers than  $^{137}\text{Cs}$ . Radioecological studies of  $^{210}\text{Po}$  also have a historical record in the Nordic countries. Revisiting the  $^{210}\text{Po}$  field can help further elucidation of  $^{137}\text{Cs}$  and reindeer radioecology as similarities and differences in routes of transfer to reindeer can help identify important factors (e.g., fungi can be an unpredictable radiocaesium source while being less important for  $^{210}\text{Po}$  intake).



**Figure 20: Reindeer running in corral used for gathering animals prior to slaughter (photo: NRPA)**



**Figure 21: Prior to slaughter, live reindeer are measured to determine whether the content of  $^{137}\text{Cs}$  comply with the Norwegian food intervention limit of 3000 Bq/kg for reindeer meat (photo: NRPA)**

### ***5.2 Some aspects of reindeer radioecology***

Reindeer have adapted physiologically to the extreme conditions they live in. Reduced metabolic rates during winter, and a winter diet consisting predominantly of lichen (being low in minerals), both contribute to relatively high radiocaesium levels in reindeer when lichen is contaminated. As spring arrives and fresh, and less contaminated, herbaceous vegetation becomes available, mineral intake and metabolic rates increases. Radiocaesium

levels in reindeer are therefore reduced during summer. Ingestion of contaminated fungi in the autumn may result in elevated contamination levels (Hove et al. 1990), before winter conditions again increases the radiocaesium contamination levels in the animals.

Lichen comprises different proportions of the reindeer's diet in different grazing areas both during winter and summer (Gaare and Staaland 1994), thus contributing to differences in contamination of reindeer in different areas. These differences reflect natural differences in pastures. However, historically, large changes in the number of reindeer, the intensity of grazing and therefore availability of lichen has occurred. The partial overgrazing of lichen carpets taking place in Northern Fennoscandia during the last years has added more variability to the natural situation. Radioecological studies have not historically focussed on the effect of grazing intensity on the long term behaviour of radiocaesium contamination. It is therefore important to assess this effect on the historical time series data, as this affects the relevance of the historical data in today's emergency preparedness, for instance the Kautokeino series (presented below).

Ingested lichen becomes less contaminated with time after fallout due to dilution by new growth and to removal of contamination by grazing. The influence of grazing on radiocaesium levels in the diet is not well described; is, for instance, "all in one time" consumption of small patches more common than a gradual grazing of larger areas over years? In autumn 2002 a reindeer herd in the Snåsa area (mid Norway) grazed in an area kept unused for many years due to high Chernobyl contamination. This resulted in  $^{137}\text{Cs}$  levels reaching  $9000 \text{ Bq kg}^{-1}$  in some animals. Another effect of grazing may be movement of fragments of contaminated lichen to nearby areas with herbaceous vegetation, thereby increasing the contamination level and prolonging the contamination situation in this fodder and reducing differences in reindeer contamination levels in winter and summer (E. Gaare, pers. comm.).

Reduced contamination levels in lichen reduce seasonal differences in contamination levels in reindeer. Ingestion of fungi (in which radiocaesium levels decreases more slowly) can then contribute significantly to contamination levels in reindeer. This has been the case in 2000 and 2002 in the Jotunheimen area (south Norway). There, reindeer could not be slaughtered in September, and slaughter was postponed till December – January; i.e. contamination levels were higher due to ingestion of fungi than lichen.

### ***5.3 Effective ecological half-lives***

#### **5.3.1 Sweden**

In Sweden a study was undertaken by Åhman et al. (2001) to investigate the role of radiocaesium origin on transfer from fallout to reindeer during 1986-2000. Five reindeer herding districts with different levels of Chernobyl fallout were studied. The results are summarized in Table 10. The half-life estimates in districts 3-5 are in agreement with earlier estimates from the same areas (Åhman and Åhman 1994). A shorter half-life of 3.1 years was estimated for these districts for the first 6 years, compared to 4.9 years for the last 6 years up to year 2000 (Åhman et al. 2001).



**Table 10: Summary of effective ecological half-life in different Swedish reindeer herding districts during 1996-2000 (adapted from Åhman et al., 2001).**

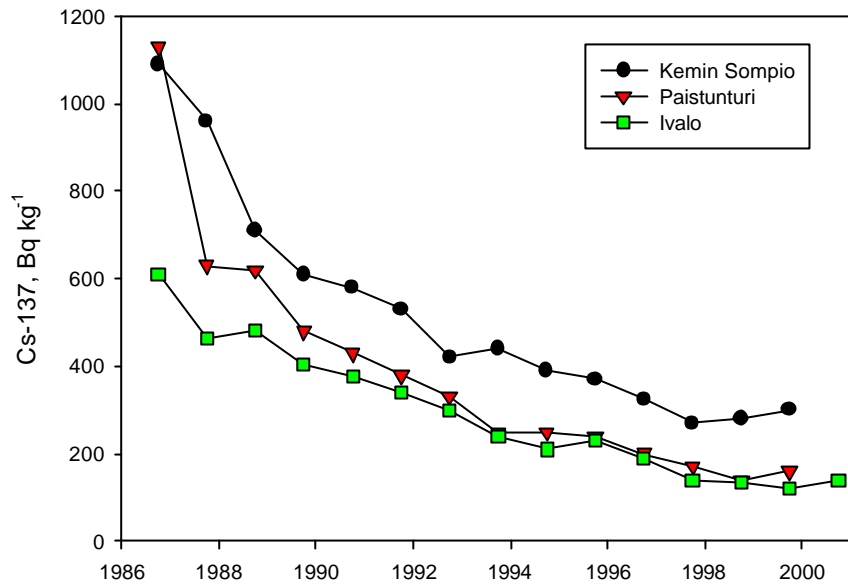
District	Latitude	Chernobyl <sup>137</sup> Cs in relation to total deposition, % (average)	Effective ecological half-life (years)		
			September	Nov-Dec	Jan-Apr
1 (Könkämä and Lainiovuoma)	68°-69°	14	No decline	8.5	9.3
2 (Stäkke and Östra Kikkejaure)	65°-66°	24	5.6	6.3	9.3
3 (Vilhelmina norra)	64°-65°	94	3.1	4.2	3.8
4 (Jiingevaerie)	63°-64°	83	4.6	3.3	3.5
5 (Tännäs and Idre)	61°-62°	51	3.5	3.9	4.2

Åhman et al. (2001) found that their results supported a theory of a gradual, but reversible, fixation of radiocaesium in soil over a mid-long term. By assuming that 44 % of the <sup>137</sup>Cs available to reindeer had a half-life equal to the physical half-life (30.2 years), and the remaining 56 % of the <sup>137</sup>Cs had a half-life of 4 years, Åhman et al. (2001) estimated that the resulting effective ecological half-life in reindeer would be 9.8 years. An effective half-life of 7.6 years was estimated assuming that 30 % of the <sup>137</sup>Cs had a half-life of 30.2 years, and 70 % a half-life of 4 years. Although this shows that radiocaesium origin is important for the effective ecological half-life estimates, the estimated half-lives are longer than those given in Table 10 for comparable fallout composition. Thus, fallout origin cannot alone explain the differences in half-lives. Other reasons were not discussed by Åhman et al. (2001). Depending on how the lichen is grazed (see above), vertical position of the contamination in the lichen may be important. The bioavailability of radiocaesium in lichen appears to be relatively constant (Hove et al. 1999).

### 5.3.2 Finland

A study by Rissanen et al. (2003) draws attention to the possible influence of overgrazing on the decline in radiocaesium contamination in reindeer meat with time. These results were also highlighted by AMAP (2002). Rissanen et al. studied contamination in animals slaughtered during October – April, Figure 22. The faster decline in Paistunturi than the two other areas was attributed to a higher grazing pressure in the first years after the Chernobyl fallout.

Estimated effective ecological half-lives are 4.9 years in Paistunturi, 6.0 years in Ivalo and 6.8 years in Kemin Sompio. Rissanen et al. (2003) give a half-life in Paistunturi during the first year of about one year, and later about 6.5 years. In Kemin Sompio contamination levels decreased during the first three years with a 3-years half-life, and, since 1989, with an eight-years half-life. An attempt was made to fit a dual exponential model to the data as part of this review (see below). For the data from Kemin Sompio, Paistunturi and Ivalo reindeer co-operatives this analysis returned non-significant parameters only.

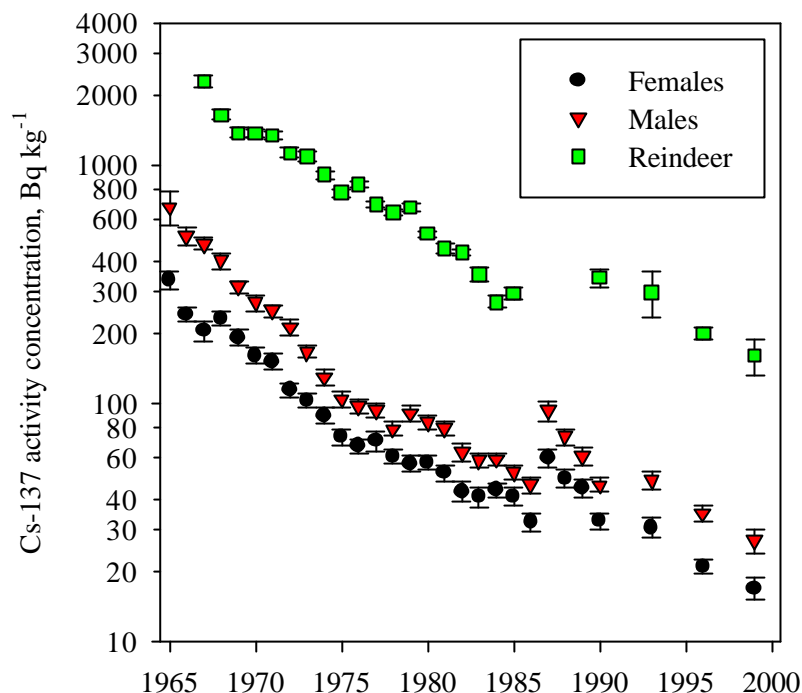


**Figure 22: Average <sup>137</sup>Cs concentrations in winter slaughtered reindeer in Kemin Sompio, Paistunturi and Ivalo reindeer co-operatives during 1986-2000. The Chernobyl fallout was higher in Kemin Sompio and Paistunturi than in Ivalo (adapted from Rissanen *et al.*, 2003).**

### 5.3.3 Norway

Estimates of effective ecological half-lives in reindeer in the Chernobyl affected areas of mid and south Norway have given results similar to those in Sweden, in the order of 3-5 years (Pedersen *et al.* 1993, Amundsen 1995, Hove *et al.* 1997, Gaare *et al.* 2000).

From Northern Norway there is only one time series available, from Kautokeino. The data are presented in Figure 23. Estimated effective ecological half-lives for <sup>137</sup>Cs in reindeer are 6.56 years (standard error interval 6.32-6.82 years) for the pre-Chernobyl period (1967-1985) and 8.1 (7.1-9.5) years post Chernobyl. The results are comparable to those from Sweden mentioned above. Unfortunately no reindeer meat data are available from Kautokeino the first years after the Chernobyl fallout.



**Figure 23:** Cs-137 activity concentrations in reindeer herding Saamis (females and males) and reindeer meat samples from Kautokeino (from Skuterud *et al.*, submitted). Monitoring in 1986 took place about one month before the Chernobyl accident.

The pre-Chernobyl Kautokeino reindeer dataset has been applied in model development by Golikov *et al.* (2002). That analysis suggested that the long term behaviour of radiocaesium in reindeer meat could be described by a dual exponential model with half-lives of 1.1 and 9.0 years (Golikov *et al.* 2002). The time series on whole body monitoring of humans in Kautokeino is more extensive than the reindeer meat data. A dual exponential model for radiocaesium in humans after the Chernobyl fallout gave results comparable to those by Golikov *et al.* (2002), i.e., half-lives of 0.9 and 10 years (Skuterud *et al.*, submitted). In their review Golikov *et al.* (2002) also obtained half-life estimates of about 1-2 and 10-11 years for reindeer time series data from the Kola peninsula and Nenets region.

### 5.3.4 Greenland

Data on  $^{137}\text{Cs}$  in reindeer from Greenland 1962 – 1997 were kindly provided for this review by Sven P. Nielsen (pers. comm.). The data are relatively scattered both geographically and in time, and killings during May – September comprise a relatively large proportion of the dataset.

Estimates based on all available winter data (averaged by year) suggest that the  $^{137}\text{Cs}$  concentrations have declined with effective ecological half-lives of about 10 years (no significant effect was obtained by excluding the years with higher global fallout from the analysis). Analysed by region, a slightly shorter half-life of 7-8 years was estimated for reindeer from the region Holsteinsborg (from which the most comprehensive time series was available).

The estimated half-lives for Greenlandic reindeer appear comparable to those estimated above in northern Sweden and Norway.

#### 4.3.5 Iceland

Data on  $^{137}\text{Cs}$  in reindeer in Iceland have not been included in this review. The animals are wild and a limited number is killed each autumn during the hunting season. A study carried out 1990 – 1992 showed significant differences between herds grazing in different regions; the difference between years was on the other hand insignificant (Pálsson et al. 1994). The most important factor was the type of vegetation selected by the reindeer in the region where they happened to be grazing in the weeks prior to the hunting season. Comparison with older data suggests a slow decline of  $^{137}\text{Cs}$  concentration in reindeer meat. These results are currently being followed-up in an ongoing study.



Figure 24: Reindeer grazing area in Iceland (photo: Skarphéðinn G. Þórisson)



Figure 25: Icelandic reindeer (photo: Skarphéðinn G. Þórisson)

## 5.4 Conclusions

This review has tried to emphasize that there are still important gaps of missing information in the understanding of reindeer radioecology, information that will help improving emergency preparedness relevant for a vulnerable indigenous population group at a regional level:

- The proportion of lichen in the reindeer's diet, during all seasons, together with fallout pattern, are the important factors determining the contamination of reindeer during the first slaughtering season after a fallout situation. In many areas the proportion of lichen in the diet is not well known. Additionally, the relevance of the present radioecological experience may be obscured by the increased grazing intensity that has occurred in Northern Fennoscandia.
- Observed difference in effective ecological half-lives are more than a factor of 2 between Chernobyl affected areas in Sweden and Norway and areas less affected by the Chernobyl fallout further north. The differences appear not to be satisfactorily explained by differences in fallout origin alone.

Although much information is available, further studies of reindeer in different areas (i.e., at a Nordic level) are required to identify and better understand the factors governing regional differences in transfer and long term behaviour of radiocaesium in the lichen – reindeer – human foodchain. These studies should also consider including other radionuclides as helpful ecological tracers, e.g.  $^{210}\text{Po}$  which also is interesting from a radiation protection viewpoint. Modelling would be a helpful tool in this work, and would probably help extracting more information from the already available datasets in the Nordic countries. The model would also fill an important gap in the other software applied in the emergency preparedness.

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## **6. Workshop on Radioecological Modelling in ECOSYS**

Risø National Laboratory, 10-11 September 2003

### ***6.1 Introduction***

The workshop was organised as a collaboration between Baltic-Danish co-operation projects on radiation protection 2001-2003 and the NKS-B project EcoDoses, which deals with improvements of radiological dose assessment for terrestrial ecosystems. The time and place of the workshop was organised back-to-back with an NKS-B mini-seminar on radioecology, which took place the 8-9 September 2003 thus enabling the seminar participants to attend also the workshop.

### ***6.2 Background***

The decision-support system ARGOS developed by the Danish Emergency Management Agency incorporates and integrates data of relevance to large-scale radioactive contamination of the environment. In 2002 ARGOS was extended with a radioecological module for estimating doses to humans from ingestion of contaminated food (FDM, Food and Dose Module). The FDM in ARGOS is intended for predicting consequences of short-term accidental releases of radioactivity. For the purpose of model validation or testing, a special version of FDM has been developed, which allows for multiple deposition events as well as full access to all model parameters. This version of the model may be used to predict levels of radionuclides in the environment from extended deposition events, eg. fallout from atmospheric nuclear weapons testing, as well as from the Chernobyl accident. Model assumptions and parameter values may be adjusted to minimize the difference between predicted and observed values, and these adjustments may then be implemented in the ARGOS FDM. FDM will be available to users of ARGOS in Poland, Estonia, Latvia and Lithuania as well as a licensed copy of the special version of FDM.

### ***6.3 Aim of Workshop***

- For participants to become acquainted with the radioecological food-and-dose module (FDM) in the ARGOS Decision support System used for predicting transfer of deposited radionuclides to foodstuffs and subsequent radiation doses to man,
- to demonstrate how assumptions and parameter values in FDM may be adjusted to improve confidence in predicted data,
- to allow participants to compare their local historical data on environmental radioactivity with corresponding data predicted by FDM.

### ***6.4 Workshop Programme***

The ARGOS food-and-dose module was demonstrated for the participants, as well as the special version of FDM, which allows for adjustment of assumptions and parameters. Following this the participants had the opportunity to become acquainted with the special FDM version and with the model adjustment technique using their own data.

## 6.5 Participants

The workshop included participants from Poland, Estonia, Latvia, Lithuania, Norway, Iceland and Denmark as listed in the table below.

**Table 11: Workshop participants**

Name	Institute	Country
Pawel Krajewski	Central Laboratory for Radiological Protection, Warsaw	Poland
Grazyna Krajewska	Central Laboratory for Radiological Protection, Warsaw	Poland
Raivo Rajamäe	Estonian Radiation Protection Centre, Tallinn	Estonia
Airi Uljas	Estonian Radiation Protection Centre, Tallinn	Estonia
Imants Bruveris	Radiation Safety Centre, Riga	Latvia
Beata Vilimaite-Silobriene	Environmental Protection Agency, Vilnius	Lithuania
Aukse Skripkiene	Radiation Protection Centre, Vilnius	Lithuania
Astrid Liland	Norwegian Radiation Protection Agency, Østerås	Norway
Håvard Thørring	Norwegian Radiation Protection Agency, Østerås	Norway
Sigurður Emil Pálsson	Icelandic Radiation Protection Institute, Reykjavik	Iceland
Steen Hoe	Danish Emergency Management Agency, Birkerød	Denmark
Søren Thykier-Nielsen	Risø National Laboratory, Roskilde	Denmark
Sven P. Nielsen	Risø National Laboratory, Roskilde	Denmark

## 6.6 Results

Following demonstrations of FDM in ARGOS and the special version of FDM, the participants had the opportunity of testing the software. Test scenarios involved use of standard model parameters and assumptions using default values and use of parameter values and assumptions adapted to site-specific conditions. Results from this work is illustrated in Figure 26 and Figure 27 showing predicted concentrations of  $^{137}\text{Cs}$  in foodstuffs from the deposition of  $100 \text{ kBq m}^{-2}$  the 1 May 1986. Figure 28 and Figure 29 compare predicted ingestion doses from different assumptions concerning intake of pork, and Figure 30 and Figure 31 show predicted vs. observed concentrations of  $^{137}\text{Cs}$  in milk and beef using standard model assumptions. A more detailed description from Dr. P. Krajewski is available in a separate document entitled “ECOSYS for Excel (V. 1.4) Test exercise on I-131 Chernobyl data in Poland”. Figure 32 shows a comparison between prediction and measurements for Icelandic milk data, where the multiple deposition feature of the program was used to base the prediction on a deposition time series.



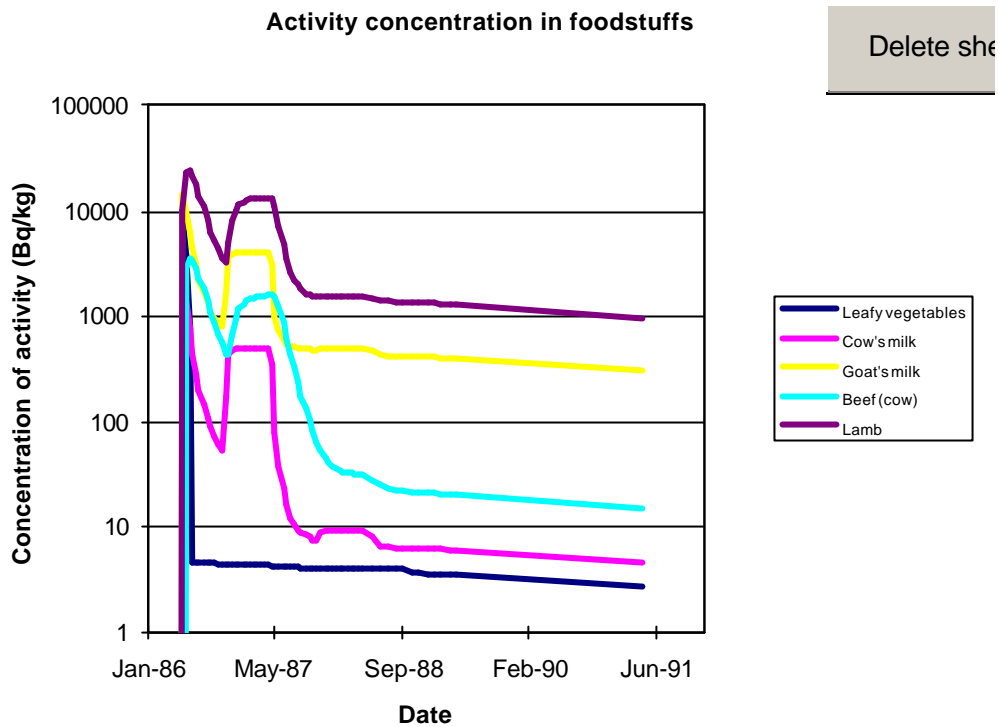


Figure 26: Model predictions from a  $^{137}\text{Cs}$  deposition of  $100 \text{ kBq m}^{-2}$  using standard parameters and assumptions.

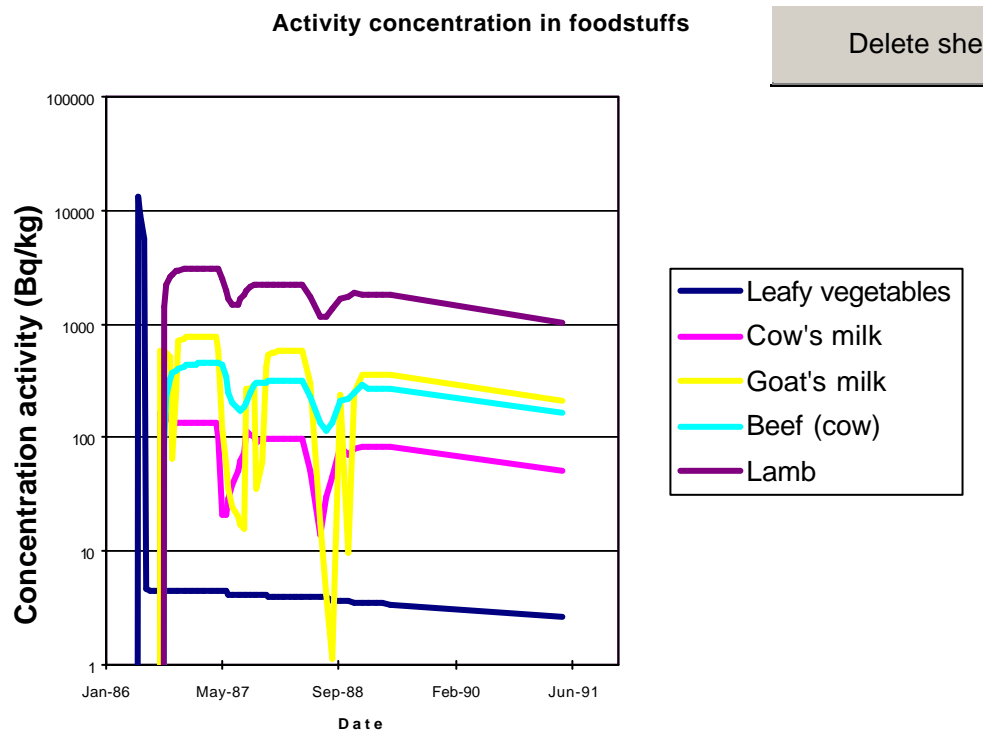


Figure 27: Model predictions from a  $^{137}\text{Cs}$  deposition of  $100 \text{ kBq m}^{-2}$  using non-standard parameters and assumptions.

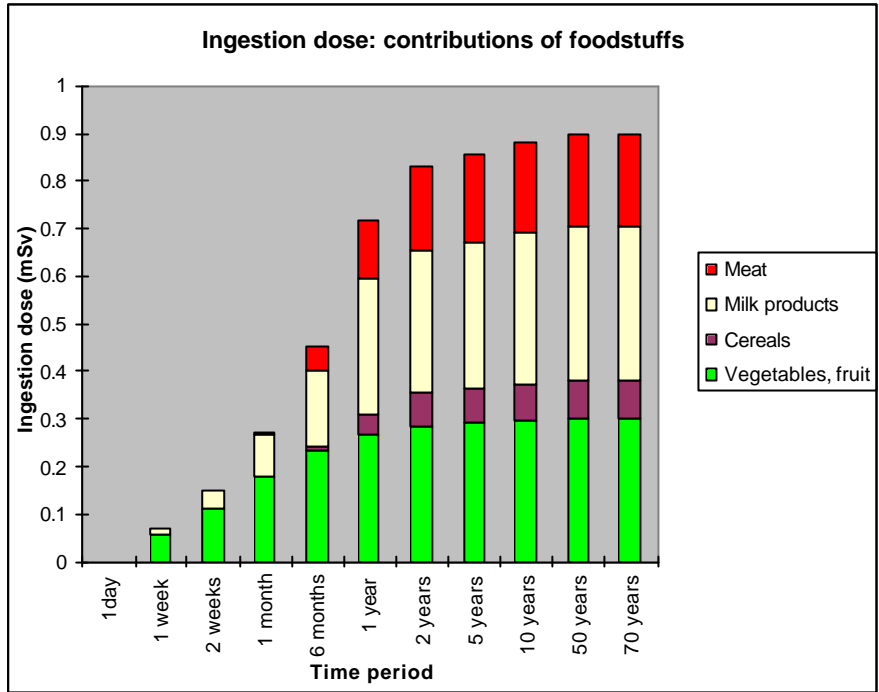


Figure 28: Predicted ingestion doses from deposition of  $40 \text{ kBq m}^{-2}$  of  $^{137}\text{Cs}$  using standard assumptions.

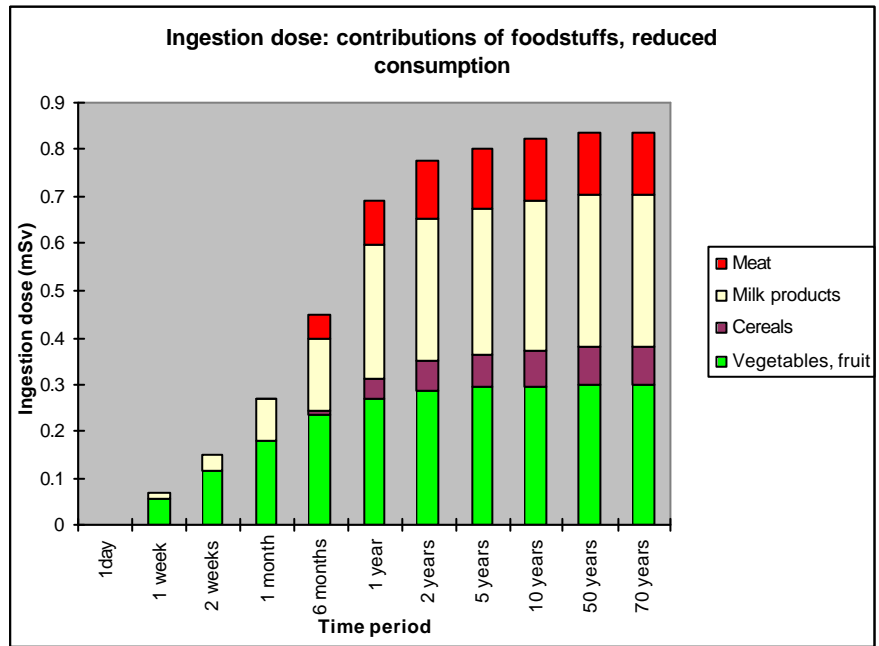


Figure 29: Predicted ingestion doses from deposition of  $40 \text{ kBq m}^{-2}$  of  $^{137}\text{Cs}$  assuming reduced intake of pork.

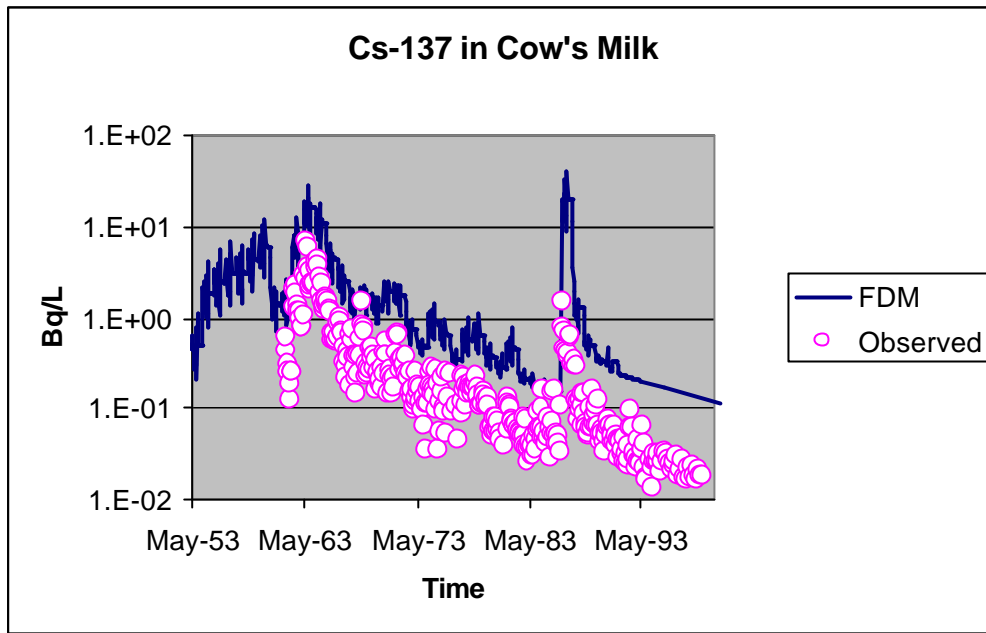


Figure 30: Predicted vs. observed concentrations of <sup>137</sup>Cs in Danish milk using standard model assumptions.

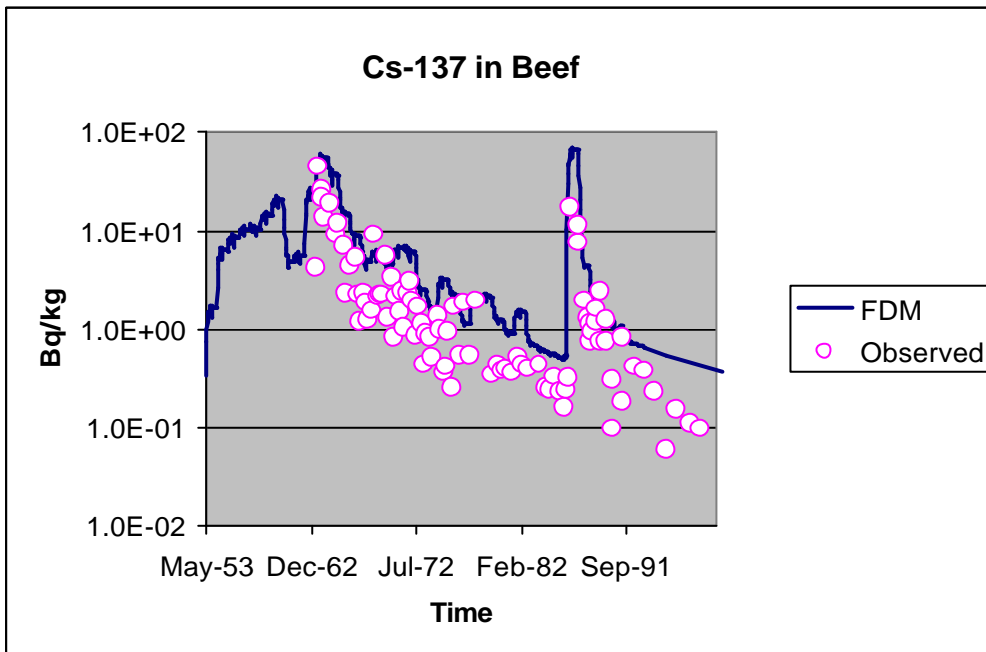
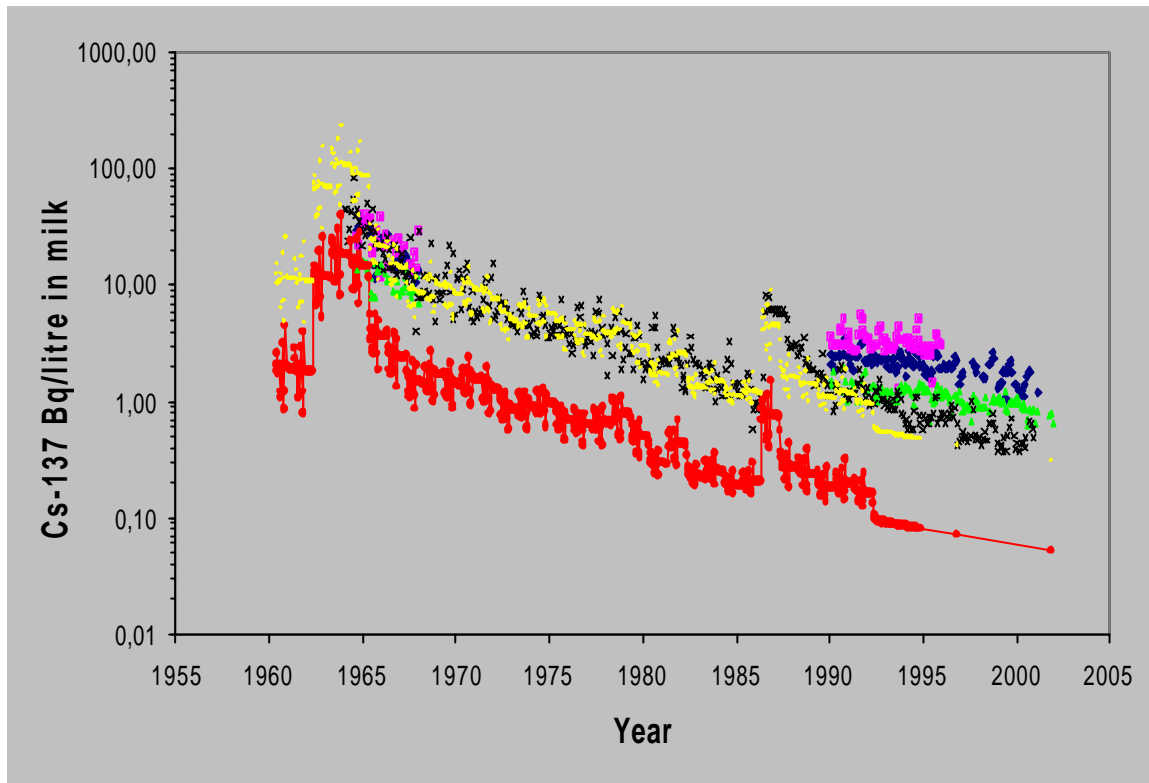


Figure 31: Predicted vs. observed concentrations of <sup>137</sup>Cs in Danish beef using standard model assumptions.



**Figure 32: Predicted vs. observed concentrations of  $^{137}\text{Cs}$  in milk in Iceland and the Faroe Islands using standard model assumptions and deposition data from Iceland. The ECOSYS prediction is given by the red curve and the same values multiplied by 6 are shown by the yellow points. Measured values in milk in Iceland are shown for 3 regions, North (green), South (blue) and West (purple). The black points are milk values from Tórshavn in the Faroe Islands, where the deposition from Chernobyl is known to have been greater than in Iceland.**

### **6.7 Conclusions**

The participants found the radioecological model FDM useful and well suited for predicting the radiological consequences from the presence of anthropogenic radioactivity in the environment. However, the participants felt that they needed more time to become familiar with details of the model and use of the modelling features. In order to assure reliability of the model applications it seemed important to specify proper assumptions of regional or local characteristics (e.g. soil type, growing season, feeding regime of animals, selection and consumption rates of human foodstuffs). Of great importance for the credibility of model predictions are demonstrations of good agreement between predicted and observed data.

## 7. Summary of work for the first project year

The work conducted the first year included a summary of the available information on nuclear weapons testing and a closer look at an empirical at an atmospheric compartmental model (UNSCEAR, 2000) for predicting global. It is evident when comparing the measurements of deposited  $^{137}\text{Cs}$  in Bodø and Tromsø with the UNSCEAR predictions, that the fallout in Norway is significantly underestimated by the global fallout model. It is likely to believe that this is the case also for the other Nordic countries. This is due to the global model not taking into account the relatively rapid deposition of radionuclides in the northern hemisphere following the Soviet tests in 1958 that mainly penetrated the troposphere and not the stratosphere.

A better approach for estimating global fallout on a regional or national scale has been developed based on a correlation between precipitation and deposition rates. Results from Iceland show very good agreement between the precipitation-based estimates and the measured deposition of  $^{137}\text{Cs}$ . The ratio of measured to estimated values is 0.98, surprisingly high considering that dry fallout has not been taken into account. A consistent methodology of soil sampling and compiling precipitation data is crucial for a successful application of this approach. The method was then used to estimate deposition in 6 stations in other Nordic countries and one station in UK. All stations, except one, showed good agreement between the precipitation-based estimates and the measured deposition. They were in the range -1 to +8 % deviation. The method thus appears to be a very good tool for estimating deposition of  $^{137}\text{Cs}$  from the nuclear weapons fallout period, both on a national and a regional scale.

Cs-137,  $^{131}\text{I}$  and  $^{90}\text{Sr}$  are easily transferred from ground deposition to humans via the food chain. Cow's milk is an important foodstuff in the Nordic countries, and extensive monitoring of radionuclide contamination in milk has been conducted since the 1960s. A database has been created covering all available data on concentrations of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{131}\text{I}$  in milk from the Nordic countries. Effective ecological half-lives have been calculated for the two former for the nuclear weapons fallout period. Traditionally, this calculation has been done using single exponential decay regression with results in the range 1-3 years for  $^{137}\text{Cs}$ . In this study we have used a dual exponential decay regression that fitted well with the experimental data. The effective ecological half-lives were similar for all regions with a fast component of about 1 year and a slow component of about 6 years. The long-term consequences of radioactive fallout thus seem to be more severe than earlier anticipated. For  $^{90}\text{Sr}$  the dual component decay regression showed important variations between regions. The short component was about 1 year for all regions, but the slow component varied between 4 and 12 years. The reason for these differences needs to be investigated further. It would also be interesting to perform dual decay regressions for the post-Chernobyl data in 2004 to see if the trend is equal for the two fallout scenarios. We would like, in the next project period, to combine the improved estimates of deposition with the milk contamination data to determine more accurate transfer factors for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  from deposition to cow's milk. The data can also be used to compare modelling results e.g. in ECOSYS with measured data.

Another important facet of Nordic radioecology is the high and persistent contamination of  $^{137}\text{Cs}$  in reindeer. This study has shown that there are still important knowledge gaps in understanding reindeer radioecology, knowledge that is important for adequate protection of vulnerable population groups in the case of nuclear fallout. The proportion of lichen in the reindeer diet and the fallout pattern are the most important factors determining the meat contamination. In many areas the proportion of lichen in the diet is not well known. Adding

to this is the possible influence of increased grazing intensity on the radioecology of reindeer. The observed difference in effective ecological half-lives is more than a factor 2 between Chernobyl and global fallout affected areas. This difference cannot be explained by differences in fallout origin alone. More studies are thus needed to identify and understand the factors governing long-term contamination in reindeer.

Modelling tools can be helpful to predict the consequences of nuclear fallout for the food-chain and for assessing doses to humans. One such model is the ECOSYS food and dose module that can be linked to various decision support systems for nuclear emergency. This model was tested in a Nordic-Baltic workshop arranged in 2003 as a collaboration between Baltic-Danish co-operation projects on radiation protection 2001-2003 and the EcoDoses project. The participants found that the model was useful and suited for predicting radiological consequences of nuclear fallout. The testing performed at the workshop showed the importance of specifying proper local or regional values for input data like growing season, feeding regime and consumption rates to get reliable results on activity concentrations in foodstuffs and doses to humans. Of great importance for the credibility of the modelled results are demonstrations of good agreement between modelled and measured values.

Accurate estimates of radionuclide deposition are important input values in any food chain models. The local and regional variations in e.g. soil types and traditions in agricultural practice must be taken into account when modelling consequences for the food chain and humans. These parameters are also crucial for understanding the differences in radionuclide transfer between regions or countries. Reliable long-time series of radioactive contamination in foodstuffs are important both to validate radioecological models and to calculate the effective ecological half-lives of radionuclides in different food chains. A good understanding of radioecology and good modelling tools are important for implementing adequate countermeasures in a nuclear emergency situation. The continuation of EcoDoses will hopefully contribute to reaching these goals in a successful manner.

Title	EcoDoses. Improving radiological assessment of doses to man from terrestrial ecosystems. A status report for the NKS-B project 2003
Author(s)	Edited by Tone D. Bergan <sup>6</sup> and Astrid Liland <sup>6</sup>  Authors: Sven P. Nielsen <sup>1</sup> , Mats Isaksson <sup>2</sup> , Ritva Saxén <sup>3</sup> , Eila Kostianen <sup>3</sup> , Kristina Rissanen <sup>3</sup> , Hans Pauli Joensen <sup>4</sup> , Sigurður Emil Pálsson <sup>5</sup> , Tone D. Bergan <sup>6</sup> , Lavrans Skuterud <sup>6</sup> , Håvard Thørring <sup>6</sup> & Astrid Liland <sup>6</sup>
Affiliation(s)	<sup>1</sup> Risø National Laboratory, Denmark, <sup>2</sup> Göteborg University, Sweden, <sup>3</sup> STUK, Finland, <sup>4</sup> Fróðskaparsetur Føroya, Faroe Islands, <sup>5</sup> Geislavarnir Ríkisins, Iceland, <sup>6</sup> NRPA, Norway
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Abstract	<p>The NKS B-programme EcoDoses project started in 2003 as a collaboration between all the Nordic countries. The aim of the project is to improve the radiological assessments of doses to man from terrestrial ecosystems. The first part, conducted in 2003, has focussed on an extensive collation and review of both published and unpublished data from all the Nordic countries for the nuclear weapons fallout period and the post-Chernobyl period. This included data on radionuclides in air filters, precipitation, soil samples, milk and reindeer. Based on this, an improved model for estimating radioactive fallout based on precipitation data during the nuclear weapons fallout period has been developed. Effective ecological half- lives for <sup>137</sup>Cs and <sup>90</sup>Sr in milk have been calculated for the nuclear weapons fallout period. For reindeer the ecological half- lives for <sup>137</sup>Cs have been calculated for both the nuclear weapons fallout period and the post-Chernobyl period. The data were also used to compare modelling results with observed concentrations. This was done at a workshop where the radioecological food-and-dose module in the ARGOS decision support system was used to predict transfer of deposited radionuclides to foodstuffs and subsequent radiation doses to man.</p> <p>The work conducted the first year is presented in this report and gives interesting, new results relevant for terrestrial radioecology.</p>
Key words	Nuclear weapons fallout, deposition modelling, food chain modelling, ecological half-lives in reindeer and milk