TECHNICAL REPORT EKO-2.3

Long ecological half-lives of radionuclides in Nordic Limnic systems

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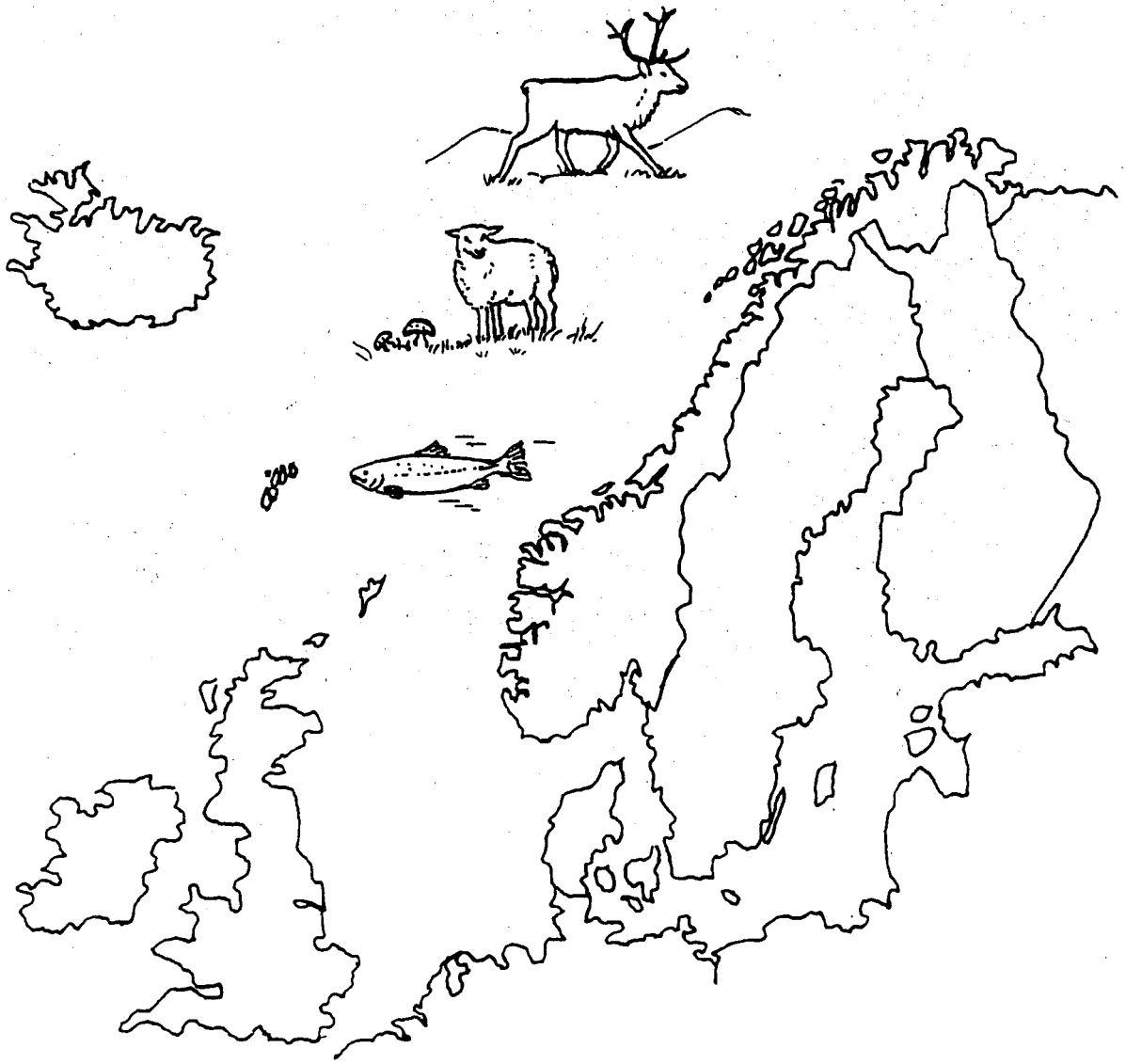

NORDISK KJERNESIKKERHETSFORSKNING

NKS Project ECO-2L

Long ecological half-lives of radionuclides in Nordic limnic systems

Progress Report to NKS (Appendix)

December 1995

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INTRODUCTION

The project aims at quantifying the physico-chemical mobility of radionuclides in soils and sediments, and the biological transfer to fish. The main focus of the project is on **radiocaesium** from the **Chernobyl** accident in Nordic (softwater) lakes. Much of the project is based on a compilation of data and models covering comparatively large scales of time (10 years) and space (10^6 km^2).

The recovery of Nordic lake ecosystems from contamination by Cs-137 originating from the Chernobyl accident is gradually slowing down. At at the same time as lakes vary widely in susceptibility and recovery rates. Accordingly, **ecological "half-lives"** are gradually increasing and cannot be treated as constants, over neither time nor space.

The main purpose of this project is to quantify such "half-lives" in different types of **landscapes** of the Northern countries, and to assess the relative importance of various control **mechanisms**. Such knowledge is necessary, since empirical knowledge may not be sufficient for an efficient radiation protection. For example, although it has long been known that the highest concentrations of Cs-137 in fish usually are found in softwater systems, liming of lakes after the Chernobyl accident did not result in any discernible reduction.

In lake ecosystems, the initial dynamic phase of contamination and equilibration after fallout of Cs-137 lasts up to five years and appears to be largely ruled by biological processes. After that, Cs-137 activities in fish approach a "steady" state, with a slow decline that is probably controlled by continuous **secondary inputs** of Cs-137 into the lakes and their food webs. The most likely sources are the loss of Cs-137 **from land to water** (external) and the recycling of Cs-137 **from sediments** (internal). While fluxes and abiotic concentrations of Cs-137 decline, Cs-137 present in the system also appears to become less and less **bioavailable**.

Sr-90 is less frequently studied, but more mobile than Cs-137, and will be included in our project. Due to the scarcity of data, comprehensive models can only be validated to a limited extent by stepwise comparisons with Cs-137.

In the following, all **subprojects** are presented with their background, their main achievements 1994-95, and their plans for 1996-97:

LONG TIME SERIES OF CS-137 IN FISH
A. MODELS AND MAPS OF LAKE SUSCEPTIBILITY TO FALLOUT
B. LOSS OF CS-137 FROM LAND TO SURFACE WATERS
C. SEDIMENTS AS A SECONDARY SOURCE OF CS-137 TO FISH
D. BIOAVAILABILITY OF PARTICULATE CS-137 TO INVERTEBRATES
OTHER PLANS
RELEVANT REPORTS BY THE PROJECT PARTICIPANTS
DATA SOURCES AND OTHER RELEVANT REFERENCES

LONG TIME SERIES OF CS-137 IN FISH

In order to identify and quantify processes controlling the temporal development of Cs-137 concentrations in fish, and to calibrate prognostic models, it is **crucial** to have access to long time series of observational data from differing systems. In this project, we are highly dependent on such time series. However, such data are increasingly **rare**, as many authorities have drastically reduced or abandoned their Chernobyl monitoring programs since 1990. **We therefore urge authorities to continue monitoring of Cs-137 in fish** in selected lakes that differ widely in biogeochemical characteristics, and in lakes where long time series already exist.

From the few data available, it is evident that a **change** in the processes controlling the contamination of fish has occurred, typically around 1992. One of the possible reasons (see also below) may be the depletion of Cs-137 in wetlands, which have constituted a major source of Cs-137 to lake ecosystems during several years. Another possible reason is the gradual burial of Cs-137 in lake sediments.

In order to calibrate our models being developed, we have started to compile available data (preferably long time series) on Cs-137 in fish, although this was not included in our initial plans. A particular problem is the striking scarcity of available data on the contamination of fish in **Norway**, where freshwater systems are comparatively poor in electrolytes, nutrients and organic matter. Accordingly, these lakes differ from the forest lakes dominating in **Sweden** and **Finland** where wetlands and humic waters are common. We will also try to obtain data from **Denmark**, where freshwaters usually are characterized by a strong influence of agriculture and a prevailing sedimentary bedrock (high nutrient and particle concentration).

A. MODELS AND MAPS OF LAKE SUSCEPTIBILITY TO FALLOUT

Relatively soon after the Chernobyl accident, it was possible to present a map that pointed out areas with a high frequency of lakes that were specially sensitive to high levels of radiocesium in fish due to Chernobyl fallout (Håkansson et al., 1988). That early lake sensitivity map was based on the empirical negative relationship between ionic strength and Cs-137 activity in fish and the geographical pattern was derived from the average ionic strength of lake waters. With increased knowledge about the causal processes behind that empirical relationship, it is now possible to separate different components and what geographical variation they may have. One model that have been developed within the frames of IAEA is the VAMP-model, a model that predicts the Cs-concentration in lake water and predatory fish using nine components meant to increase the predictive capacity of the model. Some of these components are also realistic to use as a base for Nordic maps describing the transfer of fallout to fish while others are not. The two components that we have put special emphasis on in this Nordic project are the transport of Cs-137 from the catchments to the lakes and the internal loading from sediments, which both are crucial for the long-term temporal developmentof Cs-137 in water and fish. At this stage we have not focussed on any temperature or food web induced geographical variations in the transfer, but such components could be included in a latter stage of this programme.

According to the plan, we have conducted an overview of accessible data on fallout and limnology within the Nordic countries. In Sweden, where a comprehensive database on Cs-137 concentration in fish and water already existed, the first maps and related models that describe the geographical variation in the transfer of radiocesium from fallout to water and fish have been produced. Besides data on fallout and limnological characteristics these maps are based on data on soil type and vegetation which have been sampled and compiled within the framework of the Swedish National Forest Inventory. The same methodology will then be applied on data from the other Nordic countries.

A problem for the future seems to be that the monitoring of Cs-137 in lake water and fish has been stopped in many fresh water ecosystems during the last few years, and it is not within the economic frames of ECO-2 to continue monitoring programmes. This is a problem from a radioecological aspect and will give a lack of data for validation and testing of models. However, there are freshwater systems in Finland, Sweden and Norway that have continuous data series on Cs-137 in water and fish and some of these results are shown in this report.

Transfer from soil to water

A database including a broad set of parameters describing the geographical variation in soil and vegetation characteristics and lake water chemistry has been compiled in Sweden. A fundamental problem in geographical analysis of such large data sets, is the variable distribution for different parameters, basic water chemistry is available for 6000 lakes and many soil and vegetation parameters are available for an even denser grid, major chemical constitution of mineral soils is analysed in almost 2000 stations, while data on radiocesium in water and fish is much less frequently analysed and also unevenly distributed.

However, Figure 1illustrates that several, partly overlapping geographical patterns could be identified for different parameters of interest, e.g. in large areas of the eastern parts of S. Sweden there is a high frequency of soils dominated by clay and silt, a low frequency of peaty soils, low C:N ratios in the humus of forest soils and also a high concentration of base cations in lake water. The opposite picture for these parameters is observed in the NE parts of S. Sweden and the central parts of N. Sweden.

According to the results from our soil and runoff studies, one possible method to calculate how the transfer of radiocesium from soil to runoff water is geographically distributed would

be to use the relative frequency of vegetation such as crowberry, wortleberry and heather on peaty soils. These species indicate drier conditions typical of bogs and bog components of mire complexes, and thus, a low frequency on mires would indicate fens or fen dominated mire complexes. Another approach even more in line with our field data was to let the frequency of areas with high transfer being based on the relative frequency of mires and the wetness classification in field in 4 classes depending on distance to ground water surface. In this way the frequency of mires with wet conditions (WET=ground water level at or close to soil surface) and other mires with drier conditions (DRY) was calculated for each geographic unit (here municipalitites). A simpified model based on the transfer from 3 different soil compartments was applied (Eq. 1), see next chapter for details on basic theory.

Based on field data, the Cs-137 inventory on mires in june 1986 was already reduced to about 56% of the initial fallout deposition ($I_{8606mire} = 0.56\ I_{(0)}$, $I_{(0)} = 1$). For whole catchments, this initial loss can be described as a function of the original deposition and the fraction of "wet" and "dry" wetlands:

$$Loss_{init} = (WET+DRY)\ (I_{(0)}-I_{8606mire}) \qquad (1a)$$

The relative loss over time during the following years was calculated as

$$Loss_{(t)} = 0.3\ WET\ I_{8606mire}\ e^{(-(0.3+0.02)t)} + 0{,}02\ DRY\ I_{8606mire}\ e^{(-(0.02+0.02)t)} + 0.0003\ (1\text{-}DRY\text{-}WET)\ I_{(0)}\ e^{(-(0.0003+0.02)t)} \qquad (1b)$$

based on an annual loss from "wet" and "dry" wetlands and from upland soils of about 25%, 2% and <0.03%, respectively, and a physical deacay of 2%. The annual loss of Cs-137 was calculated for each geographic unit according to Equation (1), using a yearly resolution. Figure 2 shows maps of Sweden, giving the resulting relative loss of initial fallout at some different points in time. In this approach we have chosen a constant value for the loss from mineral soils, 0.3 per mille $year^{-1}$, this value is probably not a constant but rather a variable depending on e.g. the clay content of mineral soils and the hydraulic conductivity. However, it seems unlikely that the variation in this low loss would affect the general loss pattern during the first 10 years after the fallout that could be seen in Figure 2. The initial loss during spring 1986 was very high, > 1% in large parts of Sweden and >6% in large parts of northern Sweden. Also during the first year, the loss was above 0.5 % in many areas that largely is in accordance to the areas with the highest initial loss. The map picture is than gradually changing due to that the most easily leachable stores have been emptied, but still after 10 years the highest areal losses could be found in areas with a high frequency of mires. With this geographical resolution (might be locally higher) the yearly loss, at the time being, never comprises more than a few per mille of the initial fallout.

As the deposition is fairly well known, it is also possible to estimate the absolute loss in each geographic unit, Figure 3 shows the integrated loss during the period after spring 1986 until 1993. Most areas have had a loss below 150 Bq m-2 during these years, but the loss was above 1000 Bq m-2 in areas that combine a fairly high deposition and a high loss capacity.

In order to verify the generality of our results, we performed a test of our model on data from 22 other European catchments where information on deposition, runoff and basic catchment characteristics were available and Cs-137 in water have been measured for at least 5 years. All these streams and rivers show a decreasing activity with time but they differ in amplitude and dynamics. It was a good agreement between the observed and modelled, time integrated loss for the period 1987-1993 in small streams and brooks, while the observed loss from 11 larger Finnish catchments (rivers) was significantly lower than model predictions. This test clearly verified the importance of mires for the loss but also supportsed the value used for the loss from other soils. The overestimation of the loss through rivers could be due to a retention within lakes and river pools along the river course.

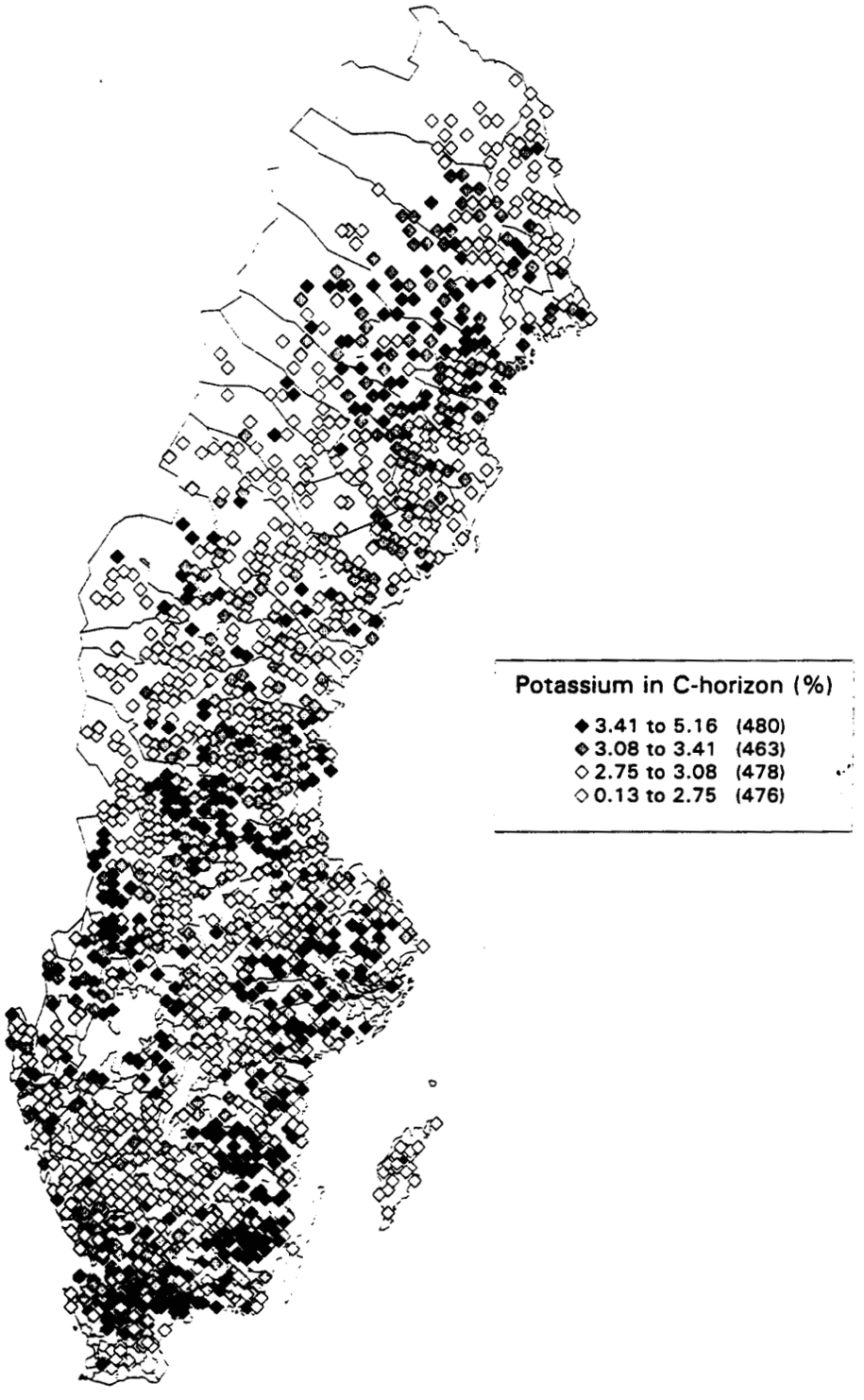
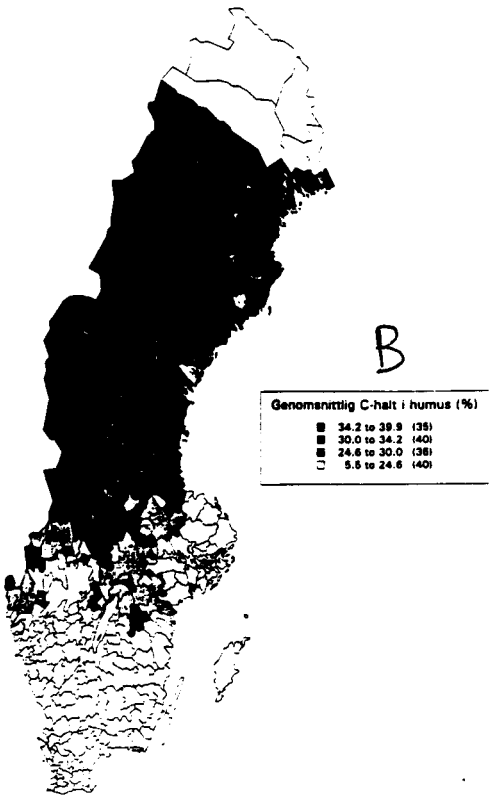
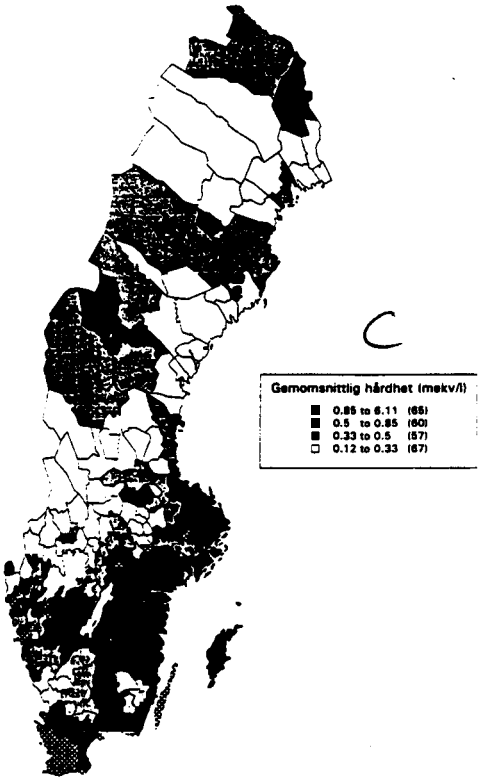
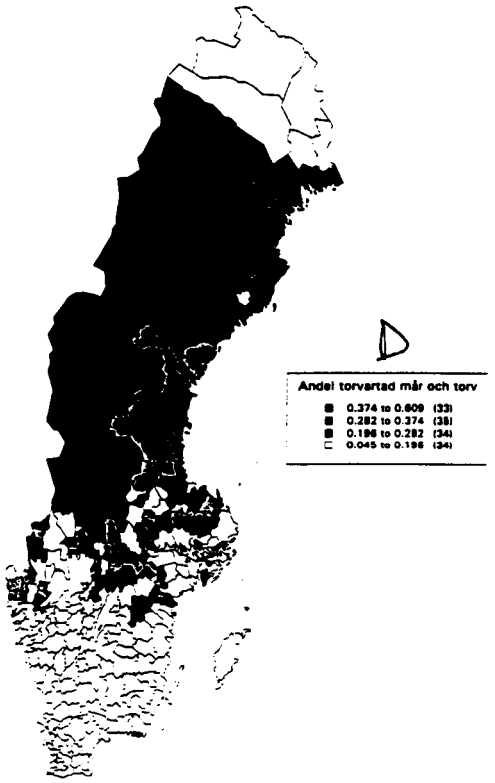


Figure A1. Maps showing the geographical variation within Sweden of A/ potassium content in mineral soils, B/ carbon in humus layer of forest soils, C/ average concentration of Ca+Mg in lake water, D/ relative frequency of peat and peaty humus layers, E/ relative frequency of soils dominated by clay or silt.







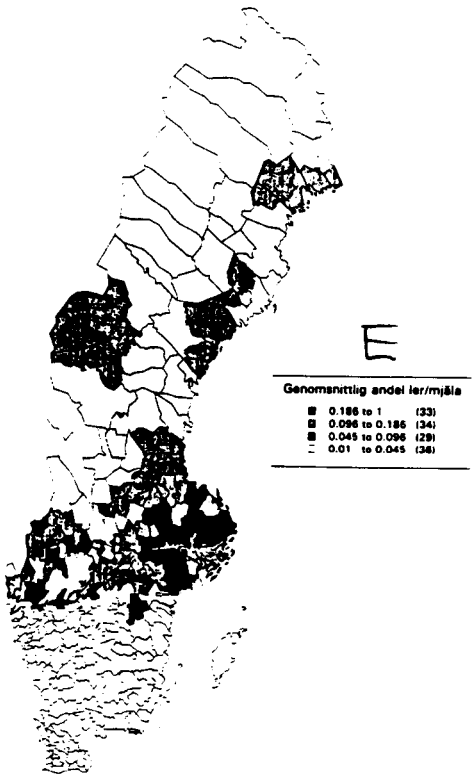


Figure A1 (cont.)

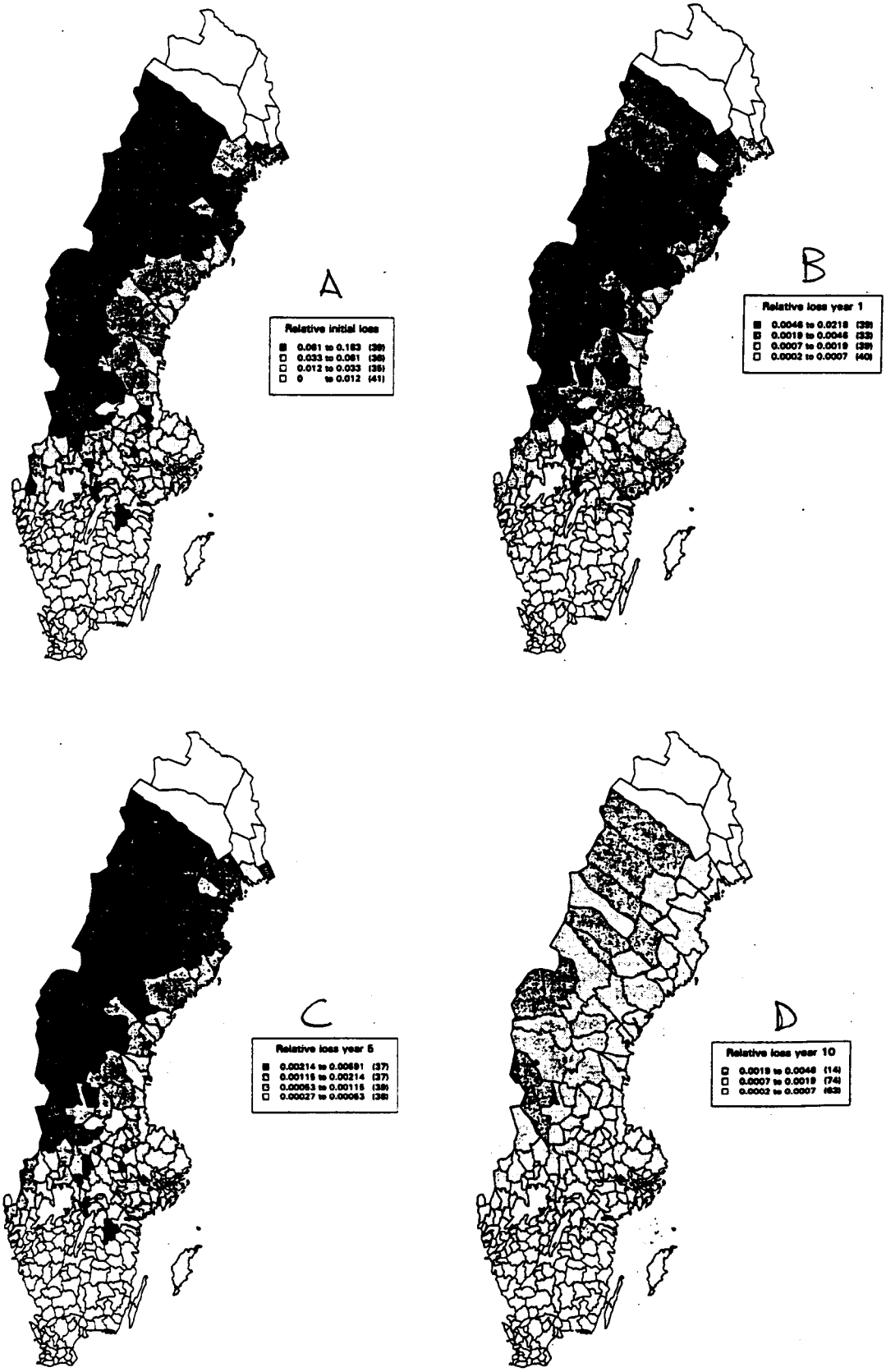


Figure A2 Geographical variation in theloss of Cs-137 in relation to fallout during first month (A), year 1 (B), year 5 (C) and year 10 (D) after Chernobyl fallout.

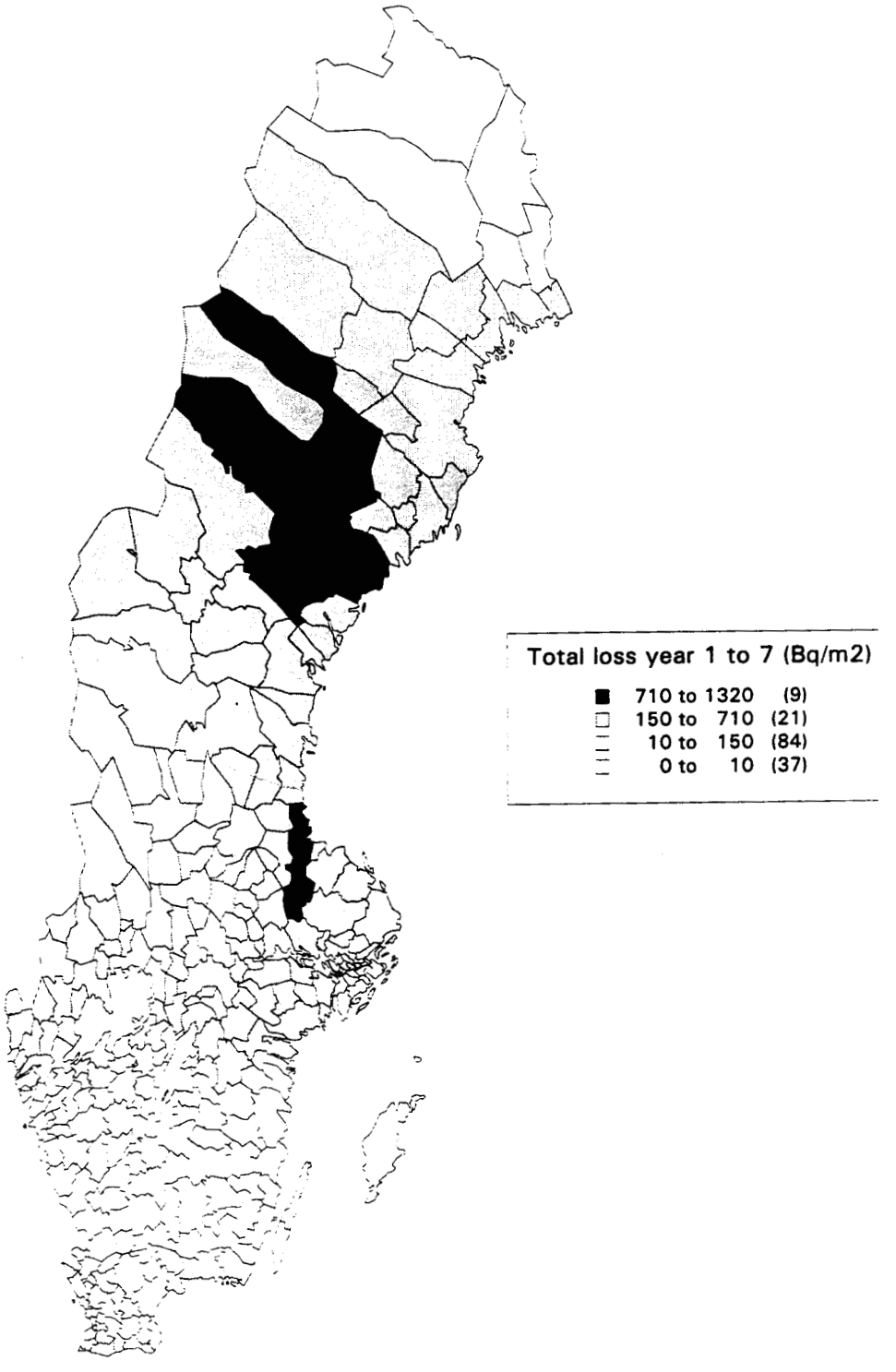


Figure A3. Integrated loss of Cs-137 during the period 1986 to 1993 (first month excluded).

Transfer to fish

The most comprehensive measurements of several fish species within the same system have been conducted in Lake Päijänne and Figure 4 shows the temporal variation for different fish species. It is interesting to note that the activity of Cs-137 not only in non-pescivores such as small perch, roach and whitefish but also in pescivores seems to have reached an "equilibrium level" during 1993 to 1995 from which the mean activity even could increase during certain years depending on high runoff. This is a temporal pattern which has been observed also in stream water and lake water and which also could be predicted theoretically from a combined loss and lake process model. A similar temporal development is observed also in trout and Arctic charr from Lake Storsjouten.

According to Rowan & Rasmussens (1994) interpretation of non-Chernobyl data and the range of potassium concentrations in Nordic lake waters, the bioaccumulation factor (BAF=Cs-137 in fish (fresh weight):Cs-137 in water) to non-pescivore fish would be in the range 1000 to 7000 in most Nordic lakes, while BAF should increase by a factor of 2 for each trophic level. Empirical and predicted concentration of Cs-137 in lake waters and small perch with a constant BAF (=4100) using catchment transfer according to Eq. 1 and a lake model based on water retention and sedimentation is shown in Figure 5. Thus, according to empirical data from Swedish softwater lakes, the bioaccumulation factor to the mainly zooplankton feeding small perch seems to have stabilized around 4000 since 1988/89, while the accumulation to pike was about 4 times higher in 1992. Until 1995 the activity in pike has probably decreased to a level closer to the activity in small perch but the available data are too scattered to draw any definite conclusions.

The bioaccumulation factor to lake trout and arctic charr was usually ranging between 10000 and 20000 without any definite temporal trend during the first 6 years after Chernobyl but was occasionally higher or lower depending on food choice and temperature climate. Arctic charr has generally lower BAF than the lake trout, but in lakes where these fish species are feeding on zooplankton they have BAF<10000, i.e close to the values for small perch, and occasionally, depending on food choice the values for trout could be below arctic charr.

In a statistical approach similar to the one used to obtain the early map showing lake sensitivity to Chernobyl fallout, the transfer coefficient from fallout to fish at a certain time was calculated. However, not only water chemistry and lake morphometry were tested, but also soil and vegetation parameters. The statistical and graphical analysis were based on mean values from a grid (50*50 km) instead of lake mean values. Using this approach, the content of potassium in the C-horizon was the parameter that gave the highest correlation coefficient (r=0.43) with TC_{fish87}. Significantly negative correlations were also obtained with the concentration of Ca+Mg in water and total ionic strength. The negative relationship between TC and potassium content of soils is well in accordance to the suggested explanation of the causal relations behind the statistical correlation of TC to the concentrations of base cations in lake water. Namely, that the concentration of base cations partly is related to the content of clay minerals, resulting in a larger relative sedimentation of radiocesium in the lakes and a higher retention in surrounding soils. The intercorrelation between K in soils and other soil paramters affecting the transfer from soils (e.g peat frequency) could also contribute to the observed statistical relationship. A positive correlation between the content of calcium in soils and TC was observed, which supports the earlier suggested hypothesis that the relation between ionic strength in lake water and TC would not be valid in areas dominated by calcite or dolomite.

Figure 6 gives an example of a predicted map of the transfer of fallout to fish using a single soil parameter (K) for prediction of the transfer (TC). In this case the time integrated transfer to pike according to Equation 2. We have not considered any geographical bias in the mean hydraulic residence time of lakes which certainly would change the picture, as it is the single most important parameter when explaining the transfer of fallout to fish in specific lakes.

$$F_{pike} = F_{pike(tmax)} + TC_{(tmax)}\ Teco/ln(2) \qquad (2)$$

where $F_{pike(tmax)}$ is the integated transfer until the time when the maximum transfer coefficient $TC_{(tmax)}$was reached. Based on the average temporal development of the Cs-137 content in pike as related to perch (Andersson et al., 1990 and Andersson&Meili, 1994), $F_{pike(tmax)}$ was formulated as $F_{pike(tmax)} = 2.1 * TC_{perch87}$ and $T_{eco} = 4$ år.

A more detailed knowledge on the geographical variation in hydraulic residence times of lakes would be of great interest for the future as a loss model must be combined to a lake model where hydraulic residence time is a key parameter. Such a mixed runoff-lake model would make it possible to predict geographical and temporal variation of lake water concentration of Cs-137 in much more detail and with higher degree of precision than existing models and maps. At this stage it is possible to present map predictions like the map in Figure 6 which instead of a statistical relationship could be based on a combined loss and lake process model. This can be reached by using geographically constant residence times (e.g. 1 yr) or scenario maps, letting each map represents lakes with a certain residence time. However, each such map could give a very divergent geographical pattern from the true pattern if there exists large differences in hydraulic residence times between different regions.

Concluding remarks and future activities

An important and interesting question for the future is , how accurate the observed geographical variation in lake sensitivity related to Chernobyl fallout would be in another, hypothetical fallout situation. It is not possible to definitely answer this question, but it is possible to relate the geographical variation to fundamental physical, chemical and biological factors that regulates the retention in soil, turnover and retention in lakes and transfer from water to fish. Thus, it also seems likely that the major geographical variation in the transfer of radionuclides to water and fish due to Chernobyl fallout will be of general validity, even if the amplitude of the transfer will depend on the timing and durability of the radioactive deposition.

It seems also clear that a large portion of the Nordic fresh water systems in an international perspective is sensitive to fallout of radioactive Cs due to a combination of relatively low retention in soils, low sedimentation rates and low concentrations of potassium and other base cations in water. Accordingly, it seems of great interest to get a very good description of the long-term development of radioactivity levels in water, sediment and aquatic biota in Nordic limnic systems with different characteristics in order to improve and validate existing models of the transfer of radionuclides. The planned activities within the other limnic subprogrammes will contribute to an increased understanding of the mechanisms behind the long-term development of radioactivity levels in water and fish. These results will be incorporated in a Nordic map based on models that describe the transfer of radioactive cesium from soils and sediments to water and fish in terms that could be determined from available geographical data on soil and lake water characteristics.

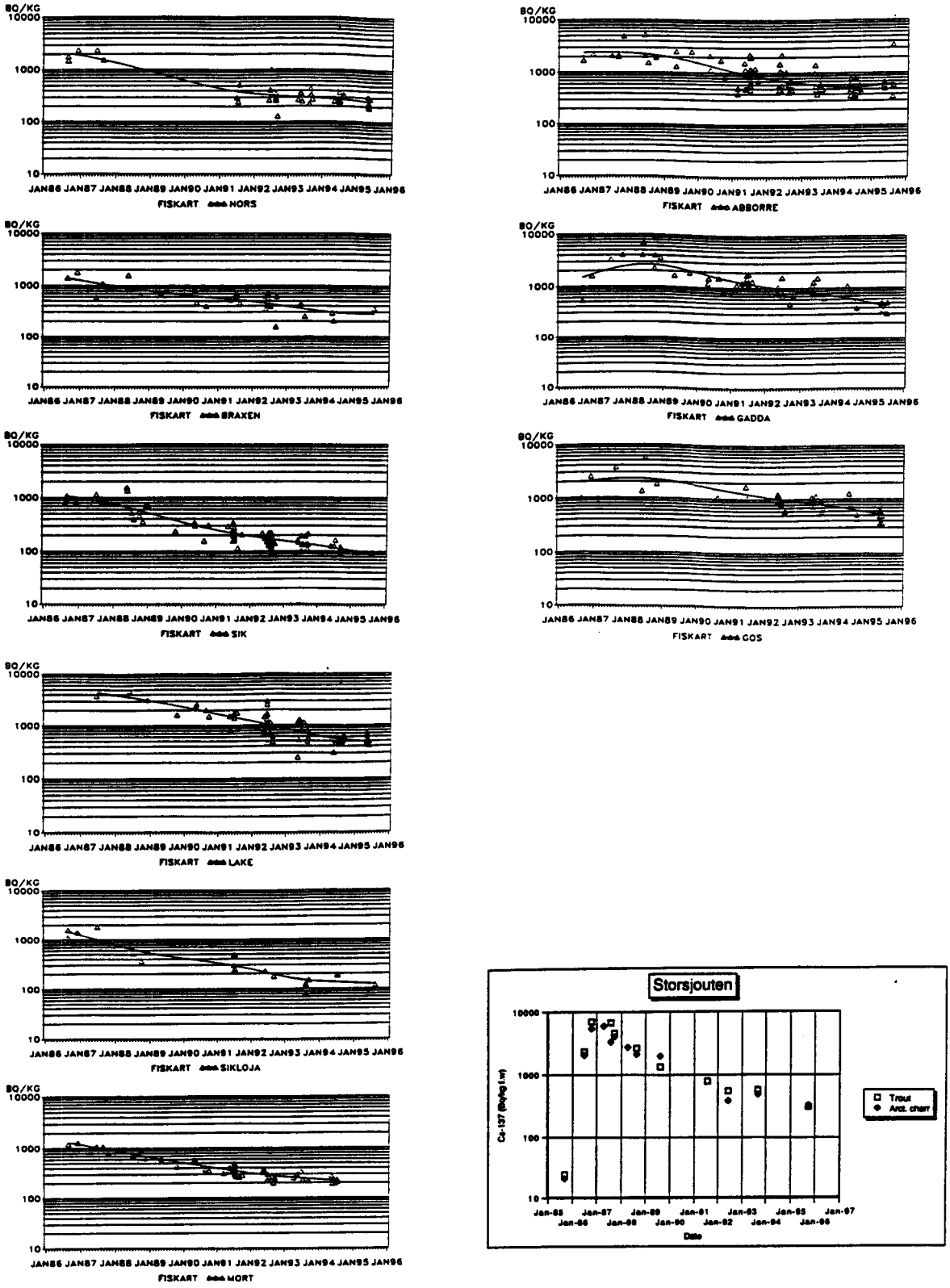


Figure A4. Temporal development of the concentration of Cs-137 in different fish species in Lake Päijänne and Lake Storsjouten until 1995.

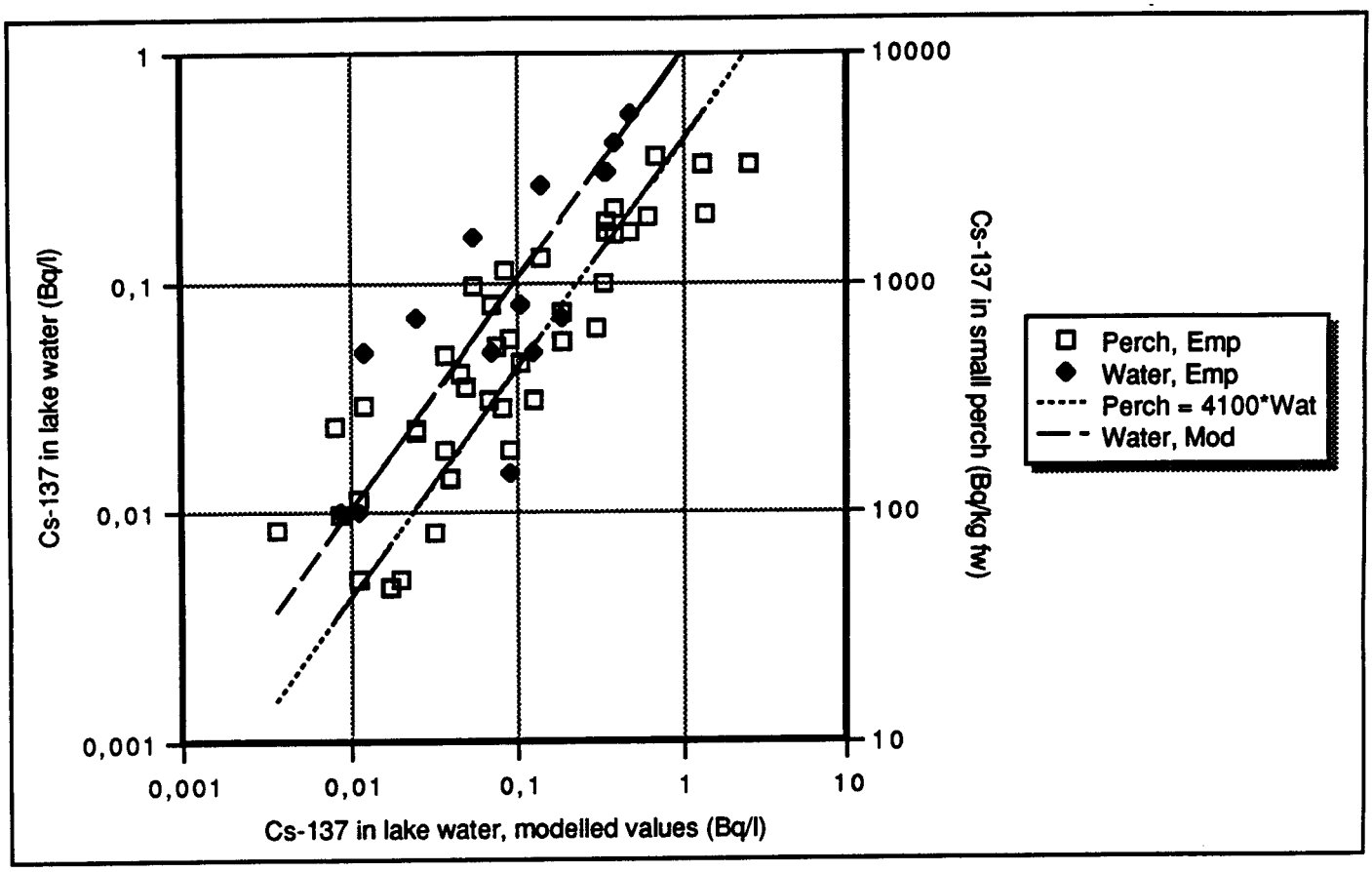


Figure A5. Observed and predicted concentrations of Cs-137 in lake water and small perch during 1989 using the catchment loss model combined to a lake model with a constant bioaccumulation factor (4100).

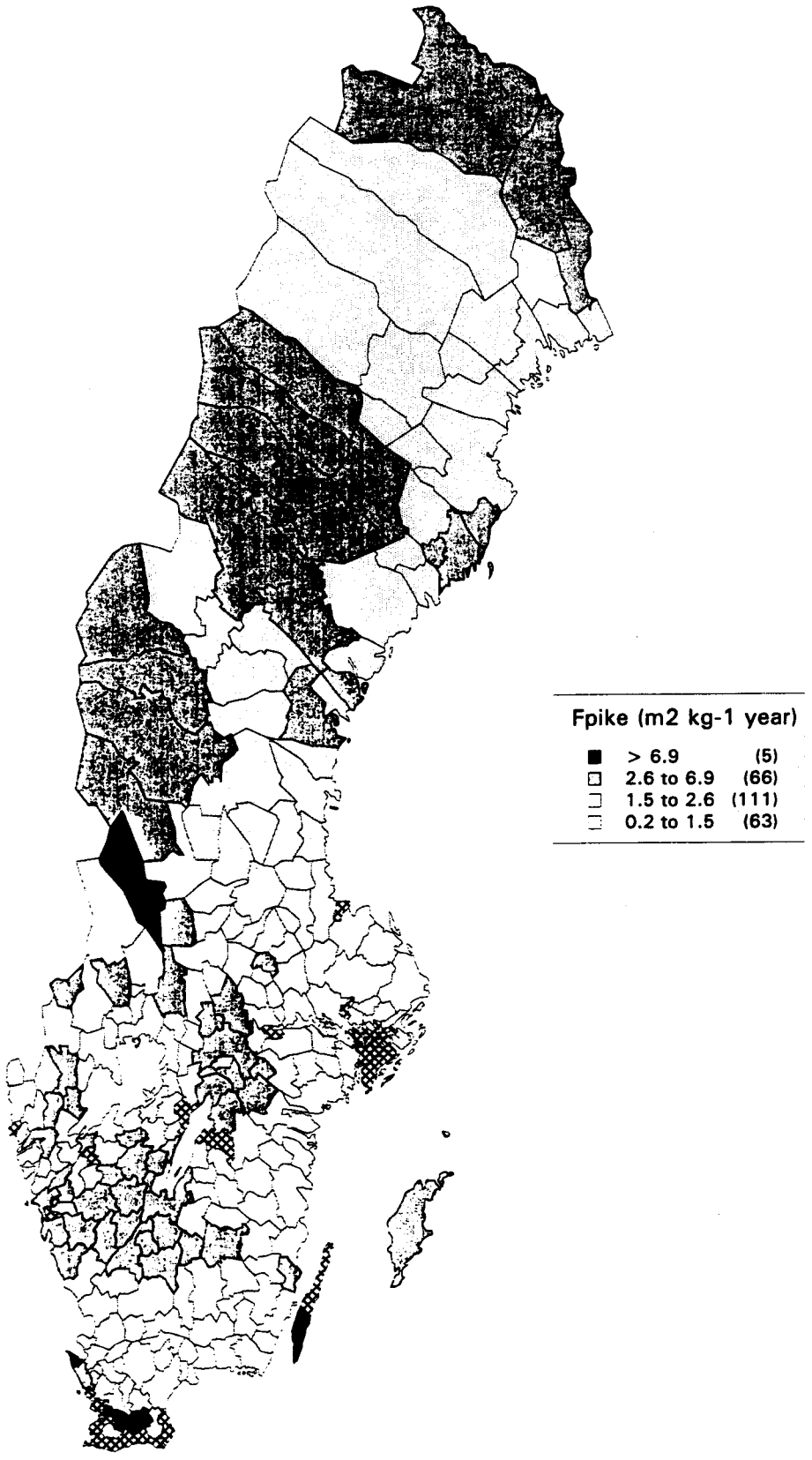


Figure A6. Geographical variation in the total transfer to pike as predicted from potassium content in mineral soils.

B. LOSS OF CS-137 FROM LAND TO SURFACE WATERS

The turnover of Cs-137 and Sr-90 in catchments is studied experimentally and theoretically. The experimental phase is carried out in Nordic catchments of of different cmpositions (eg. soiltypes, vegetation and topogtraphy) as described below (figure 1). In addition to the experimental phase a rewiew of the information on radionuclide discharge from Nordic catchments is in preparation and has been submitted as abstract to the International Symposium on Ionizing Radiation: Protection of the Natural Environment which is to be held in Stockholm in May 1996. An extended summary of this rewiew that so far mainly concerns information about Cs-137 is given below. The project is mainly carried out in national projects, financed by national institutes (e.g. FOA, LFI , SSI, Studsvik ECO-safe, STUK, UmU). Thanks to a significant contribution from NKS during 1994 to 1995 it has been possible to link the national projects together into a forum for common discussions and experimental design.

I. Experimental phase

The aim of the experimental study is to investigate the significance of Cs-137 and Sr-90 discharge on the developement of activity concentration in lakes in typical Nordic environments. Six different nordic catchments were choosen (figure 1):

-two within the hemiboreal ecosystem discharging into the lake Hillesjön in cenreal Sweden,
-one within the alpine ecosystem discharging into lake Heimdalen in central Norway,
-three in the the boreal forest ecosystem. The first two in the southern part discharging into lake Päijännä in southern Finland and the third in the northern part, located in the Svartberget experimental forest station in Vindeln, northern Sweden.

The catchments have partly been caracterized regarding soil types, vegetation, topography and runoff (Table 1, and figure 1).

This field inventory will be completed during the next summer (1996). Water is being sampled from at least one stream and location in each area. The positions has been choosen to give information about the lossrate from areas vith different characteristics (figure 2). Samples will be taken during winter, spring, summer and autumn as well as at low and high runoff intencities, if possible. In addition to the water samples a 1 mm filter is used to collect coarse particles (CPOM). Runoff intencity is monitored in the streams or taken from nearby stations. The water is filtered at 1 mm and 0.45 microns to give some information about carriers and analyzed for Cs-137 and Sr-90. In order to characterize the stream waters a few chemical parameters is determined (conductivity, K, Rb, Ca, Sr, TOC, pH, absorbance at 420 and 660 nm). In most of the catchments, these samples complete long time series that has been carried out since 1986 or earlier and will thus give a data set which will be used to calculate and modell the dynamics and origin of Cs-137 and (to some extent) Sr-90 discharge fromNnordic catchments. So far water has been sampled in all catchments at least once and in some cases, at several times since the start during the autumn in 1995. The results of Cs-137 have been included in the summary below. The results of Sr-90 is so far to few to present but earlier and resent measurements in Northern and middle Sweden and Finland and Heimdalen is available

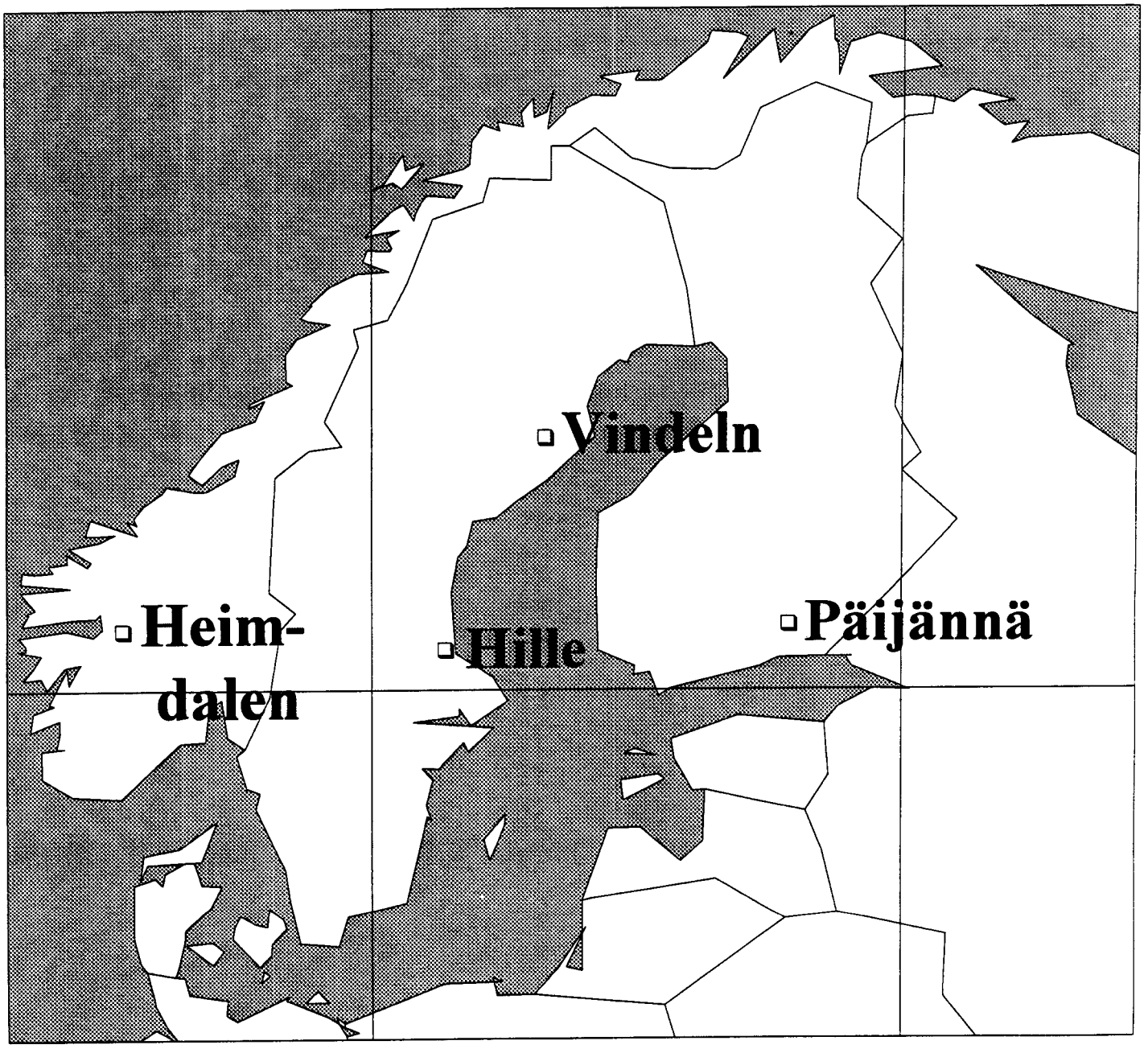


Figure B1. Location of the studied catchments.

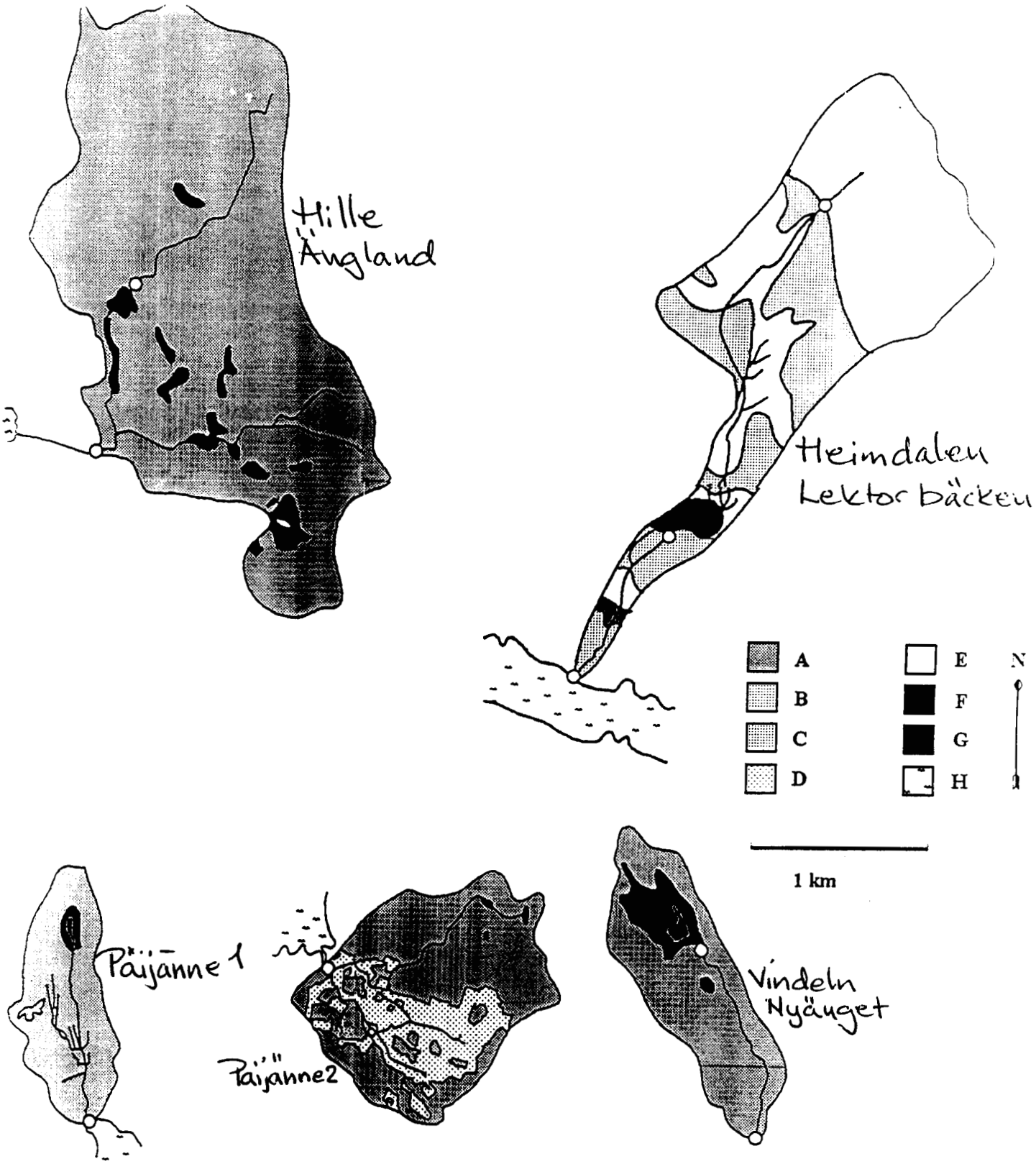


Figure B2. The studied catchments (except "Hille syd"). These drawings are preliminary and will be completed after the field inventory during the summer of 1996. A: forests on mineral soil; B, C: heat vegetation on mineral soil or parent rock; D: arable land or pasture; E areas without continuos vegetation cover or snowbed communities on mineral soil or parent rock; F: mires temporally saturated by water "dry" type; G: mires with peat saturated by flowing water in the top soil "wet" type; H: Lake; °: location for water and CPOM sampling.

II. Summary of a review on the origin and dynamics of Cs-137 discharge from land to streams rivers and lakes

This is a summary of a review that is intended to be presented in the International Symposium on Ionising Radiation: Protection of the Natural Environment which is to be held in Stockholm in May 1996. Equations, statistics and references has mainly been excluded from this version.

The significance of fish consumption as a contributor to the internal dosis of Cs-137 to man was revealed in the former NKS-project. Empirical data had shown that during a period of few years after the Chernobyl accident of maximum concentrations in fish, the activity typically decreased exponentially. The dose calculations were based on single effective ecological half-life for Cs-137 in fish of 5 years for Cs-137 resulting in an infinite time-integrated activity of 6.5 (kBq * kg^{-1} * year) for a hypothetical Scandinavian critical group. Later observations (e.g. in lake Päijänne in Finland and lake Storsjouten cf. chapter 1) has shown that the activity levels out and in the mentioned studies the specific activity 0.5 $kBq*kg^{-1}$ is a good approximation (deposition 10 - 40 $kBq*m^{-2}$). Under the assumption that the future decrease only will be governed by physical decay, the infinite time-integrated activity can be approximated to 10 to 20 (kBq * kg^{-1} * year) which then would give a significant contribution to the hypothetical critical group. It is thus important to follow the time development of Cs-137 in fish, the controlling factors and geographical abundance of critical catchments and lakes.

We believe that, apart from the physical decay, the decrease in fish will be determined by various biotic and abiotic factors. Our hypothesis is that a significant secondary contribution to the internal dose to man from fish, calculated over long time periods, is possible in:

- lakes surrounded by terrestrial ecosystems generating Cs-137 discharge in chemical forms that make biological uptake possible
- shallow lakes generating resuspension of sediments containing Cs-137 in available chemical forms.
- lakes with long residence times for the turnover of water and low sedimentation rates
- lakes with significant combinations of these factors

The factors mentioned above influencing Cs-137 in fish are being studied in the Nordic project NKS ECO-2.3 and this review concerns loss from catchments.

Pathways that might contribute to the loss of radionuclides from terrestrial ecosystems are resuspension via air, fires, harvesting by man or herbivores and discharge via drainage water. The only significant pathway among these for radioactive caesium transfer to lakes is discharge via drainage water, either as eroding particles, litter, dissolved organic ligands or cat ions. The main pathway for Cs-137 loss is via saturated surface runoff. Loss through groundwater has not been shown to give any significant contributions. An effective retention in the top organic and inorganic layers of mineral soils has been reported from numerous of investigations. Intercalation in clay lattice, effective incorporation into the biosphere and chemical binding in abiotic organic compounds in humus has been suggested as explanations for the effective retention in mineral soils.

Discharge areas, with shallow groundwater surfaces is often found in areas of thick organic soils. Thus fens and wet part of mire complexes would often be characterised as discharge areas. If they are in close vicinity to streams or lakes the saturated surface runoff from these areas should be able to generate a high discharge of radioactive caesium. Some investigations of interest for this project concerns the deposition of Cs-137 and Cs-134 in different areas:

The observed remaining fraction of the deposited Cs-137 of Chernobyl origin ($Bq*m^{-2}$) in soil is in most cases significantly lower in mires than in the surrounding mors, moulds or cultivated soils with mineral sub soils (figure 3, time < 10 years). This is also true for the

pre-Chernobyl fraction of Cs-137 (figure 3, time >10 years). The range is between 10 - 90% lower on the mire. Only in one observation, from northern England, the remaining deposition is higher in the mire. Apart from the ratio, figure 3 also combines the pre- and post-Chernobyl fractions of Cs-137 when data exists. These lines show that the relative remaining fraction of deposition on mires always is lower in the pre-Chernobyl case, for which the observations corresponds to a time further from the deposition event than in the Chernobyl case. An interpretation of figure 3 is that the decrease in activity on mires is time dependent and that at least two phases are involved. One rapid phase that in one case (t=0.09 years) seems to last no longer than some weeks, generating an effective decrease in the remaining deposition on mires. This rapid phase seems to be followed by a much slower phase of decreasing activity lasting for decades.

The processes behind these ratios might be a more effective scavenging of the fallout in forests than on areas sparse in high vegetation (which mires often are), a faster vertical migration of caesium in mires than on mineral soils or a more effective loss via drainage water from the mires.

The first assumption was tested during 1990 by Nelin and Nylén in areas of Northern Sweden identified as recharge areas. In this investigation no difference in Cs-137 deposition was found between mature forests and clear cuts. The second assumption might be valid in the parts of mires acting as recharge areas. The third assumption has been proved in Nyänget by calculations of the loss via drainage water and the remaining deposition in the differnt fractions of a mire in a forested catchment (Nyänget). The loss rate in this catchment was higher in fractions of the mire that was constantly saturated by water and explained this with the higher frequency of saturated surface runoff from these fractions of the mire.

A typical discharge area constitutes 1 to about 30% of the catchment with a spatial and temporal variation depending on topography, vegetation cover and climatic factors. It has been shown that the areal contribution to saturated surface discharge reaches its maximum of 20-30 % during spring and its minimum 1% during summer in the Nyänget catchment in Northern Sweden. If the rest of the catchment only contributes to runoff and the only fraction generating Cs-137 discharge is the mires or wetter fractions of these, then the activity concentration in the streams should be proportional to the areal contribution of mires or fractions of mires lining the streams. Further the change in activity in the stream should to some extent reflect the change in the pool of Cs-137 in the discharge areas. Fractions of the mires that constantly undergoes saturated surface discharge and are in contact with draining streams should empty their pool faster than fractions that only are saturated during snowmelt or heavy rainfall or loose its caesium via infiltration during these events. Catchments with high contributions of mires with contact to streams should generate higher activity concentrations in drainage water than catchments where mires are absent or local outstream areas.

The intensive investigation in the Nyänget catchment has shown that the initial loss of Cs-137 during the snowmelt of 1986 originated in the water saturated mire and that the amount was 44% of the original deposition. It has also been proved that the Nyänget mire has two fractions of different dynamics: the drier fraction with a loss of on average 2% * $year^{-1}$ and the wetter fraction that is constantly saturated by water and leaches 30 % * $year^{-1}$. In investigations in Northern Sweden where the positions for the soil samples has been described two classes have been identified. Drier fractions of the mire (characterised by dwarf shrubs and sphagnum forming tufts) and wetter fractions (destitute in field layer or characterised by Cyperious plants on sphagnum or peat). The relative remaining activity in these fractions are presented in figure 4. The horizontal line represents the observed 56% that remained in the Nyänget catchment after the melting of snow in 1986. This amount can be seen as the initial value for the secondary phase of relatively slow changes in remaining deposition. It is obvious from the figure that the secondary decrease in remaining deposition of caesium is faster in the wetter fractions of the mires.

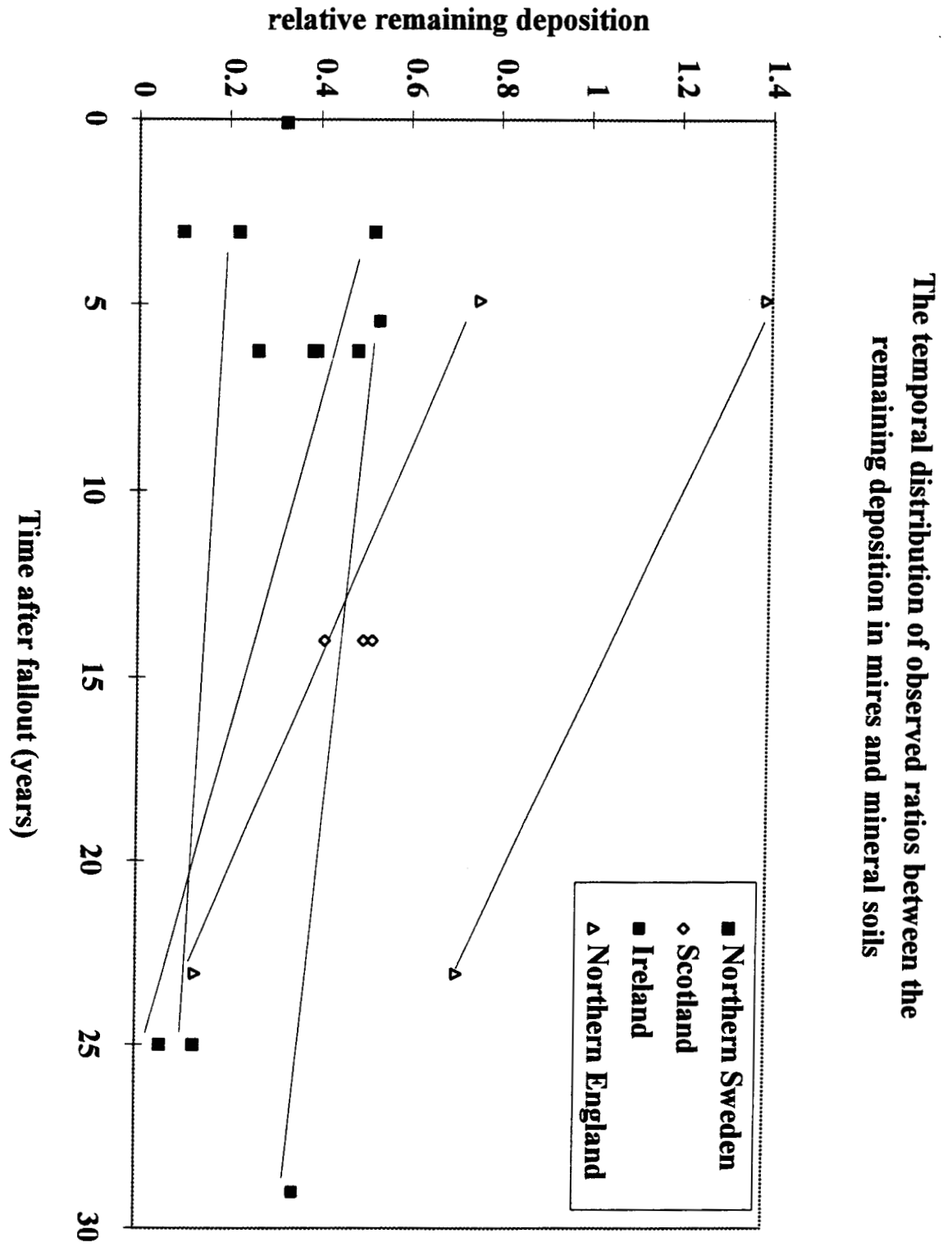


Figure B3. The ratio between deposition in mires and surrounding mineral soils. Observations were the fractions of pre- and post-Chernobyl Cs-137 has been unfolded are connected with lines. The fallout dates are in this presentation set to 1986-04-29 (Chernobyl) and 1963-06-01 (pre-Chernobyl).

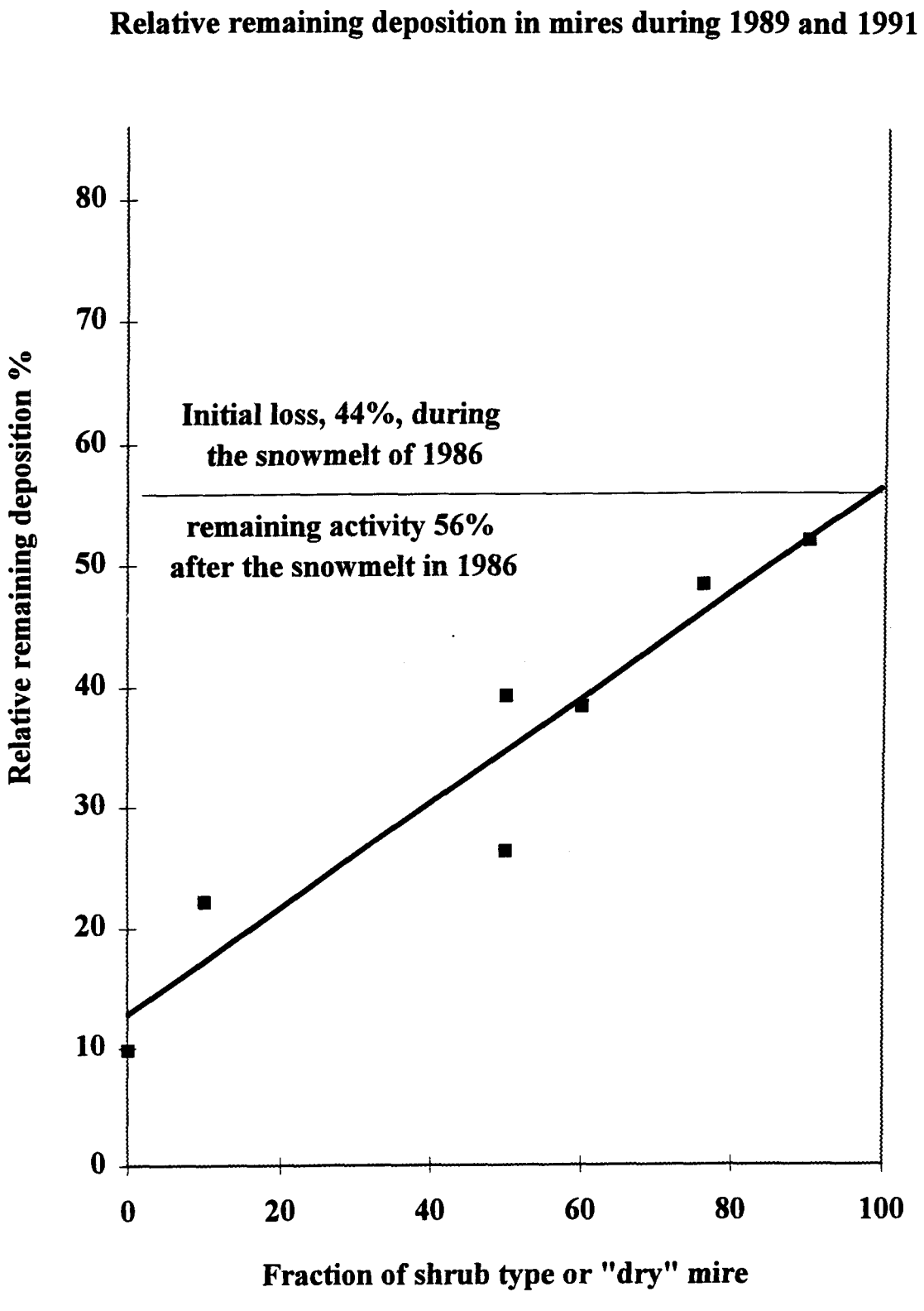


Figure B4. The relative remaining deposition of Cs-137 in mires during 1989 and 1991 related to the fraction of shrub type or drier mire in catchments of Northern Sweden. The horizontal line at 56% represents the remaining deposition on mires after the snowmelt in 1986.

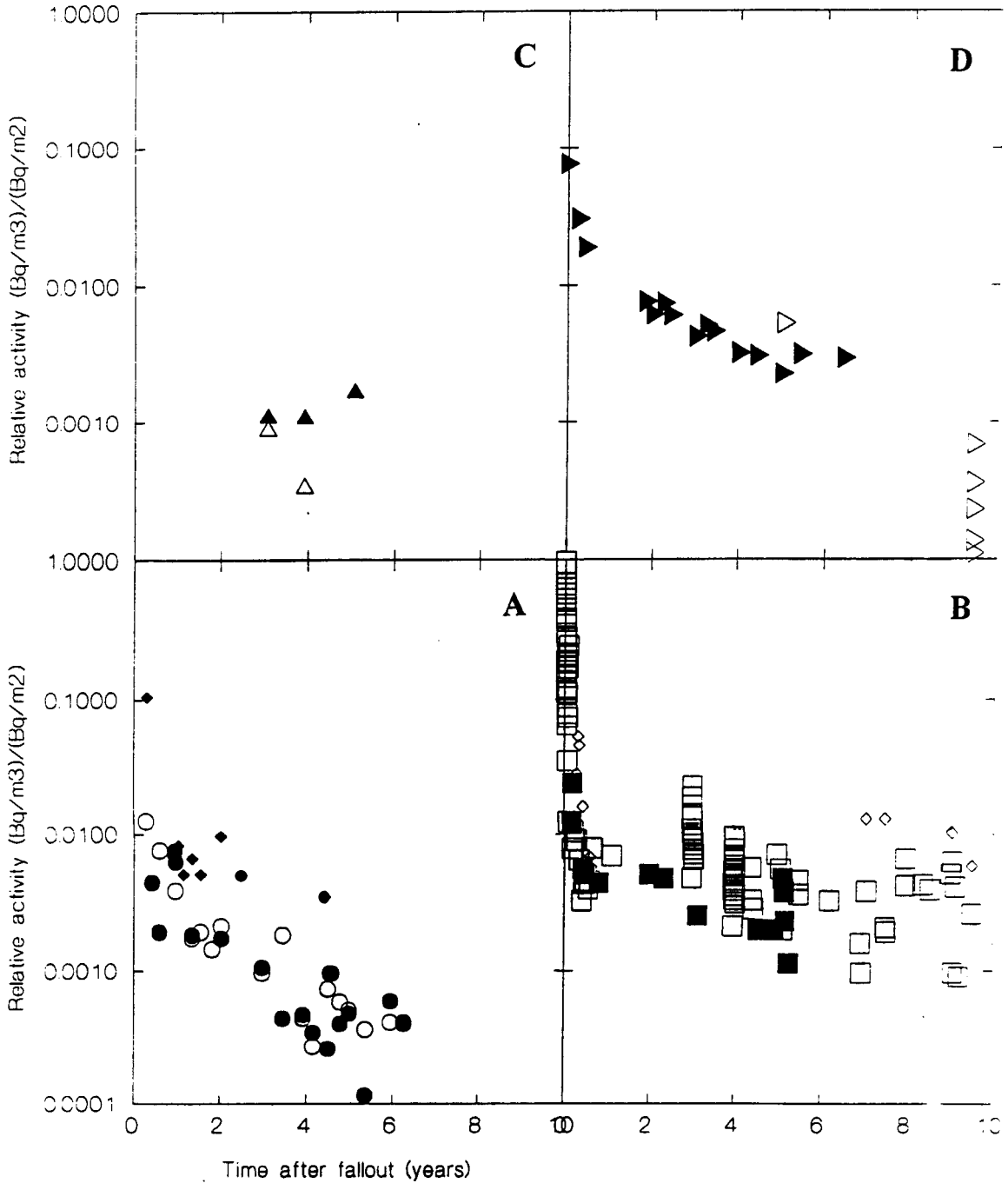


Figure B5. The relative activity concentration in Scandinavian catchment streams. a) inlets to Hillesjön (circles) and Sälgsjön (filled diamonds) in central Sweden. b) Nyänget (unfilled squares and unfilled diamonds) and Gideå (filled squares) in Northern Sweden. c) inlets to lake Heimdalen: Lektorbekken (unfilled triangles) and Brurskardsbekken (filled triangles). d) surface waters in the region of lake Päijänne (filled triangles) and five forest streams discharging into lake Päijänne (unfilled squares) in southern Finland.

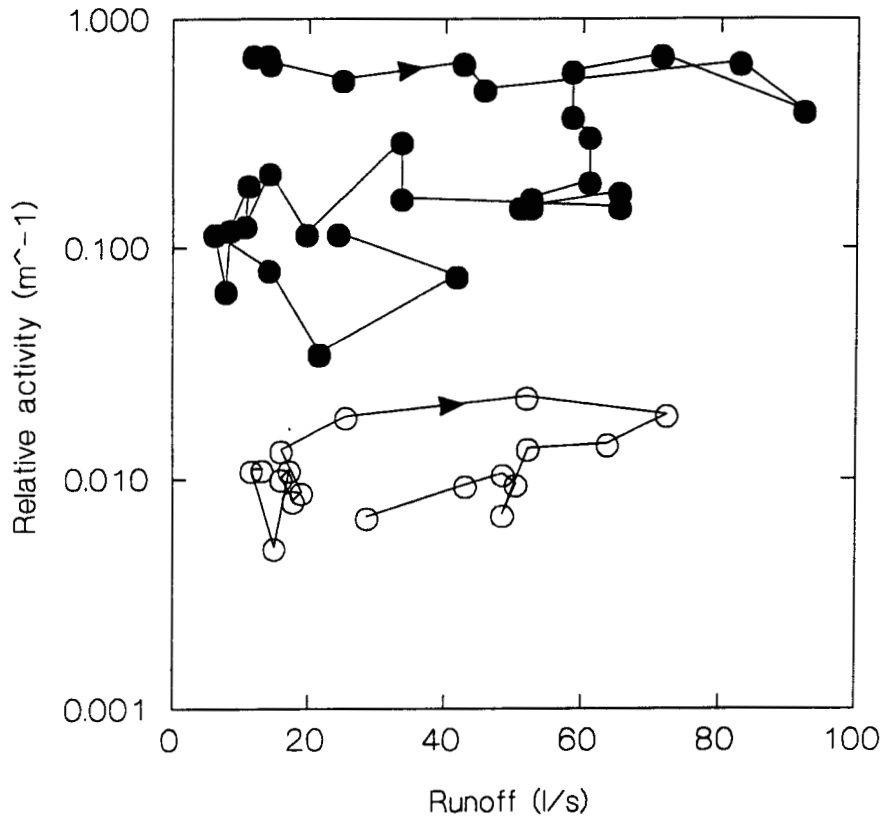


Figure B6. The changes in relative activity (m^{-1}) of Cs-137 as a function of runoff rate in the forest stream of the Nyänget catchment at two snowmelt periods: 1986 (Filled circles) and 1989 (unfilled circles). The arrows point in the direction of time progression through the event. The interval between samplings is 8, 16 or 24 hours.

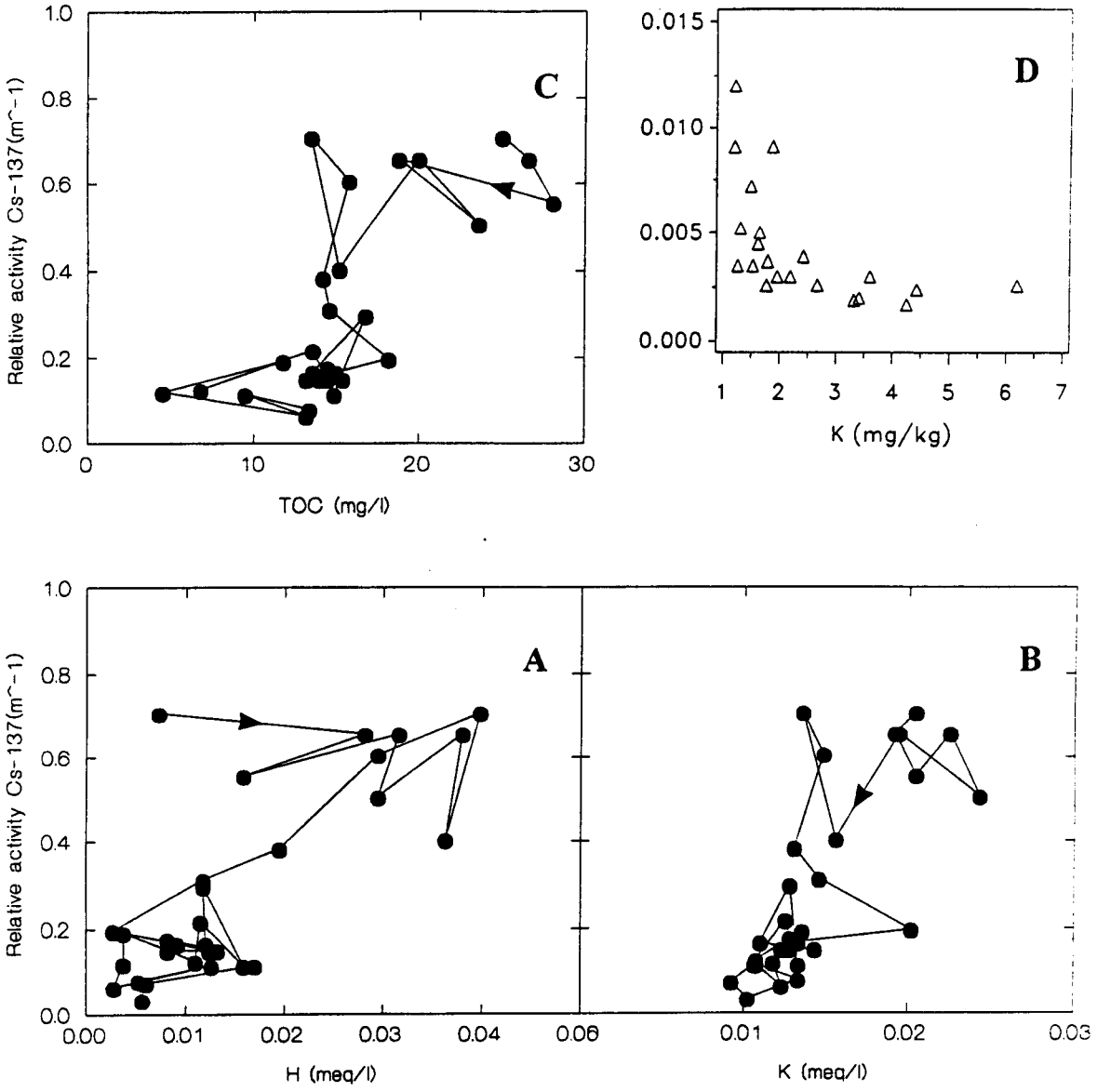


Figure B7. 7a-c: The relation between relative activity (m^{-1}) of Cs-137 and other chemical constituents in the forest stream of the Nyänget during the snowmelt of 1986. a) Cs-137 in relation to H^+ ($meq*l^{-1}$); b) Cs-137 in relation to K^+ ($meq*l^{-1}$); c) Cs-137 in relation to total organic carbon (TOC, $mg*l^{-1}$).). The arrows point in the direction of time progression through the event. The interval between samplings is 8 or 16 hours. 7 d) The relation between Cs-137 and K ($mg*kg^{-1}$) in Finnish surface waters.If this decrease is explained by discharge of Cs-137 via streams, as proven in the Nyänget catchment, then the corresponding activity in streams should follow at least two time dependent phases of decreasing activity concentration.

To be able to follow and test this hypothesis it is important to have good information about the catchments as well as long time series of activity concentration and runoff in the drainage streams. The discussion above indicate that, except for an initial decrease controlled by the contribution from discharge areas saturated during the fallout event, one might observe at least two phases of decreasing activity concentration in the streams. The loss from the constantly saturated discharge areas (typically some percent of the catchments) followed by slower changes in activity reflecting the peripheral fractions of the potential discharge areas (mires) leaching via infiltration or occasional surface runoff. In areas without deep organic soils lining the streams the base line of activity concentration in streams might also show the transport via mineral particles or groundwater.
At least two phases of decreasing relative activity in addition to a positive influence of runoff has been observed in Nordic drainage waters. Figure 5 shows the observations in the regions chosen for this study. In the hemiboreal region an important time series exists for the inlets to lake Hille and Sälgsjön (5a). The relative activity in the inlets to Hillesjön (circles) decrease more or less exponentially with some fluctuations during the first six years and has not stabilised until the end of the series. In the inlet of Sälgsjön (figure 5a filled diamonds) the base line seems to be at about 0.005 m^{-1} which is 50 times higher than the lowest observation in the Hille streams. The boreal forest stream Nyänget (figure 5b unfilled squares) shows an initial decrease from 1 to 0.01 m^{-1} during the first weeks after the Chernobyl fallout and an additional decrease with substantial variations in activity. The unfilled diamonds shows the samplings from the outlet of the mire. As suspected these values are higher reflecting the influence of the mire. Figure 5b (filled squares) show values for the near by Gideå catchment (<100 km, not included in this project) which is more or less equal in composition as the Nyänget catchment. The inlets to lake Heimdalen (figure 5c) have only been sampled at a few occasions. However the levels seems to be in the same range as in the other catchments. In Lektorbekken (unfilled triangles) the activity is similar to Hille. In figure 5d one of the time series from surface waters in the region of lake Päijänne (filled triangles) is included together with the first samplings in five forest streams for this project (unfilled triangles). The former are sampled during May and August the five for this project in June which might explain the tenfold lower activity.

One interesting observation in figure 5 is that in regions with higher fractions of mires (Sälgsjön 39%, Nyänget and Gideå 16 and 20% and Nyänget (diamonds) 54%) the relative activity continues to show high values during the period: In fact the activity during the spring peak during 1995 in the Nyänget catchment is as high as during the autumn of 1986 and that in Hille with only about 2% mire the activity seems to decrease exponentially through the series.

In humid climates the runoff intensities varies with a factor 50-100 between summer/winter and spring/autumn. As the intensities increases the discharge areas grow resulting in an increased transfer of Cs-137 through the stream water. These events of high transfer occur during spring in the northern and alpine parts of Scandinavia. In the southern parts runoff is more evenly distributed over time.

The influence of runoff on the concentration in stream water has bees studied in the Nyänget catchment. Figure 6 shows the influence of runoff at two periods of melting of snow: 1986 (filled circles) and 1989 (open circles). In both cases an hysteres effect with higher concentrations at the rising limb of runoff is evident. This can be explained with a decreasing pool of available activity between the rising and falling limb at a fixed runoff rate together with changes in the contribution to runoff from the different path ways (surface runoff and groundwater flow). The loss during these few days of high runoff constitutes the main fraction of the yearly loss in the Nyänget catchment. The values for Nyänget in figure 5b can thus be divided into values for high and low runoff. The values for runoff intensities higher than half the maximum value recorded during the period shows no significant change over the years for $t > 1$ month while the values at lower runoff rates decreases with 18% / year. This support the hypothesis that constantly saturated discharge areas has a higher loss rate. The even higher decrease in Hille (figure 5a) has not yet been

explained but might be caused by a discharge area of insignificant aerial contribution but low Kd (e.g. the streambanks or low fractions of mires).

Chemical analogues to Cs-137 might be used to improve the predictions in between samplings or regions. Until now only a few attempts has been done in this direction. Caesium in Cumbrian mires has bees shown to be easily exchanged for Calcium by simple ion exchange. Data exists for Nordic catchments but has so far not been properly analysed. However for the Nyänget catchment preliminary result comparisons between Cs-137, K^+, H^+ and total organic carbon (TOC) in the stream is presented in figure 7a to c showing the relation between these during the fallout event and snowmelt period in 1986. Also the relation between Cs-137 and potassium in Finnish surface waters from the former NKS-project (figure 7d) is included. This data together with data from the spring of 1989 and 1990 (Vindeln) together with the new data in this project will be analysed more closely during the next period.

A simple model for the discharge of radioactive caesium from catchments

Different approaches has been used, among the individual national projects, to model the loss of Cs-137 from catchments to lakes including different parameters e.g. environments, Kd, through fall and retention from vegetation, average yearly runoff and seasonal changes in runoff. In the frame of this project however the model so far is composed by four compartments (figure 8). This is to our opinion the limit of reduction. The model does not take seasonal changes into consideration. The reason for constructing such a simple model is that it can easily be included in a GIS-model (chapter 1) and generate examples of integrated yearly loss from different environments. The next step will then be to expand the model so that the dynamics of the studied catchments can be properly described.

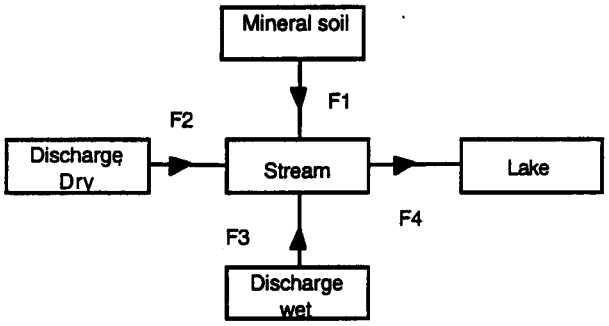


Figure B8. The reduced model generating yearly loss from catchment with different contributions of discharge areas.

The model is composed by tree terrestrial compartments (table 2). The transfer coefficients F1 to F3 is taken from the observation in the Nyänget catchment. The recharge areas discharging into the streambank with a transfer of 0.0003 $year^{-1}$ which is the detection limit for the groundwater measurements in the Nyänget catchment. The drier fraction of mire engaged in surface discharge mainly during spring and with a loss of 0.02 $year^{-1}$. The wet fraction of permanent surface saturated overland flow and loss of 0.3 $year^{-1}$. and the stream with a loss of 365 $year^{-1}$. The compartments in direct contact with the stream (which is a simplification of the real conditions). The surface of the stream is 1.5 permille of the catchment. The relative activity in the stream is calculated by dividing the yearly discharge in flow F4 with the yearly runoff. the model has been tested on. Existing data and some results can be seen in chapter 1 (här menar jag A delen).

Conclusions.

- It is important to generate long time series to be able to follow the impact of the slow leakage from catchments on the development of Cs-137 in fish.

- After an initial decrease in activity concentration of radioactive caesium in streams lasting for some weeks after the Chernobyl accident the rate in decrease will mainly be controlled by the fraction of mires in the catchments.

- In catchments with high fractions of mires no significant decrease in activity concentration have been observed since the summer of 1986 except at low flowrates.

- A seasonal variation of the transfer of radioactive caesium with high transfer during spring in the northern and alpine regions of Scandinavia has been observed. It is of interest to investigate the corresponding seasonal changes for the middle and southern parts of Scandinavia and link this seasonality to the biotic and abiotic processes in lakes.

- According to measurements in stream water certain lake systems might generate activity in fish that will decrease with a rate that is close to two (physical decay) percent per year.

Table B1. Characteristics of the studied catchments (table will be completed during 1996).

Catchment	Country	Annual precipi-tation mm*year^{-1}	Annual run off mm* year^{-1}	Altitude m.a.s	Catchment area km^2	mean annual temp °C	Cs-137 depositio Chernob kBq*m^{-2}
Lektorbekken	Norway	800		1090-1840	23.6	-1.2 to 0.4	150
Nyanget	Sweden	710	330				20
Angland	Sweden		300				100
Hille	Sweden		300				100
Paijanne 1	Finland		300				
Paijanne 2	Finland		300				

Table B2. Transfer coefficients for the GIS-model.

Compartment:	Symbol	Yearly loss t < 1 month	Yearly loss t > 1 month
Mineral soil	F1	0.0002	0.0002
Discharge dry	F2	7.1	0.02
Discharge wet	F3	7.1	0.3
Stream	F4	365	365

C. SEDIMENTS AS A SECONDARY SOURCE OF CS-137 TO FISH

In most Nordic lakes, most radiocaesium (>90%, >99%) is associated with sediments. It is thus likely that sediments are an important secondary source of Cs-137 to the lacustrine biosphere. The question is, to what extent, and during how long time. Our aims are to determine the relative importance of sediments (versus catchment soils) as a secondary source of Cs-137 in lakes, to quantify long-term trends, and to evaluate potential remedial actions.

The (abiotic) mobility of Cs-137 in sediments may be controlled by fundamentally different processes:
- Resuspension of particulate radiocaesium, especially in shallow areas (shallow lakes?)
- Diffusion of dissolved radiocaesium, especially in anoxic environments (deep lakes?)

Sediments can act as a secondary source of Cs-137 to organisms in several ways:
1) Resuspension of sediments (physical remobilization):
   - equilibration with the dissolved phase in the water followed by direct uptake in biota
   - ingestion of suspended particles by planktonic animals
2) Direct uptake from sediments (biological remobilization):
   - assimilation by rooted plants
   - ingestion of contaminated sediments by benthic animals
3) Diffusion from sediments into the water (chemical remobilization)
   - dissolution and diffusion into the water followed by direct uptake in biota

The mobility and accessibility of particulate Cs-137 in contaminated surface sediments is controlled in several ways:
1) Dilution and burial
   - new sedimentation
2) Vertical mixing of surface sediments
   - bioturbation
   - physical mixing through wind and wave action
3) Horizontal redistribution of surface sediments
   - resuspension and settling

In this project, various methods are applied in different lakes and streams:
- Horizontal and vertical (re-)distribution of radiocaesium i lake sediment
- Particulate radiocaesium in sediment traps (sedimenting material)
- Physical and chemical speciation of radiocaesium in different sediments and waters
- Diffusion studies in the field and in the laboratory

Inventories of Cs-137 in lake sediments

The mean inventory (Bq m^{-2}) varies widely among lakes. The mean inventory appears to be controlled by the water residence time and the sedimentation rate, i.e. by the initial retention during the spring flushing in 1986.

Relationship of Cs-137 inventories with water depth

Sediment studies show that in shallow lakes, the sediment inventories of Cs-137 are horizontally fairly evenly distributed across the lake. In deep lakes, however, the sediment inventory of Cs-137 is highly correlated with water depth, demonstrating a substantial net transport of sedimentary Cs-137 from littoral areas towards the profundal zone of the lake basin (Fig.). Flux estimates suggest that much of the Cs-137 has been removed from the shallow areas within the first six years after contamination, while the mean inventory in deep areas has increased substantially.

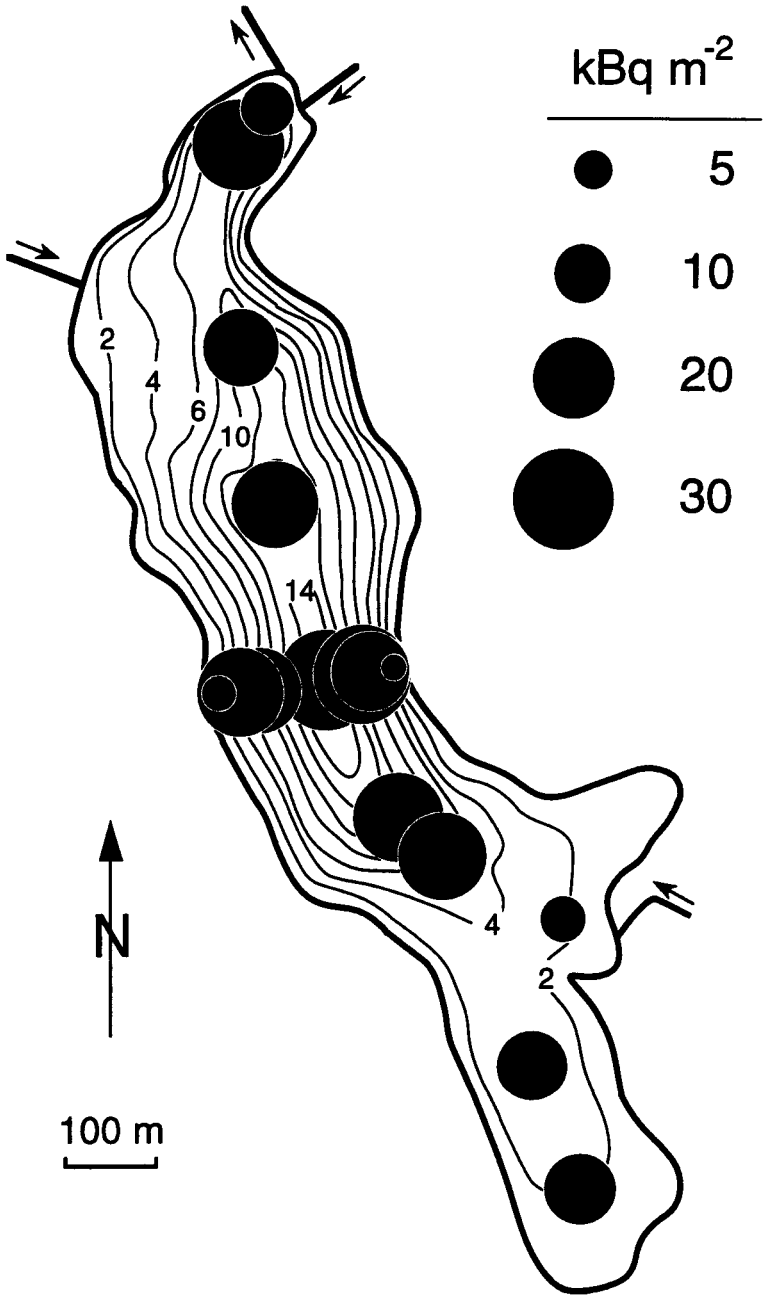


Fig.: Bathymetric map of Lake Loppesjön (Hudiksvall, central Sweden) showing the variation of area-specific total activities of 137Cs (kBq m--2) in the sediment six years after the nuclear fallout from the Chernobyl accident. The values are corrected for radioactive decay to 1 May 1986. Contour depths in meters.

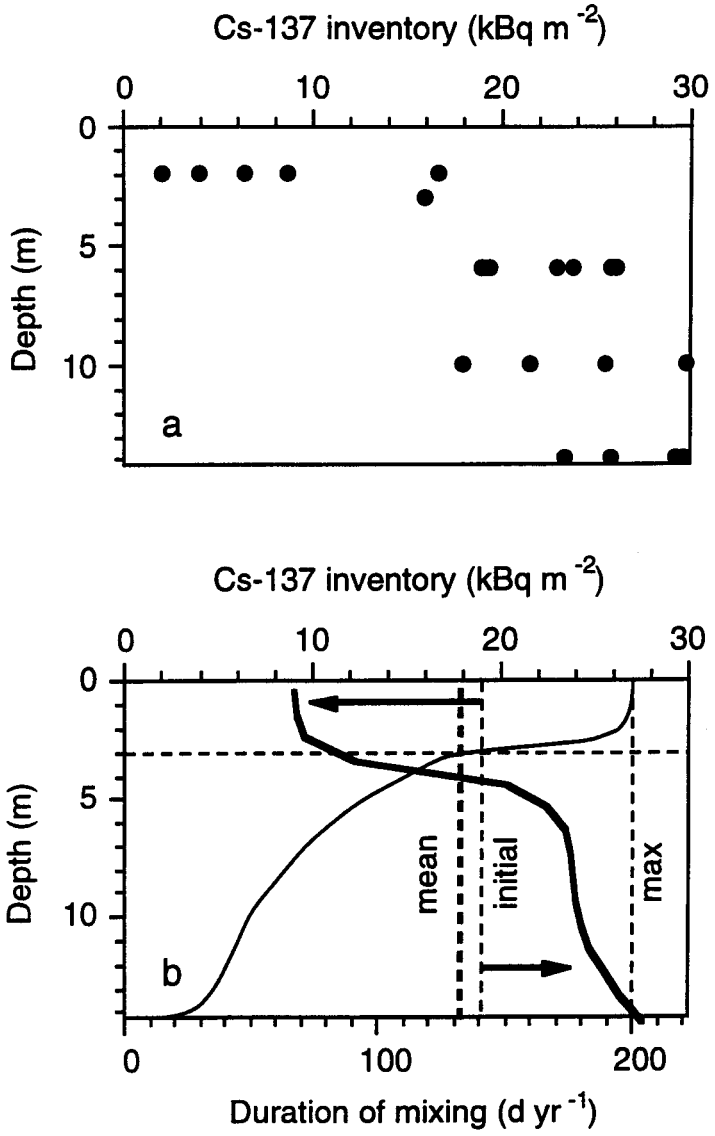


Fig. a: Total inventories of sedimentary 137Cs (kBq m-2) at different water depths in Lake Loppesjön, Hudiksvall, central Sweden in 1992. The values are corrected for radioactive decay to 1 May 1986. b: Interpolation of data as a function of depth (bold line) together with mean, maximum of 137Cs inventories (dashed vertical lines) and changes from 1986 to 1992 relative to the initial deposition (arrows). The probability of wind-induced mixing (thin line) is defined as the duration of destratification above the sediment. Epilimnetic depth observed during maximum summer stratification is shown with a dashed horizontal line.

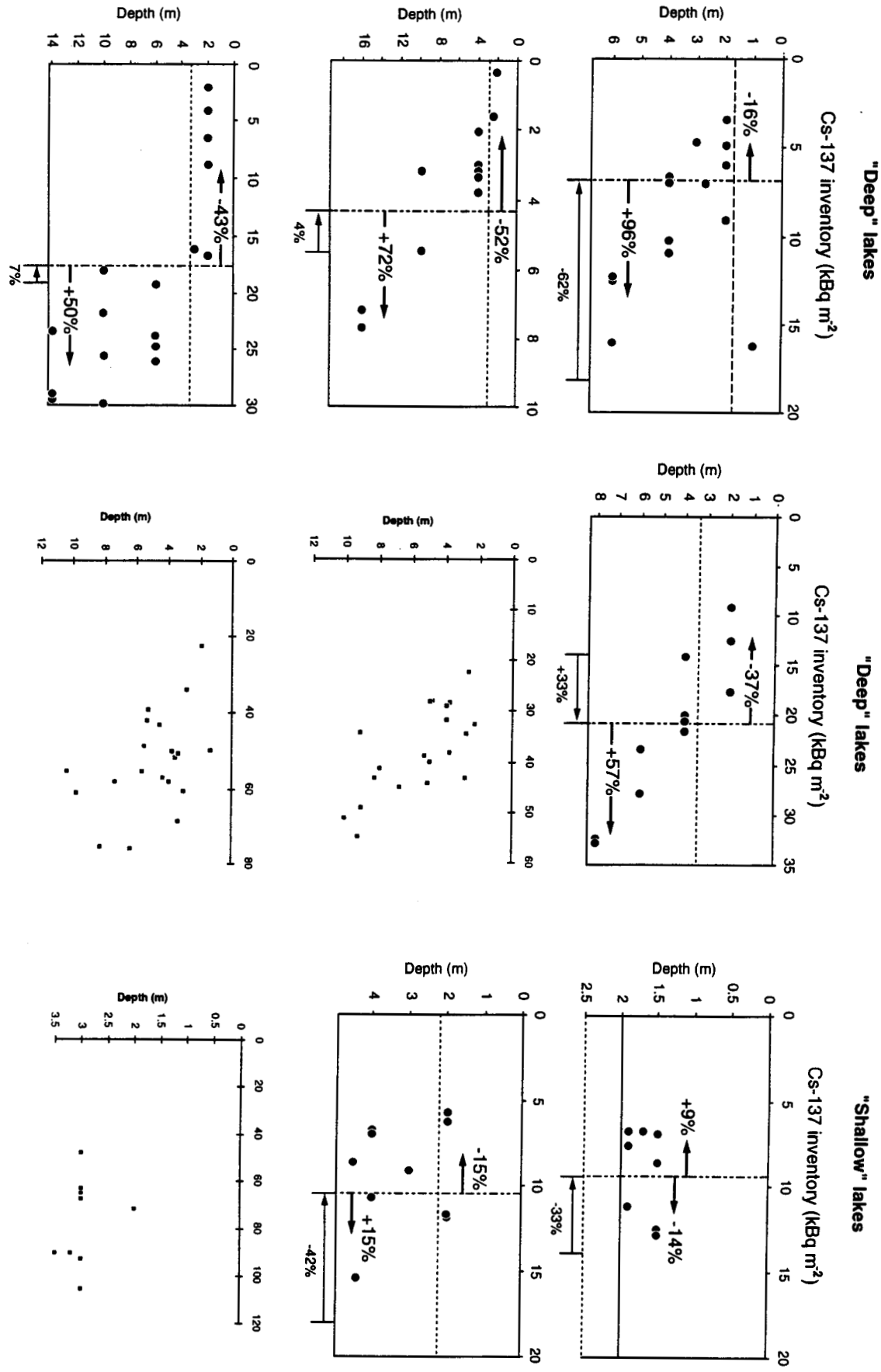


Fig. a: Total inventories of sedimentary 137Cs (kBq m-2) at different water depths in 9 lakes of different morphology. The values are corrected for radioactive decay to 1 May 1986.

Relationship of Cs-137 inventories with bottom slope

In the profundal sediments, the amount of Cs-137 stored showed no obvious relationship with slope. In the sediments of the littoral zone, however, an increasing slope appears to result in a reduced amount of Cs-137 stored in the sediment: Cs-137 inventories on flat bottoms exceeded the inventories at the steep slopes about fivefold. Thus, flat bottoms seem to allow continuous sediment accumulation, even in the presence of resuspension.

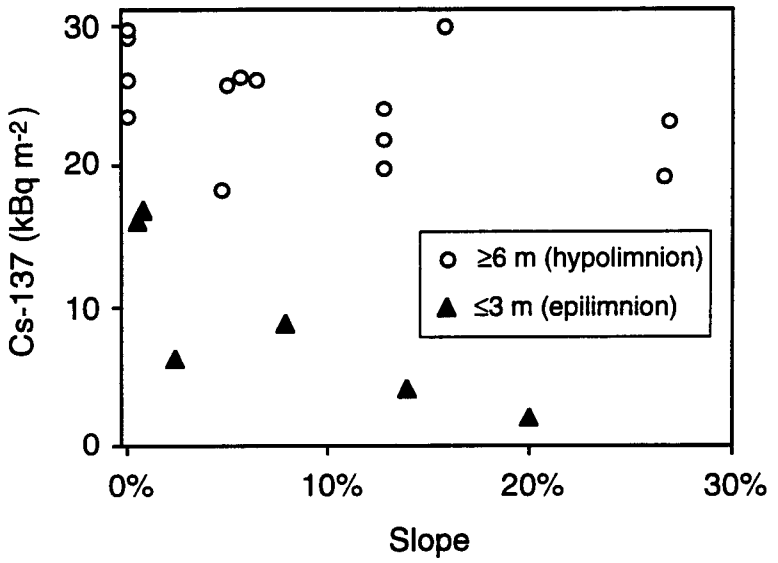


Fig.: Relationship between sedimentary 137Cs inventory (kBq m-2) and bottom slope (%) in 1992 at different depths of Lake Loppesjön, Hudiksvall, central Sweden.

Resuspension of Cs-137

The focusing of sediment Cs-137 towards deep areas can be explained by wind-driven resuspension and redeposition of surficial sediments. Cs-137 activities of settling material can be used to trace the source of the accumulating material. Mean activity concentrations of Cs-137 in surface sediments (0.5 cm) from different depth zones were all similar, suggesting a deposition of the same material throughout the whole lake basin. Moreover, settling material collected in epilimnetic sediment traps was just as contaminated as the surficial sediments, even during the period of maximal thermal stratification. The contamination of settling material during the summer season, when inflow from the watershed is almost absent, strongly suggests that a large proportion of the settling material originates from resuspended sediments.

Primarily in shallow near-shore sediments, mixing processes carry buried contaminated particles back to the surface, where they are frequently resuspended. Subsequently, they are distributed in the whole water mass and settle at random. This has resulted in a pronounced focusing of Cs-137 towards deeper lake areas.

The vertical and horizontal fluxes of Cs-137 caused by resuspension and focusing can be quantified from the undisturbed cores taken in the deepest part of some lakes. Such cores show a significant secondary deposition of Cs-137 during the years following the initial contamination. In at least some sediments, this secondary contamination is several times larger than the Cs-137 input to the lake from surrounding land and must be explained otherwise.

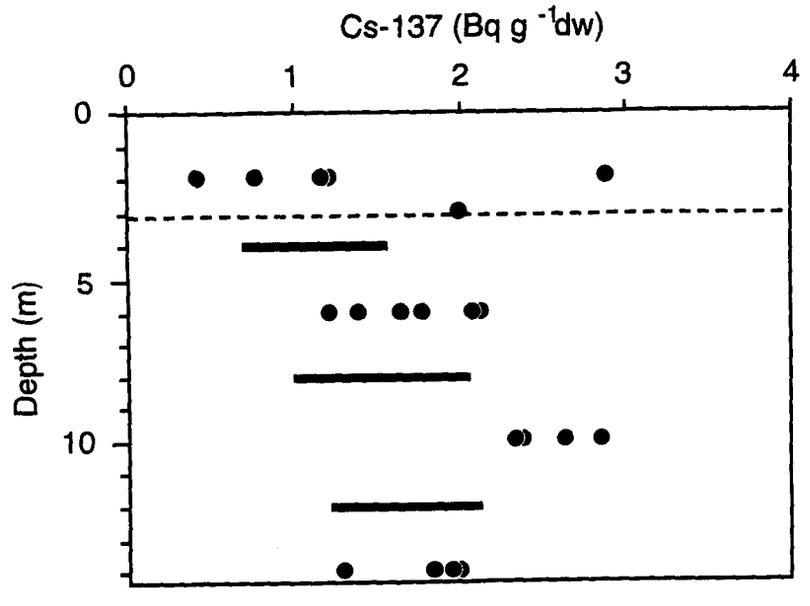


Fig. : Relationship with water depth of specific 137Cs activity concentrations (Bq g-1 dw) in surficial sediments (0-0.5 cm) 1992 (dots) and in settling material (bold lines showing ranges) during thermal stratification in 1993 for Lake Loppesjön, Hudiksvall, central Sweden.

Resuspension fluxes of Cs-137

The fluxes of sediment-associated Cs-137 in different parts of the lake can be quantified from the losses of Cs-137 relative to a total gross deposition. The areal distribution of sedimentary Cs-137 in deep lakes is strongly correlated with water depth, with inventories increasing from the littoral zones towards the deep profundal zone. Interpolated mean Cs-137 inventories can be derived for each depth (Fig. above). Horizontal and vertical annual fluxes of Cs-137 can be calculated by comparing the gross losses to the net losses or increases relative to the mean inventory or the initial sedimentation of Cs-137. Resuspension fluxes to the water column can be estimated as the gross losses from the sediment, assuming even distribution of deposition over the lake.

The procedure provides annual net rates of change at different depths, showing a transition from losses to accumulation with increasing water depth (Fig.). Shallow areas have lost up to almost 90% of the initial Cs-137 input, while the inventory in the deepest area exceeds the lake mean by up to twofold. The water depth where the inventory of Cs-137 is equal to the average inventory in the lake (where gross losses are balanced by redeposition) often coincides with the mixing depth of the water column during summer stratification. The calculated annual gross losses can reach values up to 30% in the littoral zone, which indicates a substantial erosion and remobilization of sedimentary matter. In the deep areas, substantial gross losses indicate that resuspension occurs even in deep waters, but the data also suggest that resuspension probability gradually decreases with depth. Accordingly, the gross loss rates of Cs-137 from sediments are strongly related to the intensity of water mixing (see above). The strong relationship supports wind-driven resuspension to be the cause of the strong focusing of sedimentary Cs-137.

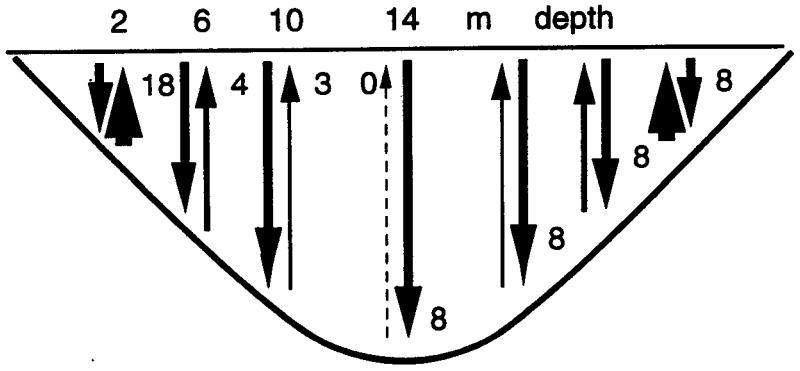


Fig.: Vertical fluxes of 137Cs (kBq m-2) to and from the sediment in Lake Loppesjön during the first six years after the nuclear fallout from the Chernobyl accident.

A prognostic lake recovery model based on sediment resuspension

The horizontal and vertical distribution of sedimentary Cs-137 in lake sediments can be used to assess the long-term immobilization of Cs-137 emitted by the nuclear accident in Chernobyl in 1986. Observed resuspension fluxes can be projected in order to make prognoses about the state of contamination in different parts of a lake ecosystem. Most biota live in shallow areas within the warm water layer which is mixed during the whole summer. In this zone, which is most exposed to wind-driven wave action, sediment resuspension results in a prolonged contamination of the water column. Accordingly, Cs-137 concentrations in fish decrease very slowly, especially in shallow lakes. At the same time, shallow areas in deep lakes are gradually cleaned by a net removal of contaminated surface sediments, which are deposited in biologically less active zones where total contamination is increasing.

A model was developed to assess the long-term immobilization of Cs-137 and to make prognoses about the state of contamination in different parts of a lake ecosystem (Fig.). The observed fluxes of sedimentary Cs-137 and their projection into the future indicate that the initial recovery of the sediments is limited to the most shallow area, where erosion results in an elimination half-time of about half a decade. Only later, decontamination progresses towards deeper zones, where contaminated particles are slowly buried.

The long-term removal of Cs-137 from the water column appears thus to be largely controlled by sediment resuspension. This process is likely to delay the recovery of many lakes from Cs-137 contamination. This suggests that the effects of the Chernobyl accident will remain a problem in Swedish lakes for several decades. On the other hand the long-term consequence of particle focusing is an eventual accumulation of Cs-137 in the least bio-active areas of the lake.

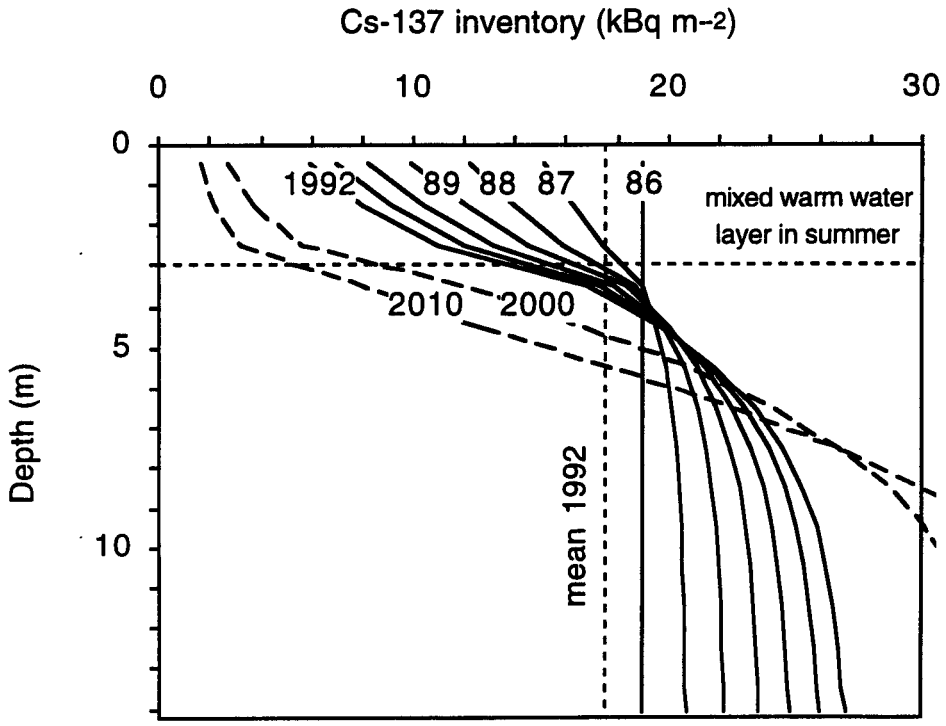


Fig.: Chernobyl Cs-137 in a Swedish lake: Observed, estimated and projected values of mean sediment inventories at different depths during the years after the nuclear fallout in 1986.

Diffusion of dissolved Cs-137 in and from lake sediments

Eight years after the Chernobyl fallout, the distribution of Cs-137 observed in the sediment of a small lake suggests that most of the Cs-137 entering the lake was rapidly deposited in the sediment. However, vertical profiles in undisturbed sediments show that Cs-137 is not irreversibly bound to particles, but slowly remobilized during the following years. These profiles show that a large fraction of the Cs-137 inventory is confined to a sediment layer of ≤0.5 cm. Below this peak layer, a tail of Cs-137 is visible to a depth >0.1 m below the peak (Fig.). Apparently, part of the radiocesium originating from the initial deposition has penetrated deeper into the sediment. Discontinuous gradients are evident on both sides around the peak, suggesting complex patterns of sorption and desorption.

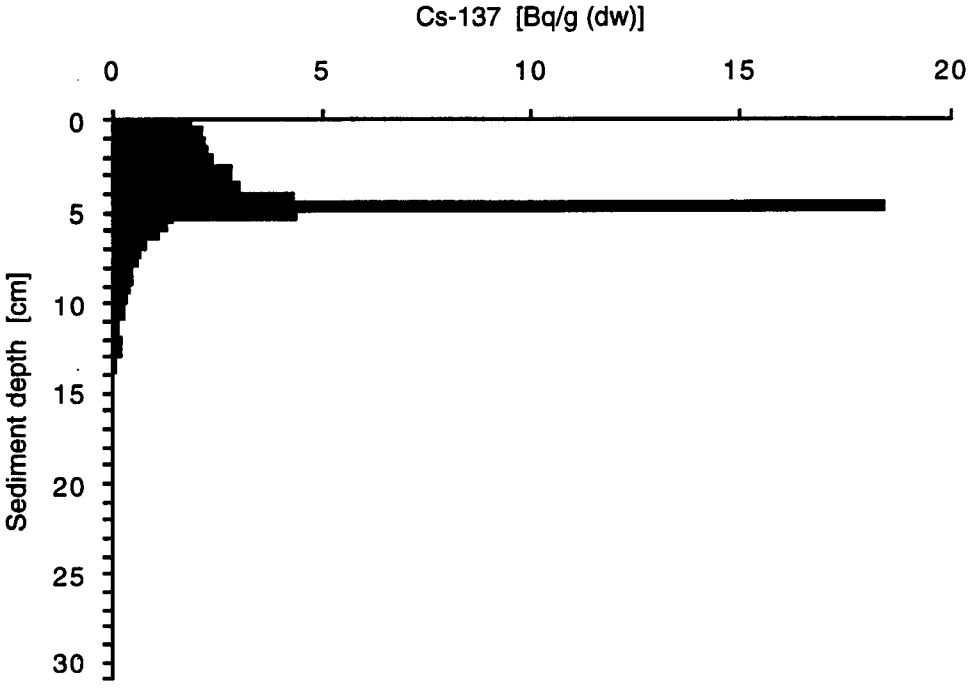


Fig.: Vertical profile of Cs-137 activity concentrations in a lake sediment core collected in Loppesjön (central Sweden) 8 years after the Chernobyl fallout in spring 1986. The profile is discontinuous, with a distinct peak and long tails.

The Cs-137 profile with both a distinct peak and long tails indicates a gradual desorption of Cs-137 from the original deposit and a vertical redistribution caused by diffusion in a matrix with different sorption properties. A three-phase model was developed that describes slowly desorbing binding sites in the initial deposit by an instantaneous equilibration changing slowly over time:

$$c_{T2}(x,t) = \frac{M}{\sqrt{4\Pi \frac{D}{1+K_d s} t}} \left[\exp\left(- \frac{(z-\varepsilon)^2}{4 \frac{D}{1+K_d s} t} \right) - \exp\left(- \frac{(z+\varepsilon)^2}{4 \frac{D}{1+K_d s} t} \right) \right]$$

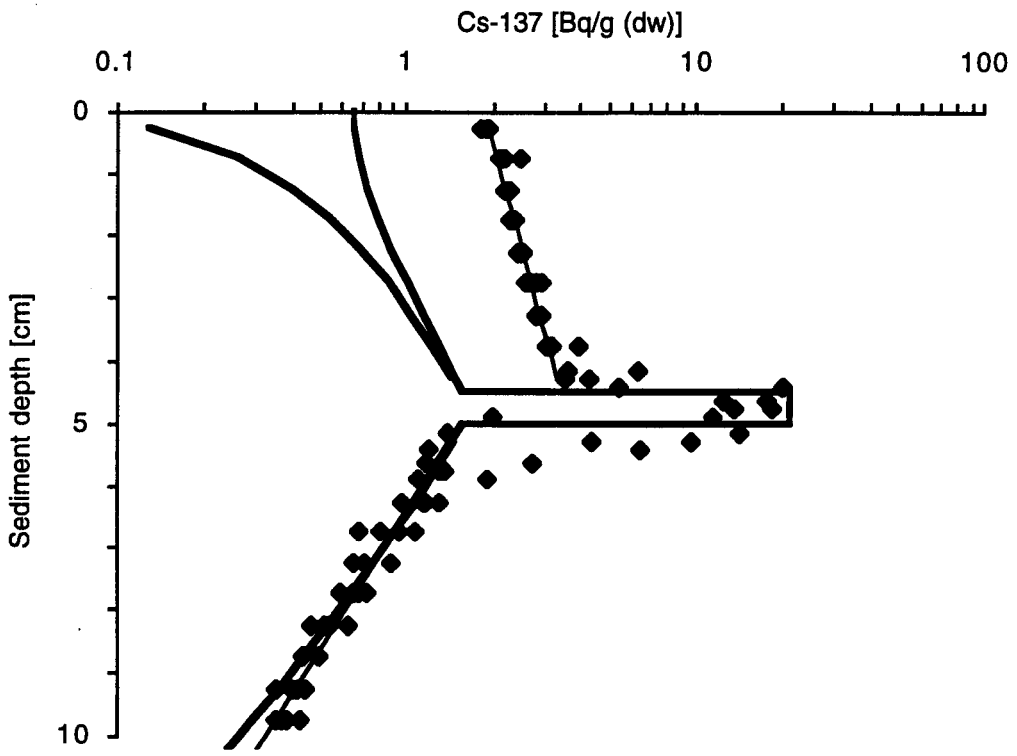


Fig.: Vertical profile of log-transformed Cs-137 activity concentrations in L. Loppesjön (central Sweden) from four sediment cores collected about 8 years after the Chernobyl fallout in spring 1986. Lines show exponential curve fits (thin) and the numerical solution of a 3-phase diffusion model with two delimiting boundary conditions at the sediment surface: Cs-137 concentration = 0 and Cs-137 flux = 0, respectively. Apart from slight variations in peak depth and in peak shape caused by the slicing, most of the variation among data from a given core depth can be explained by gamma counting statistics alone.

An explanation for the observed behaviour of rapid sorption in the water and subsequent slow desorption in the sediment can be found in the roughly hundredfold difference in Kd between these environments. The solute and particulate partitioning of the initial inventory is constituted by the environmental condition prevailing in the lake water during the fall-out. Since the conditions in the sediment are different from the lake water, the equilibrium ratio of solute and particulate Cs-137 of the inventory may successively adjust to the surrounding partitioning ratio. Such a change in Kd can be caused by ammonia ($NH4^+$) originating from the decomposition of organic matter. Ammonia ions are directly competing with Cs-137 for sorption sites, causing an inverse relationship between Kd and $NH4^+$ concentration, which usually increases with sediment depth. A hundred-fold difference in Kd between water and sediment would require a similar increase in $NH4^+$ concentration. This is not unlikely, since only 1‰ of the sediment nitrogen needs to be in the form of $NH4^+$ to meet this condition. Equilibration of intact sediment cores from the same site indeed produced $NH4^+$ concentrations in overlaying water that were thousand-fold higher than in the surface water of the lake. Accordingly, the vertical redistribution of Cs-137 in this sediment can be accounted for by ionic diffusion alone.

The slow desorption can be explained by an inert rather than an instantaneous

equilibration with the dissolved phase. This would be in accordance with the Cs-137 sorption on clay minerals during short-term experiments, suggesting a slow binding with a half-saturation time of only one year, and a subsequent redistribution in minerals that was earlier concluded to be irreversible.

Our sediment profile rather suggests a continuous loss of Cs-137 from the inert to the dissolved phase, as indicated by long tails surrounding a sharp peak, and by fairly steep tail gradients being maintained after several years of equilibration in a natural environment. This chemical remobilization may contribute to the slow recovery of lakes contaminated with Cs-137, especially in lakes with anoxic bottom waters.

Comparison of secondary inputs of Cs-137: sediments versus soils

In order to maintain observed concentrations of Cs-137 in the water of typical shallow forest lakes, surrounding soils must release 0.1-1% of their Cs-137 inventory every year. This is less than the observed loss from wetlands (>1%), but higher than the loss from upland soils (<0.03%). Consequently, at least in lakes with a catchment poor in wetlands, sediments are probably the main source of Cs-137 in lake waters and fish.

A preliminary evaluation of a study of stream sediments in a wetland-rich area indicates that these sediments contain several times less Cs-137 than the surface sediments of a downstream lake, although water exchange is rather rapid in this lake. Moreover, the total inventory of Cs-137 in the sediment of this lake has not increased over time. This suggests that even in lakes with a significant supply of Cs-137 from land, the outflow of Cs-137 may be larger than the inflow, andthe Cs-137 concentration on suspended and settling particles may be diluted rather than increased by stream input.

In other words, the relative importance of sediments versus soils in sustaining high levels of Cs-137 in waters and fish of Nordic lakes is still unclear. This will be one of the main issues in the next project period, since this is a key to both understanding and to radiation protection measures.

Future sediment studies

- compilation and evaluation of sediment and sediment trap data
- field studies to measure changes in sediment inventories of Cs-137
- field studies to compare soils, stream sediments, and lake sediments.
- Experimental studies of the diffusion of Cs-137 in sediments

Table C1: Nordic lakes where data on sediment inventories of Cs-137 are available.

Lake• = sampled more than once over time
(Lake) = 1-2 cores only

• *North-central Sweden*
Blyträsket
Blåkullträsket
Bubergssjön
Hamstasjön
Mjösjön
Rävsjön
Skavtjärn
V. Långedsjön

• *Central Sweden*
Loppesjön•
Blacksåstjärn•
Bottentjärn•
Stensjön

Alebosjön
Ecklingen
Järvsjön

• *South-central Sweden*
Flatsjön•
Siggeforasjön•
Råksjön
(Ekholmssjön•)
(Hillesjön•)

• *South-central Finland*
Päijänne•
(Ontojärvi•)
(Pielinen)
(Kallavesi)
(Konnevesi•)
(Näsijärvi•)
(Keurusselkä•)
(Pyhäjärvi•)

• *Norway*
Høysjøen
Øvre Heimdalsvatn•

• *Island*
(Thingvallavatn)?

• *Denmark*
(?)

Table C2: Nordic lakes where data on Cs-137 in sediment traps are available.

Lake• = sampled over several years and at short time intervals

• *North-central Sweden*	
?	
• *Central Sweden*	
Loppesjön•	1992-1994
Blacksåstjärn•	1986-1993
Sörsjön	1993
Bottentjärn	1986-89(92)
Bältbosjön	1986-89(92)
Ecklingen	1986-89(92)
Långsjön	1986-89(92)
Tansen	1986-89(92)
Hamstasjön	1986-89(92)
Herrbodtjärn	1986-89(92)
Lill-Bandsjön	1986-89(92)
Lill-Selssjön	1986-89(92)
Rävsjön	1986-89(92)
S.-Habborn	1986-89(92)
Selasjön	1986-89(92)
V.-Långedsjön	1986-89(92)
V.-Lövsjön	1986-89(92)
• *South-central Sweden*	
Flatsjön•	1987-1993
Siggeforasjön•	1987-1994
Råksjön•	1991-1993
Ekholmssjön	1987
• *South-central Finland*	
Päijänne ?	

D. BIOAVAILABILITY OF PARTICULATE CS-137 TO INVERTEBRATES

Resuspended as well as deposited particles are taken up by planktonic or benthic invertebrates, which are likely to assimilate associated Cs-137. Suspended particles are taken up by filtering zooplankton, whereby Cs-137 is likely to be transferred to fish. Moreover, benthic animals in the littoral sediments, which also constitute an important source of food to fish, are exposed to the Cs-137 at the sediment surface. Consequently, concentrations of Cs-137 in lacustrine food chains are likely to be controlled by the turnover and burial of contaminated sediment particles. Food chain transfer of both sedimentary and resuspended Cs-137 may thus substantially retard the natural decontamination of lacustrine fish communities.

Field and laboratry studies

One of the most important factors in this transfer is the bioavailability of particle-associated Cs-137. This bioavailability is studies in this project by compiling available field data of Cs-137 in invertebrates and the relationship to concentrations in water and sediments. In addition, laboratory experiments have been initiated to study the uptake of Cs-137 from contaminated lake sediments. Both approaches are in progress, and results will be presented in the final report.

D. OTHER PLANS

In the near future, we will try to expand our project to include the Baltic countries.

RELEVANT REPORTS BY THE PROJECT PARTICIPANTS

(peer-reviewed international publications are underlined)

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