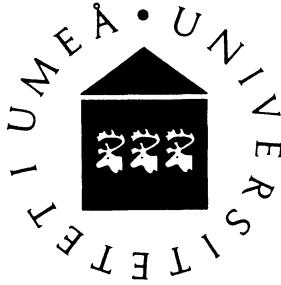


# MERCURY AND RADIOCESIUM IN SWEDISH LAKES



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Tord Andersson

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## ABSTRACT

### MERCURY AND RADIOCESIUM IN SWEDISH LAKES

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This study was initiated to create a national picture of the problems with high contents of mercury (Hg) and radiocesium ( $^{137}\text{Cs}$ ) in fish, and to investigate and determine the relationship between the content in fish and the load of the elements with respect to different abiotic lake characteristics. The work is partly based on data from lakes that were studied in connection to an evaluation of different measures (such as lake liming) to decrease the content of Hg and  $^{137}\text{Cs}$  in fish.

About 10000 Swedish lakes were calculated to have a mean Hg content in 1-kg pike (FHg) above 1 mg kg<sup>-1</sup> (wet weight) in the end of 1980's. The cumulated domestic Hg- sources of emission make the largest contribution to the presently high mercury levels in pike and particularly so in central and northern Sweden. The second most important cause is acidification and thirdly Hg emissions from European sources. The content of  $^{137}\text{Cs}$  in fish normalized to 100 g perch (FCs) was above the limit for commercial sale, 1500 Bq kg<sup>-1</sup>, in about 14000 Swedish lakes during autumn of 1987.

An empirical model including Chernobyl fallout, hydraulic residence time and ionic strength explained almost 60 % of the inter-lake variation in FCs. At the same level of fallout, this difference in lake sensitivity, gave a tenfold difference in the initial transfer from fallout to small perch. A significant relationship was demonstrated between the lake dose of  $^{137}\text{Cs}$  and the content in fish. No such clear relationship existed for Hg due to the much more complex chemical and biological behaviour of Hg, where especially factors affecting methylation and food web structure seems crucial.

The higher sedimentation of  $^{137}\text{Cs}$  in lakes with higher concentration of major base cations was due to higher particle sedimentation rates and higher  $K_d$  values in these lakes. However, the distribution and sedimentation coefficients for  $^{137}\text{Cs}$  was not notably affected of the increased mean concentration of major base cations after liming and potash addition. It is suggested that a likely causal factor rather would be the amount and nature of scavenging agents (possibly clay minerals), which in these lakes was well indicated by the natural concentration of base cations in the water.

In general, the remedial measures gave the intended water chemical response with substantially increased mean values of alkalinity, hardness and pH. Two years after the start of the remedies, the Hg concentration in small perch (Hg-pe) was reduced by about 30% on average. No obvious reduction of the Hg content in pike (Hg-pi) was obtained in limed lakes during that time but the high correlation between Hg-pi and Hg-pe indicated that a reduction of the Hg content in pike could be expected, but with a further time delay of about 2 years. None of the methods applied gave any rapid and clear reduction in the concentrations of  $^{137}\text{Cs}$  in fish.

**Keywords:** Mercury, radiocesium, lakes, fish, load, remedial measures, water chemistry, lake characteristics, sedimentation, retention.

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## ABSTRACT

Two large, nationwide monitoring data sets were compiled and statistically treated in order to create a national picture of the problems with high contents of mercury (Hg) and radiocesium ( $^{137}\text{Cs}$ ) in fish. Beside these two data sets, 75 lakes in four counties (Västernorrland, Gävleborg, Örebro and Kronoberg) were studied in connection to an evaluation of different measures to decrease the content of Hg and  $^{137}\text{Cs}$  in fish. An important objective was to investigate and determine the relationship between the content in fish and the load of the elements and how this relationship was affected by different abiotic lake characteristics. Several alternatives to measure the lake doses of Hg and  $^{137}\text{Cs}$  were evaluated (concentration in different fractions in lake water, in settling particles, and in surface sediments).

About 10000 Swedish lakes were calculated to have a mean Hg content in 1-kg pike (FHg) above  $1 \text{ mg kg}^{-1}$  (wet weight) in the end of 1980's, that is a 5-fold increase compared to the calculated preindustrial mean value. The cumulated domestic Hg- sources of emission make the largest contribution to the presently high mercury levels in pike and particularly so in central and northern Sweden. The second most important cause is acidification and thirdly Hg emissions from European sources. The content of  $^{137}\text{Cs}$  in fish normalized to 100 g perch (FCs) was above the limit for commercial sale,  $1500 \text{ Bq kg}^{-1}$ , in about 14000 Swedish lakes during autumn of 1987.

An empirical model including Chernobyl fallout, hydraulic residence time and ionic strength explained almost 60 % of the inter-lake variation in FCs. At the same level of fallout, this difference in lake sensitivity, gave a tenfold difference in the initial transfer from fallout to small perch. A significant relationship was demonstrated between the lake dose of  $^{137}\text{Cs}$  and the content in fish. No such clear relationship existed for Hg due to the much more complex chemical and biological behaviour of Hg, where especially factors affecting methylation and food web structure seems crucial. Lakes with a low relative sedimentation of Hg did also have a low relative sedimentation of  $^{137}\text{Cs}$  due to differences in particle sedimentation rates between the lakes. The sedimentation rate of radiocesium was well correlated to the natural concentration of major base cations and intercorrelated parameters such as pH, alkalinity and conductivity. The higher scavenging capacity in lakes with higher concentration of major base cations was due to higher particle sedimentation rates and higher  $K_d$  values in these lakes. However, the water chemistry was probably not causal in this respect, despite the high correlation, the distribution and sedimentation coefficients for radiocesium was not notably affected of the increased mean concentration of major base cations after liming and potash addition. It is suggested that a likely causal factor rather would be the amount and nature of scavenging agents (possibly clay minerals), which in these lakes was indicated by the natural concentration of base cations in the water.

In general, the remedial measures gave the intended water chemical response with substantially increased mean values of alkalinity, hardness and pH. Two years after the start of the remedies, the Hg concentration in small perch (Hg-pe) was reduced by about 30% on average. The sedimentation rate of Hg decreased during 1988 and 1989 (i.e. after remedial measures) in contrast to the mean concentration of total Hg in water, thus, the retention decreased. None of the methods applied gave any rapid and clear reduction in the concentrations of  $^{137}\text{Cs}$  in fish, in comparison with lakes where the water chemical or biological conditions not were changed.



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## LIST OF PAPERS

The thesis is based on following papers, which will be referred to in the text by their Roman numerals I-VI.

I. Håkanson, L., T. Andersson and Å. Nilsson, "Mercury in fish in Swedish lakes - Linkages to domestic and European sources of emission." Water, Air and Soil Pollution 50 (1990): 171-191

II. Håkanson, L., T. Andersson and Å. Nilsson. "Radioactive caesium in fish in Swedish lakes 1986-1988 - General pattern related to fallout and lake characteristics." J. Environ. Radioactivity 15 (1992): 207-229.

III. Andersson, T. and L. Håkanson. "Time resolution of mercury dose and lake sensitivity related to mercury content in fish." Water, Air and Soil Pollution 56 (1991): 169-186.

IV. Andersson, T. and L. Håkanson. "Mercury content in lake sediments and suspended matter - temporal variation and relation to water chemistry." In Sediment/Water Interactions, eds. B. T. Hart and P.G. Sly. Kluwer Acad. Publ. Hydrobiologia 235/236 (1992): 685-696.

V. Håkanson, L. and T. Andersson. "Remedial measures against radioactive Caesium in Swedish lake fish after Chernobyl." Aquatic Sciences 54 (2 1992): 141-164.

VI. Andersson, T. "Influence of abiotic lake characteristics on the distribution of Hg and  $^{137}\text{Cs}$  within lakes - Implications for the content in fish." Manuscript



# 1 Introduction

One basis for this thesis is large nationwide monitoring data sets on mercury and radiocesium contents in fish, that were compiled along with data on geographical location, regional loading measures, lake water chemistry and lake morphometry (Paper I & II). These large data sets were used to create a picture of the problems with high contents of mercury and radiocesium in fish in a national perspective. An important objective was to determine the link between emission/deposition patterns and the biological impact (the contaminant content in fish).

Another large part of the results presented in this thesis was generated within the project "Liming-Mercury-Cesium", a project where extensive field experiments were conducted in 75 lakes in order to evaluate different measures (e.g. lake liming) to decrease the content of mercury and radiocesium in fish (Andersson et al., 1991; Håkanson et al., 1991 and Paper V). The lakes studied are situated within the counties of Kronoberg, Örebro, Gävleborg and Västernorrland. The project was initiated in 1986 and originally concerned studies of mercury, but in late April - early May 1986 large areas of Sweden were afflicted by fallout from the Chernobyl nuclear accident. The counties of Västernorrland and Gävleborg received a major fallout which, for most lakes within this region, ranged between 3000 and 70000 Bq m<sup>-2</sup>. During the summer of 1986, high levels of radiocesium in freshwater fish were reported from this region (Håkanson et al., 1988), far above the guideline (1500 Bq kg<sup>-1</sup> wet weight) for commercial marketing of fish recommended by the National Food Administration. These consequences of the Chernobyl accident, together with the mass media debate, implied a major economic setback for those parts of the tourist industry based on sport fishing, and caused serious problems for, e.g. fishery management and the local population who used lake fish as part of their basic diet (Bengtsson & Hansson, 1990). Consequently, it was urgent to find a possible way to reduce the activities of radiocesium in fish as well, and the sampling schedule and planned remedies within the ongoing project "Liming-Mercury" seemed well fitted to include investigations of <sup>137</sup>Cs.

The main objective of this thesis was to study and quantify the relationship between the concentration of each respective element in fish and element loadings with respect to lake sensitivity (as regards abiotic factors) according to the concept of "effect-dose-sensitivity" with different spatial and temporal resolution of the parameters used. This concept has also been compared to the retention and distribution of the elements within lakes.

## 2 General background

### 2.1 Mercury

Mercury (Hg) is generally regarded as the most toxic among the heavy metals; no other inorganic contaminant has caused fish consumers so many problems, and Hg is not known to be essential for any metabolic process (Moore, 1990). Mercury poisoning has for centuries been known as a hazard to Hg miners ("mad hatters") and other occupations exposed to Hg vapour,

but the health risks in connection with consumption of mercury contaminated fish was first discovered in the 1950's after the well known catastrophe in Minamata, Japan (WHO, 1976). Several hundred people suffered severe symptoms of brain damage and dozens of people died after consumption of fish from Minamata Bay, where a factory producing acetaldehyde used Hg as a catalyst and released large amounts of both inorganic Hg and methylmercury to the water. Methylmercury is the most toxic among the different Hg compounds and especially the foetus is sensitive to MeHg exposure, which makes pregnant women a special risk group.

Swedish environmental problems caused by Hg were also detected during the 1950's, but here in connection with the use of Hg in seed fungicides which could be related to a decline in the bird fauna. During the 1960's high concentrations of Hg were detected in several fauna species, especially predators. At this time, high levels of Hg in fish were also recorded in several Swedish water areas (e.g., Vänern) receiving direct emissions of Hg to water, mainly from pulp and paper industries (Lindqvist et al., 1984; Lindqvist et al., 1991) As a result of these discoveries, more research on environmental Hg was initiated in Sweden during the late 1960's and early 1970's, and more important, alkylmercury was banned as a seed disinfectant and the industrial emissions of Hg to air and water were markedly reduced. These actions resulted in a clear decrease of the Hg content in fish in the affected waters within only a few years after direct emissions to the water had ceased (Andersson et al., 1987; Ohlin, 1984). However, during the 1970's and early 1980's, several reports indicated that lakes far from any known source of emission still had very high levels of Hg in fish (Lindqvist, et al., 1984). These findings, and similar observations in other parts of the northern hemisphere, i.e Finland (Verta et al., 1986), Canada (Suns et al., 1980; Wren & MacCrimmon, 1983) and United States (Akielaszek & Haines, 1981) resulted in two large projects on the behaviour of mercury in the environment and evaluation of remedies being initiated in Sweden during the mid-1980's.

### **2.1.1 Environmental behaviour**

The mercury chemistry in natural environments is very complex as Hg occurs both in different phases and oxidation states and in inorganic as well as organic compounds. Another complicating factor are the normally very low concentrations of Hg in natural systems, e.g. in remote fresh waters, which make the sampling and analytical procedures extremely important (Nelson & Campbell, 1991).

Most of the atmospheric Hg is made up of gaseous, elemental Hg. In precipitation, however, oxidized forms dominate and more than half of the total Hg is associated to particles ( $>0.4 \mu\text{m}$ ). It has been suggested that emissions of acidifying pollutants ( $\text{NO}_x$  and  $\text{SO}_2$ ) and oxidants like ozone ( $\text{O}_3$ ) would increase the wet deposition of mercury (Iverfeldt, 1991b; Lindqvist et al., 1991). The relative magnitude of dry deposition is not completely known but the relative importance increases in forested areas and shares as high as 50% of the total deposition have been reported (Iverfeldt, 1991a). A higher share of dry deposited Hg has also been reported from the northernmost parts of Scandinavia (Norway) (Iverfeldt, 1991b; Jensen & Jensen, 1991; Steinnes & Andersson, 1991). The present wet deposition of Hg in Scandinavia ranges between 4 and 40  $\text{g km}^2 \text{ yr}^{-1}$ , and the range when also including dry deposition would be about 10 to 50  $\text{g km}^2 \text{ yr}^{-1}$  (Lindqvist, et al., 1991). The deposition of Hg decreases along a south to north gradient both due to a generally decreasing precipitation and lower Hg concentration in

this direction. This general pattern is also similar to the pattern of sulfur deposition. The former deposition pattern which must also be considered due to the continuous leakage of Hg from forest soils to surface waters and, thus, also the implications for the mercury loadings to lakes (Borg & Johansson, 1989; Paper VI) and the mercury content in fish (Paper I), was somewhat different. In addition to the south to north gradient there was also an increased deposition in certain areas due to domestic large point sources. This geographical pattern of deposition is reflected in the Hg content in the uppermost forest soil horizon (the mor layer) (Lindqvist, et al., 1991; Nilsson et al., 1989) and in mosses (Lindqvist, et al., 1991). Peat bog records show that the Hg deposition in SW Scandinavia has increased with a factor 3 during the last century, while some other stations show very small changes (Jensen & Jensen, 1991). However, the records seem to be too few to draw any conclusions about regional differences in this respect.

It is well documented that mercury has a strong affinity to organic matter (Andersson, 1979; Kerndorff & Schnitzer, 1980; Lodenius et al., 1985) and most of the deposited mercury is complexed by the organic-rich material in the top soil layer. Several functional groups of the humic material are possible as complex binders of Hg, e.g. -COOH, -OH, -NH<sub>2</sub> and -RSH (Lindqvist, et al., 1991). Titration experiments with Hg and organic matter from bog water show that the complexation behaviour is comparable to that found for low molecular weight carboxylic acids such as oxalic acid and phthalic acid (Lövgren, 1990). Methylmercury (MeHg), which usually makes up <2% of the total mercury content in soils, could be expected to bind to the thiol groups (-RSH) (Lindqvist et al., 1991). It has been suggested that Hg to a higher degree would be associated with higher molecular weight fraction (Lodenius, 1987) of humics, while MeHg would prefer lower molecular weight fractions (Lee & Iverfeldt, 1991). Thus, it is logical that the transport of Hg (Mierle & Ingram, 1991) and MeHg (Lee & Iverfeldt, 1991) to surface waters has been shown to be closely connected to the loss of humic substances from the surroundings.

The cycling of Hg within natural lakes and Hg uptake in fish is governed by an array of interlocking and/or co-varying factors, the relative importance of which may differ from lake to lake. It is not possible here to relate all mechanisms that have been shown or hypothesized to affect the mercury content in lake fish, and it would be a fallacy to suggest that one mechanism can fully explain the differences in Hg uptake and accumulation in fish (Richman et al., 1988).

However, the methylation of Hg is, of course, a very important process for the Hg content in fish, as MeHg is the major form of Hg that accumulates in fish. It was early found that the rate of biological methylation was well correlated with the overall microbiological activity in the sediment (Jensen & Jernelöv, 1969). The mechanisms of methylation and demethylation have been the subject of many studies ever since, but there is still a lack of knowledge on the environmental significance of different mechanisms and how the distribution among bacterial species differ among ecosystem components (Gilmour & Henry, 1991). Sulfate-reducing bacteria appear to be the major contributors to MeHg production in lacustrine sediments, but the major primary methylators in the water column have not yet been identified, and the specific methylation mechanism by bacteria remains unknown. However, a number of factors that affect bacterial methylation and demethylation activity have been outlined; O<sub>2</sub>, temperature, pH, DOC and sulfate, of which pH and sulfate seem to contribute most to increased net methylation rates in acid-impacted aquatic ecosystems (Bloom et al., 1991; Gilmour & Henry, 1991). Another

aspect is the relative contribution of MeHg to the lakes by precipitation (Bloom & Watras, 1989) and from catchments. Measurements in Swedish forest streams have revealed fairly high concentrations of MeHg (0.04-0.64 ng l<sup>-1</sup>, Lee & Iverfeldt, 1991), which would make a very significant input of MeHg to the lakes. The uptake of Hg in fish can both occur from the food and directly from water via gills. Based on laboratory studies some authors have earlier suggested that gill uptake would be the dominant pathway (e.g. Huckabee et al., 1979) while others have advocated dietary uptake (e.g. Jernelöv & Lann, 1971). There is now considerable empirical evidence of the importance of the dietary mode of Hg uptake, Spry & Wiener (1991) expect that direct uptake accounts for perhaps <10% of the accumulated Hg in fish.

There are numerous reports on the covariation between low pH in lakes and high concentration of Hg in fish and a causal relation between lake acidification and high mercury concentrations in fish has been widely suggested. The topic was reviewed quite recently (Nelson & Campbell, 1991; Richman et al., 1988; Spry & Wiener, 1991) and there are several tentative alternative mechanisms besides the earlier mentioned possible effect on net methylation rate of a lower pH and higher sulfate concentration:

- Due to the covariation between the concentration of Hg and acidifying substances in precipitation, the Hg loading would be higher in lakes with lower pH.
- Acidification of the lake sediments and/or the surrounding soils would mobilize Hg, thus increasing the concentration of dissolved Hg.
- Low pH may favour the production of the non-volatile monomethylmercury instead of volatile dimethylmercury.
- Biological conditions that characterize acidic lakes would be important for the transfer of Hg within the system and favour biomagnification.
- Biota in acidic lakes would be more efficient to bioaccumulate Hg compared to biota in circumneutral lakes and/or more unable to excrete Hg.

Yet another explanation would be a more diffuse and indirectly acting effect of pH, due to the intercorrelation between pH and other parameters such as the concentration of base cations (Spry & Wiener, 1991), where especially Ca<sup>2+</sup> could be expected to depress the biological uptake. Two factors that at least in part are linked to pH is the concentration of allochthonous DOC and the biological productivity of the lakes. The concentration of allochthonous DOC is well correlated to the Hg concentration in lake water and at the same time inversely related to pH (Meili, 1991c; Paper IV). Indications of the existence of a similar relationship between MeHg, DOC and pH was reported by Lee & Iverfeldt (1991). A higher production tends to dilute the bioavailable Hg, thus leading to lower concentration in biota (Meili, 1991a) if the net production of MeHg is not increased. An important conclusion is that there are probably causal relationships between a low pH and a high Hg concentration in fish, which are not necessarily linked to lake acidification.

Another important water chemical variable studied in some of the project lakes, is the

concentration of selenium (Se), that has been shown to affect the Hg concentration in fish (Paulsson & Lundbergh, 1989), either due to metabolic interactions or by restricting the bioavailability of Hg, possibly by precipitating  $\text{Hg}^{2+}$  as  $\text{HgSe}$  (Björnberg et al., 1988).

## 2.2 Radiocesium

Radiocesium is a totally anthropogenic radionuclide and is either produced in nuclear weapons or in nuclear power generation. The higher amount and the slower physical decay of  $^{137}\text{Cs}$  (half-life=30.64 y) compared to  $^{134}\text{Cs}$  (2.06 y) makes  $^{137}\text{Cs}$  a more important isotope from both ecological and health aspects. The slow physical decay and the relatively large proportion in nuclear fallout has made  $^{137}\text{Cs}$  (possibly together with  $^{90}\text{Sr}$ ) the radioecologically most important and well documented radionuclide. The relation between radioactivity and mass of  $^{137}\text{Cs}$  could be exemplified with the calculated total release of  $^{137}\text{Cs}$  from the Chernobyl accident (Gudiksen et al., 1989); 89 PBq ( $P = 10^{15}$ ), that corresponds to a mass of 26 kg, or another example; the limit for freshwater fish; 1500 Bq  $\text{kg}^{-1}$ , corresponds to a concentration of  $<1 \text{ ng kg}^{-1}$ .

The first large fallout of  $^{137}\text{Cs}$  in Sweden occurred during the 1960's as a result of the nuclear arms tests by the Soviet Union and the U.S.A. The total fallout over Sweden during that period was as much as about 30% of the Swedish Chernobyl fallout. A major difference, however, was the uneven spatial distribution of Chernobyl fallout and the rapid course of deposition compared to the rather evenly distributed and more continuous deposition that occurred during the 1960's. As a result, the areas most afflicted by Chernobyl fallout in Sweden, received about 30 times higher deposition of  $^{137}\text{Cs}$  from Chernobyl compared to the accumulated fallout during the 1960's (Mattsson & Moberg, 1991). This different fallout pattern has also implications for the doses to human individuals, which varied within a much wider range after Chernobyl compared to the bomb fallout. So, even if the average Swede will receive a dose commitment of around 1mSv from Chernobyl (i.e. about the same as from bomb fallout), individuals in high fallout areas with a diet based on game animals and lake fish will receive much higher dose commitments. In this respect it is interesting to note that the total deposition of Cs-137 in Sweden was about 4.25 PBq or almost 5% of the total activity (89 PBq, Gudiksen, et al., 1989) released from the Chernobyl reactor.

### 2.2.1 Environmental behaviour

Radiocesium has been widely used both for sediment dating (Jaakola et al., 1983; Pennington et al., 1973) and as a tracer of particle transport to and within aquatic environments (Campbell et al., 1982; Santchi et al., 1988; Walling, 1989). The particle affinity of  $^{137}\text{Cs}$  has also been used to study the fate of certain metals and other particle-reactive substances (Wong & Moy, 1984) and, consequently, the same factors that affect the distribution of radiocesium may affect other substances as well (Heit & Miller, 1987). Several papers have shown that great attention has to be paid to factors influencing the distribution and behaviour of radiocesium in lakes, i.e. the size fractionation of radiocesium (Salbu et al., 1992) and the sorption of  $^{137}\text{Cs}$  to particles. These factors include, e.g. the content and type of clay minerals (Cremers et al., 1988; Evans et

al., 1983; Heit & Miller, 1987) in suspended matter and sediments, the organic content of the sediments (Longmore et al., 1983) and the concentration in water of  $\text{NH}_4^+ \cdot \text{K}^+$  (Comans et al., 1989; Evans, et al., 1983) and other competing cations.

Radiocesium is readily accumulated by biota, presumably due to the relative chemical similarity with potassium, which is a major component of the osmotic systems in organisms (Hewett & Jefferies, 1978), and radiocesium is also rather evenly distributed within the organism. The concentration of potassium in water is generally thought to determine the uptake of radiocesium in organisms due to the competing effect, and there are several reports on an inverse relation between the potassium concentration in water and the concentration of radiocesium in fish (Kolehmainen, 1972; Preston et al., 1967). Colloidal humus with a high concentration of  $^{137}\text{Cs}$  has also been suggested as an important link in the food chain transfer of radiocesium (Häsänen & Miettinen, 1963). Most studies of natural conditions show that the major uptake of  $^{137}\text{Cs}$  in fish occurs from the food (e.g. Hewett & Jefferies, 1978; Carlsson, 1978).

### 2.3 Why Hg and $^{137}\text{Cs}$ ?

One obvious reason is strictly coincidental and practical. A high fallout of radiocesium occurred in an area where a large-scale sampling and remedial programme had been initiated, and as a similar, or somewhat modified methodology seemed applicable it was practical to use the same samples also for analysis of radiocesium and modify the remedial programme with respect to  $^{137}\text{Cs}$ .

Besides lake acidification, the high levels of Hg and  $^{137}\text{Cs}$  in fish are two of the major environmental problems for lakes in Sweden (Papers I & II). Despite the chemical and biological behaviour of the two elements being different in many aspects they also have some properties in common. The problems are both related to the transfer and possible threat of the elements to humans via fish. It seems also clear that the basic transport routes to fish are similar for mercury and radiocesium, i.e. from atmospheric deposition to lake water with a certain retention within the catchment and the lake itself and with the major uptake via the food web. Both mercury and radiocesium are strongly adsorbed onto suspended matter. This means that the flux of particles within lakes must be accounted for in lake budgets of both elements (Stumm & Morgan, 1981).

The remedial measures tested in our project included both liming and addition of potash. This has made it possible to study if and how the distribution and retention of the elements were changed by the altered chemical conditions, and how and to what extent the retention was related to water chemistry in comparison with parameters being more or less unaffected by the remedies, such as hydraulic residence time, lake morphometry and fundamental particle properties.

The Chernobyl fallout coincided with the snowmelt and a high runoff in this region, which gave rise to a marked pulse of  $^{137}\text{Cs}$  to the lakes that could be followed in different lake compartments and in different biota. This situation makes it also possible to study the decline and the time needed to reach steady state. Mercury, on the other hand, is more or less

continuously transported to the lakes at a rate which, judging from the non-significant change in pike during the last decade in lakes in this region, may be assumed to be nearly constant on a yearly basis. If the Chernobyl fallout could have been postulated, the investigation strategies would have been optimized as regards, e.g., selection of lakes and overall sampling strategy. But, nevertheless, it is my belief that the results of this study should give some insight also in these matters.

### **3 Aims of this thesis**

The main objectives of the work were to:

Quantify the problems with high concentrations of the studied elements in lake fish in terms of number of affected lakes and their geographic distribution.

Study the relation between the concentration of  $^{137}\text{Cs}$  and Hg in fish and the load of the elements, with the load defined by different spatial and temporal resolution.

How the concentration of  $^{137}\text{Cs}$  and Hg in fish and the distribution of  $^{137}\text{Cs}$  and Hg within lakes could be linked to lake water chemistry and other abiotic lake characteristics.

Study the effect of remedial measures on the content in fish and suggest the mechanisms of most importance in this respect.

### **4 Material and methods**

#### **4.1 Lakes**

The national overviews in Papers I and II are based on data from more than 1000 lakes. The mean lake area is almost 5 km<sup>2</sup> but most of the lakes are smaller than 1 km<sup>2</sup>. The studied lakes are, however, somewhat larger than the average lake area of the more than 80000 Swedish lakes (Monitor, 1986). Especially the lakes in Paper II cover a rather wide range of lake types; high mountain clearwater lakes, low altitude lakes in agricultural areas, but most commonly lakes similar to the more extensively studied lakes (Papers III - VI), which are situated in the counties of Kronoberg and Örebro and along the east, central part of Sweden in the counties of Gävleborg and Västernorrland.

The catchment areas of these small drainage lakes are dominated by coniferous forests with mires as a common element, i.e., a landscape typical of the boreal forest zone. The soils are mostly podzols developed on a glacial till. Some of the lakes closest to the coast of the Bothnian Gulf also have elements of former lake and sea sediments within their catchments. These lakes are situated at the Precambrian shield and the bedrock geology of the areas was mostly classified as Ca-poor and slowly weathered (Nilsson & Håkanson, 1992).

Before any remedial actions were taken, most of the lakes were oligotrophic, with generally low values of pH, alkalinity and major base cations, and a rather high content of humic substances (measured as colour). Higher values on tot-P, pH, alkalinity and major base cations were found in lakes where the catchments had a higher proportion of soils of sedimentary origin (Nilsson & Håkanson, 1992).

## 4.2 Studied parameters

As the results and data used in this work emanate from several works, only a brief description is given here of the overall strategy to represent each lake with a mean value for each variable during a defined period of time and of the most important sources of error. A more detailed description of the analytical methods, sampling routines and information concerning the representativity and statistical reliability of the parameters used is given in each Paper and in Håkanson, Borg, & Uhrberg, 1990.

The concept of effect-dose-sensitivity is meant to be applicable for different substances and different aquatic environments (Håkanson, 1991). In this context, the effect term is given by the mean content of Hg and  $^{137}\text{Cs}$  in fish of each lake during a defined period of time according to the definition that it should be a quantitative value for a defined area and time span. Several alternative parameters to describe the lake dose of the contaminants were defined and measured. These included total concentrations of Hg and  $^{137}\text{Cs}$  in lake water as well as concentrations in operationally defined fractions and particles. More indirect measures of lake dose were given by parameters describing the deposition of Hg and  $^{137}\text{Cs}$  at the lakes and their catchments. All parameters describing lake dose are possible to translate into lake loads of the elements via physical constants. The term lake sensitivity includes a number of factors that may exert a direct or indirect influence on the relation between the lake dose and the content in fish.

In the national overviews (Papers I & II), a minimum of 3 fish individuals on each occasion was required in each lake mean value. In the more well documented project lakes (Papers III - VI) fish samples of mostly 10 to 15 individuals of each species were used to give each lake a representative value for that specific occasion. The pike (ranging around 1 kg) were caught during spring and the perch (< 10 g) during autumn (August to September). All individuals were size determined by length and weight, whereafter muscle samples were prepared and analysed with the same methods as the material from sediment traps. The Hg-content in pike was weight normalized to 1-kg pike by linear regression for each lake. Regarding the concentration of radiocesium of the actual size class (0.5-1.5 kg), any general correlation to size was covered by the individual variation (Paper V). The intra-lake variability in Cs-content for the same species and size was very high during 1986 where, e.g., the coefficient of variation (CV) for small perch ranged around an average of 0.60. The individual variation has successively decreased and the possible error ( $t_{0.95}$ ) in each yearly lake mean value has mostly been <20 % ever since. The individual variation in Hg content was generally lower, giving possible errors < 15% in most lakes.

The soil store of the Hg in the catchments was interpolated from data on the Hg-content in the mor layer of podzols similar to those occurring in the studied lake catchments. The soil concentrations of Hg being converted to quantities per unit area with data on the thickness and



specific weight of the mor layer. Samples were collected from 363 stations fairly evenly distributed over the Precambrian shield zone of the country (see (Lindqvist et al., 1991; Nilsson et al., 1989) for further details). In Paper I, a Hg deposition parameter (EFSUM) was calculated for each lake, EFSUM is based on a function of the totally emitted amount of Hg (1860-1987) from, and the distance to, the 11 largest Swedish point sources.

The atmospheric input and soil store of Chernobyl-Cs was derived from a map based on gamma-measurements of soil radiation from aircraft (distance between each flight line = 5 to 10 km) during the period May to October, 1986 (Swedish Geological Research, 1986), these aerial gamma-measurements were calibrated against measurements in soil samples. Several soil and vegetation factors could affect the measured radiation (Bergman et al., 1991; Edvarson, 1991) and certain interpolation errors are inevitable, so it seems obvious that these values of fallout give a rather crude estimate of real loading. The same also applies to the interpolated values of atmospheric deposition and soil stores, and the possible errors in each lake value are probably of at least the same order of size as those introduced by natural variability.

Lake water was sampled 3 to 6 times per year in the central part of the lakes, during ice-free conditions (April to October) and analyzed for mercury (1986 to 1989) and cesium-137 (1987 to 1989). The water samples used for Hg-analysis were collected at 0.5 m water depth according to a special clean-sampling method (Lindqvist et al., 1991). The water for analysis of radiocesium was collected in 5 dm<sup>3</sup> vessels prepared with CsCl as carrier solution and HNO<sub>3</sub> to avoid adsorption to the walls. The concentration of total-Hg in water was detected after strong oxidation with BrCl and reduction with SnCl<sub>2</sub> (Lindqvist et al., 1991), according to the method described by (Bloom & Crecelius, 1983). The fraction of reactive+non-reactive Hg (RIHg) was obtained after reduction with NaBH<sub>4</sub>, and the reactive fraction (RHg) was obtained after acidification with HCl and reduction with SnCl<sub>2</sub>. The non-reactive fraction (IHg) was computed by difference (IHg = RIHg - RHg). The analytical procedures including a description of the laboratory, equipment, calibration methods and detection limits have been described by Iverfeldt (1988) and Lindqvist, et al. (1991).

The settling material was collected in sediment traps during the summer period (about 100 d) each year from two different levels, 2 m above bottom and 2 m from top of water column in the central deepest parts of each lake. The material was freeze-dried and first analysed for <sup>137</sup>Cs with a Ge/GeLi-detector connected to a multichannel-analyzer (4096 channels), whereafter Hg was detected by an automatic FIA-AAS method after H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub> digestion (Håkanson et al., 1990). Surface sediments were collected from 3 stations at different depths in each lake and analysed for Hg (each year, 1986-89) and <sup>137</sup>Cs (1986 and 1989).

Water samples were collected monthly (1986 to 1989) in the central part of the lakes and analyzed (n=75 lakes) for pH, alkalinity, colour, Total-P, ionic strength (conductivity) and hardness (Ca+Mg). Analyses were also performed on Fe, K, Se and Ca and Mg separately in a subset of lakes. Swedish standard methods were applied, and the replication was continuously checked by intercalibrations (Håkanson, et al., 1990). The different water chemistry parameters were intended not only to show the change due to remedies, but also to give a measure of the different sensitivity of the lakes. Besides water chemistry, the lake sensitivity was also intended

to be deduced from sedimentation rates, fundamental particle characteristics as determined from the concentration of carbon, nitrogen and inorganic fraction in seston and surface sediments. All these lake characteristics have a certain variation in time and space which had to be determined and related to the lake dose and the effect parameters (Papers III & IV). Another group of lake sensitivity parameters with a constant value for each lake include the size and morphometry of all lakes and a broad set of map-derived data describing the catchment areas (Nilsson & Håkanson, 1992).

### 4.3 Remedial strategies

Suitable remedies for each individual lake were selected on a basis of, e.g., the initial water chemistry and trophic status. The strategies used were:

- To change the soil chemical conditions by liming of wetlands, hydrological discharge areas and whole catchments (full-scale liming) in order to possibly reduce the mobility and the transport of the elements into the lakes.
- To increase the scavenging of radiocesium and mercury from the water column to the sediments and thereby prevent or delay the biological uptake (lake liming and addition of selenium).
- Decrease the biological uptake of radiocesium by blocking and/or substitution with potassium and other cations. Potash treatment in combination with different liming measures.
- To change the food web structure and bioproduction of the lakes in a way that would decrease the bioavailable amount of the elements in the lakes. These measures include intensive fishing to decrease predation pressure and addition of nutrients.
- To decrease the biological bioavailability of Hg, by increased pH and interrelated parameters and addition of selenium.

The remedies and the number of lakes tested with each remedy are summarized in Table 1. The remedies were initialized during 1987 or at the latest in February 1988.

Table 1. Summary of introduced remedies and number of lakes tested with each remedy.

Method	Further measures	Number of lakes
Lake liming		18
Lake liming	Nutrient addition	3
Lake liming	Potash	6
Lake liming	Selenium	5
Lake liming	Intensive fishing	5
Wet land liming		16
Wet land liming	Potash	7
Wet land liming	Selenium	6
Catchment liming		4
Intensive fishing		5

## 5 Results and discussion

### 5.1 Geographical distribution

#### 5.1.1 Mercury

Based on samples of pike from 894 lakes during the period 1981-1988, it was estimated (Paper I) that about 10000 Swedish lakes had lake mean Hg concentration in 1-kg pike (FHg) above 1 mg kg<sup>-1</sup> (wet weight). Figure 1 illustrates the geographical variation of FHg in Sweden. The geographical distribution of areas with high FHg values could to a certain degree be linked to a loading pattern that primarily is determined by the cumulated atmospheric Hg emissions from domestic large point sources. However, high FHg mean values were also found in certain areas with few or without known major point sources, e.g., along the west coast and in the Bergslagen area. This was suggested to be due to, either, the proximity to large European industrial areas, or, as regards the Bergslagen area, several small old discharges in connection with the working of ore. Some of the elevated FHg values could be due to diffuse emissions related to population density. Only a weak positive correlation between FHg and the Hg content in the top soil horizon (the mor layer) was observed, which indicates that the soil store of Hg in a major part of Sweden is high enough to obtain high FHg values when combined with other factors such as loss rate, present deposition and lake sensitivity. The concentration and content of selenium (Se) in the mor layer was also measured, but there was a slightly positive correlation between the concentration of Se in the mor layer and FHg. Thus, nothing indicated that a high Se content in the mor layer would reduce the mobility of Hg from soils or contribute to a lower bioavailability of Hg in lakes.

A statistical evaluation showed that parameters describing lake sensitivity were just as important or even more important than the cumulated mercury loading. The parameters of most importance were pH and water colour which was used as a measure of the concentration of humic substances. A fairly good agreement was obtained between the empirical and calculated geographical distribution based on statistical models with a load factor related to the cumulated Hg emissions (EFSUM), pH and colour as input parameters. The absolute deviation between the modelled map and the empirical geographical distribution was < 0.2 mg kg<sup>-1</sup> in the major part of Sweden. The statistical models tended to overestimate the mercury content in fish close to the point emissions, which could be interpreted as an effect of the decreased emissions and direct deposition during the last decade in the immediate vicinity of point sources.

Based on the relationship between the mercury content in pike, EFSUM and lake mean pH it was possible to calculate a Swedish average background or preindustrial value of FHg to 0.22 mg kg<sup>-1</sup>. This value is close to other estimates, based on other approaches (Lindqvist et al., 1984; Verta et al., 1986). This implies that about 10000 Swedish lakes have a 5-fold increase of the mean mercury content in pike. By applying a somewhat modified acidification model (Monitor, 1986), it was possible to differentiate between the effects of acidification and domestic and European Hg emissions. Figure 2 gives the distribution of responsibility in this respect for southern, central and northern Sweden. In this context it should be noted that there are many possible sources of error both in the FHg-model and in the acidification model. However, the cumulated domestic Hg- sources of emission makes the largest contribution to

the present high mercury levels in pike all over the country (> 50% of the rise) and particularly so in central and northern Sweden. The second most important cause is the acidification due to sulfur emissions from Swedish and European sources (35 to 40%) and thirdly the Hg emissions from European sources (10 to 15 %). In southern Sweden, the discharges of Hg from the continent in combination with acidifying precipitation seem to have caused an increase of more than 0.3 mg kg<sup>-1</sup>. In northern Sweden, the foreign Hg-emissions have not had any major impact on the average concentration, but the contribution from continental sources has increased during recent years (Lindqvist et al., 1991).

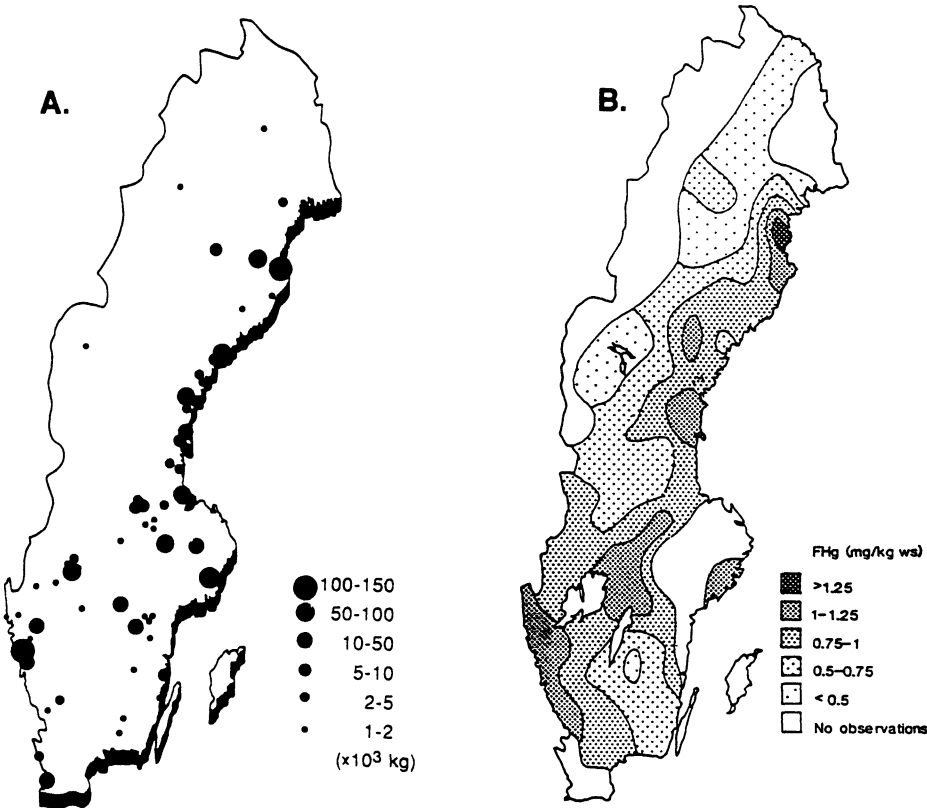


Figure 1. Maps showing: (A) accumulated Hg emissions during 1860 to 1987 larger than 1000 kg; (B) the mean Hg concentration in 1-kg pike 1980 to 1988.

Catchment budgets show that Hg input to soils from precipitation still exceeds the loss from forest soils(Aastrup et al., 1991; Borg & Johansson, 1989; Lindqvist et al., 1991), and in Paper VI it was shown that the terrestrially mediated input of Hg, on annual basis, dominated over direct atmospheric deposition in all studied lakes in the counties of Västernorrland and Gävleborg. The rather small differences between the catchments in hydrological conditions, atmospheric deposition and soil storage of mercury, resulted in these factors being of minor

importance compared to the ratio of drainage area to lake area (DA:LA), when determining the total load of Hg entering a lake (Paper VI). These results indicate that high concentrations of Hg in lake fish will remain a problem far into the next century. As a considerable reduction of Swedish Hg emissions has occurred during the last 15 years there will be a geographical

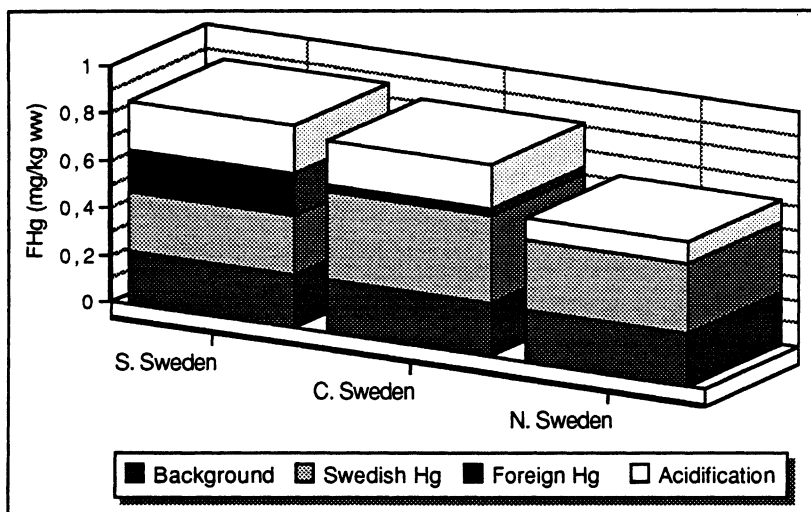


Figure 2. Histogram showing the average background concentration in 1-kg pike and the contributions from (1) Swedish atmospheric Hg emissions, (2) European Hg emissions and (3) Sulfur emissions from Sweden and other European countries, calculated for southern, central and northern Sweden.

redistribution of the "burden of guilt" from Swedish to European sources of atmospheric Hg emissions if no measures are taken, but no improvement in the lakes will take place if the total Hg emissions are not strongly decreased.

The question of differing retention times within the drainage areas are of vital importance when determining the time lag between reduced atmospheric emissions/input and any notable decrease in lake water and in fish. Despite the presumed lower mobility of Hg in soils, there were rather small differences between the leakage rates of Hg and  $^{137}\text{Cs}$  (1986 excluded) from the drainage areas, as related to the pool in the uppermost forest soil horizon (Paper VI). The most plausible explanation seems to be the continuous atmospheric deposition of Hg on the drainage areas. The average leakage rate of Hg could then be regarded as composed of a small leakage rate from the soil store covering a large part of the catchment and larger rates from smaller areas of discharge which are continuously "fed" by atmospheric deposition. Such a relationship was reported for  $^{137}\text{Cs}$  within a small catchment in Northern Sweden during 1986 (Bergman, et al., 1991) when the leakage rate from a discharge area (an open peat area close to a brook) was about ten times higher than the average yearly loss rate from the entire catchment. Due to the rapid fallout on snow cover this value seems to be an extreme, but a variation of  $\alpha_{\text{Hg}}$  within one order of magnitude depending on different hydrological conditions within the same catchment

does not seem unlikely. A parallel could also be drawn with the suggested (Bishop, 1991) capacity of the organic-rich, superficial soils and spate-specific flow pathways to "finger-print" the runoff water with high acidity and TOC (based on results from the same catchment as Bergman, et al., 1991 investigated). This type of spatial variation within a catchment is also associated with certain runoff events, that were impossible to account for in this study.

### **5.1.2 Radiocesium**

Paper II reports the results from 644 Swedish lakes. A map showing the content of  $^{137}\text{Cs}$  in fish normalized to 100 g perch (FCs) during the autumn of 1987 was constructed. To obtain the highest possible geographical resolution, and due to the large temporal, species- and size-dependent variation in activity the fish data had to be normalized to a "unit fish" caught during a defined period of time. As perch around 100 g were the most frequent, it was decided to normalize all data on  $^{137}\text{Cs}$  content in fish to 100 g perch (FCs), using empirical inter-species relationships found in lakes where data on several species existed. The chosen time, autumn 1987, seemed appropriate from two aspects. Firstly, a lot of data was available from this time and secondly, the conditions within the lakes had been stabilized so the variation between individuals were smaller which made the inter-lake as well as the inter-species relationships more reliable. The "impact" map (Figure 3) showed good agreement with the fallout map as derived from aerial measurements. In most areas with a fallout higher than  $10000 \text{ Bq m}^{-2}$ , FCs were above  $3000 \text{ Bq kg}^{-1}$  wet weight in the average lake. In the southern part of the fallout area, the maps indicate that the concentration of  $^{137}\text{Cs}$  in fish is generally lower than in the northern part at a similar level of fallout, and that the area with highest activities in fish did receive a large fallout, but not the largest. Thus, different lake types appeared to have different efficient transfer from fallout to fish.

It was calculated that about 14000 lakes had FCs above the limit for commercial sale,  $1500 \text{ Bq kg}^{-1}$ , during autumn 1987. A statistical model including Chernobyl fallout, hydraulic residence time and ionic strength could explain almost 60 % of the inter-lake variation in FCs. At the same level of fallout, this difference in lake sensitivity, gave a tenfold difference in the initial transfer from fallout to small perch. The empirical model made it possible to incorporate lakes with known data on fallout and ionic strength to derive a new interpolated map based on lakes with a more even geographical distribution which minimizes the error in the position of the interpolated isolines (Paper II).

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Figure 3. Maps showing; (A) calculated deposition of  $^{137}\text{Cs}$  in  $\text{kBq m}^{-2}$ , from Persson et al. (1987) ; (B) the average content of  $^{137}\text{Cs}$  in fish normalized to "100 g perch" during autumn of 1987.

## **5.2 Temporal variation and inter-species relations**

### **5.2.1 Mercury**

The Hg concentration in fish could show both a seasonal and a yearly variation. Seasonal changes appear to be related to temperature and metabolic processes rather than to changes in Hg concentration in the water or in the diet. In pike these seasonal changes seem to be related to sex. The concentrations are similar in both sexes during spring, but the concentrations in females could decrease by almost 30 % during summer while the concentration in males remains fairly constant (Meili, 1991c). The inter-year variation for Hg in pike is usually of minor importance in lakes that are not affected by direct Hg discharges if the sex-related seasonal variation is accounted for. This is illustrated in Figure 4, showing the lake mean Hg concentration in 1-kg pike in 63 lakes during the springs of 1987 and 1988; only a few lakes have Hg-pi values that differ by more than 20% and there is no difference in the overall mean value. The mercury levels in Swedish pike seem to have been rather constant during the 1980's, and there is no tendency for decreased levels despite the reduced Hg-emissions in Sweden (Håkanson et al., 1988). Locally, there could be deviations from this general picture,

i.e., increased levels due to increased acidification, increased Hg loadings due to damming or possibly ditching of surrounding land, or decreased levels in certain lakes with formerly high direct deposition from old point sources or in lakes and drainage areas where extensive and persistent liming has occurred.

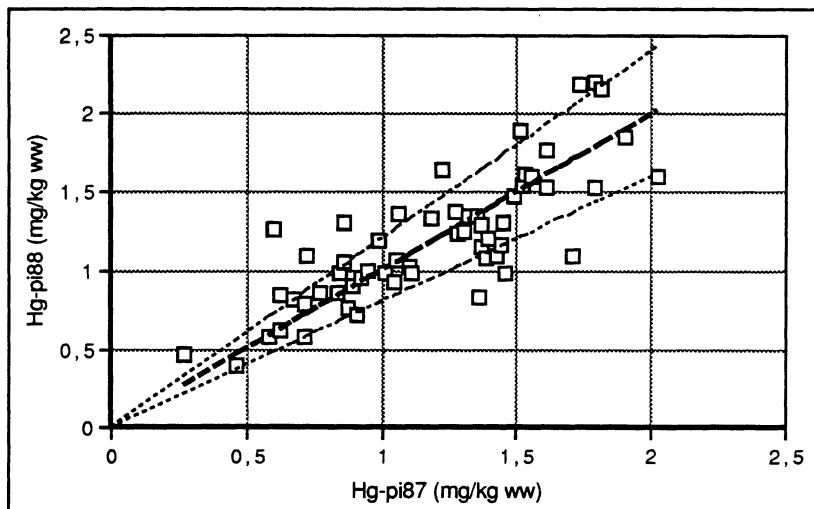


Figure 4. The mean Hg content in 1-kg pike during the springs of 1987 and 1988,  $n = 63$  lakes. The thin dashed lines gives the  $\pm 20\%$  deviation from the values of 1987.

In Paper III, it was shown that there was a rather high correlation between the lake mean Hg concentration in small perch ( $<10$  g) and in 1-kg pike and that the ratio (Hg-pi:Hg-pe) ranged around 7. These results are consistent with the findings by Meili, 1991 that the biomagnification of methylmercury could be characterized by a fairly constant enrichment factor between two trophic levels, here the predator (pike) and prey (perch). It is, however, important to note that there is a considerable variation around the average ratio (range = 4 to 10). Such a variation could, partly, be explained statistically due to the relatively large possible error in the mean value of each lake. To some extent, there seems to be a regional variation in this ratio or enrichment factor. Lakes in the county of Västernorrland generally have a higher, and lakes in Örebro have a lower, average ratio. This variation could be attributed to differences in the trophic structure of the individual lakes. Another possible explanation of such a regional difference could be that the Hg content in small perch shows a larger inter-year variation and, thus, expresses a quicker response to temporary regional differences in the Hg loading/bioavailability.

### 5.2.2 Radiocesium

As an effect of the rapid and discontinuous fallout of radiocesium in connection with the spring flow, the lakes received a pulse of  $^{137}\text{Cs}$  which was transferred along the food web of the lake ecosystem, and there was a considerable temporal variation in the radiocesium content among and between species (Papers II & V). At any time ( $t$ ) the transfer from fallout ( $\text{Cs}_{\text{soil}}$ ) to fish



(Cs<sub>fish</sub>(t)) can be described by a lake specific ratio or transfer coefficient (TC(t)) where:

$$TC(t) = Cs_{\text{fish}}(t) / Cs_{\text{soil}} \quad (m^2 \text{ kg}^{-1}) \quad [1]$$

The magnitude of this transfer coefficient was largely dependent on the actual fish species and time after fallout. This is illustrated in Figure 5, where the variation of the transfer coefficient with time is shown for small perch, pike, brown trout and Arctic charr as compiled from data from a large number of Nordic lakes. The most striking feature of Figure 5 is perhaps the wide range of transfer coefficients in different lakes for the same species and size category at a particular time. This large inter-lake variation, of course, gives rise to a wide variety of possible lake specific curves for a certain species. The different temporal development of the transfer in different lakes is illustrated with curves for pike in lakes Lövsjön and Hamstasjön and for trout in Storsjön and Grundvattnet respectively. Another measure on the spread around the mean TC(t) is given in Figure 5, by the yearly quartile values (25% and 75% , vertical lines) for small perch and pike in the most well-documented lake population (n=41 lakes).

Another conclusion that can be derived from Papers II & V and Figure 5 which includes updates from 1988 is that the maximum transfer was reached within the first three years for all species in most lakes and normally in the order: small perch - trout and charr - larger perch - pike. This sequence seems to reflect the trophic level of each species and is in accordance with the idea that the uptake of <sup>137</sup>Cs in fish occurs mainly from the food (Hewett & Jefferies, 1978) and that the activity in the predator is determined by the prey activity. Thus, the fish-feeding pike is the species with the most extended temporal development and is also the species with the highest values for the total time integrated transfer (F, see below). However, differences in the metabolic and feeding rates between different species should also be considered, e.g., within the same lake and at the same activity in their diet, small perch were observed to reach about twice as high activities as roach during 1986 (Meili, 1991b). After the maximum value on the transfer (TC<sub>(t0)</sub>) was reached, the decline can be fairly well described by an exponentially decreasing function; Cs<sub>fish</sub>(t) = Cs<sub>fish</sub>(t=0)\*e<sup>-kt</sup>, from which the apparent or ecological half-life (TE) for each lake can be derived as; TE = ln (2) / k. This makes it possible to extrapolate the future transfer coefficients and calculate the total, time-integrated value of the total expected transfer given by the transfer factor, F, as:

$$F = F(t_0) + TC_{(t_0)} * TE / \ln(2) \quad [2]$$

where F(t<sub>0</sub>) is the time integrated transfer until that the maximum transfer coefficient was reached. Planktivores, like small perch, reached their maximum transfer coefficient within a few months after the fallout and consequently F(t<sub>0</sub>) becomes small compared to the total transfer for these fish categories. From Eqn. 2 it is also obvious that the magnitude of F is determined both by the magnitude of the initial transfer and by the rate of decrease as expressed in TE.

An increase of TE with time due to an increased impact of factors controlling the secondary loading (like resuspension, Papers V & VI) is possible. Such an impact could lead to annual or seasonal fluctuations and would tend to increase TE to approach the physical halflife. However, in small perch (which in this data set have been showing a decreasing concentration of radiocesium for the longest time, > 6 years), one might see a tendency for an increase of TE

during the last 3 years compared to the figures given in Paper V ( $0.6 < TE < 2.1$  years), but no dramatic prolongation has occurred that significantly affects the calculated expected total transfer in Figure 6. From the same data it can also be noted that the future part of F from 1992, generally is rather small also when considering possible extrapolation errors. The exception is larger piscivorous species such as pike in certain lake types (see below), where up to 50 % of the expected total transfer may still remain.

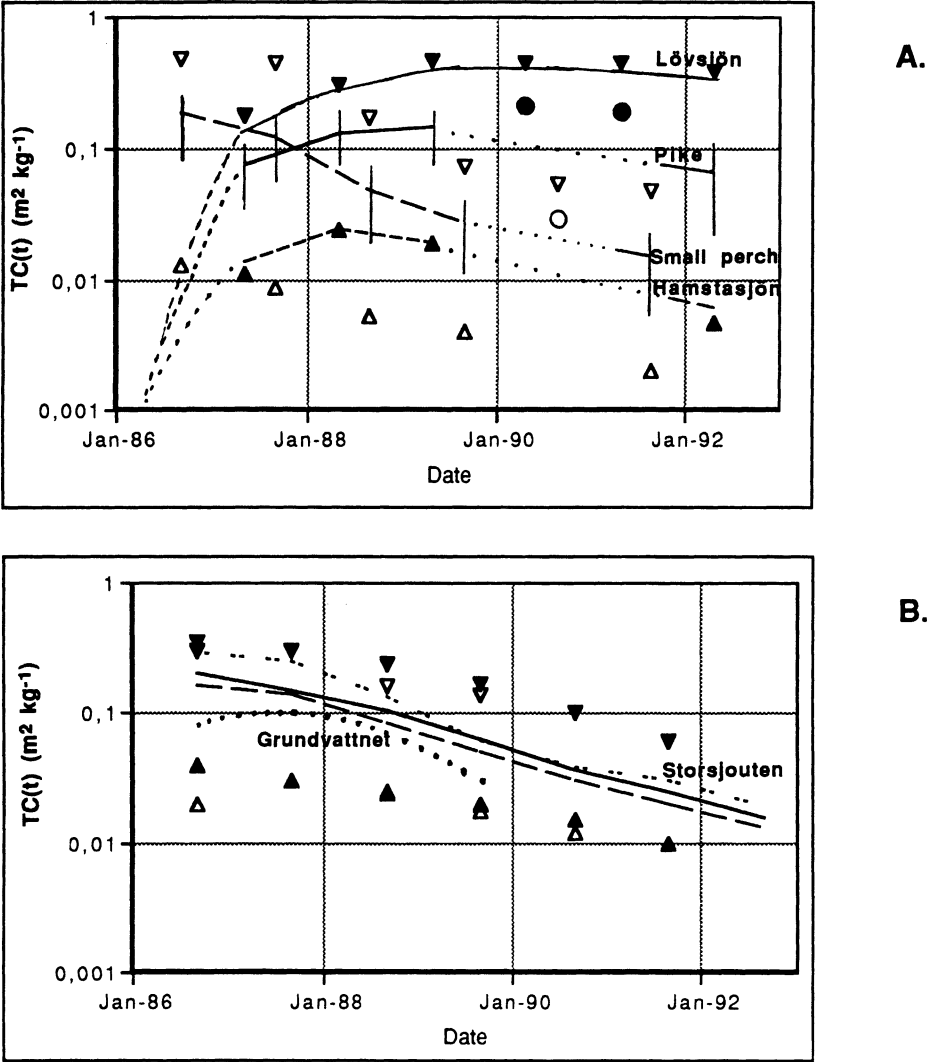


Figure 5. Ranges (arrows) and mean values (lines) for the time dependent transfer coefficient  $TC(t)$  between the fallout and the lake mean concentration of  $^{137}\text{Cs}$  in fish, for (A) small perch,  $<10$  g (unfilled arrows) and pike (filled) and (B) Arctic charr (unfilled, dashed line) and Brown trout (filled, full line), based on data from 230 Nordic lakes. Examples of lakes with a low and high total transfer to pike (A) and to trout (B), respectively.

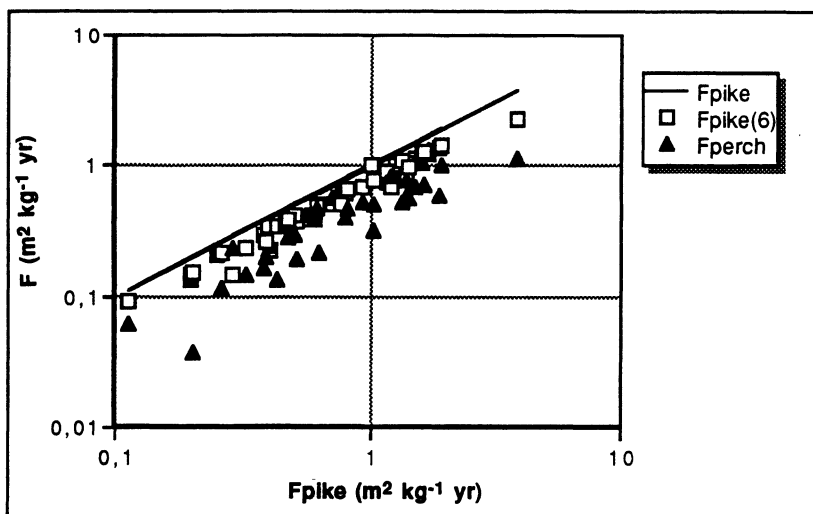


Figure 6. Relationship between the total expected transfer of Chernobyl fallout ( $F$ ) to pike and small perch ( $n=44$  lakes),  $F_{\text{pike}}(6)$  = the time integrated transfer to pike after 6 years.

There exists a very significant inter-lake relationship between the total transfer ( $F$ ) to perch and to pike, as shown in Figure 6, i.e., lakes with a high total transfer to small perch have also a high total transfer to pike. There is not enough data available for brown trout and Arctic charr to make any similar comparisons between  $F$  in different lakes, but the high correlation during autumn 1987 (Paper II,  $r^2=0.68$ ,  $n = 12$  lakes) suggests that there is a similar, but somewhat weaker, relation between the total expected transfer ( $F$ ) to trout and charr as between  $F_{\text{pike}}$  and  $F_{\text{perch}}$ .

### 5.3 Remedial measures

The remedies have generally given the intended water-chemical response. The potash treatment increased the long-term mean concentration of potassium in water to  $>20 \mu\text{eq l}^{-1}$  in most lakes compared to  $<10 \mu\text{eq l}^{-1}$  on average under natural conditions (Paper V). Mean alkalinity was increased from 0.05 to  $0.19 \text{ meq l}^{-1}$  ( $n=75$  lakes) and mean pH from 6.0 to 6.5 as an effect of the liming treatments, but the seasonal variation is considerable also after treatment, which emphasizes the importance of a frequent sampling for establishing lake mean values (Paper III). The smallest increases of long-term means were obtained, as expected, in lakes with short hydraulic residence time.

The median decrease until 1989 (i.e. two years after the remedies were initialized) for the Hg concentration in small perch was 33%. No obvious reduction of the Hg content in pike was

obtained in limed lakes during that time (Paper III, Håkanson et al., 1990). The Hg content in pike during 1989 (Hg-pi<sub>89</sub>) showed the highest correlation to an average of Hg-pe<sub>10</sub> and Hg-pe<sub>88</sub> with a best fitting equation very similar to the equation that gave the best fit for the period before remedies. This indicated that a reduction of the Hg content in pike also could be expected, but with a further time delay of about 2 years. Such a time delay is confirmed (at least partly, the mean decrease is smaller in pike) by preliminary data on the Hg concentration in pike from 1991 and 1992.

In lakes (n=11) where selenium was added, the mercury levels in small perch were decreased by 75% on average after 2 years. However, it is too early to recommend the method from an ecological point of view, selenium is toxic in rather low concentrations and a sudden decrease in the reproduction of perch was noted in 4 out of 11 lakes. None of the methods applied gave any rapid and clear reduction in the concentrations of <sup>137</sup>Cs in fish, in comparison with lakes where the water chemical or biological conditions not were changed (Paper V).

## **5.4 Lake dose and lake sensitivity vs content in fish**

### **5.4.1 Mercury**

The major cause of the problem with high Hg-levels in fish in Swedish lakes is the past and present large scale Hg-loading, which in part could explain the geographical pattern with a higher frequency of lakes with high Hg-levels in certain areas (5.1.1). In order to explain the average Hg content in fish in a certain lake these loading measures are not applicable. Several alternatives to measure the lake doses were evaluated in 25 lakes during the period 1986 to 1989, which included conditions both before and after lake remedies. The concentration of Hg was measured in different operationally defined fractions in lake water (4.2), in settling particles, and in surficial sediments.

The correlation of the concentration of Hg in different fractions in water and seston to the Hg concentration in fish was positive (Håkanson, et al., 1991; Papers III & VI), but rather weak ( $r^2 < 0.25$ , n=25 lakes). In Paper III, it was demonstrated that there was a correlation between these lake dose parameters and the Hg concentration in small perch also after the chemical conditions had been altered by remedies. The lake dose estimates should preferentially be mean values for a period that covers two summer periods in order to obtain the most relevant dose measure with respect to small perch (Paper III). A problem with all predictions that include Hg in lake water as a parameter is the low concentrations in combination with a considerable natural temporal variation, which makes it expensive and difficult to establish reliable mean values (Nelson & Cambell, 1991; Spry & Wiener, 1991; Papers III & IV). The fairly large uncertainty in each lake mean value, mainly caused by natural variation, will tend to decrease the correlation coefficient, especially as the range is quite narrow (Paper VI). However, as pointed out in Paper VI there are most likely other factors at hand which could explain the rather poor correlation between different Hg dose parameters and the Hg concentration in fish. This would be due to the much more complex chemical and biological behaviour of mercury compared to, e.g., radiocesium. The mercury content in fish is almost totally comprised of monomethylmercury, while the fraction of methylated mercury in aerobic lake water could

range up to 25% (Gilmour & Henry, 1991) but in Swedish lake waters is normally in the 1-10% range (Lee & Iverfeldt, 1991; Lindqvist et al., 1991). The Hg concentration in lake water could be estimated from the concentration of DOC or water colour (Mierle & Ingram, 1991; Meili 1991; Papers III, IV & VI) and the total Hg load to a lake could under certain circumstances (Paper VI) also be estimated simply from the ratio catchment area : lake area.

Thus, parameters affecting the methylation and biological uptake become crucial in the case of mercury. This circumstance gives rise to a paradox concerning the influence of abiotic factors on mercury content in fish (Paper VI). A factor that might be causally related to the load of Hg could also be inversely correlated to the content in fish due to a depressing effect on the bioavailability. Subsequently, a parameter which is both positively correlated to the Hg load and favouring methylation becomes a better predictor of the Hg content in fish than simply the lake dose of Hg. A typical example of such a parameter is pH, which in these lakes has a closer correlation to the concentration of Hg in fish than any of the parameters describing Hg-load (Paper IV). Not only the deposition patterns of acid rain and mercury are similar, there are also indications of common source areas within the catchments (see previous section), and more important, a low pH tends to favour methylation (Xun et al., 1987) and affects the food web structure in a way that increases the Hg content in fish (Meili, 1991c; Paper III).

Same parameters turn out to be significant in different types of models and approaches. These are; pH, colour or DOC and nutrient levels among water chemical variables, and lake area in relation to mean depth and catchment area. A problem is the nested character of these parameters which makes it difficult to separate between true causality and simply covariation with causally related variables.

Meili, 1991c showed that the concentration of methylmercury in zooplankton could be modelled rather well by accounting for the ratio between total Hg concentration in lake water and the autochthonous production of the lakes. As there seems to be a quantifiable relationship between the MeHg concentration in organisms at different trophic levels within a lake, it would also be possible to model the Hg concentration in different fish species in a similar way. In his mixed empirical/conceptual model, the influence of pH would mainly be due to the covariation with DOC and autochthonous production. The results (Papers III, IV & VI), suggest that besides mechanisms of which pH is an indicator, there are also mechanisms acting on the Hg concentration in fish which are more directly influenced by pH. The concentration of Hg in small perch was reduced by about 30 % on average in limed lakes, and the decrease was significant only in lakes where long-term lake mean pH had been increased (Paper III; Håkanson, et al., 1991). It was also shown that the concentration of Hg in seston as well as the ratio Hg:C in seston decreased after treatment, the median value for the ratio Hg:C decreased from 1.6  $\mu\text{g g}^{-1}$  during 1986 to 0.8  $\mu\text{g g}^{-1}$  during 1988 (Paper IV). The operationally defined fraction RIHg in water showed an inter-year variation similar to the Hg content in seston, in contrast to the annual mean total concentration of Hg in water which not was decreased after the treatments (IV). The relation between the Hg concentration in settling particles and in water can also be expressed with the distribution coefficient ( $K_{d,Hg}$ ) and the sedimentation coefficient,  $K_{sed,Hg}$  (Paper VI). The sedimentation coefficient  $K_{sed,x}$  is inversely related to the residence time ( $T_{sed,x}$ ) (Eadie & Robbins, 1987) of an element (x) in the water body with respect to the

removal by particle settling ( $T_{\text{sed}} \approx 1/K_{\text{sed}}$ ). The theoretical residence time of Hg in lake water ( $T_{\text{Hg}}$ ) could then be obtained as;

$$T_{\text{Hg}} = T_w * T_{\text{sed}} / (T_w + T_{\text{sed}}) \quad [3]$$

where  $T_w$  = mean hydraulic residence time.

Based on the results presented in Paper IV and from the mass balance study in Paper VI that indicates a decreased retention of Hg within the lakes after the altered chemical conditions, it is suggested that there is either a transition to less reactive Hg species and/or a decrease in the adsorption onto scavenging agents due to the increased pH. Although, the concentration of humics in lake water (measured as colour) could explain a larger part of the inter-lake variation in RIHg, the decrease in the concentration of RIHg was not related to a decrease in colour or increased nutrient levels, but rather to an increase in mean pH due to the lake liming (Paper IV).

### 5.4.2 Radiocesium

Earlier studies (Paper II) in Swedish lakes have shown that the parameters of most significance for the initial transfer of radiocesium to fish were (sign of correlation within brackets) mean hydraulic residence time (+) and any parameter from the more or less intercorrelated parameters of hardness (-), potassium concentration (-), humic content (+) and ionic strength (-). A simple and highly significant relationship was demonstrated between the lake dose expressed as concentration in lake water and the content in fish. How are these parameters related to the total transfer factor (F) and what causal relations could these correlations reflect?

At the same level of fallout, the inter-lake variation of hardness and hydraulic residence time in combination, gave a tenfold difference in the initial transfer from fallout to small perch, which was considerably larger than the inter-lake variation in apparent or ecological half-life ( $0.6 < T_E < 2$  yrs) caused by different depth conditions and humosity.

In Paper VI it is shown that the mean distribution coefficient ( $K_d$ ) for  $^{137}\text{Cs}$  varied between  $1 * 10^4$  to  $7 * 10^4 \text{ cm}^3 \text{ g}^{-1}$  in the most studied lakes ( $n=15$ ), a range that covers the values reported from Lake Zurich (Santchi et al., 1990) and Lake Päijänne (Kansanen et al, 1991) and also most of the modelled, strongly time-dependant distribution coefficients in the epilimnion of Lake Constance during 1986 (Robbins et al., 1992). The inter-lake variation of  $K_{d,\text{Cs}}$  was rather strongly correlated to the natural (i.e. before liming operations) concentration of major base cations expressed as hardness ( $r^2=0.58$ ,  $n=15$ ) and intercorrelated parameters like pH and alkalinity.  $K_{d,\text{Cs}}$  was also negatively correlated to the carbon content in settling material.

The sedimentation rate of radiocesium as expressed by the sedimentation coefficient ( $K_{\text{sed,Cs}}$ ,  $\text{d}^{-1}$ ) was well correlated to the natural concentration of major base cations ( $r^2=0.81$ ,  $n=15$ ) and intercorrelated parameters such as pH, alkalinity and conductivity. The higher scavenging capacity in lakes with higher concentration of major base cations was due to higher particle

sedimentation rates and higher  $K_d$  values in these lakes. However, water chemistry was probably not causal in this respect, despite the high correlation. The remedies caused a very significant increase in mean values of hardness (and intercorrelated parameters) in lakes with initially low concentrations of major base cations (Paper V), but this markedly increased concentration in most lakes during 1988 and 1989, did not notably affect the mean distribution and sedimentation coefficients of  $^{137}\text{Cs}$  in the lakes (Paper VI).

The lack of effect of the remedies on the distribution and sedimentation coefficients for radiocesium, suggests that a likely causal factor would rather be the amount and nature of scavenging agents, which in these lakes was well indicated by the natural concentration of base cations in the water. It was demonstrated that there was a very similar relationship between  $K_{\text{sed,Cs}}$  and the fraction of particulate inorganic matter in settling particles (PIM) before and after remedies, respectively. The higher correlation of  $K_{\text{sed,Cs}}$  to natural water hardness might reflect that the concentration of major base cations in this calcite-poor region is positively correlated to the content of clay minerals in soils and sediments. Lakes with higher sedimentation coefficients generally also had a higher bioproduction, but the correlation of  $K_{\text{d,Cs}}$  to total-P in water or the ratio C:N in settling matter was much weaker compared to the correlation to hardness.

In a model for scavenging of  $^{137}\text{Cs}$  in the epilimnion of Lake Constance (Robbins, et al., 1992), the best fit to observed activities was obtained using a substitute (particulate aluminium) for clay minerals as the "reactive phase" of total suspended matter, while the affinity to calcite was found to be negligible. Cremers et al., 1988 and several earlier works have shown that the partition of radiocesium between solid and liquid phase in soils is regulated by a small number of highly selective ion-exchange sites, located at the frayed edges of micaceous clay minerals (illites). These observations are all consistent with the observed depletion of nuclear weapon radiocesium in sediment inventories compared to the cumulative atmospheric deposition in North American softwater lakes (Heit & Miller, 1987), where low levels of binding clay minerals were suggested as a prime cause. Because of the mentioned reasons, however, this interpretation of the correlation between hardness or ionic strength to scavenging capacity seems to be restricted to lakes within the zone of boreal forests and it could not be applied in geological areas dominated by calcite or dolomite weathering.

Figure 7 shows the relationship between the mean hydraulic residence time and the theoretical residence time for  $^{137}\text{Cs}$  ( $T_{\text{Cs}}$ ) within the lake water columns for different values of the sedimentation coefficient ( $K_{\text{sed}}$ ), along with empirical values and modelled values based on natural mean water hardness (Paper VI). It may be noted that there was a considerable variation between lakes concerning their theoretical retention of  $^{137}\text{Cs}$ . This variation is directly connected to the hydraulic residence time of the lakes, but also to the factors described earlier that influence the specific sedimentation rates of  $^{137}\text{Cs}$ . The theoretical residence time of  $^{137}\text{Cs}$  was determined in 15 lakes using sediment traps. As water hardness was found to be a good indicator of the scavenging capacity of  $^{137}\text{Cs}$  from the lake water, the mean concentration of Ca+Mg in the lake waters was used to calculate the removal rate due to sedimentation (Figure 7). In this context, it is important to note that the empirical values are based on data on gross sedimentation from sediment traps without taking account of any potential resuspension effect.

The apparent residence time is almost always significantly longer than  $T_{Cs}$ , due to resuspension of, or possibly diffusion from Cs-contaminated sediments, or to input from the catchments, or a combination of these mechanisms. However, during the summer of 1986, after the large initial direct deposition and catchment derived input that occurred within the first month after Chernobyl, the lake water pools were comparatively large compared to the potential loading from resuspension and from catchments which declined very rapidly. Thus, it is likely that  $T_{Cs}$  provides a fairly realistic picture of the shape and magnitude of the pulse through the lake water columns during the first important phase.

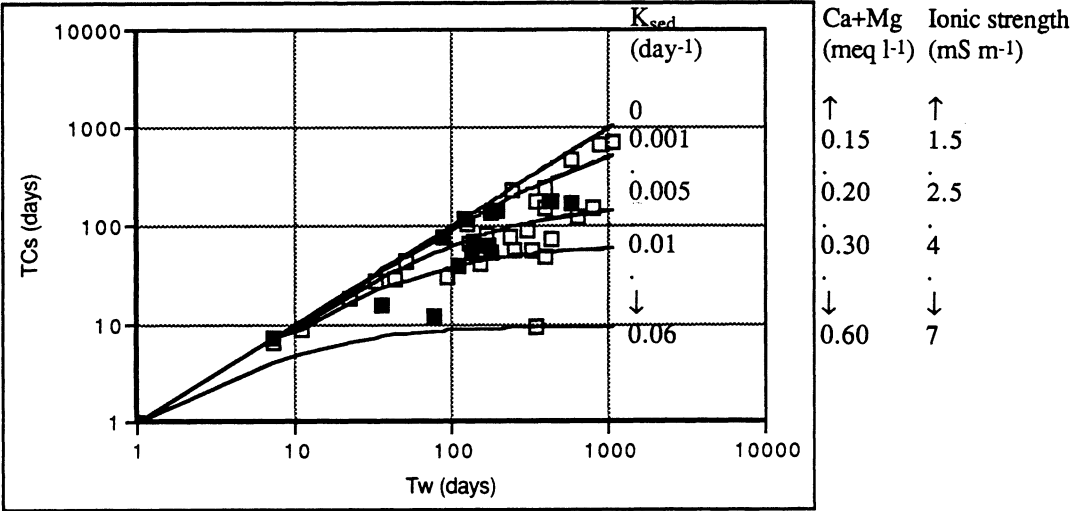


Figure 7. The relationship between the mean hydraulic residence time ( $T_w$ ) and the theoretical residence time of  $^{137}\text{Cs}$  for different values on the sedimentation coefficient ( $K_{sed}$ ), along with empirical values ( $n=15$  lakes, filled squares) and modelled values ( $n=30$  lakes unfilled squares) based on the natural mean hardness of lake water.

This suggestion is supported by Figure 8, which shows the relationship between the theoretical residence time of  $^{137}\text{Cs}$  in lake water ( $T_{Cs}$ ) and the total transfer to pike. Three lakes with high transfer to fish despite low  $T_{Cs}$  can be identified in Figure 8. In two cases these are very shallow lakes (mean depths around 1 m) and, thus, a high degree of wind/wave induced resuspension could be expected. The third lake (Lill-Selasjön) is situated immediately downstream of a much larger lake, and could in practice be regarded as a part of that larger lake with its much higher  $T_{Cs}$ -value. The relationship given in Figure 8 is therefore valid for lakes without major secondary inputs of  $^{137}\text{Cs}$  to lake waters, either from major resuspension activity (or possibly diffusion) of sediments or from major temporary traps (lakes or bogs) within the catchment. From the rather high occurrence of shallow lakes in the studied data set one would expect more outliers from the general pattern which does not take into account resuspension or sediment mediated uptake. This lack of outliers indicate that the biological availability of



sediment bound radiocesium is generally rather low, even though the effect of resuspension or sediment mediated uptake must be considered in certain lakes.

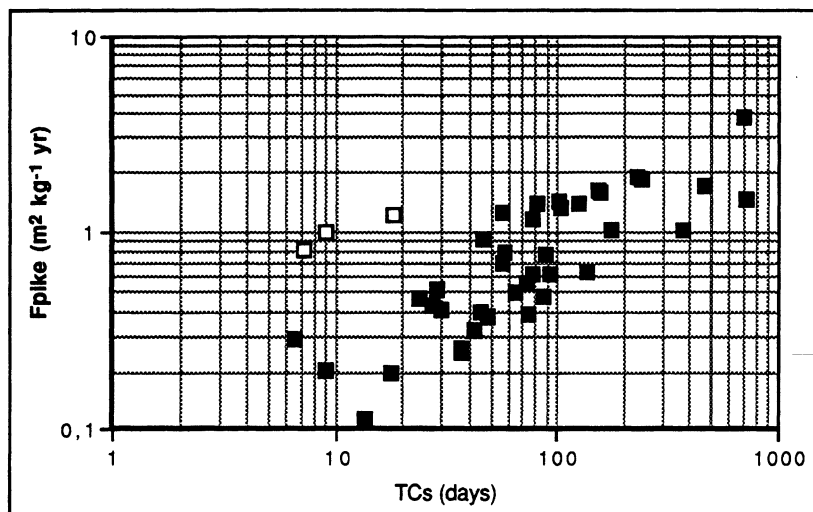


Figure 8. The relationship between the theoretical residence time (TCs) of  $^{137}\text{Cs}$  in lake water and the expected total transfer to pike (Fpi) in 45 Nordic lakes.

## 6. Conclusions

- About 10000 Swedish lakes were calculated to have a mean Hg content in 1-kg pike (FHg) above  $1 \text{ mg kg}^{-1}$  (wet weight) in the end of 1980's, that is a 5-fold increase compared to the calculated preindustrial mean value. The cumulated domestic Hg- sources of emission make the largest contribution to the high mercury levels in pike and particularly so in central and northern Sweden. The second most important cause is acidification and thirdly Hg emissions from European sources. A fairly good agreement was obtained between the empirical and the calculated geographical distribution of FHg using a factor related to the cumulated Hg emissions (EFSUM), pH and color as input parameters, the average deviation between calculated and empirical geographical distribution was  $< 0.2 \text{ mg kg}^{-1}$  in the major part of Sweden.

- The content of  $^{137}\text{Cs}$  in fish normalized to 100 g perch (FCs) was above the limit for commercial sale,  $1500 \text{ Bq kg}^{-1}$ , in about 14000 Swedish lakes during autumn of 1987. An empirical model including Chernobyl fallout, hydraulic residence time and ionic strength explained almost 60 % of the inter-lake variation in FCs. At the same level of fallout, this difference in lake sensitivity, gave a tenfold difference in the initial transfer from fallout to small perch.

- The maximum transfer of  $^{137}\text{Cs}$  was reached within the first three years for all species in most

lakes and normally in the order; small perch - brown trout and Arctic charr - larger perch - pike. This sequence seems to reflect the trophic level of each species and is in accordance with the idea that the uptake of  $^{137}\text{Cs}$  in fish occurs mainly from the food and that the activity in the predator is determined by the prey activity, even if differences in the metabolic and feeding rates between different species should also be considered. Thus, the fish feeding pike is the species with the most extended temporal development and is also the species with the highest values on the total time integrated transfer (F).

- Parameters that turn out to be significant for the Hg content in fish in different types of models and approaches are; pH, color or DOC and nutrient concentrations and lake area in relation to mean depth and catchment area. It is suggested that besides mechanisms involving DOC and autochthonous production which pH is correlating, there are also mechanisms acting on the Hg content in fish which are more directly influenced by pH. The concentration of Hg in small perch was reduced with about 30 % on average in limed lakes, and the decrease was significant only in lakes where long-term lake mean pH had been increased.

- There exists a highly significant correlation between the total transfer of  $^{137}\text{Cs}$  to perch and to pike, lakes with a high total transfer to small perch have also a high total transfer to pike. There is a rather high correlation between the lake mean Hg concentration in small perch (<10 g) and in 1-kg pike and the ratio (Hg-pi:Hg-pe) ranged around 7. These results are consistent with findings that the biomagnification of methylmercury could be characterized by a fairly constant enrichment factor between two trophic levels.

- The sedimentation rate of radiocesium was well correlated to the natural concentration of major base cations and intercorrelated parameters such as pH, alkalinity and conductivity. The higher scavenging capacity in lakes with higher concentration of major base cations was due to higher particle sedimentation rates and higher  $K_d$  values in these lakes. However, the water chemistry was probably not causal in this respect, despite the high correlation, the distribution and sedimentation coefficients for radiocesium was not notably affected of the increased mean concentration of major base cations after liming and potash addition. It is suggested that a likely causal factor rather would be the amount and nature of scavenging agents (possibly clay minerals), which in these lakes was well indicated by the natural concentration of base cations in the water. This interpretation is also in line with the non-significant effect of liming and potash addition at the radiocesium levels in fish.

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