Chernobyl radiocesium in freshwater fish: Long-term dynamics and sources of variation

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Background

The radioactive cesium isotopes ^{134}Cs and ^{137}Cs are artificial products of nuclear reactions and are not occurring naturally in the biosphere. Nevertheless, they are today globally distributed contaminants due to atmospheric nuclear bomb detonations, nuclear power plant accidents and controlled release from nuclear industry. Cesium is a very reactive and therefore found in nature only as monovalent ions, Cs^+. Because of the similar physicochemical properties of Cs^+ and K^+ radiocesium accumulates in biota, which implies potential health hazards for humans. For this reason radiocesium, especially the long-lived ^{137}Cs (half-life 30.17 years), has for decades attracted both radioecologists and health physicists. From another point of view, since ^{137}Cs is inert, not essential, has a simple chemistry and is able to measure at low levels, it has been suggested as a suitable tracer and tool for scientific research. Common applications for ^{137}Cs are the dating of recent sediments and studies of soil erosion. To a more limited extent radiocesium has been used in ecology, but has high potential in studies of bioenergetics, trophic transfer and migration in aquatic ecosystems (Davis & Foster, 1958; Kevern, 1966; Kolehmainen, 1974; Meili, 1994; Odum & Golley, 1963; Rasmussen & Lassen, 1994; Rowan & Rasmussen, 1996; Storebakken, Austreng & Steenberg, 1981; Tucker *et al.*, 1999; Tucker & Rasmussen, 1999).

When the radioactive fallout from the Chernobyl nuclear plant accident in 1986 was detected over large areas of Europe, radioecological research activities had been diminishing since the era of atmospheric nuclear bomb tests. Naturally the new and alarming situation led to the initiation of numerous research and sampling programs. Although much of the old work was substantial and of high quality it was not readily applicable to the post-Chernobyl situation. Unlike the global and more or less continuous fallout during years the 1950's and 1960's, selected areas received the nuclides from Chernobyl as a distinct pulse. As a consequence, affected ecosystems were in strong disequilibria with regard to long-lived radionuclides, such as ^{137}Cs. A dynamic equilibrium situation could no longer be assumed or justified; radioecologists and modelers were forced to focus more on dynamic situations. This proved not to be a simple task, since the equilibration rates differ substantially between different ecosystem compartments and depend much on local conditions. Taking radiocesium in freshwater fish as an example, lake chemistry, morphology and hydrological regime, the catchment's vegetation, soil types and land use, as well as climate and fish ecology, are among the factors that have been considered in post-Chernobyl dynamic models (e.g. Bergström, Sundblad & Nordlinder, 1994; Håkanson *et al.*, 1996a; Håkanson *et al.*, 1996b). In retrospect, the early attempts to predict the elimination of radiocesium from lake ecosystems were too optimistic. Recent studies report that levels in fish remain high and decline very slowly (Jonsson, Forseth & Ugedal, 1999; Saxén & Koskelainen, 1996).

There are two other problems that face both the radioecologist and the modeler that wishes to learn more about or predict radiocesium accumulation in fish. First, the shortage of data, especially from the early 1990's and later – when ^{137}Cs concentrations in fish were declining again in the late eighties, many monitoring projects were discontinued. Also the quality of data has not been sufficient. The large scale national monitoring projects often acquired samples from a multitude of sources, and little information was given about catch conditions, fish size, sex, condition or diet. Second, within lakes and even at the same sampling occasion ^{137}Cs concentrations in fish display a very high variability, often two-fold or more (e.g. Håkanson, 1999). Knowledge about the magnitude and sources of this variation is important for the reliability of data, the design of sampling routines and accurate predictive modeling of radiocesium in fish. Furthermore, for applications that use ^{137}Cs as tracer for fish bioenergetic processes (He & Stewart, 1997; Kevern, 1966; Kolehmainen, 1974; Rowan *et al.*, 1995; Sherwood *et al.*, 2000), understanding of the factors responsible for individual variation is of crucial importance.

The aim of this thesis was to investigate both the long-term temporal pattern and sources of individual variation for radiocesium in freshwater fish. The basis for the study is time series of ^{137}Cs activity concentrations in fish from three lakes in the area North-west of Uppsala, Sweden that received considerable amounts of ^{137}Cs from Chernobyl in may 1986. The lakes were Lake Ekholmssjön, Lake Flatsjön and Lake Siggeforasjön, all small forest lakes, but with different morphometrical and chemical characteristics. The data were collected regularly, usually several times per year, during 1986-2000, using consistent methods. More than 7600 fish individuals from 7 species covering wide size ranges and feeding habits were analysed for ^{137}Cs. For each fish was the length, weight, sex, and often the stomach contend recorded. The evaluation on long-term trends were based on data from all three lakes, while the study on sources of variation evaluated data from Lake Flatsjön only.

Long term trends

After the Chernobyl pulse deposition of radiocesium, the maximum activity concentrations in lake water were attained almost immediately. The following removal of ^{137}Cs from the water column was first very rapid and then gradually became slower (Saxén, Jaakkola & Rantavaara, 1996). The fast removal phase was largely dependent on lake dilution and particle setting rates (Smith, Comans & Elder, 1999). Later on, as the concentrations leveled off, secondary input from the catchment area, remobilization from sediments by diffusion (Comans *et al.*, 1989; Smith & Comans, 1996) or resuspension (Broberg, Malmgren & Jansson, 1995; Meili, Braf & Konitzer, 1997) have been pointed out as secondary sources of ^{137}Cs to lake water, thereby slowing down the net removal of ^{137}Cs. The slow sorption ^{137}Cs to clay minerals has been suggested to be the ultimate controlling factor for the long term decline of ^{137}Cs (Comans & Hockley, 1992; Hilton *et al.*, 1993; Kudelsky *et al.*, 1996).

Since fish take up cesium predominantly from the food, direct uptake being negligible, the pulse of ^{137}Cs levels in fish is significantly delayed compared to that lake water. The typical pattern is a fast increase until maximum levels are attained, followed by a relatively fast decrease and thereafter the concentrations level off along a plateau formation, probably indicating a gradual equilibration with the slowly declining concentration in water. Although there are many reports on the temporal trend of Chernobyl ^{137}Cs in fish there is no consensus on how the long-term pattern varies across species and different types of lakes. This is because most published studies covered only the first years after fallout, used small samples sizes or possessed little information of the size and diet of each fish.

In this study, a non-linear model was developed and fitted to the time series of ^{137}Cs in fish. The fish was first grouped into species and for abundant species also into different size classes. ^{137}Cs

concentrations were corrected for radioactive decay since May 1, 1986. The main benefit of the model was that it enabled an objective and consistent method for extracting the parameters of interest. These were: (1) maximum activity concentration (Csmax); (2) the time to reach maximum activity concentration (tmax); (3) the maximum rate of decline (or minimum ecological half-lives); (4) the near-steady state level (Base) and (5) the long-term ecological half-life.

The ^{137}Cs dynamics during the initial years, as well as the leveling off during the following years, were related to the type of fish. Both tmax and minimum half-life increased significantly with trophic level and weight. The effect of trophic level on tmax was stronger than the effect of weight, suggesting that retardation by the food chain was largely responsible for the delayed ^{137}Cs peak in piscivorous fish. The opposite was found for the fast-decline phase. Fish weight had stronger effect on the minimum ecological half-life than did trophic level. Small fish entered the near-steady state phase already in 1888-1989, significantly earlier than larger fish. Clearly, during this phase the biological half-life was more important than the food chain retardation, probably because the change in concentration of ^{137}Cs in water was slower after the peak than earlier.

The maximum levels increased with trophic level but decreased with weight. As mentioned above, large fish reached maximum levels later than smaller fish, which implies that their food contained less ^{137}Cs due to the overall elimination of available ^{137}Cs from the lake. The positive effect of trophic level, however, indicates the importance of biomagnification of radiocesium. This is also reflected by the near-steady state levels (Base), which increased markedly with trophic level, especially in the transition to piscivory. The effect of weight on Base was smaller. Both Csmax and Base were significantly higher for perch *(Perca fluviatilis)* than the other species. Even small non-piscivorous perch had considerably higher levels than other fish on comparable trophic positions.

Although the absolute levels varied among lakes, the relationships between the model parameters and trophic level or weight did not differ significantly. The general dynamic pattern of ^{137}Cs in lacustrine biota following a pulse contamination is thus to a large extent governed by the fish population and food web characteristics. This inertia of the food web implies that cesium dynamics may be partly uncoupled from other processes and would, if properly described and quantified, simplify the task of cross system modelers.

Long term ecological half-lives were of the same magnitude as the physical decay rate (30.17 years) and were not influenced by the fish type. In addition, the estimates of ecological half-life increased over the period, apparently approaching infinity, suggesting a gradual shift in the processes regulating the radiocesium levels in fish: From physiological, over ecological and geochemical factors, to finally radioactive decay. Ecological half-lives were long and increased over the period. If estimated over the period 1996-2000, the median ecological half-life was 20 years in Ekholmssjön, 50 years in Siggeforasjön and 80 years in Lake Flatsjön. This means that in practice the radioactive decay will determine the elimination of ^{137}Cs from these lake ecosystems. Large perch in Siggeforasjön and both perch and pike in Flatsjön had average levels above the radiation protection guidelines. Thus, fishermen in many lakes in this area ought to wait another 50-60 years before to eat or sell their catches.

Within-lake variation

During the period 1996-1999 the fishing was intensified in Lake Flatsjön. The aim was to study what factors that were responsible for the high individual variability that was observed for all species of fish. Linear models (ANOVA, ANCOVA) were used to detect the variables that could explain most of the variation. Variables of interest were species, size, condition factor, food choice, gonadal- and gastro-somatic indices, flesh water, potassium and lipid concentrations and seasonal variability. In addition, the variability that could be generated by individual variability in bioenergetic factors was calculated and compared with the observed variation.

The total variation was 22-fold among the 1338 individuals of seven fish species Individual variation was high, across species CV:s ranged from 0.16 to 0.69 with an average of 0.35. Seasonal variation accounted for only 7% of total individual variation. The amount of the variability that could be explained by the combined effects of season (sampling date) and fish length is varied among species. R2-values ranged between 0.07 and 0.59, which means that 41-93% of the variability must have been generated by other factors. Fish species, diet composition, fish size and condition factor were significant predictors of ^{137}Cs levels. Perch and pike *(Esox lucius)* had generally higher levels than cyprinid fish. ^{137}Cs levels usually increased linearly with fish length, except for perch, where levels increased with length following a more complex or stepwise pattern. The average proportion of fish in perch diet was well correlated with this pattern. Comparison of the observed variation with the variation expected from potential individual differences in bioenergetics indicated that additional factors were important for most species. Although contamination levels decreased with increasing condition factor, neither muscular lipid, potassium concentrations, sex nor gastro- and gonado-somatic indices contributed significantly to the observed variation. For perch and pike, however, individual food choice based on stomach content analyses could explain some of the exceptionally high ^{137}Cs variation in this species. Accordingly, total variation was higher for species or size classes with wide diet preferences. The results suggest that, while basic trophic structure and physiology determine the baseline ^{137}Cs levels in fish within a lake, the high variation among similar individuals often found in ^{137}Cs-contaminated lakes is to a large extent caused by individual food choice.

The average biomagnification factor between piscivores and the most common prey fish was 4.13 for large perch and 2.17 for pike. The pooled estimate of 3.17 is similar to the average piscivore biomagnification factor of 3.69 obtained by Rowan & Rasmussen (1998). Although the trophic relationships within the zooplankton and benthic communities are complex because of detritivory and microbial food webs there are more carnivorous taxa within the littoral benthos than in the water column. Assuming that biomagnification occurs also in the invertebrate food webs, it would be expected that benthivores had higher ^{137}Cs levels than zooplanktivores, which in turn would have higher levels than herbivores. Carnivorous Odonata larvae were a common prey for small perch (10-15 cm), while herbivorous crustaceans dominated the diet of perch less than 10 cm. However, there was no difference in ^{137}Cs levels between the groups. Within the non-piscivore cyprinids there was only small differences between omnivorous roach *(Rutilus rutilus)*, mainly benthivorous bream (*Abramis brama*) and white bream (*Abramis blicca*), and largely herbivorous rudd *(Scardinius erytrophtalmus)*. The results provide no evidence of a significant biomagnification of cesium along invertebrate food chain. One explanation for this may be that, due to small body size, direct uptake of ^{137}Cs is relatively more important for invertebrates than for fish, for which trophic uptake dominates. Furthermore, it has been argued that benthos and hence benthivores should accumulate more ^{137}Cs than zooplankton and zooplanktivores since they are in direct contact with the highly contaminated sediment surface. Contrary to the results of Rowan & Rasmussen (1998) no such effect was found in Lake Flatsjön suggesting the sediment-bound ^{137}Cs is not readily bioavailable.

In conclusion, although seasonal, size, and bioenergetic effects together can explain much of variation, individual food choice is of importance for species with wide diet preferences. This ought to be considered in bioenergetic models that use cesium as a tool for estimating e.g. feeding rates *in situ*. The generally high variability in popular game fish should be considered when monitoring environmental radioactivity or assessing the health risks associated with freshwater fish consumption.

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