Application of potassium chloride to a Chernobyl-contaminated lake: modelling the dynamics of radiocaesium in an aquatic ecosystem and decontamination of fish.

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Abstract

This study tests a whole-lake experiment to reduce the bioaccumulation of radiocaesium (^{137}Cs) in fish in lakes contaminated by the Chernobyl accident. In many lakes in the Chernobyl contaminated areas, radiocaesium activity concentrations in fish are still significantly higher (up to 100 times in some species) than acceptable limits for human consumption. Estimates of the long-term rate of decline of ^{137}Cs in fish in these regions, in the absence of countermeasures, show that radioactivity in fish in some lakes may remain above acceptable consumption limits for a further 50-100 years from the present date. In February 1998 we applied 15 tonnes of potassium chloride to Lake Svyatoe, Kostiukovichy. The addition of potassium chloride fertiliser to the lake resulted in a decrease in activity concentration of ^{137}Cs to around 40% of pre-countermeasure values in a number of different fish species. In contrast to Lake Svyatoe, ^{137}Cs activity concentrations in fish from four control lakes showed no systematic decrease over the study period. Simplified models for transfers of ^{137}Cs in lakes successfully “blind” predicted the changes in ^{137}Cs in water and fish resulting from this major alteration of the potassium concentration of the lake. The experiment represents the first test of a predictive model for the dynamics of radiocaesium in response to a major perturbation in potassium (its major competitor ion) in a whole lake ecosystem.

Introduction

Radiocaesium activity concentrations in water declined rapidly after the Chernobyl accident (Vakulovsky et al. 1994). Radiocaesium is, however, strongly bio-accumulated in fish and other aquatic biota (Fleishman, 1973). The level of radioactive contamination of aquatic biota is commonly defined in terms of a concentration factor (CF) where

$$\mathrm{CF} = \frac{\textit{Activity concentration per kg of fish (wet wt)}}{\textit{Activity concentration per litre of water}} \ \mathrm{l\ kg^{-1}} \qquad (1)$$

Studies following the deposition of radiocaesium during the period of atmospheric nuclear weapons testing (mainly during 1957-63), observed concentration factors in fish of between 10^2-10^4 l kg^{-1} (Fleishman, 1973). Following the 1986 Chernobyl accident, bio-accumulation of radiocaesium in fish resulted in activity concentrations (both in Western Europe and in the former Soviet Union) which were in many cases significantly above the maximum permissible level for human consumption. In many lakes in Russia and Belarus ^{137}Cs activity concentrations in fish are still significantly (up to 100 times) higher than regulatory limits. Recent evidence suggests that in some areas levels will remain above limits for many decades to come (Jonsson et al. 1999; Smith et al. 2000a).

The processes which determine the accumulation of radiocaesium in fish are complex, and lead to wide differences in contamination levels according to water chemistry, fish type, size and feeding patterns (e.g. Fleishman, 1973; Hadderingh et al. 1997). The most important pathway of radiocaesium into fish is via intake of food (e.g. Forseth et al. 1991; Elliott et al. 1992), and the high concentration factors observed are a result of accumulation of radiocaesium through the food chain.

Because of its chemical similarity to caesium, the potassium concentration of lake or river water has long been known to influence the rates of accumulation of radiocaesium in fish (Blaylock, 1982). Strong inverse relationships were observed between lake water [K^+] and ^{137}Cs activity concentrations in fish following nuclear weapons testing (Fleishman 1973, Blaylock, 1982) and the Chernobyl accident (Smith et al. 2000b). This hypothesis concerning the influence of potassium on radiocaesium accumulation in fish is confirmed by laboratory studies in which radiocaesium uptake rates were studied as a function of increasing potassium concentration (Gil Corisco & Vaz Carreiro, 1997). The artificial increase of the potassium concentration of a lake is therefore a potential method of reducing radiocaesium activity concentrations in fish.

Though the potential effectiveness of a potassium countermeasure to radiocaesium in fish has been postulated for some time, it could only be tested by carrying out a whole lake experiment. Such an experiment was carried out in Sweden after Chernobyl (Håkanson & Andersson 1992), however, because of the high water turnover in the lakes studied, it was difficult to maintain a high [K^+] content of the water and the results of the test were inconclusive. In this study we have carried out the first conclusive “whole-lake” experiment into the relationship between potassium and radiocaesium dynamics in a contaminated lake ecosystem by adding 15 tonnes of KCl fertiliser to a lake in Belarus. Lake Svyatoe in the Kostiukovichy district of Belarus was chosen for testing the potassium countermeasure. Like many lakes in this area, this lake is groundwater fed and has no outflow, thus losses of potassium by lake flushing were expected to be low.

The “natural” potassium concentration in Lake Svyatoe (Kostiukovichy) is relatively low (0.025 mEq l^{-1} or 1 mg l^{-1}) and consequently the ^{137}Cs concentration factor (CF) in fish species is relatively high (ca. 12,000 l kg^{-1} in perch, *Perca fluviatilis*, and 3,200 l kg^{-1} in other species) (Smith et al. 2000b). Activity concentrations of ^{137}Cs in small non-predatory species were approximately 15,000 Bq kg^{-1} and up to approximately 100,000 Bq kg^{-1} in larger predatory fish (perch; pike, *Esox lucius*). These activity concentrations are more than one order of magnitude above recommended maximum limits for human consumption (of approximately 1,000 Bq kg^{-1}). Although the lake is within an evacuated zone, fish are still caught and consumed by fishermen.

Methods

Sufficient [K^+] was added to the lakewater to achieve an initial increase in concentration from 0.025 mEq l^{-1} (1 mg l^{-1}) to 0.25 mEq l^{-1} (10 mg l^{-1}). The volume of L. Svyatoe is 718×10^3 m^3, requiring addition of 15 tonnes (0.05 kg per square metre of water surface) of KCl fertilizer to achieve a [K^+] concentration of 0.25 mEq l^{-1}. The application rate is comparable to volumes of fertilizer normally applied to agricultural land. The KCl was spread on the frozen lake surface during February 1998. The application method was very successful, resulting in a gradual increase in lakewater [K^+] concentration as the ice melted in spring (March-April 1998).

Field studies were carried out at L. Svyatoe and 4 other lakes in Belarus, Ukraine and the Bryansk region of Russia (Table 1). A map showing the locations of these lakes has previously been published in Smith et al. (2000b). Water samples were taken from each lake and measured for their ^{137}Cs activity concentration, suspended matter concentration, and water chemistry (in particular, potassium concentration, [K^+]). Monitoring of the water chemistry (particularly potassium concentration) of L. Svyatoe (Kostiukovichy) was carried out at approximately monthly intervals in order to assess the changes in water chemistry after the countermeasure application. Bed sediment samples were obtained using a grab sampler and the sediment porewater was removed, using a pore water press, for chemical analysis.

Fish sampling was carried out to monitor the effects of the countermeasure and in the 4 ‘control’ lakes: Tyumenskoye, Svyatoe (Chechersk), Kozhanovskoe and the Kiev Reservoir. Note that one of the control lakes, Lake Svyatoe in the Chechersk region of Belarus has the same name as the study lake, Lake Svyatoe in the Kostiukovichy region. We therefore refer to the study lake as Svyatoe (Kostiukovichy) and the control lake as Svyatoe (Chechersk).

Samples of fish were obtained using gill nets and divided by species, sex and size groups. The operculum was removed from perch and scale samples taken from other fish species for age determination. Length and total and somatic (i.e. without organs) weights were determined. Fish were prepared for gamma- radiometric analysis by removing the head, tail and organs, dried at 105 ^{o}C and ground. Dry weights were determined. For very small fish, the whole fish was measured (i.e. including head, tail and organs), and individuals were combined to give sufficient sample for counting.

In L. Svyatoe (Kostiukovichy) and three other lakes of similar physical characteristics (L. Tyumenskoe, L. Svyatskoe, L. Svyatoe, Chechersk) plankton samples were taken during 1997-99 to study possible ecological effects of the potassium treatment of L. Svyatoe (Kostiukovichy). Samples were taken with a 50 μm plankton net just below the water surface surface and hauled over a length of about 50 – 100 m behind a rowing boat.

Results

Effects of the countermeasure on ^{137}Cs and $[K^+]$ in the lake water

The countermeasure application resulted in an increase in $[K^+]$ concentration in the lake water to the target concentration of approximately 0.25 mEq l^{-1} (10 mg l^{-1}) (Figure 1). There was little loss of K^+ from the lake water, so that concentrations remained high throughout the period of study (Figure 1). Analyses of sediment pore water chemistry, prior to the countermeasure application (30/09/97), found mean $[K^+]$ in the sediment pore waters to be 0.27 mEq l^{-1}. The pore water potassium concentration (0.27 mEq l^{-1}) is approximately equal to the lakewater concentration after the potassium addition (0.25 mEq l^{-1}). There is therefore no concentration gradient between lake water and sediment pore water, so we would expect (and have observed) relatively little diffusion of potassium from the lake to the sediments. The reduction in $[K^+]$ observed during 1998-2000 (Figure 1) may be due to slow transfers to the bed sediments and to water loss via groundwater (the lake has no surface outflow). The concentration of $[NH_4^+]$ in the lake water was the same before and after the experiment, having mean value 0.03 mEq l^{-1}.

The tenfold increase in the potassium concentration of the lakewater appears, as expected, to have resulted in an increase in ^{137}Cs in the water (Figure 1), as a result of remobilisation of activity from the bottom sediments (see Discussion for further details). As discussed in greater detail in the Discussion section below, this remobilisation is attributed to competitition for sorption sites in the sediment between radiocaesium and its competitor ions, potassium and ammonium.

Effect of the countermeasure on aquatic biota

In the treated lake Svyatoe (Kostiukovichy) the water was turbid and greenish at all three sampling occasions in 1998/99, and also in the year 1997. This is caused by the blue-green algae (*Cyanophycea*), this group being dominated at all three visits by *Microcystis* sp. In August 1999 another blue-green algae *Anabaena spiroïdes* was also very abundant. In the other plankton groups large differences were found in the species diversity and abundance between the three sampling dates. For example, in the spring of 1999 many *Bacillariophycea* were present in comparison with the spring of 1998. Zooplankton was scarce in August 1999, which may be the result of consumption by fish during the summer.

As a result of the plankton sampling it can be concluded that after the potassium treatment in February 1998, all common plankton groups were present in lake Svyatoe (Kostiukovichy) in 1998 and 1999. As the sampling has been done only occasionally, possible small changes in the composition of the plankton as a result of the treatment are not discernible. Measurements of concentrations of suspended matter in L. Svyatoe (Kostiukovichy) showed that there was no significant change in total suspended solids (mainly composed of plankton and suspended organic matter) after the countermeasure application.

Assessment of the countermeasure effectiveness

Sampling results show evidence of a significant decrease in ^{137}Cs activity concentrations in fish in L. Svyatoe (Kostiukovichy) following the countermeasure addition (Figure 2). In Figure 2(b) samples of perch are separated into two different weight classes (less than and greater than 100g). This is because there is a significant difference in radiocaesium bioaccumulation in large perch compared to small, due to differences in age, physiology and dietary habits (Elliott et al. 1992; Smith et al. 2002).

Declines in radiocaesium activity concentrations were observed in all species, with ^{137}Cs activity concentrations in 1999 being approximately 40% of their value prior to the countermeasure (Table 2). The somewhat higher value for gudgeon (73.3%) than the other species may simply be a statistical anomaly.

Six different species of fish were caught in the control lakes in each of the three years of the study: in L. Tyumenskoe, rudd (*Scardinius erythrophthalmus*) and perch; in L. Svyatoe (Chechersk), roach, bream, (*Abramis brama*); in L. Kozhanovskoe, goldfish (*Carassius auratus auratus*) and pike; in the Kiev Reservoir bream, pike, goldfish, and perch. In contrast to L. Svyatoe (Kostiukovichy), there was no evidence of a systematic decrease in ^{137}Cs activity concentrations in fish from the four control lakes (Figure 3). It is noted that care had to be taken in using L. Kozhanovskoe as a control for our experiment since, during 1998, radioactively “clean” fish from an upstream reservoir were accidentally released into L. Kozhanovskoe as a result of a dam breach. These “clean” samples were identified and removed from the data set for the purposes of estimating decline in ^{137}Cs in the Kozhanovskoe fish population.

Discussion

Declines in ^{137}Cs in the absence of countermeasures

During the first few years after Chernobyl, ^{137}Cs activity concentrations in water and fish declined relatively rapidly (Smith et al. 2000a). However, studies of the long term contamination of aquatic ecosystems by ^{137}Cs (Jonsson et al. 1999, Smith et al. 2000a) have shown that in recent years activity concentrations in water and fish have declined only very slowly, with rates of decline tending towards the physical decay rate of ^{137}Cs. Observations in a lake in Norway (Jonsson et al. 2000) and two lakes in the UK showed that the effective ecological half life (T_{eff}, the time taken for the amount of radioactivity in fish to reduce by one half) of ^{137}Cs in fish was in the range 6-30 years during the period 1994-1999. Studies on two of our control lakes (Smith et al. 2001), Lake Kozhanovskoe and the Kiev Reservoir observed T_{eff} values of around 20 years in the period 1993-1997. These slow declines are consistent with our observations that ^{137}Cs activity concentrations in fish in our control lakes did not significantly decline during the study period, 1997-99 (Figure 3).

Modelling the release of ^{137}Cs from sediments after countermeasure application.

We have estimated the expected release of ^{137}Cs from bed sediments following addition of $[K^+]$ to the lakewater. The vast majority (ca. 99%) of the radiocaesium inventory in the lake is retained in the bed sediments (from measurements in Lake Svyatoe (Kostiukovichy), we estimate 3×10^9 Bq in water prior to the countermeasure and approximately 2.5×10^{11} Bq in sediments). We therefore assume that the remobilisation of ^{137}Cs has negligible effect on the total available ^{137}Cs in the sediments. The equilibrium ratio of ^{137}Cs in the dissolved phase of the lakewater ($[^{137}Cs]_W^{before}$) before countermeasure application to that after ($[^{137}Cs]_W^{after}$) can be estimated from the relative concentrations of competing ions ($[NH_4^+]$, $[K^+]$) in the lake water before and after the countermeasure application (Konoplev & Bulgakov, 2000):

$$\frac{[^{137}Cs]_W^{before}}{[^{137}Cs]_W^{after}} = \frac{K_c^{N/K}[NH_4^+]_W^{before} + [K^+]_W^{before}}{K_c^{N/K}[NH_4^+]_W^{after} + [K^+]_W^{after}} \quad (2)$$

where the concentration of ammonium is modified by the NH_4^+- K^+ selectivity coefficient, $K_c^{N/K}$. In a range of freshwater sediments $K_c^{N/K}$ was found to have values between 2.4 - 5.15 (De Preter, 1990). It should be noted that this equation gives the decrease in ^{137}Cs activity concentrations relative to the natural decrease by, for example, transfer out of the surface sediment layer by sediment accumulation. This decrease is very low for Lake Svyatoe (Kostiukovichy) as there is little net accumulation of bed sediments.

The $[NH_4^+]$ concentration of the lakewater was the same before and after the experiment, having a mean value of 0.03 mEq l^{-1}. The $[K^+]$ concentration increased from 0.025 mEq l^{-1} to 0.25 mEq l^{-1} resulting, from Equation 2, in a predicted increase in ^{137}Cs in the water of a factor of 2.3 – 3.3. $[^{137}Cs]_W^{before}$ prior to the experiment was 4.7 ± 1.3 Bq l^{-1}, giving an estimated increase in $[^{137}Cs]_W^{after}$ to 8 – 20 Bq l^{-1} (given the range in potential selectivity coefficients and initial $[^{137}Cs]_w$ estimates) after the countermeasure application (Figure 1).

The $[K^+]$ concentration in water was observed to decline exponentially (Figure 1). Using Equation 2, we have predicted the change in ^{137}Cs in the water by extrapolating the exponential decrease in $[K^+]$ over this period so that the concentration reaches the pre-countermeasure value of approximately 0.025 mEq l^{-1} after around 15 years (Figure 4). In general, the model makes good predictions of the expected changes in ^{137}Cs in the water. Since the model assumes instantaneous equilibrium between water and bed sediments, it does not predict the gradual increase in water ^{137}Cs activity concentrations over a period of months after the countermeasure application. The key long term dynamics of the ^{137}Cs in the water are, however, predicted accurately (Figure 4).

The increase in ^{137}Cs in the lake water reduces the efficiency of the countermeasure. This is an unavoidable consequence of increasing the concentration of a competitor ion (i.e. K^+) in the lake water.

Modelling the decline of ^{137}Cs in fish as a result of the countermeasure.

The change in ^{137}Cs in fish as a response to the increased potassium concentration is determined by the rate of uptake and release of ^{137}Cs by the fish (Smith et al. 2002)

$$\frac{dC_f}{dt} = k_f C_w - k_b C_f \tag{3}$$

where k_b is the rate of excretion of ^{137}Cs (d^{-1}) and k_f ($l\ kg^{-1}\ d^{-1}$), the rate of uptake of ^{137}Cs by fish, is inversely proportional to the $[K^+]$ in the lakewater:

$$k_f = \frac{y}{[K^+]} k_b \tag{4}$$

where y (mEq kg^{-1}) is an empirically determined constant.

The modelling was carried out assuming time changes in $[K^+]$ and ^{137}Cs in the lakewater as predicted above, and using a numerical solution of Equations 3 & 4. The excretion rate of ^{137}Cs in perch, pike and brown trout (*Salmo trutta* L.) was determined for a range of lakes (Crummock Water, Devoke Water, Ennerdale, Loweswater, Windermere, UK, Hillesjon, Sweden, Ijsselmeer, Holland, Iso Valkjaarvi, Finland and the Kiev Reservoir) of $[K^+]$ concentration varying between 0.01 and 0.175 mEq l^{-1} (Smith et al. 2002). In these lakes, the average excretion rate constant was $k_b = 0.0013\ d^{-1}$. This value was used for the model prediction. The uptake rate constant, k_f, was calculated using the equilbrium condition, $CF = k_f/k_b$ and using Equation (4). For L. Svyatoe (Kostiukovichy) prior to the experiment, CF = 22,130 l kg^{-1} at $[K^+]$ = 0.025 mEq l^{-1} giving, from Equation (4), y = 553 mEq l kg^{-1}.

Our model somewhat underpredicts the rate of decline in ^{137}Cs in perch after the countermeasure addition (Figure 5), but in general the prediction is good up to the present day. The model predicts that, as a result of the countermeasure, ^{137}Cs activity concentrations in fish will remain significantly reduced until around 2015.

The model predictions (Figures 4 & 5) can be considered to be “blind” predictions of ^{137}Cs in water and fish in Lake Svyatoe (Kostiukovichy) following the $[K^+]$ addition since the model parameters were independently estimated from studies of other lakes. The only required site specific input parameters were the $[K^+]$, $[NH_4^+]$ and ^{137}Cs concentrations in the lake water prior to the experiment, the ^{137}Cs activity concentration in fish prior to the experiment, and the $[K^+]$ concentration in water after the potassium was applied.

Applicability of the countermeasure

The potassium chloride treatment has been shown to effectively reduce radiocaesium activity concentrations in lake fish. It must, however, be noted that its application is limited to lakes with long water residence times (low flushing rate). As noted above, a similar experiment was carried out in Sweden after Chernobyl (Håkanson & Andersson, 1992), however, because of the high water turnover in the lakes studied, it was difficult to maintain a high $[K^+]$ content of the water and the results of the test were inconclusive. Thus, for many lake systems, repeated applications of the countermeasure would be required to maintain high potassium concentrations. In addition, it is unlikely that the countermeasure would be acceptable in lakes used for drinking water because of the increase in radiocaesium in the lake water. Though this may place a significant practical constraint on the use of the countermeasure, it is noted that lakes of low water turnover (“closed” lake systems such as those in the Chernobyl zone) tend to be more highly contaminated than those with high water turnover. Radiocaesium is more mobile in these “closed” lake systems (Bulgakov et al. 2002), so the countermeasure is more likely to be needed in “closed” lakes.

Conclusions

In this study we have developed a potentially viable countermeasure against bioaccumulation of radiocaesium in fish, and tested a model for prediction of the efficiency of the countermeasure. The experiment also represents the first test of a predictive model for the dynamics of radiocaesium in response to a major perturbation in potassium (its major competitor ion) in a whole lake ecosystem. This model allows prior assessment of the efficiency of the countermeasure in other contaminated systems, and could predict the dynamics of radiocaesium accumulation in case of future radiocaesium contamination events.

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Tables

Table 1. General description of the study lakes. The potassium experiment was carried out on Lake Svyatoe (Kostiukovichy): the table shows the potassium content of this lake prior to the countermeasure application.

Lake	Lake surface area, km^2	Depth (max), m	Lake volume, m^3	Lake $[K^+]$ mEq l^{-1}	^{137}Cs deposition* kBq/m^2
Tyumenskoye	0.085	4.5	226 720	0.0275	482.8
Svyatoye (Kostiukovichy)	0.25	5.1	717 950	0.025	1368.1
Svyatoye (Chechersk)	0.069	7.0	186 250	0.455	515.04
Kozhanovskoe	6	2.5	9×10^6	0.05	620
Kiev Reservoir	922	14.5	3.7×10^9	0.0775	very varied

* as of 01.01.1997

Table 2. Mean ^{137}Cs activity concentrations of fish from Lake Svyatoe (Kostiukovichy) in 1999 as a percentage of their value prior to the countermeasure application. Numbers in brackets show number of samples taken.

Species	**^{137}Cs in 1997 (n) Bq kg^{-1}**	**^{137}Cs in 1999 (n) Bq kg^{-1}**	**1999 value as % of 1997 value**
Roach	16020 (20)	6890 (19)	43.0 %
Perch < 100g	54870 (5)	19490 (4)	35.5 %
Perch > 100g	103850 (6)	40690 (39)	39.2 %
Rudd	14970 (10)	7760 (9)	51.8 %
Tench	10420[1] (4)	4200 (27)	40.3 %
Gudgeon	20940 (2)	15360[2] (5)	73.3 %
Goldfish	18740 (2)	7640 (7)	40.7 %
Ruffe	8270 (6)	3490 (2)	42.2 %
Pike	29320[1] (2)	16390 (6)	41.7 %
Crayfish	15170 (2)	5330 (2)	35.5 %

1. 1997 mean includes samples from May '98.
2. 1999 mean includes samples from September '98.

Figures

Figure 1. Change in K^+ and ^{137}Cs in the water of Lake Svyatoe (Kostiukovichy). Note that for illustrative purposes, units of K^+ are mg l^{-1} rather than mEq l^{-1}. Date of countermeasure application (date of ice melt, March 98) is indicated by the vertical dotted line. Range in model predicted increase in ^{137}Cs in water is shown by the two solid lines (see modelling section below).

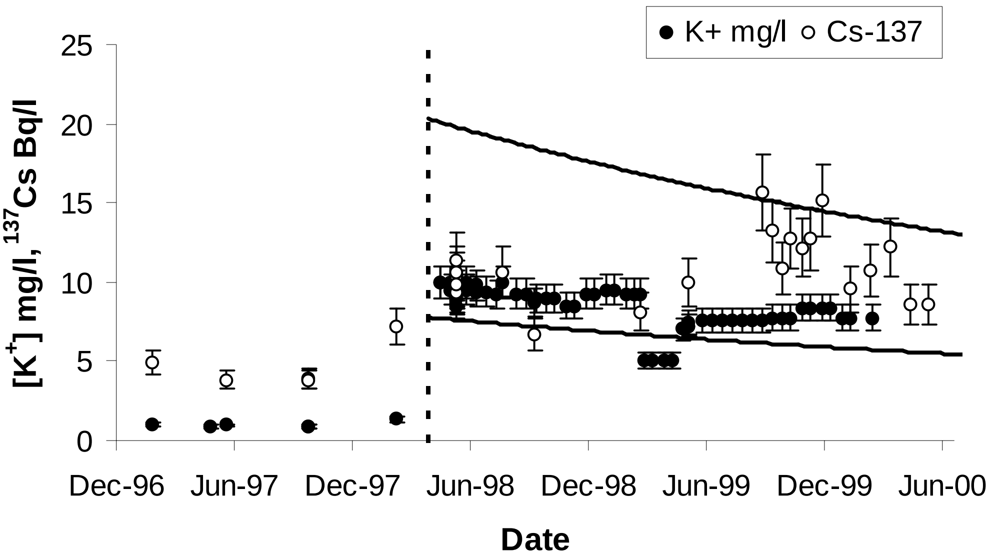

Figure 2. ^{137}Cs in (a) roach and (b) large and small perch in L. Svyatoe (Kostiukovichy) during the period 1997-99. The vertical dotted line indicates the time of countermeasure application. Error bars show 2 a range of 2 standard errors above and below the mean value, except where insufficient samples were available to estimate standard error.

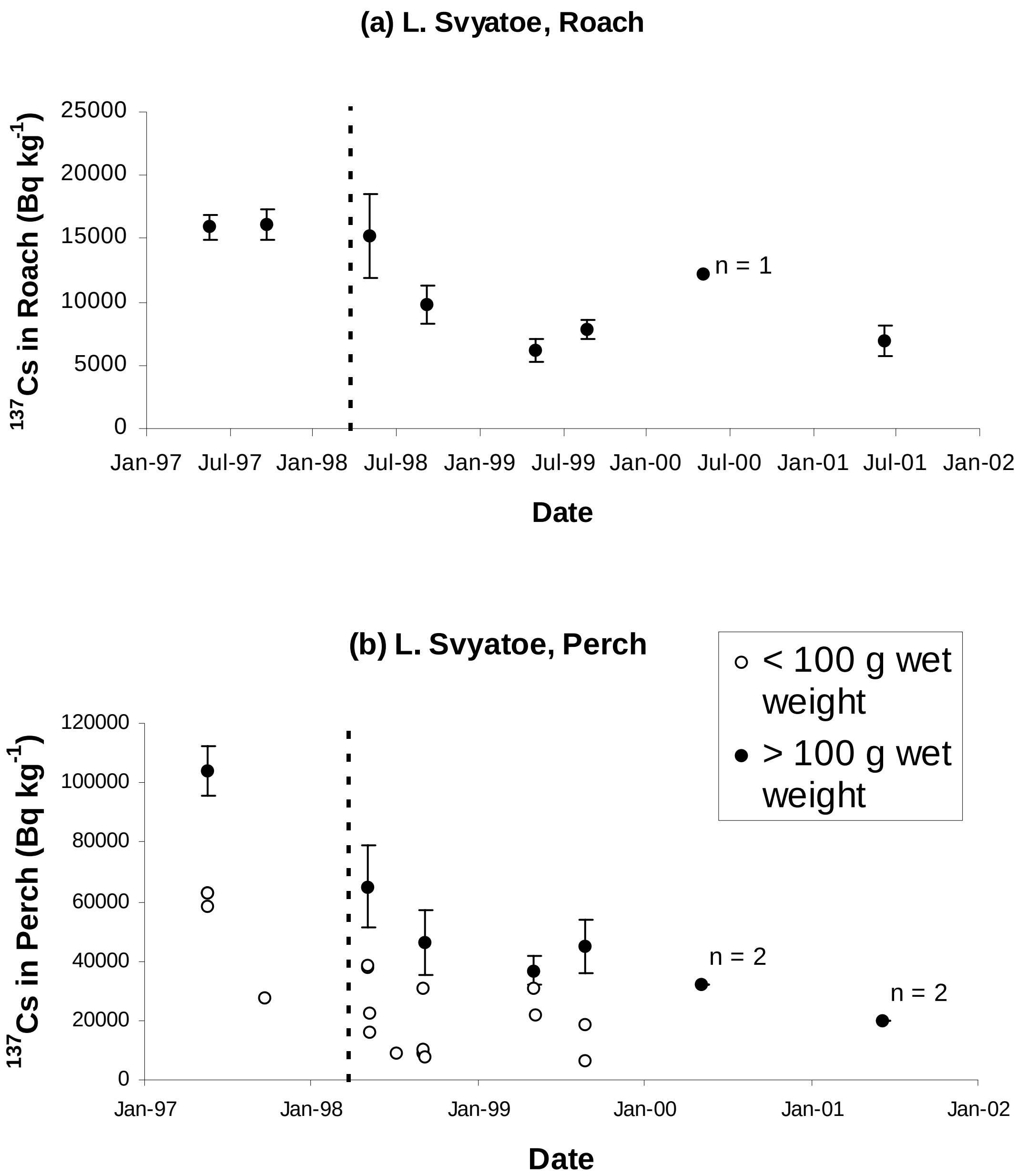

Figure 3. (a) and (b) ^{137}Cs in fish in the control lakes during the period 1997-99. The countermeasure was applied between the 1997 and 1998 sampling seasons.

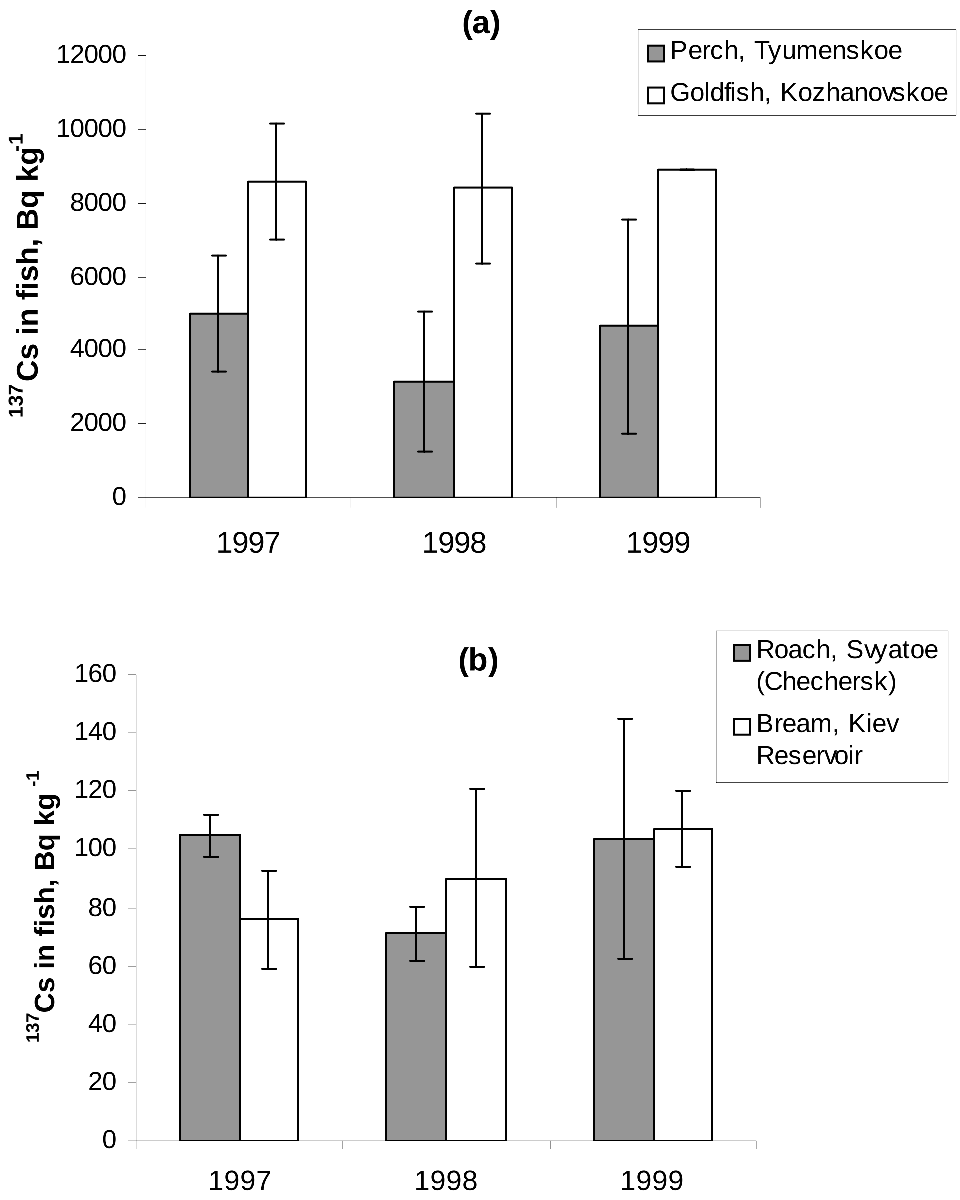

Figure 4. Predicted ^{137}Cs in water from time of countermeasure application,1998 to 2013. Measured values of ^{137}Cs and [K^+] to the year 2000 are shown. Note that for illustrative purposes, units of K^+ are mg l^{-1} rather than mEq l^{-1}. The deep solid line shows the extrapolated [K^+] concentration tending towards the pre-countermeasure value (dotted line) approximately 15 years after the application. The data and model predictions for the early period are shown in greater detail in Figure 1.

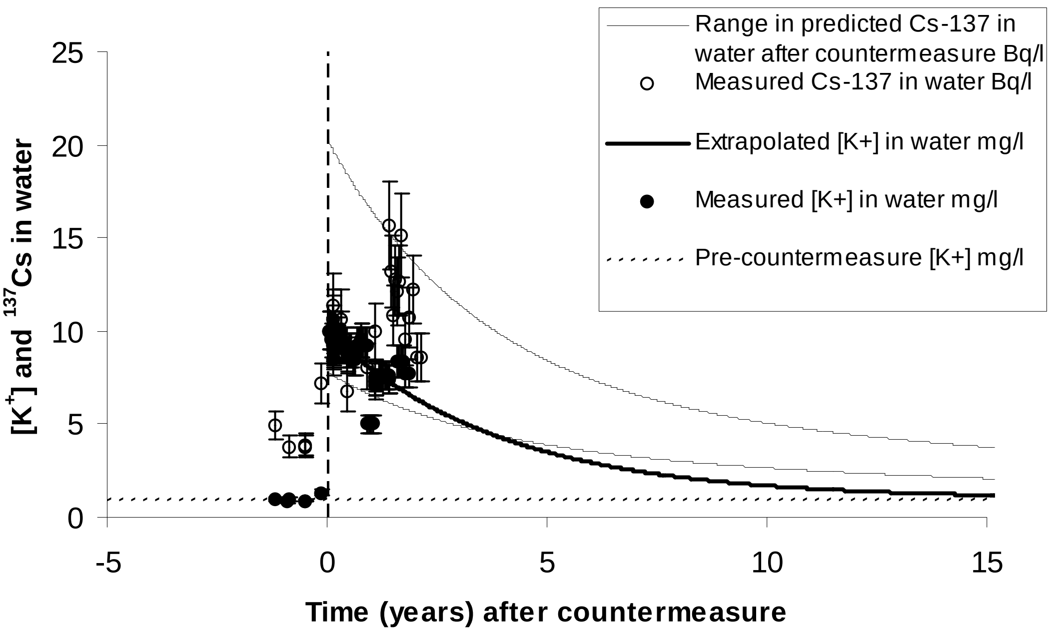

Figure 5. Predicted decrease in ^{137}Cs activity concentrations in perch in L. Svyatoe over a 15 year period after the countermeasure. Expected decrease in the absence of countermeasures is also shown assuming an effective ecological half life of 20 years, as observed in Lake Kozhanovskoe and the Kiev Reservoir (Smith et al. 2001).