

An experiment to test the addition of potassium to a non-draining lake as a countermeasure to ^{137}Cs accumulation in fish

A.V. Kudelsky, J.T. Smith¹ and A.A. Petrovich

Institute of Geological Sciences, Kuprevich Str. 7, Minsk 220141, Belarus

¹ *Centre for Ecology and Hydrology, Winfrith Technology Centre, Dorchester, Dorset DT2 8ZD, U.K.*

Abstract. Lake Svyatoye, a non-draining (closed) lake in Belarus was selected for a whole-lake experiment into the effect of potassium on ^{137}Cs accumulation in fish. The lake is in a zone of ^{137}Cs contamination of more than $1,480 \text{ kBq/m}^2$, K^+ concentration of the lake water (1.0 mg/l) prior to the experiment was relatively low, and ^{137}Cs activity concentration ranged from 3.8 to 4.9 Bq/l . Prior to the experiment, the ^{137}Cs activity concentrations in fish were amongst the highest of all aquatic systems affected by Chernobyl, being from 8.4 to 17.5 kBq/kg (wet weight) for rudd, from 12.4 to 16.7 kBq/kg for roach; from 58 to 105 kBq/kg for perch and up to 56 kBq/kg for pike. To study the effect of the dissolved K^+ concentration upon ^{137}Cs accumulation and retention in fish, $14,535 \text{ kg}$ of potassium fertilizer was spread over the ice cover of the lake. After the application of KCl fertilizer, the K^+ concentration in the lake water increased to 10 mg/l . An increase of $[\text{K}^+]$ in water resulted, as expected, in an increase in ^{137}Cs activity concentration in water (to 9.55 - 15.1 Bq/l , with average 12.17 Bq/l), as a result of ^{137}Cs desorption from bottom sediments. In spite of the increase in ^{137}Cs in the water, the K^+ addition resulted in a steady decrease of ^{137}Cs activity concentration in different fish species during the experiment (from 1998-2000) to approximately 30% to 50% of their original values.

1. INTRODUCTION

Many laboratory experiments, and field studies of weapons test fallout [e.g. 1-6] have shown that ^{137}Cs accumulation in the biota of aquatic ecosystems is directly proportional to the ^{137}Cs activity in water and inversely proportional to the $[\text{K}^+]$ concentration in lake water. This hypothesis was supported by studies of the relation between Chernobyl ^{137}Cs and $[\text{K}^+]$ in 14 contaminated lakes in Belarus, Russia and Ukraine [7]. On the basis of the inverse relationship between ^{137}Cs and $[\text{K}^+]$ we carried out an experiment to add K^+ to the water of Lake Svyatoye, Belarus, in order to decrease ^{137}Cs activity concentration in fish. Though the potential effectiveness of such a countermeasure had 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 [8] however because of the high water turnover in the lakes studied, it was difficult to maintain a high $[\text{K}^+]$ content of the water and the results of the test were inconclusive. Our test lake, Lake Svyatoye, in common with many lakes in this area of Belarus, has a long water retention time and thus allowed a large increase in the $[\text{K}^+]$ in the water to be sustained.

Initial field studies on 14 lakes in the contaminated regions of Belarus [7] were carried out to select a site for the field experiment. Many of the lakes in this area are non-draining ("closed") lakes and the ^{137}Cs in water and fish in these lakes has remained higher than in rivers, and in lakes with a rapid water turnover rate. The potassium content of the 14 lakes varied from 0.9 - 1.35 mg/l in lakes in semi-natural ecosystems to up to 20 mg/l in lakes found within regions with high agricultural activity (as a result of fertiliser runoff). Low fish-water concentration factors in the lakes with high $[\text{K}^+]$ [7] confirmed the potential effectiveness of the fertiliser experiments.

2. SITE DESCRIPTION AND METHODS

Extensive field studies were carried out on our study lake (Svyatoye, Belarus) and on 13 other lakes in Belarus and Lake Kozhanovskoe, Bryansk Region, Russia (some of this data can be seen in ref. [7]). Over a period of three years (1997-1999) samples of fish were taken using gill nets twice yearly from the lakes. In total 2173 individual fish were caught, 1022 fish being from Svyatoye lake. Among the fish species, pike comprised 0.74%, roach 22.87%, rudd 15.09%, tench 1.7%, gudgeon 5.52%, bleak 0.28%, bitterling 5.48%, crucian carp 4.33%, bream 1.2%, silver bream 0.41%, minnow 2.85%, and perch 24.1%.

The non-draining (closed) lake Svyatoye is located 30 km southeast of Kostyukovichy in a zone of ^{137}Cs contamination more than $1,480 \text{ kBq m}^{-2}$. The water surface area of the lake is 0.25 km^2 , and maximum depth is 5.2 m, the lake having total water volume $717,950 \text{ m}^3$. Seasonal water level variations in the lake are as high as 0.5 m, which is equivalent to variations in the water volume within the range $717,950 \pm 125,000 \text{ m}^3$. The water mineral content in various seasons varied from 85 to 155 mg l^{-1} : K^+ and NH_4^+ contents changed from 0.95 to 1 mg/l and from 0.2 to 0.6 mg/l , respectively. The ^{137}Cs concentration ranged from 3.8 to 4.9 Bq l^{-1} . The average specific activity of ^{137}Cs in bottom sediments in the upper 6 cm layer was 82.9 kBq kg^{-1} dry weight (0-2 cm: 147 kBq kg^{-1} ; 2-4 cm: $72.29 \text{ kBq kg}^{-1}$ and 4-6 cm: $29.42 \text{ kBq kg}^{-1}$). In 1997, prior to our experiment, ^{137}Cs activity concentration in fish varied from 8.4 to 105 kBq kg^{-1} wet weight: for perch from 58 to 105 kBq kg^{-1} ; roach from 12.4 to 16.7 kBq kg^{-1} ; rudd from 8.4 to 17.5 kBq kg^{-1} ; and in pike up to 56 kBq kg^{-1} .

To study the effect of dissolved $[\text{K}^+]$ upon the uptake and elimination processes of ^{137}Cs in fish, 14,535 kg of potassic fertilizers were spread over the ice cover of Lake Svyatoye on February 15, 1998. 94.31 % of the fertilizer mass was proper KCl, 5.69 % was identified as ammonium phosphate. KCl is 52.5 % potassium and 47.5 % chlorine with a small amount of Fe. The ^{40}K activity of fertilizer used in the experiment was 11.41 kBq/kg .

The fertiliser was added to the lake water by distribution over the ice-covered surface of the lake during February 1998. The KCl was completely dissolved in the lake water, as the ice melted, by 1 April, 1998. From that time until February, 2000, hydro- and radiochemical monitoring of lake water every 2 weeks and of fish every May and September has been carried out.

Laboratory studies were carried out to measure the ionic composition and ^{137}Cs contamination of water, preparation of fish for analysis (removing the head, tail and organs, drying, etc.) and subsequent determination of fish ^{137}Cs activity concentration using a low background "Canberra" γ -spectrometer with NaI detector.

3. RESULTS AND DISCUSSION

The range in ^{137}Cs content of water (C_w) in all the study lakes was from 0.2 to 7.2 Bq l^{-1} (in the surface water layer). Radiocaesium originated in the initial direct radionuclide fallout to the lake water surface, as well as from subsequent ^{137}Cs washout from catchments and remobilisation from bottom sediments. Groundwater in soils immediately surrounding the lakes showed rather low ($0.12\text{-}0.5 \text{ Bq l}^{-1}$) concentrations of this radionuclide and so was not considered as an important source of dissolved ^{137}Cs .

The ionic composition of the Svyatoye lake water as of May 25, 1997 was of the hydrocarbonate calcium-magnesium type with a mineral content of 101.9 mg l^{-1} and a low K^+ content (0.95 mg l^{-1}). Prior to the experiment, the ^{137}Cs concentration ranged from 3.8 to 4.9 Bq l^{-1} . After the application of KCl fertilizers the K^+ content of water increased to 10 mg l^{-1} (Figure 1) and mineral content did not significantly change (analysis of April 20, 1998). When the maximum K^+ concentration (10 mg l^{-1}) was reached, it began to decline steadily (most likely due to biological intake), but since May, 1999 has stabilized between 7.5 and 7.75 mg l^{-1} (Fig.1).

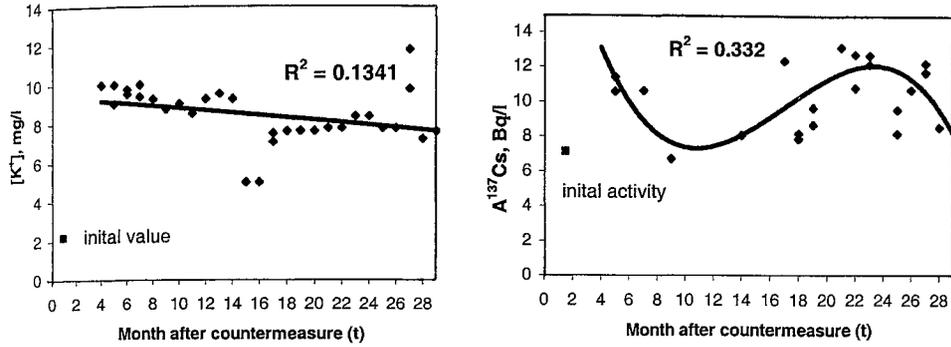


Figure 1: The $[K^+]_w$ and ^{137}Cs concentration (C_w) in water of Svyatoye lake (0-20 cm layer) after adding (on 13.02.98) potassium (sampling period February 1998 – May 2000).

The variation in time of the ^{137}Cs activity concentration (Fig.1) shows a more complicated pattern than that of $[K^+]_w$. This is due to sorption-desorption from bottom sediments and biological uptake in the water column. In addition, evaporation of the water volume and "freezing out" of ^{137}Cs from the ice cover during winter can also affect concentrations of ^{137}Cs .

An increase of $[K^+]_w$ in the water resulted in the ^{137}Cs activity concentration, C_w , increasing in water due to desorption from bottom sediments firstly to 9.3-11.4 Bq l⁻¹, and then by the end of experiment (September, 1999 – February, 2000) to 9.55-15.1 Bq l⁻¹ (with average 12.17 Bq l⁻¹). The ratio of $C_w/[K^+]_w$ firstly decreased from 4 Bq mg⁻¹ (value prior to the experiment) to 0.77-0.87, then increased during 1999 year (May-December) to 1.81-2.06 and by the end of experiment again decreased to 1.19-1.13 (Figure 2). The ^{137}Cs variation in fish (Bq kg⁻¹ w.w.) is closely associated with changes in the ratio $C_w/[K^+]_w$: observed correlations were: $^{137}Cs_{pike} = 12980(C_w/[K^+]_w)^{0.7945}$, $R^2 = 0.87$; $^{137}Cs_{roach} = 3530(C_w/[K^+]_w)^{1.06}$, $R^2 = 0.85$; $^{137}Cs_{perch} = 13000(C_w/[K^+]_w)^{1.93}$, $R^2 = 0.71$. Therefore, the relatively stable ratio of $C_w/[K^+]_w$ (Figure 2) implies that the effect of the countermeasure will continue for some years to come.

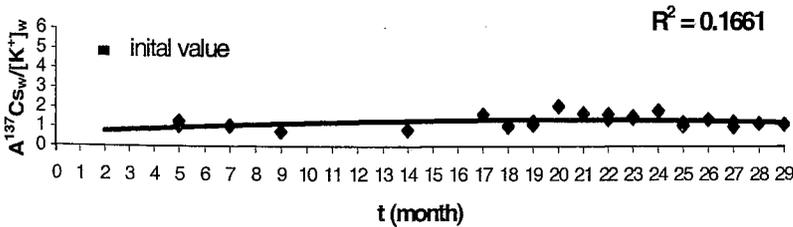


Figure 2: Behaviour of the ratio $^{137}Cs_w/[K^+]_w$ in water of Svyatoye lake (monitoring during February 1998 – May 2000).

The K^+ addition to lake Svyatoye resulted in a steady decrease of ^{137}Cs in fish. Decreases were observed in rudd (33-64%), perch (10-76%), roach (9-65%) and pike (9-56%). During the experiment the ^{137}Cs activity concentration in fish decreased to an average of 27.5% of the original value for pike, 37.5% for roach and perch and 47% for rudd (Fig.3). Thus the countermeasure reduced the activity concentration in fish by a factor of 2-3.

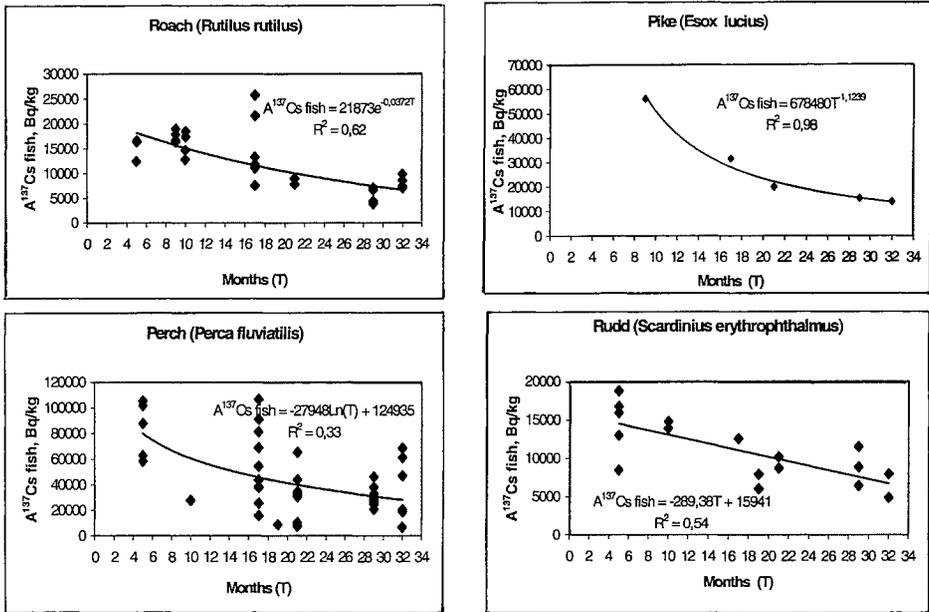


Figure 3: Rate of decrease of ^{137}Cs specific activity of fish after adding potassium (February, 1998) to the Svyatoye lake water (May 1997 – September 1999).

In the absence of countermeasures, ^{137}Cs activity concentrations in fish were expected to decline only very slowly [9, 10] over the period of study. This was confirmed by our extensive field studies in other lakes in Belarus. The results of this monitoring showed that there was no significant decrease in ^{137}Cs activity concentration in fish over the study period. This was in contrast to the significant declines observed in fish from Lake Svyatoye after the countermeasure was applied.

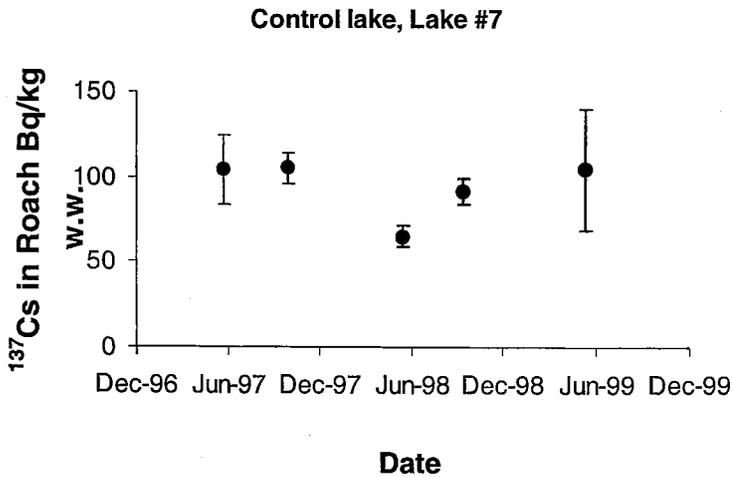


Figure 4: ^{137}Cs in roach in one of the control lakes, lake #7. No significant decrease in ^{137}Cs activity concentration in fish was observed in the control lakes during the period of study.

A directly inverse relation ($R^2 = 0.74-0.89$) between ^{137}Cs in fish and K^+ concentration in water was determined for the reference group of lakes [7]. In the absence of other factors one would expect that an increase in the $[\text{K}^+]$ concentration of a factor of approximately 10 would lead to a decrease in activity concentration in fish by a factor of 10. However, we observed, as expected, an increase of ^{137}Cs in the lakewater as a result of increased competition from $[\text{K}^+]$ and $[\text{NH}_4^+]$ and hence increased remobilisation of ^{137}Cs from bottom sediments. This reduces the efficiency of the countermeasure, but does not negate its effect.

4. CONCLUSIONS

By applying KCl fertiliser to the ice-covered surface of Lake Svyatoye, we increased the dissolved potassium concentration of the lakewater by a factor of approximately 10. This reduced the ^{137}Cs activity concentration in fish by a factor of 2-3, the efficiency of the countermeasure, as expected, being reduced by desorption of ^{137}Cs from bottom sediments. The long retention time of potassium in the lakewater meant that a single dose of fertiliser was effective for many years. The relatively stable ratio of ^{137}Cs (water phase)/ $[\text{K}^+]$ implies that the reduced ^{137}Cs in fish will continue for some years to come. Complete assessment of the countermeasure effectiveness, however, requires modelling of uptake and excretion from the fish and transfers of ^{137}Cs between water and sediments. The efficiency of the countermeasure could potentially be improved by adding a Cs specific sorbent such as a zeolite in parallel with the fertiliser addition.

Acknowledgments

The work was supported by the State Programme of the Republic of Belarus for elimination of consequences of the Chernobyl NPP accident, by the CEC Inco-Copernicus programme, and by the British Natural Environment Research Council. The authors would like to thank R.H. Haddingh, G. van Aersen, I.N. Ryabov, N. V. Belova, S. V. Ovsiannikova, V. I. Pashkevich and A. M. Petrikov for their contribution to this work and useful discussions.

References

- [1] Blaylock B.G., "Radionuclide Data Bases Available for Bioaccumulation Factors for Freshwater Biota", in *Environmental Effects* edited by R.O.Chester and C.T.Garten, Jr., (1982) pp. 427-438.
- [2] Carreiro C., Nasvit O., Corisco J., Romanenko V., Fomovsky V., Jurchuk L., Solomatina V., Belyaev V., "Experimental studies on some environmental parameters that may affect radiocaesium biological half-life" in *Experimental collaboration project №3: Modelling and study of the mechanisms of the transfer of radioactive material from terrestrial ecosystems to and in water bodies around Chernobyl* edited by U. Sansone and O.V. Voitsekhovitch, ECSC-EC-EAEC, Brussels-Luxembourg (1996) pp. 106-119.
- [3] Fleishman D.G., "Accumulation of Artificial Radionuclides in Freshwater Fish", in *Radioecology* edited by D.Greenberg, John Wiley & Sons, New York (1973) pp. 347-370.
- [4] Kudelsky A.V., Smith J.T., Pashkevich V.I., Ovsiannikova S.V., Petrovich A.A., The influence of potassium on the behaviour of ^{137}Cs in freshwaters of the radiocontaminated regions (Belarus). *Proceedings of the Academy of Sciences of Belarus*, **42**, 125-128 (1998) (in Russian).
- [5] Preston A., Jefferies D.F. and Dutton J.W.R., The Concentrations of ^{137}Cs and ^{90}Sr in the Flesh of Brown Trout Taken from Rivers and Lakes in the British Isles Between 1961 and 1966: the Variables Determining the Concentrations and their Use in Radiological Assessments, *Wat. Res.*, **1**, 475-496 (1967).
- [6] Rowan D.J., & Rasmussen J.B., Bioaccumulation of radiocaesium by fish: the influence of physicochemical factors and trophic structure. *Canadian J. of Fish. Aquat. Sci.*, **51**, 2388-2410 (1994).
- [7] Smith J.T., Kudelsky A.V., Ryabov I.N., Haddingh R.H., Radiocaesium concentration factors of the Chernobyl - contaminated fish: a study of the influence of potassium, and "blind" testing of a previously developed model. *J. Environmental Radioact.*, **48**, 359-369 (2000).

- [8] Håkanson L. and Andersson, T. Remedial measures against radioactive caesium in Swedish lake fish after Chernobyl. *Aquatic Sci.* **54**, 141-164 (1992).
- [9] Jonsson, B., Forseth, T., Ugedal, O., Chernobyl radioactivity persists in fish. *Nature* **400**, p. 417 (1999).
- [10] Smith, J.T., Comans, R.N.J., Beresford, N.A., Wright, S.M., Howard, B.J., Camplin, W.C., Chernobyl's legacy in food and water. *Nature*, **405**, p141 (2000).