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Report issued on 05/10/99

Project IC15 CT98-0205

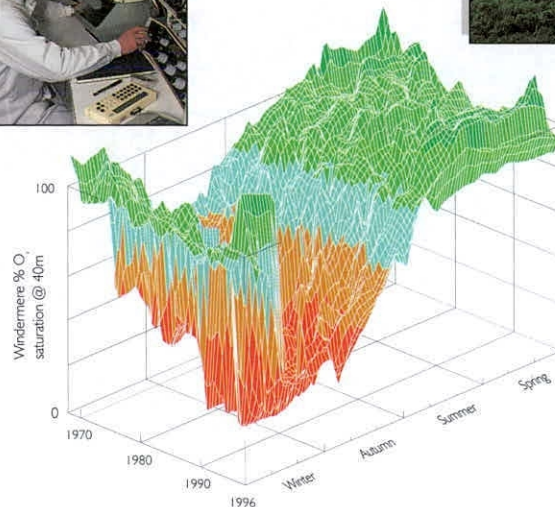
Start date: 01/09/98; duration 3 years

EU Funding: ECU 240,000

**Aquifers and Surface Waters in the Chernobyl area:
Observations and Predictive Evaluation**

AQUASCOPE

Keywords: Chernobyl; lakes; rivers.



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Executive summary

The purpose of this project is to develop general criteria and models for radionuclide mobility to and in aquatic systems. A key aspect of the project is the testing of existing aquatic radionuclide transport models against empirical data, and improvement of these models on the basis of detailed field and laboratory studies.

Measurements have been made of radiocaesium (^{137}Cs) activity concentrations in:

- catchment soils (including vertical profiles) at 12 sites;
- lake waters and lake sediments (6 lakes);
- river waters (5 rivers);
- ground waters (22 sites).

For many of these sites, long time series data is available from previous EC and CIS funded projects. These data have been collated and placed in the project database which has been complemented by data from further literature studies. Thus far, data has been collected for 25 lakes and 6 rivers in Belarus, Russia, Ukraine, UK and Germany. Additional data from rivers in Ukraine is currently being collated.

Analyses of the specific sorption capacity of ^{137}Cs (Frayed Edge Site capacity or FES), and its exchangeability, have been carried out on selected soil and sediment samples. A novel method of measuring FES, based on the Langmuir isotherm, has been developed. A further study of the influence of competing ions (particularly K^+) on radiocaesium sorption kinetics and mobility is in progress.

In lakes which have long water residence times ("closed systems"), long-term ^{137}Cs concentrations in water and fish tend to be much higher than in lakes which have more rapid flow through of water from the catchment. A model for the prediction of long-term ^{137}Cs activity concentrations in closed lake systems has been developed and is currently being tested. A model for ^{137}Cs runoff from catchments (developed in a previous EC programme, ECOPRAQ) has been tested. Initial results of a "blind" test show that the model gives reasonable predictions of time-dependent concentrations of radiocaesium in runoff waters. Modelling has also been carried out on radiocaesium activity-depth profiles in soils. Models for the change in radiocaesium transport parameters as a function of time are currently being tested.

Dissemination of results has been carried out by the production of a number of scientific papers in both CIS and Western scientific journals. In addition, the results of a meeting in Weingarten, Germany were covered by 2 local newspapers, and by German national radio. A "popular science" article has been published in the magazine of the UK Natural Environment Research Council.

1. Management progress report.

Collaboration and meetings.

Meetings involving all partners were held in the United Kingdom (September 1998), Belarus (May 9-16 1999) and in Germany (4-10 July 1999). During these meetings experimental methods and approaches were discussed and joint fieldwork carried out. Joint sediment and water sampling was carried out in Germany by FHWEIN, TYPHOON, IGSB, and UHMI and sampling methods were compared. Joint sediment sampling was carried out in Belarus (January 1999) by IGSB, TYPHOON and UHMI. Joint sediment analyses were carried out between UHMI and IGSB. Prof. Anatoly Kudelsky (IGSB) visited IFE for discussions of results (5-12 September) and attended the UK Environmental Radioactivity Forum. Dr. Alexei Konoplev visited FHWEIN for collaborative studies (July 1999) on L. Constance.

J.T. Smith (IFE) and A.V. Konoplev (IGSB) presented papers at the Radionuclide Speciation Workshop, Edinburgh, 1999. J.T. Smith (IFE) presented a paper at the UK Environmental Radioactivity Forum.

Two field trips are planned for next year: in May and September 1999, in Ukraine and Belarus. A meeting of all partners is planned in Lisbon in autumn of 2000.

Problems.

There have been no significant problems with management or administration of the contract. Difficulties have been encountered in transferring project monies to the CIS partners, however, such difficulties were not unexpected, and we hope have largely been overcome.

2. Scientific progress report.

Project global objectives.

- (1) To investigate long-term radiocaesium and radiostrontium dynamics in rivers, lakes, and groundwaters.
- (2) To develop simple, predictive, physically based and empirical models for prediction of long-term radiocaesium dynamics in these systems.
- (3) To investigate the long-term dynamics of radiocaesium solid/water distribution in suspensions of soils, sediments and clay minerals as a function of the water phase chemical composition.

Objectives for the reporting period.

- (1) To carry out sampling of water, bottom sediments and soils on catchments of rivers and lakes in Belarus, Russia, Ukraine and Germany.
- (2) To begin long-term laboratory investigations of radiocaesium fixation and exchangeable adsorption in suspensions of soils, sediments and clay minerals.
- (3) To carry out a literature review of existing experimental data and mathematical models of radiocaesium and radiostrontium behaviour in soil-water systems.
- (4) To create a database for testing mathematical models of radionuclide transfer in aquatic systems.

Progress.

Progress is outlined according to the workpackage scheme detailed in the contract Technical Annexe. WP1 (project management) is summarised in the management section above. The progress report is written as a coherent whole without specific reference to individual partners. The individual contribution of the different partners is covered in the Partner Progress Reports in the next section.

Workpackage (2): Groundwater, river and lake sampling

Field studies were carried out at 2 Belorussian (Svyatoye (Bel) and Svyatskoye), 2 Russian (Svyatoye (Rus) and Kozhanovskoye) and 2 German (Vorseesee and Constance) lakes. Samples (5-10 litre) of lake water were collected during three sampling periods (February 1999, May 1999, September 1999) at the Russian sites and during two sampling periods (February 1999, May 1999) at Belorussian sites. One water sample (1330 l) from Lake Constance and two samples from Lake Vorseesee (200 l each) were collected using the large volume water sampler "Midiya" in July 1999. Concentrations of ^{137}Cs and ^{90}Sr have been measured in the samples collected before September 1999. Concentration of ^{137}Cs in water of Lake Constance and Lake Vorseesee was equal to $0,128\pm 0,026$ mBq/l and $33,75\pm 1,13$ mBq/l correspondently. Concentrations of ^{137}Cs in Belorussian and Russian lakes are given in Table 1. Major hydrochemical

characteristics of the lake water and ^{137}Cs content on suspended matter (lakes Vorsee and Constance) were also measured.

Table 1. Concentrations of ^{137}Cs and ^{90}Sr in the water of Belorussian and Russian lakes.

Lake	Concentration of ^{137}Cs , Bq.L ⁻¹	
	February 1999	May 1999
Svyatoe (Rus)	11.7	9.5
Kozhanovskoe	7.2	3.4
Svyatoe (Bel)	8.1	12.4
Svyatskoe	4.6	6.1

Water samples were also taken from five rivers in Belarus. A continued decrease in ^{137}Cs in river waters, and catchment runoff coefficients was observed (Fig. 1). The river basins were divided into two groups with a more rapid decrease of ^{137}Cs concentrations in the Sozh, Besed and Iput and a slower decrease in ^{137}Cs concentrations in the water of the Dnieper and Pripyat. We hypothesise that the different behaviour may be due to observed differences in mean slope of the two catchments. This hypothesis will be tested in the next year of the project by comparing the Belarus catchments with catchments of different slopes in Germany and the UK.

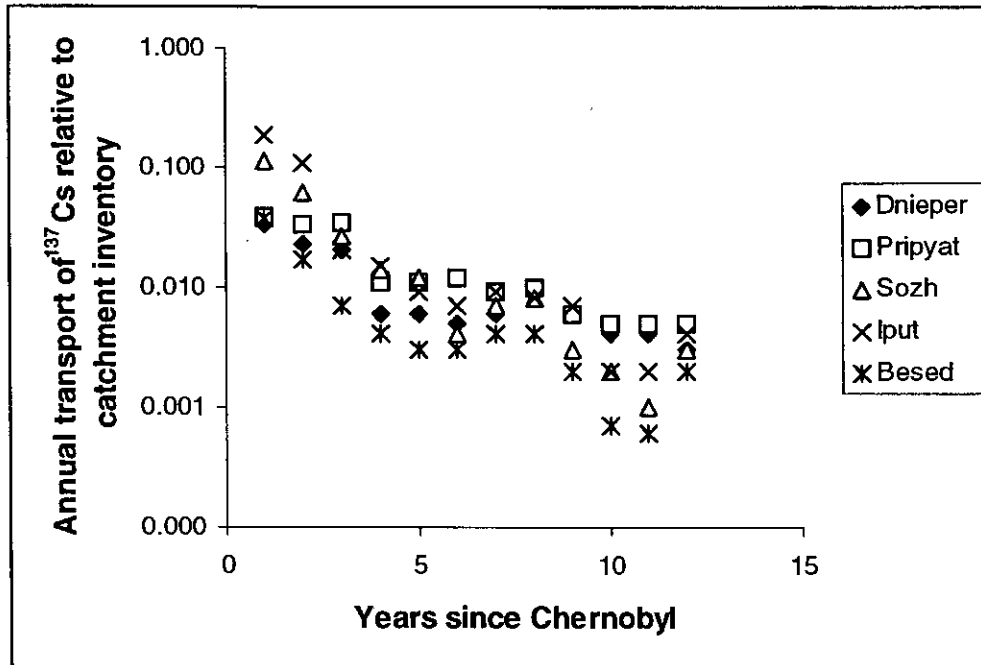


Figure 1. Transport of ^{137}Cs in Belorussian rivers as a percentage of fallout to the catchment.

Groundwaters

Groundwaters were sampled at a number of sites which have been monitored since 1987. These data have been compiled and are being put into the joint project database.

In addition, a field experiment was carried out in order to study the effect of agricultural fertilizers (KCl) on the vertical migration of ^{137}Cs to ground waters. Three sites were studied: a control, a site with KCl addition of 100 g/m^2 and one with an addition of 300 g/m^2 . Three months after fertiliser addition, soil profiles and ground water sampling was carried out at the three sites. Initial analyses have shown a significant increase in the migration of ^{137}Cs as a result of the KCl addition.

Database development.

A database of radiocaesium concentrations in surface waters has been created in EXCEL. Thus far, data has been collected (during this study, and from previous studies) for 25 lakes and 6 rivers in Belarus, Russia, Ukraine, UK, Germany. The data includes measurements of ^{137}Cs in water, lake sediments, catchments and fish, as well as key physical characteristics of the lake or river, and major ion chemistry. Where possible, literature studies have been carried out to obtain time series data of radiocaesium concentrations over the period since the Chernobyl accident.

Intercomparison of radiometric analyses

Because a number of participants are involved in providing radiometric analyses at different stages of the project, and these data to be used by co-partners “as it is“, it was decided during the first year to carry out some intercomparison exercises. The purpose of these exercises is to check the replicability between the results obtained in different labs and are not intended to be comprehensive qualitative analyses of analytical techniques used. Intercomparison consists of an interchange of environmental samples (bottom sediment, water, suspended matter etc.) processed in one lab with another followed by radiometric counting of the samples and a test to see if they fall into a defined range of gross (radio) activity. All the data sets obtained in different labs are being statistically compared against each other in order to make sure that replicability of sample measurements is acceptable. The first test between UHMI and IGS shows that the accuracy of radiometric data is sufficient ($R^2=0.96-0.99$) and does not exceed ca. 10% of standard error within the range of activity 10-100 Bq/per sample. There is a possibility to improve this further because of the systematic character of errors appearing in the lower range of activity. This will to be done after all the participants involved (namely – UHMI, IGS, TYPHOON, FHWEIN & ECN) contribute their data sets for statistical testing.

Workpackage (3): Soil radiometric analysis and measurement of RIP

Four soil monoliths were sampled in Briansk Region in the catchments of Kozhanovskoe and Svyatoe lakes. The vertical distribution of ^{137}Cs in monoliths and speciation of ^{137}Cs in the most radioactive soil layers were measured. Vertical distribution of ^{137}Cs in soils were determined in eight Belorussian sites. The main objectives of the soil sampling were:

- to refine data on ^{137}Cs fallout to the lake catchments
- to estimate the rate of ^{137}Cs transfer from soil to groundwater;
- to measure radiocaesium adsorption parameters of the soils.

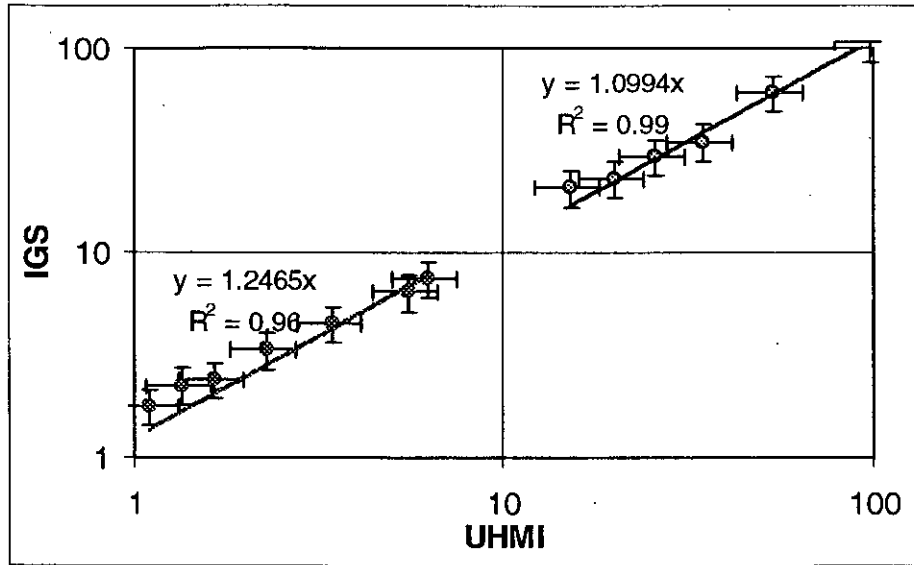


Figure 2 Intercomparison results – ^{137}Cs radiometric analysis provided by UHMI against data of IGS

A detailed study of radiocaesium distribution in the solid and aqueous phases of soil was carried out at a site on the floodplain of the river Besed. Depth distributions of competing cations, ^{137}Cs in the solid phase, K_d and predicted K_d are shown in Figure 3. Counting of ^{137}Cs activity in the pore-water from the lower parts of the profile are uncertain due to low pore water yield and low ^{137}Cs activity, so results are shown only for the upper 10 centimetres.

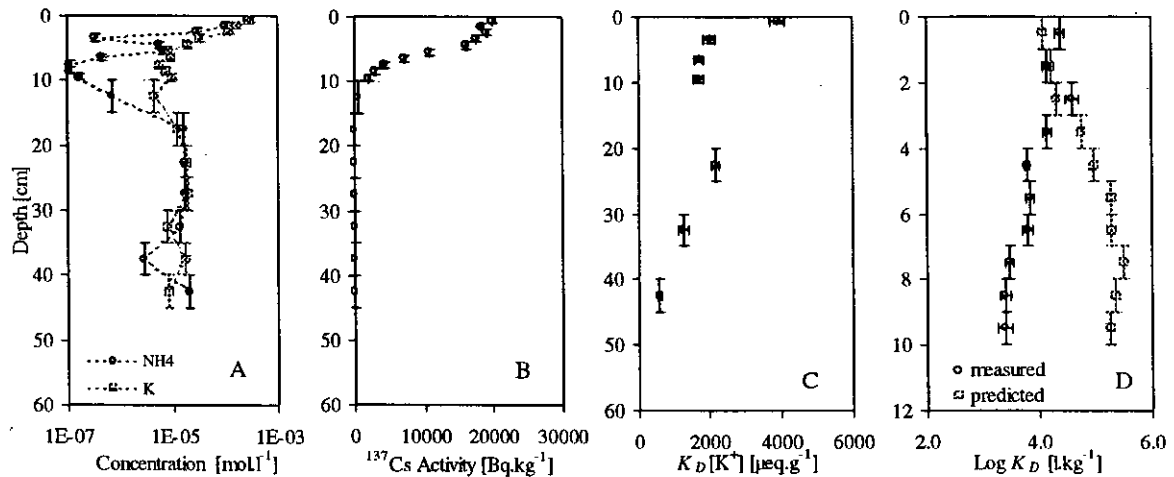


Figure 3 K^+ and NH_4^+ concentration (A), ^{137}Cs activity on the solid phase (B) and $K_D[\text{K}^+]$ as a function of depth (C). The in-situ measured and predicted K_D for the top ten centimetres of the profile are shown in D.

Workpackage (4): Sediments radiometric analysis and measurement of RIP

Sediment sampling

Samples of bottom sediments were collected in February 1999 in two Russian and two Belorussian Lakes (two cores per lake, 60 -80 cm each). One core from each lake was sliced in 2 cm layers, another one in 5 cm layers. Concentrations of ^{137}Cs were measured in the layers. Six cores of bottom sediments (from 40 cm to 1 m length) were collected from Lake Constance (Obersee) and seven cores were collected from Lake Vorsee in July 1999. One core from Lake Constance was sliced in 5 cm layers to measure ^{137}Cs vertical distribution. Three cores from Lake Constance were used in extraction experiments, one core from Lake Constance and two cores from Lake Vorsee were used for RIP^{ex}/FES measurements. In one core from each lake concentrations of ^{137}Cs , $[\text{Ca}^{2+}]$, $[\text{Mg}^{2+}]$, $[\text{K}^+]$, $[\text{Na}^+]$ and $[\text{NH}_4^+]$ in pore water were measured. In the solid phase, content of exchangeable form of ^{137}Cs and major cations are measured using a sequential extraction procedure (Pavlotskaya, 1974).

Frayed edge site (FES) measurements

The method used to the FES determinations (Cremers *et al.*, 1988) is based on the masking of the ion exchange sites using a suitable background level of silver thiourea (AgTU) being the action of the caesium ions restricted to the small number of specific adsorption sites that are inaccessible to the bulky silver thiourea complex. The experimental procedure consists firstly, on the sediments presaturation with silver thiourea (AgTU 0.015M). Afterwards, the sediments are equilibrated (24 hrs) with silver thiourea solutions containing increasing levels of caesium chloride (10^{-5} to 10^{-2} M) ^{137}Cs labelled. Finally, the suspensions are centrifuged and the caesium activity measured at the liquid phase by gamma spectrometry. The results are plotted as caesium adsorbed versus concentration in the equilibrium solution.

Figure 4 shows the radiocaesium sorption isotherms for Tejo river (Portugal) sediments. The sorption plateau values correspond to the FES capacity. However, as it can be seen mainly for A and S sediments the plateau on the sorption isotherm seems not be reached.

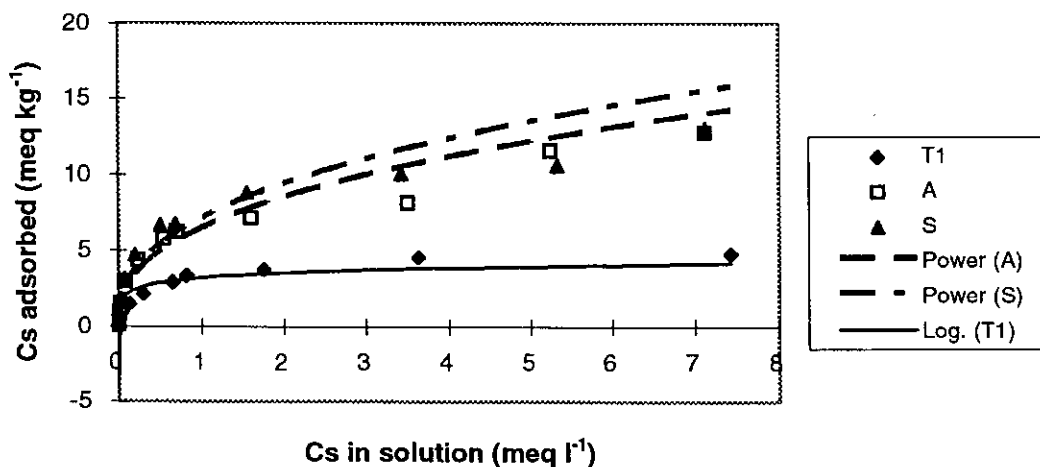


Figure 4 Caesium sorption isotherms in the frayed edge sites (FES)

Another calculation method was carried out which allows the FES determinations before the adsorption maximum has been reached. The procedure consists on the application of the Langmuir equation $X = \frac{X_m C}{k + C}$ to the radiocaesium sorption isotherms. The intercept resulting from the study of the dependence of the inverse concentration of caesium adsorbed on the inverse concentration of caesium in solution (Figure 5), this is, the linearization of the Langmuir equation, $\frac{1}{X} = \frac{k}{X_m} \left(\frac{1}{C} \right) + \frac{1}{X_m}$, corresponds to the inverse value of the maximum adsorption (FES capacity). Therefore, the FES values could be obtained from the last range of isotherm before sorption maximum, which corresponds to high caesium concentrations. The capacity of high affinity sites (HAS) can also be determined using the initial range of isotherm at low caesium concentrations.

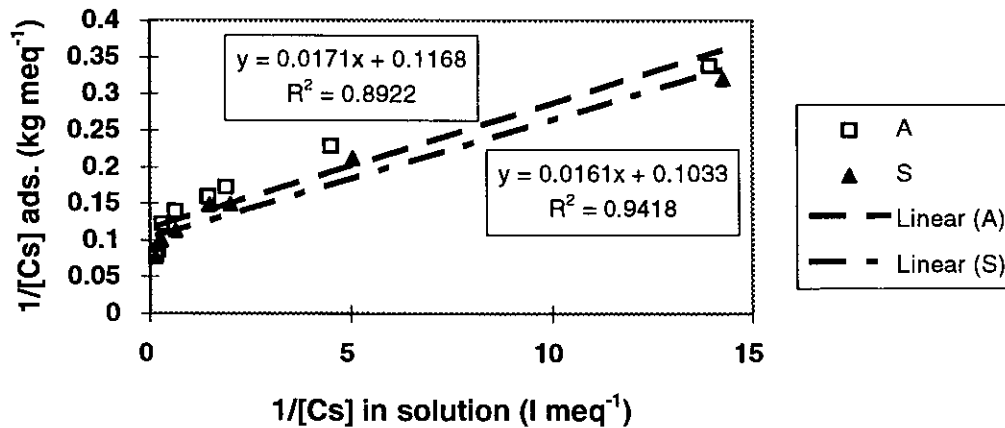


Figure 5 Inverse of the caesium concentration adsorbed in the sediments versus the inverse of the caesium concentration in solution to high caesium concentrations.

The values obtained for FES using the Cremers *et al.*, (1988) method and Langmuir calculations (FES and HAS) are presented in Table 2.

Table 2. Frayed edge sites (FES) determinations to Tejo river sediments (Portugal) using Cremers *et al.* method (1988) and Langmuir equation

Sediments	CEC	Cremers <i>et al.</i> ,1988		Langmuir
	cmol kg ⁻¹	FES meq kg ⁻¹	FES meq kg ⁻¹	HAS meq kg ⁻¹
T1	14.3	4.1±0.7	3.7	0.29 (7.9%)
A	27.9	9.9±2.7	8.6	0.63 (7.4%)
S	31.4	10.6±1.8	9.7	0.44 (4.5%)

It can be seen that FES values obtained from the Langmuir linearization are lower than those calculated from the radiocaesium sorption plateau. The HAS presented values which corresponds to about 4 to 8% of the FES.

Workpackage (5): Radiocaesium & radiostrontium sorption kinetics.

The aim of the laboratory experiment was to investigate the effects of changes in competing ion chemistry (principally K^+) on the dynamics of ^{137}Cs fixation by soils. Soils were equilibrated with water solution containing K^+ and Ca^{++} cations of different potassium adsorption ratios (PAR) as well as with natural river water collected from Besed' River (Belarus). Soil (60 g) was equilibrated three times with 1 L of solution for 1 day then dried. After passing the dry soil through a 1 mm sieve, 2 g aliquots of soil were put into 20 mL vials and equilibrated with 19.8 mL of corresponding solution for 1 day. Then 1500 Bq of ^{137}Cs in 0.2 mL was added to each vial of 120 vials (six variants x ten sampling sessions x two replicates). Then after 1, 5, 14, 33, and 63 days two vials of each variant were removed (total 12 vials each time) and the concentration of ^{137}Cs was measured in the water phase and in a 1 M CH_3COONH_4 extraction from the solid phase in order to determine the fraction of exchangeable ^{137}Cs .

The results of the ^{137}Cs dynamic in water phase of the soil:water slurries of different PAR value are shown in Fig. 6 for a mineral soil and in Fig. 7 for an organic soil. These experiments are ongoing.

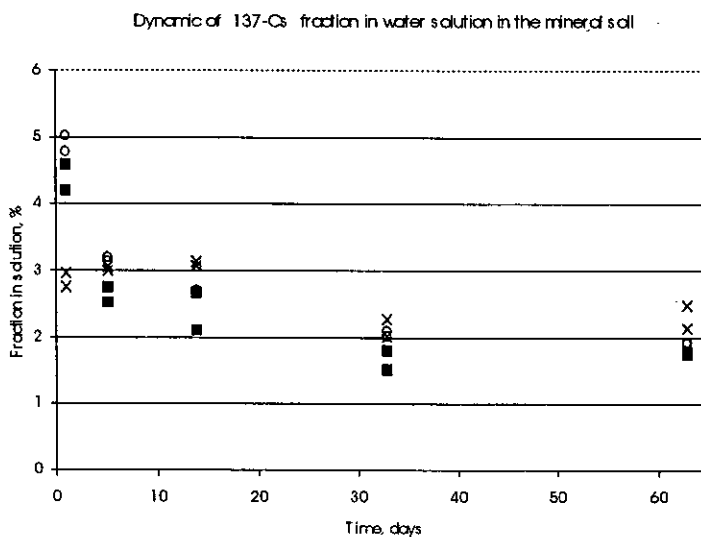


Figure 6. Dynamics of ^{137}Cs in water solution in mineral soil. Solid squares is soil - 0.1 mM K^+ + 1 mM Ca^{++} ; open circles - soil + 1 mM K^+ + 1 mM Ca^{++} ; crosses - soil + Besed' River water.

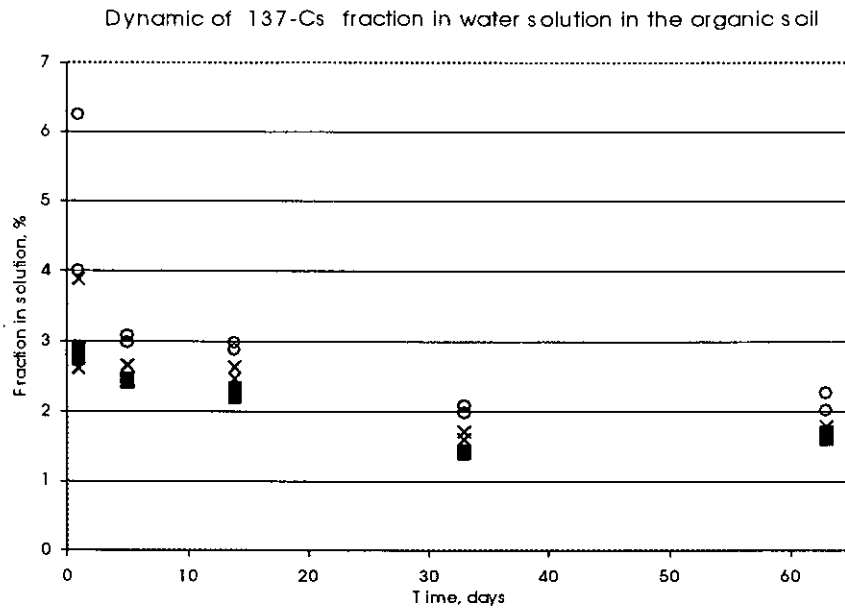


Figure 7. Dynamics of ¹³⁷Cs in water solution in organic soil. Solid squares is soil - 0.1 mM K⁺ + 1 mM Ca⁺⁺; open circles - soil + 1 mM K⁺ + 1 mM Ca⁺⁺; crosses - soil + Besed' River water.

Workpackage (6): Modelling

Model for predicting long term ¹³⁷Cs concentrations in waters of closed lake systems.

A model of radionuclide behaviour in a closed lake water and sediments was developed. The model equations were formulated and solved analytically. The following equation was obtained after simplification of the solution:

$$C_w = \frac{e^{-\lambda t} S}{K_p (\pi D_e t)^{0.5}}$$

where C_w - radionuclide concentration in water (Bq.m⁻³); S - surface density of sediments contamination (Bq.m⁻²); K_p - ratio of radionuclide concentration in the upper layer of sediments (Bq.m⁻³) to that in water (Bq.dm⁻³); D_e - effective diffusion coefficient in sediments (m².d⁻¹); λ - radioactive decay constant(d⁻¹); t - time after contamination, d.

The model is now being tested against new and previously obtained experimental data. The following data are used for the model parameter calculation (S , K_p , D_e):

- vertical distribution of ¹³⁷Cs in Svyatoe lake sediments and its concentration in water measured in 1993 during the CEC project ECP-3;
- vertical distribution of ¹³⁷Cs in Svyatoe lake sediments and its concentration in water measured in 1999 within the present project.

Calculated C_w values for ¹³⁷Cs will be compared with experimentally measured

concentration dynamics in 1992- 2000.

Modelling time-dependent ^{137}Cs activity concentrations in rivers.

Predictive models of radiocaesium transport are currently being tested using the empirical data sets developed in this project. Figure 8 shows a “blind” test of a river runoff model developed in the EC ECOPRAQ project against measurements of ^{137}Cs in the River Dnieper (Belarussian part). The model is largely driven by catchment coverage by different soil types (organic or mineral soils). The current model prediction is based on a crude estimate of percentage organic soils in the catchment. Future analyses of catchment characteristics will be carried out in order to improve this estimate.

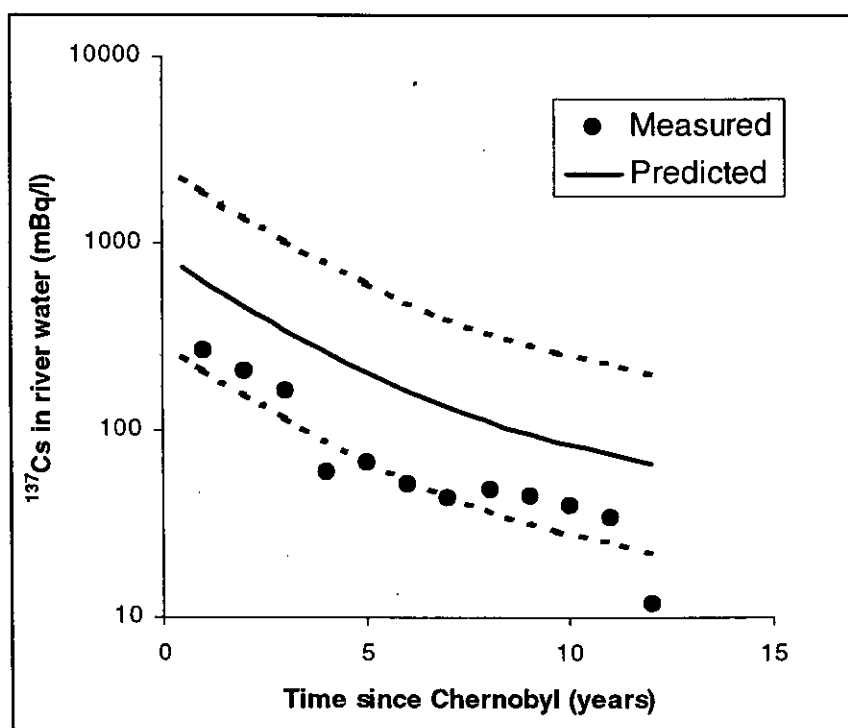


Figure 8. “Blind” prediction of ^{137}Cs activity concentration in the R. Dnieper at Rechitsa, Belarus. The model assumes a predominantly mineral catchment (95% mineral soils, 5% organic). Estimated errors are shown as dotted lines.

Modelling the mobility of ^{137}Cs and ^{90}Sr in soils and transfers to groundwaters

Following the fallout of radioactive caesium onto the soil, migration into the vertical soil profile is believed to be relatively rapid as a result of infiltration of rainwater. Sorption of radiocaesium to the soil matrix, particularly illitic clay minerals (Cremers et al., 1988), then rapidly slows the rate of migration. In the long term, most of the radiocaesium is strongly bound to the soil and is relatively immobile. After the atmospheric testing of nuclear weapons and the Chernobyl accident, the majority of deposited radiocaesium has remained in the top 30 cm of the soil profile for many years after fallout. Long term changes in the distribution of radiocaesium within the

top layers of the soil profile may, however, result in changes in external radiation exposure, transfers to vegetation, and runoff of radioactivity from contaminated soils to surface waters.

Three different models for the long term mobility of radiocaesium in soils are being tested: (i) a model which assumes that no fixation occurs, i.e. that dispersion and advection continues at a constant rate after fallout; (ii) a model which assumes that radiocaesium is completely immobile in the soil during the years after fallout; (iii) a model which assumes that radiocaesium mobility slowly changes during the years after fallout as a result of slow sorption reactions.

The 41 activity depth profiles we are using to test the different models were taken from published data, as well as measurements made during this project. The data set comprises profiles from organic and mineral soils in Cumbria, England (Horrill & Howard, 1991; Sandalls & Bennett, 1992; Smith et al., 1997), various soils in Ukraine (Ivanov et al., 1997), soils in Belarus (our measurements) and a grassland soil from Northern Italy (Chamard et al., 1993).

Analyses of these data are ongoing, but initial results show a gradual decline in mobility of radiocaesium in the soil over time after fallout (Fig. 9). There appears to be a tendency for the mobility of radiocaesium to decrease towards a constant value on long timescales after fallout. Further studies will attempt to evaluate long term (decades) mobility of ^{137}Cs and long term transfers to groundwaters.

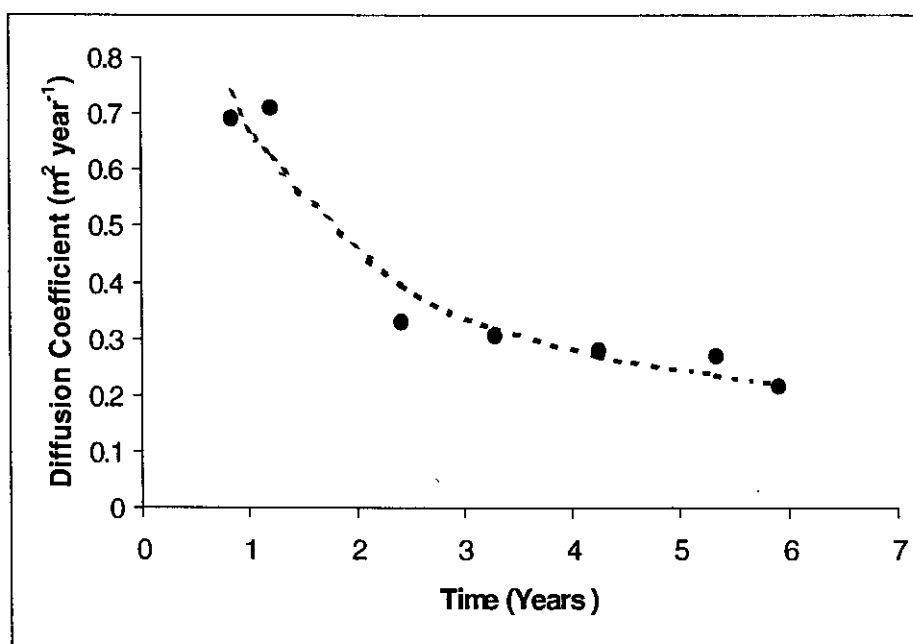


Figure 9. Change in average apparent coefficient of dispersion of ^{137}Cs in soils as a function of time since Chernobyl. Dotted line shows fit of time dependent advection dispersion equation.

Publications and papers.

Bulgakov A.A., Konoplev A.V., Scherbak, Yu.V. Shveikin. Experimental study and prediction of dissolved radionuclide wash-off by surface runoff from non-agricultural watersheds. In: I. Linkov and W.R. Shell (eds.), Contaminated Forests: Recent developments in risk identification and future perspectives, NATO ASI Series 2- Kluwer Academic Publishers, Dordrecht, 1999, pp. 103-112.

Konoplev A.V., Bulgakov A.A. Estimation and prediction of ^{90}Sr and ^{137}Cs distribution between solid and liquid phases of soil-water system. I. Transformation of speciation in soils Atomnaya Energiya (Atomic Energy), 2000, in press (In Russian)

Konoplev A.V., Bulgakov A.A. Estimation and prediction of ^{90}Sr and ^{137}Cs distribution between solid and liquid phases of soil-water system. II. Exchangeable distribution coefficient Atomnaya Energiya (Atomic Energy), 2000, in press (in Russian)

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Bulgakov A.A., Konoplev A.V. Diffusional model of radionuclide fixation in soils. 2. Parameter values estimation for some soils classes. (Submitted for Geokhimiya (Geochemistry), in Russian).

A.V. Kudelsky, J.T. Smith, O.M. Zhukova, I.I. Matveyenko, T.M. Pinchuk (1998) Contribution of river runoff to the natural remediation of contaminated territories (Belarus). Proceedings of the Academy of Sciences of Belarus, vol. 42 no. 6, 90-94 (in Russian).

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Research to be performed in the remainder of the project.

- Monitoring of ^{137}Cs concentration in lake and river water;
- Continuation of the long-term laboratory experiment
- Determination of parameter values for radiocaesium and radiostrontium vertical migration in soils and to investigate their relationships with soil properties.
- Investigation of FES and RIP correlation with content of exchangeable cations and the water phase chemistry of soils and sediments.

- Estimation of parameter values of the model of radiocaesium dynamics in a closed lake for Svyatoo Lake (Rus), Svyatoo Lake (Bel), Svyatskoe lake and the model testing against measured concentration dynamics.
- Modelling of radiocaesium transport in soils and bottom sediment.
- Continued testing and improvement of models for radiocaesium transport by surface runoff.
- Development of recommendations for the prediction of radiocaesium and radiostrontium long-term dynamics in lakes and rivers.

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3. Partner progress reports.

Partner 1 (Coordinator): Institute of Freshwater Ecology.

Objectives:

- To coordinate and monitor progress of the partners towards the project objectives.
- To participate in joint field sampling exercises
- To begin a database of radiocaesium activity concentrations in aquatic systems.
- To carry out mathematical analysis of field and experimental results.

Creation of database of ^{137}Cs activity concentrations in lakes and rivers

A database of radiocaesium concentrations in surface waters has been created in EXCEL. Thus far, data has been collected (during this study, and from previous studies) for 25 lakes and 6 rivers in Belarus, Russia, Ukraine, UK, Germany. The data includes measurements of ^{137}Cs in water, lake sediments, catchments and fish, as well as key physical characteristics of the lake or river, and major ion chemistry. Where possible, literature studies have been carried out to obtain time series data of radiocaesium concentrations over the period since the Chernobyl accident.

Modelling time-dependent ^{137}Cs activity concentrations in rivers.

We have begun analyses and predictive modelling of the assembled data set. Figure 1 shows a “blind” test of a river runoff model developed in the EC ECOPRAQ project against measurements of ^{137}Cs in the River Dnieper (Belarussian part). The model is largely driven by catchment coverage by different soil types (organic or mineral soils). The current model prediction is based on a crude estimate of percentage organic soils in the catchment. Future analyses of catchment characteristics will be carried out in order to improve this estimate.

Modelling the mobility of ^{137}Cs and ^{90}Sr in soils and transfers to groundwaters

Following the fallout of radioactive caesium onto the soil, migration into the vertical soil profile is believed to be relatively rapid as a result of infiltration of rainwater. Sorption of radiocaesium to the soil matrix, particularly illitic clay minerals (Sawhney 1972; Cremers et al., 1988), then rapidly slows the rate of migration. In the long term, most of the radiocaesium is strongly bound to the soil and is relatively immobile. After the atmospheric testing of nuclear weapons and the Chernobyl accident, the majority of deposited radiocaesium has remained in the top 30 cm of the soil profile for many years after fallout. Long term changes in the distribution of radiocaesium within the top layers of the soil profile may, however, result in changes in external radiation exposure, transfers to vegetation, and runoff of radioactivity from contaminated soils to surface waters.

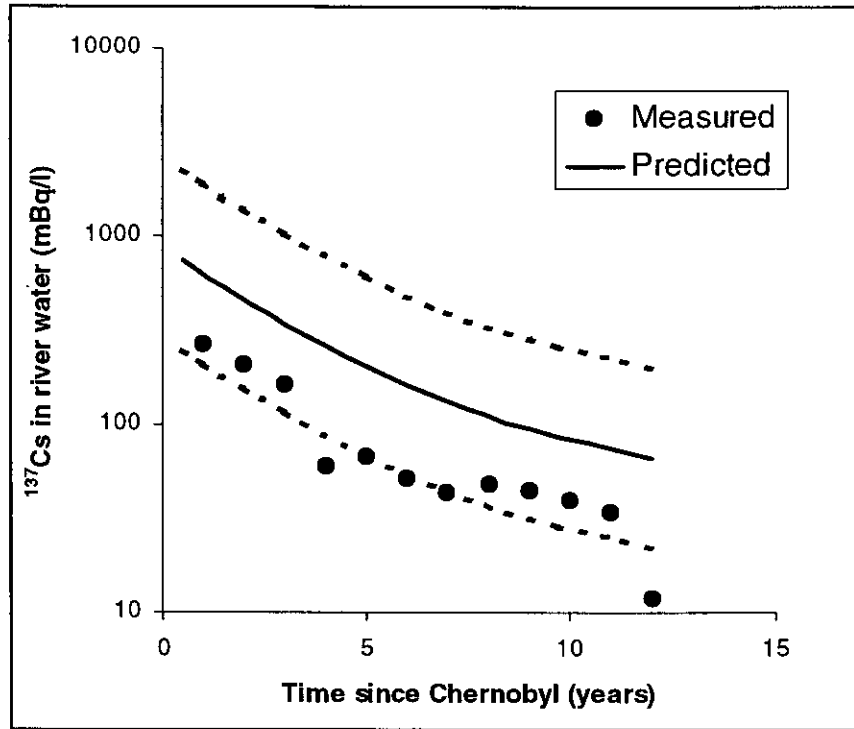


Figure 1.1 “Blind” prediction of ^{137}Cs activity concentration in the R. Dnieper at Rechitsa, Belarus. The model assumes a predominantly mineral catchment (95% mineral soils, 5% organic).

The slow sorption of radiocaesium to the soil matrix has been shown (Smith et al., 1999) to control the decline in radiocaesium activity concentrations in foodstuffs during the years after a fallout event. The purpose of the present study is to determine the extent to which sorption reactions could influence the advection and dispersion of radiocaesium in the soil. We are testing three different models for the long term mobility of radiocaesium in soils: (i) a model which assumes that no fixation occurs, i.e. that dispersion and advection continues at a constant rate after fallout; (ii) a model which assumes that radiocaesium is completely immobile in the soil during the years after fallout; (iii) a model which assumes that radiocaesium mobility slowly changes during the years after fallout as a result of slow sorption reactions.

The 41 activity depth profiles we are using to test the different models were taken from published data, as well as our own measurements. The data set comprises profiles from organic and mineral soils in Cumbria, England (Horrill & Howard, 1991; Sandalls & Bennett, 1992; Smith et al., 1997), various soils in Ukraine (Ivanov et al., 1997), soils in Belarus (our measurements) and a grassland soil from Northern Italy (Chamard et al., 1993).

Analyses of these data are ongoing, but initial results show a gradual decline in mobility of radiocaesium in the soil over time after fallout (Fig. 1). There appears to be a tendency for the mobility of radiocaesium to decrease towards a constant value on

long timescales after fallout. Further studies will attempt to evaluate long term (decades) mobility of ^{137}Cs and long term transfers to groundwaters.

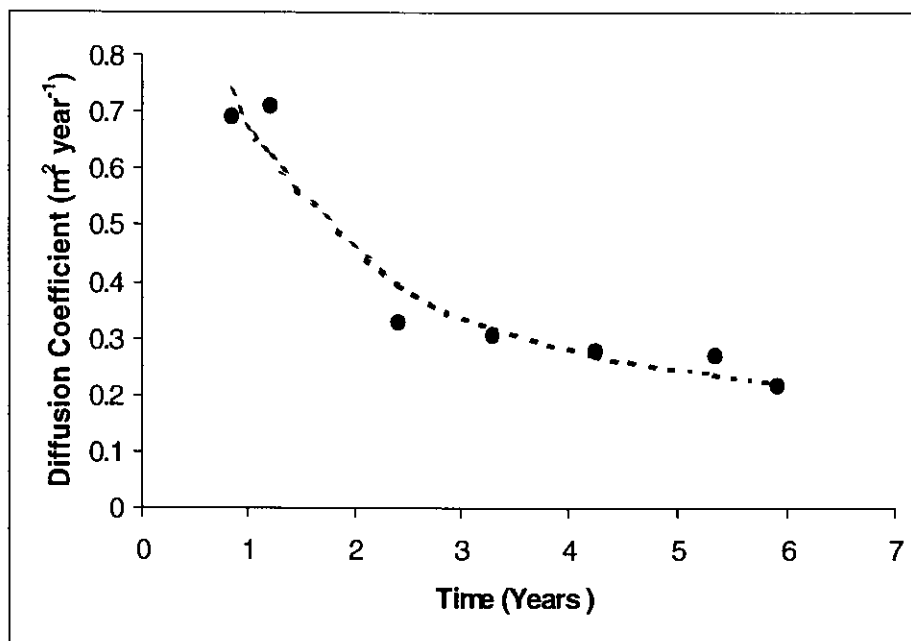


Figure 1.2 Change in average apparent coefficient of dispersion of ^{137}Cs in soils as a function of time since Chernobyl. Dotted line shows fit of time dependent advection dispersion equation.

Partner 2 (Scientific Coordinator): SPA TYPHOON.

Objectives

- To coordinate and monitor scientific and technical aspects of the project.
- To participate in joint meetings and field exercises
- Measurements of ^{137}Cs activity in Svyatoye and Kozhanovskoye lakes
- Measurements of ^{90}Sr activity in Kozhanovskoye lakes
- Measurements of ^{137}Cs activity-depth profile and ^{137}Cs speciation in soils
- Long-term measurements of rates of radiocaesium fixation by soils

Main activities and progress

TYPHOON arranged and participated in two joint meetings and field visits in Russia and Belarus. TYPHOON participated in joint meeting and field visit in FGW.

Samples of Kozhanovskoye and Svyatoye lakes water were collected during three sampling campaigns (February 1999, May 1999, September 1999). Concentrations of ^{137}Cs and ^{90}Sr have been measured in the samples collected before September 1999. Main hydrochemical characteristics of the lake water were also measured.

Samples of bottom sediments were collected in February 1999 in cooperation with URHI and BSU in two Russian and two Belorussian Lakes (two cores per lake). Speciation of ^{137}Cs in the solid phase of sediments as a function of depth were measured using sequential extraction procedure. Content of exchangeable K^+ , Na^+ , Ca^{++} , and Mg^{++} were determined in sediments as a function of depth.

Four soil monoliths were sampled in Briansk Region on catchments of Kozhanovskoe and Svyatooe lakes. Vertical distribution of ^{137}Cs in monoliths and speciation of ^{137}Cs in the most radioactive soil layers were measured. Content of exchangeable K^+ , Na^+ , Ca^{++} , and Mg^{++} were determined in sediments as a function of depth.

Table 2.1 Depth distribution of ^{137}Cs in soils of Briansk Region samples on May 1999

Soil layer	Depth of the layer, cm	Activity ^{137}Cs , Bk/g	STD, Bk/g	Activity ^{137}Cs , Bk/cm ³	Exchangeable ^{137}Cs , %
VP11S98 O _L	0-0.2	2.44	0.48	0.98	
VP11S98 O _f	0.2-2.6	44.4	9.12	5.88	3.5
VP11S98 O _h	2.6-4.0	29.86	1.25	14.51	2.31
VP11S98 E(1)	4.0-6.6	2.28	0.09	2.39	
VP11S98 E(2)	6.6-10.7	0.61	0.02	1.2	
VP11S107 A(1)	0-3.6	7.08	0.16	2.82	0.58
VP11S107 A(2)	3.6-6.4	6.57	0.33	5.85	0.64
VP11S107 A(3)	6.4-9.8	5.39	0.17	3.47	2.53
VP11S107 A(4)	9.8-15.6	2.34	0.13	2.13	
VP11S101 O _L	0-0.2	2.35	0.93	0.31	
VP11S101 O _f	0.2-1.4	57.08	8.04	7.88	4.00
VP11S101 O _h	1.4-3.1	95.34	14.27	45.35	2.73
VP11S101 E(1)	3.1-6.2	8.75	1.35	10.47	3.74
VP11S101 E(2)	6.2-9.5	0.51	0.06	0.66	
VP11S101 E(3)	9.5-14.9	0.18	0.02	0.20	
VP11S104 A(1)	0-3.6	7.08	0.16	2.82	1.32
VP11S104 A(2)	3.6-6.4	6.57	0.33	5.85	0.64
VP11S104 A(3)	6.4-9.8	5.39	0.17	3.47	
VP11S104 A(4)	9.8-15.6	2.34	0.13	2.13	

Long-term investigations of radiocaesium fixation by soils have been started. Concentrations of ^{137}Cs , K^+ , Na^+ , Ca^{++} , Mg^{++} , and NH_4^+ in the water phases and

exchangeable fraction of ^{137}Cs , K^+ , Na^+ , Ca^{++} , Mg^{++} in the solid phase were determined at the first five time points. The aim of the laboratory experiment was to investigate the dynamic of ^{137}Cs fixation by soils being preliminary equilibrated with water solution containing K^+ and Ca^{++} cations of different PAR as well as equilibrated with natural river water collected from Besed' River (Belarus). Soil (60 g) was equilibrated three times with 1 L of corresponding solution for 1 day and dried. After passing the dry soil through 1 mm sieve, 2 g aliquots of soil have been put to 20 mL vials and equilibrated with 19.8 mL of corresponding solution for 1 day. Then 1500 Bq of ^{137}Cs in 0.2 mL was added to each vial of 120 vials (six variants x ten sampling sessions x two replicates).

After 1, 5, 14, 33, and 63 days two vials of each variant were removed (total 12 vials each time) and concentration of ^{137}Cs was measured in the water phase and in 1 M $\text{CH}_3\text{COONH}_4$ extraction from the solid phase. The results of the ^{137}Cs dynamic in water phase of the soil:water slurries of different PAR value are shown in Fig. 2.1 for mineral soil and in Fig. 2.2 for organic soil.

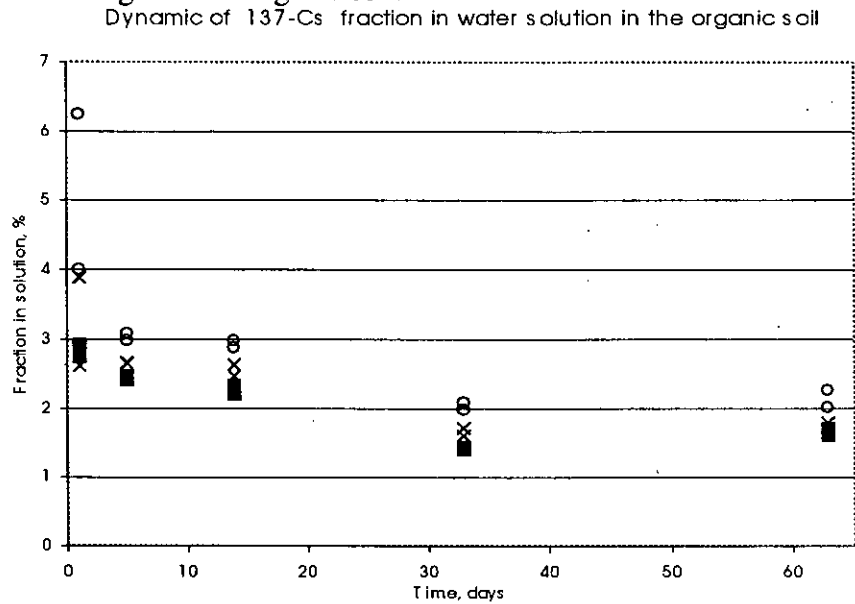


Figure 2.1 Dynamics of ^{137}Cs in water solution in mineral soil. Solid squares is soil - 0.1 mM K^+ + 1 mM Ca^{++} ; open circles - soil + 1 mM K^+ + 1 mM Ca^{++} ; crosses - soil + Besed' River water.

Dynamic of ¹³⁷Cs fraction in water solution in the mineral soil

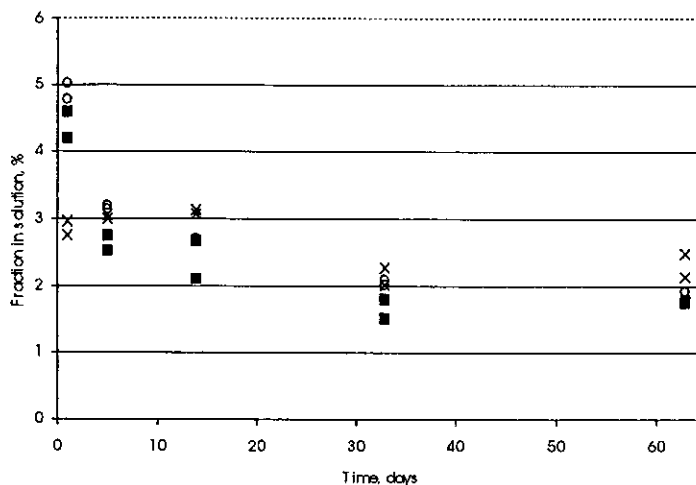


Figure 2.2. Dynamics of ¹³⁷Cs in water solution in organic soil. Solid squares is soil - 0.1 mM K⁺ + 1 mM Ca⁺⁺; open circles - soil + 1 mM K⁺ + 1 mM Ca⁺⁺; crosses - soil + Besed' River water.

Parameter values for the model of radionuclide behaviour in a closed lake were estimated using previously obtained and new experimental data. A model of radionuclide behaviour in a closed lake water and sediments was developed. The model equations were formulated and solved analytically. The following equation was obtained after the solution simplification:

$$C_w = \frac{e^{-\lambda t} S}{K_p (\pi D_e t)^{0.5}}$$

where C_w - radionuclide concentration in water (Bq.m⁻³); S - surface density of sediments contamination (Bq.m⁻²); K_p - ratio of radionuclide concentration in the upper layer of sediments (Bq.m⁻³) to that in water (Bq.dm⁻³); D_e - effective diffusion coefficient in sediments (m².d⁻¹); λ - radioactive decay constant(d⁻¹); t - time after contamination, d.

The model is now under testing against new and previously obtained experimental data. The following data are used for the model parameter calculation (S , K_p , D_e):

- vertical distribution of ¹³⁷Cs in Svyatoe lake sediments and its concentration in water measured in 1993 in frameworks of the CEC project ECP-3;
- vertical distribution of ¹³⁷Cs in Svyatoe lake sediments and its concentration in water measured in 1999 in frameworks of the present project.

Calculated C_w values for ¹³⁷Cs will be compared with experimentally measured concentration dynamics in 1992- 2000.

Problems

Concentration of ammonium in the water phase of suspensions in the long-term fixation experiment was higher than expected.

Outline plans for next year

- TYPHOON will participate in at least two joint sampling exercises in Russia and Belarus and in the meeting in Lisbon.
- Measurements of ^{137}Cs activity in Svyatoe and Kozhanovskoe lakes water will be carried out.
- The long-term fixation experiment will be continued.
- An initial analysis of the long-term fixation experiment results will be carried out.
- Values of radiocaesium sorption parameters (exchangeable selectivity coefficient, fraction of exchangeable form) for soils sampled in the first year of the project will be assessed as a function of depth.
- A joint paper on modelling of radionuclide behaviour in a closed lake will be prepared.

Partner 3 Netherlands Energy Research (ECN)

This report briefly describes the first results of the analysis of the soil core taken in Belarus. Among the results are the ^{137}Cs content of the soil sample, the ^{137}Cs concentration in the pore-water, pore-water chemistry, pH and redox potential (E_h) of the soil sample, and the specific caesium sorption capacity as a function of depth. Further work to be performed is indicated at the end of this report.

Sampling

A soil "core" has been taken in the eastern part of Belarus (near the village of Zarechye in the Kostyukovichy district) on the floodplain of the river Besedz on 11 May 1999. The location was situated in a so-called closed zone, which is an area evacuated after the Chernobyl nuclear accident because of the high ^{137}Cs and ^{134}Cs deposition there. The ^{137}Cs deposition in the zone is above $1.48 \text{ MBq}\cdot\text{m}^{-2}$.

Soil samples were taken by digging a hole of about 1 square meter, until the groundwater level was reached (groundwater level at that moment: about 1 m below the soil surface). Two litres of groundwater were taken from the water collected at the bottom of the hole. Then in the side of the hole, 1-cm slices of soil were taken down to a depth of 10 cm. Then, 5-cm slices were taken down to a depth of 45 cm. Features observed while taking the soil and groundwater samples were: the smell of rotten eggs (H_2S) coming from the hole, the coloration in the wall of the pit (small black particles, which are probably charcoal particles), and stains indicating oxidation-reduction phenomena. The soil texture differed markedly with depth; top layers were clayey and the amount of sand in the soil seemed to increase with depth.

Pore-water separation, pH, E_h , bulk density and ion concentration in pore water

Before separating the pore-water from the soil samples, pH and E_h of the soil samples was measured by inserting a glass electrode (pH measurement) and platinum electrode (E_h measurement) directly into the clay. Then, a sample was taken from the soil for analysis of soil moisture content (drying 24 hours at 105°C).

Pore-water was separated from the soil using centrifugation in home made centrifuge bottles, for 30 minutes at 4000 rpm (2952 g, 20 °C), in which the pore-water is filtered through 0.2 µm cellulose acetate membranes into a small Teflon (PTFE) collector. Generally, the pore-water yield was small; 50 ml at most for one slice, and was very small (≈ 8 ml) for some of the slices from deeper parts. Disassembling of the centrifuge bottles was done in a glove box under a nitrogen atmosphere (O₂ about 40 µg.l⁻¹). Pore-water was transferred to 50 ml PE wide mouth bottles under nitrogen atmosphere. Soil was transferred to 100-ml glass bottles with HDPE screw closure under nitrogen. Pore-water and soil were subsequently stored in a refrigerator (4 °C) until further handling.

Major ion concentrations were measured in the pore-waters by ICP-AES, flame AAS (K⁺) and ion-chromatography (NH₄⁺). Results show that the chemistry of the pore water is consistent with the measured E_h and pH of the soil samples with the lowest E_h and highest NH₄⁺ concentrations in the top of the profile (see Figure 1a).

Measurement of ¹³⁷Cs activity on solid phase and in pore-water

Counting of the ¹³⁷Cs activity on the solid phase in the pore-water was done in a low background counting room with walls of low-potassium concrete. ¹³⁷Cs and ¹³⁴Cs activity on the solid phase was counted on a coaxial Ge(Li) detector (relative efficiency 15.8 %, FWHM 1.8 keV). The ¹³⁷Cs depth-distribution is shown in Figure 1b. Total ¹³⁷Cs activity measured in the soil profile is 1.18 MBq.m⁻². Also, ⁶⁰Co and ¹²⁵Sb were present in measurable quantities (in addition to the natural radionuclides).

Aliquots of pore-water were available for radiocaesium analysis, after the addition of stable caesium as a carrier and preconcentration on ammonium molybdophosphate (AMP). Low-level ¹³⁷Cs γ-counting of the preconcentrated pore-water samples was performed in an anticoincidence array consisting of an annular NaI shield around an intrinsic germanium detector. Counting of ¹³⁷Cs activity in the pore-water from the lower parts of the profile are uncertain due to low pore water yield and low ¹³⁷Cs activity, so results are shown only for the upper 10 centimetres.

Specific caesium sorption capacity and prediction of in-situ measured K_D

In view of the observed changes in soil texture with depth (see above), the specific caesium sorption capacity is expected to vary also with depth. Results of the K_D[K⁺] measurement as a function of depth indeed confirm this expectation (Figure 3.1c). On the basis of measured K_D[K⁺] and the NH₄⁺ and K⁺ concentration, a prediction can be made of the in-situ measured K_D for the upper ten centimetres of the profile, which is shown in Figure 3.1d. In the top profile in-situ K_D is predicted very well.

Further work

Further work on these samples includes: (1) Modelling of the ¹³⁷Cs profile and estimation of ¹³⁷Cs transfer from surface to groundwater; (2) Investigation of the radiocaesium sorption reversibility in the profile (on one slice; 3 – 4 cm). (3) Investigation of ¹³⁷Cs sorption/desorption on illite in river water and at two selected PAR ratios.

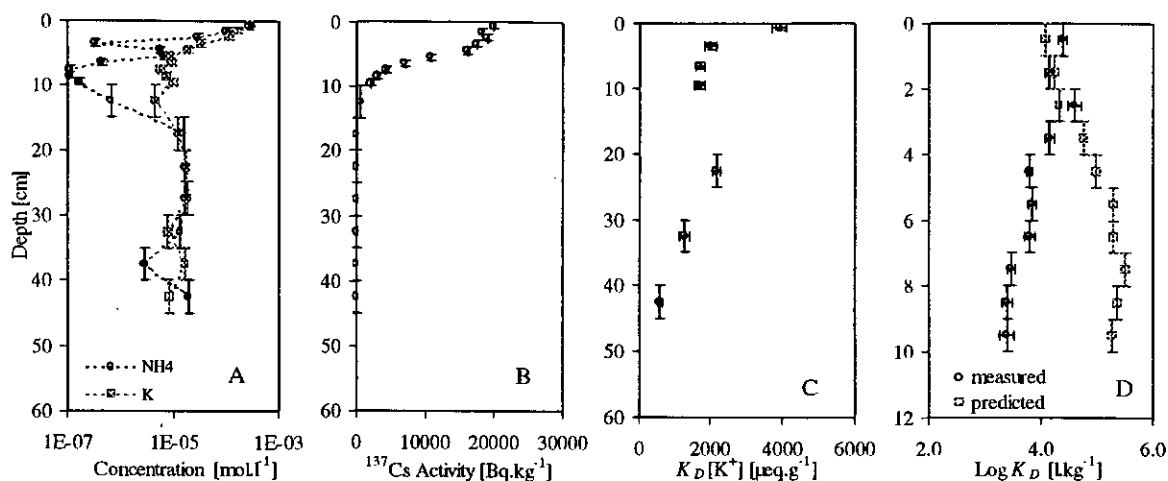


Figure 3.1 K^+ and NH_4^+ concentration (A), ^{137}Cs activity on the solid phase (B) and $K_D[\text{K}^+]$ as a function of depth (C). The in-situ measured and predicted K_D for the top ten centimetres of the profile are shown in D

Partner 4: Institute of Geological Sciences, Belarus (IGSB)

Objectives

- To organise and participate in joint meetings and fieldwork.
- To carry out chemical and radiometric analyses of lake, river and groundwaters.
- To carry out soil sampling and radiometric analyses.
- To study characteristics of river and lake catchments in relation to relative ^{137}Cs contamination.

Main activities and progress

Summary of main activities:

- Two field expeditions were undertaken for the investigation of the rivers, lakes, swamps and ground waters;
- Twice-yearly monitoring of the ^{137}Cs content in lake and river waters and measurement of lake-river waters chemistry (21 samples of water from rivers and 27 samples from lakes);
- 8 soil profiles were taken for estimation of the ^{137}Cs redistribution parameters along soil profiles and transfers to ground water. Chemistry and ^{137}Cs contamination of ground waters was studied at three sites (Strumen, Veprin and Orevichy) which have been studied since 1987;
- Three field experiments were completed, the aim of which were to study the influence of K^+ on ^{137}Cs redistribution along the soil profile and transfers to ground waters;
- Two peat-bogs (Opromokh and Maidan) have been selected for future studies.

Rivers

A continued decrease in ^{137}Cs in river waters, runoff coefficients and ^{137}Cs inventories within river catchments has been observed. On the basis of mean catchment slope, the studied river basins were divided into two groups (see Fig. 4.1) with rapid decrease of ^{137}Cs concentrations in the water (Sozh, Besed, Iput) and slower decrease in ^{137}Cs concentrations in the water (Dnieper, Pripyat).

Similar distinctions in the ^{137}Cs water runoff may be associated with different ^{137}Cs inventories and forms of its occurrence, with the differences in the structure and composition of the soil covers of the river catchments, lithology, thickness and water permeability of aeration zones, gradients of the catchment surface slopes, the territory drainage rates, and extent of bogs within catchments, etc. Each of the factors mentioned above needs special studies. However, we would like to note the highest hypsometric elevations of the river Sozh catchment within contaminated territories: 100-150 m – 48 % of the territory, 150-200 m – 52 % of the territory, river stream gradient is 0.00015, as compared with the Dnieper catchment in the region Mogilev-Rechitsa: 100-150 m – 60 % and 150-200 m – 40 % of the territory, river stream gradient is 0.000085. High levels of radiocaesium removal in the first 2-3 years after the accident are obviously due to a high hypsometric position of the contaminated catchment of the Sozh and its tributaries Iput and Besed, and are caused by intense erosion and sheet wash that preceded the ^{137}Cs penetration deep into the soil profiles. Efficiency of ^{137}Cs removal with river as against the radioactive decay within river catchments is not higher 0.69-3.18 %.

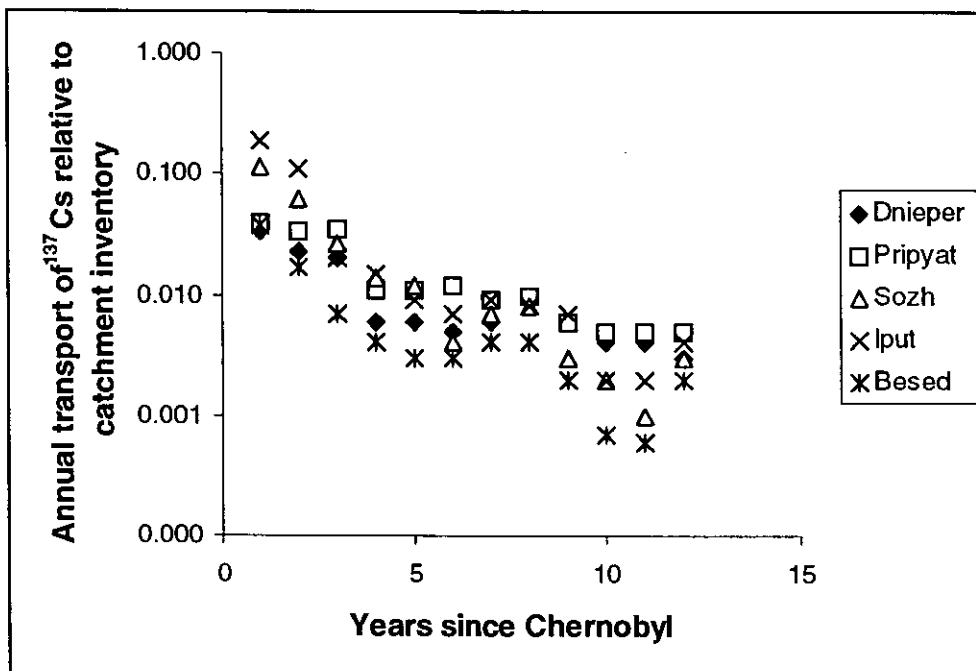


Figure 4.1 Transport of ^{137}Cs in Belarussian rivers as a % of fallout to the catchment.

Lakes

In 1996-1999 14 undrained lakes (3 of which are artificial pools) were studied in the radiocontaminated territories of the Mogilev (4) and Gomel (10) regions of Belarus, where the ^{137}Cs specific activity ranges from $65 \cdot 10^6$ to $1,385 \cdot 10^6$ Kbk/km². Most of the lakes are situated within a high terrace of the rivers Besed and Sozh and their position is associated with a zones of a large tectonic dislocations which controls over the rivers valley location. The lakes are of collapse (Karst) type and their origin and some hydrodynamic elements are related to leaching of Upper-Cretaceous carbonate rocks that occur there in a depth range of 60 to 120 m.

All the studied water bodies are undrained lakes (except the lake Perstok #15 which is intermittently drained) with both precipitation and subsurface water supply. The water balance discharge is due to the evaporation and subsurface runoff. The first variant of the lake classification was devised (see Table 4.1).

By the morphology of lakes, chemical composition of lake water and abundance of water organisms in it most of the studied water bodies are mesotrophic lakes showing oligotrophic evidences and the perch-roach type of fish populations.

Within 1996 and subsequent three years the ^{137}Cs concentration in water of the studied lakes varied between 0.2 and 7.2 Bq/l. Radiocaesium was supplied by the radionuclide fallout within the lake water area as a result of the Chernobyl accident, as well as by subsequent ^{137}Cs washout from the catchment areas, which is suggested by a close correlation ($R^2=0.9$) between the radionuclide inventory in catchments and in the water bodies of lakes. Close relations have been established between the radiocaesium and K^+ concentrations ($R^2=0.62-0.7$) and, correspondingly, between their inventories in lacustrine water ($R^2=0.78$). Groundwater was excluded as a probable source of dissolved ^{137}Cs as it shows rather low concentrations of this radionuclide (0.12-0.5 Bq/l) in the coastal zone of lakes (from the prospecting pit data).

Table 4.1

**CLASSIFICATION
of the studied lakes of Belarus**

Main indications		Character of indications		Lakes
I	Morphometric	• <i>water area</i>	to 0.1 km ²	Tyumenskoye #1, Stoyacheye #2, Art.pool #4, Svyatskoye #5, Svyatoye #7, Smerdin #10, Art.pools #12-13, Perstok #15
			0.1-0.5 km ²	Svyatoye #3, Kolpino #6, Stoyacheye #8, Svyatoye #9, Dobryn #11
		• <i>maximum depth</i>	to 1 m	Art.pool #4, Dobryn #11
			1-2 m	Art.pools #12-13
			2-5 m	Tyumenskoye #1, Stoyacheye #2, Svyatoye #3, Kolpino #6, Svyatoye #9, Perstok #15
			5-10 m	Svyatoye #7, Smerdin #10
			more 10 m	Svyatskoye #5, Stoyacheye #8
II	Genesis of the lake hollow	• <i>tectonic depression</i> • <i>karst hollows</i>		--
				Tyumenskoye #1, Stoyacheye #2, Svyatoye #3, Svyatskoye #5, Kolpino #6, Svyatoye #7, Stoyacheye #8, Svyatoye #9
		• <i>dead abandoned meander</i> • <i>part of the lake-bog system</i>		Perstok #15
			- low, including old peat-cutting hollows	Art.pools #12-13
			- high	Smerdin #10
III	Character of the lake catchment	• <i>automorphic soils</i> • <i>semi-automorphic soils</i> • <i>slightly boggy</i> • <i>boggy</i>		Svyatoye #3, Svyatskoye #5, Kolpino #6, Svyatoye #7, Stoyacheye #8, Svyatoye #9
				Perstok #15
				Smerdin #10, Art.pools #12-13
				Tyumenskoye #1
IV	Rate of total exchange of water mass a) without ¹³⁷ Cs water supply and outflow by streams or rivers		2.5 and more	Svyatskoye, #5
			2-2.5 years	Svyatoye, #3
			1.5-2	Tyumenskoye, #1
			1-1.5	Kolpino, #6
			1 and less	Perstok, #15
	b) with ¹³⁷ Cs water supply and outflow by streams or rivers		--	--
V	Stratification of water mass	• <i>slight</i> • <i>observed</i>		main part of lakes with depth less 5 m
				Svyatoye lake #5 and other lakes with depth more 5 m
VI	Chemical conditions in the bottom-side water layer	• <i>oxidizing</i> • <i>reducing</i>		main part of lakes with depth less 5 m
				Svyatoye lake #5 and other lakes with depth more 5 m

Lake sediments

The bottom sediments of Svyatoye lake #3 were studied. Two main kinds of sediments are found: organic silt in centre and sandy bottom sediments in peripheral parts of the lake (characteristics of the bottom sediments see in Table 4.2). Distribution of ^{137}Cs between solid and liquid phases showed in Table 4.3.

Table 4.2

Characteristics of the bottom sediments of Svyatoye Lake. 30.09.97

Sample of bottom sediments	Depth of layer, cm	Humidity, %	pH_{KCl}	Na_{exch}	K_{exch}	Ca_{exch}	Mg_{exch}	Organic matter, %
Sand	0-12	0.59	5.1 ± 0.1	7.5 ± 1.0	31 ± 14	340 ± 40	26 ± 2	1.2
Organic silt	0-12	88.0	4.8 ± 0.1	180 ± 20	200 ± 20	10300 ± 200	710 ± 40	48

Contents of Na_{exch} , K_{exch} , Ca_{exch} , Mg_{exch} were determined using extraction of exchangeable cations with 1M NH_4Ac . Ratios of components in the system "solid phase-solution" were 1:10 for sandy bottom sediments and 1:25 - for organic silt. Organic matter contents in the samples were established on the weight losses of absolutely dry samples after heating at 750°C .

Table 4.3

Distribution of ^{137}Cs between solid and liquid phases of Svyatoye lake bottom sediments.

Samples were taken using McCereth corer (bore-hole S2).
24.02.1999. Measurements were carried out in IGN NASB (Minsk).

N	Sample	Layer, cm	Mass of sample, g		A^{137}Cs		Kd, l/kg	Kd for core S1, (for comparison)
			Wet	Dry	Solid phase, Bq/g	Liquid phase, Bq/l		
1	S2/1	0-2	61.1	2.19	147.0	54.7	2690 ± 540	1830 ± 330
2	S2/2	2-4	61.4	3.20	72.29	56.6	1280 ± 330	1320 ± 240

3	S2/3	4-6	62.5	4.31	29.42	27.7	1060±380	870±160
4	S2/4	6-8	63.5	4.66	23.05	26.8	860±250	730±130
5	S2/5	8-10	65.4	4.97	18.19	19.6	930±340	580±100
6	S2/6	10-12	63.6	5.03	12.46	12.4	1000±420	540±100
7	S2/7	12-14	62.5	5.13	8.33	10.3	810±470	370±70
8	S2/8	14-16	62.1	5.09	6.52	13.5	480±330	290±60
9	S2/9	16-18	64.6	5.31	5.67	11.4	500±350	240±50
10	S2/10	18-20	60.9	5.05	4.40	17.3	250±200	290±80

Ground water

Radioecological conditions of the ground waters was studied on the base of radiological monitoring results of bore-holes (6) and dug-holes (16) with long time series of observations.

In 1998 was made an experiment on location for explanation of the role of agricultural fertilizers (KCl) on the increasing of the ^{137}Cs migration along soil profiles to underground waters. Within Khoinsky district were studied two places with three sites dug-holes within each place: with "natural" distribution of ^{137}Cs along the soil profile (background site); with additional bringing in the soil cover of the KCl in quantity of 100 g/m^2 ; with bringing in the soil the KCl in quantity of 300 g/m^2 . After that all sites (dug-holes) were studied (soil and water sampling) with period between observations about 3 month.

We had disclosed the increasing of ^{137}Cs migration parameters D and V (for centers of ^{137}Cs inventories) under influence KCl in 10-20 time.

We had disclosed too the very considerable increasing D and V for quick component of migration but because of very little of the component volume (near 2 % of total ^{137}Cs inventory in the soil profile) we have not the essential contamination of ground water.

Problems

No significant problems

Outline plans for next year

- IGSB will participate in the two joint sampling exercises planned for May and September 1998.
- River and lake water monitoring will be continued

- Soil sampling and analyses will be continued.

Partner 5: Ukrainian Hydrometeorological Institute (UHMI)

Introduction and objectives of the reporting period

According to the scheduled time table of the project research activity was focused on experimental collection of material need to be analysed for further understanding of main mechanisms controlling processes of radionuclide redistribution among different components of natural water systems. Results to be obtained will be used for evaluation of parameters essential for retrospective and predictive modelling of radionuclides wash-off from natural catchments, transport to and in rivers and lakes.

For reported period the main directions of UHMI studies were:

1. Field sampling exercises on collection of water, bottom sediment and soil from some tested lakes and their catchments;
2. Laboratory processing of collected material using radiometric and physico-chemical methods;
3. Preparation of digital database of the data of radiological monitoring of Ukrainian Rivers affected by Chernobyl accident in order to perform a statistical analysis of parameter of long-term variation in activity concentration in their water.

Field sampling

The objective of field sampling campaign was to collect some additional material of water, bottom sediment and soil in order to get more detailed information about properties and structure of radioactive contamination of some tested lakes.

Three sets of joint field exercises were carried out –

in February sampling of water and bottom sediment cores have been done from ice-covered Russian and Belarusian lakes - Kozhanovskoe R1, Svyatoe R2, Svyatskoe B5, Svyatoe B7. Bottom sediments have been collected using mini Mackereth corer, eight cores (duplicate for each lakes) have been then sliced in 2-cm interval for top 20 cm and in 4-cm deeper. All russian samples brought to Kiev, and belarussian ones shared for further analysis between UHMI and IGS. Water samples were taken in plastic tanks and delivered to the laboratory.

The program of field campaign in May included an additional sampling of water and soil cores from lakes with counteracted catchment area where it could be wash-off input to the lake created - Svyatoe R2, Svyatskoe B5. Water samples were also collected in Kozhanovskoe lake at three locations – input creek, lake and output creek. The choice of soil sampling points was deduced by dosimetric data in order to in the best way to reflect a spatial picture of object contamination. Sampling was carried out using sectional soil-corer up to depth 20 cm. Directly on a place soil column was sliced into

sublayers by 1 - 3 cm interval depending on visually observable distinctions in soil texture, then all subsamples were packed in labelled airtight plastic bags.

Joint intercoordination meeting in Weingarten in July was accompanied by sampling exercises as well. 6 cores of bottom sediments (from 40 cm to 1 m length) were collected from Lake Constance (Obersee) and 7 cores were collected from Lake Vorsee. 1 water sample (1330 l) from Lake Constance and 2 samples from lake Vorsee (200 l each) were collected using large volume water sampler „Midiya“. Three cores of bottom sediment from Lake Constance (2) and Lake Lugano collected in recent years and previously already analysed in FHRW are subjected for ^{210}Pb dating to be done by UHMI. All the cores selected were sliced in 0.5 cm interval, showed a nice resolution of “Chernobyl” and “weapon test” ^{137}Cs peaks and will be used in intercomparison of mathematical models.

Dating of lake's bottom sediment using ^{210}Pb profile is out of specific scope of the project, however is very useful as complementary information about temporal changes in sedimentation rate in particular places of the lake over the last century. This has become quite helpful in correct definition of responsible parameter in mathematical modelling of vertical distribution of artificial radionuclide (e.g. ^{137}Cs) in lake's bottom sediments. Among these forces are most important an acceleration of soil erosion owing to anthropogenic load on environment in last decades or seasonal (year by year) changes in hydrologically derived (riverine) input of sediments to the lake.

Laboratory analysis

In the laboratory of UHMI the following analyses were and are being performed:

1. Determination of the weight characteristics of soil samples - volumetric density, field humidity etc. Some part of samples was not dried up, and remained packed in a refrigerator for the consequent analysis of the samples in a natural state;
2. Separation of solid part and pore water in bottom sediment by highspeed centrifugation.
2. Determination of cation exchange capacity (CEC) of solid by method of barium displacement;
3. Determination of organic matter content as loss on ignition (LOI) after 2 hours at a temperature 900°C ;
4. Filtering samples of water and pore water through a membrane filter 0.45 microns and concentration of dissolved Cs-137 on a selective sorbent "ANFEZH" by ion-exchange column;
5. Radiometric measurement of Cs-137 activity in soil, bottom sediment and water samples using a semiconductor coaxial-type and well-type gamma-spectrometer • RTEC EG&G;
6. Pore water analysis for ^{137}Cs and major cations Ca, Mg, K, Na, NH_4^+ with application of AAS method;
7. Determination of Cs-137 speciation and main competing macro-cations (K^+ , Ca^{++} and Mg^{++}) in soil and bottom sediment by a method of sequential extractions.

The preliminary results of the laboratory experiments were reported on the AQUASCOPE meeting in July, 1999 in Weingarten and summarised on fig.5.1-5.2 and

in table 5.1. Laboratory analyses of remained samples have not been finished yet, but will be presented in the final report.

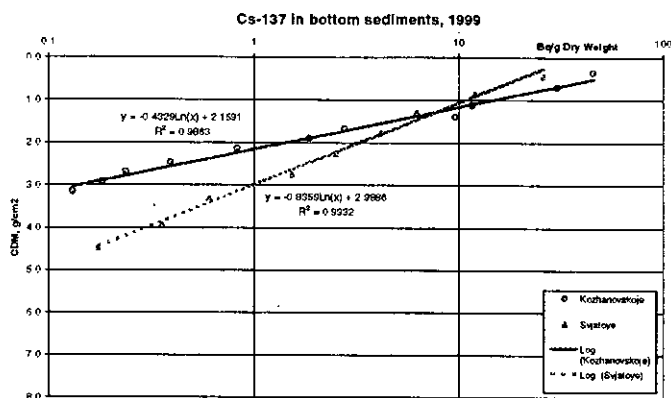


Fig.5.1 Vertical distribution of ¹³⁷Cs in bottom sediment

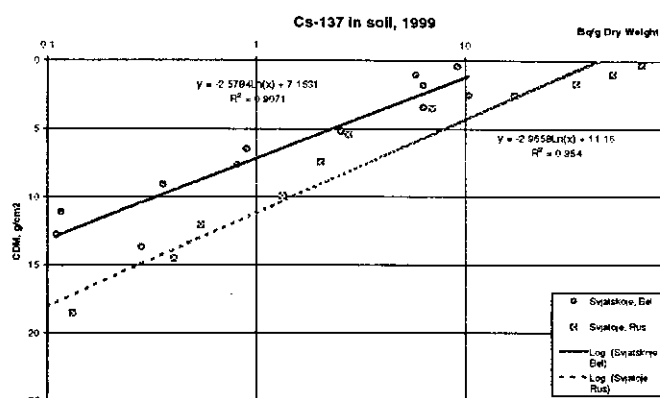


Fig.5.2 Vertical distribution of ¹³⁷Cs in soil

Table 5.1 Summary of Cs-137 contamination of system studied lakes

Lake	Cs-137 Inventory, kBq/m ²		Cs-137 concentration in water, Bq/L	Macrocation concentration in pore water, mg/L			
	Soil	Bottom Sediment		Ca	Mg	K	NH ₄
Svyatoe, Russia	1145 (1200)	613 (714)	10.5	6-16	3-10	11-30	10-30
Kozhanovskoe, Russia	(750)	460-370 (335)	8.6	15-20	6-12	7-10	9-13
Svtatoje-3, Belarus	(1370)	480	8.5			5-25	
Svyatskoje-5, Belarus	340	136	5.3				

Intercomparison of radiometric analyses

Because of a number of participants are involved in providing with radiometric analysis at different stages of the project execution and these data to be used by co-partners “as it is”, it was decided during first year to carry out some intercomparison exercises. These exercises are aimed only to check out the replicability between the results obtained in different labs and are not understood as comprehensive qualitative analysis of analytical techniques used. Intercomparison consists of interchange of environmental samples (bottom sediment, water, suspended matter etc.) processed in one lab with another followed by radiometric counting of chosen samples and check if they fall into defined range of gross (radio) activity. All the data sets obtained in different labs are being statistically compared against each other in order to make sure whether acceptable replicability take place or if there is an apparent discrepancy. First experience between UHMI and IGS shows that accuracy of radiometric data is sufficient ($R^2=0.96-0.99$) and does not exceed ca. 10% of standard error in range of activity 10-100 Bq/per sample, though allows one significantly improve even such a “good” replicability because of systematic character of errors appeared in lower range

of activity. This is to be done after all the participants involved (namely – UHMI, IGS, TYPHOON, FH R-W & ECN) contribute their data sets for arbitrary statistical test.

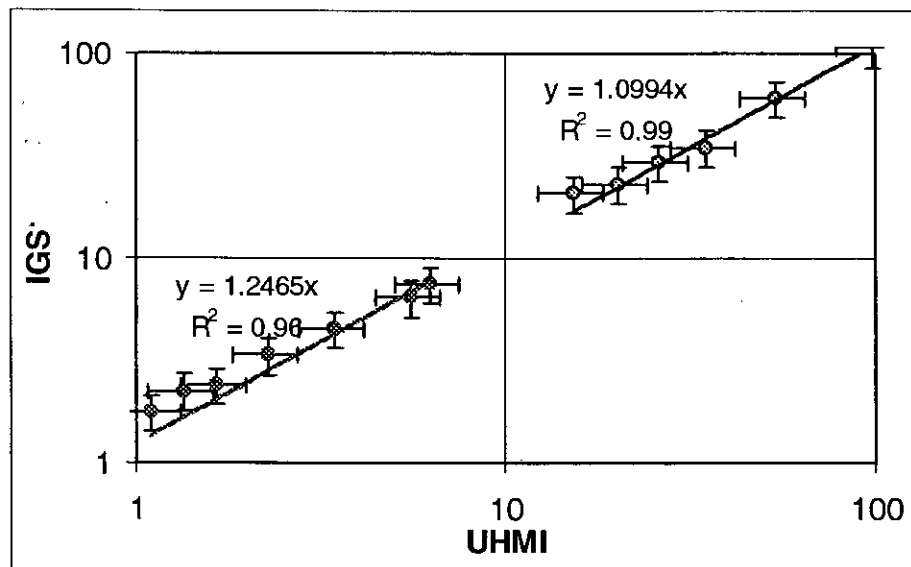


Fig.5.3 Intercomparison results – data of ^{137}Cs radiometric analysis provided by UHMI against data of IGS

Objectives for the next period

The Ukrainian Hydrometeorological Research Institute plans the following tasks for the next period of the AQUASCOPE project:

1. Completion of the laboratory analyses of samples collected during field exercises of 1999
2. Planning of experimental work for 1999-2000 and arrangement of joint sampling in Ukraine;
3. Evaluation of obtained results, definition of mechanisms and parameters controlling radionuclide distribution among abiotic components of the lakes studied;
4. Preparation of the report.

Partner 6: Department of Radiation Protection and Nuclear Safety (DPRSN)

Objectives for the reporting period

To carry out measurements of frayed edge sites (FES) and radiocaesium interception potential (RIP) of sediment samples from the Belarus and Russia (Chernobyl contaminated areas) and Germany lakes.

Progress achieved

Since the field sampling at the Chernobyl zone and German lakes was performed in May and July 1999 it was not possible to present in this report the data concerning the

characterization of these sediment samples. Instead, FES and RIP determinations were carried out to Tejo river (Portugal) sediments.

FES determinations

The method used to the FES determinations (Cremers *et al.*, 1988) is based on the masking of the ion exchange sites using a suitable background level of silver thiourea (AgTU) being the action of the caesium ions restricted to the small number of specific adsorption sites that are inaccessible to the bulky silver thiourea complex. The experimental procedure consists firstly, on the sediments presaturation with silver thiourea (AgTU 0.015M). Afterwards, the sediments are equilibrated (24 hrs) with silver thiourea solutions containing increasing levels of caesium chloride (10^{-5} to 10^{-2} M) ^{137}Cs labelled. Finally, the suspensions are centrifuged and the caesium activity measured at the liquid phase by gamma spectrometry. The results are plotted as caesium adsorbed versus concentration in the equilibrium solution.

Figure 1 shows the radiocaesium sorption isotherms for Tejo river sediments. The sorption plateau values correspond to the FES capacity. However, as it can be seen mainly for A and S sediments the plateau on the sorption isotherm seems not be reached.

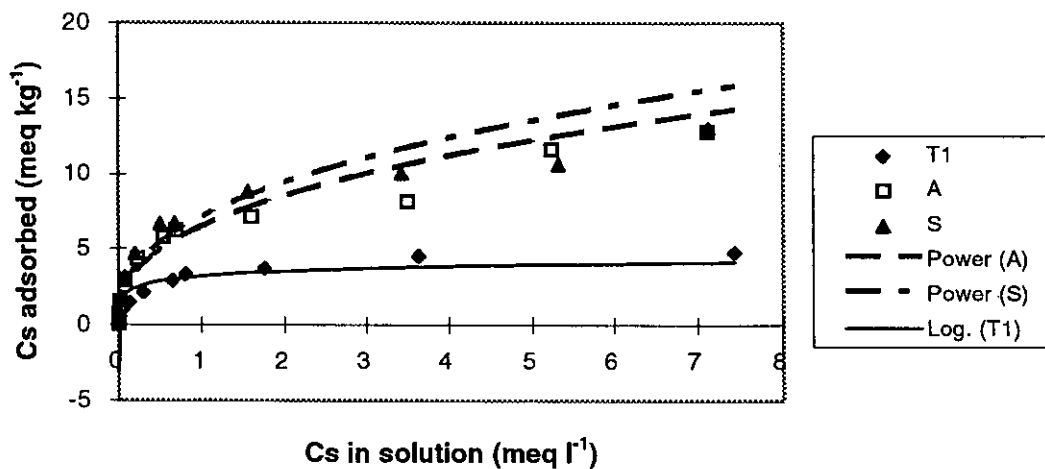


Figure 6.1 Caesium sorption isotherms in the frayed edge sites (FES)

Another calculation method was carried out which allows the FES determinations before the adsorption maximum has been reached. The procedure consists on the application of the Langmuir equation $X = \frac{X_m C}{k + C}$ to the radiocaesium sorption

isotherms. The intercept resulting from the study of the dependence of the inverse concentration of caesium adsorbed on the inverse concentration of caesium in solution

(Figure 6.2), this is, the linearization of the Langmuir equation, $\frac{1}{X} = \frac{k}{X_m} \left(\frac{1}{C} \right) + \frac{1}{X_m}$,

corresponds to the inverse value of the maximum adsorption (FES capacity). Therefore, the FES values could be obtained from the last range of isotherm before sorption maximum, which corresponds to high caesium concentrations. The capacity of

high affinity sites (HAS) can also be determined using the initial range of isotherm at low caesium concentrations. These sites having a very high selectivity to caesium will be occupied by caesium in the first place.

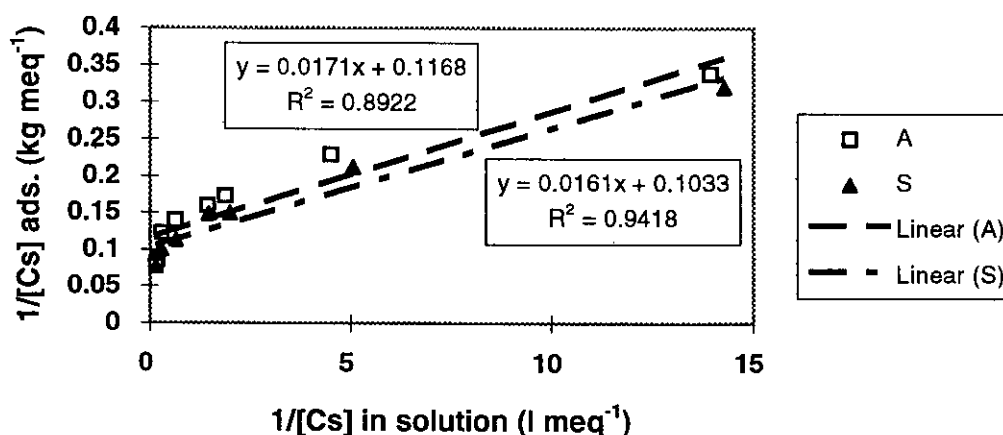


Figure 6.2 Inverse of the caesium concentration adsorbed in the sediments versus the inverse of the caesium concentration in solution to high caesium concentrations.

The values obtained for FES using the Cremers *et al.*, (1988) method and Langmuir calculations (FES and HAS) are presented in Table 1.

Table 6.1 Frayed edge sites (FES) determinations to Tejo river sediments (Portugal) using Cremers *et al.* method (1988) and Langmuir equation

Sediments	CEC cmol kg ⁻¹	<u>Cremers et al., 1988</u>		<u>Langmuir</u>	
		FES meq kg ⁻¹	FES meq kg ⁻¹	FES meq kg ⁻¹	HAS meq kg ⁻¹
T1	14.3	4.1±0.7	3.7	0.29 (7.9%)	
A	27.9	9.9±2.7	8.6	0.63 (7.4%)	
S	31.4	10.6±1.8	9.7	0.44 (4.5%)	

It can be seen that FES values obtained from the Langmuir linearization are lower than those calculated from the radiocaesium sorption plateau. The HAS presented values which corresponds to about 4 to 8% of the FES.

RIP determinations

On the other hand, the characterization of a system in terms of FES capacity and trace selectivity coefficient ($K_c(Cs/K)$) allows predictions of radiocaesium distribution coefficients in scenarios for which the FES are homoionically potassium or ammonium saturated, and could be calculated using the equation $[K_d(Cs).m_K] = K_c(Cs/K).[FES]$. Consequently, it could be expected that the K_d

potassium or ammonium concentration product would be constant and equal to the $K_c \cdot FES$ product. This quantity defined as the Radiocaesium Interception Potential (RIP) was determined to Tejo river sediments using the following experimental procedure: Firstly, sediment samples are repeatedly preequilibrated with silver thiourea solutions containing potassium or ammonium concentrations ranging from 2×10^{-4} M to 10^{-2} M. Afterwards, the same sediments are equilibrated (24 hrs) with the same solutions ^{137}Cs labelled. After phase separation, the caesium in the liquid phase is measured and the distribution coefficients calculated. Finally, in order to determine the exchangeable K_d the sediment samples are equilibrated with 1M ammonium acetate solution. After phase separation the caesium is measured at the liquid phase and the exchangeable K_d calculated. The RIP in ammonium scenario for S sediment is presented in Figure 6.3.

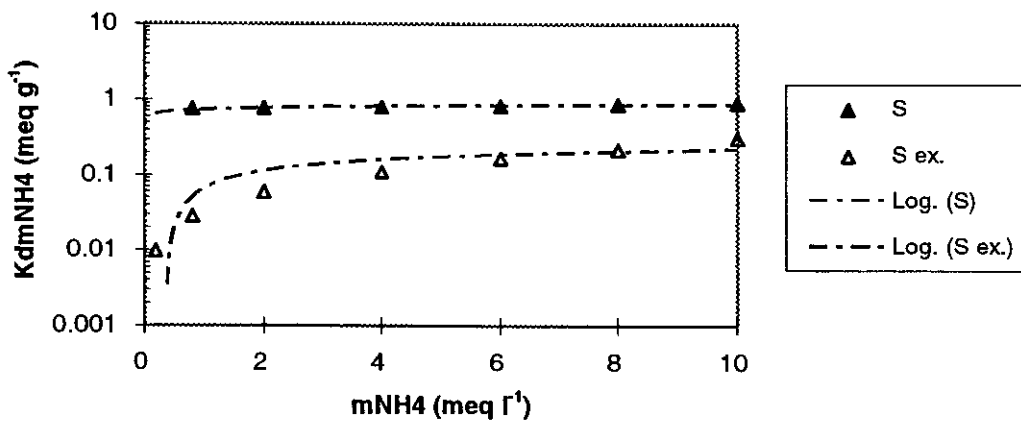


Figure 6.3 m_{NH_4} dependence of the $K_d \cdot m_{\text{NH}_4}$ product for S sediment.

It can be seen that the exchangeable RIP values using ammonium acetate extraction (lower curve) are lower than those corresponding to the RIP (first curve), by a factor of about five. The values obtained in potassium scenario for exchangeable RIP were about ten times lower than those obtained to RIP. The reason for this difference should be the caesium fixation by the sediments. This means that the radiocaesium interception potential values are overestimated if the fraction of exchangeable caesium is not taken into account.

Reference

Cremers, A., Elsen, A., De Preter, P., Maes, A. (1988) Quantitative Analysis of Radiocaesium Retention in Soils. *Nature*, Vol. 335, n°6187, 247-249.

Partner 7 Fachhochschule Weingarten (FHWEIN)

Collection of existing data from Lake Constance and Lake Vorsee

The following data were collected and stored in an EXCEL data base:

- lake characteristics
- water analyses (dissolved Cs-137, Cs-137 in suspended matter, K_d -values, major ions, temperature, pH, O_2 , in lake water, inflows and outflows)
- sediment and pore water analyses (vertical distributions of Cs-137 in bottom sediments, horizontal distributions of Cs-137 inventories, Cs-137 exchangeability,

- Cs-137 in pore waters, cations in pore waters, exchangeable cations in sediments, organic matter content, [FES] and [RIP] measurements)
 - Cs-137 in fish (also weight and length)
- „Materials and Methods“ and „References“ were also given.

Sampling on Lake Constance and Lake Vorsee

Determination of fallout to lake catchment

In March 1999, 9 soil cores were taken at three sites on meadows in the catchment of Lake Vorsee for the determination of the mean catchment fallout.

Water sampling

During the Aquascope meeting in Weingarten, Germany in July 1999, a 1330 l water sample was taken at the deepest site of Lake Constance using a large volume water sampler (Midiya system). Water samples were taken from Lake Vorsee in November 1998, June 1999 and July 1999. In 1999, sampling was carried out using the Midiya system. Also physico-chemical parameters were measured (temperature, pH, O₂).

Sediment sampling

During the Aquascope meeting in Weingarten, Germany in July 1999, 7 sediment cores were taken from the deepest site of Lake Constance using a Meischner & Rumohr gravity corer and 7 sediment cores were sampled from Lake Vorsee using a sediment corer specially constructed for soft sediments.

The following measurements will be done by the groups FHW, DGA and Typhoon:

- Analysis of the vertical distribution of Cs-137
(0.5 cm intervals at one Lake Constance sediment core, 10 cm intervals at 2 Lake Vorsee sediment cores)
The mean inventories (Bq/m²) will be determined.
- Cs-137 exchangeability in sediments
- Cs-137 in pore water of Lake Vorsee sediment cores
- Cations in pore water (Ca²⁺, Mg²⁺, Na⁺, K⁺, NH₄⁺)
- Exchangeable cations in sediments
- Organic matter in sediments (%)
- [FES] and [RIP] in sediments
- $K_c^{eff}(Cs/K)$, $K_c^{eff}(Cs/ NH_4)$
- Sorption kinetic studies
- Comparance NH₄AC extraction and infinite bath

Fish sampling

In 1998/1999 36 fish were caught in Lake Constance (26 in 1998 and 10 in 1999) and 14 fish in Lake Vorsee (5 in 1998 and 9 in 1999) for determination of the Cs-137 activity concentration.

First results for Lake Constance and Lake Vorsee

Lake catchment fallout

A mean catchment fallout of 28000 Bq/m² was calculated for Lake Vorsee catchment.

Water

The dissolved Cs-137 activity in the water of Lake Constance has further decreased. During the stratification period in July 1999, a Cs-137 concentration of 0.13 mBq/l in the surface water was measured. Three years ago, in July 1996, the Cs-137 concentration in the surface water was 0.24 mBq/l. A K_d-value of $1.98 \cdot 10^8$ l/kg was determined in July 1999.

An additional water sample should be taken during the circulation period in winter 1999/2000.

Whereas the dissolved Cs-137 concentration in the water of Lake Vorsee amounted to 90 mBq/l in November 1998, the Cs-137 concentration decreased by half in summer 1999, 47 mBq/l in June 1999 and 34 mBq/l in July 1999. K_d-values of 9213 l/kg in June 1999 and 13124 l/kg in July 1999 were determined.

Sediment

The vertical distributions of Cs-137 and NH₄ concentrations in the pore waters of 3 sediments cores from Lake Vorsee were already determined. Both, Cs-137 and NH₄ concentrations in pore waters increase with increasing sediment depth

(Core 1:

Cs-137 concentration increases from 72 +- 37 mBq/l in the 0-10 cm layer to 605 +- 56 mBq/l in the 30-40 cm layer, NH₄ concentration from 13.7 mg/l in the upper layer to 49.2 mg/l in the deepest layer.

Core 2:

Cs-137 concentration increases from 126 +- 29 mBq/l in the 0-10 cm layer to 474 +- 43 mBq/l in the 30-40 cm layer, NH₄ concentration from 2.4 mg/l in the upper layer to 52.4 mg/l in the deepest layer.

Core 3:

NH₄ concentration increases from 4,7 mg/l in the upper layer to 70 mg/l in the deepest layer.)

Fish

In 1998, Cs-137 activity concentration in planctivorous fish from Lake Constance ranged from 1 Bq/kg to 4 Bq/kg. In 1998, Cs-137 activity concentration in carps from Lake Vorsee lay between 18 Bq/kg and 40 Bq/kg and in one pike sample a Cs-137 activity concentration of 207 Bq/kg was measured.

In 1999 about 10 fish from Lake Constance will be measured in September.

In 1999, Cs-137 activity concentration in carps from Lake Vorsee ranged from 66 Bq/kg to 110 Bq/kg and in one pike sample a Cs-137 activity concentration of 216 Bq/kg was measured, 2 samples of eel had 91 Bq/kg and 115 Bq/kg.