Radioprotection, Suppl. 1, vol. **40** (2005) S759-S764

DOI: 10.1051/radiopro:2005s1-111

Observed half-lives of ^{3}H, ^{90}Sr and ^{137}Cs in hydrosphere in the Vltava river basin in vicinity of NPP Temelín (Czechia)

E.J. Hanslík[1], V. Jedináková-Křížová[2], M. Brtvová[1], D. Ivanovová[1], E. Kalinová[1], B. Sedlářová[1], J. Svobodová[1], P. Šimonek[1] and H. Tomášková[3]

[1]*T.G. Masaryk Water Research Institute, Department of Radioecology, Podbabská 30, 160 62 Prague 6, Czechia*
[2]*Institute of Chemical Technology, Technická 6, 166 28 Prague 6, Czechia*
[3]*Charles University in Prague, 128 43 Prague 2, Czechia*

Abstract. The pilot operation of the nuclear power plant (NPP) Temelín first block was started up in 2001 and of the second block in 2002. Since 1990, the systematic attention has been paid to the monitoring of the hydrosphere reference level in the NPP Temelín vicinity. The Temelín NPP waste water influence has been monitored and assessed since 2001. The monitoring has been focused especially on the tritium, strontium-90 and caesium-137 concentration changes in water, river bottom sediments, fish, and in the water plants biomass. The observed half-lives indicate that the strontium-90 and caesium-137 releases fully interfere with the residual contamination after the nuclear weapons tests and the Chernobyl accident in the last century. Only the tritium concentrations in water samples, taken in the Vltava River downstream from the NPP Temelín waste water outflow, show a measurable influence.

1. INTRODUCTION

Temelín NPP is located at the upper Vltava River in south Bohemia. The power plant consists of two PWR type reactors, each with capacity of 1000 MW.

The observation and assessment of environmental conditions involved radioactive and non-radioactive polluting substances in surface water, groundwater, precipitation water, suspended solids, bottom sediments and biomass in wider environs of the Temelín NPP. The temperature and transport conditions in the Orlík Dam and other reservoirs built downstream from the Temelín at the Vltava River were also concerned. Some of the radionuclides (such as ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs), potentially present in effluent returns from Temelín NPP, are identical to those remaining in the environment after the Chernobyl accident and the nuclear weapons tests global fallout. Therefore, the examination of the radioactive substances in the hydrosphere prior to the operation of Temelín NPP and during the operation is particularly important.

2. METHOD

The changes in radioactive substances concentrations were monitored in tributaries of the Orlík Reservoir, the Vltava, Lužnice and Otava Rivers (upstream from the waste water outflow), and in Vltava River at the outflow from the Orlík reservoir (downstream from the waste water outflow). Total radionuclides concentrations in surface water (dissolved and solid form) have been measured in the Vltava River (upstream and downstream from Temelín) and in its main tributaries since 1990. For the radionuclides concentrations determination, the evaporation residues of large-volume (about 50 l) samples, taken with frequency of four in a year, were used.

Bottom sediments were sampled by a diver at six sites involving the tributaries and the Orlík Reservoir. The samples were taken once a year from the sediments top layer (0 – 10 cm). Granularity of the samples was mostly under 2 mm. For the analyses, the samples were dried at 105 °C. Fish and aquatic flora biomass samples were also taken with annual frequency. These samples were dried and ignited. Sampling sites and radiological method were described in proceedings [1].

3. RESULTS AND DISCUSSION

3.1 Surface water

Before the pilot operation startup, ^{137}Cs, ^{90}Sr and tritium had been present in the Vltava River and its tributaries. Till present, there still have been measurable concentrations of these artificial radionuclides in this area. The reasons, the residual contamination, were meant above. For the time relation of the radionuclides concentration, the kinetic equation of the first order appears to fit. The trend was originally assessed for the entire period of the monitoring. The kinetic equation of the first order for ^{137}Cs is:

$$lnc_{137Cs} = -\lambda_{ef} \cdot t + q$$

where: c_{Cs137} is the ^{137}Cs concentration in total solids (Bq/m^3),

λ_{ef} the effective (observed) kinetic constant of the ^{137}Cs concentration decrease, includes also the physical decay constant and the ecological kinetic constant of ^{137}Cs 137; $\lambda_{ef} = \lambda + \lambda_{ec}$ (1/year),

t time (year),

q ^{137}Cs concentration at the beginning of the monitoring (Bq/m^3).

After a longer monitoring period, a lowering of the ^{137}Cs concentration decrease rate was observed. Therefore, the time changes in ^{137}Cs concentrations were further assessed separately in the period 1990-1994 and in the period 1995-2003. The example of the Vltava Hněvkovice profile (the source of technological water for NPP) is shown in the Figure 1. The Y-axis represents the ^{137}Cs concentration natural logarithm and the X-axis time (t = 0 corresponds with the monitoring beginning in 1990). The average ^{137}Cs concentration in this profile was in 2003 1.5 Bq/m^3.

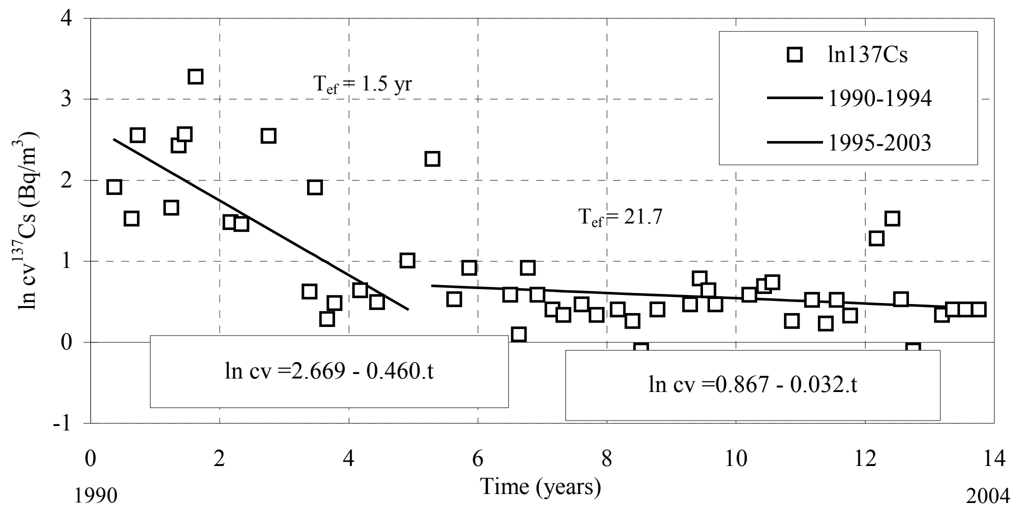

Figure 1. Time changes of ^{137}Cs concentration in the Vltava Hněvkovice profile in the period 1990-1994 and 1995-2003.

For the period 1990-1994, the effective half-life was T_{ef} = 1.5 years and 21.7 years for the period 1995-2003. Similar decreasing trends of ^{137}Cs concentrations were observed also in the water of other Orlík Reservoir tributaries, rivers Otava and Lužnice, which are not influenced by the NPP Temelín. In the Vltava Solenice profile downstream of the Orlík Reservoir, which is potentially influenced by the NPP Temelín waste water, for the period 1990-1994 the effective half-life was T_{ef} = 1.5 years and 12.6 years for the period 1995-2003.

The assessment results show that the ^{137}Cs concentrations in Vltava Solenice profile, downstream of the Orlík Reservoir, are lower than the ^{137}Cs concentrations in the Orlík Reservoir tributaries. This corresponds with the hypothesis of the suspended solids sedimentation in the reservoir and of the relatively high ^{137}Cs amounts adsorbed at them. It should be meant for the information completeness that the annual average ^{137}Cs concentrations in suspended solids (dry matter, 105 °C) in the profiles upstream of the Hněvkovice Reservoir, Vltava Pěkná and Vltava Březí, were 142 Bq/kg and 105 Bq/kg in 2003. From the monitoring also results that the significantly lower ^{137}Cs concentrations in Vltava Solenice profile downstream of the Orlík Reservoir and their decreasing trend continues even after the NPP Temelín waste water release startup. However, the fact is that the particular radionuclides concentrations in NPP Temelín waste water, excluding tritium, are mostly near MDA.

Before the NPP operation and during the pilot operation, the ^{90}Sr concentration decrease was also observed in these profiles at the tributaries and at the outflow from the Orlík Reservoir. The monitoring period was shorter in this case; therefore the ^{90}Sr data set was evaluated for all monitored profiles together (C2) (see the Figure 2). For additional information, there are given data measured by T.G.M. WRI in the Vltava basin after the nuclear weapons tests (A), results of the Danube River expedition (B) and further the results of the earlier monitoring in the NPP Temelín vicinity from 1981-1984 (C1). In the first evaluated period after the nuclear weapons tests and before the Chernobyl accident, the observed half-life was 6.8 years; in the second period 1992-2003, after the Chernobyl accident, it was 9 years. In the final stage of the monitoring in 2003, the strontium-90 concentrations were in range from 4.9 Bq/m^3 - 8.5 Bq/m^3 in the Orlík Reservoir tributaries and at the Vltava Solenice profile downstream of the reservoir. In connection to the artificial radionuclides occurrence after the Chernobyl accident in the NPP Temelín vicinity, it is needed to note that the Temelín vicinity belongs to the most contaminated areas in the Czech Republic due to the local rainfall in May 1986.

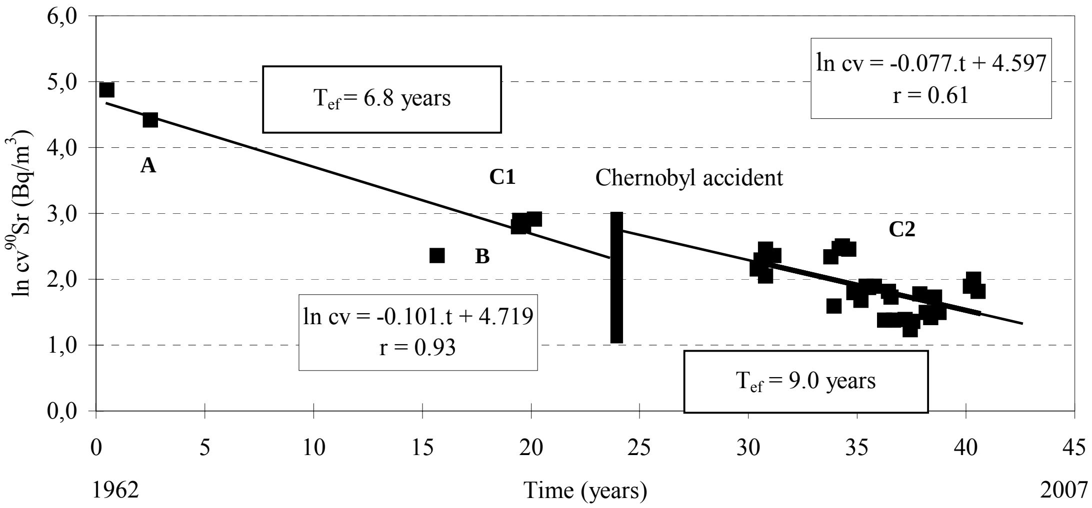

Figure 2. Time changes of ^{90}Sr concentration in the surface water in the periods 1963-1986 and 1986-2003, A - Vltava Prague-Podolí profile, B - average value of Donau Expedition 1978, C - NPP Temelín vicinity, the Orlík reservoir tributaries and outflow.

Our knowledge of the artificial radionuclides residual contamination in the hydrosphere of NPP Temelín vicinity in 2003 is elaborated by the previously published results [2]. The NPP Temelín vicinity monitoring assessment corresponds also with the results of similar studies of the changes in the water environment contamination after the Chernobyl accident made abroad. For example, Zibold et al. [3] observed the first faster phase of ^{137}Cs concentration decrease in the period 1986-1988 and the second slower phase in 1989-2000 in semi-logarithmic scale, when they assessed the results of their fresh water lakes monitoring. Based on the long-term radioecological monitoring of the radioactive contamination of the Sevastopol bays and river Dnieper respectively, Egorov et al. [4] calculated that the observed half-live was 5.9 years for ^{137}Cs in water.

In contrast to ^{137}Cs concentrations, the results of the ^{90}Sr concentrations observation showed neither the change in the slope of the decrease nor associated change in the parameters of the kinetic equation of the first order. The similar effective ^{90}Sr half-lives observed in the period after the nuclear weapons tests and the Chernobyl accident correspond with the ^{90}Sr mobility assessment made by Cross et al. [5]. At an example of Finnish rivers, they did not observe any significant differences between the mobility of ^{90}Sr from nuclear weapons tests and from the Chernobyl accident. Monte [6] published the effective half-lives (long term component) for dissolved ^{90}Sr ranges from 4 years to 6 years for the period of 240 days to approximately 2000 days for the rivers of Dnieper, Prypiat, Rhine, Teterev and Uzh after 1st May 1986. Egorov et al. [4] calculated that the half-live was 6.9 years for ^{90}Sr in water for Sevastopol bays and river Dnieper respectively.

Concerning the radionuclides balance assessment in the NPP Temelín waste water, the tritium amount released with the waste water is the most significant according to both of the project documentation and present experience. In the profiles uninfluenced by the NPP Temelín waste water, the long-term tritium concentration decrease has been observed, as it has been in the case of ^{137}Cs and ^{90}Sr.

In surface water in the Temelín vicinity, the average tritium concentration (background) was 1.5 Bq/l in 2003 and 1.1 Bq/l in 2002. The value of the effective half-life of tritium decrease was calculated as 8.2 years for the corrected tritium concentration kinetic (it means for the residual contamination after the nuclear weapons tests). In the period near the nuclear weapons tests, the observed half-life of tritium concentration decrease was about 5 years in surface water [7]. Based on the tritium monitoring results, further tritium concentration decrease can be expected at the uninfluenced profiles. Comparing the observed half-life with the tritium concentration decrease near the nuclear weapons tests, the tritium decrease rate slow down is obvious. Similar attention is paid to the tritium monitoring in rainfall water at the sites near NPP Temelín.

3.2 River bottom sediments

The sediments sampling sites include profiles uninfluenced by NPP Temelín even in future (Orlík Reservoir tributaries) and profiles potentially influenced by the NPP Temelín operation (profiles downstream of the waster water release in the Vltava Kořensko profile). All the results from the entire period are assessed together because even the results from 2001-2003 can be taken for practically uninfluenced by the NPP Temelín pilot operation due to the very low ^{137}Cs concentration in the released waste water compared to the residual sediments contamination.

The Figure 3 shows the radiocaesium isotopes concentration decrease in sediments is similar to the case of the surface water. The first order kinetic fully suits to the relation. The decrease rate slowing was not observed. For ^{137}Cs, the effective half-life of decrease was 6.1 years in the period 1990-2003. In case of ^{134}Cs, the concentrations have been mostly under the MDA since 1999. By these reasons, the trend assessment was ended in 1998 and the effective half-life was 1.6 years in the period 1990-1998. Theoretical trend of ^{137}Cs and ^{134}Cs concentrations ratio, which was derived from the Chernobyl accident primary ^{137}Cs/^{134}Cs ratio (2/1), was in a good harmony with the results of the monitoring in the Temelín vicinity until 1998. The time changes of ^{90}Sr concentration were also monitored. However, the decreasing trend of ^{90}Sr concentration in sediments has not been assessed yet, due to a small data set acquired and a frequent occurrence of values below MDA.

3.3 Fish

Fish samples were taken from the Orlík Reservoir and its tributaries and the radionuclides were measured with gamma-ray spectrometer. The time changes of the ^{137}Cs concentration are shown in the Figure 4. As in the case of the water samples, the first order equation was used to describe the ^{137}Cs

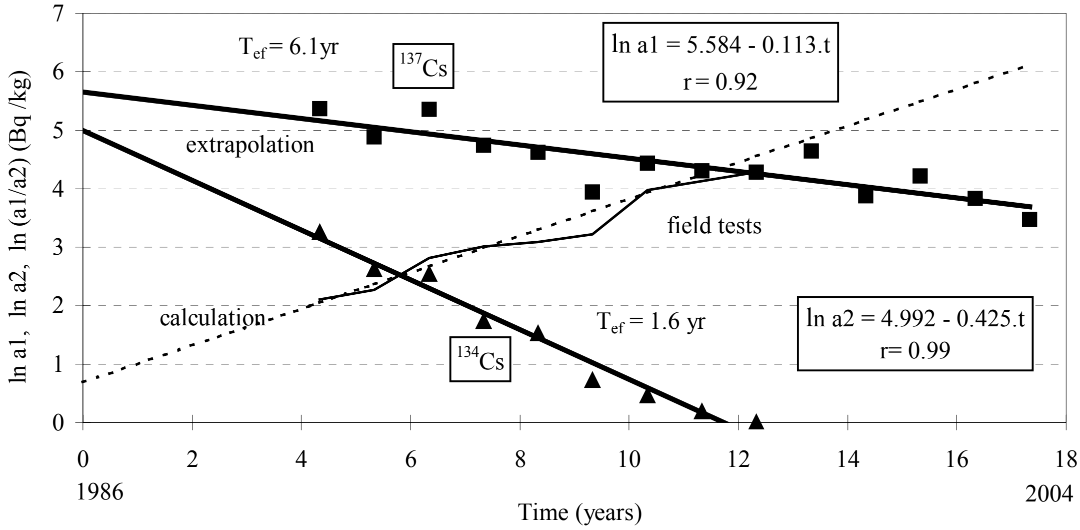

Figure 3. Time change in mean annual concentration of ^{137}Cs (a1) and ^{134}Cs (a2) in the bottom sediments in the Orlík Reservoir and their ratio, calculated from observed data and from the ratio in Chernobyl accident.

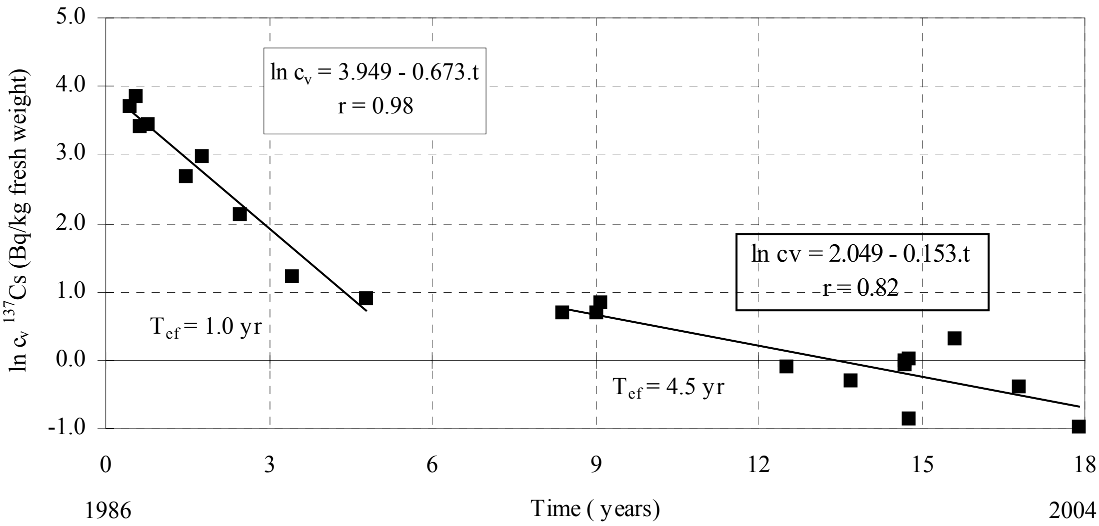

Figure 4. Time change in ^{137}Cs concentration in fish (fresh weight) sampled in Temelín NPP vicinity in the periods 1986 – 1990 and 1994 – 2002.

decrease in the fish samples. All the samples were assessed together; the piscivorious fish were assessed jointly with herbivorious fish. The assessment showed also the faster phase after the Chernobyl accident in the period 1986-1990, when the decrease half-life was 1.0 year. In the next period 1994-2003, the decrease rate was much slower; the observed half-life was 4.5 years. In 2003, the ^{137}Cs concentration in roach fish from The Orlík Reservoir was 1.5 Bq/kg in dry matter that corresponds with 0.4 Bq/kg of fresh matter. The ^{137}Cs occurrence in fish cannot be explained by the influence of the NPP Temelín because the caesium amount, released with NPP Temelín nuclear waste water during the pilot operation, was much lower than the residual contamination of water and sediments. Other radionuclides, detectable by the semiconductor gamma spectrometry, which would indicate other influence than the nuclear weapons tests and Chernobyl accident contamination, were not detected.

4. CONCLUSION

In surface water, river bottom sediments and fish samples from the NPP Temelín vicinity, the ^{137}Cs and ^{90}Sr concentrations show a decreasing trend, including the samples taken downstream from the NPP Temelín waste water outflow. In present, the detected ^{137}Cs concentrations in surfacewater draw near the MDA, despite the large volume samples (50 l) are processed. The influence of NPP Temelín to the concentration of these or any other activation and fission products (excluding tritium) was not detected. The ^{137}Cs and ^{90}Sr concentrations downstream of the NPP waste water release represent mainly the residual contamination after the nuclear weapons tests and the Chernobyl accident.

Influence of tritium, released with the NPP Temelín waste water, is measurable. The annual average tritium concentrations in the Vltava River correspond with the previously calculated prognosis for the conditions of the average and minimal guaranteed flow rates.

Acknowledgements

The authors wish to thank sponsoring organisations, the Ministry of the Environment, the Czech Power Company and Ministry of Education (project No MSM 223400008), for supporting the T.G.M. Water Research Institute and Institute of Chemical Technology research projects, whose main results are summarised in the paper.

References

[1] Hanslík E.J. Sedlářová B., Šimonek P., Temelín nuclear power plant, South Bohemia – Reference level of hydrosphere, prediction of impact, results from pre-operation period, in Proceeding of the International Congress ECORAD 2001, Aix-en-Provence, 2002, edited by F. Bréchignac (Radioprotection - colloques, Vol. 37, C1), p. C1-689-695.

[2] Hanslík E.J., Jedináková-Křížová V., Ivanovová D., Sedlářová B., Šimonek P., Observed half-lives of ^{3}H, ^{90Sr} and ^{137Cs} in hydrosphere in the Vltava River basin (Bohemia), in 7th Int. Conf. Biochemistry on Trace Elements, Uppsala, 2003, edited by G.R. Gobran and N. Lepp (Vol. 2, SLU), p. 380-381.

[3] Zibold G., Kaminsk, S., Klemt E., Smith J.T., Time-dependency of 137Cs activity concentration in freshwater lakes, measurement and prediction, in Proceeding of the International Congress ECORAD 2001, Aix-en-Provence, 2002, edited by F. Bréchignac (Radioprotection - colloques, Vol. 37, C1), p. C1-75-80.

[4] Egorov V.N., Polikarpov G.G., Mirzoyeva N.Yu., Stokozov N.A., Realization of a metrology and quality control for carrying out of radioecological monitoring in the Sevastopol bays, the Crimea, in Book of abstracts, International Symposium on In situ nuclear metrology as a tool for radioecology, Fleurus, 2002 (I.R.E)., p. 81.

[5] Cross M.A., Smith J.T., Saxen R., Timms D., An analysis of the environmental mobility of radiostrontium from weapons testing and Chernobyl in Finnish river catchments, Journal of Environmental Radioactivity 60 (2002) p.149-163.

[6] Monte L., Evaluation of radionuclide transfer function from drainage basins of fresh water Systems, Journal of Environmental Radioactivity, 26, 1995, p. 71-82.

[7] Bogen D.C., Welfrod G.A., White C.G., Tritium distribution in man and his environment, in Behaviour of tritium in the environment, Vienna, 1978 (IAEA),