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Modelling of Migration of ^{137}Cs Accidentally Released into a Small River

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ABSTRACT

Migration of ^{137}Cs accidentally released with wastewaters from a nuclear power plant into a system consisting of a wastewater channel and the Dudváh river in Slovakia was modelled. The SMC model used simulated the river by a series of mixed 'reactors' representing sections of the river. Input data for the model were obtained partially by measurements and partially by calibration of the model using several sets of data for activities of river bottom sediments. In this way the space- and time-averaged value of the coefficient of exchange between suspended solids and bottom sediments in the river was obtained and the sediment distribution along the river was refined. The relatively good agreement between the measured and calculated activities of ^{137}Cs in bottom sediments proved that the activities can be modelled reasonably well using the averaged parameters in the model.

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INTRODUCTION

Modelling radionuclide migration in surface streams requires a suitable migration model and input data for the model. There exists a wealth of mathematical models designed for this purpose; only a few of them, however, properly take into account the interaction (uptake and release) of radionuclides with suspended solids and bottom sediments (Onishi *et al.*, 1981; Codell *et al.*, 1982; IAEA, 1985; Santschi & Honeyman, 1989). Recently we discussed the importance of such interactions (Beneš *et al.*, 1989, 1992*a*; Beneš & Černík, 1992) and proposed a simple migration model (Beneš & Černík, 1990; Černík, 1991) in which both kinetic and other parameters of the interaction were considered.

A major problem in the use of mathematical models is often the lack of appropriate input data. This problem is usually encountered with rather sophisticated migration models requiring many input parameters, but it may be critical even for the application of a relatively simple model to systems which are poorly characterized in their hydrodynamic and sedimentary parameters. The problem can sometimes be overcome by calibration of the model using one or more sets of calibration data. In this paper such a calibration is carried out in order to describe the migration of radiocaesium in a small stream contaminated by an accidental release of wastewaters from a nuclear power plant.

THE MODELLED SYSTEM AND MODEL USED

The modelled system is depicted schematically in Fig. 1. It consists of two nuclear power plants (NPP) at Jaslovské Bohunice, Slovakia, the water channel Manivier and the adjacent river Dudváh. One of the power plants (V1) operates two 440-MW pressurized light water reactors, the second plant (A1) is being decommissioned. Both power plants are sources of radioactive wastewaters which are regularly released via their drainage system. The wastewaters enter the artificial water channel which carries water from the cooling towers of plant V1 5·2 km to the river Dudváh. The water channel passes through one village but its water is not used to any significant extent. The river Dudváh is a small freshwater stream with a partially regulated, shallow bed 5–11 m wide. It flows through several villages and its water is extensively used for irrigation of neighbouring fields. It also receives wastewaters from the local alcohol producer, BIOPO, carried in the water channel denoted here by the same name, and discharges into the river Váh 13·6 km from the confluence with the Manivier.

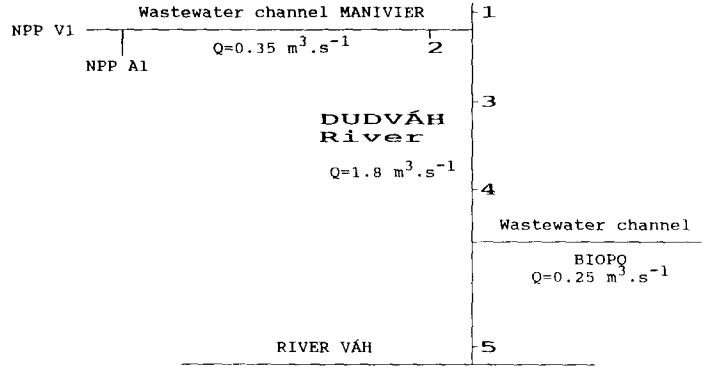

Fig. 1. Schematic picture of the system modelled (not to scale).

The regular discharges of radioactivity into the system are limited so that the activities of fission products and activated corrosion products do not exceed 37 Bq dm^{-3} in the water of the Dudváh (the limit for ^{3}H is $1{\cdot}95 \times 10^{5}$ Bq dm^{-3}). In January 1989 an accidental release of wastewaters containing about 10^{10} Bq ^{137}Cs occurred from nuclear power plant A1, which contaminated bottom sediments of the Manivier and Dudváh high above the level normally encountered. For a certain time after the accident the activity of ^{137}Cs in water in the Dudváh was mainly controlled by the release of ^{137}Cs from contaminated sediments, not by the regular discharges of wastewaters. (Measurements and calculations have shown that this time was at least several months and depended on the position of sampling site in the river — see Beneš *et al.*, 1992*c* and Černík, 1991.) Moreover, the increased activity of the sediments might have contributed to external radiation doses to local residents. Therefore, the activity of the sediments was regularly measured and modelling of radiocaesium migration was employed both to predict the activity of water and sediments in the river and to calculate the ^{137}Cs activity entering the Váh.

Modelling was limited to the Dudváh for several reasons. Information about the migration of radiocaesium in the Manivier is less important in view of its limited use. Retention of radiocaesium in the channel is relatively low because of the rapid water flow in the paved and straight bed of the channel and because of the absence of large amounts of bottom sediments. More information is available on the flow rate, concentration and forms of radiocaesium in the channel than on the same parameters in the drainage system of the nuclear power plants. Consequently, discharge from the channel can be better used as a defined source of ^{137}Cs than the outlet of the drainage system.

The SMC model used was originally developed for testing the effect of the interaction of radionuclides with suspended solids on the migration of

radionuclides in freshwater streams (Černík, 1991). It describes one-dimensional transport of a contaminant in suspended and dissolved form and thus it includes interaction of radionuclides with suspended sediments, resuspension, transport and sedimentation of sediments.

The modelled stream is divided into a series of longitudinal sections represented in the model by mixed reactors. Each section (reactor) contains a certain amount of water, suspended solids (sediments), 'active' bottom sediment (capable of resuspension) and radionuclides in three forms: dissolved, particulate (adsorbed on suspended solids) and adsorbed on 'active' bottom sediment. Water with suspended solids enters each reactor during certain time intervals called simulation periods. The period is equal to the length of stay of the water in the reactor, simulating the time of flow of water through a given section of the stream, represented by the reactor. After the simulation period, water leaves the reactor and enters the next reactor.

During the stay of water in the model reactor some of the suspended solids in the water are sedimented and completely mixed with 'active' sediment. Then part of the 'active' sediment is resuspended. This process is characterized by a coefficient of exchange of solids between water and sediment (EC) and leads to the redistribution of radionuclide between water column and bottom sediment during each simulation period. After the exchange, which is considered to occur immediately after water enters each tank (i.e. at the beginning of each simulation period), redistribution of the radionuclide between dissolved and particulate forms in water proceeds as described by the kinetic and equilibrium parameters of the uptake or release of the radionuclides by suspended solids obtained by laboratory experiments (Beneš & Černík, 1992; Beneš *et al.*, 1992*a,b*).

The model calculates changes in the distribution of the radionuclide between dissolved and particulate forms during passage of water through all sections and time changes in concentration of the radionuclide in water and sediments of each section in a series of consecutive simulation periods. For this purpose three submodels are used, describing the transport of water, sediment and radionuclides, respectively.

Hydrodynamic submodel

The hydrodynamic submodel simulates transport of water in the system. It is based on a balance equation describing the change in water volume in the ith section ΔV in time interval Δt:

$$(\Delta V/\Delta t)_i = Q_{i-1} - Q_i + QL_i \tag{1}$$

where Q_{i-1} is input flow of water from the upstream section to the ith section; Q_i is output flow of water from the ith section; and QL_i is input

flow of water to the ith section from a tributary stream. In the modelling of radiocaesium transport in the Dudváh no changes in water flow were assumed and $\Delta V/\Delta t$ was set at zero.

Sediment transport submodel

The sediment transport submodel simulates transport of sediment in water along the stream, exchange of sediment between water column and bottom of the stream, and changes in amounts of bottom sediments in sections of the stream. It uses a balance equation for the change in suspended solids content in the ith section $\Delta[S.V]$ in time interval Δt:

$$(\Delta[S.V]/\Delta t)_i = S_{i-1}.Q_{i-1} + SL_i - S_i.Q_i - SED_i + RS_i \qquad (2)$$

where S_i (S_{i-1}) is the concentration of suspended solids in section i ($i-1$), respectively; SL_i is the input of suspended solids to section i from a tributary stream; SED_i and RS_i are the sedimentation and resuspension rates (fluxes) of sediments in section i.

For this specific application of the model a constant concentration of suspended solids was assumed for the whole period modelled and, considering that $(\Delta V/\Delta t) = 0$, $(\Delta[S.V]/\Delta t)$ was set equal to zero. Further simplification, justified by the use of time-averaged input data for the model (see below), consisted in assumption of absence of changes in the distribution of bottom sediments along the stream throughout the modelling period. Normally the distribution is time-dependent and can be calculated from differences between SED_i and RS_i using eqn (2).

Radionuclide transport submodel

The radionuclide transport submodel simulates transport of a radionuclide dissolved in water and adsorbed on suspended solids. Redistribution of the radionuclide between the water and suspended solids is described by a kinetic equation for a two-step reversible reaction (Beneš *et al.*, 1992*a*; Beneš & Černík, 1992). It can also be described using a kinetic equation for a one-step reaction or by the distribution coefficient K_D. The deposition of radionuclides in the bottom sediments of the stream depends on the exchange between suspended solids and bottom sediments characterized by the exchange coefficient EC.

INPUT DATA FOR THE MODEL

Application of the SMC model requires knowledge of a number of input data. Apart from the exchange coefficient EC and the kinetic/equilibrium

parameters of radionuclide uptake or release by suspended solids mentioned above, the input data needed are: number of sections, width of stream, amounts of resuspendable sediments in individual sections expressed as thickness of the sediment layer, concentration of the radionuclide in sediments of individual sections at the start of modelling, water flow rate, linear speed of flow, length of simulation period, concentration of suspended solids and the source term, i.e. kinetics and forms of radionuclide input to the system.

For the purpose of modelling, the Dudváh was divided into 29 sections (reactors of the model) from the discharge point of the Manivier channel to its confluence with the Váh. Each section was characterized by its width and by the linear speed of water corresponding to a water flow rate of $1{\cdot}8\,m^3\,s^{-1}$ upstream of the outlet of channel BIOPO and $2{\cdot}05\,m^3\,s^{-1}$ downstream of the outlet. The length of each section varied between 350 and 600 m according to the linear speed of water, so that the residence time of water in the section (the simulation period) was always equal to 20 min.

The flow rates given above are approximate average annual flow rates at profiles shown as 3 and 5 in Fig. 1, respectively. They were calculated from hydrometeorologically derived data published by the Slovak Hydrometeorological Institute (1984) and from our own values measured in 1989–90. The actual flow rate in the Dudváh may vary from 0·8 to $3{\cdot}3\,m^3\,s^{-1}$. The variability is also caused by the release of water into the Dudváh from a dam located upstream and by the use of water from the river for irrigation during the spring and summer periods. As these actions are very irregular and unplanned, it was very difficult to predict or assess the actual water flow rate in the Dudváh. This in turn adversely affected the accuracy of modelling as the flow rate is a very important parameter. On the other hand, the use of a time-averaged water flow rate allowed the use of average values for the other input data for the model.

The average annual flow rates in both the channels are given in Fig. 1. In the case of the Manivier channel the average rates for 1989–90 as obtained from the nuclear power plants were used. However, the flow rate can vary between 0·14 and $0{\cdot}93\,m^3\,s^{-1}$. The water flow rate in channel BIOPO was calculated as the difference between reported annual flow rates at profiles 3 and 5 in the Dudváh (Slovak Hydrometeorological Institute, 1984).

The distribution of bottom sediments along the river determines the amounts of resuspendable bottom sediments in individual sections of the river. The real amounts are extremely difficult to determine, partly because the fine, resuspendable sediments are mixed with coarse, non-resuspendable sediments moving only on the bottom. Such mixing will

render a part of the fine sediments non-resuspendable due to their burial and may affect the thickness of the layer of 'active' sediments. The amounts were first roughly assessed during the measurement of other parameters of the river and then refined in the course of calibration of the model (see below). They were expressed as thickness of the layer of resuspendable sediments. Because in the real river the sediments were unequally distributed across the river bed, two parameters were determined for each section of the river: thickness of sediments in two 30-cm-wide belts adjacent to the banks of the river and thickness of the remaining sediments in the centre of the river bed.

The activities of bottom sediments in the Dudváh were determined by gamma spectrometry. The sediments were sampled along the river on 26 January, 31 January, 30 August and 1 November in 1989 and on 11 September 1990. The samples were wet sieved through a 2-mm sieve and their ^{137}Cs content was measured using a PGT HP Ge detector with relative efficiency of 33% at 1332 KeV (Slávik *et al.*, 1985). The results expressed on a dry weight basis served either as input data for calibration of the model or were used for verification of model predictions. For the sake of brevity they are presented only in figures together with the calculated values (see the following paragraphs). It is worth mentioning that the sites for sediment sampling were chosen before the river was divided into sections for modelling the radiocaesium migration. Consequently the samples of sediments analysed do not necessarily represent typical sediments of the given section of the model river. This may contribute to the inaccuracies of the calibration.

Concentration of suspended solids is also a very important input parameter as it strongly affects the interaction of radiocaesium with the solid phase (Beneš *et al.*, 1992*b*). For some rivers it can be calculated from water flow rate using an empirically determined relation between the concentration and water flow rate. This is, however, not possible in the Dudváh, where we found rather wide variation in the concentration (typically 18–72 $mg\,dm^{-3}$ with extreme values reaching 300 $mg\,dm^{-3}$) without any correlation with the water flow rate. In this work we used the average value of 34 $mg\,dm^{-3}$.

A recent study of the interaction of radiocaesium with suspended solids of the river Dudváh has shown that five out of six factors whose effects were studied, can significantly affect the interaction (Beneš *et al.*, 1992*b*). It has been recommended that the effects of contact time, temperature and concentration of suspended solids be included into a migration model in the form of suitable kinetic and equilibrium parameters. The effects of composition of suspended solids and of water can be considered in the form of variability of the parameters. It was demonstrated that the kinet-

ics of radiocaesium uptake on suspended solids can be best described by a two-step kinetic model considering two consecutive or parallel reactions.

However, large errors associated with the inaccuracy of the other input data for the model mentioned above render an exact description of the interaction unnecessary. Therefore, only a simple one-step kinetic model of the interaction (Beneš *et al.*, 1992*a*) was used here. The parameters employed were $K_d = 14\,dm^3\,g^{-1}$ and $\lambda = 0{\cdot}020\,min^{-1}$, where K_d is the equilibrium distribution coefficient and λ is the apparent rate constant of the interaction, assumed equal for the uptake and release of radiocaesium by suspended solids. The parameters were obtained by laboratory experiments with unfiltered water from the Dudváh (see Beneš *et al.*, 1992*b* for details) and were adjusted to the concentration of suspended solids equal to $34\,mg\,dm^{-3}$. The effect of variable temperature was neglected in the modelling.

The source term should contain information on the time-course of the amount and physico-chemical forms (speciation) of radiocaesium input to the Dudváh from the Manivier. Two types of source terms must be considered in connection with the modelling of radiocaesium migration. The first term is the accidental release of wastewaters which strongly contaminated the system. The amount of the released radiocaesium, time-course of the release and the speciation of radiocaesium are not exactly known. The information required can be only partially derived from measurements made in the Manivier channel and in the Dudváh by three independent groups from 25 to 31 January 1989. The results were reported earlier (Slávik & Plško, 1989). Most of them refer to profile 3 in Fig. 1, which corresponds to section 6 of the model river, and are plotted in Fig. 2

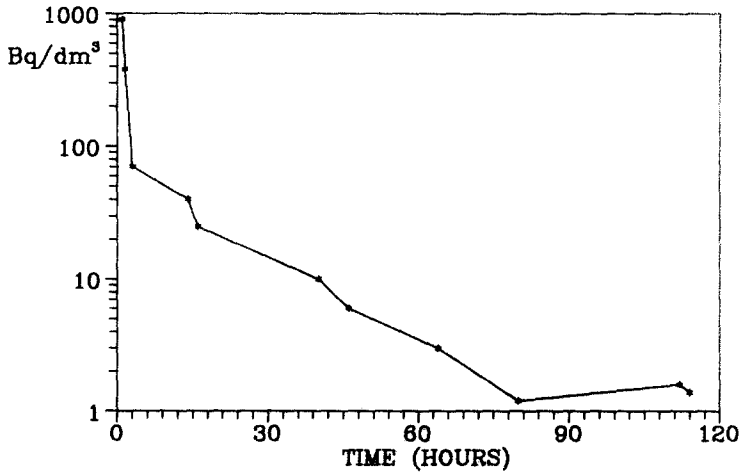

Fig. 2. Measured activity of ^{137}Cs dissolved in the water of the Dudváh river at profile 3 as a function of time elapsed from 6 pm, 25 January 1989.

as a function of time elapsed from 6 pm, 25 January 1989, when the first accidental ^{137}Cs reached section 6. From the results we calculated the amounts of radiocaesium entering section 1 of the river in 20-min intervals. The data characterize the first source term and are shown in Table 1. The total released amount derived by integration equals $1{\cdot}25 \times 10^{10}$ Bq ^{137}Cs, which is close to the value assessed earlier ($1{\cdot}3 \times 10^{10}$ Bq, Slávik & Plško, 1989).

The second source term describes the regular release of wastewaters containing a low level of radiocaesium activity and is applicable for the modelling of radiocaesium migration in normal conditions. In our case the amounts of radiocaesium entering the system were lower than those released from contaminated sediments soon after the accidental release. Nevertheless, the inputs of radiocaesium from regular releases were calculated from the data supplied by the staff of nuclear power plants A1 and V1 and included into the migration model. It was found that releases from plant V1 were much lower than those from plant A1 (more than 10 times). The average monthly release of ^{137}Cs in wastewaters from plant A1 from 5 to 20 months after the accident was 15 ± 12 MBq.

Only one result characterizing the speciation of the 'accidental' radiocaesium in the Manivier channel was available: during the passage of maximum concentration of radiocaesium through profile 2 (see Fig. 1), 18% of the radiocaesium was in particulate form (Slávik & Plško, 1989). Instead of this single value, which was not necessarily representative, we took the value obtained by calibration (20%, see the next paragraph) for the period of accidental release and 64% (as found experimentally—Beneš *et al.*, 1992*c*) for later intervals of the modelling.

The coefficient of exchange of suspended solids with bottom sediments depends on the sedimentation rate of the solids and on the rate of resus-

TABLE 1

Estimated Time-course (in 20-min intervals) of Activities of Water Entering the Dudváh During the Accident (calculated from the data in Fig. 2)

Interval no.	*Activity* ($Bq\,dm^{-3}$)	*Interval no.*	*Activity* ($Bq\,dm^{-3}$)
1	400	10	200
2	900	11	200
3	900	12	200
4	900	13	100
5	600	14	100
6	600	15	100
7	400	16	70
8	400	17	70
9	400	18	70
19–260:	activity = 97 exp ($-0{\cdot}018\,24 \times$ no. of interval)		

pension of the sediments. The SMC model used does not enable the coefficient to be predicted theoretically. Consequently, it must be determined by calibration.

MODEL CALIBRATION — ESTIMATION OF INPUT DATA

As indicated above, the modelling of radiocaesium migration using an SMC model requires knowledge of certain input data that cannot be easily or precisely determined by direct measurement or may be missing for other reasons. These parameters must then be estimated, the best value being one which maximizes the agreement between the distribution of radiocaesium in the system calculated by the model and the real distribution determined experimentally. This procedure is often called model calibration. If more than one input parameter is needed to be estimated, the calibration may become more difficult since a set of optimum parameters must be found. In such a case the parameter determined need not correspond to the actual or real situation in the system being modelled. However, if the results of the modelling are sufficiently accurate this fact does not necessarily invalidate the calibration procedure — it is simply a manifestation of the empirical nature of the model.

Three calibrations were carried out in this work with different sets of calibration data and/or different parameters to be determined. The first calibration used the data on the time-course of the accidental radiocaesium release (Table 1), the amounts of bottom sediments originally assessed for individual sections of the river from very rough measurement, and the activities of radiocaesium in sediments along the river on 31 January 1989. From this first calibration, the average value of the exchange coefficient EC was calculated (EC = 0·186), which was then used for all other calculations. In those sections where the assessed amount of bottom sediments did not permit satisfactory agreement between calculated and input activities of the sediment to be achieved, the amount was adjusted so that better agreement was obtained. As the agreement in the first two sections of the river strongly depends on the distribution of radiocaesium between the particulate and dissolved forms in the Manivier channel, the optimum distribution was also calculated (80% dissolved). The final agreement between the real and calculated activities of radiocaesium in bottom sediments is shown in Fig. 3, where the activities are compared. This calibration is valid for the first part of the river (sections 1–6) and for the first 6 days after the accident.

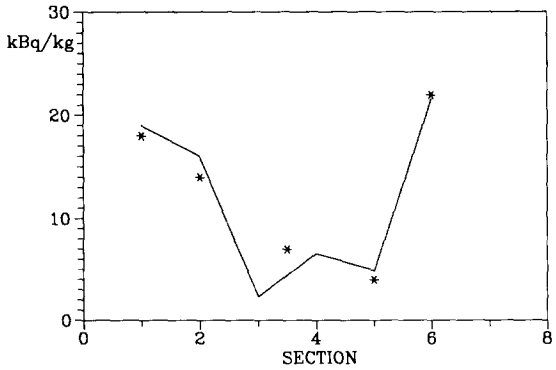

Fig. 3. Comparison of experimentally determined (*) activities of ^{137}Cs in bottom sediments of the Dudváh river on 31 January 1989 with those calculated in the calibration of the model (interconnected by the line).

Further calibrations served for extension of the validity to longer periods of time and to the whole length of the river. Calculation of the change in activity of sediments in profile 3 (section 6), with time from January 1989 to September 1990 revealed that additional radiocaesium entered the river, without doubt due to its release from contaminated sediments in the drainage system of the nuclear power plant A1 or in the Manivier channel. The calculated activities decreased with time more rapidly than the measured activities, if only radiocaesium input from regular releases reported by the nuclear power plants was considered. The additional concentration of ^{137}Cs in water of the Manivier due to its release from sediments was determined by calibration as $16\,mBq\,dm^{-3}$ for the time period studied.

The third calibration aimed at optimization of the distribution of resuspendable sediments along the river. It was based on the comparison of activities of the sediments sampled on 31 January and 1 November 1989. The distribution of sediments was varied so as to obtain the best agreement between the measured and calculated activities of sediments along the river. The agreement is shown in Figs 4 and 5, and the optimized distribution of resuspendable sediments is presented in Table 2. As can be seen, the calibration procedure using two sets of activity data resulted in worse agreement for sections 1–6 (Fig. 4) than the procedure using only one set of activities (Fig. 3), despite the optimized distribution of sediments. This is due to simplifications made in the calibration procedures. The values given in Table 2 only roughly reflect the distribution of real sediments and their accuracy has no relation to the possible accuracy of determining the actual distribution of resuspendable sediments by measurement.

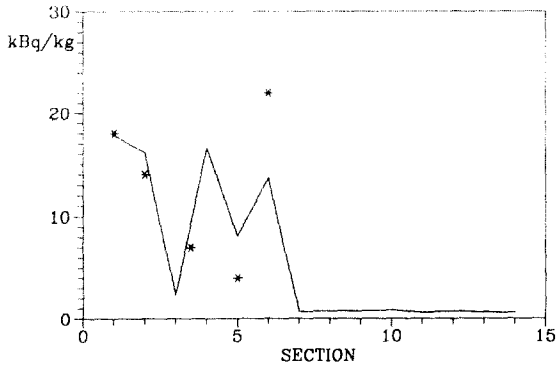

Fig. 4. Comparison of experimentally determined (*) activities of ^{137}Cs in bottom sediments of the Dudváh river on 31 January 1989 with those calculated in the calibration of the model (interconnected by the line).

It is clear that the input data obtained by the calibration represent only rough approximations to the data needed for accurate description of the modelled system. Probably the most ill-defined parameter is the exchange coefficient, due to the implicit assumption that it is independent of water flow rate and constant along the river. The actual distribution of sediments along the river can also vary in time. However, the simple model and its calibration do not permit a much better description of the system unless more precise data on water flow rates and the distribution of sediments are available.

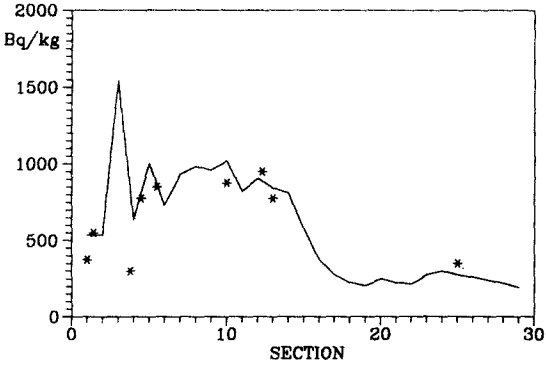

Fig. 5. Comparison of experimentally determined (*) activities of ^{137}Cs in bottom sediments of the Dudváh river on 1 November 1989 with those calculated in the calibration of the model (interconnected by the line).

TABLE 2
Thickness of the Layer of Resuspendable Bottom Sediment in the Mid-stream and Along the Banks (30-cm belts) of the Dudváh River Determined by Calibration

Section no.	*Thickness (cm)*		*Section no.*	*Thickness (cm)*	
	Mid-stream	*Banks*		*Mid-stream*	*Banks*
1	0	3	16	8	10
2	0	5·7	17	6	10
3	5	10	18	6	10
4	0	5	19	6	10
5	0·3	7	20	4	10
6	0	4	21	4	10
7	15	15	22	4	10
8	10	15	23	2	10
9	5·5	6	24	1	10
10	5	5	25	0·6	10
11	6	3·6	26	0·5	10
12	5	3·6	27	0·5	10
13	5	15	28	0·5	10
14	3	10	29	0·3	10
15	5	10			

VERIFICATION OF THE CALIBRATED MODEL

The quality of the input data obtained by the calibration can be most easily checked by comparison of the measured distribution of radiocaesium in the system with the distribution calculated using the calibrated model. Several such comparisons were carried out. The first one compared the activities of radiocaesium in sediments along the river on 11 September 1990, i.e. about 20 months after the accident. As can be seen in Fig. 6, the character of the distribution is quite well predicted by the model, although the calculated activities in certain sections of the river are significantly lower than those measured. Given the uncertainties discussed above, the results of the calculation seem acceptable. Still better agreement was found when time changes in the activity of sediments in river profile 3 were compared, as shown in Fig. 7.

It can therefore be concluded that the activities of bottom sediments in the Dudváh after the accident can be reasonably well modelled using time-averaged parameters of the model. On the other hand, such parameters do not enable accurate simulation of actual (time-dependent) distribution of the radionuclide between dissolved and particulate forms in the same river

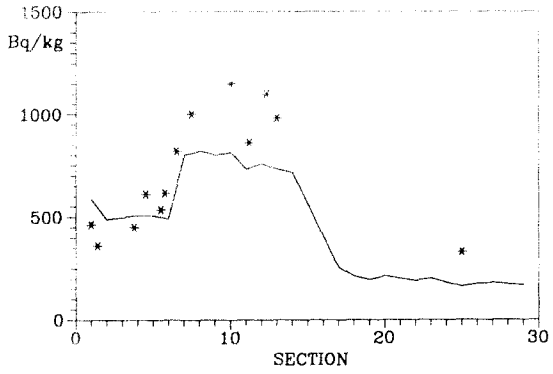

Fig. 6. Comparison of experimentally determined (*) activities of ^{137}Cs in sediments of the Dudváh river on 11 September 1990 with those calculated by the calibrated model (represented by the line).

(Beneš *et al.*, 1992*c*). Such a simulation would require time- and site-dependent input data for an actual state of the river and different method of model calibration.

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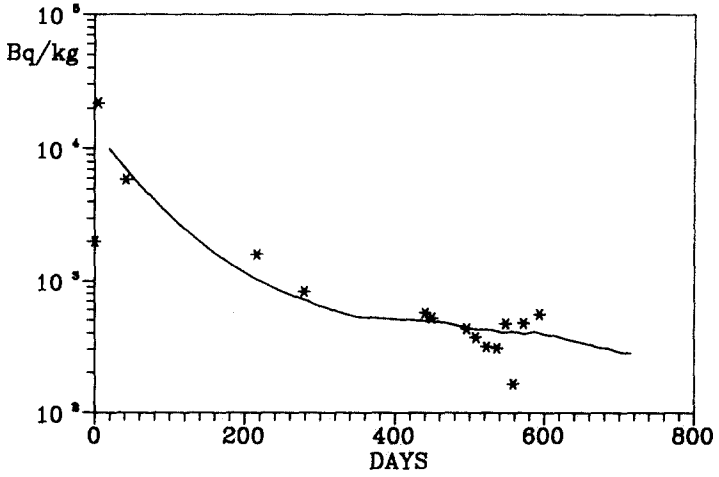

Fig. 7. Comparison of experimentally determined (*) and calculated (represented by the line) activities of ^{137}Cs in bottom sediments of the Dudváh river at profile 3 (section 6) as a function of the time elapsed from the accident.

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