The "AQUASCOPE" simplified model for predicting ^{89,90}Sr, ^{131}I and ^{134,137}Cs in surface waters after a large-scale radioactive fallout.

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Abstract

Simplified dynamic models have been developed for predicting the concentrations of radiocaesium, radiostrontium and ^{131}I in surface waters and freshwater fish following a large-scale radioactive fallout. The models are intended to give averaged estimates for radionuclides in waterbodies and in fish for all times after a radioactive fallout event. The models are parameterised using empirical data collected for many lakes and rivers in Belarus, Russia, Ukraine, UK, Finland, Italy, The Netherlands and Germany. These measurements span a long time period after fallout from atmospheric nuclear weapons testing and following the Chernobyl accident. The models thus developed were tested against independent measurements from the Kiev Reservoir and Chernobyl Cooling Pond (Ukraine) and the Sozh River (Belarus) after the Chernobyl accident, from Lake Uruskul, (Russia), following the Kyshtym accident in 1957 and from Haweswater Reservoir, (UK) following atmospheric nuclear weapons testing. The AQUASCOPE models (implemented in EXCEL® spreadsheets) and model documentation are available free of charge from the corresponding author.

INTRODUCTION

Many models have been developed for the transfer of radiocaesium (and to a lesser extent radiostrontium) to and in aquatic systems. Models for radionuclides in runoff water and lakes have been developed in, for example, the IAEA VAMP (IAEA 2000) and EU MOIRA (Monte 2001), ECOPRAQ (Håkanson 2000) and RODOS (Heling et al. 1997) projects. These models are relatively complex, being implemented in specialised software packages and (in general) having a relatively large number of required input parameters and model processes. It is believed that, in parallel with these more complex models, there is a requirement for simplified models for radiation protection which are implemented in more standard software systems. A main objective of the AQUASCOPE project (Smith et al. 2001), therefore, was to develop models which are simple in structure and are implemented in EXCEL® spreadsheets. These simplified models are clearer, more understandable and user-friendly than more complex approaches. In general, they also require much less model customisation with site-specific data to make predictions. The identification of only the key processes by analysis of extensive empirical data sets maintains the high predictive power of the models.

A key aspect of the models is that they are based on a very large database of field measurements of radionuclides in surface waters in a wide range of European countries. Long time series measurements of radionuclides in surface waters were collated from the AQUASCOPE project (Smith et al. 2001) and from previous EC and CIS funded projects. These measurements were complemented by data from literature studies. The measurements span a long time period after fallout from atmospheric nuclear weapons testing (NWT) and following the Chernobyl accident. On the basis of this extensive empirical data set, models for prediction of radionuclide activity concentrations in both rivers and lakes were developed. The target variables were radioactivity concentration in water (dissolved phase) and in non-predatory and predatory fish. Estimations were made for whole fish, though in the case of strontium, corrections may be made for activity concentrations in the flesh or bony parts separately.

It has been observed that in relatively shallow 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 (Bulgakov et al. 2002). In the models presented here, a closed lake is defined as a lake in which the water residence time is greater than 1 year and the mean depth is less than 7 m. In such closed lake systems, long term radioactivity in the lake is dominated by resuspension and remobilisation from the sediments, whereas in open lake systems (with much faster water turnover), inputs of radioactivity from the catchment control the long term contamination of the lake. Separate models have therefore been developed for closed and open lake systems.

The models presented here are not in themselves novel. Rather, they represent a synthesis of many previous studies into modelling the environmental transport of radioactivity in rivers and lakes. The novel aspect of the work presented here is the synthesis and parameterisation of these many models, and the development and "blind" testing of a simplified operative software tool for predicting the fate of radionuclides in rivers and lakes.

MATERIALS AND METHODS

River/runoff model

The models used to predict time changes in radionuclide runoff are based on those described in Monte (1995) and Smith et al. (2000a). The radionuclide concentration in runoff or river water, C_R (Bq m^{-3}), is given by:

$$C_R(t) = D_c.(\alpha.e^{-(\lambda+k_1)t} + \beta.e^{-(\lambda+k_2)t} + \gamma.e^{-(\lambda+k_3)t}) \quad (1)$$

where λ (y^{-1}) is the decay constant of the radionuclide and D_c is the radionuclide deposition to the catchment (Bq m^{-2}). α, β, γ (m^{-1}) and k_1, k_2, k_3 (y^{-1}) are empirically determined (radionuclide-specific) constants. The k values may be expressed as effective ecological half-lives, T_{eff}, where $T_{eff} \approx \ln 2/(k+\lambda)$. The three exponential terms in equation (1) represent, respectively: a fast "flush" of activity as a result of rapid washoff processes; a slow decline as a result of soil fixation and redistribution processes; and the very long term "equilibrium" situation. The time-dependent behaviour of ^{137}Cs in rivers is illustrated using measurements from the Pripyat river in the long-term after the Chernobyl accident (Figure 1).

Model for open lakes

A number of models of varying complexity have been developed to describe the mobility of radionuclides (most commonly ^{137}Cs) in lakes (Monte et al. 2003). Following a fallout event, radionuclides are removed from the lake to bottom sediments and through the outflow. Transfers to bottom sediments occur as a result of attachment to and settling of suspended particles and via direct diffusion across the sediment/water interface. In the longer term, radionuclides may potentially be remobilised from bottom sediments (particularly, for Cs, under anoxic conditions, see Evans et al. 1983) or resuspended by wave action.

Models have been developed for ^{137}Cs transport in lakes which incorporate all of the above processes (e.g. IAEA 2000; Monte 2003), however the objective here is to develop simplified models for aquatic systems which only model the key processes. In view of this, a simplified semi-empirical model is used. The structure of this model has been presented previously and has been extensively calibrated and tested for Chernobyl-derived ^{137}Cs in European lakes (Smith et al., 1997; 1999a). The reader may refer to these publications for more detail of this model.

It is assumed that direct deposition to the lake surface results in a short duration "spike" input to the lake, leading to an initial mean lakewater concentration $C_L(0)$ (Bq m^{-3}):

$$C_L(0) = \frac{D_L}{d} \quad (2)$$

where D_L (Bq m^{-2}) is the deposition per square metre of lake surface and d (m) the mean depth of the lake, calculated by dividing lake volume, V_L, (m^3} by lake area, A_L, (m^2).

In the short term, sediments act as a sink for radionuclides. In the longer term, however, when catchment inputs dominate over the direct deposition component, sediments act both as a sink for radionuclides and as a source (by physical resuspension and chemical remobilisation). It is assumed in the model that, in the long term, the dissolved-phase activity concentration in the lake is similar to that in the inflowing rivers or streams (i.e. no net uptake to or release from sediments). The model therefore assumes uptake by sediments of that component of radioactive contamination due to lake surface deposition and of the short term catchment runoff component. It assumes no net uptake by sediments during the period when the secondary catchment processes dominate.

Direct measurements of radiocaesium in streams flowing into lakes in Cumbria, UK, test the hypothesis that there is no significant net transfer of radioactivity to sediments in the long term. In Brotherswater, Devoke Water, and Loweswater it was shown (Smith et al. 1997) that activities in inflow streams are of the same order as the lakewater activities (Table 1). This similarity of inlet and lakewater activity concentrations implies that catchment loss is the dominant contamination process and that there is no significant net transfer of dissolved phase radioactivity to or from sediments during the secondary phase of contamination. This simplifying assumption, for open lakes, is also expected to hold for radiostrontium since transfers of strontium to bottom sediments are lower, and rates of catchment remobilisation are higher than for radiocaesium.

The model for predicting radionuclide activity concentrations in open lakes predicts the removal from the lake water of inputs from atmospheric fallout to the lake surface and inputs from the catchment:

$$C_L = \frac{D_L}{d} e^{-(K+\lambda)t} + \frac{D_c e^{-\lambda t}}{T_w}\left[\frac{\alpha(e^{-k_1 t} - e^{-Kt})}{(K - k_1)} + \frac{\beta(e^{-k_2 t} - e^{-t/T_w})}{(1/T_w - k_2)} + \frac{\gamma(e^{-k_3 t} - e^{-t/T_w})}{(1/T_w - k_3)}\right] \quad (3)$$

where α, β, γ, k_1, k_2, k_3 are the same parameters and have the same values as in the runoff model (equation 1) above. D_c is the deposition to the catchment and D_L is the deposition to the lake surface (Bq m^{-2}) and d is the lake mean depth. The rate of removal of radionuclides to the lake bed sediments and outflow occurs at rate K, where

$$K = 1/T_w + 1/T_L \quad (4)$$

T_w (y^{-1}) is the water residence time of the lake defined as the ratio of lake volume to rate of water inflow or outflow, estimated from the net rainfall to the lake and catchment:

$$T_w \approx \frac{V_L}{R.A_c} \quad (5)$$

where R (m y^{-1}) is the net rainfall, V_L (m^3) is the lake volume and A_c (m^2) is the catchment area.

T_L, the rate of removal of radionuclides to bed sediments (y^{-1}), is given by:

$$\frac{1}{T_L} = \frac{f_p v_p}{d} \tag{6}$$

where f_p is the particulate sorbed fraction, v_p (m y^{-1}) is the particle settling velocity and d (m) is the mean depth. The particulate sorbed fraction, f_p can be calculated from the radiocaesium distribution coefficient, K_d and the suspended solids concentration (s, mg l^{-1}) of the lake water.

Notice that the first term in square brackets in equation (3) differs from the second and third terms in that it accounts for uptake to sediments of the radioactivity transferred from the catchment in the short term after fallout ($K = 1/T_w + 1/T_L$). After this initial period it is assumed that there is no net transfer of radioactivity to or from sediments (in most cases a conservative assumption).

Closed lakes

For "closed" lake systems (defined here as lakes with water residence time > 1 year and mean depth < 7 m) it is assumed that the input of ^{137}Cs from the catchment can be neglected. It can be assumed (Bulgakov et al. 2002) that the remobilisation rate of radiocaesium from contaminated sediments declines inversely with time after fallout. We have simulated this decline over time using a series of two exponential equations, similar to the equations for river/runoff water used above:

$$\eta_1 . e^{-(\lambda + k_2)t} + \eta_2 . e^{-(\lambda + k_3)t} \tag{7}$$

Where the rate constants k_2, k_3 and the physical decay constant, λ, (all in y^{-1}) are as defined above. The constants η_1, η_2 (units: m^{-1}), are empirically determined from data on long term activity concentrations in closed lakes per unit of deposition to the lake surface, D_L.

The model for a closed lake system is a linear combination of the model for removal of direct deposition to the lake surface and the double exponential function (equation 7) describing remobilisation from sediments:

$$C_L = \frac{D_L}{d} e^{-(\mathrm{K}+\lambda)t} + D_L(\eta_1 . e^{-(\lambda + k_2)t} + \eta_2 . e^{-(\lambda + k_3)t}) \tag{8}$$

where k_2, k_3 and D_L are defined as in the open lake model above. η_1, η_2 are empirically determined constants and $\eta_1 + \eta_2 \ll 1$. The deposition to the lake surface D_L, appears in the second term describing remobilisation from sediments since the constants η_1, η_2, are estimated empirically from measurements normalised to unit deposition to the lake surface.

Models for fish uptake and excretion of radionuclides

It is known that the bioaccumulation of radioactivity in fish is determined by numerous ecological and environmental factors such as the trophic level of the fish species, the length of the food chain, water temperature and the water chemistry. Uptake may be via ingestion of contaminated food or direct transfers from the water via the gills. For most radionuclides the food chain is the primary uptake pathway, so a food uptake model will be used to estimate uptake rates. For strontium isotopes, a model for direct uptake via the water (Chowdhury and Blust 2001) will be used to estimate the intake rate. Though other studies (Kryshev, 2003) assume most uptake via the food pathway, in practice both assumed uptake routes lead to similar assimilation rates (Smith et al., unpubl. res.).

Note that uptake of radioactivity to biota does not significantly influence the amount of radioactivity in the water, so was ignored in the development of the physical transport models above.

The level of radioactive contamination of aquatic biota is commonly defined in terms of a concentration factor (*CF*) where

$$\mathrm{CF} = \frac{Activity\ concentration\ per\ kg\ of\ fish\ (wet\ wt)}{Activity\ concentration\ per\ cubic\ metre\ of\ water} \qquad \mathrm{m^3\ kg^{-1}} \qquad (9)$$

Previous studies on the accumulation of radiocaesium in fish have focused on the prediction of *CF* (sometimes termed the bioaccumulation factor, *BAF*, or aggregated concentration factor, *ACF*). Some models (e.g. Rowan and Rasmussen, 1994; Smith et al. 2000b) predict the water-fish *CF* for certain radionuclides using relationships with the water chemistry, for example using an inverse relation between the radiocaesium *CF* and the potassium concentration of the surrounding water.

For non-equilibrium conditions (e.g. in the short term after an accidental release), activity concentration of a radionuclide in fish, C_f (Bq kg^{-1}) may be modelled by a simple "two-box" model describing uptake from the water C_w (Bq m^{-3}) and release from the fish (Figure 2):

$$\frac{dC_f}{dt} = k_f C_w - (k_b + \lambda) C_f \qquad (10)$$

where k_f (m^3 kg^{-1} y^{-1}) is the rate constant describing transfers of ^{137}Cs to fish and k_b (y^{-1}) is the backward rate constant describing excretion of radioactivity from the fish. The ratio of these rate constants gives the equilibrium concentration factor, *CF* (m^3 kg^{-1}), of the radionuclide in fish relative to water:

$$\frac{k_f}{k_b + \lambda} = \frac{C_f}{C_w}(at\ equilibrium) = CF \qquad (11)$$

MODEL PARAMETERISATION

Estimation of runoff parameters

Radionuclide runoff is often (e.g. Helton et al. 1985) parameterised as the fraction of the total radioactivity in a catchment which is removed (i.e. transported to a river) each year. The model of Helton et al. (1985) assumes a "rapid" loss of activity from the catchment during the first year (λ_a, dimensionless) followed by a lower, constant rate of loss, λ_b (y^{-1}), in subsequent years. This differs from the model presented above (equation 1) which predicts a steadily declining loss rate rather than a constant rate in the long term. Helton et al. (1985), however, state (in agreement with equation 1) that "the assumption that λ_b remains constant rather than decreasing with time is probably a conservative one". A further difference in the model of Helton et al. (1985) and that presented in equation (1) is that equation (1) represents the fractional removal of activity from a catchment as a function of the flux of water out of the catchment (or the net rainfall) rather than a constant fractional removal independent of water flux (as in the Helton et al. (1985) model).

The data presented in Helton et al. (1985) also measures total runoff in dissolved and particulate phases whereas here only runoff in the dissolved phase is considered. These data will therefore lead to an over-estimate of dissolved phase activity concentrations considered here. Much of the radionuclide removal data is, however, reported in terms of the model presented by Helton et al. (1985). Equation (1) will be related to the Helton et al. (1985) model in order to utilise these valuable data.

During the first year after fallout (in the model presented here) the cumulative loss of activity from the catchment is given by:

$$\lambda_a = R \int_0^1 \alpha e^{-(k_1+\lambda)t} + \beta e^{-(k_2+\lambda)t} + \gamma e^{-(k_3+\lambda)t}\, dt \qquad (12)$$

where R (m y^{-1}) is the net annual rainfall (ie accounting for evapotranspiration) to the catchment. This is approximately equal to the annual flow of water from the catchment per unit surface area. As a conservative assumption, α will be estimated by assuming that all of the first year's runoff occurs during the initial fast loss phase (leading to a maximum overestimate of around a factor of 2). Integration of equation (12) then allows the initial runoff parameter in equation (1), α, to be related to the values of λ_a quoted by Helton et al. (1985):

$$\alpha \approx \frac{k_1 \lambda_a}{R} \qquad (13)$$

assuming that $k_1 + \lambda \gg 1\ y^{-1}$ (this assumption is validated below). This equation allows us to relate the runoff measurements reported by Helton et al. (1985) to equation (1) and hence to use their measurements in model parameterisation.

Estimation of runoff parameter values for Cs-137, Cs-134

Measurements of the change in ^{137}Cs activity concentrations as a function of time after fallout from Chernobyl were obtained for four European rivers (Voitsekhovitch et al. 1991; Monte 1995). These give (Table 2) a mean value of $k_1 = 13.2$ y^{-1} for the initial rate of decline in radiocaesium concentrations in rivers.

Data in the review of Helton et al. (1985) gives λ_a values in the range 0.001-0.02 which, using equation (13) with mean rainfall, $R \sim 1$ m y^{-1} and k_1 as above gives $\alpha \sim 0.013$ - 0.26 m^{-1}. The R value may be an overestimate for some of the catchments studied, so a slightly conservative value of $\alpha = 0.3$ m^{-1} will be chosen for the model.

The values of the decay constants, k_2, k_3 are estimated from studies of the time dependence of radiocaesium activity concentrations in many European rivers after both weapons testing and Chernobyl (Smith et al. 1999b; 2000a; 2000c). Estimated values are: $k_2 = 0.41$ y^{-1} and $k_3 = 0.02$ y^{-1}.

Work has shown (Hilton et al. 1993) that high long term runoff rates of radiocaesium (i.e. high β ,γ values) are related to boggy peat soils in the catchment. It is therefore proposed that models for the estimation of these parameters be based on the percentage coverage of the catchment by these boggy, organic soils (Hilton et al. 1993; Smith et al. 1998). The following linear relations have been used in the model:

$$\beta = \beta_1.(f_{\min}) + \beta_2.(f_{org}) \tag{14}$$

$$\gamma = \gamma_1.(f_{\min}) + \gamma_2(f_{org}) \tag{15}$$

where $\beta_{1,2}$ and $\gamma_{1,2}$ are empirically determined constants, f_{org} is the fraction (by area) of the catchment which is covered by boggy, organic soils and f_{min} (= 1 – f_{org}) is the fraction not covered by boggy, organic soils.

The values of β and γ have been determined by fitting equation (1) to measurements of radiocaesium runoff in various catchments of different coverage by organic boggy soils. Equation (1) was fitted to measurements of ^{137}Cs activity concentration in water, C_R, draining each of the catchments, after normalisation to unit fallout (by dividing C_R by deposition, D_c). The fit is shown in Figure 3 and gives the following estimated parameters for ^{137}Cs runoff:

$$\beta = 0.003(f_{\min}) + 0.05(f_{org}) \tag{16}$$

$$\gamma = 0.0002(f_{\min}) + 0.007(f_{org}) \tag{17}$$

Parameter values for the runoff model are summarised in Table 3.

Estimation of runoff parameters for Sr-90 and Sr-89

Measurements from Voitsekhovitch et al. (1991) give a value of the rate of decline of ^{90}Sr activity concentrations in rivers of $k_1 = 24.4$ y^{-1} during the weeks after the Chernobyl accident. This compares with a calculation made by Monte (1995) of

approximately 16 y^{-1} for ^{90}Sr in the River Dnieper. In the model it is (conservatively) assumed that $k_1 = 16\ y^{-1}$, the lower value of the two estimates.

Measurements of k_2 for ^{90}Sr concentrations in 11 Italian Rivers after weapons testing were similar, having value $k_2 = 0.13\ y^{-1}$ (Monte 1997). This can be compared with $k_2 = 0.09\ y^{-1}$, estimated from ^{90}Sr measurements in 5 catchments in Finland (Cross et al. 2001, from data in Salo et al. 1984) and with a mean of $0.09\ y^{-1}$ for 9 rivers in the Chernobyl area (Smith et al. 2001, from data supplied by the Ukrainian Hydrometeorological Institute, Kiev). Degradation of fuel particles in the vicinity of Chernobyl may have influenced this latter result, tending to decrease the rate of decline (Kashparov et al. 1999) in ^{90}Sr activity concentrations. For the model, a value of $k_2 = 0.09\ y^{-1}$ is chosen. In the Finnish catchments, equilibrium in ^{90}Sr activity concentrations appeared to have been achieved approximately 15 years after the peak in weapons test activity. It will therefore be assumed that in the very long term (> 15 y after fallout) radiostrontium activity concentrations decline only by physical decay (i.e. $k_3 = 0$).

The percentage of deposited Sr-90 removed from watersheds in the first year after fallout has been found to vary in the range 0.5 - 12.2 % (Helton et al. 1985), though the studies giving values at the high end of this range appear to have been carried out over shorter periods than one year after the fallout. It is not clear that these values have been corrected by accounting for changes in removal rates over time during the first year. It is assumed, therefore, that the values at the high end of this range are an over-estimate of the annual mean value for the first year. According to equation (13), a removal of 0.5 - 12.2 % in the first year gives $\alpha \sim 0.08 - 2.0\ m^{-1}$ for net rainfall, $R = 1.0\ m\ y^{-1}$ and $k_1 = 16\ y^{-1}$ as above. In a study of Sr-90 concentrations in 11 Italian Rivers, Monte (1997) observed values of $\alpha \sim 0.2\ m^{-1}$ in eight catchments, with 3 catchments showing a factor of 2 higher than this, $\alpha \sim 0.4\ m^{-1}$. Values of β for Sr-90 are around $0.03\ m^{-1}$ (from data in Salo et al 1984, Monte 1997), though as with α, variation is around one order of magnitude (Helton et al 1985).

The variation in ^{90}Sr river concentrations has been shown to be correlated with catchment characteristics, in particular the soil moisture content, organic carbon content and percentage of surface water and saturated bog in the catchment (Cross et al. 2002; Smith et al. 2004). From studies of NWT derived ^{90}Sr (Smith et al. 2004), catchments have been categorised as "organic" or "mineral" on the basis of their organic carbon content where organic catchments are defined as those with organic carbon content > 10 $kg\ m^{-2}$. Such catchments are likely to be high in organic boggy soils, for example those in upland soils in the UK and northern Scandinavia. The values of α, β, γ were determined for wet/organic and dry/mineral catchments separately by fitting equation (1) to measurements of NWT ^{90}Sr (Figure 4a,b), giving:

"Organic" catchments: $\alpha = 0.8\ m^{-1}$; $\beta = 0.03\ m^{-1}$; $\gamma = 0.005\ m^{-1}$.

"Mineral" catchments: $\alpha = 0.8\ m^{-1}$; $\beta = 0.005\ m^{-1}$; $\gamma = 0.003\ m^{-1}$.

Where there is doubt about the classification of a catchment, the default parameters are those for "organic" catchments, though this will lead to an over-estimate in most cases. Parameter values for the runoff model are summarised in Table 3.

Estimation of runoff parameters: I-131

Measurements made of I-131 in the River Calder after the Windscale fire (Jackson and Jones 1992) gave a maximum value of 1036 Bq l^{-1} in the river water and a maximum deposition of 1.1 x 10^6 Bq m^{-2}, leading to an estimate of $\alpha = 1.0$ m^{-1}. Because of its rapid rate of physical decay, it can be assumed for ^{131}I that $\beta = 0$, $\gamma = 0$.

Measurements from Voitsekhovitch et al. (1991) give a value of the rate of decline of activity concentrations in the early phase of $k_1 = 17.7$ y^{-1} for ^{131}I, which compares with calculations made by Monte (1995) which give $k_1 = 8.5$ y^{-1} for ^{131}I in the River Po, Italy. In the model it is assumed that $k_1 = 8.5$ y^{-1} for ^{131}I. Parameter values for the runoff model are summarised in Table 3.

Rate of removal of radiocaesium from lakewater, *K*

The process which determines radionuclide removal to lake sediments is still open to question: although most models consider particulate settling only (Robbins et al. 1992; Carlsson 1978) other workers (Santschi et al. 1986; Hesslein 1987) have shown that direct diffusional uptake may also be important. For open lake systems, direct diffusion to bottom sediments (and remobilisation) is ignored since correct modelling of this process is complex (Smith and Comans 1996). In addition, studies on ^{137}Cs transport in Cumbrian (Smith et al. 1997) and other European (Smith et al. 1999a) lakes suggest that it is of minor importance in open lake systems.

The radiocaesium removal rate, K, is a combination of removal to sediments and removal to the lake outflow (at rate $1/T_w$, where T_w is the lake water residence time). Relationships for predicting K (Smith et al. 1999a) are given in Table 4. Parameters required for the open lake model are summarised in Table 5.

Rates of removal (K) of Sr-89, Sr-90 and I-131 from the lakewater

The water-sediment distribution coefficient (K_d) of radiostrontium and ^{131}I are, in general, in the range 10^{-3} – 10^{0} m^3 kg^{-1} (Coughtrey and Thorne 1983; IAEA 1994; Mundschenk 1996) which typically gives a fraction in the solid phase, $f_p < 0.05$ for lakes (assuming that suspended solids concentration, $s < 50$ mg l^{-1}).

For 5 lakes in Cumbria, UK, the average value of the particle settling velocity, v_p was 365 m yr^{-1} (Smith et al. 1997), so if the sorption and particle settling values are unknown, the following estimate is made for the removal rate for radiostrontium using equations (4,6):

$$K = \frac{1}{T_w} + \frac{f_p v_p}{d} = \frac{1}{T_w} + \frac{3.65}{d} \qquad (18)$$

where it has been assumed that $K_d = 1$ m^3 kg^{-1} and using a typical suspended solids concentration, $s = 10$ mg l^{-1} giving $f_p = 0.01$. T_w is the water residence time of the lake and d is the lake mean depth.

In a study of two closed (i.e. $1/T_w$ is close to zero) experimental lakes in Canada, Hesslein (1987) presented measurements of the ^{89}Sr removal rate due to transfers to sediments of K = 3.65 y^{-1} at location (L226NE) and 0.73 y^{-1} at location (L226SW) giving an average removal rate to the sediments of $1/T_L$ = 2.2 y^{-1}. If lake mean depth information is not available, it is recommended, conservatively, that the removal rate parameter value is 0.73 y^{-1} so:

$$K = \frac{1}{T_w} + 0.73 \tag{19}$$

For I-131 the conservative assumption will be made that there is no transfer of activity to the bottom sediments since the reduction in activity concentration in the lakewater will be dominated by radioactive decay and transfers througth the outflow, hence $K = 1/T_w$. Parameters required for the open lake model are summarised in Table 5.

Radiocaesium in "closed" lakes

Parameters for radiocaesium in closed lakes were obtained by fitting the sediment remobilisation model (equation 8) to the long term average measured concentration (per unit of fallout) in nine closed lakes (see Table 6), assuming a rate of change over time inversely proportional to $\sqrt{t}$ (Bulgakov et al. 2002). This gave η_1 = 0.04 m^{-1}; η_2 = 0.0085 m^{-1} and k_2, k_3 parameter values as determined in the runoff model (i.e. k_2 = 0.41 y^{-1} and k_3 = 0.02 y^{-1}).

The rate constant for removal of radioactivity to the bed sediments, K, is determined from the models given above (Table 4). If the water residence time is unknown (as is often the case for long water residence time "closed" lakes) a default value of T_w = 10 years may be used. Parameters required for the closed lake model are summarised in Table 7.

Radiostrontium in closed lakes

For "closed" lake systems (lakes with water residence time > 1 year and mean depth < 7 m) it is assumed that the input of radiostrontium from the catchment can be neglected. A similar model to that for radiocaesium is used, except that, as observed by Konoplev and co-workers in Lake Svyatoe, Russia (Konoplev et al., in Sansone and Voitsekhovitch 1996) there is only a single component long-term decline in lakewater activity concentration as a result of sediment interactions:

$$C_L = \frac{D_L}{d} e^{-(K+\lambda)t} + D_L . \eta_1 . e^{-(\lambda+g_1)t} \tag{20}$$

The constants η_1 and g_1 are empirically determined and K is determined as in the open lake model above (equation 18). If the water residence time is unknown (as is often the case for long water residence time "closed" lakes) a default value of T_w = 10 years may be used.

In Lake Svyatoe, Russia, it was observed (Konoplev et al., in Sansone and Voitsekhovitch, 1996) that, in the long term, the ^{90}Sr activity concentration declined only at twice the rate of physical decay of ^{90}Sr (i.e. $\lambda + g_1 \approx 0.05$ y^{-1}). An estimated value of the rate of long term decline of radiostrontium in closed lakes is therefore: $g_1 = 0.025$ y^{-1} ($\lambda = 0.025$ y^{-1} for ^{90}Sr).

The value of η_1 is estimated from measurements (Vakulovsky, 1994; Sansone and Voitsekhovitch, 1996) of long-term ^{90}Sr activity concentrations in 14 closed lakes and slow flowing water bodies in the Chernobyl region (Table 8). From the measurements in Table 8, a slightly conservative estimate of $\eta_1 = 0.05$ m^{-1} is used, since the values estimated in Table 8 do not account for the presence of fuel particles in the sediments of many of these water bodies. Parameter values for the closed lake models are summarised in Table 7.

I-131 in closed lakes

For closed lakes it is assumed that transport of ^{131}I to bottom sediments and through the outflow is negligible in comparison with physical decay. The simplified AQUASCOPE model for prediction of ^{131}I in the water of closed lakes (lakes with water residence time > 1 year and mean depth < 7 m) is therefore:

$$C_L = \frac{D_L}{d} e^{-\lambda t} \tag{21}$$

Parameter values for uptake and excretion of radiocaesium in fish

The rate of uptake of radiocaesium is determined by the potassium concentration ([K^+], mmol m^{-3} ≡ μmol l^{-1}) of the lake (Blaylock 1982; Smith et al. 2000b). The uptake rate constant in fish, k_f, is given by:

$$k_f = \frac{Y k_b}{[K^+]} \quad (\text{y}^{-1}) \tag{22}$$

where k_f (m^3 kg^{-1} y^{-1}) is the rate constant describing transfers of ^{137}Cs to fish and k_b (y^{-1}) is the backward rate constant describing excretion of radioactivity from the fish (see equation 10 above). Y (mmol kg^{-1}) is an empirically determined constant.

For predatory fish, the parameter values (given in Table 9) of this model were empirically determined from measurements of radiocaesium in different species of predatory fish after Chernobyl (Smith et al. 2000b; 2002). The parameter values chosen were those determined for perch, since a large empirical data set was available for perch and comparisons with pike and trout suggest that the perch model will give good predictions for these species also. Observations after Chernobyl (Zibold et al. 2002) showed that radiocaesium activity concentrations in non-predatory fish reached equilibrium with activity concentrations in the water phase within less than one month after the accident. Values of Y and k_b (Table 9) for non-predatory fish are calculated such that equilibrium is reached on a timescale of the order of one month (i.e. $\ln 2/k_b = 0.083$ y so $k_b = 8.4$ y^{-1}) and such that:

$$CF\ (\mathrm{m^3\ kg^{-1}}) = \frac{k_f}{k_b} = \frac{Y}{[K^+]} = \frac{61.3}{[K^+]} \Rightarrow Y = 61.3\ \ (\mathrm{mmol\,kg^{-1}}) \qquad (23)$$

since it has been observed that for non-predatory fish, $CF \approx 61.3/[K^+]$ (Smith et al. 2000b). Note that $[K^+]$ is here measured in μmol l^{-1} (≡ mmol m^{-3}) rather than mg l^{-1}.

Accounting for the "size effect" in radiocaesium accumulation

The "size effect" of radiocaesium accumulation in fish results in an increasing contamination (per unit weight of fish) with increasing fish size (Elliott et al., 1992; Koulikov and Ryabov, 1992; Hadderingh et al., 1997). In the Kiev Reservoir and an old channel of the River Pripyat, near Chernobyl, Hadderingh et al. (1997) found that perch and pike (predatory species) showed a size effect in ^{137}Cs accumulation, whereas non-predatory species did not. Elliott et al. (1992) showed a logarithmic relationship between fish ^{137}Cs accumulation and wet weight. To account for the size effect, the fish uptake model is therefore modified (Smith et al. 2002) using a power law relationship between ^{137}Cs activity concentration in fish, C_f, and fish wet weight, w (g): $C_f \propto w^n$, where n is an empirically determined coefficient (Table 9). For non-predatory fish, it was assumed that there was no "size effect" of increasing bioaccumulation with increasing weight, so n was set at 0.

Parameters for uptake and excretion of radiostrontium in fish

There is less quantitative information available for uptake and retention rates of ^{90}Sr in fish than ^{137}Cs. A direct uptake model (Figure 2) is used for predicting uptake rates for ^{90}Sr. Like calcium, strontium is primarily absorbed in the bony parts of the fish (skeleton, head, fins, scales). Measurements made by Vanderploeg et al. (1975), quoted in Blaylock (1982) have determined relationships between fish-water CF (m^3 kg^{-1}) for ^{90}Sr, and [Ca] (mmol m^{-3}) in the surrounding water:

$$CF(muscle) = \exp(2.2 - 1.2\ln[Ca]) \qquad (24)$$

$$CF(bone) = \exp(6.7 - 1.2\ln[Ca]) \qquad (25)$$

Assuming that 20% of the wet weight of a fish is composed of bony parts, this gives a whole fish CF:

$$CF(whole\ fish) = \exp(5.1 - 1.2\ln[Ca]) \qquad (26)$$

Using these concentration factors, it is estimated that 95% of the strontium in a fish is found in the bony parts (skeleton, scales, fins) and only 5% in the soft tissues.

The Chowdhury and Blust (2001) model estimates the stable strontium uptake rate, j_{Sr} (mmol kg^{-1} hr^{-1}) as a function of the H^+ and Ca^{2+} concentration:

$$j_{Sr} = J_{\max Sr} \frac{\beta_{Sr}(H^+) + K_{iH}}{(H^+) + K_{iH}} \frac{(Sr^{2+})}{(Sr^{2+}) + K_{mSr}[1 + (Ca^{2+})/K_{iCa}]} \qquad (27)$$

where the constants of the model have values given in Chowdhury and Blust (2001) and Sr^{2+}, H^{+}, Ca^{2+} are measured in mmol m^{-3} (≡ μmol l^{-1}). The Sr uptake rate, j_{Sr}, is converted to the uptake rate constant in the model developed here, k_f (m^3 kg^{-1} y^{-1}):

$$k_f = \frac{24.365.j_{Sr}}{(Sr^{2+})} \tag{28}$$

where the uptake rate is divided by the Sr^{2+} concentration to convert the absolute Sr^{2+} uptake rate (i.e. mmol m^{-3} of Sr per hour per kg of fish) to the uptake rate per unit concentration (in Bq or mmol m^{-3}) in water. The factor 24 × 365 accounts for the change of units from h^{-1} to y^{-1}.

The Chowdhury and Blust (2001) model is somewhat complex and requires a number of water chemical variables ([Ca], [Sr], $[H^{+}]$) to be input. This model has been simplified by estimating possible ranges in the water chemical parameter values and carrying out a Monte Carlo analysis of the effect of variation in these parameters on the ^{90}Sr uptake rate. It was assumed that the stable Sr concentration ranged between 0.09 and 4.1 μmol l^{-1} and the Ca concentration between 50 and 3000 μmol l^{-1} as observed in a range of UK catchments (Hilton 1997; Neal and Robson 2000) of different surface geology. A range in pH of between 5 and 9 was assumed, and a random distribution of values of each chemical parameter within these ranges. Three hundred sets of possible parameter values were randomly generated. The ^{90}Sr uptake rate predicted by equations (27) and (28) was calculated for each set of chemical parameter values. The 300 calculated uptake rates were then correlated with each water chemical variable to determine the model sensitivity to each variable. It was found that $[Ca^{2+}]$ was the most important factor determining the strontium uptake rate (Figure 5). The strong negative correlation between $k_{f(Sr)}$ and $[Ca^{2+}]$ means that $k_{f(Sr)}$ can be estimated using the following simplified form of equation (28):

$$k_f = \frac{391}{[Ca]^{0.95}} \tag{29}$$

The excretion rate for strontium, k_b, is estimated from the uptake rate (determined from equation 29) and the concentration factor (Equation 26):

$$k_b + \lambda = \frac{k_f}{CF} \tag{30}$$

Parameter values of uptake and excretion of Iodine-131 in fish

Literature values for the concentration factor of ^{131}I are given in Table 10. There is less quantitative information available for uptake and retention rates of ^{131}I in fish than ^{137}Cs and ^{90}Sr, so the uptake rate can be estimated using a simplified model for uptake via the food chain. It was assumed that fish feed at their maximum daily rate and calculations were made for trout, a predatory fish about which there is good data

on feeding rates. For ^{131}I, the model does not differentiate between predatory and non-predatory fish, nor does it differentiate between fish of different weights (i.e. $n = 0$).

In cases where uptake is principally via ingestion, the uptake rate can be estimated by:

$$k_f = \frac{C_{food} \times D_{\max} \times \phi \times 365 \times 10^{-3}}{C_w \times w \times 10^{-3}} = \frac{CF_{food} \times D_{\max} \times \phi \times 365}{w} \quad (31)$$

where D_{max} (g d^{-1}) is the maximum daily intake (wet weight) of food by trout, w is the wet weight of fish in grammes and ϕ is the assimilation efficiency (the fraction of amount ingested which is absorbed by the fish). The factor 10^{-3} in the numerator is required to convert D_{max} from g d^{-1} to kg d^{-1} and the factor 10^{-3} in the denominator is required to convert w from grammes to kg. The factor 365 converts from d^{-1} to y^{-1}. C_{food} is the activity concentration of the food and CF_{food} is the concentration factor of the food (e.g. plankton for herbivores or herbivorous fish for piscivores). It is assumed that uptake to the prey of predatory fish is instantaneous, thus the activity concentration in the prey, C_{food}, is estimated from the concentration of radioactivity in the water, C_w, using the concentration factor, CF_{food}. CF_{food} is assumed in the model to be equal to the concentration factor of the fish (i.e. $CF_{food} = CF_{fish} = 0.040$ m^3 kg^{-1}).

It was assumed that fish feed at their maximum daily rate and calculations were made for trout, a predatory fish about which there is good data on feeding rates. Elliot (1975) developed an empirical model which estimates trout feeding rate for fish of different wet weight w (grammes) at different water temperatures, T :

$$D_{\max} = (4 \times 10^{-3}) A_D \times w^{b_1} \times \exp(b_3 T) \quad (32)$$

where A_D, b_1 and b_3 are empirically determined constants whose values are given in Elliot (1975). The factor 4 converts the dry weight feeding rates estimated by the Elliot (1975) model to feeding rate expressed in terms of wet weight as used in equation 31. The factor 10^{-3} converts D_{max} in mg per day estimated by the Elliot model to g per day.

For uptake of ^{131}I, the assumption is made that the assimilation efficiency is $\phi = 1.0$ (i.e. all ^{131}I ingested is assimilated) and that $CF_{food} = CF_{fish}$. Using equation (31) this gives a value of $k_f = 0.593$ m^3 kg^{-1} y^{-1} for a 1000 g fish at water temperature of 17^{o}C and, from equation (30), $k_b = k_f/CF_{fish} = 14.8$ y^{-1}.

RESULTS AND DISCUSSION

The models were "blind" tested using data which was not used to develop the models. The input data for each of the test scenarios is given in Table 11.

Radiocaesium runoff model

An example of the "blind" testing of the AQUASCOPE river/runoff model for ^{137}Cs in the river Sozh, Belarus, is shown in Figure 6(a). Model parameter values are given

in Table (3). The model was run using estimates of the ^{137}Cs deposition (D_c = 220 kBq m^{-2}) and fraction of organic soils (f_{org}=0.11) in the catchment derived from maps held by the Institute of Geological Sciences, Minsk. The model tends to over-estimate long-term activity concentrations in the river, though the measurements are within the estimated errors in the model. Errors in the model are due to errors in estimating deposition to the catchment and fraction of organic soils in the catchment, as well as uncertainties in the estimate of runoff from the different soil types.

Radiostrontium open lake model

The model for ^{90}Sr in open lakes was tested against measurements in Haweswater Reservoir, Cumbria, UK following fallout from nuclear weapons testing (Linsley et al. 1985). Since the catchment of Haweswater is dominated by shallow upland organic soils, runoff parameter values for "organic" soils (Table 3) were used. Model parameter values for the open lake model are given in Table (5) and user input parameters for this scenario are given in Table (11). Since the weapons test fallout occurred over a number of years, the model was modified to predict ^{90}Sr concentrations from each year's fallout. This was then summed to give the output for each year. As shown in Figure 6(b) the model gives good predictions for this scenario: the ^{90}Sr activity concentrations in water and their change over time are predicted well. It should be noted that the initial variation over time is strongly influenced by variation in fallout, so it is not unexpected that this is predicted well. However, the scenario is a good test of the long-term time changes, and of the model's ability to predict the level of ^{90}Sr activity concentrations in the reservoir. The scenario is also a good test of the runoff model since, for ^{90}Sr in open lakes, the long term activity concentration in water reflects the changing activity concentration of runoff water from the catchment.

Radiostrontium closed lake model

Following the 1957 Kyshtym accident, Lake Uruskul (a closed lake) was highly contaminated with ^{90}Sr and other radionuclides. Figure 6(c, d) shows the result of a "blind" test of the AQUASCOPE model against measurements of ^{90}Sr in water and fish of Lake Uruskul (Monte et al. 2002). Model parameter values for the closed lake model are given in Table (7) and values of user input parameters are given in Table (11). The predictions for water are very good, the underestimate at the start being largely due to measurement error as a result of the influence of other, short lived, radionuclides (Kryshev and Sazykina, 1999). The fish model predicts the maximum fish concentration quite well (to within a factor of two), but underpredicts the long-term continuing contamination. This may be because it underestimates continuing accumulation and retention of Sr in bony tissues.

Model for ^{131}I uptake in fish

The ^{131}I fish uptake model has been tested against measurements in fish in the Kiev Reservoir following Chernobyl (Kryshev and Ryazantsev 2000), as shown in Figure 6(e). The whole fish CF_{fish} (= 0.040 m^3 kg^{-1}) has been used (Table 10). Uptake and excretion rates are: k_f = 0.593 m^3 kg^{-1} y^{-1} and $k_b = k_f/CF_{fish}$ = 14.8 y^{-1}. Activity concentration of ^{131}I in water is calculated using data from Kryshev and Ryazantsev (2000). Note that the model predicts whole-body average concentrations: ^{131}I is expected to be relatively strongly accumulated in the fish thyroid. The model

predictions for ^{131}I are generally very good, though the first measurement made is significantly under-predicted. It appears that, in this case, ^{131}I is much more rapidly absorbed by fish than was expected.

Cooling Pond scenario

Finally, the AQUASCOPE models have been tested against the Cooling Pond scenario, developed by Kryshev and coworkers (Kryshev et al. 1996). Although measurements of radiocaesium and radiostrontium in the Chernobyl Cooling Pond have been published in the open literature (Kryshev et al. 1996; Kryshev 1995; Vakulovsky et al. 1994), this is a valuable test scenario for the model. It should also be noted that, whilst data from the Cooling Pond was not used for model calibration, prior to the test it was known that the concentration factor of ^{137}Cs in fish followed the inverse relationship with potassium used in the model (Smith et al. 2000b).

The Cooling Pond is classed as a closed lake. Although it has inflows from the NPP discharge canal, there is no surface outflow, with losses through groundwater flows into the Pripyat River being 1.2×10^5 m^3 y^{-1} (Kryshev et al. 1996). This is a small fraction of the total water volume (1.5×10^8 m^3) implying a long water residence time. The model default water residence time of 10 years has therefore been assumed. Model parameter values for the closed lake model are given in Table (7) and values of user input parameters are given in Table (11).

From data in Kryshev (1996) the average fallout to the Cooling Pond surface was estimated to be 7.7×10^6 Bq m^{-2} of ^{137}Cs and 2.5×10^6 Bq m^{-2} of ^{90}Sr. The majority of the release of ^{90}Sr, $^{141,144}Ce$, Pu isotopes and ^{241}Am was in the form of fuel particles (Kashparov et al., 1999), and within 30 km of the plant most (approximately 50-75%) of the ^{137}Cs was in fuel particles (Krouglov et al. 1998; Kashparov et al. 1999). It is assumed that fuel particle associated radionuclides were rapidly transferred to sediments so (in the early period) all radionuclides in the sediment were in the form of fuel particles. Therefore, from measurements of the relative amounts in bed sediments and water (Kryshev et al. 1996), 65% of ^{137}Cs and 89% of ^{90}Sr was estimated to be in the form of fuel particles. For the model, therefore fallout of chemically and biologically available radionuclides were estimated to be 2.7×10^6 Bq m^{-2} ($=7.7 \times 10^6 \times 0.35$) of ^{137}Cs and 2.75×10^5 Bq m^{-2} ($=2.5 \times 10^6 \times 0.11$) of ^{90}Sr.

Model predictions for the Cooling Pond scenario (Figure 7) are in good agreement with the empirical data. All of the measured values are within the range of errors in model predictions and for four of the five target variables, time trends in the data are predicted accurately. The ^{90}Sr activity concentration in fish, however, appears to increase over time where the model predicts a decrease. Though there are insufficient data to confirm the time trend, it is possible that, as with the Uruskul scenario (Figure 6d), the model may be under-predicting long term ^{90}Sr accumulation in bony tissues.

Model uncertainty

Estimating uncertainty of predictive models is in itself an uncertain and error-prone process. Often information of the distribution of uncertainties in parameter values is not available. In addition, error propagation is difficult to quantify accurately. The best method of assessing uncertainty is by comparison of model predictions with

empirical data. This is the approach which was taken here, and the error ranges shown in Figures 6 and 7 are estimated on this basis.

CONCLUSIONS

It has been shown that simplified models can give useful predictions of radioactivity in surface waters and fish following a number of different fallout events and scenarios. Note that the model tests carried out were "blind" predictions and the models were not calibrated to fit the data. Although environmental systems are often very complex, their behaviour is not necessarily best predicted by complex models (Smith, 2000d). The relative success of this simplified approach for aquatic systems suggests that such an approach may also be useful for predicting the behaviour of radionuclides in terrestrial systems.

The AQUASCOPE models (implemented in a series of EXCEL® spreadsheets) and model documentation are available free of charge from the corresponding author.

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Zibold G, Kaminski S, Klemt E, Smith JT. Time-dependency of the 137Cs activity concentration in freshwater lakes, measurement and prediction. Radioprotection – Colloques 37:75-80; 2002.

Table 1 Comparison of mean ^{137}Cs activity concentration in inflow streams with those in the lake water of Brotherswater (in 1992), Devoke Water (in 1988) and Loweswater (in 1988).

Lake	Mean inlet activity Bq m^{-3}		Mean lake activity Bq m^{-3}	
	Aqueous phase	Solid phase	Aqueous phase	Solid phase
Brotherswater	0.86	0.36	1.3	0.14
Devoke Water	70.3	-	81.0	-
Loweswater	12.0	-	12.2	-

Table 2 Estimates of the initial rate of decline of radiocaesium in rivers after Chernobyl.

River	k_1 (y^{-1})	
Pripyat	23	Voitsekhovitch et al. 1991
Dnieper	28	Voitsekhovitch et al. 1991
Po	7.3	Monte (1995)
Rhine	20.5	Monte (1995)
Mean	**13.2**	

Table 3 Parameters for runoff model (equation 1).

User input parameters						
D_c Bq m^{-2}	Deposition to catchment required for all models.					
Cs model only	f_{org} – fraction of catchment covered by organic soils ($f_{min} = 1 - f_{org}$)					
Sr model only	Classify catchment as "organic" or "mineral"					
Fixed model parameters						
	α (m^{-1})	β (m^{-1})	γ (m^{-1})	k_1 (y^{-1})	k_2 (y^{-1})	k_3 (y^{-1})
Cs model	0.3	eq (16)	eq (17)	13.2	0.41	0.02
Sr (organic)	0.8	0.03	0.005	16	0.09	0
Sr (mineral)	0.8	0.005	0.003	16	0.09	0
I-131 model	1.0	0	0	8.5	0	0

Table 4. Models for predicting K (y^{-1}) for four different levels of input data (adapted from Smith et al. 1999a): (1) T_w known; (2) T_w, d known; (3) T_w, d, s, v_p known; (4) T_w, d, s, v_p, $[K^+]$ known.

Model	ψ_i	θ_i
(1) $K = 1/T_w + \psi_1$	2.0 y^{-1}	
(2) $K = 1/T_w + \psi_2/d + \theta_2$	8.0 m y^{-1}	1.0 y^{-1}
(3) $K = 1/T_w + f_p v_p/d$, $K_d = \psi_3$.	36 m^3 kg^{-1}	-
(4) $K = 1/T_w + f_p v_p/d$, $K_d = \psi_4/[K^+]$	1200 μM m^3 l^{-1} kg^{-1}	-

Table 5 Required parameters for open lake model (Equation 3). Other parameters are as defined in the model for runoff (Table 3)

User input parameters	
D_c Bq m^{-2}	Deposition to catchment
D_L Bq m^{-2}	Deposition to lake: usually ≈ D_c
d (m)	Lake mean depth
T_w (y)	Lake water residence time – can be estimated from Eq. (5)
K	Removal rate of radionuclides from the lake water. Cs - see Table (4); Sr – see Eq. (18); ^{131}I – $K = 1/T_w$.

Table 6 Long term radiocaesium activity concentration in the water of 9 "closed" lakes.

Lake	^{137}Cs Bq m^{-3} in water during1997	^{137}Cs Deposition* kBq m^{-2}	^{137}Cs in water per unit deposition m^{-1}
Tyumenskoe #1	1300	291	4.46×10^{-3}
Stoyacheye #2	200	108	1.85×10^{-3}
Svyatoe #3	4200	1413	2.97×10^{-3}
Svyatskoe #5	4900	207	23.6×10^{-3}
Svyatoe #7	1000	226	4.43×10^{-3}
Smerdin #10	1100	75	14.7×10^{-3}
Petrovschina #12	500	312	1.60×10^{-3}
Kozhanovskoe	4300	1497	2.87×10^{-3}
Iso Valkjaarvi**	1100	70	15.7×10^{-3}
Mean			**8.0×10^{-3}**

*Deposition to lake surface, as measured in 1997 (i.e. at t = 11 years). **Measurements extrapolated to 1997 from measured value of 1850 Bq m^{-3} in 1990 using the observed rate of long term decline of ^{137}Cs in this lake (from VAMP data set: IAEA, 2000).

Table 7 Parameters for closed lake model: Equations (8) for Cs, (20) for Sr, (21) for I-131.

User input parameters	
D_L, d, K	As for open lakes (Table 5)
T_w (y)	As for open lakes (Table 5) or use default value of 10 years.
Fixed model parameters	
Cs model	$\eta_1 = 0.04$ m^{-1}; $\eta_2 = 0.0085$ m^{-1}
Sr model	$\eta_1 = 0.05$ m^{-1}; $g_1 = 0.025$ y^{-1}
^{131}I model	None needed.

Table 8 Radiostrontium in closed lakes and slow flowing water bodies in the period 1989-94, from data in Vakulovsky (1994) and (*) Sansone & Voitsekhovitch (1996). The ^{90}Sr deposition to the lake surface is estimated from the sediment inventory for all lakes except Svyatoe, Russia for which an estimate of ^{90}Sr deposition to the catchment is used.

Water body	^{90}Sr in water C_L Bq m^{-3}	^{90}Sr deposited D_L Bq m^{-2}	Ratio C_L/D_L m^{-1}
Murovka	2.60×10^{3}	1.90×10^{5}	1.37×10^{-2}
Krasnyansk 1	2.96×10^{4}	8.14×10^{6}	3.64×10^{-3}
Krasnyansk 2	6.48×10^{4}	5.92×10^{6}	1.09×10^{-2}
Krasnyansk 3	7.40×10^{4}	6.29×10^{6}	1.18×10^{-2}
Glubokoye	2.96×10^{5}	2.44×10^{7}	1.21×10^{-2}
Vershina	2.59×10^{5}	1.85×10^{7}	1.40×10^{-2}
Pojmennoe 1	2.78×10^{5}	4.80×10^{6}	5.79×10^{-2}
Pojmennoe 2	3.33×10^{5}	2.00×10^{7}	1.67×10^{-2}
Pojmennoe 3	3.33×10^{5}	8.14×10^{6}	4.09×10^{-2}
Beloe	9.25×10^{4}	4.44×10^{6}	2.08×10^{-2}
Ashbuchia	1.85×10^{5}	9.62×10^{6}	1.92×10^{-2}
Pripyat Creek	1.30×10^{5}	8.14×10^{6}	1.59×10^{-2}
Semikhoda Creek	3.33×10^{4}	7.40×10^{6}	4.50×10^{-3}
Svyatoe, Russia*	6.00×10^{2}	3.96×10^{4}	1.52×10^{-2}
Mean			**1.84×10^{-2}**

Table 9 Model parameters for predatory and non-predatory fish. For non-predatory fish n = 0 since it is assumed that there is no influence of weight on bioaccumulation.

Type	***Y* (mmol kg^{-1})**	**k_b (y^{-1})**	***n* [ ]**
Predatory	462	0.511	0.34
Non-predatory	61.3	8.4	0

Table 10 Summary of literature values and recommended value for ^{131}I in whole fish. Note that the different review estimates may include some of the same data.

Recommended *CF* used in model m^3 kg^{-1}	Ranges and other estimates of *CF* m^3 kg^{-1}	Notes and references
0.04	0.04 (0.02 – 0.6)	IAEA (1994). Estimate and range.
	0.04 (0.01 – 0.132)	Blaylock (1982). Estimate and range for fish muscle
	0.01	Kryshev (1995) measured in Kiev reservoir.
	0.03	Coughtrey & Thorne vol 3 (1983)

Table 11 Description of model "blind" test scenarios. Parameter values input by the user are given for each scenario where required.

	River Sozh	*Haweswater Reservoir*	*Lake Uruskul*	*Kiev Reservoir*	*Cooling Pond*	*Cooling Pond*
Model tested	Runoff model	Open lake model	Closed lake	Fish uptake	Closed lake	Closed lake
Radionuclide	^{137}Cs	^{90}Sr	^{90}Sr	^{131}I	^{137}Cs	^{90}Sr
Target variables	Water	Water	Water, fish	Fish	Water, Fish	Water, Fish
Fallout event	Chernobyl	Weapons testing	Kyshtym	Chernobyl	Chernobyl	Chernobyl
Deposition, Bq m^{-2}	D_c= 220,000	D_c=D_L= 3780	D_L= 2.0×10^7	++	D_L= 2.7×10^{6} **	D_L= 2.8×10^{5} +
Organic soils in catchment	f_{org} = 11%	Classed as "organic"	N/A	N/A	N/A	N/A
Lake mean depth, d (m)	N/A	17.4	2	++	6.6	6.6
Water res. time, T_w (y)	N/A	0.85	10*	++	10*	10*
[K^+], µmol l^{-1}	N/A	N/A	N/A	N/A	100	N/A
[Ca^{2+}] µmol l^{-1}	N/A	N/A	244	N/A	N/A	1180
Fish weight, w	N/A	N/A	N/A	1000g*	1000g*	N/A

*Default value; ** Assuming 65% in fuel particles; + Assuming 89% in fuel particles; ++ Best fit to measured water concentrations used as input; N/A – not applicable.

List of figures

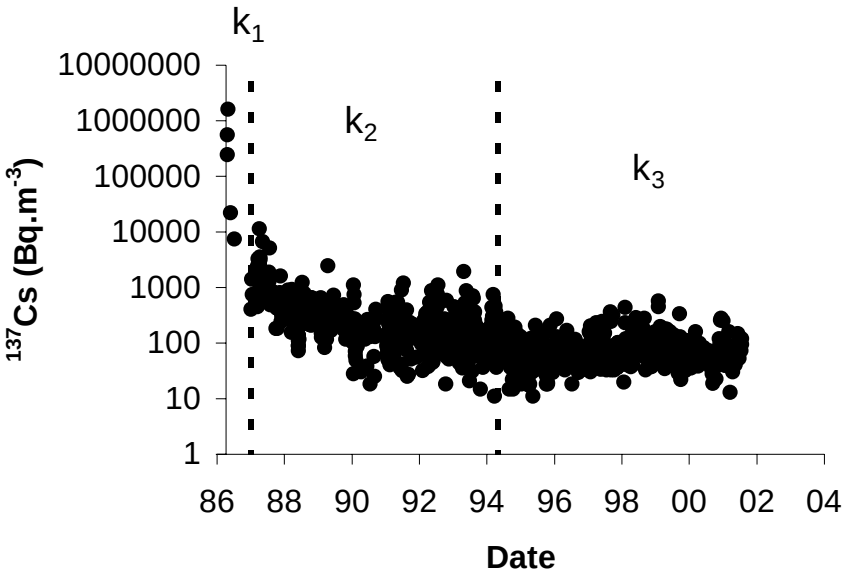

Figure 1

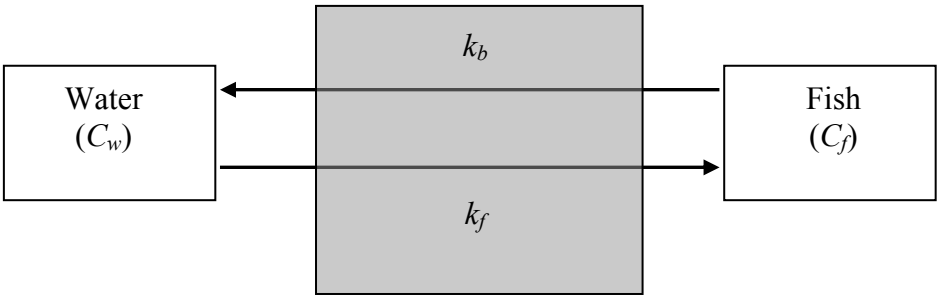

Figure 2

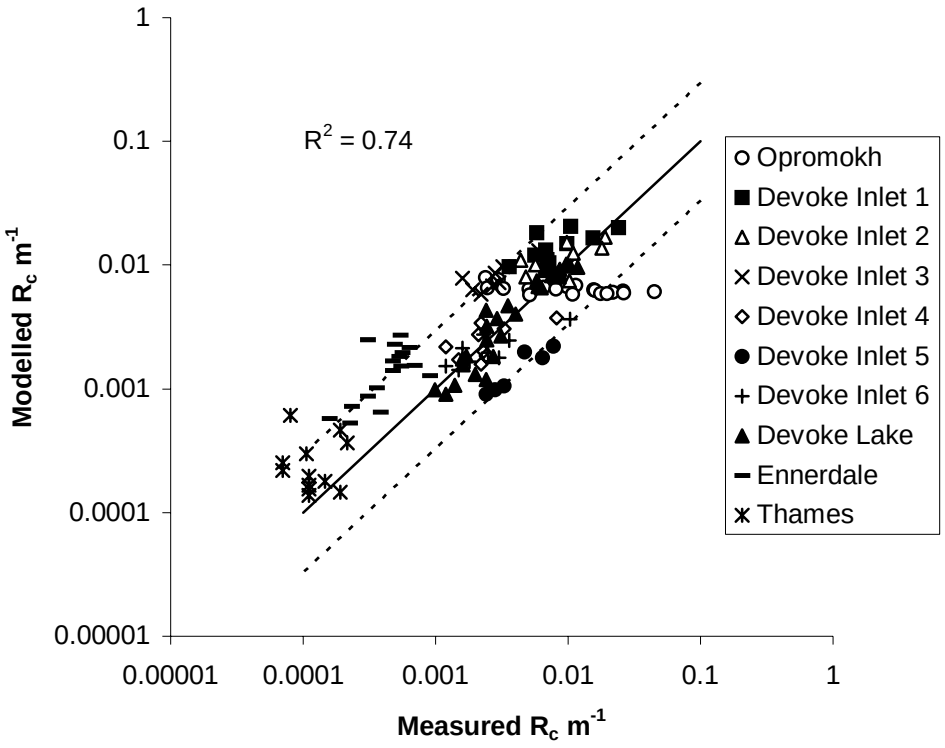
1
0.1
0.01
0.001
0.0001
0.00001
R2 = 0.74
Modelled Rc m-1
0.00001
0.0001
0.001
0.01
0.1
1
Measured Rc m-1
Opromokh
Devoke Inlet 1
Devoke Inlet 2
Devoke Inlet 3
Devoke Inlet 4
Devoke Inlet 5
Devoke Inlet 6
Devoke Lake
Ennerdale
Thames

Figure 3

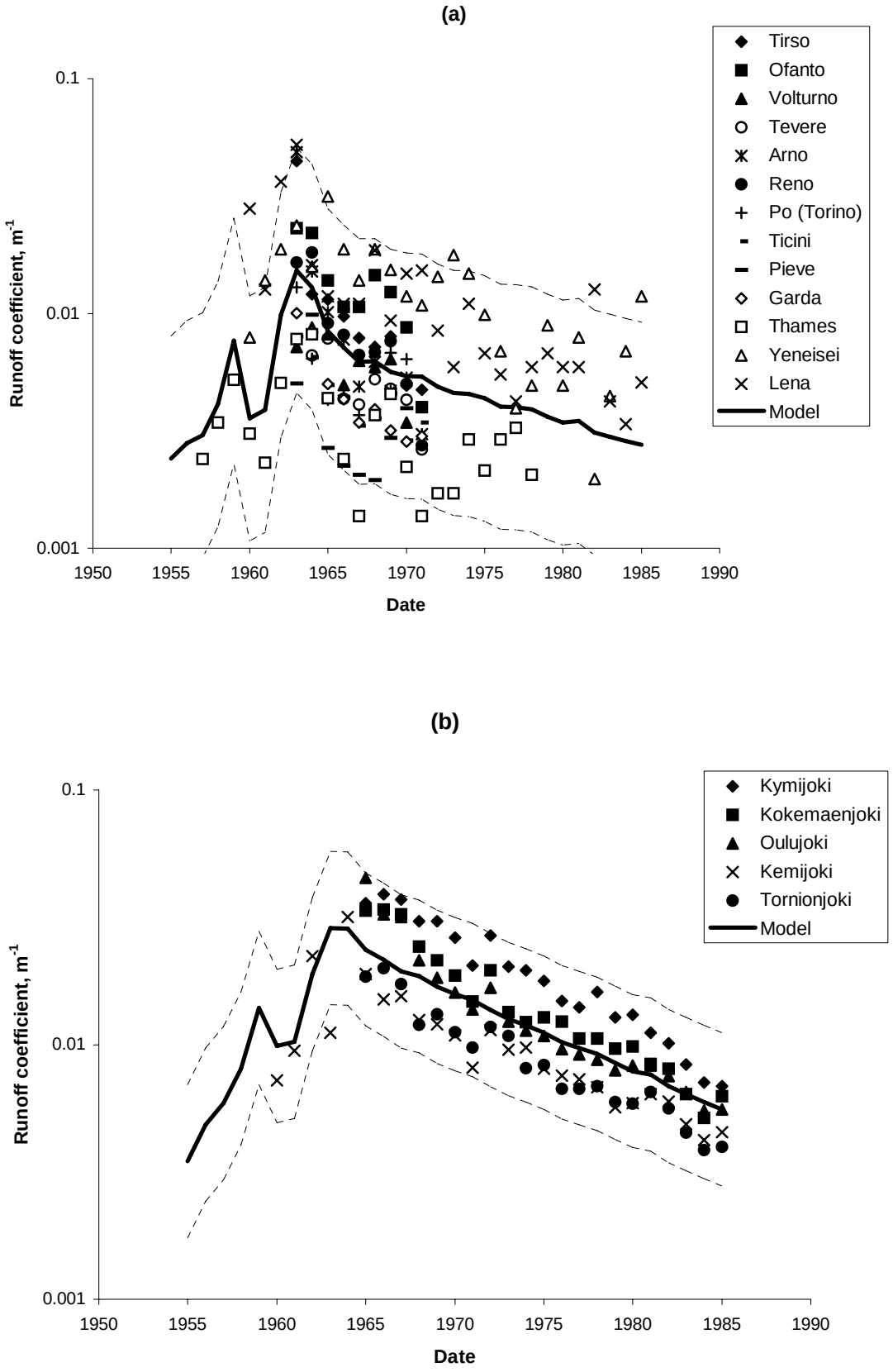
(a)
Tirso
Ofanto
Volturno
Tevere
Arno
Reno
Po (Torino)
Ticini
Pieve
Garda
Thames
Yeneisei
Lena
Model
0.1
0.01
0.001
Runoff coefficient, m-1
1950
1955
1960
1965
1970
1975
1980
1985
1990
Date
(b)
Kymijoki
Kokemaenjoki
Oulujoki
Kemijoki
Tornionjoki
Model
0.1
0.01
0.001
Runoff coefficient, m-1
1950
1955
1960
1965
1970
1975
1980
1985
1990
Date

Figure 4

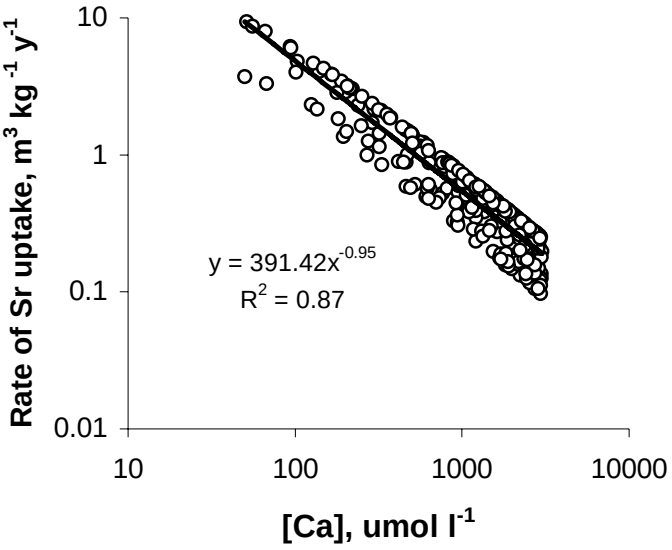
Rate of Sr uptake, m3 kg-1 y-1
10
1
0.1
0.01
y = 391.42x-0.95
R2 = 0.87
10
100
1000
10000
[Ca], umol l-1

Figure 5

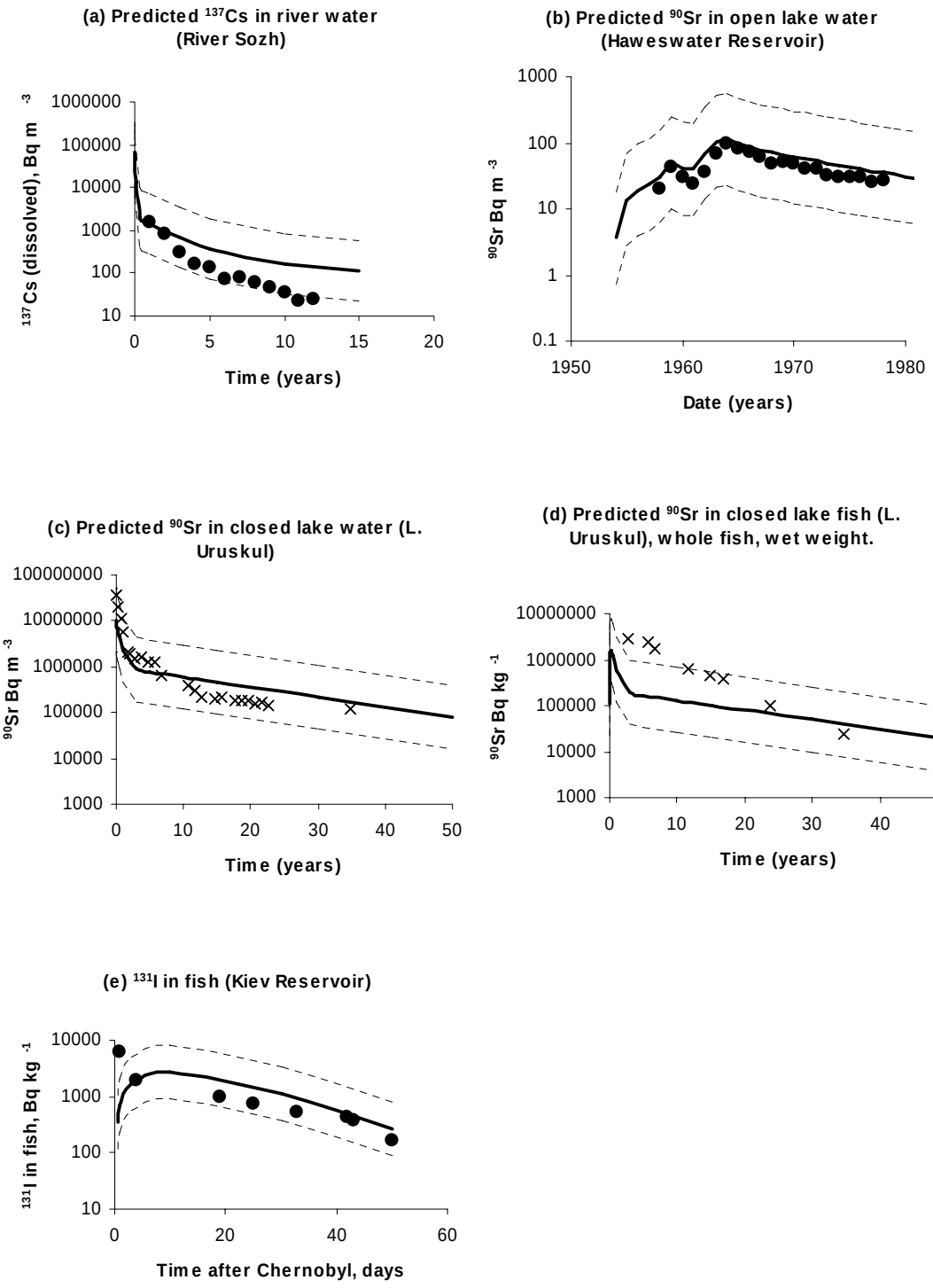
(a) Predicted 137Cs in river water (River Sozh)
137Cs (dissolved), Bq m -3
1000000
100000
10000
1000
100
10
0
5
10
15
20
Time (years)
(b) Predicted 90Sr in open lake water (Haweswater Reservoir)
90Sr Bq m -3
1000
100
10
1
0.1
1950
1960
1970
1980
Date (years)
(c) Predicted 90Sr in closed lake water (L. Uruskul)
90Sr Bq m -3
100000000
10000000
1000000
100000
10000
1000
0
10
20
30
40
50
Time (years)
(d) Predicted 90Sr in closed lake fish (L. Uruskul), whole fish, wet weight.
90Sr Bq kg -1
10000000
1000000
100000
10000
1000
0
10
20
30
40
50
Time (years)
(e) 131I in fish (Kiev Reservoir)
131I in fish, Bq kg -1
10000
1000
100
10
0
20
40
60
Time after Chernobyl, days

Figure 6

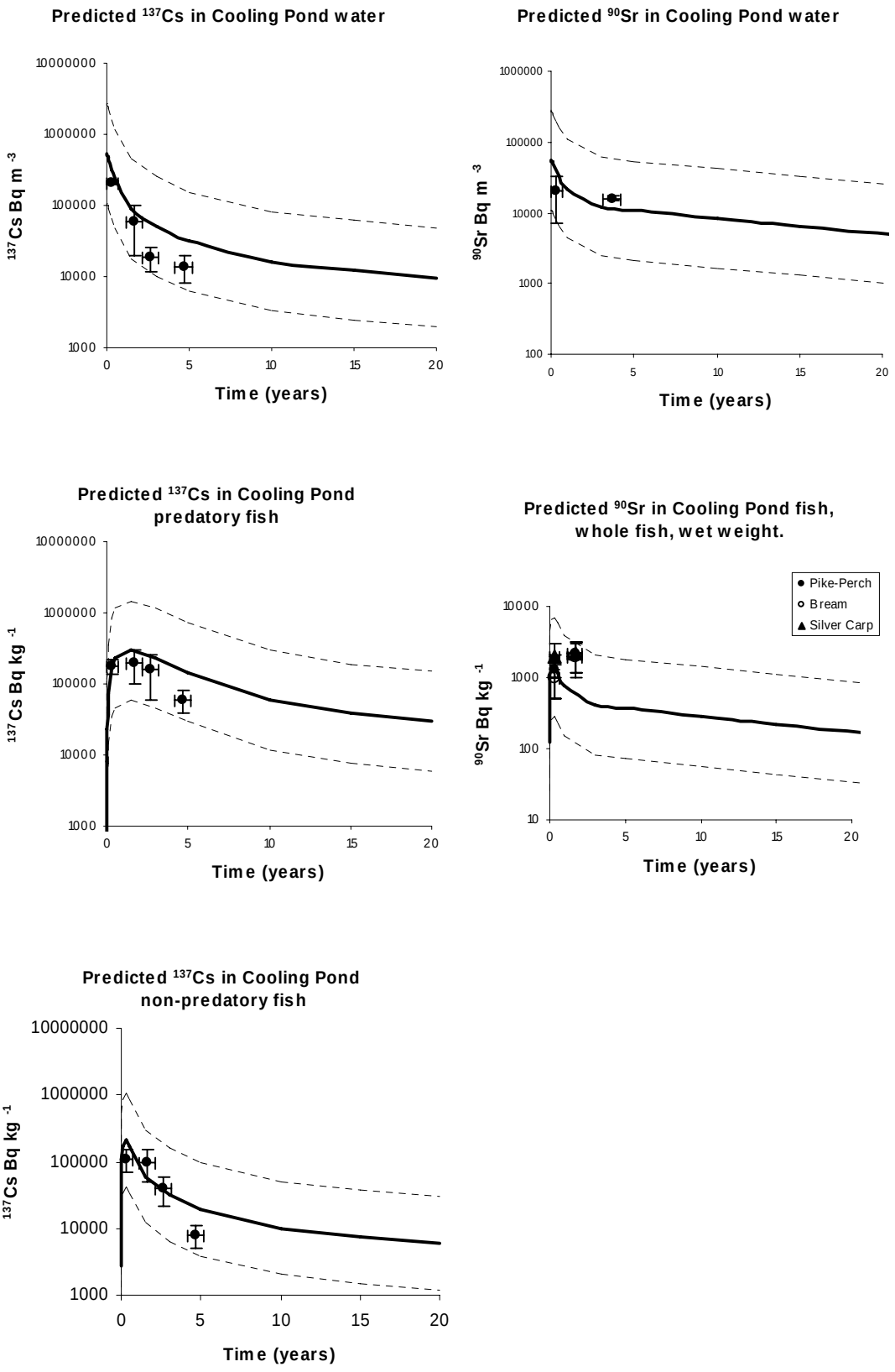
Predicted 137Cs in Cooling Pond water
137Cs Bq m -3
10000000
1000000
100000
10000
1000
0
5
10
15
20
Time (years)
Predicted 90Sr in Cooling Pond water
90Sr Bq m -3
1000000
100000
10000
1000
100
0
5
10
15
20
Time (years)
Predicted 137Cs in Cooling Pond predatory fish
137Cs Bq kg -1
10000000
1000000
100000
10000
1000
0
5
10
15
20
Time (years)
Predicted 90Sr in Cooling Pond fish, whole fish, wet weight.
Pike-Perch
Bream
Silver Carp
90Sr Bq kg -1
10000
1000
100
10
0
5
10
15
20
Time (years)
Predicted 137Cs in Cooling Pond non-predatory fish
137Cs Bq kg -1
10000000
1000000
100000
10000
1000
0
5
10
15
20
Time (years)

Figure 7