Modelling the dispersion of radionuclides following short duration releases to rivers: Part 2. Uptake by fish.

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ABSTRACT

This paper evaluates and generalises state-of-the-art approaches for dynamic modelling of bioaccumulation in fish resulting from short duration liquid discharges of radionuclides (^{3}H, ^{14}C, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{65}Zn, ^{89}Sr, ^{90}Sr, ^{125}I, ^{131}I, ^{241}Am, isotopes of Pu and U) to rivers. Based on a review of model parameter values, predictions are made of maximum and time-integrated activity concentrations in fish. A simplified version of the model was developed and presented as "look-up" graphs. The influence of various environmental parameters on model output was evaluated by sensitivity analysis. Maximum and time-integrated concentrations in fish may be predicted for rivers on the basis of the river volumetric flow rate and water temperature. It is demonstrated that the dynamic model gives lower and more realistic predictions of maximum concentrations in fish than the simpler "Concentration Factor" approach. However, for time-integrated concentration in fish, and estimation of radiation dose to humans from consumption of the fish, the Concentration Factor approach gives similar predictions to the dynamic model.

Key words: radioactivity, river, fish, bioaccumulation, uptake.

INTRODUCTION

In Part I of this study (Smith et al., in press) we presented a generalised model for the assessment of concentrations of radionuclides (^{3}H, ^{14}C, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{65}Zn, ^{89}Sr, ^{90}Sr, ^{125}I, ^{131}I, ^{241}Am, isotopes of Pu and U) in water and sediment following hypothetical short-duration releases to rivers. In Part II, linked models are presented for predicting activity concentrations of these radionuclides in fish, particularly focusing on predictions for chemical conditions prevailing in the River Thames, though the model can relatively easily be generalised to other rivers. The purpose of this paper is not to develop novel models (since many such models are available in the literature), but to evaluate, parameterise and simplify existing models for practical application to risk assessment for short term radionuclide releases to rivers.

The level of radioactive contamination of aquatic biota is commonly defined in terms of a concentration factor (*CF*) (wet weight, w.w., of fish) where

$$CF = \frac{Activity\ concentration\ per\ kg\ of\ \ fish\ (w.w.)}{Activity\ concentration\ per\ litre\ of\ \ water} \quad \mathrm{l\,kg^{-1}} \qquad (1)$$

Many studies on the accumulation of radionuclides in fish have focused on the prediction of *CF* (sometimes termed the bioaccumulation factor, *BAF*, or aggregated concentration factor, *ACF*). There are models to predict the water-fish *CF* for radiocaesium and radiostrontium using an inverse relation between the *CF* and, respectively, the potassium and calcium concentration of the surrounding water (e.g. Blaylock, 1982; Rowan and Rasmussen, 1994; Smith et al., 2000). For most radionuclides, however, single "best estimate" *CF* values are commonly assumed to apply to all freshwater bodies (IAEA, 1994).

Estimates of the fish-water *CF* may be made from measurements (in the lab or field) reported in the literature. Alternatively, in appropriate cases, *CF* may be estimated from the concentration factor of the stable isotope:

$$CF \approx \frac{mass\ of\ \ stable\ isotope\ \ per\ kg\ of\ \ fish\ (w.w)}{mass\ of\ \ (available)\ stable\ isotope\ \ per\ l\ of\ \ water} \quad \mathrm{l\,kg^{-1}} \qquad (2)$$

The equilibrium *CF* modelling approach is appropriate for cases in which the radionuclide activity concentration in fish can be assumed to be in equilibrium with that in water, for example at long times (years) after radionuclide fallout, or for continuous releases of radionuclides to a river. At short times after radioactive contamination of an aquatic system, or where activity concentrations in water are changing relatively rapidly, a dynamic modelling approach may be more appropriate. Dynamic models for radiocaesium accumulation in freshwater fish have previously been developed using the results of controlled laboratory experiments (for example, Garnier-Laplace et al., 1997) and in field studies (Sazykina, 2000; Smith et al., 2002; Kryshev, 2003).

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 assumed to be the primary uptake pathway. For strontium isotopes, a model for direct uptake via the water (Chowdhury and Blust, 2001) has been developed to estimate the intake rate. Other studies, however, have assumed that the majority of ^{90}Sr uptake is via the food pathway (Kryshev, 2003).

MODELLING AND METHODS

Summary of main symbols with units as used

C_f	Concentration of the RN in fish	Bq kg^{-1}
CF	Fish-water concentration factor	l kg^{-1}
CF_{food}	Food-water concentration factor	l kg^{-1}
C_{food}	Concentration of the RN in prey fish	Bq kg^{-1}
C_w	Activity concentration of the RN in water	Bq l^{-1}
k_f	Rate constant of the RN uptake in fish	l kg^{-1} d^{-1}
k_b	Rate constant of RN excretion from fish	d^{-1}
D_{max}	Maximum daily intake of food by fish	g d^{-1}
α	Radionuclide assimilation efficiency	[ ]
w	Wet weight of fish	g

The 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 l^{-1}) and release from the fish (Figure 1):

$$\frac{dC_f}{dt} = k_f C_w - (k_b + \lambda)C_f \tag{3}$$

where k_f (l kg^{-1} d^{-1}) is the rate constant describing transfers of ^{137}Cs to fish through its food, k_b (d^{-1}) is the backward rate constant describing excretion of radioactivity from the fish and λ is the decay rate of the radionuclide (d^{-1}, note the change in units of λ from Part 1 of this study where λ is in s^{-1}). The ratio of these rate constants gives the equilibrium concentration factor, CF (l kg^{-1}), of the radionuclide in fish relative to water:

$$\frac{k_f}{k_b + \lambda} = \frac{C_f}{C_w}(at\ equilibrium) = CF \tag{4}$$

For a constant activity concentration in the water phase, C_w, Equation (3) has solution:

$$C_f = \frac{k_f C_w}{k_b + \lambda}(1 - \exp(-(k_b + \lambda)t)) \tag{5}$$

Estimating uptake and excretion rates

In cases where uptake of radionuclides is principally via ingestion, we can estimate the uptake rate by:

$$k_f = \frac{C_{food} \times D_{\max} \times \alpha \times 10^{-3}}{C_w \times w \times 10^{-3}} = \frac{CF_{food} \times D_{\max} \times \alpha \times 10^{-3}}{w \times 10^{-3}} \quad (6)$$

where D_{max} (g d^{-1}) is the maximum daily intake (wet weight) of food by fish, w is the wet weight of fish in grammes and α 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. 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). Note that this model has previously been applied to uptake of ^{131}I in fish by Smith et al. (2005).

In this study, model predictions will be made for piscivorous fish. It is assumed that uptake of radionuclides to the prey of piscivorous fish is effectively 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 in most cases assumed to be equal to the concentration factor of the predatory fish unless it is known to be significantly different (for example, for ^{137}Cs, it is known that the CF of predatory fish is approximately two to four times higher than their herbivorous prey, Rowan and Rasmussen 1994; Rowan et al., 1998).

It was (conservatively) assumed that fish feed at their maximum daily rate. Elliot (1975a) developed an empirical model which estimates feeding rate for fish (brown trout) of different wet weight w (grammes) at different water temperatures, T :

$$D_{\max} = (4 \times 10^{-3}) A_D \times w^{b_1} \times \exp(b_3 T) \quad (7)$$

where A_D, b_1 and b_3 are empirically determined constants whose values are given in Table 1. The factor 4 converts the dry weight feeding rates estimated by the Elliot (1975a) model to feeding rate expressed in terms of wet weight as used in Equation 6. The factor 10^{-3} converts D_{max} in mg per day estimated by the Elliot (1975a) model to g per day used in Equation (6).

For strontium isotopes, which may be absorbed through the gills (Chowdhury and Blust, 2001) or via the food pathway (Kryshev, 2003), we compare uptake rates for models based on both pathways. A previous study (Smith et al., 2005) has simplified the Chowdhury and Blust (2001) model for gill uptake of radiostrontium. It was found by these workers that the calcium concentration [Ca^{2+}] was the most important factor determining the strontium uptake rate. The strong negative correlation between $k_f(Sr)$ and [Ca^{2+}] meant that $k_f(Sr)$ could be estimated using the following simplified form of the model presented in Chowdhury and Blust (2001):

$$k_f = \frac{50.4}{[Ca]^{0.95}} \quad (8)$$

Note the change in the constant from Smith et al. (2005): this is due to the different units of k_f and [Ca] used in the present study.

The excretion rates of radionuclides from fish, k_b, were estimated from the uptake rate and the concentration factor (Equation 4):

$$k_b + \lambda = \frac{k_f}{CF} \tag{9}$$

Model implementation and simplified version

Equation (3) was solved numerically within the advection-dispersion equation (ADE) model code described in Part I. Estimates of pollutant maximum and time integrated concentrations were also determined using simplified relationships between the key model parameters. The simplified relationships (outlined below) were then tested against the output of the numerical model.

Simplified method of estimating fish concentrations

Where the excretion rate of radionuclides in fish has a significantly longer time scale than the travel time of the plume, i.e.:

$$\frac{3600 \times 24}{k_b} >> L/v \tag{10}$$

(where 3600×24 changes k_b to units of s^{-1}, L is the reach length, 10,000 m in this case, and v is the flow velocity), the maximum activity concentration in fish may be estimated by:

$$C_f^{\max}(x) \approx \Sigma C_w(x).k_f \tag{11}$$

(i.e. it is assumed that there is only accumulation and no excretion). The excretion rate condition (Equation 10), for the scenarios considered here, holds for all radionuclides except tritium, for which a concentration factor approach may be more appropriate (see below).

Given the above condition (Eq. 10), the time integrated activity concentration in fish, $\Sigma C_f(x)$, may be estimated by:

$$\Sigma C_f(x) \approx \frac{\Sigma C_w(x).k_f}{k_b + \lambda}\left(1 - \exp(-(k_b + \lambda)\tau)\right) \tag{12}$$

where $\Sigma C_w(x)$ is the time integrated water concentration (see Part I) and τ is the time (days) over which the concentration is to be integrated.

Testing the simplified model

The simplified model output was compared with that of the numerical model for 55 model runs at different sites under different environmental conditions and for different radionuclides. For all radionuclides other than ^{3}H, comparison showed good agreement between the simplified model and the numerical model ($r^2 > 0.99$). Maximum (but not time-integrated) concentrations of ^{3}H in fish, however, are over-estimated by the simplified model since the concentration factor approach was used for this radionuclide (see below).

Parameter values for the fish uptake model

Table 2 shows values of the *CF* obtained from a review of the literature. These are generally conservative estimates, particularly for those radionuclides about which there is relatively little information available. Uptake and excretion rates were determined for each radionuclide for a 500 g piscivorous fish (using Equations 6 and 9), as described below and shown in Table 3. Table 3 gives parameter values for a water temperature of 12°C – variation in feeding rates (and hence radionuclide uptake rates) with temperature is discussed below.

Caesium-134, 137

Rates of uptake of radiocaesium in fish have been extensively studied both in the laboratory and in the field following the Chernobyl accident. The principal uptake route is via food, there being relatively low rates of transfer via the gills (e.g. Coughtrey and Thorne 1983). For piscivorous fish eating herbivorous food (e.g. small roach) a value of CF_{food} is taken as $CF/2$ since typically the concentration factor of herbivorous fish is 25-50% that of piscivorous fish, depending on the size of fish (Rowan and Rasmussen, 1994; Smith et al., 2000). The assimilation efficiency, α, is taken as the average value for atlantic salmon and brook trout of $\alpha = 0.44$ (Tucker and Rasmussen 1999).

Strontium-89, 90

There is less quantitative information available for uptake and retention rates of ^{90}Sr in fish than ^{137}Cs. Uptake via the gills has been demonstrated (Chowdhury and Blust, 2001), however Kryshev (2003) assumes that "the major part of ^{90}Sr in freshwater ecosystems enters the fish with contaminated food". Uptake rates will here be calculated using both models and their values will be compared.

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* for ^{90}Sr, and [Ca] (mg l^{-1}) in the surrounding water:

$$CF(muscle) = \exp(5.2 - 1.2\ln[Ca]) \tag{13}$$

$$CF(bone) = \exp(9.7 - 1.2\ln[Ca]) \tag{14}$$

Assuming that 20% of the wet weight of a fish is composed of bony parts (I.I. Ryabov, Severtsov Institute, Moscow, pers. commun.) this gives a whole fish *CF*:

$$CF(whole\ \ fish) = \exp(8.13 - 1.2\ln[Ca]) \quad (15)$$

The *CF* value used here (60 l kg^{-1}) reflects Sr activity concentrations in whole fish since *CF* values for edible portions (muscle) are predicted to be significantly lower than this value for the high calcium content River Thames (Table 2).

The uptake rate for radiostrontium is estimated using a model for transfers by direct uptake across the gills (Chowdhury and Blust, 2001) for [Ca = 121 mg l^{-1}]; [Sr (stable) = 0.358 mg l^{-1}] and pH (= 8.1) in the Thames (Neal and Robson (2000) gives k_f = 0.675 l $kg\ d^{-1}$. The simplified model (Equation 8: Smith et al., 2005), using [Ca] concentration only, gives a similar value of k_f = 0.53 l $kg\ d^{-1}$.

The uptake and excretion rates for radiostrontium were also calculated assuming uptake via the food pathway (Eqs. 6 and 9). The rates for both pathways are presented in Table 3 and show that there is no major difference in the rates of uptake and excretion predicted by the two methods. Since in the accumulation model (Eq. 11), maximum radionuclide concentration in fish is directly proportional to uptake rate, we will conservatively assume uptake via the food pathway which gives greater predicted uptake rates (Table 3).

Iodine-125, 131

A previous study (Smith et al., 2005) estimated bioaccumulation of ^{131}I in fish assuming (conservatively) that the assimilation efficiency is α = 1.0 (i.e. all ^{131}I ingested is assimilated) and that $CF_{food} = CF_{fish}$.

Carbon-14

The *CF* of ^{14}C in fish has previously (I.L. Ophel, quoted in Blaylock 1982) been estimated from the stable carbon content of fish (typically 10% of body wet weight, Vinogradov 1953) and the carbon (HCO_3 + CO_3) content of water. This (Blaylock, 1982) gave estimates of $CF = 5 \times 10^3$ l kg^{-1} for waters of high mineral content and 5×10^4 l kg^{-1} for waters of low mineral content. This compares with an estimate of 22×10^3 l kg^{-1} used in this study for the Thames from stable carbon in fish (10% of wet weight, Vinogradov, 1953) and DOC in the Thames of 4.4 mg l^{-1} (Neal and Robson 2000). This latter is likely to be an over-estimate of the *CF* (under-estimate of available carbon) since it does not include organic particulate carbon and inorganic carbon in the water. The assimilation efficiency, α, of stable carbon in food may be estimated from data presented in Elliott (1975b) on growth rates of brown trout. For a 500g trout at temperature 12 ^oC, equations given in Elliott (1975b) estimate a growth rate of 1.65 g dy^{-1} (wet weight) at a maximum feeding rate of 11.8 g dy^{-1} (wet weight). Assuming that the carbon content of food is approximately equal to that of the fish, this gives an assimilation efficiency, $\alpha \approx 14\%$.

Phosphorus-32

The phosphorus content of fish is approximately 0.3% of wet weight (Vinogradov, 1953) and soluble reactive phosphorus (SRP) is 0.91 mg l^{-1} for the Thames (Neal and

Robson, 2000), giving a *CF* of 3300 l kg^{-1}. A value of 10^4 l kg^{-1} was conservatively chosen for use in the model. Uptake and excretion rates are estimated by assuming that $CF_{food} = CF_{fish}$ and assuming, conservatively, an assimilation efficiency of α = 1.0 (i.e. all radioactivity ingested is assimilated).

Cobalt-60

There is less quantitative information available for uptake and retention rates of ^{60}Co in fish than ^{137}Cs. Laboratory studies, by Baudin et al. (2000) observed assimilation of ^{60}Co from food at a rate approximately 6 times lower than ^{137}Cs. We will make the (slightly conservative) assumption that the assimilation efficiency is α = 0.1, approximately four times lower than for ^{137}Cs, and that $CF_{food} = CF_{fish}$.

Tritium

We here consider uptake of tritium as tritiated water, though it is noted (as discussed below) that organically bound tritium may have higher accumulation in the aquatic food chain (Williams et al., 2001). The biological half life of tritium absorbed as tritiated water is < 1d so the concentration of ^{3}H in fish will closely follow the concentration of ^{3}H in ambient water (Vanderploeg et al., 1975 quoted in Blaylock, 1982). Assuming a biological half life of 1 d gives k_b = 0.69 d^{-1} and, using Equation 9, a value of k_f = 0.69 d^{-1}. We have thus far found no empirical data against which to test ^{3}H uptake models. Since the turnover of tritium is so rapid in fish, we assume equilibrium between activity concentrations in water and those in fish. For the simplified model, we have therefore used the *CF* model (Eqs. 18 and 19, below) for predicting tritium concentrations in fish. This is expected to lead to a slight over-estimate of maximum concentrations in fish compared to the dynamic model.

Americium-241, Uranium-234, 235, 238, Plutonium-238, 239, 240

Concentration factors of these radionuclides are given in Table 2. Uptake and excretion rates are estimated by assuming that $CF_{food} = CF_{fish}$ and assuming an assimilation efficiency of α = 1.0 (i.e. all radioactivity ingested is assimilated). This is likely to be a significant over-estimate of the uptake rate, as these radionuclides are in general not strongly bioaccumulated. For example, a study of plutonium accumulation by various marine organisms found that "concentrations in benthic [bottom-dwelling] biota are generally 1-2 orders of magnitude lower than in surface sediments. Furthermore, a significant part of this plutonium is probably not metabolised but rather associated with particles in the guts and adhering to the surface structure of the animals" (Dahlgaard et al., 2001)

RESULTS

Testing the radionuclide uptake model

For four of the radionuclides considered, it was possible to test the model against empirical data obtained in the field or in the laboratory. Activity concentrations of radionuclides in water were simulated using a series of exponential equations and uptake to fish was modelled using a solution to Eq. (3) (see Smith et al., 2002). The

^{137}Cs uptake model was tested against measurements of ^{137}Cs in trout, pike, eel and perch in Windermere following Chernobyl (Camplin et al., 1989), as shown in Figure 2. The model of Rowan and Rasmussen (1994) was used (see Table 2) to predict the *CF* for Windermere for $[K^+]$ = 0.6 mg l^{-1} and s = 1.1 mg l^{-1}: changes in activity concentration in water are determined from Smith et al. (1997), and water temperature (required for estimating D_{max}) is estimated from Davison et al. (1993).

The ^{90}Sr uptake model (Chowdhury and Blust, 2001) was tested against measurements of ^{90}Sr in bream and pike-perch in the Kiev Reservoir following Chernobyl (Kryshev and Ryazantsev 2000), as shown in Figure 3. These data are for whole fish, so the predicted whole fish CF_{fish} (= 53 l kg^{-1}, for [Ca] = 32mg l^{-1} in the Kiev Reservoir using Eq. 15) has been used. Activity concentration of ^{90}Sr in water was simulated using data from the Ukrainian Hydrometeorological Insitute (Voitsekhovitch, 2001).

Smith et al. (2005) have previously tested the ^{131}I uptake model against measurements in fish in the Kiev Reservoir following Chernobyl (Kryshev and Ryazantsev 2000), as shown in Figure 4. In this study (Smith et al., 2005) the whole fish CF_{fish} (= 40 l kg^{-1}) was used (Table 2).

The ^{60}Co uptake model was tested against laboratory measurements by Baudin and Fritsch (1989), as shown in Figure 5. The whole fish CF_{fish} (= 300 l kg^{-1}) has been used (Table 2) in the calculation. Temperature of the experiments was 20 ^{o}C (Baudin and Fritsch 1989) for estimation of feeding rate. Activity concentration of ^{60}Co in water was approximately constant during the experiment having value 5×10^4 Bq l^{-1}. Average fish mass during the experiment was approximately 1.4 g.

Maximum concentrations in fish

Figure 6 gives the estimated maximum activity concentrations in whole fish (per kg wet weight, 0.5 kg fish) as a function of volumetric flow rate for all of the radionuclides except tritium (see below). The estimates apply to a short duration release of radionuclides to the river of any duration $\leq$ 24 h, for all distances downstream x up to 10000 m, and for a water temperature of 12^{o}C. For other water temperatures, the maximum concentrations should be multiplied by the correction factors given in Table 4. The graph may be applied to other scenarios by multiplying the output for a 1 MBq release by the actual release amount in MBq.

Figure 6 gives estimates for maximum radioactivity concentrations in fish where there is no sorption to suspended sediments (f_p = 0). For the three sites studied here, it was conservatively assumed, when calculating activity concentrations in fish, that f_p = 0 for all radionuclides (see Part 1 of this study). In cases where f_p > 0, these estimates should be multiplied by (1-f_p): $C_f(x) \rightarrow C_f(x) \times (1 - f_p)$.

Estimates of maximum concentration of tritium (H-3) in fish may be made by multiplying the maximum concentration of tritium in water by the fish-water concentration factor. Since, for tritium, *CF* = 1.0 l kg^{-1} for all water temperatures, the maximum activity concentration in fish (Bq kg^{-1}) is equal to the maximum water concentration (Bq $l^{-1} \approx$ Bq kg^{-1}).

Time integrated activity concentrations in fish

Figure 7 gives the estimated time integrated activity concentrations (for integration times of one week and one year) in whole fish (per kg wet weight for a 500 g fish) as a function of volumetric flow rate. The graphs apply to all the radionuclides except tritium (see below). The estimates apply for all release durations ≤ 24 h, for all distances x, up to 10 km, and for a water temperature of 12°C. The graphs may be applied to other scenarios by multiplying the output for a 1 MBq release by the actual release amount in MBq.

For water temperatures of 7 and 17 °C , the integrated concentrations should be multiplied by the correction factors given in Table 5. These correction factors are different to those in Table 4 (particularly for the one year integration time) because different physical decay rates and excretion rates affect the time integrated concentration but have less effect on the maximum activity concentration.

Figure 7 gives estimates for integrated concentrations in fish where there is no sorption to suspended sediments (f_p = 0). For the three sites studied here, it was assumed, when calculating activity concentrations in fish, that f_p = 0 for all radionuclides. For cases where $f_p > 0$, these estimates should be multiplied by $(1\text{-}f_p)$: $\sum C_f(x) \rightarrow \sum C_f(x) \times (1 - f_p)$.

Estimates of the time integrated concentration (all integration times > 3 days) of tritium (H-3) in fish may be made by multiplying the time integrated concentration of tritium in water by the fish-water concentration factor. Since, for tritium, CF = 1.0 l kg^{-1} for all water temperatures, this is equal to the integrated water concentration.

The time integrated activity concentration in fish (Bq d kg^{-1}) may be used to determine the time integrated dose:

$$Estimated\ dose = \sum C_f . e_r . I_f . H_{e(ing)} \qquad (16)$$

where e_r is the efficiency of removal of the radionuclide during food processing and preparation (e_r is conservatively assumed to be 1.0 which could lead to an over-estimation by the model since food processing can often reduce radioactivity in fish). I_f is the food ingestion rate and $H_{e(ing)}$ is the effective dose per unit intake by ingestion of the particular radionuclide.

DISCUSSION

Key parameters

It can be seen from Equation (11) that the maximum activity concentration in fish is dependent only on the time-integrated activity concentration in water (ΣC_w) and on the rate of uptake of the radionuclide, k_f. Since ΣC_w is dependent only on the river volumetric flow rate (see Part 1 of this study; Smith et al., 2006), the model presented

in Figure 6 and Table 4 can be applied to all rivers (though note potential variation in bioaccumulation as a function of water chemistry, discussed below). Similarly, the time-integrated activity concentration in fish (ΣC_f) is dependent only on the time integrated activity concentration in water (and hence on river volumetric flow rate) and the uptake and excretion parameters (Eq. 12). Importantly, neither maximum nor time-integrated activity concentrations in fish are influenced by the duration of the radionuclide release for release periods up to 24 hours (given the same total amount of radioactivity released).

The attachment of radionuclides to suspended and bed sediments can potentially reduce the uptake of radionuclides to the aquatic food chain. In the model, however (see Part I of this study, Smith et al., 2006), it is conservatively assumed that there is no absorption of radionuclides to suspended matter – i.e. that all radioactivity is available for uptake to the aquatic food chain. This could lead to an over-estimate of predicted activity concentrations in fish in some cases. The review presented in Part I of this study (Table 2, Smith et al., 2006) suggests that for some radionuclides, particulate sorbed fractions, f_p, could be up to 0.5. This could lead to a factor of two over-estimate in maximum and time-integrated activity concentrations in fish if this particulate radioactivity is unavailable for uptake to fish.

Model tests

The model for ^{137}Cs uptake gives good, somewhat conservative, "blind" predictions of the post-Chernobyl activity concentrations in fish from Windermere (Figure 2). The mean ratio of modelled÷measured values was 2.0 (i.e. on average, the model over-estimates measured values by a factor of 2) and 95% of predictions were within the range 0.9 – 5.4 times measured values. A similar model (Smith et al., 2002; 2005) has been observed to give good predictions in other aquatic systems. The model for ^{90}Sr in fish (Figure 3) also gives good blind predictions, though the time resolution of the data is such that the time changes are only tested to a limited extent. The average model over-estimate of annual mean measurements was a factor 1.2 with range in predictions from 0.3-3 times measured annual averages. The Chowdhury and Blust (2001) model (in both original and simplified forms) and the food uptake model (Eq. 6) each gave similar predictions.

A previous test of the ^{90}Sr uptake model under-estimated ^{90}Sr in fish from Lake Uruskul, Siberia (Smith et al., 2005) possibly due to very long term continuing accumulation of ^{90}Sr in bony tissues. In this latter study, however, the maximum ^{90}Sr concentration in fish was predicted to within a factor of two, implying that - in the short term - the model performed reasonably well.

As shown in Figure 5, the model significantly over-estimates ^{60}Co activity concentrations in fish. Model predictions were typically between 10 and 20 times higher than measured values. This may be because (as is typical for laboratory studies) very small fish (initial body mass 1 g) were used in the Baudin and Fritsch (1989) study, and we have used an estimated CF_{fish} largely from field data where larger fish are typically studied. In addition, it is appropriate to use a conservative estimate of bioaccumulation in this case because it is also known (Baudin and Fritsch 1989) that water is also an important uptake pathway for ^{60}Co.

It has not been possible to validate the model against data for transuranium elements because we have not found appropriate validation data. As noted above, it is likely that the model significantly over-estimates activity concentrations of these elements in fish.

Assumptions and errors

The estimates give maximum concentrations for an average fish at a particular location in the river. Maxima for individual fish will vary according to feeding behaviour and movement of the fish within the river. The rate of uptake and concentration factors of radionuclides in fish also depend on water chemistry: the values used in the model assume water chemical conditions prevailing in the Thames, a hard water river of relatively high nutrient status. Fish-water concentration factors, and therefore uptake rates may be much higher (around one order of magnitude) for rivers of different water chemistry. In particular, radiostrontium concentration factors will be much higher in soft water rivers and radiocaesium concentration factors and uptake rates will be much higher in rivers of lower potassium concentration (Table 2). There is also evidence that concentration factors of other radionuclides are strongly influenced by water chemistry, as illustrated in Table 6. Whilst this variation has to some extent been accounted for by the generally conservative estimates of *CF* used in the model (Table 6), some changes in the model parameter values may be required for other rivers. Such correction may in particular be required for radiostrontium and radiocaesium (previously developed models for Cs and Sr *CF*, presented again here, can be used).

When considering model uncertainty, it should be noted that, due to uncertainty of model parameter values, conservative assumptions of many parameters have been made, particularly for radionuclides (such as the actinides) about which the empirical evidence of accumulation rates is relatively sparse. Thus there is a strong expectation that the model would over-estimate activity concentrations of these radionuclides in a real situation. For example, assimilation efficiencies of ^{241}Am, and isotopes of Pu and U are likely to be to be much lower than the value $\alpha = 1.0$ assumed in the model. Such conservatism in the model is, however, felt necessary due to the large uncertainties in uptake of some radionuclides (Kryshev and Sazykina, 1994) and potential importance of sorption of radionuclides to exterior surfaces of biota (Kryshev and Sazykina, 1994).

It should also be noted that the *CF* for tritium is estimated assuming that tritium is present as tritiated water. Tritium released from nuclear power plants is "likely to be almost entirely in the form of $^{3}H_2O$" (Williams et al., 2001 quoting Kirchmann et al., 1979). In circumstances where tritium is discharged bound to organic molecules it could, however, be significantly more accumulated in the food chain than tritiated water (Williams et al., 2001). This study reports fish-seawater *CF*s of approximately 10^4 compared to the value of 1 assumed here for tritiated water. The model presented here therefore may significantly under-estimate accumulation of tritium in fish in some circumstances.

Seasonal variation in fish uptake rates via the food pathway

Rates of radionuclide uptake by fish are strongly influenced by feeding rates which in turn are strongly influenced by water temperature. As an example of the influence of seasonal variation on uptake rates, we have calculated ^{137}Cs, ^{32}P and ^{131}I activity concentrations in a 500 g fish at three different temperatures representative of the water temperature of the Thames during winter (7 ^{o}C), spring or autumn (12 ^{o}C), and summer (17 ^{o}C). Figure 8 shows the results of this analysis for a constant water temperature, and constant radionuclide concentration in water of 1 Bq l^{-1}. These are compared with equilibrium activity concentrations predicted by the Concentration Factor (CF) model (see Eqs. 18, 19 below).

The activity concentrations of ^{137}Cs tend towards those predicted using the *CF* model (Figure 8), but for ^{131}I and ^{32}P the steady state concentrations are significantly lower than those predicted using the *CF* model. This is because the *CF* model does not account for radioactive decay of the radionuclide in the fish. From Equation (5) it can be seen that at steady-state:

$$C_f = \frac{k_f C_w}{k_b + \lambda} \tag{17}$$

so in cases where λ is of the order of or greater than k_b (as for ^{131}I, ^{32}P), the *CF* model ($C_f = CF.C_w$) will over-estimate the steady state activity concentration. Note, however, that where *CF* has been determined from field measurements of a given radionuclide *in situ*, it is likely that decay is implicitly accounted for. The over-estimation is only likely to occur when a stable analogue or longer lived radionuclide is used to estimate the *CF* of a short-lived radionuclide.

The water temperature significantly influences the maximum activity concentration in fish, C_f(max) because fish feeding rates are much lower at lower water temperatures. At a water temperature of 17^{o}C, C_f(max) is predicted to be approximately five times higher than at a water temperature of 7^{o}C for all radionuclides except tritium (Table 4). The water temperature has less effect on time integrated activity concentrations in fish, there being less than a factor of three decline in ΣC_f as temperature changes from 17^{o}C to 7^{o}C (Table 5).

Influence of fish size on radionuclide accumulation

Fish feeding rates (Elliot, 1975a) and, for radiocaesium and radiostrontium at least, accumulation rates (Elliott et al., 1992; Koulikov and Ryabov, 1992; Kryshev, 2003) are strongly influenced by fish size. We have studied the sensitivity of the model to fish size by calculating maximum and time-integrated activity concentrations in fish of different sizes for selected illustrative radionuclides (Figure 9). For the analysis a river volumetric flow rate of 10 m^3 s^{-1} and a water temperature of 12^{o}C were assumed.

It is seen (Fig. 10a) that there is a small, but significant influence of fish size on maximum predicted activity concentrations in fish due to a more rapid food ingestion (per unit weight of fish) in small fish. Maximum predicted activity concentrations in a 50 g fish are 1.7 times higher than in a 500 g fish. The influence of fish size on time-

integrated activity concentrations is less significant and depends on the excretion and decay rates of the radionuclide. For Cs-137, the time integrated activity concentration in a 50 g fish is 1.17 times higher than that of a 500 g fish. For P-32, the value for a 50 g fish is 1.4 times higher than for a 500g fish.

Comparison of dynamic model with "concentration factor" approach

Using the concentration factor approach, the maximum concentration in fish is given by:

$$C_f^{\max}(x) \approx C_w^{\max}(x).CF \tag{18}$$

This model leads to significant over-estimates of the maximum activity concentrations in fish for short term releases. A model scenario (vol. flow rate 10 m^3 s^{-1}, duration of release 3 hours, T = 12^{o}C, 500 g fish, Pangbourne-Reading reach of the Thames) illustrates this over-estimation. For all radionuclides studied, the maximum concentration in fish predicted by the *CF* approach was 340 – 3400 times higher than the maximum concentration predicted by the dynamic model (Eq. 3).

The time integrated activity concentration in fish is estimated by the *CF* approach using:

$$\Sigma C_f(x) \approx \Sigma C_w(x).CF \tag{19}$$

This usually only slightly over-estimates integrated activity concentrations since, for long integration times ($\tau \times (k_b + \lambda) >> 1$) and $k_b >> \lambda$ so Equation (12) tends to Equation (19). A hypothetical model scenario (vol. flow rate 10 m^3 s^{-1}, any release duration, T = 12^{o}C, 500 g fish) illustrates this. For all radionuclides studied, the one year time-integrated concentration in fish predicted by the *CF* approach was 1.0 – 4.7 times higher than the time-integrated concentration predicted by the dynamic model (Eq. 5). Of all the radionuclides studied, only I-131 and P-32 showed a greater than factor 2 difference between the two models, due primarily to their short half-lives.

CONCLUSIONS

A simplified model has been presented for predicting radionuclide activity concentrations in fish following a short duration release to a river. The model is generally applicable to all rivers, though it is noted that radionuclide bioaccumulation rates have been estimated on the basis of water chemical conditions prevailing in the River Thames. The general applicability of the model for radionuclides in fish would be strengthened by estimates of fish uptake parameters for river chemical conditions which are significantly different to those of the Thames (particularly for low nutrient, low mineral content rivers). It is also noted that for some radionuclides, for which bioaccumulation is relatively uncertain, the model makes highly conservative assumptions, thus potentially leading to significant over-estimates in model predictions.

It was demonstrated that the maximum and time-integrated activity concentrations in fish were (given the same total amount of radioactivity released) independent of the duration of the radionuclide release for the release durations of up to 24 hours studied here. Both water temperature and fish size had a significant influence on model output, particularly in the predictions of maximum activity concentration in fish. Time-integrated activity concentration in fish was much less sensitive to the values of the uptake and excretion parameters. This was illustrated by comparison of the dynamic model with the simple Concentration Factor approach. This showed, for the radionuclides with half life greater than that of ^{32}P (14.3 days) that assuming instantaneous uptake and release (as in the *CF* approach) had only a minor influence on model predictions compared to the much slower uptake and release assumed in the dynamic model. Whilst, for short-duration releases, maximum activity concentrations in fish were significantly over-estimated by the Concentration Factor model, the two models showed relatively little difference in predictions of time-integrated activity concentrations.

The radiation risk to humans from the water-fish pathway is a function of the total radioactivity ingested and hence is directly proportional to the time-integrated activity concentration in fish (assuming averaged rates of ingestion). Therefore, the radiation dose to humans from short-duration releases to rivers (via the water-fish pathway) can be predicted with reasonable accuracy on the basis of the simple Concentration Factor model (Eq. 19). Dynamic models are only required if the maximum activity concentration in fish needs to be predicted.

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Table 1. Values of constants for different temperature ranges in model for estimating fish feeding rates (from Elliot 1975a).

$T\,^oC$	A_D	B_1	B_3
3.8 - 6.6	0.654	0.762	0.418
6.6 - 13.3	3.384	0.759	0.172
13.3 - 18.4	5.956	0.767	0.126

Table 4. Correction factors for converting estimates of maximum fish concentration from a water temperature of 12°C to other temperatures

	Ratio: $\frac{C_f(x)\ at\ 7^oC}{C_f(x)\ at\ 12^oC}$	Ratio: $\frac{C_f(x)\ at\ 17^oC}{C_f(x)\ at\ 12^oC}$
Correction Factor	0.42	2.0

Table 5. Correction factors for converting estimates of time integrated fish concentration from a water temperature of 12°C to other temperatures

Radionuclide	Ratio: $\frac{\Sigma C_f(x)\ at\ 7^oC}{\Sigma C_f(x)\ at\ 12^oC}$	Ratio: $\frac{\Sigma C_f(x)\ at\ 17^oC}{\Sigma C_f(x)\ at\ 12^oC}$
One week integration time		
All RNs	0.45	2.0
One year integration time		
Cs-134, Cs-137	0.65	1.24
P-32	0.52	1.52
I-125	0.70	1.21
I-131	0.49	1.66
Sr-89	0.68	1.22
Sr-90	0.98	1.0
Co-60	0.53	1.44
C-14	0.57	1.30
Pu, U, Am	1.0	1.0
Zn-65	0.88	1.07

Table 6 Concentration factors of freshwater fish (muscle) in waters of low and high mineral content (adapted from Blaylock, 1982).

Element	Water of low mineral content	Water of high mineral content	*CF* used in the model
Hydrogen	1	1	1
Carbon	5×10^4	5×10^3	2.2×10^4
Cobalt	1×10^3	50	300
Ruthenium	100	10	-
Iodine	50	5	40
Radium	150	10	-
Uranium	20	2	50
Plutonium	50	5	50

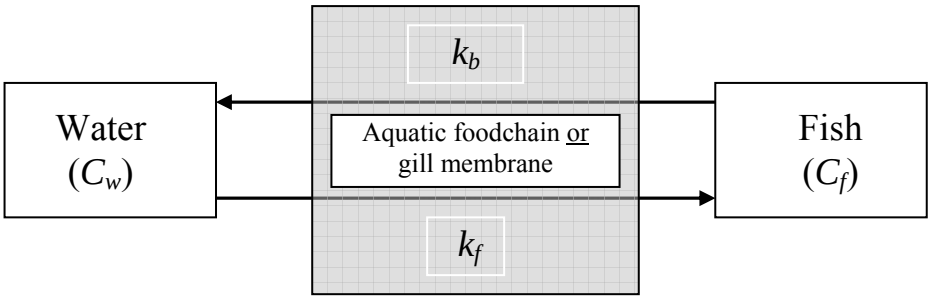


Figure 1. Illustration of model for uptake in fish via the aquatic food chain or gills. Mathematically, both models are identical, but the uptake parameters are determined differently.

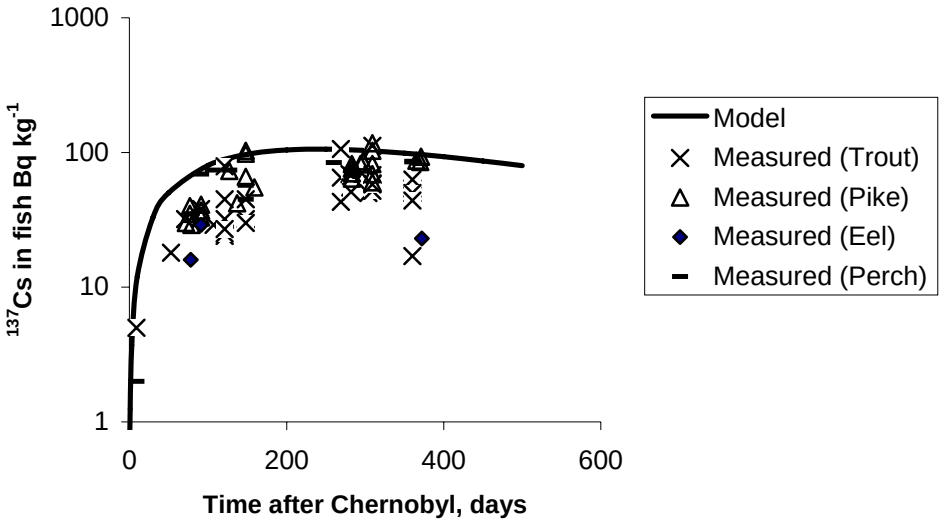


Figure 2. Test of the ^{137}Cs uptake model against measurements in Windermere after Chernobyl (data from Camplin et al., 1989).

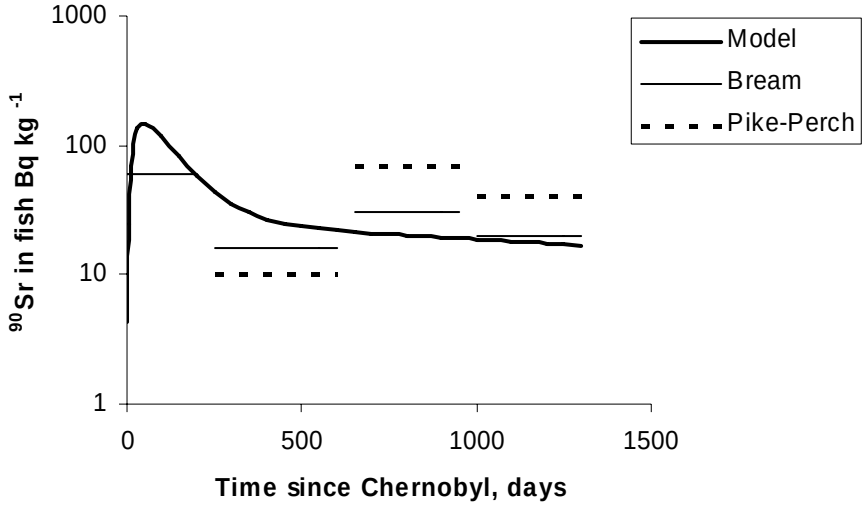


Figure 3. Test of the ^{90}Sr uptake model against measurements in the Kiev Reservoir after Chernobyl. Data from Kryshev and Ryazantsev (2000).

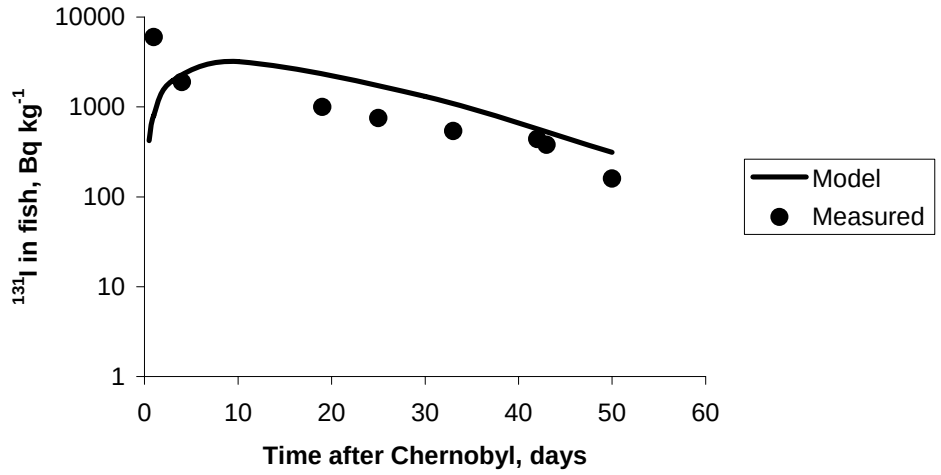


Figure 4. Test of the ^{131}I uptake model against measurements in the Kiev Reservoir after Chernobyl (Kryshev and Ryazantsev, 2000). Adapted from Smith et al. (2005).

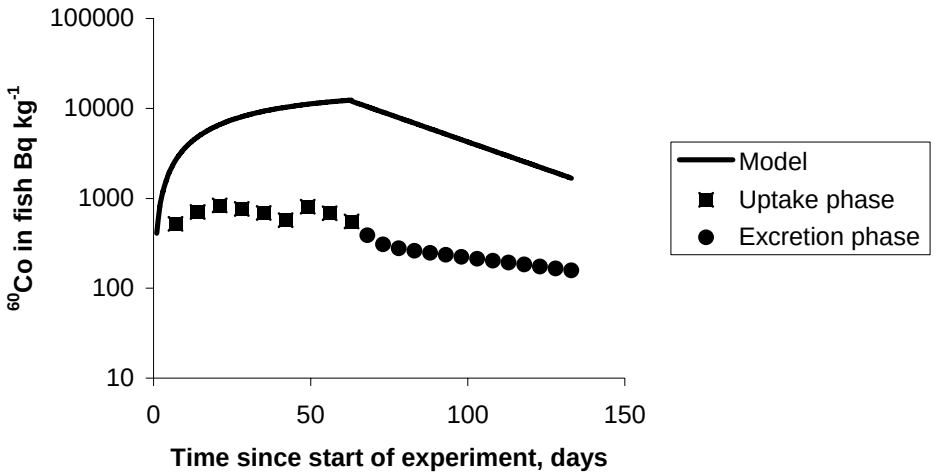


Figure 5. Test of the ^{60}Co uptake model against measurements in a laboratory experiment (data from Baudin and Fritsch 1989).

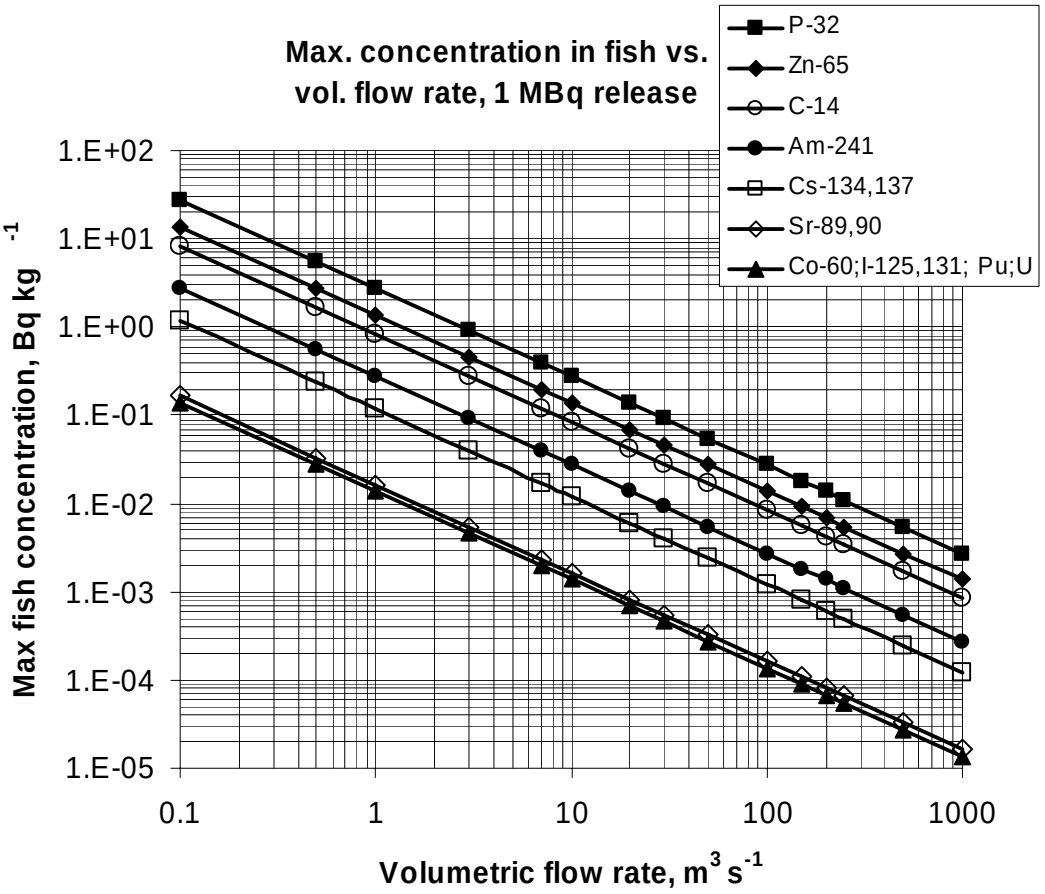
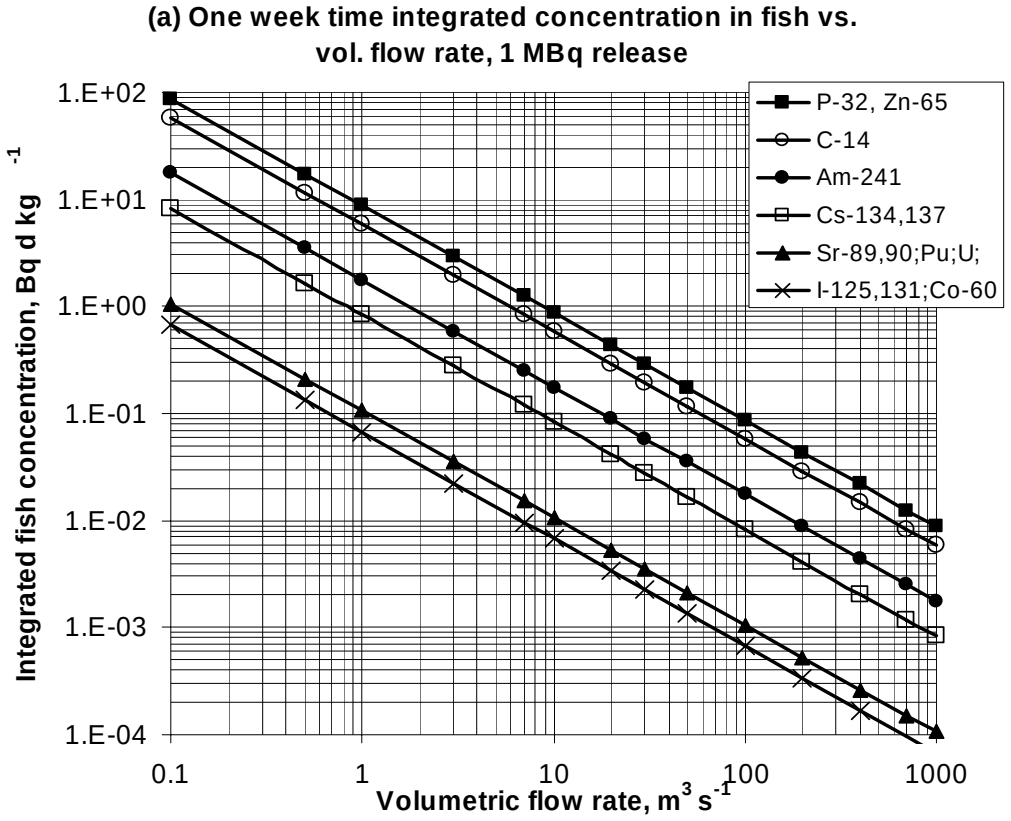


Figure 6. Estimated maximum concentrations in fish for a 1MBq release of different radionuclides at a water temperature of 12°C. For other temperatures, the maximum concentrations should be multiplied by the correction factors given in Table 4. The results apply to all distances <10 km downstream of the release for all release durations up to 24 h.



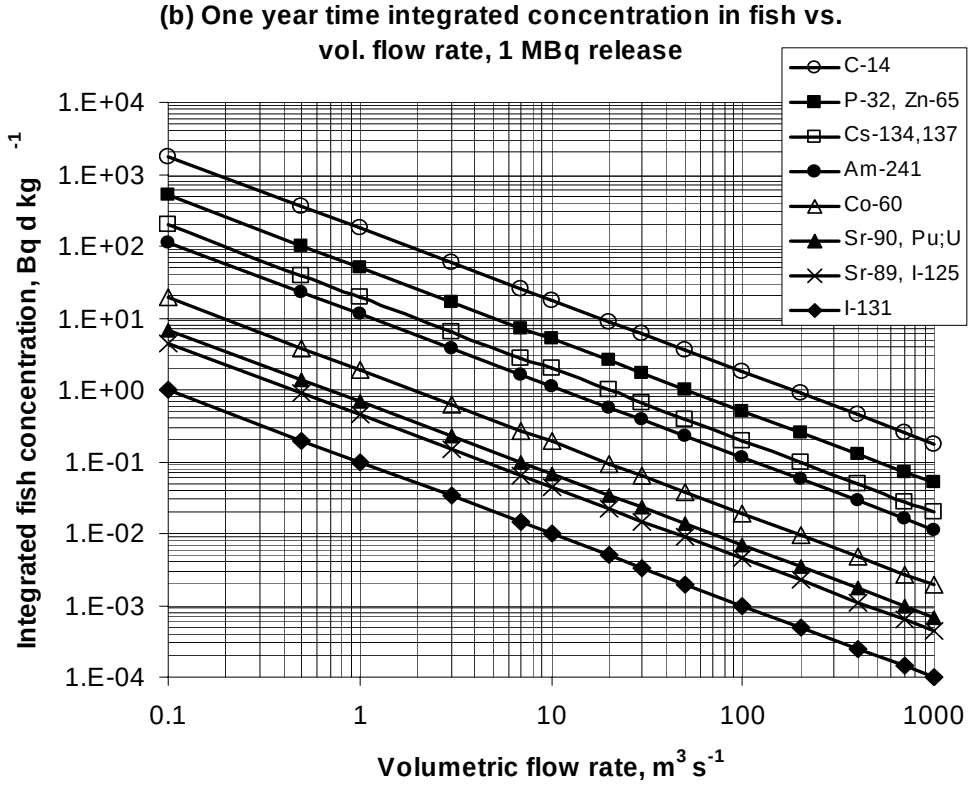


Figure 7. Estimated (a) one week; (b) one year time integrated activity concentrations in fish for a 1MBq release, water temperature of 12°C. For other temperatures, the integrated concentrations should be multiplied by correction factors given in Table 5. Results apply to all distances <10 km downstream and for all release durations.

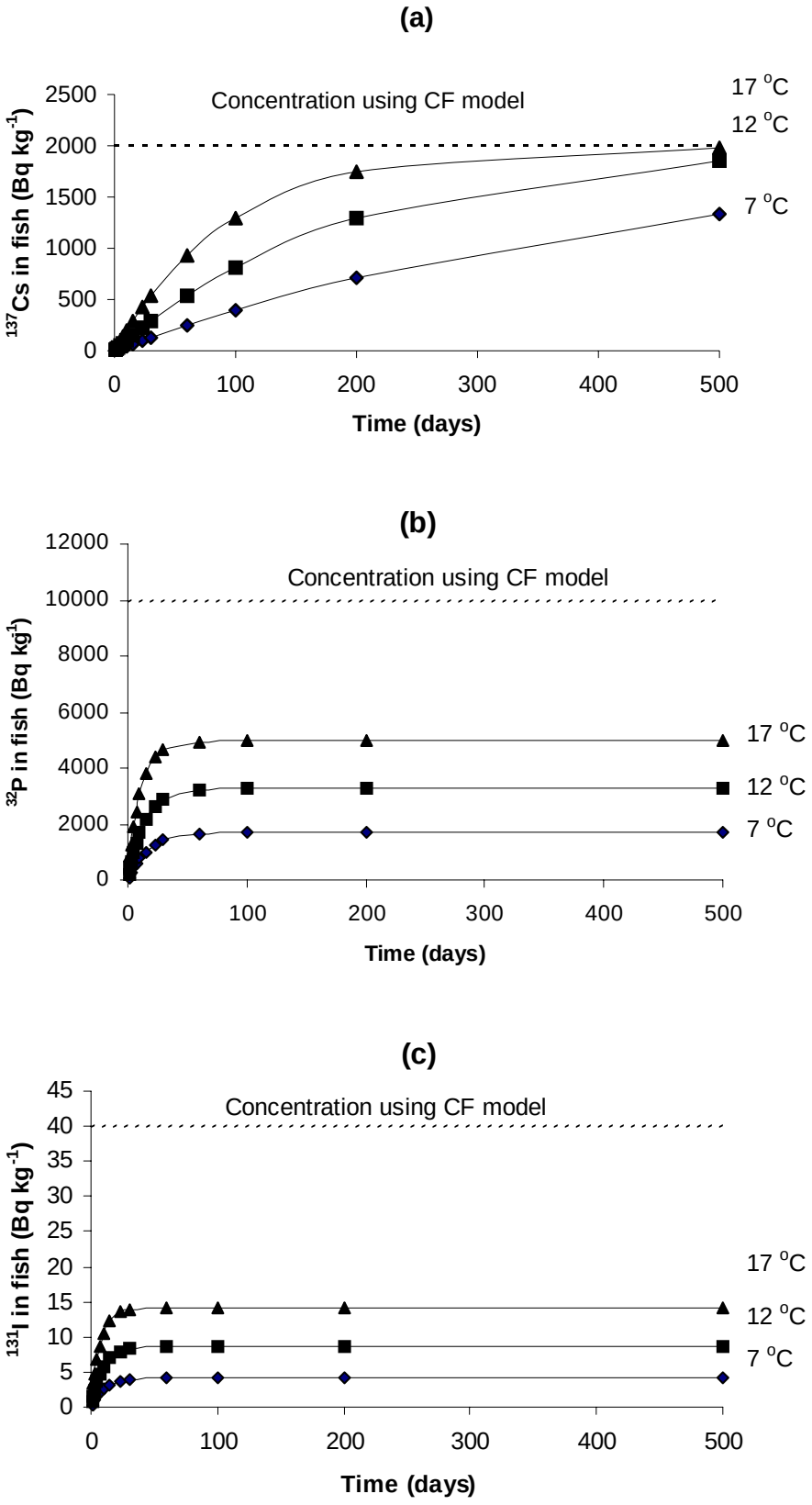


Figure 8. Variation in uptake of (a) ^{137}Cs; (b) ^{32}P and (c) ^{131}I in fish as a function of water temperature.

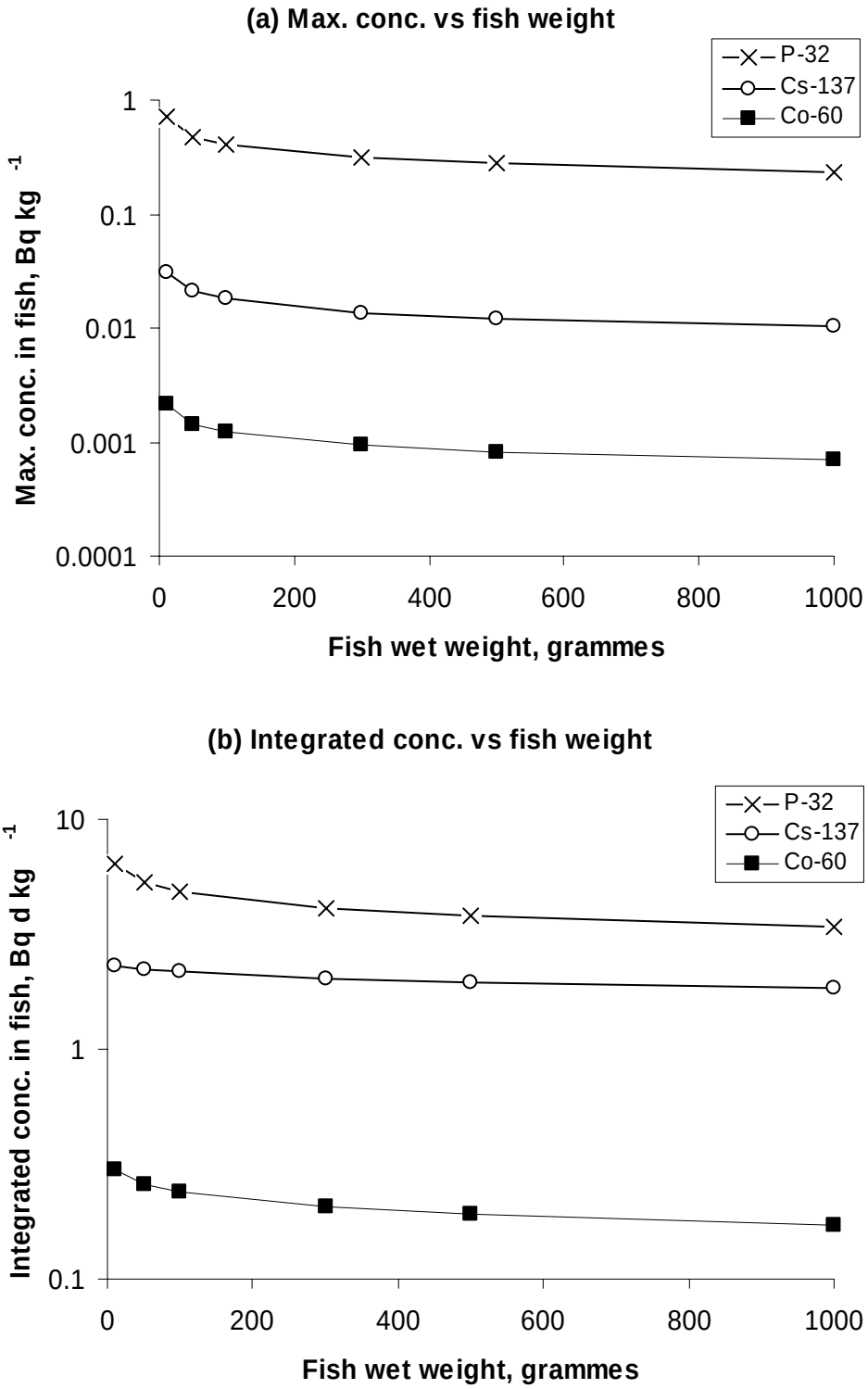


Figure 9. Sensitivity of model output to fish wet weight for selected radionuclides: (a) maximum concentration in fish and (b) one-year time integrated concentration in fish. The scenario assumes a 1 MBq release, river volumetric flow rate of 10 m^3 s^{-1}, a temperature of 12^oC and that all of the radionuclide remains in the dissolved phase.

Table 2. Concentration factors of radionuclides in whole fish. Note that some reviews quoted (Blaylock, 1982; Coughtrey and Thorne, 1983; IAEA, 1994) are likely to have been based on some of the same empirical data. Concentration factors (*CF*) are given for wet weight of freshwater (whole) fish unless stated otherwise. Where distinction between piscivorous and non-piscivorous fish has been possible, values for piscivorous fish were chosen. *CF* values were chosen for the Thames which has high mineral and nutrient content of the water. *CF* values for rivers of lower nutrient and/or mineral content may be higher.

Element	Uptake pathway	Estimated CF used in model, l kg^{-1}	Ranges and other estimates of CF, l kg^{-1}	Notes and references
^{3}H	Water/food	**1**	1 (0 – 10)	IAEA (1994) Estimate and range.
^{14}C	Food	**2.2×10^4**	2.2×10^4 5×10^4 5×10^3 - 5×10^4	Estimate from stable carbon in fish (10% of wet weight, Vinogradov, 1953) and DOC in the Thames of 4.4 mg l^{-1} (Neal & Robson 2000). Likely to be over-estimate. IAEA (1994) Blaylock (1982), value for fish muscle, high value is for low mineral content water, low value for high mineral content water.
^{32}P	Food	**1×10^4**	5×10^4 (3×10^3 - 1×10^5) 3300 4020	IAEA (1994) Estimate and range. This study, from P = 0.3% of wet weight (Vinogradov, 1953) and SRP 0.91 mg l^{-1} for the Thames (Neal and Robson, 2000). Denyer (2001) estimate for Grand Union Canal for stable P.
$^{134,137}Cs$	Food.	**2×10^3**	2×10^3 (3×10^1 - 3×10^3) $3 \times 10^3/[K^+]$ 450^* $4880/[K^+]$ 730^+ $\log(CF) = 3.3 - 0.72\log[K^+] + 0.29 - 0.23\log[s]$ 570^{**}	IAEA (1994). Estimate and range. Coughtrey & Thorne (1983), for turbid water. *Estimate assuming $[K^+]$ = 6.7 mg l^{-1} in the Thames (Neal & Robson, 2000). Smith et al.(2000), inverse relation with $[K^+]$. $^+$Estimate assuming $[K^+]$ = 6.7 mg l^{-1} in the Thames (Neal & Robson, 2000). Rowan & Rasmussen (1994), $[K^+]$ is in mg l^{-1}, s is the suspended solids conc. in mg l^{-1}. **Estimate assuming $[K^+]$ = 6.7 mg l^{-1}, s = 13.0 mg l^{-1} in the Thames (Neal & Robson, 2000).
$^{89,90}Sr$	Sr is taken up from water by the same mechanisms as Ca which (Vinogradov, 1953) is primarily absorbed direct from water via the gills. Other studies, however have assumed uptake via the food chain (Kryshev, 2003). It is likely that both	**60**	60 ($1 - 1 \times 10^3$) $CF = \exp(5.2 - 1.2 \ln[Ca^{2+}])$ 0.54^* 56 (0.82 – 198) 3.9 (0.74 – 10) 10.8	IAEA (1994) Best estimate and range. Vanderploeg et al. (1975), quoted in Blaylock (1982) for edible portions. Edible portions of fish have much lower CF's than whole fish. *Estimate for $[Ca^{2+}]$ (121 mg l^{-1}) in the Thames (Neal & Robson, 2000) for edible portions. Blaylock (1982) for edible portions of fish; calcium concentration in water < 20 mg l^{-1}.Estimate and range. Blaylock (1982) for edible portions of fish; calcium concentration in water in range 20-60 mg l^{-1}. Estimate and range. Estimate from relations given by Vanderploeg et al. (1975),

	uptake mechanisms operate.			quoted in Blaylock (1982) (see text) for whole fish for [Ca^{2+}] (121 mg l^{-1}) in the Thames (Neal & Robson, 2000).
^{65}Zn	Coughtrey & Thorne (1983) cite 3 reports referring to the importance of zinc intake from food, but note other reports have referred to passive sorption via the gills.	**5×10^3**	1×10^3 ($1 \times 10^2 - 3 \times 10^3$) 2×10^3 ($2.8 \times 10^2 - 2 \times 10^4$) 4600 1250	IAEA (1994) Estimate and range. Coughtrey & Thorne (1983). Estimate and range. CF likely to be related to stable Zn concentration. This study, from Zn(stable) = 0.003% of wet weight of fish (from data in Vinogradov, 1953) and [Zn] 6.5 x 10^{-6} g/l for the Thames (Neal and Robson, 2000). Denyer (2001) estimate for Grand Union Canal from stable Zn concentration.
$^{125,131}I$	Limited data, but uptake is expected to be rapid (Kryshev 1995)	**40**	40 ($20 - 6 \times 10^2$) 40 (10 – 132) 10 30	IAEA (1994). Estimate and range. Blaylock (1982), Estimate and range for fish muscle Kryshev (1995) Coughtrey et al. (1983) *CF expected to be related to stable element concentration.* *Expected to concentrate somewhat in the thyroid.*
^{60}Co	Evidence for significant uptake from both water and food (Baudin & Fritsch 1989).	**3×10^2**	3×10^2 (10 – 300) 2.5×10^2 5-280 20	IAEA (1994). Estimate and range. Coughtrey & Thorne (1983) Blaylock (1982), whole fish. Blaylock (1982), value for fish muscle. *Greatest accumulation in internal organs, particularly kidneys.*
U	Not known	**50**	10 (2 – 50) 0.3 - 0.6 8 0.7-38	IAEA (1994) Estimate and range. Blaylock (1982) planktivorous fish L. Michigan. Blaylock (1982) planktivorous fish, Lake Issyk-kul, Soviet Union. Blaylock (1982) omnivorous fish, Zirovski mining area, Yugoslavia.
Pu	Not known	**50**	30 (4 – 300) 35 (10 – 1000) 0.4 – 7	IAEA (1994) Estimate and range. Coughtrey et al. (1984a) Estimate and range. Blaylock (1982) *Accumulation decreases with increasing trophic level. Edible portions have lower CF's than whole fish.*
^{241}Am	Not known	**1000**	30 (30 – 300) 840 (700-1000)	IAEA (1994) Estimate and range. Coughtrey et al. (1984b) Vol 5. Best estimate and range.

Table 3. Estimated uptake and excretion rates of radionuclides in a 500 g fish at 12 ^{o}C. All isotopes are assumed to be assimilated via the food pathway except radiostrontium. For radiostrontium, uptake and excretion rates are compared between (a) uptake via the gills and (b) via the food chain. For ^{3}H, no uptake mechanism is specified (measured uptake rates are used rather than an uptake model).

Element	Pathway	Relevant water chemistry	*CF(fish)* l kg^{-1}	*CF(food)* l kg^{-1}	Assimilation efficiency, α	Feeding rate, w.w. g dy^{-1} @ 12 ^{o}C	Uptake rate k_f l kg^{-1} d^{-1} @ 12 ^{o}C	Excretion rate, k_b d^{-1} @ 12 ^{o}C
^{134,137}Cs	Food	[K] 6.7[1] mg l^{-1}	2×10^{3}	= *CF(fish)/2*	0.44[2]	11.8[3]	10.4	0.0052
^{89,90}Sr	Water[a] Food[b]	[Ca] 121[1] mg l^{-1} [Sr] 0.36[1] mg l^{-1} pH 8.1[1]	60	N/A[a] = *CF(fish)*[b]	N/A[a] 1.0[b]	N/A[a] 11.8[3 b]	0.68[a] 1.42[b]	0.011[a] 0.024[b]
^{125,131}I	Food		40	= *CF(fish)*	1.0	11.8[3]	0.94	0.024
^{60}Co	Food		3×10^{2}	= *CF(fish)*	0.1	11.8[3]	0.71	0.0024
^{3}H	N/A		1.0	1.0	N/A	N/A	0.69	0.69
^{32}P	Food	[SRP] stable = 0.91[1] mg l^{-1}	1.0×10^{4}	= *CF(fish)*	1.0	11.8[3]	236.0	0.024
^{14}C	Food	DOC 4.4[1] mg l^{-1}	2.2×10^{4}	= *CF(fish)*	0.14[4]	11.8[3]	72.7	0.0033
Pu, U	Food		50	= *CF(fish)*	1.0	11.8[3]	1.18	0.024
^{65}Zn	Food	[Zn] stable = [1]6.5×10^{-3} mg l^{-1}	5000	= *CF(fish)*	1.0	11.8[3]	118.0	0.024
^{241}Am	Food		1000	= *CF(fish)*	1.0	11.8[3]	23.6	0.024

1. Neal and Robson, 2000; 2. Tucker and Rasmussen (1999); 3. Elliott (1975a); 4. From data in Elliott (1975b).