Smith, J.T.; Bowes, M.J.; Cailes, C.R.. 2011 A review and model assessment of 32P and 33P uptake to biota in freshwater systems. *Journal of Environmental Radioactivity*, 102 (4). 317-325. 10.1016/j.jenvrad.2010.12.006

A review and model assessment of ^{32}P and ^{33}P uptake to biota in freshwater systems

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Abstract

Bioaccumulation of key short-lived radionuclides such as ^{131}I and ^{32,33}P may be over-estimated since concentration ratios (*CR*s) are often based on values for the corresponding stable isotope which do not account for radioactive decay during uptake via the food chain. This study presents estimates for bioaccumulation of radioactive phosphorus which account for both radioactive decay and varying ambient levels of stable P in the environment. Recommended interim *CR* values for radioactive forms of P as a function of bioavailable stable phosphorus in the water body are presented. Values of *CR* are presented for three different trophic levels of the aquatic food chain; foodstuffs from all three trophic levels may potentially be consumed by humans. It is concluded that current recommended values of the *CR* are likely to be significantly over-estimated for radioactive phosphorus in many freshwater systems, particularly lowland rivers. Further research is recommended to field-validate these models and assess their uncertainty. The relative importance of food-chain uptake and direct uptake from water are also assessed from a review of the literature. It can be concluded that food-chain uptake is the dominant accumulation pathway in fish and hence accumulation factors for radioactive phosphorus in farmed fish are likely to be significantly lower than those for wild fish.

Keywords: bioaccumulation; phosphorus, water, fish, P-32, P-33.

1. Introduction

Variability in the bioaccumulation of radionuclides from water to aquatic organisms is an important source of uncertainty in generic models for the prediction of radiation doses both to humans and aquatic biota (Monte et al., 2003; Yankovich et al., 2010). Bioaccumulation factors (here termed the concentration ratio, *CR*) for key short-lived radionuclides such as ^{131}I and ^{32,33}P may be over-estimated since they are often based on values for the corresponding stable isotope which do not account for radioactive decay during uptake via the food chain. This study presents estimates for bioaccumulation factors of radioactive phosphorus which account for both radioactive decay and varying ambient levels of stable P in the environment. The concentration ratio for aquatic biota is defined here as:

$$CR = \frac{\text{Activity concentration in biota (Bq kg}^{-1}\text{ fresh weight)}}{\text{Activity concentration in water (Bq l}^{-1}\text{)}} \quad \text{l kg}^{-1} \qquad (1)$$

The *CR* assessment carried out here will use the River Cam at Cambridge, UK, as a case study, but generalised interim *CR* values will also be determined for other rivers, based on uptake of stable P.

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 water chemistry (for example, Smith et al., 2002). For radionuclides which have a suitable stable element (for example, ^{14}C) or a close stable element analogue (for example, potassium for ^{134,137}Cs) the bioaccumulation of the stable element is a valuable guide to that of the radioisotope. If the rate of accumulation of a radionuclide is rapid in comparison to the radioactive decay time (i.e. if equilibrium is reached before much of the radionuclide has decayed), then the *CR* for the radioactive element will be close to that of the stable element. However, it is known (for example, Elliott

et al., 1992; Smith et al., 2005) that radionuclide uptake in aquatic food chains can take place over timescales of months or greater. Therefore, for short-lived radionuclides, such as ^{131}I ($T_{1/2}$ = 8.02 days) and $^{32,33}P$ ($T_{1/2}$ = 14.26 and 25.34 days respectively), *CR* values based on stable analogue elements are likely to give overestimates of the bioaccumulation of the radioactive element.

Phosphorus is an essential nutrient for all organisms, being a key component of phospholipid cell membranes, DNA and RNA, and of ATP, used to transport energy within the cell. Phosphorus is also deposited in the bones of vertebrate animals. Kryshev (2004) quotes Saurov et al. (Saurov et al., 1973) who found in experiments that 70% of ^{32}P is accumulated in the bones and 30% in soft tissues. Phosphorus is homeostatically regulated in organisms: in other words, P is assimilated to the level needed and excess P is excreted from the body. This means that body concentrations of P remain relatively stable. Bioaccumulation of radioactive phosphorus is determined by the availability of stable P, with high accumulation of radiophosphorus being observed in low stable P systems (for example, Poston and Klopfer, 1986).

In evaluating the uptake of radionuclides in farmed fish, is also important to consider the accumulation pathway of the radio-element since (presuming that they are fed radioactively "clean" food), uptake to farmed fish via the food chain will be of minor importance. Therefore, if the food chain is the dominant radiophosphorus accumulation pathway (compared to direct uptake from water), then recommended *CR* values for wild fish are likely to over-estimate uptake of radionuclides to farmed fish. The relative importance of these two uptake pathways will therefore be considered in this study.

1.1. Current recommended value of the radiophosphorus-water CR

The International Atomic Energy Agency recommended value (IAEA, 1994) for the radiophosphorus concentration ratio in "edible portions" of fish was previously 5×10^4 l kg^{-1}, with a range 3×10^3 - 1×10^5 l kg^{-1}. The recent revision to this document (IAEA, 2010) recommends a value of 1.4×10^5 l kg^{-1} (range 1.2-1.7 $\times 10^5$ l kg^{-1}) for fish muscle, this value presumably being based on the stable P *CR*. It is notable that the latter (IAEA, 2010) study recommends a much narrower range of possible values than the former (IAEA, 1994). Using

the recommended correction factor to account for decay of short-lived radionuclides (IAEA, 2010), the stable P *CR* of 1.4×10^5 l kg^{-1} would lead to recommended *CR*s of 4.5×10^4 l kg^{-1} for ^{32}P and 6.4×10^4 l kg^{-1} for ^{33}P, values very close to that recommended in the earlier (IAEA, 1994) review.

It is likely that these conservative estimates are for relatively low stable P concentration systems in which bioaccumulation of the available P is particularly high. Many lowland rivers have elevated P concentrations from anthropogenic sources (primarily diffuse agricultural runoff and point sources from sewage treatment works) (Bowes et al., 2010; Foy, 2007; White and Hammond, 2009). The IAEA (1994) best estimate and range appears to be based on a literature review by Poston et al. (1986) and an earlier review by Kahn and Turgeon (1980; cited; Poston and Klopfer, 1986). According to Poston and Knopfler (1986), Kahn and Turgeon (1980) "recommend a *CR* of 3000 [l kg^{-1}] for ^{32}P and 70,000 [l kg^{-1}] for stable P", the difference being due to decay of ^{32}P as it accumulates up the aquatic food chain. It was not possible to consult the Kahn and Turgeon (1980) report directly, but a later paper by these authors (Kahn and Turgeon, 1984) describes the derivation of the concentration factor for stable and radioactive P. The stable P *CR* is obtained (using Equation 2, below) from a review of phosphorus concentrations in fish muscle (assumed value 2.2 g/kg f.w) and in the dissolved phase of river waters (assumed value 0.03 mg l^{-1}). In their later paper, Kahn and Turgeon (1984) use a model for phosphorus turnover in fish to infer a *CR* which is 20 times lower than that for stable P (i.e. 3500 l kg^{-1}). This latter value is close to that quoted by Poston and Knopfler (1986) based on the earlier Kahn and Turgeon (1980) study.

2. Methods

2.1. Literature search

A literature search was carried out to critically assess the available information on the bioaccumulation of both stable and radioactive phosphorus. Estimates of the fish-water *CR* may be made from measurements (in the lab or field) reported in the literature. Alternatively, in appropriate cases, *CR* may be estimated from the concentration factor of the stable isotope (assuming instantaneous uptake of P by the organism, and therefore, for the moment, ignoring radioactive decay):

$$CR(P-32{,}33) \approx CR(stable\ P) = \frac{Conc.\ of\ \ stable\ P\ in\ organism\ (kg\,kg^{-1})}{Conc.\ of\ available\ stable\ P\ in\ water\ (kg\,l^{-1})} \qquad (2)$$

where we have ignored the minor effects of P mass discrimination.

The equilibrium *CR* 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.

2.2. Modelling

The modelling approach is similar to those presented in, for example, Kryshev (2004) and Smith et al. (Smith, 2006; 2002), being based on an evaluation of concentrations and uptake/excretion rates of stable phosphorus. For comparison with other modelling approaches, a review of models for uptake of radionuclides in different biotic components of lake ecosystems is given by Monte et al. (2003).

In this study, a relatively conservative approach was taken in which the aquatic food chain was divided into three levels. The first level consisted of macrophytes, phyto- and zoo-plankton, algae, and macroinvertebrates. The second level consisted of small, omnivorous fish and the third of larger predatory fish. It was assumed (see *Uptake and Excretion Rates* section, below) that the transfer of radioactive phosphorus from water to the first trophic level is effectively instantaneous. This conservative approach under-emphasises the important role detritus plays in the food chain of many river systems. A significant contribution of detritus in the food web of a river will tend to retard radiophosphorus uptake to fish much more than is predicted by the simplified model used here. Assimilation of radioactive P in detritus at the bottom of the river, breakdown of that detritus and uptake by bacteria, invertebrates and detritivorous fish is likely to significantly retard uptake of radioactive P by fish and hence reduce activity concentrations in predatory fish (compared to those predicted by the simplified food chain model used here).

The concentration in water, C_w (Bq l^{-1}) and the three biota compartments C_1, C_2, C_3 (Bq kg^{-1}) are mediated by the forward, k_{fn}, and backward, k_{bn}, rate constants where n is the trophic level (see Figure 1). Note that k_{f1} has dimensions of [length^3 mass^{-1} time^{-1}], but all other rate constants have dimensions of [time^{-1}]. The equations describing the concentrations in each trophic level 1-3 are as follows:

$$\frac{dC_1}{dt} = k_{f1}C_w - (k_{b1} + \lambda)C_1 \quad (3)$$

$$\frac{dC_2}{dt} = k_{f2}C_1 - (k_{b2} + \lambda)C_2 \quad (4)$$

$$\frac{dC_3}{dt} = k_{f3}C_2 - (k_{b3} + \lambda)C_3 \quad (5)$$

For a time-constant activity concentration in water, C_w, and where the activity concentrations in biota are zero at time $t = 0$, the analytical solution to these simultaneous equations is as follows:

$$C_1 = \frac{k_{f1}C_w}{(k_{b1} + \lambda)}\left(1 - \exp(-(k_{b1} + \lambda)t)\right) \quad (6)$$

$$C_2 = \varphi\left[\frac{\left(1 - \exp(-(k_{b2} + \lambda)t\right)}{(k_{b2} + \lambda)} + \frac{k_{b2}\left(\exp(-(k_{b1} + \lambda)t) - \exp(-(k_{b2} + \lambda)t)\right)}{(k_{b2} - k_{b1})}\right] \quad (7)$$

$$C_3 = k_{f3}\varphi\left[\frac{\left(1 - \exp(-(k_{b3} + \lambda)t\right)}{(k_{b2} + \lambda)(k_{b3} + \lambda)} - \frac{\left(\exp(-(k_{b2} + \lambda)t) - \exp(-(k_{b3} + \lambda)t)\right)}{(k_{b2} + \lambda)(k_{b3} - k_{b2})}\right.$$
$$\left. + \frac{k_{b2}\left(\exp(-(k_{b1} + \lambda)t) - \exp(-(k_{b3} + \lambda)t)\right)}{(k_{b2} - k_{b1})(k_{b3} - k_{b2})} - \frac{k_{b1}\left(\exp(-(k_{b1} + \lambda)t) - \exp(-(k_{b3} + \lambda)t)\right)}{(k_{b2} - k_{b1})(k_{b3} - k_{b1})}\right] \quad (8)$$

where:

$$\varphi = \frac{k_{f2} k_{f1} C_w}{(k_{b1} + \lambda)} \qquad (9)$$

It is useful to consider the behaviour of these equations at equilibrium (i.e. as time $\rightarrow \infty$). For constant activity concentration in water, at equilibrium, we get:

$$CF(\textit{trophic level } 1) = \frac{C_1}{C_w} = \frac{k_{f1}}{k_{b1} + \lambda} \tag{10}$$

$$CF(\textit{trophic level } 2) = \frac{C_2}{C_w} = \frac{k_{f2}k_{f1}}{(k_{b1} + \lambda)(k_{b2} + \lambda)} \tag{11}$$

$$CF(\textit{trophic level } 3) = \frac{C_3}{C_w} = \frac{k_{f3}k_{f2}k_{f1}}{(k_{b1} + \lambda)(k_{b2} + \lambda)(k_{b3} + \lambda)} \tag{12}$$

and

$$\frac{CF(\textit{trophic level } 2)}{CF(\textit{trophic level } 1)} = \frac{C_2}{C_1} = \frac{k_{f2}}{(k_{b2} + \lambda)} \tag{13}$$

$$\frac{CF(\textit{trophic level } 3)}{CF(\textit{trophic level } 2)} = \frac{C_3}{C_2} = \frac{k_{f3}}{(k_{b3} + \lambda)} \tag{14}$$

These equilibrium solutions allow us, in a simple way, to investigate the influence of uptake and excretion rate parameters on the equilibrium concentration factor of radioactive phosphorus. The solutions equal those for stable phosphorus uptake for the special case: $\lambda = 0$.

2.4. Model parameterisation

2.4.1. Stable P in freshwater organisms

The uptake and excretion rates of radioactive phosphorus were estimated by evaluating the accumulation of stable phosphorus in aquatic biota. A literature review was conducted to

obtain conservative values of the stable P concentration in biota in the three trophic levels, and values of the stable phosphorus assimilation efficiency (Table 1). It is noted that there

was significant variation in stable P concentrations. These data imply that the highest P concentrations are found in relatively large fish, though size-related bio-magnification of P concentrations appears not to be high. Based on the information in Table 1, the stable P concentrations in organisms that are used in the modelling are presented in Table 2, using the trophic level scheme described in the *Modelling* section above.

In addition to the data for whole fish presented in Table 1, a review of stable P content in fish muscle by (Kahn and Turgeon, 1984) estimated a geometric mean of 2.2 g kg^{-1} f.w. This is lower than the value we have assumed here (10 g kg^{-1} f.w. for predatory fish) for whole fish (i.e. including skeleton). P concentrations in muscle are expected to be significantly lower than those in bone, which has a mean P concentration of 24 g kg^{-1} f.w. (Kahn and Turgeon, 1984), giving a bone:muscle concentration ratio of 11. This compares with a study by Yankovich (2009) who reported a literature value of bone:muscle *CR* of 16 compared to a mean value in Perch Lake, Canada of 46.3.

2.4.2. Stable P concentration in water

Since phosphorus is homeostatically controlled in organisms, the fish-phosphorus *CR* (for both stable and radioactive forms) is strongly dependent on available stable P concentration in water. Stable phosphorus concentrations vary widely in surface waters due to differing catchment geology, soil types, catchment and waterbody characteristics and human impacts. Concentrations in different water bodies can range from a few µg l^{-1} to a few mg l^{-1}. Phosphorus may be measured as Total Phosphorus (TP, the P both in the dissolved phase and the less readily bio-available P absorbed to or incorporated in suspended particles), Total Dissolved Phosphorus (inorganic PO_4^{3-} and HPO_4^{2-}, plus some low molecular weight organic phosphorus) or Soluble Reactive Phosphorus (SRP, the readily bioavailable inorganic phosphate and hydrogen phosphate, plus some colloidal particulate P with a particle diameter of less than 0.45µm). For the purposes of estimating *CR* values of radioactive P, measurements of dissolved phosphorus or SRP were used, as these are estimates of the fractions that are readily available for bio-accumulation.

In the River Cam, P concentrations are relatively high: the average stable P from 2001 to 2005 at two sites (Bottisham Lock, Dimmocks Cote Road Bridge) was approximately 0.49 mg l^{-1} (S.E. 0.03 mg l^{-1}) (from data provided by Environment Agency; J.Titley, pers.

commun.). Note that this was measurement of phosphate (PO_4) and the concentrations are here presented as mg l^{-1} of P.

2.4.3. Phosphorus assimilation efficiency

Phosphorus assimilation at low trophic levels is expected to be very high, for example, ^{32}P assimilation efficiency of freshwater insect larvae (*Chaoborus trivittatus*) has been found to be approximately 80% (Giguere, 1981). The assimilation of radioactive phosphorus by fish is expected to be dependent on the availability of stable phosphorus in the diet. In an experiment in which fish (brown trout) were fed pellets containing ^{32}P, only a small fraction (0.2-0.4%) was incorporated into muscle (Winpenny et al., 1998). It may be that this low assimilation efficiency was due to a relatively high stable P concentration in the commercial feed pellet (8.3 mg g^{-1}). The report (Winpenny et al., 1998) cites an earlier study by Nelson (Nelson, 1961) which showed assimilation efficiencies approximately 25 times higher than this.

It is possible also to estimate assimilation efficiency on the basis of food ingestion and growth rates of fish, if the P content of feed can be estimated. For a 500g brown trout at a water temperature of 12^{o}C, equations given in Elliott (Elliott, 1975) estimate a growth rate of 1.65 g d^{-1} (fresh weight) at a maximum feeding rate of 11.8 g d^{-1} (fresh weight). Assuming the phosphorus content of the feed to be approximately half that of the trout (see Table 2), this implies a P assimilation efficiency of approximately 28%. This method of estimating assimilation efficiency will tend to under-estimate actual assimilation efficiencies as it accounts only for that fraction of assimilated stable P contributing to growth. The fraction which is assimilated and then subsequently excreted is not accounted for. Direct laboratory measurements of P assimilation efficiency (Nakashima and Leggett, 1980) have found a significantly higher assimilation efficiency of 72% (S.D. 7.2, N = 12) in yellow perch.

In view of the wide range in assimilation efficiencies measured, and of the potentially very high P assimilation efficiency (Nakashima and Leggett, 1980), we here conservatively

assume that all P ingested by the fish is assimilated (i.e. that the assimilation efficiency is 100%).

2.4.4. Uptake and excretion rates

A previous study by Smith (2006) presented a method for determining uptake and excretion rates of radionuclides in fish. Rates of uptake and excretion of radionuclides, k_{fn}, k_{bn} may be estimated using field and/or laboratory experiments, or using estimates of fish feeding rates and stable P content.

The rate of uptake of phosphorus to aquatic vegetation, algae, phyto- and zoo-plankton is relatively rapid, but highly variable between species. For freshwater phytoplankton, Seip and Reynolds (Seip and Reynolds, 1995) report population growth rates of 1.3-5.5 d^{-1} (population doubling times of 0.13-0.53 days) implying very rapid growth and hence very rapid assimilation and turnover of phosphorus. By inspection of Equations (2, 10), it can be seen that, if the rate of excretion of P is greater than the radioactive decay rate ($k_{b1} >> \lambda$), then the concentration factor of radioactive P is close to that of stable P. We have therefore here assumed a rapid excretion rate, k_{b1}, of 1 d^{-1} for the first trophic level leading (relative to the ^{32}P or ^{33}P physical decay rate) to effectively instantaneous equilibrium between water and the first trophic level. This may in some cases lead to over-estimation of radioactive P concentration factors in fish (for example, for fish feeding primarily on aquatic insects or slower-growing aquatic macrophytes), but it is an appropriate conservative approach at this stage.

For phosphorus uptake to fish via ingestion of food, we can estimate the uptake rate using the following equation (adapted from Smith (2006)):

$$\text{[illegible]} \quad \text{[illegible]} \quad \text{[illegible]} \quad \text{[illegible]} \qquad (15)$$

where D_{max} (g d^{-1}) is the maximum daily intake (wet weight) of food, 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).

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

$$D_{\max} = (4 \times 10^{-3}) A_D \times w^{b_1} \times \exp(b_3 T) \qquad (16)$$

where A_D, b_1 and b_3 are empirically determined constants whose values are given by Elliott (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 (16). The factor 10^{-3} converts D_{max} in mg per day estimated by the Elliot model to g per day used in Equation (15).

The excretion parameters for the second and third trophic levels, k_{b2}, k_{b3}, may be estimated using the relations between these parameters and the equilibrium concentration factors of <u>stable</u> P (Equations 13, 14 with λ =0):

$$\frac{CF(\textit{trophic level } 2)}{CF(\textit{trophic level } 1)} = \frac{C_2}{C_1} = \frac{k_{f2}}{k_{b2}} \qquad (17)$$

$$\frac{CF(\textit{trophic level } 3)}{CF(\textit{trophic level } 2)} = \frac{C_3}{C_2} = \frac{k_{f3}}{k_{b3}} \qquad (18)$$

Brown trout were chosen as a fish for which there are good data on feeding rates at different water temperatures and weights of fish. The species is common in England and Wales, though it tends to be found in rivers which are relatively unpolluted and unimpacted by river engineering. It may not, therefore, be found at all sites for which assessments of radiophosphorus uptake is required. For the purposes of this study, however, it needs only to serve to provide realistic growth rates for fish species typically found. Since the feeding rates

given here are maximum values, it is believed that these will be representative of other species of fish at the sizes (10 g for trophic level 2 and 500 g for trophic level 3) appropriate to this study.

2.4.5. Summary of model input parameters and assumptions

It is assumed that:

1. The phosphorus assimilation efficiency is 100%;
2. All of the radiophosphorus discharged to the river is in bioavailable forms and none is retarded by bed sediments;
3. The average weight of trophic level 2 and 3 fish is 10g and 500g, respectively;
4. Model output is predicted mean activity concentration in a given population of fish at a particular time;
5. Model output is presented as Bq kg^{-1} (f.w.) ^{32}P or ^{33}P in whole fish (i.e including bone and scales) or muscle tissue.

3. Results and Discussion

For the example scenario of a river with average dissolved P equal to 0.49 mg l^{-1} (the average for the River Cam), changes in ^{32}P and ^{33}P activity concentrations in aquatic biota at different trophic levels were calculated, assuming a constant 1 Bq l^{-1} activity concentration of each of these radionuclides in water. It was assumed that, at time zero, activity concentrations in water and aquatic organisms were equal to zero. Example model predictions for this scenario are shown in Figures 2 and 3 which assume average water temperatures of 12 (spring) and 17^{o}C (summer). Accumulation of radioactive phosphorus is more rapid at higher water temperatures leading to higher *CR* values at higher water temperature. The resulting ^{33}P *CR* is greater (at all water temperatures) than that of ^{32}P, owing to the slower radioactive decay rate of ^{33}P.

3.1. Recommended interim CR values for ^{32}P and ^{33}P

The IAEA (1994, 2010) recommended *CR* for radioactive P (ca. 50,000 l kg^{-1}), is likely to be a significant over-estimate for many aquatic systems and particularly for lowland rivers in England and Wales (and other rivers impacted by P from intensive agriculture and urban development). Firstly, stable P concentrations in lowland rivers of England and Wales tend to be significantly higher than the 0.03 mg l^{-1} assumed by Kahn and Turgeon (Kahn and Turgeon, 1984). A survey of English rivers draining to the North Sea in the mid- to late-1990's found annual average SRP concentrations to range from 0.06 to 1.8 mg l^{-1} with the range in lowland rivers being 0.14 – 1.8 mg l^{-1} (Neal and Robson, 2000); stable dissolved P in the River Cam averages around 0.49 mg l^{-1}. The Environment Agency phosphorus water quality target for lowland rivers on alluvial or clay catchments in England and Wales is an annual average of 0.10 mg l^{-1} (Mainstone and Parr, 2002). Secondly, as noted by Kahn and Turgeon (1984), *CR* values determined by using the stable isotope analogue need to be corrected to account for the decay of ^{32}P and ^{33}P during assimilation through the aquatic food chain.

Using the phosphorus uptake model, interim *CR* values for ^{32}P and ^{33}P were estimated as a function of bioavailable stable P in the river water. These *CR* values are presented in Figure 4. Predicted *CR* values for a stable P concentration of 0.49 mg l^{-1} are shown as a vertical dashed line and presented in Table 3. Using the River Cam as an example, the model predicts interim *CR* values (Trophic Level 3; muscle tissue) of 1500 l kg^{-1} for ^{32}P and 2200 l kg^{-1} for ^{33}P at an average water temperature of 17^{o}C. These estimates are more than one order of magnitude lower than the recommended value of *ca*. 50,000 l kg^{-1} (IAEA, 1994, 2010) for fish "edible portions".

3.2. Model comparison with available empirical data

Field data of radioactive phosphorus bioaccumulation are relatively sparse. Measurements were, however, made in both the Soviet Union and the U.S.A. from discharges of radioactive phosphorus created as an activation product in the cooling water of certain nuclear reactors. These data provide limited testing of the model.

Annual geometric mean *CR* values measured in whitefish in the Colombia River (Kahn and Turgeon, 1984) ranged between 3,000 and 300 l kg^{-1} though individual values at this site and

at other sites ranged between 100 and 12,000 l kg^{-1}. It is not known, however, whether the variation in values was influenced by relatively rapidly changing ^{32}P activity concentrations in water which can make *CR* estimates inaccurate. Kahn and Turgeon (Kahn and Turgeon, 1984) recommended an average *CR* for stable P of approximately 70,000 l kg^{-1} but noted that this would vary according to stable P availability in the water. They suggested that the ^{32}P concentration factor would be approximately one-twentieth of this value (3,500 l kg^{-1}), noting that "measured ^{32}P bio-accumulation factors are of this magnitude, but vary considerably". A further experimental study of phosphorus uptake in fish, Kahn et al. (1987) observed that "at a mean [stable] phosphorus bioaccumulation factor of 70,000 [l kg^{-1}, fish muscle] the factors for ^{32}P are 6,000 [bluegill] and 12,000 [catfish] respectively"

During operation of the Krasnoyarsk Mining and Chemical Industrial Complex (KMCIC), in Russia, significant quantities of ^{32}P were discharged to the Yenisei River. Table 4 presents measurements of ^{32}P in water and fish (roach, *Rutilus rutilus*) of the Yenisei River and an estimate of the mean *CR*. The Yenisei River is relatively low in stable P (though having large seasonal variations) leading to high bioaccumulation. A review by (Vakulovsky et al., 2004) has estimated the water-fish *CR* of stable P in the Yenisei River to be in the range $10^4 – 10^5$ l kg^{-1}, one- to two orders of magnitude greater than that measured for radioactive ^{32}P (Table 4).

The available field data, therefore, strongly support a significantly lower *CR* for radioactive phosphorus than that derived for stable phosphorus. However, tentative quantitative comparisons with these data suggest that the model may still significantly over-estimate uptake of radioactive phosphorus to fish in some aquatic systems. The stable P concentration in the Yenisei river varies seasonally between 0.01 – 0.5 mg l^{-1} (A.I. Kryshev, pers commun.). From Figure 4, the model predicts a *CR* of ^{32}P for this system in the region $4 \times 10^3 – 2 \times 10^5$ l kg^{-1} (whole fish) significantly higher than that estimated from measurements (1.2×10^3 l kg^{-1}, see Table 4). For the Colombia River, Kahn and Turgeon (1984) give a range in stable P in water of 0.01 – 0.05 mg l^{-1} which, from Figure 4, would give *CR* values in the range $1 \times 10^4 – 7 \times 10^4$ l kg^{-1} for ^{32}P (fish muscle). Again this is significantly higher than the range in reported values ($1 \times 10^3 – 1 \times 10^4$ l kg^{-1}) (Kahn and Turgeon, 1984). These over-estimates may be due to the conservative assumptions inherent in the model, in particular, the rate of uptake of stable and radioactive phosphorus through the food chain may have been over-estimated. However, without more precise empirical data on both radioactive

and stable phosphorus in the Yenisei and Colombia Rivers, it is difficult to draw a strong conclusion on this issue.

3.3. Phosphorus accumulation pathway

There is good evidence that the primary uptake pathway for P in fish is via food rather than direct uptake from water. Poston and Knopfler (Poston and Klopfer, 1986) state that "there was a strong indication from laboratory experiments that accumulation of ^{32}P from food was more significant than adsorption from the water column". Kahn and Turgeon (Kahn and Turgeon, 1984) estimated a stable P intake via the food pathway which was approximately 40,000 times higher than that via the water pathway. They observed a *CR* value of approximately 1 via the water pathway. A later experimental study (Kahn et al., 1987) found that phosphorus uptake was two orders of magnitude lower via the water pathway compared to the food pathway. Laboratory experiments measuring the assimilation of ^{32}P in brown trout also showed relatively little uptake direct from the water, in comparison with the food uptake pathway (Winpenny et al., 1998).

3.4. Radio-phosphorus accumulation in farmed fish

As discussed above, it seems clear that the accumulation of phosphorus occurs predominantly via food and that the water pathway is of minor importance. This implies that farmed fish, receiving a diet comprised of (presumably uncontaminated) fish pellets, would accumulate much less radioactive phosphorus than wild fish. Kahn and Turgeon (Kahn and Turgeon, 1984) suggest a *CR* via the water pathway of approximately 1.0 l kg^{-1}, several orders of magnitude lower than that via the food pathway. Here, we do not give a recommended *CR* for farmed fish as further research would be necessary before a recommendation could be made. Specifically, an assessment of food sources of farmed fish would need to be made to confirm that no significant proportion of wild food is ingested by farmed fish (many fish farms are in, or linked to, natural rivers or lakes).

In spite of these caveats, it can be concluded that accumulation factors for radioactive phosphorus in farmed fish are likely to be significantly lower than those for wild fish.

4. Conclusions

The model presented here has been used to estimate the *CR* value for radioactive phosphorus as a function of available stable P in the water body. In lowland rivers in England and Wales, and, likely, in many rivers worldwide, this results in significantly lower estimated *CR* values than the IAEA (1994, 2010) recommended values. For example, in the River Cam, the predicted *CR* value for radioactive phosphorus is approximately 5×10^3 l kg^{-1} (whole fish) or 2×10^3 l kg^{-1} (muscle tissue).

The current recommended *CR* of ^{32}P and ^{33}P of ca. 5×10^4 l kg^{-1} (IAEA, 1994, 2010) for fish "edible portions" (assumed to be primarily muscle tissue) is likely to be a significant over-estimate for many rivers. This is primarily a result of a conservative assumption inherent in this recommended value: that the stable phosphorus concentration of the aquatic system is relatively low. This is accounted for somewhat in the IAEA (1994) report since a large range in the recommended value is given: 3×10^3 - 1×10^5 l kg^{-1}. The range in values in IAEA (2010) (*ca.* $4 – 6 \times 10^4$ l kg^{-1}, corrected for ^{32}P decay) is, however, much too narrow.

The relative importance of food-chain uptake and direct uptake from water has been assessed in a literature review. It can be concluded that food-chain uptake is the dominant accumulation pathway in fish and hence accumulation factors for radioactive phosphorus in farmed fish are likely to be significantly lower than those for wild fish. Some further research is recommended to estimate suitable conservative *CR* values for farmed fish.

4.4. Scope and use of the model

It should be noted that, owing to the sparsity of field data on bioaccumulation of radioactive phosphorus in aquatic systems, the model could only be tested in a somewhat limited way. Comparisons with the available empirical data from the Yenisei and Colombia rivers support the general conclusion of a significantly lower *CR* for radioactive compared to stable phosphorus. The limited quantitative model test we have made here suggests the possibility that the model still over-estimates *CR* for radioactive phosphorus in fish. More field studies

of the bioaccumulation of radioactive phosphorus in fish would improve confidence in the model outputs.

Once fully validated, the model presented here can be used to assess bioaccumulation of ^{32}P and ^{33}P in fish in any river. The inputs required are the estimated or measured activity concentration in water, plus the concentration of bioavailable stable P in the water. It is important that an appropriate measure of bioavailable P be used (either "dissolved" P or "soluble reactive" P, SRP).

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Acknowledgements

We would like to thank Rob Allott, David Copplestone and John Titley for discussions during the course of this study, and for comments on this manuscript. We also acknowledge financial support of the Environment Agency of England and Wales.

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Figure 3 Predicted time changes in ^{33}P in biota of the River Cam: (a) 12^{o}C and (b) 17^{o}C water temperature.

Figure 4 Recommended interim values for (a) ^{32}P and (b) ^{33}P concentration ratio in different trophic levels as a function of bio-available stable P in the water (water temperature 17^{o}C). The *CR*s account for radioactive decay during the bioaccumulation process. The vertical dashed lines indicate the *CR* values for a dissolved P concentration of 0.49 mg l^{-1} (equal to the average for the River Cam).

Table 1. Stable P concentrations (dry weight basis) in some different aquatic organisms.

Organism	**Stable P conc. g kg^{-1} d.w.**	**Notes and references**
Macrophytes: *Carex riparia, Glyceria maxima, Sparganium erectum, Schoenoplectus lacustris*	2.0 – 8.4	House et al. (2001). R. Thame. Roots, shoots and rhizomes sampled. P (as TP) in river water varied from 400-1990 ug/l.
Algae (various species)	0.5 – 33	Hecky and Kilham (1988)
Zooplankton: *Acanthodiaptomus, Heterocope, Holopedium, Bosmina, Diaphanosoma, Daphnia*	4 – 18	Andersen and Hessen (1991). Field measurements in a lake.
Zooplankton: *Daphnia magna*	4.0-6.4	Boersma and Kreutzer (2002). Experiment in which *Daphnia* were fed with low and high P content food. Low end of range corresponds to low P food, high end to high P food.
Fish: minnows: *Phoxinus eos, Phoxinus neogaeus, Margariscus margarita, Pimphales promelas.*	14.9 ± 4.5 (1 S.D.)	Sterner and George (2000). Small (length approx. 20-100 mm) non-piscivorous fish. Whole fish analysed.
Two temperate species: roach *Rutilus rutilus* perch *Perca fluviatilis*	21.1 – 42.8	Dantas and Attayde (2007).
Six tropical species: Nile tilapia *Oreochromis niloticus,* tucunaré *Cichla monoculus,* pescada *Plagioscion squamosissimus,* pirambeba *Serrasalmus rhombeus,* curimatã *Prochilodus brevis,* traíra *Hoplias malabaricus*	Mean for all species: 30.5 ± 8.2	Length 31-370 mm; wet weight approx. from a few g up to 600g. Bonier species tended to have highest P concentration. Whole fish. d.w./f.w. ratio 0.24-0.32.
Temperate species	24.5	Tanner et al. (2000), reported in Dantas and Attayde (2007)

Table 2. Assumed stable P concentrations for each trophic level.

Trophic level	**Assumed stable P concentration in biota g kg^{-1} d.w. (c.f. Table 1)**	**f.w./d.w. ratio**	**Stable P conc. in biota g kg^{-1} f.w.**
1 Plants, plankton, insects	10	5	2
2 Small non- pred. fish	15	3	5
3 Large pred. fish			
Muscle tissue			2.2*
Whole fish	30	3	10

*(Kahn and Turgeon, 1984)

Table 3. Estimated *CR* values for a concentration of dissolved P in water of 0.49 mg l^{-1} (equal to the average for the River Cam) at different water temperatures.

Trophic level, TL	**Assumed stable P concentration factor in biota (l kg^{-1})**	**Estimated *CR* for ^{32}P l kg^{-1}**	**Estimated *CR* for ^{33}P l kg^{-1}**
	Water Temp 7 ^{o}C		
TL 1	4100	3900	4000
TL 2 (whole fish)	10000	1700	2700
TL 3 (muscle)	4500	240	540
TL 3 (whole fish)	20000	320	840
	Water Temp 12 ^{o}C		
TL 1	4100	3900	4000
TL 2 (whole fish)	10000	3200	4600
TL 3 (muscle)	4500	740	1300
TL 3 (whole fish)	20000	1300	2800
	Water Temp 17 ^{o}C		
TL 1	4100	3900	4000
TL 2 (whole fish)	10000	4800	6300
TL 3 (muscle)	4500	1500	2200
TL 3 (whole fish)	20000	3200	5800

Table 4. Measurements of ^{32}P in water and fish (roach) of the Yenisei River at different distances[1] downstream of KMCIC, August 1991. From data in Vakulovsky et al. (2004).

Distance (km)	**^{32}P in water Bq l^{-1}**	**Distance (km)**	**^{32}P in roach Bq kg^{-1}**	**Estimated mean *CR*, l kg^{-1}**
180	2.2	200	2900	
250	1.9	260	2135	
290	1.1	340	1100	
Mean	**1.73**		**2045**	**1180**

Notes: [1] Measurements obtained within 100 km of the discharge point were not used as the discharge was not yet fully mixed across the river.