

NATIONAL DOSE ASSESSMENT WORKING GROUP

PAPER 14-05 MODELLING THE BIO-ACCUMULATION OF ^{32}P AND ^{33}P IN FRESHWATER SYSTEMS

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1 Introduction

Radioactive isotopes of phosphorus may be discharged to freshwater systems, under authorisation, by a variety of non-nuclear establishments. The consumption of fish in river environments is a potential pathway for the transfer of radioactive phosphorus to consumers. The accumulation by fish of radioisotopes of phosphorus (^{32}P and ^{33}P) is commonly modelled using an equilibrium concentration factor (*CF*) approach. This approach, however, may significantly over-estimate accumulation of radio-phosphorus in fish for the following reasons:

1. *CF* values for radio-phosphorus are commonly estimated using the *CF* of stable P. This approach tends to over-estimate radio-phosphorus activity concentrations in fish because it does not account for radioactive decay as (relatively short-lived, see Table 1) radio-phosphorus bio-accumulates through the aquatic food chain and within the fish;
2. Conservatively, current *CF* values for radio-phosphorus may be up to 1×10^5 l kg^{-1} : the IAEA (1994) recommended value is 5×10^4 l kg^{-1} . It is likely that these conservative estimates are for oligotrophic (i.e. low stable P) systems in which bioaccumulation of P is particularly high. Most lowland rivers in England and Wales have elevated P concentrations from anthropogenic sources (primarily diffuse agricultural runoff and point sources from sewage treatment works), hence bioaccumulation of stable P is expected to be lower than the IAEA recommended values;
3. A fraction of radio-phosphorus discharged into rivers may be absorbed to suspended and bed sediments which delays bioaccumulation and hence, by radioactive decay, reduces uptake to fish;
4. Application of literature *CF* values is likely to significantly over-estimate uptake of radio-phosphorus to farmed fish since farmed fish tend to accumulate much less radioactivity via the food chain than wild fish (e.g. Smith et al. 2001).

The purpose of this study is to evaluate current approaches to predicting the accumulation of radioactive phosphorus isotopes in wild and farmed fish. Current estimates of *CF* values will be assessed by both a critical review of the literature and using a dynamic model simulating the transfer of phosphorus through the aquatic food chain. Recommendations will be made for further modelling work to better evaluate uncertainty as well as further field studies for model validation.

Table 1 Physical half-lives and decay constants of ^{32}P and ^{33}P

Radionuclide	Half life, $T_{1/2}$	Decay constant, λ
P-32	14.262 days	0.0486 d ⁻¹
P-33	25.34 days	0.0274 d ⁻¹

Current recommended value of the radiophosphorus-water CF

The IAEA recommended value (IAEA 1994) for the radiophosphorus concentration factor is $5 \times 10^4 \text{ l kg}^{-1}$, with a range $3 \times 10^3 - 1 \times 10^5 \text{ l kg}^{-1}$. This 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 Knopfler, 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 *CF* is obtained (using Equation 2, below) from a review of phosphorus concentrations in fish muscle (assumed value 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 *CF* 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 et al. (1986) based on the earlier Kahn and Turgeon (1980) study.

2 Methods

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

$$CF = \frac{\text{Activity concentration per kg of fish (wet wt)}}{\text{Activity concentration per litre of water}} \quad \text{l kg}^{-1} \quad (1)$$

A literature review has been carried out to critically assess the available information on the bioaccumulation of both stable and radioactive phosphorus. The *CF* assessment and models will use the River Cam at Cambridge as a case study, but generalised interim *CF* values will be determined for typical stable P concentrations in lowland rivers in England and Wales (based on stable P uptake).

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:

$$CF \approx \frac{\text{mass of stable isotope per kg of fish (w.w)}}{\text{mass of (available) stable isotope per l of water}} \quad (2)$$

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.

Freshwater food chains (often called food webs because of their many interconnections) are complex and include detritus, bacteria, phyto- and zoo-plankton, algae,

macrophytes, macroinvertebrates, herbivorous, omnivorous and predatory fish of various sizes. The transfer of radionuclides to fish may therefore take many different pathways. For the purposes of this study, it is important to consider the food chain because it retards the uptake of radioactive phosphorus to fish and thus (by physical decay) reduces radioactivity concentrations in fish compared to what would be expected if the uptake was instantaneous.

Modelling

Uptake of radio-phosphorus through the aquatic food chain was modelled using a “box” model to predict the dynamics of transfer from water to the first (here defined as macrophytes, phytoplankton, invertebrates), second (here defined as small, non-predatory fish) and third (here defined as larger predatory fish) trophic levels. A simplified model of the aquatic food chain is used for this assessment.

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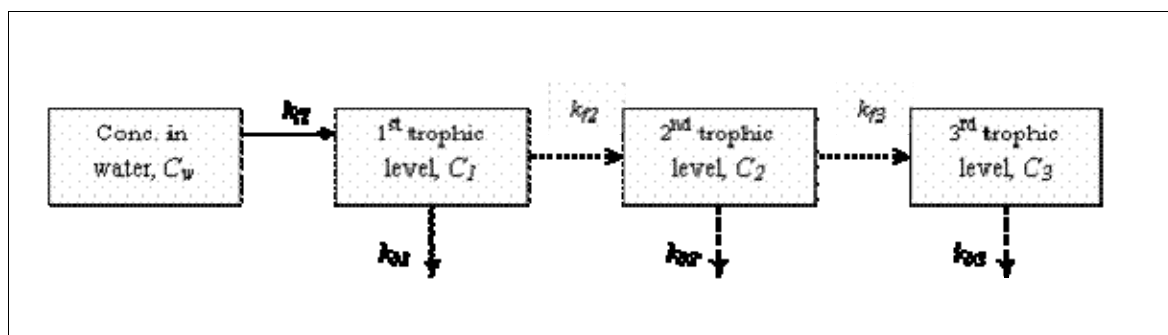


Figure 1 Illustration of phosphorus uptake model.

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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_{fn} has dimensions of $[\text{length}^3 \text{ mass}^{-1} \text{ time}^{-1}]$, but all other rate constants have dimensions of $[\text{time}^{-1}]$.

The transfer of phosphorus between the different trophic levels was modelled as a series of first order differential equations. These were solved both analytically and (as a check) numerically using the finite-difference method.

Determination of model parameter values

A previous study by Smith and co-workers (Smith et al. 2002) 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.

A literature review was conducted to obtain conservative values of the stable P concentration in biota in the three trophic levels (Table 3), and values of the stable phosphorus assimilation efficiency. In view of the wide range in assimilation efficiencies measured, and of the potentially very high P assimilation efficiency (Nakashima and Leggett 1980), we will conservatively assume that all P ingested by the fish is assimilated (i.e. that the assimilation efficiency is 100%).

Table 2 Assumed stable P concentrations for each trophic level.

Trophic level	Assumed stable P concentration in biota g kg^{-1} (c.f. Table 2)	f.w./d.w. in ratio d.w.	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	30	3	10

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 (1995) report population growth rates of $1.3\text{-}5.5 \text{ d}^{-1}$ (population doubling times of 0.13-0.53 days) implying very rapid growth and hence very rapid assimilation and turnover of phosphorus. It was therefore assumed that there was 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). It also under-emphasises the important role detritus plays in the food chain of many river systems. 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).

Rates of uptake of phosphorus to fish were estimated using the rate of intake of food (from the first trophic level). 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. The empirical model of Elliot (1975) was used to calculate the feeding rate for fish of different wet weight w (grammes) at different water temperatures. Based on the concentration of stable P in fish and in their food, rates of uptake of phosphorus were calculated from the food intake rates.

Summary of model input parameters and assumptions

It is assumed that:

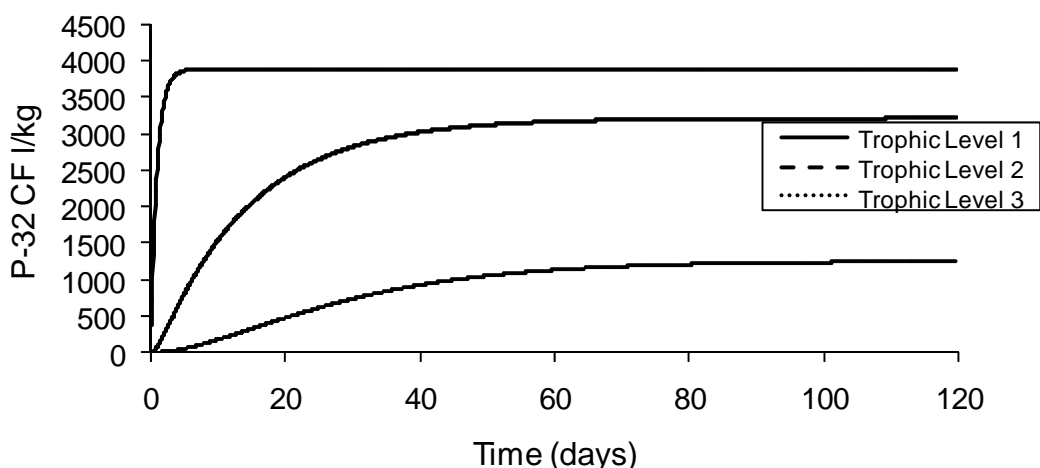
1. The phosphorus assimilation efficiency is 100%;
2. All of the radiophosphorus discharged to the river is in available forms and none is retarded by bed sediments;
3. The average weight of trophic level 2 and 3 fish is 10g and 500g, respectively;
4. Model output is predicted mean activity concentration 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.

3 Results

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 Figure 2 which assumes an average water temperature of 12°C . Accumulation of radioactive phosphorus is more rapid at

higher water temperatures leading to higher *CF* values at higher water temperature. P-33 accumulation is greater than that of ³²P, owing to the slower radioactive decay rate of ³³P.

(a) Predicted P-32 *CF* for biota of the River Cam for a constant water concentration of 1 Bq/l, water temperature 12°C



(b) Predicted P-33 *CF* for biota of the River Cam for a constant water concentration of 1 Bq/l, water temperature 12°C

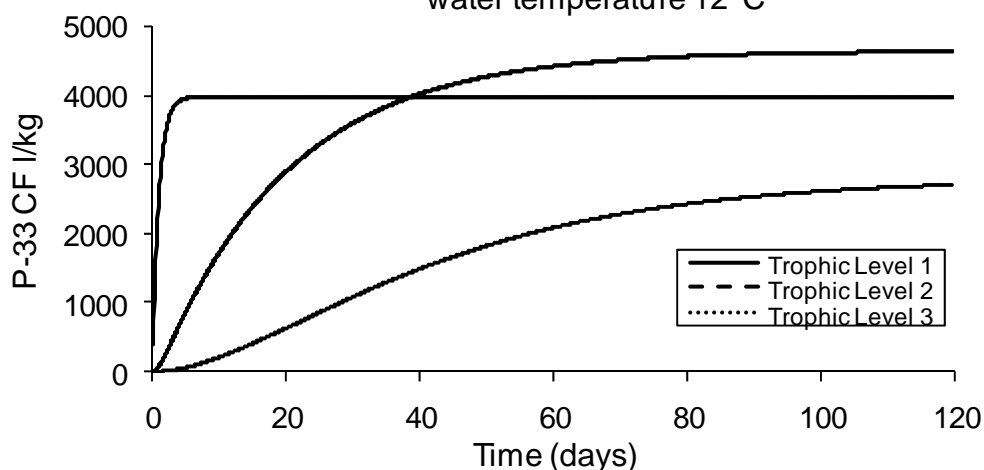


Figure 2 Predicted time changes in radioactive phosphorus in biota at a stable P concentration of 0.49 mg l⁻¹ (equivalent to that of the River Cam): (a) P-32 (b) P-33.

Recommended interim CF values for ³²P and ³³P

Using the phosphorus uptake model, interim *CF* values for ³²P and ³³P were estimated as a function of bioavailable stable P in the river water. These *CF* values are presented in Figures 3 and 4. Predicted *CF* values for a stable P concentration of 0.49 mg l⁻¹ (equal to the average dissolved P in water of the River Cam) are shown as a vertical dashed line.

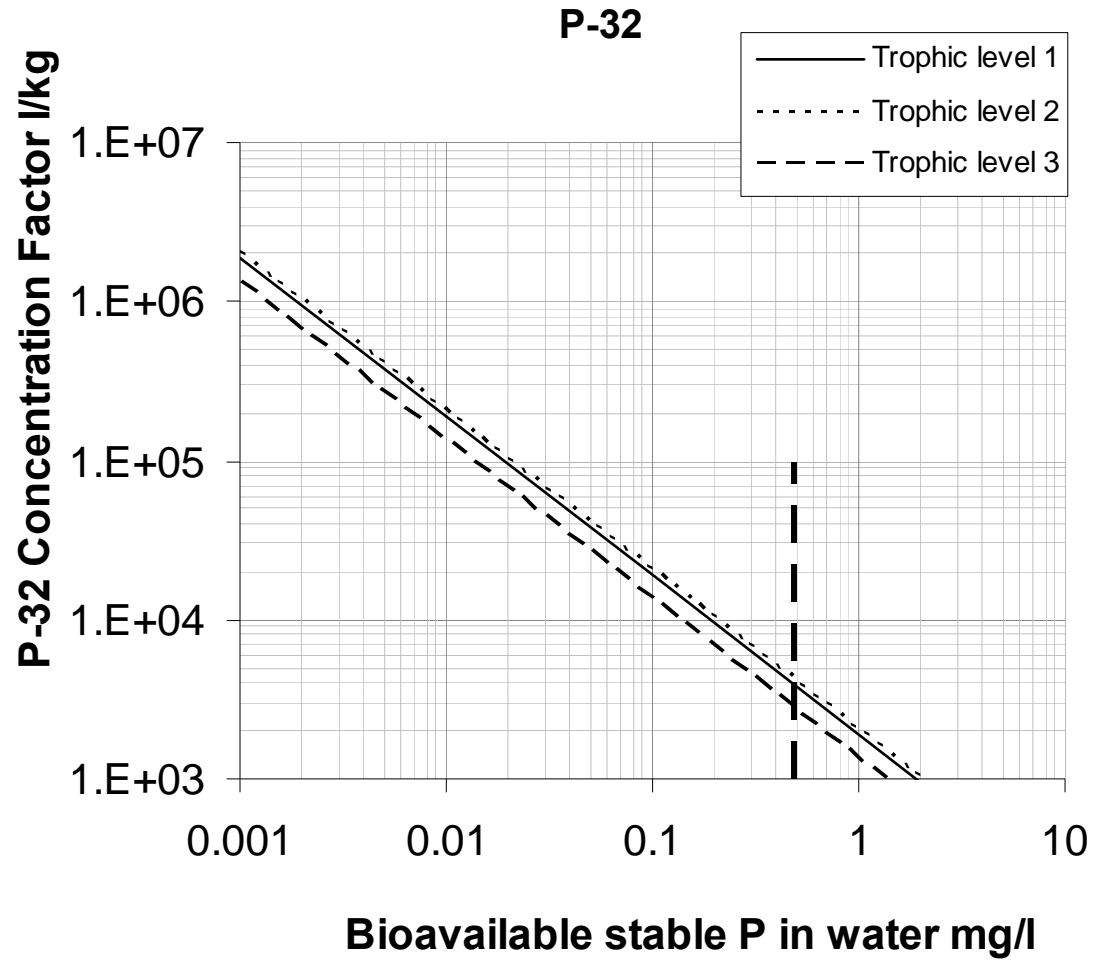


Figure 3 Recommended interim values for P-32 concentration factor in different trophic levels as a function of bio-available stable P in the water (water temperature 17°C). The *CFs* account for radioactive decay during the bioaccumulation process. The vertical dashed lines indicate the *CF* values for a dissolved P concentration of 0.49 mg l⁻¹ (equal to the average for the River Cam).

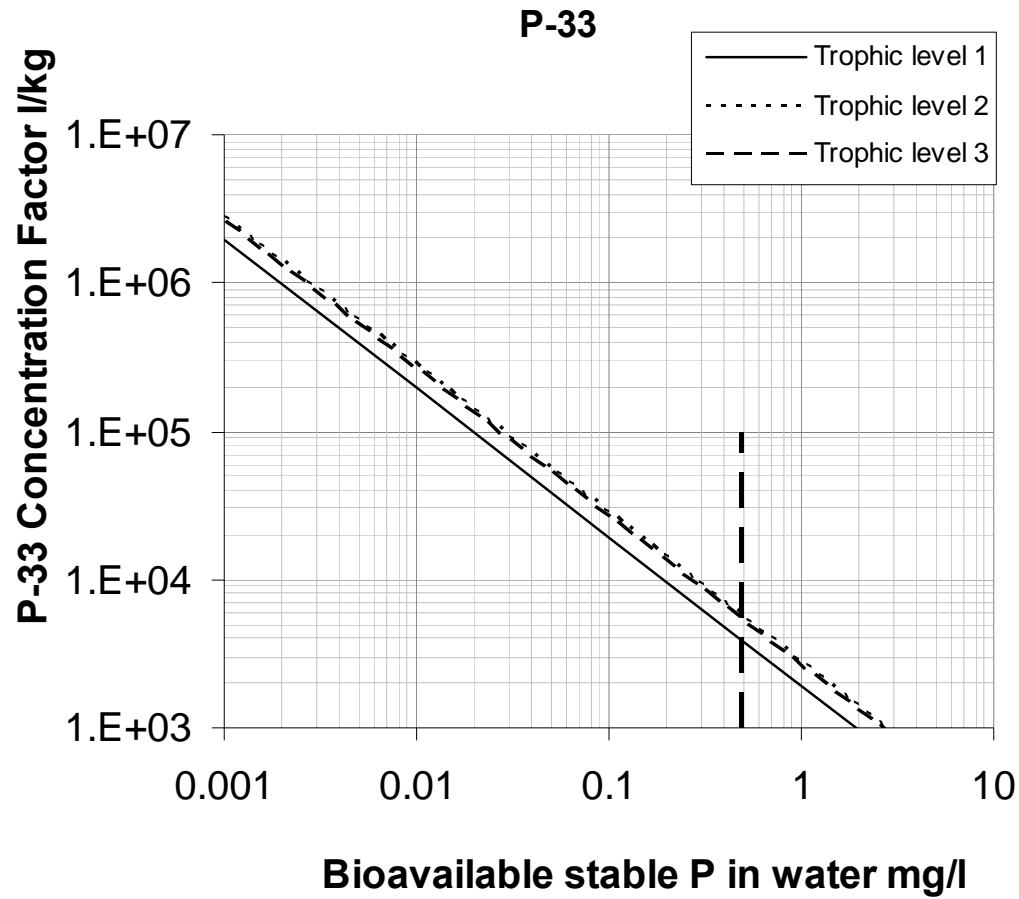


Figure Error! No text of specified style in document. **Recommended interim values for P-33 concentration factor in different trophic levels as a function of bio-available stable P in the water (water temperature 17°C). The CFs account for radioactive decay**

during the bioaccumulation process. The vertical dashed lines indicate the *CF* values for a dissolved P concentration of 0.49 mg l^{-1} (equal to the average for the River Cam).

4 Conclusions and recommendations

The IAEA (1994) recommended CF for radioactive P ($50,000 \text{ l kg}^{-1}$), which is of the same order as that recommended by Kahn and Turgeon (1984) for stable P ($70,000 \text{ l kg}^{-1}$), is likely to be a significant over-estimate for many aquatic systems and particularly for lowland rivers in England and Wales. 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 (1984): for example, stable dissolved P in the River Cam averages around 0.49 mg l^{-1} . Secondly, CF 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. These factors, to a large extent, account for the significantly lower CFs typically observed for radioactive P than that recommended in the IAEA (1994) review.

Using models for the transfer of phosphorus through the aquatic food chain, recommended interim CF values for radioactive forms of P were estimated as a function of (bioavailable) stable phosphorus in the water body. The models predicted significantly lower CF values compared to those recommended in IAEA (1994). For the example of the River Cam, at an average water temperature of 17°C , the model predicts interim CF values for ^{32}P in the range $2800\text{-}3900 \text{ l kg}^{-1}$ (depending on trophic level) and for ^{33}P in the range $4000\text{-}5300 \text{ l kg}^{-1}$. These estimates are approximately one order of magnitude lower than the recommended value of $50,000 \text{ l kg}^{-1}$ (IAEA, 1994).

It is recommended that this model be validated, where possible, by field measurements. Within the scope of this project it has not been possible to assess model uncertainty though, where appropriate, conservative assumptions have been made. It is further recommended that a model uncertainty analysis be carried out.

It is predicted that, assuming a constant input of radiophosphorus to the river, the highest activity concentrations of P-32 and P-33 in aquatic biota would be observed during the summer and early autumn. This is due firstly to the generally lower rates of dilution during the summer low flow period, and, secondly, to the higher rates of uptake of phosphorus during the warmer period.

Scope and use of the model

Once validated, the model presented here can be used to assess bioaccumulation of P-32 and P-33 in fish in all rivers in England and Wales. 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). If the measurement of stable P in water included some biologically unavailable P on suspended sediment, the bio-available stable P in the water would be over-estimated and hence bioaccumulation would be under-estimated.

5 References

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