Development and Evaluation of a Regression-Based Model to Predict Cesium-137 Concentration Ratios for Saltwater Fish

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ABSTRACT

Data from published studies and World Wide Web sources were combined to develop a regression model to predict ^{137}Cs concentration ratios for saltwater fish. Predictions were developed from 1) numeric trophic levels computed primarily from random resampling of known food items and 2) K concentrations in the saltwater for 65 samplings from 41 different species from both the Atlantic and Pacific Oceans. A number of different models were initially developed and evaluated for accuracy which was assessed as the ratios of independently measured concentration ratios to those predicted by the model. In contrast to freshwater systems, were K concentrations are highly variable and are an important factor in affecting fish concentration ratios, the less variable K concentrations in saltwater were relatively unimportant in affecting concentration ratios. As a result, the simplest model, which used only trophic level as a predictor, had comparable accuracies to more complex models that also included K concentrations. A test of model accuracy involving comparisons of 56 published concentration ratios from 51 species of marine fish to those predicted by the model indicated that 52 of the predicted concentration ratios were within a factor of 2 of the observed concentration ratios.

1. Introduction

The ratio of the mean concentration of a radionuclide in a fish to its mean concentration in the water, when measured under equilibrium conditions, is an important parameter used to assess the bioavailability of radionuclides such as ^{134}Cs and ^{137}Cs in aquatic environments. This ratio may alternatively be termed the concentration ratio (hereafter, C_r), the concentration factor, or bioaccumulation factor and has units of L kg^{-1}. Numerous compilations of previously observed C_r for Cs isotopes have been developed (*e.g.*, Vanderploeg et al., 1975; Blaylock, 1982; Fesenko et al., 2011; Yankovich et al., 2012; Psaltaki et al., 2013; Tagami and Uchida, 2013) for use in accident assessments, and models have also been developed to predict C_r for Cs isotopes in fish using aspects of fish biology (*e.g.*, diet) and water quality parameters such as K concentrations (*e.g.*, Rowan and Rasmussen, 1994), Maximum Entropy Methods of analyses of previously compiled C_r (Hosseini et al., 2008), and Residual Maximum Likelihood extrapolations of known C_r among similar species (Beresford et al., 2015).

Rowan and Rasmussen (1994) developed a predictive model applicable to both freshwater and saltwater systems that has been shown to predict C_r for ^{137}Cs within a factor of 2 for a majority of cases (Smith et al., 2000). The model is based on 1) a classification of fish as either piscivorous or nonpiscivorous and 2) measures of K and suspended sediment concentrations in the water column. The model predicts 1) greater C_r for piscivorous fish, 2) smaller C_r in waters with greater K concentrations and 3) smaller C_r in waters with greater suspended sediment concentrations. This model is most appropriately applied to predict C_r when equilibrium conditions exist between the fish and the water.

Recently, an alternative form of the Rowan and Rasmussen (1994) model (hereafter referred to as the "freshwater fish model") was developed and evaluated (Pinder et al., 2014) that predicts C_r for ^{137}Cs in freshwater fish using the data from Rowan and Rasmussen (1994) but replaces their nonpiscivorous and piscivorous classification with a species-specific, numerical trophic level (hereafter, TL) obtained from the online database *fishbase.org* (Froese and Pauly, 2011). Information compiled in this database on fish diets and food items are used to compute numeric estimates of mean ($\pm$ Standard Error, hereafter SE) TL. These TLs range from 2, which indicates a herbivorous diet, through 3, which indicates a primarily carnivorous diet composed of herbivorous species, and to 4 and above that indicates a diet composed primarily of other carnivorous species. An advantage of this alternative, model using a continuous range of TL is that it predicts a separate C_r for each species rather than predictions for only the two discrete groups of piscivorous species and nonpiscivorous species.

This alternative, predictive model considered only freshwater fish. It did not include saltwater species, and it is the purpose of this analysis is to 1) extend the approaches used in Pinder et al., (2014) to develop a predictive model of C_r for saltwater species (hereafter termed the saltwater fish model) and 2) to test this model using two sources of independent data. The first source involves published data not incorporated into the Rowan and Rasmussen (1994) analysis. The second source involves those data that are appropriate for estimating C_r from the releases of Cs isotopes at the Fukushima Daiichi Nuclear Power Plant (hereafter, FDNPP). An ability to predict C_r for saltwater species based on their TLs may become a useful asset as the FDNPP ^{137}Cs releases continue to be dispersed across the northern Pacific Ocean (Buesseler et al., 2012; Kamenik et al., 2013; Otosaka and Kato 2014; Povinec et al., 2013; Kawamura et al., 2014; Ramzaev et al., 2014).

Instead of incorporating freshwater and saltwater fish into a common model, a separate model was developed for saltwater fish. The use of separate, independent models for fresh and salt waters was suggested by the differing relative ranges of variation in the important predictor variables of TL and K concentrations between fresh and salt water. The TLs of *fishbase.org* range over a factor of approximately two from somewhat > 2 to somewhat > 4 in both fresh and salt waters. In contrast, the ranges of maximum to minimum K concentrations in these environs range over factors of approximately 1.2 for the ocean (*i.e*., from 8783 to 10058 µM L^{-1}; Rowan and Rasmussen, 1994) but over a factor of approximately 30 in freshwaters (8 to 249 µM L^{-1}; Rowan and Rasmussen, 1994). These relative ranges suggest that K concentrations may be a more important predictor of C_r in fresh water but that TL may be a more important predictor of C_r in salt water.

2. Materials and methods

Separate data sources were employed in model development and model evaluation.

2.1. Model development

Four data sources were employed in model development including: 1) the TL estimates from *fishbase.org*; 2) C_r data from Rowan and Rasmussen (1994) for saltwater fish; 3) concentrations of K in salt waters from Rowan and Rasmussen (1994); and 4) assessments of fish species as being primarily pelagic species or primarily demersal species in *fishbase.org*

2.1.1. TL data

The TL estimates were obtained from *fishbase.org* where data on fish biology and ecology have been compiled for > 30,000 species from > 45,000 references. Several alternative methods are used in *fishbase.org* to estimate a mean $\pm$ SE TL depending on the type of available data. Where only lists of the food items consumed are available, the TL is estimated using a randomized resampling of those items to produce a mean $\pm$ a SE TL (see Pinder et al., 2014 and *fishbase.org* for details of this resampling process). In those cases where data are available on the proportions of food items consumed, an additional estimate of the TL is also computed using these proportions. For both methods, the TL of the fish is computed as 1 plus the mean TL computed for its diet (*i.e*., a fish whose diet has a mean TL of 2.5 would have a TL of 3.5). These estimations of TLs have been shown to agree with those computed from stable isotopic ratios (Kline and Pauly, 1988; Vander Zanden et al., 1997). Where data are lacking on diets, a fish's TL is inferred from taxonomically related species of a similar size. Because TL estimates from the random resampling procedures were available for the majority of the species involved in this study, they have been used in the models to predict most C_r. Where random resampling estimates were not available, the taxonomically related estimates were used instead.

2.1.2. The ^{137}Cs data for fish and their environment

The development of the predictive model was based on the ^{137}Cs data for saltwater fishes compiled by Rowan and Rasmussen (1994; Table 1) which included: 1) the fish's scientific name; 2) the 0 or 1 nonpiscivorous or piscivorous classification; 3) the location of the study; 4) the K concentration in the water (µM K L^{-1}); 5) the ^{137}Cs concentration (mBq L^{-1}) in the water; and 6) the wet mass ^{137}Cs concentration (Bq kg^{-1}) in either the fish's whole body or its muscle. The K concentrations in saltwater reported by Rowan and Rasmussen (1994) were interpolated from salinities using the conversion factor of 283.3 µM K L^{-1} per g $kg^{-1}$of salinity (Broecker and Peng, 1982). The Rowan and Rasmussen (1994) study included 71 measures of C_r for 42 species from 18 open ocean locations which included seas such as the Irish Sea, the North Sea and the Sea of Japan which are open to flow through circulation of ocean waters. Data from the more enclosed Baltic Sea and the Gulf of California were not used because of known variation in K concentrations in gradients of fresh waters to salt waters in the Baltic Sea and the possibility for similar gradients in other similarly enclosed waters. In the estimation of C_r from the data of Rowan and Rasmussen (1994) it was assumed that the concentrations in fish and water were at or near equilibrium, and this assumption of equilibrium is reasonable because the principal source of the ^{137}Cs in the oceans was global fallout from past weapons testing.

Concentration ratios were computed from these data as the ratio of concentrations in fish to those in water did not (and in some cases could not) differentiate between concentrations for whole fish and those for only muscle tissue. As a consequence, no distinctions with regard to muscle versus whole-body concentrations have been made in the use of these data to predict C_r in fish.

2.1.3. Classification of fish species as either demersal or pelagic species

Fish species were classified as either pelagic or demersal species based on descriptions in *fishbase.org*. Although the classification of species as pelagic or demersal may be a subjective classification, demersal species 1) have been documented to accumulate greater ^{137}Cs concentrations than co-occurring pelagic species (Rowan et al., 1998), 2) have sometimes had their ^{137}Cs concentrations ratios underestimated by predictive models (*e.g*., Pinder et al., 2014). Moreover, it has been suggested that rates of decline of ^{137}Cs concentrations for demersal fishes at Fukushima may be slower than that for pelagic fishes and other components of the marine biota (Tateda et al., 2013; Wada et al., 2013). For the purposes of this analysis, demersal species included both benthic species, which live in contact with the bottom, and benthopelagic species which live in the water column near to, or sometimes in contact with, the bottom.

2.1.4. Development of the predictive models

Preliminary predictive models were developed using simple and multiple regression procedures (Draper and Smith, 1981) that related concentration ratios to the variables of TLs and K concentrations as well as the interactions of these variables. Regression procedures may be used to 1) determine which independent variables from a set of preferably uncorrelated variables have important relationships with the dependent variable (*e.g*., Rowan and Rasmussen, 1994), 2) to estimate parameter values such as uptake and loss rates (*e.g*., Smith et al., 2002), or 3) as in the case of this study, to simply construct a predictive model (in the sense of Pedhazur, 1997)that relates the values of some predictor variables to the value of an important criterion variable (*e.g.*, Pinder et al., 2014).

Because prediction, and not explanation, is the objective in this modeling, the predictor variables only need to be correlated with the criterion variable, and this correlation may be due to the predictor variable is strongly correlated with a more difficult to measure or expensive to obtain causative agent. The validity of a predictive model is not evaluated in the use of its independent variables but in its ability to accurately predict the criterion variable in an independent data set (Pedhazur, 1997; Pinder et al., 2014).

The regression models in this study used the logarithm of the C_r as the criterion variable, but the predictive equations will be expressed in arithmetic forms as

$$\text{Predicted } C_r = 10^{[(b_0 + 0.5*EMS) + b_i*X_i + \ldots]} \quad (1)$$

where b_0 = the intercept of the regression equation, b_i = the regression coefficient for the *ith* predictor variable and EMS = the Error Mean Square of the regression analysis of variance. The value, 0.5*EMS, is added to b_0 in Eq. 1 to compensate for a bias after logarithmic transformation that would result in under prediction of the C_r (Beauchamp and Olson, 1973).

2.2. Assessing the accuracy of the models using regression statistics, observed-to-fitted or observed-to-predicted ratios

The accuracy of a regression model maybe assessed using; 1) regression statistics such as the coefficient of determination, R^2, which is the proportion of the variance in the criterion variable explained by the predictor variables or 2) the adjusted R^2 (Draper and Smith, 1981; Eq. 2.6.11.b; hereafter aR^2) which is R^2 adjusted for the number of predictor variables. including the intercept b_0. In contrast to the R^2, aR^2 only increases if the added variable makes a unique contribution to the model's predictability. Alternatively, accuracy for a predictive model may be more rigorously assessed using analyses of the frequency distributions of the ratios of independently measured values of the criterion to those predicted by the model. Such analyses may include the median ratio, the range of the ratios, or the inter-quartile range of the ratios (*i.e.*, the range from the 25th percentile largest ratio, hereafter termed Q_1, to the 75th percentile largest ratio, hereafter, Q_3). Analyses of observed values-to-those fitted by the regression or of independently observed values-to-those predicted by the model may prove to be more informative in cases where errors of under prediction (*i.e*., observed values greater than predicted values and ratios > 1) and over prediction (*i.e*., observed values less than predicted values and ratios < 1) are not equally acceptable. This may be the case for predicting concentration ratios that may subsequently be used to estimate radiation exposures. In developing and initially evaluating the models in this study, statistical significance levels, R^2, aR^2 and observed-to-fitted ratios are reported, but the primary assessments of model accuracies will be made using the ratios of independently measured C_r to those predicted by the model.

In using observed-to-predicted or observed-to-fitted ratios in evaluating models, two separate aspects of evaluation may be considered. The first can be termed accuracy and is measured as the proportion of the ratios that indicate the observed C_r is within some limits of the predicted C_r such as within a factor of 2 (*i.e*., observed-to-predicted ratios from 0.5 to 2) or 3 *(i.e*., observed-to-predicted ratios of 0.33 to 3). Because under prediction of the C_r, with resulting observed-to-predicted or observed-to-fitted ratios greater than 1, may incur greater consequences than over prediction, the second aspect of the evaluation can be termed conservatism which is expressed as the proportion of observed-to-fitted or observed-to-predicted ratios that are less than some upper limit such as 2 or 3.

What constitutes acceptable standards of accuracy or conservatism is a decision for those employing the model, but for the purposes of discussion within this analysis, an acceptable level of accuracy will be defined as $\geq$ 80 % of the observed-to-fitted or observed-to-predicted ratios being within factors of 2 or 3. An acceptable level of conservatism will be defined as $\geq$ 90 % of the ratios being < 2 or < 3.

Whether these levels of accuracy and conservatism are sufficient is a user-driven decision, but obtaining levels of accuracy within factors of 2 and conservatisms < 3 using the predictive model approach may be limited by several sources of biological variation and statistical errors. The sources of biological variation may include 1) uncontrolled biological effects such as seasonal differences in fish concentrations, 2) methodological differences such as whole-body versus muscle-only concentrations, 3) size or age dependent variations in fish concentrations (Smith et al., 2002), and 4) among site variations in fish community compositions (*e.g.*, Rasmussen et al., 1990) that can cause a species TL to vary among sites. The statistical sources of errors may include sampling errors in measuring 1) the predictor variables or 2) the Cs concentrations in fish or water that results in sampling errors for measuring C_r. For the purpose of this analysis, models have been constructed assuming that concentrations ratios were measured without error, which is clearly not the case.

2.3. Assessing model accuracy using independent measures of concentration ratios

Two data sets were used to evaluate model accuracies. The first of these were published measures of C_r for saltwater species that were not included in the compilation of Rowan and Rasmussen (1994). For the second set, temporal non-linear regression models were fitted to measures of ^{137}Cs concentrations in fish and water from the coastal waters of Fukushima Prefecture following the tsunami and the release of radionuclides from the FDNPP. As is described below, these models provide estimates of an uptake and a loss rate parameter for fish species, and the ratio of these two parameters provides an estimate of the equilibrium C_r. This model has been commonly employed to describe the dynamics of radionuclides in fish and other biota in aquatic environs (*e.g.*, Smith et al., 2002; Pinder et al., 2011; Martinez et al., 2014).

2.4. Assessing the accuracy of Model I using C_r from published results.

Data from six published studies from both the Atlantic and Pacific oceans were used in testing Model I. These test data were either 1) published after the Rowan and Rasmussen (1994) publication or 2) were published before the Rowan and Rasmussen (1994) study but not included in their analyses (*i.e*., the ^{133}Cs data of Suzuki et al., 1973). The six studies included Suzuki et al. (1973), Cuntha et al. (1993), Tateda and Koyanagi (1996), Kasamatsu and Ishikawa (1997), Godoy et al. (2003), Hong et al., (2011), and Antovic and Antovic (2011). Where multiple measures of observed C_r were reported for a species within a study, either 1) the mean C_r reported by the authors was used to test the model or 2) the median of the multiple measures was used to test the model.

2.5. Assessing model accuracy using data from the Fukushima releases

A simple, temporal model of uptake and loss was fitted to publically available data for ^{137}Cs concentrations in water and fish from FDNPP releases for the coastal region of Fukushima Prefecture for the first 750 days following the tsunami. The model (Eq. 2) has the form

$$\frac{\mathrm{F(t)}}{\mathrm{dt}} = \mu * \mathrm{W(t)} - k * \mathrm{F(t)} \qquad (2)$$

where F(t) is the Cs concentration in whole fish or specific fish tissues such as muscle, W(t) is the dissolved Cs concentration in water, μ is an uptake constant with units L kg^{-1} d^{-1} , and k is a first-order loss rate constant with units d^{-1}. The parameter μ may include absorption of Cs from the water column but may function more as integrator of Cs assimilation from the various components of the diet. The parameter *k* is a measure of the rate of decline of the concentration in the population and includes: 1); 1) the physiological elimination of Cs from individuals; 2) growth dilution; 3) mortality, which includes predation; and 4) the physical decay of the radionuclide. Due to the relatively long half-life of ^{137}Cs, its physical decay has been considered negligible. Adjustment for the physical decay of ^{134}Cs is described below. The rate of decline may also reflect the rates of decline of the ^{137}Cs concentrations in the foods consumed by the species. For this open system, where fish are free to move from one area of study to another, *k* is also affected by emigration from the area of study and immigration into the area of study from areas of differing ^{137}Cs concentrations.

This model has been successfully applied to fish and other aquatic biota for ^{133}Cs, ^{137}Cs and ^{131}I (Smith et al., 2002; Pinder et al., 2009 and 2011; Martinez et al., 2014). The important aspect of this model in its use here is that the ratio μ / *k* is an estimator of the equilibrium C_r that does not require a constant ratio between F(t) and W(t).

The first step in the estimation of *u* and *k* involves modeling the decline in water concentrations using a multi-component exponential equation (Whicker and Shultz, 1982) of the form:

$$\mathrm{W(t)} = \sum_{\mathrm{i}} \mathrm{a_i} * e^{(-\mathrm{b_i} * \mathrm{t})} \qquad (3)$$

where a_i = the initial concentration of the ith component and b_i = the rate constant for exponential decline in the ith component. The second step involves estimating *μ* and *k* by fitting the following equation (Whicker and Shultz, 1982) to the time series of Cs concentrations in fish

$$\mathrm{F(t)} = \mu * \sum_{i} \frac{\mathrm{a_i}}{(k - \mathrm{b_i})} * (e^{(-\mathrm{b_i} * \mathrm{t})} - e^{(-k * \mathrm{t})}) \qquad (4)$$

where the a_i and b_i are from Eq. 2. For this analysis, estimates of μ and k and their asymptotic Standard Errors (SE) were obtained using the PROC NLIN of the Statistical Analysis System (SAS Institute Inc., 1989). In fitting Eq. 4, the initial Cs concentrations in the fish on the first day following the tsunami (*i.e.*, F(1)) were assigned a concentration of 1 Bq ^{137}Cs kg^{-1} based on the before tsunami data of Kasamatsu and Ishikawa (1997).

Other studies have postulated that certain processes are occurring and certain models are appropriate for quantifying these processes for the FDNPP releases (*e.g.*, Vives i Batlle, 2015; Tateda et al., 2013). Rather than proposing specific controlling processes, Eq. 1 is used as a descriptor of the temporal patterns in concentrations and reduces that temporal pattern to two parameters (and their SEs) where the ratio of these parameters estimates the equilibrium C_r for that species.

2. 6. Sources of ^{137}Cs data from the Fukushima releases

Both the ^{137}Cs concentrations for fish and water during the first 750 days following the tsunami were drawn from publically accessible websites. Surface water data were obtained from the Japanese Atomic Energy Agency for TEPCO's Fukushima Dai-chi NPP monitoring station at Futaba (Station T-1) which is approximately 3 km north of the FDNPP. This station had a more continuous record of monitoring ^{137}Cs concentrations with results consistent with those at other nearby stations within Fukushima Prefecture at Okuma (Station T-2) and Tomioka (Station T-3). Measurements of ^{137}Cs concentrations (Bq L^{-1}) were began on 23 March 2011 and data for the period from this date through 31 March 2013 were obtained as file 2079022502_00.csv(1) from

http://emdb.jaea.go.jp/emdb/en/portals/20760202/

The ^{137}Cs concentrations in seawater were measured once in the morning and once in the afternoon until 28 June 2011. Thereafter, they were measured in the morning on 6 days per week with one set of replicate morning measures being obtained each week.

The ^{137}Cs concentrations in numerous species of fish (Wada et al., 2013) from the Fukushima region have been collected by various procedures including nets (*i.e.*, seines and trawls), bait fishing, cages and diving. The ^{137}Cs concentrations were variously measured in $\geq$ 5 kg samples of whole bodies, muscles, and muscles with skin but not scales, and by various analytical facilities (Wada et al., 2013). The fish data for this analysis, which were obtained from

http://www.jfa.maff.go.jp/e/inspection/index.html

as MAFF, 2013; 201103-201203e1.xls and MAFF, 2014; eigo250329.xls for the period from April 2011 through March 2013. These files provided information on fish species, region of capture and analytical facility, but only limited information on sample processing. Because of this, the effects of sample processing and tissues have been assumed to be negligible.

In analyzing the Fukushima data for water and fish, those data reported as "nondetectable" in the form "(Nondetectable < detection limit)" were replaced with the reported "detection limit". Using this detection limit rather than the more often employed alternative procedure of using ½ the detection limit (Newman et al., 1989) should result in more conservative mean and median concentrations. Where sample results were reported as "Nondetectable" without a reported detection limit, the sample was deleted from the analysis. Within the first months following the tsunami, concentrations in fish were sometimes reported as the sum of both ^{134}Cs and ^{137}Cs (*i.e.*, $^{134+137}$Cs). Where this occurred, the ^{137}Cs concentration was estimated using Eq. 5,

$$^{137}\mathrm{Cs} = {}^{134+137}\mathrm{Cs}\ e^{-0.000857*t} \qquad (5)$$

where t is the days elapsed since the tsunami and the coefficient of t is from Wada et al. (2013).

3. Results

Not all of the 71 samples in the data of Rowan and Rasmussen (1994) from open ocean locations were used in the development of predictive models. All four of the samples for *Thunnus alalnga* (Albacore tuna) were deleted because their C_r were markedly greater than those for other tuna species. Two other samples that were each from different species and different locations were deleted because their C_r were markedly greater than those for other samples from their species. These deletions resulted in 65 observations on 41 species.

Concentration ratios for these 65 samples ranged over more than a factor of 8 from 16 to 139 L kg^{-1} with a median of 57 L kg^{-1} (Fig. 1). This range in C_r for saltwater fish was less than the factor of 15 ranges in C_r observed for freshwater fish in Rowan and Rasmussen (1994). Concentration ratios for the 23 samples from species classified as pelagic species ranged from 16 to 105 L kg^{-1} with a median of 53 L kg^{-1}. The range for the 42 samples from species classified as demersal fishes was from 18 to 139 L kg^{-1} with a median of 62 L kg^{-1}. As suggested by the broad overlap in ranges of C_r between pelagic and demersal species, there was no significant difference in mean C_r between the two classifications ($P > 0.50$ for 20,000 random rearrangements of C_r between pelagic and demersal species; PROC MULTTEST of the Statistical Analysis System; Richter and Higgins, 2006).

The TLs for the 41 species ranged from 2.48 to 4.29 with a median of 3.56, and there was considerable overlap in TL values within the Rowan and Rasmussen (1994) piscivorous and nonpiscivorous classifications. For the piscivorous classification, TL ranged from 3.23 to 4.29 with a median of 3.76. For those in the nonpiscivorous classification, the range was 2.48 to 3.92 with a median of 3.30. From the descriptions in *fishbase.org*, 21 of the species were classified as demersal species and

20 were classified as pelagic species. The range in TL for the demersal species was 2.48 to 4.29 with a median of 3.57 and completely overlapped the range in TL for pelagic species of 2.56 to 4.18 with a median of 3.46.

The Spearman rank correlations (r_s; Conover, 1971) in Table 1 indicated a statistically significant (*i.e.*, $P < 0.05$) correlation of C_r with TL. The correlation between C_r and K concentration was not significant. Moreover, the r_s value for this correlation was > 0 implying that C_r increased with increasing K. This increase in C_r with increasing K concentrations is not consistent with 1) the negative correlations between C_r and K concentrations observed for freshwater fish (Rowan and Rasmussen, 1994; Pinder et al., 2014) or 2) the experimentally induced declines in ^{137}Cs C_r observed following the additions of potassium chloride to a Chernobyl contaminated lake (Smith et al., 2003). Whatever the explanation for this statistically insignificant, but positive correlation, K concentrations were still included in the following model development. TLs and K concentrations were not significantly correlated.

3.1. Selection of a predictive model for the concentration ratio of saltwater fish

Initial analyses compared the model accuracies for all 7 of the possible combinations of the predictor variables and their interactions, and these models were evaluated for their accuracies in fitting the observed C_r and their ease of use. The three most appropriate models, arranged in order of increasing complexity, are hereafter referred to as Models I, II, and III. In Model I, the only predictive variable was the TL obtained from *fishbase.org*. This model has the advantage that it only requires readily available information from *fishbase.org* for its predictions. In Model II the predictive variables are the TL and the $\log_{10}$(K) concentration in the water. Model III used the predictive variables of the TL and $\log_{10}$(K) and the interaction term of TL and $\log_{10}$(K). Note that K concentrations could be used as a predictor variable because they can be computed from salinities that are readily available for various oceanic locations (*e.g.,* http//:www.nasa.gov/images/content/505786 main_mean_salinity_2005-full.jpg).

Table 2 compares the R^2, the aR^2, the error mean square, and the regression parameter estimates, with their standard errors, for Models I, II and III. The frequency distributions of their observed-to-fitted ratios are compared in Table 3. All three models were statistically significant, but their R^2 and aR^2 were relatively small (*i.e.*, < 0.15).

Although the R^2 are small, the frequency distributions of the model's observed-to-fitted ratios (Table 3) suggest that they are reasonably accurate predictors of the observed C_r. Their median observed-to-fitted ratios were near 1, and their ranges of Q_1 to Q_3, which represent 50% of the ratios, were within the range of 0.5 to 2.0. None of the models had ratios that are < 0.33 or > 3. Each model had only 2 ratios > 2, and $\leq$ 8 ratios < 0.5. Those ratios greater than 2 occurred for the same two samples in all three models. The accuracy percentages for being within a factor of 2 were in excess of 80% for all three models. The conservatism percentages for being less than a factor of 2 are all > 90%. The accuracy and conservatism percentages for factors of 3 are all 100 %.

Importantly, there was a clear trend for the more conservative and less egregious over predictions of C_r to be more prevalent (*i.e*., $\leq$ 8) than the less acceptable under predictions (*i.e*., > 2). The oberved accuracy percentages would be far less acceptable, from ecological and human health perspectives, if most of the errors were under predictions rather than over prediction.

Because of their similar accuracy and conservatism percentages, the decision of which model to employ may be based on 1) their relative ease of use and 2) a reasonable interpretation of their regression parameter estimates. On these bases, Model I is preferable because: 1) it is the simplest in requiring only the one readily available predictor variable of TL; and 2) its regression parameters are statistically significant and readily interpretable. The additions of the other predictor variables in Models II and III do not markedly improve either aR^2 (Table 2) or model predictability (Table 3) and have less readily interpretable regression parameters which are mostly not statistically significant. Model I has the form:

$$\text{Predicted } C_r = 10^{[(1.222 + 0.5*0.0398) + 0.147*TL]}. \qquad (6)$$

3.2. *The comparable accuracies of Model I in fitting C_r for pelagic and demersal species*

The fitted C_r from Model I developed using both the data from pelagic and demersal species demonstrated similar accuracies in fitting the C_r for the separate groups of pelagic and demersal species in the modelling data as shown in Fig. 2. The frequency distributions of the observed-to-fitted ratios had median values of 0.88 for pelagic and 1.05 for demersal species, and there was no statistically significant difference between the two frequency distributions of ratios (Kruskal-Wallis χ^2 = 2.36; df = 1; P > 0.10; Conover, 1971). Both distributions had accuracy percentages > 80% and conservative percentages > 90% for factors of 2. A suggestive trend in Fig. 2 for an apparently greater number of ratios > 1.5 for the demersal species (*i.e.*, 7) than for pelagic species (*i.e.*, 3) may be more a reflection of the greater number of ratios for demersal (n = 42) than for pelagic (n = 23) species rather than a difference in model accuracies.

3.3. *Assessing Model I accuracy from published results.*

The test data obtained from published results included 56 separate measures of observed C_r for either ^{133}Cs or ^{137}Cs from 51 species whose TL ranged from 2.28 to 4.37 and whose range in observed C_r was from 20 to 162 L kg^{-1} with a median of 50 L kg^{-1}. This range is similar to that for the modelling data. Thus, the set of test data was of comparable size and diversity to that of the modeling data.

Comparisons of predicted and observed C_r for the 55 demersal and the single pelagic fish in the test data are shown in Fig. 3. There little indication of increasing or decreasing ratios with increasing predicted C_r. There were 3 observed-to-predicted ratios < 0.5 and one ratio > 2. The distributions of the observed-to-predicted ratios are consistent with accuracies of 94% and 98% for factors of 2 and 3, respectively. The distributions are also consistent with conservatisms of 98% for both factors of 2 and 3.

3.4. Assessing Model I accuracy using the C_r computed as the ratio of μ to k from the temporal model for FDNPP releases

3.4.1. Temporal pattern of ^{137}Cs concentrations in Fukushima seawater

The ^{137}Cs concentrations in surface waters at Futaba (Fig. 4) initially declined until early April and then increased rapidly to a maximum of 68,000 Bq L^{-1} on 7 April in response to continuing releases. Following this second maxima, concentrations declined rapidly at first and then more slowly until April, 2012 when numerous measures of concentrations became less than the detection limits of 1.4 to 1.6 Bq L^{-1}. A three component version of Eq. 3 was fitted to the data using the Gradient method within PROC NLIN (SAS, 1989) with approximated intercept parameters, A_i and estimated decline rate parameters, b_i. These parameters and their SE are listed in Table 4. The standard errors for these parameters are greater than the parameter estimates. Normally, these large SE could suggest considerable error in the fitted model, but the > 700 ratios of observed water concentrations to fitted water concentations, which are a more appropriate evaluation of using this form of Eq. 3 as an input for estimating μ and *k* in Eq. 4, had a median of 1.08 with 80 % of the ratios occurring within the range of 0.55 to 1.88.

Previous applications of the simple water model in Eq. 3 have been confined to the dynamics of radionuclides in freshwater lakes where there were limited rates of loss due to mixing and and outfalls (*e.g.*, Smith et al., 2002; Pinder et al., 2016; Martinez et al., 2014). The use of Eq. 3 in this instance involves the complexities of strong lateral currents, near infinite relative mixing volumes, tides and wind effects (Buesseler et al., 2012;Kamenik et al., 2013; Rossi et al., 2013) as well as continuing radionuclide releases. These factors can complicate the selection and use of simple models, and the three component model employed here can only be a less than perfect approximation to the rapidly changing and variable water concentrations. In particular, the water model does not account for the brief period in early April 2011 when water concentrations were affected by additional releases.

3.4.2. The ^{137}Cs concentrations in fish and other biota.

Weekly sampling of the $^{134+137}$Cs, ^{137}Cs and ^{131}I concentrations in marine fishes and invertebrates began within a month following the tsunami and resulted in more than 8,000 measurements of either $^{134+137}$Cs or ^{137}Cs in all forms of biota from the region of the Fukushima Prefecture. Among these samples, there were 38 species of fish with n $\geq$ 50 measurements of either $^{134+137}$Cs or ^{137}Cs concentrations in the first 750 days following the initial releases. The TLs for these species ranged from 3.10 to 4.37 with a median of 3.60. The number of species, the number of samples for species, and the range of TLs all suggest a sufficiently large and sufficiently diverse set of test data.

However, the estimations of μ and k for these species were compromised by two factors. Firstly, there were fewer reported $^{134+137}$Cs or ^{137}Cs concentrations reported in the first 200 days after the tsunami than in the subsequent 100 day intervals (Wada et al., 2013). The effect of this sampling pattern was further complicated by the occurrence of maximum concentrations in most fish species within these first 200 days as is indicated by 1) the distributions of data illustrated in Fig. S1 of Wada et al. (2013) and 2) the regression lines fitted to these data in Table S2 of Wada et al. (2013) and similar regression lines in Iwata et al. (2013).

3.4.3. Fitting the temporal model to the fish data.

The combination of fewer early measurements and the possible occurrence of maximum concentrations within this early period resulted in uncertainties in the estimation of μ and, consequently, the underestimation of C_r. These uncertainties in fitting the models are illustrated in Fig. 5 for the benthopelagic species *Nibea mitsururii.* This species is illustrated because it had: 1) a representative number of total samples in the first 750 days, n = 93; 2) a representative number of samples during the first 200 days, n = 10; 3) an intermediate TL of 3.5 $\pm$ 0.37; and 4) a predicted C_r of 57 L kg^{-1} from Model I. The fitted parameters of μ and its SE were 0.00367 $\pm$ 0.00034 L kg^{-1} d^{-1} and k and its were SE of 0.00676 $\pm$ 0.00046 d^{-1}. These μ and k result in a predicted maximum concentration of 560 Bq L^{-1} occurring on day 28 following the tsunami, and the timing of this statistically estimated peak concentration is similar to that estimated for fish by D-Dat modelling (Vives i Batile, 2015). If samples with measurable concentrations greater than 200 Bq kg^{-1} had occurred in the first 100 days, the estimated μ may have been greater. If samples with measurable concentrations less than 200 Bq kg^{-1} had occurred in the first 100 days, the estimated μ may have been smaller. The lack of these early data may also have contributed to a lack of fit in the model where observed concentrations occurring before day 300 were consistently overestimated and concentrations after day 300 were largely underestimated.

3.4.4. The estimated C_r from the temporal models.

Despite the complications arising from the lack of early data, the model presented in Fig. 5 provides a reasonable approximation to the observed data. However, the ratio of μ to k produces a completely inappropriate estimated C_r of 0.543 L kg^{-1} which is a factor of 100x less than the predcited C_r from Model I of 57 L kg^{-1}. The potential errors in estimating μ due to the lack of early data are insufficient to account for an error of this 100X magnitude. Similar inappropriate estimates of C_r also occurred for the other species. Fitting Eq. 4 to the data for the 38 species of fish (including *N. mitsururii*

) resulted in estimated C_r ranging from 0.074 L kg^{-1} to 1.754 L kg^{-1} with a median of 0.232 L kg^{-1}. All of these C_r were far less than the minimum 16 L kg^{-1} observed by Rowan and Rasmussen (1994).

A possible explanation for these small C_r would be differences in the extent of the spatial scales from which concentrations in water and fish were estimated. Most monitoring of water concentrations occurred near FDNPP (Fisheries Agency of Japan, 2014), but fish were harvested from both farther north and south along the coast as well as father offshore from FDNPP. However, using alternative models with smaller A_1 terms that might be more representative of 1) more remote coastal areas north and south of FDNPP or 2) more distant offshore locations did not result in C_r that occurred within the ranges predicted by Model I or observed by Kasamatsu and Ishika (1997).

The uncertainties concerning the concentrations in fish from the first 200 days and extremely small estimated μ and C_r suggests questions concerning the appropriateness of this model for these data. Where this model has been successfully applied to fish and other biota involving either experimental or accidental releases of radionuclides (*e.g.*, Smith et al., 2002; Pinder et al., 2006, 2009, 2011), the rapid increases in cesium concentrations in the water were $\leq$ 1000-fold, and In contrast, the ^{137}Cs concentrations in Fukushima waters increased by a factor of > 20,000,000 from approximately 0.3 mBq L^{-1} (Kasamatsu and Ishikawa, 1997) to 68,000 Bq L^{-1} (Fig. 4), and the maximum concentrations in fish, which occurred before large-scale sampling began, increased by at least a factor of 10,000-fold from 0.3 Bq kg^{-1} (Kasamatsu and Ishikawa, 1997) to 3000 Bq kg^{-1} (Wada et al., 2013). Whether these disparities in magnitudes and timings are, or are not, responsible for the small μ and their small projected C_r, the small C_r resulting from the application of the temporal model to these Fukushima data are clearly inappropriate for testing the accuracy of Model I.

The underestimation of C_r appeared to be more the result of small values of μ (Table S1), which ranged from 0.000085 L kg^{-1} d^{-1} to 0.0279 L kg^{-1} d^{-1} with a median of 0.001175 L kg^{-1} d^{-1} than the values of *k* which ranged from 0.00106 d^{-1} to 0.0196 d^{-1} with a median of 0.00371 d^{-1} and were consistent with the decline rates reported by Wada et al. (2013; Table S2). If the estimates of μ had been in the range of of 0.082 L kg^{-1} d^{-1} for psicivorous fish and 0.12 L kg^{-1} d^{-1} for carnivorous and for planktivorous fish as reported by Tateda et al. (2015), the C_r obtained from Eq. 2 would have been in the range of 30 L kg^{-1} (29. L kg^{-1} = 0.11 L kg^{-1} d^{-1}/0.00371 d^{-1}).

4. DISCUSSION

A potential limitation of this analysis involves the inability to use the extensive data from the FDNPP releases to test the predicted C_r from Model I. A more comprehensive assessment of Model I's accuracy may have been possible if the application of the temporal model to the Fukushima data had been more appropriate and had produced more reasonable C_r. Despite this potential limitation, the existing accuracy assessment suggests that Model I could still prove to be an accurate and useful predictor of ^{137}Cs C_r for saltwater fish. Thus, the model could be used to 1) predict concentration ratios for species of saltwater fish or 2) to scale the relative levels of contamination to be expected among an assemblage of saltwater species at a specific location. However, Model I is not appropriate for predicting the C_r for saltwater species in the Baltic Sea due to the variation in K concentrations in its water as a result of the mixing of fresh and salt waters (Pinder, unpublished analyses).

Although Model I may be considered accurate, the smaller range in C_r among saltwater species than for freshwater species (*i.e.*, a factor of 8 as opposed to 15) may reduce the importance of the saltwater model relative to that for the freshwater model. This smaller range of C_r for saltwater species is likely due to the relative constancy of salinities and corresponding K concentrations across oceans as opposed to the large variation in K concentrations among freshwater bodies. This consistency in salinities and K concentrations may allow the trophic status of a species to largely determine its C_r in salt water, whereas in freshwater systems the trophic status may merely determine the relative magnitude of a species C_r within a fish community where the absolute magnitudes of C_r may be predominantly affected by the water's K concentration.

Because of this smaller range of C_r among species, it may also prove acceptable to apply the median C_r for saltwater species observed by Rowan and Rasmussen (1994) of 57 L kg^{-1} (or more conveniently 60 L kg^{-1}) to saltwater species in general rather than using Model I to predict individual C_r. This median value is within a factor of 2 of many of the larger C_r observed by Rowan and Rasmussen (1994), and this range of 2 for the median is similar to the two-fold factor of accuracy for Model I. The C_r of 60 L kg^{-1} is similar to the range of recently reported, recommended or employed Cr for marine fish of 30 to 36 L kg^{-1} reported by Tagami and Uchida (2013), the range of 31 to 87 L kg^{-1} recommended by Psaltaki et al. (2013), or C_r of 86 L kg^{-1} employed by Vives I Batlle (2015).

Moreover, the wide range and variety of saltwater species used as human food sources may support the convenience of using a simplified default value. Such a default value might also be employed to estimate the mean ^{137}Cs concentration for an assemblage of fish were the relative abundances of the species within the assemblage is unknown, but the ^{137}Cs concentrations in the water from which they were obtained is known.

5. Summary

Models have been developed which allow potentially accurate predictions of Cs concentration ratios for saltwater species that requires only 1) readily-available, numerical TLs from the FishBase Global Information System (*fishbase.org*) and, possibly, 2) estimates of the K concentration in the water which may be derived from also readily-available measures of ocean salinities.

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The authors are very impressed and deeply appreciative of the Japanese government and its agencies for their full, open and timely public release of their monitoring data. We are also deeply saddened by the heart wrenching impacts of the tsunami on the peoples and the economies of Fukushima and its surrounding areas. We have both hope and faith that the resilience and the determination of the Japanese peoples will eventually win through in restoring the economies, cultures and spirits of Fukushima and its surrounding communities.

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Table 1. Spearman rank correlation coefficients (r_s) among the variables TLs (TL), K concentrations, and ^{137}Cs concentration ratios for 65 samples of fish from 41 species from 18 open ocean locations. * = P $\leq$ 0.05.

Variables	Spearman rank correlations for the variables	
	K Concentrations	Concentration ratios
TLs	0.060	0.261*
K concentrations	---------	0.229

Table 2. Parameter estimates and their standard errors, R^2, aR^2 = the adjusted R^2, and the EMS = Error Mean Square for Models I, II and III fitted to the 65 samples of concentration ratios for the 41 species. TL = random resampling TL from *fishbase.org*. The $\log_{10}(K)$ = logarithm to base 10 of the K concentration in saltwater. Interaction = interaction between TL and $\log_{10}(K)$. * = $P \leq 0.05$.

Model	R^2	aR^2	EMS	Parameter estimates $\pm$ standard errors			
				Intercept	TL	$\log_{10}(K)$	Interaction
I	0.099*	0.085	0.398	1.222 $\pm$ 0.202*	0.147 $\pm$ 0.056*	------	------
II	0.141*	0.113	0.389	-8.204 $\pm$ 5.461	0.140 $\pm$ 0.055*	2.378 $\pm$ 1.577	------
III	0.146*	0.104	0.389	23.597 $\pm$ 50.91	-8.587 $\pm$ 13.891	-5.630 $\pm$ 12.802	2.198 $\pm$ 3.498

Table 3. Frequency distribution parameters for observed-to-fitted ratios for both pelagic and demersal species where Q_1 and Q_3 represent the first and third quartiles of the distribution.

Model	Frequency distribution parameters				
	Minimum	Q_1	Median	Q_3	Maximum
I	0.378	0.673	1.000	1.317	2.413
II	0.334	0.738	0.998	1.304	2.516
III	0.337	0.735	1.016	1.276	2.500

Table 4. Parameter estimates and their standard errors for the three component model fitted to the decline in surface water concentrations at Futaba.

	Parameters	
	Estimated Value	Standard Error
	Intercept parameters (Bq L^{-1})	
A_1	20,000	24,156
A_2	110	2514
A_3	4	2383
	Rate parameters (d^{-1})	
b_1	0.11	0.102
b_2	0.013	0.640
b_3	0.002	1.136

Figure Legends

Figure 1. The frequency distribution of the 65 concentration ratios (L kg^{-1}) in the modeling data as obtained from Rowan and Rasmussen (1994).

Figure 2. The distribution of the observed-to-fitted concentration ratios for demersal and pelagic species in the modelling data.

Figure 3. A comparison of observed-to-predicted ratios for 55 demersal and the one pelagic fish from Model I for the test data where: 1) ratios of observed-to-predicted concentrations ratios > 2 are indicated by the upper dashed line; 2) ratios of 1 are indicated by the solid line; and 3) observed-to-predicted ratios < 0.5 are indicated by the lower dashed line.

Figure 4. The ^{137}Cs concentrations in surface waters from the Futaba sampling station with a three component exponential model fitted to the declining concentrations. Horizontal patterns at concentrations of 24, 9 and 1.5 Bq L^{-1} are reported values of detection limits applicable at these times rather than measured concentrations. The median ratio of the > 700 observed-to-fitted water concentrations was 1.08.

Figure 5. A comparison of the 93 observed ^{137}Cs concentrations (Bq kg^{-1} wet mass) for *Nibea mitsukurii* with the concentrations fitted from using the water model illustrated in Fig. 4. The observed data begin on the 136^{th} day after the tsunami and end on the 710^{th} day.

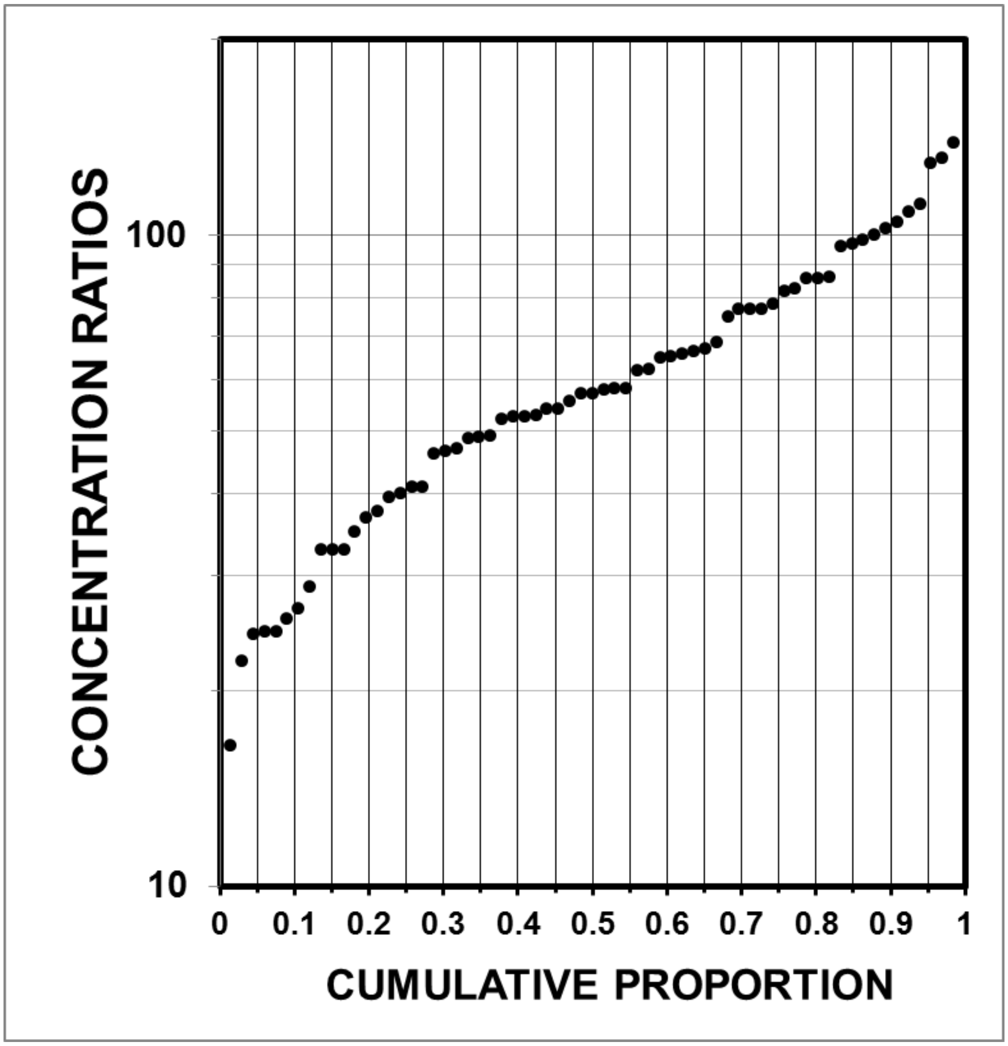

CONCENTRATION RATIOS
100
10
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
CUMULATIVE PROPORTION

Fig. 1

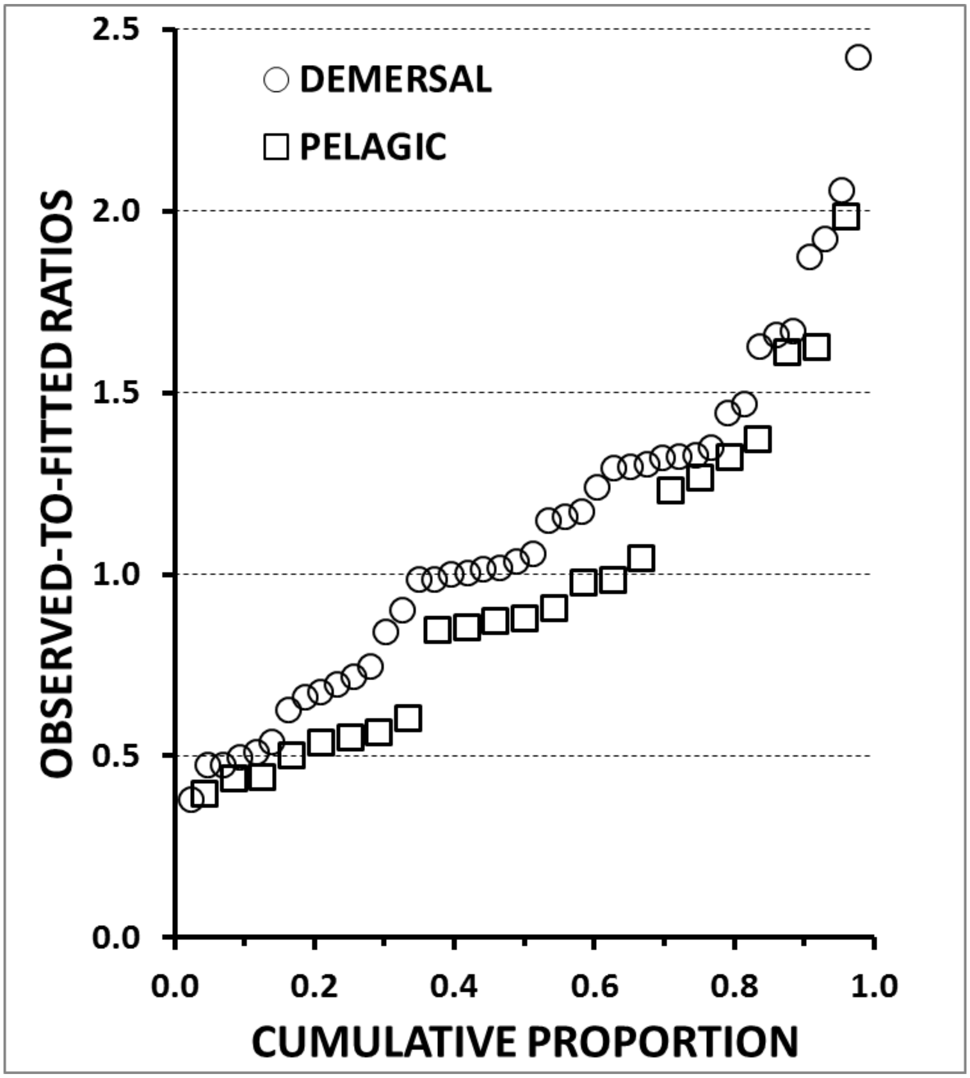

DEMERSAL
PELAGIC
OBSERVED-TO-FITTED RATIOS
2.5
2.0
1.5
1.0
0.5
0.0
0.0
0.2
0.4
0.6
0.8
1.0
CUMULATIVE PROPORTION

Fig. 2

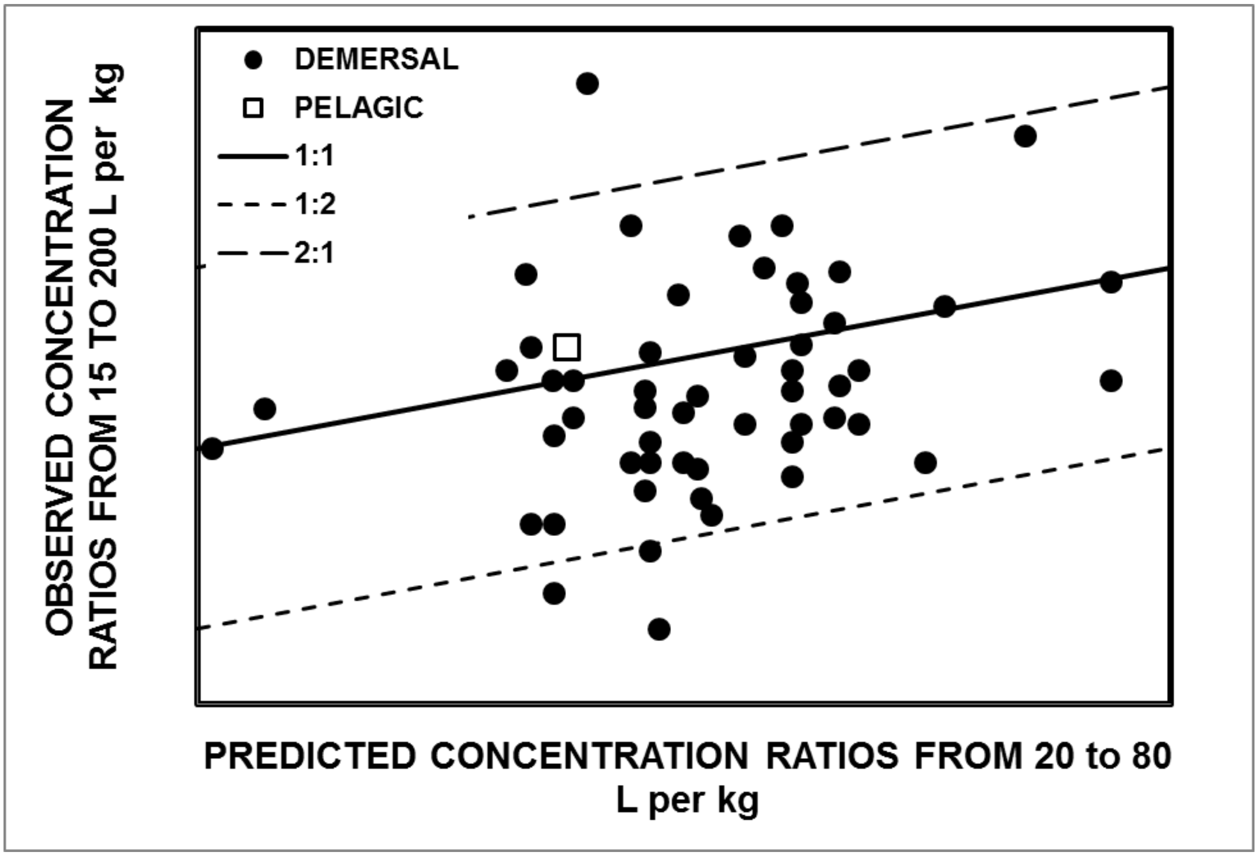
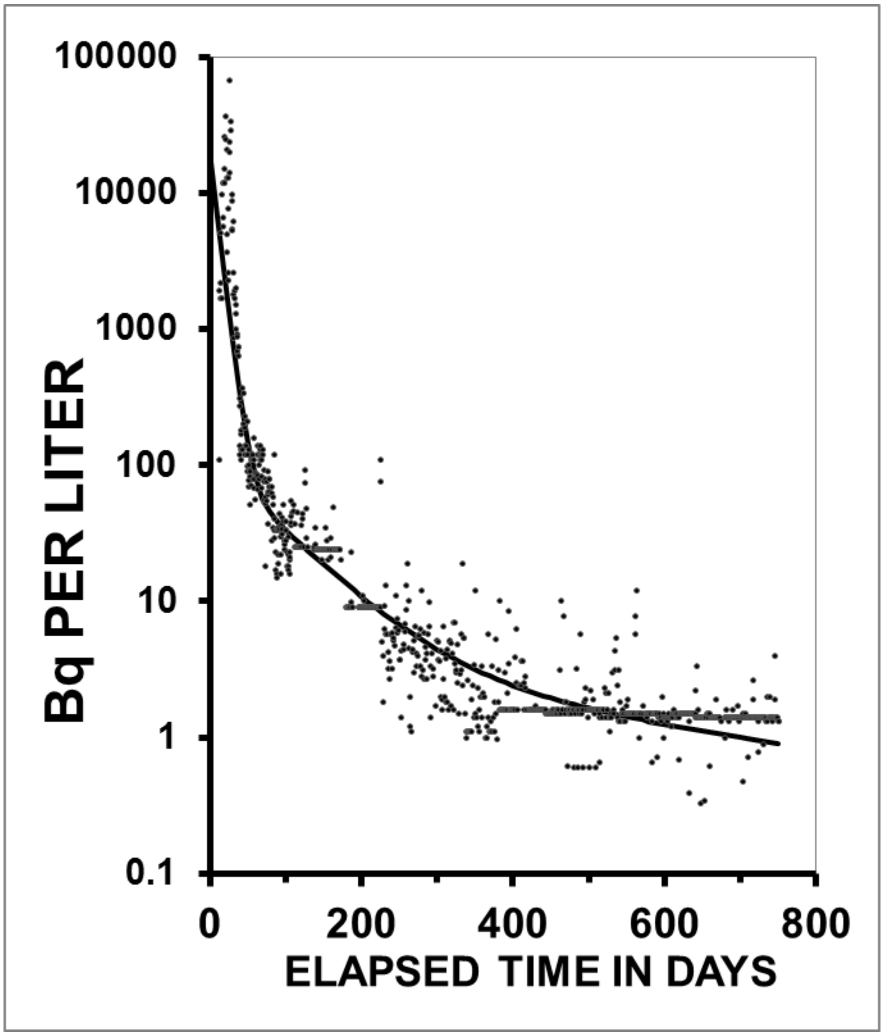

DEMERSAL
PELAGIC
1:1
1:2
2:1
OBSERVED CONCENTRATION RATIOS FROM 15 TO 200 L per kg
PREDICTED CONCENTRATION RATIOS FROM 20 to 80 L per kg

Fig. 3



100000
10000
1000
100
10
1
0.1
Bq PER LITER
0
200
400
600
800
ELAPSED TIME IN DAYS

Fig. 4

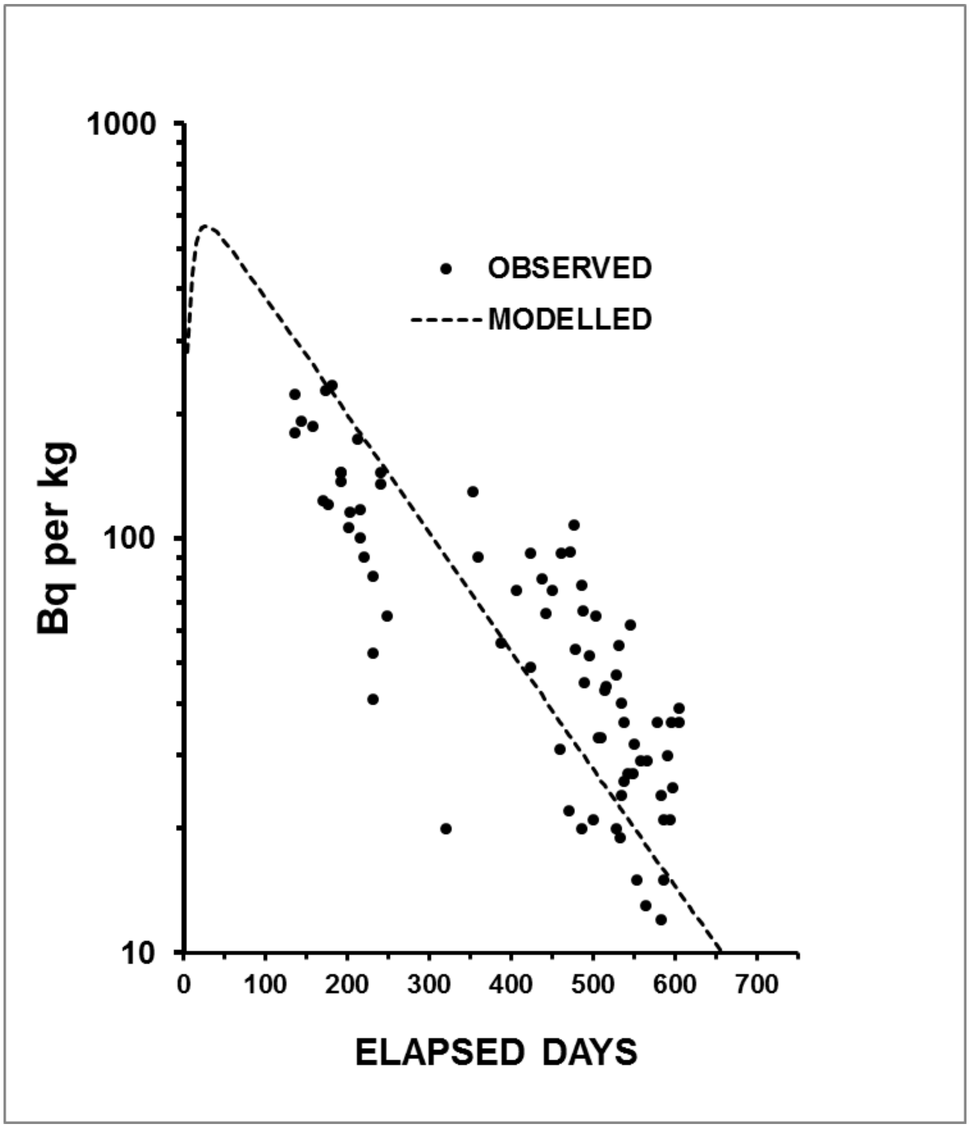


Fig. 5

Table A.1

The species names, trophic levels, habitats and observed concentration ratios for the 56 data sets from published studies used to test the accuracy of Model I. Trophic levels and habitats are from *fishbase.org*. Species nomenclature conforms with that used in the cited references with the exception of Kasamatsu and Ishikawa (1997) where nomenclature from fishbase.org replaces the use of common names.

Literature Cited	Genus	Species	Trophic Level	Habitat	Observed C_r
1	*Chelon*	*labrosus*	2.59	BP	47
2	*Cynoscion*	*jamaicensis*	3.84	D	54
2	*Micropogonias*	*furnieri*	3.27	D	162
2	*Sardinella*	*brasiliences*	3.10	D	54
3	*Hexagrammos*	*decagrammus*	3.59	D	91
3	*Hexagrammos*	*lagocephalus*	3.46	D	72
3	*Myoxocephalus*	*polyacanthocephalus*	3.71	D	75
4	*Achirus*	*klunzingeri*	3.40	D	27
4	*Arctoscopus*	*japonicus*	3.51	D	33
4	*Cleisthenes*	*pinetorum*	3.40	D	58
4	*Conger*	*myriaster*	3.98	D	38
4	*Cynoglossus*	*joyneri*	3.50	D	37
4	*Dentex*	*tumifrons*	3.80	D	51

Table A.1. Continued.

Literature Cited	Genus	Species	Trophic Level	Habitat	Observed C_r
4	*Dexistes*	*rikuzenius*	3.53	D	31
4	*Eopsetta*	*grigorjewi*	3.50	D	49
4	*Gadus*	*macrocephalus*	4.37	D	76
4	*Glossanodon*	*semifasciatus*	3.14	BP	78
4	*Hexagrammos*	*otakii*	3.79	D	45
4	*Hippoglossoides*	*dubius*	3.20	D	42
4	*Kareius*	*bicoloratus*	3.15	D	59
4	*Lateolabrax*	*japonicus*	3.36	D	94
4	*Lepidion*	*inosimae*	3.50	BP	52
4	*Pagrus*	*major*	3.70	D	50
4	*Paralichthys*	*olivaceus*	3.72	D	70
4	*Paraplagusia*	*japonica*	3.20	D	30
4	*Pennahia*	*argentata*	3.47	BP	46
4	*Pleurogrammus*	*monopterygius*	3.64	D	80
4	*Sebastes*	*schlegelii*	3.80	D	79
4	*Sebasticus*	*marmoratus*	3.60	D	57
4	*Theagra*	*chalcogramma*	3.68	BP	94
5	*Mylio*	*macrocephalus*	3.24	D	52
5	*Argyrosomus*	*argentatus*	3.47	D	38

Table A.1. Continued.

Literature Cited	Genus	Species	Trophic Level	Habitat	Observed C_r
5	*Branchiostegus*	*japonicus*	3.40	D	38
5	*Dasyatis*	*akajei*	3.84	D	44
5	*Ditrema*	*temminicki*	3.39	D	34
5	*Gadus*	*macrocephalus*	4.37	D	52
5	*Lateolabrax*	*japonicus*	3.36	D	38
5	*Mugil*	*cephalus*	2.50	D	40
5	*Pagrus*	*major*	3.70	D	36
5	*Paralichthys*	*olivaceus*	3.72	D	61
5	*Scomber*	*japonicus*	3.42	P	50
6	*Ditrema*	*temminicki*	3.39	D	49
6	*Hexagrammos*	*otakii*	3.79	D	65
6	*Hippoglossoides*	*dubius*	3.20	D	52
6	*Kareius*	*bicoloratus*	3.15	D	30
6	*Limanda*	*yokohamae*	3.24	D	45
6	*Microstomus*	*achne*	3.39	D	50
6	*Oncorhynchus*	*keta*	3.42	BP	20
6	*Paralichthys*	*olivaceus*	3.72	D	60
6	*Parapristipoma*	*trilineatum*	3.40	BP	41
6	*Scombrus*	*boops*	4.19	BP	133

Table A.1. Continued.

Literature Cited	Genus	Species	Trophic Level	Habitat	Observed Cr
6	*Sebastes*	*baramenuke*	3.70	D	54
6	*Sebastes*	*inermis*	4.02	D	69
6	*Sebastes*	*marmoratus*	3.60	D	44
6	*Sebastes*	*pachycephalus*	3.70	D	41
6	*Siganus*	*fusecenns*	2.28	D	43

1. Antovic and Antovic (2011)
2. Cunha *et al.* (1993)
3. Hong *et al*. (2011)
4. Kasamatsu and Ishikawa (1997)
5. Suzuki *et al*. (1973)
6. Tateda and Koyanagi (1996)