Modeling Bioaccumulation for the Prediction of Tissue and Sediment Concentrations

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Introduction

Biota-sediment accumulation factors (BSAFs) or biota-sediment accumulation regressions (BSARs) are statistical tools used to:

- Estimate chemical concentrations in tissue when empirical tissue data are lacking
- Estimate sediment preliminary remediation goals (PRGs) from target tissue concentrations

Early applications of BSAFs focused on the prediction of tissue concentrations using simple ratios of tissue and sediment concentrations that assume that tissue concentrations change in a constant proportion with sediment concentrations. These BSAFs do not account for non-sediment sources of contamination to tissue concentrations (e.g., water, background inputs). BSARs allow the nature of the relationship between tissue and sediment concentrations (e.g., linear, log linear, or non-linear) to be investigated and for the contributions of non-sediment sources of contamination to be explicitly determined (via the intercept of the regression model). It is important to consider both the shape of the relationship and the contributions of non-sediment sources when developing tissue concentration predictions or PRGs. Here, tissue and sediment chemistry datasets for three chemicals are used to explore how predicted tissue concentrations and sediment PRGs can be affected by the model chosen and whether tissue or sediment is used as the dependent variable.

Methods

Data from the US Environmental Protection Agency (EPA) Mid-Continent Ecology Division's (MED's) BSAF database were used to explore tissue-sediment relationships. This dataset includes approximately 20,000 BSAFs from 20 locations (mostly Superfund sites) for non-ionic organic chemicals, including polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polycyclic aromatic hydrocarbons (PAHs), pesticides, and other compounds. Fresh, tidal, and marine species are included in the dataset.

A total of 352 chemical datasets for 6 species that had more than 1 sample were selected (Table 1). Of those, 265 datasets had a sample size of 4 or greater. Those 265 datasets were tested for linear relationships between untransformed (arithmetic) tissue and sediment concentrations, arithmetic tissue and log (10) transformed sediment concentrations, and log (10) tissue and log (10) sediment concentrations.

Table 1. Datasets investigated from the EPA MED database

Species	No. of Datasets (Chemicals)	No. of Datasets with n≥ 4
Alewife floater (*Anodonta implicate*)	6	6
Benthic invertebrates	8	3
Blue mussel (*Mvtilus edulis*)	42	33
Hard clam (*Mercenaria mercenaria*)	56	52
Hard clam (*Pitar morrhuana*)	52	38
Crayfish (unid)	188	133
Total	**352**	**265**

Note: The dataset included data for 201 PCB congeners, total PCBs, 25 PAHs, 17 PCDD/PCDF congeners, 12 pesticides, 2 phenols, and tributyltin.

EPA – US Environmental Protection Agency
MED – Mid-Continent Ecology Division
PAH – polycyclic aromatic hydrocarbon
PCB – polychlorinated biphenyl
PCDD – polychlorinated dibenzo-*p*-dioxins
PCDF – polychlorinated dibenzofurans

BSAF Equation

BSAFs were derived using Equation 1.

$$BSAF = \frac{(C_{tiss,LN})}{(C_{sed,OC})} \qquad \textbf{Equation 1}$$

Where:

$BSAF$ = site-specific fish BSAF
$C_{tiss,LN}$ = organism tissue concentration, lipid-normalized (mg/kg lipid dry weight [dw])
$C_{sed,OC}$ = surface sediment concentration, organic carbon (OC)-normalized (mg/kg OC dw)

BSAR Equation

For organic chemicals, sediment concentrations were normalized based on OC content, and tissue concentrations were normalized based on lipid content before BSAR regressions were performed. For non-organic chemicals, regressions were performed using the total sediment chemical concentration and the total tissue chemical concentrations (unadjusted).

The following linear regressions were considered for each organism-chemical dataset:

- Untransformed tissue concentrations vs. sediment concentrations
- Untransformed tissue concentrations vs. log-transformed sediment concentrations
- Log-transformed tissue concentrations vs. log-transformed sediment concentrations

A regression model was screened in for further evaluation if the slope was significant ($p < 0.05$) and the coefficient of determination (r^2) was > 0.3 (i.e., at a minimum, a weak relationship was established). The strengths of the relationships were evaluated as follows:

- No relationship: where $0.0 \leq r^2 < 0.3$
- Weak relationship: where $0.3 \leq r^2 < 0.5$
- Moderate relationship: where $0.5 \leq r^2 < 0.7$
- Strong relationship: where $0.7 \leq r^2 < 1.0$

Regression models that were screened in were further evaluated for goodness of linear fit through a consideration of the distribution of residuals and evidence of outliers.

Results

Tissue vs. Sediment Regressions

Of the datasets and relationships tested, only 15 to 30% of the relationships met the screening criteria of statistical significance and $r^2 > 0.3$ (Table 2). Approximately one-third of the relationships were negative.

Table 2. Number of datasets with different classes of linear relationship between tissue and sediment

Relationship Class	No. of Datasets by Type of Relationship: Tissue (µg/kg Lipid) and Sediment (µg/kg OC)	Tissue (µg/kg Lipid) and Log(Sediment (µg/kg OC))	Log(Tissue (µg/kg Lipid)) and Log(Sediment (µg/kg OC))
r < 0.0 (negative slope)	81	69	82
r^2 < 0.3 (not even a weak relationship)	56	83	78
$r^2 \geq 0.3$ (some relationship)	128	113	105
Total	**265**	**265**	**265**
$r^2 \geq 0.3$, $p < 0.05$ (passing relationship)	82	50	53

OC – organic carbon

Only a limited suite of chemicals in hard clam and crayfish had relationships that passed the criteria (Table 3).

Table 3. Species and chemicals with tissue sediment regressions that pass regression screening criteria

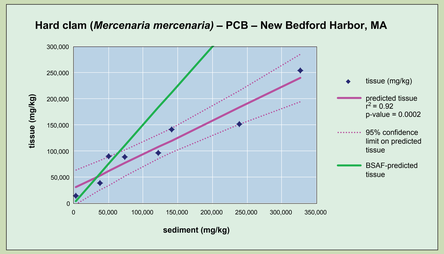
Species	Chemicals with Relationships Between Sediment and Tissue
Hard clam (*Mercenaria mercenaria*)	PCB congeners and total PCBs, chrysene
Hard clam (*Pitar morrhuana*)	chrysene
Crayfish (unid)	PCDD/PCDF congeners, dieldrin, PCB congeners

PCB – polychlorinated biphenyl
PCDD – polychlorinated dibenzo-*p*-dioxin
PCDF – polychlorinated dibenzofuran

Of the datasets that passed the screening criteria, most would not be considered "good" linear relationships based on the distribution of the residuals.

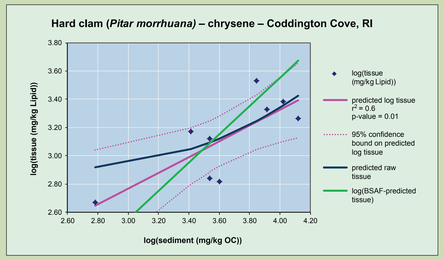
Examples

Example 1 illustrates the importance of considering the intercept in characterizing the tissue-sediment relationship. BSAFs, and BSARs with the intercept forced through the origin, are biased to under-predict tissue concentrations at low sediment concentrations and to over-predict at high sediment concentrations. Thus PRGs developed using BSAFs or by forcing a BSAR through the origin are liable to overestimate the reduction in tissue chemical concentration possible with sediment remediation because they fail to account for non-sediment sources (e.g., water) and tend to have steeper slopes. Note that this relationship seems quite linear across two orders of magnitude and also has fairly well-distributed residuals.



Example 1. Value of including an intercept in the model

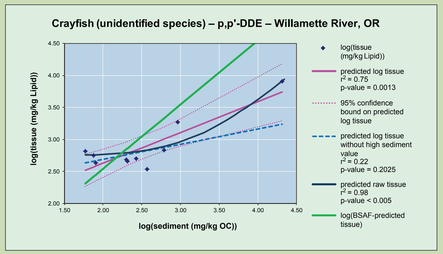
Example 2 illustrates the importance of considering the nature of the relationship being investigated (e.g., linear, log linear, or non-linear). In this case, considering only a linear relationship would probably lead to unnecessarily low sediment PRGs. The relationship presented in Example 2 is not very strong and just barely meets the screening criteria. The predictions of tissue from sediment or vice versa would be strongly affected by the choice of the tissue-sediment model.



Example 2. Ability to investigate the shape of the relationship

Example 3 illustrates several points:

- **Influence of a single high tissue-sediment pair on the tissue sediment relationship** – The high pair in this dataset has a much greater effect on the regression (as measured by outlier statistics, including leverage, Cook's Distance, and DFBeta's) than any of the other data pairs.
  - A high pair could indicate the presence of more than one population of tissue-sediment relationships in the dataset. Multiple populations could be created by fine- or large-scale heterogeneity in physical or biological factors.
  - If a high pair is not part of the same population as the rest of the data but retained in the dataset, tissue concentrations probably would be overpredicted, and PRGs might overestimate the reduction in tissue chemical concentration that would be achieved in the majority of the population with sediment remediation.
  - If a high pair appears to be valid and is the sole cause of a significant relationship, data transformations or other types of models should be considered.
- **Effect of model choice on predictions of tissue concentrations** – A model that uses raw tissue and sediment data would lead to tissue concentration predictions that are higher or lower than those from a log-log model, depending on the sediment concentration.
- **Use of relationship when linear model is not really a good fit** – The distribution of model residuals is usually used to assess the goodness of fit of a linear relationship and also to determine whether transformations of the independent and/or dependent variable improve linearity. In general, the residuals around the log-log model are more evenly distributed around the predicted values than those of a linear model using the raw data or log transformed sediment; however, residuals of even the log-log regression have a distinct pattern that could have many causes. If the highest data pair is removed, the relationship between tissue and sediment is not significant. What are the risks of assuming a linear relationship when there is evidence that other factors may be affecting the relationship?
- **Potential issues if non-detect data are not included in dataset** – Several datasets with both high tissue and non-detected tissue concentrations at the highest sediment values have been encountered. If non-detects are not included in a dataset (as the EPA database), it is possible to overestimate the strength of a relationship.



Example 3. Consideration of the effect of outliers on predictions

In Example 3, the removal of the highest sediment tissue pair results in a non-significant relationship. Does the high pair indicate that more than one population is present in the dataset, that other factors are affecting the relationship, or that the relationship between sediment and tissue is not linear? With the outlier, the residuals indicate that a linear model is not a good fit, but the reason why it is not must be explored.

Discussion

PRGs and associated confidence intervals can differ depending on the model used, as well as the sediment or tissue value used to predict the other variable. Consequently, it is very important to find a model fit that best characterizes the relationship between tissue and sediment. This involves:

- Evaluating model residuals to look for any patterns or unequal variances along the range of predicted values
- Evaluating tissue-sediment pairs that strongly influence the model or lie further from the model than do other data

Both types of information are needed to determine if:

- A linear fit is appropriate
- Subpopulations of the data exist
- Other factors are affecting the tissue-sediment relationship
- Either a BSAR or a BSAF is appropriate or other approaches are required

Other Potential Approaches

Model II-Type Regressions

When the tissue-sediment relationship is truly linear with normally distributed residuals, Model II-type regression models, which consider residual variability in both x and y, will be useful for describing uncertainty in sediment concentrations through confidence intervals around PRG calculations (Burkhard 2006). However, Model II-type approaches will not eliminate the uncertainty in PRG calculations that arises from truly weak or non-linear relationships between sediment and tissue concentrations or lead to tighter confidence intervals around the predicted PRGs.

Non-Linear Regressions or Models that Consider Other Factors

Non-linear or hierarchical models that incorporate information about other factors that could be affecting the tissue-sediment relationship may be helpful if there is a theoretical basis for assuming a non-linear relationship and/or data about other factors and adequate sample size to explore alternative models.

Reverse Regressions (Sediment vs. Tissue) for Developing PRGs

The use of reverse regressions (i.e., sediment vs. tissue) was also investigated. This approach makes sense if BSARs are to be used for developing PRGs because the sediment concentration (i.e., the PRG) is the predicted variable. Also, if it is assumed that there is greater certainty in tissue than in sediment concentrations (e.g., tissue samples based on composites, sediment samples based on individual grabs), this approach is appropriate because it acknowledges the uncertainty in sediment concentrations. There is also great uncertainty in the sediment concentration as a result of uncertainty regarding the exposure area for organisms. Even if there are good estimates of home range, the use of localized habitat is often dependent on numerous factors (e.g., substrate qualities, organism size, population density) that are not well characterized.

If the tissue-sediment relationship is truly linear, residuals are normally distributed, and the residual variance is low, an increased precision in estimating a PRG using a reverse regression could be anticipated. However, as with most of the hundreds of tissue-sediment relationships examined, predicting sediment from tissue does not consistently produce tighter confidence intervals around the PRG because of the fact that **few tissue-sediment relationships appear to meet the assumptions required to fit a regression and calculate valid confidence intervals.** This is a topic that deserves further investigation, but the bottom line is that if regression modeling assumptions are met, then reverse regression makes more sense for developing PRGs.

Recommendations

- Based on the extant data compiled by EPA MED in its BSAF database, few tissue-sediment relationships meet the assumptions required to fit a regression and calculate valid confidence intervals. Better guidance on sampling design should be developed to improve the quality of BSAFs and BSARs.
- BSARs have an advantage over BSAFs in that they allow for the consideration of non-sediment contributions to tissue burdens.
- An acceptable strength of relationship (r^2, or percent of variability in y explained by x) and significance level (p-value) should be established prior to the development BSARs.
- In selecting a BSAR, the nature of the relationship (e.g., linear, log linear, or non-linear) should be considered.
- An evaluation of potential outliers, including the development of criteria for the exclusion of data (e.g., a specified leverage value), should be performed.
- The pattern of residuals should be examined to determine goodness of fit and the possibility of contributions from other factors.
- Given the profound differences in tissue concentration predictions and estimated PRGs that can result from different statistical approaches, it is important to be extremely clear about:
  - Informational goals (is tissue or sediment being predicted?) and how informational needs change through the risk evaluation and remediation process
  - The strength of the relationship between tissue and sediment that would be needed to make a prediction meaningful
  - The extent to which modeling the uncertainty in both variables is considered necessary for the predictions to be meaningful

References

Burkhard LP. 2006. Estimation of biota sediment accumulation factor (BSAF) from paired observations of chemical concentrations in biota and sediment. EPA/600/R-06/045. Ecological Risk Assessment Support Center, US Environmental Protection Agency, Cincinnati, OH.