BIOACCUMULATION-BASED SEDIMENT QUALITY CRITERIA FOR THE PROTECTION OF HUMAN HEALTH

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1. INTRODUCTION

Bottom sediments are an important sink and sometimes source for hydrophobic organic contaminants such as PCBs, chlorinated pesticides, and dioxins/furans. Once in the sediments, these contaminants can pose a direct toxic threat to sediment-dwelling organisms and may also represent an indirect risk to humans who consume fish that have become contaminated via the sediment and food chain transfer. While there are several tools available to evaluate potential toxic impacts to benthic organisms themselves, very little work has been done to define sediment quality criteria designed to limit fish bioaccumulation and attendant human health risk.

One approach to developing "bioaccumulation-based sediment quality criteria" that shows promise relies upon equilibrium partitioning theory. The approach is based on the observation that organic contaminants like PCBs preferentially associate with the organic carbon component of sediments and the lipid fraction of fish. This phenomenon can be used to translate an allowable contaminant level in fish flesh to an associated contaminant concentration in the sediment based on simple partitioning. The factor that is used to relate the contaminant level in the organic carbon fraction of the sediment to the contaminant level in the lipid of the fish is referred to as the biota-to-sediment accumulation factor (BSAF). Formally, the BSAF is defined as the ratio of the concentration of chemical in the fish normalized to the lipid content to the chemical concentration in the sediment normalized to organic carbon. The use of the BSAF in the development of bioaccumulation-based sediment quality criteria is illustrated in the derivation that follows.

2. METHODS

For known or probable human carcinogens, excess lifetime cancer risk can be estimated as the product of lifetime average daily dose (LADD) and the cancer potency slope (q_1^*).

$$Risk = LADD \times q_1^* \qquad (1)$$

In the above equation, the units of LADD are mg/kg/d and those of potency slope are 1/(mg/kg/d). Therefore, cancer risk is a unitless value.

Lifetime average daily dose, in turn, is defined as the product of lifetime average daily exposure rate (LADE) times a gastrointestinal absorption efficiency factor, AF, divided by the body weight of the human receptor, BW.

$$LADD = \frac{LADE \times AF}{BW} \qquad (2)$$

The units of LADE are expressed as mg/d, absorption factor is unitless, and the units of body weight are in kilograms.

In the case of human exposure through consumption of contaminated fish flesh, LADE can be computed as follows:

$$LADE = \frac{C_f \times CR \times ED \times (100 - RF)/100}{LT} \qquad (3)$$

where the following terms and units are defined:

C_f = concentration of contaminant in the edible (fillet) portion of the fish, mg/kg
CR = fish consumption rate, kg/d
ED = duration over which exposure is assumed to occur, yr
LT = lifetime duration, yr
RF = percent reduction in contaminant concentration in the fish due to trimming and cooking losses, %

Equations 1, 2, and 3 can be combined and rearranged to solve for the concentration in the fish, C_f. The resulting expression is:

$$C_f = \frac{Risk \times BW \times LT}{q_1^* \times AF \times CR \times ED \times (100 - RF)/100} \qquad (4)$$

The above equation can be used to estimate a contaminant concentration in the fillet portion of the fish which corresponds to any pre-defined risk level, potency slope, and combination of exposure factors (BW, LT, CR, ED, and RF).

The next step in the formulation is to assume that the contaminant partitions itself in the whole body and fillet portion of the fish based upon the lipid content of the whole body (lc_{wb}) and lipid content of the fillet (lc_f), respectively. Formally, we set the lipid-normalized contaminant concentration in the fillet equal to the lipid-normalized concentration in the whole fish.

$$\frac{C_f}{lc_f} = \frac{C_{wb}}{lc_{wb}} \qquad (5)$$

The units of lc_f and lc_{wb} should be expressed as grams of lipid per grams of fillet and grams of lipid per grams of whole fish, respectively. For example, a fillet sample reported as 2 % lipid would be the same as 2 grams of lipid per 100 grams of fillet or a numeric value of 0.02 gram lipid per gram of fillet.

Rearranging equation 5 to solve for C_f:

$$C_f = \frac{C_{wb} \times lc_f}{lc_{wb}} \tag{6}$$

Now, setting equation 4 equal to equation 6, we obtain the following:

$$\frac{C_{wb} \times lc_f}{lc_{wb}} = \frac{Risk \times BW \times LT}{q_1^* \times AF \times CR \times ED \times (100 - RF)/100} \tag{7}$$

We now seek to introduce an expression which couples fish tissue contaminant concentration to sediment contaminant concentration. As noted in the introduction, this is done through the biota-to-sediment accumulation factor (BSAF). By definintion,

$$BSAF = \frac{C_{wb}/lc_{wb}}{C_{sed}/f_{oc}} \tag{8}$$

where the following new terms are introduced:

C_{sed} = bulk concentration of the contaminant in the surficial sediment, ng/g or ppb
f_{oc} = fraction of organic carbon in the surficial sediment, gm o.c./gm sed

Combining equations 7 and 8 and solving the resulting expression for C_{sed}, we obtain the final, desired expression for computing a bioaccumulation-based sediment quality criterion for known or probable human carcinogens:

$$C_{sed} = \frac{Risk \times BW \times LT \times f_{oc} \times 1000}{q_1^* \times AF \times CR \times ED \times BSAF \times lc_f \times (100 - RF)/100} \tag{9}$$

By extention, it can easily be shown that the applicable equation to compute a bioaccumulation-based sediment quality criterion for non-carcinogenic effects is:

$$C_{sed} = \frac{RfD \times BW \times f_{oc} \times 1000}{AF \times CR \times BSAF \times lc_f \times (100 - RF)/100} \qquad (10)$$

All the terms in the above equation have been previously defined except RfD, which is the reference dose associated with the non-carcinogenic effect of interest. The units of reference dose are mg/kg-d.

The factor of 1000 in the numerator of equation 9 and 10 is a unit conversion factor that ensures that C_{sed} is expressed in units of ng/g (i.e., ppb), assuming the other terms are expressed in the units previously noted. For convenience, the units to use in the above equations are summarized in the table that follows.

Risk and AF	unitless
BW	kg
LT and ED	yrs
f_{oc}	(gm o.c./gm sed)
q_1^*	$(mg/kg\text{-}d)^{-1}$
CR	kg/d
BSAF	(ug/gm lipid)/(ug/gm sed o.c.)
lc_f	(gm lipid/gm fish fillet)
RF	percent

3. RESULTS

As an example, equations 9 and 10 of the previous section have been used to compute bioaccumulation-based sediment quality criteria for carcinogenic and non-carcinogenic effects due to PCBs. A target risk level of 1 additional cancer case in a population of 100,000 individuals (i.e., 10^{-5}) has been assumed. This risk level is consistent with Delaware's Hazardous

Substance Cleanup requirements but is ten times less restrictive than the risk level used in Delaware's Surface Water Quality Standards. A cancer potency slope of 2.0, based on positive evidence of carcinogenicity for several Aroclors, was used as representative for all PCBs. An RfD of 2E-5 was used for the non-carcinogenic effects of PCBs based upon studies using Aroclor 1254. Both the potency slope and reference dose were taken from EPA's Integrated Risk Information System (IRIS).

Three separate human receptor groups were considered in the derivation of the criteria: average adults, women of child-bearing age, and children between 0 and 6 years old. Body weights for these three groups were assumed to be 70 kg, 64 kg, and 14.5 kg, respectively. Fish consumption rates for these three groups were taken from a creel study of Delaware anglers conducted in 1992/1993. That study covered the area of the Delaware Estuary between the PA/DE border down to Cape Henlopen. The average fish consumption rate for the three groups was reported as 0.0175 kg/d, 0.0159 kg/d, and 0.0059 kg/d.

Lifetime duration for all three receptor groups was assumed to be 75 years based upon Bureau of Vital Statistics data. For purposes of the carcinogenicity assessment, exposure duration for the two adult groups was assumed to be 30 years, which is a standard risk assessment assumption. Exposure duration for the child receptor group was 6 years.

The gastrointestinal absorption factor was set to 1 and the contaminant reduction factor for proper trimming and cooking of the fish was assumed to be zero.

The fraction of organic carbon was assumed to be 2.5 % (i.e., 0.025 gm o.c./gm sediment) based on the mean value in the Delaware Estuary as reported in a study conducted by Arthur D. Little on behalf of the Delaware Estuary Program in 1993. The lipid content in the fillet portion of an average fish was assumed to be 2 % (i.e., 0.02 gm lipid/gm fillet). This value is representative of those reported by the U.S. Department of Agriculture for estuarine and marine fish species. Finally, a value of 1.85 was specified for the BSAF based upon values reported in the technical literature.

The table that follows summarizes the results of the calculations for PCBs in sediment.

	Average Adult	**Women of Child-Bearing Age**	**Children**
Carcinogenic Effects	33.8 ppb	34.0 ppb	103.8 ppb
Non-carcinogenic Effects	54.1 ppb	54.4 ppb	33.2 ppb

The figures in the above table indicate that a bulk PCB concentration of 33 ppb or less is necessary in order to keep cancer and non-cancer health risks to adults and children to *de Minimus* levels. Because sediment contamination exhibits a great deal of spatial variability, the 33 ppb target concentration should be applied as an areal mean (or median for skewed distributions). Note that the values in the above table can be adjusted upward or downward based upon site-specific data.

4. DISCUSSION

The actual concentration of PCB in the surficial sediments of the tidal Delaware River is roughly an order of magnitude greater than the desired level of 33 ppb presented in the previous section. Recall that the risk level used in the derivation of the bioaccumulation-based sediment quality criteria was 10^{-5} and that fish consumption rates were taken as broad averages. If the methodology used to derive the sediment criteria is at all accurate and predictive, then the cancer risk associated with consuming fish from the tidal Delaware River should also be an order of magnitude greater than the target risk level used in the development of the criteria. In other words, the cancer risks associated with consuming fish from the Delaware Estuary should be approximately 10^{-4}, assuming the same exposure factors are considered. As an example, consider the case of the average adult fisherman who consumes 0.0175 kg of fish per day from the Delaware. For a typical PCB concentration of 1 ppm in the edible muscle of striped bass, channel catfish, or white perch, lifetime cancer risk is computed as 2×10^{-4}. We conclude from this example that the methodology used to derive the bioaccumulation-based sediment quality criteria is sound and that it provides answers accurate to at least an order of magnitude, and perhaps even better.

Despite the fact that the methodology appears reasonable for the case of PCBs in the tidal Delaware River, it is important to recognize that there are many assumptions and uncertainties to the approach. Central to this is the use of a "no-threshold" cancer model. A related issue is the assumption that the relationship between dose and tumor incidence in the human population is linear under low dose. In addition, questions remain regarding the relevence of cancer incidence in animals relative to the human population.

5. SUMMARY AND CONCLUSIONS

Equilibrium partitioning theory has been used to derive bioaccumulation-based sediment quality criteria for the protection of human health. PCBs in the Delaware Estuary have been used as an example. Based on the approach, average bulk concentrations of PCBs in surficial sediments should fall below 33 ppb in order to protect potential human receptors from adverse health effects.

Based upon available sediment and fish tissue PCB data from the tidal Delaware River, the methodology appears reasonably accurate and predictive.