Integrated Environmental Assessment and Management — Volume 5, Number 1—pp. 138–149

Empirical Estimation of Biota Exposure Range for Calculation of Bioaccumulation Parameters

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(Received 5 May 2008; Accepted 2 October 2008)

ABSTRACT

Bioaccumulation factors (BAFs) and biota–sediment accumulation factors (BSAFs) are frequently used to predict contaminant bioaccumulation in risk assessments. Development of these parameters is often hindered by uncertainty regarding the spatial scale of contaminant transfer from sediments to biota. We present a simple statistical method for optimizing bioaccumulation parameters (BAF and BSAF) in aquatic species, such as fish, whose exposure history may occur over broad spatial scales. For 6 finfish species sampled in San Francisco Bay, San Diego Bay, or the Southern California Bight, California, USA, the spatial scale of correlation was optimized using regression analysis. The ranges identified for pairing biota and sediment observations generally corresponded to the known life histories of the species and with laboratory tests comparing relationships observed for 28-d *Macoma* spp. This procedure may be useful for identifying appropriate species and spatial scales to predict bioaccumulation and for developing data sets of corresponding sediment and tissue residues.

Keywords: Bioaccumulation factor Exposure range BSAF Sediments

Original Research

INTRODUCTION

Legacy pollutants have severely impacted natural aquatic systems, necessitating costly risk assessments and cleanup actions. The bioavailability of organic pollutants, and thereby the potential for bioaccumulation, has been shown to vary widely among estuarine and coastal water bodies potentially at risk (Boese et al. 1995; Boese et al. 1997; Mason and Lawrence 1999; Kraaij et al. 2002; Battelle et al. 2005). Bioaccumulation, the net increase of a chemical by an organism because of uptake from all environmental sources, is frequently modeled using bioaccumulation factors (BAFs) and biota–sediment accumulation factors (BSAFs). Bioaccumulation factors are the ratio of biota to sediment contamination concentration (Eqn. 1).

$$\mathrm{BAF} = \frac{C_t}{C_s} \quad (1)$$

Biota–sediment accumulation factors are the same ratio (Eqn. 2), corrected for lipid content of the biota and organic carbon content of the sediment (reviewed in Wong et al. 2001; Burkhard et al. 2004).

$$\mathrm{BSAF} = \frac{(C_t/f_L)}{(C_s/f_{OC})} \quad (2)$$

where C_t is the tissue concentration, C_s is the sediment concentration, f_L is the fraction of lipid in tissue, and f_{OC} is the fraction of organic carbon in sediment (USEPA 2000). For organic pollutants, the use of lipid and organic carbon normalization rests on the principle that pollutants are predominantly associated with these matrices, producing more reliable relationships (Clark et al. 1988).

The use of BSAFs and BAFs to predict biota exposure from sediment-associated pollutants relies on several key assumptions, which should be considered before their application. These include the assumptions that currently monitored sediments are in steady state with the organism and are the primary source of contamination to the species being modeled. For the selected fish species, contamination is assumed to be primarily due to bioaccumulation from contaminated benthic prey, such as invertebrates and smaller fish, closely associated with the sediment. The exposure to contaminants from waterborne sources other than the sediments, including uptake from ambient water, respiratory surfaces (e.g., gills and external body), and prey not associated with sediments (e.g., phytoplankton, zooplankton, and pelagic forage fish), are assumed to be relatively small. These assumptions have been shown to generally apply when assessing bioaccumulation in chemicals of higher hydrophobicity (K_{ow}), as indicated by log octanol–water partitioning coefficients between 6 and 7 (Burkhard, Cook, et al. 2003). As a result, sediments as the ultimate source of organic contaminant exposure to benthic fish and invertebrates have been indicated in many recent modeling approaches (e.g., Morrison et al. 2002; Burkhard, Cook, et al. 2003). Pelagic species are less attractive for sediment risk assessment and decision-making than benthic species because there is more uncertainty regarding the indirect (i.e., food web mediated) contribution of sediments to the contaminant burden of pelagic species.

Biota–sediment accumulation factors are widely applied in the scientific literature (e.g., Boese et al. 1995; Tracey and Hansen 1996; Burkhard, Cook, et al. 2003; Burkhard, Endicott, et al. 2003) and have commonly been used in sediment risk assessments (e.g., Byron et al. 2003; USEPA 2003). For PCBs, a BSAF of 4 is expected for finfish, whereas benthic invertebrates typically have values around 1 (Ankley et al. 1992; Maruya et al. 1997; Kraaij et al. 2002). However, substantial variation exists among locations, as observed in syntheses undertaken at national and global scales (Wong et al. 2001; Burkhard et al. 2005). Differences often arise as a result of multiple factors. Food web structure and resulting

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Published on the Web 10/15/2008.

trophic position influence contaminant biomagnification as do dietary variation, organism lipid content, and spatial movement (Kidd et al. 1998; Linkov et al. 2002). Within the abiotic matrices, sediment organic carbon and sediment versus water column disequilibrium can be important (Burkhard, Cook, et al. 2003). As a result, BAFs and BSAFs can have a range of 2 to 3 orders of magnitude among species and contaminants (Tracey and Hansen 1996). Consequently, when performing ecological risk assessments in support of sediment remediation, site-specific BAF and BSAF values are needed.

The spatial scale of transfer between sediment and biota is expected to increase with factors such as food web trophic level as well as organism longevity and mobility. Contaminant concentrations in sediments and sessile benthic invertebrates are expected to reflect local conditions because of limited movement. Higher trophic level organisms, including fish and wildlife, move and forage at multiple locations over a longer life span and, thus, integrate their exposure over broader spatial scales than an individual sediment-monitoring station (Linkov et al. 2002; Moore et al. 2005). Sampling of sediment and biota across the immediate home range ("exposure area") of target organisms is needed for successful measurement of bioaccumulation parameters (Burkhard 2006).

In addition to some fish, benthic invertebrates are also well suited for predicting bioaccumulation. The bent-nosed clam (*Macoma nasuta*) burrows in and ingests sediments, and is, therefore, a good indicator of bioavailable sediment-associated contaminants. *Macoma* spp. have been recommended previously for bioaccumulation evaluations based on known tolerance, exposure history, and data availability (e.g., Lee et al. 1993; USEPA 2000). *Macoma nasuta* has also been shown to reach steady state in some laboratory experiments, although results depend on the compound being examined and conditions of the experiment (Pruell et al. 1993; Boese et al. 1997; Moore et al. 2005). *Macoma nasuta* has been used extensively in laboratory bioaccumulation experiments because of life history factors that cause high sediment exposure (Pruell et al. 1993; Boese et al. 1997; MEC Analytical Systems 2003; Werner et al. 2004). Nevertheless, concerns regarding extrapolations between field exposure and laboratory test conditions warrant collection of field biota for risk assessment purposes (e.g., Ankley et al. 1992; Pruell et al. 1993; Tracey and Hansen 1996).

It must be recognized that empirical BAFs and BSAFs do not determine the relative contribution of sediment contamination to biota contaminant burden. Rather, local source data collection and mechanistic contaminant fate and bioaccumulation modeling are needed to determine rates of direct (i.e., dermal and respiratory exposure and sediment consumption) and indirect (i.e., food web mediated) exposure to sediment-associated contaminants (Clark et al. 1988). Although such methods can identify the complex interactions among sediments, the water column, and organisms, they can be costly and time-consuming to implement. We suggest that a need also exists for considering more straightforward methods of evaluating currently available contaminant data.

The objective of this study was to demonstrate a statistical method for developing bioaccumulation parameters (BAF and BSAF) using aquatic species in California, USA. The statistical procedure focused on development of biota–sediment contaminant relationships in aquatic species whose exposure may be derived over broad spatial scales of unknown size. The challenge in establishing biota–sediment relationships from field data is that the exposure area and duration of exposure for the fish is unknown and unlikely to be causally related to a sediment concentration from a single point in time and space (i.e., an individual sediment sample). Therefore, the field-collected data used in this study were tested for the most appropriate spatially averaged scale for sediments to pair with each fish tissue concentration value. For comparison, laboratory *Macoma nasuta* 28-d test results were evaluated to examine bioaccumulation parameters in the absence of spatial scale variability.

METHODS

Bioaccumulation data

Data sets of sediment and tissue chemistry were obtained from the California Sediment Quality Objectives database. The database consists of raw data compiled from more than 100 dredging, monitoring, and research studies conducted in California from 1980 to 2003 (Myre et al. 2006). Further information on the studies and screening criteria for the database can be found at (http://www.sccwrp.org/data/2006_sqo.html). The data were collected from bays, estuaries, and coastal locations that ranged geographically from San Francisco Bay to San Diego Bay. Sediment analyses all focused on surficial (<15 cm) sediments. Additionally, fish and sediment data from the Southern California Bight (SCB) and laboratory bioaccumulation data from Newport Bay not available through the California Sediment Quality Objectives database were obtained from original study authors. To ensure that the data used for our analysis were comparable, they were examined for consistency. This evaluation included verification of species studied and chemicals measured, comparable detection and reporting limits, geographic coordinates and description, and presence of individual contaminant data necessary for summing (e.g., 6 DDT isomers to calculate tDDTs). Samples that did not contain the necessary individual contaminant data for summing were excluded. Laboratory exposure tests employed standard 28-d test methods for exposure of *M. nasuta* to field contaminated (not laboratory-spiked) sediments. Analyses focused on PCBs, legacy organochlorine pesticides (DDTs, chlordanes, and dieldrin), and PAHs. Summation procedures followed that of the Regional Monitoring Program for Water Quality in the San Francisco Estuary (e.g., SFEI 2005), with individual congeners or compounds below detection converted to 0.

Statistical analyses were performed on data aggregated by species, contaminant, and water body. Bioaccumulation data were separated into 3 data sets for analysis; the first examined laboratory bioaccumulation in benthic invertebrates; the other two used data on fish and sediments from marine embayment and coastal locations, respectively. Data for each analysis were paired using the spatial optimization procedure described below, which was developed in Matlab Version 7.1 (http://www.mathworks.com/products/matlab) and may be obtained by contacting the authors.

Laboratory bioaccumulation

The majority of laboratory bioaccumulation data in California have been performed on species evaluated for bioaccumulation potential in contaminated dredged sediments (USEPA 1994a, 1994b). Laboratory bioaccumulation data sets for *M. nasuta*, *Neanthes virens*, and *Nephtys caecoides*

were initially screened for use in this study. However, the majority of data for *N. virens* and *N. caecoides* were found to be below detection limits. Previous laboratory experiments using these species have also documented generally low tissue contaminant concentrations, which has been attributed to species not reaching steady state within the duration of the standard 28-d tests (Pruell et al. 1993). Therefore, to demonstrate the statistical procedure using laboratory bioaccumulation data, only the *M. nasuta* data set was used. Data used for this example followed guidance outlined in the Ocean Testing Manual (USEPA and US Army Corps of Engineers 1991) and US Environmental Protection Agency (1994a). Homogenized sediment of known field concentrations were administered to test organisms in a laboratory environment, with test conditions monitored throughout the experiments. Tissue analyses were subsequently performed after 28 d to determine the availability of sediment contaminants taken up by the test organisms.

Sediment and *M. nasuta* chemistry data were paired where spatial coordinates matched between samples. For each contaminant class and sampling location, sediment data were averaged and then matched to the average *M. nasuta* contaminant concentration at that location. Relationships were subsequently examined using regression analysis in SAS Version 9.1 (SAS Institute, Cary, NC, USA). Residuals were checked for normality and variance homoscedasticity, and biota or sediment concentrations were log or square root transformed, if necessary (Draper and Smith 1998). The procedure was applied using lipid-normalized or wet weight tissue data and organic carbon–normalized or dry weight sediment data.

Marine embayment bioaccumulation

The finfish analyses focused on determining an appropriate spatial scale for BSAF or BAF development. The underlying assumption of this technique is that, although the true fish exposure area is unknown, the long-term averaging nature of organochlorine bioaccumulation will yield a maximum coefficient of determination with the spatial scale closest to the true spatial scale of exposure. Exposure is the combined effect of spatial distributional histories of predator and prey in relation to the underlying sediment contamination and is also influenced by the ever-changing mix of potential prey items, which may change seasonally, as well. The complexity of these interacting factors dictates the site-specific nature of the analyses of exposure range as described below.

Fish tissue (filet muscle, wet weight) concentrations were averaged at each sampling location. Analyses focused on PCBs and legacy pesticides; PAHs were excluded because they are rapidly metabolized by fish (Eisler 1987; van der Oost et al. 2003). Sediment chemistry data were pooled over 2 spatial areas that represented the largest bay and estuary data sets available (San Francisco Bay and San Diego Bay) and averaged by sampling location. To provide a comparable approach to the Offshore Coast assessment (see details below), in addition to averaging sediment at discrete sampling locations, data were also spatially averaged using kriging. The discrete and spatially averaged sediment data were paired to fish tissue concentrations in separate analyses.

Average sediment concentrations located in a circle of varying size, centered at the spatial coordinates of each fish sampling location, were paired with the average of fish tissue concentrations at that location. The spatial area (size of the circle) at which sediment concentrations were averaged was varied at increasing radial distance to evaluate the strength of statistical association between biota and sediment concentrations (e.g., Figure 1) and to identify the spatial scale at which the coefficient of determination (r^2 of the linear regression) was greatest. Analyses were conducted to compare fish tissue concentrations to surrounding sediment concentrations at 1-km-radius increments at spatial scales from 0 to 10 km (e.g., Figure 2). Regression analysis using lipid-normalized or wet weight tissue data and organic carbon–normalized or dry weight sediment data were conducted as described above.

Offshore coast bioaccumulation

The average fish concentrations for PCBs and DDTs were determined from all discrete coastal sampling locations in the Southern California Bight. This coastal region was selected for analysis because it comprised a high density of sediment chemistry and fish tissue samples. Sediment data were generally sparse around the Channel Islands and the offshore shallow banks. As a result of this heterogeneous sediment sample distribution, kriging was performed to estimate sediment contaminant concentrations in areas not sampled. Kriging results were interpolated onto a regularly spaced grid using Surfer Version 7 (Golden Software, Golden, CO, USA). Subsequently, average sediment concentrations calculated from kriging results were paired with the average of organism tissue concentrations at each fish sampling location using the spatial procedure. The same method for varying the spatial area described for marine embayments was used in this analysis, and regression analysis was conducted to evaluate the maximum degrees of association (r^2) for paired results.

Bioaccumulation parameters

Bioaccumulation parameters, including BAFs and BSAFs, were calculated for species with tissue concentrations showing significant, positive correlations to that of sediment (see Eqns. 1 and 2). For *M. nasuta*, both BAFs and BSAFs were calculated to facilitate a comparison of bioaccumulation parameters. For fish species, either BAFs or BSAFs was calculated, depending on the biota–sediment relationship that was strongest (smallest p value and highest r^2). Bioaccumulation factors and BSAFs were calculated after pairing biota samples from specific locations with average sediment concentrations over the spatial scale that produced the highest r^2 in the optimization routine. Each paired observation was back-transformed, the ratio of contaminant concentration in biota to that of sediment was calculated for that observation, and subsequently, the mean of all ratios in that water body was determined. Estimation of variability was determined using the standard deviation of the mean in BAF and BSAF values. It is recognized that back-calculation of log-transformed values yields predictions of the geometric rather than arithmetic mean. However, geometric means are better predictors of central tendency for log-normal data, as frequently occurs with contaminant data.

RESULTS AND DISCUSSION

Spatial optimization of biota–sediment relationships

The spatial optimization procedure developed for this study was demonstrated using 3 example data sets. To some extent, life history differences may explain the variability

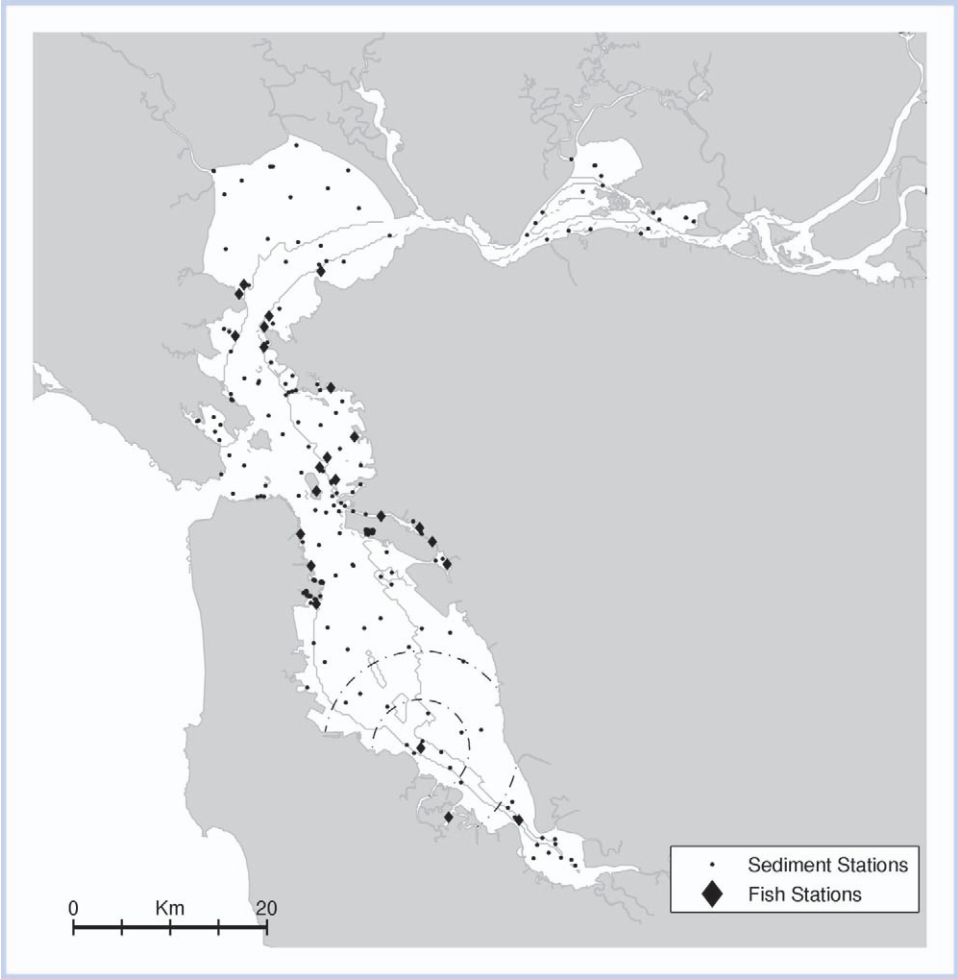

Figure 1. Graphical illustration of pairing fish and sediment data in the spatial optimization procedure. Dashed line represents circles of increasing radial distance from the fish station. Sediment concentrations within each circle were averaged and paired to the corresponding average fish concentration for that location.

observed among species in statistical significance, regression r^2, and spatial scale of the strongest results. Fish with benthic dietary associations (e.g., California halibut [*Parilichthys californicus*] and white croaker [*Genyonemus lineatus*]) often exhibited the strongest correlation to sediment contamination. However, in many cases, correlations were weak (e.g., $r^2 < 0.4$), suggesting that other factors may impede strong, consistent relationships. Therefore, caution is warranted in interpreting individual results.

Laboratory bioaccumulation was evaluated in the bentnosed clam (*Macoma nasuta*) for bioaccumulation of trace organic contaminants in San Francisco Bay, Newport Bay, and San Diego Bay. Normalization of tissue data for lipid content produced relatively weaker regressions that those based on raw data (Table 1). Generally, results indicated significant, positive correlations to sediment concentrations for each contaminant and water body evaluated. For example, PCBs in San Diego Bay sediments were highly correlated ($r^2 = 0.93$, $p < 0.001$) to *M. nasuta* exposed to those sediments (Figure 3). Previous analyses of *M. nasuta* laboratory bioaccumulation data have documented similar correlations to sediment concentrations (Naber et al. 2007).

To examine bioaccumulation in marine embayments, fish species from San Francisco Bay and San Diego Bay were evaluated for various trace organics. Five different fish species each showed at least one statistically significant correlation between tissue and sediment chemistry data averaged at discrete locations (Table 2), as well as for sediment data averaged by kriging (results not presented). Our observation of significant correlations was consistent with many literature examples of significant correlation between aquatic organism bioaccumulation and sediment chemistry for PCBs, DDTs, and other chlorinated organic compounds (Table 3). In southern California, statistically significant relationships have been shown for trace organic contaminants in sanddabs (*Citharichthys* spp.) and other flatfishes (e.g., Schiff and Allen 2000; Allen et al. 2004), as well as white croaker.

In shiner surfperch (*Cymatogaster aggregata*), concentrations of 4 classes of organic contaminant each exhibited a significant correlation to sediment at spatial scales of 1 km (e.g., Figure 4). The small spatial scale of the correlations suggests that surfperch likely bioaccumulate the majority of their contaminant exposure from invertebrates foraging within San Francisco Bay sediments. Shiner surfperch has previously shown strong spatial patterns with trace organic contaminants in San Francisco Bay (Davis et al. 2002; Greenfield et al. 2005). The limited variation explained by the biota–sediment regressions ($r^2 = 0.25$–0.44) could be attributed to spatial movement, with warmer months spent in nearshore shallow water and movement offshore into deeper

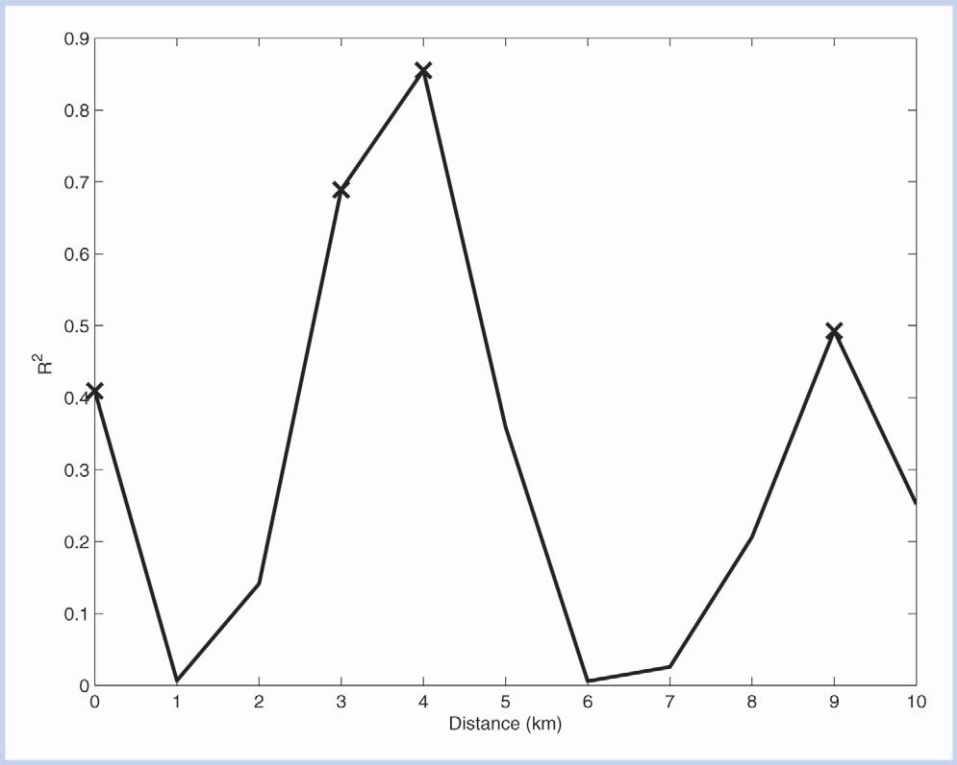

Figure 2. Example of model output from spatial association procedure. Results are presented for total PCBs (wet weight) in California halibut (*Paralichthys californicus*) from San Diego Bay ($n = 11$, SD = 0.27). Xs indicate significant relationships ($p < 0.05$) at the given spatial scale. Note that this is the same relationship as presented in Table 2.

water during the fall and winter (Bane 1970; Shaw et al. 1974; Emmett et al. 1991). Partial exposure from foraging outside of contaminated sediments has been shown to strongly influence bioaccumulation (Linkov et al. 2002). Another potential explanation for relatively low r^2 is that surfperch likely accumulate contaminants from multiple routes of exposure, which may have introduced additional variability not explained by the biota–sediment regressions. Despite the relatively high unexplained variation, the significant correlations between tissue and sediment concentrations are consistent with dietary studies indicating shiner surfperch forage on benthic invertebrates (Bane 1970; Jahn 2008), resulting in indirect exposure to sediment contamination. The small optimum scale of exposure (1 km) combined with a

Table 1. Results of linear regression analysis of log-transformed sediment concentrations (dry wt or organic carbon normalized) versus log-transformed *Macoma nasuta* tissue concentrations (wet wt or lipid normalized). All samples were analyzed in 28-d laboratory bioaccumulation tests performed with California sediments. In all cases, N = number of locations sampled for both sediment and *M. nasuta* in a given water body. All slopes were significant and positive with $p < 0.001$, except normalized tPAHs in San Francisco Bay where $p = 0.22$. Data necessary for normalizations were not available for Newport Bay[a]

		Raw data[b]			Normalized to sediment organic carbon and tissue lipid[b]		
Water body	Contaminant	*N*	r^2	BAF ± SD	*N*	r^2	BSAF ± SD
Newport Bay	*p,p′*-DDE	11	0.74	0.3 ± 0.23	—	—	—
San Diego Bay	tHPAHs	14	0.75	0.2 ± 0.23	14	0.73	0.6 ± 0.6
San Diego Bay	tPCBs	14	0.93	0.2 ± 0.14	14	0.92	0.4 ± 0.22
San Francisco Bay	tChlordanes	37	0.74	1 ± 0.76	37	0.53	2 ± 1.2
San Francisco Bay	Dieldrin	38	0.49	2 ± 3.5	37	0.40	2 ± 2.4
San Francisco Bay	tDDTs	38	0.42	1 ± 1.0	37	0.29	1 ± 0.56
San Francisco Bay	tHPAHs	75	0.23	0.3 ± 0.49	37	0.04	Not calculated
San Francisco Bay	tPCBs	37	0.72	0.7 ± 0.53	37	0.62	0.8 ± 0.47

[a] tPAH = total polycyclic aromatic hydrocarbon; *p,p′*-DDE = *p,p′*-dichlorodiphenyldichloroethane; tHPAH = total hydroxylated polycyclic aromatic compounds; tPCB = total polychlorinated biphenyl.
[b] BAF = bioaccumulation factor; BSAF = biota–sediment accumulation factor.

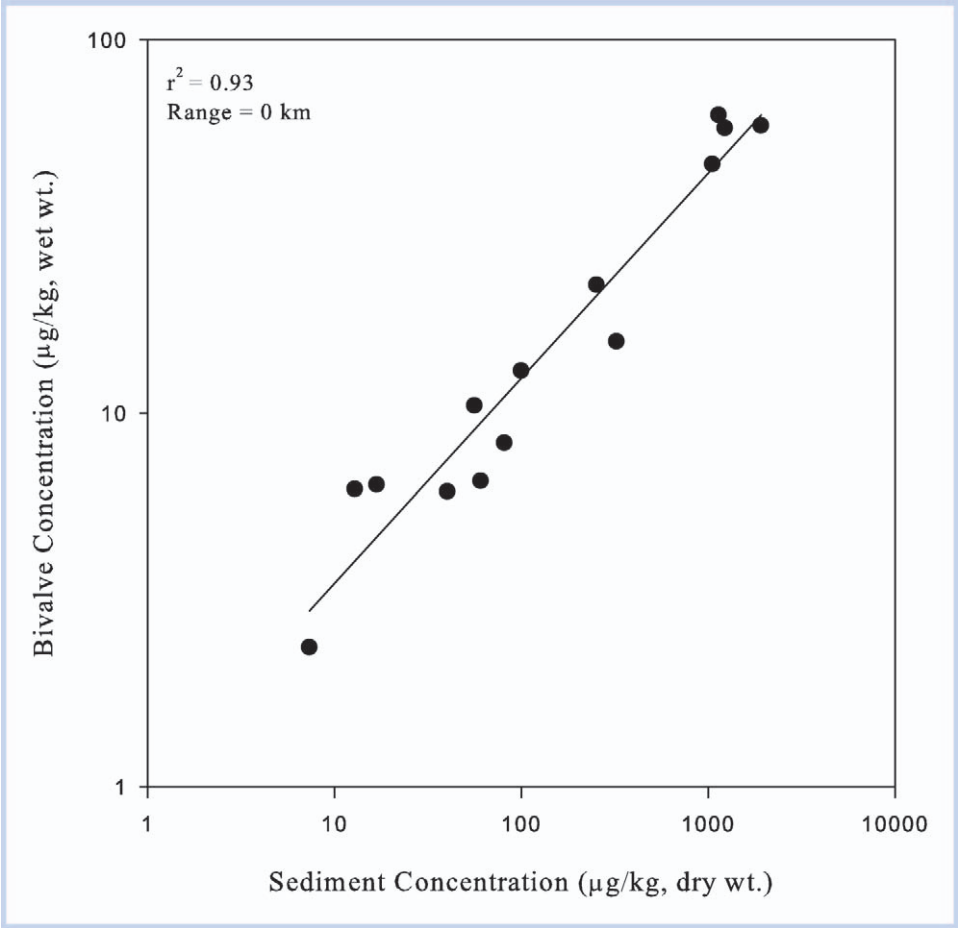

Figure 3. Linear regression of total PCBs in San Diego Bay sediments versus *Macoma nasuta* exposed to those sediments in 28-d laboratory tests. Note log scale.

benthic association suggests that shiner surfperch would be an appropriate candidate species for estimating bioaccumulation for San Francisco Bay sediment contaminants.

In the final example (offshore coastal bioaccumulation), the statistical procedure was used to evaluate biota–sediment relationships from the Southern California Bight using data on 2 fish species. White croaker and kelp bass (*Paralabrax clathratus*) both exhibited statistically significant, positive correlations between sediment concentrations and corresponding fish tissue concentrations for DDTs and PCBs (Table 4). Kelp bass relationships were optimal at spatial scales of 2 km, whereas white croaker relationships were optimal at 10 km (Figure 5), suggesting that white croaker from the SCB may be exposed to contamination over broader areas than kelp bass. Close relationships between fish tissue and sediments in the SCB have been shown previously for DDTs and PCBs in sanddabs and kelp bass (Allen, Moore, et al. 2002; MSRP 2002; Allen et al. 2004). The relatively low r^2 for kelp bass, as compared with white croaker in the SCB, is consistent with dietary studies indicating kelp bass to be a piscivorous species with more pelagic food sources than white croaker (Emmett et al. 1991; Connolly and Glaser 1997).

A number of species examined in San Francisco Bay did not show significant relationships for some contaminants (Table 2). This was observed both with the discrete averaging and kriged sediment data. This may indicate that fish foraging ranges were large enough to obscure spatial patterns in contaminant exposure at the scales examined (<10 km). For example, in contrast to the SCB example (Table 4), results for white croaker in San Francisco Bay varied considerably by contaminant, with biota concentrations generally showing nonsignificant relationships to sediment. When sediment data from San Francisco Bay were spatially averaged using kriging (as performed for SCB sediments) and paired to white croaker, the same nonsignificant relationships were found (results not presented). Evidence suggests that white croaker are resident in bays and estuaries for the majority of the year, with some emigration to the coastal ocean during winter months (Fleming 1999). White croaker are also known to feed at multiple trophic levels that include fish, squid (*Loligo* spp.), and benthic crustaceans (Emmett et al. 1991), which may have introduced other sources of variation to their relationships with bulk sediment concentrations.

The statistically significant SCB white croaker regressions may have been driven by exposure to strong spatial gradients of organochlorine contamination in sediments and associated prey items of the Palos Verdes Shelf (USEPA 2003), compared with less-variable conditions and possibly different food items in San Francisco Bay. Although spatial variation in PCBs and legacy pesticides is present in San Francisco Bay, the ambient range of concentrations does not approach the 4 orders of magnitude variability seen in the SCB (USEPA 2003; Connor et al. 2007; Davis et al. 2007).

Biota–sediment relationships in California halibut were statistically significant from San Diego Bay, but not San Francisco Bay. Despite the potential for offshore movement,

Table 2. Results of linear regression analysis of log-transformed sediment concentrations (dry wt) versus log-transformed fish tissue concentrations (wet wt). An asterisk (*) indicates a significant positive relationship ($p < 0.05$). N = number of locations. All significant slopes were positive and were used to calculate BAFs; BSAFs were not calculated for these data because regressions were generally not statistically significant[a]

Water body	Species[b]	Contaminant	N	r^2	p value	Spatial scale (km)	BAF ± SD
San Diego Bay	California halibut	tDDTs	11	0.63	0.003*	4	3 ± 0.6
San Diego Bay	California halibut	tPCBs	11	0.86	<0.001*	4	4 ± 1.3
San Francisco Bay	California halibut	Dieldrin	23	0.15	0.07	2	Not calculated
San Francisco Bay	California halibut	tDDTs	18	0.18	0.08	1	Not calculated
San Francisco Bay	California halibut	tPCBs	18	0.19	0.07	1	Not calculated
San Francisco Bay	English sole	Dieldrin	12	0.17	0.18	10	Not calculated
San Francisco Bay	English sole	tDDTs	12	0.41	0.03*	5	3 ± 2
San Francisco Bay	English sole	tPCBs	11	0.15	0.24	2	Not calculated
San Francisco Bay	Shiner surfperch	Chlordanes	36	0.25	0.002*	1	8 ± 8.7
San Francisco Bay	Shiner surfperch	Dieldrin	41	0.33	<0.001*	1	4 ± 4.0
San Francisco Bay	Shiner surfperch	tDDTs	41	0.44	<0.001*	1	5 ± 4.5
San Francisco Bay	Shiner surfperch	tPCBs	39	0.33	<0.001*	1	9 ± 6.8
San Francisco Bay	Pacific staghorn sculpin	tDDTs	22	0.04	0.4	1	Not calculated
San Francisco Bay	Pacific staghorn sculpin	tPCBs	22	0.73	<0.001*	1	5 ± 2.4
San Francisco Bay	White croaker	Chlordanes	33	0.17	0.02*	5	18 ± 10
San Francisco Bay	White croaker	Dieldrin	17	0.36	0.01*	1	11 ± 4.2
San Francisco Bay	White croaker	tDDTs	16	0.00	0.91	1	Not calculated
San Francisco Bay	White croaker	tPCBs	15	0.36	0.02*	1	16 ± 5.7

[a] BAF = bioaccumulation factor; BSAF = biota–sediment accumulation factor.
[b] California halibut = *Paralichthys californicus*; English sole = *Pleuronectes vetulus*; shiner surfperch = *Cymatogaster aggregata*; Pacific staghorn sculpin = *Leptocottus armatus*; white croaker = *Genyonemus lineatus*.

DDTs and PCBs from San Diego Bay halibut were significantly related to contaminants in nearby sediments (Table 2). Both California halibut and English sole (*Pleuronectes vetulus*) exhibited varied results in San Francisco Bay, with only the relationship of DDT in English sole being significant. Although flatfishes are benthic-dwelling and sediment-foraging organisms (Emmett et al. 1991), they are known to vary their foraging range depending on stage of development and time of year. For example, juvenile California halibut (<8 inches in length) are thought to remain relatively localized in bays and estuaries (Frey 1971), whereas adult halibut generally migrate to deeper waters, with individual average lifetime movements of 13 km (Domeier and Chun 1995). In general, adult flatfishes limit movements to seasonal onshore–offshore migrations, being generally resident within a given season (Emmett et al. 1991). California halibut and English sole both indicated biota–sediment relationships that were optimal at intermediate spatial scales (4–5 km), consistent with the relatively broad foraging ranges for the species.

Normalization of tissue data for lipid content or sediment data for TOC did not improve the biota–sediment correlations, except for analyses conducted in example 3 (Offshore Coast). Nonsignificant correlations between lipids and PCBs have also been observed for salmonid (*Oncorhynchus* spp.) species (Stow 1995, Stow et al. 1997). These findings may be attributable to low within-species variability in tissue lipid content for the fish species examined, but the lipid-determination method may also be a source of inconsistency (Landrum and Fisher 1999). Lipid methods have previously been identified as a significant source of variation when combining multiple data sets for development of biota–sediment relationships (Naber et al. 2007) and subsequent comparison of BSAF among species (Pruell et al. 1993).

Another consideration often ignored when selecting species for predicting bioaccumulation is that fish and invertebrates with benthic life histories may feed selectively within the sediment matrix. Consequently, such species may bioaccumulate contaminants that do not represent bulk sediment concentrations. Boese et al. (1996) found *M. nasuta* to ingest sediment particles that are higher in TOC and contaminants than the bulk sediment as a function of selective deposit feeding. This may explain why correlations made with TOC-normalized concentrations explained slightly less variation than those based on wet weight (Table 1). Boese et al. (1997) found BSAF to be more variable than BAF for PCBs in laboratory experiments with *M. nasuta*. Furthermore, fractions of a contaminant associated with the bulk sediment may actually be retained and, hence, not available for assimilation. Incorporation of the nonbioavailable fraction of contaminants

Table 3. Selected literature sources indicating significant relationships between sediment and fish tissue contaminant concentrations

Species[a]	Contaminants with significant sediment association	Source
Shiner surfperch	DDTs	Lee et al. (1994)
Sanddab guild, California halibut	PCBs, DDTs	Allen, Groce, et al. (2002); Allen, Moore, et al. (2002)
White croaker	PCBs, DDTs, chlordanes	Connolly and Glaser (1997)
Shorthorn sculpin	PCBs	Kuzyk, Hodson, et al. (2005); Kuzyk, Stow, et al. (2005)
White croaker, English sole	Hg, lead	Meador et al. (2005)
White croaker, four-horn sculpin, flathead sole, English sole, starry flounder, hornyhead turbot, barred sand bass, and black croaker	PCBs, DDTs, chlordanes, dieldrin, PAHs, hexachlorobenzene	Brown et al. (1998)
White sucker, carp, sea bass, and other species	PCBs, dioxins, DDTs, chlordanes	Burkhard et al. (2005); Wong et al. (2001)
Longjaw mudsucker	PCBs, DDTs	Hwang et al. (2006)
Pacific staghorn sculpin, yellowfin goby, and chameleon goby	PCBs	Battelle et al. (2005)

[a] Shiner surfperch = *Cymatogaster aggregata*; sanddab guild = *Citharichthys* spp.; California halibut = *Paralichthys californicus*; white croaker = *Genyonemus lineatus*; shorthorn sculpin = *Myoxocephalus scorpius*; English sole = *Pleuronectes vetulus*; four-horn sculpin = *Myoxocephalus quadricornis*; flathead sole = *Hippoglossoides elassodon*; starry flounder = *Platichthys stellatus*; hornyhead turbot = *Pleuronichthys verticalis*; barred sand bass = *Paralabrax nebulifer*; black croaker = *Cheilotrema saturnum*; white sucker = *Catostomus commersonii*; carp = *Cyprinus carpio*; sea bass = *Lateolabrax japonicus*; longjaw mudsucker = *Gillichthys mirabilis*; Pacific staghorn sculpin = *Leptocottus armatus*; yellowfin goby = *Acanthogobius flavimanus*; chameleon goby = *Tridentiger trigonocephalus*.

into predictions of bioaccumulation introduces an additional explanation for observed variation (Kristensen and Tyle 1991; van der Oost et al. 2003). Normalization of invertebrate PAH concentrations for soot carbon has been used to account for variable BSAFs (Thorsen et al. 2004) and may explain why the organic carbon normalizations of PAH in our analyses with *M. nasuta* indicated a nonsignificant relationship.

Bioaccumulation parameters

Bioaccumulation factors and BSAFs in fish can vary because of multiple factors causing a lack of equilibrium to sediments and the water column (Burkhardt et al. 2003a). These factors include variation in trophic transfer, benthic-pelagic coupling, and metabolic breakdown of contaminants (Morrison et al. 1996; Wong et al. 2001; Burkhard et al. 2004). The effects of differing conditions, parameters, and feeding habits upon the values of BSAFs were captured in a survey by Wong et al. (2001), where measured BSAFs for white suckers (*Catostomus commersonii*) ranged from 1.7 to 27 (with a median value of 8.8) for *p,p′*-dichlorodiphenyldichloroethene across 36 different ecosystems. Because of the high variability among systems, site-specific BAFs or BSAFs are desirable because they incorporate local processes influencing bioaccumulation at the site.

In some instances, bioaccumulation factors have been developed by pairing sediment and organism samples collected from the same location (e.g., Ankley et al. 1992; Schiff and Allen 2000), which does not account for the exposure range of the organism. In this study, BAFs and BSAFs were calculated based on samples within sediment areas specified by a statistical optimization routine. Our assumption was that this method would provide a more precise prediction of bioaccumulation because observations that were more representative of exposure area were used to calculate bioaccumulation parameters. The ensuing values differed among species and contaminants (Tables 1, 2, and 4). Correlations based on the strongest statistical relationships (e.g., California halibut in San Diego Bay and white croaker in SCB) exhibited BAFs of 4 to 5 in fish and of <2 in *M. nasuta* (Boese et al. 1995). Higher BAF and BSAF values and greater variability were shown for species exhibiting weaker biota–sediment relationships and less physical or trophic connection to sediments. The best example is kelp bass in SCB, which is a pelagic piscivore (Emmett et al. 1991), and exhibited higher average BSAF and larger standard deviation, compared with white croaker (Table 4). This likely resulted from generally higher and more variable biomagnification because of elevated trophic position (Kidd et al. 1998). These results highlight the value of selecting species with benthic diets and life histories for sediment risk assessments.

Results for *M. nasuta* were indicative of species that are predominantly exposed to contaminated sediments, having concentrations that are closer to equilibrium with sediment conditions (BAFs = 1; Boese et al. 1995). BSAFs are expected to range from 1 to 2 when thermodynamic equilibrium is reached (Ankley et al. 1992; Moore et al. 2005). The BAFs and BSAFs calculated for *M. nasuta* in this study were generally lower than that expected based on equilibrium (i.e., <1; Table 1). This may reflect the limited time duration of the 28-d tests, such that thermodynamic equilibrium was not reached (Pruell et al. 1993; Boese et al. 1995).

The low variability in BAFs for fish species showing strong biota–sediment regressions suggests that some of the uncertainty can be reduced through the use of the optimization

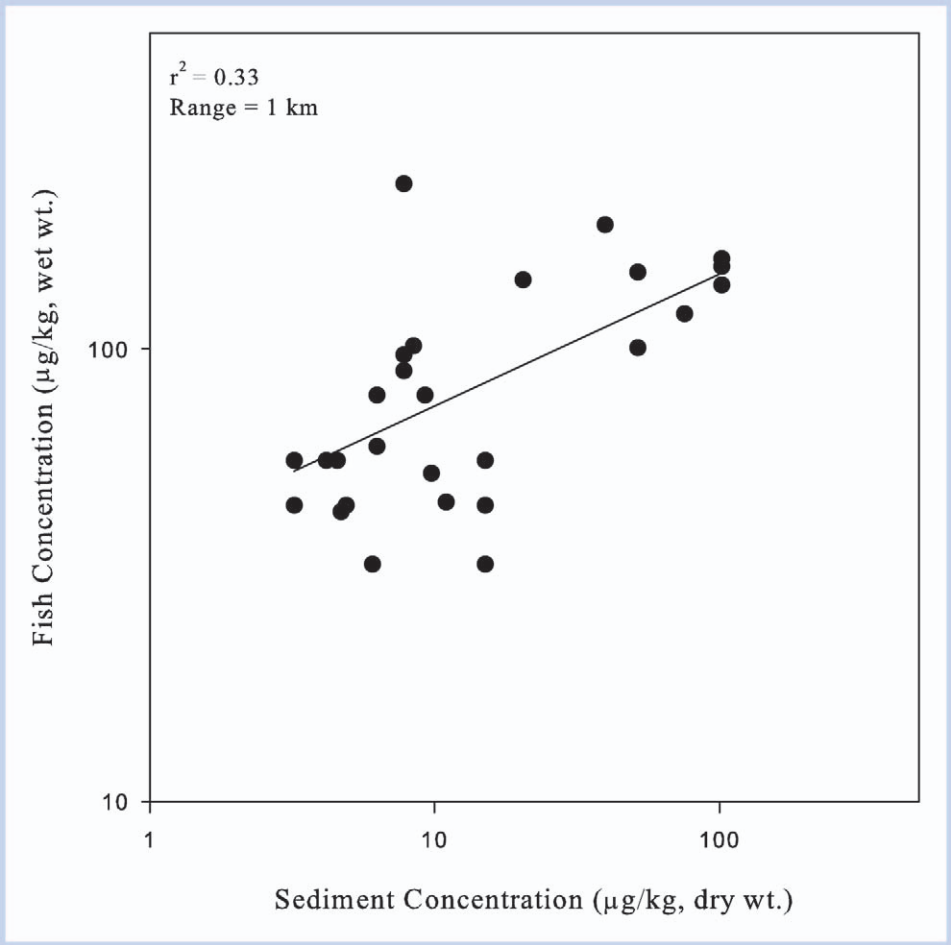

Figure 4. Linear regression of sediment and shiner surfperch (*Cymatogaster aggregata*) total PCBs in San Francisco Bay. Note log scale.

procedure. Variability in BAF, except for a few cases, was reduced to within an order of magnitude (Table 2). Furthermore, standard deviations of BAFs at the optimal scale for biota–sediment relationships were often lower than the variability of BAFs at other spatial scales (Table S1; *Supporting Information*, http://dx.doi.org/10.1897/2008-033.S1). For example, white croaker tPCBs from San Francisco Bay exhibited a coefficient of variation (CV) of 34% at 1 km (the optimal scale), 186% at 2 km, and 70% at 5 km.

Table 4. Results of linear regression analysis of log-transformed sediment concentrations versus log-transformed fish tissue concentrations in the Southern California Bight. N = number of locations. All slopes were significant and positive with $p < 0.001$

Species[a]	Contaminant	N	r^2	Spatial scale	BSAF ± SD[b]
White croaker	tDDTs	220	0.77	10 km	4 ± 5.5
White croaker	tPCBs	199	0.64	10 km	5 ± 5.5
Kelp bass	tDDTs	153	0.37	2 km	8 ± 13.2
Kelp bass	tPCBs	153	0.31	2 km	27 ± 59

[a] White croaker = *Genyonemus lineatus*; kelp bass = *Paralabrax clathratus*.
[b] BSAF = biota–sediment accumulation factor.

Although exceptions existed in some cases, variability was generally found to be lowest when the biota–sediment relationship was optimal (specifically, in 6 of 11 comparisons). These results suggest the utility of the procedure for calculation of bioaccumulation parameters with less variability than those based on a presumed correlation between samples.

CONCLUSIONS

Using data from multiple water bodies in California, this study has shown that significant biota–sediment relationships may be obtained by optimizing the spatial scale of exposure to fit the most likely exposure area of biota. The procedure identified spatial scales that appear appropriate based on the known life-histories of the species examined. Nevertheless, the correlations were often weak, suggesting that this procedure is not a panacea for the substantial complexity of contaminant transfer between sediments, the overlying water column, and food webs.

Biota–sediment accumulation factors have been commonly used for regulatory decision making and environmental risk assessment (Kraaij et al. 2002; USEPA 2003). However, our results, based on degree of correlation between biota and sediment concentrations, corroborated the findings of Boese et al. (1997) that BAF can be a less-variable estimate of bioaccumulation. This is particularly the case when combining data from multiple studies that employ multiple lipid-determination methods (Landrum and Fisher 1999).

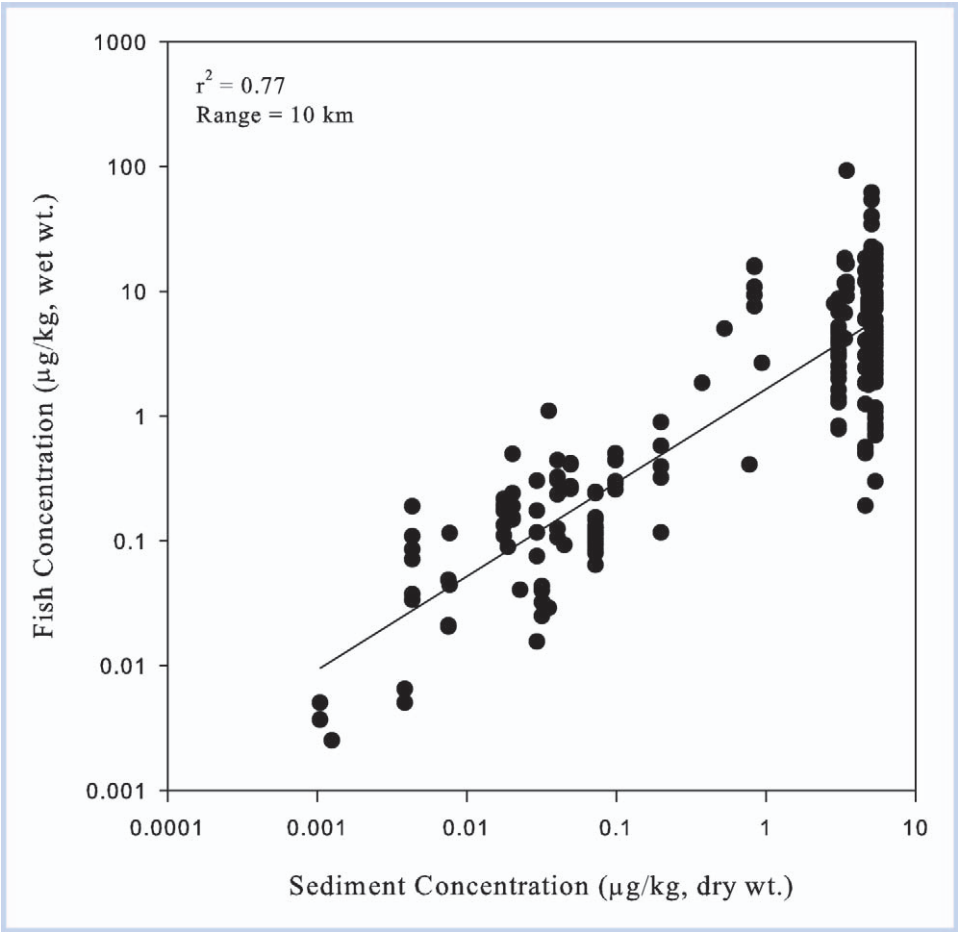

Figure 5. Linear regression of sediment and white croaker (*Genyonemus lineatus*) total DDTs in the Southern California Bight. Note log scale.

We propose that this spatial optimization procedure may have 3 benefits for estimation of BAFs and BSAFs in risk assessments and regulatory programs: 1) identification of the appropriate spatial scale of biota exposure to sediments, particularly when life history data are lacking and when biota may be expected to range across a large area; 2) identification of species with relatively strong spatial association to sediment contamination, based on relatively strong correlation between sediment and biota concentrations; and 3) development of a data set for determining empirical BAFs or BSAFs when biota and sediment sampling were not colocated (as is frequently the case when combining sediment and fish chemistry databases). As in all contaminant risk assessments, this approach should be considered as one of many potential tools that may be employed, depending on factors such as the contaminants of concern, assessment endpoints, and available resources (Bridges et al. 2005).

SUPPORTING INFORMATION

Table 1S. Comparison of bioaccumulation factors (BAFs) at varying spatial scales of biota–sediment association.

Found at DOI: 10.1897/2008-033.S1 (10 KB PDF).

Acknowledgment—We thank Christine Arenal (CH2M HILL), Harry Ohlendorf (CH2M HILL), John Oram (SFEI), and Fred Shauffler (EPA Region 9) for review and advice in implementing the model. We are grateful to M.S. Connor (SFEI), S. Klosterhaus (SFEI), and C. Lutz (US Army Corps of Engineers) for their constructive reviews of the draft manuscript. The manuscript was enhanced by the comments of two anonymous reviewers. Funding for this study was provided by CH2M HILL and the Southern California Coastal Water Research Project.

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