EPA Response to Integral Consulting Inc. Memorandum
Portland Harbor PDI; fish tissue-sediment relationship exploration
Dated August 28, 2019

Response dated September 6, 2019

Following is the United States Environmental Protection Agency's (EPA's) response to the document titled *Portland Harbor PDI; fish tissue-sediment relationship exploration* (Memorandum) prepared by Integral Consulting Inc. (Integral) on behalf of the Portland Harbor Pre-Remedial Design Group (Pre-RD Group).

1 Introduction

The *PDI Evaluation Report* (AECOM Technical Services [AECOM] and Geosyntec Consultants, Inc. [Geosyntec] 2019) and an August 28, 2019 memorandum (Integral 2019) prepared by the Pre-RD Group suggest that contaminant concentrations in fish tissues are unrelated to contaminant concentrations in sediment. In their analysis, sediment data were paired with fish tissue data by averaging over relatively large areas including data from both sides of the river irrespective of the location where the fish were captured, or on the other extreme, averaging sediment samples within 100 feet. To evaluate the approaches used by the Pre-RD Group, EPA developed a series of maps showing interpolated sediment concentrations and fish tissue concentrations throughout the Portland Harbor Superfund Site (site) and performed statistical analyses linking contaminants in fish tissue to those in sediment. The site maps were developed for the five focused contaminants of concern (COCs) that are bioaccumulative organic chemicals and show that the highest sediment concentrations for a COC also have elevated fish tissue concentrations (**Supplemental Data Figures**). The statistical analyses were based on averaging of sediment sample locations over spatial scales ranging from within 100 feet up to 5200 feet (i.e., within 1 mile of the capture location), and restricted averaging to the side of the river, where the relevant fish were captured (East, West or Navigation Channel). The statistical models fit to the data treated fish tissue contaminant concentrations as a log-log relationship with sediment contaminant concentration, fish lipid content and sediment organic carbon content. The methods and results of our analysis are described in the following sections.

2 Methods

Simple bioaccumulation models for organic contaminants usually relate lipid normalized contaminants in fish to organic carbon normalized contaminants in sediment. This normalization is intended to reflect the differential accumulation rates as they vary as a function of the ratio of lipid in tissue to organic carbon in sediment. Fish exposed to contaminants in sediments with lower organic carbon content are expected to exhibit proportionally higher body burden, relative to fish exposed to contaminants in sediments with higher organic carbon content. Similarly, for fish exposed equally to contaminants, wet weight contaminant concentrations in tissue are expected to be proportionally greater for fish with greater lipid content. The biota to sediment accumulation factor (BSAF) reflecting these relationships is usually given by the following equation:

$$\frac{C_f/f_L}{C_s/f_{OC}} = BSAF \qquad \text{Equation 1}$$

Where

C_f = the contaminant concentration in fish tissue
C_s = the contaminant concentration in sediment
f_L = the fraction of lipid in the fish specimen
f_{oc} = the fraction of organic carbon in sediment

2.1 Generalization of BSAF

Hebert and Keenleyside (1995) found that normalized relationships have some implicit assumptions of a linear regression through the origin that may not hold, and further showed that the relationships can be masked when either tissue contaminant concentrations are independent of lipid content, or when organic carbon is independent of sediment contaminant concentrations. Generally, they recommended restructuring normalized relationships as regression models and testing for the requisite relationships before arbitrarily normalizing measurements. EPA followed this advice by noting that the BSAF relationship can be decomposed into a multiple linear regression model which provides a framework for testing the contribution of each term in the BSAF model for relevance in predicting fish tissue concentrations from sediment exposure. By taking natural logarithms of both sides of the BSAF relationship the equation can be expressed in more general form:

$$LN\left(C_f\right) = LN(BSAF) + \beta_1 LN(f_L) + \beta_2 LN(f_{OC}) + \beta_3 LN(C_s) \qquad \textit{Equation } 2$$

When $\beta_1 = -\beta_2 = \beta_3 = 1.0$, this multiple regression simplifies to the usual BSAF equation. However, when coefficients differ from 1.0, the usual BSAF relationship is inappropriate, because the relationship may be non-linear. When $\beta_1 = 0$ or $\beta_2 = 0$, accumulation should be expressed independently of fish lipid content or sediment organic carbon content respectively. Finally, a relationship between fish tissue contaminant concentration and sediment contaminant concentration is evaluated by testing the null hypothesis (H_0: $\beta_3 = 0$). Reorganizing Equation 2, the accumulation relationship is similar in form to the BSAF with allowance for non-linear relationships and potential that some variables may not be informative in estimating fish tissue concentrations from exposure to contaminated sediments.

$$C_f = (BSAF) \times f_L^{\beta_1} \times f_{OC}^{\beta_2} \times C_s^{\beta_3} \qquad \textit{Equation } 3$$

In this form, concentrations in fish tissue are proportional to powers of lipid, organic carbon, and sediment concentrations. The BSAF is the special case of this relationship found by setting $\beta_1 = -\beta_2 = \beta_3 = 1.0$ as described above.

EPA fit Equation 2 to data using the maximum likelihood estimation assuming that the log of mean fish tissue concentration is linear in logarithm of fraction lipid, fraction organic carbon, and contaminant concentration in sediment. Maximum likelihood estimation is needed for these models because usual least squares fitting would result in biased estimates of the mean and would more tightly constrain the residuals to be log-normally distributed.

2.2 Scale of Fish to Sediment Relationships

The sediments to which fish are exposed is uncertain because the life history of the fish that were sampled is unknown. EPA averaged sediment samples over a range of distances from the capture location from 100 feet up to 5,280 feet (i.e., 1-mile radius or 2 river miles overall) in increments of 100 feet and tested the null hypothesis (H_0: $\beta_3 = 0$) for each scale of averaging. EPA then plotted the statistical level of significance against scale of averaging to evaluate what scale of averaging provided the strongest evidence of a relationship between fish tissue concentration and sediment concentration. EPA applied this approach to the five focused COCs that are bioaccumulative organic contaminants at the site and present the results in the following section. Finally, EPA tabulated the model results for the scale of averaging that was best for each model.

3 Results

The results from the statistical analyses for the five bioaccumulative organic focused COCs is included in the subsections below.

3.1 Association Between Fish and Sediment Contaminant Concentrations

For all five organic contaminants, EPA found statistically significant positive associations between fish tissue concentrations and average sediment concentrations (i.e., $\beta_3 > 0; p < 0.05$) when sediments included for averaging were restricted to the side of the river where the fish was captured, and when the radius of averaging was relatively small—on the order of 100 to 600 feet (**Figure 1**). The strongest relationships were found for averaging areas less than 500 feet for all 5 contaminants suggesting that fish exposures were dominated by sediment concentrations proximal to capture locations as opposed to averages representing larger exposure areas.

Tissue concentrations of 1,2,3,7,8-PeCDD were unrelated to sediment contaminant concentrations when averaging radii were greater than approximately 600 feet (p>0.05). For the other 4 organic contaminants fish tissue contaminant concentrations were also associated with sediment concentrations averaged over larger sediment averaging areas, although strength of relationships decreased with increasing averaging area size.

3.2 Best Models

The models fit to the data provide a framework to explain contaminant variability in tissue due to variation in exposure to contaminants in sediment, as well as co-variation with lipid content in fish and organic carbon content in sediment. Both lipid and organic carbon are expected to influence accumulation of organic contaminants. Our analysis provided a way to account for these components of variation, which in general results in more precise estimation of the relationship between tissue and sediment contaminant concentrations. For the spatial averaging scales found to result in the strongest relationships, EPA summarized the full models in **Tables 1** through **5** below. For all five organic contaminants, tissue concentrations were independent of lipid content, indicating that traditional lipid normalization is unnecessary and could tend to mask relationships between tissue and sediment concentrations based on traditional BSAF-like analyses, or linear regressions between normalized concentrations (Hebert and Keenleyside, 1995).

3.2.1 1,2,3,7,8-PeCDD

Concentrations of 1,2,3,7,8-PeCDD in fish tissue were positively associated ($\beta_3 = 0.46; p < 0.001$) with sediment concentrations and negatively associated with organic carbon content ($\beta_2 = -0.39; p = 0.03$). The coefficient for sediment differed from 1.0, and that for organic carbon differed from -1.0, indicating that contaminant accumulation in tissue is nonlinear in both sediment concentration and organic carbon, in contrast to the usual assumptions of linearity with the BSAF model. The coefficient on sediment (β_3) is less than 1.0 indicating that the ratio of fish to sediment concentrations declines with increasing contaminant concentrations. For organic carbon, because the coefficient is greater than negative 1 (as assumed in the BSAF model) accumulation per unit of organic carbon is less than would be predicted by the BSAF model.

3.2.2 2,3,4,7,8-PeCDF

Concentrations of 2,3,4,7,8-PeCDF in fish tissue were positively associated with sediment concentrations ($\beta_3 = 0.67; p < 0.001$) and negatively associated with organic carbon content ($\beta_2 = -0.46; p = 0.008$). As with 1,2,3,7,8-PeCDD, both coefficients differed from 1.0 and -1.0, respectively indicating a non-linear relationship between tissue concentrations, sediment concentrations, and organic carbon. Again, these data indicate that accumulation is less at higher sediment contaminant concentrations than would be predicted with a linear accumulation model. This non-linearity in orgainic carbon also indicates that changes in accumulation are less responsive to changes in organic carbon than a linear model would predict.

3.2.3 2,3,7,8-TCDD

Tissue 2,3,7,8-TCDD concentration was positively associated with sediment concentration content ($\beta_3 = 0.16; p = 0.005$) but unrelated to both fish lipid (p=0.34) and sediment organic carbon (p=0.28). As with the other orgainc contaminants, accumulation is lower at higher sediment concentrations than would be predicted by a linear regression or BSAF-based accumulation model.

3.2.4 Total DDx

Total DDx concentrations in fish tissue were positively associated with DDx in sediment ($\beta_3 = 0.23; p = 0.002$) but unrelated to fish lipid content (p=0.68) or sediment organic carbon (p=0.67).

3.2.5 Total PCBs

As with total DDx, fish tissue total PCB concentrations were positively associated with sediment total PCB concentrations ($\beta_3 = 0.23; p = 0.002$) but unrelated to fish lipid and sediment organic carbon. As with the other organic contaminants, accumulation from sediment is lower at higher levels of sediment PCBs than would be predicted with a linear regression or BSAF accumulation model.

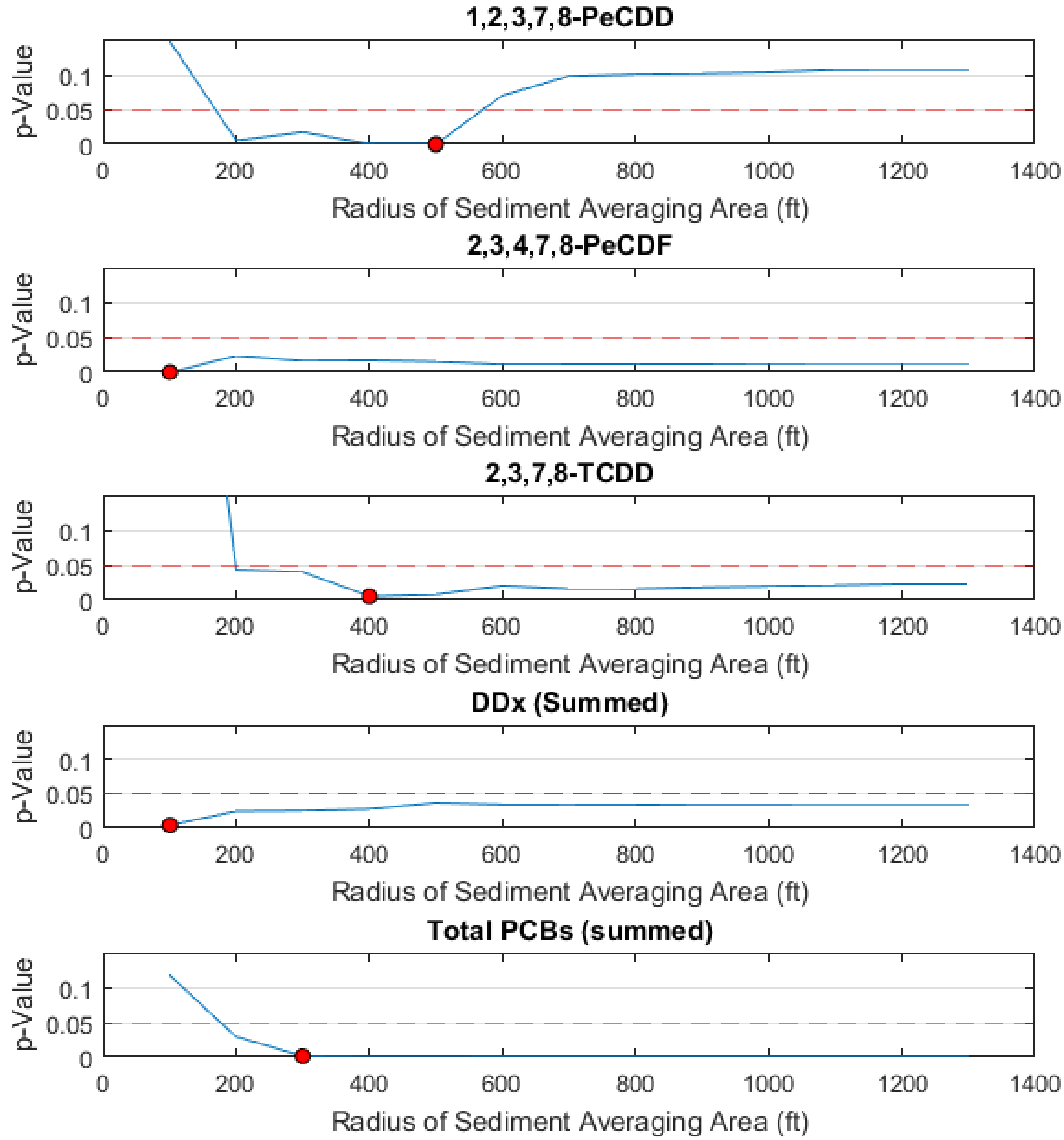

Figure 1. Significance of sediment concentration as a predictor of tissue concentration versus sediment averaging radius restricted to the side of the river where fish were captured at the site, the Downtown Reach, and the Upriver Reach (reference area). The blue line shows the statistical significance of the relationship at different averaging distances. The red dot represents the sediment averaging distance where the statistical significance of the relationship was strongest (i.e., lowest p-value).

Table 1. Generalized linear model coefficient estimates and tests of association and linearity for fish lipid, sediment organic carbon, and sediment contaminant concentration for 1,2,3,7,8-PeCDD averaged over points within 500 feet of the fish capture location on the same side of the river.

			Test of Association		Test of Linearity	
Parameter	Estimate	SE	T-Stat	Significance	T-Stat	Significance
(Intercept)	-7.06	1.58	-4.47	<0.001		
Log(Lipid)	-0.63	0.45	-1.40	0.166	-3.61	<0.001
Log(OC)	**-0.39**	0.18	-2.20	**0.030**	3.49	**0.001**
Log(Cs)	**0.46**	0.12	3.88	**<0.001**	-4.49	**<0.001**

Table 2. Generalized linear model coefficient estimates and tests of association and linearity for fish lipid, sediment organic carbon, and sediment contaminant concentration for 2,3,4,7,8-PeCDF averaged over points within 100 feet of the fish capture location on the same side of the river.

			Test of Association		Test of Linearity	
Parameter	Estimate	SE	T-Stat	Significance	T-Stat	Significance
(Intercept)	-6.67	1.85	-3.61	0.0009		
Log(Lipid)	-0.98	0.58	-1.70	0.10	-3.43	0.002
Log(OC)	-0.46	0.16	-2.80	**0.0081**	3.28	**0.002**
Log(Cs)	**0.67**	0.09	7.20	**0.0000**	-3.47	**0.001**

Table 3. Generalized linear model coefficient estimates and tests of association and linearity for fish lipid, sediment organic carbon, and sediment contaminant concentration for 2,3,7,8-TCDD averaged over points within 400 feet of the fish capture location on the same side of the river.

			Test of Association		Test of Linearity	
Parameter	Estimate	SE	T-Stat	Significance	T-Stat	Significance
(Intercept)	-6.88	0.89	-7.74	0.0000		
Log(Lipid)	-0.24	0.25	-0.96	0.3419	-4.97	0.0000
Log(OC)	0.10	0.09	1.09	0.2782	12.4	0.0000
Log(Cs)	**0.16**	0.05	2.89	**0.0047**	-15.31	0.0000

Table 4. Generalized linear model coefficient estimates and tests of association and linearity for fish lipid, sediment organic carbon, and sediment contaminant concentration for total DDx averaged over points within 100 feet of the fish capture location on the same side of the river.

			Test of Association		Test of Linearity	
Parameter	Estimate	SE	T-Stat	Significance	T-Stat	Significance
(Intercept)	2.87	2.00	1.44	0.1584		
Log(Lipid)	-0.27	0.63	-0.42	0.6763	-2.00	0.0516
Log(OC)	-0.08	0.19	-0.44	0.6656	4.71	<0.001
Log(Cs)	**0.30**	0.10	3.07	**0.0037**	-7.18	<0.001

Table 5. Generalized linear model coefficient estimates and tests of association and linearity for fish lipid, sediment organic carbon, and sediment contaminant concentration for total PCBs averaged over points within 300 feet of the fish capture location on the same side of the river.

			Test of Association		Test of Linearity	
Parameter	Estimate	SE	T-Stat	Significance	T-Stat	Significance
(Intercept)	4.40	1.44	3.05	0.0027		
Log(Lipid)	-0.36	0.41	-0.90	0.3705	-3.36	0.0009
Log(OC)	0.05	0.15	0.34	0.7354	6.91	<0.001
Log(Cs)	**0.23**	0.07	3.21	**0.0016**	-10.88	<0.001

4 Discussion

These analyses indicate that bioaccumulative contaminants in fish are best described by the concentration in sediments near where they were captured. This contrasts with the conclusions drawn in the *PDI Evaluation Report* based on the contaminant data and acoustic fish tracking study (AECOM and Geosyntec 2019). The acoustic fish tracking data suggest that the home ranges of smallmouth bass within the lower Willamette River appear to be larger than the optimal sediment averaging areas, but nonetheless the scale of sediment averaging is smaller than the home range sizes. This at first may seem contradictory, although there are several reasons why contaminant exposure areas may not align fully with the size of an organism's home range.

Visual inspection of the capture locations shows that smallmouth bass which were caught by hook and line (i.e., while they were feeding) were consistently along the shallower shoals where sediment contaminant concentrations are highest as opposed to within the deeper navigation channel where contaminants are generally lower. The acoustic fish tracking data indicate that smallmouth bass home ranges were generally larger than the 100 to 600-foot radius sediment averaging areas evaluated in our statistical analyses, including time spent in deeper water in the navigation channel. These observations are consistent with previous studies on smallmouth bass behavior in the lower Willamette River (Pribyl et al. 2004) which show that the fish generally cycle between feeding in shallow waters that have more contaminated sediments and retreating to deeper adjacent waters that have more consistent temperatures year-round. With such a cycle, and with a substantial component of tissue contaminant burden likely originating from dietary exposure rather than aqueous exposure (Streit 1988), one would expect the sediments in feeding areas on the shoals where food items are exposed to better reflect tissue concentrations in the fish—consistent with our findings that intermediate to small scales of averaging provide the best predictor of tissue concentrations.

Developing empirical relationships in contaminant concentrations in tissue and sediment are plagued by several difficulties: 1) the sediments which provide a source directly and indirectly to fish exposure are unknown; 2) measurements of average concentration in these sediments even when known are variable; and 3) the degree to which exposure is apportioned between dietary uptake, direct exposure to contaminated sediments, and aqueous uptake are also unknown. Because of these uncertainties, it is expected that relationships estimated from sample data would be uncertain. In general, uncertainty in measurements tends to dampen regression relationships, which would result in accumulation functions that underpredict tissue concentrations based on the available sediment data. Nonetheless, statistically identifiable empirical relationships between sediment and fish tissue contaminant levels have been found for the five focused COCs that are known bioaccumulative organic compounds. Because of the uncertainties described above, EPA expects that these empirical estimates approximate the actual underlying mechanistic relationships that have been studied and developed through controlled experimentation and models and underpredict the resulting tissue concentrations. The *PDI Evaluation Report* argues that the mechanistic food web model (FWM) used to develop sediment cleanup levels at the site overpredicts tissue accumulation because their empirical models indicate less accumulation. It is important to note that the empirical relationships summarized in this memo and the *PDI Evaluation Report* are estimated and are insufficient to replace the mechanistic FWM used to develop sediment cleanup levels at the site. The mechanistic FWM is based on extensive peer-reviewed literature that identifies and quantifies the underlying mechanisms driving bioaccumulation. Furthermore, the mechanistic FWM has been empirically calibrated to site conditions to account for uncertainty in the data. It is our overall conclusion that bioaccumulative contaminants in fish tissue are associated with contaminant concentrations in sediment near the areas where these fish were caught. Smallmouth bass contaminant exposure is driven primarily through dietary uptake and therefore the sediment contaminant concentrations in foraging areas along the shoals are most representative of exposure.

5 Conclusions

These results indicate four primary findings:

1. In contrast to the findings in the *PDI Evaluation Report*, all five bioaccumulative focused COCs in tissue were positively associated with contaminant concentrations in sediment provided that averaging was restricted to the side of the river where fish were collected.
2. The strongest relationships for all contaminants were identified when fish tissue concentrations were paired with sediment concentrations found near fish capture locations.
3. Analyses based on lipid and organic carbon normalized data should be interpreted cautiously and may be counterproductive because they tend to mask relationships between tissue and sediment concentrations.

4. The empirical relationships between fish tissue and sediment concentrations modeled in this memo and in the *PDI Evaluation Report* are an estimate and, in contrast to a FWM, do not incorporate empirical calibration to site conditions, or underlying mechanisms driving bioaccumulation. As, such, the relationships are informative, but insufficient to replace the mechanistic FWM used to develop sediment cleanup levels at the site.

6 References

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