APPENDIX G. TECHNICAL MEMORANDUM REGARDING THE IMPLICATIONS OF THE UPDATED TOXICITY VALUES FOR BENZO(A)PYRENE



TECHNICAL MEMORANDUM: IMPLICATIONS OF UPDATED TOXICITY VALUES FOR BENZO(A)PYRENE

Prepared for

Lower Duwamish Waterway Group

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Acronyms

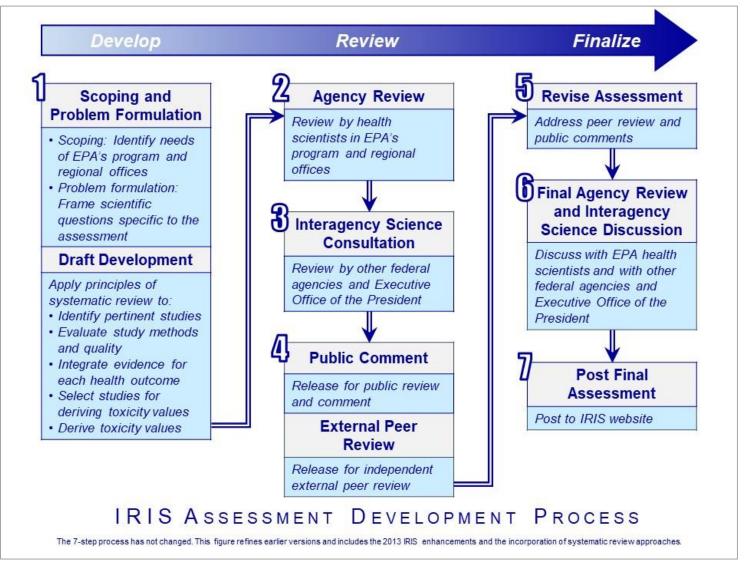
95UCL	95 upper confidence limit
ВаР	benzo(a)pyrene
coc	chemical of concern
сРАН	carcinogenic polycyclic aromatic hydrocarbon
dw	dry weight
EPA	US Environmental Protection Agency
EPC	exposure point concentration
ESD	explanation of significant differences
HHRA	human health risk assessment
HQ	hazard quotient
IRIS	Integrated Risk Information System
LDW	Lower Duwamish Waterway
PAH	polycyclic aromatic hydrocarbon
РСВ	polychlorinated biphenyl
PDI	pre-design investigation
PEF	potency equivalency factor
RAL	remedial action level
RBTC	risk-based threshold concentration
RfD	reference dose
RI/FS	remedial investigation/feasibility study
RM	river mile
RME	reasonable maximum exposure
ROD	Record of Decision
TEQ	toxic equivalent
TTL	target tissue level
ww	wet weight

1 Introduction

The purpose of this technical memorandum is to evaluate the effect of the updated toxicity values for benzo(a)pyrene (BaP) on the cleanup plan for the Lower Duwamish Waterway (LDW). On January 19, 2017, the US Environmental Protection Agency (EPA) finalized its toxicological review of BaP and published updated toxicity values for BaP on the Integrated Risk Information System (IRIS) website (https://www.epa.gov/iris). EPA updated the slope factor for BaP, which is used to estimate excess lifetime cancer risk from exposures to carcinogenic polycyclic aromatic hydrocarbons (cPAHs). EPA also published a reference dose for BaP for non-cancer based on the developmental endpoint (neurobehavioral changes). Both of these toxicity values were published on EPA's IRIS website and documented in the toxicological review for BaP (EPA 2017). The process for updating these values involved more than five years of research, the results of which showed that BaP is less toxic than previously thought for people who contact or ingest the chemical. The IRIS update process is described in Figure 1. For BaP, this process included four draft documents released for agency or public comment from 2011 to 2016.1

¹ Details regarding the BaP review process can be found here: https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=136#tab-3.





Source: https://www.epa.gov/sites/production/files/2015-09/iris_process_figure_2015.jpg

Figure 1. IRIS assessment development process



1.1 BACKGROUND INFORMATION

The 1984 BaP slope factor was used in the LDW human health risk assessment (HHRA) (Windward 2007) to evaluate risks from exposure to cPAHs, identify contaminants of concern (COC), and develop risk-based threshold concentrations (RBTCs) for sediment and tissue. The cPAH RBTCs used in the LDW remedial investigation/feasibility study (RI/FS) (Windward 2010; AECOM 2012) were adopted as sediment cleanup levels and target tissue levels (TTLs) in the Record of Decision (ROD).

Estimations of risks from exposures to cPAHs are calculated using cPAH toxic equivalents (TEQs). The cPAH TEQ is calculated by applying the relative potency equivalency factors (PEFs) that estimate the cancer potency of each of the other cPAH compounds relative to the cancer potency of BaP. Summing the cPAHs after applying the PEFs results in a TEQ with potency equivalent to that of BaP alone. The cPAH TEQ is multiplied by the BaP slope factor to estimate excess cancer risk; thus, changes to its slope factor may have a large effect on cPAH TEQ risk.

1.2 POTENTIAL UPDATES TO THE ROD

Figure 2 presents a flow chart showing how the updated slope factor and new reference dose for BaP could affect the cPAH COC designation, cPAH RBTCs adopted in the ROD as sediment cleanup levels (ROD Table 19²) and TTLs (ROD Table 21³), and the development of the sediment remedial action levels (RALs) (ROD Table 28⁴).

⁴ ROD Table 28 is titled *Remedial Action Levels, ENR Upper Limits, and Areas and Depths of Application* (EPA 2014).



² ROD Table 19 is titled *Cleanup Levels for PCBs, Arsenic, cPAHs, and Dioxins/Furans in Sediment for Human Health and Ecological COCs (RAOs 1, 2 and 4)* (EPA 2014).

³ ROD Table 21 is titled *LDW Resident Fish and Shellfish Target Tissue Concentrations* (EPA 2014). The ROD used the terms "target tissue level" and "target tissue concentration" interchangeably; the term target tissue level (i.e., TTL) is used in this technical memorandum.

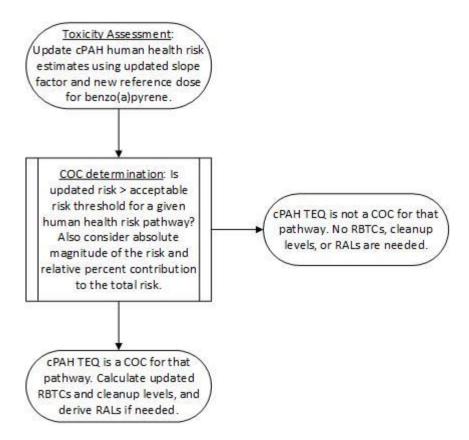


Figure 2. cPAH update flow chart

EPA is considering whether changes to the LDW ROD (EPA 2014) are needed to reflect the updated BaP slope factor and new reference dose. This technical memorandum presents updated cPAH risk estimates and their associated values (e.g., RBTCs and cleanup levels) based on these risk estimates, as well as potential implications for the sediment remedy, to provide context for EPA in the consideration of next steps. The remaining sections of this technical memorandum, which explore potential changes to the ROD, are organized as follows:

- ◆ Section 2 Updated risk calculations and risk conclusions for the scenarios evaluated in the LDW HHRA
- Section 3 Updated RBTCs for these scenarios
- Section 4 Implications for the sediment remedy
- Section 5 Summary of the changes described in Sections 2 to 4
- Section 6 References

2 Updated Risk Calculations and Conclusions

As described in Section 1, the January 19, 2017, update to the BaP toxicity values included the following (Windward 2007; EPA 2019):

- ◆ A revised slope factor EPA changed the slope factor, which is the toxicity value used to calculate excess cancer risks, from the prior value of 7.3 (mg/kg-day)⁻¹ to 1 (mg/kg-day)⁻¹.
- New reference dose EPA published a new reference dose, which is the toxicity value used to calculate non-cancer hazard quotients (HQs), equal to 3 × 10⁻⁴ mg/kg-day. No reference dose was available for BaP prior to the 2017 update.

This section presents updated risk estimates and corresponding COC determinations for cPAHs based on the updated BaP slope factor and new reference dose.⁵

Updated cPAH excess cancer risks and BaP non-cancer HQs for the human health seafood consumption and direct contact scenarios are presented in Tables 2-1 and 2-2, respectively, using the new BaP toxicity values and data from the FS.

Table 2-1. Updated cPAH TEQ and BaP risk estimates for the human health seafood consumption RME scenarios

	Risks as Presente	ed in the LDW FS	Updated Risks with New BaP Toxicit Values (Using FS Dataset) ^a		
Scenario	cPAH Excess Cancer Risk	BaP HQ	cPAH Excess Cancer Risk	BaP HQ	
Adult tribal RME – Tulalip data	8 × 10 ⁻⁵	na ^b	1 × 10 ⁻⁵	0.02	
Child tribal RME – Tulalip data	8 × 10 ⁻⁵	na ^b	1 × 10 ⁻⁵	0.05	
Adult API RME	3 × 10 ⁻⁵	na ^b	4 × 10 ⁻⁶	0.02	

Note: cPAH risk estimates shown in **bold and red** are above the excess cancer risk threshold of 1×10^{-6} . No HQs were above the threshold of 1. Details regarding scenario parameterization were presented in Tables B.3-7 to B.3-12 of the HHRA (Windward 2007) and in Table 1 of the errata to the HHRA (Windward 2009).

- The FS dataset includes tissue data collected from 1990 to 2010 in the LDW. The FS dataset was used as the basis for conclusions in the FS and ROD. EPCs for cPAHs were those presented in Table B.3-38 of the HHRA (Windward 2007). EPCs for BaP, which were calculated for this memorandum, were 0.37 μg/kg for benthic fish fillet, 1.4 μg/kg for whole-body benthic fish, 12 μg/kg for clams, 0.59 μg/kg for edible meat crab tissue, 0.63 μg/kg for whole-body crab tissue, and 0.59 μg/kg for whole-body pelagic fish.
- b No reference dose was available for PAHs including BaP at the time the HHRA and FS were conducted.

API – Asian Pacific Islander HQ – hazard quotient

BaP – benzo(a)pyrene LDW – Lower Duwamish Waterway

cPAH – carcinogenic polycyclic aromatic hydrocarbon na – not applicable

EPC – exposure point concentration PAH – polycyclic aromatic hydrocarbon FS – feasibility study RME – reasonable maximum exposure

⁵ Details regarding the methods used to calculate these risk estimates can be found in Section B.5.1 of the HHRA (Windward 2007). The methods used in this memorandum (including exposure



assumptions) are the same as those used in the HHRA.

Table 2-2. Updated cPAH TEQ risk estimates for the human health direct contact RME scenarios

	EPC (µg/kg	_	Risks as Presented in the FS		Updated Risks with New BaP Toxicity Values (Using FS Dataset)	
Scenario	cPAH TEQ	BaP	cPAH Excess Cancer Risk	BaP HQ	cPAH Excess Cancer Risk	BaP HQ
Netfishing RME	570	380	1 × 10 ⁻⁶	na ^b	2 × 10 ⁻⁷	0.0007
Tribal clamming RME	770	370	5 × 10 ⁻⁶	na ^b	7 × 10 ⁻⁷	0.001
Beach play RME – Area 1	380	280	4 × 10 ⁻⁶	na ^b	6 × 10 ⁻⁷	0.003
Beach play RME – Area 2	7,000	5,000	8 × 10 ⁻⁵	na ^b	1 × 10 ⁻⁵	0.06
Beach play RME – Area 3	1,500	1,400	1 × 10 ⁻⁵	na ^b	2 × 10 ⁻⁶	0.02
Beach play RME – Area 4	1,400	810	1 × 10 ⁻⁵	na ^b	2 × 10 ⁻⁶	0.009
Beach play RME – Area 5	2,200	850	3 × 10 ⁻⁵	na ^b	3 × 10 ⁻⁶	0.009
Beach play RME – Area 6	7,100	4,900	8 × 10 ⁻⁵	na ^b	1 × 10 ⁻⁵	0.05
Beach play RME – Area 7	98	65	1 × 10 ⁻⁶	na ^b	1 × 10 ⁻⁷	0.0007
Beach play RME – Area 8	270	180	3 × 10 ⁻⁶	na ^b	4 × 10 ⁻⁷	0.002

Note: cPAH risk estimates shown in **bold and red** are above the excess cancer risk threshold of 1×10^{-6} . No HQs were above the threshold of 1. Details regarding scenario parameterization were presented in Tables B.3-15 to B.3-26 of the HHRA (Windward 2007).

BaP – benzo(a)pyrene HQ – h

cPAH – carcinogenic polycyclic aromatic hydrocarbon

dw - dry weight

EPC – exposure point concentration

FS – feasibility study

HHRA - human health risk assessment

HQ - hazard quotient

LDW - Lower Duwamish Waterway

na - not applicable

RME - reasonable maximum exposure

TEQ - toxic equivalent

Table 2-3 presents a summary of reasonable maximum exposure (RME) scenarios for which cPAHs or BaP would be a COC based on the new BaP toxicity information (using the FS dataset). Non-cancer HQs for BaP are well below the threshold of 1 for all scenarios (Table 2-1 and 2-2), and therefore, BaP is not a COC. Because excess cancer risks for cPAHs for netfishing and tribal clamming using the new BaP slope factor are less than 1×10^{-6} (Table 2-2), cPAHs would no longer be a COC for these pathways. cPAHs would remain a COC for seafood consumption RME scenarios and for the beach play direct contact scenario.

^a EPCs for cPAHs were those presented in the HHRA for netfishing and clamming (Table B.3-43) or the FS for beach play areas (Table B-1 of Appendix B). EPCs for BaP in this table were calculated for this memorandum to estimate non-cancer HQs for BaP using the new reference dose.

b No reference dose was available for cPAHs or BaP at the time the HHRA and FS were conducted.

Table 2-3. Identification of cPAHs as a human health COC by scenario

	ВаР	Identified as a COC?	cPAHs Identified as a COC?					
Scenario	FS	Updated BaP Toxicity Values	Updated BaP FS Toxicity Values		Change in Status of cPAHs as a COC			
Seafood consumption RME scenarios								
Adult tribal RME – Tulalip data	na	no	yes	yes				
Child tribal RME – Tulalip data	na	no	yes	yes	No: cPAHs are still a COC (no change from ROD).			
Adult API RME	na	no	yes	yes				
Direct contact RME	scenario	S						
Netfishing RME	na	no	yes	no	Yes: cPAHs are no longer a COC			
Tribal clamming RME	na	no	yes	no	(cancer risks below 1 x 10 ⁻⁶ threshold) using updated BaP toxicity values.			
Beach play RME	na	no	yes	yes	No: cPAHs are still a COC (no change from ROD).			

Note: Shading indicates identification as a COC.

API – Asian Pacific Islander FS – feasibility study
BaP – benzo(a)pyrene na – not applicable
COC – chemical of concern ROD – Record of Decision

cPAH – carcinogenic polycyclic aromatic hydrocarbon RME – reasonable maximum exposure



3 Updated RBTCs for cPAH TEQ

RBTCs, which represent the concentrations in sediment and seafood tissue that equate to specific risk thresholds, were developed in Section 8 of the RI (Windward 2010) for human health COCs. RBTCs were calculated for excess cancer risk thresholds of 1×10^{-4} , 1×10^{-5} , and 1×10^{-6} . In the ROD, RBTCs were an important component in the derivation of sediment cleanup levels (EPA 2014), and tissue RBTCs were used to derive TTLs for seafood consumption COCs. This section provides updated RBTCs for the seafood consumption and beach play scenarios for which cPAHs are a COC. Consistent with the RI, FS, and ROD, RBTCs are not calculated for chemicals that are not COCs for a given scenario. The changes discussed in this section apply to cPAHs only and do not impact RBTCs for other COCs.

3.1 UPDATED BEACH PLAY SCENARIO RBTCs

RBTCs for human health COCs were presented for all three direct contact exposure scenarios in the RI (Windward 2010) for various risk thresholds. Updated RBTCs are presented here for only the beach play scenario, because cPAH TEQ is not a COC for the other two direct contact scenarios based on the updated toxicity values for BaP (i.e., risks for the netfishing and tribal clamming scenarios are already below the target risk thresholds). Using the updated BaP slope factor, the beach play sediment RBTCs would increase by approximately a factor of 7.3 (e.g., from 90 to 650 μ g/kg dry weight [dw] at the 1 × 10-6 risk level) (Table 3-1).

Table 3-1. Sediment RBTCs for human health direct contact RME scenarios

	cPAH TEQ Sediment RBTC (μg/kg dw)							
		RI		Updated with New BaP Slope Factor				
Scenario	Target Risk of 1 × 10 ⁻⁶	Target Risk of 1 × 10 ⁻⁵	Target Risk of 1 × 10 ⁻⁴	Target Risk of 1 × 10 ⁻⁶	Target Risk of 1 × 10 ⁻⁵	Target Risk of 1 × 10 ⁻⁴		
Netfishing RME	380	3,800	38,000	not calculated (not a COC based on updated toxicity values)				
Beach play RME	90	900	9,000	650	6,500	65,000		
Tribal clamming RME	150	1,500	15,000	not calculated (not a COC based on updated toxicity values)				

Note: Information regarding the development of RBTCs can be found in Section 8.1 of the RI (Windward 2010).

BaP – benzo(a)pyrene

COC - chemical of concern

cPAH – carcinogenic polycyclic aromatic hydrocarbon

dw - dry weight

RBTC - risk-based threshold concentration

RI - remedial investigation

RME - reasonable maximum exposure

TEQ - toxic equivalent

3.2 SEAFOOD CONSUMPTION SCENARIOS

For the seafood consumption scenarios, the RI/FS developed RBTCs for cPAHs in seafood tissue (Windward 2010; AECOM 2012). Corresponding sediment RBTCs for cPAHs could not be developed because clam tissue-to-sediment relationships were too uncertain (Windward 2010; EPA 2014).

Because clam consumption accounts for more than 90% of the total seafood ingestion risk for cPAHs, the ROD cPAH TTL is only for clam tissues. As in the FS (AECOM 2012), species-specific RBTCs for eastern softshell clams were calculated for three target risk levels. The 1 \times 10-6 clam-specific RBTC was used as the basis for the TTL presented in the ROD for cPAHs (ROD Table 21) (EPA 2014). The cPAH RBTC (and thus TTL) for the 1 \times 10-6 target risk level for the consumption of clams (based on the adult tribal RME) would increase from 0.24 to 1.8 µg/kg wet weight (ww) based on the updated toxicity values (Table 3-2).

Table 3-2. Tissue RBTCs for cPAHs and the human health RME seafood consumption scenarios

		сРА		rn Softshell C s (µg/kg ww)	Softshell Clam (μg/kg ww)			
	RI/FS				ited with New Slope Factor			
Scenario	Target Risk of 1 × 10 ⁻⁶	Target Risk of 1 x 10 ⁻⁵	Target Risk of 1 x 10 ⁻⁴	Target Risk of 1 x 10 ⁻⁶	Target Risk of 1 x 10 ⁻⁵	Target Risk of 1 × 10 ⁻⁴		
Adult tribal RME (Tulalip data)	0.24	2.4	24	1.8	18	180		
Child tribal RME (Tulalip data)	0.26	2.6	26	1.8	18	180		
Adult API RME	0.61	6.1	61	4.6	46	460		

Note: Information regarding the development of RBTCs can be found in Section 8.2 of the RI (Windward 2010).

API - Asian Pacific Islander

BaP - benzo(a)pyrene

cPAH - carcinogenic polycyclic aromatic hydrocarbon

FS - feasibility study

RBTC - risk-based threshold concentration

RI - remedial investigation

RME - reasonable maximum exposure

TEQ - toxic equivalent

ww - wet weight

4 Implications for the Sediment Remedy

This section presents a discussion of potential implications for the LDW sediment remedy based on the updated BaP toxicity values for each of the human health risk scenarios. The changes discussed in this section reflect only the changes in the BaP toxicity values: The scenarios and target risk levels have not changed. The higher RBTCs, sediment cleanup levels, RALs, and TTLs reflect the finding that BaP is less toxic than originally thought, and therefore target risk levels can be achieved with cleanup levels established at higher cPAH TEQs.

4.1 DIRECT CONTACT - NETFISHING

Question 1 – How might the change in the BaP slope factor affect the risk conclusions and RBTCs/cleanup levels for the netfishing scenario?

The ROD lists cPAHs as a COC for the netfishing scenario and presents a site-wide cleanup level for cPAH TEQ of 380 μ g/kg dw for the 0–10-cm sediment interval (ROD Table 19). Using the updated BaP slope factor, cPAHs would no longer be a COC for the netfishing scenario, so no cleanup level would be applicable. In other words, as shown in Section 2, risks for this scenario are already below the acceptable risk thresholds using the updated toxicity values, so a cleanup level would not be needed.

Question 2 - How might this change affect the cPAH RALs?

The ROD presents a cPAH human health RAL of 1,000 μ g/kg dw for the 0–10-cm sediment interval (ROD Table 28). This RAL, which was based on the netfishing scenario, would no longer be applicable based on the updated risk conclusions. Thus, there would no longer be a cPAH TEQ RAL for the LDW for the 0–10-cm sediment interval (Figure 2).

Question 3 – How might the remedial footprint change without a cPAH RAL for the 0–10-cm sediment interval?

The areas where sediments require active cleanup (e.g., dredging, capping, or enhanced natural recovery) may change without the 0–10-cm cPAH human health RAL of 1,000 μ g/kg dw. To assess the potential impact of removing the cPAH RAL, the RI/FS data were reviewed to identify locations with cPAH TEQs greater than 1,000 μ g/kg dw (i.e., the current RAL). This dataset was used because it is the basis for the remedial footprint presented in the ROD (Figure 18) (EPA 2014).⁶ Based on the RI/FS data, elimination of the 1,000- μ g/kg dw cPAH RAL would impact less than 1% of the sampling locations in the LDW, as described below:



- Approximately 7% of the RI/FS surface sediment locations (49 out of 711 locations) had cPAH TEQs that were greater than the 0–10-cm cPAH human health RAL of 1,000 μg/kg dw (Table 4-1).
- ◆ Of those 49 locations with a cPAH human health RAL exceedance, 35 were co-located with locations of RAL exceedances for other chemicals. None of the other RALs are changing. Thus, approximately 2% of the surface sediment locations (14 of 711 locations) had only a cPAH human health RAL exceedance (purple-circled locations on Map 1). In subsurface sediment, less than 1% of the locations (1 of 151 locations) had only a cPAH human health RAL exceedance (purple-circled locations on Map 2).
- ◆ Of the 14 locations with a cPAH-only human health RAL exceedance, only 4 (i.e., 0.6% of the 711 locations) were in areas without nearby exceedances of other RALs (Map 1).

Ultimately, based on this evaluation, more than 90% of the locations (45 of 49 locations) with current RAL exceedances would likely be remediated. For the four locations without a nearby exceedance, the shapes and sizes of these remediation polygons (i.e., those shown on ROD Figure 18) could be affected by the absence of the cPAH RAL. The sizes and shapes of the remedial polygons on ROD Figure 18, which were determined based on rules defined in ROD Figures 19, 20, and 21,7 are likely to change also as a result of the sampling for the remedial design.

Table 4-1. Summary of RAL exceedances in sediment based on the RI/FS dataset

	Count of Sediment Locations		
Statistic	Surface Sediment	Subsurface Sediment	
Locations analyzed for PAHs ^a	711	151	
Locations with cPAH TEQ > 1,000 µg/kg dw (may also have RAL exceedances for other contaminants)	49	11	
Locations with cPAH TEQ > 1,000 μg/kg dw and benthic PAH RAL exceedance	23	6	
Locations with <u>ONLY</u> cPAH TEQ > 1,000 µg/kg dw (range of cPAH TEQs for these locations shown in parentheticals)	14 (cPAH TEQs for these 14 locations ranged from 1,060–2,400 µg/kg dw)	1 (cPAH TEQ for this location was equal to 1,200 μg/kg dw)	
Locations with <u>ONLY</u> cPAH TEQ > 1,000 µg/kg dw and without nearby RAL exceedances for other chemicals	4	0	

Only locations within areas with recovery categories were considered for RAL compliance (i.e., early action areas were excluded). RM 4.75 to RM 5 was assumed to be Recovery Category 2 for this exercise (no recovery category was designated for this area in the FS).

⁷ ROD Figure 19 is titled *Intertidal Areas – Remedial Technology Applications*. ROD Figure 20 is titled *Subtidal Areas – Remedial Technology Application*. ROD Figure 21 is titled *Intertidal and Subtidal Areas – Natural Recovery Application*.



cPAH – carcinogenic polycyclic aromatic hydrocarbon dw - dry weight FS - feasibility study PAH – polycyclic aromatic hydrocarbon

RAL – remedial action level RI - remedial investigation RM - river mile TEQ - toxic equivalent

In understanding the potential impact of the elimination of the 0-10-cm cPAH RAL, it is important to recognize that polycyclic aromatic hydrocarbons (PAHs) would still be considered in the development of the remedy. Exceedances of PAH benthic RALs are based on Washington State Sediment Management Standards for the protection of the benthic community and are shown as yellow-circled locations on Maps 1 and 2. These RALs are not changing and will still be used in determining the eventual site-wide remedial footprint. Of the 49 locations with cPAH human health RAL exceedances using the RI/FS dataset, 23 locations also had a PAH benthic RAL exceedance (Table 4-1).8 Furthermore, all locations with cPAH TEQs greater than 2,500 µg/kg dw were co-located with locations of benthic PAH RAL exceedances, meaning that the highest cPAH TEQs would still be addressed during remediation.

4.2 **DIRECT CONTACT - TRIBAL CLAMMING**

Question 1 – How might the change in the BaP slope factor affect the risk conclusions and RBTCs/cleanup levels for the tribal clamming scenario?

The ROD lists cPAHs as a COC for the tribal clamming scenario and presents a site-wide cPAH TEQ cleanup level of 150 μg/kg dw in intertidal (0-45-cm) sediment (ROD Table 19) (EPA 2014). Using the updated BaP slope factor, cPAHs would no longer be a COC for the tribal clamming scenario, so a cleanup level would not be applicable. In other words, as discussed in Section 2, risks for this scenario are already below the acceptable risk thresholds using the updated toxicity values.

Question 2 – How might this change affect the cPAH RALs?

The intertidal RAL for cPAHs presented in the ROD for 0-45-cm sediment was based on the beach play scenario, so the elimination of cPAHs as a COC for tribal clamming would not affect this RAL. However, the intertidal cPAH RAL would no longer be applicable to the clam collection areas, because cPAH risks associated with clamming in these areas are already below acceptable risk levels. Note that there would still be a cPAH RAL in beach play area (see Section 4.3).

Question 3 – How might the remedial footprint change without a cPAH RAL for clam tissue collection areas?

cPAH TEQ would no longer be a COC for tribal clamming, meaning that the 0-45-cm RAL for cPAHs would not apply in intertidal areas outside of beach play areas. cPAHs would still be addressed in the beach play portions of the intertidal area (Section 4.3) and, as discussed for the netfishing scenario, PAH benthic RALs would still apply in



the 0–10-cm sediment interval throughout the entire LDW. Furthermore, as shown on Map 3, which presents the cPAH RAL exceedances in the intertidal areas, nearly all of the cPAH exceedances are in a beach play area (Section 4.3) or located near other RAL exceedances, indicating minimal change to the overall remedial footprint. Of the 22 intertidal locations in clam collection areas analyzed for PAHs with concentrations that exceed the current RAL of 900 μ g/kg dw (including both 0–10- and 0–45-cm samples), all would be expected to be remediated because of either other RAL exceedances for that location or other nearby RAL exceedances.

4.3 DIRECT CONTACT - BEACH PLAY SCENARIO

Question 1 – How might the change in the BaP toxicity value affect the risk conclusions and RBTCs/cleanup levels for the beach play scenario?

The ROD lists cPAHs as a COC for the beach play scenario and presents a cleanup level of 90 μ g/kg dw (as TEQ) in intertidal (0–45-cm) sediment applied to each beach play area (ROD Table 19). Using the updated BaP slope factor, cPAHs would still be a COC for the beach play scenario (Table 2-4), and the 1 × 10-6 sediment RBTC would increase from 90 to 650 μ g/kg dw (Table 3-1). As a result of this change, if the updated slope factor were applied, the 1 × 10-6 RBTC would increase from 90 to 650 μ g/kg dw, and the cleanup level would change accordingly to match the updated RBTC.

Question 2 – How might this change affect the cPAH RALs?

The cPAH RAL for 0–45-cm intertidal sediment in the ROD is equal to 900 μ g/kg dw, which is based on the 1 × 10⁻⁵ RBTC for beach play (AECOM 2012; EPA 2014). Based on the updated BaP slope factor, EPA could update this RAL to be based on the new 1 × 10⁻⁵ RBTC for beach play of 6,500 μ g/kg dw (Table 3-1) and make the RAL specific to beach play areas.

Question 3 – How might the remedial footprint change as a result of the change in beach play cleanup level and intertidal RAL?

The areas included in the remedial footprint may change with an increase of the 0–45-cm RAL from 900 to 6,500 μ g/kg dw in the beach play areas. To assess the potential impact of the change, the FS sediment data for beach play areas were compared with the cPAH TEQ RAL of 900 μ g/kg dw. A total of eight intertidal locations (including both 0–10- and 0–45-cm samples) in beach play areas exceeded this cPAH RAL. cPAH TEQs for these locations ranged from 900 to 4,800 μ g/kg dw, meaning that all were below the potential new RAL of 6,500 μ g/kg dw. To evaluate the impact of this change on the remedial footprint, the eight intertidal locations that exceeded the current RAL were further assessed. All would be expected to be remediated because of other RAL exceedances at that location or because of the presence of other nearby RAL exceedances.



Design sampling within intertidal areas will identify locations with exceedances of applicable RALs (both human health and benthic protection) in accordance with the RAL application flow charts in the ROD (EPA 2014).

4.4 SEAFOOD CONSUMPTION SCENARIOS

No cPAH sediment cleanup level was set for seafood ingestion in the ROD because, as described in the ROD (EPA 2014):

Seafood consumption excess cancer risks for ...(cPAHs)... were largely attributable to eating clams. However, data collected during the RI/FS showed little relationship between concentrations of ... cPAH in sediment and their concentrations in clam tissue. EPA will define the sediment cleanup footprint based on other cleanup levels, then use the clam target tissue levels (Section 8.2.3) to measure reduction in ... cPAH concentrations in clams. Research will be conducted during the remedial design phase to study the relationships between sediment concentrations (of) ... cPAHs and concentrations in clam tissue and methods to reduce concentrations of these contaminants in clams. If EPA determines, based on these studies, that additional remedial action is needed to reduce clam tissue ... cPAH concentrations for the purpose of achieving RAO 1, EPA will document and select those actions in a future decision document.

The Pre-Design Studies collected additional cPAH data to further assess the relationships among sediment, clam tissue, and porewater. This section presents the results of this investigation with respect to the seafood consumption pathway (Remedial Action Objective 1), the details of which are presented in Attachment 1.

Question 1 – How might the change in the BaP slope factor affect the risk conclusions and RBTCs for the seafood consumption scenarios?

The ROD lists cPAHs as a COC for the seafood consumption scenarios and presents a risk-based TTL of 0.24 μ g/kg ww in eastern softshell clams (ROD Table 21). Using the updated BaP slope factor, cPAHs would still be a COC for the seafood consumption scenarios (Table 2-3), and the 1 × 10⁻⁶ tissue RBTC (i.e., the basis for the TTL in the ROD) would increase from 0.24 to 1.8 μ g/kg dw (Table 3-2).

Question 2 – How might the change in the BaP slope factor affect the comparison of the 95 upper confidence limit on the mean (95UCL) for cPAH TEQ in clam tissue with the TTL?

To evaluate progress toward the TTL for cPAHs, the 95UCL for clam tissue can be compared with the TTL using both the RI/FS dataset and the baseline dataset:

• RI/FS dataset – The site-wide 95UCL used in the HHRA was 20 μg/kg ww based on clams collected from the LDW in 2004. This 95UCL was approximately two orders of magnitude higher than the risk-based TTL in the ROD and about one order of magnitude higher than the TTL calculated using the updated BaP slope factor.



• Baseline dataset – The site-wide 95UCL calculated using clams collected from the LDW in 2018 is 5.94 μg/kg ww,⁹ which is about 70% lower than the 2004 95UCL. The 2018 95UCL is above the TTL in the ROD by a factor of 25 and above the TTL calculated using the updated BaP slope factor by a factor of 3.3.

Thus, while the site-wide 95UCL has not achieved the TTL, cPAH TEQs in LDW clam tissue were about 70% lower in 2018 than in 2004.

Clam tissue monitoring will be continued as part of long-term monitoring of the site. It is expected that the more-sensitive ultra-trace analytical method for cPAHs will be used to detect cPAH compounds at lower concentrations in clam tissues, thus avoiding the low detection frequencies encountered during the initial analysis of the baseline samples.

Question 3 – Do the post RI/FS data sufficiently improve our understanding of the relationship between cPAHs in clams and sediment to support the development of a cleanup level?

Although the relationships among surface sediment (0–10 cm), clam tissue, and porewater for cPAHs were further investigated as part of the Pre-Design Studies, these data did not support the development of a clam tissue and sediment relationship. Thus, it is still not possible to develop a sediment cleanup level for this exposure pathway. The clam tissue-sediment-porewater relationship analysis is summarized in Questions 3a and 3b, with additional details provided in Attachment 1.

Question 3a - Did the 2018 cPAH investigation data demonstrate a reasonable correlative relationship between cPAHs in clam tissue and sediment?

Sixteen cPAH investigation areas were established where co-located clam tissue and surface sediment samples were collected across a range of cPAH TEQs (Windward 2018a, 2019). The sediment cPAH TEQ values ranged from 9.55 to 11,600 μ g/kg dw, with values of 208 μ g/kg dw or less reported for 13 of the samples. The tissue cPAH TEQ values were similarly skewed, with a range of 1.8 to 52.8 μ g/kg ww and 13 samples with values of 2.6 μ g/kg ww or less. The relationship between cPAH TEQ values in sediments and those in tissues is heavily influenced by the three samples with the highest values (Figure 3). Any reasonable model that could describe this relationship will need to be based on transformed data and/or a culled dataset, to reduce or remove the influence of these three data points.

⁹ The 95UCL for the baseline dataset was calculated using the validated analytical results from the ultra-trace method analysis of cPAHs, which was able to detect cPAH compounds at lower concentrations.



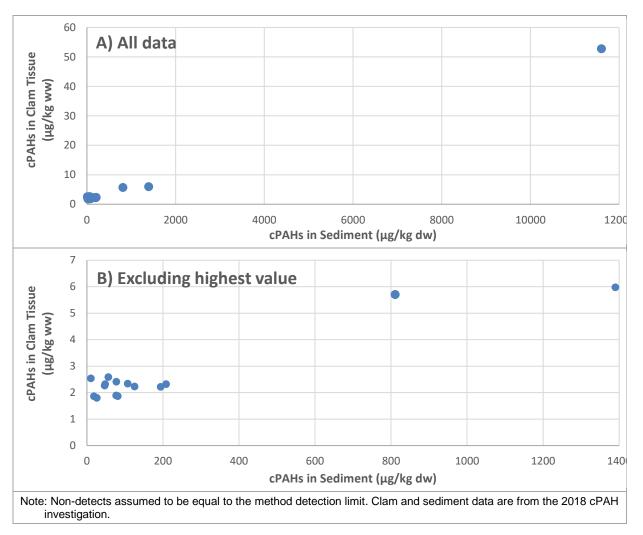


Figure 3. cPAH TEQs in sediment and clam tissue

The clam tissue-sediment relationship was evaluated using both the cPAH TEQs and several of the individual cPAHs that were more frequently detected (i.e., benzo(b) fluoranthene, chrysene, and benzo(a)anthracene). Several forms of regression models were considered in evaluating these data, including linear regression on untransformed scales, linear regression on log-log scales, and segmented (hockey stick) regression on log-log scales. Details of this evaluation are presented in Attachment 1.

Based on the results of this evaluation, tissue concentrations did not co-vary with sediments when sediment cPAH TEQs were less than approximately 200 μ g/kg dw. For the three samples with sediment cPAH TEQs greater than 800 μ g/kg dw, the cPAH concentrations were greater in clam tissue, but insufficient data were available in this range to estimate this relationship with confidence (Figure 3).

Question 3b - Did porewater help to improve our understanding of the relationship between cPAHs in clams and sediment?

At 10 of the clamming locations, the co-located sediment collected with the clams was equilibrated $ex\ situ$ with passive samplers for 28 days in order to estimate C_{free} cPAH porewater concentrations (Windward 2018a). These data, which were presented in the final clam data report (Windward 2019), were evaluated relative to cPAH concentrations in clam tissue and sediment to determine if porewater improved the ability to understand clam tissue-sediment relationships. Ultimately, as described in Attachment 1, the results of this evaluation indicated that porewater was not useful in understanding the clam tissue-sediment relationship for cPAHs.

Question 4 – Based on this new information, is the relationship between cPAHs in clams and sediment sufficiently certain to develop a sediment RAL for seafood consumption?

As described in response to Question 2, the 2018 cPAH investigation indicates that cPAH concentrations in clam tissue show no clear response to increasing concentrations in sediment up to sediment cPAH TEQs of 200 μ g/kg dw. Above this sediment concentration, clam tissue concentrations are higher, but there are too few data to adequately define any relationship. Although efforts were made in the planning of this investigation to target clams (and co-located sediment collection) from areas with the desired range of cPAH TEQs based on the RI/FS dataset, concentrations in the target areas were approximately 50% lower (on average) than anticipated. Based on the RI/FS dataset, target areas had concentrations ranging from 13 to 3,000 μ g/kg dw, with estimated TEQs greater than 200 μ g/kg dw in nine samples. In actuality, sediment concentrations ranged from 10.2 to 11,600 μ g/kg dw, and only three locations sampled had sediment cPAH TEQs above 200 μ g/kg dw (Windward 2019).

While additional data in the sediment cPAH TEQ range of 200 to 2,000 μ g/kg dw may further elucidate relationships between cPAHs in sediment and clam tissue, uncertainty in the clam exposure pathways precludes the ability to define a sediment cleanup value. In other words, even if additional co-located clam tissue and sediment data could be collected in the desired cPAH TEQ range, other uncertainties in understanding clam exposure may continue to prevent the development of a clam tissue-sediment relationship. These uncertainties include the following:

◆ Appropriate depth of sediment exposure – The sediment depth to which Eastern softshell clams are primarily exposed is likely to be shallow. The 0–10-cm surface sediment samples collected as part of the cPAH investigation might not accurately represent the depth of the sediment to which the clams are primarily exposed. In other words, rather than being exposed to sediment from the 0–10-cm-depth, as was collected during the RI/FS and baseline sampling, the majority of sediment exposure for clams could occur within a thinner layer of surface sediment where the siphon of the clam is located, a detail that could be particularly important given the observed



- variability of cPAH concentrations in sediment (Windward 2018b, Sections 2.3.1.1 and 2.3.2.1).
- ◆ Exposure to suspended particulates or colloids in the water column or to fine materials on the sediment surface The feeding habits of clams may suggest that fine particulates or colloids could be important for understanding the uptake of cPAHs in clam tissue. Eastern softshell clams are considered filter-feeding clams, which feed by extending their siphon into the water (e.g., Aller and Yingst 1985; Lin and Hines 1994; Wade 1967, 1969). Similarly, deposit-feeding clams feed by extending their siphon across the sediment surface and thus are exposed to a thin layer of surface sediment or materials on the sediment surface.

Based on these uncertainties and declining cPAH TEQs in clam tissue, development of a sediment cleanup level for cPAHs based on seafood consumption is not recommended at this time. Because concentrations of cPAHs in clam tissue and sediment (both site wide and in intertidal areas) are decreasing and seafood consumption risks for cPAHs are nearing the 1×10^{-6} TTL (based on the updated BaP slope factor), continued monitoring of clam tissue is recommended to evaluate progress toward meeting the TTL for cPAHs in clams.

Question 5 – If a sediment RAL aimed at reducing cPAHs in clam tissue cannot be developed at this time, how will risks to human health from the consumption of clam tissue be addressed?

As EPA stated in the ROD, if a cPAH TEQ cleanup level and sediment RAL for seafood consumption are too uncertain, "EPA will define the sediment cleanup footprint based on other cleanup levels, then use the clam target tissue levels to measure reduction in ... cPAH concentrations in clams." Therefore, current trends in clam and sediment cPAH concentrations, as well as the anticipated influence of other COC RALs on further reducing cPAH concentrations in sediment, were evaluated.

As described in response to Question 1, concentrations of cPAHs in clam tissue in the LDW have decreased by more than 60% between 2004 and 2018. This decrease is consistent with observed decreases in the cPAH TEQ in surface sediment of the LDW over time. Evidence of decreases in surface sediment concentrations of cPAH is based on the RI/FS and 2018 baseline surface sediment data and source-related data summarized in the draft data evaluation report (Windward 2018b). Preliminary analyses suggest that surface sediment spatially weighted average concentrations based on 2018 data are 62% lower than those based on RI/FS data, and the cPAH concentrations in suspended solids entering the LDW have decreased by about 40%. ¹⁰

¹⁰ This estimate is based on the base-case lateral bed composition model input value based on source solids data presented in the draft data evaluation report (Windward 2018b).



Based on the evaluation described in Section 4.1 (Table 4-1 and Map 1), it is expected that even with the potential changes to the cPAH RALs discussed herein, approximately 90% of the RI/FS sample locations with cPAH TEQs greater than 1,000 μ g/kg dw in surface sediment samples would be addressed as part of the sediment remediation. Thus, cPAH TEQs in sediment are expected to continue to decrease. Clam tissue monitoring will continue to assess changes in cPAH concentrations in clams as remediation progresses.

5 Summary and Conclusions

EPA will make the decisions regarding any changes to cPAH cleanup levels and RALs in the ROD based on updated BaP toxicity information. Changes may be presented in an explanation of significant differences (ESD) document. Key conclusions from this technical memorandum are as follows.

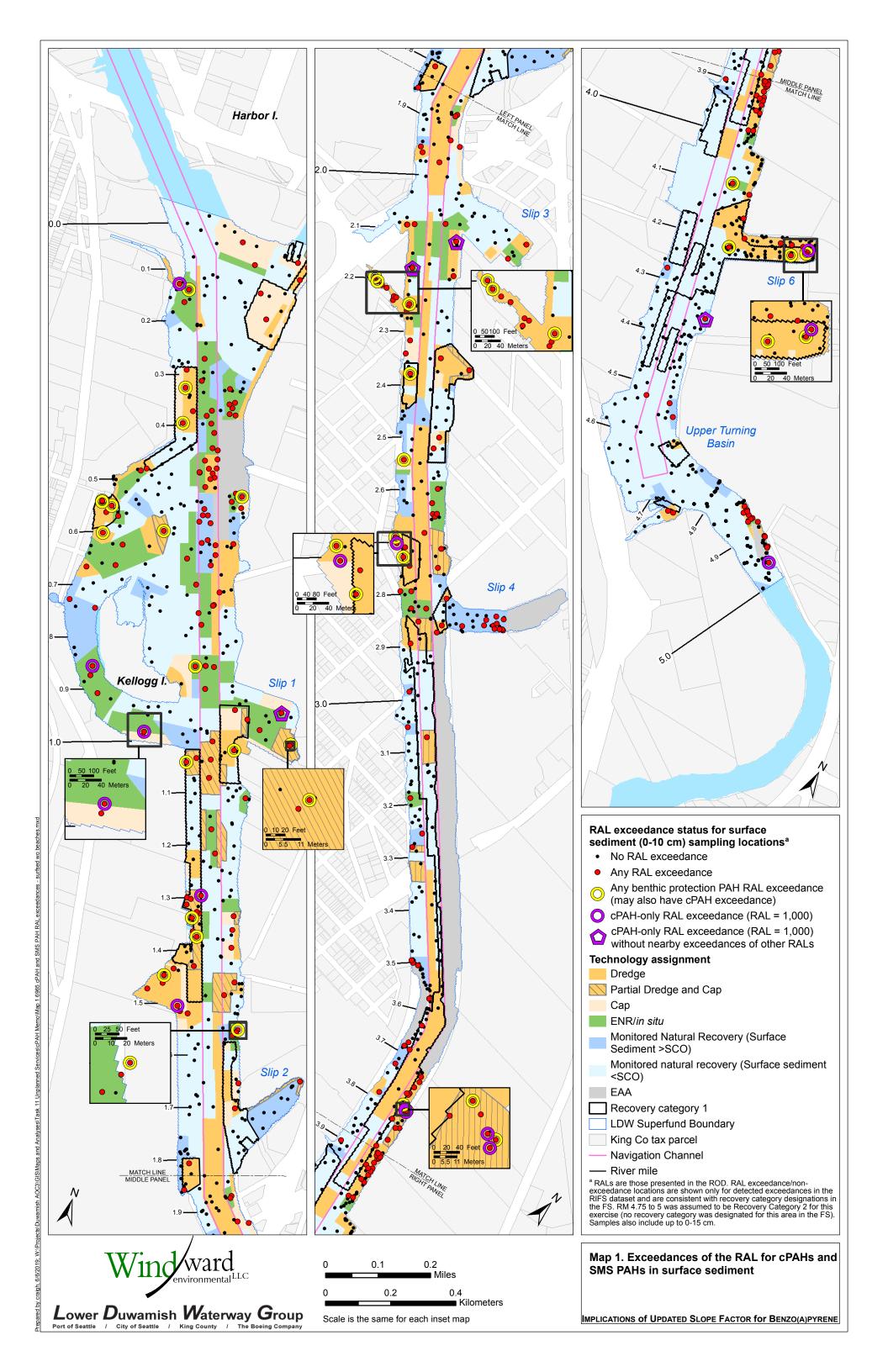
- Changes to the sediment remedy are expected to be minimal as a result of the change in the BaP slope factor. cPAHs in LDW sediments generally will be addressed by the presence of co-located or nearby RAL exceedances for other chemicals as well as benthic RALs for PAHs.
- cPAHs would no longer be a COC for direct contact exposure from netfishing and tribal clamming. An ESD would likely remove cPAH cleanup levels for the netfishing and clamming exposure scenarios and associated RALs from the ROD.
- cPAHs would remain a COC for the beach play scenario. An ESD would likely revise the cPAH RAL for the 0-45-cm intertidal sediment from 900 to 6,500 µg/kg dw and apply the cPAH RAL to only the beach play areas.
- cPAHs would remain a COC for the seafood consumption scenarios, and an ESD would likely revise the cPAH TTL for clams from 0.24 to 1.8 μg/kg ww.
- cPAH TEQs in both clam tissue and sediment appear to be decreasing. This
 trend is likely to continue as remediation progresses. Clam tissue
 monitoring will continue as part of long-term monitoring for the site.

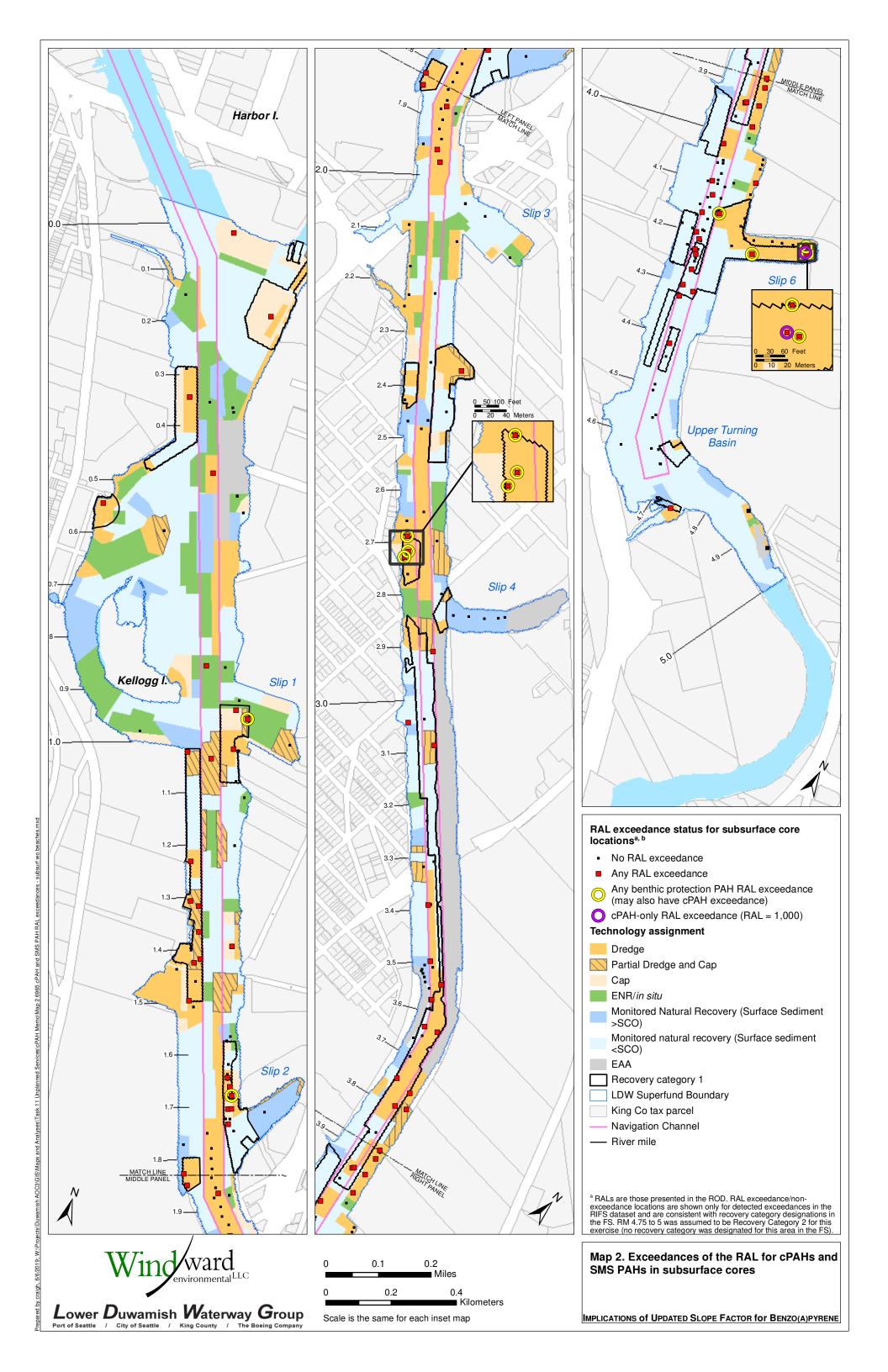
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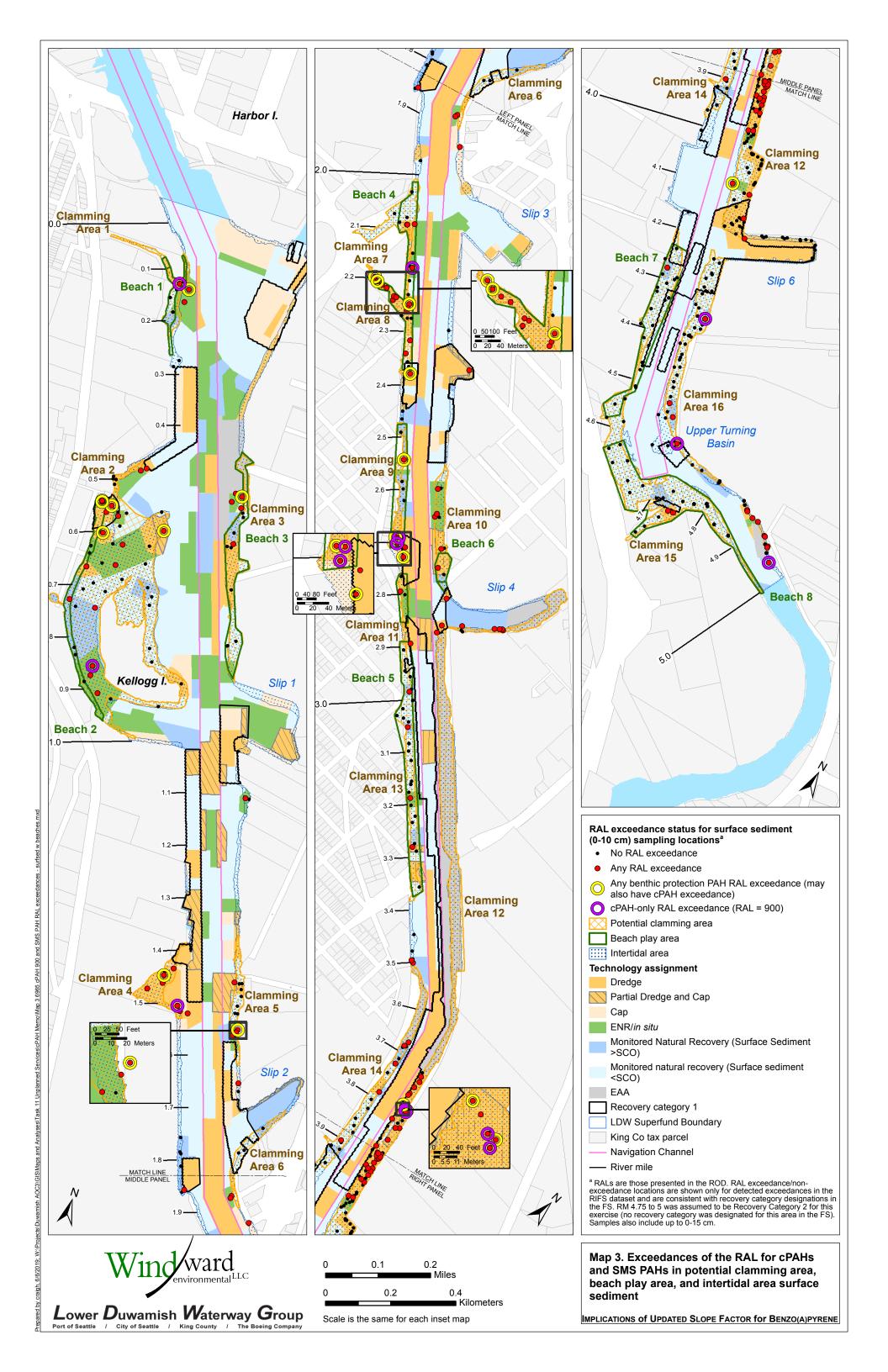
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ATTACHMENT 1. SUPPLEMENTAL INFORMATION

1 Overview

This attachment provides supplemental information regarding:

- Carcinogenic polycyclic aromatic hydrocarbon (cPAH) risks calculated using the 2017/2018 Pre-Design Studies baseline dataset
- Details of the evaluation of the cPAH clam tissue, sediment, and porewater relationships
- A summary of remedial investigation/feasibility study (RI/FS) data with cPAH toxic equivalent (TEQ) remedial action level (RAL) exceedances

2 Risk Calculations Using Baseline Dataset

For informational purposes, this section presents a summary of the cPAH excess cancer risk estimates for the seafood consumption and direct contact scenarios; these estimates were calculated using the baseline dataset and the updated benzo(a)pyrene (BaP) slope factor (Table 1). All of the exposure assumptions are the same as those used in the Lower Duwamish Waterway (LDW) human health risk assessment (HHRA). The risks calculated using this dataset are lower than those calculated using the FS dataset and updated BaP slope factor. No changes to the identification of chemicals of concern (COCs) would result from using the baseline data rather than the FS data.

Table 1. cPAH TEQ risk estimates for the human health seafood consumption RME scenarios using the baseline dataset compared to FS dataset

	cPAH EPCs (μg/kg)		Cance Calculated	Q Excess er Risk d using the lope Factor	Change to COC Designation Relative to the FS Dataset (Both			
Scenario	FS Dataset			Baseline Dataset ^a	Using Updated BaP Slope Factor)?			
Seafood consumption RME	Seafood consumption RME scenarios							
Adult tribal RME – Tulalip data	varies ^b	varies ^b	1 × 10 ⁻⁵	4 × 10 ⁻⁶	No: Risks are lower, but cPAH TEQ would still be a COC.			
Child tribal RME – Tulalip data	varies ^b	varies ^b	1 × 10 ⁻⁵	4 × 10 ⁻⁶				
Adult API RME	varies ^b	varies ^b	4 × 10 ⁻⁶	1 × 10 ⁻⁶				

	cPAH EPCs (μg/kg)		cPAH TEQ Excess Cancer Risk Calculated using the New BaP Slope Factor		Change to COC Designation Relative to the FS Dataset (Both	
Scenario	FS Dataset	Baseline Dataset ^a	FS Dataset	Baseline Dataset ^a	Using Updated BaP Slope Factor)?	
Direct contact RME scenario	os					
Netfishing RME	570	226	2 × 10 ⁻⁷	8 × 10 ⁻⁸	No: Risks are still below the 1 x 10 ⁶ threshold.	
Tribal clamming RME	770	913	7 × 10 ⁻⁷	9 × 10 ⁻⁷	No: Risks are still below the 1 x 10 ⁶ threshold.	
Beach play RME – Area 1	380	600	6 × 10 ⁻⁷	9 × 10 ⁻⁷	No: cPAH TEQ would still be a	
Beach play RME – Area 2	7,000	696	1 × 10 ⁻⁵	1 × 10 ⁻⁶	COC (risks are above the 1×10^{-6} threshold for at least 1 beach).	
Beach play RME – Area 3	1,500	325	2 × 10 ⁻⁶	5 × 10 ⁻⁷	However, the baseline data	
Beach play RME – Area 4	1,400	93.4	2 × 10 ⁻⁶	1 × 10 ⁻⁷	suggest that risks may be above	
Beach play RME – Area 5	2,200	5,310	3 × 10 ⁻⁶	8 × 10 ⁻⁶	this threshold for fewer beaches (i.e., only beaches 5 and 6) than	
Beach play RME – Area 6	7,100	1,650	1 × 10 ⁻⁵	3 × 10 ⁻⁶	was thought based on the FS	
Beach play RME – Area 7	98	63.4	1 × 10 ⁻⁷	1 × 10 ⁻⁷	dataset (for which beaches 2, 3, 4, 5, and 6 had risks above the	
Beach play RME – Area 8	270	232	4 × 10 ⁻⁷	4 × 10 ⁻⁷	threshold).	

Note: cPAH risk estimates shown in **bold and red** are above the excess cancer risk threshold of 1 x 10⁻⁶. No BaP non-cancer HQs were above the threshold of 1, and thus those values are not presented here.

- Baseline data were collected in 2017 and 2018 as part of the Pre-Design Studies. These data were collected to update the FS dataset after the early actions but before the full sediment remedy.
- ^b EPCs for the seafood consumption scenarios varied by consumption category. For these risk estimates, EPCs were equal to those used in the LDW HHRA (Windward 2007), with the exception of the EPC for clams, which account for over 90% of the risk associated with cPAHs. For clams, the EPC of 5.94 μg/kg ww calculated using the ultra-trace baseline dataset was used for the baseline dataset risks. For the FS dataset risks, the EPC used in the HHRA (20 μg/kg ww) was used.

BaP - benzo(a)pyrene

cPAH – carcinogenic polycyclic aromatic hydrocarbon

EPC – exposure point concentration

FS - feasibility study

HHRA - human health risk assessment

HQ - hazard quotient

LDW - Lower Duwamish Waterway

RME - reasonable maximum exposure

TEQ - toxic equivalent

ww - wet weight

3 Evaluation of cPAH Investigation Clam Tissue, Sediment, and Porewater Data

As described in Section 4.4 (Question 2) of the memorandum, the relationships among surface sediment (0–10 cm), clam tissue, and porewater for cPAHs were further investigated as part of the Pre-Design Studies. Co-located clam tissue and surface sediment samples were collected at 16 locations across a range of cPAH TEQs (Windward 2018, 2019). At 10 of the clamming locations, the co-located sediment collected with the clams was equilibrated ex situ with passive samplers for 28 days in order to estimate C_{free} cPAH porewater concentrations (Windward 2018). This section presents analyses of the data in the three media: tissue, porewater, and sediment.

3.1 RELATIONSHIP BETWEEN CLAM TISSUE AND SEDIMENT FOR CPAHS

This section presents details regarding the evaluation conducted to answer Question 2a of the memorandum: "Did the 2018 cPAH investigation data demonstrate a reasonable correlative relationship between cPAHs in clam tissue and sediment?" Evaluation of the relationship between cPAHs in clam tissue and sediment required consideration of two key factors: the influence of high values and the detection frequency of individual cPAHs.

First, the relationship between cPAH TEQ values in sediments and those in clam tissue is heavily influenced by the three samples with the highest values (Figure 1). Any reasonable model that could describe this relationship will need to be based on transformed data and/or a culled dataset, to reduce or remove the influence of these three data points. Second, the detection frequency of several individual cPAHs was less than 25% in the clam tissue samples. For example, 3 out of 16 tissue samples had detected values for benzo(a)pyrene (BaP), which is the reference compound (and most toxic contributor) for cPAH TEQ (i.e., has a potency equivalency factor [PEF] of 1.0).¹

To avoid the confounding influence of numerous non-detects on the tissue cPAH TEQ values, models were considered for individual cPAHs with high detection frequencies in tissue samples. Data were evaluated for three cPAHs: benzo(b)fluoranthene (detected in 15 of 16 tissue samples), chrysene (detected in 13 of 16 tissue samples), and benzo(a)anthracene (detected in 12 of 16 tissue samples).² All of these cPAH compounds were detected in all of the sediment samples. The same patterns observed in cPAH TEQ values were observed for these three compounds. All three compounds were detected in the same three samples in both sediment and tissue at much higher concentrations relative to the remainder of the dataset. Consequently, these three samples were highly influential in determining the model parameter estimates (e.g., slope or intercept) in any of the model forms.

² The PEFs for these cPAHs are 0.1 for benzo(b)fluoranthene, 0.01 for chrysene, and 0.1 for benzo(a)anthracene.



¹ PEFs for the other cPAHs included in the TEQ range from 0.4 to 0.01.

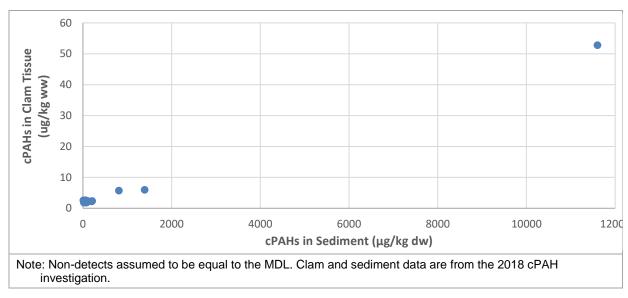


Figure 1. cPAH TEQs in sediment and clam tissue

Several forms of regression models were considered in evaluating these data, including linear regression on untransformed scales, linear regression on log-log scales, and segmented (hockey stick) regression on log-log scales. Among the models considered, two different response variables (tissue and log-tissue) and three different datasets (using all data or omitting one or more of the influential data points) were identified. The validity of each of the fitted models is based on the behavior of the residuals from diagnostic plots: If the patterns displayed by the residuals violate the underlying assumptions about a model, then that model is not considered to be valid. Results for co-located tissue and sediment data that had detected concentrations³ of benzo(b)fluoranthene in both media are summarized in Table 2. Benzo(b)fluoranthene is shown because it had the highest number of detected tissue concentrations, and these results are indicative of the patterns observed in both chrysene and benzo(a)anthracene.

³ Models that attempted to incorporate the tissue method detection limits (MDLs) sacrificed goodness-of-fit to the remainder of the data in order to accommodate these extreme (low) values.



Table 2. Comparison of model diagnostics for clam tissue and sediment regressions for benzo(b)fluoranthene

Model	Dataset	Residual Diagnostics	Does Model Approximate Trends?
	all detected data (n=15)	Pattern of residuals indicates model is inappropriate. ^b Line is entirely determined by 3 points with sediment > 143 µg/kg dw.	no
Linear (un- transformed) ^a	detected tissues and sediment ≤ 789 µg/kg (n=14)	Pattern of residuals indicates model is inappropriate. ^b Line is largely determined by 2 points with sediment > 143 µg/kg dw.	no
	detected tissues and sediment ≤ 143 µg/kg (n=12)	Residuals are adequate. An influential data noint (sediment	
Linear (log-log) ^c	all detected data (n=15)	Pattern of residuals indicates model is inappropriate. ^b Two highly influential points (sediment at 6.54 and 5,030 µg/kg dw) are present.	no
	detected tissues and sediment ≤ 789 µg/kg (n=14)	Pattern of residuals indicates model is inappropriate. ^b Three highly influential points (sediment at 6.54 and two samples with sediment > 143 µg/kg dw) are present.	no
	detected tissues and sediment ≤ 143 µg/kg (n=12)	Residuals are adequate. An influential data point (sediment at 6.54 $\mu g/kg$ dw) is present.	yes
	all detected data (n=15)	Residuals are adequate. Model is largely driven by 3 points with sediment > 143 μ g/kg dw.	yes
Segmented (log-log) ^d	detected tissues and sediment ≤ 789 µg/kg (n=14)	Model fails to converge.	no
	detected tissues and sediment ≤ 143 µg/kg (n=12)	A segmented model is not supported by the data in this range.	na

^a $Tissue = \beta_0 + \beta_1 \times Sediment$

dw - dry weight

na - not applicable

Three models appear to approximate the patterns in this dataset (indicated by a "yes" in the last column of Table 2). Two of these models include only sediment data with concentrations less than or equal to $143 \, \mu g/kg$ dry weight (dw). The one model that includes all data is the segmented (hockey stick) regression defined by:

Segment 1: $\ln(Tissue) = 0.69 + 0.035 \times \ln(Sediment)$ when $Sediment < 227 \,\mu\text{g/kg}$ Segment 2: $\ln(Tissue) = -4.33 + 0.88 \times \ln(Sediment)$ when $Sediment \ge 227 \,\mu\text{g/kg}$

If the form of the model is appropriate for the dataset (e.g., a linear model with normally distributed errors), then the pattern of residuals will be approximately normal: randomly distributed around zero (i.e., no particular pattern). If either of these conditions are seriously violated, it indicates that the model is inappropriate for these data.

 $[\]ln(Tissue) = \beta_0 + \beta_1 \times \ln(Sediment)$

 $[\]ln(Tissue) = \beta_0 + \beta_1 \times \ln(Sediment) \ when Sediment < Inflection Point$, and $\ln(Tissue) = \beta_2 + \beta_3 \times \ln(Sediment) \ when Sediment \ge Inflection Point$

This model is shown in the top panel of Figure 2. In this model formulation, the inflection point is the concentration at which the relationship between tissue and sediment changes. As a rule, statistical models are not reliable if they are being driven by just a few data points, as is the case with this dataset. Based on residual diagnostics for benzo(b)fluoranthene, the segmented regression model was the only model that appeared valid for all samples. However, preference for this model form was driven by the three samples with the highest sediment and tissue concentrations. If just the most extreme sample is omitted from this dataset, a log-linear model offers an alternative fit to the data (the orange regression line in Figure 2).⁴ Both model forms suffer from the presence of influential data points and insufficient data at higher sediment concentrations. Similar results were observed for the other individual cPAH compounds with the highest detection frequencies (13 detects for chrysene, and 12 detects for benzo(a)anthracene). These models are shown in the middle and bottom panels, respectively, of Figure 2.

 $^{^4}$ Omitting the sample with sediment concentrations > 5,000 µg/kg dw for each of these cPAHS, the log-linear models had residual sums of squares (RSS) that were similar to the full segmented regression models' RSS calculated without this extreme sample. However, the log-linear models had poor residual patterns, with influential data points at both the low and high ends of the sediment scale.

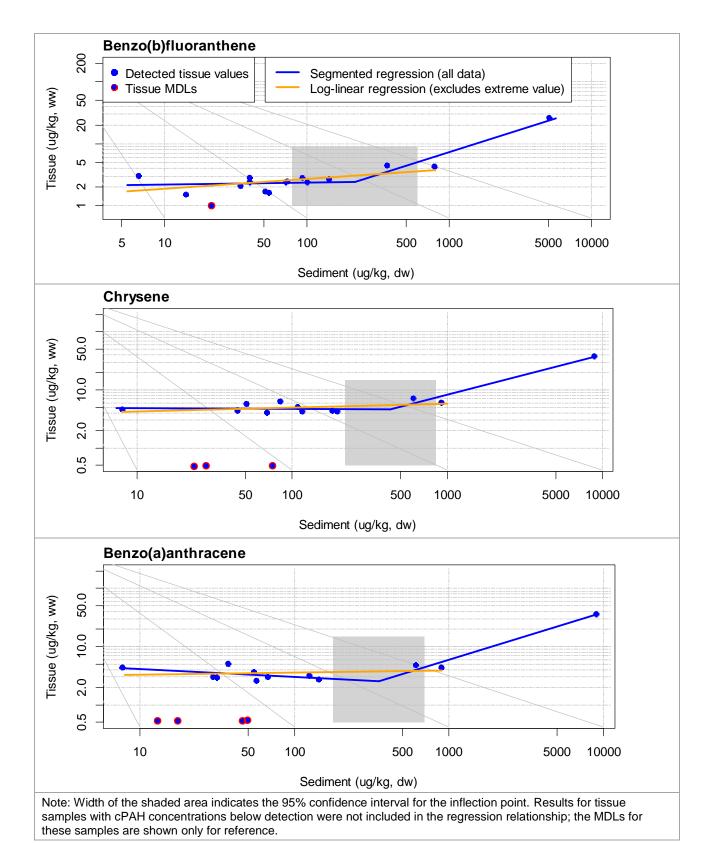


Figure 2. Relationship between individual cPAHs in sediment and clam tissue for benzo(b)fluoranthene, chrysene, and benzo(a)anthracene

Based on the results of this evaluation, tissue concentrations did not co-vary with sediment concentrations when the latter were less than approximately 200 μ g/kg dw (i.e., for each compound, Segment 1 had a slope not significantly different from 0; Figure 2). The cPAH concentrations were greater in clam tissue associated with sediment with higher concentrations, but data in this range were insufficient to estimate this relationship with confidence.

The tissue concentrations associated with the lower sediment concentrations were generally within laboratory analytical variability: Locations with sediment cPAH concentrations less than approximately 200 $\mu g/kg$ dw had tissue concentrations for these three cPAHs (i.e., benzo(b)fluoranthene, chrysene, and benzo(a)anthracene) that were within +/- 35% of the mean of these samples for all but one analyte in one sample. The three samples with the higher sediment concentrations showed a tendency to also have higher tissue concentrations: a 26 to 92% increase over the mean for the two moderate sediment concentration locations, and an order of magnitude increase for the highest sediment concentration location.

Regression models using organic carbon (OC)-normalized sediment concentrations and lipid-normalized tissue concentrations were also investigated. The correlations between dry weight and OC-normalized sediment concentrations and between wet weight and lipid-normalized tissue concentrations were very good (Pearson correlations of 0.96 or better for sediments, and of 0.99 or better for tissues). The OC-and lipid-normalized results looked very similar to those presented for dry weight sediment and wet weight for tissues, respectively.

These data suggest that higher cPAH concentrations in sediment tend to be associated with higher concentrations in clam tissue; however, this relationship is poorly defined by these data, because a tissue response that exceeds analytical variability is limited to just three data points (or fewer). At lower sediment concentrations, there was no trend in the detected tissue concentrations with increasing sediment concentrations, and all of these samples maintained a cPAH TEQ that was close to the target tissue level (values ranged from 1.8 to $2.6 \,\mu\text{g/kg}$ wet weight [ww]).

Other factors (e.g., exposure to surface water), as discussed in Section 4.4 in the memorandum, may play a more substantial role in determining clam tissue concentrations at lower sediment concentrations. Clams' exposure to cPAH from those other factors may not be directly related to the exposure of co-located sediments.

⁶ The slopes of the line segments at these lower sediment concentrations were not significantly different from zero (p-values > 0.14).



⁵ Based on Table 4-10 in the clam quality assurance project plan (Windward 2018), +/- 35% of the mean comprises the laboratory performance-based precision limits for cPAHs.

3.2 POREWATER AND THE RELATIONSHIP BETWEEN CPAHS IN CLAM TISSUE AND SEDIMENT

This section presents details regarding the evaluation conducted to answer Question 2b of the memorandum: "Did porewater help to improve our understanding of the relationship between cPAHs in clams and sediment?"

For the 10 samples for which C_{free} cPAH porewater concentrations were available, the individual cPAHs with minimal non-detects in tissue samples were the same cPAHs described above: benzo(b)fluoranthene, chrysene, and benzo(a)anthracene. Several methods were used to evaluate these data:

- Scatterplots and pairwise correlations were reported for tissue vs. sediment (both dry weight and OC-normalized), tissue vs. porewater, and porewater vs. sediment (both dry weight and OC normalized) (Figures 3 and 4).
- ◆ Spearman's rank correlation was used to summarize the relationships among these data pairs. The Spearman's rank correlation coefficient summarizes any monotonic correlation (e.g., Y tends to increase whenever X increases) and, unlike Pearson's linear correlation, does not rely on linearity or bivariate normality. Spearman's rank correlation is thus less influenced by extreme values (because ranks are used instead of concentrations). Like Pearson's, possible values of Spearman's rank correlation range from -1 to +1. A rank correlation of 0.5 indicates moderate association between the two variables. Unlike a linear (Pearson) correlation, which is directly associated with a linear regression, Spearman's rank correlation is not directly tied to a specific model form and requires only that the model be monotonic (i.e., linear, log-linear, etc.).

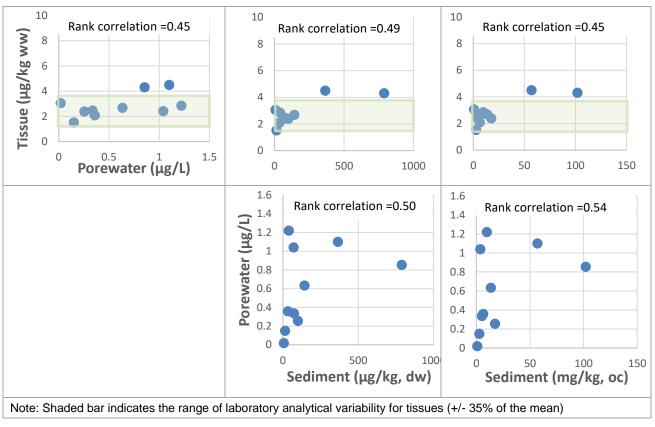


Figure 3. Benzo(b)fluoranthene compared among porewater (C_{free}), sediment, and clam tissue

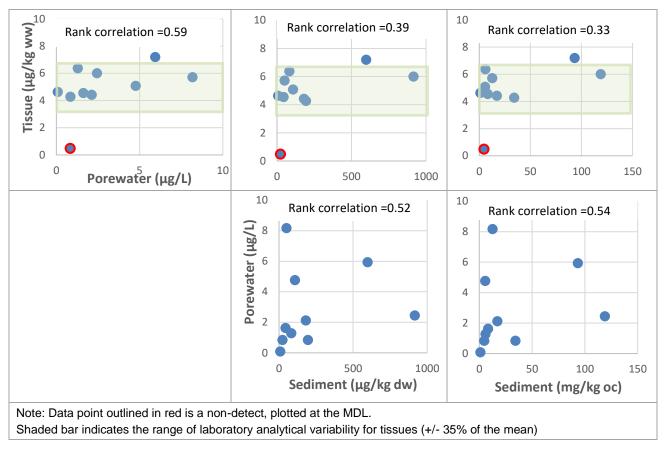


Figure 4. Chrysene compared among porewater (Cfree), sediment, and clam tissue

The correlations among the endpoints for the cPAH data are moderate at best (Table 3). Results for benzo(b)fluoranthene and chrysene are presented in Figures 3 and 4; results for benzo(a)anthracene show similar relationships. Porewater data were only moderately correlated with sediment concentrations (correlations \geq 0.5, Table 3), so the unique contribution of porewater over sediment cannot be isolated, and vice versa. Perhaps the most important aspect of these data is that at the 13 locations with sediment cPAH TEQ values less than approximately 200 μ g/kg dw, the variability of tissue concentrations was relatively low (i.e., within analytical variability), so correlations within this region may be spurious.

Table 3. Spearman's rank correlation coefficients (and p-values) for individual cPAHs with high detection frequency in clam tissues and co-located sediments and porewater

	Spearman's Rank Correlation Coefficients (p-values)				
Paired Data ^a	Benzo(b)fluoranthene	Chrysene	Benzo(a)anthracene		
Tissue vs. porewater	0.45 (p=0.19)	0.59 (p=0.08)	0.43 (p=0.22)		
Tissue vs. sediment (dry weight)	0.49 (p=0.15)	0.39 (p=0.26)	0.26 (p=0.47)		
Tissue vs. sediment (OC normalized)	0.45 (p=0.19)	0.33 (p=0.35)	0.39 (p=0.26)		
Porewater vs. sediment (dry weight)	0.50 (p=0.14)	0.52 (p=0.13)	0.53 (p=0.12)		
Porewater vs. sediment (OC normalized)	0.54 (p=0.11)	0.54 (p=0.11)	0.50 (p=0.14)		

Sample size is 10 for all pairwise comparisons.
 cPAH – carcinogenic polycyclic aromatic hydrocarbon

OC – organic carbon

4 RI/FS Data with cPAH RAL Exceedances

This section presents a summary of the RI/FS data with cPAH RAL exceedances. Table 4 presents the locations with exceedances of the site-wide cPAH RAL of 1,000 μ g/kg dw, and Table 5 presents the intertidal locations with exceedances of the cPAH intertidal RAL of 900 μ g/kg dw. As discussed in the memorandum, the majority of these locations will be remediated because of RAL exceedances for other chemicals that are either co-located or nearby, or because of benthic RAL exceedances for PAHs.

Table 4. RI/FS locations with exceedances of the site-wide cPAH RAL

Location Name	Sample Year	River Mile	cPAH TEQ (μg/kg dw)	Intertidal Location?	Other RAL Exceedances at this Location? ^a
DUD043	1995	0.4	1010 J	no	yes
NFK006	1994	5	1060 J	yes	no
TRI-036	2006	1	1100	no	no
DR124	1998	1.5	1100	no	no
DR047	1998	0.9	1100	yes	no
TRI-047T	2006	1.3	1100	no	yes
DUD037	1995	0.7	1180 J	no	yes
R44	1997	4.2	1200 J	no	yes
SB-12	2004	4.1	1200 J	yes	yes
R41	1997	4.2	1200	no	no
K-02-1	1991	0.7	1200 J	no	yes
DR112	1998	2.1	1200	no	no
LDW-SS9	2005	0.1	1200	yes	no
SS-1	1993	1.3	1250	no	yes
DR040	1998	0.5	1300	no	yes
DR050	1998	1	1300	no	yes

Location Name	Sample Year	River Mile	cPAH TEQ (μg/kg dw)	Intertidal Location?	Other RAL Exceedances at this Location? ^a
DR120	1998	1.4	1300	no	yes
LDW-SS48	2005	1.3	1400	no	yes
EAA2-SED-1	2007	2.2	1400 J	yes	yes
DR141	1998	2.4	1400 J	no	yes
R63	1997	4.4	1400	yes	no
TRI-045	2006	1.3	1400	no	no
LDW-SS311	2006	0.6	1500	yes	yes
LDW-SS112	2005	3.7	1500	yes	yes
DR017	1998	1	1500	no	no
DR174	1998	2.7	1600	yes	yes
DR044	1998	0.6	1600	no	yes
LDW-SS46	2005	1.3	1600 J	no	yes
LDW-SS335	2006	2.7	1700 J	yes	no
DR038	1998	0.4	1700	no	yes
B4a	2004	1.4	1700	yes	yes
LDW-SS114	2005	3.8	1800 J	yes	yes
DR033	1998	0.1	1900	no	yes
SB-3	2004	4.2	1900 J	no	yes
DR136	1998	2.2	1900	no	no
LDW-SS157	2005	3.8	1900	yes	no
DUD010	1994	0.6	1910 J	no	yes
B3b	2004	1	2200	no	yes
SB-1	2004	4.2	2300 J	no	yes
LDW-SS115	2005	3.8	2400	yes	no
LDW-SS509	2009	0.5	2900 J	yes	yes
LDW-SS35	2005	1	3000	no	yes
LDW-SS24	2005	0.5	3000	yes	yes
LDW-SS95	2005	2.7	3100 J	no	yes
R22	1997	3.8	3500	yes	yes
LDW-SS312	2006	0.6	4200	yes	yes
LDW-SS530	2009	2.7	4400 J	yes	yes
EAA2-SED-2	2007	2.2	4800 J	yes	yes
R23	1997	3.8	6600	yes	yes

^a Other RAL exceedances include both human health and benthic RALs.

cPAH – carcinogenic polycyclic aromatic hydrocarbon dw – dry weight

FS – feasibility study

J – estimated concentration

RAL – remedial action level RI – remedial investigation TEQ – toxic equivalent



Table 5. RI/FS locations with exceedances of the intertidal cPAH RAL

Location Name	Sample Year	River Mile	cPAH TEQ (μg/kg dw)	Beach Play Area?	Clamming Area?	Other RAL Exceedances at this Location? ^a
R77	1997	4.6	920 J	no	yes	no
DR189	1998	2.5	990	yes	yes	yes
LDW-SS93	2005	2.7	1000 J	yes	yes	no
NFK006	1994	5	1060 J	no	no	no
DR047	1998	0.9	1100	no	yes	no
SB-12	2004	4.1	1200 J	no	no	yes
LDW-SS9	2005	0.1	1200	yes	yes	no
EAA2-SED-1	2007	2.2	1400 J	no	yes	yes
R63	1997	4.4	1400	no	yes	no
LDW-SS311	2006	0.6	1500	no	yes	yes
LDW-SS112	2005	3.7	1500	no	yes	yes
DR174	1998	2.7	1600	no	yes	yes
LDW-SS335	2006	2.7	1700 J	yes	yes	no
B4a	2004	1.4	1700	no	yes	yes
LDW-SS114	2005	3.8	1800 J	no	yes	yes
LDW-SS157	2005	3.8	1900	no	yes	no
LDW-SS115	2005	3.8	2400	no	yes	no
LDW-SS509	2009	0.5	2900 J	yes	yes	yes
LDW-SS24	2005	0.5	3000	yes	yes	yes
R22	1997	3.8	3500	no	yes	yes
LDW-SS312	2006	0.6	4200	no	yes	yes
LDW-SS530	2009	2.7	4400 J	yes	yes	yes
EAA2-SED-2	2007	2.2	4800 J	yes	yes	yes
R23	1997	3.8	6600	no	yes	yes

Other RAL exceedances include both human health and benthic RALs.

 $\label{eq:cpah} cPAH-carcinogenic\ polycyclic\ aromatic\ hydrocarbon$

dw - dry weight

FS – feasibility study

J - estimated concentration

RAL – remedial action level

RI – remedial investigation

TEQ – toxic equivalent

5 References

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