Verification of 1998 303(d) PCB Listing, Inner Budd Inlet

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Verification of 1998 303(d) PCB Listing Inner Budd Inlet

by
Steven Golding

Environmental Assessment Program
Olympia, Washington 98504-7710

April 2003

Waterbody No. WA-13-0030, 390KRD
Budd Inlet (Inner)

Publication No. 03-03-016

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Abstract

Inner Budd Inlet was placed on the 1998 303(d) list for polychlorinated biphenyls (PCBs) based on a single composite sample of mussels collected from a culvert at the mouth of Moxlie Creek where it enters East Bay. That sample had a total PCB concentration of 21 µg/Kg wet weight (ww). This concentration exceeded the 303(d) listing criterion of 1.4 µg/Kg ww which was in effect at the time of sampling. In 1998, EPA revised their health criteria for PCBs. Based on EPA's reassessment of the cancer potency of PCBs, the new 303(d) listing criterion was set at 5.3 µg/Kg ww.

For this current project, more intensive sampling of mussels was conducted on September 6, 2002, to determine if the Inner Budd Inlet PCB listing continues to be warranted. Samples of blue mussel (*Mytilus trossulus*) from the sites in East Bay were found to have PCB concentrations ranging from 7.0 to 9.6 µg/Kg ww. All of these concentrations exceed the current 303(d) listing criterion of 5.3 µg/Kg ww. The PCB mixture found was in a form most closely resembling PCB 1254. A fourth site at Priest Point Park had an estimated PCB 1254 concentration of 3.0 µg/Kg ww.

The new PCB data for mussels in East Bay are sufficient to support the continued listing of the Inner Budd Inlet waterbody segment. Only the reference site, north of the listed segment at Priest Point Park, had a PCB concentration lower than the criterion.

Acknowledgements

The author appreciates the work of Dale Norton and Art Johnson in reviewing this report. Randy Coots provided excellent ArcView support for this project. Paul Anderson assisted in the field.

Samples for this study were analyzed by the Ecology Manchester Laboratory. Myrna Mandjikov performed PCB Aroclor analyses. Members of the Organics Extraction Unit performed lipids analysis.

The final report was edited and formatted by Joan LeTourneau.

Introduction

Inner Budd Inlet is included in the 1998 federal Clean Water Act 303(d) list for polychlorinated biphenyl (PCB) concentrations exceeding the listing criterion in edible shellfish tissue (Table 1). The listing is based on an analysis of blue mussels (*Mytilus trossulus*; formerly known as *M. edulis*) collected by the Washington State Department of Ecology (Ecology) in 1995 from the head of East Bay at the mouth of Moxlie Creek (Johnson and Davis, 1996). The listing criterion is based on EPA bioaccumlation factors applied to the EPA National Toxics Rule criterion for human health.

Table 1. 303(d) Listings for Budd Inlet.

	1996 (303)d List	1998 (303)d List
WRIA	13	13
Waterbody Name	Budd Inlet (Inner)	Budd Inlet (Inner)
Parameter	PCBs	PCB 1254
Latitude	--	47° 2' 42"
Longitude	--	122° 53' 42"
New ID#	--	390KRD
Old ID#	WA-13-0030	WA-13-0030

Inner Budd Inlet (segment ID WA-13-0030/390KRD) is on the 1998 303(d) list for PCB concentrations in edible shellfish tissue that exceed the EPA National Toxics Rule criterion for human health. The listing is based on analysis of bay mussels (*Mytilus* sp.) collected by Ecology in 1995 from the head of East Bay at the culvert at the mouth of Moxlie Creek (Johnson and Davis, 1996).

The Johnson and Davis study was a screening analysis of pesticides and PCBs in mussels collected from six marine locations. The sampling sites ranged from background areas (such as Padilla Bay) to areas known to be contaminated (such as the Hylebos and Duwamish waterways). Table 2 shows the PCB data from this study.

The Budd Inlet sample had a total PCB concentration of 21µg/Kg wet weight (ww). The PCB mixture most closely resembled Aroclor 1254. PCBs found in the environment tend to become altered through weathering and/or metabolic processes, resulting in changes in their constituent PCB congeners.

Each sample consisted of the entire soft parts from 33-84 individual mussels. The Budd Inlet sample was prepared from 30 mussels with a mean shell length of 52 mm. The samples were analyzed by the California Department of Fish & Game, Water Pollution Control Laboratory, using GC/ECD methods described in Rasmussen and Blethrow (1991) and Magoon (1993).

Table 2. PCB Concentrations (µg/Kg, wet weight) Measured in Marine Mussels Collected by Ecology on May 25, 1995.

Aroclor Equivalent	Hylebos Waterway	Duwamish Waterway	**Budd Inlet**	Chambers Creek	Ilwaco (Col. R.)	Padilla Bay
PCB 1016	nd	nd	**nd**	nd	nd	nd
PCB 1221	nd	nd	**nd**	nd	nd	nd
PCB 1232	nd	nd	**nd**	nd	nd	nd
PCB 1242	nd	nd	**nd**	nd	nd	nd
PCB 1248	18	nd	**nd**	nd	nd	nd
PCB 1254	46	32	**21**	6	6 N	2 J
PCB 1260	6 J	12 J	**nd**	2 J	nd	nd
Total PCBs	70 (est.)	44 (est.)	**21**	8 (est.)	6 (est.)	2 (est.)

From Johnson and Davis (1996)
nd = not detected
J = estimated value
N = tentatively identified

At the time the 1998 303(d) list was developed, the EPA human health criterion for PCBs was 1.4 µg/Kg ww for a 10^{-6} cancer risk. The PCB-1254 concentration in Budd Inlet mussels at the mouth of Moxlie Creek exceeded the listing criterion by a factor of 15.

In 1998, EPA revised their health criteria for PCBs (40 CFR 131, Water Quality Standards: Establishment of a Numeric Criteria for Priority Toxic Pollutants: States' Compliance – Revision of Polychlorinated Biphenyls (PCBs) Criteria). Based on EPA's reassessment of the cancer potency of PCBs, the new criterion was set at 5.3 µg/Kg ww.

This current project provides more intensive sampling of mussels from the listed segment in East Bay to better represent the water segment. The primary objective of this study was to determine if the 303(d) listing for PCBs in Inner Budd Inlet is still warranted.

Sampling Design

Ecology Water Quality Program Policy 1-11 (effective August, 1993; revised September 2002) states that sampling to obtain data for 303(d) listing considerations "should represent the waterbody segment as a whole – spatially and over time – rather than limited or isolated conditions."

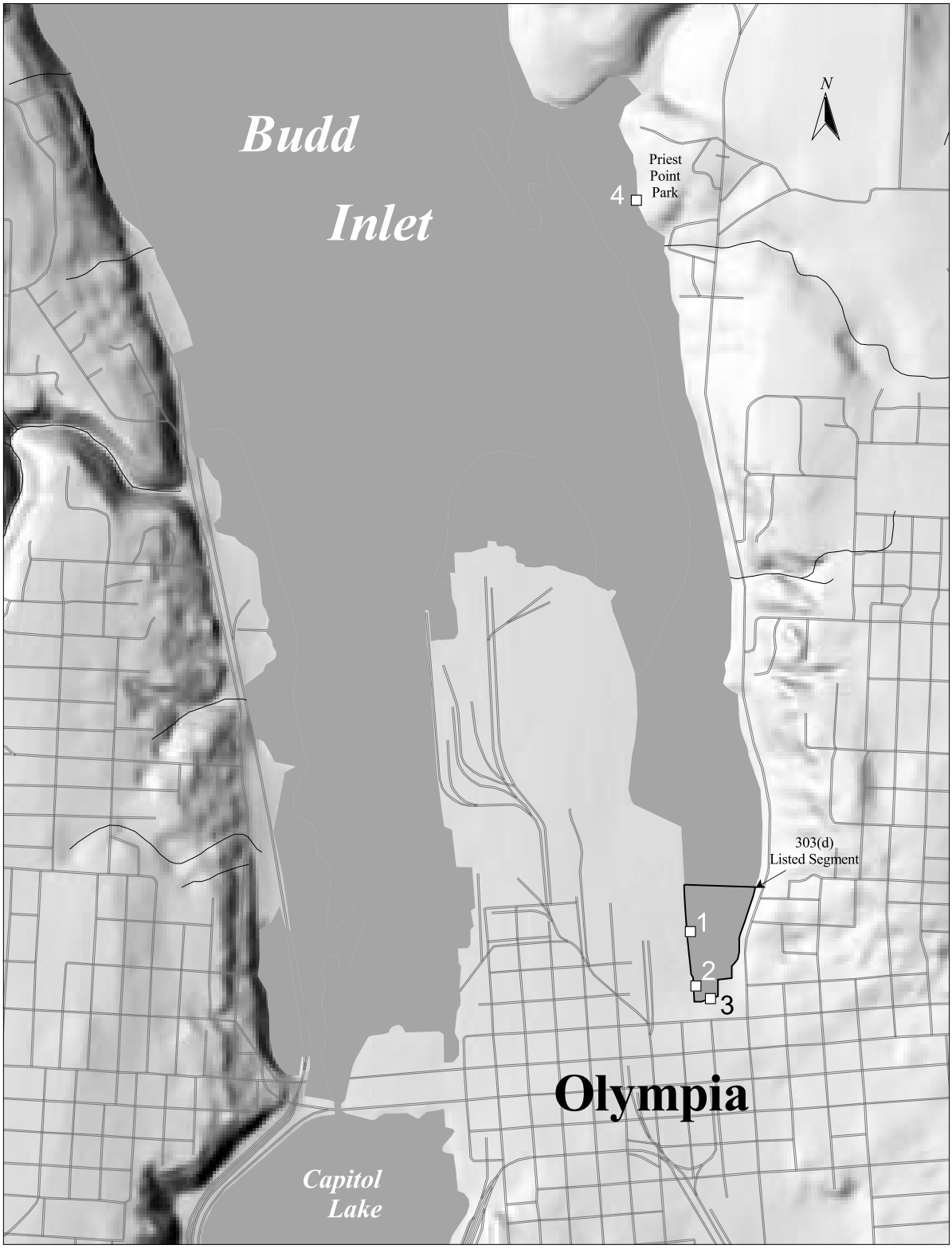
Three shoreline sites in Inner Budd Inlet from East Bay were selected for sampling of blue mussels (*Mytilus trossulus*). Mussels were also collected from a reference site at Priest Point Park near the northern boundary of the city of Olympia. Locations of the four sites are shown in Figure 1.

Sites 1 through 3 are distributed spacially within the 303(d) listed segment, which includes approximately ½ mile of shoreline. Mussels were collected from rocks and non-woody debris at Sites 1, 2, and 4, and from the exterior surface of the Moxlie Creek culvert entering East Bay at Site 3, the same site sampled in 1995. All mussels collected were at a distance of greater than 50 feet from treated wood. Descriptions and locations of the sampling sites are included in Appendix A.

Samples were collected on September 6, 2002 at low tide. Because PCBs are persistent compounds and organics integrate concentrations in tissues over time, the single collection date was considered to provide a temporally representative sample.

Policy 1-11 states that a single composite sample made up of at least five separate individuals provides sufficient data for 303(d) listing considerations. Four composite samples of 46-72 mussels were analyzed for this study.

Analysis of the samples included PCB Aroclors-equivalents and % lipids. Sample collection, handling, and tissue preparation methods were identical to the procedures used in 1995 to ensure the results were comparable.



□ – mussel monitoring site

Figure 1. Budd Inlet Mussel Sampling Locations.

Methods

Field

Approximately 50-75 individual mussels were collected from each of the four shoreline sites. Mussels were collected by hand and placed in laboratory cleaned one-gallon glass jars with Teflon lids. All personnel wore powder-free nitrile gloves. The samples were placed on ice in a cooler and then kept in a secure freezer at Ecology headquarters for later processing.

Processing

The samples were processed within the 6-month holding time for PCB analysis of frozen samples (Ecology, 2002). The number of individuals in each composite and range of shell lengths are shown in Table 3. Equipment used to dissect and homogenize the samples were cleaned with deionized water, laboratory-grade acetone, and hexane. The composite samples consisted of the entire soft tissue from each mussel. A summary of parameters, collection containers, preservation, and holding times appears in Table 4.

Table 3. Mussel Size Distribution by Site.

Site	# of Mussels Per Sample	Size Range
1	54	41 – 52 mm
2	63	39 – 64 mm
2 REP	72	37 – 73 mm
3	46	44 – 56 mm
4	51	33 – 56 mm

Table 4. Sample Size, Container, Preservation, and Holding Time by Parameter.

Parameter	Sample Size	Container	Preservation	Holding Time
PCB Aroclors	250 g	8 oz., organic-free	Freeze	6 months
Lipids (%)	20 g	(from PCB container)	Freeze	NA

Laboratory

The mussel tissue samples were extracted into a solvent mixture of 50/50 methylene chloride /hexane using the Soxhlet extraction procedure. Each extract was then eluted through a macro Florisil® column with 100% hexane. The extracts were then solvent exchanged to iso-octane, volume adjusted, and treated with concentrated sulfuric acid to remove interferences.

The extracts were analyzed for PCB Aroclors by dual column GC-ECD at the Ecology Manchester Laboratory using modifications of EPA SW-846 methods 3540, 3620, 3665, and 8082. GC/ECD also was used to analyze the 1995 sample.

Lipids analyses followed EPA-600 (EPA, 1980)

Data Quality

Manchester Laboratory prepared written case narratives on the quality of the PCB and lipids data for this project (Appendix B). The reviews include an assessment of compliance with holding times, instrument calibration, procedural blanks, surrogate recovery, matrix spike and matrix spike duplicate recoveries, and duplicate sample analyses. Matrix spike and matrix spike duplicate recoveries were within established quality control limits of 50 - 150%, and relative percent differences (RPDs) were within 15%. No problems compromising the accuracy, validity, or usefullness of the data were encountered.

Analytical variability in the data is indicated by laboratory duplicate results. The overall variability in data reported here is indicated by variability of paired field replicates. The field replicates for this project were obtained by dividing the mussels collected at a site into two groups for separate analysis.

Results of field replicates and laboratory duplicates are shown in Table 5. RPDs for PCBs were 10% for the laboratory duplicate and 28% for the field replicate.

Table 5. Results of Analyses of Mussel Samples from Inner Budd Inlet, September 2002.

Lab Duplicate Results

Site #	Lipids %	PCB 1016 Equivalent µg/Kg (ww)	PCB 1221 Equivalent µg/Kg (ww)	PCB 1232 Equivalent µg/Kg (ww)	PCB 1242 Equivalent µg/Kg (ww)	PCB 1248 Equivalent µg/Kg (ww)	**PCB 1254** Equivalent µg/Kg (ww)	PCB 1260 Equivalent µg/Kg (ww)
3	**0.30**	2.3 U	2.3 U	2.3 U	2.3 U	2.5 U	**6.6**	0.98 U
	0.45	2.2 U	2.2 U	2.2 U	2.2 U	0.89 U	**7.3**	0.89 U
RPD	40%	--	--	--	--	--	10%	--
RSD	28%	--	--	--	--	--	7%	--

Field Replicate Results

Site #	Lipids %	PCB 1016 Equivalent µg/Kg (ww)	PCB 1221 Equivalent µg/Kg (ww)	PCB 1232 Equivalent µg/Kg (ww)	PCB 1242 Equivalent µg/Kg (ww)	PCB 1248 Equivalent µg/Kg (ww)	**PCB 1254** Equivalent µg/Kg (ww)	PCB 1260 Equivalent µg/Kg (ww)
2	**0.35**	2.5 U	2.5 U	2.5 U	2.5 U	1.0 U	**9.5**	2.5 U
	0.24	2.3 U	2.3 U	2.3 U	2.3 U	0.90 U	**7.2**	2.3 U
RPD	37%	--	--	--	--	--	28%	--
RSD	26%	--	--	--	--	--	20%	--

RPD - Relative percent difference
RSD - Relative standard deviation
U - The analyte was not detected at or above the reported value.
bold - Detected analyte

Table 5 shows variability as RPD, the average deviation from the mean expressed as a percentage of the mean, as well as relative standard deviation (RSD), the standard deviation expressed as a percentage of the mean. The RSD of 28% between the lipid sample and lab duplicate for Site 3 is twice the 14% (RSD) for data from Seiders and Yake (2002). This higher RSD may indicate a lack of complete mixing of the prepared sample. PCB Aroclor-equivalent RPDs of 10% for the laboratory duplicate and 28% for the field replicate are within expected precision. The higher RPD for the PCB field replicate than for the laboratory duplicate may indicate heterogeneity of the mussels being sampled.

Results and Discussion

Results of PCB and lipid analyses for mussels at the four sites in Budd Inlet are shown in Table 6.

Table 6. Results of Analyses of Mussel Samples from Inner Budd Inlet, September 2002.

Name	East Bay	East Bay	East Bay	Priest Pt
Site	1	2	3	4
Date	09/06/02	09/06/02	09/06/02	09/06/02
Sample #	2458000	2458001	2458003	2458004
Lipids (%)	**0.36**	**0.35**	**0.30**	**0.48**
PCB as Aroclor Equivalent (µg/Kg wet wt)				
PCB 1016	2.5 U	2.5 U	2.5 U	4.8 U
PCB 1221	2.5 U	2.5 U	2.5 U	4.8 U
PCB 1232	2.5 U	2.5 U	2.5 U	4.8 U
PCB 1242	2.5 U	2.5 U	2.5 U	4.8 U
PCB 1248	1.0U	1.0U	2.5 U	0.97 U
PCB 1254	**9.6**	**8.4**	**7.0**	**3.0 J**
PCB 1260	2.5 UJ	2.5 U	0.98 U	0.97 U
TOTAL PCBs	**9.6**	**8.4**	**7.0**	**3.0 J**

U - The analyte was not detected at or above the reported limit.

UJ - The analyte was not detected at or above the reported estimated limit.

J - The analyte was positively identified. The associated numerical result is an estimate.

bold - Detected value

Exceeds 303(d) listing criterion of 5.3 (µg/Kg wet wt).

Lipids levels were similar among the three East Bay samples, ranging from 30 to 36%. The 48% lipid level of the Priest Point Park sample was significantly higher, at a confidence limit of 95% (2-tail Student's t).

The mussels had total PCB concentrations ranging from 7.0 to 9.6 µg/Kg ww. The highest concentrations were found at the north end of the shoreline. The PCB concentrations are less than half the 21µg/Kg ww concentration measured in mussels collected from the mouth of Moxlie Creek in 1995. The PCB concentration at the Moxlie Creek culvert site in 2002 was 7.0 µg/Kg ww. This is lower than the concentration of the 1995 sample by a factor of 3.

The estimated PCB 1254 concentration of 3.0 µg/Kg ww for the sample at Priest Point Park was significantly lower than those at the three other sites at a confidence limit of 90% (2-tail Student's t).

Mussel samples from the three sites in the 303(d) listed area were found to have total PCB concentrations of 7.0 - 9.6 µg/Kg ww. This exceeds the criterion of 5.3 µg/Kg ww. Only the mussel sample from Site 4, Priest Point Park north of the 303(d) listed area, had total PCB concentrations lower than the criterion.

For perspective, PCB concentrations reported in shellfish from several locations in the Puget Sound are shown in Table 7.

Table 7. Concentrations of Total PCBs Reported in Studies on Puget Sound Shellfish µg/Kg ww (median values).

Location	Reference	Sample date	Species	n =	Total PCBs
Budd Inlet	current study	2002	blue mussel	4	3.0-9.6
Budd Inlet (Moxlie Creek)	Johnson and Davis (1996)	1995	blue mussel	1	21
Budd Inlet	Mearns (2001)	1997-98	*Mytilus* sp.	1	36*
Duwamish Waterway	Johnson and Davis (1996)	1995	*Mytilus* sp.	1	32
Duwamish Head, Elliott Bay	Mearns (2001)	1997-98	*Mytilus* sp.	1	106*
South Seattle	Mearns (2001)	1997-98	*Mytilus* sp.	1	71*
Padilla/Fidalgo Bay	Johnson (2000)	1999	oyster, clam sp.	6	<0.025-2.8

* Estimated wet weight concentration based on 80% water (20% solids) rule of thumb for mussels (Mearns, 2001).

Concentrations of total PCBs up to 2.8 µg/Kg ww in samples collected in the Padilla Bay area during 1999 (Johnson, 2000) were similar to the 3.0 µg/Kg ww for the Priest Point Park sample in this current study. Total PCBs found in the Padilla Bay study and in the current study were considerably higher than the concentration for butter clams at the Samish Island reference area (<0.25 µg/Kg ww). Several earlier Ecology studies found no detected PCBs in shellfish but at relatively high detection limits.

Conclusions and Recommendations

Mussel samples from all three sites in the 303(d) listed area of the East Bay of Inner Budd Inlet have higher total PCB concentrations than the health-based criterion of 5.3 µg/Kg ww. This finding supports the continued listing of Inner Budd Inlet for total PCBs.

In light of the apparent downward trend in mussel tissue PCB concentrations that has occurred in the seven years since the 1995 sampling, it is recommended that, rather than a total maximum daily load study, follow-up sampling be conducted in five years.

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Appendices

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Appendix A

Descriptions and Locations of Sampling Sites

Site #	Location Description	Latitude	Longitude
1	On the western shoreline of East Bay, approximately 700 feet from the south end of East Bay.	47º 02’ 59” N	122º 53’ 40’ W
2	On the western shoreline of East Bay, approximately 100 feet from the south end of East Bay.	47º 02’ 52” 'N	122º 53’ 38” W
3	At the south end of East Bay, the exterior of the Moxlie Creek culvert emptying into East Bay.	47º 02’ 51” N	122º 53’ 37” W
4	Along the south shore of Ellis Cove, approximately 200 feet from the mouth of the cove.	47º 04’ 16” N	122º 53’ 52” W

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Appendix B

Laboratory Case Summaries

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Manchester Environmental Laboratory

7411 Beach Dr E, Port Orchard, Washington 98366

Case Narrative

February 10, 2003

Subject: Budd Inlet Mussel PCB

Samples: 02458000-02458004

Officer: Steve Golding

By: J. Daiker, K. Donegan, C. Milne
Organic Extractions Unit

Lipids Analysis

Analytical Method(s)

These samples were prepared and analyzed following Manchester Laboratory's standard operating procedure for the extraction of percent lipids in tissue using a 50:50 mixture of hexane and methylene chloride. The extracts were transferred to a calibrated centrifuge tube and brought to a volume of 10ml. One ml of sample was transferred to a pre-weighed beaker. Solvent was allowed to evaporate off in a hood overnight. Beakers were placed in a drying oven for four hours and then placed into a desiccator until completely cooled. Beaker and residue were weighed.

Holding Times

The method has no sample preservations or holding times.

Blanks

The "U" qualifier included in the results indicates no mass gains from solvent were detected in the laboratory method blanks.

Laboratory Duplicates

Sample 02458003 was analyzed in duplicate. The relative percent difference between the sample and the duplicate is 40%.

Comments

The data are useable as reported.

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Manchester Environmental Laboratory
7411 Beach Dr E, Port Orchard, Washington 98366

Case Narrative

March 5, 2003

Subject: Budd Inlet Mussel Study

Samples: 02458000 - 02458004

Officer: Steven Golding

By: M. Mandjikov

Polychlorinated Biphenyl Analysis

Analytical Method(s)

The Mussel tissue samples were extracted into a solvent mixture of 50/50 methylene chloride/hexane using the Soxhlet extraction procedure. Each extract was then eluted through a macro Florisil® column with 100% hexane. The extracts were then solvent exchanged to iso-octane, volume adjusted, and treated with concentration sulfuric acid to remove interferences before analysis by dual column GC-ECD.

These methods are modifications of EPA SW- 846 methods 3540, 3620, 3665, and 8082.

Holding Times

All samples were prepared and analyzed within the method holding times.

Calibration

The initial calibrations for all analytes are acceptable and within the established QC limits.

Blanks

No target analytes are detected in the procedure blanks.

Surrogates

Each sample was spiked with Decachlorobiphenyl (DCB) prior to extraction. All the surrogate recoveries are within the established QC limits.

Sample Duplicate

Sample 02458003 was prepared in duplicate to access the precision of this method. The relative percent difference between the duplicate samples is 10%.

Matrix Spike and Matrix Spike Duplicate

Four replicates of Sample 02458003 were prepared. Two replicates were spiked with the PCB Aroclors 1016 and 1260. All spike recoveries are within the established QC limits. All relative percent differences between the spiked samples are below 15%.

Comments

The Aroclor result for sample 02458004 is qualified, "J" as an estimate. Although the PCB congeners present in this sample identify the Aroclor as 1254, due to weathering the ratios of the congeners poorly match the pattern of the Aroclor 1254 standard.

The reporting limits of the Aroclors vary somewhat due to the amount of interference from Aroclor 1254 in the sample. In samples where the interference is great enough to obscure the congeners from Aroclors 1248 and 1260, the reporting limit has been raised to level above which these Aroclors can be detected independently from Aroclor 1254.

The data are useable as qualified.