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RESEARCH ARTICLE

Uptake of PCBs into sediment dwellers and trophic transfer in relation to sediment conditions in the Salish Sea

Brenda J Burd[a*], Christopher J Lowe[b], and Carmen Morales-Caselles[c]

[a]Ecostat Research Ltd., North Saanich, BC V8L 5P6, Canada; [b]Capital Regional District, Victoria, BC V8W 2S6, Canada; [c]Biology department, University of Cadiz, 11510 Cadiz, Spain

*bburd@telus.net

Abstract

We examined uptake of polychlorinated biphenyls (PCBs) into various marine sediment feeders relative to physical and geochemical factors and transfer to higher trophic levels. PCBs exceeding Canadian Council Ministers of the Environment Guidelines by 6–55× were found in industrialized harbours and some near-outfall sediments, indicating ongoing land input. Sediment PCBs were correlated with organic flux and content. Tissue PCBs were >10× sediment PCBs in all samples and highest in Victoria Harbour infauna, suggesting considerable uptake from these extremely contaminated, organically enriched, chronically disturbed sediments. Sediment PCBs were the primary predictor of tissue lipid PCBs followed by %fines. This results in generally higher tissue PCBs in more depositional regions. The lipid/sediment PCBs (uptake rate) declined with increasing sediment PCBs, acid volatile sulfides and benthos biomass turnover. PCB homologue composition did not change with uptake from sediments or at higher trophic levels, suggesting minimal metabolization in tissues. Trophic bio-magnification occurs since lipid PCBs were 2–100× higher in seal blubber than sediment feeders. PCBs were compared with polybrominated diphenyl ethers (PBDEs) for the same samples. PCBs were highest in industrialized harbours, whereas PBDEs were elevated in harbours but highest near wastewater discharges. This reflects differences in usage history, sediment dynamics, and affinities. PCBs appear to be more bio-accumulative and persistent at higher trophic levels than PBDEs.

Key words: benthos, PCBs, trophic transfer, sediment geochemistry

Introduction

Polychlorinated biphenyls (PCBs) are industrial chemicals that were manufactured from 1929 until banned in most of the industrial world in the late 1970s–1980s. PCBs are still ubiquitous globally as a consequence of their persistence and cycling in the environment, leakage from historically contaminated sites, and delivery via atmospheric processes from distant regions where regulatory controls may not yet be in place (Ross et al. 2004). The highest concentrations of PCBs in marine sediments and mussels are typically found near estuaries of large rivers flowing through urban and industrial regions (Olenycz et al. 2015), where carbon and nitrogen stable isotope analyses have shown that even low-flow creeks through landfills can be significant contamination vectors to the marine environment. Garrett and Ross (2010) described the distribution of PCBs in the south coastal inland region of British Columbia (BC), known as the northern Salish Sea, with the highest concentrations detected in Vancouver, Victoria, and Esquimalt harbours (see also DesForges et al. 2014). Cullon et al. (2012)

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also described Puget Sound (southern Salish Sea) as a regional "hotspot" for PCB contamination, making it a serious source of historic and (or) continued release from past industrial activities (Garrett and Ross 2010).

Studies suggest that PCBs are mostly accumulated in invertebrates and fish through ingestion of food particles with adsorbed PCBs (Gobas and Morrison 1999; Mearns et al. 2013) and particularly by digestion of contaminated lipids in the gut. Apex predators such as killer whales (*Orcinus orca*) and harbour seals (*Phoca vitulina*) are especially vulnerable to high accumulations of PCBs as a result of trophic bio-accumulation, high body lipid levels, long life span, and a limited ability to metabolize them (Muir et al. 1988; Ross et al. 2000, 2004; Rayne et al. 2004; Shaw et al. 2005; Hickie et al. 2007).

Strongly declining trends in concentrations of PCBs in harbour seals from the northern Salish Sea were found from 1984 through the 1990s (Ross et al. 2013). However, there was little change from 2003 to 2009, and similar patterns of steep declines in the 1980s followed by a levelling out after 2000 have been noted in the southern Salish Sea (West et al. 2017). Similarly, tissue PCB levels in various marine mammals in European waters are not declining and are much higher than in North America (Jepson et al. 2016). Epidemiological evidence and field sampling of PCBs in seals have led to the conclusion that these chemicals pose an ongoing reproductive risk (Reijnders et al. 2010; Murphy et al. 2015) as well as immuno-suppression (Hall et al. 2018). DeForges et al. (2018) modeled the continued effects of sediment PCBs on global killer whale populations, predicting that many populations feeding at the apex of the food chain and (or) living near industrial centers are at risk of population collapse. PCBs therefore remain a global problem due to their persistence, bioavailability for uptake, and resulting toxicity in higher trophic level organisms (Ross et al. 2013).

Arblaster et al. (2015) suggested that if the PCB sediment concentrations are equal to current Canadian sediment quality guidelines (SQGs: CCME 2001), tissue concentrations in most marine mammals can be expected to exceed health and consumption guidelines (see also Gobas and Arnot 2010). Alava et al. (2016) modelled this theory using a food-basket approach, showing that existing SQGs are inadequate to protect higher trophic levels in the BC marine ecosystem, particularly if climate change leads to an increase in contaminant exposure or if contamination leads to an increase in climate change susceptibility.

Despite being banned in Canada for almost 50 years, the persistent high levels of tissue PCBs in urban and industrialized areas of the Salish Sea on the west coast of Canada suggest that they are continuing to enter the marine food chain, most likely through sediment feeding. Otherwise, levels would be slowly declining in the Salish Sea as faunal mortality, sedimentation, and burial remove older and more refractory chemicals from the ecosystem.

To understand PCB persistence in marine fauna, we examined sediment physical and geochemical factors relative to uptake of PCBs into marine sediment feeders as well as transfer to higher trophic-level fauna. We compare and contrast this with results from a companion study of polybrominated diphenyl ether (PBDE) uptake in the same sediments and tissue samples (Burd et al. 2019).

Methods

Study area

All tissue and sediment samples were collected from southern BC (**Fig. 1**). Most of the samples were from the Strait of Georgia (SoG), a relatively isolated glacial depression (up to 400 m deep) that makes up the northern half of the Salish Sea, with restricted water flow at the north and south ends and low–moderate bottom currents except at the tidal channels. The oceanography (Thomson 2014) and high sediment flux (Johannessen et al. 2003) in the SoG is driven by the massive freshwater discharge of the

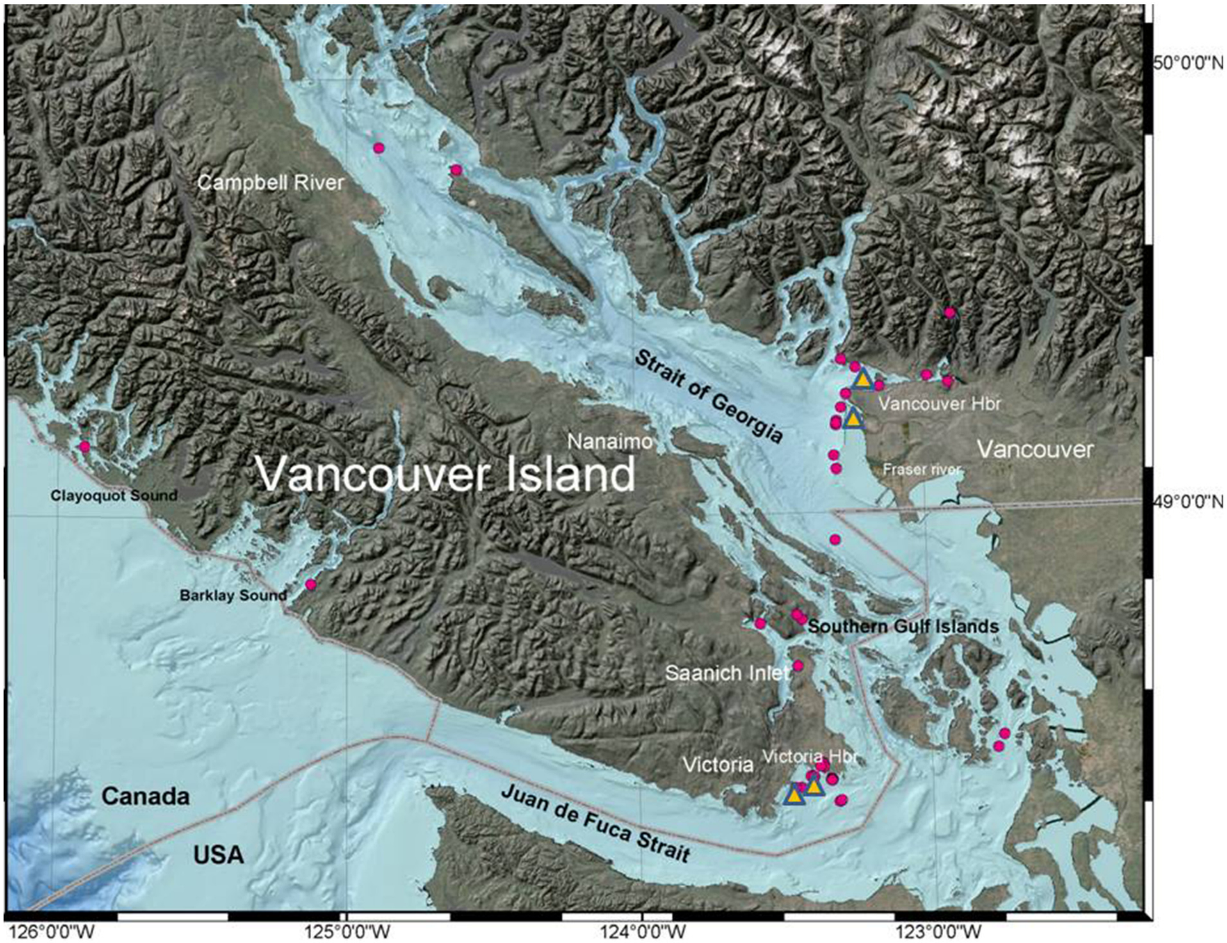

Fig. 1. Locations of matched tissue/sediment polychlorinated biphenyl samples (see Table 1 and Supplementary material S1) from the Salish Sea, southwestern coast of Canada. The Canadian portion of the Salish Sea includes most of the Strait of Georgia and half of Juan de Fuca Strait (see Canada/USA border). The four largest marine wastewater treatment plant outfalls are shown as orange triangles (Lion's Gate furthest north, Iona next south, Macaulay and Clover Points off southern Victoria). The southern Salish Sea is not shown but includes Puget Sound off the coast of Washington State, USA. Base map generation is from ESRI, USA ArcGis 9 with all added data points and levels generated from this study. Projection is NAD83.

Fraser River. The SoG is therefore an inland sea with high sedimentation of fines and high water column and sediment production (Burd et al. 2008a; Burd 2014).

The primary treatment Iona wastewater outfall off Vancouver (Fig. 1) discharges into the southeast quadrant of the SoG, along the deposition gradient for the Fraser River. Vancouver Harbour to the north (Burrard Inlet) also has relatively fine sediments, with more variable currents and reduced influence from the Fraser River (Burd 2014). Multiple industrial and urban discharges and shipping activities in Burrard Inlet include another primary treatment municipal wastewater outfall (Lions Gate) and various combined sewage overflows. The north end of the SoG is a shallower basin with minimal influence from Fraser River particulates (Johannessen et al. 2003). Sediments are less productive than in the southern basin (Burd 2014), with nutrient input dominated by reworked, fine marine detritus derived from phytoplankton production. Samples collected in Juan de Fuca Strait off the south end of Victoria and in the southern Gulf islands (Fig. 1), as well as on the west coast of Vancouver Island, are from sediments dominated by coarser sands with typically high currents in exposed areas. This includes the sample locations surrounding the pretreated (screened only) Macaulay and Clover Point municipal wastewater outfalls off Victoria (Fig. 1). These substrates are naturally productive (Burd 2014) due to a steady supply of suspended particulates from seasonal offshore upwelling and strong mixing around the southern Gulf islands (Thomson 2014). Nearby Victoria Harbour is a shallow and organically enriched sheltered bay, with various urban and industrial inputs and considerable bottom disturbance from shipping and dredging (UMA and Morrow 2007).

Samples

Sediment conventional data (total organic carbon (%TOC), acid volatile sulfides (AVS), δ15N (sediment δ15N is used to identify the source and quality of sediment organic material utilized by the sediment feeders and thus feeding dynamics as they relate to contaminant uptake as described in Burd et al 2014), grain size (%fines), and sedimentation ($g/cm^2/y$) and organic flux (in mg $C/cm^2/y$ rates) were compiled for 55 sample locations, all with one or more matched PCB sediment and (or) tissue sample (see Supplementary material S1). Locations for matched tissue/sediment samples are shown in Fig. 1 and geo-referenced in Table 1.

Table 1. Sediment factors, latitudes, and longitudes for matched tissue/sediment samples (see also Supplementary material S1).

Sample	Latitude	Longitude	Depth (m)	Moisture (%)	Organic flux (mg C/cm^2/yr)	Sediment flux (g/cm^2/yr)	AVS (umol/g)	% TOC	δ15N	% fines
AH10	48.37695	−123.4499	83	36.2	2.74	0.38	0.20	0.59	6.3	27
AH5	48.3778	−123.4699	74	35.4	2.74	0.38	0.3	0.72	7.4	27
AM10	49.8445	−124.8868	309	77.1	2.5	0.062	4	3.68	7.13	92
AM6	48.9367	−123.3133	186	61.2	37.8	2.7	1.3	1.3	5.8	85
C0	48.3943	−123.3459	65	22.9	1.9	0.38	0.62	0.5	7.6	10*
C1E	48.3944	−123.3442	70	44	3.572	0.38	11.3	0.94	7.4	31.2
C1NE	48.3960	−123.3445	59	39.6	2.926	0.38	1.06	0.77	7.9	26.7
C1NW	48.3961	−123.3479	59	33.3	3.686	0.38	5.96	0.97	7.7	35.4
C1S	48.3930	−123.3458	75	33	2.736	0.38	3.61	0.72	7.9	17.5
C1SE	48.3935	−123.3445	75	30.2	2.128	0.38	9.89	0.56	8.2	14.8
C1SW	48.3935	−123.3474	69	35.6	3.42	0.38	2.89	0.9	8.1	13.9
C1W	48.3943	−123.3477	70	32.1	4.18	0.38	5.96	1.1	7.3	13.5
C1W	48.3943	−123.3477	70	33.9	4.18	0.38	5.96	1.1	7.3	13.5
C1W	48.3943	−123.3477	70	31.2	4.18	0.38	5.96	1.1	7.3	13.5
C2E	48.3943	−123.3431	68	39.2	2.736	0.38	11.9	0.72	7.6	20.4
C2S	48.3924	−123.3458	78	29.5	2.698	0.38	0.201	0.71	8.0	20.8
CB1	48.3440	−123.3180	67	35.8	2.74	0.38	0.55	0.7	8.7	22.8
CB2	48.3449	−123.3165	65	39	2.74	0.38	1.13	0.83	8.3	33
CB3	48.3433	−123.3197	63	39.8	2.74	0.38	1.79	0.89	8.6	24.6
CB4	48.3467	−123.3135	63	37.6	2.74	0.38	2.67	0.88	8.2	21.2
FC1	48.4574	−122.7733	70	34.8	2.60	0.39	1.41	0.69	7.8	30.9

(*continued*)

Table 1. (*concluded*)

Sample	Latitude	Longitude	Depth (m)	Moisture (%)	Organic flux (mg C/cm^2/yr)	Sediment flux (g/cm^2/yr)	AVS (umol/g)	% TOC	δ15N	% fines
FC2	48.4859	−122.7527	59	25.8	2.60	0.39	0.2	0.66	8	11.5
IO15	49.1304	−123.3114	80	37.42	40	6	1	0.95	3.7	77.5
IO200N	49.2388	−123.2820	80	n/a	15	1.2	8.00	1	1.9	70
IO200S	49.2692	−123.2641	80	n/a	9.8	0.81	6.00	1.3	2.3	68
IO400N	49.2075	−123.2300	80	36.9	18.0	1.10	10.0	1.13	01.9	75
IO400S	49.1992	−123.3012	80	38.7	9.70	0.76	3.36	0.77	02.5	74
LG45	49.3488	−123.2782	52	60.3	3.78	0.21	1	1.82	4.3	71.6
M0	48.4026	−123.4104	60	39.1	36.9	0.84	26	4.4	5.2	30*
MAC 200M SE	48.4017	−123.4083	60	44.1	21.8	0.84	12.6	2.6	4.2	28
MAC200NW	48.4035	−123.4126	60	33.2	10.92	1.1	2	0.96	5.14	36
MAC400M N	48.4027	−123.4158	60	37.4	7.5	0.79	1.5	0.754	5.45	36
MAC400M S	48.4007	−123.4060	60	38.7	19	0.79	5.44	2.4	4.93	28.7
IO8	49.2085	−123.3000	80	46.83	22.78	1.3	16.5	1.13	1.75	80.4
IONA ZERO	49.2038	−123.3006	80	42.7	13.0	1.3	3.06	0.76	2.0	63.2
LG12	49.3299	−123.2283	58	59.8	4.25	0.21	3.05	2.03	4.3	94.7
VH1	48.4331	−123.3767	8	64.7	37	0.8	7.2	4.639	7.1	98.4
VH2	48.4295	−123.3724	8	41.9	35	0.8	31	4.413	6.5	49.04
VH3	48.4230	−123.3721	7.6	60.6	40	0.8	21.1	5.042	7	97.6
VH4	48.4239	−123.3832	8.3	46.9	24	0.8	3.07	3.075	6.5	71.7
1A1	49.4463	−122.8922	16.8	51.40	7.00	0.27		2.61	1.00	49.00
Dix0	48.8500	−125.1232	20	42.10	2.40	0.08	1.46	1.51	7.70	35.20
FR1	49.1000	−123.3034	10	26.80	60.00	8.00	1.3	0.19	4.90	11.10
GINP1A	48.7587	−123.4345	15.60	36.30	16.90	0.70	0.24	1.3	7.70	8.20
GINP2A	48.7693	−123.4512	13.3	25.40	16.90	0.70	0.21	0.67	6.00	18.90
PatBay	48.6536	−123.4535	23	19	13.9	0.37	0.2	0.29	7.1	7.9
PMV3	49.3071	−122.9816	12.2	28	—	—	0.2	0.34	3.7	7.9
PMV6	49.2852	−123.1485	20.4	47.6	4.25	0.21	1.65	0.92	4	72.3
WCVI3	49.1593	−125.9084	5	20.60	—	—	0.2	0.14	7.50	2.10
HG4	52.5142	−131.6110	21	63.7	—	—	4	5.23	—	60.8
NWC2	54.3329	−130.4228	4	29	—	—	0.2	0.67	—	28.1
PNEV1	50.5377	−125.9705	13	58.4	—	—	14.7	1.95	—	79.9
Blubber Bay	49.7935	−124.61355	3.5	21.22	—	—	—	0.18	—	20
Cowichan Bay	48.7505	−123.5791	4	21.15	—	—	—	0.25	—	20
Moody Arm	49.2904	−122.9076	15.5	60.46	—	—	—	3.66	—	90

Note: AVS, acid volatile sulfides; %TOC, percentage of total organic carbon; δ15N, stable nitrogen isotopes; %fine, grain size
*%fines value for M0 is artificial—15% actually, but about 30% gravel—readjusted after removing gravel. The same was done for C0.

Most of the sediment conventional data was collected using consistent methods, and concurrently with sediment PCB samples. For conventional sediment analysis methods see crd.bc.ca/about/document-library/documents/plans-reports/wastewater-stormwater; Burd et al. 2012a, 2014; USEPA Method 9060A (Total Organic Carbon; United States Environmental Protection Agency 2004) and Forestry Canada Method NOR-X319 (Kalra and Maynard 1991). Sedimentation and organic flux rates were estimated from proximate Pb^{210} dated core samples based on the method of Johannessen and Macdonald (2012) (for regional data and patterns see Burd et al. 2012a, 2012b, 2014; Johannessen and Macdonald 2012). Sediment AVS was used to infer geochemical conditions resulting from metabolism of organic material by microbes, as described in Burd et al. (2008b).

A description of all tissue types sampled, including feeding method, typical taxa and habitat type is given in **Table 2**. In total, 295 tissue PCB samples were collected, along with 58 sediment PCB samples

Table 2. Taxa descriptions and trophic types, along with text signifiers used throughout.

Text signifier	Description	Trophic type	Typical taxa	Sample type	Comments
Whole	Subtidal, infaunal mixed whole benthos assemblage	Mixed, primarily deposit feeders	Polychaetes, bivalves some crustaceans, brittle stars	O,B,H	Proportions vary; crustaceans and echinoderms rare in contaminated sediments
Subtidal bivalves	Subtidal, infaunal burrowing	Deposit feeders	Mixed, mainly *Axinopsida, Macoma*	O,B	—
Intertidal bivalves	Intertidal, burowing bivalves	Filter feeders	*Saxidomus gigantea* (butter clam) and *Mya arenaria* (soft shell clam)	B	Sequester toxins in siphons for long periods
Mytilus	Intertidal, epifaunal attached bivalves	Filter feeders	*Mytilus edulis*	O,B,H,R	—
Horse mussel	Subtidal, epifaunal attached bivalves	Filter feeders	*Modiolus modiolus*	O,B	—
Scallops	Subtidal epifaunal, mobile bivalves	Filter feeders	*Crassodoma gigantea*	B	—
Heteromastus	Subtidal, infaunal polychaete	Redox boundary deposit feeder	*H. filobranchus*	O	Opportunistic species near Iona outfall (low O_2 sed)
Polychaetes	Subtidal, infaunal	Deposit feeders and predators	Mixed taxa	O,B	—
Echinoderm	Subtidal, infaunal holothurians	Deposit feeders	*Brisaster, Molpadia*	B	—
Cerebratulus	Subtidal, infaunal nemertean	Nemertean predator	*C. californiensis*	O	—
Midshipman	Subtidal, epifaunal fish	Predator	*Porichthys notatus*	B	—
English sole	Subtidal, epifaunal fish	Predator	*Parophrys vetulus*	B,O	Whole, carcass, liver, muscle
Irish Lord (Sculpin)	Subtidal, Epifaunal fish	Predator	*Hemilepidotus hemilepidotus*	B,O	Whole
Crab hepato	Subtidal, epifaunal, mobile crustacean hepatopancreas	Predator	*Metacarcinus magister*	B,R,H,O	Hepatopancreas or muscle
Salmon	Pelagic fish	Predator	*Oncorhynchus* spp.	B	—
Seal	Marine mammal	Predator	*Phoca vitulana*	B	—

Note: All tissues are whole body samples unless otherwise indicated. B, background; O, near outfall/organically enriched; H, harbour; EH, harbour and organically enriched.

for 55 sample locations. A complete listing of all samples (matched and unmatched sediment and tissue samples) and general locations is given in **Supplementary material S1**, with a summary in **Table 3**. Overlap of some sediment samples with more than one tissue sample increased the total matched tissue/sediment sample set to 85 (see **Supplementary material S1**). Matched tissue/sediment samples were collected from (*i*) background areas over a range of depths throughout southern BC (**Fig. 1**), (*ii*) gradients (near- to far-field) away from 3 municipal wastewater outfalls, and (*iii*) the two largest urban/industrial harbours in coastal BC (Vancouver and Victoria Harbours). Only sediment samples that match one or more tissue sample locations are included in the analysis. Most of the unmatched tissue samples were from mobile fauna that could not be matched to exact sediment locations. Salmon and seal blubber samples were all collected from background areas in southern BC, rather than near expected PCB sources.

Twenty-eight matching PCB sample pairs for sediment and whole benthos (mixed community dominated by infaunal polychaetes and bivalves) were collected between March 2012 and January 2015 throughout the study region. Sediment and tissue field sampling procedures and preservation methods for whole benthos are described in detail in Dinn et al. (2012a, 2012b) and Burd et al. (2014). Whole benthos samples were collected from three replicate $0.1m^2$ Van Veen grabs and screened through 1 mm mesh stainless steel trays that were first cleaned with acetone (to remove organic material) and rinsed with distilled water. During screening, visible individual organisms were removed with clean forceps, placed in amber glass jars, weighed, and frozen. As many organisms as possible from a variety of available taxonomic groups and sizes were picked to meet the minimum analytical biomass threshold (10 g). During the screening and picking process, the composition of the community (identification of dominant organisms to the lowest level possible without a microscope) was recorded by trained taxonomic experts from Biologica Environmental Services, Victoria, BC. Where there was adequate material, some sub-samples of specific taxa types were separated out of the benthos for individual analyses (bivalves, polychaetes, echinoderms) in addition to whole benthos samples.

Nineteen matched tissue/sediment sample pairs of epibenthic filter feeders (swimming scallops (*Chlamys hastata*) and horse mussels (*Modiolus modiolus*)) were collected near the Clover Point municipal wastewater outfall and in background areas off Victoria. The sediment and horse mussel samples were collected and processed by the Capital Regional District (CRD) as part of their routine outfall monitoring program in 2012/2013 (crd.bc.ca/about/document-library/documents/plans-reports/wastewater-stormwater). Sample collection and processing methods for sediments and horse mussel tissue samples were described in detail in deBruyn et al. (2009). The scallop samples with matched sediments were collected and processed in January 2015 in the same manner as the horse mussels.

CRD also provided PCB tissue data for 89 samples of English sole (whole or tissues), Dungeness crab (muscle or hepatopancreas) or Sculpin (Irish Lord) collected in 2017 from near the Macaulay Point outfall, the Clover Point outfall (both off south end of Victoria, BC) and a background location near Sidney, BC. Because these samples were collected by trawl, no site-specific sediment data could be matched to them.

Eighteen sample pairs of intertidal blue mussels (*Mytilus edulis*) and nearby sediments collected as part of the coast-wide Pollution Tracker Program (Vancouver Aquarium, Vancouver, BC, Canada) in 2015 and 2016 were included in the dataset. Older (2005) tissue sample data provided by the Fisheries and Oceans archive (LEACA laboratory—see description in next section) from butter clams (*Saxidomus* sp.), soft-shell clams (*Mya* sp.), blue mussels (*Mytilus* sp.), crabs, salmon and seal blubber collected from various parts of the BC coast have also been included for comparison of congener and trophic patterns, but most of these did not have matching sediment data. The crab samples were split

Table 3. Faunal types and number of tissue and sediment matches for all taxa (Table 1).

Sample type (Table 2)	Faunal type	Biota	Sediment	Extra matches
B	Subtidal bivalves	2	1	—
O	Subtidal bivalves	1	2	—
B	Intertidal clam	11	1	—
O	Cerebratulus	3	y	3
B	Crab Hepato	3	1	—
R	Crab hepato	6	—	—
H	Crab hepato	4	Y	2
O	Crab hepato	2	—	—
B	Crab muscle	16	1	—
H	Crab muscle	3	y	1
R	Crab muscle	1	—	—
O	Crab muscle	2	—	—
B	Echino	5	1	3
B	English sole whole	32	—	—
O	English sole whole	27	—	—
B	English sole liver	4	—	—
O	English sole liver	3	—	—
B	English sole muscle	4	—	—
O	English sole muscle	4	—	—
O	Heteromastus	2	Y	2
B	Horse mussels	6	4	2
O	Horse mussels	12	12	—
B	Midshipman	1	Y	1
B	Mytilus	28	8	3
h	Mytilus	9	5	2
R	Mytilus	1	1	—
B	Polychaete	1	Y	1
O	Polychaete	1	y	1
B	Salmon filet	33	1	—
B	Scallops	2	1	1
B	Seal	30	—	—
B	Irish Lord	6	—	—
O	Irish Lord	3	—	—
B	Whole	7	5	3
h	Whole	5	4	1
O	Whole	15	10	1
Total		**295**	**58**	**27**

Note: See Supplementary material S1 for detailed listings. "Y" indicates a matched sediment sample that has already been counted.

into hepatopancreas and muscle tissues, the salmon samples were only muscle (fillet), and the seal blubber samples were only subcutaneous fat tissues.

Laboratory PCB processing

All sediment and tissue PCB samples were subject to identical extraction, cleanup, and quantification procedures. High-resolution PCB analyses of all tissue and sediment samples were conducted by SGS AXYS Analytical or LEACA (Laboratory for Expertise of Aquatic Chemical Analysis) at the Institute of Ocean Sciences (Fisheries and Oceans Canada), Sidney, BC, Canada. Both laboratories follow US EPA Method 1668 (well-labs.com/docs/epa_method_1668_1997.pdf) to measure PCBs by gas chromatography e-mass spectrometry (GCeMS) and are certified by the Canadian Association for Laboratory Accreditation to analyze 209 PCB congeners. Detailed methods for sample extraction, lipid determination, cleanup, and quantification are reported elsewhere (Ikonomou et al. 2001; Christensen et al. 2005; Frouin et al. 2013). Sample analysis batches included two procedural blanks, one standard reference material, and one duplicate sample. Recommended criteria for laboratory quality assurance and quality control (QA/QC) for high-resolution PBDE analytical analyses are also described in Golder (2017). There are 209 theoretically possible congeners that vary by degree of chlorination, location of chlorine atoms, as well as physicochemical properties and toxicology (Shiu and Mackay 1986; Safe 1994). Some of these congenors co-extract and had to be combined. The final combined congener list for PCBs is in **Supplementary material S2**.

All data are shown as blank-corrected dry weight pg/g or lipid weight unless otherwise stated. Any data anomalies were concurrently identified, and the original data and QA/QC material checked. The steps for data QA/QC and processing have been standardized as part of the Vancouver Aquarium Ocean Wise program under the former direction of Dr. Peter Ross (see acknowledgements). For the calculation of total contaminant concentrations, all nondetects were replaced by the detection limit minus blank correction. The same approach was used for comparisons of congener homologue groups.

Statistical evaluations of relationships between tissue and sediment PCBs or conventionals were done using linear correlations, least squares regression, multiple regression (Wessa 2017), and principal components analyses (PCAs; using software PRIMER 6; Clarke and Gorley 2006). PCB data for multiple regressions were log $(x + 1)$ transformed, and PCA was log $(x + 1)$ transformed to reduce multi-collinearity and meet normality requirements, as recommended for the statistical models or for proportional comparisons.

Results

Sediment conventional and geochemical measurements for matched tissue/sediment PCB samples are listed in **Table 1**. Not all parameters were available for all locations. Log sediment PCBs were moderately correlated with sediment AVS and %TOC (**Table 4**). Mixed-tissue log PCBs (dry wt or lipid wt) were both positively correlated with log sediment PCBs. Log lipid-tissue PCBs were also positively correlated with sediment %fines. The multivariate model suggests that variance in log lipid-weight PCBs is best explained by log sediment PCBs and %fines (adjusted $R^2 = 0.68$; $p < 0.0001$; **Table 5**). Percent TOC and AVS were removed from the multivariate model, since both co-vary with sediment PCBs and %fines and do not contribute to the variance explained.

Homologue group total PCBs for each sample are listed in **Supplementary material S3**. Total PCBs (dry wt or lipid wt) were highest in whole benthos tissues and sediments from the harbours (**Fig. 2**). Other taxa samples from harbours (*Mytilus*, crab muscle, and crab hepatopancreas) and seal blubber had the next most extreme lipid and dry weight tissue values. PCB values from near

Table 4. Pearson's correlations for total polychlorinated biphenyls (PCBs) and sediment factors.

A	Sample pairs	Whole benthos dry wt PCBs	Mixed tissue dry wt PCBs	Mixed tissue lipid wt PCBs	Sediment PCBs
Sediment PCBs	72	0.09	0.13	0.12	
Sed/%TOC	72	0.13	0.18	0.17	**0.99**
Depth (m)	72	−0.14	−0.16	−0.14	−0.29
Organic flux (mg C/cm^2/yr)	62	0.10	0.12	0.11	0.36
Sediment flux (g/cm^2/yr)	63	−0.02	−0.02	−0.02	−0.04
AVS	67	−0.03	−0.01	−0.01	**0.68**
%TOC	72	0.17	0.20	0.20	**0.57**
δ15N	65	0.07	0.07	0.07	0.13
%fines	72	0.13	0.14	0.14	0.11

B	Log sediment PCBs	Depth (m)	AVS	%TOC	%fines
Depth (m)	−0.22	—	—	—	—
AVS	**0.59**	−0.12	—	—	—
%TOC	**0.73**	−0.14	**0.59**	—	—
%fines	0.48	0.27	0.25	0.45	—
Log lipid tissue PCBs	**0.78**	−0.07	0.34	**0.56**	**0.62**
Log dry tissue PCBs	**0.67**	0.38	0.31	**0.57**	0.31

Note: The first table (A) is linear correlations with untransformed factors. The second table (B) is log tissue PCBs related to a subset of sediment factors that are most related. Bold text shows correlations (>0.55). %TOC, percentage of total organic carbon; AVS, acid volatile sulfides; δ15N, stable nitrogen isotopes; %fine, grain size.

Table 5. Multiple regression model for sediment factors that best explain log lipid tissue weight of total polychlorinated biphenyls.

	b	B	$B \times r_{xy}$
logsediment	0.4474	0.6336	0.4966
fines	0.0079	0.3127	0.1929
Multiple $R^2 = 0.6895$			
Adjusted multiple $R^2 = 0.6805$			
Standard error of multiple estimate	0.4025		

ANOVA Table					
Source	SS	df	MS	F	P
Regression	25.544	2	12.772	76.62	<.0001
Residual	11.5009	69	0.1667	—	—
Total	37.0449	71	—	—	—

Note: The multiple regression equation is of the general form; $Y = a + b_1X_1 + b_2X_2 + \cdots + b_kX_k$ where a is a starting-point constant analogous to the intercept, in a simple two-variable regression, and b_1, b_2, etc., are the unstandardized regression weights for X_1, X_2, etc., each analogous to the slope in a simple two-variable regression. In the present analysis, $a = 3.5486$ and the values of b are as indicated below. Values listed as B are the standardized regression weights.

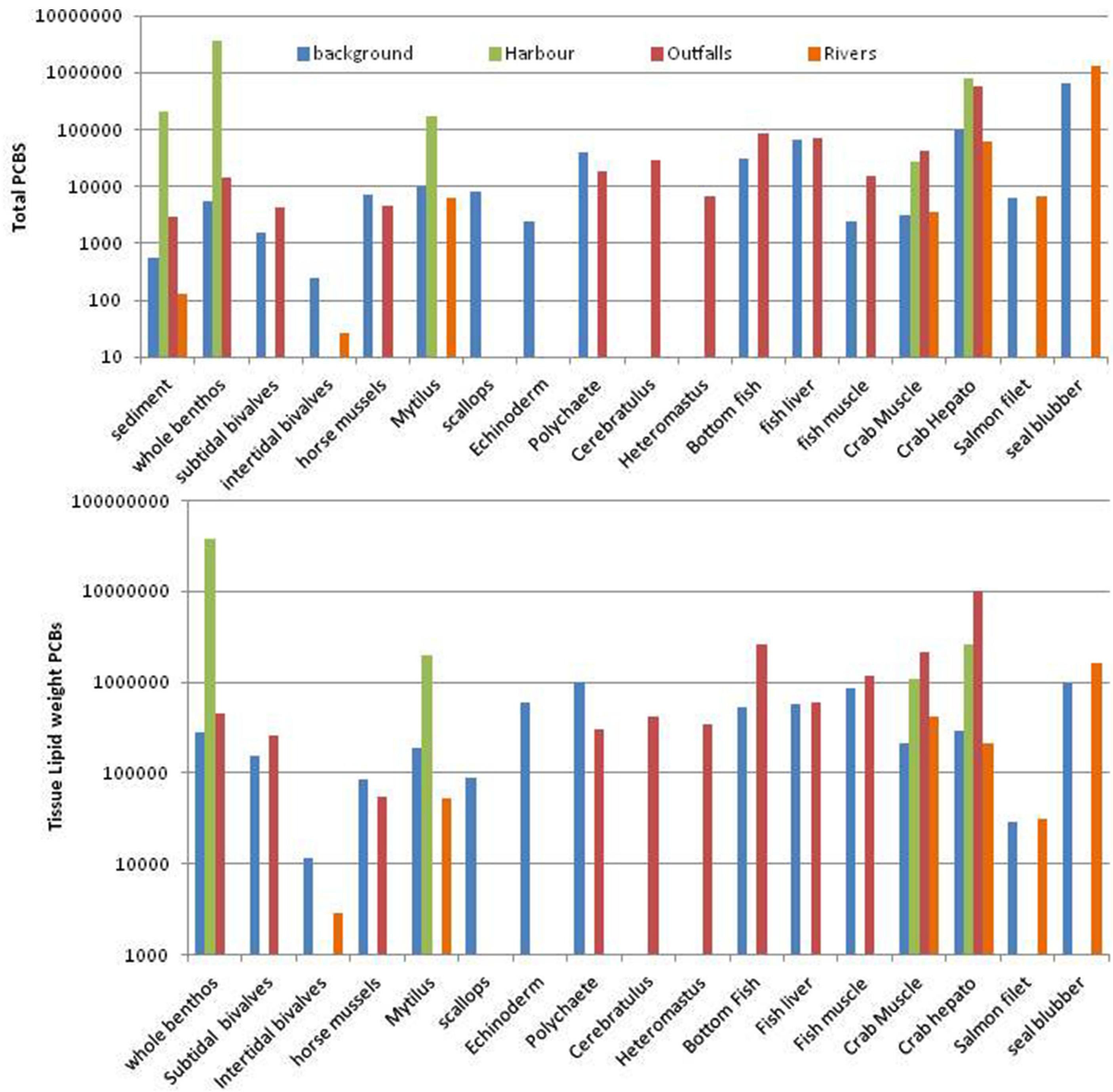

Fig. 2. Total dry weight and lipid-weight polychlorinated biphenyls (PCBs) averaged for each tissue type and habitat category. Note "Bottom Fish" includes Midshipman, English sole, and Irish lord.

wastewater outfalls tended to be slightly higher than background in sediments and whole benthos, with variable values in other taxa. Lowest PCB values were found in intertidal bivalves and river sediments, although the latter had a small sample size. When the harbour samples are ignored, the high lipid tissues (crab and seal) had the highest PCBs overall.

The log/log relationship between dry weight tissue and sediment PCBs for whole infaunal benthos, *Mytilus* and horse mussels (the largest matched groups) was typically flat, except for the high values in Victoria Harbour (**Fig. 3a**). The same relationship for lipid weight tissue and sediment dry weight PCBs appeared to be more linear. **Figure 3b** shows the log/log regressions for dry-weight and lipid-weight tissue PCBs and sediment dry-weight PCBs for the different habitat types for all tissue types. This function showed a more convincing linear trend. As a result, the correlations shown in **Table 4** indicate that lipid-weight PCBs were more closely related than dry weight to sediment PCBs.

The log ratio of lipid-weight tissue/sediment PCBs declined with increasing sediment log PCBs, resulting in reduced tissue accumulation ratios in the samples with the highest sediment levels, particularly in harbours. However, all ratios were greater than 10 (**Fig 4A**). Ratios also decreased with increasing %TOC and AVS (AVS only shown; **Fig 4B**), both of which were correlated with sediment PCB levels.

PCB homologue groups in most sediments and taxa were dominated by penta- and hexa-CBs, followed by tetra-CBs (**Fig. 5A**). Overall, little change in homologue proportions occurred with sediment uptake by the direct deposit or filter feeders, except for a modest increase in hexa-CBs in some taxa (**Fig. 5B**). There was some change in composition of higher taxa, including the three benthic fish taxa (midshipman, English sole, and Irish lord) and seal blubber. The intertidal clams showed the most unusual homologue profile, with some samples having much higher proportions of the more volatile compounds (di- and tri-CBs) than other taxa. This also occurred in one polychaete sample from near the Fraser River discharge. Unfortunately there were no matching sediment PCB values for these unusual samples.

Discussion

Sediment PCBs

Grant et al. (2011) described the geographic distribution and characteristics of surface sediment PCBs in the SoG. They found total PCBs to be negatively correlated with sedimentation rate. Johannesssen et al. (2008) have shown that PCB concentrations in sediment cores collected in the SoG showed a peak at depth and a decreasing trend toward the sediment surface, consistent with the history of release. Thus, PCB concentrations in the top 2 cm surface of the grab sediment samples in areas without any expected bottom disturbance and low bioturbation would be expected to resemble current seston concentrations of PCBs. In our study, extremely elevated PCBs were found in industrialized harbours, particularly Victoria Harbour (see also Hickie et al. 2007; Johannesssen et al. 2008; Grant et al. 2011; Morales-Caselles et al. 2017), with values exceeding Canadian Sediment Quality Guidelines (SQG: 21,500 pg·g^{-1}dw) by 6–55×. Extremely high PCB levels are also still evident in sediments from moderate–heavily industrialized areas in the southern Salish Sea (Puget Sound: Cullon et al. 2012; West et al. 2017). Monitoring by King County in Washington State (King County 2014; eopugetsound.org/magazine/IS/atmospheric_deposition) suggests that an urban plume of PCBs from old construction materials, paints, transformers, and fluorescent light fixtures that can volatilize into the air before attaching to surfaces or particles on or near the ground, covers Seattle and Lake Washington, as described for San Francisco, Chicago, Camden (New Jersey) and Toronto, Canada.

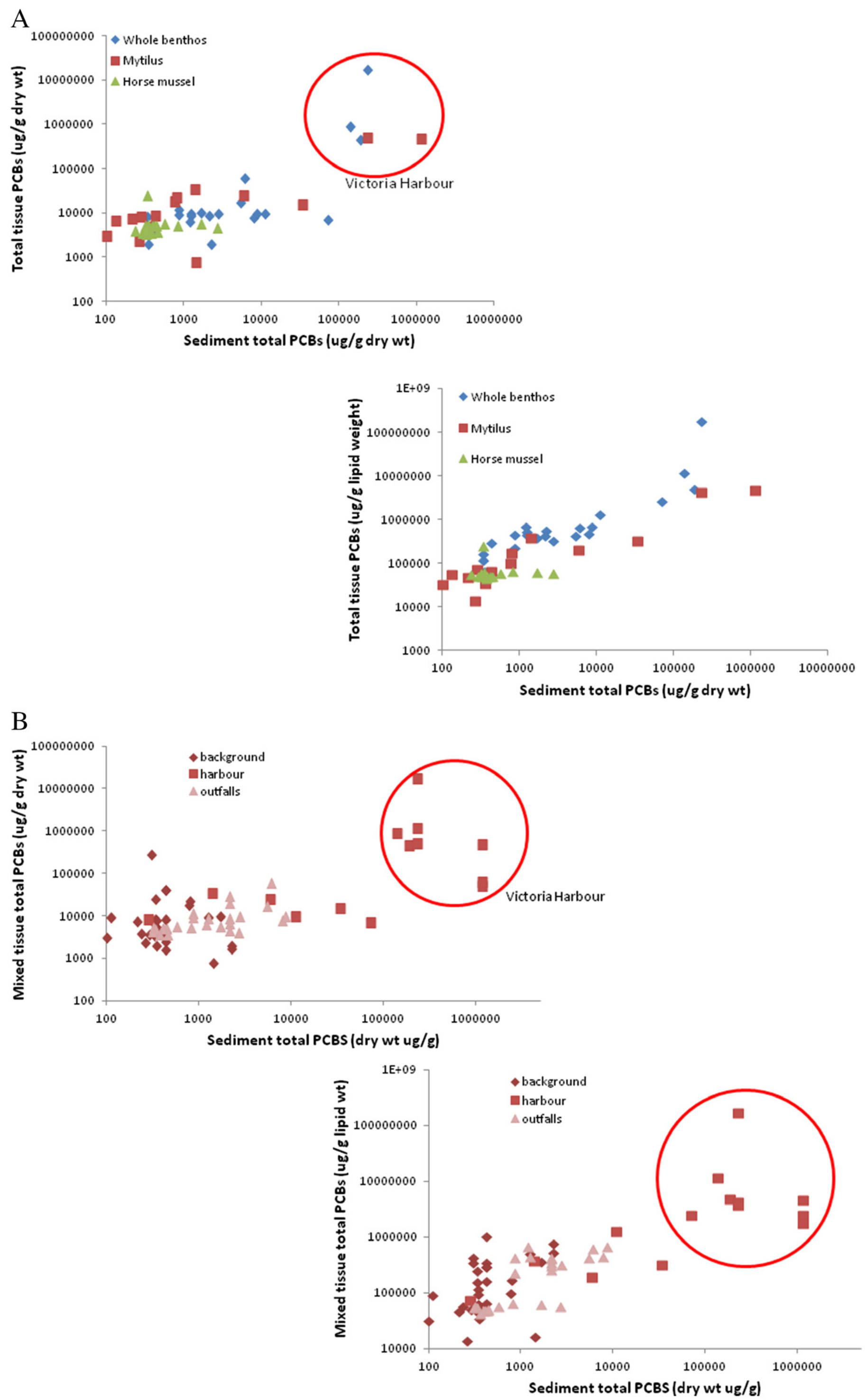

Fig. 3. Total polychlorinated biphenyls (PCBs) for the three most abundant matched taxa/sediment sample types as well as for all matched taxa/sediment samples for dry weight tissues and lipid-weight tissues for (A) the three most common taxa categories and (B) all sediment-dwelling taxa from different habitat types.

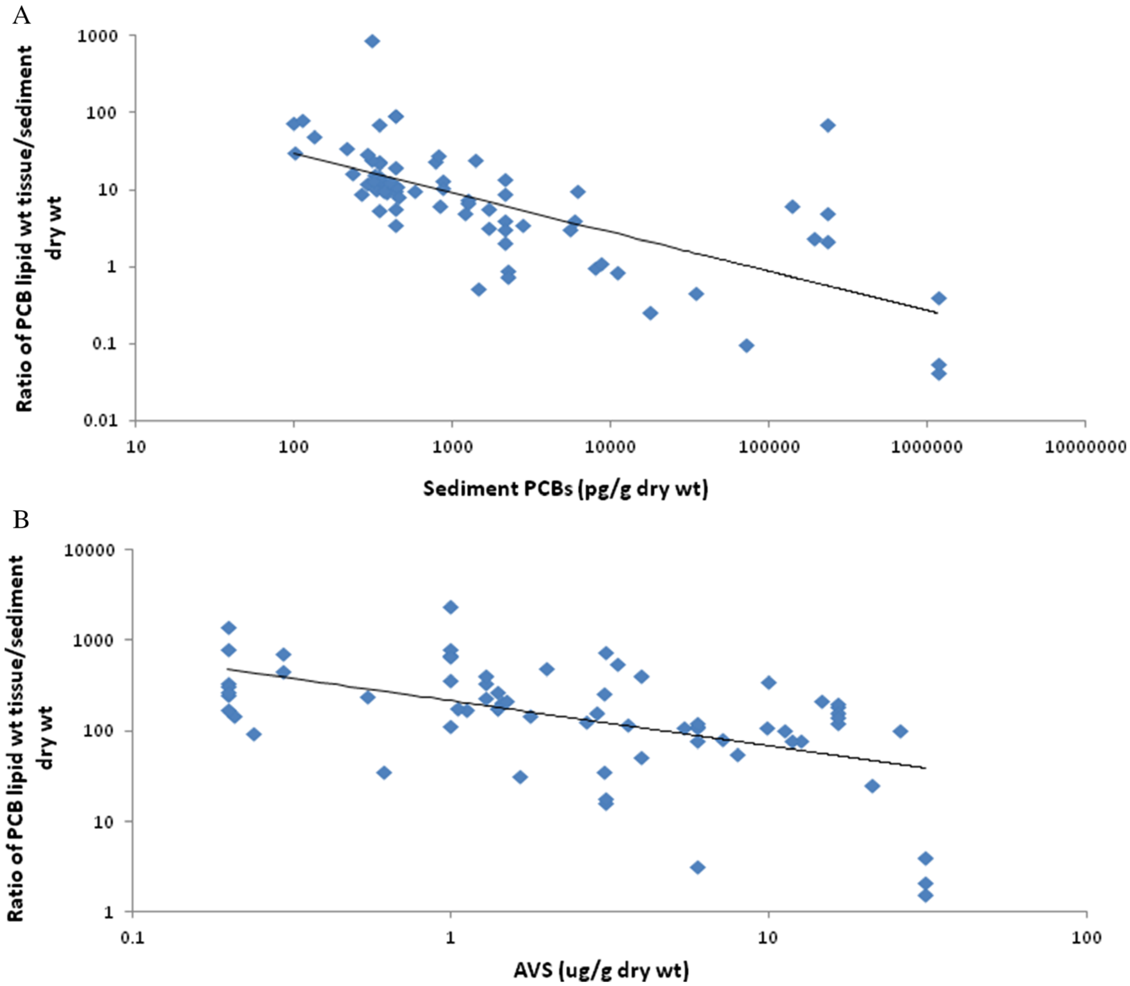

Fig. 4. Polychlorinated biphenyl (PCB) uptake rate (tissue/sediment ratio) relative to total sediment PCBs and sediment acid volatile sulfides (AVS), for both (A) dry weight and (B) lipid-weight tissue values.

These results suggest ongoing input of PCBs to urban sediments from land-based sources. Yang et al. (2012) suggested that the inadvertent introduction of PCBs into marine areas adjacent to industrial zones may be related to e-waste from recycling of electronics off the coast of China, which may have resulted in higher sediment levels after the year 2000. This may be relevant in the Pacific northwest as well. Although there are no landfill sources near Vancouver Harbour, the most extreme sediment PCB values in Victoria Harbour may be related to ongoing inputs from contaminated fill and historical industrial practices in the near- and fore-shore. Several recent water quality samples from Victoria Harbour (November 2018) identified total PCBs ranging from 2.5 to 20.8 ng/L which is 20–200× above the short-term acute water quality guidelines (BC water quality objectives - www2.gov.bc.ca/gov/content/environment/air-land-water/water/water-quality/water-quality-objectives). There is thus a need to identify previously undocumented sources of PCB contamination entering marine waters around the Salish Sea.

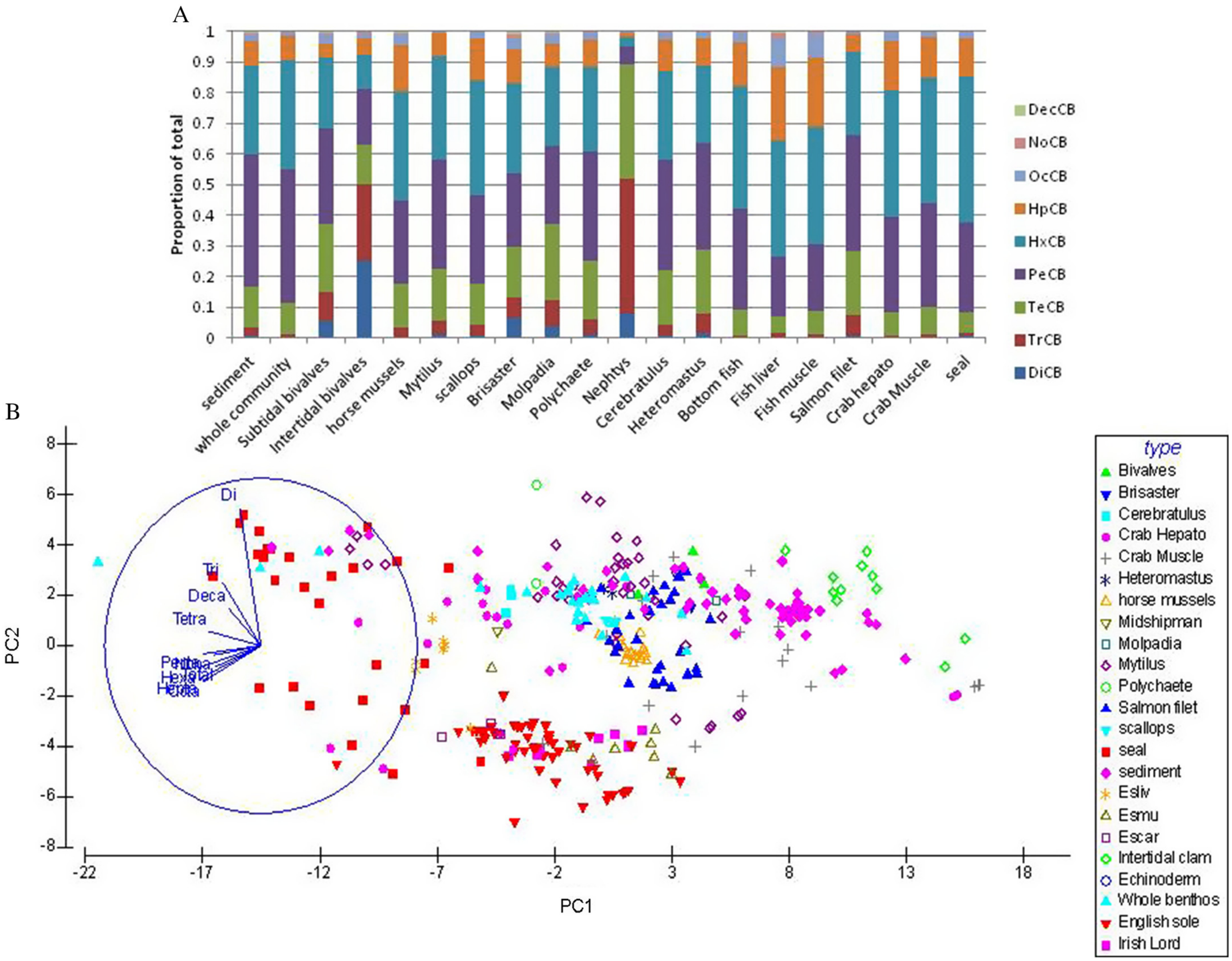

Fig. 5. Proportion of polychlorinated biphenyl (PCB) homologue groups in all sediment and tissue samples, shown as (A) averages for each taxa type, and (B) a principal components plot for all separate samples standardized. Esmu, Esliv, and EScar are acronyms for English sole muscle, liver, and carcass, respectively.

PCBs are typically hydrophobic and tend to bind strongly to organic carbon and particulates (Cullon et al. 2012). PCBs in sediments in the present study were most correlated with organic flux and content (AVS, %TOC, less so with organic carbon or OC flux), as found by Grant et al. (2011). Despite this, PCBs were modestly elevated in sediments near outfalls, suggesting that municipal wastewater is a minor source (Johannesssen et al. 2008). The higher organic content in sediments near outfalls tends to be from relatively "newly fixed" (low N values—see Burd et al. 2013) and recently deposited organic material and therefore is likely to have lower levels of the legacy PCBs. However, the high organic content and input (and therefore high AVS) in the shallow Victoria Harbour (UMA and Morrow 2007) adsorbs the above-described continuing PCB input from land-based sources. The PCBs are then incorporated into pelagic and benthic algal and bacterial production, and ultimately ends up in sediments. This highlights the contamination hazard of any shallow, organically enriched harbour with surrounding historical input sources of PCBs.

In the current study, sediment %fines did not correlate with sediment PCBs. This has been found in other marine studies (Yang et al. 2002; Grant et al. 2011; Li et al. 2012; Gao et al. 2013). The lack of correlation between sediment PCBs and %fines may be related to the fact that organic matter tends to be associated with specific sediment size fractions rather than the finest particles, which are often inert clay. For example, Zhao et al. (2010) found that sediment PCBs and %TOC were not associated with %fines but rather with different specific particle size ranges in different hydrographic conditions. Adeyinka and Moodley (2019) found that the sorption of PCB congeners onto particulates was dependent on the amount of organic matter, particle surface area, pH, and pore size distribution.

The source of organic matter may also influence PCB association with sediment grain size. Kuzyk et al. 2010 found elevated $\sum$PCB concentrations and more highly chlorinated PCB signatures in surface sediments underlying eutrophic regions and lower PCB concentrations and weathered signatures in oligotrophic regions dominated by "old" marine organic matter in Hudson's Bay, Canada. Similarly, Grant et al. (2011) found that sediment surface PCBs were highest in "older" organic sediments in the SoG. Staniszewska et al. (2011) suggested that black carbon seems to be more important for PCB absorption in sediments than %TOC, which is why charcoal is often used for remediation (for review of methods see Gomes et al. 2013; Galran et al. 2015). The structure of non-natural inorganic particles in sediments may affect PCB adsorption. Parks et al (2014) indicated that nanotubules may be effective remediators as they adsorb well to PCBs, and also reduce bioavailability to benthic organisms. These studies highlight the importance of understanding not only organic content of sediments relative to PCB distributions, but also the type, source, and age of organic material (see also Frouin et al. 2013).

In more remote areas far from source, there is rarely a close association between sediment PCBs and sediment organic matter or structure. Ameur et al. (2011) found no significant correlation between total PCB concentrations and organic carbon content or sediment %fines (<40 um), suggesting that sediment properties did not play an important role in controlling the PCB levels in sediments off Tunisia. Hong et al. (2012) studied the spatial distribution and potential source of PCBs in surface sediments from Bering Sea, Chukchi Sea, and Canada Basin. They found that sedimentary properties including grain size, water content, loss on ignition, TOC, and black carbon showed no apparent relationships with sediment PCBs. This indicated that the distribution of PCBs away from source is controlled mainly by atmospheric transport and deposition, sedimentation rates, mixing, partitioning, and sorption in the water column and sediments. This may have relevance in some of the background sample locations far from source in the current study.

Other important factors affecting sediment PCB levels include sediment resuspension due to tides, waves, dredging, anchor dragging, bottom trawling, or bioturbation. Joseffsson et al. (2011) described a study in which the sediment-to-water flux of PCBs was inversely related to the burial depth (2–10 cm) and inversely related to the hydrophobicity of the congener. The flux was therefore most pronounced for less hydrophobic contaminants and was linked to the bio-irrigating behaviour of deep burrowing polychaetes. Contaminants previously considered buried at a "safe" depth can thereby be remobilized to surface sediments and (or) the overlying water column.

In the shallow Victoria Harbour, considerable sediment disturbance is caused by dredging, anchor chains, and prop wash, all of which can remobilize buried PCBs and other contaminants (UMI and Morrow 2007; Morales-Caselles et al. 2017) from remnant-contaminated sediment "pockets" (Goossens and Zwolsman 1996; National Research Council 2007). This is likely a factor influencing Victoria and Vancouver Harbour sediments. Monitoring studies suggest that recurrent natural disturbances such as tides and waves may cause the majority of contaminant release from sediments in shallower marine environments (Roberts 2012). This may explain why persistent bioaccumulation of organic contaminants can occur despite dredging efforts targeted at removing contaminated sediments (Voie et al. 2002), such as those in certain areas of Victoria Harbour.

Tissue PCBs relative to sediments

The sediment dry-weight PCB concentrations were considerably lower (>10×) than tissue concentrations across all taxa for background, harbours, and outfalls. Therefore, the sediment-dependent food chain in the Salish Sea still represents a significant and refractory repository of PCBs. Desforges et al. (2014) indicated that phytoplankton levels of legacy PCBs were uniform across sites in southern BC, suggesting that biotic uptake has become spatially uniform in coastal BC due to recycling over time. In the present study, salmon, intertidal bivalves, scallops, and horse mussels had the lowest lipid-weight PCBs. Since these are pelagic feeders, it suggests lower availability of water column PCBs (particulate-bound) than sediment availability in the SoG. Trophic level bio-magnification of PCBs is likely, as a result of either the pelagic or benthic food chain transfer, since lipid-weight concentrations were highest in seal blubber, as well as in echinoderms and polychaetes from background areas. These organisms showed an average apparent bio-magnification relative to whole benthos at a rate of 2–3×. This magnification rate would be even higher for a pelagic food chain.

In this study, dry weight PCBs in sediment dwellers were poorly related to sediment PCB levels (**Fig. 3a** and **3b**, **Table 4**; Kuzyk et al. 2005; deBruyn et al. 2009). The disconnect between dry-weight tissue and sediment PCBs suggests at first that fauna are no longer taking up PCBs from sediments but just recycling them within the food chain. However, the highest tissue PCBs (dry weight or lipid weight) were found in a few Victoria Harbour whole infaunal community samples, with values much higher than in local sediments, crab tissues, and *Mytilus*. Since the infauna are typically annuals, this suggests that there is still considerable and rapid uptake and bio-concentration of PCBs directly into the food chain from these extremely contaminated, organically enriched, and chronically disturbed sediments. In addition, lipid-weight PCBs were more clearly related to sediment PCBs, reflecting the strong lipid-affinity of these chemicals. This also suggests that the long-term storage of PCBs is primarily associated with body fats. Most marine organisms lose considerable lipids during reproduction (typically annually). For example, the lipid-rich reproductive byproducts of benthic infauna and epifauna typically rise to the ocean surface, where they become part of the pelagic food chain. However, ultimately most of the PCBs would settle back to sediments since they are strongly bound to organic particulates. The only way to remove them from the food chain is to bury them with clean sediments too deeply for bioturbation or other disturbances to remobilize.

The multiple regression model illustrates that sediment PCBs are the primary predictor of tissue lipid PCB levels, but that this relationship is positively enhanced by increasing sediment %fines. Since sediment PCB levels were not as strongly related to %fines as tissue lipid levels were, this may suggest that areas with high fines are more susceptible to sediment resuspension (Grant et al. 2011). Multi-species mesocosm experiments have shown that contaminants remobilised by burrowers and irrigators are bioavailable to co-occurring organisms, leading to greater body burdens of contaminants in nearby infaunal species (Roberts 2012). Since elevated sediment %fines are an indicator of depositional environments, this results in higher tissue PCBs in general in the SoG relative to Juan de Fuca Strait (see Burd et al. 2019).

The ratio of lipid/sediment PCBs (uptake rate) declined with increasing sediment PCBs and increasing AVS or reducing conditions in sediments (**Fig. 4**). These patterns imply that in very high sediment concentrations, some of the PCBs are not bio-available, possibly because they are bound to nonlabile particles (such as black carbon or nano-plastics—see next section). PCB uptake rate is further diminished by reducing conditions in sediments, which is related to increased or pulsed organic input to sediments. Although the geochemical data collected in this study were insufficient to show

how this uptake dynamic might work, pulsed or seasonal input of organics from algal blooms are particularly prevalent in the extremely high PCB content, organically enriched, shallow Victoria Harbour sediments, whereas organic input is more constant and different in source and composition near the primary or untreated wastewater outfalls. It is impossible from the current study to know how the seasonality and composition of such inputs could affect sediment organism uptake of contaminants, particularly in light of the changing metabolic requirements surrounding reproductive cycles. There are several possible explanations that can by hypothesized for uptake patterns, one of which is that Matturro et al. (2016) point out that the only known way to dechlorinate PCBs in marine sediments to less toxic and bio-accumulative congenors is via anaerobic bacteria in reducing conditions (see also Quensen et al. 1988; Abramowicz et al. 1995) such as those found in Victoria Harbour and around the outfalls. Although the data were not collected to test this hypothesis, it is possible that the most bio-accumulative PCBs (penta, hexa, hepta), which dominated all taxa in the current study, are rapidly being broken down in anaerobic sediments with high PCB levels, making them less available for uptake by sediment-feeding fauna.

A further comparison was made between mixed tissue lipid uptake ratios and limited matched data available for infaunal production and biomass (from Burd et al. 2012a, 2012b; 2013). This limited comparison is suggestive that increasing infaunal biomass turnover (ratio of production to biomass (P/B)) may reduce uptake rate of tissue PCBs in local taxa (**Fig. 6**). This makes sense if we assume that high P/B implies that smaller, more rapid turnover fauna dominate benthos. These fauna have a short time to accumulate sediment contaminants before they become part of the food chain or are recycled in sediments. Rapid biomass turnover promotes a higher rate of bacterial breakdown of organic matter in sediments, which would contribute to more rapid metabolization and dechlorination of PCBs. This is most likely to occur in areas of high organic input (oxygen-reducing) or frequently disturbed sediments. Therefore, the concurrent patterns of increasing biomass turnover, sediment AVS and PCBs collectively result in reduced PCB uptake rates into benthos.

The filter-feeding *Mytilus* tended to higher tissue PCBs similar to direct sediment feeders but higher than other filter-feeding bivalves. *Mytilus* lives in high current, rocky areas likely to have considerable sediment resuspension, making PCBs more bio-available (Roberts 2012). Sediment resuspension in intertidal rocky areas is likely to be intense due to wave and tidal action and can enhance the growth of water column bacteria and protozoa through release of nutrients. Mobilised organic contaminants may be accumulated by these microorganisms and subsequently taken up by nearby filter-feeding organisms (Latimer et al. 1999; Zarull et al. 1999; Eggleton and Thomas 2004). Similarly, bioaccumulation studies by Voie et al. (2002) showed that PCB concentrations in mussels and lipid-containing

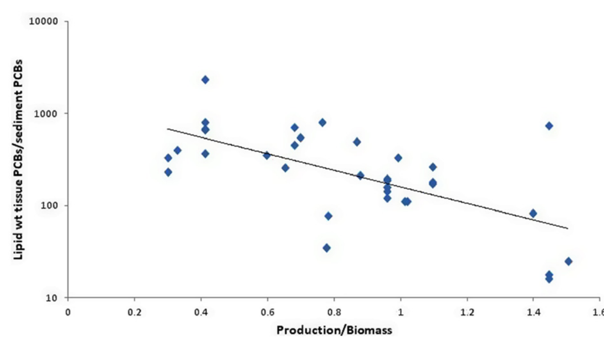

Fig. 6. Relationship of log lipid-weight polychlorinated biphenyl (PCB) uptake rates for mixed sediment dwellers versus infaunal biomass turnover (P/B) in samples with matched values (P/B values from Burd et al. 2012a; 2012b; 2013). P/B is ratio of production to biomass.

semipermeable membrane devices transplanted 1 m above a contaminated sediment site increased during remediation dredging and up to 6 months after dredging activities had ceased.

As the correlations, multiple regressions, and figures in this study suggest (see **Figs. 4**, **6**; **Tables 4** and **5**), the relationship between sediment conditions and PCB uptake in sediment feeders is complex. Sediment homologue profiles are affected by source and recent input of historical deposits (for which we have no real data) and by sediment geochemistry. Uptake rate seems to be related to sediment homologue composition, resuspension, sediment structure (fines), long-term oxygen conditions (AVS), biotic community functioning (P/B ratios which reflect complex trophic and biomass interactions), and other possible unknown factors that are beyond the scope of this paper. With the few samples and limited goechemical and biophysical data available it is only possible to show uptake patterns and hypothesize about reasons for them.

Trophic accumulation

West et al. (2017) found that PCBs in Puget Sound (southern Salish Sea) herring and English sole declined after 1997 in undeveloped areas, but not in the moderate to heavily industrialized basins. Some PCB increases in English sole were noted after that date, and tissue levels were much higher than for conspecific sole from densely populated areas of the Baltic Sea. Notably, the southern SoG PCB (see stations on the US side near the Canadian/US border of the Salish Sea—**Fig. 1**) values for English sole described in West et al. (2017) were similar to that for a single specimen of Midshipman, as well as the English Sole from the current study. As in Puget Sound, values in the current study were much higher for whole bottom fish and muscle tissue near outfalls than in background areas. However, the salmon tissue levels in the current study were <1/5 those of pelagic fish measured in Puget Sound (West et al. 2017). These results collectively suggest that source control of PCBs in Puget Sound has not been particularly effective, and levels of concern are still evident in Puget Sound's pelagic prey base.

Hickie et al. (2007) predicted it would take 14–57 years for PCB concentrations to fall below an effects threshold of 17 mg total PCBs/kg blubber lipids (= 17000 pg/g lipid-weight) for southern resident killer whales in Puget Sound. Lipid-based PCB concentrations in all faunal tissues except intertidal bivalves in the current study were much higher than the effects threshold, so 50–60 years may be more realistic. Lachmuth et al. (2010) did extensive model simulations with recommendations of a protective sediment PCB limit of 200 pg/g dry wt (about 1/4 mean background levels measured in the current study) and suggested that disposal of dredged material from highly contaminated harbour sediments will likely add to the body burden of southern resident killer whales in critical habitats. Current CCME marine sediment PELs are about 16,000 pg/g dry wt (CCME factsheet: st-ts.ccme.ca/en/index.html?factsheet = 173), which is only exceeded in harbour and a few near-outfall sediment samples in the current study.

Congenor patterns

Highly chlorinated or hydrophobic PCBs tend to adsorb to suspended particulate matter and deposit close to the source (Gao et al. 2013; Hong et al. 2003). These tend to desorb from sediment particulates slowly, in the order of years (Eggleton and Thomas 2004), and become more resistant to desorption over time (Chen et al. 1999). The lighter PCB congeners (particularly di- and tri-CBs) disperse more readily through atmospheric and oceanographic processes (Ross et al. 2004; Grant et al. 2011). The fact that a lighter congenor mix doesn't show up in the sediment homologue composition for Victoria Harbour samples suggests that there is a fairly constant supply of legacy PCBs with the classic (heavier) industrial configuration, entering the marine system. In summary, proximity to source and duration in sediments influences the PCB mixture to which marine fauna are exposed (Hong et al. 2012).

In the present study the congenor group patterns were very similar between all taxa and sediments (see also Burd et al. 2014), with the exception of one *Nephtys*/polychaete sample from near the Fraser River discharge. The lighter PCB homologues predominant in the remote intertidal filter-feeding bivalves are assumed to reflect the untransformed parent congenor patterns from sediments (Porte and Albaiges 1993), which are likely less chlorinated far from source due to the long residence time of legacy PCBs with no recent input source (Grant et al. 2011). Unfortunately, no matching sediment PCB data were available for these distant clam samples. In addition, these clams had the lowest tissue PCB levels of any samples in the study. Abramowicz et al. (1995) suggested that the lower chlorinated PCBs may be less bio-accumulative.

The tetra-, penta- and hexa-CB congenor groups dominated all other samples (tissues and sediments), similar to the configuration reported elsewhere (Porte and Albaiges 1993), where deposition of land-based PCBs is still proximate. This suggests considerable spatial sorting and distribution of PCBs over time throughout the BC coast, as well as remarkable stability in congenor composition in sediments and the sediment-based food chain. Homologue composition does not seem to shift notably at higher trophic levels, suggesting that very little PCB metabolization or selective uptake occurs at any trophic level.

PCBs versus PBDEs (from Burd et al. 2019)

The differences in historical usage of PBDEs and PCBs and their divergent distributions in marine sediments in the SoG are discussed in detail in Johannesssen et al. (2008). Sediment PBDEs are still predominantly entering marine systems through wastewater outfalls and combined sewer overflows. In contrast, the legacy PCBs have dispersed much more broadly, but also appear to still be entering marine systems from historical sources in industrially developed harbours. **Figure 7** shows the matched total PCB and PBDE dry weight sediment and tissue values from this study and from Burd et al. (2019). There is a reasonable log/log relationship ($R^2 = 0.57$, $n = 283$; $p < 0.001$), reflecting similar long-term dispersion patterns of these contaminants in southern BC. Despite the longer legacy period, PCB tissue levels were considerably higher (up to 100×) than PBDE levels in many samples. The ratio of PCBs/PBDEs was almost always >1 in harbours, lower near outfalls than in other areas, and mixed for background and near-river samples. Ratios in sediment, whole benthos and *Mytilus* were highest overall in all background salmon and some crab muscle and hepatopancreas samples (>10), and always >1 in background scallops, seal blubber and bottom fish. PCBs therefore appear to be more persistent than PBDEs in higher trophic levels. The notably high ratios in salmon suggest a much stronger persistence of PCBs than PBDEs in the pelagic food chain. The opposite appears to be true for intertidal clams taken from remote areas. Reduced ratios near outfalls are in line with findings from previous research (Johannesssen et al. 2008; Burd et al. 2019), indicating that PBDEs are still being discharged from wastewater outfalls in the southern SoG, whereas historical sediment PCBs are being progressively diluted by wastewater outfall deposition.

Tissue PBDEs and PCBs both increased in a predictable way with sediment values, but otherwise responded differently to geochemical conditions in sediments (Burd et al. 2019), reflecting similar particle affinities of both classes of contaminants but different source concentrations.

Whole benthos tissue PBDEs relative to sediment levels reflect more efficient contaminant uptake from ongoing, fresh organic input from urban waste, as well as rapid biomass turnover (Burd et al. 2013). By contrast, the legacy PCBs are recycling through the ecosystem absorbed to existing,

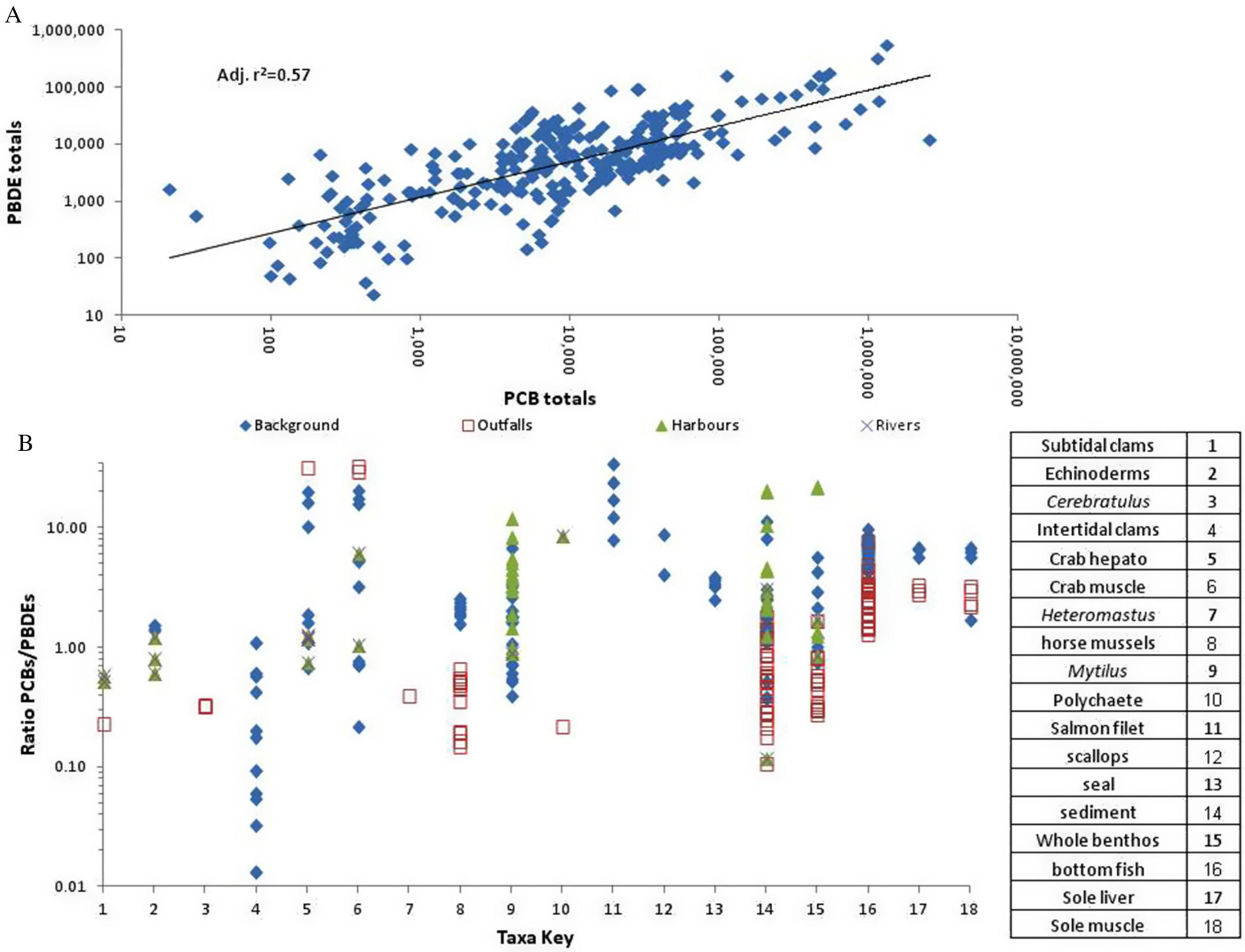

Fig. 7. (A) Log/log comparison of polychlorinated biphenyl (PCB) and PBDE totals in matched sediment and tissue samples from this study and Burd et al. (2019), and (B) ratio of PCBs/PBDEs for different taxa and habitat types. One extremely high value found in VH whole benthos is not shown in this figure (ratio ~1500). Bottom fish includes Midshipman, English sole, and Irish Lord.

refractory or "old" carbon sources, with "new" supplies disconnected from fresh organic food resources, coming instead from fine, silty river runoff or stormwater runoff through industrial harbours.

The uptake rate (tissue dry wt/sediment dry wt) of total PCBs and PBDEs showed a similar range (about 0–100+ times). Tissue/sediment PBDEs and PCBs (rate of uptake) both declined with increasing sediment contaminant levels, suggesting that in extremely contaminated areas, not all sediment contaminants are bio-available. This could be due to adsorbtion of contaminants to black carbon or other refractory particles such as microplastics or engineered nanotubules (Parks et al. 2014). In fact, these types of materials have been shown to significantly reduce bioaccumulation of PCBs in marine polychaetes (Janssen et al. 2010; Beckingham and Ghosh 2017) and are thus useful for remediation of contaminated marine sediments. With PBDEs, bio-dilution (ratio <1) occurred at extreme sediment PBDE concentrations (>10,000 pg/g dry wt) in urban harbours. PCBs did not show bio-dilution, with tissue levels always at least 10× higher than sediment levels. This further supports the finding described above that PCBs (particularly penta-, hexa- and hepta-) are much more bio-accumulative in marine tissues than PBDEs.

Uptake rate of PBDEs decreased with increasing %fines (Burd et al. 2019). In contrast, uptake rate of PCBs declined relative to increasing sediment AVS, less so organic carbon, but not %fines. As discussed above, this is likely related to the more rapid dechlorination of heavier, more toxic, and strongly organic-bound PCB congenors, by anaerobic microbes in reducing sediments.

There is clearly a high initial accumulation of both PBDEs and PCBs from sediment to infaunal whole benthos, despite low tissue lipid content. A profound change to PBDE congener composition occurs at the point of sediment uptake (Burd et al. 2019), a pattern seen in primary consumer zooplankton as well (Frouin et al. 2013). However, tissue PCBs tend to be very similar in composition to surrounding sediments (Kobayashi et al. 2010). Considerable accumulation of dry-weight PBDEs and PCBs both occurred in the higher trophic level organisms, but the lipid-normalized accumulation in these animals relative to direct sediment feeders was very low for PBDEs (ratio close to 1), suggesting no bio-magnification (rather lipid accumulation). In contrast, the PCBs showed modest bio-magnification relative to the background whole infauna in some larger benthic taxa (echinoderms, large polychaetes) and higher magnification in seal blubber (2-3×).

Accumulation patterns for both contaminant groups could be interpreted very differently depending on the relative importance of benthic versus pelagic food chains, since the pelagic feeders typically had the lowest tissue PCB and PBDE levels. Added to this complexity, understanding the PCB and PBDE trophic accumulation patterns will continue to be confounded by the difficulty in determining whole-body burdens and the lack of research on temporal accumulation in larger, long-lived organisms.

Conclusions

PCBs appear to be much more bio-accumulative than PBDEs in marine sediment-dependent fauna and show remarkable persistence in higher trophic levels despite being banned in North America decades ago. There is strong evidence that PCBs continue to leech into urban harbours in southern BC but are slowly being buried by wastewater outfall particulate deposition. The strong lipid association of PCBs in living tissues and clear lack of metabolization at any trophic level makes it very difficult to erradicate them from the marine food chain, particularly in harbours where sediment concentrations remain extreme. This continued input and resulting uptake in sediment-dwelling organisms remains a problem which is clearly keeping PCB levels high throughout the food chain in the more populated regions of the Salish Sea.

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Author contributions

BJB conceived and designed the study. CJL and CM-C performed the experiments/collected the data. BJB analyzed and interpreted the data. CJL contributed resources. BJB drafted or revised the manuscript.

Data availability statement

Data generated or analyzed during this study are provided in full within the published article and its Supplementary Materials. More detailed data generated or analyzed during this study are available from the corresponding author upon reasonable request.

Competing interests

The authors affirm that there are no competing interests in the data collection, analysis, or production of this publication.

Supplementary material

The following Supplementary Material is available with the article through the journal website at doi:10.1139/facets-2021-0032.

Supplementary Material S1

Supplementary Material S2

Supplementary Material S3

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