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SPECIAL ISSUE: INDICATORS OF OCEAN POLLUTION

Effects of Feeding Strategy, Sediment Characteristics, and Chemical Properties on Polychlorinated Biphenyl and Polybrominated Diphenyl Ether Bioaccumulation from Marine Sediments in Two Invertebrates

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Abstract Shellfish and sediment invertebrates have been widely used to assess pollution trends over space and time in coastal environments around the world. However, few studies have compared the bioaccumulation potential of different test species over a range of sediment-contaminant concentrations and profiles. The bioavailability of sediment-related contaminants was evaluated using sediments collected from sites ($n = 12$) throughout the Salish Sea, British Columbia, Canada. Two benthic marine invertebrates—the Baltic clam *Macoma balthica* and the polychaete worm *Neanthes arenaceodentata*—were exposed for 28 days in a controlled environment to these field-collected coastal sediments. The congener-specific uptake of legacy polychlorinated biphenyls (PCBs) and emergent polybrominated diphenyl ethers (PBDEs) was determined using high-resolution gas chromatography/mass spectrometry in sediments and in invertebrates after the experimental exposure. The polychaete *Neanthes* accumulated lower concentrations of PCBs but higher concentrations of PBDEs. The present study indicates that differences in bioaccumulation between these two invertebrates shape the accumulation of PCB and PBDE congeners, reflect differences in feeding strategies, and reveal the physicochemical properties of the contaminants and sediment properties. Because biota–sediment accumulation factor values are often calculated for environmental monitoring or site-specific impact assessments, our results provide insight into potentially confounding factors and the need for caution when selecting indicator species for coastal marine pollution.

Marine environmental biomonitoring programs are typically designed to provide an integrated assessment of contaminants over space and time, at times capturing biochemical, physiological and morphological measures (Tosti and Gallo 2012). Bivalves, such as mussels in the "Mussel Watch" program (Goldberg et al. 1978), and polychaetes (Dean 2008), have been used as indicators for monitoring coastal ocean pollution due to their wide distribution, easy sampling, and ready accumulation of a wide range of contaminants. The use of invertebrates as indicators of environmental quality has been typically assessed by means of conventional life history or physicochemical parameters (Zhou et al. 2008). The bioaccumulation of pollutants by biota is also influenced by the chemical properties of the compounds and the surrounding environmental conditions, features that are not always fully understood or incorporated into study design.

Estuarine and coastal waters receive contaminants from a combination of inputs including local activities and riverine inputs. Persistent organic pollutants (POPs), which include legacy compounds such as polychlorinated

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biphenyls (PCBs) as well recent chemicals of concern such as polybrominated diphenyl ethers (PBDEs), remain significant management issues in Canada and around the world. As a signatory of the Stockholm Convention (2004), Canada has worked to eliminate the use and production of POPs listed under the terms of the treaty. Disposal at Sea (DaS) is one activity that is subject to scrutiny by Environment and Climate Change Canada (ECCC), which ensures that the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, 1972 (London Convention) and its 1996 Protocol (London Protocol) is adhered to through a permitting system under the Canadian Environmental Protection Act (CEPA 2001) and, in particular, the Disposal at Sea Regulations (Porebski and Osborne 1998). As part of its administration of DaS activities, ECCC monitors sediment concentrations for several contaminants of concern at dredging and disposal sites. More recently, the Species at Risk Act (1999) has documented threats to the recovery of listed wildlife species, which in some cases include pollutants in aquatic food webs and those found in adjacent sediments. The subject of bioavailability of sediment-associated contaminants, therefore, remains a question of concern.

Marine-bedded sediments tend to accumulate contaminants released through point-source and nonpoint-source discharges into coastal waters over time and act as a long-term sink of environmental pollutants (CCME 2001). However, contaminants found in sediments can also be made available to sediment-dwelling or -dependent biota, thereby serving as a potential source of contaminants to adjacent food webs. The bioavailability and bioaccumulation of contaminants in an aquatic environment is mainly dependent on the partitioning behaviour or binding strength of the contaminant to the sediment (Voie et al. 2002). Contaminants partition between aqueous (porewater, overlying water) and solid phases (sediment, suspended particulate matter, and biota) (Luoma 1983). Generally, only dissolved fractions of lipophilic contaminants in the porewater and in the water overlying contaminated sediments are considered to be bioavailable to aquatic organisms irrespective of their mode of life (Di Toro et al. 1991). However, with aqueous-exposure pathways, the bioavailability of organic contaminants decreases with increasing octanol/water partitioning coefficient (K_{ow}) due to the hydrophobic properties of the chemical. Ingestion of sediment particles then becomes the major route for the uptake of lipophilic contaminants (Ruus et al. 2005), and that uptake from different sources is additive (Landrum and Robbins 1990).

Recent studies have characterized spatial and temporal variation in the sediment concentrations of PCBs and PBDEs in the Salish Sea in southern British Columbia, Canada (Grant et al. 2011; Johannessen et al. 2008). PCBs were banned in Canada in 1977, but their legacy persists in a number of environmental compartments. Flame retardants, notably PBDEs, represent another threat to the marine environment. Although some PBDE formulations have been banned or are no longer used in Canada, they are persistent and are transported atmospherically from areas that continue to use them. A prolonged period of exponential increases in PBDEs in biota in the Northeast Pacific region has been observed (Rayne et al. 2003, Ross et al. 2013). Risks to endangered southern resident killer whales (Ross 2006) resulting from the uptake of sediment-associated PCBs by marine food webs were recently evaluated using a combination of measurements and food web-modelling tools (Alava et al. 2012; Lachmuth et al. 2010; Ross 2010).

To strengthen the utility of using marine invertebrates as indicators of habitat and sediment quality, we performed a study of PCB and PBDE uptake from sediment samples collected from sites in the Salish Sea. A standard 28-day laboratory bioaccumulation procedure was used to derive biota-sediment accumulation factors (BSAFs) which represent a ratio of the concentration of a given contaminant in tissue relative to that in the sediment (Nendza 2002). Two marine benthic organisms currently used in sediment monitoring programs were selected for study: the Baltic clam (*Macoma balthica*), a facultative deposit-feeding bivalve living in the sediment, and the polychaete (*Neanthes arenaceodentata)*, a deposit feeder living in the sediment. *Macoma* burrows below the sediment surface and feeds on particles of small size, low specific gravity, and high organic content (Hylleberg and Gallucci 1975). This clam has been determined to be a facultative feeder, switching between suspension-feeding and deposit-feeding modes depending on the food quality and quantity in the water phase (Lin and Hines 1994). The nereid polychaete *Neanthes* ingests large quantities of sediment/food relative to its body weight and is often used as a marine test organism for sediment-quality assessments. This species is a widely distributed annelid found in shallow marine and estuarine environments throughout the world. It deposit-feeds on sediment particles $\leq$ 70-µm diameter with a preference for sediment particles of approximately 12 µm (Bridges and Farrar 1997). Polychaetes represent 30–75% of benthic macro-invertebrates and serve as food for higher trophic organisms.

Our objectives were to characterise the presence and availability of PCB and PBDE congeners in sediments from coastal British Columbia (Canada) in order to (1) explore the influence of contaminant and sediment properties on bioavailability to two marine benthic invertebrates used routinely in monitoring efforts; (2) characterise congener-specific uptake profiles for PCBs and PBDEs by these indicator organisms; and (3) evaluate the influence of normalization procedures for organic carbon (OC) and lipid on results.

Materials and Methods

Organisms

The two benthic invertebrate species selected for sediment bioavailability evaluation are recommended in the Inland Testing Manual [United States Environmental Protection Agency/United States Army Corps of Engineers (USEPA/USACE) 1998] and the Ocean Testing Manual (USEPA/USACE 1991) and used routinely in the North Atlantic Region.

In the first sediment-bioavailability evaluation, the test organisms consisted of Baltic clams (*Macoma)* collected by Harris Industrial Testing Services at Walton, Nova Scotia, Canada. They were held at 15 ± 2 °C in the collection sediment with overlying seawater until used for testing 5 days later. No mortality was observed at the start of the test.

In the second sediment-bioavailability evaluation, the test organisms were cultured juvenile marine polychaete (*Neanthes)* purchased from Aquatic Toxicology Support. The organisms were placed in pails of aerated seawater and adjusted to and held at 20 ± 2 °C until used for testing 2 days later. No mortality was observed on holding.

Sediment Sampling and Characterization

Sediment samples were collected from 12 sites in or near 3 marine harbours in coastal British Columbia, Canada (Fig. 1). Samples were collected using a Petit Ponar grab sampler deployed from a small vessel between September 28 and December 8, 2011. Sample penetration was typically 7–12 cm with the entire sediment sample retrieved used for this study. Each of the sediment samples was homogenized before use in each assay using a drill with a stirring attachment. Samples were stored at −20 °C, shipped frozen by ground transport to the evaluation facility in Nova Scotia, and thawed before bioavailability evaluation.

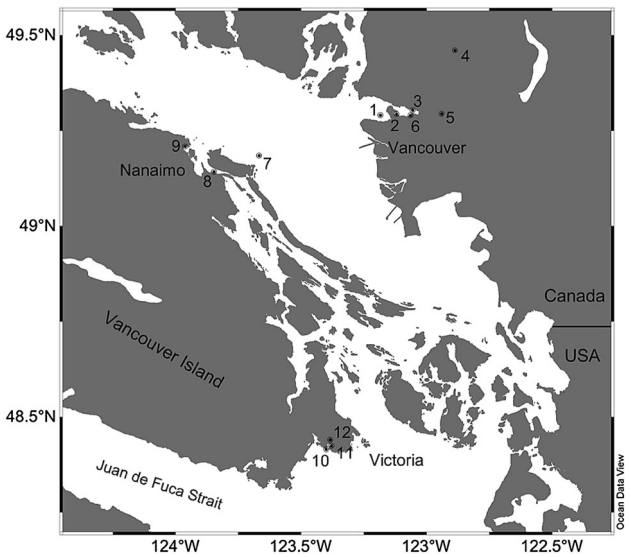

Fig. 1 Surficial sediment samples for contaminant analysis were collected at stations in coastal British Columbia (Canada) near Vancouver (VA), Nanaimo (NA), and Victoria (VI) harbours

A sediment sample was collected from the same location as the Baltic clams at Walton Wharf, Nova Scotia, Canada. This sediment was stored at 4 °C until used for testing. This was used as the control sediment for both tests. A portion of this sediment was also spiked with PCB 153 (2,2′,4,4′,5,5′-hexachlorobiphenyl). A 500-mg/L stock of PCB 153 was prepared in isooctane. Four milliliters of this solution were added to 20 kg of sediment and thoroughly mixed to five a 0.1 mg/kg PCB 153 in sediment.

Bioaccumulation-Evaluation Setup

This method is based on ASTM E1688-10 the Standard Guide for Determination of the Bioaccumulation of Sediment-Associated Contaminants by Benthic Invertebrates (ASTM International 2010). Both bioaccumulation evaluations took place at the Atlantic laboratory for Environmental Testing (ALET; Moncton, New-Brunswick, Canada).

One day before placing the organisms into the sediments, the sediments were homogenized and added to the test chambers to allow settling. The aquaria and bottles were precleaned and rinsed with seawater three times before use. Any indigenous organisms and large twigs or rocks were removed. The tests were performed in 8-L glass aquaria (*Macoma*) or 2-L glass bottles (*Neanthes*). To each aquarium or bottle, 5 cm of sediment was added, then carefully filled with 5 µm filtered seawater with a salinity of 28–32 ppt.

There were 3 (*Macoma*) and 4 (*Neanthes*) replicates for each treatment. The treatments consisted of 12 test sediments, control sediment (from the *Macoma* test collection site used for both *Neanthes* and *Macoma*), and a positive control (the control sediment spiked with PCB 153). The aquaria or jars were placed in a controlled bath at 20 ± 1 °C using a randomized block design. The overlying water was aerated through the test with an airline fitted with a glass pipette tip. The tip was secured so the airline tubing was not in the water. Fluorescent lighting of 100 to 500 lx with a 16 to 8-h light-to-dark phase, including a 30-min transition phase, was used.

Twenty-Eight-Day Organism Exposure

A water sample was taken from 1 replicate of each treatment to measure for ammonia, salinity, dissolved oxygen, temperature, and pH at the start of the evaluation. For the

first laboratory exposure, the clams were removed from their holding sediment by filtering through a 60-µm sieve. The clams were rinsed in seawater several times to remove collection-site sediment and sorted into plastic trays. The clams were held under seawater to minimize exposure to air. Twenty-two clams were added to each replicate with a total of 66 clams per treatment. For a random sample, the shell size and weight of 20 clams were measured to determine average size of the population at the start of the test (19.8 ± 1.8 mm; 3–10 years old Cardoso et al. 2006). The next day, each tank was checked to ensure that the clams had buried, and any that had not were replaced. For the second laboratory exposure, 35 polychaetes were added to each replicate, which allows for a total of 140 worms/treatment. The average weight of the worms at the start of the test was 0.01 g.

Each day the temperature in one replicate of each treatment was measured. Daily observations were made of animals at surface, mortality, and aeration. Three times a week (Monday, Wednesday, and Friday), a sample of the overlying water was measured for: dissolved oxygen, pH, and salinity. The overlying water was renewed twice a week on Mondays and Fridays; approximately 80% was siphoned and replaced with fresh seawater. Before replacing the water, 200 ml (*Macoma*) or 75 ml (*Neanthes*) of new sediment was added to the surface at least once a week as per the recommendation of the test method, and then refilled with seawater, in order o reduce the depletion of target analytes during the exposure duration (28 days). To provide nutrition for the animals, six (*Macoma*) or five (*Neanthes*) additions of sediment were made throughout the experiment. After 28 days of exposure, each treatment was sieved through a 100-µm (*Macoma*) or 500-µm (*Neanthes*) sieve, and the clams or worms were removed.

For the first exposure test, the clams were rinsed several times in seawater and then placed into clean prerinsed mason jars with approximately 1L of fresh seawater and aerated overnight to permit gut purging of sediment. After the 24-h gut purging, the animals were removed from the seawater. The length of the shell and weight of each clam was measured. The clams were opened and the tissue removed. The tissue was transferred into a tared glass vial, and the weight of the tissue was recorded. All tissues from the three replicate tanks were combined. Throughout the experiment every effort was made to ensure there was no cross contamination between treatments by using only new or cleaned (solvent-rinsed) glass or metal apparatus. No plastic devices were used. Tissue samples were frozen at −20 °C after collection and shipped to the former Laboratory of Expertise for Aquatic Chemical Analysis (LEACA; Fisheries, Oceans & the Canadian Coast Guard, Sidney, BC) on dry ice.

For the second exposure test, the worms were rinsed several times in seawater and placed into tared vials. The total weights of the worms were recorded. A depuration period was planned; however, with the difficulties in recovering the worms and the stress this caused on the animals, it was decided this step would not happen due to potential of loss of the tissues. All tissues from the four replicate tanks were combined. Every attempt was made to ensure there was no cross-contamination between treatments. No plastic devices were used; all of the equipment was metal or glass and appropriately cleaned. The tissue samples were frozen and shipped to LEACA on dry ice.

Tissue Analysis

A total of 28 biota samples (14 *Macoma* and 14 *Neanthes*) exposed to sediments from the Salish Sea were submitted to LEACA. Samples were analysed for congener-specific PCBs ($n = 182$ congeners) and PBDEs ($n = 66$ congeners). A typical analytical batch consisted of 9 and 10 samples, respectively; one replicate; two laboratory procedural blanks; and one certified reference material (CIL Herring EDF-2524e). Detailed analytical methods are described elsewhere (Ikonomou et al. 2001). One sample (*Neanthes* VA-03) was lost during the extraction.

Tissues samples were analysed according to USEPA protocols 1668 and 1614, and analytes were identified only when high-resolution gas chromatography/mass spectrometry (HRGC/HRMS) data satisfied all quality assurance/quality control (QA/QC) criteria described elsewhere (Ikonomou et al. 2001). Briefly, samples were spiked with ^{13}C-labelled surrogate standards and then ground with anhydrous sodium sulphate. Samples were transferred to a Soxhlet thimble; surrogate standard was added; and samples were refluxed for 16 h with dichloromethane (DCM). The extract was eluted through a gel permeation column with 1:1 DCM and hexane. The extract was applied to a partially deactivated Fluorisil column and eluted with hexane followed by 15:85 DCM and hexane. Eluates were then combined and eluted with 1:1 DCM and hexane and each fraction concentrated. Samples were analyzed using an Ultima HRMS (Micromass, Manchester, UK) equipped with a Hewlett Packard 5890 gas chromatographer (Agilent, Wilmington, Delaware, USA) and a DB-5 Durabond capillary column (60 m × 0.25 mm, 0.10-µm film). Percent lipid in samples was determined using the gravimetric lipid determination by weight-of-extract method with dichloromethane.

Due to the small amount of available tissue, it was only possible to obtain lipid and moisture determinations for 14 biota samples (13 *Macoma* and only 1 *Neanthes*). For

Neanthes, the lipid percentage obtained for a single sample was applied to other samples (a minimal variation was assumed between samples).

Sediment Analysis

A total of 12 sediment samples from Salish Sea were submitted to LEACA. Samples were analysed for congener-specific PCBs ($n = 182$ congeners) and PBDEs ($n = 66$ congeners). A typical analytical batch consisted of 8 samples, 1 replicate, 2 laboratory procedural blanks, and 1 certified reference material (UMEA B sediment). In each class of analytes, the 10th batch included re-extractions along with two procedural blanks.

Sediment-sample analyses were performed according to USEPA protocols 1668 and 1614 (USEPA 2003, 2007), and analytes were identified only when HRGC/HRMS data satisfied all QA/QC criteria described elsewhere (Ikonomou et al. 2001). Samples were spiked with ^{13}C-labelled surrogate standards and then ground with anhydrous sodium sulphate. Samples were transferred to a Soxhlet thimble; surrogate standard was added; and samples were refluxed for 16 h with DCM. The extract was eluted through a gel permeation column with 1:1 DCM and hexane. The extract was applied to a partially deactivated Fluorisil column and eluted with hexane followed by 15:85 DCM and hexane. Eluates were then combined and eluted with 1:1 DCM and hexane and each fraction concentrated. Samples were analyzed using an Ultima HRMS (Micromass) equipped with a Hewlett Packard 5890 GC (Agilent) and a DB-5 Durabond capillary column (60 m × 0.25 mm, 0.10-μm film). Sediment-particulate OC was determined with a Leemans Elemental analyzer after removal of carbonates by acid-fuming with HCl for 24 h in a closed container.

QA/QC Measures

LEACA analyses were performed using the USEPA protocols 1668 and 1614 (USEPA 2003, 2007). Each batch of nine samples included one procedural blank. Procedural blanks were run in each of analytical batches. Procedural blank values were subtracted from the analytical values of all of the samples. Analytes were identified only when the HRGC/HRMS data satisfied all criteria reported elsewhere (Ikonomou et al. 2001).

To reduce the influence of the nondetected congeners on the total analyte concentrations, the following substitutions were applied (see Table 1):(1) congeners that were detected in <70% of the samples were not included in the calculations; (2) where congeners were detected in ≥70% of the samples, a limit of detection subtraction was applied. No substitutions were made for PCB and PBDE congeners detected in all samples at all times.

Data-Analysis Methods

Both congener-specific and total concentrations (sum of all detected congeners) for PCBs and PBDEs are presented. BSAFs, which is a ratio of the concentration of a contaminant in tissue relative to that in the sediment, were calculated for each species-contaminant-sediment combination. PCB and PBDE concentrations in tissues and sediment were normalized to lipid (lipid-normalization was performed based on wet weight) and TOC content, respectively. However, BSAFs were also calculated based on wet- (biota) and dry-weight (sediment) concentrations (not normalized) to assess the influence of normalization procedures on outcomes.

Results

Survival, Weight, and Lipid

Survival of *Macoma* was >80% in all sediment tests except for the positive control (Table S1). There was only 6% survival in the positive control; either the concentration of PCB-153 was too high, or there was an effect of the associated solvent. All clams buried themselves on the first day, with six being replaced after 24 h. Aside from the positive control, survival rates were high, and there was no difference in size (Table S2). Water-quality parameters were satisfactory throughout the experiment, and the ammonia levels were low at the start of the test (Table S3). Approximately 11–16 g of wet-tissue weight was obtained for each of the treatments. Survival was not affected at the highest concentrations of ΣPCBs or ΣPBDEs bioaccumulated by Baltic clams (Fig. 2a, b).

Neanthes survival varied across the different sediment tests ranging from 31 to 87% (Table S4). There was low survival in samples VA-01, VA-06, NA-08, and VI-10. Sediment samples had some gravel for both VA-06 and NA-08. For the positive control, there was high survival (78%), which compares dramatically with the very low survival for the clams in the same treatment. Water-quality parameters were satisfactory throughout the experiment, and the ammonia levels were within an accepted range at the start of the test (Table S5). Only approximately 1–2 g of wet weight was obtained for each of the treatments. We observed a negative correlation between PBDEs and survival of *Neanthes*, but this was not true for PCBs (PBDEs $R^2 = 0.58$, $p < 0.05$, Fig. 2c, d).

In both *Macoma* and *Neanthes*, weight was similar between exposed and control organisms after the 28-day

Table 1 PCB and PBDE congeners were detected at varying frequencies in sediment, *Macoma*, and *Neanthes*

Congeners detected	Sediment		*Macoma*		*Neanthes*	
	PCBs (%)	PBDEs (%)	PCBs (%)	PBDEs (%)	PCBs (%)	PBDEs (%)
0%	13 (7.1)	15 (22.7)	22 (12.1)	23 (34.8)	42 (23.1)	41 (62.1)
0 ≤ 70%	36 (19.8)	20 (30.3)	42 (23.1)	25 (37.9)	95 (52.2)	21 (31.8)
70 ≤ 100%	47 (25.8)	18 (27.3)	86 (47.3)	17 (25.8)	37 (20.3)	3 (4.6)
100%	86 (47.3)	13 (19.7)	32 (17.5)	1 (1.5)	8 (4.4)	1 (1.5)
Total no. of congeners analysed	182	66	182	66	182	66

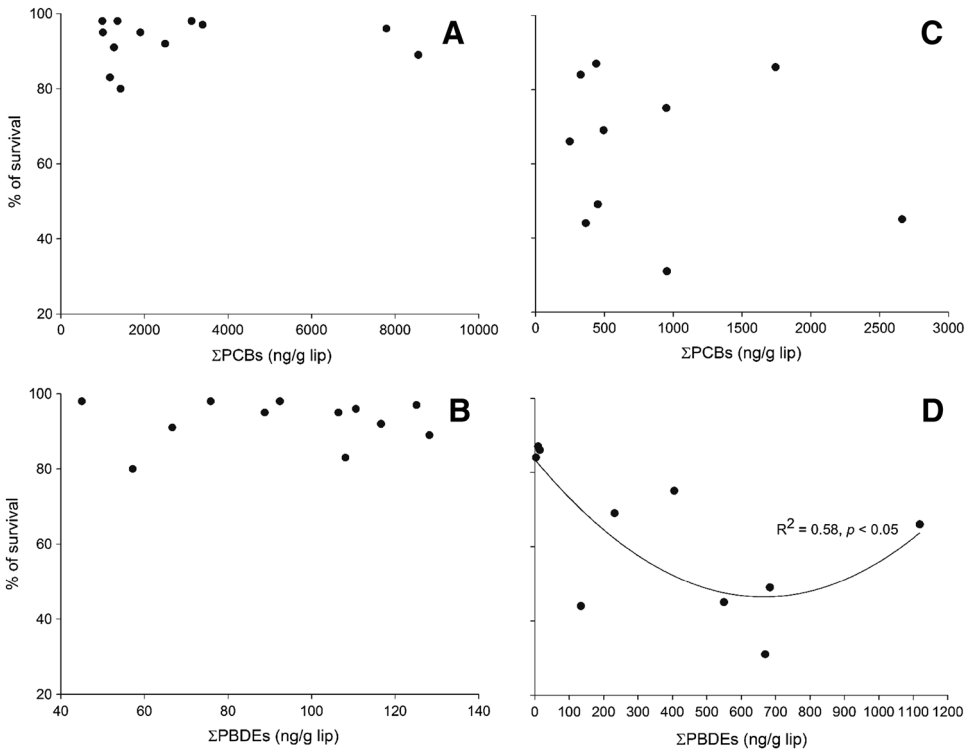

Fig. 2 Survival of *M. baltica* in each experiment against their ΣPCB (**a**) or ΣPBDE (**b**) tissue concentrations. Survival of *N. arenaceodentata* in each experiment against their ΣPCB (**c**) or ΣPBDE (**d**) tissue concentrations

exposure. The lipid contents of *Macoma* were in the range of 0.7–0.94% (Table 2). The lipid content of *Neanthes* was analyzed for only one sample due to the low-weight samples of *Neanthes* (tissue weight range = 0.58–2.06 g; tissue average weight = 1.5 ± 0.4 g) and was 0.76% (Table 2).

Bioaccumulation of PCBs and PBDEs

PCB residues in all sediment tests were highest in *Macoma* and lowest in *Neanthes* whether or not concentrations were normalized (TOC basis for sediment and lipid basis for organisms) (Fig. 3a, b). PBDE residues, in contrast, were highest in *Neanthes* and lowest in *Macoma* for most sediment tests whether or not concentrations were normalized (TOC basis for sediment and lipid basis for organisms) (Fig. 3c, d). Tissue and sediment concentrations were correlated for PCBs but not for PBDEs. Interestingly, PBDEs increased in *Macoma* until a threshold was attained, and the organisms did not continue to accumulate the chemical.

The concentrations of PCBs and PBDEs in all samples are listed in Table 2, and the overall concentration comparisons are shown in Fig. 4. The PBDE concentrations were lower—i.e., Student *t* test: sediments = 2384 ± 1009 ng/g PCB TOC basis and 196 ± 57 ng/g PBDE TOC basis ($p = 0.04$); *Macoma* = 3029 ± 807 ng/g PCB lipid basis and 92 ± 8 ng/g PBDE lipid basis ($p = 0.001$); *Neanthes* = 1116 ± 336 ng/g PCB lipid basis and 348 ± 112 ng/g PBDE lipid basis ($p = 0.04$)—than those of PCBs.

CB-153 was the most dominant PCB congener in both species (Figure S1; *Neanthes* 32% vs. *Macoma* 22%) except in two samples (VA-02 and VA-06). The congener pattern in

Table 2 Sample characteristics and concentrations of total PCBs and PBDEs in surficial sediments and two organisms coincubated with these sediments for 28 days (*M. balthica* and *N. arenaceodentata*)

Sediment sample ID	Sediments			*Macoma balthica*			*Neanthes arenaceodentata*		
	TOC (%)	PCBs (ng/g dw)	PBDEs (ng/g dw)	Lipid (%)	PCBs (ng/g ww)	PBDEs (ng/g ww)	Lipid (%)	PCBs (ng/g ww)	PBDEs (ng/g ww)
VA-01	1.16	10.17	1.49	0.81	9.47	0.87	0.76	2.77	1.02
VA-02	2.16	133.24	4.97	0.88	68.49	0.97	0.76	13.24	0.11
VA-03	1.01	0.51	0.54	0.81	11.52	0.46	0.76	N/A	N/A
VA-04	3.17	3.06	0.46	0.89	8.81	0.67	0.76	2.49	0.03
VA-05	2.00	21.20	5.42	0.91	22.83	1.07	0.76	3.35	0.07
VA-06	3.03	130.19	9.87	0.91	30.88	1.14	0.76	20.23	4.18
NA-07	2.14	8.01	1.63	0.94	11.91	0.62	0.76	1.89	8.50
NA-08	17.22	118.05	1.26	0.88	12.00	0.40	0.76	7.25	5.09
NA-09	0.81	3.10	0.34	0.73	7.37	0.65	0.76	3.75	1.76
VI-10	1.26	5.94	1.40	0.79	15.01	0.84	0.76	3.43	5.19
VI-11	2.65	310.44	11.66	0.87	74.25	1.11	0.76	27.68	0.04
VI-12	5.94	143.93	38.74	0.70	20.81	0.62	0.76	7.22	3.08
Control		0.41	0.33	0.94	15.05	0.51	0.76	0.56	0.81
Positive control		189.69	0.35	0.84	171.3	0.16	0.76	234.7	1.01

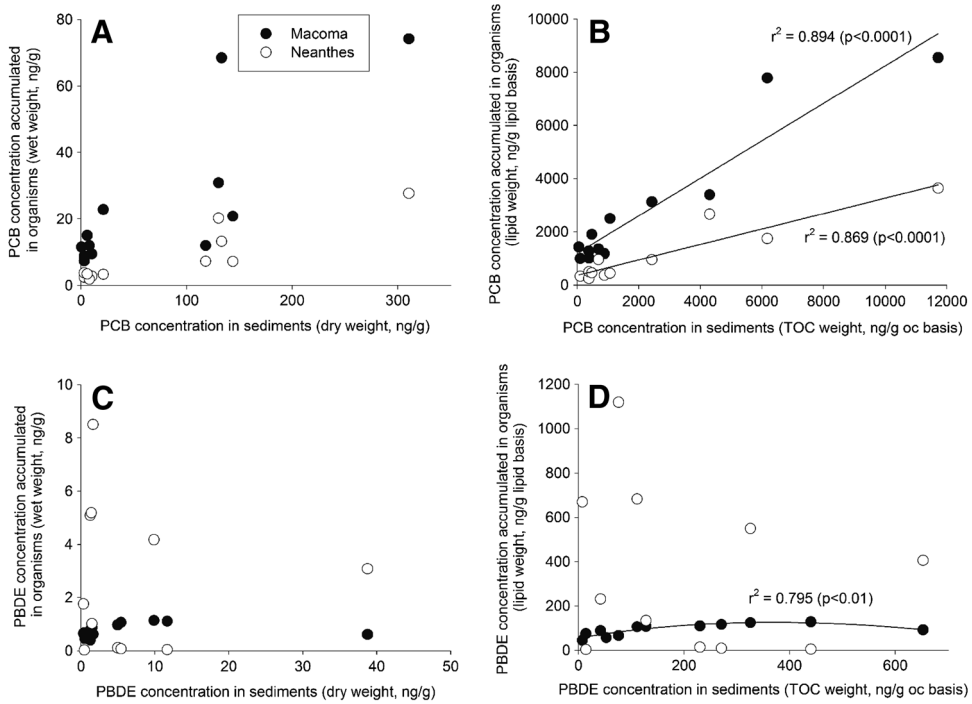

Fig. 3 Accumulated concentrations in *Macoma* (*closed circle*) and *Neanthes* (*open circle*) after 28-day exposure to sediments with different levels of PCBs and PBDEs. Concentrations were expressed as not normalized (**a** PCB; **c** PBDE) or TOC/lipid normalized (**b** PCB; **d** PBDE)

both invertebrates was similar to the pattern observed in the sediments. BDE-47 dominated the PBDE profile in both species (Figure S2; *Neanthes* 44% vs. *Macoma* 57%) except in three *Neanthes* samples (VA-01, VA-05, and VI-12) followed by congeners BDE-99 and BDE-100.

BSAFs were calculated as the ratio of tissue (lipid weight-corrected) to sediment concentrations (TOC-corrected) ΣPCBs and ΣPBDEs (Tables S6 and Table S7). The BSAFs calculated with dry-weight sediment and wet-weight biota concentrations are also presented in those tables. Some PBDE congeners in *Neanthes* samples NA-07 and NA-08 were excluded as outliers (Table S6). In those cases, BSAFs were very high due to the low concentrations and many nondetects in the *Neanthes*.

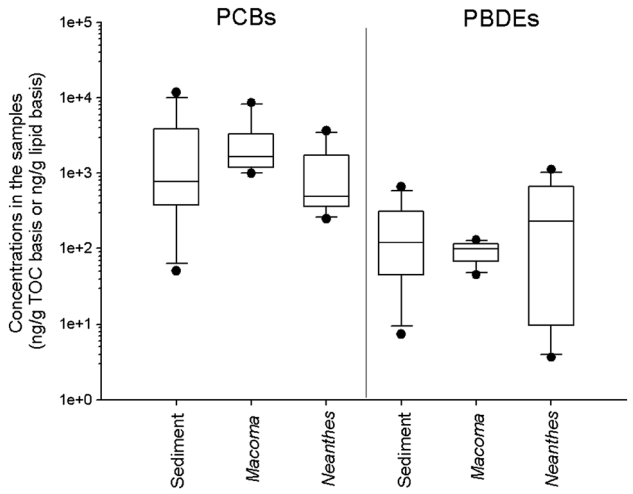

Fig. 4 *Box-plots* of ΣPCBs and sum ΣPBDEs for all sediment at all locations and both species exposed to all sediments on TOC-weight basis (sediment) and lipid-weight basis (biota). Whiskers indicate 5th to 95th percentile; box indicates 25th to 75th percentile; *line* shows the median; *dots* represent outliers

The influence of physicochemical properties of the two contaminant classes in shaping uptake and accumulation is evident when plotting BSAFs against log K_{ow} values for the individual PCB and PBDE congeners (Figs. 5, 6). BSAFs of PCBs versus log K_{ow} in *Neanthes* showed a peak at a log K_{ow} of 6. The BSAFs for PCBs (range 0.01–30) were similar to those for PBDEs (range 0.01–30).

PCB and PBDE BSAFs differed between *Macoma* and *Neanthes* (Fig. 7). PCBs exhibited higher bioaccumulation in *Macoma* than *Neanthes*. For PBDEs, BSAF differences between species were less obvious due to the low number of detected congeners in *Neanthes*.

Discussion

Benthic invertebrates have been widely used in the assessment of coastal contamination around the world, but different species-specific factors (choice of test species, feeding ecology, age/size, growth rate), sampling protocols, and analytical instrumentation severely constrain the ability to compare results across studies and regions. In the case of PCBs and PBDEs, where both contaminant classes have as many as 209 theoretically possibly congeners in environmental mixtures, interpretation and reporting is subject to considerable variation among studies. Budget-conscious monitoring programs may opt for total concentrations for these two contaminants of concern. However, congener-specific concentrations using high-resolution instrumentation offer valuable opportunities to inform source, transport, fate, and biological uptake functions through the use of fingerprinting techniques such as multivariate statistical models (Grant et al. 2011; Ross et al. 2004). Characterizing the congener-specific uptake of PCB and PBDE congeners by invertebrates using real world sediment samples (mixtures) here can enable guidance on the design of marine pollution indicators.

Sediment samples were taken near or in urban harbours, which represent hot spots for both PCBs and PBDEs (Grant et al. 2011), whereas PBDEs are also found at high concentrations near municipal outfalls (DeBruyn et al. 2009). Sediment concentrations were in the range of PCB and PBDE concentrations from the same area (Grant et al. 2011). Hot spots observed in Victoria and Vancouver harbours are influenced by local waste discharges, thus reflecting direct input pathways by way of urban and industrial runoff (Johannessen et al. 2008). The PCB and PBDE concentrations in the Victoria area (VI-11 and VI-12) were higher than those in the Vancouver area.

This study was designed to look at biological uptake of PCB and PBDE congeners and not biological end points, but basic aspects of biological condition were noted. Higher relative mortality of *Neanthes* versus *Macoma* may be due to higher concentrations of ΣPBDEs bioaccumulated by *Neanthes*. Interestingly, the low survival observed in *Neanthes* (31%), occurred in the sample with the higher TOC concentration (17.22%). A possible explanation is that this sample generated sufficiently high sulphide in porewater during the test to affect the survival of *Neanthes* (Vismann 1990). Dillon et al. (1993) showed that survival of juvenile worms may be adversely affected if test conditions involve exposure to ≥0.7 mg/L unionized ammonia or ≥5 mg/L hydrogen sulfide. The same investigators have also observed that the grain size had no significant effect, whereas the number of worms placed in each exposure vessel was critical. Polycyclic aromatic hydrocarbons (PAHs) are among the contaminants of concern in industrial harbours (Yunker et al. 2011) and near marine municipal-sewage discharge (Chapman et al. 1996). Chapman et al. (1996) observed that some of the gravel found in sediments actually consisted of coal and coke. In two sediment samples associated with low survival of *Neanthes*, gravel was present. It is therefore, possible that other parameters (e.g., PAHs) affected the survival of *Neanthes*. The high mortality observed in the *Macoma* positive control test may have initially triggered PCB-153-associated toxicity (or its solvent) but subsequently exacerbated by the release of ammonia by dead clams.

Our bioaccumulation findings are consistent with those of others who have documented the general nature of sediments as a source of benthic-invertebrate contamination (Foster et al. 1987; Janssen et al. 2010; Josefsson 2011). In all of our tests, PCBs and PBDEs were readily accumulated by both species, but differences in bioaccumulation patterns between species were evident. The

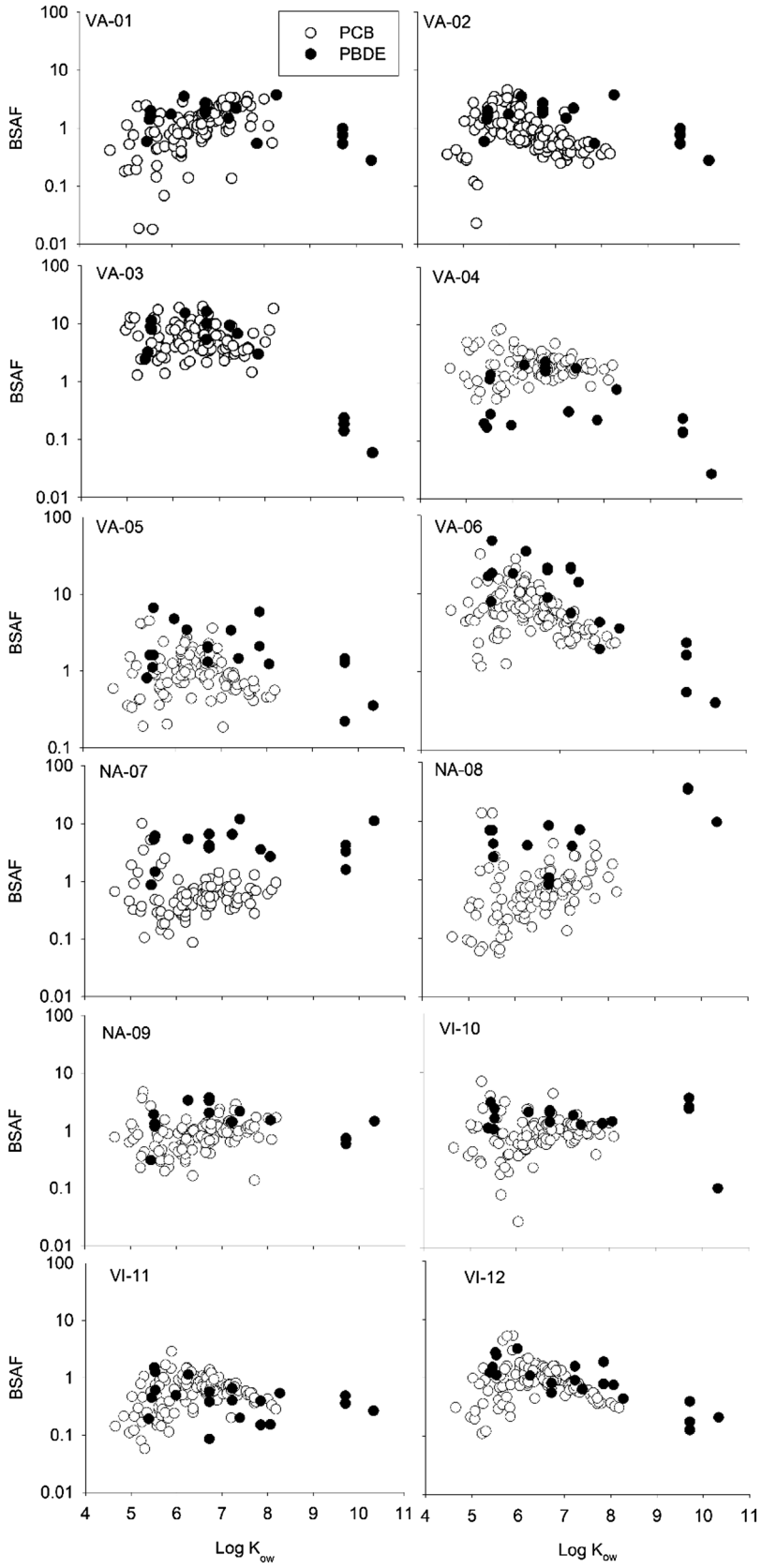

Fig. 5 BSAFs normalized to lipid and OC in *Macoma* for PCBs (*open circle*) and PBDEs (*closed circle*) against log K_{ow}. The log K_{ow} values of the congeners were taken from selected literature (from Braekevelt et al. 2003 for PBDEs and from Hawker and Connell 1988 for PCBs)

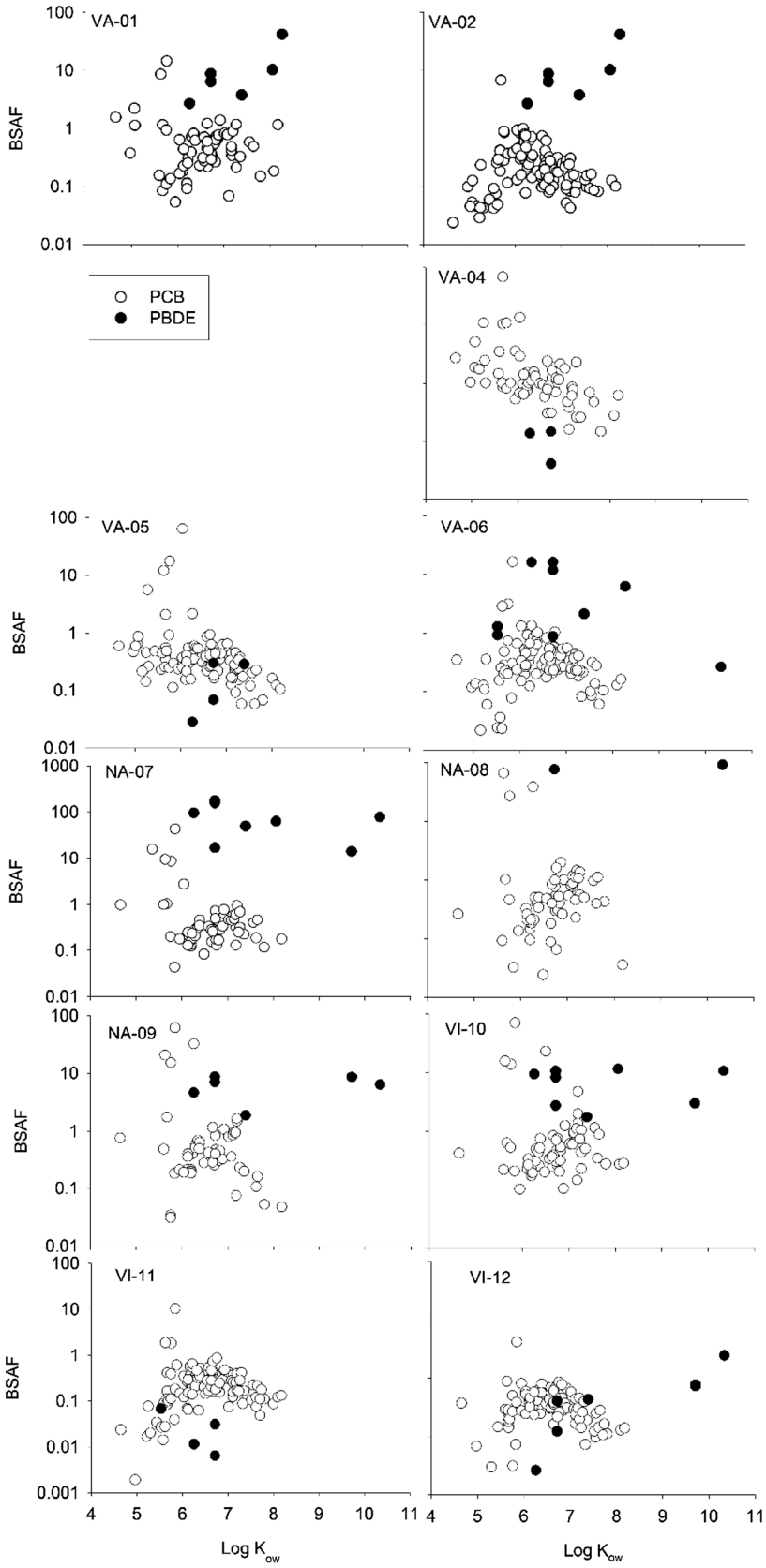

Fig. 6 BSAFs normalized to lipid and OC in *Neanthes* for PCBs (*open circle*) and PBDEs (*closed circle*) against log K_{ow}. The log K_{ow} values of the investigated POPs congeners were taken from selected literatures (from Braekevelt et al. 2003) for PBDEs and from Hawker and Connell 1988 for PCBs)

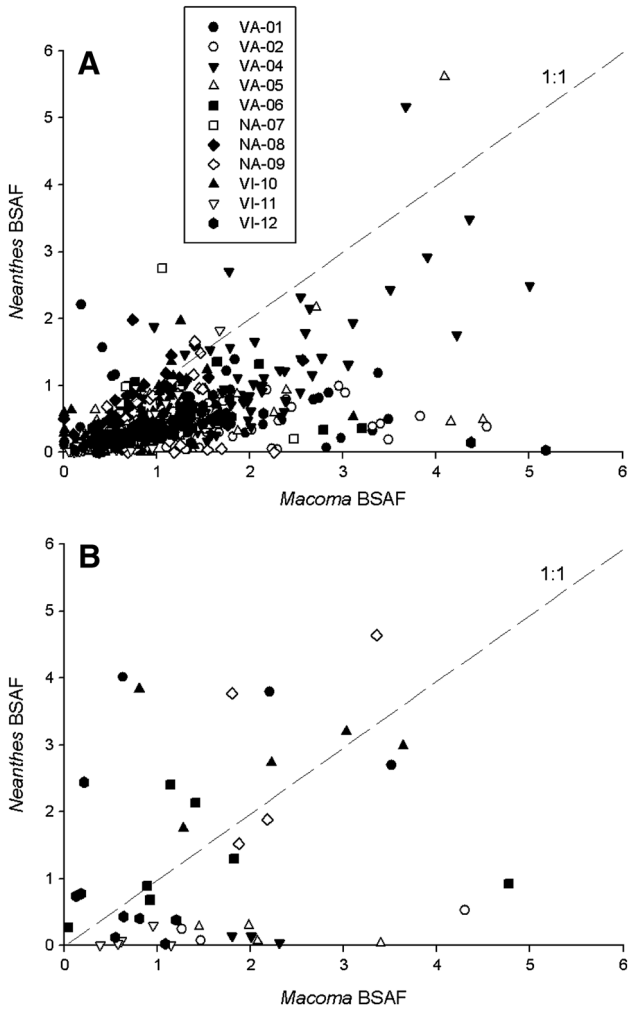

Fig. 7 Comparison of BSAF (ratio of normalized to lipid and OC) for **a** PCBs and **b** PBDEs for *Macoma* and *Neanthes* derived from 28-day laboratory exposure

polychaete *Neanthes* acquires food by bulk deposit feeding; this invertebrate accumulated the lowest concentration of PCBs but the highest concentration of PBDEs. PCB concentrations in lipids were approximately three times lower, whereas PBDE concentrations in lipids were approximately 5 times higher in this deposit feeder compared with the facultative deposit-feeding clam *Macoma*. Interestingly, the biodynamic model developed by Janssen et al. (2011) suggested that *Neanthes* accumulates approximately 20 times more PCBs in its lipids than *Macoma*. In our study, contaminants associated with retained sediment in the gut of *Neanthes* may have led to an overestimate of the concentration in tissue.

Limited correlations observed between sediment- and biota-contaminant concentrations in our study may be in part due to the differences in sediment conditions (e.g., granulometry, OC, and black carbon) from the different sites from which the samples were collected. Age and sex may also affect uptake. Lotufo et al. (2000) showed that sex had a large influence on contaminant uptake kinetics in *N. arenaceodentata* with mature males having a more efficient uptake clearance rate and faster elimination rate than mature females. Differential bioaccumulation between the two invertebrates may be attributed to differences in functional feeding groups and in feeding rates. Exposure and uptake can vary widely among filter versus deposit feeders based on their relative interaction with the supra-sediment water and the extent to which they consume sediment. The high absorption efficiencies by polychaetes is due to the strong surfactancy in deposit-feeder guts (Ahrens et al. 2001). In addition, the presence of a barrier (e.g., shell) can limit the organism's passive exposure to contaminated environments.

The importance of bioturbation by biorrigator/gallery-diffusor species on the remobilization of POPs from sediment has been described previously by Josefsson (2011). The presence of tubes created by *Neanthes* could increase the sediment-water interface area and facilitate the movement of colloids with associated POPs (Bosworth and Thibodeaux 1990). Because more hydrophobic POPs are more resistant to desorption from the sediment particles and the distribution to colloids (and particles) generally increases with increased hydrophobicity, *Neanthes* would be more exposed to more hydrophobic POPs. In contrast, the bivalve *Macoma* has a limited effect on POP flux between water and sediments. This invertebrate lives few centimetres beneath the water-sediment interface, extending its two siphons to the surface where it can switch between surface deposit- and suspension-feeding. The clams can mix particles from the surface into the sediments when they feed and move around, thus causing particles to fall into the space created around their shells. In addition, species-specific differences in the uptake and elimination of certain congeners could also be occurring. Magnusson et al. (2006) showed that the interspecific variations in bioaccumulation of PCB congeners in marine benthic infauna did not correlate with differences in feeding strategies but rather may be caused by differences in biotransformation and in the age and size of the analysed specimens.

Our results show that PCB accumulation in both *Macoma* and *Neanthes* is related to concentrations in sediment. However, the variation in accumulated levels between both species is important. Although the driver(s) of this variation remains unclear, one factor that might have played a role is the time of year in which the different experiments were performed. Seasonal variation in PCB content has been observed in mussels, with highest levels found in February, because this species is a spring spawner (Hummel et al. 1990). *Macoma* is also a spring spawner (Philippart et al. 2003). The 28-day exposure

experiment started in February, so it is reasonable to assume that this species will concentrate highest levels during this period of time. However, the seasonal variability in uptake by mussels in situ involves many factors (e.g., temperature), which may vary during laboratory testing of bioaccumulation. Lipid-normalization of tissue PCB and PBDE concentrations, therefore, reduces only part of the variation between species. Differences observed between PCB and PBDE congener BSAF values between *Neanthes* and *Macoma* may be most likely due to differences in feeding behaviour. However, it is also possible than the presence of sediment in *Neanthes* affected the comparison between the two species. The *Macoma* were depurated, whereas the *Neanthes* were not. For example, depuration experiments showed that the larger part of the BDE209 content in mussels was associated with ingested particles (Booij et al. 2002). *Macoma* showed a PCB distribution that was dominated by less-chlorinated congeners compared with *Neanthes* (Figure S1). Aside from the shift to a lower molecular-weight distribution, *Macoma* did not appear to be selectively depleted in the concentrations of individual congeners. This indicates a lack of metabolism of PCB congeners and is consistent with what has generally been reported for bivalves (Boon et al. 1989).

BSAF calculated with TOC-normalized sediment and lipid-normalized biota concentrations appeared to provide a clearer evaluation of resultant data because the relationships became significant compared with uncorrected values. Consequently, we subsequently relied on BSAFs derived from normalised PCB and PBDE concentrations. It has previously been shown that refractory nonpolar trace contaminants concentrate in the OC fraction of sediments and in animal lipids (Hartley and Graham-Bryce 1990). The use of lipid- and OC-normalized concentrations makes the BSAF an approximate fugacity ratio (Burkhard et al. 2005). When bioaccumulation was examined on a congener-specific basis, BSAFs between different sediments differed. This consolidates our view that variation in BSAFs among sediments are shaped by such sediment properties as granulometry or type of OC. It has been recently shown that in benthic invertebrates near submarine municipal outfalls, uptake of individual PBDE congeners was determined by sediment properties (OC, grain size), whereas PCB congener uptake was governed by physicochemical properties (Dinn et al. 2012). It has been demonstrated that although the distribution of hydrophobic organic contaminants increases in the small-particle size fraction (particles <63-μm diameter), the BSAF is often negatively correlated with the fraction of organic size carbon in fine particles (Kukkonen et al. 2005). This pattern shows that as OC increases in the fine material, which is in the size range ingested by the organisms, bioavailability decreases (Menone et al. 2006). Interestingly, in *Neanthes,* the lower relative BSAFs for PCB congeners along with high log K_{ow} values suggest that those compounds may be more strongly adsorbed to organic matter and may therefore be less bioavailable. Different TOC fractions, black carbon being a notable example, have been shown to enhance sorption and reduce the bioavailability of contaminants. A recent study showed that bioaccumulation rates of PCBs in *N. arenaceodentata* were reduced after the addition of a highly sorbent activated carbon (Janssen et al. 2010).

Because estimates of bioaccumulation, such as those reflected in BSAFs, can be both species- and site-specific, this study highlights the need for and use of standardised approaches to correction factors and an understanding of factors affecting bioaccumulation by invertebrates. The expression of BSAFs using lipid-normalized concentration in tissue and OC-normalized concentrations in sediment appears preferable to noncorrected data. This likely reflects the fact that the transport, bioavailability, and bioaccumulation of PCBs and PBDEs are governed by thermodynamic relationships of these chemicals in the marine environment.

We did not seek to determine a steady-state equilibrium for PCBs and PBDEs in this study, which would allow a more complete evaluation of the partitioning of these contaminants from sediment to tissues. Some congeners may require more than the 28 days used here to achieve steady-state tissue residue (functionally defined as at least 80% of the actual equilibrium concentration). Meloche et al. (2009) suggested that after a 28-day exposure, higher log K_{ow} PCB congener concentrations in *M. balthica* had not reached steady state with concentrations in sediments. Determination of steady-state equilibrium is important in the context of risk estimation because tissue concentrations that have not attained steady-state with sediments may underestimate risk. Tissues should be sampled at various time points to determine the uptake and elimination kinetics of multiple contaminant classes of concern (PCBs and PBDEs). The uptake rates of contaminants into test organisms should be directly measured, and a kinetic model should be used to indirectly determine elimination rates. This kinetics information could be then used to derive useable information such as the time (in days) required for analytes to reach steady state, the fraction of steady state acquired after the standard 28-day exposure duration, and BSAFs.

In this study, bioaccumulation was evaluated using two commonly employed test organisms during an exposure time of 28 days. Our results suggest that exposure time required to attain steady state is organism- and compound-specific and that *Neanthes* seemed take longer than *Macoma* to reach stable tissue residues for PCBs but not for PBDEs. Our study suggests that species-specific differences in bioaccumulation and bioavailability from

sediments vary with contaminant properties and sediment type. To improve sediment-based evaluation of food-web bioavailability and/or toxic risk, the collection of detailed information on sediment properties should be undertaken in addition to high-resolution analysis. The value, relevance, and comparability of invertebrate monitoring data are therefore highly dependent on site- (sediment properties) and species-specific (feeding ecology) features. The more complete incorporation of such features into contaminant assessments will contribute to the possibility of expanded comparative evaluations across disparate regions and enable more meaningful pollution indicator assessments using invertebrates.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

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