Persistent organic pollutants in the Scheldt estuary: Environmental distribution and bioaccumulation

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ARTICLE INFO

ABSTRACT

Levels of polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and organochlorine pesticides (OCPs) were determined in the sediment and several species (European flounder, *Platichthys flesus*; common sole, *Solea solea*; Chinese mitten crab, *Eriocheir sinensis*; shore crab, *Carcinus maenas*; brown shrimp, *Crangon crangon*; blue mussel, *Mytilus edulis* and bristle worms, Polychaeta) from 7 locations in the Scheldt estuary (SE, the Netherlands–Belgium). Overall POP levels in the sediment were low. The average PCB and PBDE concentrations were respectively 31.5 and 115 ng/g dry weight (dw). Highest sediment loads were measured in the vicinity of Antwerp (368 ng PCBs/g dw), a location with intense harbor and industrial activities. Pollution concentrations in the tissues of biota were species-specific. Blue mussels contained the highest lipid concentrations (2.74 ± 0.55%) and reached the highest contamination levels (from 287 to 1688 ng PCBs/g ww, from 2.09 to 12.4 ng PBDEs/g ww). Lowest tissue loads were measured in brown shrimp (from 3.27 to 39.9 ng PCBs/g ww, from 0.05 to 0.47 ng PBDEs/g ww). The PCB congener profile in most of the species was similar with the pattern found in the sediment. PCB 153 was the most abundant congener (16.5–25.7% in biota, 10.4% in sediment). In the sediment, the total amount of PBDEs consisted for more than 99% of BDE 209. Congener BDE 47 had the highest concentrations in all sampled species (38.5–70.1%). Sediment POP loadings and tissue concentrations were poorly correlated, indicating that a simple linear or non-linear relationship is insufficient to describe this relationship, possible caused by the complexity of the bioaccumulation processes and the variability in exposure. Because of the high PCB levels, regular consumption of fish and seafood, especially mussels, from the Scheldt estuary should be avoided.

1. Introduction

Due to their intensive production and use over the last century, persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and organochlorine pesticides (OCPs), can be detected worldwide in various environmental compartments (Dominguez et al., 2011; Fuoco et al., 2009; Lammel et al., 2011). The chemical properties of industrially used POPs, such as their chemical stability and hydrophobicity, make them persistent in the environment and give them the ability to bioaccumulate easily into the tissues of biota, to enrich throughout food chains and to eventually cause toxicological effects (Naso et al., 2005). Since human consumption of contaminated animals can also create a potential health risk, a good understanding of bioaccumulation processes of POPs and how they affect aquatic ecosystems at various levels, is necessary.

In aquatic ecosystems such as estuaries and rivers, POPs are mostly concentrated in the sediments which act like a sink for organic compounds (Covaci et al., 2005; Voorspoels et al., 2004b). In addition to food and water (including suspended particulate matter, SPM), sediments represent an important potential exposure pathway for POPs to aquatic species. The bioaccumulation of POPs into biota is influenced by the chemical characteristics of the compounds and the surrounding environmental conditions, but also depends on the characteristics of the species itself (lipid content, metabolic processes) (Dominguez et al., 2011). Because of differences in structural organization, habitat preferences and feeding strategies, all species will be exposed in various ways through the different possible exposure routes. This results in different exposure levels and influences the final bioaccumulation levels of the pollutants in their tissues, making bioaccumulation species-specific. Because of these many influences, bioaccumulation is not always directly linked with total water or sediment loadings and is therefore difficult to predict across a diversity of species.

Despite the inherent complexity of the system, models have been developed to estimate tissue concentrations of exposed organisms, just by measuring environmental concentrations. Accurate estimation methods would indeed make labor-intensive biomonitoring less needed for risk assessment purposes. Biota–sediment accumulation factors (BSAFs) are used in this way, to calculate the potential bioaccumulation

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of organic pollutants in aquatic systems. An accurate application of this approach is only possible when certain conditions are fulfilled, i.e. a steady-state equilibrium and the BSAF independency of exposure concentrations (Iannuzzi et al., 2011; Wong et al., 2001). Given the complexity of the system in terms of exposure concentrations, pathways and biological diversity it is important to assess the impact of differences in structural and functional organization of the organisms on the bioaccumulation levels reached and how these differences can be explained.

This study was conducted in the Scheldt estuary (SE, The Netherlands–Belgium), a frequently used recreational fishing area (Anon, 2010). It receives large volumes of water, originating from densely populated and industrialized areas such as Brussels, Antwerp and Ghent, enriching the estuary with pollutants, including trace metals (Baeyens et al., 2005), volatile organic compounds (VOCs) (Huybrechts et al., 2003) and POPs (Steen et al., 2001; Voorspoels et al., 2003). Main sources of POPs in the estuary are wastewater, industrial activities, agriculture runoff and harbor activities.

The aims of the present study were (1) to determine POP concentrations in sediment of the SE and aquatic animals, representative for different groups and trophic levels, (2) to investigate the relationship between sediment loadings and the bioaccumulation of POPs into different species in the SE, taking into account sediment and water characteristics, (3) to study seasonal differences in bioaccumulation of POPs and (4) to estimate the potential human health risk through consumption of contaminated edible species.

2. Material and methods

2.1. Sampling

Samples were collected at seven locations along the SE (Fig. 1), covering the marine region (L1, Vlissingen) until the freshwater region (L7, Kastel) of the river. Two fish species, European flounder (*Platichthys flesus*) and common sole (*Solea solea*), two crab species, shore crab (*Carcinus maenas*) and Chinese mitten crab (*Eriocheir sinensis*), and brown shrimp (*Crangon crangon*), were collected by fyke fishing (INBO, Research Institute for Nature and Forest) and trawl fishing with the vessel *Zeeleeuw* (VLIZ, Flanders Marine Institute). Blue mussel (*Mytilus edulis*) and bristle worms (Polychaeta) were gathered by hand at low tide. From fish, crabs and shrimps, muscle tissues were separated for analysis during dissection. A caudal part of the fish muscle tissue was sampled. For the smaller fish, it was necessary to take the whole muscle tissue. From crabs, all muscle tissue was sampled and homogenized. From blue mussels and bristle worms, the whole soft body was taken. Tissues from brown shrimp, blue mussels and bristle worms were pooled to get an adequate sample size. Animals were not depurated. All samples were stored at −20 °C until analysis.

The top layer (10 cm) of the surface sediment was sampled manually from the shores at low tide and with a Van Veen grab in the middle of the river during the cruises with vessel *Zeeleeuw* (VLIZ, Flanders Marine Institute) and vessel *Luctor* (NIOO, Netherlands Institute of Ecology). Where possible, at least three sediment samples from each location were pooled. Shore sediment and samples from the middle section were treated separately. At each location, water temperature, pH and conductivity (μS/cm) were measured using a WTW Multiline P4 meter. At the laboratory, the sediment samples were divided into subsamples for contaminant analysis, organic matter content (TOC, total organic carbon) and clay content determination. TOC was determined through Loss on Ignition (LOI). To this, the sediment subsamples were incinerated at 550 °C for 4 h and weight loss was determined (Heiri et al., 2001). Clay content was measured with laser diffraction (Malvern Mastersizer S., Worcestershire, UK). Because of difficulties during sampling, sediment from the middle of the river could not always be collected, leaving gaps in the database. Calculations and correlations were therefore performed using only the shore sediment data.

This sampling campaign was repeated three times: in March, July and September 2010. Due to ecological features, not all species could be collected at each location and during every season. For example,
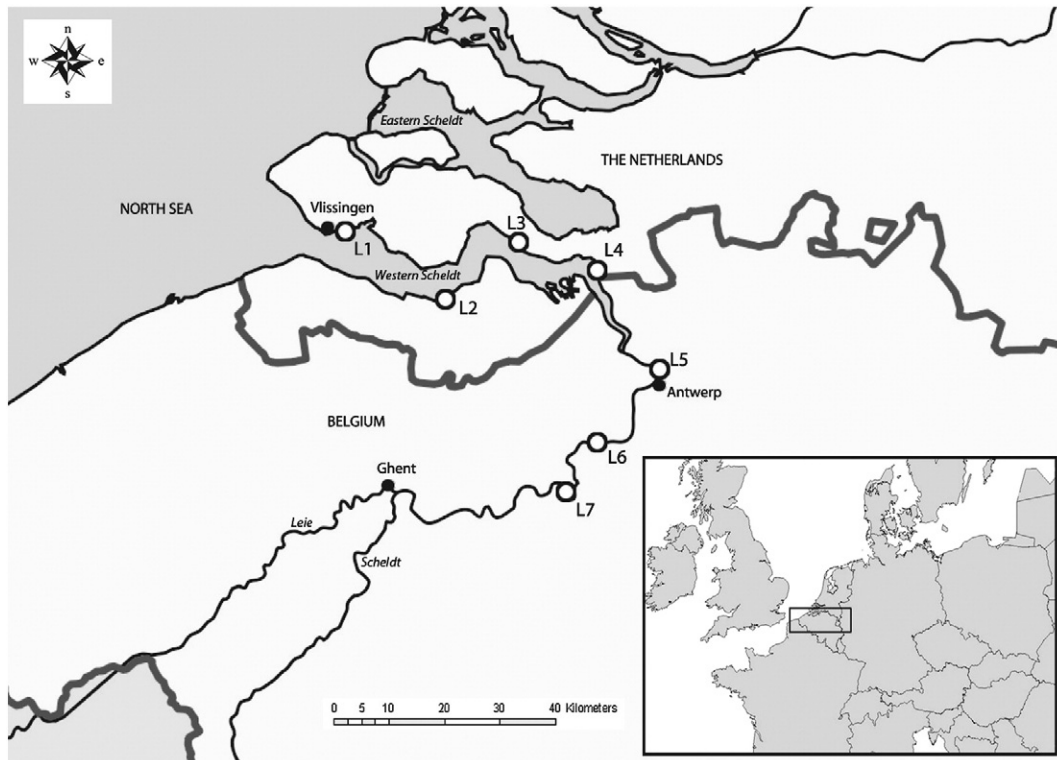

Fig. 1. Sampling locations along the Western Scheldt: 1, Vlissingen; 2, Terneuzen (Griete); 3, Waarde; 4, Bath (the Netherlands); 5, Antwerpen; 6, Steendorp; and 7, Kastel (Belgium).

because the Chinese mitten crab is a brackish and freshwater inhabitant, it could only be collected at the sampling locations more upstream of the estuary (L3–L7). Blue mussel was only collected at the two first sampling locations. In March, common sole could not be collected, probably because cold temperatures kept the animals into deeper waters of the North Sea. An overview of the collected samples is presented in Table 1.

2.2. Sample preparation and clean up

In all samples, 33 PCB congeners (IUPAC numbers: CB 18, 28, 44, 49, 52, 87, 95, 99, 101, 105, 110, 118, 128, 138, 146, 149, 151, 153, 156, 170, 171, 172, 174, 177, 180, 183, 187, 194, 195, 199, 205, 206, 209), 7 PBDEs (IUPAC numbers: BDE 28, 47, 99, 100, 153, 154, 183), DDXs (*o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDT, *p,p′*-DDT), chlordanes (TC, CC, TN, OxC), HCHs (α-, β-, γ-hexachlorocyclohexane) and HCB (hexachlorobenzene) were targeted for analysis. PBDE 209 was measured only in the sediment samples. Although higher brominated BDEs, such as BDE 209, are present in estuarine sediments, the uptake in biota appears negligible (Voorspoels et al., 2003). A combination of fast metabolism and low uptake, caused by the very high log K_{ow} and the size of the molecule, which impede the compound from crossing membranes, is related to their absence in organisms (Boon et al., 2002; Dominguez et al., 2011; Kierkegaard et al., 1999; Voorspoels et al., 2004b).

The method used for the preparation and clean-up of the samples has been previously described in detail (Covaci et al., 2005; Roosens et al., 2008) and is briefly presented below. For the animal samples, an adequate amount of tissue (between 1 and 5 g) was ground with anhydrous Na_2SO_4, spiked with internal standards (CB 143, BDE 77, and ε-HCH) and extracted for 2 h by hot Soxhlet with 80 ml hexane/acetone (3/1; v/v). After lipid determination (on an aliquot of the extract), the extract was cleaned-up on 8 g acid silica and analytes were eluted

with 20 ml hexane and 15 ml dichloromethane. The eluate was concentrated and reconstituted in 100 μl *iso*-octane.

For sediments, the same procedure was followed, but 5 g of activated copper powder was added and mixed with the sample (3 g), followed by the addition of internal standards (CB 143, BDE 77, ^{13}C-BDE 209, and ε-HCH), before Soxhlet extraction. Copper powder (2 g) was also added on top of the acid silica during the clean-up step. The rest of the clean-up procedure for sediments was similar to biota, except for lipid determination.

2.3. Analysis

For the detection of PBDEs, HCHs and chlordanes, an Agilent 6890-5973 gas chromatograph was used, coupled with a mass spectrometer (GC–MS) and equipped with a capillary column (30 m × 0.25 mm × 0.25 μm DB-5). The GC–MS was operated in an electron capture negative ionization (ECNI) mode and used in the selected ion-monitoring (SIM) mode. For the measurements of PCBs, DDXs, and HCB a similar GC–MS system was operated in electron ionization (EI) mode and equipped with a 25 m × 0.22 mm × 0.25 μm HT-8 capillary column.

2.4. Quality assurance/quality control (QA/QC)

Quality control during extraction, clean-up and analysis procedures was assured by regular analysis of procedural blanks and the use of standard reference materials: SRM 1945 — PCBs, PBDEs and OCPs in whale blubber (NIST, National Institute of Standards and Technology) and CRM 536 — PCBs in freshwater sediment (BCR, Community Bureau of Reference of the European Commission). Measured values were deviating less than 20% from the certified values. For each compound, the mean procedural blank value was used for subtraction. After blank subtraction, the limit of quantification (LOQ) was set at 3 times the standard deviation of the procedural blanks, taking into account the sample intake. For analytes that were not detected in procedural blanks, LOQs were calculated for a ratio S/N equal to 10. LOQs ranged between 1 and 4 ng/g lipid weight. The QC scheme is assessed through regular participation to inter-laboratory comparison exercises organized by the US National Institute of Standards and Technology.

2.5. Statistical analysis

Statistical analyses were conducted using GraphPad Prism 5.04 (GraphPad Software, Inc.) and SigmaPlot 11.0 (Systat Software Inc.). The level of statistical significance was defined at $p < 0.05$. For concentrations below the LOQ, a value of LOQ∗f (detection frequency) was used. After testing the normality and homogeneity of variances, data were log-transformed where necessary. Differences between locations and sampling periods were detected by using two-way ANOVA with Tukey test. To compare bioaccumulation among species, both wet weight based and lipid weight based tissue concentrations were compared, using one-way ANOVA with Tukey test. Pearson's correlation coefficients were calculated between pollution levels in sediment and tissues. Multiple regression was used to explore the relationship between POP tissue concentrations and POPs in the sediment and environmental conditions (TOC, pH, conductivity and clay content) as additional independent variables. Multiple regression was calculated for both non-transformed and log-transformed data. For all detected compounds, biota–sediment accumulation factors (BSAFs) were calculated as the ratio of the tissue concentration (lipid weight basis) to the sediment concentrations in the shore samples (Bervoets et al., 2005). Both TOC-normalized as non-normalized sediment concentrations were used. One-way ANOVA with Tukey test was used to compare the BSAFs between species and compounds.

Table 1
Overview of the number of samples collected. For brown shrimps, blue mussels and bristle worms the number of pooled samples is presented.

Location	Bristle worms	Blue mussel	Brown shrimp	Shore crab	Chinese mitten crab	Common sole	European flounder
Vlissingen							
March	1						1
July		2	2	2		5	2
September	1		2	1			
Terneuzen							
March							
July	1	2	2	3		2	4
September		2	2	2		4	1
Waarde							
March							
July			2	2	2	2	
September	1		2	1		1	
Bath							
March	1		1		2		3
July	1		2		2	5	5
September			3	1			4
Antwerpen							
March					2		1
July					2	5	6
September	1						5
Temse							
March							1
July							
September					2		1
Kastel							
March					1		
July							
September							1

Table 2
Concentration ranges and median concentrations of PCBs, PBDEs and OCPs in sediment (ng g^{-1} dw) and biota (ng g^{-1} ww); ranges and median lipid levels of the biota samples of the Scheldt estuary. Values indicated in bold, are exceedances of EU regulation limits (75 ng sum 6 ICES PCBs/g ww; European Commission, 2011).

	Sediment	Bristle worms	Blue mussel	Brown shrimp	Shore crab	Chinese mitten crab	Common sole	European flounder
N	36	7	6	18	12	13	24	35
Lipids (%)	–	0.96–1.55	2.04–3.68	0.50–0.93	0.30–0.68	0.38–0.97	0.42–1.25	0.44–1.75
	–	1.26	2.67	0.69	0.51	0.70	0.69	0.81
PCB 18	0.06–2.51	0.09–1.49	0.71–2.17	<LOQ	<LOQ	0.02–1.21	0.11–0.36	0.11–2.83
	0.06	0.09	1.28			0.02	0.11	0.41
PCB 28	0.07–5.11	0.17–2.32	1.60–4.64	0.01–0.78	0.19–1.47	0.31–4.56	0.14–1.10	0.14–4.74
	0.07	0.32	3.78	0.01	0.39	1.32	0.19	0.97
PCB 52	0.11–18.9	0.17–2.85	8.94–35.1	0.14–1.69	0.18–3.57	0.58–4.01	0.26–5.37	0.29–8.66
	0.20	1.06	20.7	0.25	0.98	1.58	1.02	3.12
PCB 49	0.11–16.4	0.17–1.81	5.58–20.9	<LOQ	0.19–3.42	0.52–2.76	0.18–2.77	0.18–4.39
	0.38	0.61	12.5		1.00	1.31	0.43	1.49
PCB 44	0.10–9.88	0.17–1.63	3.731–12.3	0.02–0.63	0.08–0.33	0.08–1.69	0.15–2.05	0.15–2.24
	0.14	0.26	7.11	0.02	0.08	0.30	0.25	0.75
PCB 95	0.07–26.6	0.28–7.28	16.8–89.0	0.14–2.13	0.13–1.78	0.30–3.16	0.49–9.79	0.40–11.7
	0.34	2.29	45.6	0.39	0.82	0.76	1.70	4.46
PCB 101	0.08–31.6	0.71–4.87	25.0–147	0.21–2.05	0.92–21.3	1.34–9.90	0.67–15.0	0.67–26.0
	0.54	3.43	67.3	0.41	3.29	4.86	2.49	7.13
PCB 99	0.06–11.1	0.66–4.82	12.1–71.5	0.08–1.31	0.94–14.6	0.62–6.51	0.33–6.45	0.37–10.6
	0.15	2.39	30.9	0.21	2.90	2.18	1.57	3.12
PCB 87	0.07–8.61	0.09–1.17	3.45–20.8	0.06–0.38	0.09–0.56	0.30–2.18	0.10–2.76	0.12–4.85
	0.23	0.49	10.7	0.10	0.25	0.93	0.58	1.35
PCB 110	0.07–31.0	0.39–5.23	15.9–102	0.09–1.21	0.18–3.12	0.61–5.90	0.47–8.48	0.41–20.0
	0.39	2.25	47.2	0.18	0.84	2.00	1.70	5.37
PCB 118	0.07–22.4	0.48–4.14	13.2–71.6	0.10–2.39	0.40–10.3	0.87–12.7	0.35–6.81	0.60–14.0
	0.34	2.24	31.7	0.50	1.40	3.94	1.84	4.08
PCB 105	0.04–7.20	0.13–1.08	2.86–16.9	0.02–0.41	0.10–2.76	0.27–2.85	0.10–2.00	0.13–3.89
	0.04	0.55	7.64	0.02	0.33	1.08	0.43	1.15
PCB 151	0.07–8.19	0.19–3.34	8.62–55.7	0.12–2.18	0.12–1.31	0.13–1.74	0.27–6.30	0.53–10.8
	0.22	1.27	24.9	0.55	0.55	0.31	0.99	3.32
PCB 149	0.08–32.5	1.02–10.2	35.6–227	0.27–2.39	0.71–18.2	0.70–6.73	0.81–18.1	0.66–23.3
	0.51	5.46	99.7	0.47	3.04	1.77	2.85	7.77
PCB 146	0.06–7.24	0.52–2.96	12.7–77.2	0.25–2.90	0.32–8.56	0.58–13.2	0.34–7.64	0.61–13.4
	0.14	1.87	31.8	1.14	1.01	2.38	1.55	3.58
PCB 153	0.07–35.7	2.39–23.2	50.7–362	0.32–6.38	2.58–55.2	2.60–41.0	1.37–37.3	1.91–53.9
	0.50	11.9	150	1.07	8.46	10.2	6.65	16.7
PCB 138	0.08–34.4	1.15–12.1	26.1–182	0.17–3.39	1.14–21.5	1.74–23.9	0.82–23.3	0.89–34.5
	0.43	5.83	75.8	0.47	3.57	6.95	3.81	10.4
PCB 128	0.04–2.90	0.13–0.90	2.90–12.0	0.01–0.22	0.10–2.04	0.20–2.47	0.10–2.88	0.10–2.26
	0.04	0.66	7.31	0.01	0.35	0.72	0.46	1.02
PCB 156	0.04–1.84	0.09–0.33	1.23–5.19	0.06–0.62	0.04–1.44	0.15–2.53	0.09–1.02	0.11–2.17
	0.04	0.24	3.56	0.21	0.15	0.69	0.25	0.63
PCB 187	0.06–5.28	0.64–3.21	14.3–62.0	0.27–2.73	0.41–9.45	0.63–8.31	0.43–9.45	0.74–10.9
	0.13	2.20	37.8	1.15	1.30	2.78	2.11	3.92
PCB 183	0.05–2.38	0.12–1.76	3.31–20.0	0.08–0.68	0.18–4.71	0.20–1.37	0.11–4.44	0.23–4.31
	0.05	0.79	12.7	0.20	0.55	0.73	0.53	1.61
PCB 174	0.05–9.72	0.10–0.87	0.72–1.56	0.06–0.49	0.04–2.21	0.10–1.84	0.11–6.89	0.10–5.84
	0.09	0.47	1.04	0.10	0.29	0.13	0.43	2.09
PCB 177	0.05–5.63	0.25–1.29	6.91–37.7	0.09–1.07	0.12–2.90	0.28–4.37	0.11–5.27	0.19–4.85
	0.05	1.02	17.0	0.35	0.41	1.34	0.56	1.58
PCB 171	0.05–2.79	0.13–1.03	2.05–13.2	0.05–0.34	0.08–1.19	0.08–0.67	0.09–2.06	0.09–2.85
	0.05	0.45	6.00	0.06	0.20	0.29	0.25	0.85
PCB 172	0.03–1.63	0.10–0.31	0.55–1.89	0.06–0.47	0.07–1.01	0.07–1.68	0.03–1.61	0.08–2.30
	0.03	0.21	1.41	0.16	0.07	0.35	0.15	0.66
PCB 180	0.07–10.7	0.52–4.58	6.35–28.0	0.23–5.20	0.25–9.08	1.14–20.2	0.26–17.6	0.98–20.9
	0.19	2.54	18.0	1.48	1.08	4.61	2.19	7.14
PCB 170	0.05–5.35	0.28–1.60	1.99–8.01	0.11–2.08	0.09–1.87	0.49–6.94	0.10–6.94	0.45–7.61
	0.08	1.09	5.23	0.42	0.34	1.97	1.00	2.84
PCB 199	0.04–3.55	0.14–0.42	0.07–0.63	0.08–0.71	0.08–1.23	0.18–2.52	0.09–4.16	0.13–4.64
	0.04	0.30	0.17	0.27	0.12	0.61	0.33	0.99
PCB 195	0.03–1.58	0.09–0.31	0.06–0.69	0.07–0.20	0.04–0.15	0.04–0.28	0.08–1.44	0.08–1.57
	0.03	0.17	0.17	0.07	0.04	0.12	0.10	0.38
PCB 194	0.04–2.82	0.15–0.52	0.09–3.02	0.07–0.52	0.07–0.33	0.07–0.80	0.09–3.26	0.12–3.63
	0.04	0.36	1.40	0.16	0.07	0.24	0.18	0.91
PCB 205	0.01–0.65	<LOQ	0.07–0.41	<LOQ	<LOQ	<LOQ	0.01–0.13	0.01–0.17
	0.01		0.21				0.01	0.01
PCB 206	0.03–1.38	0.06–0.14	0.07–0.97	<LOQ	<LOQ	0.01–0.45	0.05–0.61	0.05–0.68
	0.03	0.10	0.22			0.01	0.05	0.13
PCB 209	0.03–0.83	0.01–0.12	0.09–0.44	<LOQ	<LOQ	0.01–0.43	0.02–0.29	0.02–0.29
	0.03	0.01	0.31			0.01	0.02	0.02
Sum PCBs	1.91–368	11.6–106	287–1690	3.27–39.9	10.9–205	16.0–172	9.47–213	12.9–285
	6.43	52.8	782	10.6	36.5	60.8	39.3	105
Sum 6 ICES PCBs	0.47–136	5.12–49.2	**119–759**	1.09–17.6	5.29–**112**	8.44–96.9	3.71–97.3	5.32–**137**
	1.88	24.2	**337**	3.65	18.5	31.8	17.2	46.1
HCB	0.01–1.29	0.07–0.26	0.09–0.76	<LOQ	0.04–0.17	0.04–0.27	<LOQ	0.05–0.44

Table 2 (*continued*)

	Sediment	Bristle worms	Blue mussel	Brown shrimp	Shore crab	Chinese mitten crab	Common sole	European flounder
	0.01	0.14	0.29		0.04	0.16		0.16
op-DDE	0.01–1.46	0.04–0.15	0.08–1.72	<LOQ	<LOQ	<LOQ	<LOQ	0.01–0.22
	0.01	0.04	0.58					0.01
pp-DDE	0.05–12.1	0.14–3.51	10.1–38.9	0.08–1.03	0.20–6.66	0.63–9.55	0.19–4.08	0.32–7.40
	0.05	1.68	21.7	0.25	0.72	1.71	0.93	2.32
op-DDD	0.03–1.57	0.07–1.06	1.77–8.17	<LOQ	<LOQ	<LOQ	0.05–0.12	0.05–0.62
	0.03	0.62	4.59				0.05	0.19
op-DDT	0.01–0.34	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	0.01							
pp-DDD	0.04–6.39	0.09–1.45	3.50–16.3	0.02–0.20	0.02–0.18	0.02–0.36	0.09–1.63	0.09–2.17
	0.04	0.96	8.49	0.02	0.02	0.02	0.29	0.83
pp-DDT	0.01–1.21	0.01–0.42	0.09–1.17	0.02–0.14	0.04–0.25	0.04–0.17	0.02–0.13	0.02–0.39
	0.01	0.01	0.72	0.02	0.04	0.04	0.02	0.02
OxC	<LOQ	<LOQ	0.04–0.13	<LOQ	0.06–0.21	0.22–2.45	<LOQ	0.00–0.10
			0.04		0.06	0.41		0.00
TN	<LOQ	0.03–0.11	0.46–0.77	<LOQ	<LOQ	0.05–1.03	<LOQ	0.01–0.30
		0.03	0.73			0.17		0.01
CN	<LOQ	<LOQ	0.20–0.48	<LOQ	<LOQ	0.01–0.22	<LOQ	0.00–0.11
			0.36			0.01		0.00
TC	0.01–0.22	0.06–0.20	0.44–1.27	<LOQ	<LOQ	<LOQ	<LOQ	0.02–0.30
	0.01	0.11	1.01					0.02
CC	<LOQ	0.01–0.12	0.50–2.61	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
		0.01	0.80					
a-HCH	0.02–0.36	0.03–0.12	0.25–0.46	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	0.02	0.03	0.32					
b-HCH	0.01–0.40	<LOQ	0.15–0.30	<LOQ	0.02–0.10	0.02–0.10	<LOQ	<LOQ
	0.01		0.21		0.02	0.02		
g-HCH	0.04–0.27	0.06–0.31	0.43–1.07	<LOQ	<LOQ	<LOQ	0.05–0.16	0.05–0.40
	0.04	0.12	0.79				0.05	0.12
BDE 28	0.00–0.08	0.01–0.09	0.05–0.70	<LOQ	0.02–0.05	0.02–0.12	<LOQ	0.01–0.13
	0.00	0.01	0.27		0.02	0.06		0.01
BDE 47	0.03–1.33	0.33–0.85	0.28–5.16	0.02–0.23	0.05–0.75	0.70–2.32	0.05–0.59	0.13–2.71
	0.10	0.52	1.43	0.02	0.11	1.57	0.17	0.69
BDE 100	0.01–0.33	0.05–0.30	0.72–2.78	0.01–0.05	0.03–0.05	0.03–0.25	0.04–0.51	0.04–0.77
	0.01	0.17	1.35	0.01	0.03	0.14	0.06	0.22
BDE 99	0.03–1.60	0.21–0.60	0.05–3.35	0.02–0.20	0.04–0.29	0.20–0.75	0.04–0.26	0.04–0.58
	0.04	0.38	0.36	0.02	0.04	0.33	0.08	0.11
BDE 154	0.01–0.20	0.01–0.05	0.20–0.58	<LOQ	<LOQ	0.00–0.05	0.02–0.27	0.02–0.25
	0.01	0.01	0.35			0.00	0.02	0.06
BDE 153	0.01–0.21	0.01–0.16	0.05–0.45	<LOQ	<LOQ	0.01–0.08	<LOQ	0.01–0.07
	0.01	0.01	0.14			0.01		0.01
BDE 183	0.01–0.13	<LOQ	0.04–0.27	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	0.01		0.09					
BDE 209	0.43–1200	–	–	–	–	–	–	–
	6.80							
Sum PBDEs	0.52–1200	0.87–1.81	2.09–12.4	0.05–0.47	0.14–0.96	1.02–3.24	0.17–1.53	0.25–3.97
	6.90	1.20	3.80	0.05	0.20	2.15	0.38	1.19

<LOQ — below limit of quantification.

3. Results and discussion

3.1. Sediment

3.1.1. POP concentrations

The TOC and clay content in the sediment ranged respectively from 0.40% to 9.91% and from 0.58% to 13.79%, with a positive trend for both with increasing distance from Vlissingen (L1). Sediment concentrations of PCBs, PBDEs and OCPs expressed in ng/g dw, are presented in Table 2. All analyzed compounds were detectable, except for chlordanes, where only low concentrations of TC (*trans*-chlordane) were measured. A positive correlation between POP concentrations and the TOC of the sediment was found (N = 36; for $\sum$ PCBs: p < 0.0001, r^2 = 0.397; for $\sum$ PBDEs: p = 0.0001, r^2 = 0.359, for $\sum$ DDXs: p = 0.0003, r^2 = 0.333). Also a positive correlation between POP concentrations and the clay content was detected (N = 36; for $\sum$ PCBs: p < 0.0001, r^2 = 0.499; for $\sum$ PBDEs: p = 0.0009, r^2 = 0.280, for $\sum$ DDXs: p < 0.0001, r^2 = 0.413).

With an average concentration of 31.5 ng/g dry weight (dw), PCB levels were low. The highest PCB concentrations (368 and 128 ng/g dw) were detected in Antwerp (L5) and Temse (L6). Since the area of Antwerp is highly urbanized and industrialized, higher levels of pollution in its vicinity were expected. PCB congeners 153, 149 and 138 were the most dominant ones (respectively 10.4%, 9.3% and 8.9% of total PCBs).

Similar levels of PCB concentrations (from 151 to 400 ng/g dw) were measured by Covaci et al. (2005) in sediment cores obtained in 2000 from Kruibeke, located between Antwerp and Temse, indicating that PCB levels did not decrease rapidly in the last 10 years. Also in sediment of the harbors of Amsterdam and Rotterdam (The Netherlands), comparable PCB levels (up to 120 ng/g dw) were found (de Boer et al., 2001). In sediments from 3 Spanish estuaries (Santoña, Mogro and Tina Menor), sampled in 2006, PCB concentrations were lower (up to 1.47 ng PCB 153/g dw and 0.92 ng PCB 138/g dw) (Gomez-Lavin et al., 2011). However, in the Seine estuary (France), higher average PCB levels were reported (10 ng PCB 153/g dw, 9 ng PCB 138/g dw; this study: 3.28 ng PCB 153/g dw and 2.79 ng PCB 138/g dw) (Durou et al., 2007). Also in Italy (Pialassa Baiona coastal lagoon), higher PCB concentrations have been reported (up to 192 ng PCB 118/g dw and 25 ng PCB 105/g dw) (Guerra, 2012).

In the present study, all 8 analyzed PBDEs were detected but in relatively low concentrations. The total amount of PBDEs consisted for more than 99% of BDE 209. The other abundant congeners were BDE 99 and BDE 47. The same profile was described previously by Voorspoels
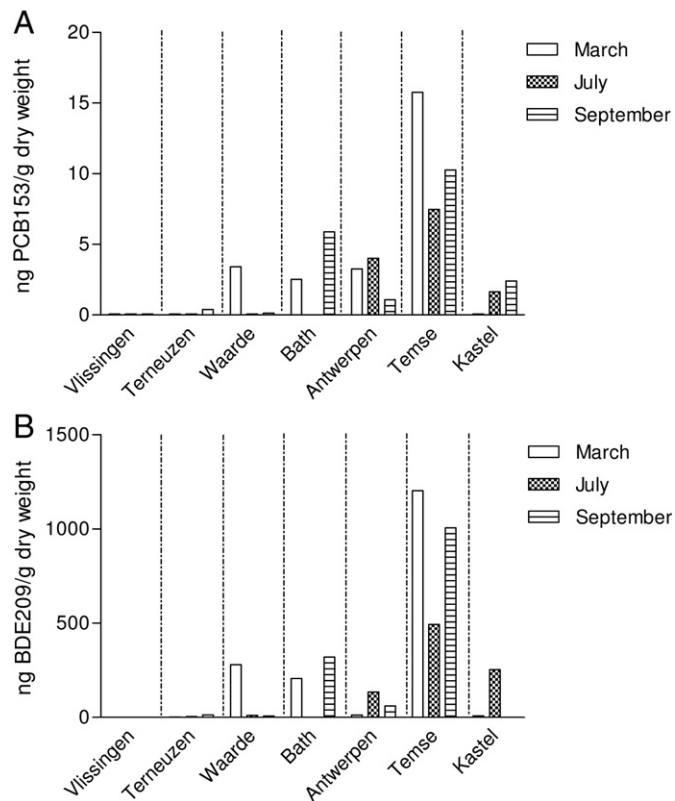
Fig. 2. Geographical and temporal variation of A) CB153 and B) BDE209 concentrations in the sediment (not normalized), starting from L1 on the left to L7 on the right.

Fig. 3. Average PCB (A) and PBDE (B) congener profiles in all samples. Only the 7 ICES PCBs are presented. BDE 209 is excluded.

et al. (2004b) for Belgium and by Zegers et al. (2003) for other West European countries. An average concentration of 0.61 ng ΣPBDE/g dw (without BDE 209) and 114 ng BDE 209/g dw was measured. At sampling location 6 (Temse), PBDE concentrations in the sediment taken at the shores of the estuary were remarkably high (up to 1205 ng/g dw). The same trend as noted by de Boer et al. (2003) was found, i.e. higher levels of BDE 209 in the east part of the estuary and a decreasing trend towards the sea. In the present study however, lower concentrations were detected at Terneuzen (average 4.8 ng BDE 209/g dw) comparing to the above-mentioned study (110 ng BDE 209/g dw). In the Clyde estuary (UK), PBDE levels were higher (from 1 to 2337 ng BDE 209/g dw) (Vane et al., 2010). Comparison with the PBDE concentrations measured in samples from the Scheldt obtained in 2001 (Voorspoels et al., 2004b), suggest a slight decrease in PBDE levels over the last ten years (up to 6.1 ng BDE 47/g dw, 6.1 ng BDE 99/g dw; this study: up to 1.3 ng BDE 47/g dw and 1.6 ng BDE 99/g dw).

OCP levels were very low. In case of DDTs, p,p′-DDE (48%) and p,p′-DDD (32%) were most abundant. In contrast with Covaci et al. (2005), HCHs were detected in the present study, yet at very low concentrations (up to 0.78 ng/g dw).

For some locations, large differences in concentrations between the shore samples and the samples from the middle of the river were found, however no constant trend could be indicated. For example, in Antwerp (L5) shore samples contained an average value of 23.7 ng PCB 153/g dw, but in the middle of the river an average of 2.78 ng PCB 153/g dw was measured. In Bath (L4), samples from the middle of the river contained significantly more PCB 180 than the shore samples (2.51 and 0.11 ng PCB 180/g dw, respectively). These differences can be explained by large local variability in sedimentation and erosion. Local hydromorphology, sediment properties and biological properties all influence the sedimentation and erosion characteristics in a different way (Grabowski et al., 2011), which leads to complex erosion patterns.
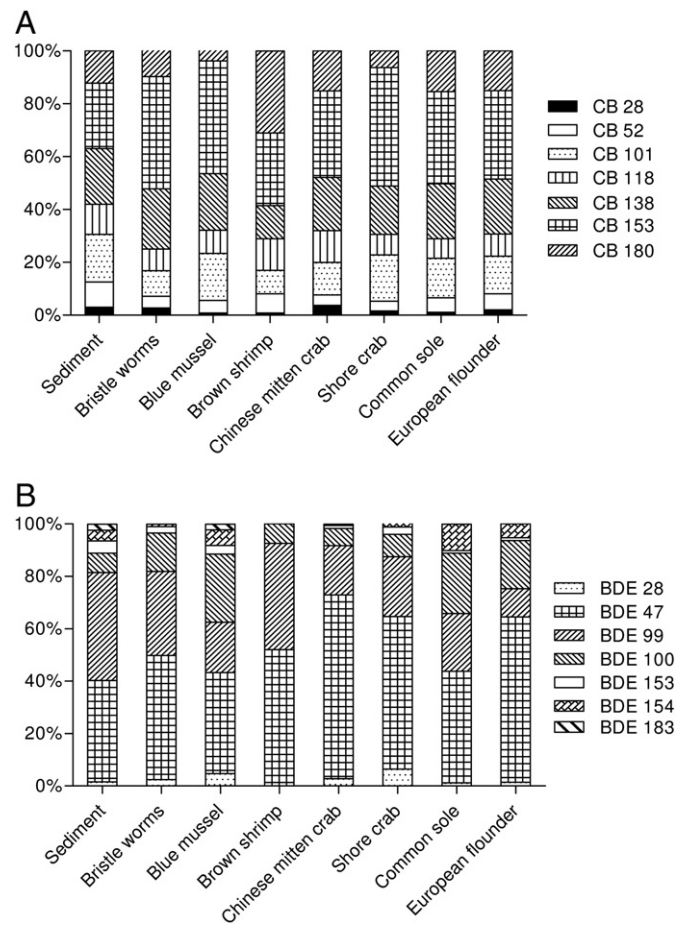
3.1.2. Geographical and temporal variation

For PCB and PBDE concentrations in the sediment, significant differences were found between the sampling locations (p ≤ 0.008) (Fig. 2). Higher concentrations were measured towards more industrialized areas (L5, Antwerp). Regarding temporal variation, differences in concentrations were not large enough to indicate a seasonal effect on sediment POP loadings (0.219 < p < 0.991).

3.2. Biota

3.2.1. POP concentrations

POP concentrations and lipid content in sampled biota from the Scheldt estuary are summarized in Table 2. The lipid percentage varied from 0.53 ± 0.12% in shore crab to 2.74 ± 0.55% in blue mussel (mean ± SD).

Except for PCB 205 and BDE 183, all analyzed PCB and PBDE congeners were detected in most species. Total PCB levels were highest in blue mussel, where concentrations ranged from 287 to 1688 ng/g wet weight (ww) and lowest in brown shrimp with concentrations from 3.27 to 39.9 ng/g ww. The maximum limit for the sum of the 6 indicator PCBs (75 ng/g ww), as set by European regulation (European Commission, 2011), was exceeded by levels in blue mussel, shore crab and European flounder. Also total PBDE levels were highest in blue mussel (from 2.09 to 12.4 ng/g ww) and lowest in brown shrimp (from 0.05 to 0.47 ng/g ww).

The average PCB and PBDE congener profiles are presented in Fig. 3. CB 153 was the most dominant PCB congener in all species (16.5–25.7%), except in brown shrimp, where CB 180 was most detected (13.8%). The congener pattern in most of the sampled species was similar
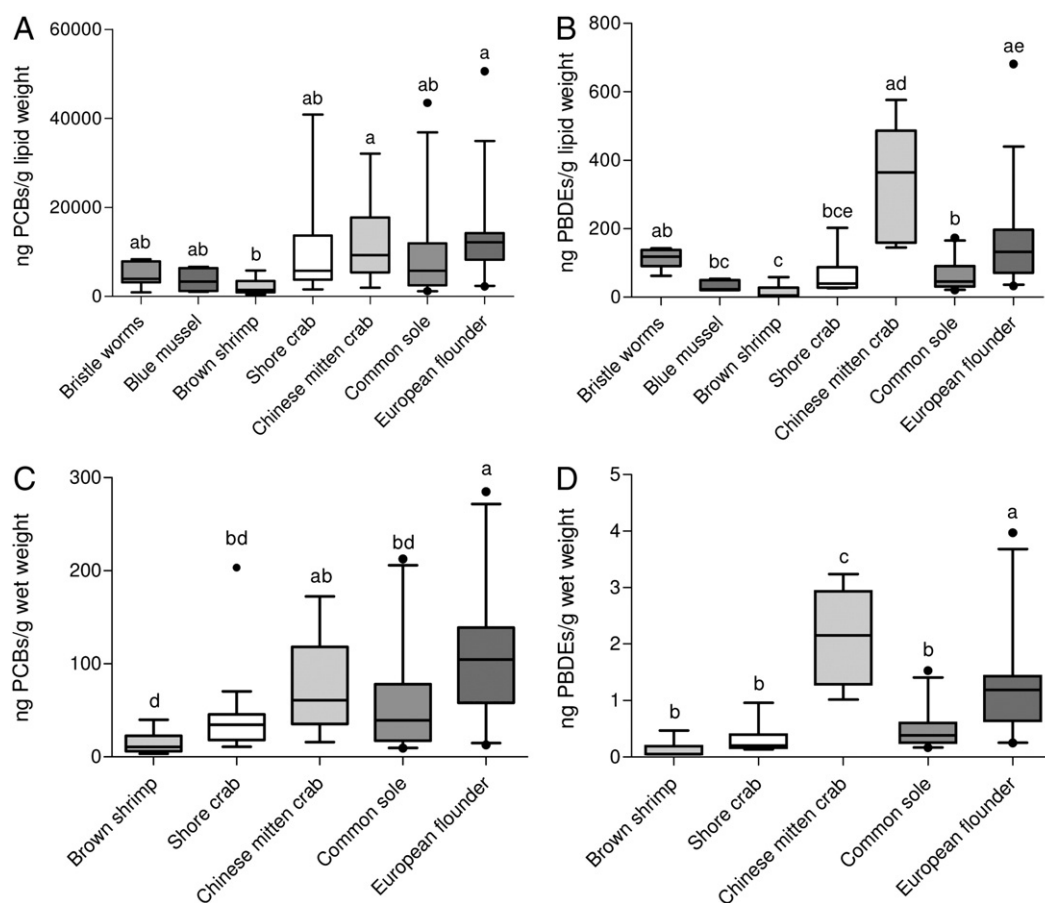
Fig. 4. Boxplots of sum PCB and sum PBDE for all species at all locations on lipid weight basis (A and B) and on wet weight basis (C and D). Whiskers indicate 5–95 percentile; box indicates 25–75 percentile; line shows the median; dots represent outliers. Boxes indicated with the same letter are not significantly different.

with the pattern found in the sediment. Relatively higher concentrations of hepta- and octa-PCB congeners in brown shrimp were also described by Voorspoels et al. (2004a). From the present PBDEs, congener BDE 47 had the highest concentrations in all species (38.5–70.1%), followed by congeners BDE 99 or BDE 100. This typical PBDE congener profile is found worldwide due to the former use of the commercial penta-BDE mixture, mainly as flame retardant in consumer products (Luross et al., 2002; Roosens et al., 2008).

Although no chlordanes were detected in the sediment, except low concentrations of TC, they were present in the tissues of bristle worms, blue mussel, Chinese mitten crab and European flounder, yet at low concentrations (from not detected to 5.11 ng/g ww).

Table 3
Concentration ranges of PBDEs (sum of tri- to hepta-BDEs) and PCBs (sum 7 ICES PCBs) in *Mytilus* sp. from various studies.

Location	PBDE (ng/g ww)	Reference
Belgium	2.09–12.4	Present study
France	0.1–2	Johansson et al. (2006)
Denmark	0.08–0.8	Christensen and Platz (2001)
UK	8	Allchin et al. (1999)
Korea	0.04–9.5	Ramu et al. (2007)
Japan	0.03–2	Ueno et al. (2010)
US (San Francisco)	1.3–3.7	Oros et al. (2005)

Location	PCB (ng/g ww)	Reference
Belgium	132–830	Present study
Spain	1.04–624	Bellas et al. (2011)
Italy	1.32–18.5	Bayarri et al. (2001)
Denmark–Poland	n.d.–412	Mikoszewski and Lubelska (2010)

n.d. – not detected.

In the present study, tissue concentrations were not correlated with the size of the fish and crabs or with the octanol–water partition coefficient (log K_{ow}) (data not shown).

When comparing the tissue concentrations of the different POPs in the species on wet weight basis, some trends were observed (Fig. 4). Shrimp tissues always contained the lowest concentrations of POPs, as also noticed by Voorspoels et al. (2004a). These authors suggested that the low levels in shrimp were partially caused by their more pelagic nature and their feeding habits. The other sampled species live in closer contact with the sediment, resulting in higher pollution loads. Brown shrimp feed mainly on preys of lower trophic levels, like amphipods or mysids (Oh et al., 2001). Although European flounder and common sole feed on the same prey species and in the same niche (Stevens et al., 2006), European flounder contained significantly higher POP concentrations than common sole (0.0001<p<0.002). Individuals of European flounder were however bigger in size than individuals of common sole (medians of 21.2 and 15.4 cm, respectively). From the two crab species, Chinese mitten crab bioaccumulated the highest concentrations, although there was no size difference. Chinese mitten crab was mostly collected more upstream in the estuary, where pollution levels are higher, but even when caught at the same sampling location, shore crabs clearly contained less pollutants. As the Chinese mitten crab is a catadromous crab (Dittel and Epifanio, 2009) and the shore crab a saltwater inhabitant, the two crab species probably experience a different exposure pattern when integrated over time.

In case of blue mussels and bristle worms, the whole soft body was analyzed, so that comparison with the other species, where only muscle tissue was analyzed, was not possible on a wet weight basis. The lipid content in blue mussel was significantly higher compared to the other species (average 2.74%). Because of this, wet weight based POP
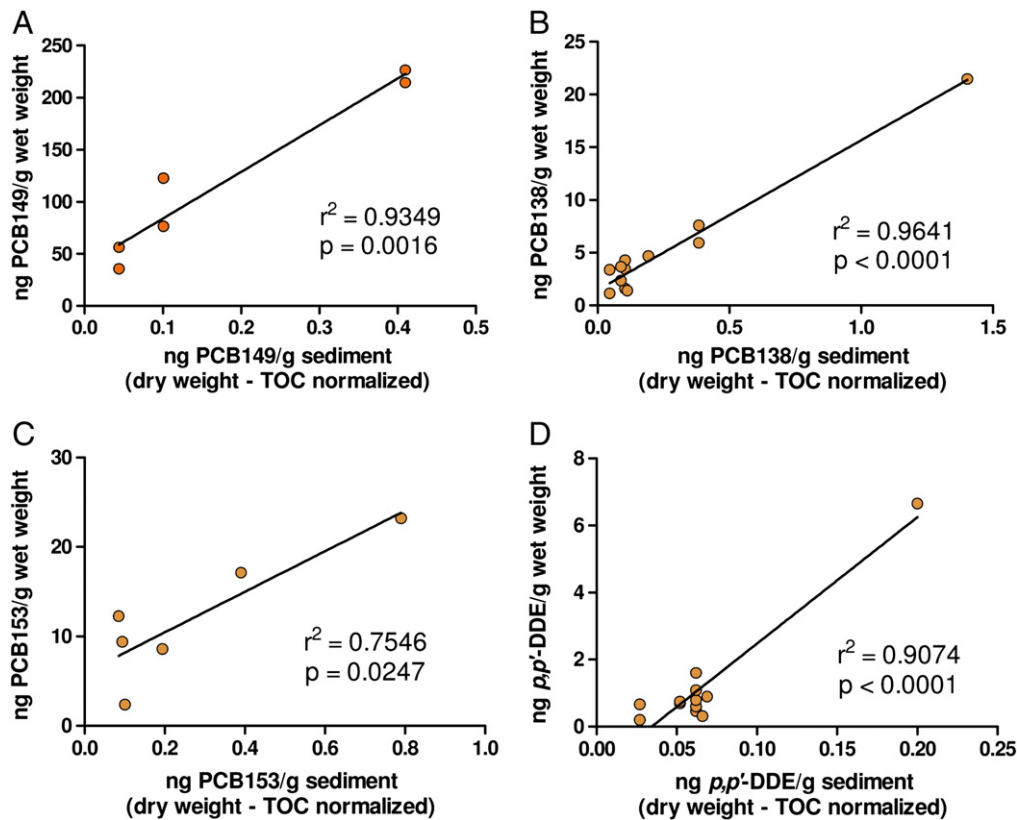
Fig. 5. Pearson's correlation between sediment loadings and tissue concentrations for A) CB 149 in blue mussel, B) CB 138 in shore crab, C) CB 153 in bristle worms and D) *p,p′*-DDE in shore crab.

concentrations of blue mussel were very high. Bivalves are known for their ability to concentrate lipophilic substances. They exhibit a low biotransformation capacity and as filter feeders, they achieve high filtration rates and accumulate chemicals from water, as well as from particulate material (Doerr and Liebezeit, 2009; Edgar et al., 2006). Because of these abilities, bivalves like *M. edulis* are widely used as biomonitors (Christensen and Platz, 2001; Johansson et al., 2006; Wepener et al., 2008).

Pearson correlation showed a significant, although not so strong correlation ($0.144 < r^2 < 0.529$) between lipid content and POP concentration in biota. It is previously stated that the lipid content of an organism has an influence on the accumulation of pollutants (Deribe et al., 2011; Gewurtz et al., 2006; Kidd et al., 1998; Waszak and Dabrowsak, 2009). In the present study the selected species did not vary much in lipid content, except for blue mussel, so the relationship is not that strong.

When tissue concentrations were compared on a lipid weight basis, the trends stayed the same as when compared on a wet weight basis (Fig. 4). The concentrations in brown shrimp were still the lowest. The Chinese mitten crab bioaccumulated the highest concentrations of PBDEs. However, on a lipid weight base, the concentrations in blue mussel and bristle worms did not differ significantly from the other species.

Compared to the results from other studies and regions, POP concentrations in biota from the present study were relatively high. PCB concentrations in European flounder from the present study were 10 times higher than concentrations measured in European flounder from the Gulf of Gdansk, Baltic Sea (up to 8.18 ng 7 ICES PCBs/g ww) (Napierska and Podolska, 2008). BDE47 concentrations in European flounders of France, were higher in the Seine estuary (0.42–4.15 ng/g ww), but lower in the Loire estuary (0.06–0.5 ng/g ww) (Bragigand et al., 2006), when compared to concentrations from this study (0.13–2.71 ng/g ww). Common sole from the Loire estuary and the North Sea contained comparable amounts of BDE47 (0.09–0.39 ng/g ww; 0.04–0.34 ng/g ww; in this study: 0.05–0.59 ng/g ww) (Bragigand et

al., 2006; Voorspoels et al., 2003). The blue mussels in this study contained very high PBDE and PCB levels when compared to literature (Table 3). Also POP concentrations in bristle worms were relatively high. PCB concentrations were at least ten times higher than bristle worms from the Gwangyang Bay in South Korea (average 215 ng/g lw for 78 PCB congeners; this study: 925–7858 ng/g lw for 33 PCB congeners) (Hong et al., 2011).

Table 4
Average BSAF of PCBs and PBDEs from the present study compared with BSAFs reported in other studies.

Species	BSAF PCBs	Other studies	BSAF PCBs
Bristle worm	1180	Bristle worms	0.01–2.68[a]
Blue mussel	1090	Blue mussel	0.5–2.5[a]
Brown shrimp	606		
Shore crab	2330	Blue crab	0.22–1.7[b]
Chinese mitten crab	2900		
Common sole	2340	Various fish	0.01–30[a]
European flounder	2370	White perch	0.34–1.5[b]
		Mummichog	0.16–1.03[b]

| Species	BSAF PBDEs	Other studies	BSAF PBDEs
Bristle worm	1090		
Blue mussel	304	Northern horse mussel	0.95–527[c]
Brown shrimp	162	Greasyback shrimp	up to 12.2[d]
Shore crab	598		
Chinese mitten crab	2520		
Common sole	1210	Northern pike	4.6–36.0[a]
European flounder	2010	Large yellow croaker	Up to 58[d]
		Bartail flathead	Up to 41[d]

[a] U.S. Army Corps of Engineers BSAF database (http://el.erdc.usace.army.mil/bsafnew/bsaf.html).
[b] Iannuzzi et al. (2011).
[c] deBruyn et al. (2009).
[d] Xiang et al. (2007).
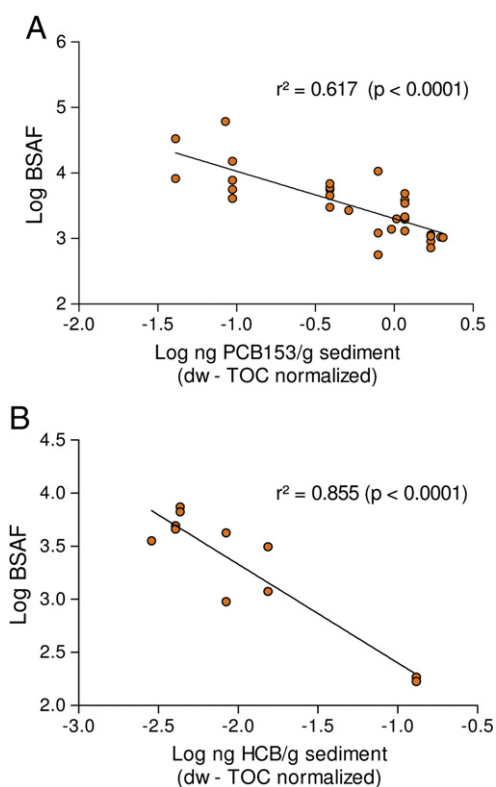
Fig. 6. Exposure concentration versus BSAF for A) CB 153 in flounder and B) HCB in Chinese mitten crab.

3.2.2. Geographical and temporal variation

For most of the PCB congeners, significant differences in lipid weight normalized concentrations between sampling locations were found in tissues of European flounder ($0.006 \leq p \leq 0.033$, concentrations higher in L5, Antwerp than in L2, Terneuzen), common sole ($p \leq 0.006$; lower concentrations in L1, Vlissingen), brown shrimp ($p < 0.001$, increasing concentrations going from L1, Vlissingen, to L4, Bath), blue mussel ($0.002 \leq p \leq 0.018$, lower concentrations in L1, Vlissingen) and bristle worms ($p < 0.001$, increasing concentrations going from L1, Vlissingen, to L5, Antwerp). Differences in the concentrations in time were only observed for some PCB congeners in common sole ($0.013 \leq p \leq 0.040$, lower concentrations were detected in September), brown shrimp ($p \leq 0.040$, lower concentrations were detected in September) and blue mussel ($0.002 \leq p \leq 0.011$, lower concentrations were detected in July). Since sole size was not smaller

Table 5
Comparison between the MRLs (ATSDR, 2010) and the mean concentrations of total PCBs, PBDEs, DDXs and HCB found in sole, flounder and blue mussel.

	PCB	PBDE	DDX	HCB
MRL (ng/kg/day)	30	7000	500	100
MRL (ng/day) for a person of 70 kg	2100	490,000	35,000	7000
Mean concentration in sole (*S. solea*) (ng/g)	54.9	0.48	1.70	n.d.
Maximum edible amount of sole per day (g) for a person of 70 kg	38.2	1,017,257	20,570	–
Mean concentration in flounder (*P. flesus*) (ng/g)	106	1.3	3.6	0.2
Maximum edible amount of flounder per day (g) for a person of 70 kg	19.8	390,865	9630	41,291
Mean concentration in mussel (*M. edulis*) (ng/g)	934	6.1	39.9	0.3
Maximum edible amount of mussel per day (g) for a person of 70 kg	2.2	79,953	878	21,261

n.d. — not detected.

in September, lower PCB concentrations were not correlated with fish size. Average surface water temperatures were 7.9, 18.6 and 16.8 °C for March, July and September, respectively. For the other measured compounds, differences between sampling locations or time were absent or statistically not significant.

3.3. Relationship between sediment loadings and tissue concentrations

Tissue concentrations and sediment loadings were poorly correlated. After TOC-normalization of the sediment concentrations, significant relationships were only found between the PCB concentrations in bristle worms, blue mussel and shore crab and the sediment concentrations at the same sampling location (Fig. 5). In common sole and brown shrimp, only some of the PCB congeners showed such correlation. For the other analyzed POPs, no relationships were found, except for p,p′-DDE in shore crab.

Multiple linear regressions showed that in most cases tissue concentrations did not depend on the sediment concentrations and/or environmental variables (TOC, conductivity, pH, clay content and tissue lipid content). However, in blue mussel, PCB and PBDE tissue concentrations could be predicted from a linear combination of sediment loading, tissue lipid content and clay content ($0.989 \leq r^2 \leq 0.996$) (log-transformed data). For PCB 153: [tissue] ng/g ww $= 0.992 + (0.442*$[sediment] ng/g dw$) + (2.184*\%$ lipid$) + (1.701*\%$ clay$)$. In common sole, the PCB concentrations in the tissues could be predicted from the sediment loading and the conductivity of the water ($0.703 \leq r^2 \leq 0.746$) (log-transformed data), where the conductivity is in function of the distance from the sea. For PCB 180: [tissue] ng/g ww $= 1.351 - (1.709*$[sediment] ng/g dw$) + (1.535*\%$ clay$) - (2.324*$conductivity (μS/cm)$)$.

Poor correlations between sediment and biota concentrations are possibly caused by the complexity of bioaccumulation processes and the variability in exposure. Next to the sediment, organisms get exposed through other media, like water, suspended material and their food. For species at low trophic levels, such as plankton and filter feeders, direct uptake of POPs from the environment (water, particulate matter) is the most important way of exposure. Dietary intake gets more important than direct absorption at higher trophic levels (Vallack et al., 1998). This may explain why the PCB concentrations in species at lower trophic levels in the present study (blue mussels and bristle worms) are better correlated with sediment loadings than concentrations in predators, such as European flounder. Moreover, blue mussel and bristle worm are less mobile species, so they will better reflect local conditions, while other species can move freely or even migrate (European flounder, common sole).

A simple empirical way to describe the relationship between chemical sediment concentrations and tissue concentrations of aquatic organisms is to express it by a specific factor, the biota–sediment accumulation factor (BSAF). BSAFs were calculated as the ratio of tissue concentration (lipid weight) to sediment concentration (dry weight) of a pollutant. The BSAFs calculated with TOC-normalized sediment concentrations are presented as Supplementary Information. TOC normalization did not change any of the relationships. If values were below the detection limit, LOQ*f was used to calculate BSAFs. The ranges of calculated BSAFs within species and locations were large. Compared with other field based studies, BSAFs were very high (Table 4), due to the low concentrations and many non-detects in the sediment.

To investigate the differences in bioaccumulation of different classes of chemicals, the BSAFs from CB 153, CB 180, BDE 47, p,p′-DDE and HCB were compared. In shore crab ($p < 0.0001$), blue mussel ($p < 0.0001$) and bristle worms ($p = 0.0041$), the BSAFs for CB 153 were significantly higher than the BSAFs from the other compounds. No other significant differences were indicated. European flounder bioaccumulated more BDE 47 and HCB from the sediment, when compared to common sole. BSAFs were not correlated with the octanol–water partition coefficient (log K_{ow}).

Because of the simplicity of the model, BSAFs are sometimes used to predict tissue concentrations from sediment concentrations (Iannuzzi et al., 2011). Regulatory agencies can use these values to set sediment quality guidelines for certain aquatic areas or for use in risk assessment. An important condition for the use of this concept is that the BSAFs should be stable with changing sediment concentrations and varying environmental factors (Bervoets et al., 2005). BSAFs are most accurate in steady-state systems, where the ratio between chemical concentrations in sediment and organism do not change remarkably over time (U.S. EPA, 2000). Under the field conditions in our study these conditions are not met since BSAFs were not independent of sediment concentrations but increased most of the time with decreasing exposure concentrations (Fig. 6).

In the present study, sediment was sampled on the shores and in the middle of the river. For some locations, large differences in concentrations between the shore samples and the samples from the middle of the river were found, but no constant trend could be indicated. This may have implications for the monitoring of POPs as well as for determination of BSAFs. To do this a representative sample is needed. In field studies, where local differences can be large, variability can pose a problem and questions can be raised in relation to the accuracy of field based BSAFs.

A single bioaccumulation factor or linear regression is insufficient to describe the relationship between sediment and biota concentrations (Iannuzzi et al., 2011). Therefore, the BSAF model is a poor predictor of bioaccumulation of micropollutants in aquatic ecosystems, and it should only be used as a first screening tool in risk assessment to separate sediments with low and high potential for bioaccumulation (Wong et al., 2001).

Because correlations and simple models are not accurate enough to predict possible bioaccumulated concentrations in certain situations, the use of in situ biomonitoring stays a more reliable method to get a clear image of bioaccumulation into biota (Belpaire et al., 2011; Bervoets et al., 2005). Also much more complex food web models are being developed. In these models more variables, such as species specific biotransformation and clearance rates, POP concentrations in prey species, pore water concentrations, assimilation efficiency (Bodiguel et al., 2009; Gobas and Arnot, 2010), are taken into account, which make them more accurate in predicting bioaccumulation.

3.4. Risk for human health

As fish and seafood accumulate POPs and are a common element in the diet of most individuals, they represent a major exposure route for persistent and bioaccumulative compounds from the aquatic systems into humans (Dorea, 2008). As mentioned above, some PCB values in blue mussel, shore crab and European flounder exceeded the European maximum limit for the sum of 6 ICES PCBs (75 ng/g ww) (European Commission, 2011). Since some of the currently investigated species are edible, a question can be raised about the health risks when consuming them. Therefore, the average POP concentrations found in common sole, European flounder and blue mussel in this study were compared to the minimum risk levels (MRLs) for oral intake of these compounds (ATSDR, 2010). The maximum amounts which are recommended to eat without risk for an average person of 70 kg, are calculated (Table 5). For PCBs, these amounts are very low, especially in mussels (2.2 g/day). Because of the high PCB levels, the MRLs are easily exceeded, as an average Belgian consumer above fifteen years consumes 23.9 g of fish or seafood a day (WIV, 2006). To exceed the MRL for DDXs, an average person must eat more than 878 g of these mussels per day. Other POPs, such as PBDEs, are less of a threat. Although the Scheldt is not a highly commercial fishing area, some fishing is going on. Moreover, local fishermen frequently collect fish in the estuary, so their fish consumption may exceed the average Belgian diet. Since exposure to POPs can possibly lead to adverse health effects, like endocrine disruption and respiratory diseases (Li et al., 2006; Sunyer et al., 2010), regular consumption of fish and seafood from the SE should be avoided.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2012.06.017.

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