



Effect of abiotic factors and environmental concentrations on the bioaccumulation of persistent organic and inorganic compounds to freshwater fish and mussels

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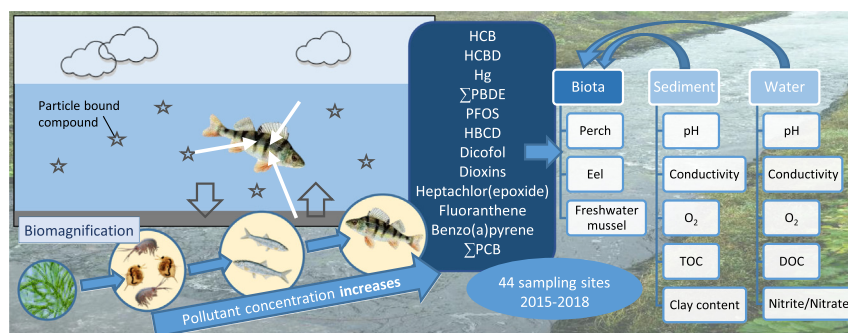
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HIGHLIGHTS

- Twelve persistent (in)organic compounds measured in biota, sediment and water.
- PFOS and benzo(a)pyrene water concentrations predicted biota concentrations.
- Organic content in sediment affected HCB, Σ PCB and Σ PBDE predicted fish concentrations.
- pH and nitrite negatively affected HCB and PFOS concentrations in eel respectively.
- Extrapolation between perch and eel was possible for Hg, Σ PBDE, PFOS, HBCD and Σ PCB.

GRAPHICAL ABSTRACT



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ABSTRACT

Many aquatic ecosystems are under persistent stress due to influxes of anthropogenic chemical pollutants. High concentrations can harm entire ecosystems and be toxic to humans. However, in case of highly hydrophobic compounds, their low water solubility precludes direct measurement in water, and thus alternative monitoring strategies are needed. In the present study, we investigated the extent to which bioaccumulated concentrations of persistent compounds can be predicted by concentrations in environmental compartments (water and sediment). Due to their high biomagnification potential, Hg and PFOS were included in this analysis as well. At 44 field locations in Flanders (Belgium), we monitored the concentrations of 11 priority compounds and their derivatives, included in the Water Framework Directive, in both sediment and water (where feasible) and biota (European perch, European eel and freshwater mussels). Besides, some sediment (i.e. total organic carbon (TOC) and clay content) and water characteristics were measured (i.e. pH, oxygen level, conductivity, nitrate, nitrite and dissolved organic carbon (DOC)). Measurements of HCB, HCBd, cis-heptachlorepoide, HBCD and PFOS in sediment and Σ PCB in water showed a lower detection frequency than in fish samples. While PCB profiles were comparable between all matrices, for PBDE clear differences were detected between sediment and fish profiles, with BDE99 contributing the most for sediment (34%) and BDE47 for fish ($\geq 44\%$), followed by BDE99 for

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perch (28%) and BDE100 for eel (25%). Water concentrations for PFOS and benzo(a)pyrene were predictive of respective bioaccumulated concentrations. HCB, \sum PCB and \sum PBDE, concentrations in fish were dependent on sediment concentrations and negatively related to organic compound levels ($p < 0.05$). Furthermore, pH and nitrite were negatively associated with accumulated concentrations in eel for HCB and PFOS, respectively ($p < 0.05$). Strong relationships between bioaccumulation and sediment and/or water concentrations strengthened the basis for surrogate monitoring methods. Finally, the extrapolation potential of Hg, \sum PBDE, PFOS, HBCD and \sum PCB between both fish species offered new opportunities in extrapolating different European monitoring frameworks.

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1. Introduction

Persistent organic pollutants (POPs) and metals may harm entire aquatic ecosystems due to losses of habitat and biodiversity and can cause chronic or acute toxicity to aquatic organisms (European Commission, 2008). Originating from various anthropogenic activities (i.e. industry, agriculture, side products of combustion), pollutants may end up in the environment via discharge, leaching, erosion and atmospheric deposition (Schweitzer and Noblet, 2018). Although the use and production of many of the pollutants have declined substantially over the last decades, due to the Stockholm Convention, historical contamination is still omnipresent in the aquatic environment (Belpaire and Goemans, 2007b; Maes et al., 2008).

Hydrophobic organic compounds (HOCs) will not easily dissolve in the water and therefore are not to be measured so straightforwardly in the water phase (Belpaire and Goemans, 2007b; Jürgens et al., 2013). Additionally, weak or non-existent relationships between environmental concentrations and accumulated levels in aquatic organisms can lead to an underestimation of the risk. Therefore, monitoring water or sediment does not guarantee sufficient protection of the aquatic environment (Weltens et al., 2002). Whereas concentrations of these pollutants in abiotic compartments, especially in the water column, are often below the detection limit, they are still ubiquitous and easily detectable in biota (Belpaire et al., 2008; Weltens et al., 2002). Mercury and PFOS (perfluorooctanesulfonic acid), on the other hand, are known to have a high affinity for proteins and are less hydrophobic (Amlund et al., 2007; Zhong et al., 2019). However, through their chemical characteristics, HOCs, mercury and PFOS accumulate and biomagnify through the food chain, eventually reaching high, harmful concentrations in top predators and potentially being toxic to humans via consumption (European Commission, 2013; Lavoie et al., 2013; Van Ael et al., 2013; Wu et al., 2009).

Both water variables (e.g. oxygen content, pH, conductivity, dissolved organic carbon (DOC)) and sediment characteristics (e.g. clay content, total organic carbon (TOC)) can influence the bioavailability of pollutants. High DOC or TOC levels might result in higher organic complexation reducing the bioavailability of lipophilic compounds (Dittman and Driscoll, 2009; Moeckel et al., 2014; Li et al., 2015). Salinity and pH are known factors to influence chemical speciation of metals and organic compounds and induce structural and morphological modifications in organisms, affecting bioavailability and accumulation efficiency of these compounds (Dittman and Driscoll, 2009; Spry and Wiener, 1991). Furthermore, high environmental acidity reduces biodiversity and the general quality of the ecosystem (Driscoll et al., 2001; Watras et al., 1998).

In the present study, European perch (*Perca fluviatilis*) and European eel (*Anguilla anguilla*) in its yellow eel stage were selected as suitable monitoring species. They fulfil the essential monitoring purpose requirements (Belpaire and Goemans, 2007a; Teunen et al., 2020). Both species accumulate high HOCs concentrations because of their high position in the aquatic food chain. Furthermore, they are known resident species with a broad habitat range because of low sensitivity to environmental pollution and poor water quality. Their restricted home range allows for local contamination monitoring (Ovidio et al., 2013). Usually,

eel concentrations are among the highest recorded in freshwater biota because of their high lipid content and bottom-dwelling lifestyle (Belpaire and Goemans, 2007a; Palstra et al., 2006). Because of the absence of a reproductive cycle during its juvenile yellow eel stage, seasonal fluctuation in accumulation patterns is limited (Belpaire and Goemans, 2007a).

Concerning polycyclic aromatic hydrocarbons (PAHs), their high elimination rate in fish, however, might underestimate the accumulated concentrations present in the food chain (European Commission, 2013). Hence, PAHs are recommended to be measured in bivalves or crustaceans instead (European Commission, 2013). Active biomonitoring, using translocated individuals, often has been used for monitoring bioaccumulative pollutants (Babut et al., 2020; Catteau et al., 2021). This standardized sampling technique is based on the exposure of a particular species - with controlled low background concentrations and sizes or conditions - to different sampling locations, reflecting the local pollution load. The *Dreissena* bivalve genus has often been used for this purpose (Bashnin et al., 2019; Teunen et al., 2021; Potet et al., 2018).

The effect of general abiotic factors, such as DOC/TOC, on bioavailability and thus bioaccumulation of lipophilic compounds has been studied in the past (Dittman and Driscoll, 2009; Moeckel et al., 2014). Contrastingly, to the best of our knowledge, no detailed studies looking into the effects of numerous abiotic factors and environmental concentrations on bioaccumulation of a large set of priority hydrophobic organic compounds on a vast collection of sample locations have ever been performed. In Flanders (Belgium), an extensive physicochemical and biota monitoring network allowed us to investigate the influence of environmental concentrations and characteristics on the accumulation of HOCs in the aquatic food chain - mainly focussing on the highest concentrations reached in top predators through biomagnification - in a very broad range of aquatic ecosystems. Our study's general innovative aspect is evaluating the relationship between environmental and accumulated concentrations of a large group of hydrophobic priority compounds over a vast number of sample locations with varying backgrounds and contamination levels, taking into account water quality parameters and sediment characteristics. All priority hydrophobic organic compounds (HOCs) that are of interest for monitoring in fish (and mussels), according to the Water Framework Directive (WFD), were included in the present study. Furthermore, PCBs (polychlorinated biphenyls) were included due to their highly lipophilic properties and high accumulation in predatory fish (Belpaire and Goemans, 2007b; Masset et al., 2019).

To determine the bioavailability and potential toxicity of hydrophobic organic compounds to aquatic organisms, it is imperative to identify the most relevant sample matrix to avoid over- or underestimation of environmental quality and potential human health risk assessment. The primary purpose of the present study was (1) to identify the major environmental exposure paths that affect the bioavailability of hydrophobic pollutants by relating concentrations in biota to sediment and water concentrations; (2) to evaluate the role of abiotic characteristics of water and sediment to the bioaccumulation of these pollutants and (3) to compare PCB and PBDE (polybrominated diphenyl ether) profiles between biota, water and sediment.

2. Material and methods

2.1. Sample location and target species

A total of 44 sampling locations were selected in Flanders (Belgium), reflecting extensive water body types (canals, rivers and streams) from fresh and brackish water environments (Fig. 1; Table SI-1).

2.1.1. Fish

European eel (*A. anguilla*) in its sedentary 'yellow eel' stage and European perch (*Perca fluviatilis*) were collected from the different sites between 2015 and 2018. In total, 132 eels and 515 perches were caught using electrofishing (Deka 7000 or Deka 3000) and fyke fishing. The specific fishing method was dependent on the water body type. For a more detailed description, we refer to Belpaire et al. (2000). Fish were identified on the field, measured (total length) and weighed. Furthermore, they were sacrificed using MS-222 (Acros Organics, Geel, Belgium) and frozen for transport. A mean length between 45 and 55 cm of juvenile yellow eel was targeted. However, this was not possible for all locations. In order to have sufficient muscle tissue for analysis, the largest perches were collected. Unfortunately, it was not possible to catch both species at every sample location.

2.1.2. Mussels

Freshwater mussels (zebra mussel *Dreissena polymorpha* or quagga mussel *Dreissena bugensis*) were collected from reference sites. An alternative species, Asiatic clams (*Corbicula fluminea*), was exposed in locations with high salinity (mean EC20: >2.4 mS cm⁻¹), since *Dreissena* sp. cannot survive these brackish waters. Zebra mussels were collected from the recreational lake the Blaarmeerse in Ghent (2015) and at the drinking water reservoir of the Antwerp Drinking Water Company (Waterlink) in Duffel (2016). Since zebra mussel stocks decreased, quagga mussels were used from 2017. Simultaneous exposure of both species showed a minimal variation in accumulated concentrations of benzo(a)pyrene, while fluoranthene showed a larger variation up to a factor 2 (Teunen et al., 2020). Quagga mussels were collected from the recreational lake the Nekker in Mechelen (2017 and 2018). Finally, Asiatic clams were collected from the recreational lake the Blaarmeerse in Ghent (2015–2018). Low background concentrations of organic micropollutants (PCBs, PBDEs, organochlorine pesticides and metals) were measured in mussels from these reference locations (Bervoets et al., 2005).

The mussels were acclimated to the current environmental temperature in a semi-natural pond (mesocosm structure, University of

Antwerp, Belgium; dechlorinated tap water), at least two weeks prior to exposure. Background concentrations were monitored on a subset of 5–10 mussels (Table SI-4). The exposure took place during autumn and winter, as to reduce the risk of spreading these exotic species in the sampling locations since mussel reproduction is reduced at water temperatures below 12 °C (Wong et al., 2012).

The mussels were exposed for six weeks to each of the sampling locations. The set-up existed of two polyethylene cages, each consisting of two attached pond baskets (11x11x22 cm; mesh size of 2 × 4 mm), positioned 1 m below the water surface, allowing free water circulation (Bashnin et al., 2019; Bervoets et al., 2005). Per location, a total of 70–75 and 25–30 individuals were exposed of *Dreissena* sp. and *Corbicula fluminea*, respectively. The cages were attached to bridges or solid structures on the river banks using metal chains and locks. After recollection, mussels were depurated for at least 15 h in particle-free water from the respective sampling site at 15–20 °C.

2.2. Sample preparation

Fish were again weighed (Sartorius CP4202S, 0.01 g accuracy, Göttingen, Germany) and measured (total length, 1 mm accuracy) before dissection. Muscle tissue was dissected from the fish over the entire length of the body. Per location, a maximum of 20 perch and 3 eels were targeted and pooled per species (Table SI-1). A total of 50 g per pool was needed to be able to perform all the analyses. Pools were homogenised in 50 mL polypropylene tubes using a stainless steel kitchen mixer (Bosch, MSM65PER) and stored at -20 °C before analysis.

The exposed mussels were dissected and pooled per location. Pools were homogenised in 50 mL polypropylene tubes using a Qiagen TissueRuptor (Qiagen, Hilden, Germany) and stored at -20 °C before analysis.

2.3. Analysis of biota samples

The analytical methods are reported in detail in the Supplementary Information, including quality assurance/control. The compounds measured in biota were hexachlorobenzene (HCB), hexachlorobutadiene (HCBd), mercury (Hg), PBDEs, PFOS, hexabromocyclododecane (HBCD), dicofol, dioxins, heptachlor, trans- and cis- heptachlorepoxide, PCBs, benzo(a)pyrene and fluoranthene.

In the present study, PCB was considered as the sum of congeners PCB28, PCB52, PCB101, PCB118, PCB138, PCB153 and PCB180 (PCB ICES 7; further referred to as \sum PCB) and PBDE as the sum of BDE28,

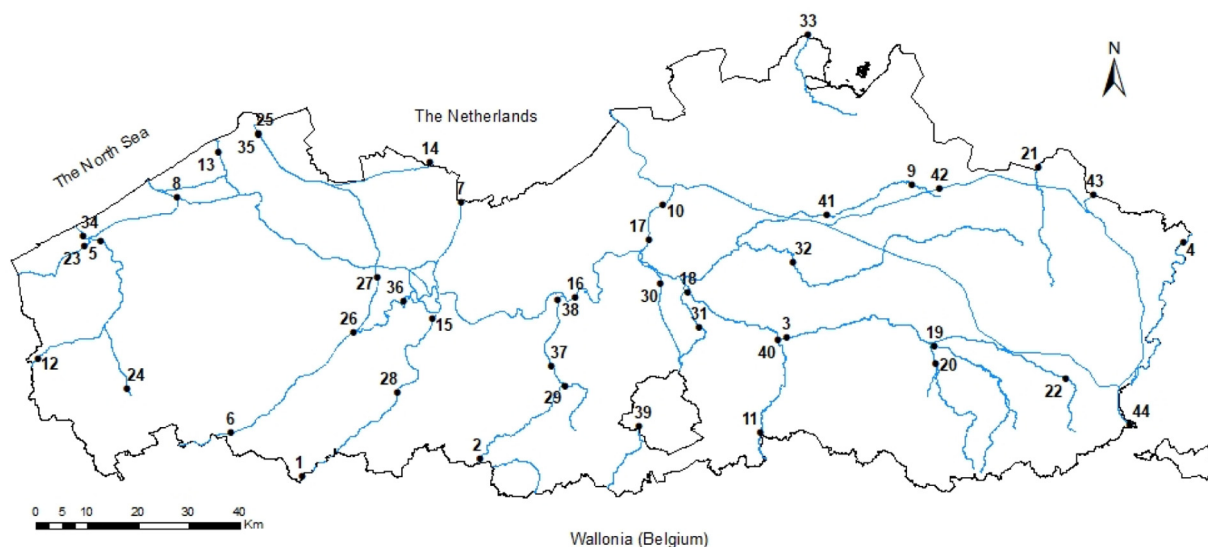


Fig. 1. Map of Flanders (northern part of Belgium) with 44 sampling locations (2015–2018). Detailed information on sample points are indicated in Table SI-1.

BDE47, BDE99, BDE100, BDE153 and BDE154 (PBDE ICES 6; further referred to as \sum PBDE). Furthermore, HBCD was calculated as the sum of α -, β - and γ -HBCD. If at least one of the congeners showed a detectable concentration, a value of $\frac{1}{2}$ LOQ was used for the congeners with concentrations <LOQ (Bervoets et al., 2004; Custer et al., 2000). Dioxins were calculated as the sum of polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofuranes (PCDFs) and dioxin-like polychlorobiphenyls (PCB-DL).

2.4. Analysis of water and sediment samples

The same compounds as analysed in fish were measured in water and sediment at the same locations between 2009 and 2019. These data were provided by the Flanders Environment Agency (<http://geoloket.vmm.be/Geoviews/>) and were available as part of their routine monitoring network. This is a licensed laboratory (as specified in the Compendium for Water sampling, measurements and Analysis (WAC)) holding a BELAC accreditation to ISO/IEC 17025 for environmental monitoring (including water and sediment) of organic compounds and metals. Furthermore, physical and chemical characteristics of water were also recorded by the Flanders Environment Agency (oxygen content, conductivity, pH, nitrate, nitrite and dissolved organic carbon (DOC)) and taken into account as predictive water variables. As for sediment, oxygen content, conductivity, pH, total organic carbon (TOC) and clay content were included. Water samples were collected and measured monthly, while for sediment, this was done yearly. To account for the effect of seasonal fluctuation, measurements of all (abiotic) environmental characteristics and concentrations were calculated as geometric means per location. No measurements were available for PBDE, HBCD, dicofol and dioxins in water. Heptachlor and dioxins data are lacking for sediment. To compare environmental concentrations and physico-chemical characteristics with accumulation of PAHs in mussels, calculations were performed on samples taken within the year of exposure were used for sediment parameters and on samples taken during the exposure period or one year difference within the same season for water parameters. This approach resulted in an adjustment of all environmental data to the short-term exposure of the mussels.

2.5. Statistics

Statistical analyses were performed using the software package R (R version 4.0.4; R Core Team, 2021). A Spearman correlation was performed between the abiotic parameters of water and sediment. Further statistical analysis was only performed for compounds and matrices with at least 50% of measurements above the detection limit. For each of the measured compounds, multiple regression models were constructed to establish the links between the concentration of compounds in biota and in sediment or water, as well as the influence of abiotic factors. Using stepwise elimination of non-significant factors, a model with factors contributing to accumulation in biota was identified. Because of the skewed nature of concentration data in each compartment, they were transformed using the logarithmic function. For testing extrapolation possibilities between both fish species, a linear regression model was constructed. With this the potential to predict the accumulated concentration in one species by analysing the other one was investigated. A Kruskal-Wallis test was used to compare results for accumulated concentrations in mussels, because different species and populations were used for exposure between sample years. Significant outliers were removed using the Grubbs test in Graphpad and adjusted datasets were used for statistics and figures. Significance levels were interpreted at a p -value < 0.05.

3. Results

3.1. Results and detection frequency in different matrices

Measurements of both pollutants and characteristics in biota, sediment and water column are displayed in Table 1 and Table SI-2

to SI-4. For a standardized comparison between different matrices, biota as well as sediment concentrations were displayed per dry weight (dw).

The percentage of quantifiable concentrations (>LOQ) was determined for biota (pooled per location and per species), water and sediment (Table 2). For abiotic measurements, a geometric mean was determined per location and matrix for each compound and characteristic. Only locations with all measurements below the detection limit were scored as <LOQ (Table SI-6). For some compounds in specific matrices, a significant number of sample locations resulted in values below LOQ. This was the case for HCB in water (90%) and sediment (50%), PFOS in sediment (49%), HBCD in sediment (100%) and \sum PCB in water (67%). Hg concentrations in water showed a large seasonal fluctuation, with 48% of all individual measurements being <LOQ. This was also the case for PAHs, to a lesser extent. HCB, heptachlor and trans-heptachlorepoxyde had a very low detection frequency in all tested matrices. Cis-heptachlorepoxyde was easily detected in eel samples only. On the other hand, only 24% of sediment samples had dicofol concentrations above the LOQ, in contrast to biota samples (0%).

The correlation tests showed a relation between O_2 ($r^2 = 0.42$; $p < 0.05$), pH ($r^2 = 0.73$; $p < 0.001$) and conductivity (EC20; $r^2 = 0.99$; $p < 0.001$) measured in water and sediment (Table SI-8). Furthermore a strong correlation was found between pH_{sediment} and clay content ($r^2 = 0.42$; $p < 0.05$), between TOC and clay content ($r^2 = 0.74$; $p < 0.001$), between $O_{2, \text{water}}$ and pH_{water} ($r^2 = 0.49$; $p < 0.001$), between $O_{2, \text{water}}$ and nitrite ($r^2 = -0.64$; $p < 0.001$), between $EC20_{\text{water}}$ and nitrate ($r^2 = -0.31$; $p < 0.05$), between $EC20_{\text{water}}$ and nitrite ($r^2 = -0.32$; $p < 0.05$), between nitrate and nitrite ($r^2 = 0.56$; $p < 0.001$) and between nitrate and DOC ($r^2 = 0.48$; $p < 0.001$).

3.2. PCB and PBDE profiles

All measured matrices showed the highest concentrations for congeners PCB153, PCB138 and PCB180. In general, profiles of bioaccumulated and environmental concentrations were very comparable (Fig. 2). Small differences could be detected in the contribution of lower halogenated congeners to the \sum PCB (water: PCB153 > PCB138 > PCB180 > PCB101/52/28 > PCB118; sediment: PCB153 > PCB138 > PCB180 > PCB101 > PCB 118 > PCB 52 > PCB28; perch: PCB 153 > PCB138 > PCB180 > PCB 101 > PCB 52 > PCB 118 > PCB 28; Eel: PCB153 > PCB138 > PCB180 > PCB 118 > PCB 101 > PCB 52 > PCB 28).

For both perch and eel, BDE47 was the main BDE congener (Fig. 2). In sediment the highest concentrations were measured for BDE 99. Furthermore, a large variation existed for PBDE profiles between matrices (Sediment: BDE99 > BDE47 > BDE153 > BDE100 > BDE28 > BDE154; Perch: BDE47 > BDE99 > BDE100 > BDE154 > BDE153 > BDE28; Eel: BDE47 > BDE100 > BDE154 > BDE153 > BDE99 > BDE28).

3.3. Relationship between environmental and accumulated concentrations

Further statistical analyses and interpretations were only performed on compounds with more than 50% of bioaccumulated and environmental concentrations above LOQ. This included HCB (in eel), Hg, \sum PBDE, PFOS and \sum PCB (measured in fish) and benzo(a)pyrene and fluoranthene (measured in mussels).

Based on the correlation test results, Eq. (1) was used in a multiple regression model. Stepwise deletion was performed until all non-significant factors were removed. In the case of logic correlations - such as O_2 , pH and conductivity in water and sediment and nitrate and nitrite - one of both variables was used in the multiple regression models. Water parameters were used for the characteristics measured in both abiotic matrices, since a more extensive dataset was available for water.

$$\log(\text{conc}_{\text{biota}}) = \log(\text{conc}_{\text{water}}) + \log(\text{conc}_{\text{sediment}}) + O_2 + pH + EC20 + TOC + clay + nitrite + DOC \quad (1)$$

Table 1

Ranges (and median) of measured concentrations in biota ($\mu\text{g kg}^{-1}$ dw), sediment ($\mu\text{g kg}^{-1}$ dw) and water (ng L^{-1}). Abiotic parameters are included such as pH (-), O_2 (mg L^{-1}), conductivity (EC20; $\mu\text{S cm}^{-1}$), TOC (g C kg^{-1} dw), DOC (mg C L^{-1}), Clay (%), nitrate (mg N L^{-1}) and nitrite (mg N L^{-1}).

Parameter	<i>Perca fluviatilis</i>	<i>Anguilla anguilla</i>	Sediment	Water
HCB	<LOQ-2.5 (0.24)	0.48-33 (9.1)	<LOQ-1.4 (0.18)	<LOQ-1.3 (1.25)
HCBD	<LOQ-4.0 (1.3)	<LOQ-8.4 (<LOQ)	<LOQ-2.7 (0.5)	<LOQ
Hg	160-735 (286)	83-1526 (407)	21-2404 (94)	6.4-65 (17)
Σ PBDE	<LOQ-8.4 (3.4)	0.90-285 (22)	<LOQ-8.3 (0.74)	-
PFOS	13-270 (48)	4.4-220 (27)	<LOQ-8.0 (0.25)	0.8-14 (2.7)
HBCD	<LOQ-4.7 (1.3)	<LOQ-1106 (25)	<LOQ	-
Dicofol	<LOQ	<LOQ	<LOQ-19 (2.5)	-
Dioxins ^a	0.002-0.020 (0.007)	0.006-0.103 (0.03)	-	-
Heptachlor	<LOQ	<LOQ	-	<LOQ
tHpChlepx	<LOQ	<LOQ	<LOQ	<LOQ
cHpChlepx	<LOQ-7.8 (0.60)	<LOQ-46 (0.76)	<LOQ	<LOQ
Σ PCB	3.5-700 (100)	25-4001 (1277)	<LOQ-318 (8.0)	<LOQ-9.4 (4.8)
Benzo(a)pyrene ^b	<LOQ-270 (23)	NA	<LOQ-2800 (100)	<LOQ-40 (2.6)
Fluoranthene ^b	<LOQ-1073 (169)	NA	<LOQ-7500 (250)	<LOQ-73 (11)
pH	-	-	6.8-9.1 (7.9)	7.1-8.3 (7.9)
O_2	-	-	2.8-13 (8.9)	4.4-11 (8.5)
EC20	-	-	254-10,770 (848)	339-16,233 (832)
TOC	-	-	1.2-50 (13)	-
DOC	-	-	-	3.7-14 (6.8)
Clay (%)	-	-	1.7-36 (6.9)	-
Nitrate	-	-	-	0.4-6.5 (3.1)
Nitrite	-	-	-	0.01-0.3 (0.09)

^a Concentrations in $\mu\text{g WHO-TEQ}_{2005} \text{ kg}^{-1}$ dw.

^b Measured in freshwater mussels instead of fish. LOQs are indicated in Table SI-9. tHpChlepx: trans-heptachlorepoixide; cHpChlepx: cis-heptachlorepoixide.

Where $\text{conc}_{\text{biota}}$ is the bioaccumulated concentration of a specific compound in biota ($\mu\text{g kg}^{-1}$ dw), $\text{conc}_{\text{water}}$ is the concentration of the same compound measured in the water column (ng L^{-1}), $\text{conc}_{\text{sediment}}$ is the concentration of that compound measured in the sediment ($\mu\text{g kg}^{-1}$ dw). Furthermore, parameters added to the multiple regression models were O_2 (oxygen content; $\text{mg O}_2 \text{ L}^{-1}$ water), pH, EC20 (electrical conductivity at 20 °C; $\mu\text{S cm}^{-1}$), TOC (g C kg^{-1} dw in sediment), clay content (%), nitrite concentration (mg N L^{-1}) and DOC (mg C L^{-1}).

In these multiple regression models, accumulated Σ PBDE and Σ PCB concentrations in biota showed a positive relation with concentrations in the sediment ($p \leq 0.003$; Table 3, Fig. 3; Table SI-10). For PFOS, concentrations in fish could be related to water concentrations ($p \leq 0.002$). The same was true for benzo(a)pyrene concentrations in mussels and water ($p < 0.001$). Furthermore, DOC or TOC contributed significantly to the described relationship between sediment and both fish species for Σ PBDE (≤ 0.012) and Σ PCB ($p < 0.001$), and for eel in HCB ($p = 0.003$). Nitrite concentration ($p = 0.028$) and magnitude of conductivity ($p = 0.012$) contributed to the relationship for PFOS in eel and pH ($p = 0.036$) for HCB in eel. These abiotic characteristics

contributed negatively to the relationship between concentrations in the environment (water or sediment) and the bioaccumulated concentrations. However, the effect of conductivity on PFOS concentrations in eel was minimal (slope of 0.0001), and therefore not included in the graphs of Fig. 3. For Hg and fluoranthene, no significant ($p > 0.05$) relationships with bioaccumulated concentrations could be identified for abiotic parameters or environmental concentrations.

3.4. Extrapolation between fish species

Linear regression models were used to test for extrapolation potential between accumulated concentrations in both fish species (Table 4, Table SI-11). These analyses were only performed when for both species, the detection exceeded 50%. Despite the 100% detection in both perch and eel, no statistical analyses could be done for dioxins since only one species was analysed per location. We identified an equation to extrapolate concentrations between perch and eel for all other included compounds. Furthermore, it was clear that the highest concentrations were mainly measured in eel, especially in a location with a

Table 2

Percentages of measured locations with concentrations >LOQ of hydrophobic compounds in biota (perch and eel), water and sediment and the number of measurements (N) and sample locations (n).

Compound	Perch (2015-2018)			Eel (2015-2018)			Sediment (2009-2019)			Water (2009-2019)		
	% > LOQ	N	n	% > LOQ	N	n	% > LOQ	N	n	% > LOQ	N	n
HCB	42	65	33	100	67	41	50	148	44	10	1467	30
HCBD	6.1	65	33	2.4	67	41	4.5	86	43	0	72	2
Hg	100	65	33	100	67	41	100	151	44	100^a	2283	42
Σ PBDE	85	65	33	100	67	41	98	149	44	NA	NA	NA
PFOS	100	65	33	100	67	41	51	56	41	100	302	26
HBCD	61	65	33	95	67	41	0	55	42	NA	NA	NA
Dicofol	0	40	29	0	26	24	24	44	38	NA	NA	NA
Dioxins	100	28	28	100	16	16	NA	NA	NA	NA	NA	NA
heptachlor	0	65	33	0	67	41	NA	NA	NA	0	1450	30
tHpChlepx	0	65	33	0	67	41	0	45	31	0	7	3
cHpChlepx	21	65	33	93	67	41	0	45	31	0	1461	30
Σ PCB	100	65	33	100	67	41	98	133	44	33	1073	27
Benzo(a)pyrene ^b	86	2369	43	NA	NA	NA	89	150	44	100^a	2015	28
Fluoranthene ^b	98	2369	43	NA	NA	NA	84	150	44	100^a	2014	28

^a Large seasonal variation results in 52%, 73% and 74% of all measurements > LOQ for Hg, benzo(a)pyrene and fluoranthene respectively.

^b Compounds measured in mussels instead of fish. Percentages in bold contain at least 50% of measurements > LOQ.

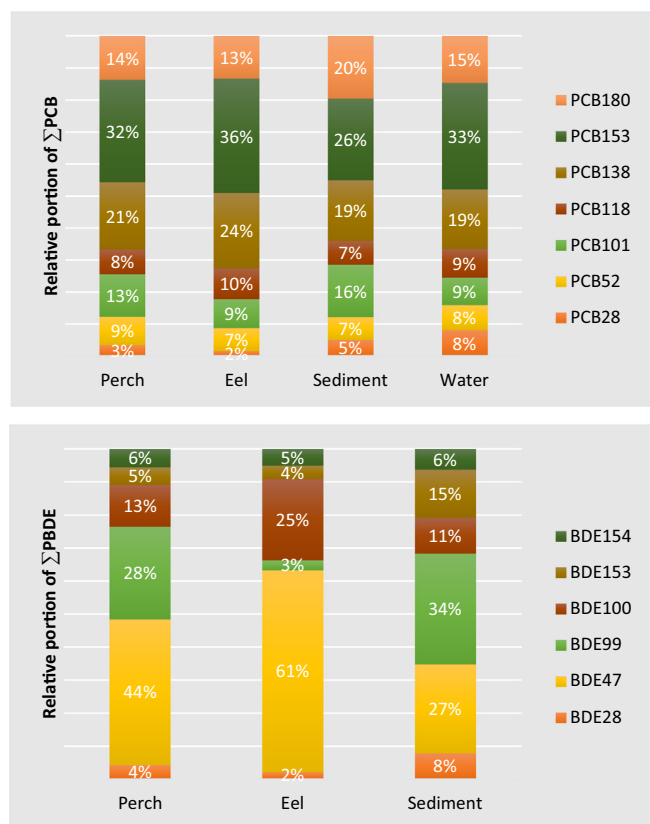


Fig. 2. Profiles of PCBs (UPPER) and PBDEs (LOWER) contributions to Σ PCB and Σ PBDE in water (2009-2019), sediment (2009-2019), perch and eel (2015-2018).

lower pollution loading (intercept >1). However, for PFOS, Hg and Σ PCB, the difference between both species decreased at highly polluted areas (slope < 1), potentially with higher accumulated concentrations in perch.

4. Discussion

4.1. Accumulated concentrations in biota

For comparison with literature, biota concentrations were also reported per wet weight (ww; Table 5). These calculations were performed using dry/wet weight ratios which were determined for each sample (Table SI-1 and SI-4). Results on bioaccumulated concentrations

Table 3

Significant ($p < 0.05$) equations for HCB, PFOS, Hg, Σ PBDE, Σ PCB, benzo(a)pyrene and fluoranthene after stepwise deletion in multiple regression models. Significance levels of the independent parameters are indicated with letters: ^A $p < 0.05$, ^B $p < 0.01$, ^C $p < 0.001$.

Compound	Significant equation	R ² (DF)
HCB	NA	NA
PFOS	$\text{Log}(\text{eel}) = 11.62 + 0.53 \cdot \text{log}(\text{sediment})^B - 0.04 \cdot \text{TOC}^B - 1.01 \cdot \text{pH}^A$	0.29 (37)
	$\text{Log}(\text{perch}) = 3.14 + 0.80 \cdot \text{log}(\text{water})^C$	0.41 (20)
Hg	$\text{Log}(\text{eel}) = 3.33 + 0.59 \cdot \text{log}(\text{water})^B - 0.0001 \cdot \text{conductivity}^A - 6.04 \cdot \text{nitrite}^A$	0.38 (21)
	NS	NS
Σ PBDE	$\text{Log}(\text{perch}) = 2.16 + 0.29 \cdot \text{log}(\text{sediment})^B - 0.15 \cdot \text{DOC}^B$	0.48 (28)
	$\text{Log}(\text{eel}) = 5.03 + 0.48 \cdot \text{log}(\text{sediment})^C - 0.06 \cdot \text{clay}^C - 0.16 \cdot \text{DOC}^A$	0.53 (36)
Σ PCB	$\text{Log}(\text{perch}) = 5.32 + 0.40 \cdot \text{log}(\text{sediment})^B - 0.29 \cdot \text{DOC}^C$	0.59 (27)
	$\text{Log}(\text{eel}) = 6.30 + 0.71 \cdot \text{log}(\text{sediment})^C - 0.08 \cdot \text{TOC}^C$	0.53 (37)
Benzo(a)pyrene	$\text{Log}(\text{mussel}) = 2.28 + 0.58 \cdot \text{log}(\text{water})^C$	0.48 (20)
	$\text{Log}(\text{Dreissena}) = 2.61 + 0.45 \cdot \text{log}(\text{water})^B$	0.37 (16)
Fluoranthene	NS	NS
	NS	NS

NS: not significant. NA: insufficient data above LOQ to perform statistics. Mussel includes a combination of all exposed mussels; Dreissena refers to both *Dreissena polymorpha* and *Dreissena bugensis*.

reported in this study are in line with reported ranges of HOCs in Flemish and European monitoring studies of perch and eel in freshwater systems. Measured concentrations of flame retardants (HBCD and PBDEs) seemed to be remarkably lower in eel and perch from Italy (Tavoloni et al., 2021) and eel from Poland (Szlinder-richert et al., 2014) compared to the present study. Accumulated concentrations of PFOS were lower in eel from Italy (Giari et al., 2015) and perch from Sweden (Åkerblom et al., 2017). Also, Σ PCB concentrations in fish from Belgium (including the present study) showed to be often higher compared to other European countries (Blanchet-letrouvé et al., 2014; Ferrante et al., 2010; Flidner et al., 2018; Mchugh et al., 2010; Szlinder-richert et al., 2014, 2010). Finally, mercury concentrations in perch from Scandinavia were much higher than those measured in the present study (Miller et al., 2013; Negm, 2015; Sonesten, 2002). Variation in accumulation patterns and concentrations between countries, on the other hand, was to be expected to a certain level due to different pollution sources (e.g. point sources, atmospheric deposition). Furthermore, the year of sampling can be an important factor influencing results, since most of the target compounds have been banned or restricted over the past decades. Finally, due to the biomagnifying nature of these compounds, age and therefore the duration of exposure, usually shows a positive relationship with accumulated concentrations (Gewurtz et al., 2011). Furthermore, the reproductive stage is also considered during the spawning of lipid-rich eggs by females (Weis and Ashley, 2007). This could result in a lower accumulated concentration in females during the reproductive season. In the present study, however, this could only be the case for perch, since eel were collected in their juvenile, non-reproductive yellow eel phase.

Accumulated fluoranthene concentrations in mussels from the present study (< 5 – $107 \mu\text{g kg}^{-1}$ ww; median: $22 \mu\text{g kg}^{-1}$ ww) were higher than concentrations measured in zebra mussels from a pilot study in Flanders (9.9 – $10 \mu\text{g kg}^{-1}$ ww; median: $10 \mu\text{g kg}^{-1}$ ww; De Jonge et al., 2014), but lower than those previously measured in zebra mussels from the Netherlands (33 – $250 \mu\text{g kg}^{-1}$ ww; Hendriks et al., 1998). Although the pilot study and the present study partly covered the same sampling locations, differences in accumulated PAH concentrations might be due to the small number of locations ($N = 2$) in the pilot study, rather than differences in environmental concentrations and exposure. It should also be taken into account that in the present study, mussels collected from the drinking water reservoir, which were exposed in 2016 (Table SI-4), showed higher background fluoranthene concentrations ($21 \mu\text{g kg}^{-1}$ ww) than mussels from other reference locations ($< 5 \mu\text{g kg}^{-1}$ ww). This might overestimate the local pollution load. Exposed mussels from 2016, however, showed accumulated concentrations lower than the background concentrations at some locations,

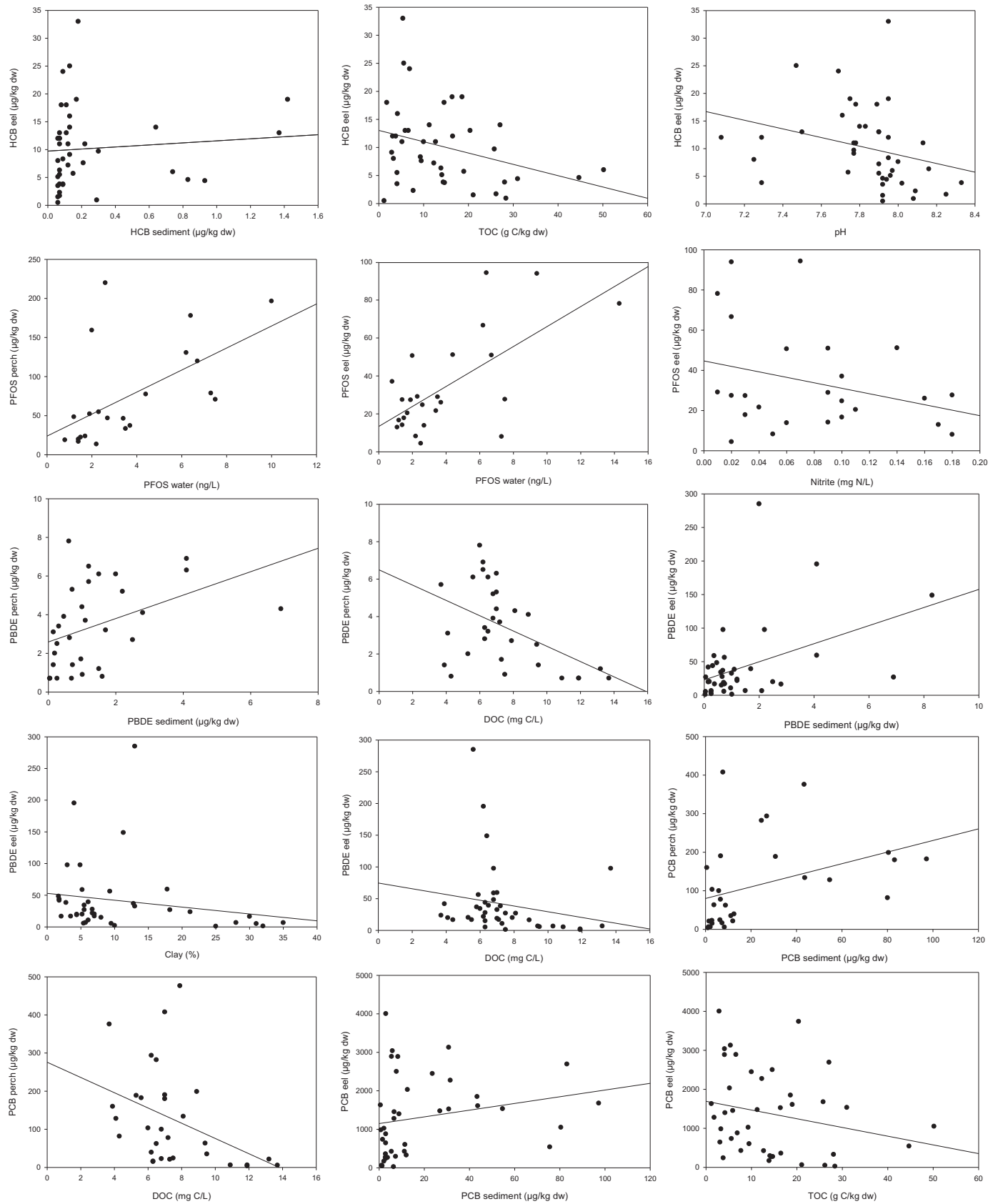


Fig. 3. Scatterplots of abiotic factors or environmental concentrations (2009-2019) in relation to bioaccumulated concentrations in biota (2015-2018) with regression lines. Independent variables were included in the above graphs in case of significant contribution according to multiple regression models in Table 3. Significance levels of independent parameters are given in Table 3.

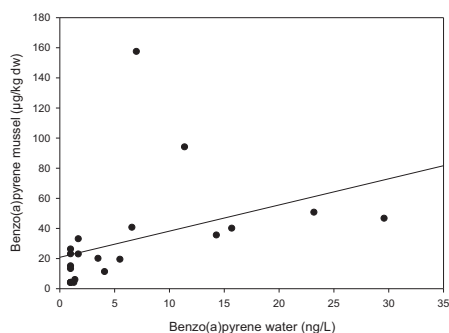


Fig. 3 (continued).

indicating the potential for active metabolism and elimination of fluoranthene by freshwater mussels (Thorsen et al., 2004). Furthermore, no significant difference between results of different sample years ($H_{(3)} = 2.34$; $p = 0.51$), including all mussel species, was found. For benzo(a)pyrene, concentrations reported in the present study (<1 – $27 \mu\text{g kg}^{-1}$ ww; median: $3.0 \mu\text{g kg}^{-1}$ ww) were higher compared to the Flemish study (0.66 – $1.3 \mu\text{g kg}^{-1}$ ww; median: $0.98 \mu\text{g kg}^{-1}$ ww; De Jonge et al., 2014), but comparable to the Dutch results (6.0 – $15 \mu\text{g kg}^{-1}$ ww; Hendriks et al., 1998).

4.2. Detection frequency in different matrices

In the present study, water concentrations of HCB and \sum PCB were often below LOQ (Table 2). For HCB, PFOS and HBCD, this was the case in sediment. Cis-heptachlorepoxyde was not detected in environmental samples. Accumulated concentrations of these compounds in biota, however, were well within the detectable range. Therefore, we conclude that the accuracy of the current method for environmental samples is not suitable for predicting the bioaccumulated concentrations, since environmental concentrations were below the LOQ. For dicofol, the opposite was true; it could only be quantified in sediment samples. This might be a result of a higher LOQ value for biota compared to sediment rather than the absence of dicofol in biota. Previous studies showed higher accumulated dicofol concentrations in fish compared to sediment (Singh et al., 2015). HCBd, heptachlor and trans-heptachlorepoxyde showed a very low detection rate in all three matrices. We did, however, have a very small sample size for HCBd and trans-heptachlorepoxyde measurements in water. In general, it should be made clear that the variation in magnitude of LOQs (Table SI-9) can significantly impact the detection in different matrices. In a monitoring study reporting data collected between 2000 and 2006 in two tributaries of the Nete basin in Flanders, the frequency of detection of HCB and \sum PCB in the environment (water and sediment) was even lower than the present study, despite lower LOQ values (Belpaire et al., 2008). This might indicate an increased presence of these compounds in the environment. However, in the present study, different large water basins were included, and the \sum PCB was interpreted instead of

individual congeners. Van Ael et al. (2012) found a high detection rate of PBDEs and PCBs in the Scheldt basin sediment.

Furthermore, high detection rates of PCBs and organochlorine pesticides (such as HCB) have been reported in eel (Belpaire et al., 2008; Belpaire and Goemans, 2007a; Weltens et al., 2002) and in perch (Bremle and Ewald, 1995) compared to environmental matrices. Furthermore, in line with the results of the present study, a 100% detection rate in eel has been found in previous Flemish studies for mercury and PBDEs (Belpaire and Goemans, 2007a). General low detection rates for hexachlorobutadiene in fish, as was the case for the present study, have been reported before (Macgregor et al., 2010; Roose et al., 2003). To our knowledge, no publications on dicofol concentrations in perch or eel are available.

The only compounds with a high detection frequency in water in the present study were mercury, PFOS and PAHs. However, both for mercury and PAHs a large seasonal variation was observed with a noticeable amount of concentrations at each location below LOQ. In sediment, mercury, PFOS, \sum PBDE, \sum PCB and PAHs were often detected. As stated before, mercury and PFOS show a high affinity for proteins and are less hydrophobic (Amlund et al., 2007; Zhong et al., 2019) than the other priority compounds, which have a pronounced lipophilic character. The water solubility of perfluoroalkyl substances (PFAS) is inversely proportional to the carbon chain length (Labadie and Chevreuil, 2011). Short-chain PFAS and PFOS, previously showed a high detection frequency in water and biota, whereas longer chain compounds are only to partition to sediment particles, with PFOS being the predominant compound in sediment as well (Loi et al., 2011; Xu et al., 2014). Due to the very low water solubility of PCBs and PBDEs, on the other hand, sediment particles are considered a sink for these pollutants (Kuosmanen et al., 2001; Watanabe and Sakai, 2003).

4.3. Relation between biota and environmental samples

In the present study, a direct relationship to accumulated concentrations in biota was identified for water concentrations of PFOS and benzo(a)pyrene and sediment concentrations of HCB, \sum PBDE and \sum PCB (Table 3). As stated before, PFOS shows a larger solubility in water compared to the other priority compounds. Therefore, a more direct relationship between dissolved concentrations in water and bioaccumulated concentrations was to be expected. However, Houde et al. (2008) reported a relationship between PFOS concentrations in invertebrates and sediment rather than between invertebrates and water concentrations. Mussels are exposed mainly to the water column as filter feeders, and therefore a direct relationship for accumulated benzo(a)pyrene to dissolved concentrations is logical. However, kinetic models based on uptake and elimination variability proved to describe the relation between PAH concentrations in zebra mussels and environmental concentrations better than simple equilibrium partitioning (Bourgeault and Gourlay-Francé, 2013). Sediment concentrations of flame retardants (\sum PBDE and HBCD) showed a comparable pattern to the concentrations measured in eel in a monitoring study on 18 sampling locations in the Scheldt basin and three reference locations between 2000 and 2001 (Belpaire et al., 2003). However, Van Ael et al. (2012) found that concentrations of PCBs and PBDEs in sediment were poorly correlated to accumulated concentrations in species from different trophic levels and sediment in the Scheldt estuary. The strongest relations were found for organisms on lower trophic levels, which is expected because they are more likely to ingest sediment or particle-bound hydrophobic compounds. A correlation between eel concentrations and the sum of sediment and dissolved concentrations of PCBs was reported in a pilot study for water quality assessment in Flanders (Weltens et al., 2002). Furthermore, less seasonal fluctuation in PCB concentrations was observed in eel compared to sediment. Both Hg concentrations in fish and fluoranthene in mussels were not significantly affected by abiotic compartments. The absence of a relationship for Hg might be explained by the fact that even at lower trophic levels Hg, in its organic form (methylmercury), is

Table 4

Significant ($p < 0.05$) extrapolation equations for PFOS, Hg, HBCD, \sum PBDE and \sum PCB between both species. Significance levels of the independent parameters are indicated with letters: ^A $p < 0.05$, ^B $p < 0.01$, ^C $p < 0.001$.

Compound	Significant equation	R ² (DF)
PFOS	$\text{Log}(\text{eel}) = 1.67 + 0.41 \cdot \text{log}(\text{perch})^{\text{B}}$	0.26 (28)
Hg	$\text{Log}(\text{eel}) = 1.74 + 0.76 \cdot \text{log}(\text{perch})^{\text{B}}$	0.20 (28)
HBCD	$\text{Log}(\text{eel}) = 2.68 + 1.15 \cdot \text{log}(\text{perch})^{\text{A}}$	0.16 (27)
\sum PBDE	$\text{Log}(\text{eel}) = 1.90 + 1.08 \cdot \text{log}(\text{perch})^{\text{C}}$	0.39 (28)
\sum PCB	$\text{Log}(\text{eel}) = 3.91 + 0.68 \cdot \text{log}(\text{perch})^{\text{C}}$	0.49 (28)

Table 5

Ranges (and median) muscle concentrations of HOCs in perch and eel as measured during the present study compared to literature data from European monitoring studies.

Species	HCB	HCBd	Hg	∑PBDE	PFOS	HBCD	Dioxins ^a	∑PCB	Country	Study
<i>Anguilla anguilla</i>	0.12-10 (3.1)	<0.5-2.1 (0.25)	32-332 (132)	0.25-106 (7.4)	1.5-65 (8.3)	<0.3-412 (9)	0.001-0.04 (0.008)	5.3-1320 (385)	Belgium, Flanders	Present study
	<LOQ-62		10-535	6.9-5284 ^d					Belgium, Flanders	(Belpaire and Goemans, 2007a)
							0.057		Belgium, Flanders	(Belpaire et al., 2011)
									Belgium, Flanders	(Byer et al., 2013)
	<2-19 (6.2)	<2-5	49-324 (194)	14-15	7.2-34 (24)	110-430			Belgium, Flanders	(De Jonge et al., 2014)
			93-173						Belgium, Flanders	(Malarvannan et al., 2014)
								5-2600 (75) ^b	Belgium, Flanders	(Maes et al., 2005)
	0.002-192		5.0-1185						Belgium, Flanders	(Maes et al., 2008)
		<LOQ-6.9 (0.2)							Belgium, Flanders	(Roose et al., 2003)
	2.1-5.6 (3.9)			7.5-18 (8.8) ^c					Belgium, Flanders	(Van Ael et al., 2013)
			10-708 (97)						Belgium, Flanders	(Van Ael et al., 2014)
								3.5-279	France	(Blanchet-Ietrouvé et al., 2014)
	3.4-50		69-314	0.1-18 ^c	18-39			29-746 ^b	France	(Couderc et al., 2015)
	1.9-2.5			9.2-242	8.3-49			165-1630 ^b	Germany	(Guhl et al., 2014)
					37-83 (77)		0.006-0.045		Germany	(Hölzer et al., 2011)
								Great Britain	(Jürgens et al., 2013)	
								Italy	(Giari et al., 2015)	
<LOQ-21 (1.2)								37-518 (159)	Italy	(Ferrante et al., 2010)
				0.27-0.93 (0.50)		0.16-1.1 (0.54)			Italy	(Tavoloni et al., 2021)
				1.0-7.1 ^e		1.2-15		1.9-18	Ireland	(Mchugh et al., 2010)
0.4-23.8							0.0007-0.008	4.0-534	Poland	(Szlander-richert et al., 2010)
				0.07-8.2		0.18-16	0.001-0.015	1.7-289	Poland	(Szlander-richert et al., 2014)
<3-7.2)	<3.3-3.9)							(5.6-10,487)	Scotland	(Macgregor et al., 2010)
					7-52				The Netherlands	(Kwadijk et al., 2010)
				8.3-151		<0.1-230			The Netherlands	(Van Leeuwen and De Boer, 2008)
<i>Perca fluviatilis</i>	<0.1-0.52 (0.05)	<0.5-0.79 (0.25)	32-148 (58)	<0.3-1.4 (0.73)	2.4-54 (10)	<0.3-1.1 (0.29)	0.0003-0.005 (0.001)	0.75-140 (18)	Belgium, Flanders	Present study
	<2	<2	42-926 (97)						Belgium, Flanders	(De Jonge et al., 2014)
							0.42-1.6		Czech Republic	(Pulkrabová et al., 2007)
	0.26-0.33		131-509	0.7-1.4	8.1-12		0.0007-0.0015	8.2-16 ^b	Germany	(Fliedner et al., 2018)
					39-150 (96)				Germany	(Hölzer et al., 2011)
			260-310						Norway	(Braaten et al., 2014)
			221-448	<LOQ-0.024 ^f			<LOQ-0.027		Finland	(Miller et al., 2013)
					5.4-17				Italy	(Tavoloni et al., 2021)
		20-2420						Italy	(Squadrone et al., 2015)	
		263-550						Sweden	(Sonesten, 2003)	
		160-830		<LOQ-0.93				Sweden	(Miller et al., 2013)	
								Sweden	(Åkerblom et al., 2017; Negm, 2015)	

Concentrations in µg kg⁻¹ ww.^a Concentrations in µg WHO-TEQ₂₀₀₅ kg⁻¹ ww.^b PCB as sum of 6 congeners (without PCB118).^c PBDE as sum of 7 congeners (including BDE 183).^d PBDE as sum of 10 congeners.^e PBDE as sum of 11 congeners.^f PBDE as sum of 15 congeners.

primarily ingested via food rather than absorbed from the water and is strongly bioaccumulative (Bradley et al., 2017). Accordingly, no relationship was found between environmental concentrations in water or

sediment and the trophic magnification slope (TMS) of mercury (Lavoie et al., 2013). In contrast to the present study, De Jonge et al. (2014) did not find a significant relationship between accumulated HCB

concentrations in eel and water or sediment concentrations, although it should be taken into account that their sample size was much smaller. For Hg, on the other hand, they found a relationship between perch and water concentrations. However, this might have been due to very high tissue concentrations at one of the sampling locations.

As a measure for hydrophobicity, the octanol-1-water partition coefficient (K_{OW}) does not provide a straightforward explanation for relationships between biota and environmental matrices. Compounds with a $\log K_{OW} < 5$ are considered to biomagnify less (Kim, 2019). Although the $\log K_{OW}$ for PFOS is relatively low (4.49; <https://pubchem.ncbi.nlm.nih.gov/>). It should, however, be taken into account that this is an estimated value. Due to the surface-active properties of PFOS, the $\log K_{OW}$ cannot be measured accurately (<http://www.atsdr.cdc.gov/toxprofiles/index.asp>). For benzo(a)-pyrene, the $\log K_{OW}$ value is considerably higher (6.13; De Maagd et al., 1998). This is in contrast to the relationship found between water and accumulated concentrations for benzo(a)pyrene in the present study. On the other hand, the $\log K_{OW}$ for HCB (5.5), PCBs (5.6-6.6) and PBDEs (5.9-7.9) do explain the relationship with the sediment compartment due to a higher hydrophobicity (Braekevelt et al., 2003; Larsen et al., 1992; Mackay et al., 1992). PCBs and PBDEs typically show a parabolic relationship between TMF and $\log K_{OW}$ of different congeners, with TMF increasing until $\log K_{OW} = 7$, before a depression for higher hydrophobicity (Bremle and Ewald, 1995; Wu et al., 2009). This decrease in biomagnification potential is probably due to the large molecular size slowing down transport and subsequent fast elimination (Fisk et al., 1998). However, other studies found a linear increase, even for PCBs with $\log K_{OW} > 7$ (Van Ael et al., 2013). The $\log K_{OW}$ values for PCBs and PBDEs generally increase with degree of halogenation. However, a stronger biomagnification effect for PCBs than PBDEs with the same halogenation degree has been described (Van Ael et al., 2013; Wu et al., 2009).

4.4. PCB and PBDE profiles

In the present study, PCB profiles in biota were comparable to those in the environmental matrices, with PCB153 contributing the most to the \sum PCB, followed by PCB138 and PCB180. Belpaire et al. (2008) stated that contamination profiles are location specific, suggesting a different input of pollutants. Furthermore, they found that higher-chlorinated PCBs had a higher detection rate in sediment. Bremle and Ewald (1995) reported PCB patterns in water and sediment being comparable, with perch accumulating more higher-chlorinated PCBs than the abiotic compartments. Accordingly, higher-chlorinated PCBs show a stronger biomagnification, resulting in the largest contribution of PCB153 in eel and PCB28 being more prominent in sediment than in biota (Van Ael et al., 2012; Weltens et al., 2002). Similar patterns were found in eel from the North Rhine-Westphalian basin (Guhl et al., 2014) and the Scheldt river (Roosens et al., 2008). Eels from different European countries show the highest contribution for PCB138 and PCB153, with Belgian eels typically characterised by the highest PCB153 contribution followed by PCB 138 (Belpaire et al., 2011; Malarvannan et al., 2014).

The largest PBDE portion in the present study consisted of BDE47, followed by BDE99 in perch and BDE100 in eel. In sediment, BDE99 and BDE47 respectively contributed to the total sum the most. BDE47 is considered the main congener in biota (Roosens et al., 2008; Van Ael et al., 2012), mainly due to the previous elaborate use of penta-BDE in many countries. On the other hand, carp has been shown to metabolise BDE 99 to lower brominated congeners (e.g. BDE47) (Stapleton et al., 2004). To a lesser extent, this was also seen for American eel (*Anguilla rostrata*; Ashley et al., 2007). Roberts et al. (2011) reported a species-dependent metabolization rate, with carp metabolizing 10-100 times faster than salmonid fish. This slow metabolization effect might also be true for perch, leading to a larger contribution of BDE99 to the \sum PBDE. Comparable patterns to the present study were found in eel from Belgium (Malarvannan et al., 2014;

Roosens et al., 2008) and Germany (Guhl et al., 2014). A large contribution of BDE47 and BDE100 respectively was also reported in perch from the Scheldt, although BDE99 contribution to the total \sum PBDE was comparable to BDE100 (Roosens et al., 2008). Voorspoels et al. (2004) indicated that profiles of PBDE, observed in fish samples, did not correspond to profiles in sediment from the Scheldt estuary. Their sediment samples collected from the Scheldt basin showed a comparable PBDE profile to the present study (BDE99 > BDE47 > BDE154 > BDE100 > BDE153 > BDE28). BDE209 has been identified as an important congener in sediments due to its widespread use of deca-BDE as a fire protection surfactant (Van Ael et al., 2012; Voorspoels et al., 2004). However, it was not included in the present study as uptake by aquatic invertebrates or fish is hindered by its physical properties (i.e. high molecular mass, high $\log K_{OW}$), slow uptake, and rapid metabolization (Kierkegaard et al., 1999; Stapleton, 2003).

4.5. Effects of abiotic characteristics on bioaccumulation

No significant effect of environmental characteristics could be linked to bioaccumulation of mercury or fluoranthene. Lavoie et al. (2013) found an increase in the TMS of mercury with increasing DOC levels. Other studies, however, showed that DOC could limit the uptake of Hg and lipophilic compounds by reducing bioavailability due to complexation (Dittman and Driscoll, 2009; Li et al., 2015). This effect was found in the present study for some HOCs, with a negative relationship of DOC or TOC relative to accumulated HCB, \sum PBDE and \sum PCB concentrations in biota being identified. Furthermore, negative effects were found of pH on the accumulation of HCB in eel, of nitrite on PFOS accumulation in eel and of clay on \sum PBDE accumulation in eel. Although extreme pHs have been shown to be toxic to fish (Wood, 2001), no direct relationships to bioaccumulation have been identified. Watras et al. (1998) stated that low pH potentially slows down the growth rate and, therefore, biodilution, resulting in higher accumulated concentrations at a low pH and thus a negative relation between accumulated concentrations and pH. In the present study, however, pH values ranged from neutral to more basic (6.8-9.1). Furthermore, clay content in the sediment was correlated to TOC levels and is generally considered a sorbent for immobilization and detoxification of hazardous substances (Awad et al., 2019). High concentrations of nitrate, internally converted to nitrite, are toxic to wildlife and humans by reducing the oxygen-binding capacity of haemoglobin and decreasing of total haemoglobin (Monsees et al., 2017; Yang et al., 2019). However, no clear explanation could be found for the negative relation with PFOS accumulation. High nitrite content might even be expected to reduce the fish metabolism, potentially reducing the elimination rate of pollutants. Other factors not included in the present study, might influence bioaccumulation and biomagnification efficiency (e.g. food web structure, food availability, overall quality of ecosystems). For example, biomagnification of Hg has been shown to be the highest in cold and low productivity environments (Lavoie et al., 2013). On the other hand, due to the spread of sampling locations within the relatively small area of Flanders, the abiotic environmental characteristics did not show extreme situations. Therefore, studies in more variable and extreme environments might reveal stronger effects of these factors.

4.6. Extrapolation between perch and eel concentrations

Finally, our results showed an extrapolation possibility for bioaccumulated concentrations of PFOS, Hg, HBCD, \sum PBDE and \sum PCB between perch and eel. Weltens et al. (2002) also found a positive relationship between PCB concentrations in eel and other biota (including predator fish). Extrapolation between species can have important implications for future monitoring studies. Monitoring studies on a regular basis as e.g. demanded for the WFD (European Commission, 2008) require significant efforts in field work in order to collect the required specimens for analysis. Practical constraints and limits in the

distribution and abundance of the targeted species often impede the collection of sufficient suited fish samples. In our study in respectively 25 and 7% of the sites perch and eel could not be sampled (sufficiently). Extrapolation from one species to another can now complement the missing gaps. Our equations may also be useful when comparing and intercalibrating datasets from different monitoring networks (e.g. different European countries). On the other hand, European eel stocks have been declining over the last decades, probably due to high pollutant levels (Palstra et al., 2006; ICES, 2020), resulting in an IUCN red list status 'critically endangered' (Jackoby and Gollock, 2014). Due to their high fat content, eels tend to accumulate very high levels of lipophilic compounds, facilitating detection and analysis. The present study revealed the European perch (IUCN red list status: 'low concern'; Freyhof and Kottelat, 2008) as a valid alternative.

However, in order to extrapolate accumulation between multiple species from different (European) monitoring programs, species-specific, lifestyle-based traits should be taken into account (e.g. lipid content, trophic level). In the guidance document of the EU on biota monitoring, a standardization based on lipid content, trophic level (TL) and dry weight was proposed (European Commission, 2014). Concentrations of hydrophobic compounds should be standardized for an individual with a TL of 3–4, containing 5% lipid content. For mercury and PFOS, on the other hand, a standardization for TL and 26% dry weight is recommended, due to their affinity for proteins.

Trophic levels of perch and eel in the Flemish water bodies were not significantly different (Teunen et al., 2021). Higher concentrations of HOCs in eel compared to perch, on the other hand, were probably a result of the high lipid content in eel (Table SI-1).

5. Conclusions

Accumulated HOCs concentrations in aquatic biota, especially high trophic levels, are generally much higher than concentrations measured in abiotic environmental matrices, allowing for easier detection and analysis of these compounds. Bioaccumulated concentrations are not related merely to environmental concentrations. Frequently, other abiotic parameters (such as organic content) can affect bioavailability and metabolism processes. In the present study, we found a positive relation between bioaccumulated concentrations and dissolved water concentrations for PFOS and benzo(a)pyrene and between bioaccumulated concentrations and sediment concentrations for HCB, Σ PBDE and Σ PCB. Furthermore, an additional negative effect of TOC or DOC was detected for the latter group. PCB profiles between all matrices were comparable, while PBDE profiles showed evidence of metabolism of higher halogenated congeners by the fish. In general, we advise using biota over environmental sampling for monitoring purposes since bioaccumulation and magnification of hydrophobic compounds is a complex process with numerous mediating factors at play. Therefore, modelling concentrations in top predators based on environmental measurements are likely to underestimate or misinterpret effective body burdens. Furthermore, due to seasonal variation, especially for water concentrations, elaborate environmental sampling is needed to predict accumulated concentrations in biota. Finally, we observed an extrapolation potential between perch and eel for PFOS, Hg, HBCD, Σ PBDE, and Σ PCB concentrations. This allows for implementing missing gaps in datasets when field surveys failed to collect suitable fish samples. Equations in bioaccumulation between species may offer new opportunities in calibration exercises between monitoring frameworks of different European countries.

CRedit authorship contribution statement

Lies Teunen: Conceptualization, Resources, Formal analysis, Writing – original draft. **Maarten De Jonge:** Resources, Writing – review & editing. **Govindan Malarvannan:** Resources, Writing – review & editing. **Adrian Covaci:** Resources, Writing – review & editing. **Claude**

Belpaire: Resources, Writing – review & editing. **Jean-François Focant:** Resources, Writing – review & editing. **Ronny Blust:** Writing – review & editing. **Lieven Bervoets:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary information

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.149448>.

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