Dispersal of persistent organic pollutants from fiber-contaminated sediments: biotic and abiotic pathways

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Abstract

Purpose Numerous sites contaminated with fiber emissions from pulp and paper industries are found in coastal areas of the Baltic Sea, but there is limited knowledge about the magnitude of dispersal of persistent organic pollutants (POPs) from these anthropogenic, organic-rich sediments called fiberbanks. The aim of this study was to quantify and compare different POP dispersal pathways from such fiberbanks. Dispersal mechanisms studied included abiotic and biotic routes (dissolved in water, particle-bound, and bioaccumulation).

Materials and methods Contaminated fibrous sediments located in Ångermanälven River estuary in north-eastern Sweden were studied in sediment types representing different fiber content (i.e., fiberbanks, fiber-rich sediments, and less fiber impacted sediments). Sediment-to-water fluxes of dissolved contaminants (polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), and dichlorodiphenyltrichloroethane (DDT)) were measured in situ using benthic flux chambers. Particle resuspension was measured by sampling bottom water, before and after disturbing the sediment surface. Benthic biota was collected to determine the body burden of contaminants and to determine biota-pore water accumulation factors (BAF_{PW}) and biota-sediment accumulation factors ($BSAF$s). In addition, concentrations of dissolved POPs in the water column were measured in field using passive samplers. Instrumental analysis was performed using gas chromatography coupled to a triple quadrupole mass spectrometer (GC-MS/MS).

Results and discussion The flux of dissolved Σ_{20}PCBs was approximately two times higher from one of the investigated fiberbanks (3.4 ng m^{-2} day^{-1}) compared to the other. The average particle burden of PCBs was also higher at this fiberbank after artificial disturbance (15 ng g^{-1} particle), which indicates that larger amounts of contaminants are likely to disperse via particle resuspension from this site compared to the other fiberbank (4.8 ng g^{-1} particle). The difference might be associated with a layer of recently settled minerogenic material that covers one of the fiberbanks, which probably functions as a protective barrier. The lack of benthic biota implies that contaminant release by bioturbation is negligible in the studied fiberbanks. However, benthic biota from fiber-rich sediment showed bioaccumulation and biomagnification of contaminants.

Conclusions The importance of diffusive flux from fiberbanks under undisturbed conditions became apparent when the different dispersal pathways were quantified. However, no dispersal pathway could be judged as irrelevant, since even under undisturbed conditions, advective particle transport was significant. Additionally, the uptake by biota and trophic transfer can be considerable. Quantification of dispersal routes and understanding of the relative importance of various pathways is critical for proper risk assessment and management of contaminated sediments.

Keywords Fiberbank · Resuspension · Benthic uptake · Sediment-to-water flux · Pulp and paper emissions

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1 Introduction

The aquatic environment can be polluted by persistent organic pollutants (POPs) that exist at contaminated sediment sites by transport via various mechanisms such as diffusion, advection (particle resuspension), and bioturbation (Schwarzenbach et al. 2003). However, the importance of each of these potential dispersal pathways is rarely quantified, which restricts comparisons between and within sites. Fibrous sediment is a highly contaminated anthropogenic type of sediment (Apler et al. 2014; Apler et al. 2020; Dahlberg et al. 2020) formed through previous emissions of wood fiber material from the pulp and paper industry. Knowledge about the dispersal of POPs is limited, and quantitative information is essentially lacking. This includes the dispersion of contaminants as well as fiber particles into the environment and the environmental consequences that these processes may have.

The pulp and paper industry has historically been a major source of aquatic contaminants due to the release of insufficiently treated wastewater that contained high levels of, e.g., organic matter, POPs, and heavy metals (Ali and Sreekrishnan 2001; Hall 2003; Svrcek and Smith 2003). The extensive discharge of fibers, which were emitted together with wastewater, was deposited on the sea floor near outlets and led to the formation of large fiber deposits (*fiberbanks*). Further away from the fiberbanks in sediment accumulation zones, *fiber-rich sediments* are formed when fibers are mixed with natural, more minerogenic sediment (Apler et al. 2014). The pulp and paper production grew rapidly during the twentieth century, with North America and northern European countries being the dominating pulp and paper producers still today. Besides Sweden, fiber-contaminated sediments are present in many pulp and paper production areas such as Finland (Kokko et al. 2018), Canada (Biberhofer et al. 2011), and the USA (WSDE 2012). Despite their abundance and unique characteristic of having relatively high organic content (8.6–37% TOC (Dahlberg et al. 2020)), low density, and generally high gas content, fiberbanks have been poorly investigated. In Sweden, fiberbanks have shown to be highly contaminated with heavy metals and POPs, such as polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), dichlorodiphenyltrichloroethane (DDT), and its related transformation products, dichlorodiphenyldichloroethene (DDE) and dichlorodiphenyldichloroethane (DDD) (Apler et al. 2014; Norrlin and Josefsson 2017; Dahlberg et al. 2020). In a recent study, it was shown that although organic carbon-rich fiberbanks sorb hydrophobic pollutants were almost 20 times stronger than natural (minerogenic) sediment (Dahlberg et al. 2020), the PCB levels in the pore water of the fiberbanks were higher (approx. 4 times) than corresponding levels in the natural sediment due to the high level of contamination of the fiberbanks (Dahlberg et al. 2020). Hypothetically, the higher concentrations of freely dissolved POPs in the pore water of the fiber-rich compartments cause a net diffusive flux of pollutants to the overlying water, but fluxes of POPs from these organic-rich sediments have not yet been empirically quantified.

Apart from diffusion, dispersal of POPs from sediment to the surrounding aquatic environment can also occur through advective processes, such as colloidal transport (Sen and Khilar 2006) and particle resuspension due to, e.g., wave motion and currents (Roberts 2012), gas ebullition (Yuan et al. 2009), and bioturbation (Josefsson et al. 2010; Mustajarvi et al. 2019) as well as less frequent, but large-scale events, such as submarine landslides. Signs of past submarine landslides have been observed in fiberbanks deposited in coastal areas with steep slopes (Apler et al. 2014) and probably caused major resuspension of particles. While the resuspended particles are suspended in the water column, solubilization of contaminants can occur (Eggleton and Thomas 2004; Schneider et al. 2007). The contaminated particles can be transported by the water and redeposit in other areas. Although there are several potential mechanisms that may lead to resuspension and subsequent dispersion of polluted fiberbank material, little is known about the importance of this process on contaminant transport because it has not been quantified specifically for fiberbanks. Another route of dispersal of POPs associated with fiberbanks and fiber-rich sediments is trophic transfer. Benthic organisms accumulate POPs via ingestion and diffusion, and the POPs can then biomagnify in the food web (Nfon et al. 2008). To our knowledge, the magnitude of bioaccumulation of POPs associated with fiberbanks and fiber-rich sediments has not yet been investigated.

The overall aim of this study was to quantify and compare different dispersal mechanisms for PCBs, HCB, DDT, and its related transformation products (DDE and DDD), at three fiberbank sites located in a coastal area in north-eastern Sweden. The dispersal mechanisms included abiotic routes (dissolved and particle-bound) and a biotic route (bioaccumulation). Briefly described, we determined sediment-water fluxes of dissolved contaminants, the effect of particle resuspension, biota-sediment accumulation factors (*BSAF*s), and biota-pore water accumulation factors (*BAF_PW*). The concentration of dissolved POPs in the free water column was also measured at two sites. Our study provides novel information on the magnitude of pollution dispersion from contaminated sediments, more specifically in fiberbank sites, and allows for a quantification of different dispersal pathways. This knowledge is critical for risk assessment and risk management of contaminated sediment, including fiberbank areas, in the Baltic Sea and globally.

2 Materials and methods

2.1 Study area

Field sampling was performed at three fiberbank sites. These are located close to Väja kraft pulp mill (V), Sandviken old

kraft pulp mill (S), and for biota also Kramfors old sulfite pulp mill (K) in the Ångermanälven River estuary located in the county of Västernorrland, NE Sweden (Fig. 1, Figs. S1–4). This part of Sweden is known for its expansive pulp and paper industry since the late nineteenth century. Detailed descriptions of the fiberbanks (e.g., size, fiber material, and thickness) have been presented previously (Dahlberg et al. 2020), but a summary is provided in the Supporting information (Table S1). Samples were also taken along the river estuary, from the fiberbank areas to the outer estuary, to study potential pollution gradients (transect, T, Fig. S5).

2.2 Benthic flux chambers

Fluxes of dissolved POPs through the sediment-water interface were measured in situ using benthic flux chambers (BFCs) that contained samplers in the form of semipermeable membrane devices (SPMDs). The flux chambers have been developed by the Norwegian Geotechnical Institute (NGI) and are described in detail by Eek et al. (2010). Briefly, each BFC encloses a volume of water (3.4 L) above the sediment surface (area 0.049 m^2) and

is equipped with a triolein-filled (4.8 mL) SPMD (0.92 m length, 534 cm^2 surface area, 75–90 μm-thick LDPE, Exposmeter, Sweden). The SPMD works as an "infinite-sink" that collects organic pollutants as they diffuse from the sediment to the bottom water just above the seabed. It absorbs dissolved pollutants transported through the sediment-water interface by diffusion or by advective processes affecting the sediment pore water, including gas ebullition. It should, however, be noted that the enclosure caused by the BFC can lead to oxygen deficiency, changes in redox conditions, and decreased bioturbation inside the chamber. Fluxes measured with BFCs thus tend to be underestimated when the natural activity of benthic organisms is suppressed by the oxygen deficiency.

The BFCs ($n = 6$) were deployed on the fiberbanks in Väja and Sandviken and at different distances from the fiberbanks in summer 2015. At each site, one BFC was deployed on the fiberbank, the fiber-rich sediment, and the more natural (minerogenic) sediment. (Table S2, Figs. S1–2). The BFCs were placed on the seabed and fastened with a line to a buoy. The BFCs were collected after 32–34 days. In addition to the BFCs, SPMDs were also deployed in the water column in
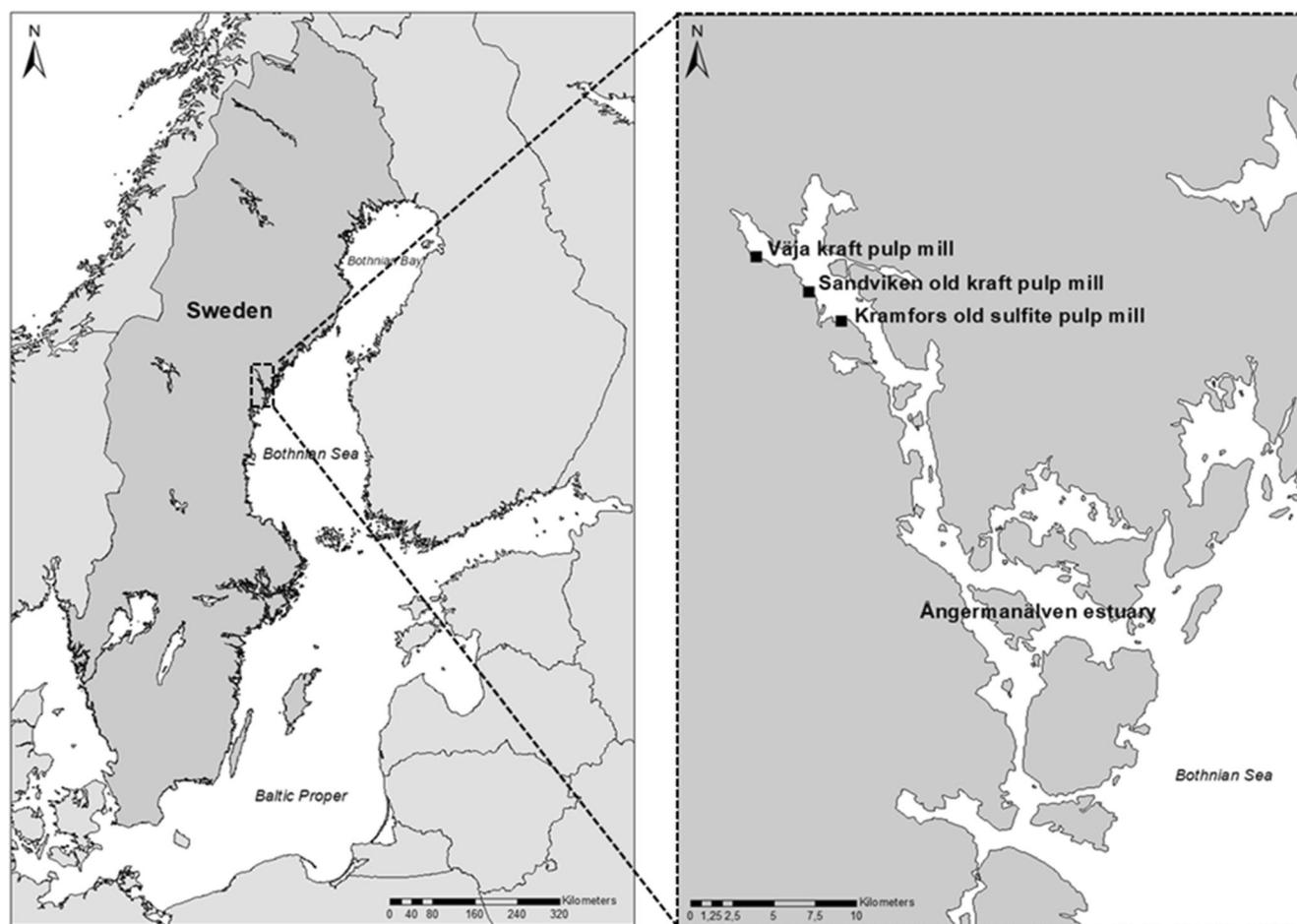
Fig. 1 Maps showing the location of the Ångermanälven River estuary (Sweden) and the sampling area. The three sampled contaminated sites are located next to Väja kraft pulp mill and Sandviken and Kramfors old pulp mills

order to measure POPs in the free water body in Sandviken at two sites (Table S2, Figs. S1–2). The SPMDs were mounted on a stainless-steel spider in a stainless-steel canister (Exposmeter, Sweden, Ø 16 cm, height 15 cm) attached to a line at two water depths (2 m above the sediment and 7 m below the water surface, respectively) for 32–34 days. The SPMDs were deployed in duplicate at each site and depth (in total 8 SPMDs).

2.3 Suspended particles

The amount of suspended particles was measured by sampling bottom water at Väja (V, $n = 11$) and Sandviken (S, $n = 8$) in autumn 2017, using a Ruttner water sampler (2 L) mounted on a camera frame. Water was collected approximately 30 cm above the sediment surface when (i) the sediment surface was undisturbed and (ii) after artificially disturbing the sediment surface with a weight suspended underneath the camera frame. Detailed information about the sampling sites and water depths are given in the Supporting information (Table S3, Figs. S1–2).

Each water sample was divided into two aliquots (1 L each) and filtered through a GF/F glass microfiber filter (0.7 μm, Whatman). One of the two filters was dried at 105 °C for gravimetrical determination of the mass of particles. The other filter was stored frozen (at − 18 °C) until chemical analysis of the particle-bound POPs. Prior to filtration, all filters were pre-washed with MilliQ-water and dried in an oven at 400 °C before used.

2.4 Benthic biota

Benthic biota was collected to determine their body burden of contaminants and to calculate BAF_{PW} and $BSAF$s. The benthic macroorganisms (*Marenzelleria sp.* and *Saduria entomon*) were collected in summer 2014 and 2015, using a boxcore sampler. Sediment was sampled from Väja (V, $n = 8$), Sandviken (S, $n = 7$), Kramfors (K, $n = 5$), and the river estuary transect (T, $n = 6$). The sampled area at each sampling site was 0.17 m^2. The benthic organisms were collected after sediment sieving (mesh size 1 mm). The organisms were placed in glass jars with water from the sampling site and then stored cold (+ 4 °C, ~ 24 h) for depuration. After depuration, the water was removed by blotting on filter paper and the organisms were stored frozen (− 18 °C) until chemical analysis. Detailed information about the sampling sites are given in the Supporting information (Table S4, Figs. S3–5).

2.5 Sediment

In addition to benthic organisms, sediment samples were collected at the same sites for chemical analysis of POPs in both sediment and pore water. The measured POP levels in

sediment and pore water from Väja, Sandviken, and Kramfors were published by Dahlberg et al. (2020), whereas original results from sediment ($n = 6$) and pore water collected along the river transect (T, $n = 6$) sampled in summer 2015 are presented here (Table S5, Figs. S5). The sediment samples were stored cold (+ 4 °C) prior to sediment pore water extraction and then stored frozen (− 18 °C) until chemical analysis.

2.6 Target compounds

The targeted POPs included 20 polychlorinated biphenyls (PCBs: CB-28, CB-52, CB-77, CB-81, CB-101, CB-105, CB-114, CB-118, CB-123, CB-126, CB-138, CB-153, CB-156, CB-157, CB-167, CB-169, CB-170, CB-180, CB-189, and CB-209). The sum of all target PCBs are abbreviated Σ_{20}PCBs, and the sum of the seven indicator PCBs (CB-28, CB-52, CB-101, CB-118, CB-138, CB-153, and CB-180) are abbreviated as Σ_7PCBs. Hexachlorobenzene (HCB), dichlorodiphenyltrichloroethane (*o,p′*-DDT and *p,p′*-DDT), and their transformation products dichlorodiphenyldichloroethene (*o,p′*-DDE and *p,p′*-DDE) and dichlorodiphenyldichloroethane (*o,p′*-DDD, *p,p′*-DDD) were also analyzed. The sum of the six DDT-related compounds is referred to as Σ_6DDX. Information about the authentic reference compounds, isotope-(^{13}C)-labeled internal standards (ISs) and isotope-(^{13}C)-labeled recovery standards (RSs), used is provided in the Supporting information (Table S6 and S7).

2.7 Chemical analysis

2.7.1 SPMDs

The SPMDs were carefully wiped clean from particles using a wet paper tissue and weighted prior to extraction. Each SPMD was placed in an amber glass jar and extracted with *n*-hexane (150 mL + 150 mL) for 2 × 24 h, while continuously vertically shaken using a modified method by Eek et al. (2010). Prior to extraction, ISs were added (Table S7) to the first aliquot of solvent in the glass jar. The extracts from the two extraction cycles were combined and evaporated down to ~ 1 mL. The extract was then cleaned up on a multilayer-clean up column composed of (from bottom) activated silica (SiO$_2$, 3 g), sulfuric acid-treated silica (40% H$_2$SO$_4$: SiO$_2$, 6 g), and sodium sulfate (Na$_2$SO$_4$, 3 g), similar to Josefsson et al. (2010). The analytes were eluted with 60 mL *n*-hexane, the extract was thereafter evaporated to 100 μL, and RSs were added (Table S7).

2.7.2 Suspended particles

The GF/F glass microfiber filters were Soxhlet extracted with acetone : *n*-hexane (225 mL, 1 : 1, *v/v*) for approx. 24 h and

cleaned up as described above (Section 2.7.1). Prior to extraction, ISs were added to each sample (Table S7).

2.7.3 Benthic biota

Each biota sample was weighed, and an aliquot was taken for dry and lipid weight (dw and lw) determination. The remaining aliquot was mixed and mortared with sodium sulfate (Na_2SO_4). This mixture was then added to glass columns fitted with glass wool. ISs were added on top (Table S7), and the sample was then extracted using acetone : n-hexane (75 mL, 5:2, v/v) followed by n-hexane : diethyl ether (75 mL, 9 : 1, v/v). Each sample extract was evaporated to ~1 mL and cleaned up on a multilayer silica column composed of (from bottom) activated silica (SiO_2, 3 g), sulfuric acid-treated silica (40% $H_2SO_4 : SiO_2$, 6 g), sulfuric acid-treated silica (20% $H_2SO_4 : SiO_2$, 3 g), and sodium sulfate (Na_2SO_4, 3 g) similar to Josefsson et al. (2010). The analytes were eluted with n-hexane : dichloromethane (60 mL, 4 : 1, v/v). The extract was thereafter evaporated to 100 µL, and RSs were added (Table S7).

The dry weight was gravimetrically determined after drying an aliquot of the sample at 60 °C for 24 h. The lipid weight of *Marenzelleria* sp. was determined by re-hydrating the dried sample with water, mortaring it with sodium sulfate (Na_2SO_4, 4 g), and transferring it to a glass column fitted with glass wool. The lipids were then eluted with acetone : n-hexane (6 mL, 5 : 2, v/v) and n-hexane : diethyl ether (6 mL, 9 : 1, v/v). The extract was evaporated to dryness, and the lipid weight was determined gravimetrically. For *Saduria entomon*, an aliquot of the extract obtained from the chemical extraction using acetone : n-hexane (75 mL, 5 : 2, v/v) followed by n-hexane : diethyl ether (75 mL, 9 : 1, v/v) was instead used for gravimetric lipid determination.

2.7.4 Sediment

The sediment samples were extracted using Soxhlet with acetone : n-hexane (225 mL, 1 : 1, v/v) and cleaned up as recently described by Dahlberg et al. (2020). In short, ISs were added to each sample prior to extraction (Table S7). The extract was treated with activated copper to remove sulfur and then further cleaned up on a multilayer column composed of (from bottom) activated silica (SiO_2, 3 g), sulfuric acid-treated silica (40% $H_2SO_4 : SiO_2$, 6 g), and anhydrous sodium sulfate (Na_2SO_4, 3 g) similar to Josefsson et al. (2010). The analytes were eluted with 60 mL n-hexane. The samples were thereafter evaporated to 100 µL, and RSs were added (Table S7).

An aliquot of sediment was also used to gravimetrically determine the dry weight of each sediment sample after heating (105 °C, 24 h). Dry combustion (1350 °C) and elemental analysis using a TruMac instrument (LECO Corporation, St. Joseph, USA) were used to determine the sediment content of total carbon (TC) and total organic carbon (TOC), following the Swedish standard procedure SS-ISO 10694.

2.7.5 Pore water

The freely dissolved pore water concentration was determined by equilibrating sediment with a polyoxymethylene (POM) strip (76 µm in thickness, CS Hyde, Lake Villa, IL, USA) as recently described by Dahlberg et al. (2020). Each POM strip was extracted with acetone : n-hexane (1 : 1, v/v, 20 + 20 mL) for 2 × 24 h using an end-over-end shaker. Before extraction, IS were added (Table S7). After extraction, the POM strip was retrieved and the weight of the dry POM strip was determined gravimetrically. The extract was thereafter evaporated to 100 µL, and RSs were added to each sample (Table S7).

2.8 Calculations

BSAF describes the ratio between the concentration in biota and sediment (using TOC normalized sediment concentrations) whereas BAF_{PW} expresses the ratio between the dissolved fraction measured in pore water and the concentration in biota. BAF_{PW} and *BSAF* of each target compound and organism were calculated using spatially and temporally paired concentration measurements of biota, pore water, and sediment using Eq. (1) and (2) (Burkhard 2003), respectively.

$$BAF_{PW} = (C_{BIOTA}/f_{LIPID})/C_{PW} \qquad (1)$$

$$BSAF = (C_{BIOTA}/f_{LIPID})/(C_{SED}/f_{TOC}) \qquad (2)$$

where C_{BIOTA} is the chemical concentration in the organism (ng g^{-1} wet weight), f_{LIPID} the lipid fraction of the organism (g lipid g^{-1} wet weight), C_{SED} is the chemical concentration in sediment (ng g^{-1} dw), f_{TOC} is the fractional amount of organic carbon in the sediment (g TOC g^{-1} dw), and C_{PW} is the sediment pore water concentration (ng mL^{-1}). The measured sediment and pore water concentrations used have been presented by Dahlberg et al. (2020).

The sediment to water flux of each target compound ($F_{COMPOUND}$; ng m^{-2} day^{-1}) was calculated using Eq. (3) where M_{SPMD} is the amount of the target compound accumulated in the SPMD in the benthic flux chamber (ng), A_{SED} is the sediment surface area covered by the chamber (m^2), and $t_{DEPLOYMENT}$ is the sampling time (days).

$$F_{COMPOUND} = (M_{SPMD})/(A_{SED} \; x \; t_{DEPLOYMENT}) \qquad (3)$$

The water concentration measured by the SPMDs deployed in the free water column was calculated using the United States Geological Survey's (USGS) SPMD calculator v5.1. Differences in field conditions at the different sites (e.g., water

flow, biofouling, and temperature) can affect the sampling rates and were accounted for through the use of a performance reference compound (PRC) (CB-54), which enable estimation of in situ sampling rates as described by Huckins et al. (2006).

2.9 Instrumental analysis and quality assurance/quality control

All samples were analyzed using a gas chromatograph (GC; Agilent Technologies, 7890 A) coupled to a triple quadrupole mass spectrometer (Agilent Technologies, 7010, GC-MS/MS Triple Quad). The MS was operated using multiple reaction monitoring (MRM) mode, in which two transitions were monitored for each compound. The software Agilent MassHunter Quantitative Analysis (for QQQ) was used for data evaluation. Detailed information about the instrumental settings has been presented previously (Dahlberg et al. 2020) but is also given in brief in the Supporting information. Authentic reference standards were used for identification and quantification.

The limit of detection (LOD) was set to a signal-to-noise ratio of 3. The lowest calibration solution that could be quantified reliably by the GC-MS/MS was set as the limit of quantification (LOQ). The ISs were used to correct for losses during sample extraction and clean up. The recovery of the ISs was on average $105 \pm 7\%$ for PCBs, $111 \pm 5\%$ for HCB, and $94 \pm 17\%$ DDX in sediment. For biota, the recoveries were on average $95 \pm 14\%$ for PCBs, $77 \pm 27\%$ for HCB, and $80 \pm 26\%$ for DDX, and for the passive samplers (SPMDs), the recovery was on average $79 \pm 20\%$ for PCBs, $59 \pm 34\%$ for HCB, and $86 \pm 18\%$ for DDX. For the POMs, the recovery was on average $114 \pm 9\%$ for PCBs, $114 \pm 19\%$ for HCB, and $94 \pm 10\%$ for DDX. Blanks showed little or no contamination, but average blank levels were subtracted from samples when present. The only exception was for HCB in POM blanks, where blank levels were similar to those in sediment-exposed POMs. Due to this, HCB pore water concentrations from the river estuary transect were excluded from the data evaluation.

3 Results and discussion

3.1 Sediment-to-water flux

The sediment-to-water flux (ng m^{-2} day^{-1}) of dissolved Σ_7PCBs, Σ_{20}PCBs, Σ_6DDX, and HCB at the different sediment types in Väja and Sandviken are shown in Fig. 2 and Table S8. The flux of Σ_7PCB was approximately two times higher in Väja fiberbank (3.0 ng m^{-2} day^{-1}) compared to Sandviken fiberbank (1.3 ng m^{-2} day^{-1}). The fluxes from fiberbanks were elevated compared to the fluxes measured in the fiber-rich sediment areas (Väja 0.71 ng m^{-2} day^{-1} and Sandviken 0.29 ng m^{-2} day^{-1}) and in the reference sites placed in areas with more natural minerogenic sediment (Väja 0.035 ng m^{-2} day^{-1} and Sandviken 0.38 ng m^{-2} day^{-1}). For corresponding fluxes of Σ_{20}PCB, see Fig. 2 and Table S8. The flux of DDT and its transformation products could only be determined at levels above LOQ in the fiberbank in Väja (2.6 ng m^{-2} day^{-1}) and Sandviken (0.23 ng m^{-2} day^{-1}), not in the fiber-rich sediment or the reference sites. On the other hand, the flux of HCB was similar across the three sediment types in both Väja (0.29–0.33 ng m^{-2} day^{-1}) and Sandviken (0.20–0.37 ng m^{-2} day^{-1}). The fiberbank PCB fluxes were similar to those measured from another contaminated (but not fiber-containing) sediment area in Sweden (Σ_7PCBs: 7.5 ng m^{-2} day^{-1}) using a similar (closed) benthic flux chamber (Mustajarvi et al. 2017). The higher fluxes from the fiberbanks compared to the fiber-rich sediments are
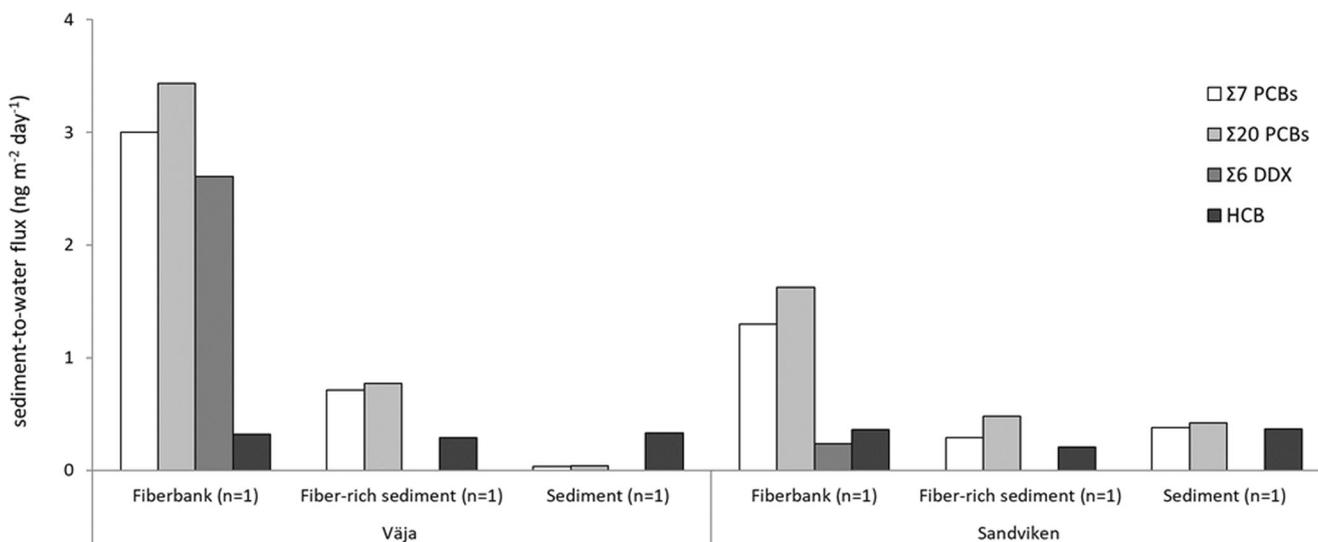

Fig. 2 Sediment-to-water flux (ng m^{-2} day^{-2}) of Σ_7PCBs, Σ_{20}PCBs, Σ_6DDX, and HCB from different sediment types in Väja and Sandviken

reasonable based on elevated PCB and DDX levels in fiberbank sediment and pore water as previously reported (Dahlberg et al. 2020). As expected, the sediment-to-water flux of Σ_7PCBs and Σ_6DDX was found to be significantly correlated to the sediment pore water concentration (Σ_7PCBs Pearson, $p < 0.05$, $r^2 = 0.70$; Σ_6DDX Pearson, $p < 0.05$, $r^2 = 0.84$) but not to the sediment concentration (ng g^{-1} TOC) (Fig. S6). The HCB levels in pore water and sediment showed low variation across sediment types (Dahlberg et al. 2020), and accordingly, the flux of HCB could not be correlated to neither concentrations in sediment nor pore water (Fig. S6). The higher flux of PCBs in Väja fiberbank compared to Sandviken fiberbank (Fig. 2), despite showing similar pore water levels (Dahlberg et al. 2020), indicate that additional factor(s) influence the contaminant flux. For example, additional sediment sampling revealed that the Sandviken fiberbank is covered by a ~ 10 cm natural cap of recently settled fine-grained material characteristic of more natural sediment accumulation zones, whereas Väja lacked a visible layer of such material (Apler et al. 2020). It is possible that this natural layer of clay at Sandviken functions as an effective isolation barrier that restricts contaminant flux. This protection would explain the observed difference between Väja and Sandviken. A previous study also indicated that the cellulose material that Väja fiberbank is composed of is a poorer sorbent for POPs than the coarser wood material found in Sandviken fiberbank, which is surprising (Dahlberg et al. 2020). Furthermore, numerous pockmarks have been observed on the surface of the Väja fiberbank (see photo, Fig. S7) (Norrlin and Josefsson 2017). These pockmarks are the result of extensive gas ebullition. Gas ebullition, induced by degradation of organic material in the sediment, has been shown to boost contaminant transport by enhanced pore water advection and particle resuspension (Yuan et al. 2007).

Different sampling techniques can be used to study diffusive flux of hydrophobic organic contaminants at the sediment-water interface, which are reviewed elsewhere (Liu et al. 2014). The advantage of the benthic flux chamber approach used in this study is that the condition present at the sediment-water interface is undisturbed during the flux measurement. However, the closed chamber design and the long sampling time (32–34 days) can lead to anoxic conditions and underestimation of biota-driven contaminant flux in sediments where benthic biota is normally present. Due to the prevailing anoxic conditions at fiberbank sites, biota is absent anyway, but fluxes from fiber-rich sediment and minerogenic sediment where biota is present may have been underestimated.

3.2 Resuspension of particles and particle-bound contaminants

The amount of suspended particles (mg L^{-1}) in the collected bottom water is presented in Fig. S8 and Table S9. A similar

quantity of particles was found in the bottom water collected above the undisturbed seafloor in Väja fiberbank (geometric mean (GM): 4.3 mg L^{-1}), Väja fiber-rich sediment (GM: 4.6 mg L^{-1}), Sandviken fiberbank (GM: 5.3 mg L^{-1}), and at the reference site (S17) not affected by fibers (4.8 mg L^{-1}). When artificially disturbing the sediment (to mimic wave and current erosion), the amount of suspended particles increased as intended at all investigated sites: Väja fiberbank (GM: 11 mg L^{-1}), Väja fiber-rich sediment (GM: 11 mg L^{-1}), Sandviken fiberbank (GM: 9.1 mg L^{-1}), and reference site (S17, 6.2 mg L^{-1}). However, no significant difference between the amount of particles released from the fiber-contaminated sites and the minerogenic reference site was found.

Similar amounts of POPs per gram particle were found in the bottom water collected above the undisturbed seafloor at all sites (Fig. 3 and Table S9). However, when the sediment surface was disturbed, the particle burden of Σ_{20}PCBs increased significantly (t-test, $p < 0.05$) in Väja fiberbank (GM: 15 ng g^{-1} particle) compared to Sandviken fiberbank (4.8 ng g^{-1} particle). This significant difference (t-test, $p < 0.05$) was also observed when the levels of particle-bound contaminants per unit of bottom water (pg L^{-1}) were studied (Fig. S9). This result indicates that larger amounts of contaminants can be dispersed via particle resuspension from Väja compared to Sandviken. It is possible that the natural cap covering the fiberbank in Sandviken hinders resuspension of the more contaminated fibers found below it.

The water sampling was performed at depths ranging from 11 to 69 m, and there is a risk that the sampling ship Ocean Surveyor might have caused unintentional resuspension during the water sampling.

3.3 Benthic organisms

3.3.1 Distribution of macrofauna in fibrous sediment

No benthic macro-organisms were found in any of the fiberbank samples (Table S10–11). Underwater camera photos showed large areas of the Väja fiberbank surfaces to be covered by the sulfur-oxidizing bacteria *Beggiatoa* sp. (see photo, Fig. S7), which illustrates the reducing conditions prevailing in the sediment. The lack of oxygen makes the fiberbanks hostile environments for macrofauna. Furthermore, no macro-organisms were found in the fiber-rich sediment in Väja ($n = 2$), whereas the fiber-rich sediment area in Sandviken ($n = 4$) contained both *Marenzelleria* sp. and *Saduria entomon*. In Kramfors, three out of four sampled sites contained *Marenzelleria* sp. whereas *S. entomon* was only found at one site (Table S10–11). The biomass of *Marenzelleria* sp. was not significantly different (unpaired t-test, $p > 0.05$) between the fiber-rich sediment ($n = 10$, 0–42 g ww m^{-2}) and more natural sediment ($n = 10$, 3.0–28 g ww m^{-}
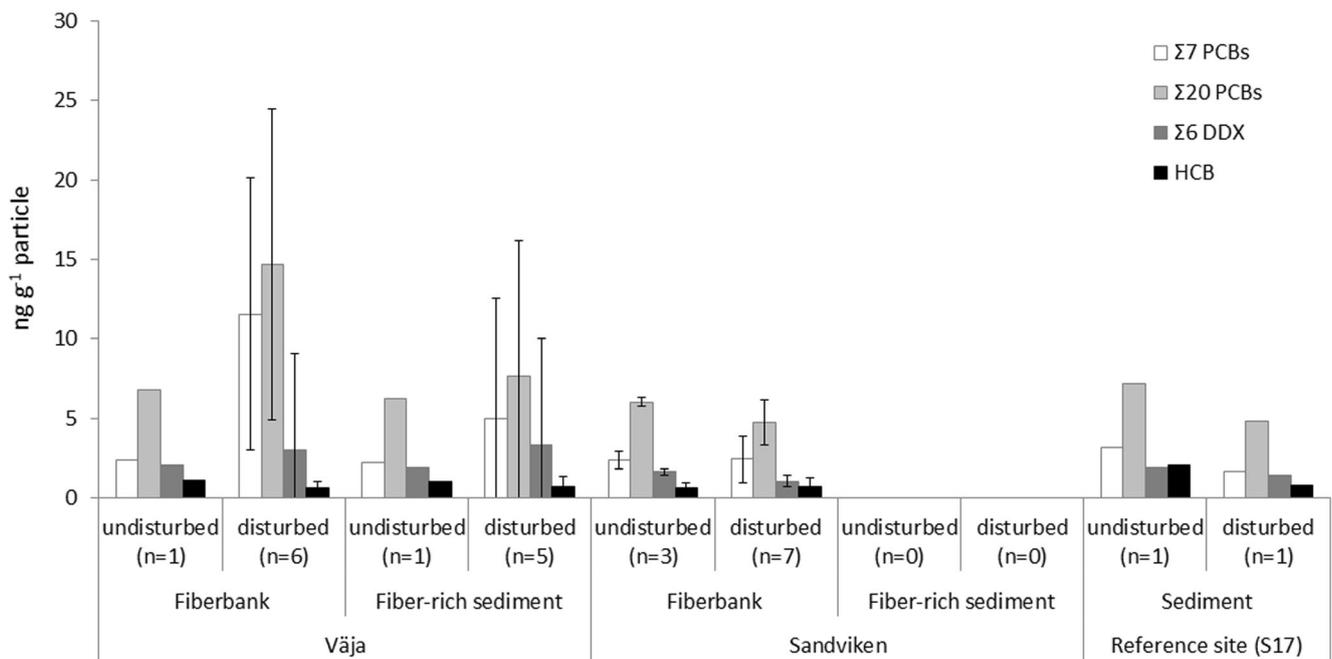
Fig. 3 Particle concentration of POPs (ng g^{-1} particles) in bottom water collected above the undisturbed seafloor (undisturbed) and after suspending the sediment (disturbed). Site S17 is a reference site, where more natural minerogenic sediment has accumulated

^{2}). For *S. entomon*, the biomass was 0–3.5 g ww m^{-2} in fiber-rich sediment ($n = 10$) and 0–22 g ww m^{-2} in more natural sediment ($n = 10$). The highest biomass of *S. entomon* was found at the off-shore site outside the Ångermanälven River estuary, where the bottom water is more saline. In the estuary transect, the species *Monoporeia affinis* was also found and retrieved when sieving the sediment but was not analyzed for pollutants. The presence of benthic macrofauna can enhance the sediment-to-water flux of contaminants (Meysman et al. 2006; Josefsson et al. 2010; Apell et al. 2018; Mustajarvi et al. 2019), but the lack of benthic macro-organisms in the three fiberbanks investigated suggests that contaminant release caused by bioturbation/bioirrigation is negligible at these sites.

3.3.2 Concentrations of POPs in macrofauna

The concentrations (ng g^{-1} lw) of Σ_7PCBs, Σ_{20}PCBs, Σ_6DDX, and HCB in biota are presented in Fig. 4 and Table S10–11. *S. entomon* was found to contain higher average (GM) levels of Σ_7PCBs (270 ng g^{-1} lw) and HCB (26 ng g^{-1} lw) than *Marenzelleria* sp. (Σ_7PCBs 130 ng g^{-1} lw and HCB 9.2 ng g^{-1} lw), whereas the levels of Σ_6DDX were similar in the two species (*S. entomon*: 36 ng g^{-1} lw; *Marenzelleria* sp.: 39 ng g^{-1} lw). The concentration of POPs in biota showed a weak positive relationship with contaminant levels in sediment (PCBs and DDX) and pore water (PCBs) for both *S. entomon* and *Marenzelleria* sp., but significant correlations were only found for Σ_{20}PCBs in *Marenzelleria* sp. (for sediment and pore water) and *S. entomon* (for sediment normalized on TOC) (Fig. S10).

3.3.3 Bioaccumulation factors (*BAF*s)

The bioaccumulation factor (*BAF$_{PW}$*) in this study describes the partitioning of compounds between organism and pore water. For the calculations of *BAF$_{PW}$*, only compounds detected above LOQ in both biota and pore water in spatially and temporally paired samples were included. The pore water concentrations are presented in Table S12 (for estuary transect samples) and in our previous study (Dahlberg et al. 2020). The average log *BAF$_{PW}$* in *Marenzelleria* sp. were in the range of 6.1–8.0 (PCBs), 4.6–6.5 (DDX), and 5.9 (HCB) for samples collected from fiber-rich sediment (Sandviken and Kramfors, $n = 6$) and 6.2–7.9 (PCBs), 5.8–7.4 (DDX), and 5.8 (HCB) for samples collected from more natural minerogenic sediment (Väja, Sandviken, and river transect $n = 10$). For *S. entomon*, the average log *BAF$_{PW}$* were in the range of 5.8–8.5 (PCBs), 5.1–6.7 (DDX), and 6.3 (HCB) in organisms collected from fiber-rich sediment (Kramfors and Sandviken, $n = 4$) and 5.5–8.4 (PCBs) and 6.2–8.0 (DDX) for organisms collected from more natural minerogenic sediment (river transect, $n = 3$). As expected, the *BAF$_{PW}$* were found to be well correlated with the octanol-water partitioning coefficient (K_{ow}) in both *Marenzelleria* sp. and *S. entomon* as shown in Fig. 5. The regression lines for *Marenzelleria* sp. and *S. entomon* collected from fiber-rich sediment differed significantly in intercepts ($p < 0.05$) but not their slopes, indicating slightly higher bioaccumulation across the compounds' hydrophobicity (K_{ow}) range in *S. entomon* compared to *Marenzelleria* sp. This difference in bioaccumulation between species may depend on different processes such as the route of

Fig. 4 Concentrations (ng g^{-1} lipid) of Σ_7PCB, Σ_{20}PCB, and Σ_6DDX in *Marenzelleria* sp. (upper graph) and *Saduria entomon* (lower graph) from fiberbank (FB), fiber-rich sediment (FRS), and minerogenic sediment (SED)
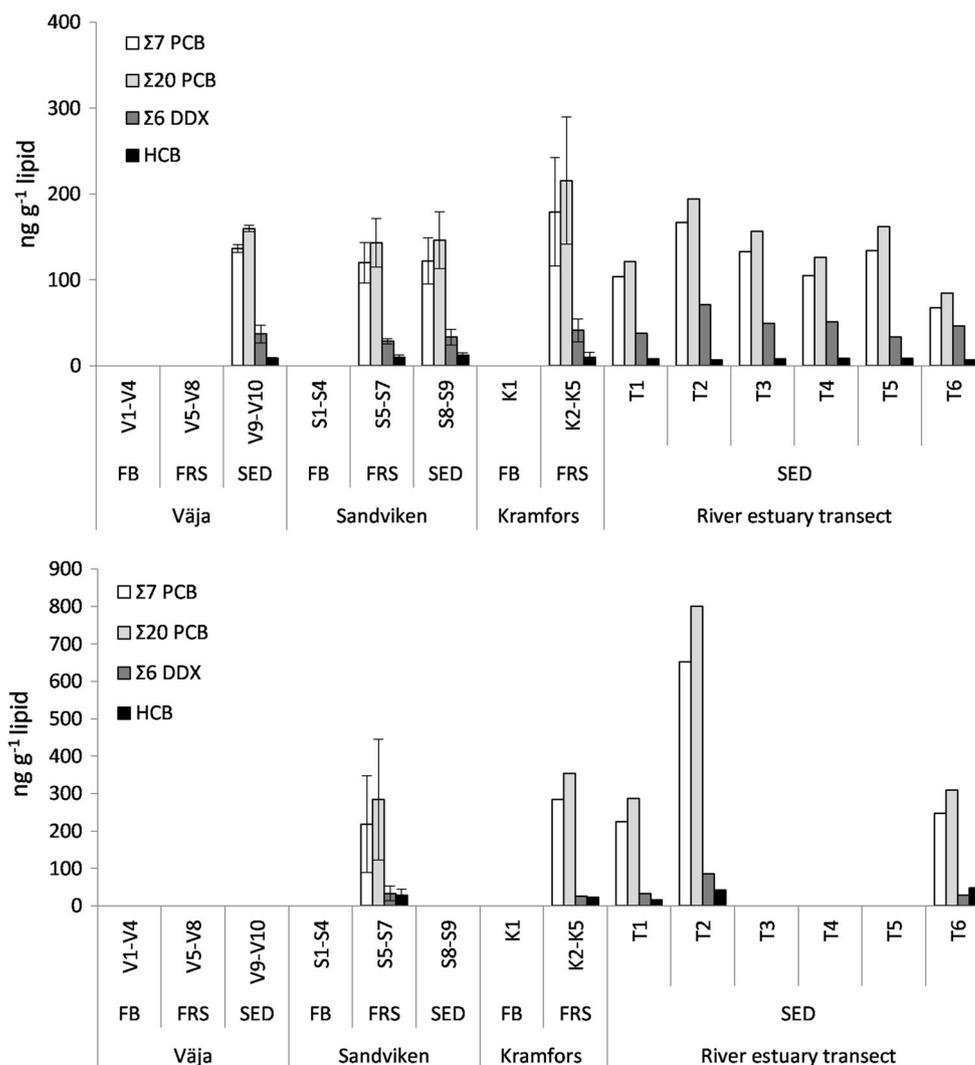
exposure and biomagnification, which will result in elevated concentrations at higher trophic levels. *Marenzelleria* sp. is a burrowing polychaete that feeds on phytoplankton and organic material in the sediment (Livetihavet 2019a), whereas *S. entomon* is a predator and scavenger that lives on the sediment surface and feeds on other benthic organisms (Livetihavet 2019b). *S. entomon* is food for many fish species in the Baltic Sea (e.g., cod and sculpin) (Livetihavet 2019b) and thus an important vector to transfer these contaminants to higher trophic levels.

3.3.4 Biota-sediment accumulation factors (*BSAF*s)

The biota-sediment accumulation factor (*BSAF*) describes the partitioning of compounds between organism (on a lipid weight basis) and sediment (on an organic carbon content basis). For the calculation of *BSAF*s, only compounds detected above LOQ in both biota and sediment in spatially and temporally paired samples were included. The sediment concentrations are presented in

Table S13 (for river transect samples) and in our previous study (Dahlberg et al. 2020). The average *BSAF* in *Marenzelleria* sp. ranged between 0.88–3.2 (PCBs), 0.21–3.1 (DDX), and 2.0 (HCB) in samples collected from fiber-rich sediment (Sandviken and Kramfors, $n = 6$) and 0.61–3.0 (PCBs), 0.76–4.6 (DDX), and 1.5 (HCB) in samples collected from more natural minerogenic sediment (Väja, Sandviken and river transect, $n = 10$). For *S. entomon* average *BSAF* collected from fiber-rich sediment (Kramfors and Sandviken, $n = 4$) ranged between 0.31–7.8 (PCBs), 0.92–3.5 (DDX), and 4.2 (HCB), whereas for *S. entomon* collected from more natural minerogenic sediment (river transect, $n = 3$), the average *BSAF* ranged between 0.21–12 (PCBs), 1.4–8.4 (DDX), and 6.2 (HCB). A bell-shaped correlation between *BSAF*s and log K_{ow} was observed for both species (Fig. 5), in agreement with other studies showing decreased *BSAF*s for PCBs above and below the log K_{ow} range of 6.0–7.3 in benthic species (Tracey and Hansen 1996) and in accordance with reduced bioavailability of higher chlorinated (larger) congeners (Bremle et al. 1995).
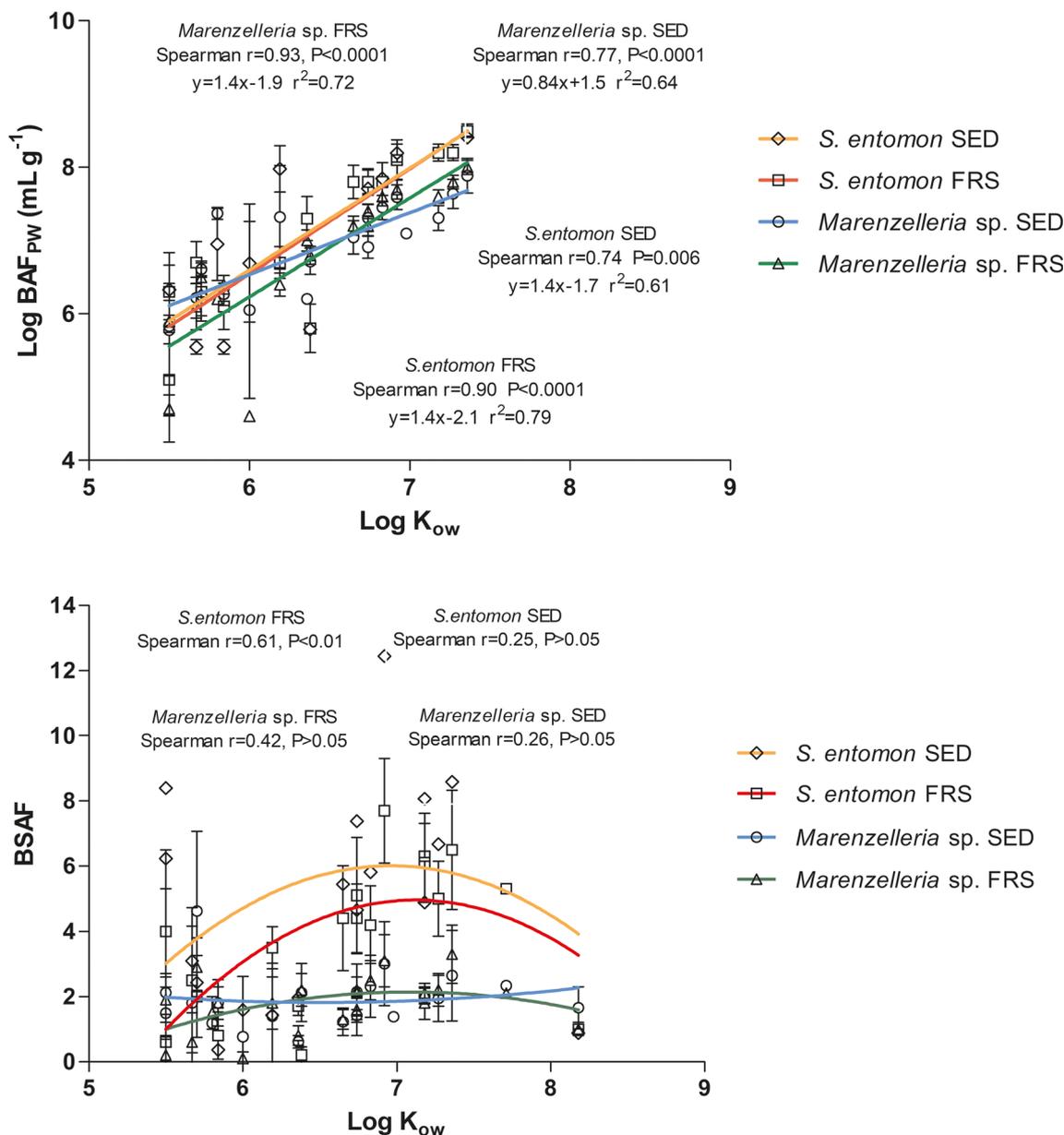
Fig. 5 Bioaccumulation factors (BAF_{PW}, upper graph) and biota-sediment-accumulation factors ($BSAF$s, lower graph) for PCBs, DDX, and HCB in *Marenzelleria* sp. and *S. entomon* collected from fiber-rich sediment (FRS) and minerogenic sediment (SED). Data are expressed as arithmetic means, and error bars show the standard deviations

3.3.5 Water column concentrations of POPs

Figure S11 and Table S14 show the concentration of dissolved contaminants in water (pg L^{-1}) sampled at two different depths above the Sandviken fiberbank (WS1, 7 and 12 m) and above the reference site with more natural, minerogenic sediments (WS2, 7 and 64.2 m). The bottom water above the seafloor of the fiberbank contained higher levels of Σ_7PCBs (5.5 pg L^{-1}) than water closer to the water surface (2.5 pg L^{-1}), which agrees with the sediment being a source of contaminants to the water column (Fig. 2). However, the bottom water at the minerogenic reference site contained levels of

Σ_7PCBs (4.3 pg L^{-1}) that were similar to the bottom water above the fiberbank. It was also unexpected to find that the surface water at the reference site contained higher concentrations of Σ_{20}PCBs (5.6 pg L^{-1}) than the bottom water above the fiberbank. The surface water at the reference site was also found to have higher concentrations of Σ_6DDX (4.3 pg L^{-1}) and HCB (8.7 pg L^{-1}) than the bottom water collected above the fiberbank (Σ_6DDX 1.9 pg L^{-1} and HCB 5.5 pg L^{-1}) and at the reference site (Σ_6DDX 2.9 pg L^{-1} and HCB 6.3 pg L^{-1}). The elevated concentrations of pollutants in water at the reference site might reflect that there are other contaminated areas along the Ångermanälven River (Apler et al. 2014);

Table 1 Quantitative comparison of the different dispersal pathways using Σ_7PCB as an example

Site	Sediment type	Area	Diffusive transport		Advective transport			Benthic uptake			
			Sediment to water flux of dissolved contaminants	Daily mass load of $_7$PCB	Particle associated contaminants at undisturbed conditions	Mass of particles corresponding to dissolved flux	Daily mass load of particles	Biomass of *Marenzelleria sp.*	Bioaccumulated contaminants in *Marenzelleria sp.*	Amount of biota needed to be eaten each day to correspond to the dissolved flux	Amount of biota that would correspond to total dissolved flux
		m²	ng m⁻² day⁻¹	ng day⁻¹	ng g⁻¹ dw particle	g dw particle m⁻² day⁻¹	kg dw particle day⁻¹	g ww m⁻²	ng g⁻¹ ww	%	kg ww biota day⁻¹
Väja	Fiberbank	70,000	3.0	210,000	2.4	1.3	88	0	n.a.	n.a.	n.a.
	Fiber-rich sediment	800,000	0.71	568,000	2.2	0.33	260	0	n.a.	n.a.	n.a.
	Sediment	70,000ᵃ	0.035	2450	n.s.	n.s	n.s.	9.0	1.8	0.19	12
Sandviken	Fiberbank	55,000	1.3	70,950	2.4	0.54	30	0	n.a.	n.a.	n.a.
	Fiber-rich sediment	550,000	0.29	159,500	n.s.	n.s.	n.s.	30	1.7	0.57	95
	Sediment	55,000ᵃ	0.38	20,900	3.2	0.12	6.6	19	1.6	1.3	13

ᵃ Sediment area estimated to be the same as corresponding fiberbank to simplify comparison; n.s. = not sampled; n.a. = not applicable; ww = wet weight; Σ_7PCB: sum of CB-28, CB-52, CB-101, CB-118, CB-138, CB-153, and CB-180

hence, the water is likely affected by additional pollution sources and not only the fiberbanks studied here. Furthermore, the difference in concentrations at the different depths may be affected by the pycnocline, present at around 12 m water depth, which prevents mixing of the upper and lower water masses at the reference site. The pycnocline is present due to different salinity and temperature in surface water (16 °C, 0 psu) and bottom water (4 °C, ~ 5 psu) causing stratification of the water mass in the river (Apler et al. 2019). Many fiberbanks are deposited at depths shallower than the pycnocline (12 m), and it is possible that the contaminants are dispersed into the upper layer of the waterbody.

3.4 Comparison of different transport processes

The lack of biota in the fiberbanks investigated indicates that there is no bioturbation/bioirrigation, at least not from macrofaunal movements, in the fiberbanks. However, there was still a high sediment-to-water flux of dissolved contaminants in the fiberbanks, especially in Väja, which might be caused by the lack of natural isolation cap that functions as protective barrier, and microbial gas production resulting in extensive gas ebullition that may enhance contaminant transport. Based on the current estimated size of the fiberbank in Väja (70,000 m^2) and Sandviken (55,000 m^2), the daily load was estimated to Σ_7PCB (210 µg/day), Σ_6DXX (180 µg/day), and HCB (22 µg/day) from Väja and Σ_7PCB (71 µg/day), Σ_6DXX (13 µg/day), and HCB (20 µg/day) from Sandviken. In Table 1, sediment-to-water flux was compared with the other dispersal pathways quantified in this study using Σ_7PCBs as an illustrative example. The amount of particle resuspension that corresponds to the sediment-to-water flux of Σ_7PCBs in Väja fiberbank during undisturbed conditions (as a more conservative approach than using disturbed conditions) was 1.3 g dw particle/m^2/day or 88 kg dw particle/day. In Sandviken fiberbank, 0.54 g dw particle/m^2/day or 30 kg dw particle/day corresponds to the sediment-to water flux of Σ_7PCBs. The flux of particles required for this advective pathway to exceed the diffusive pathway from sediment to water is very low. Sediment can probably be remobilized, during periods of high flow, illustrating how important particulate pathways are for hydrophobic contaminants.

In fiber-rich sediment, the sediment-to-water flux of Σ_7PCBs was lower than in the corresponding fiberbanks (Väja: 0.71 ng/m^2/day; Sandviken: 0.29 ng/m^2/day). However, due to their larger area, the fiber-rich sediment shows a higher daily load of Σ_7PCBs (Väja: 570 µg/day; Sandviken: 160 µg/day). Hence, more particle needs to be resuspended each day to correspond to this load (Väja; 260 kg dw particles/day) although the amount of particles per square meter is slightly smaller (Väja: 0.33 g dw particle/m^2/day). In the fiber-rich sediment, the macrofaunal species *S. entomon* and *Marenzelleria sp.* were found and

constituted a potential biotic transport route of contaminants. In Sandviken, the distribution of *Marenzelleria sp.* (GM 30 g ww m^{-2}) and the concentration of Σ_7PCBs (GM 1.7 ng g^{-1} ww) give an amount of 51 ng Σ_7PCBs m^{-2}. This value is approx. 200 times higher than the daily flux of dissolved Σ_7PCBs from the same area (0.29 ng m^{-2}) and indicates that less than 1% of the benthic biomass has to be ingested each day for the trophic transfer to be equivalent to the diffusive flux from the fiber-rich sediment in Sandviken. The fraction that is ingested is, of course, dependent on the extent of predation, but that 1% would be eaten each day is not unreasonable (e.g., Virnstein 1977; however, please note that predation may be very site-specific and also vary throughout the year).

Our study demonstrated the high importance of diffusive sediment-to-water flux as a dispersal pathway in fiberbanks and fiber-rich sediment under undisturbed conditions. This result is of great importance for risk assessment of this kind of sediments because the freely dissolved contaminants are more bioavailable than the particle-bound. However, the potential dispersal due to advective particle transport is not negligible, even under undisturbed conditions. It may however not be ruled out that gas ebullition may have an influence on our measurements. We emphasize that the magnitude of these processes is site-specific and that our calculations do not include particle resuspension under disturbed conditions. Ship traffic and sudden events like submarine landslides, which have been documented to have occurred in Väja (Apler et al. 2014), may result in severe particle resuspension and contaminant dispersal with elevation of dissolved concentrations as a secondary effect. In fiber-rich sediment, benthic uptake and potential transfer to higher trophic levels were also found to be of high importance.

4 Conclusions

This study quantified pollution dispersion from contaminated fibrous sediments. By comparing three different pathways of dispersal (sediment-to-water flux of dissolved contaminants, resuspension of contaminated particles, and uptake in benthic biota), none of the dispersal pathways could be ruled out as irrelevant. The flux of dissolved contaminants was significantly higher in the fiberbanks than in fiber-rich sediments or more natural minerogenic sediments. It was also higher at one of the fiberbank sites compared to the other, which is believed to be due to the lack of natural isolation cap and more gas ebullition at that site. In some fiber-rich sediment, where biota was present, uptake in biota and trophic transfer could also be considerable. The higher *BSAF*s in a predatory species compared to a deposit feeder also illustrate the first step of biomagnification from the benthic biota to organisms higher up in the food web. However, the lack of benthic biota on the fiberbanks excluded this dispersal pathway in the most contaminated sediment.

Quantification of dispersal routes and a better understanding of the relative importance of various pathways is critical for a proper risk assessment and risk management of contaminated sediment, including fiberbank areas, in the Baltic Sea and globally.

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