Chemosphere 85 (2011) 1444–1451

Contents lists available at SciVerse ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Influence of contaminant burial depth on the bioaccumulation of PCBs and PBDEs by two benthic invertebrates (*Monoporeia affinis* and *Marenzelleria* spp.)

Sarah Josefsson [a,*], Kjell Leonardsson [b], Jonas S. Gunnarsson [c], Karin Wiberg [a,1]

[a] *Department of Chemistry, Umeå University, SE-901 87 Umeå, Sweden*
[b] *Department of Wildlife, Fish and Environmental Studies, Swedish University of Agricultural Sciences, SE-901 83 Umeå, Sweden*
[c] *Department of Systems Ecology, Stockholm University, SE-106 91 Stockholm, Sweden*

A R T I C L E I N F O

Article history:
Received 24 March 2011
Received in revised form 30 June 2011
Accepted 6 August 2011
Available online 15 September 2011

Keywords:
Bioturbation
BSAFs
Sediment pollution
Baltic Sea
Invasive species

A B S T R A C T

The bioaccumulation of buried polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) added to specific depths in sediment (2.0–2.5, 5.0–5.5 and 10.0–10.5 cm) was studied in two infaunal species with similar feeding habits (surface deposit-feeders) but different bioturbation modes. The deep-burrowing polychaetes *Marenzelleria* spp. (*Mz*) displayed up to 36 times higher tissue concentrations of buried (spiked) contaminants than the surface-dwelling biodiffusing amphipod *Monoporeia affinis*. The differences in bioaccumulation were most pronounced for less hydrophobic contaminants due to the bioirrigating activity of *Mz*. Contaminants buried at shallow depths displayed higher accumulation than more deeply buried contaminants. In contrast, the bioaccumulation of unspiked (native) contaminants with a uniform vertical distribution in the sediment was similar between the species. For *Mz*, the BSAFs increased with increased K_{OW} for the uniformly distributed contaminants, but decreased for the buried contaminants, which indicates that the dominant uptake routes of the buried contaminants can differ from the uniformly distributed contaminants. The surface sediment concentration of buried contaminants increased in *Mz* treatments, showing that *Mz* bioturbation can remobilize historically buried contaminants to the biologically active surface layer and increase the exposure for surface-dwelling species.

1. Introduction

Contaminated sediments function as sources of persistent organic pollutants (POPs) to sediment-living biota (infauna) and to the overlying water column and its biota. Benthic organisms play an important role in the transport of POPs from sediment to pelagic biota. Due to their bioturbation, i.e. mixing of particles and pore-water in the sediment, they can increase the flux of dissolved and particle-associated POPs to the water phase (Karickhoff and Morris, 1985; Thibodeaux, 2005). They can also transfer contaminants to other species in the benthic-pelagic food web when preyed upon (Kobayashi et al., 2011).

As a result of the close contact between infauna and sediment, concentrations of POPs in infauna can be high. The two most important routes for uptake in infauna are uptake from pore-water (Oliver, 1987; Shaw and Connell, 1987) and from ingested particles (Boese et al., 1990; Forbes et al., 1998). The relative importance of various uptake routes is believed to be largely governed by the hydrophobicity of the chemical (Landrum, 1989; Thomann et al., 1992; Lu et al., 2004). For less hydrophobic compounds, uptake from sediment pore-water is an important route, with the uptake rate controlled by the kinetics of desorption from sediment, while for more hydrophobic compounds, uptake from ingestion of sediment particles becomes more dominant. The importance of feeding strategies for the accumulation of POPs has also been demonstrated (e.g. Kaag et al., 1997; Magnusson et al., 2003).

The release of buried contaminants from sediment and their subsequent uptake by aquatic organisms is becoming an increasingly important issue, since the primary sources of many POPs have been reduced substantially during the last decades (e.g. Quass et al., 2004). As a consequence, the concentrations can be elevated in deeper layers of sediment compared to the sediment surface layer (Jonsson et al., 2000; Isosaari et al., 2002). However, the influence of contaminant burial depth on uptake in infauna has so far received little attention. Buried contaminants can be considered unavailable to the aquatic ecosystem, but conditions may change with the arrival of new benthic species. For instance, the recent invasion of the deep-burrowing polychaetes *Marenzelleria* spp. in the Baltic Sea has raised concerns for increased remobilization of previously buried contaminants, as *Marenzelleria* spp. have become

---

[*] Corresponding author. Present address: Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, SE-750 07 Uppsala, Sweden. Tel.: +46 18 673032; fax: +46 18 673156.

E-mail address: sarah.josefsson@slu.se (S. Josefsson).

[1] Present address: Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, SE-750 07 Uppsala, Sweden.

0045-6535/$ - see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.chemosphere.2011.08.024

dominating species in the benthic community in many areas of the Baltic Sea (Zettler et al., 1995; Zmudzinski, 1996).

We hypothesized that the accumulation of buried POPs by benthic species depends on the vertical distribution of both the contaminants and the species, and examined this by studying the accumulation of polychlorinated biphenyl (PCB) and polybrominated diphenyl ether (PBDE) congeners by two infaunal surface deposit-feeders, *Marenzelleria* spp. (*Mz*) and *Monoporeia affinis* (*Mo*). Even though the two species feed at the same location in the sediment (Dauer et al., 1981; Byren et al., 2006), they differ in their vertical distribution. The amphipod *Mo* lives near the sediment surface (Albertsson and Leonardsson, 2000), while the spionid polychaetes *Mz* can construct mucus-lined tubes down to 50 cm (Dale and Miller, 2008). Our main research objective was to investigate if the bioaccumulation of POP congeners that were buried (spiked) at different sediment depths differed between the surface-dwelling *Mo* and the deep-burrowing *Mz*. We also examined the bioaccumulation of native (unspiked) PCBs, the transport of buried POPs in the sediment profile, and the importance of different contaminant uptake routes.

2. Materials and methods

A detailed description of materials and methods is available in a companion paper (Josefsson et al., 2010), but essential or new information is presented below.

2.1. Preparation of sediment and aquaria

Three layers of contaminated sediment were placed in each aquarium, at depths of 2.0–2.5, 5.0–5.5, and 10.0–10.5 cm below the sediment surface. Each layer was spiked with different PCB and PBDE congeners, with one congener from each of the homologue groups *tri*- to *hepta*-CBs and *tri*-, *penta*- and *hepta*-BDEs (Fig. S1 and Table S1 in Supplementary material). In total, each layer contained 5 PCB congeners and 3 PBDE congeners at elevated concentrations (1.0 μg per aquarium for *penta*- to *hepta*-CBs; 1.5 μg per aquarium for PBDEs, *tri*- and *tetra*-CBs), and the different congeners were specific for one of the contaminated layers.

The layers were prepared by spiking field-collected sediment with unlabelled (^{12}C) PCB and PBDE congeners, uncommon in environmental samples, and storing it in dark and cold for 30 d for the POPs to equilibrate between sediment and pore-water (Landrum et al., 2002). Thereafter, the sediment was mixed and frozen in thin layers (0.5 cm) before being placed at the specific depths in the experimental aquaria. Sediment not spiked with contaminants (i.e. containing only native contaminants) was placed between the layers. The total sediment depth was 12 cm and the height of the overlying water column was 13 cm.

2.2. Experimental setup and biota characteristics

The experiment consisted of two treatments: (i) *M. affinis* (*Mo*) at a density of 220 individuals m^{-2} ($n = 4$), and (ii) *Marenzelleria* spp. (*Mz*) at a density of 1200 individuals m^{-2} ($n = 5$). The densities corresponded to normal-high densities in the field (northern Baltic Sea) at the time of the experiments, but low for *Mo* when compared to earlier decades. The animals used in the experiment were collected from the same location as the sediment. *Mz* is an invading genus in the Baltic Sea, consisting of three morphologically similar sibling species: *M. viridis*, *M. neglecta* and *M. arctia*. The population in this study consisted of both *M. viridis* and *M. neglecta* (Josefsson et al., 2010).

At the density used in this experiment, *Mo* inhabits and bioturbates approx. the top 2 cm of sediment in the field, while at extremely high densities (10 000 individuals m^{-2}), the mixed layer may extend down to 7 cm (Albertsson and Leonardsson, 2000). As mentioned, *Mz* construct tubes that can extend down to 50 cm (Dale and Miller, 2008). In addition to having different burrowing depths, the species also have different bioturbation modes. *Mo* can be classified as a biodiffusor, i.e. randomly mixing sediment particles over short distances. *Mz* is a bioirrigator/gallery-diffusor, mainly impacting the sediment by digging and irrigating its burrows.

2.3. Sampling

The experiment was terminated 56 d after the animals had been added to the aquaria. The sediment was sampled and sliced into 1 cm layers using a small core sampler (ID 15 mm; 5 cores pooled from each aquarium). The sediment layer was 12 cm when placed in the aquaria at the start of the experiment, but compacted to 11.2 ± 0.3 cm (average ± 95% confidence intervals; $n = 13$) after 1.5 weeks. An additional 1.5 weeks later, just before the addition of animals, the thickness was 10.8 ± 0.2 cm. During the 8-week animal exposure period, the sediment depth did not change markedly, and measured 10.8 ± 0.3 cm also at the end of the experiment. During the sampling at the end of the experiment, the core sampler compacted the sediment and the bottom sediment was difficult to retrieve. Therefore, the original 12 cm of sediment was divided into nine 1 cm slices at the sampling.

After the sediment sampling, animals were sieved from the sediment and put in clean water for 24 h to empty their guts. The wet weight of each species for each aquarium was determined after removing excess water on a dry filter paper. The average survival was 75% (range 70–80%) for *Mo* and 85% (range 80–94%) for *Mz*. Animals not exposed to the spiked sediment were also analyzed. In addition, the water column was sampled during the experiment to determine the fluxes of contaminants from sediment to water, see results in Josefsson et al. (2010).

2.4. Analytical procedures

Sediment was Soxhlet–Dean–Stark extracted for 24 h, and before extraction ^{13}C-labelled PCBs and PBDEs were added as internal standards. The cleanup followed the description by Josefsson et al. (Josefsson et al., 2010), with additional sulphur removal with activated copper. Biota samples were mixed with Na_2SO_4, and extracted with acetone/*n*-hexane (5/2 v/v) and *n*-hexane/diethylether (9/1 v/v). The lipid content was determined gravimetrically. Instrumental analysis was performed with GC-HRMS, and the isotope dilution method was used for quantification.

2.5. QA/QC

The average recovery of the internal standards was 93% for PCBs and 87% for PBDEs. For a positive detection and quantification, a signal-to-noise ratio >3 of the chromatographic peak was required. As this limit of detection (LOD) was high for spiked *hepta*-BDEs, and they were only detected in 2 of 9 biota replicates, this homologue group was excluded from the results. In laboratory blanks, 3 of 24 spiked PCB and PBDE congeners were detected, but the amounts never exceeded 20% of the amounts in samples. The spiked congener CB 182 coelutes with the native congener CB 187 on the columns used (Frame, 1997); this congener (CB 182 + 187) was included in the results, but excluded from further discussions as it cannot be classified as a solely spiked or native congener. Amounts of spiked or native congeners detected in blanks were subtracted from amounts in samples, and the native CB 52 was excluded due to high background levels. Amounts detected in unexposed animals (sampled just before placement in