IN VIVO BIOACCUMULATION OF CONTAMINANTS FROM HISTORICALLY POLLUTED SEDIMENTS – RELATION TO BIOAVAILABILITY ESTIMATES

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Abstract

Many contaminants are recalcitrant against degradation. Therefore, when primary sources have been discontinued, contaminated sediments often function as important secondary pollution sources. Since the management and potential remediation of contaminated marine sediments may be very costly, it is important that the environmental risks of contaminants present in these sediments and benefits of remediation are evaluated as accurately as possible. The objective of this study was to evaluate the bioavailability of common organochlorine contaminants and polycyclic aromatic hydrocarbons (PAHs) in selected polluted sediments from Norway by simple generic sorption models (free energy relationships), as well as by pore water concentration measurements. Furthermore, the aim was to predict bioaccumulation from these bioavailability estimates for comparison with *in vivo* bioaccumulation assessments using ragworm (*Nereis virens*) and netted dogwhelk (*Hinia reticulata*). Predicted biota-to-sediment accumulation factors (BSAFs) derived from pore water concentration estimates were in better agreement with the bioaccumulation observed in the test organisms, than the generic BSAFs expected based on linear sorption models. The results therefore support that site-specific evaluations of bioaccumulation provide useful information for more accurate risk assessments. A need for increased knowledge of the specific characteristics of benthic organisms, which may influence the exposure, uptake and elimination of contaminants, is however emphasized.

Key Words: Bioavailability, Bioaccumulation, Organochlorine compounds, Polycyclic Aromatic Hydrocarbons, Sediment

1. INTRODUCTION

In Norway, several estuaries and harbours have hosted a varied mix of industries and therefore have been the recipients of a range of different environmental contaminants. Many of the primary sources and discharges to the marine environment have been discontinued or markedly reduced *inter alia* because of international agreements, such as the Stockholm Convention on Persistent Organic Pollutants (POPs) of the United Nations Environment Program (UNEP), resulting in banning or phasing out of several environmental contaminants. However, since many of these contaminants degrade slowly and because of their nonpolar properties have a high affinity for particles, contaminated sediments can still remain as important secondary pollution sources for a long time after primary sources have been stopped.

Among environmental challenges in Norway are thus the management and potential remediation of these contaminated marine sediments. The Norwegian Food Safety Authorities have indeed issued food consumption advisories for specific seafood items in several fjord localities along the coast. Efforts are now being made to eliminate or reduce the potential of contaminated sediments as sources of contaminants to the ecosystem. Risk assessment guidelines for contaminated marine sediments have been developed (Bakke et al., 2010; SFT, 2007) to provide an accessible tool for authorities, stake holders, consultants and environmental managers (especially the Climate and Pollution Agency, Klif; formerly Norwegian Pollution Control Authority, SFT) to assess the present environmental and human health risks from sediments. The rationale is that remedial actions should be based on sound risk assessment. Since sediment remediation is very costly it is important that the environmental benefits are predictable and measurable. The quality criteria of the guidelines are harmonized with the risk assessment principles of the European Union (EC,

2003). In order to benefit from already available knowledge (in terms of structure and coherence of methods), existing guidance systems were reviewed during the development of the Norwegian guidelines. Besides the EU technical guidelines, examples of reviewed systems were Dutch tools for risk assessment of dispersion of contaminants from sediments, as well as US and Canadian guidelines for risk assessment. A common feature of the reviewed methods is that they integrate physical, chemical and biological elements in the risk assessment.

Risk assessment of contaminated sediments needs to be feasible for a large number of actors, thus the guidelines are of a generic nature. The initial approach of the risk assessments is to compare total contaminant concentrations in the sediments (normalised to the solid phase) with fixed environmental quality standards (Bakke et al., 2010; EC, 2003). However, risk may be overestimated, as limit values are deduced from generic sorption parameters, only considering sorption of contaminants to natural organic matter (Ruus et al., 2010; van der Heijden and Jonker, 2009). Many studies, however, have shown that different sedimentary organic matter can be composed of different sorption domains within the particle phase resulting in varying degrees of binding strengths (Arp et al., 2009; Cornelissen et al., 2006a). Thus, a cost-effective approach to ensure sound remediation plans may include measurements of bioavailable concentration estimates or bioaccumulation (Lu et al., 2003; Lu et al., 2011). To this end the use of passive samplers and solid phase extraction techniques has been a much used approach (Cornelissen et al., 2006a; Cornelissen et al., 2006b; Gschwend et al., 2011; Lu et al., 2011; van der Heijden and Jonker, 2009). This is also suggested in the Norwegian guidelines for risk assessment of contaminated sediments (SFT, 2007). A recent evaluation of the most influential factors for the dispersion of contaminants from sediments in the guidelines identified the sediment:water

partition coefficient (K_d) and the bioconcentration factor (BCF) among the parameters of which accurate estimates are required for a sound risk assessment (Saloranta et al., 2011).

The objective of this study was to estimate the bioavailability of common organochlorine contaminants and polycyclic aromatic hydrocarbons (PAHs) in selected polluted sediments from Norway by simple generic sorption models (free energy relationships). Measurements of pore water concentrations were previously performed in the same sediments (Allan et al., 2012), and the aim was to predict bioaccumulation from both above mentioned bioavailability estimates for comparison with *in vivo* bioaccumulation assessments.

In vivo bioaccumulation assessments were performed on ragworm (*Nereis virens*, Polychaeta) and netted dogwhelk (*Hinia reticulata*, Gastropoda), while measurements of freely dissolved pore water concentrations were done using polyethylene passive samplers (Allan et al., 2012). The selected sediments originated from different localities in Norway, representative of different types of pollution sources and organic matter content.

2. MATERIAL AND METHODS

2.1 Sediment sampling

Sediments from Aker Brygge (59° 54.277 N, 10° 42.985 E; 15 m depth; South-Eastern Norway), Kristiansand harbour (58° 07.495 N, 07° 58.632 E; 28 m depth; Southern Norway), the Frierfjord (59° 06.768 N, 09° 36.963 E; 48 m depth; South-Eastern Norway) and the Outer Oslofjord (59° 29.035 N, 10° 36.949 E; 32 m depth; South-Eastern Norway) were collected using a box corer (USNEL 0,25 m^2 box corer). Triplicate box cores were collected from each site and brought

intact/undisturbed to NIVA's marine research facility at Solbergstrand (Berge et al., 1986) for the experiment.

Aker Brygge is the site of a former shipyard in the Inner Oslofjord (South-Eastern Norway). Frierfjord (the Grenlandfjord area; South-Eastern Norway) has a 50-year long pollution history, where main emissions were organochlorine compounds from a magnesium smelter and PAHs from a ferro-manganese plant. The Kristiansand harbour area (Southern Norway) was contaminated with organochlorine compounds from a metal refinery and PAHs from an electrode paste factory using coal tar pitch. The Outer Oslofjord (South-Eastern Norway) sediment was from a relatively clean site and served as control/reference.

2.2 Chemicals

Solvents and other chemicals used are listed in Supplementary Material.

2.3 Organisms

In vivo bioaccumulation assessments were performed using *Nereis virens* (Polychaeta) and *Hinia reticulata* (Gastropoda). *Nereis virens* were purchased from Seabait Ltd. (Ashington Northumberland, UK), and brought to Solbergstrand by air freight and car. *Hinia reticulata* were collected at a site in the Outer Oslofjord, described earlier (Ruus et al., 2005). After an acclimation period of ≥ 7 days, the organisms were added to the experimental boxes (triplicate boxes form each site).

These generally abundant species intimately interact with the sediment (Hayward and Ryland, 1995). *N. virens* and *H. reticulata* both prefer sandy or muddy sediment. *H. reticulata* is primarily a scavenger, whereas *N. virens* is omnivorous.

Before and during the experiments, the organisms were fed Skretting advanced fish feed (Coarse fish - 23. Skretting, Roman Island, Westfort Co., Mayo, Ireland).

2.4 Passive sampling measurements of freely dissolved concentrations in pore water

The measurement of freely dissolved pore water concentrations was undertaken using low density polyethylene (LDPE) membrane in batch experimental exposure of 3 to 50 days. Concentrations in the pore water (C_{PW}) and resulting total organic carbon-normalised sediment-water partition coefficients (K_{OC}) for the three sediments under investigation (one box from each site) are given in Allan et al. (2012), along with the methods applied (preparation of LDPE passive samplers, LDPE exposure in sediment slurries, extraction and analyses of LDPE samplers).

2.5 Experimental setup

In the mesocosm, the boxes with intact/undisturbed sediments (triplicates per site) were submersed to 1-2 cm below the rim in a basin continuously flushed with seawater with salinity, temperature and dissolved oxygen ranging from 33.6 to 34.9 PSU, 6.3-8.3 ˚C, and 7-9 mg O_2 L^{-1}, respectively. Salinity, temperature and oxygen saturation were logged with WTW (Wissenschaftlich-Technische Werkstätten GmbH, Weilheim, Germany) electrodes every minute in the primary header tank. The seawater was supplied through a pipe-line from 60 m depth in the Oslofjord outside the sill at Drøbak. Separate flows of the same source water was used for

continuous exchange of the overlying water in each box-cosm and an air-lift system (Schaanning et al., 2006) was used to ensure a well-mixed, oxygen saturated overlying water. Each box (area: 0.25 m^2) contained 62-75 L of sediment, with 25-38 L of overlying water. Twenty-two ragworms and 50 netted dog whelk were added to each box (together), and the duration of the exposure was 28 days, as recommended by Lee et al. (1991).

Upon termination of the experiment the overlying water in each box was carefully removed. The organisms from each box were carefully retrieved, and the sediment (within each box) was thoroughly mixed. Aliquots of sediments were taken for chemical analysis and LDPE exposure (pore water concentration measurements; see Allan et al. (2012) for details), as well as analyses of other sediment properties; sediment porosity, fine fraction (% dry wt. <63 µm) and Total Organic Carbon (TOC) content.

The soft parts of *H. reticulata* were removed from their hard shells using a nut-cracker. The soft parts, as well as individuals of *N. virens* were then rinsed in seawater and transferred to glass containers before storage at -20 °C until chemical analysis.

2.6 Extraction and analyses of organisms and sediments

Sediment samples were mixed with Hydromatrix, while adding internal standards (see below), and extracted using Accelerated Solvent Extraction (Dionex ASE-200; Dionex Corp. Sunnyvale, CA, USA; temperature and pressure of 100°C and 2000 psi, respectively), using a mixture of cyclohexane and dichloromethane (1:1, vol:vol). Clean-up of samples was done by Gel Permeation Chromatography (GPC), using dichloromethane as mobile phase (applies to both PAHs and organochlorine compounds). For organochlorine compounds, extracts were further

175 treated with concentrated sulphuric acid (H_2SO_4) prior to analysis. Analysis by gas
176 chromatography and mass spectrometry (GC-MS; PAHs) or electron capture detection (GC-
177 ECD) was done as described for the organisms, below (see Supplementary material for details).
178
179 The organisms (*N. virens* and *H. reticulata*) were each homogenised, using an ultra Turrax™.
180 Internal standards (for PAHs: naphthalene-d8, acenaphthene-d8, phenanthrene-d10, chrysene-
181 d12, perylene-d12, and anthracene-d10; for organochlorine compounds: PCB-30, -53 and -204)
182 were added. For PAH analysis, the samples were saponified, and the PAH compounds were
183 extracted with *n*-pentane and dried over sodium sulphate, before the solvent volume was reduced
184 and exchanged to dichloromethane. The resulting extracts were then cleaned by GPC and the
185 solvent exchanged to cyclohexane. The organochlorine compounds were extracted twice with
186 cyclohexane and acetone (4:3, vol:vol) by ultrasonication (3 min). The extracts were then washed
187 with saline water (0.5%) before the extraction volume was reduced and the solvent exchanged to
188 dichloromethane. After GPC cleanup, the solvent was exchanged to cyclohexane. Further cleanup
189 was done with concentrated H_2SO_4.
190
191 Extracts for PAH and organochlorine determination were analysed by GC-MS and GC-ECD,
192 respectively, as previously described (Ruus et al., 2005; Ruus et al., 2010; see Supplementary
193 material for details). The detection limit was defined as >3 times signal noise and was from <0.05
194 to <2 ng g^{-1}(wet wt.; biota) or <0.5 to <50 ng g^{-1}(dry wt.; sediment), dependent on compound and
195 matrix. Further quality control (QA/QS) details are as follows: The laboratory is accredited by the
196 Norwegian Accreditation as a testing laboratory according to the requirements of NS-EN
197 ISO/IEC 17025 (2000). Furthermore, analytical quality control of the laboratory is also ensured
198 by the participation in international calibration tests, including QUASIMEME (Quality

Assurance of Information for Marine Environmental Monitoring in Europe) twice per year. The certified reference material used was SRM 1944 (National Institute of Standards and Technology, Gaithersburg, MD, USA) and an in-house reference material (blue mussel) was also used to ensure reproducibility. Recoveries were 84-125 % (PAHs in biota), 82-130 % (organochlorine compounds in biota), 36-122 % (PAHs in sediment) and 37-135 % (organochlorine compounds in sediment).

2.7 Analysis of lipid, total dry matter, organic carbon and particle size fraction.

After lipid extraction (cyclohexane and acetone), aliquots of the homogenised organism material were used to determine the lipid content gravimetrically. Total dry matter in sediment sub-samples was also analysed gravimetrically. TOC was obtained by catalytic combustion (1800 °C; Carlo Erba 1106 elemental analyser; Carlo Erba SpA, Rodano, Italy) of freeze-dried, crushed and acidified (1N HCl) sediment sub-samples. Proportion (% dry wt.) of particles with size <63 µm was analysed according to the methods described by Krumbein and Pettijohn (1938).

2.8 Calculation of Biota-to-Sediment-Accumulation-Factors (BSAFs)

Biota-to-Sediment-Accumulation-Factors (BSAFs) were predicted from generic (linear, one domain) sorption models, following Karickhoff et al. (1979) and Schwarzenbach et al. (2003) (for deduction, see Supplementary Material):

$BSAF_{Predicted\ (Karickhoff)} = 1.61$

or

$BSAF_{Predicted\ (Schwarzenbach)} = 2.08K_{OW}^{0.02}$ (for PAHs)

or

$BSAF_{Predicted\ (Schwarzenbach)} = 0.71K_{OW}^{0.26}$ (for organochlorine compounds)

Furthermore, BSAFs were predicted from pore water concentration measurements as follows:

$$\mathrm{BSAF}_{\mathrm{Predicted\,(Porewater)}} = \frac{C_{lipid}}{C_{OC}} = \frac{K_{lipid} \cdot C_{PW}}{\left(\frac{C_S}{f_{OC}}\right)}$$

where C_{lipid} is the lipid normalised concentration in the organism (here estimated from K_{lipid} and C_{PW}), C_{OC} the organic carbon normalised concentration in the sediment, C_{PW} the concentration in porewater, C_S is the concentration of the compound in the sediment (µg kg^{-1} dry wt.), f_{OC} is the fraction of organic carbon content in the sediment (dry:dry), and $K_{lipid} = C_{lipid}/C_{PW} = K_{OW}$.

Finally, BSAFs were calculated from observed concentration in the organisms (*N. virens* and *H. reticulata*) applied in the experiments:

$$\mathrm{BSAF}_{\mathrm{Observed}} = (C_{organism}/f_{lipid})/(C_S/f_{OC})$$

where $C_{organism}$ is the dry weight concentration in the organism, and f_{lipid} is the fraction of tissue lipid (dry wt.)

A conceptual sketch of the approaches to deduce and compare BSAFs is given in Figure S1 (see Supplementary Material).

3. RESULTS AND DISCUSSION

The sediments tested in this study contained a wide range of PAH and especially organochlorine concentrations, with high abundance of some compounds very specific to the pollution source (Tables S1-S2, Supplementary Material). All three sediments displayed a PAH contamination of pyrogenic origin, as the proportions of the larger molecules were high, relative to the lighter compounds (Table S1, Supplementary Material; Neff, 2002). In Aker Brygge, however, the sediments showed a higher influence of petrogenic PAH sources (lower proportions of larger molecule compounds), when compared with the other two contaminated sites (e.g. indicated by a pyrene:benzo(k)fluoranthene-ratio of 5, as compared with 3). This is in agreement with its history as a harbour and site of a former shipyard. Regarding the organochlorine compounds, high concentrations of hexachlorobenzene (HCB) were found in the Kristiansand and Frierfjord sediments, reflecting the contamination by the metal refinery and magnesium smelter, respectively. High concentrations of PCB-209 (as well as proportion of this congener relative to the other PCBs), pentachlorobenzene and octachlorostyrene are also a signature of the magnesium smelter in the Frierfjord. Contaminant concentrations in the outer Oslofjord sediments (control/reference) were low, or below limits of detection (Tables S1-S2, Supplementary Material). The total organic matter content of the contaminated sediments was in the range 3.75-7.01 % dry wt., while TOC in the outer Oslofjord sediment was lower (~1 % dry wt.; Table S3, Supplementary Material). Thus the control/reference sediment had somewhat different characteristics than the other test sediments, a sub-optimal feature that should be kept in mind.

The range of contaminant concentrations was largely reflected in pore water and organisms (Tables S4-S9, Supplementary Material). However, the bioavailability of PAHs (in terms of

occurrence in pore water) was seemingly lower in the Kristiansand and especially Frierfjord sediments, as compared with sediments from Aker Brygge, indicating the presence of stronger sorbents in the sediments (Tables S1 and S4, Supplementary Material). Black carbon is known to contribute to such sorption behaviour (Koelmans et al., 2006; see also below), however, other sorbents have been shown to be responsible for high sediment-water partition coefficients (Cornelissen et al., 2006a). In a previous study from Norway, it was shown that total sorption was most adequately described when other (nonlinear sorbing) carbonaceous geosorbents, such as unburned coal and kerogen, were taken into account (Cornelissen et al., 2006a). The inter-sediment/location differences in pore water concentrations were not equally expressed by the bioaccumulation in *N. virens* and *H. reticulata*, however, where concentrations differed less than among the pore water measurements (Tables S4-S9, Supplementary Material). Nevertheless, biota-to-sediment accumulation factors predicted from the pore water measurements corresponded fairly well (mainly within one order of magnitude, with some exceptions) with the data from the *in vivo* bioaccumulation experiments, particularly when compared with predictions from generic (linear, one domain) sorption models, following Karickhoff et al. (1979) and Schwarzenbach et al. (2003; see below).

Assuming $K_{lipid} = K_{OW}$ and a free energy relationship between K_{OC} or K_{OW} following Karickhoff et al. (1979) or Scwarzenbach et al. (2003), predicted generic BSAFs equaled approximately 1 – 3 (for PAHs) or 1 – 100 (for organochlorine compounds; the higher BSAFs following Schwarzenbach et al., 2003), for the compounds in question (log K_{OW} ~ 4 – 8). Biota-to-sediment accumulation factors predicted from the pore water concentration measurements equalled 0.0002 – 2.3 (median: 0.04), and the highest BSAFs were predicted for organochlorine compounds. Thus, for PAHs, the generic BSAFs, following Karickhoff et al. (1979) or Scwarzenbach et al.

(2003) were in general two orders of magnitude higher than those predicted from the pore water concentration measurements. For the organochlorine compounds, the generic BSAFs following Karickhoff et al. (1979) agreed fairly well with those predicted from the pore water concentration measurements (that were approximately an order of magnitude lower), while the generic organochlorine BSAFs following Schwarzenbach et al. (2003) were in general two orders of magnitude higher than the BSAFs predicted from the pore water concentration measurements.

Biota-to-sediment accumulation factors observed in the *in vivo* bioaccumulation experiment corresponded to 0.005 – 2 (median: 0.02) for *N. virens* and 0.0008 – 33 (median 0.02) for *H. reticulata*. Again, the higher BSAFs were observed for the organochlorine compounds and the very highest (BSAF = 33) was observed for PCB-52 in *H. reticulata* exposed to Kristiansand-sediment. More specifically, BSAFs (for both species) for the organochlorine compounds were generally in the order 0.1-1, while BSAFs for the PAHs were lower, and generally in the order 0.001-0.01 (not shown, but can be deduced from Tables S1-S3 and S6-S10 in Supplementary Material). As such, the *in vivo* BSAFs for PAHs were comparable to or slightly lower than previous BSAFs measured in *N. diversicolor and H. reticulata* exposed to a range of PAH-contaminated sediments (Ruus et al., 2010; Ruus et al., 2005) and comparable with BSAFs measured in *N. diversicolor* exposed to pyrene-spiked marine sediments (Granberg and Selck, 2007). Furthermore, the BSAFs for organochlorine compounds corresponded well with those previously observed for *N. virens* exposed to PCB-contaminated sediments (Ruus et al., in press), for *N. diversicolor* exposed to sediments collected outside a Norwegain navy base (Ruus et al., 2005), for *Limnodrilus sp.* (Jonker et al., 2004) and *Lumbriculus variegatus* (You et al., 2006; Oligochaeta) exposed to spiked lake sediments, as well as for grass shrimp (*Palaemonetes pugio*) from a contaminated tidal creek system (Maruya and Lee, 1998).

The BSAFs predicted from the pore water concentration measurements corresponded (as mentioned) fairly well with the BSAFs deduced from the *in vivo* bioaccumulation results (in general within an order of magnitude; note that pore water concentration estimates may underestimate observed bioaccumulation by a factor ~10 for certain compounds and sediments; Figures 1-2). This applies to both PAHs and organochlorine compounds. The variability was also larger than the expected uncertainties in the pore water concentration estimates (a factor of ~2; Allan et al., 2012). There was some evidence that the pore water concentrations over predicted observed bioaccumulation for the higher molecular weight PAH compounds (Figure 1 e. and f.). Barthe et al. (2008) have previously attributed this phenomenon to some steric hindrance of biological membrane permeation by the larger molecules. The possibility of equilibrium not fully reached in the organisms for the most hydrophobic compounds can, however, not be ruled out. For PAHs, the generic BSAFs, following Karickhoff et al. (1979) or Schwarzenbach et al. (2003), represented large overestimates (up to several orders of magnitude), when compared with observed (*in vivo*) ones (Figure 1). The *in vivo* BSAFs for sediments from Aker Brygge generally showed the least discrepancy with the generic BSAFs, following Karickhoff et al. (1979) or Schwarzenbach et al. (2003; Figure 1). For the organochlorine compounds, the generic predicted BSAFs, following Karickhoff et al. (1979) corresponded fairly well with (generally slightly overestimating) the *in vivo* BSAFs (Figure 2). PCBs in the Kristiansand sediment displayed the highest *in vivo* BSAFs, most markedly in *H. reticulata*, for which the observed (*in vivo*) BSAFs were in fact mostly higher than those predicted from the Karickhoff et al. (1979) free energy relationship (Figure 2). On the other hand, generic organochlorine BSAFs following Schwarzenbach et al. (2003) were generally two orders of magnitude higher than those observed in the *in vivo* bioaccumulation experiment (Figure 2). The exception was for PCBs in *H.*

reticulata exposed to Kristiansand sediments, for which the BSAFs following Schwarzenbach et al. (2003) generally represented only slight overestimates (Figure 2).

PAHs emitted during the ferro-manganese plant activity in the Frierfjord and the electrode paste factory in Kristiansand may be entrapped in soot and coal particles during their formation, as previously pointed out (Allan et al., 2012; Arp et al., 2009; Jonker et al., 2005; Ruus et al., 2010). In addition, there have been discharges of soot from the Mg plant. This is likely to result in a large proportion of sediment-sorbed PAHs not available for partitioning to sediment pore water and for bioaccumulation. Strong sorption behaviour of coal tar pitch is known, although individual PAH compounds in coal tar pitch can show distinct sorption behaviour (Ghosh and Hawthorne, 2010). Highest logK_{OC} for PAHs were found for sediments from Frierfjord with values ranging from ~6 to ~9. Lowest logK_{OC} were for sediments from Aker Brygge with values ranging from ~5 to ~7 (Table S4, Supplementary Material; Allan et al., 2012).

During the last twenty years a growing number of observations indicate that the model describing a single partition coefficient for organic matter is too simple and that at least a dual model is needed, involving non-linear sorption, expressed through Freudlich coefficients (e.g. Accardi-Dey and Gschwend, 2003; Koelmans et al., 2006):

$$K_d = f_{AOC}K_{AOC} + f_{BC}K_{BC,F}C_{PW}^{nF-1}$$

where nF is the Freundlich exponent describing the non-linear sorption, f_{AOC} is the proportion of amorphous organic carbon in the sediment (proportion of TOC that is not black carbon), K_{AOC} is the partition coefficient between amorphous organic carbon and water, f_{BC} is the proportion of

black carbon in the sediment and $K_{BC,F}$ is the partition coefficient between black carbon and water. Studies also suggest, however, that quantitative models to assess bioavailability through a combination of amorphous organic carbon and black carbon sorption is not applicable among field sites with a wide range of black carbon fractions (e.g. Thorsen et al., 2004). Furthermore, Hawthorne et al. (2011) found that utilizing a two carbon model (including black carbon) did not improve predictions over a one-carbon TOC model in their data from 53 different sediments. They found that a Raoult's Law model could predict average K_{TOC} values, and that predictions were further improved by introducing a weathering factor that accounted for depletion of lower molecular weight compounds.

The strong sorption behaviour is obviously a likely explanation of the lower observed BSAFs of PAHs in *N. virens* and *H. reticulata*, compared with the predicted generic BSAFs, following Karickhoff et al. (1979) or Schwarzenbach et al. (2003). Furthermore it is a likely explanation of the generally lower BSAFs in the Frierfjord and Kristiansand sediments, than in the Aker Brygge sediments (with a mixture of petrogenic and pyrogenic sources of PAHs). Organochlorine compounds unlike PAHs were likely not enclosed in soot or black carbon-type particles, but rather adsorbed on their surface, thus lower sediment-water partition coefficients were measured as a result (Allan et al., 2012). These differences in sorption processes of the two classes of chemicals are thus likely responsible for the higher *in vivo* BSAFs for organochlorine, compared with PAH compounds. The reason for the higher bioaccumulation of PCBs from the Kristiansand sediments, especially in *H. reticulata*, as compared with the other sediments, however, is not known. It must be noted that concentrations of PCBs in *H. reticulata* exposed to the outer Oslofjord sediment (i.e. the control/reference) were high, when compared with those exposed to the other sediments and when compared to the outer Oslofjord sediment concentrations (below

389 limit of detection; Tables S2 and S9, Supplementary Material; BSAF of e.g. PCB-138 could
390 correspond to ≥3.9). Thus, some PCB residual/background contamination of the *H. reticulata*
391 employed in the experiment cannot be ruled out.
392
393 Bioaccumulation is the net result of uptake (all exposure routes including dietary absorption,
394 transport across respiratory surfaces and dermal absorption) and elimination routes and rates.
395 Since metabolism of certain PAHs is shown in Nereid species (e.g. Christensen et al., 2002;
396 Jorgensen et al., 2005; Rust et al., 2004), low observed *in vivo* BSAFs could theoretically be a
397 result of elimination of PAHs from the polychaetes. As such, Rust et al. (2004) advised against
398 the use of *N. virens* for the assessment of PAH bioaccumulation. This explanation would also
399 imply an equivalent metabolic capability in the gastropod (*H. reticulata*), considering the similar
400 concentrations of accumulated compounds. Results by Ruus et al. (2010), however, suggested
401 that bioaccumulation in *N. diversicolor* and *H. reticulata* was highly influenced by the
402 bioavailable fraction of compounds in sediment pore water, with metabolic capability of the
403 species being less important.
404
405 **4. CONCLUDING REMARKS**
406
407 Allan et al. (2012) emphasize that organic carbon/water partition coefficients for the three
408 sediments in question were all high, and that simple predictive relationships (e.g. based on
409 logK_{OW}) failed to predict partitioning accurately. The present results also show that predicted
410 BSAFs derived from the pore water concentration estimates provided a better agreement with the
411 bioaccumulation observed in the test organisms, than the generic BSAF estimated based on linear
412 sorption models (and that this especially applies to PAHs). The results therefore support that site-

specific evaluation of bioaccumulation provides information useful to more accurately providing a basis for cost-effective risk assessment and remediation plans. This is more difficult based on total sediment concentrations (Arp et al., 2009; Ghosh and Hawthorne, 2010; Hawthorne et al., 2006; Ruus et al., 2010; van der Heijden and Jonker, 2009). The present study demonstrated apparently more similarities than differences in bioaccumulation behaviour among compounds, between the chosen test species. These were, however, only two of a vast number of different relevant organisms. Therefore, one must also consider possible influence of biological aspects when extrapolating results from such assessments to field conditions. In a recent probabilistic modelling study aiming at explaining differences between bioaccumulation measurements in laboratory and field data, the importance of ingestion of sediments and sediment quality and composition was drawn attention to (Selck et al., 2012). Different benthic invertebrates exhibit different modes of living, which may have a large impact on the degree of exposure to and bioaccumulation of sediment associated contaminants (Meador, 2003). Increased knowledge of the specific characteristics of the vast number of (relevant) species potentially inhabiting contaminated sediments, which may influence the exposure, uptake and elimination of contaminants, would therefore be beneficial.

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Supplementary Material

Supplementary material related to this article can be found on-line at doi:10.1016/j.scitotenv.XXXX.XX.XXX;

Overview of chemicals used, methods for PAH and organochlorine determination, and calculation of predicted biota-to-sediment accumulation factors (BSAFs) from generic (one-domain) sorption models.

Figure S1. Conceptual sketch of the approaches to deduce and compare BSAFs.

Table S1. Concentrations (ng g^{-1} dry wt.) of polycyclic aromatic hydrocarbons (PAHs) in sediments.

Table S2. Concentrations (ng g^{-1} dry wt.) of organochlorine compounds in sediments.

Table S3. Amount total dry matter (TDM; %), fraction of particles smaller than 63 µm (P<63µm; % dry wt.) and amount of total organic carbon (TOC; % dry wt.) in sediments.

Table S4. Octanol-water partition coefficients (log transformed; log K_{OW}), total organic carbon-water partition coefficients (log transformed; log K_{OC}) and pore water concentrations (measured using low density polyethylene (LDPE) passive samplers and solid phase extraction; data from Allan et al., 2012) of PAHs in sediments.

Table S5. Log K_{OW}, log K_{OC} and pore water concentrations (measured using LDPE passive samplers; data from Allan et al., 2012) of organochlorine compounds in sediments.

Table S6. Concentrations (ng g^{-1} wet wt.) of PAHs in *Nereis virens* (Polychaeta) exposed to test sediments.

Table S7. Concentrations (ng g^{-1} wet wt.) of PAHs in *Hinia reticulata* (Gastropoda) exposed to test sediments.

Table S8. Concentrations (ng g^{-1} wet wt.) of organochlorine compounds in *Nereis virens* (Polychaeta) exposed to test sediments.

Table S9. Concentrations (ng g^{-1} wet wt.) of organochlorine compounds in *Hinia reticulata* (Gastropoda) exposed to test sediments.

Table S10. Amount total dry matter (TDM; %) and amount lipid (% wet wt.) in *N. virens* and *H. reticulata* exposed to test sediments.

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590 **Figure legends**

591

592 **Figure 1.** Ratio between observed (*in vivo*) biota-to-sediment accumulation factors (BSAFs; in
593 organisms exposed to test sediment) and predicted BSAFs for polycyclic aromatic hydrocarbons
594 (PAHs). Upper figures (**a.** and **b.**): BSAFs predicted from generic sorption model from
595 Karickhoff et al. (1979; based on organic carbon-water partitioning and linear free energy
596 relationship between K_{OW} and K_{OC}; $\log K_{OC} = \log K_{OW} - 0.21$); middle figures (**c.** and **d.**): BSAFs
597 predicted from generic sorption model from Schwarzenbach et al. (2003;
598 $\log K_{OC} = 0.98 \log K_{OW} - 0.32$); bottom figures (**e.** and **f.**): BSAFs predicted from measurements
599 of dissolved concentrations of PAHs in sediment pore water, using passive samplers (low density
600 polyethylene, LDPE) and solid phase extraction (data from Allan et al., 2012). Left figures (**a.**, **c.**
601 and **e.**): *Nereis virens*; right figures (**b.**, **d.** and **f.**): *Hinia reticulata*. Solid line: 1:1 relationship
602 ($BSAF_{Observed}/BSAF_{Predicted} = 1$). Stapled lines: One order of magnitude below and above the 1:1
603 relationship, respectively.

604

605 **Figure 2.** Ratio between observed (*in vivo*) biota-to-sediment accumulation factors (BSAFs; in
606 organisms exposed to test sediment) and predicted BSAFs for organochlorine compounds. Upper
607 figures (**a.** and **b.**): BSAFs predicted from generic sorption model from Karickhoff et al. (1979;
608 based on organic carbon-water partitioning and linear free energy relationship between K_{OW} and
609 K_{OC}; $\log K_{OC} = \log K_{OW} - 0.21$); middle figures (**c.** and **d.**): BSAFs predicted from generic
610 sorption model from Schwarzenbach et al. (2003; $\log K_{OC} = 0.74 \log K_{OW} + 0.15$); bottom figures
611 (**e.** and **f.**): BSAFs predicted from measurements of dissolved concentrations of PAHs in
612 sediment pore water, using passive samplers (low density polyethylene, LDPE) and solid phase
613 extraction (data from Allan et al., 2012). Left figures (**a.**, **c.** and **e.**): *Nereis virens*; right figures

(**b.**, **d.** and **f.**): *Hinia reticulata*. Solid line: 1:1 relationship ($BSAF_{Observed}/BSAF_{Predicted} = 1$). Stapled lines: One order of magnitude below and above the 1:1 relationship, respectively.

Figure 1.

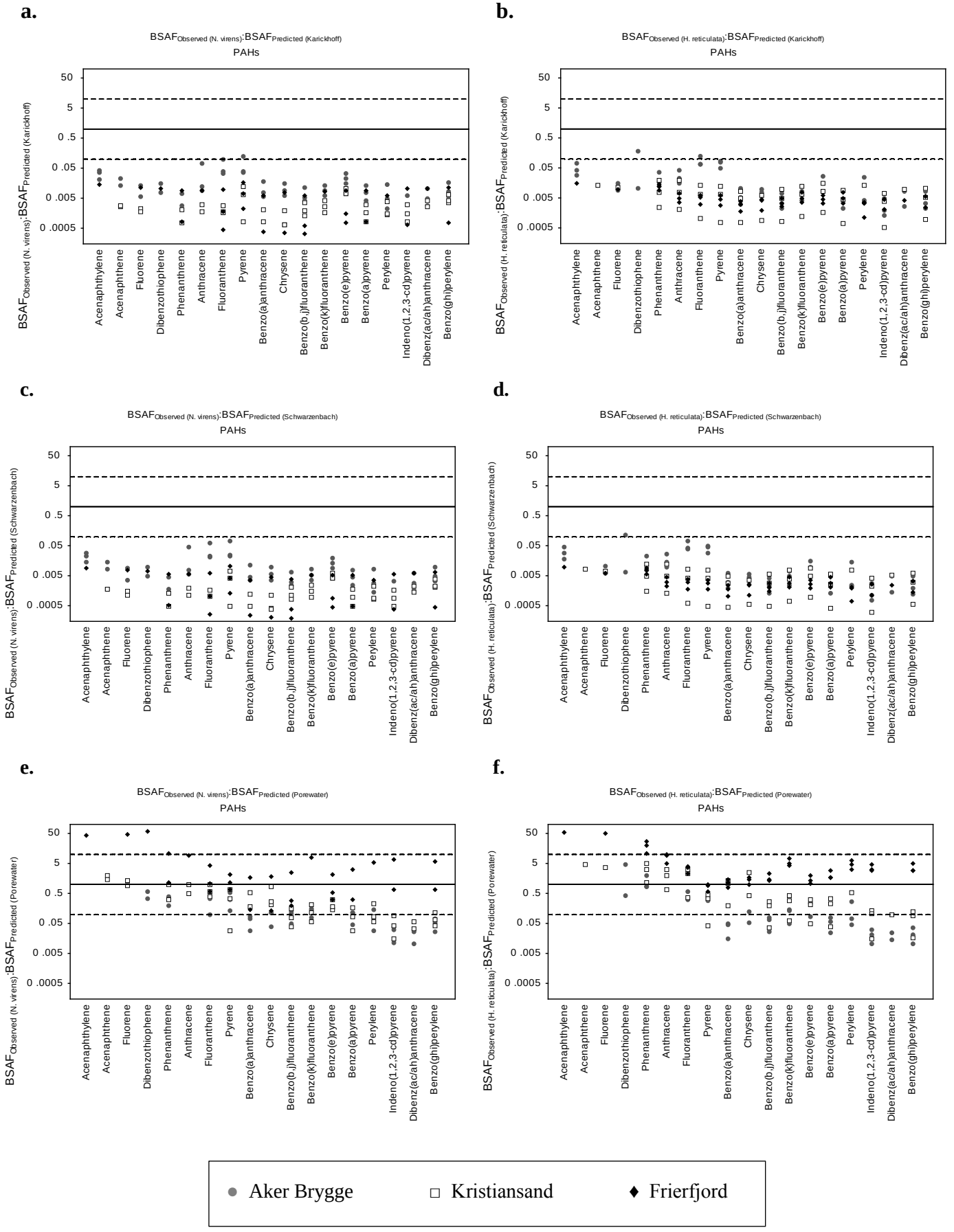
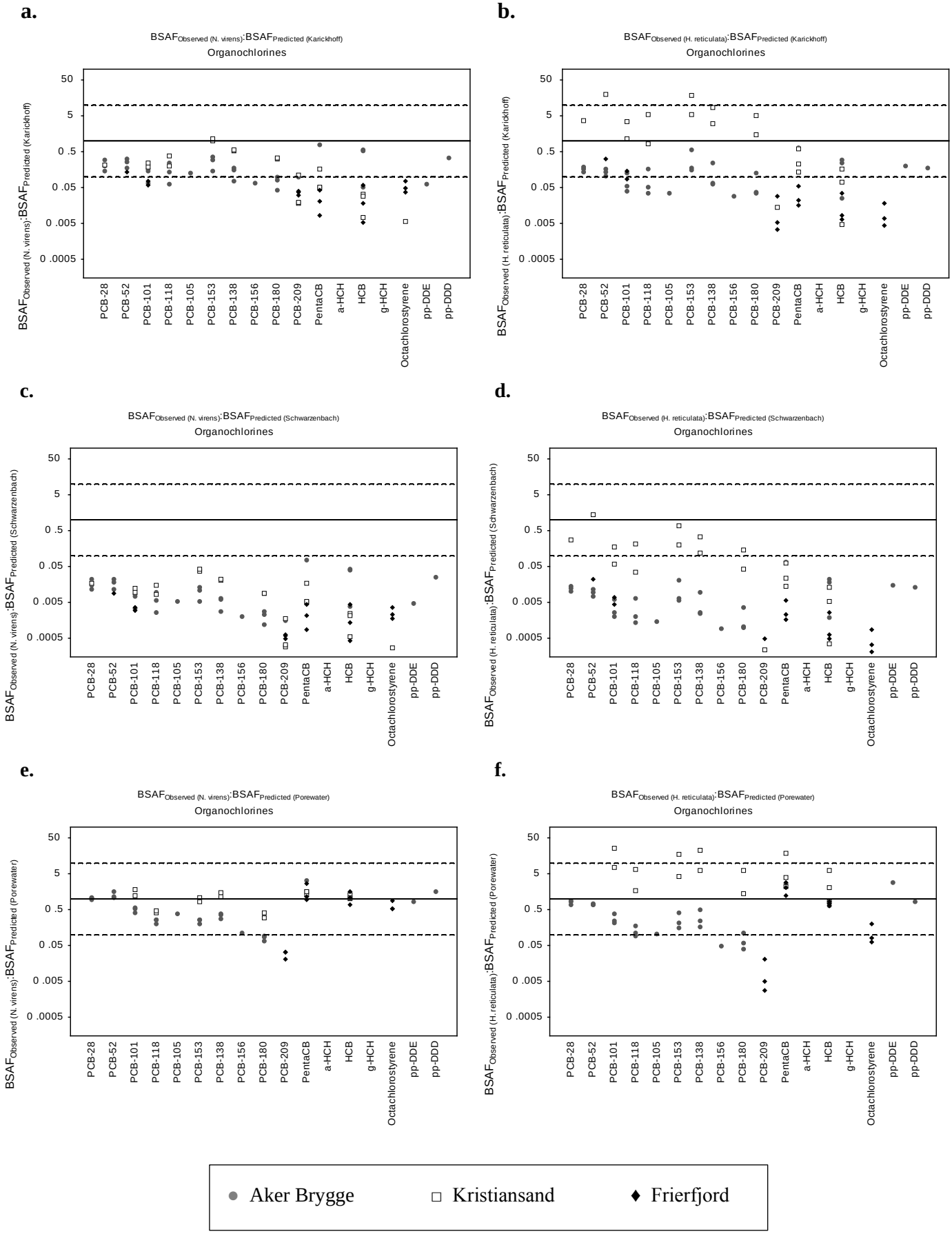

a.
BSAFObserved (N. virens):BSAFPredicted (Karickhoff)
PAHs
BSAFObserved (N. virens):BSAFPredicted (Karickhoff)
50
5
0.5
0.05
0.005
0.0005
Acenaphthylene
Acenaphthene
Fluorene
Dibenzothiophene
Phenanthrene
Anthracene
Fluoranthene
Pyrene
Benzo(a)anthracene
Chrysene
Benzo(b,j)fluoranthene
Benzo(k)fluoranthene
Benzo(e)pyrene
Benzo(a)pyrene
Perylene
Indeno(1,2,3-cd)pyrene
Dibenz(ac/ah)anthracene
Benzo(ghi)perylene
b.
BSAFObserved (H. reticulata):BSAFPredicted (Karickhoff)
PAHs
BSAFObserved (H. reticulata):BSAFPredicted (Karickhoff)
c.
BSAFObserved (N. virens):BSAFPredicted (Schwarzenbach)
PAHs
BSAFObserved (N. virens):BSAFPredicted (Schwarzenbach)
d.
BSAFObserved (H. reticulata):BSAFPredicted (Schwarzenbach)
PAHs
BSAFObserved (H. reticulata):BSAFPredicted (Schwarzenbach)
e.
BSAFObserved (N. virens):BSAFPredicted (Porewater)
PAHs
BSAFObserved (N. virens):BSAFPredicted (Porewater)
f.
BSAFObserved (H. reticulata):BSAFPredicted (Porewater)
PAHs
BSAFObserved (H. reticulata):BSAFPredicted (Porewater)
Aker Brygge
Kristiansand
Frierfjord

Figure 2.


a.
BSAFObserved (N. virens):BSAFPredicted (Karickhoff)
Organochlorines
BSAFObserved (N. virens):BSAFPredicted (Karickhoff)
b.
BSAFObserved (H. reticulata):BSAFPredicted (Karickhoff)
Organochlorines
BSAFObserved (H. reticulata):BSAFPredicted (Karickhoff)
c.
BSAFObserved (N. virens):BSAFPredicted (Schwarzenbach)
Organochlorines
BSAFObserved (N. virens):BSAFPredicted (Schwarzenbach)
d.
BSAFObserved (H. reticulata):BSAFPredicted (Schwarzenbach)
Organochlorines
BSAFObserved (H. reticulata):BSAFPredicted (Schwarzenbach)
e.
BSAFObserved (N. virens):BSAFPredicted (Porewater)
Organochlorines
BSAFObserved (N. virens):BSAFPredicted (Porewater)
f.
BSAFObserved (H. reticulata):BSAFPredicted (Porewater)
Organochlorines
BSAFObserved (H. reticulata):BSAFPredicted (Porewater)
50
5
0.5
0.05
0.005
0.0005
PCB-28
PCB-52
PCB-101
PCB-118
PCB-105
PCB-153
PCB-138
PCB-156
PCB-180
PCB-209
PentaCB
a-HCH
HCB
g-HCH
Octachlorostyrene
pp-DDE
pp-DDD
Aker Brygge
Kristiansand
Frierfjord

SUPPLEMENTARY MATERIAL

IN VIVO BIOACCUMULATION OF CONTAMINANTS FROM HISTORICALLY POLLUTED SEDIMENTS – RELATION TO BIOAVAILABILITY ESTIMATES

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Chemicals

Hydromatrix (Varian Inc., Palo Alto, Ca, USA), cyclohexane (J.T. Baker, Deventer, Holland), dichloromethane (Rathburn chemicals Ltd, Walkerburn, Scotland), sulphuric acid (H_2SO_4; Merck, Darmstadt, Germany), *n*-pentane (Rathburn), sodium sulphate (Merck), acetone (Rathburn), saline water (0.5 %; Merck), hydrochloric acid (HCl; 1N; Merck), Analytical standards (PAHs: Chiron, Trondheim, Norway; organochlorine compounds: Dr. Ehrenstorfer GmbH, Augsburg, Germany), internal standards (for PAHs: Chiron): naphthalene-d8, acenaphthene-d8, phenanthrene-d10, chrysene-d12, perylene-d12, and anthracene-d10; (for organochlorine compounds: Dr. Ehrenstorfer GmbH): PCB-30, -53 and -204.

Purities of standards (analytical standard) were >99% (>99.5% for deuterated PAHs). Solvents were of HPLC grade or better.

Details regarding methods and chemicals used for passive sampling measurements of freely dissolved concentrations in pore water is given in Allan et al. (2012).

PAH and organochlorine determination

Extracts for PAH determination were analysed by GC-MS (HP/Agilent 6890N; Agilent Technologies, Wilmington, DE, USA) with the MS detector (HP/Agilent 5973) in selected ion monitoring mode (SIM). The GC was equipped with a 30 m J&W DB-5MS (stationary phase of 5% phenyl polysoxilane) column (0.25 mm i.d. and 0.25 µm film thickness; Agilent JW Scientific, Santa Clara, USA), and splitless injection. The initial column temperature was 60 °C (raised in steps to 310 °C). Injector, transfer line, ion source and quadruple temperatures were 300, 280, 230 and 150 °C, respectively. Quantification of individual compounds was performed by using the relative response of the internal and external (standard curve) standards.

Extracts for organochlorine determination were analysed by GC-ECD (HP/Agilent 6890N). Analytes were separated on a 60 m DB-5 column (0.25 mm i.d. and 0.25 µm film thickness, Agilent JW Scientific) with hydrogen as carrier gas (1 mL min^{-1}). The injector was operated in splitless mode (splitless time of 1.25 min and split flow of 60 mL min^{-1}) with a septum purge flow of 5 mL min^{-1} and at a temperature of 255 °C. Make-up gas was nitrogen at a flow rate of 30 mL min^{-1}. The detector temperature was 285 °C. The initial column temperature was 90 °C (raised in steps to 310 °C). Quantification of individual compounds was performed by using the relative response of the internal and external (standard curve) standards.

Calculation of predicted Biota-to-Sediment Accumulation Factors (BSAFs) from generic (one-domain) sorption models

Assuming that the partition coefficient between organism lipids and water equals K_{OW} ($K_{lipid} = K_{OW}$; e.g. Ruus et al., 2010), predicted generic Biota to Sediment Accumulation Factors ($BSAF_{Predicted\ (Karickhoff)}$ or $BSAF_{Predicted\ (Schwarzenbach)}$) were calculated as follows:

$$\mathrm{BSAF}_{\text{Predicted (Karickhoff/Schwarzenbach)}} = \frac{C_{lipid}}{C_{OC}},\ K_{lipid} = \frac{C_{lipid}}{C_{PW}} = K_{OW},\ K_{OC} = \frac{C_{OC}}{C_{PW}} \text{ and } C_{OC} = \frac{C_S}{f_{OC}},$$

where C_{lipid} is the lipid normalised concentration in the organism, C_{OC} is the organic carbon normalised concentration in the sediment, C_{PW} is the concentration in sediment pore water, C_S is the concentration in sediment (total, dry wt.) and f_{OC} is the fraction of organic content in the sediment (dry:dry).

Furthermore,

If: $\log K_{OC} = \log K_{OW} - 0.21$ or $K_{OC} = 0.62K_{OW}$ (Karickhoff et al., 1979)

Then: $\mathrm{BSAF}_{\text{Predicted(Karickhoff)}} = \dfrac{K_{OW} \cdot C_{PW}}{0.62 \cdot K_{OW} \cdot C_{PW}} = 1.6$

If: $\log K_{OC} = 0.98\log K_{OW} - 0.32$ or $K_{OC} = 0.48K_{OW}^{0.98}$ (Schwarzenbach et al., 2003; for PAHs)

Then: $\mathrm{BSAF}_{\text{Predicted(Schwarzenbach)}} = \dfrac{K_{OW} \cdot C_{PW}}{0.48 \cdot K_{OW}^{0.98} \cdot C_{PW}} = 2.08K_{OW}^{0.02}$ (for PAHs)

If: $\log K_{OC} = 0.74\log K_{OW} + 0.15$ or $K_{OC} = 1.4K_{OW}^{0.74}$ (Schwarzenbach et al., 2003; for organochlorine compounds)

Then: $\mathrm{BSAF}_{\text{Predicted(Schwarzenbach)}} = \dfrac{K_{OW} \cdot C_{PW}}{1.4 \cdot K_{OW}^{0.74} \cdot C_{PW}} = 0.71K_{OW}^{0.26}$ (for organochlorine compounds)

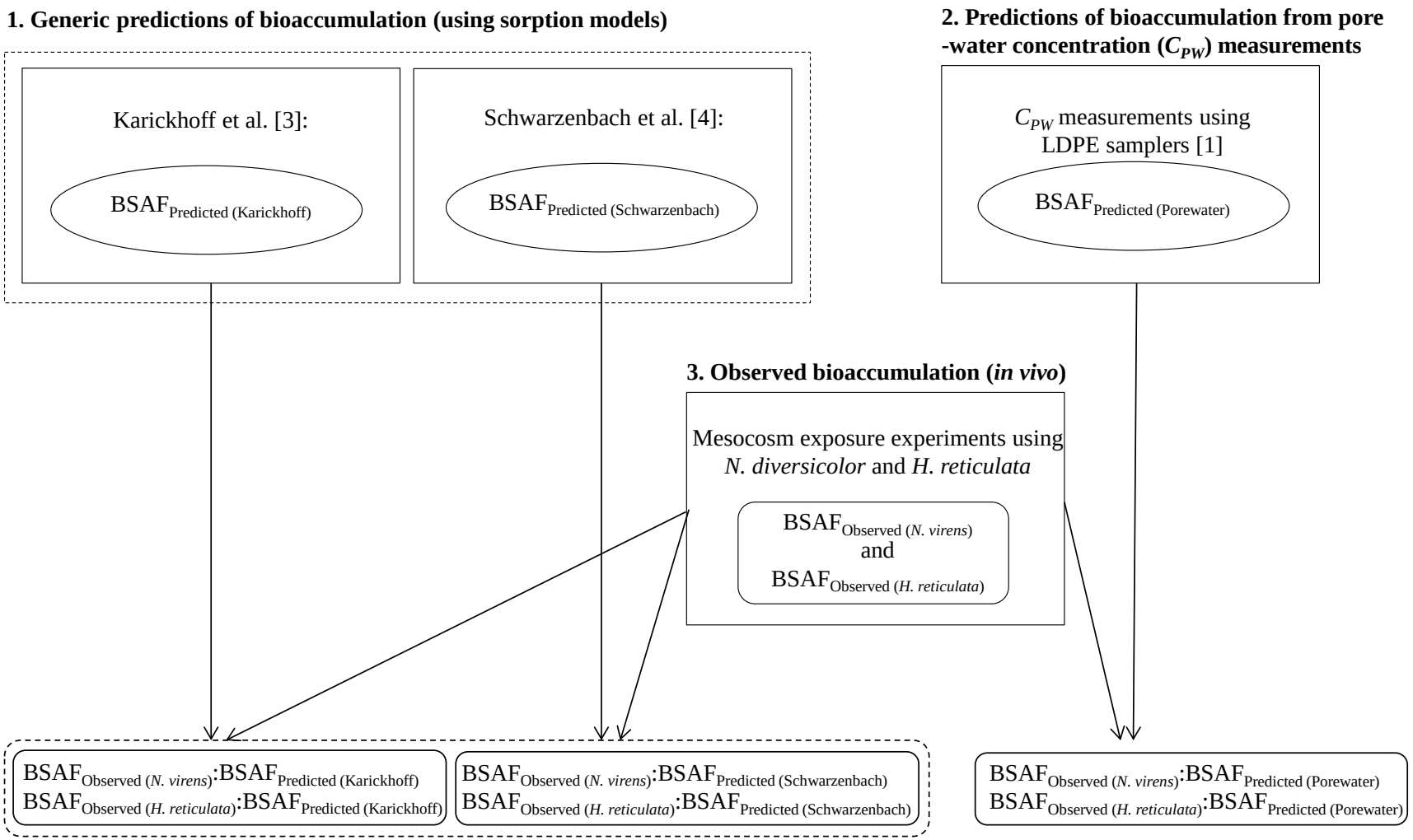

Figure S1. Conceptual sketch of the approaches to deduce and compare biota-to-sediment accumulation factors (BSAFs). Bioaccumulation (given as BSAF) is predicted from simple generic sorption models following Karickhoff et al. (1979) and Schwarzenbach et al. (2003). In addition bioaccumulation is predicted from sediment pore water concentrations measured using low density polyethylene passive samplers (data from Allan et al., 2012). The different predictions of bioaccumulation are compared with observed bioaccumulation in *Nereis virens* (Polychaeta) and *Hinia reticulata* (Gastropoda) exposed to the sediments in mesocosm exposure experiments.

Table S1. Concentrations (ng g^{-1} dry wt.) of polycyclic aromatic hydrocarbons (PAHs)* in sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	Nap.	Acy.	Ace.	Fle.	Dbthi.	Phe.	Ant.	Flu.	Pyr.	B(a)A
Aker brygge	**1**	380	58	120	200	150	900	810	6600	6400	2700
Aker brygge	**4**	360	63	130	170	97	800	590	4300	5200	3000
Aker brygge	**11**	520	110	160	280	200	1400	2200	15000	14000	6800
Kristiansand	**2**	210	11	330	270	130	1900	500	3300	2900	2100
Kristiansand	**6**	760	33	1300	920	410	6400	1800	9700	8400	6100
Kristiansand	**7**	490	49	730	570	250	4100	1100	6700	5800	4100
Frierfjord	**5**	330	100	46	170	190	1800	1500	3900	4800	4600
Frierfjord	**9**	610	200	43	180	170	1500	990	2400	3300	3400
Frierfjord	**10**	470	110	33	160	170	1200	700	1600	2300	2300
Outer Oslofj.	**3**	6	<2	<2	3.6	<2	<15	3	<25	<30	<8
Outer Oslofj.	**8**	<5	<2	<2	3.6	<2	<15	4.6	<25	<30	8.1
Outer Oslofj.	**12**	5.5	<2	<2	5	<2	<15	4.9	<25	<30	14

Sediment	Box id.	Chr.	B(bj)F.	B(k)F.	B(e)P.	B(a)P.	Per.	I(123-cd)P.	Db(ac/ah)A.	B(ghi)P.
Aker brygge	**1**	2300	3500	1200	2200	1900	530	2000	370	1800
Aker brygge	**4**	2100	4100	1400	2500	2400	630	2400	490	2100
Aker brygge	**11**	5000	7800	2300	4200	4000	990	3900	840	3300
Kristiansand	**2**	1800	2900	1100	1700	2300	570	1800	370	1400
Kristiansand	**6**	5100	8200	3000	4700	6700	1600	5100	1000	3900
Kristiansand	**7**	3400	5700	2000	3200	4400	1100	3400	700	2700
Frierfjord	**5**	5400	4400	1200	2800	1900	1900	2000	610	2000
Frierfjord	**9**	3700	4700	1100	3000	1800	1200	1800	550	2100
Frierfjord	**10**	2200	3600	930	2000	1600	540	1100	310	1100
Outer Oslofj.	**3**	<10	50	19	29	16	64	36	3.6	45
Outer Oslofj.	**8**	17	70	22	36	20	44	57	5.3	57
Outer Oslofj.	**12**	18	77	25	40	22	38	57	5.3	62

* Naphthalene (Nap.), acenaphthylene (Acy.), acenaphthene (Ace.), fluorene (Fle.), dibenzothiophene (Dbthi.), phenanthrene (Phe.), anthracene (Ant.), fluoranthene (Flu.), pyrene (Pyr.), benzo(a)anthracene (B(a)A.), chrysene (Chr.), benzo(b,j)fluoranthene (B(bj)F.), benzo(k)fluoranthene (B(k)F.), benzo(e)pyrene (B(e)P.), benzo(a)pyrene (B(a)P.), perylene (Per.), Indeno(1,2,3-cd)pyrene (I(123-cd)P.), dibenz(ac/ah)anthracene (Db(ac/ah)A.) and Benzo(ghi)perylene (B(ghi)P.).

Table S2. Concentrations (ng g^{-1} dry wt.) of organochlorine compounds* in sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	PCB-28	PCB-52	PCB-101	PCB-118	PCB-105	PCB-153	PCB-138	PCB-156	PCB-180
Aker brygge	**1**	31	88	95	46		40	88		52
Aker brygge	**4**	53	140	200	140		80	170		99
Aker brygge	**11**	54	130	150	100	69	60	130	21	86
Kristiansand	**2**	<0.5	<0.5	3.3	3.3		2.6	3.9		2.9
Kristiansand	**6**	<5	<5	<20	<20		<20	<20		<20
Kristiansand	**7**	0.74	0.8	5	2.9		4.1	6.7		5.1
Frierfjord	**5**	<5	5.5	32	<30		<30	<30		<30
Frierfjord	**9**	<30	<30	36	<30		<30	<30		<30
Frierfjord	**10**	<50	<50	<50	<50		<50	<50		<50
Outer Oslofj.	**3**	<0.5	<0.5	<0.5	0.87		<1	<0.5		<0.5
Outer Oslofj.	**8**	<2	<2	<2	<2		<2	<2		<2
Outer Oslofj.	**12**	<0.5	<0.5	<2	<2		<2	<2		<2

Sediment	Box id.	PCB-209	PentaCB	α-HCH	HCB	γ-HCH	OCS	p,p′-DDE	p,p′-DDD
Aker brygge	**1**	1.3			6				
Aker brygge	**4**	2.8			46				
Aker brygge	**11**	1.2	1.3		5.9			38	100
Kristiansand	**2**	10	25		92				
Kristiansand	**6**	6.9	16		1085		230		
Kristiansand	**7**	18	38		162				
Frierfjord	**5**	1035	372		298		911		
Frierfjord	**9**	1196	85		2686		935		
Frierfjord	**10**	1400	432		1701		927		
Outer Oslofj.	**3**								
Outer Oslofj.	**8**								
Outer Oslofj.	**12**								

* Polychlorinated biphenyl (PCB) -28, -52, -101, -118, -105, -153, -138, -156, -180, and -209, and pentachlorobenzene (PentaCB), α-hexachlorocyclohexane (α-HCH), hexachlorobenzene (HCB), γ-hexachlorocyclohexane (γ-HCH), octachlorostyrene (OCS), 1,1-dichloro-2,2-bis(4-chlorophenyl)ethene (p,p′-DDE) and 1,1-dichloro-2,2-bis(4-chlorophenyl)ethane (p,p′-DDD).

Table S3. Amount total dry matter (TDM; %), fraction of particles smaller than 63 µm (P<63µm; % dry wt.; Krumbein and Pettijohn, 1938) and amount of total organic carbon (TOC; % dry wt.) in sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	TDM	P<63µm	TOC
Aker brygge	**1**	22.3	86	5.53
Aker brygge	**4**	22.1	50	5.68
Aker brygge	**11**	25.3	37	6.75
Kristiansand	**2**	47.8	62	3.75
Kristiansand	**6**	40.5	57	7.01
Kristiansand	**7**	43.8	68	5.2
Frierfjord	**5**	36.5	47	4.86
Frierfjord	**9**	29.4	54	6.47
Frierfjord	**10**	30.5	59	6
Outer Oslofj.	**3**	55.1	93	0.94
Outer Oslofj.	**8**	56.4	92	1.01
Outer Oslofj.	**12**	52.2	89	1.06

Table S4. Octanol-water partition coefficients (log transformed; *log* K_{OW}), total organic carbon-water partition coefficients (log transformed; log K_{OC}) and pore water concentrations (measured using low density polyethylene (LDPE) passive samplers and solid phase extraction; data from Allan et al., 2012) of polycyclic aromatic hydrocarbons (PAHs)* in sediments from Aker brygge, Kristiansand and Frierfjord.

Sediment	Parameter		Box id.	Nap.	Acy.	Ace.	Fle.	Dbthi.	Phe.	Ant.	Flu.	Pyr.	B(a)A
	log K_{OW}			3.37	4.00	3.92	4.18	4.00	4.57	4.54	5.22	5.18	5.91
Aker brygge	**log K_{OC}**	L kg^{-1}						5.35	6.13		5.77	5.71	6.51
Kristiansand	**log K_{OC}**	L kg^{-1}		6.04		6.48	6.67	6.65	6.96	6.76	7.52	6.66	8.04
Frierfjord	**log K_{OC}**	L kg^{-1}			7.45	6.85	7.64	7.60	7.59	7.58	7.91	7.29	8.36
Aker brygge	C_{PW}	ng L^{-1}	**1**					11.23	12.86		245.04	276.11	21.54
Kristiansand	C_{PW}	ng L^{-1}	**7**	8.43		4.93	2.34	1.11	8.58	3.73	3.71	23.58	0.70
Frierfjord	C_{PW}	ng L^{-1}	**9**		0.09	0.11	0.07	0.08	0.73	0.52	0.60	3.35	0.28

Sediment	Parameter		Box id.	Chr.	B(bj)F.	B(k)F.	B(e)P.	B(a)P.	Per.	I(123-cd)P.	Db(ac/ah)A.	B(ghi)P.
	log K_{OW}			5.86	5.90	5.90	6.00	6.04	6.00	6.50	6.75	6.50
Aker brygge	**log K_{OC}**	L kg^{-1}		6.60	6.86	6.82	6.85	6.91	6.92	7.20	7.15	7.01
Kristiansand	**log K_{OC}**	L kg^{-1}		8.26	7.29	7.34	7.22	7.52	7.52	7.71	7.66	7.49
Frierfjord	**log K_{OC}**	L kg^{-1}		8.32	8.40	8.75	8.34	8.47	9.16	9.34		9.24
Aker brygge	C_{PW}	ng L^{-1}	**1**	13.02	11.96	4.16	7.06	5.73	1.44	2.89	0.67	3.92
Kristiansand	C_{PW}	ng L^{-1}	**7**	0.36	5.45	1.73	3.65	2.52	0.61	1.26	0.28	1.62
Frierfjord	C_{PW}	ng L^{-1}	**9**	0.34	0.32	0.04	0.22	0.11	0.02	0.01		0.02

* Naphthalene (Nap.), acenaphthylene (Acy.), acenaphthene (Ace.), fluorene (Fle.), dibenzothiophene (Dbthi.), phenanthrene (Phe.), anthracene (Ant.), fluoranthene (Flu.), pyrene (Pyr.), benzo(a)anthracene (B(a)A.), chrysene (Chr.), benzo(b,j)fluoranthene (B(bj)F.), benzo(k)fluoranthene (B(k)F.), benzo(e)pyrene (B(e)P.), benzo(a)pyrene (B(a)P.), perylene (Per.), Indeno(1,2,3-cd)pyrene (I(123-cd)P.), dibenz(ac/ah)anthracene (Db(ac/ah)A.) and Benzo(ghi)perylene (B(ghi)P.).

Table S5. Octanol-water partition coefficients (log transformed; *log* K_{OW}), total organic carbon-water partition coefficients (log transformed; log K_{OC}) and pore water concentrations (measured using low density polyethylene (LDPE) passive samplers and solid phase extraction; data from Allan et al., 2012) of organochlorine compounds* in sediments from Aker brygge, Kristiansand and Frierfjord.

Sediment	Parameter		Box id.	PCB-28	PCB-52	PCB-101	PCB-118	PCB-105	PCB-153	PCB-138	PCB-156	PCB-180
	log K_{OW}			5.67	5.84	6.38	6.74	6.65	6.92	6.83	7.18	7.36
Aker brygge	**log K_{OC}**	L kg^{-1}		6.17	6.35	6.81	6.84	6.98	6.71	7.06	7.25	7.22
Kristiansand	**log K_{OC}**	L kg^{-1}				6.94	6.64		6.57	6.93		7.12
Frierfjord	**log K_{OC}**	L kg^{-1}										
Aker brygge	C_{PW}	ng L^{-1}	**1**	0.52	0.89	0.38	0.23	0.12	0.20	0.19	0.02	0.08
Kristiansand	C_{PW}	ng L^{-1}	**7**			0.01	0.01		0.02	0.01		0.01
Frierfjord	C_{PW}	ng L^{-1}	**9**									

Sediment	Parameter		Box id.	PCB-209	PentaCB	α-HCH	HCB	γ-HCH	OCS	p,p′-DDE	p,p′-DDD
	log K_{OW}			8.26	5.20	3.80	5.70	4.20	6.50	5.70	5.80
Aker brygge	**log K_{OC}**	L kg^{-1}			5.66		5.93			6.68	6.29
Kristiansand	**log K_{OC}**	L kg^{-1}			6.31		7.50				
Frierfjord	**log K_{OC}**	L kg^{-1}		7.99	6.97		7.45		7.41		
Aker brygge	C_{PW}	ng L^{-1}	**1**		0.05		0.11			0.13	0.85
Kristiansand	C_{PW}	ng L^{-1}	**7**		0.24		0.27				
Frierfjord	C_{PW}	ng L^{-1}	**9**	0.23	0.59		1.05		0.68		

* Polychlorinated biphenyl (PCB) -28, -52, -101, -118, -105, -153, -138, -156, -180, and -209, and pentachlorobenzene (PentaCB), α-hexachlorocyclohexane (α-HCH), hexachlorobenzene (HCB), γ-hexachlorocyclohexane (γ-HCH), octachlorostyrene (OCS), 1,1-dichloro-2,2-bis(4-chlorophenyl)ethene (p,p′-DDE) and 1,1-dichloro-2,2-bis(4-chlorophenyl)ethane (p,p′-DDD).

Table S6. Concentrations (ng g^{-1} wet wt.) of polycyclic aromatic hydrocarbons (PAHs)* in *Nereis virens* (Polychaeta) exposed to sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	Nap.	Acy.	Ace.	Fle.	Dbthi.	Phe.	Ant.	Flu.	Pyr.	B(a)A
Aker brygge	**1**	<0.5	1.1	1.2	1.2	0.99	2.9	26	290	340	22
Aker brygge	**4**	<0.5	0.62	<0.5	<0.5	<0.5	<0.5	2.5	66	86	8
Aker brygge	**11**	<0.5	1.7	0.88	0.7	0.67	1.7	12	250	240	23
Kristiansand	**2**	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	4.7	1.9	<0.5
Kristiansand	**6**	1	<0.5	1.3	0.79	<0.5	5	2.1	10	37	4.8
Kristiansand	**7**	<0.5	<0.5	0.99	0.5	<0.5	1.6	1	5.6	19	1.7
Frierfjord	**5**	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	0.8	4.9	<0.5
Frierfjord	**9**	0.56	<0.5	<0.5	<0.5	<0.5	0.61	<0.5	2	11	0.64
Frierfjord	**10**	2.1	0.74	<0.5	0.91	0.88	5.4	3	7.7	19	6.8
Outer Oslofj.	**3**	0.89	<0.5	<0.5	<0.5	<0.5	0.81	3.7	41	58	5.3
Outer Oslofj.	**8**	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	0.61	0.94	<0.5
Outer Oslofj.	**12**	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5

Sediment	Box id.	Chr.	B(bj)F.	B(k)F.	B(e)P.	B(a)P.	Per.	I(123-cd)P.	Db(ac/ah)A.	B(ghi)P.
Aker brygge	**1**	16	17	7.2	32	11	3.2	5.2	1.7	13
Aker brygge	**4**	5.7	7.6	4	17	4.5	0.67	1.7	0.64	5.4
Aker brygge	**11**	20	18	8.9	40	13	2	5	1.8	12
Kristiansand	**2**	1	3.2	1.5	9.7	1.5	0.68	1.2	<0.5	4.1
Kristiansand	**6**	3.5	11	4.7	18	7.7	2.2	5.6	1.5	9.6
Kristiansand	**7**	1.1	5.3	2.6	11	3.7	0.85	2.6	0.88	5.4
Frierfjord	**5**	<0.5	0.67	<0.5	1	<0.5	<0.5	<0.5	<0.5	<0.5
Frierfjord	**9**	0.63	1.4	<0.5	2.2	0.71	<0.5	0.55	<0.5	0.77
Frierfjord	**10**	8.1	11	3.9	8.8	7.2	1.6	5.4	1.6	6.1
Outer Oslofj.	**3**	4.1	4.6	2	7	3.2	0.83	1.5	<0.5	2.5
Outer Oslofj.	**8**	<0.5	<0.5	<0.5	0.53	<0.5	<0.5	<0.5	<0.5	<0.5
Outer Oslofj.	**12**	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5

* Naphthalene (Nap.), acenaphthylene (Acy.), acenaphthene (Ace.), fluorene (Fle.), dibenzothiophene (Dbthi.), phenanthrene (Phe.), anthracene (Ant.), fluoranthene (Flu.), pyrene (Pyr.), benzo(a)anthracene (B(a)A.), chrysene (Chr.), benzo(b,j)fluoranthene (B(bj)F.), benzo(k)fluoranthene (B(k)F.), benzo(e)pyrene (B(e)P.), benzo(a)pyrene (B(a)P.), perylene (Per.), Indeno(1,2,3-cd)pyrene (I(123-cd)P.), dibenz(ac/ah)anthracene (Db(ac/ah)A.) and Benzo(ghi)perylene (B(ghi)P.).

Table S7. Concentrations (ng g^{-1} wet wt.) of polycyclic aromatic hydrocarbons (PAHs)* in *Hinia reticulata* (Gastropoda) exposed to sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	Nap.	Acy.	Ace.	Fle.	Dbthi.	Phe.	Ant.	Flu.	Pyr.	B(a)A
Aker brygge	**1**	2.3	0.68	<0.5	0.51	<0.5	5.4	5.6	70	79	4.4
Aker brygge	**4**	2	1	<0.5	0.58	6.5	5.1	5	190	160	3.5
Aker brygge	**11**	2.8	1.1	0.74	1	0.73	6.1	12	360	240	13
Kristiansand	**2**	2.4	<0.5	1	0.68	<0.5	8.7	2.2	10	8.4	4.1
Kristiansand	**6**	2.8	<0.5	<0.5	<0.5	<0.5	4.7	1.1	3	2	1.4
Kristiansand	**7**	<2	<2	<2	<2	<2	10	2.6	14	12	6.1
Frierfjord	**5**	3.4	0.6	<0.5	0.66	<0.5	8.5	2.1	4.7	5.5	3.2
Frierfjord	**9**	2.8	<0.5	<0.5	<0.5	<0.5	5.9	1.9	4.1	5.5	3.7
Frierfjord	**10**	2.1	<0.5	<0.5	<0.5	<0.5	3.7	1.2	2.9	3.7	2.4
Outer Oslofj.	**3**	2.5	<0.5	<0.5	<0.5	<0.5	5.4	1.2	1.1	0.78	<0.5
Outer Oslofj.	**8**	1.5	<0.5	<0.5	<0.5	<0.5	4.4	<0.5	1.6	0.88	<0.5
Outer Oslofj.	**12**	2.8	<0.5	<0.5	<0.5	<0.5	4	0.64	1.2	0.76	<0.5

Sediment	Box id.	Chr.	B(bj)F.	B(k)F.	B(e)P.	B(a)P.	Per.	I(123-cd)P.	Db(ac/ah)A.	B(ghi)P.
Aker brygge	**1**	2.8	4	2.4	9.4	2.6	2.1	1.6	0.5	2.5
Aker brygge	**4**	3.1	3.3	2.1	7.6	2	0.86	1.2	<0.5	1.7
Aker brygge	**11**	17	9.6	6.9	24	5.5	1.5	2.8	0.78	4
Kristiansand	**2**	4.1	6.3	3.2	6	4.9	1.7	2.9	0.81	3.4
Kristiansand	**6**	1.4	2	1.1	2.3	1.4	<0.5	0.78	<0.5	1.1
Kristiansand	**7**	6.6	8.8	4.1	8	6.2	<2	4.1	<2	4.5
Frierfjord	**5**	4.2	4.5	1.7	3.6	2.6	0.88	1.6	<0.5	2
Frierfjord	**9**	4.6	6.3	2.3	5	3.8	1.1	2.3	0.64	3.2
Frierfjord	**10**	3.2	4.3	1.6	3.2	2.8	0.65	1.8	<0.5	2.2
Outer Oslofj.	**3**	0.74	0.62	<0.5	0.7	<0.5	<0.5	<0.5	<0.5	<0.5
Outer Oslofj.	**8**	<0.5	<0.5	<0.5	0.51	<0.5	<0.5	<0.5	<0.5	<0.5
Outer Oslofj.	**12**	<0.5	<0.5	<0.5	0.55	<0.5	<0.5	<0.5	<0.5	<0.5

* Naphthalene (Nap.), acenaphthylene (Acy.), acenaphthene (Ace.), fluorene (Fle.), dibenzothiophene (Dbthi.), phenanthrene (Phe.), anthracene (Ant.), fluoranthene (Flu.), pyrene (Pyr.), benzo(a)anthracene (B(a)A.), chrysene (Chr.), benzo(b,j)fluoranthene (B(bj)F.), benzo(k)fluoranthene (B(k)F.), benzo(e)pyrene (B(e)P.), benzo(a)pyrene (B(a)P.), perylene (Per.), Indeno(1,2,3-cd)pyrene (I(123-cd)P.), dibenz(ac/ah)anthracene (Db(ac/ah)A.) and Benzo(ghi)perylene (B(ghi)P.).

Table S8. Concentrations (ng g^{-1} wet wt.) of organochlorine compounds* in *Nereis virens* (Polychaeta) exposed to sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	PCB-28	PCB-52	PCB-101	PCB-118	PCB-105	PCB-153	PCB-138	PCB-156	PCB-180
Aker brygge	**1**	3.9	10	7.1	4.9	2.7	6.2	6.6	0.55	2.3
Aker brygge	**4**	3.6	11	6	4	2.7	5.3	5.7	0.43	1.9
Aker brygge	**11**	4.7	18	9.2	6	3.7	7.9	8.6	0.6	3
Kristiansand	**2**	0.09	6.7	0.5	0.55	0.18	2.2	1.7	0.1	0.75
Kristiansand	**6**	0.09		0.63	0.59	0.18	2.6	2	<0.1	0.85
Kristiansand	**7**	0.08		0.61	0.55	0.19	2.4	1.9	<0.1	0.84
Frierfjord	**5**	0.13	0.34	1.1	0.46	0.16	2.1	1.4	<0.1	0.65
Frierfjord	**9**	0.2	0.48	1	0.47	0.16	2.1	1.4	<0.1	0.73
Frierfjord	**10**	0.31	0.52	1.8	0.59	0.18	2.2	1.6	0.22	0.77
Outer Oslofj.	**3**	0.58	1.6	1.1	1	0.47	2.4	2.1	0.13	0.76
Outer Oslofj.	**8**	0.06	0.25	0.33	0.45	0.17	1.7	1.3	0.07	0.45
Outer Oslofj.	**12**	<0.05	0.22	0.27	0.4	0.14	1.4	1.1	0.06	0.4

Sediment	Box id.	PCB-209	PentaCB	α-HCH	HCB	γ-HCH	OCS	p,p′-DDE	p,p′-DDD
Aker brygge	**1**	<0.05	0.35	<0.05	1.4	<0.05	0.19	0.83	11
Aker brygge	**4**	<0.05	0.34	<0.05	1.1	<0.05	0.18	0.45	8.8
Aker brygge	**11**	0.05	0.43	<0.1	1.4	<0.05	0.21	1	15
Kristiansand	**2**	0.15	0.98	0.06	2.5	<0.05	0.17	<0.05	0.18
Kristiansand	**6**	0.28	0.99	0.08	2.9	<0.05	0.48	<0.05	0.13
Kristiansand	**7**	0.18	0.97	0.05	2.3	<0.05	0.21	<0.05	0.12
Frierfjord	**5**	19	1.5	0.42	8.1	0.08	16	0.13	0.31
Frierfjord	**9**	17	1.7	0.45	7	<0.2	22	0.18	0.41
Frierfjord	**10**	25	4.5	0.63	15	<0.2	35	0.22	0.54
Outer Oslofj.	**3**	<0.05	0.17	<0.05	0.46	<0.05	0.06	0.16	1.6
Outer Oslofj.	**8**	<0.05	0.16	<0.05	0.35	<0.05	<0.05	<0.05	<0.1
Outer Oslofj.	**12**	<0.05	0.13	<0.05	0.32	<0.05	<0.05	<0.05	<0.1

* Polychlorinated biphenyl (PCB) -28, -52, -101, -118, -105, -153, -138, -156, -180, and -209, and pentachlorobenzene (PentaCB), α-hexachlorocyclohexane (α-HCH), hexachlorobenzene (HCB), γ-hexachlorocyclohexane (γ-HCH), octachlorostyrene (OCS), 1,1-dichloro-2,2-bis(4-chlorophenyl)ethene (p,p′-DDE) and 1,1-dichloro-2,2-bis(4-chlorophenyl)ethane (p,p′-DDD).

Table S9. Concentrations (ng g^{-1} wet wt.) of organochlorine compounds* in *Hinia reticulata* (Gastropoda) exposed to sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Box id.	PCB-28	PCB-52	PCB-101	PCB-118	PCB-105	PCB-153	PCB-138	PCB-156	PCB-180
Aker brygge	**1**	0.94	2.4	1.9	1.2	0.48	3.7	3.5	0.21	1.1
Aker brygge	**4**	2.6	5.4	2.9	1.8	0.79	4.5	4	0.25	1.4
Aker brygge	**11**	3.2	6.5	2.9	1.8	0.87	3.9	3.2	0.22	1.1
Kristiansand	**2**	0.09	0.43	0.86	0.64	0.21	3.2	2.7	0.16	0.99
Kristiansand	**6**	0.15	0.86	1.7	1.2	0.38	7.1	5.4	0.33	2.3
Kristiansand	**7**	0.83	5	5.5	4.8	1.6	24	18	1.1	8.1
Frierfjord	**5**	0.2	0.69	1.1	0.75	0.26	4.1	3.4	0.19	1.4
Frierfjord	**9**	0.1	1.3	1.4	0.81	0.22	6.9	5.7	0.33	2.8
Frierfjord	**10**	0.12	0.68	1.5	0.95	0.28	6.2	5	0.31	2.3
Outer Oslofj.	**3**	0.08	0.77	1	0.79	0.27	3.4	2.7	0.17	1
Outer Oslofj.	**8**	0.11	0.55	1	0.77	0.25	4.3	3.6	0.21	1.5
Outer Oslofj.	**12**	0.2	0.84	1.04	1.04	0.33	4.74	3.59	0.26	1.6

Sediment	Box id.	PCB-209	PentaCB	α-HCH	HCB	γ-HCH	OCS	p,p′-DDE	p,p′-DDD
Aker brygge	**1**	<0.05	0.2	<0.05	0.29	<0.05	<0.05	2	1.6
Aker brygge	**4**	<0.05	0.36	<0.05	0.43	<0.05	<0.05	2.9	3.5
Aker brygge	**11**	<0.05	0.31	<0.05	0.51	<0.05	0.07	2.8	6.5
Kristiansand	**2**	<0.05	0.8	<0.05	1.5	<0.05	<0.05	1.7	<0.1
Kristiansand	**6**	<0.05	1.1	<0.05	1.5	<0.05	<0.05	2.1	<0.1
Kristiansand	**7**	0.08	7.1	0.3	8.6	0.17	0.24	11	0.25
Frierfjord	**5**	1.4	3.3	<0.05	4.1	<0.05	1.6	2.1	0.16
Frierfjord	**9**	9.6	1.3	<0.05	4.8	<0.05	4.8	1.6	<0.1
Frierfjord	**10**	2.7	2.5	<0.05	5.1	<0.05	2.3	1.9	<0.1
Outer Oslofj.	**3**	<0.05	0.07	<0.05	0.08	<0.05	<0.05	2.1	<0.1
Outer Oslofj.	**8**	<0.05	0.21	<0.05	0.08	<0.05	<0.05	1.9	<0.1
Outer Oslofj.	**12**	0.14	0.08	0.03	0.11	0.02	0.03	2.02	0.09

* Polychlorinated biphenyl (PCB) -28, -52, -101, -118, -105, -153, -138, -156, -180, and -209, and pentachlorobenzene (PentaCB), α-hexachlorocyclohexane (α-HCH), hexachlorobenzene (HCB), γ-hexachlorocyclohexane (γ-HCH), octachlorostyrene (OCS), 1,1-dichloro-2,2-bis(4-chlorophenyl)ethene (p,p′-DDE) and 1,1-dichloro-2,2-bis(4-chlorophenyl)ethane (p,p′-DDD).

Table S10. Amount total dry matter (TDM; %) and amount lipid (% wet wt.) in *Nereis virens* (Polychaeta) and *Hinia reticulata* (Gastropoda) exposed to sediments from Aker brygge, Kristiansand, Frierfjord and Outer Oslofjord (control). Triplicate experimental boxes.

Sediment	Species	Box id.	TDM	Lipid
Aker brygge	***N. virens***	**1**	12	1.5
Aker brygge	***N. virens***	**4**	13	1.6
Aker brygge	***N. virens***	**11**	13	1.8
Kristiansand	***N. virens***	**2**	13	1.9
Kristiansand	***N. virens***	**6**	13	1.6
Kristiansand	***N. virens***	**7**	13	1.6
Frierfjord	***N. virens***	**5**	12	1.4
Frierfjord	***N. virens***	**9**	13	1.9
Frierfjord	***N. virens***	**10**	13	1.8
Outer Oslofj.	***N. virens***	**3**	14	2
Outer Oslofj.	***N. virens***	**8**	14	2.1
Outer Oslofj.	***N. virens***	**12**	13	1.7

Sediment	Species	Box id.	TDM	Lipid
Aker brygge	***H. reticulata***	**1**	26	0.55
Aker brygge	***H. reticulata***	**4**	24	1.3
Aker brygge	***H. reticulata***	**11**	27	1.5
Kristiansand	***H. reticulata***	**2**	28	0.53
Kristiansand	***H. reticulata***	**6**	27	1.3
Kristiansand	***H. reticulata***	**7**	27	1
Frierfjord	***H. reticulata***	**5**	25	1.2
Frierfjord	***H. reticulata***	**9**	24	1.1
Frierfjord	***H. reticulata***	**10**	21	1.3
Outer Oslofj.	***H. reticulata***	**3**	23	1.3
Outer Oslofj.	***H. reticulata***	**8**	23	1.1
Outer Oslofj.	***H. reticulata***	**12**	25	1

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