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1 **IN VIVO BIOACCUMULATION OF CONTAMINANTS FROM**
2 **HISTORICALLY POLLUTED SEDIMENTS – RELATION TO**
3 **BIOAVAILABILITY ESTIMATES**

4
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11 **Abstract**

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

29
30

31 **Key Words:** Bioavailability, Bioaccumulation, Organochlorine compounds, Polycyclic Aromatic

32 Hydrocarbons, Sediment

33

34 **1. INTRODUCTION**

35

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

45

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

58 2003). In order to benefit from already available knowledge (in terms of structure and coherence
59 of methods), existing guidance systems were reviewed during the development of the Norwegian
60 guidelines. Besides the EU technical guidelines, examples of reviewed systems were Dutch tools
61 for risk assessment of dispersion of contaminants from sediments, as well as US and Canadian
62 guidelines for risk assessment. A common feature of the reviewed methods is that they integrate
63 physical, chemical and biological elements in the risk assessment.

64
65 Risk assessment of contaminated sediments needs to be feasible for a large number of actors, thus
66 the guidelines are of a generic nature. The initial approach of the risk assessments is to compare
67 total contaminant concentrations in the sediments (normalised to the solid phase) with fixed
68 environmental quality standards (Bakke et al., 2010; EC, 2003). However, risk may be
69 overestimated, as limit values are deduced from generic sorption parameters, only considering
70 sorption of contaminants to natural organic matter (Ruus et al., 2010; van der Heijden and Jonker,
71 2009). Many studies, however, have shown that different sedimentary organic matter can be
72 composed of different sorption domains within the particle phase resulting in varying degrees of
73 binding strengths (Arp et al., 2009; Cornelissen et al., 2006a). Thus, a cost-effective approach to
74 ensure sound remediation plans may include measurements of bioavailable concentration
75 estimates or bioaccumulation (Lu et al., 2003; Lu et al., 2011). To this end the use of passive
76 samplers and solid phase extraction techniques has been a much used approach (Cornelissen et
77 al., 2006a; Cornelissen et al., 2006b; Gschwend et al., 2011; Lu et al., 2011; van der Heijden and
78 Jonker, 2009). This is also suggested in the Norwegian guidelines for risk assessment of
79 contaminated sediments (SFT, 2007). A recent evaluation of the most influential factors for the
80 dispersion of contaminants from sediments in the guidelines identified the sediment:water

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

83
84 The objective of this study was to estimate the bioavailability of common organochlorine
85 contaminants and polycyclic aromatic hydrocarbons (PAHs) in selected polluted sediments from
86 Norway by simple generic sorption models (free energy relationships). Measurements of pore
87 water concentrations were previously performed in the same sediments (Allan et al., 2012), and
88 the aim was to predict bioaccumulation from both above mentioned bioavailability estimates for
89 comparison with *in vivo* bioaccumulation assessments.

90
91 *In vivo* bioaccumulation assessments were performed on ragworm (*Nereis virens*, Polychaeta)
92 and netted dogwhelk (*Hinia reticulata*, Gastropoda), while measurements of freely dissolved pore
93 water concentrations were done using polyethylene passive samplers (Allan et al., 2012). The
94 selected sediments originated from different localities in Norway, representative of different
95 types of pollution sources and organic matter content.

96

97 **2. MATERIAL AND METHODS**

98

99 **2.1 Sediment sampling**

100 Sediments from Aker Brygge (59° 54.277 N, 10° 42.985 E; 15 m depth; South-Eastern Norway),
101 Kristiansand harbour (58° 07.495 N, 07° 58.632 E; 28 m depth; Southern Norway), the Frierfjord
102 (59° 06.768 N, 09° 36.963 E; 48 m depth; South-Eastern Norway) and the Outer Oslofjord (59°
103 29.035 N, 10° 36.949 E; 32 m depth; South-Eastern Norway) were collected using a box corer
104 (USNEL 0,25 m² box corer). Triplicate box cores were collected from each site and brought

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

107
108 Aker Brygge is the site of a former shipyard in the Inner Oslofjord (South-Eastern Norway).
109 Frierfjord (the Grenlandfjord area; South-Eastern Norway) has a 50-year long pollution history,
110 where main emissions were organochlorine compounds from a magnesium smelter and PAHs
111 from a ferro-manganese plant. The Kristiansand harbour area (Southern Norway) was
112 contaminated with organochlorine compounds from a metal refinery and PAHs from an electrode
113 paste factory using coal tar pitch. The Outer Oslofjord (South-Eastern Norway) sediment was
114 from a relatively clean site and served as control/reference.

115

116 **2.2 Chemicals**

117 Solvents and other chemicals used are listed in Supplementary Material.

118

119 **2.3 Organisms**

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

126

127 These generally abundant species intimately interact with the sediment (Hayward and Ryland,
128 1995). *N. virens* and *H. reticulata* both prefer sandy or muddy sediment. *H. reticulata* is
129 primarily a scavenger, whereas *N. virens* is omnivorous.

130

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

133

134 **2.4 Passive sampling measurements of freely dissolved concentrations in pore water**

135 The measurement of freely dissolved pore water concentrations was undertaken using low
136 density polyethylene (LDPE) membrane in batch experimental exposure of 3 to 50 days.

137 Concentrations in the pore water (C_{PW}) and resulting total organic carbon-normalised sediment-
138 water partition coefficients (K_{OC}) for the three sediments under investigation (one box from each
139 site) are given in Allan et al. (2012), along with the methods applied (preparation of LDPE
140 passive samplers, LDPE exposure in sediment slurries, extraction and analyses of LDPE
141 samplers).

142

143 **2.5 Experimental setup**

144 In the mesocosm, the boxes with intact/undisturbed sediments (triplicates per site) were
145 submersed to 1-2 cm below the rim in a basin continuously flushed with seawater with salinity,
146 temperature and dissolved oxygen ranging from 33.6 to 34.9 PSU, 6.3-8.3 °C, and 7-9 mg O₂ L⁻¹,
147 respectively. Salinity, temperature and oxygen saturation were logged with WTW
148 (Wissenschaftlich-Technische Werkstätten GmbH, Weilheim, Germany) electrodes every minute
149 in the primary header tank. The seawater was supplied through a pipe-line from 60 m depth in the
150 Oslofjord outside the sill at Drøbak. Separate flows of the same source water was used for

151 continuous exchange of the overlying water in each box-cosm and an air-lift system (Schaanning
152 et al., 2006) was used to ensure a well-mixed, oxygen saturated overlying water. Each box (area:
153 0.25 m²) contained 62-75 L of sediment, with 25-38 L of overlying water. Twenty-two ragworms
154 and 50 netted dog whelk were added to each box (together), and the duration of the exposure was
155 28 days, as recommended by Lee et al. (1991).

156
157 Upon termination of the experiment the overlying water in each box was carefully removed. The
158 organisms from each box were carefully retrieved, and the sediment (within each box) was
159 thoroughly mixed. Aliquots of sediments were taken for chemical analysis and LDPE exposure
160 (pore water concentration measurements; see Allan et al. (2012) for details), as well as analyses
161 of other sediment properties; sediment porosity, fine fraction (% dry wt. <63 µm) and Total
162 Organic Carbon (TOC) content.

163
164 The soft parts of *H. reticulata* were removed from their hard shells using a nut-cracker. The soft
165 parts, as well as individuals of *N. virens* were then rinsed in seawater and transferred to glass
166 containers before storage at -20 °C until chemical analysis.

167

168 **2.6 Extraction and analyses of organisms and sediments**

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

175 treated with concentrated sulphuric acid (H₂SO₄) 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 H₂SO₄.

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

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

205

206 **2.7 Analysis of lipid, total dry matter, organic carbon and particle size fraction.**

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

213

214 **2.8 Calculation of Biota-to-Sediment-Accumulation-Factors (BSAFs)**

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

$$218 \text{BSAF}_{\text{Predicted (Karickhoff)}} = 1.61$$

219 or

$$220 \text{BSAF}_{\text{Predicted (Schwarzenbach)}} = 2.08K_{OW}^{0.02} \quad (\text{for PAHs})$$

221 or

$$222 \text{BSAF}_{\text{Predicted (Schwarzenbach)}} = 0.71K_{OW}^{0.26} \quad (\text{for organochlorine compounds})$$

223

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

225

226
$$\text{BSAF}_{\text{Predicted (Porewater)}} = \frac{C_{\text{lipid}}}{C_{\text{OC}}} = \frac{K_{\text{lipid}} \cdot C_{\text{PW}}}{\left(\frac{C_s}{f_{\text{OC}}} \right)}$$

227

228 where C_{lipid} is the lipid normalised concentration in the organism (here estimated from K_{lipid} and

229 C_{PW}), C_{OC} the organic carbon normalised concentration in the sediment, C_{PW} the concentration in

230 porewater, C_s is the concentration of the compound in the sediment ($\mu\text{g kg}^{-1}$ dry wt.), f_{OC} is the

231 fraction of organic carbon content in the sediment (dry:dry), and $K_{\text{lipid}} = C_{\text{lipid}}/C_{\text{PW}} = K_{\text{OW}}$.

232

233 Finally, BSAFs were calculated from observed concentration in the organisms (*N. virens* and *H.*

234 *reticulata*) applied in the experiments:

235

236
$$\text{BSAF}_{\text{Observed}} = (C_{\text{organism}}/f_{\text{lipid}})/(C_s/f_{\text{OC}})$$

237

238 where C_{organism} is the dry weight concentration in the organism, and f_{lipid} is the fraction of tissue

239 lipid (dry wt.)

240

241 A conceptual sketch of the approaches to deduce and compare BSAFs is given in Figure S1 (see

242 Supplementary Material).

243

244 **3. RESULTS AND DISCUSSION**

245
246 The sediments tested in this study contained a wide range of PAH and especially organochlorine
247 concentrations, with high abundance of some compounds very specific to the pollution source
248 (Tables S1-S2, Supplementary Material). All three sediments displayed a PAH contamination of
249 pyrogenic origin, as the proportions of the larger molecules were high, relative to the lighter
250 compounds (Table S1, Supplementary Material; Neff, 2002). In Aker Brygge, however, the
251 sediments showed a higher influence of petrogenic PAH sources (lower proportions of larger
252 molecule compounds), when compared with the other two contaminated sites (e.g. indicated by a
253 pyrene:benzo(k)fluoranthene-ratio of 5, as compared with 3). This is in agreement with its history
254 as a harbour and site of a former shipyard. Regarding the organochlorine compounds, high
255 concentrations of hexachlorobenzene (HCB) were found in the Kristiansand and Frierfjord
256 sediments, reflecting the contamination by the metal refinery and magnesium smelter,
257 respectively. High concentrations of PCB-209 (as well as proportion of this congener relative to
258 the other PCBs), pentachlorobenzene and octachlorostyrene are also a signature of the
259 magnesium smelter in the Frierfjord. Contaminant concentrations in the outer Oslofjord
260 sediments (control/reference) were low, or below limits of detection (Tables S1-S2,
261 Supplementary Material). The total organic matter content of the contaminated sediments was in
262 the range 3.75-7.01 % dry wt., while TOC in the outer Oslofjord sediment was lower (~1 % dry
263 wt.; Table S3, Supplementary Material). Thus the control/reference sediment had somewhat
264 different characteristics than the other test sediments, a sub-optimal feature that should be kept in
265 mind.

266
267 The range of contaminant concentrations was largely reflected in pore water and organisms
268 (Tables S4-S9, Supplementary Material). However, the bioavailability of PAHs (in terms of

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

285
286 Assuming $K_{lipid} = K_{OW}$ and a free energy relationship between K_{OC} or K_{OW} following Karickhoff
287 et al. (1979) or Schwarzenbach et al. (2003), predicted generic BSAFs equaled approximately 1 –
288 3 (for PAHs) or 1 – 100 (for organochlorine compounds; the higher BSAFs following
289 Schwarzenbach et al., 2003), for the compounds in question ($\log K_{OW} \sim 4 - 8$). Biota-to-sediment
290 accumulation factors predicted from the pore water concentration measurements equalled 0.0002
291 – 2.3 (median: 0.04), and the highest BSAFs were predicted for organochlorine compounds.
292 Thus, for PAHs, the generic BSAFs, following Karickhoff et al. (1979) or Schwarzenbach et al.

293 (2003) were in general two orders of magnitude higher than those predicted from the pore water
294 concentration measurements. For the organochlorine compounds, the generic BSAFs following
295 Karickhoff et al. (1979) agreed fairly well with those predicted from the pore water concentration
296 measurements (that were approximately an order of magnitude lower), while the generic
297 organochlorine BSAFs following Schwarzenbach et al. (2003) were in general two orders of
298 magnitude higher than the BSAFs predicted from the pore water concentration measurements.
299

300 Biota-to-sediment accumulation factors observed in the *in vivo* bioaccumulation experiment
301 corresponded to 0.005 – 2 (median: 0.02) for *N. virens* and 0.0008 – 33 (median 0.02) for *H.*
302 *reticulata*. Again, the higher BSAFs were observed for the organochlorine compounds and the
303 very highest (BSAF = 33) was observed for PCB-52 in *H. reticulata* exposed to Kristiansand-
304 sediment. More specifically, BSAFs (for both species) for the organochlorine compounds were
305 generally in the order 0.1-1, while BSAFs for the PAHs were lower, and generally in the order
306 0.001-0.01 (not shown, but can be deduced from Tables S1-S3 and S6-S10 in Supplementary
307 Material). As such, the *in vivo* BSAFs for PAHs were comparable to or slightly lower than
308 previous BSAFs measured in *N. diversicolor* and *H. reticulata* exposed to a range of PAH-
309 contaminated sediments (Ruus et al., 2010; Ruus et al., 2005) and comparable with BSAFs
310 measured in *N. diversicolor* exposed to pyrene-spiked marine sediments (Granberg and Selck,
311 2007). Furthermore, the BSAFs for organochlorine compounds corresponded well with those
312 previously observed for *N. virens* exposed to PCB-contaminated sediments (Ruus et al., in press),
313 for *N. diversicolor* exposed to sediments collected outside a Norwegian navy base (Ruus et al.,
314 2005), for *Limnodrilus* sp. (Jonker et al., 2004) and *Lumbriculus variegatus* (You et al., 2006;
315 Oligochaeta) exposed to spiked lake sediments, as well as for grass shrimp (*Palaemonetes pugio*)
316 from a contaminated tidal creek system (Maruya and Lee, 1998).

317
318 The BSAFs predicted from the pore water concentration measurements corresponded (as
319 mentioned) fairly well with the BSAFs deduced from the *in vivo* bioaccumulation results (in
320 general within an order of magnitude; note that pore water concentration estimates may
321 underestimate observed bioaccumulation by a factor ~10 for certain compounds and sediments;
322 Figures 1-2). This applies to both PAHs and organochlorine compounds. The variability was also
323 larger than the expected uncertainties in the pore water concentration estimates (a factor of ~2;
324 Allan et al., 2012). There was some evidence that the pore water concentrations over predicted
325 observed bioaccumulation for the higher molecular weight PAH compounds (Figure 1 e. and f.).
326 Barthe et al. (2008) have previously attributed this phenomenon to some steric hindrance of
327 biological membrane permeation by the larger molecules. The possibility of equilibrium not fully
328 reached in the organisms for the most hydrophobic compounds can, however, not be ruled out.
329 For PAHs, the generic BSAFs, following Karickhoff et al. (1979) or Schwarzenbach et al. (2003),
330 represented large overestimates (up to several orders of magnitude), when compared with
331 observed (*in vivo*) ones (Figure 1). The *in vivo* BSAFs for sediments from Aker Brygge generally
332 showed the least discrepancy with the generic BSAFs, following Karickhoff et al. (1979) or
333 Schwarzenbach et al. (2003; Figure 1). For the organochlorine compounds, the generic predicted
334 BSAFs, following Karickhoff et al. (1979) corresponded fairly well with (generally slightly
335 overestimating) the *in vivo* BSAFs (Figure 2). PCBs in the Kristiansand sediment displayed the
336 highest *in vivo* BSAFs, most markedly in *H. reticulata*, for which the observed (*in vivo*) BSAFs
337 were in fact mostly higher than those predicted from the Karickhoff et al. (1979) free energy
338 relationship (Figure 2). On the other hand, generic organochlorine BSAFs following
339 Schwarzenbach et al. (2003) were generally two orders of magnitude higher than those observed
340 in the *in vivo* bioaccumulation experiment (Figure 2). The exception was for PCBs in *H.*

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

343
344 PAHs emitted during the ferro-manganese plant activity in the Frierfjord and the electrode paste
345 factory in Kristiansand may be entrapped in soot and coal particles during their formation, as
346 previously pointed out (Allan et al., 2012; Arp et al., 2009; Jonker et al., 2005; Ruus et al., 2010).
347 In addition, there have been discharges of soot from the Mg plant. This is likely to result in a
348 large proportion of sediment-sorbed PAHs not available for partitioning to sediment pore water
349 and for bioaccumulation. Strong sorption behaviour of coal tar pitch is known, although
350 individual PAH compounds in coal tar pitch can show distinct sorption behaviour (Ghosh and
351 Hawthorne, 2010). Highest $\log K_{OC}$ for PAHs were found for sediments from Frierfjord with
352 values ranging from ~6 to ~9. Lowest $\log K_{OC}$ were for sediments from Aker Brygge with values
353 ranging from ~5 to ~7 (Table S4, Supplementary Material; Allan et al., 2012).

354
355 During the last twenty years a growing number of observations indicate that the model describing
356 a single partition coefficient for organic matter is too simple and that at least a dual model is
357 needed, involving non-linear sorption, expressed through Freundlich coefficients (e.g. Accardi-
358 Dey and Gschwend, 2003; Koelmans et al., 2006):

359
360
$$K_d = f_{AOC}K_{AOC} + f_{BC}K_{BC,F}C_{PW}^{nF-1}$$

361
362 where nF is the Freundlich exponent describing the non-linear sorption, f_{AOC} is the proportion of
363 amorphous organic carbon in the sediment (proportion of TOC that is not black carbon), K_{AOC} is
364 the partition coefficient between amorphous organic carbon and water, f_{BC} is the proportion of

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

374
375 The strong sorption behaviour is obviously a likely explanation of the lower observed BSAFs of
376 PAHs in *N. virens* and *H. reticulata*, compared with the predicted generic BSAFs, following
377 Karickhoff et al. (1979) or Schwarzenbach et al. (2003). Furthermore it is a likely explanation of
378 the generally lower BSAFs in the Frierfjord and Kristiansand sediments, than in the Aker Brygge
379 sediments (with a mixture of petrogenic and pyrogenic sources of PAHs). Organochlorine
380 compounds unlike PAHs were likely not enclosed in soot or black carbon-type particles, but
381 rather adsorbed on their surface, thus lower sediment-water partition coefficients were measured
382 as a result (Allan et al., 2012). These differences in sorption processes of the two classes of
383 chemicals are thus likely responsible for the higher *in vivo* BSAFs for organochlorine, compared
384 with PAH compounds. The reason for the higher bioaccumulation of PCBs from the Kristiansand
385 sediments, especially in *H. reticulata*, as compared with the other sediments, however, is not
386 known. It must be noted that concentrations of PCBs in *H. reticulata* exposed to the outer
387 Oslofjord sediment (i.e. the control/reference) were high, when compared with those exposed to
388 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 $\log K_{OW}$) 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-

413 specific evaluation of bioaccumulation provides information useful to more accurately providing
414 a basis for cost-effective risk assessment and remediation plans. This is more difficult based on
415 total sediment concentrations (Arp et al., 2009; Ghosh and Hawthorne, 2010; Hawthorne et al.,
416 2006; Ruus et al., 2010; van der Heijden and Jonker, 2009). The present study demonstrated
417 apparently more similarities than differences in bioaccumulation behaviour among compounds,
418 between the chosen test species. These were, however, only two of a vast number of different
419 relevant organisms. Therefore, one must also consider possible influence of biological aspects
420 when extrapolating results from such assessments to field conditions. In a recent probabilistic
421 modelling study aiming at explaining differences between bioaccumulation measurements in
422 laboratory and field data, the importance of ingestion of sediments and sediment quality and
423 composition was drawn attention to (Selck et al., 2012). Different benthic invertebrates exhibit
424 different modes of living, which may have a large impact on the degree of exposure to and
425 bioaccumulation of sediment associated contaminants (Meador, 2003). Increased knowledge of
426 the specific characteristics of the vast number of (relevant) species potentially inhabiting
427 contaminated sediments, which may influence the exposure, uptake and elimination of
428 contaminants, would therefore be beneficial.

429

430

431 **Acknowledgements**

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433 172531).

434

435

436 **Supplementary Material**

437 Supplementary material related to this article can be found on-line at

438 doi:10.1016/j.scitotenv.XXXX.XX.XXX;

439

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

443

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

445

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

448

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

450

451 **Table S3.** Amount total dry matter (TDM; %), fraction of particles smaller than $63 \mu\text{m}$ ($P < 63 \mu\text{m}$;
452 % dry wt.) and amount of total organic carbon (TOC; % dry wt.) in sediments.

453

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

458

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

461
462 **Table S6.** Concentrations (ng g⁻¹ wet wt.) of PAHs in *Nereis virens* (Polychaeta) exposed to test
463 sediments.

464
465 **Table S7.** Concentrations (ng g⁻¹ wet wt.) of PAHs in *Hinia reticulata* (Gastropoda) exposed to
466 test sediments.

467
468 **Table S8.** Concentrations (ng g⁻¹ wet wt.) of organochlorine compounds in *Nereis virens*
469 (Polychaeta) exposed to test sediments.

470
471 **Table S9.** Concentrations (ng g⁻¹ wet wt.) of organochlorine compounds in *Hinia reticulata*
472 (Gastropoda) exposed to test sediments.

473
474 **Table S10.** Amount total dry matter (TDM; %) and amount lipid (% wet wt.) in *N. virens* and *H.*
475 *reticulata* exposed to test sediments.

476
477
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588

589

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 ($BSAF_{Observed}/BSAF_{Predicted} = 1$). Staped 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

614 **(b., d. and f.):** *Hinia reticulata*. Solid line: 1:1 relationship ($BSAF_{\text{Observed}}/BSAF_{\text{Predicted}} = 1$).

615 Staped lines: One order of magnitude below and above the 1:1 relationship, respectively.

616

Figure 1.

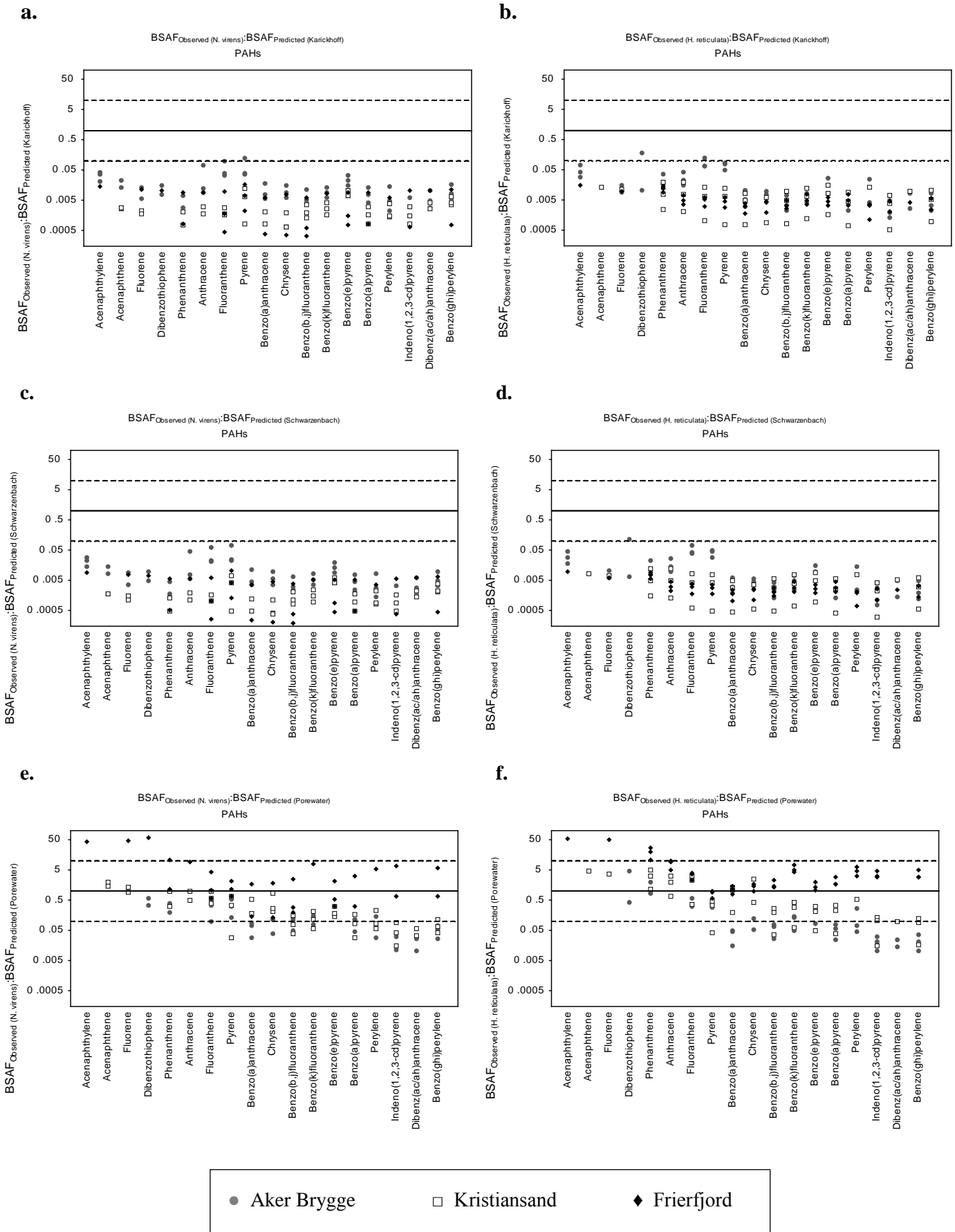
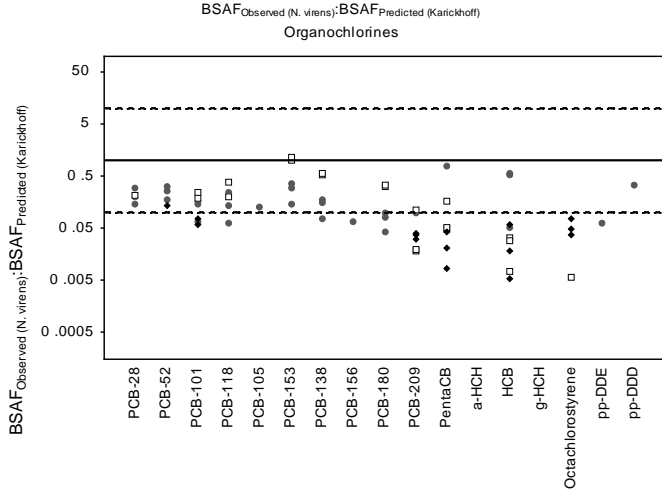
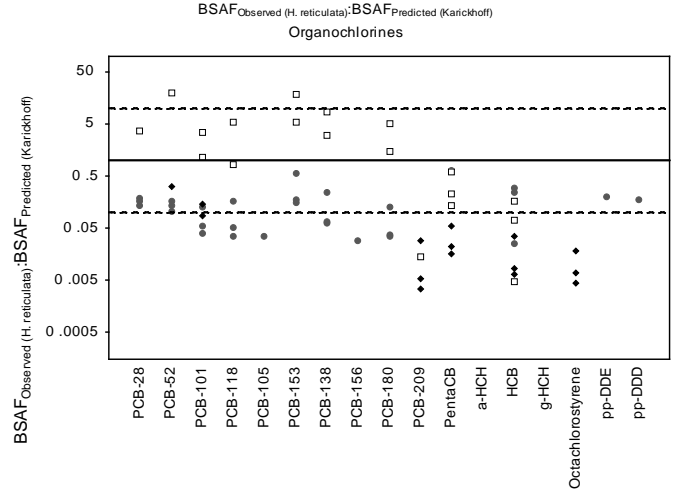


Figure 2.

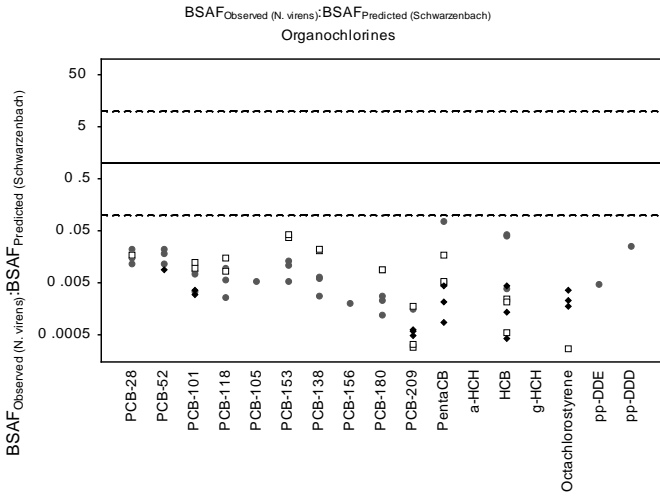
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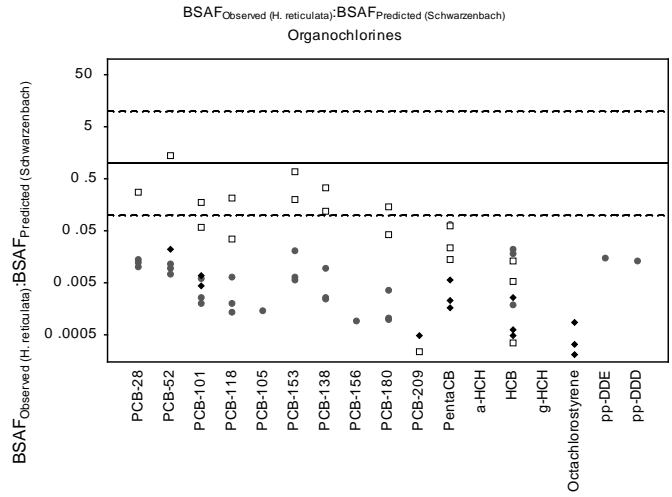
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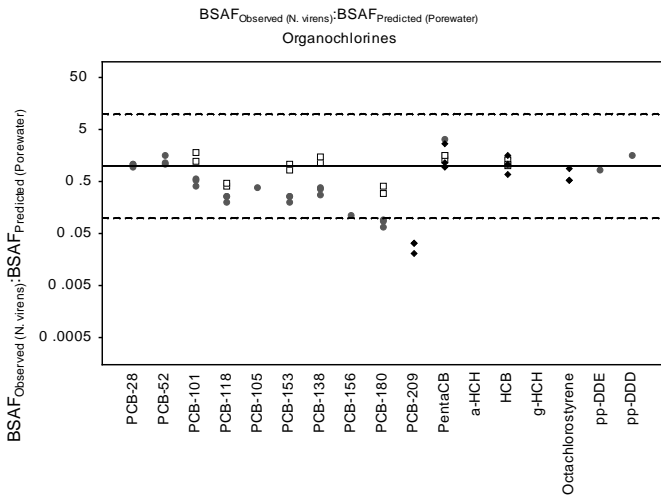
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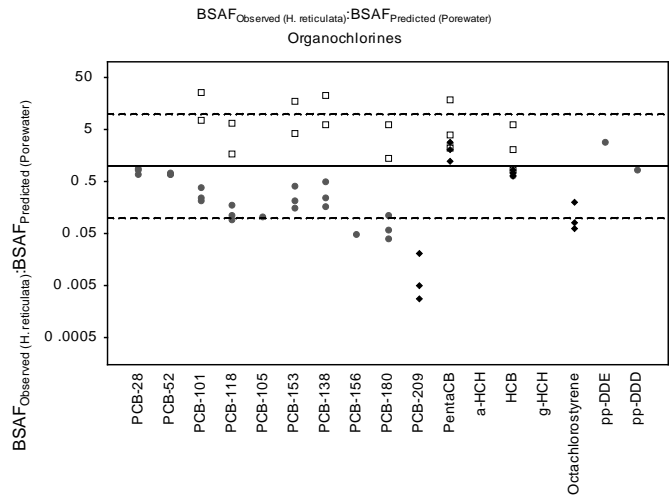
d.



e.



f.



● 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 (H₂SO₄; 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 polyoxilane) 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:

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

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)

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

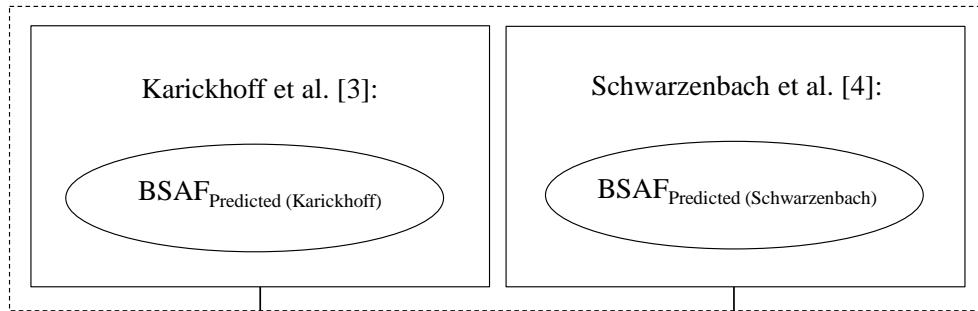
If: $\log K_{OC} = 0.98 \log K_{OW} - 0.32$ or $K_{OC} = 0.48K_{OW}^{0.98}$ (Schwarzenbach et al., 2003; for PAHs)

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

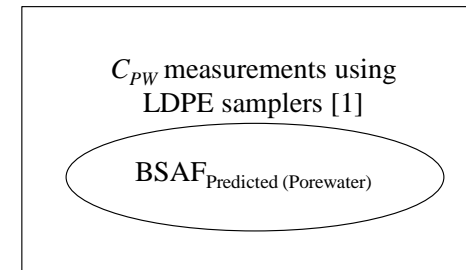
If: $\log K_{OC} = 0.74 \log K_{OW} + 0.15$ or $K_{OC} = 1.4K_{OW}^{0.74}$ (Schwarzenbach et al., 2003; for organochlorine compounds)

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

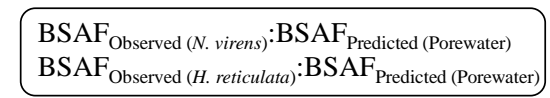
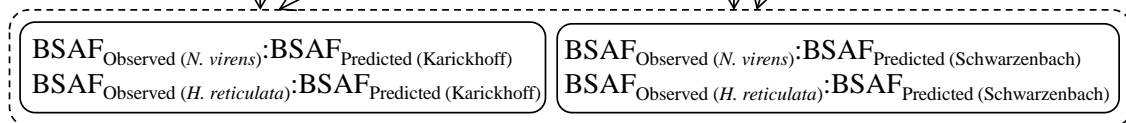
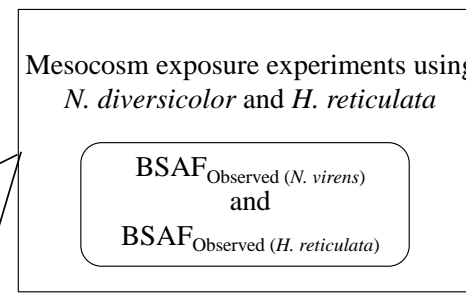
1. Generic predictions of bioaccumulation (using sorption models)



2. Predictions of bioaccumulation from pore-water concentration (C_{PW}) measurements



3. Observed bioaccumulation (*in vivo*)



4. Comparisons between predicted bioaccumulation and observed (*in vivo*) bioaccumulation (Figures 1 and 2)

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 ($P < 63\mu\text{m}$; % 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	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	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	<i>N. virens</i>	1	12	1.5
Aker brygge	<i>N. virens</i>	4	13	1.6
Aker brygge	<i>N. virens</i>	11	13	1.8
Kristiansand	<i>N. virens</i>	2	13	1.9
Kristiansand	<i>N. virens</i>	6	13	1.6
Kristiansand	<i>N. virens</i>	7	13	1.6
Frierfjord	<i>N. virens</i>	5	12	1.4
Frierfjord	<i>N. virens</i>	9	13	1.9
Frierfjord	<i>N. virens</i>	10	13	1.8
Outer Oslofj.	<i>N. virens</i>	3	14	2
Outer Oslofj.	<i>N. virens</i>	8	14	2.1
Outer Oslofj.	<i>N. virens</i>	12	13	1.7

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

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