

**Running head:****Free concentrations and activated carbon amendments**

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**Bioaccumulation of native PAHs from sediment by a polychaete  
and a gastropod: Freely dissolved concentrations and activated  
carbon amendment**

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1 **Abstract**

2 The present paper describes a study on the bioaccumulation of native PAHs from three  
3 harbors in Norway in the polychaete *Nereis diversicolor* and the gastropod *Hinia reticulata*.  
4 First, BSAFs were measured for the original sediments. Median BSAFs were 0.004-0.01 (ten  
5 PAHs; six organism-sediment combinations), which was a factor of 89-240 below the  
6 theoretical BSAF based on total sediment contents (which is around one). However, if BSAF-  
7 values were calculated on the basis of measured freely dissolved PAH concentrations in the  
8 porewater (measured with polyoxymethylene passive samplers), it appeared that these  
9 “BSAF<sub>free</sub>” values were much closer to the actually measured BSAF values, within a factor of  
10 1.7-4.3 (median values for ten PAHs and six organism-sediment combinations). This means  
11 that freely dissolved porewater concentrations are a much better measure for bioaccumulation  
12 than total sediment contents.  
13 Second, we tested the effect of 2 wt-% activated carbon (AC) amendments on BSAF. BSAFs  
14 were significantly reduced (factor 6-7) for *N. diversicolor* in two sediments, i.e. two out of six  
15 organism-sediment combinations, while no significant reduction was observed for *H.*  
16 *reticulata*. This implies that site-specific evaluations of AC amendment are necessary, using  
17 several site-relevant benthic organisms.

18

19 **Key words:**

20 Sediment, Bioaccumulation, PAH, Freely Dissolved Concentrations, Activated Carbon  
21 Amendment

22

## 23 Introduction

24 A steadily growing volume of studies has shown that carbonaceous geosorbents (CG) can  
25 show much stronger sorption of hydrophobic organic chemicals (HOCs) than amorphous  
26 organic carbon (AOC) [1-3]. Examples of CG include unburned coal particles, kerogen, and  
27 soot and charcoal (the residuals of incomplete combustion, commonly termed “black carbon”  
28 or BC). Strong sorption to CG carbon, CGC (exceeding sorption to AOC by one to three  
29 orders of magnitude) has been shown for PAHs, PCBs, polychlorinated dibenzo-*p*-dioxins  
30 and -furans (PCDD/Fs), polybrominated diphenylethers (PBDEs), diuron, benzene,  
31 chlorobenzenes and chlorinated short-chain aliphatic compounds [3].

32 Because bioaccumulation of HOCs is regarded as an equilibrium situation between sediment,  
33 porewater and organisms’ lipids, strong sorption to CGC will reduce concentrations of these  
34 compounds in aquatic and benthic organisms. Thus, with variation in geosorbent quality (e.g.,  
35 CGC and AOC contents), the bioaccumulation of organic compounds can be expected to  
36 vary. This has led to the notion that freely dissolved concentrations provide a better measure  
37 for uptake in organisms and thus environmental risk than total sediment contents [3-6]. From  
38 an engineering perspective, an interesting consequence of strong CGC sorption is that  
39 amendment of such materials to HOC-contaminated sediments should reduce the *in situ* freely  
40 dissolved concentrations [7-9] and thus the actual risk of contaminated sediments. In some  
41 cases, activated carbon (AC) amendment has been shown to effectively reduce the uptake by  
42 benthic organisms of especially PAHs [10] and PCBs from sediments [11,12]. However, in  
43 another case, lipid-normalized contents in organisms were not reduced by amendment of coal  
44 and charcoal [13]. In the latter study, the overall contents in the test organisms were lowered  
45 by the amendments (factor 1.2-8.5), but this effect was completely offset by reductions in  
46 lipid contents hypothesized to be caused by adverse effects of coal and charcoal on habitat  
47 quality.

48 In the present study, native PAHs were studied in sediments from three contaminated and  
49 CGC-rich harbors in Norway, and the tested organisms were the gastropod *Hinia reticulata*  
50 and the polychaete *Nereis diversicolor*. The following two aims were pursued: i) comparison  
51 of BSAFs based on total sediment contents with ones based on freely dissolved  
52 concentrations, in order to evaluate the effect that CGC has on bioaccumulation from CGC-  
53 rich sediments and ii) testing the effect of AC amendments on bioaccumulation, and  
54 comparing this with their effect on freely dissolved concentrations as measured in a previous  
55 study [9]. This is the first study in which these aims are simultaneously pursued, and the one  
56 employing most simultaneous sediment-organism combinations on either aim. The obtained  
57 results will be useful in the contexts of improved risk assessment (freely dissolved  
58 concentrations and bioaccumulation) and of novel *in situ* remediation strategies (AC  
59 amendments).

60

## 61 **Methods**

62 **Sediments.** Samples were taken from three Norwegian harbors with variable degrees of  
63 native PAH pollution (sum-PAH 8.9-161 mg/kg dry weight, dw; Table 1): Oslo (OS), Bergen  
64 (BG) and Tromsø (TR). Sampling locations and characteristics of the sediments (Table 1)  
65 have been described in detail earlier [9,14]. The sediments contain significant amounts of  
66 carbonaceous materials: BC is 6-12% of TOC [14], and total contents of CGC (unburned +  
67 partly combusted carbonaceous carbon) amount to 28-56% of TOC [9]. Due to the presence  
68 of these strongly sorbing materials, total sorption of the native PAHs is much stronger than  
69 according to linear free energy relationships (LFERs) that are based on AOC sorption only  
70 and do not take into account this strong sorption: for example, PHE showed TOC-water  
71 partitioning coefficients ( $K_{\text{TOC}}$ ) almost two orders of magnitude above the LFER- $K_{\text{TOC}}$  value

72 (Table 1). Similar observations were made for the other PAHs that showed sorption 10-100  
73 times stronger than predicted by LFER-values [9].

74 **AC amendments.** Pilot experiments showed that the currently used AC (Sigma-Aldrich  
75 “untreated powder 100-400 mesh”, i.e. 37-149  $\mu\text{m}$ ) would sink in water at 22 °C within 10  
76 min [9]. In contrast to methods described in [7,8,11,12], where the AC was boiled in water  
77 prior to use, we therefore added dry AC to our experimental systems, as boiling the AC is  
78 probably cumbersome and cost-increasing for field applications. AC (2% of sediment dry  
79 weight) was added to 5-kg sediment batches. Water was added until slurries were obtained  
80 (30-50% dry matter) that could be well-mixed by end-over-end shaking (6 rpm;  $20 \pm 1^\circ\text{C}$ ).  
81 These slurries were shaken for 78 d. In our previous study, a time series experiment showed  
82 that 31 d was long enough for redistribution of native PAHs between sediment and AC [9].

83 **Bioaccumulation studies.** These experiments were carried out exactly according to the  
84 procedures described in [15]. The test organisms were the netted dog whelk *Hinia reticulata*  
85 (Gastropoda) and the ragworm *Nereis diversicolor* (Polychaeta). In addition to being  
86 important prey to several bottom-dwelling fish species [16], both organisms are common in  
87 large parts of Europe [17] and able to live in aquaria for extended periods [15]. In spite of the  
88 gastropod’s siphon ventilating to the overlying water, the mode of living of these organisms is  
89 such that they are intimately associated with the sediment so that equilibrium between  
90 organism and pore water can be expected [15]. *H. reticulata* is a scavenger and predator, but it  
91 can also utilize OM in the sediment. *N. diversicolor* uses OM as its sole food source [15].  
92 There are to our knowledge no specific studies regarding the ability of *H. reticulata* to  
93 metabolize PAHs. *N. diversicolor* has been shown to metabolize PAHs to a certain extent;  
94 however, the metabolization rate constants are most likely considerably lower than the uptake  
95 rate constants so that PAH biotransformation does not influence steady-state uptake [15,18].

96 In addition, *N. diversicolor* is one of the most studied marine invertebrates with regard to  
97 bioaccumulation [15,18,19].

98 All of the test organisms were collected in May 2005 at a fixed location in the outer Oslofjord  
99 where concentrations of metals and organic pollutants are on background levels for  
100 Norwegian fjord and coastal sediments. PAH contents in the reference sediment (16-PAH)  
101 were  $\sim 100 \mu\text{g}/\text{kg dw}$ , about 2-3 orders of magnitude lower than the PAH concentrations in  
102 the test sediments. Adult organisms (both sexes) were picked to avoid growth dilution. The  
103 size of the *H. reticulata* individuals was approximately 15 mm in length and 1.3 g in weight  
104 (0.3 g without the shell); the employed specimens of *N. diversicolor* were 40-80 mm long.  
105 The organisms were acclimatized in the laboratory for  $\geq 14$  d in an aquarium with flowing  
106 clean seawater.

107 Briefly, homogenized sediments were placed in 5-L all-glass aquaria. Twenty-one aquaria  
108 each holding 1.5 kg sediment were deployed: aquaria for the original sediments (three  
109 triplicates), for the three AC-amended ones (three triplicates) and for the reference sediment  
110 (one triplicate). This reference sediment originated from the place where the organisms were  
111 collected. *H. reticulata* (10 individuals) and *N. diversicolor* (22 individuals) were added to  
112 each aquarium after 24 h. To provide oxygen, clean seawater (as described above) was  
113 circulated over the sediment in each aquarium at a rate of 150 mL/min. Temperature ( $7.2 \pm$   
114  $0.2^\circ\text{C}$ ), salinity ( $34.8 \pm 0.2$  practical salinity units), pH ( $8.00 \pm 0.05$ ) and oxygen ( $83 \pm 3\%$   
115 saturation) in the water were monitored and found to vary little throughout the experiment.

116 At the termination of the experiment (28 days), all *H. reticulata* and on average 14 out of the  
117 22 individuals of *N. diversicolor* could be retrieved from the aquaria. No significant  
118 differences in mortality were observed between the 18 test sediments and the three reference  
119 sediments (t-test, 95%), so the mortality of *H. reticulata* was probably not caused by the  
120 contamination of the test sediments. The soft parts of the gastropods were separated from



121 their hard shell, and rinsed in seawater. The polychaetes were held in seawater for 12 h to  
122 empty all sediment remnants from their guts. All individuals from one species in each  
123 aquarium were pooled into one sample (3-6 g wet weight). An exposure time of 28 d results in  
124 steady-state tissue residues [20].

125 **Analysis of organism tissue.** To obtain measurable amounts of lipid, small aliquots of the  
126 organism quantity from the triplicate aquaria were pooled into one sample for lipid analysis.  
127 Lipids were determined gravimetrically following Folch et al [21].

128 PAH analysis of the organisms was accomplished as described previously [15]. Briefly,  
129 samples (about 3-6 g wet weight) were homogenized, six deuterated PAHs were added as  
130 internal standards and the mixture was subsequently saponified. The PAHs were extracted  
131 with *n*-pentane and the extracts were dried over sodium sulfate. The extraction volume was  
132 reduced, solvent-exchanged to dichloromethane, and the extracts were cleaned by gel  
133 permeation chromatography and solvent-exchanged to cyclohexane. The extracts were  
134 analyzed by GC-MS (Agilent GC 6890 with MSD 5973) in selected ion monitoring mode.  
135 Measured PAH contents in organisms exposed to the test sediments were not corrected for  
136 initial PAH contents (i.e., the PAH contents in organisms exposed to the reference sediment),  
137 as it was assumed that the organisms in the test sediments would have reached a new steady  
138 state independent of their initial PAH body burden, provided that these initial internal  
139 concentrations were relatively low. Therefore results are reported on the condition that PAH  
140 contents in organisms exposed to test sediment were more than twice the PAH contents in  
141 organisms exposed to the reference sediment.

142

## 143 **Results and discussion**

144 **BSAF of original sediments based on free vs. total concentrations.** Measured BSAFs were  
145 calculated from the data as

$$146 \quad \text{BSAF}_{\text{measured}} = \frac{C_{\text{lipid}}}{C_{\text{TOC}}} \quad (1)$$

147 where  $C_{\text{lipid}}$  (mg/kg lipid) is the lipid-normalized concentration in the organisms and  $C_{\text{TOC}}$  the  
 148 TOC-normalized concentration in the sediment (mg/kg organic carbon). Median  $\text{BSAF}_{\text{measured}}$   
 149 values were 0.004-0.01 for all PAHs in both organisms in all three sediments (Table 2),  
 150 around two orders of magnitude lower than the “theoretical” value ( $\text{BSAF}_{\text{theoretical}}$  being  
 151 slightly above one because lipids are usually a bit more hydrophobic than AOC [22,23]). It  
 152 should be noted that the effects of strong PAH sorption to CGC are not considered in this  
 153 theory. As indicated in the Methods section as well as in Table 1 and Cornelissen et al. [9],  
 154 the presence of CGC caused  $K_{\text{TOC}}$  of our sediments to be more than two orders of magnitude  
 155 higher than LFER- $K_{\text{OC}}$  values based on AOC sorption only.

156 To compare measured BSAFs to theoretical ones, ratios between these two values are  
 157 reported (Table 2). Median  $\text{BSAF}_{\text{theoretical}} : \text{BSAF}_{\text{measured}}$  ratios were as high as 89-240. We  
 158 hypothesize that strong sorption (to CGC) is the reason for the low observed BSAFs. To test  
 159 this hypothesis, BSAFs were also estimated on the basis of the freely dissolved aqueous  
 160 concentrations for the same sediment batches,  $C_{\text{w,free}}$  (determined using the polyoxymethylene  
 161 passive sampler method and reported in Cornelissen et al. [9]). This way, overall TOC  
 162 sorption (to AOC and CGC) is taken into account. Such  $\text{BSAF}_{\text{free}}$  values were determined as

$$163 \quad \text{BSAF}_{\text{free}} = \frac{K_{\text{lipid}} \cdot C_{\text{w,free}}}{C_{\text{TOC}}} \quad (2)$$

164 where  $K_{\text{lipid}}$  is the lipid–water partition coefficient (L/kg; approximated as equal to octanol–  
 165 water partition coefficient ( $K_{\text{ow}}$ ) values from Mackay et al. [24]). It should be pointed out that  
 166 equation (2) is no attempt to correct  $\text{BSAF}_{\text{measured}}$  to the theoretical value. Instead it provides  
 167 an independent chemical method for estimating BSAF that takes into account strong sorption.  
 168  $\text{BSAF}_{\text{free}}$  was compared to the empiric  $\text{BSAF}_{\text{measured}}$  by calculating  $\text{BSAF}_{\text{free}} : \text{BSAF}_{\text{measured}}$   
 169 ratios. These appeared to be close to one (medians 1.7-4.3; Table 2), although estimations for

170  $K_{lipid}$  were used (estimated as being equal to  $K_{OW}$ ). In our previous paper [9], we showed that  
171 the low freely dissolved PAH concentrations (used in eqn. 2) in these sediments could be  
172 explained by strong sorption to CGC. Thus, strong sorption to CGC is probably the reason for  
173 the low BSAF values observed for these sediments. This implies that BSAFs are better  
174 described by freely dissolved aqueous concentrations than by total sediment contents.  
175 In addition, biological/ecological factors can influence the magnitude of BSAF. For example,  
176  $K_{lipid}$  can vary among organisms, and sediment ingestion and gut fluid characteristics may  
177 influence the uptake kinetics and thus the rate at which steady-state is reached [25]. In  
178 addition, the trophic structure of the ecological system can influence BSAFs [26]. However, it  
179 is not likely that variations in ecological factors can explain the observed 100-fold deviations  
180 from  $BSAF_{theoretical}$  such as observed here. In addition, the two tested organisms showed very  
181 similar  $BSAF_{measured}$  for the original sediments. Therefore strong sorption rather than  
182 biological/ecological factors most likely explains the low  $BSAF_{measured}$ . Any noteworthy PAH  
183 biotransformation has likely not influenced our results because of i) the similar findings for  
184 different organisms, and ii) the good agreement between  $BSAF_{free}$  and  $BSAF_{measured}$  (Table 2).  
185 Theoretically, it would be expected that significant PAH biotransformation would lead to  
186  $BSAF_{free}$  being higher than  $BSAF_{measured}$ . In fact, it was observed that  $BSAF_{free}$  slightly  
187 exceeded  $BSAF_{measured}$  by a factor of 1.7-4.3 (Table 2). One possibility is that this difference  
188 can be attributed to (limited) biotransformation, but it is also possible that the “real”  $K_{lipid}$  for  
189 the organisms is below  $K_{OW}$  (in the calculation of  $BSAF_{free}$ , eqn. 2, it was assumed that  $K_{lipid}$   
190 =  $K_{OW}$ ).

191 The present findings are in agreement with earlier observations by Kraaij et al. [27] who  
192 observed that  $C_{W,free}$  measured by Solid-Phase Micro-Extraction (SPME) was a good  
193 predictor for BSAFs of laboratory-added PAHs and PCBs in one specific sediment and for  
194 one particular organism (the oligochaete *Tubifex sp.*). Other observations where strong

195 sorption to CGC probably explained variations in BSAF-values include e.g. i) variation in  
196 BSAFs of organic compounds for deposit-feeders in New York Harbor sediments [26], ii)  
197 BSAFs for mussels being lower for pyrogenic PAHs than for petrogenic ones [28,29], iii)  
198 improved BSAF data modeling for PAH uptake in invertebrates through the inclusion of CGC  
199 [30], iv)  $K_{TOC}$  values being higher [31] and BSAFs being lower [32] in the rainy season than  
200 in the dry period due to higher surface runoff of BC, and v) BSAF values of native PAHs in  
201 six sediments decreasing by approximately a factor of 20 when native BC contents increased  
202 from 0.1 to 0.45% [33].

203 **Effect of AC amendment.** Lipid contents did not show a clear difference between organisms  
204 in the original sediments and in the AC-amended ones (Table 3). Neither were there any clear  
205 differences between organisms exposed to the clean reference sediment and the tested  
206 contaminated ones, with the exception of *H. reticulata*, where the lipid contents were higher  
207 in the test sediments. Unfortunately no statistical analysis can be offered to substantiate the  
208 above comparisons due to the manner in which lipids were quantified (samples for the three  
209 replicate aquaria per sediment had to be pooled in order to obtain enough lipid mass for an  
210 accurate analysis). In spite of this, it can be stated that our results are not in agreement with  
211 those of Jonker et al. [13], who observed that individuals of the aquatic oligochaete  
212 *Limnodrilus sp.* exposed to sediments amended with 1.5 wt-% coal or charcoal for 28 d  
213 possessed an order of magnitude lower lipid contents than worms exposed to original  
214 sediment. We did not see this adverse effect on habitat quality of our AC (charcoal) additions.  
215 Our previous chemical study of the current sediments [9] showed that the presently used AC  
216 was very effective in reducing freely dissolved PAH concentrations in the porewater ( $C_{w,free}$ )  
217 for these sediments:  $C_{w,free}$  was reduced 17-fold (OS), 33-fold (BG) and 55-fold (TR) by 2 wt-  
218 % AC amendment (medians for all PAHs; Table 4 and ref. [9]). The present measurements  
219 showed that bioaccumulation (BSAF) was less strongly reduced by AC amendment than

220  $C_{W,free}$  (Table 4; significant difference for 44 out of 49 comparisons; t-test on individual  
221 PAHs, 95%). BSAFs were significantly reduced for *N. diversicolor* in two of the sediments  
222 (OS and TR; median reduction factor 6 and 7, respectively; Table 4). A t-test (95%) showed  
223 that the reductions in BSAF for *N. diversicolor* in these two sediments were significant for 9  
224 out of 14 comparisons of individual PAHs. For the other four sediment-organism  
225 combinations (*N. diversicolor* in BG sediment, *H. reticulata* in all three sediments)  
226 amendment of 2 wt-% AC resulted in significant BSAF reductions in 4 out of 30 cases (Table  
227 4; t-test on individual PAHs, 95%), so AC amendment appeared hardly effective in those  
228 cases.

229 These results show that differences in the effectiveness of AC amendment probably exist  
230 between sediments and organisms. One must, however, consider the possibility that the  
231 absence of an effect of AC amendment for the snail *H. reticulata* may partly be accounted for  
232 by foldings in its body surface [34]: fine AC particles could adhere in these foldings and  
233 inadvertently be analyzed as taken up by the organism. PAHs sorbed to such particles would  
234 be quantified as bioaccumulated, but in fact they are present in a nonbioavailable form, likely  
235 also for organisms that prey on *H. reticulata*. Unfortunately no organism material was left  
236 after the chemical analyses to test this hypothesis. To a lesser extent, this process of AC  
237 sorbing to the organisms' surface could have played a role for *N. diversicolor* as well. That  
238 leaves, however, unexplained why reduction factors in BSAF were so different between the  
239 three sediments for this organism.

240 Earlier studies provide contradicting information on the effect of amendment of carbonaceous  
241 materials. For example, the addition of 1.5% coal and charcoal led to a seemingly extensive  
242 reduction (factor 1.5-8) of total concentrations of sediment-bound PCBs in *Limnodrilus sp.*  
243 (aquatic worms) [13]; however, when normalizing the results on lipid contents, actually an  
244 increase of BSAFs was observed as the organisms' lipid contents were drastically reduced

245 upon coal/charcoal amendment. Other studies indicate that BSAF is lowered by CGC and AC  
246 additions: i) BSAFs of PCBs were reduced by factors of 3 and 7 for *Neanthes*  
247 *arenaceodentata* (polychaeta) and *Leptocheirus plumulosus* (amphipoda), respectively, upon  
248 the addition of 3.4% AC [11]; ii) the carbonaceous resin Ambersorb was observed to  
249 significantly reduce the bioaccumulation of native PAHs in *Lumbriculus variegatus*  
250 (oligochaeta) [10]; iii) the amendment of 3.4% AC lowered PCB uptake by factors of 14 and  
251 10 for *N. arenaceodentata* and *L. plumulosus*, respectively [12]; iv) uptake of benzo[*a*]pyrene  
252 (BaP) by *Macoma baltica* (bivalvia) was lower from coke, char, anthracite and activated  
253 carbon than from wood and diatoms [36], AC-bound HOCs being up to 60 times less  
254 bioavailable than coke, char and anthracite-bound HOCs; v) BSAFs for sediment amended  
255 with BC-bound pyrogenic PAHs were six times lower than those of the same sediment  
256 amended with BC-bound petrogenic PAHs [35].

257 The present study is the most extensive one so far on the effect of native CGC and amended  
258 AC on bioaccumulation, with three sediments, two organisms and native compounds. In  
259 addition, it is the first one where these two processes are simultaneously studied. In the first  
260 part of the study, it was shown that the effect of CGC on BSAFs very closely follows its  
261 effect on sorption for all organism-sediment combinations, so that freely dissolved  
262 concentrations are accurate predictors of actual bioaccumulation in all cases. In contrast, the  
263 effects of AC amendments on BSAFs were observed to be site- and organism-specific  
264 (significant reductions of BSAFs upon 2% AC amendments for two out of six tested  
265 organism-sediment combinations). This implies that site-specific evaluations of the effect of  
266 AC amendment are necessary, for several benthic organisms that are relevant to the  
267 potentially remediated site.

268

269 **Literature**

- 270 1. Huang W, Peng P, Yu Z, Fu J. 2003. Effects of organic matter heterogeneity on  
271 sorption and desorption of organic contaminants by soils and sediments. *Appl*  
272 *Geochem* 18:955.
- 273 2. Allen-King RM, Grathwohl P, Ball WP. 2002. New modeling paradigms for the  
274 sorption of hydrophobic organic chemicals to heterogeneous carbonaceous  
275 matter in soils, sediments and rocks. *Adv Wat Res* 25:985.
- 276 3. Cornelissen G, Gustafsson Ö, Bucheli TD, Jonker MTO, Koelmans AA, Van  
277 Noort PCM. 2005. Extensive Sorption of Organic Compounds to Black Carbon,  
278 Coal, and Kerogen in Sediments and Soils: Mechanisms and Consequences for  
279 Distribution, Bioaccumulation, and Biodegradation. *Environ Sci Technol* 39:  
280 6881.
- 281 4. Escher BI, Hermens JLM. 2004. Internal Exposure: Linking Bioavailability to  
282 Effects. *Environ Sci Technol* 38:455A-462A.
- 283 5. National Research Council. 2002. *Bioavailability of contaminants in soils and*  
284 *sediments: processes, tools and applications*; National Academies Press:  
285 Washington DC.
- 286 6. Ehlers LJ, Luthy RG. 2003. Contaminant bioavailability in soil and sediment.  
287 *Environ Sci Technol* 37:295A.
- 288 7. Werner D, Higgins CP, Luthy RG. 2005. The sequestration of PCBs in Lake  
289 Hartwell sediment with activated carbon. *Wat Res* 39:2105-2113.
- 290 8. Zimmerman JR, Ghosh U, Luthy RG, Millward RN, Bridges TS. 2004. Addition  
291 of Carbon Sorbents to Reduce PCB and PAH Bioavailability in Marine  
292 Sediments. Physicochemical Tests. *Environ Sci Technol*, 38:5458.

- 293 9. Cornelissen G, Breedveld GD, Kalaitzidis S, Christanis K, Kibsgaard A, Oen  
294 AMP. 2005. Strong Sorption of Native PAHs to Pyrogenic and Unburned  
295 Carbonaceous Geosorbents in Sediments. *Environ Sci Technol*, in press.
- 296 10. West CW, Kosian PA, Mount DR, Makynen EA, Pasha MS, Sibley PK, Ankley  
297 GT. 2001. Amendment of sediments with a carbonaceous resin reduces  
298 bioavailability of polycyclic aromatic hydrocarbons. *Environ Toxicol Chem* 20:  
299 1104-1111.
- 300 11. Millward RN, Bridges TS, Ghosh U, Zimmerman JR, Luthy RG. 2005. Addition  
301 of Activated Carbon to Sediments to Reduce PCB Bioaccumulation by a  
302 Polychaete (*Neanthes arenaceodentata*) and an Amphipod (*Leptocheirus*  
303 *plumulosus*). *Environ Sci Technol* 39:2880.
- 304 12. Zimmerman JR, Werner D, Ghosh U, Millward RN, Bridges TS, Luthy RG.  
305 2005. Effects of dose and particle size on activated carbon treatment to sequester  
306 polychlorinated biphenyls and polycyclic aromatic hydrocarbons in marine  
307 sediments. *Environ Toxicol Chem* 24:1594-1601.
- 308 13. Jonker MTO, Hoenderboom A, Koelmans AA. 2004. Effects of sedimentary  
309 sootlike materials on bioaccumulation and sorption of polychlorinated biphenyls.  
310 *Environ Toxicol Chem* 23:2563.
- 311 14. Oen AMP, Cornelissen G, Breedveld GD. Relation between PAH and Black  
312 Carbon contents in size fractions of Norwegian harbor sediments. *Environ. Poll.*  
313 In press, doi:10.1016/j.envpol.2005.08.033.
- 314 15. Ruus A, Schaanning M, Øxnevad S, Hylland K, 2005. Experimental results on  
315 bioaccumulation of metals and organic contaminants from marine sediments.  
316 *Aquatic Toxicol* 72:273-292.



- 317 16. Ruus A, Uglund KI, Skaare JU. 2002. Influence of trophic position on  
318 organochlorine concentrations and compositional patterns in a marine food web.  
319 *Environ Toxicol Chem* 21:2356-2364.
- 320 17. Moen FE, Svensen E. 2000. *Dyreliv I havet*. 2<sup>nd</sup> ed. Kom forlag , Kristiansund,  
321 576 pp. (in Norwegian).
- 322 18. Giessing AMB, Mayer LM, Forbes TL. 2003. 1-Hydroxypyrene glucoronide as  
323 the major aqueous pyrene metabolite in tissue and gut fluid from the marine  
324 deposit feeding polychaete *Nereis diversicolor*. *Environ Toxicol Chem* 22: 1107-  
325 1114.
- 326 19. Goerke H. 1984. Testing the fate of xenobiotics in *Nereis diversicolor* and *Nereis*  
327 *virens*. In: Persoone G, Jaspers E, Claus C (Eds.) Ecotoxicological testing for the  
328 marine environment. University Gent, Gent, Belgium pp. 53-66.
- 329 20. Lee H, Boese BL, Pelletier J, Winsor M, Specht DT, Randall RC, 1991.  
330 Guidance manual: bedded sediment bioaccumulation tests. EPA/600/x-89/302.
- 331 21. Folch J, Lees M, Stanley G. 1957. A simple method for the isolation and  
332 purification of total lipids from animal tissues. *J Biol Chem* 226:497-509
- 333 22. Di Toro DM, Zabra CS, Hansen DJ, Berry WJ, Swartz RC, Cowan CE, Pavlou  
334 SP, Allen HE, Thomas NA, Paquin PR. 1991. Technical basis for establishing  
335 sediment quality criteria for nonionic organic chemicals using equilibrium  
336 partitioning. *Environ Toxicol Chem* 10, 1541-1583.
- 337 23. Schwarzenbach RP, Gschwend PM, Imboden DM. 2003. *Environmental Organic*  
338 *Chemistry*, 2<sup>nd</sup> ed, John Wiley and Sons Inc., New York.
- 339 24. Mackay D, Shiu WY, Ma KC. 1991. *Illustrated Handbook of physical-chemical*  
340 *properties and environmental fate for organic chemicals*, Vol. I, Lewis Publ.  
341 Inc., Chelsea, MI, pp. 290-488.

- 342 25. Lamoureaux EM, Brownawell BJ. 1999. Chemical and biological availability of  
343 sediment-sorbed hydrophobic organic contaminants. *Environ Toxicol Chem*  
344 18:1733.
- 345 26. Moermond CTA, Zwolsman JJG, Koelmans AA, 2005. Black carbon and  
346 ecological factors affect in situ biota to sediment accumulation factors for  
347 hydrophobic organic compounds in flood plain lakes. *Environ Sci Technol* 39:  
348 3101-3109.
- 349 27. Kraaij H, Mayer P, Busser F, Van het Bolscher M, Seinen W, Tolls J, Belfroid  
350 AC. 2003. Measured pore-water concentrations make equilibrium partitioning  
351 work-a data analysis. *Environ Sci Technol* 37:268.
- 352 28. Farrington JW. 1986. Fossil fuel aromatic hydrocarbon biogeochemistry in the  
353 marine environment: research challenges. In: Giam CS, Dou HJM, eds,  
354 *Strategies and advanced techniques for marine pollution studies: Mediterranean*  
355 *Sea*. Springer-Verlag, Berlin, Germany, pp. 113-142.
- 356 29. Farrington JW, Goldberg ED, Risebrough RW, Martin JH, Bowen VT. 1983. US  
357 "Mussel Watch" 1976-1978: An overview of the trace-metal, DDE, PCB,  
358 hydrocarbon, and artificial radionuclide data. *Environ Sci Technol* 17:490.
- 359 30. Baumard P, Budzinski H, Garrigues P. 1998. Polycyclic aromatic hydrocarbons  
360 (PAHs) in sediments and mussels of the western Mediterranean sea. *Environ*  
361 *Toxicol Chem* 17:765.
- 362 31. Maruya KA, Riseborough RW, Horne AJ. 1996. Partitioning of Polynuclear  
363 Aromatic Hydrocarbons between Sediments from San Francisco Bay and Their  
364 Porewaters. *Environ Sci Technol* 30:2942.

- 365 32. Maruya KA, Riseborough RW, Horne AJ. 1997. The Bioaccumulation of  
366 Polynuclear Aromatic Hydrocarbons by Benthic Invertebrates in an Intertidal  
367 Marsh. *Environ Toxicol Chem* 16:1087.
- 368 33. Sundelin B, Eriksson-Wiklund A-K, Lithner G, Gustafsson Ö. 2004. Evaluation  
369 of the role of black carbon in attenuating bioaccumulation of PAHs from field-  
370 contaminated sediments. *Environ Toxicol Chem* 23:2611.
- 371 34. Hayward PJ, Wigham GD, Yonow N. 1995. Molluscs (Phylum Mollusca). In:  
372 Hayward PJ, Ryland JS (Eds), *Handbook of the marine fauna of North-West*  
373 *Europe*. Oxford University Press, New York, USA, pp. 484-628.
- 374 35. Thorsen WA, Cope WG, Shea D. 2004. Bioavailability of PAHs: Effects of Soot  
375 Carbon and PAH Source. *Environ Sci Technol* 38:2029.
- 376 36. McLeod PB, Van den Heuvel-Greve MJ, Allen-King RM, Luoma SN, Luthy RG.  
377 2004. Effects of Particulate Carbonaceous Matter on the Bioavailability of  
378 Benzo[*a*]pyrene and 2,2',5,5'-Tetrachlorobiphenyl to the Clam, *Macoma balthica*.  
379 *Environ Sci Technol* 38:4549.

**Table 1:** Sediment characteristics of Oslo (OS), Bergen (BG) and Tromsø (TR) sediments. Chemical parameters are presented for the example compound PHE. Chemical data for the other PAHs are in [9].

	<b>OS</b>	<b>BG</b>	<b>TR</b>
<b>Latitude</b>	59° 54' 21" N	60° 23' 26" N	69° 39' 03" N
<b>Longitude</b>	10° 45' 01" E	05° 18' 15" E	18° 15' 42" E
<b>Total Organic Carbon (TOC; %)<sup>a</sup></b>	4.2	7.4	1.7
<b>Total Organic Nitrogen (TON; %)<sup>a</sup></b>	0.26	0.27	0.13
<b>Black Carbon (BC; %)<sup>a</sup></b>	0.25	0.90	0.12
<b>BC:TOC<sup>a</sup></b>	6.1	12.1	6.7
<b>CGC:TOC<sup>b</sup></b>	50.9	55.8	27.9
<b>Total-PAH (mg/kg dw)<sup>c</sup></b>	31.1 ± 1.5	161 ± 31	9.0 ± 1.1
<b>C<sub>SED</sub> (PHE; mg/kg dw)<sup>d</sup></b>	1.10 ± 0.06	6.2 ± 2.4	1.1 ± 0.4
<b>C<sub>w,free</sub> (PHE; ng/L)<sup>e</sup></b>	16.8 ± 2.5	128 ± 21	59 ± 12
<b>log K<sub>TOC</sub> (PHE; L/kg)</b>	6.19 ± 0.09	5.79 ± 0.17	6.13 ± 0.12
<b>log K<sub>AOC</sub> (PHE; L/kg)<sup>f</sup></b>	4.2	4.2	4.2

<sup>a</sup> from [14]

<sup>b</sup> CGC, Carbonaceous Geosorbent Carbon, i.e., the total carbonized OM + the unburned coal carbon, derived from organic micropetrography [9].

<sup>c</sup> dw, dry weight

<sup>d</sup> total PHE concentration in the sediment [9]

<sup>e</sup> Freely dissolved PHE concentration [9]

<sup>f</sup> AOC, amorphous organic carbon. Value derived from a Linear Free Energy Relationship for PAHs:  $\log K_{OC} = 0.98 \log K_{OW} - 0.32$  [36], with  $\log K_{OW}$  from [23].

**Table 2:** BSAF of original Oslo (OS), Bergen (BG) and Tromsø (TR) sediments ( $\text{kg}_{\text{OC}}/\text{kg}_{\text{lipid}}$ ), as well as ratios between theoretical and empiric BSAFs ( $\text{BSAF}_{\text{theory}} / \text{BSAF}_{\text{measured}}$ ) and between BSAFs calculated on the basis of freely dissolved concentrations and empiric ones ( $\text{BSAF}_{\text{free}} / \text{BSAF}_{\text{measured}}$ ).

		<i>Nereis diversicolor</i> (Polychaeta)			<i>Hinia reticulata</i> (Gastropoda)		
		BSAF <sub>measured</sub>	Ratio BSAF <sub>theory</sub> / BSAF <sub>measured</sub>	Ratio BSAF <sub>free</sub> / BSAF <sub>measured</sub>	BSAF <sub>measured</sub>	Ratio BSAF <sub>theory</sub> / BSAF <sub>measured</sub>	Ratio BSAF <sub>free</sub> / BSAF <sub>measured</sub>
OS	PHE	b.d. <sup>a</sup>	x <sup>b</sup>	x	b.d.	x	x
	FLU	b.d.	x	x	0.0115 ± 0.0038	87 ± 29	1.9 ± 0.6
	PYR	0.095 ± 0.010	10.5 ± 1.1	0.30 ± 0.02	0.077 ± 0.009	13 ± 2	0.40 ± 0.02
	CHR	b.d.	x	x	b.d.	x	x
	BBF	0.0057 ± 0.0019	175 ± 57	2.3 ± 0.6	0.0067 ± 0.0038	150 ± 85	2.0 ± 1.0
	BEP	0.038 ± 0.015	26 ± 10	1.5 ± 0.4	0.052 ± 0.021	19 ± 8	1.1 ± 0.3
	BAP	0.0042 ± 0.0015	240 ± 86	3.4 ± 1.0	0.0064 ± 0.0037	156 ± 89	2.2 ± 1.1
	PER	0.0041 ± 0.0011	241 ± 62	1.8 ± 0.4	0.0075 ± 0.0038	133 ± 67	1.0 ± 0.5
	IND	0.0022 ± 0.0005	450 ± 106	10 ± 2	0.008 ± 0.007	119 ± 96	3 ± 2
	BGP	0.0036 ± 0.0014	274 ± 105	6 ± 2	0.012 ± 0.009	81 ± 59	1.8 ± 1.3
	<b>Median</b>	<b>0.0042</b>	<b>240</b>	<b>2.3</b>	<b>0.010</b>	<b>103</b>	<b>1.8</b>
<b>IQR<sup>c</sup></b>	<b>0.0038-0.021</b>	<b>100-258</b>	<b>1.6-4.7</b>	<b>0.006-0.018</b>	<b>66-137</b>	<b>1.1-2.0</b>	
BG	PHE	b.d.	x	x	0.010 ± 0.006	96 ± 56	2.5 ± 0.5
	FLU	0.008 ± 0.004	115 ± 53	3.1 ± 0.6	0.007 ± 0.003	143 ± 67	3.8 ± 0.8
	PYR	0.11 ± 0.04	9 ± 4	0.36 ± 0.05	0.032 ± 0.012	31 ± 11	1.3 ± 0.1
	CHR	0.002 ± 0.002	474 ± 420	6 ± 4	0.002 ± 0.001	473 ± 225	6.0 ± 1.1
	BBF	0.010 ± 0.008	94 ± 70	1.2 ± 0.4	0.007 ± 0.002	138 ± 68	1.7 ± 0.2
	BEP	0.06 ± 0.02	16 ± 7	0.7 ± 0.2	0.032 ± 0.012	31 ± 12	1.4 ± 0.2
	BAP	0.006 ± 0.003	167 ± 72	2.2 ± 0.7	0.006 ± 0.002	156 ± 46	2.1 ± 0.4
	PER	0.010 ± 0.002	99 ± 21	0.9 ± 0.2	0.012 ± 0.002	85 ± 19	0.8 ± 0.1
	IND	0.007 ± 0.006	155 ± 145	5 ± 2	0.016 ± 0.010	63 ± 40	2.0 ± 0.3
	BGP	0.009 ± 0.006	114 ± 72	2.5 ± 0.9	0.018 ± 0.007	56 ± 22	1.2 ± 0.2
	<b>Median</b>	<b>0.0094</b>	<b>107</b>	<b>1.7</b>	<b>0.011</b>	<b>90</b>	<b>1.8</b>
<b>IQR</b>	<b>0.0070-0.051</b>	<b>35-145</b>	<b>0.8-2.9</b>	<b>0.007-0.017</b>	<b>58-142</b>	<b>1.3-2.4</b>	
TR	PHE	b.d.	x	x	b.d.	x	x
	FLU	0.028 ± 0.008	36 ± 5	1.0 ± 0.2	0.016 ± 0.004	62 ± 14	1.7 ± 0.1
	PYR	0.058 ± 0.013	17 ± 6	0.45 ± 0.03	0.015 ± 0.004	69 ± 21	1.8 ± 0.3
	CHR	b.d.	x	x	b.d.	x	x
	BBF	0.011 ± 0.002	93 ± 25	2.8 ± 0.5	0.0057 ± 0.0028	174 ± 85	5 ± 2
	BEP	0.024 ± 0.008	42 ± 13	1.7 ± 0.2	0.011 ± 0.005	85 ± 39	3.4 ± 0.8
	BAP	0.011 ± 0.007	86 ± 50	6 ± 2	0.008 ± 0.007	120 ± 96	8 ± 5
	PER	0.0029 ± 0.0013	347 ± 161	3.0 ± 0.6	0.006 ± 0.007	181 ± 253	1.6 ± 1.8
	IND	0.010 ± 0.008	97 ± 74	11 ± 7	0.012 ± 0.007	85 ± 51	10 ± 5
	BGP	0.007 ± 0.004	146 ± 86	8 ± 4	0.010 ± 0.006	96 ± 56	5 ± 2
	<b>Median</b>	<b>0.011</b>	<b>89</b>	<b>2.9</b>	<b>0.0094</b>	<b>91</b>	<b>4.3</b>
<b>IQR</b>	<b>0.0077-0.027</b>	<b>36-109</b>	<b>1.4-6.2</b>	<b>0.006-0.012</b>	<b>81-133</b>	<b>1.8-5.9</b>	

<sup>a</sup> b.d., below detection, defined as the situation where  $C_{\text{biota}}$  was less than twice the  $C_{\text{biota}}$  in the reference sediment.

<sup>b</sup> x, cannot be calculated because  $C_{\text{biota}}$  was below detection.

<sup>c</sup> IQR, Interquartile range.

**Table 3:** Lipid contents in the presence and absence of AC, of Oslo (OS), Bergen (BG) and Tromsø (TR) sediments as well as clean reference sediment.

Sediment	Lipid content (%)			
	<i>N. diversicolor</i> (Polychaeta)		<i>H. reticulata</i> (Gastropoda)	
	No AC	2% AC	No AC	2% AC
Reference sediment <sup>a</sup>	0.73		0.78	
OS	1.3	0.51	1.3	1.2
BG	0.68	0.94	1.2	1.2
TR	0.82	0.62	1.3	1.6

<sup>a</sup> clean sediment from outer Oslofjord (see text).

**Table 4:** BSAF-values after AC addition (2%), and reduction factors of  $C_{w,free}$  and BSAF due to 2% AC amendment, for Oslo (OS), Bergen (BG) and Tromsø (TR) sediments. For example,  $C_{w,free}$  of PHE in OS sediment was reduced 17-fold by the addition of 2% AC.

		<i>N. diversicolor</i>			<i>H. reticulata</i>	
		Reduction factor $C_{w,free}$ <sup>a</sup>	BSAF <sub>AC</sub> <sup>b</sup>	Reduction factor $C_{biota, lipid}$ <sup>c</sup>	BSAF <sub>AC</sub>	Reduction factor $C_{biota, lipid}$
OS	PHE	17	x <sup>d</sup>	x	x	x
	FLU	53	x	x	0.011 ± 0.003	1.6 ± 0.9
	PYR	46	0.0048 ± 0.0033	29 ± 20	0.010 ± 0.002	11 ± 2
	CHR	31	x	x	x	x
	BBF	18	0.0012 ± 0.0008	7 ± 6	0.0071 ± 0.0017	1.4 ± 0.4
	BEP	17	0.006 ± 0.003	9 ± 5	0.022 ± 0.005	3.4 ± 1.2
	BAP	11	0.0023 ± 0.0016	2.5 ± 2	0.007 ± 0.002	1.3 ± 0.8
	PER	9	0.0043 ± 0.0004	1.4 ± 0.4	0.0095 ± 0.0013	1.1 ± 0.6
	IND	3	0.0030 ± 0.0007	1.1 ± 0.4	0.012 ± 0.002	1.0 ± 0.9
	BGP	3	0.0006 ± 0.0002	9 ± 6	0.014 ± 0.003	1.3 ± 1.1
	<b>Median</b>	<b>17</b>	<b>0.0030</b>	<b>7</b>	<b>0.010</b>	<b>1.3</b>
<b>IQR<sup>e</sup></b>	<b>9-28</b>	<b>0.0018-0.0045</b>	<b>2-9</b>	<b>0.009-0.012</b>	<b>1.2-2.0</b>	
BG	PHE	33	x	x	0.018 ± 0.017	0.7 ± 0.6
	FLU	127	0.009 ± 0.009	1 ± 1	0.008 ± 0.008	1.1 ± 1.1
	PYR	113	0.008 ± 0.006	19 ± 14	0.009 ± 0.008	4 ± 3
	CHR	61	0.003 ± 0.003	0.9 ± 1.1	0.003 ± 0.003	0.8 ± 0.6
	BBF	32	0.008 ± 0.008	1.7 ± 1.2	0.009 ± 0.008	1.0 ± 0.7
	BEP	39	0.018 ± 0.015	4 ± 3	0.025 ± 0.017	1.6 ± 1.0
	BAP	30	0.007 ± 0.006	1.1 ± 1.2	0.007 ± 0.005	1.1 ± 0.8
	PER	16	0.013 ± 0.008	1.0 ± 0.7	0.014 ± 0.009	1.0 ± 0.7
	IND	8	0.02 ± 0.03	0.4 ± 0.5	0.018 ± 0.018	1.1 ± 0.7
	BGP	8	0.014 ± 0.012	0.8 ± 0.7	0.018 ± 0.014	1.2 ± 0.7
	<b>Median</b>	<b>33</b>	<b>0.010</b>	<b>1.2</b>	<b>0.012</b>	<b>1.1</b>
<b>IQR</b>	<b>19-56</b>	<b>0.008-0.014</b>	<b>0.9-3.7</b>	<b>0.008-0.018</b>	<b>1.0-1.2</b>	
TR	PHE	27	x	x	x	x
	FLU	236	x	x	0.03 ± 0.04	1.2 ± 1.8
	PYR	147	0.003 ± 0.002	43 ± 33	0.02 ± 0.03	2 ± 2
	CHR	76	x	x	x	x
	BBF	64	0.005 ± 0.003	5 ± 3	0.03 ± 0.03	0.4 ± 0.6
	BEP	46	0.005 ± 0.001	11 ± 2	0.02 ± 0.02	1.1 ± 1.3
	BAP	66	0.010 ± 0.004	2.0 ± 1.2	0.05 ± 0.06	0.4 ± 0.6
	PER	25	0.0012 ± 0.0003	5.0 ± 1.1	0.012 ± 0.011	1 ± 2
	IND	20	0.0032 ± 0.0005	7 ± 4	0.06 ± 0.07	0.4 ± 0.6
	BGP	18	0.009 ± 0.001	1.5 ± 0.7	0.04 ± 0.04	0.6 ± 0.8
	<b>Median</b>	<b>55</b>	<b>0.0046</b>	<b>6</b>	<b>0.03</b>	<b>0.9</b>
<b>IQR</b>	<b>25-74</b>	<b>0.0031-0.0070</b>	<b>4-19</b>	<b>0.02-0.04</b>	<b>0.4-1.2</b>	

<sup>a</sup> from [9]; <sup>b</sup> BSAF after AC amendment; <sup>c</sup> reduction factor in  $C_{biota}$  (lipid weight basis); <sup>d</sup> x, cannot be calculated because  $C_{biota}$  was below detection (less than twice the concentrations in reference organisms); <sup>e</sup> IQR, Interquartile range.

