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A Combined Field and Laboratory Study on Activated Carbon-Based Thin Layer Capping in a PCB-Contaminated Boreal Lake

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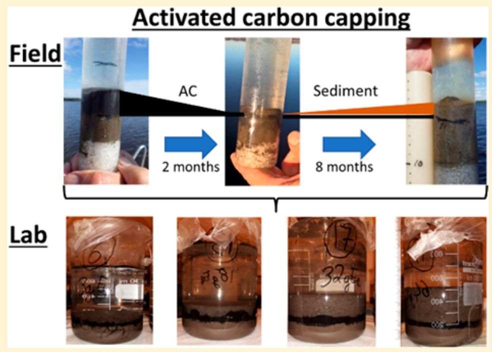
Supporting Information

ABSTRACT: The *in situ* remediation of aquatic sediments with activated carbon (AC)-based thin layer capping is a promising alternative to traditional methods, such as sediment dredging. Applying a strong sorbent like AC directly to the sediment can greatly reduce the bioavailability of organic pollutants. To evaluate the method under realistic field conditions, a 300 m^2 plot in the PCB-contaminated Lake Kernaalanjärvi, Finland, was amended with an AC cap (1.6 kgAC/m^2). The study lake showed highly dynamic sediment movements over the monitoring period of 14 months. This led to poor retention and rapid burial of the AC cap under a layer of contaminated sediment from adjacent sites. As a result, the measured impact of the AC amendment was low: Both the benthic community structure and PCB bioaccumulation were similar on the plot and in surrounding reference sites. Corresponding follow-up laboratory studies using *Lumbriculus variegatus* and *Chironomus riparius* showed that long-term remediation success is possible, even when an AC cap is covered with contaminated sediment. To retain a measurable effectiveness (reduction in contaminant bioaccumulation), a sufficient intensity and depth of bioturbation is required. On the other hand, the magnitude of the adverse effect induced by AC correlated positively with the measured remediation success.



1. INTRODUCTION

Thin layer capping with activated carbon (AC) and other sorbents is a promising method for the *in situ* remediation of contaminated sediments. It relies on the high affinity of hydrophobic organic contaminants (HOCs) to the sorbent particle surfaces. The resulting adsorption is strong enough to lower the pollutant's bioavailability and mobility significantly, thus limiting its uptake into organisms and release from the sediment into the water phase.[1] The costs and labor requirements of AC-based remediation are generally lower compared to traditional remediation methods, such as dredging or capping with nonactive materials like sand or clay. In addition, these conventional methods can cause major disruptions, leading to the deterioration of the local benthic ecosystem.[2,3]

The remediation potential of AC thin layer caps has been clearly demonstrated in a wide range of laboratory trials, with reductions in HOC bioaccumulation and sediment to water fluxes reaching over 90%.[4−6] Field trials on the remediation method have been focusing mostly on the release of contaminants into the water column, which has been shown to be significantly reduced.[7] Especially for long-term remediation success, AC capping has been shown to be more effective than traditional capping methods.[8] Bioaccumulation data for this remediation method under field conditions is less readily available. Samuelsson et al.[9] reported a reduction of HOC uptake by benthic invertebrates of 40%−97% in field-collected sediment cores amended with AC thin layer caps. Most available field studies measuring HOC body burdens of benthic organisms utilize a different application method for the AC, where the sorbent particles are mixed actively into the sediment. Several studies (field and semifield trials) have found greatly reduced contaminant uptake into benthic, as well as pelagic, biota with this mixing treatment.[10,11] For thin layer capping, the remediation potential upon initial application may not be equally high.[6] In the long run, however, burying activities of benthic organisms (bioturbation) can lead to a similar dispersion of AC particles within the biologically active layers of sediment[4,6] and thus comparable remediation efficiencies.

In contrast to the promising reports on the remediation potential of AC amendments, several studies have reported adverse effects of the sorbent material itself to the benthic fauna. The magnitude of these secondary effects induced by AC vary depending on the affected species and have been reviewed by Rakowska et al.[12] and Janssen and Beckingham.[13] The most sensitive species were found to be sediment dwelling organisms, such as for example *Lumbriculus variegatus*, *Chironomus riparius*, or *Arenicola marina*.[14−18] Observed effects in these organisms are most often reduced growth or loss in biomass, reduced emergence rate (*C. riparius*) or lowered feeding rates. Acute

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toxicity (mortality) has been observed in a few cases involving high doses of AC for *Gammarus pulex*[15] and *L. variegatus*.[6] There are also reports of benthic fauna species that are less sensitive to AC amendments, such as *Neanthes arenaceodentata*[19] and *Leptocheirus plumulosus*.[20] Reports about adverse effects under field conditions are contradictory. Reduced species richness, abundance, and biomass were observed in some studies,[7,21] while others found only a limited negative impact of AC amendments or none.[22,23]

The long-term effects of AC amendments can be strongly influenced by sedimentation on the treated site. The deposition of clean sediment settling on AC is intended and supports the remediation success.[24] It could further reduce the adverse effects of the amendment by spatially isolating the benthic fauna from the sorbent layer, which might assist the long-term recovery of benthic communities after the AC application observed in some studies.[25] However, this is not always a realistic scenario. In many cases, water bodies can only be treated partially (hotspot treatment), leaving larger areas with lower contamination levels untreated. Especially in more turbulent waters these untreated sites can become a source for contaminated sediment that can redeposit on top of an applied AC thin layer cap. The same applies to situations where the original source of contamination remains active, e.g., by a diffuse input to larger water bodies. Cornelissen et al.[8] found that AC can retain its remediation efficiency even after being buried under newly deposited, contaminated material and could have a significant advantage over traditional remediation methods. In general, however, data on the remediation potential of AC in such a scenario is sparse.

The aim of this study is to evaluate the applicability of AC thin layer caps for sediment remediation under suboptimal environmental conditions, such as the aforementioned frequent dynamic sediment movements. The incorporation of both field and laboratory trials allows for a thorough and realistic evaluation. The measured parameters were chosen to reflect both the method's risks (adverse effects) and benefits (remediation potential).

2. MATERIALS AND METHODS

2.1. Field Test Site. The central component of this study is the first field trial on AC-based sediment remediation in Finland, which was established in August 2015 in the PCB-contaminated Lake Kernaalanjärvi. (60°85′44′′ N, 24°64′ 21′′ E). A continuous discharge of PCB oils between 1956 and 1984 from a paper mill, located at the lake's tributary Tervajoki River, was the original source of contamination. The PCB load of the sediment in the lake is spatially heterogeneous. The measured concentration in the south part of the lake (where the mouth of the tributary river is located) varied from 4.2 to 10.7 mg/kg according to a 1999 study.[26] The large surface area (4.448 km^2) of the lake, combined with its relatively shallow depth (ca. 1 m around the test site) means that the sediment is frequently affected by water turbulences caused by winds. The potential impact on the long-term success of an AC thin layer cap is one of the central questions in this study. The two major factors in this are likely the aforementioned redeposition of contaminated material from untreated areas of the lake, but also the stability of the cap after its application.

2.2. Field Trial Setup. For the setup of the field trial, a 300 m^2 (10 m × 30 m) test plot in the southern part of Lake Kernaalanjärvi (Map S1) was amended with approximately 1000 kg of pressed SediMite pellets consisting of an AC:clay (1:1) mixture (AC particle size distribution as determined by wet sieving: 69% < 100 μm; 25% 100−200 μm; 6% > 200 μm). The pellets were applied by hand in 5 m × 5 m subplots to ensure an even spread This equals an anticipated AC dose of 1.6 kg/m^2, although the dose practically reached was expected to be lower due to a loss of sorbent particles via drift within the water column. The application of AC in a mixture with clay brings three major advantages. First, the application of the sorbent particles is simplified, as the clay adds bulk to the sorbent material, increasing its density and thus allowing the pressed pellets to sink faster through the water column. This enables the application of the pellets from the water surface, although limiting the use to shallow areas due to the rapid disintegration of the pellets. Furthermore, it has been shown that the addition of clay to the AC can increase the long-term stability of a thin layer cap and lower its adverse effects to the benthic fauna.[6,7] The success of the amendment method (formation of an even, thin layer cap) was controlled with sediment cores taken from the whole plot area 1 day post AC amendment.

2.3. Monitoring of the Field Trial. Monitoring of the field trial was conducted at 2, 10, and 14 months post AC amendment. Sediment cores were taken from the plot to visually examine the development of the AC thin layer cap over time. For the evaluation of adverse effects of the AC material itself and its remediation potential, benthic fauna surveys were conducted at a total of six sites in Lake Kernaalanjärvi: two sites were situated in the AC amended plot (PL-1 in the plot center; PL-2 toward the plot margin) and four surrounding reference sites (Ref-1 to Ref-4) in equal distances (ca. 75 m) from the plot (Map S1). Analyzed parameters for each of the sites were taxa richness, abundance, and biomass (adverse effects), as well as PCB body burdens (remediation potential). The first three parameters were normalized to the total mass of sediment collected from the respective site, and PCB body burdens were adjusted to the background concentrations in the sediment from which the organisms were collected. Lipid normalization of PCB body burdens could not be conducted due to insufficient biota sample sizes. However, the sampled organisms were predominantly either Chironomidae or Oligochaeta, which both have similar, minimal lipid contents (ca. 1% of organism wet weight).[6,27] Background PCB concentrations were normalized to the sediment's total organic carbon content (TOC). A subsample of sediment was taken for each site, for which sediment dry weight (dw) content, TOC, and black carbon (BC) content were determined (n = 3). For TOC measurements, the inorganic carbon was removed from the samples with 1 M H_3PO_4, and BC samples were chemically oxidized with 0.1 M $K_2Cr_2O_7$.[28] The prepared samples were analyzed with an N/C analyzer (Analytik Jena N/C 2100, Jena, Germany). The two month sampling was restricted to PL-1 and Ref-1 sites only.

2.4. Sampling. Bulk sediment samples were collected using 4−5 Ekman grabs (0.122−0.153 m^2; depth approximately 5 cm; 5−9 kg ww) stored at 4 °C and handled within 7 days for the main benthic fauna survey. The samples were sieved (400 μm), and the organisms were sorted into taxonomic groups, counted, and weighed for wet weight (ww; SI-234 analytical balance, Denver Instrument, Bohemia, NY, USA). The following taxonomic groups were chosen as lowest classification level: Chironomidae, Oligochaeta, Copepoda, Hirudinae, Hydrachnidia, Gammeridae, and Trichoptera. Thereafter,

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biota samples containing sufficient biomass for PCB extraction were frozen (−20 °C) and stored for later PCB analysis.

In order to increase the amount of biomass available for PCB analysis, a sediment pump was used to collect larger amounts of surface layer sediment during the 10 and 14 month sampling. These organisms were solely used for PCB measurements (in addition to the aforementioned organisms) and were not included in the benthic fauna survey to maintain a single sampling method. The samples were sieved *in situ* (400 μm) and stored with local lake water until further handling in the laboratory, where organisms (Chironomidae) were collected, weighed for ww, and frozen (−20 °C).

2.5. Sediment Traps. To monitor the amount and quality of new material depositing onto the AC plot, three sediment traps were installed 1 day post AC application. The traps consisted of two submerged PVC tubes (Ø 105 mm), floating at ca. 50 cm above the sediment. At each of the site visits 10 and 14 months post setup, one of the traps was retrieved, while the third one was lost. Upon retrieval, the contained sediment was transferred into glass jars and allowed to settle. The overlying water was then discarded and the sediment weighed (ww). Additionally, the dw content, TOC, and BC of the material were determined ($n = 3$).

2.6. Laboratory Follow-Up Experiments. To assess the effect of the aforementioned dynamic sediment redistribution, an array of laboratory tests was conducted to investigate its impact on both the adverse effects and remediation potential of an applied AC thin layer cap. The tests were conducted in microcosms with a base sediment layer covered by an AC cap. The treatments consisted of sediment layers of varying thicknesses covering the AC (Picture S1). For reference, untreated control microcosms containing only the sediment were included in the tests. Natural, PCB-contaminated sediment from Lake Kernaalanjärvi (area of sampling point Ref-1, see Map S1) was used for both layers in the laboratory trials to create a simplified, but accurate, replication of the field site scenario. The sediment dw content and TOC were determined from a subsample ($n = 3$).

After the base layer of sediment had been added to glass vessels and covered with artificial freshwater (pH 6.5−7.5),[29] powdered AC (particle size <100 μm) was applied as a slurry with a pipet to create a thin layer cap (dose: 1.2 kg AC/m^2). Although slightly finer AC material was used than in the field, the adverse effects and remediation potential have been found to be of similar magnitude if AC particle sizes fall below 200 μm.[6] The top layer sediment was applied with the same method after the AC cap had settled with doses ranging from 1.3 to 63.5 kg/m^2 (sediment ww), which equaled thicknesses of <1 to 40 mm. The actual range of doses used depended on the measured parameters and used test organism. A detailed overview of applied doses and resulting layer thicknesses in each test setup is given in Table S1.

Two different benthic species were chosen to run the tests with: the oligochaete *Lumbriculus variegatus* and chironomid larva *Chironomus riparius*. Although both organisms are sediment dwellers, *L. variegatus* generally inhabit deeper sediment layers than *C. riparius* larvae. Both species are highly important in many freshwater ecosystems. They are widely used in sediment ecotoxicology testing due to their ease of handling and the availability of comprehensive test guidelines.[30,31] The rearing method for the organisms used for the experiments of this study has been described earlier (for *C. riparius* in Waissi-Leinonen et al.[32] and for *L. variegatus* in Abel et al.[6]).

The *C. riparius* tests were conducted using first instar larvae, starting the exposure at 1−3 days post hatching. Egg sacs were transferred from the rearing culture aquaria to glass beakers containing artificial freshwater and monitored on a daily basis to determine the hatching date. Experimental microcosms for the exposure were set up in 1 L glass beakers and contained 40 larvae ($n = 3$). This setup allows a minimum of a 2 cm^2 sediment surface area for each larva as recommended by the OECD guideline.[31] The measured endpoints were the PCB bioaccumulation, growth (final biomass) and survival over an exposure time of 12 days. This time span was set on the basis of preliminary experiments to allow for a maximum exposure time without the risk of any individuals reaching their adult stage. On the final day of the experiment, surviving larvae were sieved out from the sediment (200 μm sieve), counted, and transferred to clean artificial freshwater to depurate their gut contents for 6 h. The organisms were then weighed for ww using a fine scale (Analytic AC 210 P, Sartorius, Göttingen, Germany) and stored at −20 °C in 10 mL test tubes for later PCB extraction.

The measured endpoints for the *L. variegatus* tests were growth as a change in biomass and PCB bioaccumulation over an exposure time of 28 days ($n = 3$). The used range (Table S1) of top sediment layer thicknesses was extended to also include thicker layers covering the AC cap, as preliminary tests showed a much deeper dwelling activity of *L. variegatus* compared to *C. riparius*. Before the organisms were added to the microcosms, they were weighed for initial biomass (ww) and acclimatized overnight in artificial freshwater. On the last day of the exposure period, the worms were removed from the sediment (200 μm sieve) and transferred to clean artificial freshwater for 8 h, allowing them to empty their guts. The organisms were then weighed (ww) into 10 mL test tubes and stored at −20 °C for later PCB extraction.

C. riparius larvae were fed throughout the experiment using a suspension of finely ground TetraMin. This external feeding is required to allow the survival of *C. riparius* in laboratory microcosms but should be limited to the lowest possible level, to reduce its impact on the test results.[33] A feeding level of 0.25 g TetraMin/larva/day was found to be optimal for the experimental conditions (preliminary test). No feeding was necessary to support *L. variegatus*.

2.7. PCB Analysis. For samples obtained from the benthic fauna survey, a modified microscale method described in Jones et al.[34] was used due to the very low biomass of samples. The preweighed tissue (<100 mg ww) was homogenized in 5 mL *n*-hexane for 6 min (repeated twice) with a sonicator. The extracts were cleaned with silica gel column chromatography[35] followed by a sulfuric acid cleanup according to US EPA method 3665A.[36]

Sediment samples were sieved to 1 mm prior to analysis, dried with Na_2SO_4, and Soxhlet-extracted for 2 h with acetone:hexane (1:1 v/v).[27] The extracts were passed through granular Na_2SO_4 and cleaned with concentrated sulfuric acid following Mäenpää et al.[37] Extracts (homogenized using sonication) from biota samples obtained with the sediment pump were cleaned following the same protocol.

The PCB content (21 target congeners) of all samples was measured using GC-MS (Hewlett-Packard series 6890 gas chromatography system coupled with a Hewlett-Packard 5973 mass selective detector) with a method described in Figueiredo

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et al.[27] and Abel et al.[6] Additional details on the used PCB methods can be found in the SI.

2.8. Statistics. All results are expressed as mean ± standard deviation. Statistical analyses were performed using SigmaPlot 13.0 (Systat Software). The data was tested for significant differences between the treatments using one-way ANOVA followed by Dunnett's posthoc tests (α = 0.05). When assumptions for equal variances (Brown−Forsythe test) or normality (Shapiro−Wilk test) were not met, a nonparametric Kruskal−Wallis test followed by Dunn's posthoc test was used instead.

Poor growth of *C. riparius* larvae in the laboratory trial made it necessary to merge organisms from multiple microcosms in some treatment groups for the PCB analysis. This reduced the number of replicates to $n = 1$ and $n = 2$ in the two lowest doses of the applied top sediment layer (1.3 and 2.6 kg/m^2 [0−1 and 1−2 mm], respectively), thus reducing the reliability of the statistical significance found. This did not affect data from the adverse effects bioassay or *L. variegatus* experiments.

The extent and quality (available amount of sample replicates) of the data obtained during the field trial did not allow for thorough statistical analyses. Data from the monitored test sites was merged into a treatment group (plot sites PL-1 and PL2) and a control group (reference sites Ref-1 to 4). The two groups were compared using an independent t test.

3. RESULTS AND DISCUSSION

3.1. General Conditions and Developments at the Field Site. The visual examination of the sediment cores taken from the test plot showed that 1 day post amendment an even layer of AC had formed and capped the sediment. The layer thickness varied between the cores (few cm thick on average) with no cores revealing gaps in the AC cap (Picture S2A). Sediment cores taken two months later showed a significant decrease in AC thickness (Picture S2B). While parts of the cap might have been worked into the underlying sediment via bioturbation,[4] the major factor for the loss is likely one or multiple storm events. However, the AC layer was still visible in most cores taken, and sediment BC values on the plot were significantly elevated (Figure S1 and Table S2). Another effect of the strong winds affecting the lake's bottom is the deposition of large quantities of sediment on top of the AC layer (Picture S2B). This could have further been amplified by the sediment input via the tributary Tervajoki River. The loss of AC from the cap and deposition of sediment continued at a high rate. Only a very thin distinctive AC layer was visible in sediment cores taken 10 months post amendment, with some cores showing no visible traces of AC. Sediment BC contents confirm these visual observations, with values at PL-1 decreasing at after 10 and 14 months (Figure S1). Although samples obtained at PL-2 retained a slightly higher BC level, they are not significantly higher than in all reference sites. This can be caused by a rapid loss of AC due to storm events and the dilution effect of the sediment deposition. In comparison to the AC layer, the amount of newly deposited sediment was high (>20 mm, Picture S2C). The average sedimentation rate measured with the sediment traps were 34.2 ± 1.5 g dry matter/m^2/day. Accumulation rates of this magnitude suggest that the majority of the deposited material was relocated from adjacent, untreated sites due to wind-induced water turbulences.[38] The analysis of the sediment deposited post AC amendment showed a material with higher TOC (52.5−63.5 g/kg, Table S2) and lower dw content (19.7−17.5%). The highly organic sediment that settled on top of the remaining AC layer showed comparatively high PCB concentrations (Table S2 and Figure S2) due to the selective transport of the lighter, organic particles with a high affinity to HOCs.[39] This is a problem that can be expected for many waterbodies in which sediments are susceptible to wind-induced turbulence. One potential solution is the treatment of larger areas of the lake with AC, ensuring no untreated, contaminated sediment could settle in other sites.

The combination of a rapid loss of sorbent particles from the site during storms and the dilution effect of the high sedimentation rates resulted in low AC concentrations (BC values). Nevertheless, it has to be noted that both remediation potential and adverse effects can be observed in very low AC concentrations.[6] While the BC contents measured at the six field sites show that the AC levels are under the intended values, this does not necessarily rule out any impact of the applied sorbent.

The strong spatial variation of PCB and TOC levels in the six monitored sites (Map S1 and Table S2) is not static. Reviewing preceding studies on PCB levels found in the lake showed that the concentrations can change strongly over time. Reported contaminant levels that have been measured in just the south end of the lake (close to the tributary Tervajoki River; same area as this study) were up to 10.7,[26] 4.5,[37] 3.4,[40] and 0.798 mg/kg.[41] This data would further support the theory that sediments in Lake Kernaalanjärvi are frequently relocated (dynamic sediment system), creating changing patches of higher and lower PCB levels. The PCB congener profile was relatively homogeneous among the sediment from the six sampling sites and the sediment traps (Figure S2). The main constituents were tri-, tetra-, penta-, and hexa-CBs.

3.2. Remediation Success. *Field Trial.* The poor retention of sorbent particles on the test plot and the rapid coverage by contaminated sediment led to the assumption that no great remediation success of the AC thin layer cap was to be expected. This was confirmed by the obtained data on PCB bioaccumulation in field-collected benthic invertebrates (Figure 1; homolog-specific data is shown in Table S3). Organisms sampled two months post AC amendment showed lower PCB bioaccumulation compared to an untreated reference site, indicating an initial remediation success. However, after 10 and 14 months, no statistically significant reduction of PCB uptake

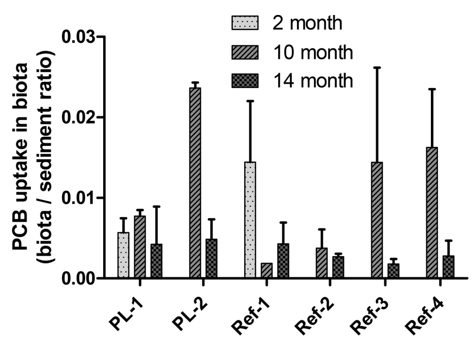

Figure 1. Average bioaccumulation of PCBs, given as ratios of PCB concentrations in biota over TOC-normalized sediment concentrations, measured in field-collected benthic organisms at 2, 10, and 14 months after an AC thin layer cap was applied. For the two month sampling point, data is only available for the sites PL-1 and Ref-1. Sampled organisms were predominantly Chironomids and Oligochaetes (see Figure S3 for more detailed, taxonomic group-specific bioaccumulation data).

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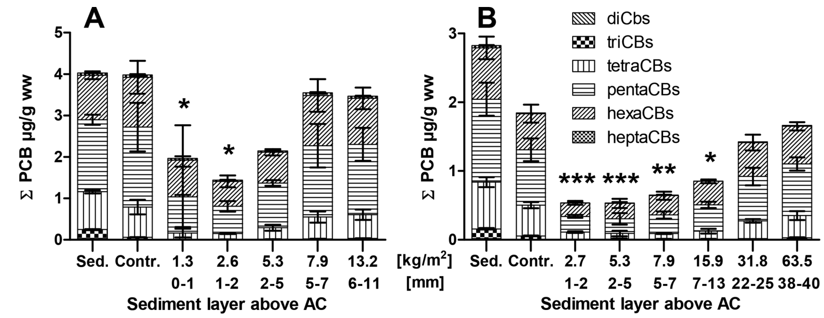

Figure 2. PCB bioaccumulation (laboratory tests) in *Chironomus riparius* (A) and *Lumbriculus variegatus* (B) exposed to sediments amended with an activated carbon thin layer cap that has been covered by increasing amounts of contaminated sediment. Sediment PCB concentrations shown as μg/g dw for comparison (not included in statistical analysis). Significance levels: *** $p < 0.001$, ** $p < 0.01$, * $p < 0.05$.

was found at the AC amended plot, suggesting that high rates of sedimentation have compromised the remediation efficiency of the applied AC cap. Due to the uncertainty in the actual retained AC concentrations at the amended plot (BC measurements), the conclusiveness of the field data is relatively low and should only be evaluated in conjunction with the laboratory follow-up trials.

Laboratory Trial. The hypothesis that the low remediation success observed in the field trial was caused by newly settling, contaminated sediment particles, could generally be confirmed with the laboratory experiments. As seen in the field, the remediation efficiency was diminished with large amounts of contaminated sediment covering the AC layer. With *C. riparius*, the impact of the top sediment layer thickness on PCB uptake by the organism was apparent (Figure 2A). With less than 1 mm of sediment covering the AC layer, the PCB bodyburden in the test organism was reduced by ca. 81% compared to individuals in the unamended control without AC. Although not directly comparable, due to a different AC application method, this is in a comparable range as reported for high doses of AC (2.5% of sediment dw) mixed into the sediment.[42] However, when the amount of covering sediment was increased, the PCB body burdens rose correspondingly, and with 5−7 mm of sediment above the AC (5.3 kg/m^2 or 2−5 mm), there was no longer a statistically significant effect of the AC on PCB uptake.

For *L. variegatus*, the reduction of AC remediation efficiency occurs at significantly higher amounts of sediment covering the sorbent layer (Figure 2B). A high reduction in PCB bioaccumulation of 64%−70% was observed in top sediment layer doses up to 5.3 kg/m^2 or 2−5 mm. These values are in good conformity with previously published results for *L. variegatus* in sediments applied with AC thin layer caps and no further sediment coverage.[4,6] Therefore, it was assumed that the top sediment layer had little to no effect in these doses. A slight increase in PCB uptake was observed when the sediment cover thickness was increased to 7−13 mm (7.9 kg/m^2), although with a 54% decrease in PCB bodyburdens; this still meant a significant reduction compared to the unamended control. Only with at least 22−25 mm (31.8 kg/m^2) sediment covering the AC layer, there was no longer a statistically significant reduction in PCB bioaccumulation. Comparing this to the 2−5 mm (5.3 kg/m^2) that had the same effect on *C. riparius*, it shows that there is a clear difference between the two test organisms. This could be due to the more pronounced sediment dwelling activity of *L. variegatus*, which is known to bury to relatively deep sediment layers (e.g., down to 45 mm observed by Abel et al.[6] under laboratory conditions). Chironomids, on the other hand, inhabit mostly the sediment surface.[43]

These results are comparable to trends that were observed during the field. The sediment layer that deposited within the first two months was still relatively low (5−10 mm; Picture S2). This is comparable to the maximum top sediment layer thickness that showed a lowered PCB bioaccumulation for both organisms during the laboratory follow-up trial. To some extent, this confirms the initial remediation success seen in the field within the first two months after the AC amendment. After 10 months, however, a sediment layer of >20 mm had deposited on the plot. This exceeds the amounts that still enabled a measurable remediation success with either test organism in the laboratory. In addition, the higher share of Chironomids in the samples at 10 and 14 months (due to the use of the sediment pump samples) could have had an impact in masking the remediation efficiency: although not significant, the higher remediation efficiency for Oligochaetes compared to Chironomids was also indicated in the field trial (Figure S3).

The dwelling depth of the two tested organisms provides a sound explanation to the different impact of AC on PCB bioaccumulation. The deeper dwelling activity of *L. variegatus* means that they are exposed to sediment particles which are in closer proximity to the AC layer, even if larger amounts of sediment are deposited. For *C. riparius*, with its habitat being more restricted to the sediment surface, the major impact of an AC thin layer cap is the physical separation from the underlying, contaminated sediment. In addition, the diffusion of contaminants to the newly settled sediment and the water column is prevented by the sorbent layer. If the material deposited after the sorbent application is contaminated, however, the barrier created by the AC cap is no longer sufficient in reducing PCB uptake by the organism. In such cases, the remediation success relies mostly on the passive diffusion of contaminants from the deposited sediment to the sorbent particles beneath.[44] Increasing distances between sorbent and sediment particles slow this process, leading to the increasing PCB uptake seen with increasing top sediment layer thicknesses in the experiments. This can become problematic in remediation scenarios, where sedimentation rates are high or sudden deposition of large amounts of particles can occur, as was the case in Lake Kernaalanjärvi. The diffusion may then become too slow in relation to the input rate of newly settled sediment to sufficiently enable adsorption to the AC.

3.3. Adverse Effects of AC Amendments.

Field Trial. At the 10 and 14 month sampling, the abundance, biomass, and diversity of the sampled invertebrates were low in all six

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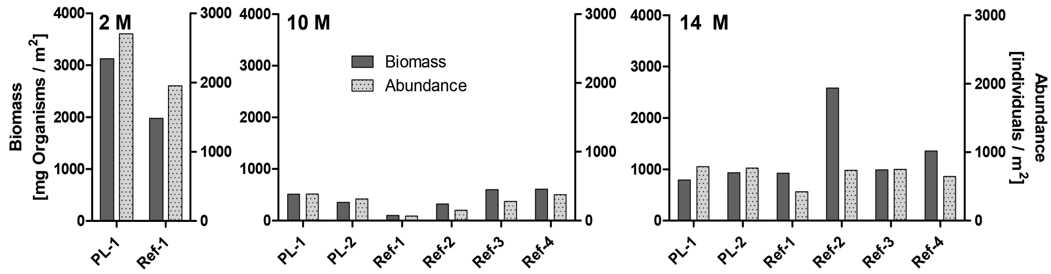

Figure 3. Abundance and biomass (all taxa merged) at 2, 10, and 14 months after an activated carbon thin layer cap was applied at the field site. Two sites within the treated plot (PL-1, PL-2) were sampled and compared to four surrounding reference sites (Ref-1 to Ref-4). Only data for PL-1 and Ref-1 were available for the first sampling. The individual abundance and biomass data of the different taxa are listed in Tables S4−S7.

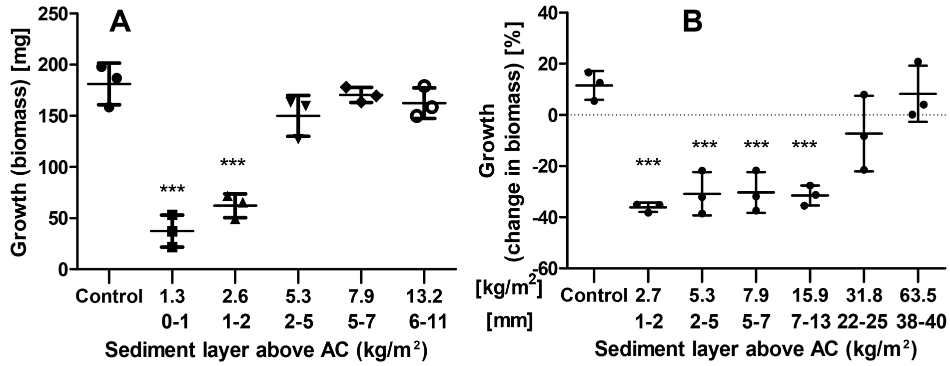

Figure 4. Growth of *Chironomus riparius* (A) and *Lumbriculus variegatus* (B) during the laboratory tests with AC thin layer caps that have been covered with increasing amounts of PCB-contaminated sediment. Unamended sediment was chosen for the control group. (Horizontal lines: mean; whiskers: standard deviation; ***: significant at $p < 0.001$)

investigated sites (Figure 3). At two months, biomass and organism abundance were higher, indicating a strong, yearly fluctuation in the condition of the lake's benthic fauna. This variability has also been observed in benthic fauna surveys conducted earlier (2006−2015) as part of the Figueiredo et al.[27] study and for the online Environmental Information System database (Hertta) of the Finnish Environmental Administration (Table S4).

The vast majority of invertebrates sampled throughout the monitored time period could be classified as Chironomidae or Oligochaeta (Tables S5−S8). These taxa occurred in all sampled sites, while other taxa were found only sporadically. After 2 and 14 months the taxonomic richness (diversity) was slightly higher (4−5) than at 10 months (2−3). The overall low diversity, biomass, and abundance of organisms makes clear conclusions hard to draw. However, the variation between all six sites appears to be higher than between the two sites that were amended with AC and the four reference areas. The adverse impact of AC treatments, such as a reduction in benthic species abundance, biomass, and diversity that has been reported in other field studies, could not be seen from the obtained data.[7,21,22] This can be attributed to the initial loss of AC and thus low retained dose, as well as a potential alleviating effect of the clay addition to the sorbent layer.[6] No statistically significant difference was seen for total biomass or number of organisms between the plot and reference sites at any of the monitoring dates (Figure 3), indicating no adverse impact of the AC amendment over the monitoring period in Lake Kernaalanjärvi. The sampling conducted two months post AC application showed higher abundance and biomass of benthic organisms at the plot site, but the data for this sampling point is not extensive enough to allow for robust statistical analysis. Kupryanchik et al.[15,25] have shown that benthic communities can recover after six months or more post AC application. As the first extensive sampling in this study was conducted at 10 months after the sorbent cap was applied, this recovery could have already taken place. Another explanation could be the aforementioned problems with the AC cap stability and coverage with untreated sediments from adjacent sites. Cornelissen et al.[7] applied a thin layer cap consisting of an AC−clay mixture in a Trondheim harbor channel at 4−6 m depth and reported a loss of around 40% of the initially applied AC after 12 months. The considerably shallower depth at the field site in Lake Kernaalanjärvi, and thus more exposed location, can explain the even higher losses of AC. In combination with the burial of the remaining sorbent under new sediment, these losses could have lowered the general impact on the environment.

Laboratory Trial. Both *L. variegatus* and *C. riparius* were able to survive and increase their biomass over the exposure period in the unamended control sediment and showed strong adverse effects to the AC thin layer cap. These effects were of high magnitude for both organisms when the sorbent layer was covered with only minimal amounts of sediment (Figure 4). While *C. riparius* showed a significantly lower final biomass (growth) than observed in the control, *L. variegatus* showed even a loss in biomass. This is in line with previous publications that showed significant adverse effects of AC to these two test organisms.[6,40,45] However, as soon as the amount of sediment covering the AC layer was increased, adverse effects on the growth and development of *C. riparius* were quickly decreasing. With a layer of 2−5 mm (5.3 kg/m^2) sediment on top of the AC, there was no statistically significant difference in growth rates to the unamended control sediment (ANOVA, Dunnett's posthoc). The survival rate of the larvae followed the same trend as the biomass. While almost no mortality was observed

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in the control, only 22% and 32% of all larvae survived with the two lowest top sediment layers. With 5.3 kg/m^2 (2−5 mm) sediment covering the AC, this was increased to 77.5%. The two highest doses of 7.9 and 13.2 kg/m^2 (5−7 and 6−11 mm) showed good survival rates of 86% and 89% that were no longer significantly lower than in the control.

In contrast, *L. variegatus* showed comparably strong adverse effects (loss of biomass), even when the layer of sediment above the AC cap was increased 7−13 mm (15.9 kg/m^2). Only with more than 20 mm of sediment (31.8 kg/m^2) covering the sorbent layer did the adverse effects started to decrease, and the control level was reached at ca. 40 mm (63.5 kg/m^2) of a top sediment layer.

The adverse effects of AC seem to be closely linked to its remediation potential. The pattern of increased growth strictly follows the increase in PCB uptake for each of the used test species (Figures 2 and 4). This finding suggests that the sediment coverage first and foremost reduces the overall impact of the sorbent on the test organism, both for adverse and beneficial effects. Subsequently, the deeper dwelling activity of *L. variegatus,* which led to the reduced PCB uptake, also caused an increased exposure to the sorbent particles, which in turn causes the observed adverse effects. Vice versa, the stricter spatial isolation from the AC particles of *C. riparius* protected them from the adverse effects at the cost of increased PCB uptake.

This interdependency can help explain the stark contrast concerning the observation of adverse effects in the laboratory and field trial. The overall lower impact of the AC in the field, due to the high sedimentation rate, would not only have resulted in the lower remediation efficiency but also milder adverse effects of the AC itself. The presence of clay in the SediMite pellets used in the field could have further lowered the adverse effects.[6,7] The magnitude of the observed adverse effects in the laboratory could thus be exaggerated.

Another possible cause for the correlation of adverse and beneficial effects is a potential dependence between them. The lowered PCB bioaccumulation can at least partially be a result of the adverse effects caused by the activated carbon, rather than a reduction in PCB bioavailability due to the contaminant binding to the AC particles.[5,46] Reduced feeding rates[6,14,16] or inhibited nutrient assimilation efficiency[20] have been proposed as potential causes for the adverse effects of AC, such as reduced growth. A lowered sediment ingestion rate (feeding), however, would predominantly affect the exposure of the test organisms from the active PCB uptake pathway. The passive (dermal) uptake pathway, on the other hand, is not affected by the sediment ingestion.[47] Nevertheless, the partial dependency between the two parameters can result in an overestimation of the AC's remediation efficiency. The magnitude of this overestimation is influenced by the relative importance of the active uptake pathway, which rises with increasing contaminant hydrophobicity.[48,49] For Lake Kernaalanjärvi sediment, where lower chlorinated and thus less hydrophobic PCB congeners make up the major share of the PCB contamination, the dependency of the remediation efficiency on the level of adverse effects is assumed to be relatively low. Figure 2 and Figure S3 show that organisms in the unamended control sediment take up most homolog groups from the sediment with similar assimilation efficiencies. With AC applied and low sediment deposition, a shift occurs, and the uptake of homologs with the highest degrees of chlorination in the sediment (hexa and hepta-CBs) rises. The trend is reversed with increasing sediment layer thickness above the AC layer. This indicates that homologs favored by the active uptake pathway are accumulated at higher rates, even though feeding is presumably lowered in these treatment groups. However, this could also be caused by a relatively higher adsorption efficiency of the AC for lower chlorinated homologs due to a quicker sequestration.[1,50,51]

4. IMPLICATIONS

A field study by Cornelissen et al.[8] showed that AC-based sediment remediation is generally possible in sites where the input of contaminated sediments cannot be avoided. This could be confirmed by the results from the laboratory trials in this study. However, the comparison of the two test organisms has shown that in these environments the presence of deeper dwelling benthic organisms is vital to enable the long-term success of the remediation works. Only if a sufficient amount of sorbent particles is constantly transported upward via bioturbation into newly settled sediment layers can the remediation success be assured in the long run. The manual coverage of an applied AC layer with clean material would be ill-advised under these conditions, as the physical separation of the biologically active sediment layers from the sorbent particles should be avoided under all circumstances. The importance of this implication can be shown with the low measurable success of the remediation works conducted in Lake Kernaalanjärvi. Besides the overall low biological activity, the composition of the benthic fauna (dominated by shallow dwelling Chironomidae) made AC thin layer application unsuitable for the present conditions at this dynamic, shallow water boreal lake site.

The results obtained in this study indicate that field sites, which are dominated by shallow dwelling organisms, could theoretically recover more quickly after the initial disturbance caused by an AC thin layer cap. This would be highly beneficial in cases where the contaminant source could be reliably shut down, resulting in only clean sediment covering the sorbent layer. The AC cap would remain as a relatively undisturbed layer separating the old, contaminated, and newer clean sediment, thus preventing the flux of contaminants into biologically active sediment layers. A high abundance of deeper dwelling organisms can still be beneficial for speeding up the contaminant sequestration by mixing the cap into the underlying sediment.[52] Additionally, this process helps stabilize the layer physically against drift from the site by water turbulences. This stabilization would otherwise have to be achieved by manually covering the thin layer cap with an additional layer of clean sand or sediment[7] or actively mixing the sorbent into the sediment.[53] However, larger amounts of AC might have to be applied when deeper bioturbation is expected in a site, as low amounts of AC might get diluted too quickly when they are mixed into the sediment.[5]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b05114.

Map of the field site, photos illustrating results and experimental setup, additional graphs and tables on sediment quality parameters, PCB bioaccumulation and benthic fauna survey. (PDF)

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