

# Carbon Amendments and Remediation of Contaminated Sediments



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**Abstract** Sediments are archives of human activities and other environmental changes in the aquatic environment. In many cases they reflect past and present activities in their catchment. Therefore, elevated concentrations of many types of micropollutants, including hydrophobic organic compounds, are found in sediments. However, sediment is not necessarily the final sink for contaminants, as they can pose a threat to local biota as well as to human health. In cases where sediments are toxic or the contaminants bioaccumulate in organisms and/or biomagnify in food chains, remedial actions are considered. This chapter introduces briefly the most common sediment remediation methods including monitored natural recovery and environmental dredging and capping, but the focus is on more recently introduced activated carbon-based sediment amendment technology. All methods come with advantages but also with problems, and these may be contaminant and site specific.

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Overall, carbon amendment technology is shown to be a worthy alternative to the traditional methods. Further development is ongoing to broaden the applicability, for example, and promote biodegradation of the contaminants and environmental conditions under which it can be applied.

**Keywords** Activated carbon, Bioavailability, Contamination, Remediation, Sediment

## 1 Introduction

Sediments are an important part of the aquatic environments, offering habitats for a wide variety of organisms and playing an important role in the control of biogeochemical cycles. Sediments consist of material eroded from the watershed and materials produced within the water body itself. In many cases materials from land dominate. These reflect both natural and anthropogenic processes in the watershed. The solid matter in sediments is a mixture of organic and mineral particles of varying type and size. The quality of sediment varies both among different aquatic systems and within a water body. Heavier and bigger particles are more likely to settle in high-energy environments, whereas lighter and finer materials are deposited in low-energy environments. Seasonal and episodic variations between low- and high-energy conditions (e.g., high flow vs. low flow, storm events) create dynamic sediment conditions at certain sites.

In aquatic systems, sediments form archives of human activities and other environmental changes. They offer a sink for many types of micropollutants but especially for hydrophobic organic chemicals (HOCs) characterized by low water solubility and in most cases also persistence. Once in the sediment, these compounds are well preserved due to low temperature, darkness, and often anoxic conditions, where bio-, photo-, and chemical degradation are negligible. Since the sediments collect pollutants, they may also pose risks to local organisms. Problems can be manifested in two primary ways: (1) the sediment is toxic to benthic fauna, and/or (2) the contaminants bioaccumulate in organisms and can biomagnify in food webs, leading to toxic effects at the top of the food web. Thus, high chemical concentrations can be found, for example, in predatory fish, which can eventually lead to human health risks if the fish from the area are used as a human food source.

Often the total concentration of a chemical in sediment does not directly correlate with the bioaccumulated concentration in organisms or with the observed toxicity. The fraction of chemical that is responsible for the uptake and effects in biota is called the bioavailable fraction. Sorption to sediment (natural) organic matter was noted to play a major role in controlling bioavailability of many organic compounds [1]. However, this could not conclusively explain all variations in sorption [2, 3]. HOCs were shown to have orders of magnitude higher sorption to

sedimentary carbonaceous materials (e.g., black carbon) than to amorphous organic matter. In addition, sorption to carbonaceous materials was found to be stronger than to the amorphous organic matter, emphasizing the importance of black carbon-type materials as a sorbent phase in sediments [4].

These observations led to an investigation of different carbonaceous materials and their influence on bioavailability of HOCs in sediments [5, 6]. Activated carbon (AC), a manufactured carbonaceous material, was found to have a strong sorptive ability [7], and thereafter its effects in sediments were tested [8]. It was quickly deduced that this strong sorption of HOCs to black carbon and AC could be utilized for active remediation of contaminated sites. As summarized in the benchmark paper by Ghosh et al. [9], and discussed further in the latter parts of this chapter, contaminated sediments can be artificially enriched with these strong sorbents to lower the bioavailability and therefore the risks of HOCs.

In the water phase of aquatic systems, remediation of HOC contamination is “easy.” Stopping the discharges will result in the contaminants being degraded, diluted, and/or transported elsewhere. With sediments, things are much more complicated. Due to the ability of sediments to accumulate and store contaminants, high concentrations can be found decades after the termination of the discharge. As the problem rarely solves itself, sediment remediation often needs to be considered. There’s no universal remedy that would fit all sites, contaminants, and situations. Therefore, we need to continue developing new approaches and refine the old ones so that we have methodologies for all kinds of contamination situations.

This chapter will briefly introduce commonly used sediment remediation methods, with special focus on the use of AC amendments as a stabilizing method for contaminants in sediments. The chapter reviews recent scientific literature on the topic, including possible ecological risks and remediation potential, i.e., sorption capacity of AC amendments, and outcomes of field applications.

## **2 Sediment Remediation**

### ***2.1 Need for Remediation***

The need for sediment remediation can arise for several reasons. The fundamental issues are the protection of ecological integrity and human health. The concentration of one or several contaminants exceeding the limits set in national legislation (e.g., sediment quality guidelines) can trigger further evaluation and lead to remedial actions. However, in many countries such guidelines do not exist [10], and, without binding legislation, decisions on whether to take actions are made case by case. Common reasons for initiating remediation include toxicity of the contaminated sediment to benthic organisms, bioaccumulation of the contaminants in the aquatic food chain, and associated human health risks through organisms used as food. The question as to whether remediation is required needs proper risk assessment and expert analysis of weight of evidence. In addition, the financial costs of the

remediation are always an issue. When the decision to remediate has been made, the most suitable remediation method is chosen after weighing the risks and benefits of the alternatives.

## **2.2 *Traditional Methods***

There are three common sediment remediation methods used for sediments contaminated with HOCs: (1) monitored natural recovery (also referred to as monitored natural attenuation), which entails closely following natural processes (physical, chemical, biological) that transform, degrade, immobilize, and/or isolate contaminants in the sediment; (2) dredging or excavation, whereby contaminated sediment is removed, treated, and landfilled; and (3) in situ capping, in which clean material is applied to cover the contaminated sediment on site, physically isolating it from the surrounding environment and reducing exposure of aquatic organisms as well as contaminant fluxes from sediment to water. Common to all three methods is that continuous monitoring is required to ensure that the remediation effort is effective in short and in long term. These methods do not work for new or ongoing contamination, and therefore the first step of all remedial actions should be to eliminate the active sources of the contamination.

All three approaches have their advantages, but none is trouble-free [11]. Dredging is the only method that removes the contaminants from the site. Monitored natural recovery causes the least disturbance, whereas capping and dredging disturb the habitat. Monitored natural recovery may entail restrictions in the use of the area, which could have economic impacts. Despite the possible disadvantages, for several years there has been a clear emphasis on in situ methods, especially capping and monitored natural recovery, for example, in the USA [12].

### **2.2.1 *Monitored Natural Recovery***

Monitored natural recovery is the least invasive remediation method, and it is based on natural processes [13]. In monitored natural recovery, the site is actively monitored, and the progress is assessed to ensure that the risks are reduced over time. Physical, chemical, and biological processes affect either the contaminants themselves or the surrounding environment. Physical processes include burial through ongoing sedimentation of clean material, which forms a natural cap. Concentrations at the site can also decrease as a result of downstream transport of particle-associated contaminants. Chemical attenuation processes can be divided into two types. Contaminants can be sorbed by sediment particulate matter, which reduces their bio-availability. Other chemical processes (e.g., hydrolysis) can lead to degradation or transformation of the chemicals to less harmful breakdown products. Microbial biodegradation can be an important biological process that transforms the

contaminants into more water-soluble and less toxic compounds, in some cases leading to full mineralization.

This method comes with some concerns [14]. Depending on the site characteristics and the remediation goals, recovery can take decades. Thus, this approach will affect and restrict the use of the area for a long period of time, during which the local organisms and humans are still at risk of being exposed to the contaminants. Also, environmental changes or extreme weather events may cause movement of the deposited sediment beds, re-exposure of the highly contaminated layers, and transport of the contaminated sediment into new areas. In addition, a method, which does not contain active remediation, can also encounter resistance among the public. To speed up the natural remedial processes, enhanced monitored natural recovery approaches can be applied, combining environmental engineering with monitoring. These enhancements include the locally restricted, active treatment of highly contaminated hotspots, for example, by removal (dredging) or isolating (capping) of contaminants. This can help in reducing the direct toxic effects of the contaminant to the local micro- and macrofauna, allowing contaminant-degrading organisms to thrive [15].

### 2.2.2 Dredging

Environmental dredging refers to removal of contaminated sediments from predetermined areas and relocation and treatment of the dredged material. Typical equipment for dredging includes grabs, buckets, or different types of suction dredgers. In theory, environmental dredging is straightforward, but practice has shown that it is not always successful. In addition, the method disturbs the local ecosystems [16, 17]. Increased turbidity of water due to suspended sediments and their deposition to different sites throughout the water body can result in habitat changes. Dredging can also re-expose contaminants buried deep in the sediment, and resuspended contaminated sediment particles can increase the concentrations of contaminants in water. This can lead to an increase in exposure and thus increased tissue concentrations in organisms. Longer-lasting problems can be caused by dredging residuals, i.e., contaminated material that is not dredged but is instead left on site or material that escapes from the dredge [17, 18]. Despite the disadvantages, environmental dredging is still an actively used remediation method, especially when persistent chemicals need to be removed from the aquatic environment.

### 2.2.3 Capping

Traditional capping is a remediation method where contaminated sediment is covered with clean materials on site. The capping materials are inert, such as easily obtainable rock, gravel, and clean sediment, which are used to create up to 50 cm thick layers on top of the contaminated sediment. Along with the natural materials, synthetic materials such as geotextiles can be used. The cap physically separates the

contaminated sediment from the surrounding environment, preventing resuspension and transport of the contaminated sediment. At the same time, clean habitats are created for a new succession of benthic communities and other organisms.

The limitations of the traditional *in situ* caps are, for example, that the thick caps are not suitable for shallow sites. Also, the geotextiles can contain persistent and toxic compounds [19]. On the one hand, capping has been shown to be efficient in reducing the diffusion of contaminants through the caps due to the increased diffusion distance [20]. However, the caps do not stop the migration of the contaminants through the cap in all cases due to advection with groundwater seepage [21]. To overcome some of these challenges, active capping and thin layer capping have been developed [22], where the capping materials contain sorbents such as AC with a high affinity for the contaminants (see Sect. 3.1).

### 3 Sediment Remediation with Carbon Amendments

Remediation with sorbent amendments is a relatively new *in situ* remediation option that has been extensively studied since the early 2000s. In this remediation approach, sorbent material is added on top of or mixed into the sediment. The aim is to stabilize the contaminants in the sediment by reducing their freely dissolved concentrations. The contamination is not physically removed, but rather the treatment targets to the reduction of the bioavailable and bioaccessible fractions. In general, the term bioavailability includes internal processes in the organism in addition to processes in the environment [23, 24]. However, here the bioavailable fraction is defined as the fraction of a chemical in the environment that is free to be taken up by organisms and pass biological membranes. In the aquatic environment, it is equal to the freely dissolved fraction. The bioaccessible fraction can be defined as the fraction of a contaminant in sediment that can desorb and thus become bioavailable.

There is a general consensus within the scientific and regulatory community that the bioavailable and bioaccessible fractions of a sediment-associated contaminant are the key drivers for the observed effects of a substance in the environment [25]. Bulk sediment concentrations, which are measured by total, exhaustive extraction of the contaminated sediment with solvents, are far less indicative of the actual risk they pose. Measuring the bioaccessible fraction relies on non-exhaustive extraction of the sediment, using adsorbents instead of organic solvents. The adsorbent is mixed with a sediment-water slurry for different durations to assess fast (<24–48 h mixing) and slower desorbing fractions of a contaminant. The total capacity of the applied sorbent must exceed the fraction of interest, since the saturation of the adsorbent will lead to underestimation of chemical concentrations. The most widely used sorptive phase in this application are Tenax<sup>®</sup> beads, which provide a high capacity for organic contaminants and are easily separable from the sediment slurry. Numerous studies have demonstrated that Tenax extraction can be used reliably to assess the bioaccessible contaminant fraction in sediments (as reviewed by Lydy et al. [26]).

Bioavailability can be approximated by focusing the measurement on porewater concentrations or chemical activity of a contaminant. Several approaches have been proposed and validated over the recent decades. Solid-phase microextraction (SPME) and other passive sampling methods, such as silicone-coated jars [27], have been widely used due to their simplicity and comparably low costs. The key difference to desorption methods, e.g., Tenax extraction, is the non-depletive nature of the passive sampling [28]. Instead, a thermodynamically stable equilibrium is formed between the chemical activities of a compound between the sampler medium (SPME fibers, silicone, or other polymers) and the aqueous phase. Distribution coefficients, specific to the used sampling material, are then used to calculate the porewater concentration of the contaminant. An overview on the underlying concepts of different approaches to passive sampling has been given by Mayer et al. [29].

Several materials have been tested to constrain contaminant fluxes from sediment to water, for example, apatite, chitosan, and thiols to sequester metals [30–32] and organoclays and AC for organic contaminants [17, 33]. Likely due to its established use in water and wastewater treatment, AC amendments have received most attention as a new in situ remediation technique for contaminated sediments [17, 34–36].

In the years since AC-based sediment remediation has been proposed, the remediation efficiency of the method has been evaluated in numerous laboratory and field studies using passive sampling approaches and a range of organisms. The applicability and suitability of AC treatments have been demonstrated for sediments containing polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polychlorinated dibenzodioxins (PCDDs), pesticides, and other similar chemicals [37–43]. In addition, organo-metal-contaminated sediments have been successfully remediated with AC amendments [44, 45]. In many of these studies, the application of AC reduced the contaminant bioavailability by up to 95%.

### ***3.1 Activated Carbon as a Sorbent***

AC is produced by activating a carbonaceous base material, such as charcoal (biogenic ACs or activated biochars) or black coal (petrogenic ACs). Most commonly, this activation is achieved by applying high temperature (>500°C) reinforced with a controlled amount of oxidizing gas (e.g., steam, oxygen, or carbon dioxide). Alternatively, activation may be carried out chemically, using corrosive reagents [46]. Both activation methods aim at opening the pore space in the carbonaceous base material, leading to an end product with highly porosity and a high specific surface area that can easily exceed 1000 m<sup>2</sup>/g [37].

The increase in surface area increases the number of potential sorption sites for chemicals. Therefore, the highly porous AC, with high specific surface area, has a strong affinity for HOCs. Before remediation the contaminants are presumably at an equilibrium between (organic) sediment particles and the sediment porewater. Once AC is brought into the system, the contaminants begin to redistribute from their

previous compartments to the introduced sorbent particles. This redistribution is characterized by an initial, fast phase, in which the fast-desorbing fraction of the contaminant load is sequestered by the AC. This leads to quickly decreasing aqueous (porewater) concentrations of the contaminants within a few days to a month [47, 48]. The slower desorbing fraction of HOCs, however, may require several months before it is transferred to the applied AC [8]. In general, the share of this slowly desorbing fraction rises with increasing hydrophobicity of the contaminant [49].

For an accurate assessment of AC remediation efficiency, it can be important to consider this two-stage mass transfer of contaminants to the AC particles. When only the quickly reduced aqueous concentrations are measured, the actual success of the remediation might be overestimated. The remaining, slow-desorbing fraction that is still bound to sediment particles might not be reflected in the aqueous concentrations. Nonetheless, it can still become bioavailable to organisms that ingest the contaminated sediment particles due to an increased solubility of the contaminant in the presence of digestive fluids [50, 51]. This is reflected to some extent by the – on average – higher measured remediation efficiencies of AC amendments in studies, which focus on passive sampling methods (Fig. 3). McLeod et al. [52, 53] found that the physical mass transfer of contaminants from sediment particles to sorbent particles is a more important mechanism behind the beneficial impact of AC amendments, compared to the reduction of aqueous concentrations. Therefore, the AC amendment's efficiency in reducing concentrations in biota exposed primarily through sediment ingestion may be lower than expected based on the reduction in the freely dissolved concentration. Hence, measuring the freely dissolved porewater concentrations (passive sampling) or freely dissolved and bioaccessible fractions (e.g., different extraction methods) can be suitable methods to approximate bioaccumulation reductions after sorbent remediation. However, directly measuring the uptake into benthic organisms will yield a much more accurate evaluation.

### 3.2 *Applicability*

The high porosity and light texture of AC poses technical challenges to its application in situ. Two different application methods for AC amendments have been used: (1) mixing AC into the sediment on site or applying it premixed with inert material, such as sediment or sand, and (2) direct application as a thin-layer cap. Mixing AC into the sediment during application helps to stabilize the amendment and provides homogenous distribution of AC particles in the biologically active upper sediment layers. Active mixing can significantly accelerate the mass transfer of contaminants, thus yielding fast control of HOC fluxes. On the other hand, mixing technologies may increase the operational costs of the amendment, especially in deeper water [36]. In suitable areas, such as tidal zones [39] or relatively shallow water bodies [64], mixing AC into the sediment can provide quick and efficient contaminant sequestration.



In thin-layer capping, AC is applied to the sediment surface, which can reduce the need for heavy or specialized equipment and thus make the remediation more cost-effective. To enhance the settling, and to improve the placement accuracy, inert binders (e.g., clay) or weighting agents can be added. With direct application, the addition of foreign material to the site is minimized. Therefore, thin layer AC capping may be especially suitable at sites where retaining the water depth is vital, as AC caps generally are only few centimeters thick [43]. Thin-layer capping with AC can create a closed barrier preventing contaminant fluxes from the sediment to the overlying water [65], which can be a strong advantage, especially for heavily contaminated sites where an immediate improvement is desired.

Long-term monitoring of in situ field studies has shown that AC can be well retained in the amended sites. No losses of AC were observed in the lower Grasse River (NY, USA), from plots amended by mixing or by thin-layer capping, during 3-year monitoring [40, 64]. Burial of the amendment due to newly deposited sediment was observed. In addition, downward fluxes of PCBs from water to sediment were reported during the postplacement monitoring period, suggesting that AC can reduce PCB fluxes from sediment in the long term. High stability of a thin-layer powdered AC cap was observed also on active harbor in Bremerton (Washington, USA), after 33 months of deployment [43]. Slightly increasing performance of the amendment over time was reported with increasing reductions of PCB availability to a clam *Macoma nasuta* and polychaete worm *Nephtys aecoides*. Similarly, post-amendment monitoring in Greenland fjords (Norway), 5 years after thin-layer capping, showed increased effectiveness of powdered AC containing caps in reducing polychlorinated dibenzodioxin and dibenzofuran (PCDD/F) fluxes from sediment to water [65]. However, thin layer caps are susceptible to disturbance by environmental factors such as currents or winds, and losses of AC from the amended sites have been reported. The AC levels can be reduced by lateral mixing with untreated surrounding sediment carried into or the AC spread outside the remediated areas [54, 66]. In addition, ongoing sources of contamination, such as newly settling contaminated sediment particles, may hinder the remediation efficiency [54, 67]. On sites with low sediment stability, deep water, or slopes, additional measures such as covering the AC amendment with a layer of sand, clay, or gravel may be required [36].

In addition to the amendment stability, it is important that applied AC gets mixed into the sediment, as this reduces the diffusion distances for the sediment-associated contaminants and facilitates a more complete mass transfer to the AC particles. In thin-layer capping, incorporation of AC into the sediment relies on natural process such as bioturbation, sediment deposition, or other physical processes. For example, in tidal estuarine wetland in upper Canal Creek in Maryland (USA), close to 100% of AC was retained after 10 months of deployment, but the vertical mixing of AC into the wetland sediment proved to be slow [66]. The burrowing activity of the local benthic fauna plays a key role in incorporating AC into the sediment [54, 66, 68, 69]. Thus, AC amendment-induced effects to bioturbating benthic fauna are an important factor in thin-layer cap stability.

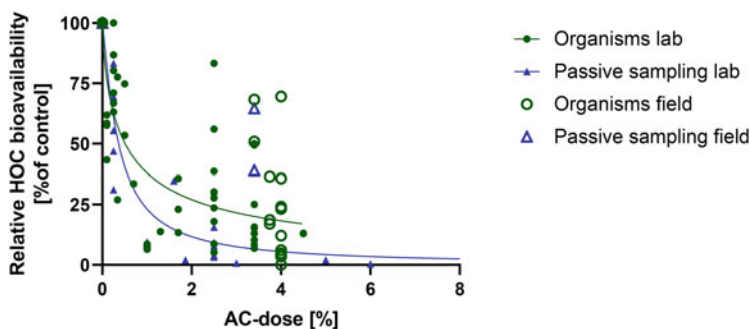
In general, AC amendments are considered less costly compared to traditional dredging and capping. The main costs of the AC amendments are the material and the operational costs. The materials costs are relatively low, approximately 2–3 €/kg AC [36]. Operational costs, however, are highly dependent on the amendment technique and site characteristics. The selection of the most suitable approach is always determined by site-specific characteristics, as well as the ecological and health risks posed by the contaminated site [36, 70].

### 3.3 Remediation Efficiency

The raw material used to produce the AC seems to be of only minor importance for the remediation efficiency. Experimental studies have shown both equal [55] and lower performances of biogenic ACs compared to petrogenic ACs in HOC bioavailability reduction [71, 72]. The difference in performance is likely explained by the pore structure of the ACs in synergy with the molecular size of the target contaminant. Certain biogenic ACs may exhibit smaller pores, which can lead to fast pore blockage by large molecules and thus reduced adsorption capacities [71, 72].

The most dominant factor determining the contaminant binding potential is the particle size of the ACs. It is evident that the remediation efficiency markedly increases with decreasing sorbent particle size. As reviewed by Rakowska et al. [34], finer powdered ACs (PACs) have two major advantages. Firstly, the distribution within the sediment is denser for PAC, given that the same doses are applied. This reduces the required contaminant diffusion pathways between sediment and AC particles. Secondly, the external surface area becomes larger in proportion to the quantity of applied sorbent. The accessibility of pores within the AC particles can be obstructed and, thus, especially for larger molecules, hamper their adsorption [71]. This issue can be further exacerbated when the outer pores of larger AC particles (granular AC; GAC) are clogged by sediment particles, reducing the available pore space for contaminant adsorption significantly [56]. In the most severe published cases, this has caused GAC to bind insufficient or even no measurable amounts of contaminants [37, 52, 56].

The remediation efficiencies of AC amendments show a clear dose-response relationship (Fig. 1). In laboratory studies it has been shown that even small PAC doses below 0.5% of the amended sediments' dry weight (dw) can lead to a measurable reduction in the bioaccumulation of organic contaminants [55, 78]. Increasing the dose is initially followed by a fast increase in remediation efficiency, as reviewed, for example, by Janssen and Beckingham [35]. The maximum measured remediation efficiency reported in the literature varies and seems to depend on the utilized test setups. The highest efficiencies with the least variation are measured when contaminant bioavailability is determined with passive sampling methods (Fig. 1). When actual bioaccumulation is measured, the observed variation in the results depends largely on the test species used. In most laboratory studies, the achieved reduction of freely dissolved contaminant concentrations and



**Fig. 1** Published remediation efficiencies vs. applied activated carbon doses (powdered activated carbon particle sizes  $<300\ \mu\text{m}$ ; mixed into the sediment; doses as sediment dry weight %). The datasets were grouped by sampling methods. Studies using organisms measure the actual bioaccumulation from the amended sediment into test organisms, while passive sampling approaches measure freely dissolved porewater concentrations of a contaminant. Closed symbols show results from laboratory (lab) studies, while field results are shown as open symbols. Due to the short span of applied AC doses in field trials, no dose-response curve was fitted.  $R^2$ -values for the remediation efficiencies obtained from laboratory studies are 0.46 (organisms) and 0.84 (passive sampling). Data from [37, 40, 52, 53, 55, 57–61, 73–77]

bioaccumulation in organisms reaches up to 95%. The AC doses needed to reach these drastic reductions in contaminant bioavailability often lie below 2.5%. Only few laboratory studies have found significant benefits from further increasing the AC concentrations within the contaminated sediment. Under field conditions, however, higher doses are often required to reach similar remediation efficiencies. In part, this is due to the risk of loss of applied AC under environmental conditions. In such cases, the effective AC dose can be significantly lower than the applied target dose [54, 68]. This is especially critical for field applications, as the time required for AC amendment to develop its full remediation potential can be much longer (up to several years) than in a confined laboratory microcosm [40, 41, 65]. Nonetheless, if a sufficient amount of sorbent is retained over longer time periods, AC amendments have proven to yield similar effectiveness in field trials as shown in laboratory studies [70].

One of the fundamental principles of the remediation method is that the reduction of contaminant bioavailability and bioaccumulation in benthic organisms will lead to reduced HOC transport in the aquatic food chain [9, 70]. Based on laboratory studies, it has even been suggested that AC treatment could reduce HOC transport from aquatic to terrestrial food chains if the concentrations in benthic invertebrates going through metamorphosis (developing from sediment dwelling larvae stage to flying adults) decrease as a consequence of reduced HOC bioavailability [58]. However, experimental field-scale evidence to support the reduction of HOC food chain transfer is scarce. Kupryianchyk et al. [74] demonstrated reduction in bioaccumulation of PCBs in a food chain (macrophytes, zooplankton, macroinvertebrates, and fish) with 4% sediment dw PAC treatment, and an order of magnitude decrease in concentration was observed in fish. However, the

experiment was executed in artificial 15-m long ditches, with benthic invertebrate communities and fish established at the beginning of the experiment. The influence of the interrelated surrounding areas is a much more important factor in the field than in (semi-) closed experimental systems. Thus, the effects of AC amendment on higher trophic levels may be less apparent in the field or require longer time to be established.

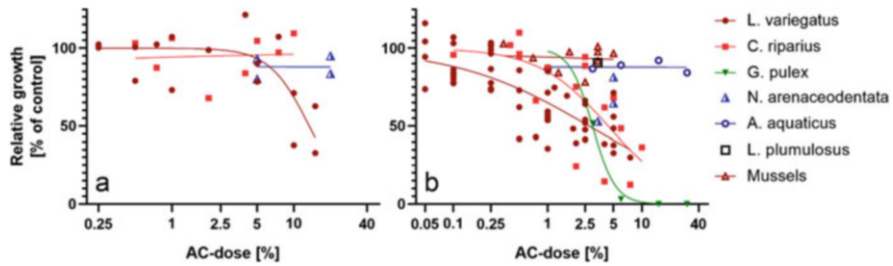
### 3.4 Secondary Ecological Effects

Organisms that exhibit severe adverse reactions to an untreated contaminated sediment will generally only see benefit from AC amendments. This can be the case for sites with either highly sensitive benthic fauna or high contaminant levels. Although there may be hidden adverse effects caused by the AC itself, the benefit of a reduced contaminant toxicity outweighs the risk in these cases. Tomaszewski et al. [75], for example, found a notably increased survival rate of the mussel *Mytilus edulis* after contaminated sediment was amended with AC. Kupryianchuk et al. [73] found that highly toxic sediment (PAH content >1,100 mg/kg) became habitable again for the benthic macroinvertebrates *Gammarus pulex* and *Asellus aquaticus*; there was no survival of the test organisms in the untreated sediment.

Nevertheless, a large share of sites that are remediated with AC does not show these extremely high levels of contamination. For such sites, there are indications that AC treatments may cause direct adverse effects. Undesired, negative side effects of the AC amendments are most strongly seen in deposit-feeding benthic species.

Analogous to the remediation efficiency, the particle size and dose of AC are the major factors determining the magnitude of adverse effects [35]. Finer PAC particles can cause severe effects to organisms, while the adverse effects of the coarse GAC particles only manifest at unrealistically high doses or not at all (Fig. 2a). Sediment-dwelling benthic worm *Lumbriculus variegatus* has been observed to avoid ingesting PAC-amended sediment [57, 78], and AC exposure which induced internal damages on *L. variegatus* and *Chironomus riparius* gut wall microvilli layer has been reported [57, 79]. Co-sequestration of nutrients by the AC is another suspected cause of the observed adverse effects on benthic organisms, although it is not considered to be the main contributor [80]. Despite several studies reporting adverse effects of PAC amendments, the mechanism causing the negative effects is not fully understood.

The sensitivity of benthic organisms to PAC exposure varies considerably between different species. Figure 2b shows an overview of 15 studies that measured biological effects of PAC amendments on 9 species. Clear adverse effects were reported in all available studies for *L. variegatus*, *C. riparius*, and *G. pulex*. Millward et al. [59] found *Neanthes arenaceodentata* to be similarly sensitive. However, the published results for this organism are not as consistent and might be influenced by experimental factors such as external feeding of the test organism during laboratory bioassays [80]. On the other hand, organisms such as *A. aquaticus* and *Leptocheirus*

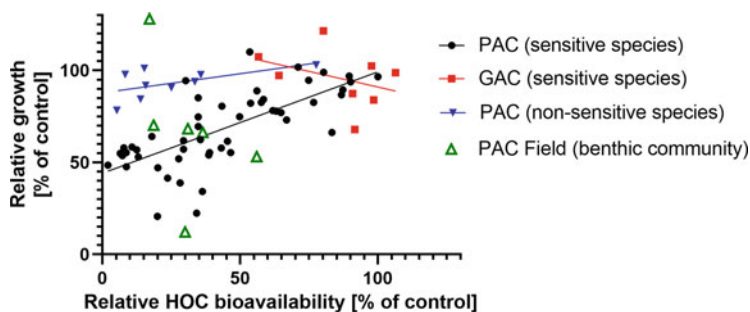


**Fig 2** Adverse effects of (a) granular activated carbon (GAC; particle size  $>300\ \mu\text{m}$ ) and (b) powdered activated carbon (PAC;  $<300\ \mu\text{m}$ ) in relation to the applied doses (mixed into the sediment, based on sediment dry weight %). The graphs show that (1) GAC produces significantly lower adverse effects in comparable doses and (2) adverse effects are dependent on species. While some species show strong adverse reactions to already small PAC doses (closed symbols), others are significantly less sensitive (open symbols). The fitted curves show a clear dose-response reaction to PAC (b) for the sensitive species *Lumbriculus variegatus* ( $R^2 = 0.60$ ), *Chironomus riparius* ( $R^2 = 0.56$ ), and *Gammarus pulex* ( $R^2 = 0.98$ ). Adverse reaction to GAC was observed with *L. variegatus* only ( $R^2 = 0.56$ ), with dosages much higher than generally applied for sediment remediation. No significant increase of adverse effects with increasing AC dose was found for *Leptocheirus plumulosus*, *Neanthes arenaceodentata*, *Asellus aquaticus*, and any of the examined mussel species. Data from [52, 53, 55–61, 73, 75, 78–81]

*plumulosus* and different mussel species show limited or no adverse effects, even at high doses of PAC (Fig. 2).

For AC amendments on a field scale, observed adverse effects depend, to a large extent, on the composition of the benthic fauna at the treated site. Given that the benthic fauna varies widely between different aquatic ecosystems and even within a single lake [54], the observed effects of AC amendments can vary accordingly. The results reported for field studies range from significant disturbances following the application of AC [82] to no measurable adverse effects [39]. The magnitude of the adverse effect can further depend on the general habitat or sediment quality prior to the AC application, where organisms that are already living on the margin of their habitable conditions are more negatively affected [78, 80].

The strong dependency of both adverse effects and remediation potential on the AC particle size can complicate the material selection. This is especially the case when benthic communities at a potential treatment site are dominated by sensitive species. When minimizing the risk for adverse effects at such sites is prioritized and coarser GACs are used, the success of the remediation might be compromised due to the low contaminant binding potential of these materials. The direct comparison of remediation potential and adverse effects of different ACs illustrates this trade-off (Fig. 3).



**Fig. 3** AC-induced adverse effects correlate strongly with the beneficial effect of reduced contaminant bioavailability for sensitive species (*Lumbriculus variegatus*, *Gammarus pulex*, *Chironomus riparius*). The reduced bioavailability of hydrophobic organic contaminants with powdered activated carbon (PAC; particle size  $<300\ \mu\text{m}$ ) treatment is high, with also strongest secondary effects to sensitive organisms ( $R^2 = 0.51$ ,  $p < 0.01$ ). The opposite holds true for granular activated carbon (GAC;  $>300\ \mu\text{m}$ ), to which even sensitive species rarely show adverse effects, but also the remediation efficiency is low (insignificant correlation). Significant correlation of adverse effects and remediation potential is not found for nonsensitive species. Field studies focus on benthic communities, which can be dominated by either sensitive or resistant species. Results vary and can be positioned in either groups on the graph. All data points represent the adverse effects and remediation efficiency measured for a single AC treatment. The data for the graph was collected from studies that report both values [37, 40, 41, 52–63]

## 4 Concluding Remarks and Future Prospects

Considering the extent of anthropogenic chemical pollution and the complexity and diversity of contaminated sites, AC amendment methods are a valuable supplement to traditional sediment remediation approaches. Results from both laboratory and field studies have laid a foundation for AC amendment-based sediment remediation. AC amendments have been proven to efficiently reduce freely dissolved concentrations, bioaccumulation, and fluxes of HOCs, and it is currently the most widely used in situ stabilization amendment worldwide. In addition, the carbon produced under controlled temperature conditions and absence of oxygen (slow-pyrolysis biochars) have shown to be extremely persistent and have higher carbon sequestration potentials compared to the naturally produced pyrogenic carbon (wildfire charcoal) [83]. However, to choose the optimal site-specific remediation approach, balanced consideration of the potential benefits, ecological effects, and costs is required.

Several field-scale studies have confirmed the remediation potential of AC amendments, and in long-term monitoring, AC has proven to be a stable sorbent, retaining its ability to reduce bioavailable HOC concentrations over several years. In addition, due to ongoing mass transfer, the effectiveness of AC application can even increase over time, attaining its full treatment capacity years after application. Despite the many examples of successful remediation with AC amendments, site-specific factors such as high organic matter content or dynamic sedimentation or sediment transport processes can lower the remediation efficiency.

The direct (i.e., secondary ecological) effects of AC amendments on benthic organisms have been extensively studied. The results have varied from no observed effects to significant effects on abundance, biomass, and number of species. In general, the observed effects on benthic organisms have been less severe in the field compared to laboratory experiments, and often (but not in all cases) adverse effects of AC amendment in the field have diminished within 1 to 2 years after the amendment. Nevertheless, the effects on benthic fauna should not be overlooked when planning remediation project. When multiple remediation approaches are viable at a given site, their potential adverse effects should be evaluated and weighed against each other. Adverse effects to benthic fauna can endanger the stability of the AC amendment, because if the benthic fauna disappear, then the incorporation of the AC into the sediment will slow. Depending on the areal extent of the AC treatment, the areas surrounding the AC placement can serve as reservoirs for recolonization. AC treatments are often limited to a small area within a water body [9], thus limiting the adverse effects to a relatively low fraction of the total ecosystem fauna. The beneficial effects, however, could extend throughout the food chain and over time lower the contaminant burden in the whole ecosystem.

Balancing the AC dose and particle size of the amendments is a key factor in seeking to reduce negative effects to benthic fauna. Where higher dose and smaller particle size may provide more efficient or quicker sequestration of HOCs, they are also more likely to induce greater stress to organisms. New innovations can provide future solutions to reduce the adverse effects of the sorbent material by altering the AC properties so that it is less disruptive to benthic organisms [56]. Developing AC remediation products further could allow this remediation method to move from a purely passive, bioavailability-reducing role towards active contaminant removal. Magnetized AC, which can be retrieved from the sediment [60, 61], allows for the extraction of contaminants. AC can function as a substrate for microbial growth, and biofilms formed on the particle surfaces can enhance degradation of chemicals [84]. AC has been shown to stimulate the diversity of PAH-degrading microbes, and under anaerobic conditions, AC amendment was shown to significantly induce naphthalene degradation [85]. The novel approach of inoculating AC particles with contaminant-degrading bacteria combines enhanced biodegradation with sorption [86, 87]. The advantage of such bioamended AC is that, in addition to reduction of the bioavailable fraction, it can also reduce the total concentrations of the contaminants in the sediment. Development of AC amendments in this direction could promote the wider adoption of the method, as in many countries, the regulations on the reuse and management of contaminated sediment are based on total concentration [9, 36]. Although bioavailability is being increasingly incorporated into risk assessment [88], legislation in many cases still lags behind. In addition, the acceptance of the remediation approach among the general public could improve if the method also provides contaminant removal.

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