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REVIEW ARTICLE

In situ remediation of contaminated marine sediment: an overview

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Abstract Sediment tends to accumulate inorganic and persistent hydrophobic organic contaminants representing one of the main sinks and sources of pollution. Generally, contaminated sediment poses medium- and long-term risks to humans and ecosystem health; dredging activities or natural resuspension phenomena (i.e., strongly adverse weather conditions) can remobilize pollution releasing it into the water column. Thus, ex situ traditional remediation activities (i.e., dredging) can be hazardous compared to in situ techniques that try to keep to a minimum sediment mobilization, unless dredging is compulsory to reach a desired bathymetric level. We reviewed in situ physico-chemical (i.e., active mixing and thin capping, solidification/stabilization, chemical oxidation, dechlorination, electrokinetic separation, and sediment flushing) and bio-assisted treatments, including hybrid solutions (i.e., nanocomposite reactive capping, bioreactive capping, microbial electrochemical technologies). We found that significant gaps still remain into the knowledge about the application of in situ contaminated sediment remediation techniques from the technical and the practical viewpoint. Only activated carbon-based technologies are well developed and currently applied with several available case studies. The environmental implication of in situ remediation technologies was only shortly investigated on a long-term basis after its application, so it is not clear how they can really perform.

Keywords Marine environment · Sediment · In situ technologies · Remediation · Toxicity · Activated carbon

Introduction

Marine sediment can accumulate persistent hydrophobic organic contaminants (HOCs) such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), dichloro-diphenyl-trichloroethane (DDT), and heavy metals (Nikolaou et al. 2009a, 2009b; Lofrano et al. 2016). Sediment-bound pollutants pose major concerns for human health and the environment, showing combined effects that are still largely unknown (USEPA, US Environmental Protection Agency 2005; Libralato et al. 2009, 2010a, 2010b; Mamindy-Pajany et al. 2010; Hurel et al. 2016). As a result, remediation of contaminated sediments has raised a great deal of scientific and public concern around the world representing a huge actual challenge both under a technical and technological viewpoint.

Sediment remediation techniques are commonly classified as in situ (i.e., treatments operating where the contamination is present with no sediment dredging) and ex situ (i.e., treatments including sediment dredging or resuspension phenomena to some extent). Nevertheless, dredging still remains an important issue; like for hotspots, dredging activities can heavily remobilize sediment like as the associated pollution via washing out events (Arizzi Novelli et al. 2006; Libralato et al. 2008; Krull et al. 2014; Chakraborty et al. 2014).

From 26 dredging projects carried out by the National Research Council (NRC, National Research Council 2007),

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systematic difficulties were observed in achieving target cleanup thresholds in addition to the impairment of sediment-associated benthic ecosystem. During the remediation of sediment in large-scale contaminated sites like Hunters Point Naval Shipyard (San Francisco, CA, USA), ex situ techniques were ineffective: economic, environmental, and technical goals were not met (Zimmerman et al. 2004).

The development of cost-effective sediment management strategy requires a multi-approach assessment including in situ treatment alternatives, unless dredging is compulsory to reach a desired bathymetric level. Since they allow sediment remediation avoiding excavation and transport, remediation footprint and cost savings could be significantly optimized. The main disadvantages are related to long-lasting procedures (months or years), uncertainty about the treatment uniformity due to the variability of sediment and aquifer characteristics, and the overall efficiency of the process is more difficult to verify.

This paper proposed an overview of existing in situ sediment remediation treatments: (i) providing a synthetic overview of the physico-chemical performance of contaminants' reduction, (ii) refining papers on the basis of the description of detailed experimental design and the application of standardized methods, and (iii) supporting a "mind-the-gap" approach stressing on missing data to support hazard assessment. A special focus was devoted to three main technological clusters identified for in situ remediation: (i) physical and chemical treatments, (ii) biological treatments, and (iii) hybrid solutions. Results were discussed considering the characteristics of contaminants and their removal efficiency, how long the remediation might last, the role of natural organic matter (NOM) and of potential by-products, and their ecotoxicological implications, including remediation design and general costs as well.

Techniques and technologies for in situ sediment remediation

Apart from the no-action in situ approach, like monitored matural attenuation/recovery (MNA/R) (USEPA, US Environmental Protection Agency 2014), only direct or active interventions might produce a significant reduction in sediment contamination level in a reasonable time (e.g., from months to few years).

Physical and chemical treatments

In situ amendment: active mixing and thin capping

Two main approaches could be used to remediate contaminated sediment: (i) active mixing and (ii) thin capping. Active mixing consists of mixing contaminated sediments with natural substrates or other inert materials. In both cases, the bioactive surface layer of sediment is able to transfer contaminants from sediment to strongly binding sorbent particles, reducing their bioavailability to benthic organisms and contaminant flux into the water column and thus the potential general accumulation in the aquatic food web (Ghosh et al. 2011).

Thin capping consists of one or more layers of amendment (e.g., sand and NOMs) actively reducing the overall cap thickness required, for example when compared to conventional sand cap (Wessels Perelo 2010).

In the last two decades, several authors evaluated in situ amendment introducing various sorbents such as activated carbon (AC), organoclay, apatite, biochar, coke, zeolites, and zerovalent iron (ZVI) into contaminated sediments (USEPA, US Environmental Protection Agency 2013a). Amendments tend to modify sediment geochemistry increasing contaminant binding and stability in order to reduce its risk to human health and the environment. Among all, AC, organoclay, and apatite were identified as particularly promising sorptive amendments for in situ sediment remediation (USEPA, US Environmental Protection Agency 2013b). But several data about their potential side effects are still missing. Except for AC and ZVI, (eco-)toxicity data are scarce or still unavailable.

As shown in Tables 1 and 2, most studies are referred to AC administration. Several laboratory experiments and recent field studies demonstrated that AC showed significant reductions in chemical concentration and biological availability of polychlorinated biphenyls (PCBs) (Zimmerman et al. 2004; Werner et al. 2005; Cho et al. 2009; Beckingham and Ghosh 2011; Cho et al. 2012), polyaromatic hydrocarbons (PAHs) (Hale et al. 2010; Cornelissen et al. 2011; Hale et al. 2012; Meynet et al. 2012), and dichlorodiphenyltrichloroethane (DDT) (Tomaszewski et al. 2007) by active mixing and thin capping.

Considering a series of differentially polluted sediment samples, Hale et al. (2010) and Hale and Werner (2010) highlighted that 1–5% AC can reduce the pore water concentration of PCBs, PAHs, DDT, dioxins, and furans from 70 up to 99%. Organoclay effectively removed soluble organics and non-aqueous phase liquids (NAPLs) such as oils, chlorinated solvents, and PAHs (Alther 2002a, 2002b). Apatite facilitated the immobilization of metals including Cu, Pb, and Zn (Knox et al. 2008). Laboratory results demonstrated that the effectiveness of sorbents in lowering contaminant bioavailability increased with decreasing amendment particle size, growing dose, greater mixing, and contact time (Zimmerman et al. 2005; Ghosh et al. 2011), but it could vary for various amendments with similar surface areas (Tomaszewski et al. 2007).

As shown in Table 3, some patented commercial products are available yet and some of them were applied at full-scale remediation projects like for capping (Table 2), but every technological approach must be considered on a case-by-

Table 1 Summary of active mixing findings with amendments

Amendment	Sampling site	Experimental conditions	Scale	Target contaminants	Main findings	Reference
AC	South Basin at Hunters Point Naval Shipyard (San Francisco, USA)	Contaminated sediment was contacted with 3.4% (dry weight) AC on a roller for 1 month	Laboratory scale	PCB	87% reductions in aqueous equilibrium PCB concentrations after 1 month and over 90% after 6 m	Zimmerman et al. (2004)
AC	South Basin at Hunters Point Naval Shipyard (San Francisco, USA)	Contaminated sediment was contacted with 3.4% (dry weight) AC on a roller for 1 month	Laboratory scale	PCB,	After 28 day, sediment exposure tests that PCB bioaccumulation in polychaetes (*Neanthes arenaceodentata*) is reduced by 82% following 1-m contact of sediment with AC and by 87% following 6-month contact of sediment with AC. PCB bioaccumulation in an amphipod (*Leptocheirus plumulosus*) is reduced by 70 and 75% following 1- and 6-month contact of sediment with activated carbon, respectively. Reductions in aqueous PCB concentrations in equilibrium with the sediment were similar to reductions in PCB bioaccumulation	Millward et al. (2005)
AC	South Basin at Hunters Point Naval Shipyard (San Francisco, USA)	Contaminated sediment was contacted with 3.4% (dry weight) AC on a roller for 1 month	Laboratory scale	PCB	84% reductions in the bioaccumulation of PCBs by the clam *Macoma balthica*	McLeod et al. (2007)
TOG®-NDS	Hunters Point Shipyard (San Francisco, USA)	A large-scale device to mix 500 kg of AC into a 34.4-m^2 plot to a depth of 1 ft	Pilot scale	PCB at 2 mg/kg	34% less PCB uptake into semi permeable membrane devices and 24% less PCB bioaccumulation in *M. nasuta* after 1 month Seven months after the AC treatment occurred, the differences further increased up to 62% less in SPMD uptake and 53% less in clam bioaccumulation, which implies the possibility of the long-term effectiveness of AC	Cho et al. (2007)
■ TOG® ■ Filtrasorb® 400 (F400) ■ Aquacarb® 830 (AC830) ■ Aquacarb RS (ACRS)	Lauritzen Channel Hunters Point Shipyard (San Francisco, USA)	Large glass bottles were filled with surface sediment followed by the addition of 0.8, 1.6, and 3.2 wt% AC on a dry mass basis. The bottles were rolled at 2–3 rpm for 31 days, and then, sediment was used for SPMD and aqueous equilibrium	Laboratory scale	DDT (up to 252 mg/kg)	Amendment of field-collected residual sediments with AC resulted in large reductions (>80%) in aqueous equilibrium concentration and SPMD uptake reduced from 91 to 99% during 1 to 26 months treatment with 3.2 wt% re-AC (Aquacarb RS, ACRS)	Tomaszewski et al. (2007)

Table 1 (continued)

Amendment	Sampling site	Experimental conditions	Scale	Target contaminants	Main findings	Reference
		tests.				
AC	Hunters Point Shipyard (San Francisco, USA)	The study design comprised four test plots having a surface area of 34.4 m^2 each. Two field equipment was compared: a barge-mounted rotovator system for direct mixing of AC into sediment and a crawler-mounted AC-slurry injector system.	Pilot scale	PCB	Successful incorporation of AC into sediment to a nominal 30-cm depth. AC treatment did not adversely impact sediment resuspension or PCB release into the water column as well as the composition of the macrobenthic community richness and diversity. About 50% reduction in PCB uptake in AC-treated sediment and in pore water even after 13 months post-treatment.	Cho et al. (2009)
AC	Hunters Point Shipyard (San Francisco, USA)	Non-amended reference sediment, reference sediment with fresh AC, and sediment containing field-aged AC were investigated. The clean AC additions were at levels of 0, 2, 4, and 8% (dw) in duplicate. Wet sediment (3.2 g dw) was combined with Grasse River water (30 mL), NaN_3 (1 mg) and spiking solution (20 μL acetone, containing 2000 ng of PCB-14, PCB-65, and PCB-166).	Laboratory scale	HOCs, PCBs	Reference sediment with 2% fresh GAC reduced HOC concentration in pore water by 87–98% compared to non-amended sediment at low spiked PCB concentrations. When increasing the spiking concentration, the decrease in CPW dropped to 52% for PCB-103, whereas the percentage was almost the same for the two smaller PCB congeners with 98% (tri PCB-29) and 81% (tetra PCB-69).	Oen et al. (2012)
AC	Hunters Point Shipyard (San Francisco, USA)	Sediment column studies were conducted using the two study sediments with variations in initial AC mixing regimes, AC distribution, AC particle size, pore water movement, and AC-sediment contact time.	Laboratory scale	PCBs	After 2 years of stagnant contact, the contaminant uptake in polyethylene passive samplers embedded in the columns was reduced by 95–99% for PAHs and 93–97% for PCBs with 5 and 4 wt% AC dose, respectively, when AC was initially applied by mechanical mixing.	Choi et al. (2014)

AC activated carbon

Table 2 Summary of thin-layer capping findings

Amendment	Sampling site	Experimental setup	Scale	Target contaminants	Main findings	Reference
crushed limestone and gneiss	Bjørvika (Oslo harbor, Norway)	Laboratory microcosm test	Laboratory scale	PAHs and PCBs	The flux from sediments capped with 1-cm mineral cap (crushed limestone or crushed gneiss (0–2 mm)), observed during the first 410 day, was 3.5–7.3% of the flux from uncapped sediments.	Eek et al. (2008)
AC	Trondheim harbor (Norway)	Five experimental field sites were established: (1) reference site, no capping; (2) site capped with AC only; (3) site capped with AC mixed with bentonite clay (AC + clay); (4) site capped with AC and covered by 5 mm of sand (AC + sand); and (5) capped with sand only (5 mm) to discern the effect of AC from that of sand. After 12 months from capping, sediment cores were collected and sliced in 1-cm sediment sections down to 5-cm depth and then in 2-cm sections between 5- and 11-cm depth.	Pilot scale	PAHs	Compared to reference field, AC capping reduced fluxes by a factor of 2–10. Pore water PAH concentration profiles were measured in situ using a new passive sampler technique and yielded a reduction factor of 2–3 compared to reference field. Benthic macrofauna composition and biodiversity were affected by AC amendments, AC + clay having a lower impact on benthic taxa than AC-only, or AC + sand. In addition, AC + clay gave the highest AC recoveries (60 vs 30% for AC only and AC + sand) and strongest reductions in sediment-to-water PAH fluxes and pore water concentrations.	Cornelissen et al. (2011)
AC	Grenlandsfjords (Norway)	One very large field was established in the Eidangerfjord (size 200 × 200 m, 40,000 m^2; clay + AC) at 100-m water depth. Target dimensions for cap thickness were 5 cm (limestone and clay only) and 2.5 cm (clay + AC), with an AC amendment of 2 kg/m^2 on the clay + AC fields.	Large scale field experiment	Polychlorinated dibenzodioxin and dibenzofuran (PCDD/F)	In situ AC profiles revealed that the AC was mixed into the sediment to 3- to 5-cm depth in 20 months. Only around 25% of the AC was found inside the pilot fields. Sediment-to-water PCDD/F fluxes measured by in situ diffusion chambers were significantly lower at the capped fields than at reference fields in the same fjord, reductions being largest for the limestone (50–90%) followed by clay (50–70%) and the AC + clay (60%). Also, reductions in overlying aqueous PCDD/F concentrations measured by passive samplers were significant in most cases (20–40% reduction), probably because of the large size of the trial fields. The AC was less effective in the field than in the laboratory, probably due to prolonged sediment-to-AC mass transfer times for PCDD/Fs and field factors such as integrity of the cap, new deposition of contaminated sediment particles, and bioturbation.	Cornelissen et al. (2012)
AC	Wyckoff/Eagle Harbor (WA, USA)	Laboratory column experiments were performed using contaminated sediments and capping materials (sand + 2% GAC, sand + peat and sand only).	Laboratory scale	PAHs	The most dramatic result was observed for caps amended with AC at a dose of 2% by dry weight. PAH concentrations in the pore water of the AC-amended caps were 3–4 orders of magnitude lower (0.04 ± 0.02 μg/L for pyrene) than concentrations in the pore water of the source sediments (26.2 ± 5.6 μg/L for pyrene) even after several hundred pore volumes of flow.	Gidley et al. (2012)

Table 2 (continued)

Amendment	Sampling site	Experimental setup	Scale	Target contaminants	Main findings	Reference
AC and kraft lignin	Frierfjord (Norway)	The influence of cap thickness (0.5–5 cm) and different cap materials (passive material: coarse or fine limestone or a marine clay; active material: AC or kraft lignin) was tested using a three-factor experimental design (boxcosm).	Laboratory scale	PCDD/Fs, HCB, OCS	AC was more efficient than lignin, and a ~90% reduction of fluxes and bioaccumulation was achieved with 3-cm caps with 3.3% AC	Josefsson et al. (2012)
AC (SediMite)	Custom Plywood (Anacortes, WA, USA)	Six plots (5.5 × 5.5 m) were established to determine whether the contaminated sediments can be remediated with thin layers (less than 0.6 cm) of sand amended with activated carbon, without adverse impacts to existing marine habitat.	Pilot and full scale	Dioxins/ Furans	Evaluate AC/cap effects in sensitive seelgrass environments	WDOE (Washington State Department of Ecology) (2012)
AC (AquaGate)	Puget Sound Naval Shipyard, (Bremerton, WA, USA)	The contaminated area (58 × 35 m) was amended with powdered activated carbon (PAC) using the AquaGate™ composite aggregate system to improve delivery and physical stability and decrease the bioavailability of contaminants.	Full scale	PCBs, Hg	The project showed and validated the placement, stability and performance of AC amendments for treating contaminated sediments in an area with elevated PCB and Hg contamination.	Johnston et al. (2013)
AC (BioBlok)	Kirkebukten Bergen Harbor (Norway)	The aim of this paper is to present the design, manufacture, placement, and available performance monitoring results for two types of caps tested in situ (both of which incorporated BioBlok capping products amended with PAC).	Pilot scale	PCBs, BT	It appears that the capping has been effective with respect to reducing leaching of PCB from the sediments. A ~90% reduction of fluxes was achieved.	Hjartland et al. (2013)
AC (BioBlok)	Sandefjord Harbor (Norway)	Pilot test has been used to showcase the effectiveness and costs for implementing of a thin-layer capping using conventional materials as well as innovative materials and engineered products.	Pilot scale	PCBs, BT, PAHs	The study was focused on key elements related to controlled material placement as well as at evaluating short- and long-term remedy effectiveness (reductions in contaminant bioavailability, benthic recolonization, erosion protection, etc.).	Lundh et al. (2013)
AC	Trondheim harbor (Norway)	Three types of thin-layer caps with AC were tested in situ in experimental plots (10 × 10 m), using AC + clay, AC only, or AC +sand. One year after capping, intact sediment cores were collected from the amended plots for ex situ surveys of the capping efficiency in reducing the PAH and PCB aqueous concentrations and bioaccumulation.	Pilot scale	PAHs and PCBs	AC + clay reduced bioaccumulation of PAH and PCB congeners between 40 and 87% in the worms and between 67 and 97% in the clams. Sediment capped with AC only also led to reduced bioaccumulation of PCBs, while AC + sand showed no reduction in bioaccumulation. Thus, the best thin-layer capping method in this study was AC mixed with clay	Samuelsson et al. (2015)

AC activated carbon

case basis. In fact, organoclay® MRM (Table 3) can enhance the production of methyl mercury in presence of sulfate-reducing bacteria. Short-, medium-, and long-term monitoring surveys should be carried out after remediation activities to verify the amendment stability in sediment within real exposure scenarios.

Considering the *Hunters Point Shipyard* case study (Table 1), Choi et al. (2016) focused on the importance of developing and applying decision-making frameworks for in situ sediment AC remediation, including a modeling approach supporting long-term prediction and engineering design. The modeling framework compared various design alternatives for treatment optimization and estimation of long-term effectiveness over 10–20 years under slow mass transfer condition in order to identify the best efficient and cost-effective solution for HOC-contaminated sediment treatment.

Solidification/stabilization

In situ solidification/stabilization (S/S) treatment involves the addition of chemicals and/or cements to encapsulate contaminated sediment and/or convert pollutants into less soluble, less mobile, or less toxic forms (Scanferla et al. 2009; Wang et al. 2015). Unless *Portland* cement and quicklime are the most commonly used materials for S/S, recently, new additives are available from the market (Table 3). The mixture of reagents and additives used for S/S is commonly referred as the binder and can range from a single compound to a multi-component system.

The S/S process proved to be efficient for treating sediments contaminated with heavy metals, PAHs, and PCBs (EPA 2009). S/S is most often selected for metals including lead, arsenic, and chromium because these contaminants form insoluble compounds when combined with appropriate additives. In applying S/S for treating organics, the use of some kind of organophilic clay and AC, either as pre-treatment or as additives in cement, can improve contaminant immobilization. Generally, the depth of contaminated sediment can limit its application. A series of bench tests should be performed stabilizing the best mix design capable of reducing the leaching of contaminants from the solidified mass. Mixing conditions, pH, water/binder ratio, and curing temperature were identified as the principal factors influencing solidified sediment strength and leaching behavior (Malviya and Chaudhary 2006). Since mixing and temperature are difficult to control in situ, the process can be less effective than other in situ treatments. Few examples of full-scale in situ S/S of contaminated sediments were reported (Robb et al. 2015). Small-scale immobilization has been used at *Manitowoc harbor* in Wisconsin (USA), where cement and fly ash slurry were added to sediment using a proprietary mixing tool and slurry injector (EPA 1994). The in situ mixing of cement with sediment for enhancing primarily compressive strength has not been proved or accepted for treatment of contaminated marine sediments in USA (EPA 1993). The use of in situ S/S process should be carefully evaluated since the treated site characteristics could be significantly modified. Some processes might result in substantial volume increase up to two times the original one. No data are available about the ability of the process to keep contaminants immobilized over time in real environmental conditions. A generalized lack of aging effects of the technology was evidenced.

Chemical oxidation

In situ chemical oxidation involves the introduction of chemical oxidant agents into the subsurface in order to transform sediment contaminants into less harmful chemical species. Contaminants amenable to treatment by chemical oxidation include benzene, toluene, ethylbenzene, and xylenes (BTEX); methyl tert-butyl ether (MTBE); total petroleum hydrocarbons (TPH); chlorinated solvents (ethenes and ethanes); PAHs; polychlorinated biphenyls (PCBs); chlorinated benzenes (CBs); phenols; organic pesticides (insecticides and herbicides); and munitions constituents (e.g., cyclotrimethylenetrinitramine, trinitrotoluene, octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine) (Flotron et al. 2005; Brosillon et al. 2015; Shih et al. 2016).

The most commonly employed chemical oxidants include hydrogen peroxide, ozone, sodium permanganate, and sodium persulfate (Shih et al. 2016). These oxidants have been able to cause rapid and complete chemical destruction of many toxic organic chemicals; other organics are amenable to partial degradation as an aid to subsequent bioremediation.

So far, most applications focused on ex situ treatments but presenting limited potentiality for in situ activities due to the difficulty related to direct addition of chemicals to sediment and the consequent environmental side effects.

Dechlorination

Nanoscale zero-valent iron (nZVI) can reduce some organic contaminants to less toxic by-products. Laboratory studies evidenced that also micrometer-scale ZVI effectively promoted sediment reductive dechlorination of PCBs, but including PCB congeners potentially more toxic than the parent compounds and increasing degradation time for larger congeners (Gardner 2004; USEPA, US Environmental Protection Agency 2013a). Additional limitations related to ZVI can include alterations to sediment geochemistry, passivation of iron by the formation of a thin layer of iron oxide, and the high cost of microscale and nanoscale iron. At now, the use of ZVI to treat contaminated sediment is limited to bench-scale studies with no pilot- or full-scale applications for in situ sediment remediation to the best of our knowledge.

Table 3 Summary of marketed sorbent products

Marketed products	Description	Target	Matrix	Technological application	Reference
Organoclay® MRM	A granular, permeable, high-capacity adsorptive material	NAPLs, dissolved low-solubility organics mercury and arsenic	WC, WSI, S	Can be applied: ■ In thick layers over sediments ■ As solidification/stabilization additive to Portland	1
Organoclay® PM-199	A granular, permeable, high-capacity adsorptive material	Oils, greases, NAPLs, high-molecular-weight/low-solubility organics	WC, WSI, S	Can be applied: ■ In thick layers over sediments ■ As solidification/stabilization additive	1
Organoclay® PM-200	A granular, permeable, high-capacity adsorptive material	NAPLs, dissolved PAHs and PCBs	S	Bulk active capping	1
Organoclay® SS-199	A semi granular adsorption media	Oils, greases, NAPLs, high-molecular-weight/low-solubility organics	S	Solidification/stabilization additive	1
Reactive Core Mat® (RCM)	An innovative product consisting of a variety of active materials encapsulated between carrier textiles, which are adhered together to provide product integrity. RCM is manufactured in rolls with typical panel size of 15 ft by 100 ft.		WC, WS	- Especially designed to provide a method of placing active materials into sub-aqueous sediment caps. - The quantity of active material encapsulated into the mat is specific with a maximum product thickness of approximately 1 cm.	1
	with apatite	Metals			
	with organoclay®	NAPLs and low-solubility organics			
	With granular activated carbon (GAC)	Organics			
	with (GAC) and sand	Organics			
Triton Marine Mattress	The mattress is constructed of an advanced geo-grid designed to be integrated with available fill and is used to simplify construction and provide protection in highly erosive conditions		S	The Triton Marine Mattress system is used in conjunction with reactive core mat.	2
Bentoblock®	A natural sodium bentonite screened to 3/8 in. (0.95 cm) to 3/4 in. (1.90 cm) in size		WC, WSI	Especially formulated for mixing with aggregate and deploying as a low-permeability sediment cap	1
Econo-Hp Reactive Core Mat™	A permeable composite of geotextiles and a non-swelling granular clay compound.	Oil and similar organic	WC, WSI	Especially designed for in situ subaqueous cap for contaminated sediments	1
AquaGate+™	Amendments in a thin coating around an inner core.		WC, WSI, S	The proprietary AquaGate product typically includes a dense aggregate core, along with clay-sized materials, polymers, and powdered AC additives. AquaGate serves as a delivery mechanism to reliably place reactive capping materials into aquatic environments.	3
	with organoclay	NAPLs, PAHs, and PCBs			
	With PAC	Organics			
	With ZVI				
	With Provect-IRM™	Metal			
	With sulfur compounds				

Table 3 (continued)

Marketed products	Description	Target	Matrix	Technological application	Reference
	With aluminum sulfate			Especially designed for in situ subaqueous cap for contaminated sediments.	
	With linoptilolite				
	With organic carbon	Organics			
	With sorbster™	Mercury and metal			
	With microbes	Organics			
	With iron oxide				
	With EHC-M™	Metals			
AquaBlok™				AquaBlok™ will form a low-permeability isolation cap over contaminated sediments in a layer thickness of 6-in. or less	3
SediMite™	An agglomerate composed of a treatment agent (typically AC), a weighting agent (to enable it to sink and resist resuspension), and an inert binder.	PCBs, Hg, CH_3-Hg, dioxins, furans, PAHs, DDT, and other hydrophobic chemicals.		Application directly over the sediment surface for eventual mixing into surficial sediments. as a layered active cap and as an amended sand cap.	4
Quik-Solid® 50	A custom blend of granular cross-linked polyacrylate superabsorbent polymer and ORGANOCLAY®.	Organics	S	Solidification/stabilization additive	1
TOG®-NDS	A virgin GAC	Organics	WC, WSI, S	Amendment not specifically designed for sediment remediation	5
Filtrasorb® 400 (F400)	A virgin GAC	Organics		Amendment not specifically designed for sediment remediation	5
Aquacarb® (ACRS)	Reactivated carbon	Organics		Amendment not specifically designed for sediment remediation	6
Aquacarb® 830 (AC830)	A virgin GAC	Organics		Amendment not specifically designed for sediment remediation	6

WC water column, *WSI* water sediment interface, *S* sediment; *1* www.cetco.com, *2* www.tensarcorp.com, *3* www.aquablok.com, *4* http://sedimite.com/, *5* www.calgoncarbon.com, *6* www.siemens.com

Electrokinetic separation

Electrokinetic separation is an emerging technology relying on the application of a low-voltage direct current through sediment to separate and extract heavy metals, radionuclides, and organic contaminants (SMOCS 2012; Hahladakis et al. 2013; Iannelli et al. 2015). The promotion of electro-migration involves (i) positively charged chemical species (e.g., metals, ammonium ions, and some organic compounds) moving toward the cathode and (ii) negatively charged chemicals (e.g., chloride, cyanide, fluoride, nitrate, and negatively charged organic species) migrating toward the anode. Enhancing agents (e.g., surfactants reducing the interfacial tension) and co-solvents could be used in order to increase the reduction performance (Wan et al. 2009). The addition of solubilizing agents can change the characteristics of sediment particles as well as of pore water, potentially affecting the electro-osmotic flow and, consequently, the removal process of contaminants. Cyclodextrins are frequently used being biodegradable and non-toxic increasing the solubility of contaminants like PAHs by lowering their sorption at the same time (Hahladakis et al. 2013). Inert electrodes, such as carbon or graphite, or platinum, must be used since metallic electrodes may dissolve as a result of electrolysis introducing further contamination into sediment (Hahladakis et al. 2013).

Sediment flushing

In situ sediment flushing treatment removes harmful chemicals by injecting water or chemicals into sediment washing it out and conveying hydrophilic contaminants toward the extraction wells. Elutriates are pumped and treated in *on-site* wastewater treatment plants (SMOCS 2012). Environmentally sustainable surfactants could be used to increase the solubility of organic compounds, the flushing solution could significantly alter the physico-chemical properties of sediment. This technology can offer the potentiality of recovering metals mobilizing a wide range of organic and inorganic contaminants especially from coarse-grained soils, but it could be highly non-effective in presence of heterogeneous mixtures of contaminants because no universal flushing solution is available: a case-by-case basis approach is strongly required. The flushing technique could be effective, but current in situ applications are mainly related to soil (SMOCS 2012).

Bioremediation

Bioremediation consists of biologically driven processes to remove and/or detoxify environmental pollutants. Frequently, the complexity of sediment-water ecosystem can limit the effectiveness of in situ bioremediation, which is generally more successful when environmental conditions are carefully controlled and adequately adjusted to enhance biotransformation processes. Under this viewpoint, ex situ treatments are easier to manage.

Several treatment approaches are included in contaminated sediment bioremediation (Wessels Perelo 2010) like (i) monitored natural attenuation/recovery (NA/NR) (i.e., the action is “no action” meaning that no other activities than environmental monitoring are required leaving natural remediation to occur), (ii) biostimulation (i.e., indigenous populations eliminating pollutants are stimulated removing the factors limiting their growth), (iii) bioaugmentation (i.e., introduction of allocthonous species for the degradation of contaminants, and (iv) phytoremediation (i.e., using macrophytes and/or algae to degrade and/or remove contaminants from sediment). Wessels Perelo (2010) reviewed in situ bioremediation of organic pollutants in sediment evidencing how it is promising (i.e., lower impact and more cost-efficient) than traditional management strategies, but with some major drawbacks. In situ bioremediation takes longer (from months to several years) and is less predictable than traditional methods being valuable only to treat low-risk sites with low level or diffuse contamination not immediately impairing human health and the environment (Magar and Wenning 2006). Akcil et al. (2015) stated that when considering how to reduce the risk of contaminated sediment, it is important to recognize the capacity of natural processes to achieve remediation objectives without human intervention.

NA processes could reduce pollutants under different environmental conditions (i.e., pH and nutrient levels) (Röling and van Verseveld 2002). Frequently, NA is driven by physical mechanisms such as mixing and in-place burial of contaminated sediment with progressively cleaner sediment delivered by the watershed. Natural sedimentation process can reduce contaminants’ (bio)availability of environmental concern (CEC) limiting their downstream transport as well.

Other potentially significant mechanisms include chemical processes such as adsorption and redox reactions coupled with biodegradation. According to Wang and Tam (2012), no significant decline in total PAHs, TBT, heavy metals, and contamination “hotspots” could be detected 1 year after the removal of an old floating dock in Hong Kong SAR (South China). However, the profile of 16 PAHs changed 6 months in the impacted stations after the dock removal showing a decrease of some low-molecular-weight PAHs suggesting the presence of on-going in situ biodegradation events.

In confined hydrocarbon-rich environments, anoxic conditions tend to prevail already few centimeters below the water-sediment and the increase of oxygen availability would support biodegradation processes. The use of oxygen-releasing compounds (ORC) based on Ca or Mg peroxide has been proposed to ensure long-lasting release of oxygen in contaminated subsurface environments (Bellagamba et al. 2016). But drawbacks are present such as (i) the poor control of the oxygen rate released and the resulting oxygen availability, (ii) the need for repeated injections due to oxygen consumption/scavenging by biotic and abiotic side reactions, and (iii) ORC could have secondary harmful effects on aquatic biota (Abdallah et al. 2009). Apart from several case studies (Yu et al. 2011; Akcil et al. 2015; Matturro et al. 2015), no great additional information is available after Wessels Perelo (2010) investigation of in situ sediment bioremediation.

Hybrid solutions

Nanocomposite reactive capping

In order to enhance current capping technologies, Choi et al. (2009) focused on the development of granular AC (GAC) impregnated with reactive iron/palladium (Fe/Pd) bi-metallic nanoparticles (NPs) (i.e., reactive AC (RAC)). RAC is a smart composite for dechlorination of PCBs. Due to its high adsorption capacity, RAC actively attracts hydrophobic PCBs from sediment matrix. The concept of a "reactive" cap/barrier composed of RAC pellets contained between thin geo-textile membranes is proposed in Fig. 1.

In 2 days, 5 g/L of RAC achieved 86% dechlorination of 2-chlorobiphenyl (2-ClBP, 4.08 mg/L), while almost complete dechlorination was achieved in just 1 day at 20 and 100 g/L of RAC. Its scale-up and field application will take some time due to the high cost of RAC mainly due to Pd doping and synthesis time. In general, GAC showed PCB adsorption capacity in presence of ZVI as a strong reducer, while Pd speeds up the dechlorination kinetic. Critical issues like Fe and Pd leaching, adsorption, and dechlorination capacity and yield of RAC, as well as their aging and oxidation, are still under investigation along with the initial demonstration of PCB-contaminated sediment remediation using RAC (Choi et al. 2009).

Bioreactive capping

The use of physico-chemical active caps is a promising option, but possible limitations (e.g., high material costs, sorption, and reaction capacities) lead to the consideration of in situ bio-reactive caps. Specifically, biotransformation of contaminants is designed to occur within the cap matrix producing environmentally friendly reaction by-products. Biologically based active caps have the potential to keep reactivity over long time periods serving as potentially sustainable remedial options especially when degrading microorganisms are present and the necessary metabolic requirements are met (Himmelheber et al. 2011). Previous studies about the activity of microbial populations within a sediment cap demonstrated that indigenous microorganisms in the underlying sediment, including organisms capable of contaminant

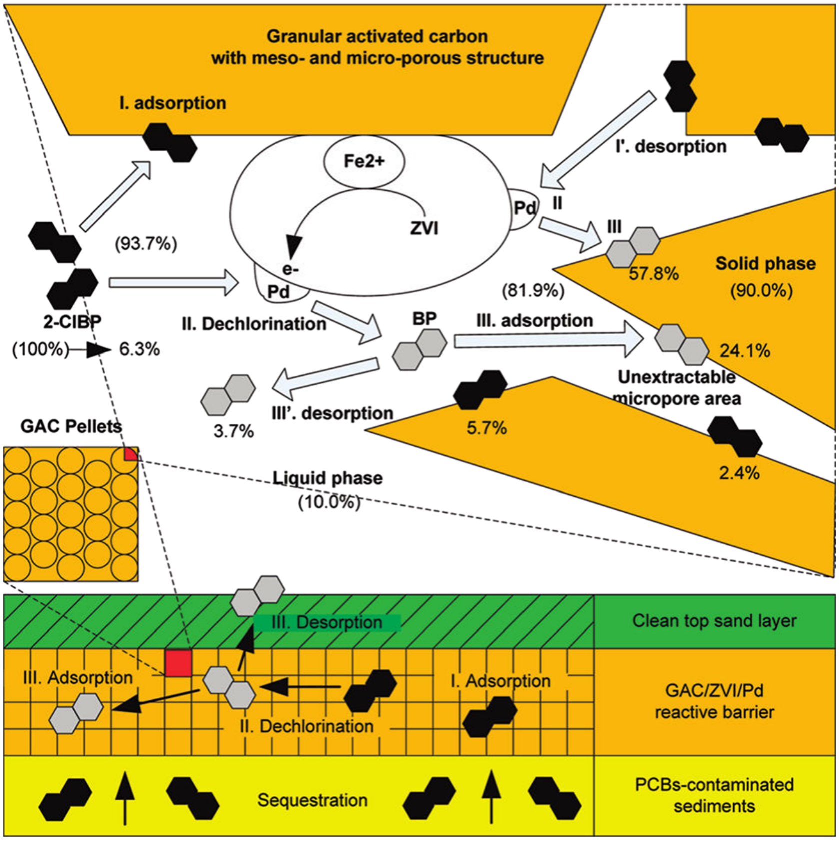

Fig. 1 Conceptual diagram of "reactive" capping barrier composed of GAC impregnated with Fe/Pd bimetallic particles (noted as RAC) for the adsorption and simultaneous dechlorination of PCBs in sediment (Choi et al. 2009)

biotransformation, could colonize the overlying cap and, possibly, participate to contaminant bioattenuation process (Himmelheber et al. 2009).

Some recent examples of bioreactive capping were related to ZVI (Sun et al. 2010) and AC (Wang et al. 2014). An integrated ZVI–sorbent–microorganism remediation system as an in situ active capping technique was proposed to remediate nitrobenzene-contaminated (NB-contaminated) sediment; NB was reduced by ZVI to aniline that is more biodegradable than the parental compound (Sun et al. 2010). A novel bio-reactive capping barrier composed of polysulfone/GAC (PS/GAC) hybrid membrane immobilized with microorganism was developed to remediate NB-polluted sediment (Wang et al. 2014).

Microbial electrochemical technologies

Microbial electrochemical technologies (METs) are increasingly considered for remediating contaminated sediment. They rely on the capability of microorganisms to use (directly or indirectly) solid electrodes; suitably deployed in the contaminated environment, they seemed to require low maintenance and being virtually an inexhaustible source of electron donors/acceptors for the reductive/oxidative degradation of contaminants (Aulenta et al. 2011; Li and Yu 2015; Wen-Wei and Han-Qing 2015; Majone et al. 2015; Bellagamba et al. 2016) suggesting the creation of benthic microbial fuel cells (BMFC) (Wen-Wei and Han-Qing 2015).

Apart from creating an efficient electron transport route for bridging the natural redox reactions, BMFC can be used to directly supply electron donors or acceptors to sediment through tuning the local electrode potential. Thus, in some cases, the cathode can also be buried into the sediment, while an external voltage can be applied to provide the desired electrode potential for contaminant removal (Aulenta et al. 2007; Chun et al. 2013).

Bellagamba et al. (2016) explored the possibility to electrochemically manipulate the redox potential of a crude oil-contaminated marine sediment establishing in situ conductive conditions to contaminants' biodegradation by autochthonous microbial communities. They showed that low-voltage electrolysis (2 V) applied continuously or intermittently accelerated (up to 3 times) the biodegradation of hydrocarbons from crude oil in marine sediment.

Discussion

Factors affecting technology selection should generally include (i) contaminant characteristics; (ii) removal of target contaminants (i.e., performance criteria); (iii) contaminant transformation and release; (iv) material/reagent placement, time, mechanisms, physical and chemical stability, and monitoring; (v) ecotoxicological implications; and (vi) costs.

Characteristics of contaminants

Metals and metalloids according to sediment geochemical fraction can significantly change their mobility. Since sediment composition varies from site to site due to geological, hydrographical, climatic, and socio-economic characteristics, metals and metalloids can distribute among various geochemical phases with various grades of adsorption and retention. Thus, their partitioning must be determined on a case-by-case basis before any intervention (Akcil et al. 2015). Organic pollutants may associate temporarily to the particulate matter, establishing equilibrium relations at the water-sediment interface. These sorption and desorption processes can substantially influence compound bioavailability (Viganò 2000). The direct transfer of chemicals from sediments to organisms is now considered to be a major route of exposure for many species. To evaluate the contaminant release from sediment through desorption processes, both the characteristics of sediment and overlying water column must be considered (Zoumis et al. 2001).

Over limited concentration ranges, the partitioning of HOCs between sediment and water can be described in simple terms according to a solid-water distribution coefficient (K_d) based on the assumption that partitioning depends primarily upon sediment total organic carbon (TOC) as stated in Eq. 1:

$$\frac{C_s}{C_w} = K_d = f_{oc} K_{oc} \tag{1}$$

where C_s = sediment contaminant concentration (mg/kg), C_w = water contaminant concentration (mg/L), K_d = solid-water distribution coefficient (L/kg), f_{oc} = fraction of sediment OC, and K_{oc} = OC-normalized partition coefficient (L/kg OC). For HOCs, K_{oc} is often correlated with octanol-water partition coefficient (K_{ow}) of pollutants.

Removal of target contaminants

Remediation technologies should be evaluated on the basis of their compliance with site-specific goals on a site-specific basis. Due to the complexity of field activities and the lack of proper controls, like as temporal or spatial replicates, incomplete results are available for in situ treatment of contaminated marine sediment (Mayer-Pinto et al. 2010). Additional focused field-scale demonstrations would be helpful to evaluate site-specific HOCs such as dioxins, furans, and methyl mercury whose treatment effectiveness has been either variable or slow to develop (Patmont et al. 2014).

Technological efficiency

The effectiveness of in situ remediation technologies can vary on the basis of the selected technological options. Generally, physico-chemical processes are quicker (from months to years), but with a potential immediate environmental impact, while bioremediation is slower (from years to decades) having a little impact that is diluted over time.

Sediment treated with 3.4% AC showed a decrease of the total aqueous PCB concentrations up to 87 and 92% for 1- and 6-month contact times, respectively. With active mixing, the effect of AC addition to sediment on PCB aqueous equilibrium concentration is manifested relatively quickly and is not lost with time. Similarly, adding AC to sediment reduced the water equilibrium of total PAH by 74 and 84% for 1- and 6-month contact periods, respectively (Zimmerman et al. 2004).

Adsorption kinetic studies of NB onto pure polysulfone and polysulfone/GAC hybrid membranes at 20 mg/L initial concentration showed that most NB adsorption rapidly occurs in the first 2 h, reaching the adsorption equilibrium after 6 h (Wang et al. 2014). This was due to the fact that adsorbents get adsorbed into the meso-pores during the initial stages; adsorbents need to move deeper into the pores encountering larger resistance (Himmelheber et al. 2011).

Microbial reductive dechlorination of PCBs in contaminated sediment is characterized by long lag periods and low rates ranging from months to years (Wiegel and Wu 2000). This is a significant difficulty to use anaerobic bioremediation as sediment cleanup technology. However, it has been proved that the direct addition of controlled amounts of ZVI to sediment could be an effective way to reduce the lag period prior to dechlorination at PCB-impacted sites (Wessels Perelo 2010).

Influence of natural organic matter

The content of NOM can seriously affect the performance of in situ treatments. In laboratory trials, some authors demonstrated a decrease in sorbent capacity due to NOM (Koelmans et al. 2009; To et al. 2008; Cho et al. 2012). Moreover, there could be an influence of NOM from the surrounding test area such as from overlying water and deposited sediment (Cho et al. 2009). Additional NOM could also be formed in intertidal sediment under field conditions from algae biomass decay. The adsorption of pollutant molecules by sorbent could result in greater competition for the finite number of sorption sites over time. Long-term effects and fouling mechanisms need to be further evaluated especially under field conditions.

Chemical oxidation treatments are also sensitive to NOM. Ozone decomposes back to oxygen rapidly in presence of OM. In general, OM reduces the efficiency of chemical oxidation processes scavenging considerable amounts of non-selective oxidants (Cho et al. 2009).

By-products

All in situ chemical methods have the potential to generate secondary impacts with the application of treatment reagents considering their direct related toxicity and generated post-treatment by-products. Consequently, in situ chemical treatments could be applied when the contaminated area is contained while operating or water flow can be diverted for the whole treatment duration.

Ecotoxicological assessments

Current knowledge about in situ sediment remediation techniques lacks principally of their ecotoxicological implications (Libralato et al. 2008; Han et al. 2015; Libralato et al. 2016).

Several bioindicators were used to assess the potential ecotoxicological implications of AC like bacteria, annelids, molluscs, and crustaceans. Van der Mei et al. (2008) exposed *Escherichia coli* and *Raoultella terrigena* observing cell viability as endpoint. After 30 min, mortality ranged between 83 and 96 and 54–56% for acid and basic AC, respectively, and between 76 and 78 and 32–37% for positively and negatively charged AC, in that order. Jonker et al. (2009) did not observe any adverse effect on the structure of microbiological communities after exposing them to freshwater and marine sediment treated with AC (2, 4, 10, and 20% *w*/*w*). Jonker et al. (2009) demonstrated that powdered ACs can exert adverse effects to aquatic invertebrates (*Lumbriculus variegatus*, *Daphnia magna*, and *Corophium volutator*) based on different mechanisms stating that it should be preferably washed prior to their application as well as removed, when exhausted, preventing pollutants to be released back to the environment.

Millward et al. (2005) investigated AC (3.4 and 8.5%) effect to the marine polychaete *Neanthes arenaceodentata* observing an average body weight reduction of approximately 50% caused by the ingested AC interfering with nutrients' uptake due to its affinity to lipids, carbohydrates, and proteins. At the same time, AC showed a great ability to sequester PCB. Indeed, the average PCB content in *N. arenaceodentata* was reduced by 82% after 1 month and 87% after 6 months of contact time. In the case of the marine polychaete *Nereis diversicolor*, Cornelissen et al. (2006a,b) evidenced that 2% AC reduced the PAH biota-sediment accumulation factor by 6.7-fold after 28 days. Similarly, no or slight effects were evidenced by Ho et al. (2004), McLeod et al. (2007, 2008), and Janssen et al. (2012) considering other species.

The potential effects of organo-clay, apatite, and biopolymers were investigated with several biological models like annelids (Paller and Knox 2010; Rosen et al. 2011), molluscs (Paller and Knox 2010), crustaceans (Paller and Knox 2010; Rosen et al. 2011), and fish (Rosen et al. 2011). Adverse effects were absent or slight, but sometimes measurable when in presence of biopolymers.

Also, nZVI was investigated several times (Libralato et al. 2016), including bacteria (Lee et al. 2008; Diao and Yao 2009), fungi, yeasts (Diao and Yao 2009; Otero-González et al., 2013), algae (Keller et al. 2012), rotifers (Nogueira et al. 2015), molluscs (Kadar et al. 2009, 2010), crustacean (Libralato 2014), and fish (Li et al. 2009), but effect data on in situ treated soil or sediment are substantially absent. Results are very case specific, generally with slight effects at the considered exposure concentrations.

Technology footprint and general costs

The way in which amending agents are administered in situ is of extreme importance to optimize the whole remediation performance (Cho et al. 2012). Amendments can be contained in a mat, applied in bulk onto the sediment surface, mixed into the sediment, or added as part of a sand cap or as a layer within a sand cap (USEPA, US Environmental Protection Agency, 2013a, b). When amendments are mixed into the sediment, heavy equipment has to be used, potentially producing stressing events to benthic communities along with high handling costs during placement. Analogously, when applying in situ chemical treatments, it is necessary to ensure that treatment reagents are completely mixed with the contaminated sediment layer by the introduction of one or more reagents, additives, and/or nutrients onto the sediment by spreading and settling or injecting them inside it through tubes, pipes, or other devices. Further methods to administer in situ treatments consist in isolating sediment from the surrounding environment especially when reagents or process conditions used can be harmful to the environment.

In general, using in situ treatment appears to be less expensive than ex situ treatment or disposal of contaminated sediment. It has already been estimated that the cost of a typical AC treatment is at least an order of magnitude lower than sediment dredging and disposal (Ghosh et al. 2011). However, the full technological cost must be evaluated through pilot studies on a case-by-case basis.

The cost of passive and reactive supplies depends on the type of material, purity, size, delivery, source, material processing needs, and means of application. McDonough et al. (2007) reported that cap placement costs for large-scale site (circa 405 ha) at about 25/0.836 \$/m^2, excluding the material cost. The breakup of cap placement costs is approximately as follows: (a) mobilization/demobilization 1/0.836 \$/m^2, (b) cap placement 10/0.836 \$/m^2, (c) project management 2/0.836 \$/m^2, (d) monitoring 10/0.836 \$/m^2, and (e) miscellaneous (site preparation, construction management, design, and permit) 2/0.836 \$/m^2.

Conclusions

Apart from the kind of amendment potentially useful in active capping or mixing, little is known regarding amendment application techniques, application rates, and amendment combinations that could maximize the immobilization of contaminants. As with other remedial alternatives (such as capping and dredging), the long-term permanence of amendments and their ability to retain contaminants over time are not well understood. Significant gaps remain between the current understanding of the in situ technology and the level of engineering know-how necessary for widespread implementation as an alternative remedial action mainly related to contaminant reduction efficiency under a spatial and temporal scale. Most of the times, due to the lack of real scale applications, costs are not available like as the full range of drawbacks.

The effectiveness of some amendments, such as AC and Organoclay™, has been demonstrated just in a small number of field applications, while other amendments, such as zerovalent iron (ZVI), phosphate additives, and biopolymers, are still in the bench-scale or pilot-testing phase. Even though some of these materials have been used in other environmental applications, such as groundwater and off-gas treatments, there are a limited number of projects and available performance data on their effectiveness in treating contaminated sediments.

The use of physico-chemical-based active barriers seems to be promising; however, the adsorption and/or reaction capacities of reactive materials are limited, and the contaminants cannot be completely removed from the environment. Capped sediment can also represent a future risk by excessive contaminant breakthrough due to diffusion or advection. Hence, the enhancement of biodegradation by effective microorganisms into the capping system is necessary. Biologically based active caps could have the potential to maintain reactivity over long periods of time serving as a sustainable remedial option also hosting microorganisms able to biotransform contaminants.

Future research efforts should (i) investigate long-term effects of treatment activities (i.e., years after the end of the remediation); (ii) develop database about pros and cons of adsorptive materials considering both their physico-chemical and ecotoxicological implications coupling contaminant removal and environmental compatibility issues; potentially, the database should be extended to all new proposed in situ remediation technologies; and (iii) provide cost-benefit analysis considering life cycle impact analysis as well.

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