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A review of the in-situ capping amendments and modeling approaches for the remediation of contaminated marine sediments

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Abstract

Contaminated sediments can pose long-term risks to human beings and ecosystems as they accumulate inorganic and organic contaminants becoming a sink and source of pollution. Compared to ex-situ technologies (i.e., dredging activities and off site treatments), in-situ capping (ISC) intends to minimize contaminated sediment mobilization and impact into the water column whilst treating contamination. Literature shows that numerous types of ISC amendments in presence of both organic and inorganic pollutants are investigated, although a few are contributions whose experiments have been designed and conducted with a view to future engineering. Against this background of shortcomings, this review paper intends to investigate ISC reliability, applicability and its long-term effectiveness, by also comparing reactive and physical ISCs. Additionally, an examination of the main numerical simulations applied to ISC technology was carried out. We found that activated carbon and organoclay resulted the most studied amendments for organically contaminated sediment, whereas biochar, clay minerals, and industrial-by products were more employed in presence of sediment contaminated by metal(loids). There is no better ISC system in absolute terms, since technological performance depends on many factors and only a few experimental investigations included a long-term modeling phase to predict ISC long-term efficiency. Most of numerical models included simplified transport equations based on diffusion and adsorption, and the goodness of fitting between experimental and modeled data was not always computed. The review finally

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discusses new research directions such as the need for long-term applications on field-scale and cap effectiveness in presence of site-specific tidal forces and currents.

Keywords

Adsorption; Contaminant fate and transport; Environmental remediation; Long-term simulation; Organic and inorganic pollution.

1. Introduction

Sediments represent the main receptor of organic and inorganic pollutants entering the aquatic system including heavy metals, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), etc., which are principally released into the environment by industrial and anthropic activities (Chen *et al.*, 2020). Aquatic sediments pose further challenges when compared to terrestrial soils, since their higher organic content increase contaminant adsorption, allowing them to enter more easily through water transport phenomena into the food chain and human consumption (Meric *et al.*, 2012). This effect has entailed the implementation of environmental regulations and development of management programs to improve environmental quality. In Europe, a set of used standards is represented by the so-called “Dutch Standards”, environmental pollutant threshold limits and intervention values (i.e., concentrations in environmental matrices) for environmental investigation and remediation. Furthermore, the European Directive 2000/60/EC (European Directive 60/EC, 2000) established a framework for the protection of inland surface waters, coastal waters, transitional waters, and groundwater. The Environmental Quality Standards Directive (EQSD) (European Directive 105/EC, 2008) sets out environmental quality standards (EQSs) concerning the presence of priority pollutants in surface water because of the significant risk they pose to, or via, the aquatic environment.

During last years, differing remediation technologies have been developed to reduce, clean up and restore contamination in sediments (Lofrano *et al.*, 2017; Bianco *et al.*, 2020). They can be generally classified in (i) in-situ treatments, which are applied on site, such as capping and Monitored Natural Recovery (MNR), and (ii) ex-situ treatments, which entail sediment dredging and off-site treatment or disposal (Lofrano *et al.*, 2017). Several concerns are posed on disposal or reuse of dredged material due to their contamination content (Mymrin *et al.*, 2017). Additionally, dredging operations involve the resuspension of contaminated particles and high CO_2

emissions, representing a serious risk to the ecosystem. Due to its high cost, dredging is often applied limitedly to small areas or hot spot zones (Abel, Nybom and Akkanen, 2020). However, the Superfund program, administered by the US EPA, demonstrated the feasibility of dredging at the so-called “megasites” (where the cost of remedial activities exceeds $50 million) (NRC, 2007). In this context, In-Situ Capping (ISC) is a reliable alternative technology. It entails the placement of a clean material layer (e.g., sediments or a combination of materials, etc.) on the top of contaminated sediments. Several field and bench-scale capping applications are presented in literature (Cornelissen *et al.*, 2012; Park *et al.*, 2018; Bortone *et al.*, 2020), demonstrating how ISC controls contamination in sediments and, subsequently, to the overlying water. The main advantages of an ISC-based remediation can be summarized as (i) relatively lower costs compared to other sediment remedial technologies (Lofrano *et al.*, 2017), (ii) suitability for sequestering both organic and inorganic pollutants (Sharma and Reddy, 2004), and (iii) all the intrinsic advantages of being an in-situ technique, such as the reduction of resuspension phenomena (Ting *et al.*, 2020). For instance, in the Hudson River remediation project, ISC application allowed a cost reduction of one order of magnitude compared to conventional dredging (Ghosh *et al.*, 2011). According to Jansen et al. (2016), ISC has both community and sustainability advantages if compared to dredging and off site treatments, where the overall costs might be even higher than ISC. A disadvantage of ISC systems is related to their long-term effect of remediation activities (generally years after the cap placement) and possible uncertainties related to the variability of natural environmental conditions (e.g., pH and nutrient levels).

The literature shows substantial advances in new materials for ISC systems, although critical insights into the engineering of remediation (e.g., time duration of capping) are limited. Studies concerning the modelling of ISC systems, which should discuss the duration of remediation, are rare and in many cases with variable methodological approaches. In this review paper, several types of capping amendments are investigated and compared, to assess the most effective materials according to the type of contamination and environmental conditions, specifically referring to organic and inorganic compounds. Additionally, the main processes and parameters influencing ISC design and treatment and their combined role are also analyzed, together with revising how existing numerical models assist with their interpretation and prediction over time. It is intended to highlight the need for a long-term modeling assessment of the ISC contaminant environmental fate and transport. Specifically, the estimation of the model uncertainties is a crucial task to demonstrate the reliability of results.

2. In-Situ capping technology

Conventional ISC approaches commonly use clean sediment or sand as barrier to physically separate contaminated sediment from the overlying water layer, known as (physical) capping.

However, ISC remediation can also involve the placement of reactive amendments that bind or adsorb the sediment pore water contaminants to reduce contaminant spreading and mobility, known as reactive capping (**Table 1**) (Meric *et al.*, 2012).

On the top of ISC, an armor layer of gravel, cobbles, and/or large boulders can be placed in order to restrain the capping system. In some cases, a habitat layer designed to reinstate the native sediments and promote recovery of local benthic communities may also be added (US EPA, 2015) (**Fig. 1**).

The main functions of ISC systems can be summarized as (Sharma and Reddy, 2004):

1. separation of the contaminated sediment area from the benthic environment,
2. prevention of resuspension and transport phenomena of contaminated sediments,
3. reduction of dissolved contaminant fluxes into the overlying water column.

The clear advantage of using ISC lies in the fact that they do not require contaminated sediment excavation or movement, by preventing further contamination of the surrounding matrices.

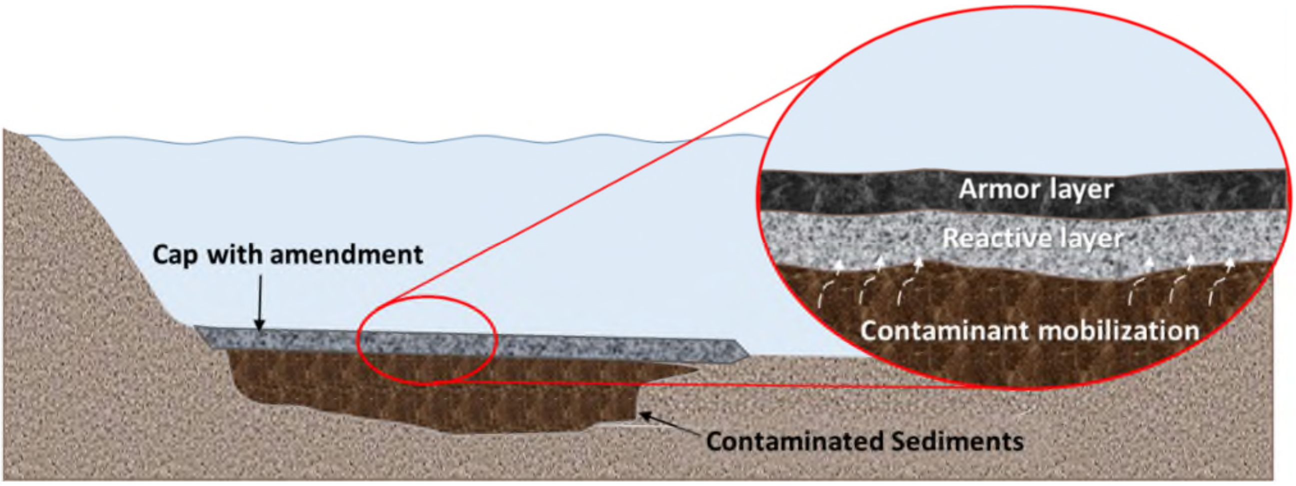

Figure 1. ISC technique with reactive and armor layers**.**

Table 1. Differences between physical and reactive capping technologies (adapted from Zhang *et al.*, 2016).

	(Physical) Capping	**Reactive Capping**
Materials	Crushed rock debris, clean dredged sediments, sand, silt, and clay (natural materials).	Activated carbon, organoclay, zero-valent iron, zeolite, apatite, biopolymers, etc. (reactive amendments).
Tasks	Containment: 1. Contaminated sediment physical isolation and stabilization. 2. Reduction of the dissolved contaminant flux into the overlying water.	Containment and treatment: 1. Chemo-physical isolation of contaminated sediment. 2. Sorption and degradation of pollutants in sediment. 3. Reduction of the dissolved contaminant flux into the overlying water even in complex environments.
Thickness	< 100 cm	< 30 cm
Field testing	Advanced	In progress
Average cost	15 €/ton*	500 €/ton**

*(Platias *et al*., 2014; Yoo *et al.*, 2014)
**(Todaro *et al.*, 2021)

Capping by using natural material such as sand or clean sediment, has been widely investigated in various studies and mostly utilized for the large availability and ease of placement of materials (Jiao *et al.*, 2020).

In Bortone *et al*. (2018) a sand cap was investigated to reduce the exposure of the aquatic ecosystem to PCB-contaminated sediments in Lake Hartwell, US. Numerical simulations were performed by using COMSOL Multiphysics® and compared two sand-cap scenarios, revealing their effectiveness in the long-term. Specifically, it was demonstrated that PCB transport was extremely dependent on the cap and material characteristics, and that the cap thickness could further be reduced by using a more sorbent sand (i.e. with higher retardation factor). A minimum 20cm-thick sand cap allowed reducing the contaminant transport to the overlying water. In the study by Meric *et al.* (2014a), a 7.5 cm thick sand cap reduced the bioavailability of PCBs by a factor of 100 compared to the no-capping scenario, but it did not influence the bioavailability of naphthalene. However, sand as capping material has been mainly applied to decrease nutrient concentrations in water (Kim *et al.*, 2007; Jiao *et al.*, 2020).

Nevertheless, a conventional sand cap might partially allow dissolved contaminants into the overlying water column, and consequently still pose a risk to the benthic environment and the trophic chain. In these cases, reactive materials can be added to the ISC system. Murphy and Lowry (2004) demonstrated that a thin layer of adsorptive amendment (i.e., activated carbon) can have a more performing pollution containment capacity for PCBs, equal to over 100 times sand adsorption effectiveness. Additionally, reactive amendments may represent unsuitable habitats for benthic communities and as such, a clean armor layer often serves as habitat restoration space (Lampert, 2010).

One current concern about ISC is the possible breach of the containment layer due to erosional and tidal forces, but also strong sea currents influenced by subaqueous ground water discharge, wave action or wave pumping. Therefore, the main purpose of the armor layer (**Fig. 1**) is to provide protection to the capping layer from being eroded away (Mohan *et al.*, 2000). Additionally, laboratory investigations estimated that a traditional cap thickness is approximately 100 cm including an armor layer, but further studies may be necessary in the field-scale at high water depth (Zhang *et al.*, 2016).

In this regard, an innovative use of reactive ISC (**Fig. 2a**) entails encapsulation of the reactive amendments (e.g., activated carbon, organoclay, apatite, etc.) (**Fig. 2c**) between two geotextile layers (**Fig. 2b**), by further reducing ISC total layer thickness to about 1 cm (Meric *et al.*, 2014b; Bortone *et al.*, 2020). It includes additional benefits such as uniform consolidation, defined mass per area, and drainage. Reactive Core Mat (RCM) technology has been adopted for several US EPA Superfund site projects (CETCO, 2019), where a 15 cm thick-sand layer was set over the reactive mat both for the ISC anchoring and allowing benthic organisms colonization. Meric *et al*. (2014b) tested RCMs and demonstrated that the geotextile layers added a beneficial filtering for low advective fluxes (at early stages of stress application) to capture Hydrophobic Organic Compounds (HOCs), while the reactive core provided a strong HOC sorption during high advective fluxes (at later stages of stress application, with a final backpressure of 200 kPa).

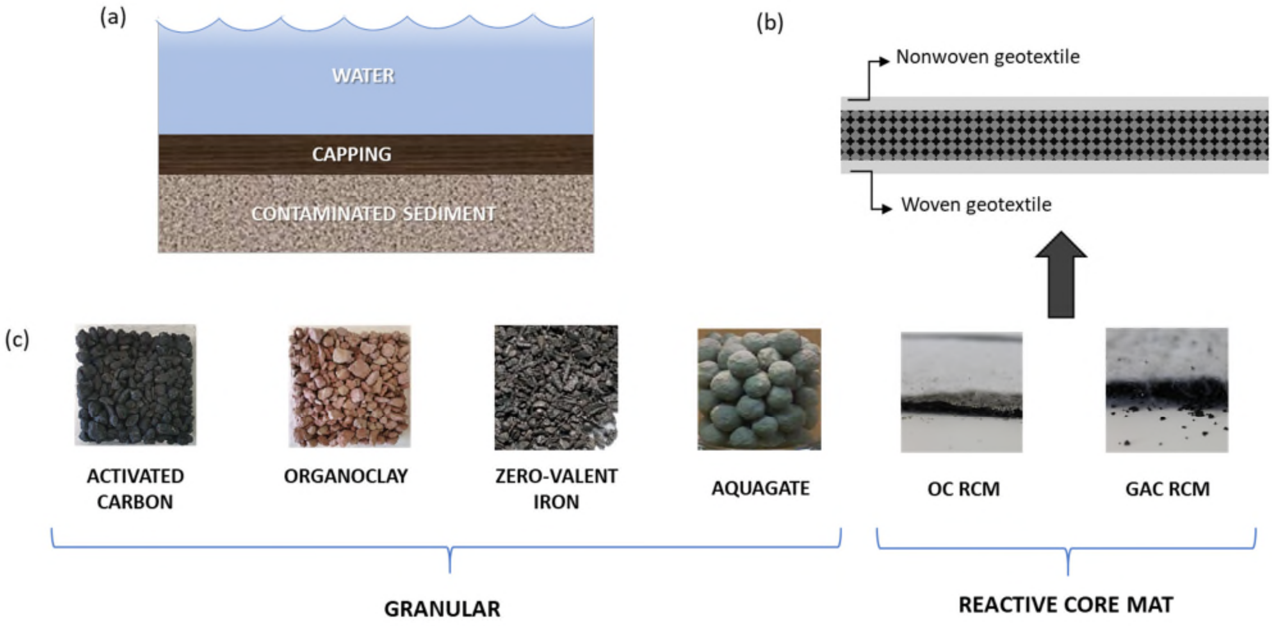

Figure 2. (a) Capping layer schematization; (b) RCM schematization; (c) main capping materials in literature.

Alternatively, an innovative reactive amendment is represented by AquaGate® (**Fig. 2c**), an AquaBlok® patented system (AquaGate, 2020) where the reactive amendments cover an inner inert core more stable and slough off the aggregate during hydration, enabling mixing with the contaminated sediment. For the thin reactive layer powdered activated carbon (PAC), organophilic clay or other sorbent materials have been tested and monitored, confirming their long-term performance. AquaGate has been successfully used for application in sites contaminated by PCBs, PAHs, and pesticides (Johnston *et al.*, 2014; Kirtay *et al.*, 2016). It allows for in-situ treatment to be applied to deep water depths and high energy surface water systems and may be used for freshwater as well as marine environments. Its innovative formula permits a natural mixing of the reactive additives with the sediments after placement.

Therefore, the use of reactive materials can significantly reduce the capping thickness compared to conventional caps (Wessels Perelo, 2010).

Common reactive amendments applied in pilot and laboratory experiments include: apatites (Knox *et al.*, 2012a; Xing *et al.*, 2016; Zhang *et al.*, 2016) and zeolites (Kang and Park, 2015; Gu *et al.*, 2019), which adsorb and treat complex different metals; organophilic clay (OC) (Olsta, 2010; Erten *et al.*, 2012; Bortone *et al.*, 2020), biochar (BC) or activated carbon (AC) (Janssen and Beckingham, 2013; Ting *et al.*, 2020; Bianco *et al.*, 2021), which effectively adsorb organic and inorganic compounds; zero-valent iron (ZVI) for treating nitroaromatics, metals, DDT, and nutrients (Chapman *et al.*, 2020; Hu *et al.*, 2020). In general, amendments modify sediment geochemistry by improving contaminant stability and binding capacity, reducing contaminant flux into the water column and consequent bioavailability to benthic organisms and bioaccumulation in the food chain, while allowing upward groundwater flow (Ghosh *et al.*, 2011).

Many innovative bio-materials have great potential as ISC amendments and are still under study. In order to remove nitrobenzene from sediment, Wang *et al.* (2014) developed a bio-reactive cap including polysulfone (PS), granular activated carbon, and immobilized microorganisms. Another bio-reactive amendment was investigated in the study by Sun *et al.* (2010), where a mixture of indigenous microorganisms from Songhuajiang River sediment (China) was added to simulate the real in-situ environment. Atashgahi *et al.* (2014) demonstrated that the organohalide respiration of chlorinated ethenes in bioreactive caps can be improved by incorporating solid polymeric organic materials, such as wood chips, shrimp waste, hay, straw, and tree bark, which were used as electron donors for cis-dichloroethene (cDCE) and vinyl chloride (VC) degradation.

2.1. Design aspects and modeling approaches of an ISC system

The transport of different types of contaminants through engineered porous containment capping systems has been modeled extensively over the past 40 years (Rowe and Booker, 1985; Malusis and Shakelford, 2002; Silvani, *et al.*, 2017; Bortone *et al.*, 2020). Numerical simulations represent a useful tool to evaluate the effectiveness of an ISC treatment, predicting concentrations and fluxes of contaminants over time in sediment, capping, and water layers.

Conventional modelling aims at simulating porous media transport processes such as advection, diffusion, dispersion, sorption, and reaction (**Fig. 3**). However, there are several unconventional processes at the near-surface sediment influencing contaminant behavior, such as deposition, bioturbation, consolidation, multispecies reactions, and exchange with the overlying water (Shen *et al.*, 2018).

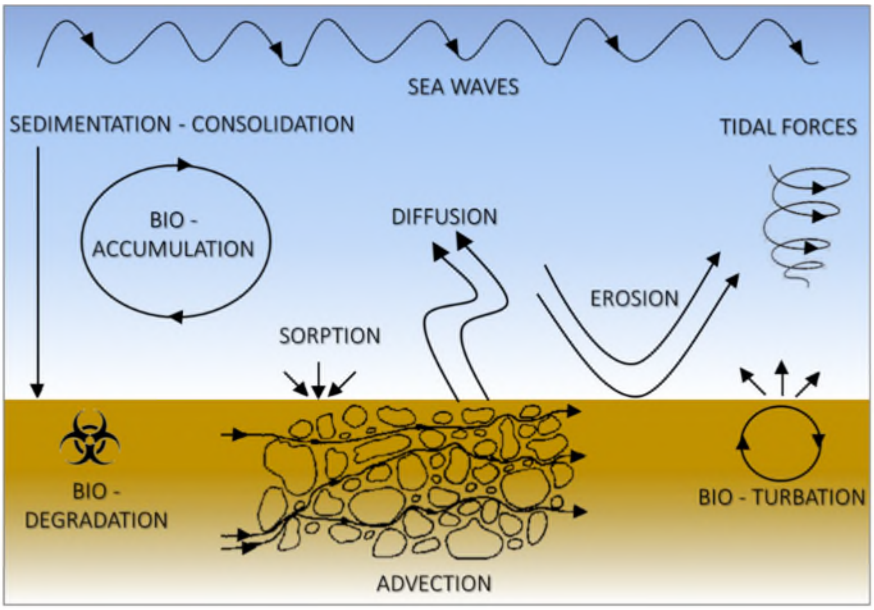

Figure 3. Main processes involved in sediments, water, and contaminants at the sediment-water interface.

Since the mid-20th century, numerous researchers have worked on the diffusion-advection-dispersion equation through analytical approximations (Craig and Rabideau, 2006; Wang and Zhan, 2015; Yan *et al.*, 2019), numerical simulations (Tompson and Gelhar, 1990; Craig and Rabideau, 2006; Go *et al.*, 2009; Boso *et al.*, 2013; Bortone *et al.*, 2020), and experimental investigations (Starr *et al.*, 1985; Willingham *et al.*, 2008; Silvani *et al.*, 2017) of fully saturated sediment environments.

The sediment system is generally conceptualized as one-dimensional and composed of stratified multiple layers with various chemo-physical properties according to sediment, capping and water characteristics (Palermo, 1998). All layers can be simulated as saturated porous media with immobile solid particles with the exception of the bioturbation zone, where benthic organisms and sea currents move the particles. Contaminant mass transport through porous media is typically described by the conventional transport model for the one-dimensional multi-layered, one-species porous media system with the capacity to account for physical (advection, diffusion) and chemical-biological phenomena (sorption and degradation) (**Eq. 1**) (Shen, 2017; Shen *et al.*, 2018):

$$R_i \frac{\delta C_{w,i}}{\delta t} = D_i \frac{\delta^2 C_{w,i}}{\delta z^2} - u_z \frac{\delta C_{w,i}}{\delta z} \pm \Sigma r_i \tag*{(Eq. 1)}$$

$$R_i = 1 + \frac{\rho_{s,i}}{\mathcal{E}_i} K_{d,i} \tag*{(Eq. 2)}$$

$$K_{d,i} = \frac{\delta C_{s,i}}{\delta C_{w,i}}$$ **(Eq. 3)**

where:

R_i= retardation factor
$C_{w,i}$= porewater concentration
$C_{s,i}$ = individual solid concentration
D_i= dispersion coefficient
ε_i = porosity of the i-th layer
u_z= water velocity
$\rho_{s,i}$ = bulk density of the i-th layer
r_i = summation of chemical and biological reaction rates in the i-th layer
$K_{d,i}$ = partitioning coefficient in the i-th layer

Contaminants can migrate within the layers and across the i-th layer interfaces. Further details including initial and boundary conditions, are described in Shen (2017) and Shen *et al.* (2018).

The adsorption/desorption phenomena taking place in the sediment and capping layers usually do not involve local reversible reactions in the time scale of transport processes, allowing the assumption of local equilibrium. In this way, the interaction between the solute and the solid phase of the porous medium is described by equilibrium isotherms (e.g., linear, Freundlich, Langmuir, etc.) involving the calculation of a retardation factor R_i (**Eq. 2**) (Bear, 1972), which is correlated to the contaminant partitioning coefficient (**Eq. 3**) (Acar and Haider, 1990). However, the assumption of sorption phenomena and local equilibrium is a strong simplification of the real solid-liquid interaction processes. For example, for some organic compounds (e.g., HOCs) sorption/desorption phenomena could be slow with strongly sorbing phases or solids (Accardi-Dey and Gschwend, 2002; Yang *et al.*, 2012). In this case, linear/nonlinear sorption/desorption kinetics better describe the phenomenon involved. In the study by Bortone *et al.* (2020), the adsorption/desorption phenomena in the sediment layer were described by considering time-varying kinetic properties of different materials to represent PAH partitioning and transport over time as shown in **Eq. 4** (Di Natale *et al.*, 2015; Minick and Anderson, 2017)

$$\Sigma r_i = (1-\varepsilon_i)\rho_{s,i}\frac{\delta C_{s,i}}{\delta t} = k_{c,i} \cdot a_i \left(C_{w,i} - C_{eq,w,i}\left(C_{s,i}\right)\right)$$ **(Eq. 4)**

where $k_{c,i}$ is the mass transfer coefficient between water and particles, a_i is the specific area of the sorbent particles in the i-th layer and is equal to $6/d_{p,i}$, where $d_{p,i}$ is the particle average diameter.

It is well-known that the effective diffusion coefficient of the single contaminant takes into account the tortuosity and porosity of the porous media (Millington and Quirk, 1961). Hydrodynamic dispersion and advection are related to the transport of pollutants forced by hydraulic flows and they both are dependent on the water velocity u_z (**Eq. 5**) (Shen *et al.*, 2018):

$$u_z = V_{darcy} + V_{con,0}e^{-k_{com}t} + V_{oscillation} * \sin(\frac{2\pi t}{t_c}) \qquad \textbf{(Eq. 5)}$$

where u_z includes the term of the Darcy velocity by groundwater upwelling, the pore water velocity due to consolidation of solid particles, and periodic movements related to periodic tidal waves/drainage flows. Several researchers modeled periodic groundwater flow velocity as a sinusoidal function versus time where t_c is the period for a full tidal cycle (Depner and Rasmussen, 2016). In general, consolidation is modeled as an increase of the water upwelling decreasing over time as the sediment seabed reaches the equilibrium. However, a modeling study demonstrated that consolidation-induced advection can involve an acceleration of contaminant breakthrough through the cap by many orders of magnitude (Alshawabkeh *et al.*, 2005). Furthermore, bioturbation occurs through the activities of benthic organisms living at the sediment-water interface (∼5–15 cm). A common modeling approach assumes the mixing process randomly and the bioturbation flux as a diffusion process governed by Fick's second law (Fick, 1855; Cornelissen *et al.*, 2012; Shen *et al.*, 2018). Furthermore, the deposition and accumulation of sediment can be modeled as a new layer with increasing thickness over time (Walker *et al.*, 2013).

Numerical modeling can be useful to design the right capping thickness once all phenomena involved are identified. The cap design must consider its effectiveness both in the short and long-term chemical isolation of contaminants (Palermo, 1998).

Numerous studies have shown the potential of different amendments for addressing both organic and inorganic contaminants present in the environmental matrices. The following sections will discuss ISC interventions by showing specific case studies and limitations of their effectiveness.

3. ISC amendments for organic contamination

The most common organic contaminants in sediments include PCBs, PAHs, pesticides, phenols, BTEX, halogenated and non-halogenated volatile organic compounds (VOCs) (USEPA, 1997). Several researches demonstrated that high PAH concentrations in the environment cause a high incidence of mutagenic events and cancer for exposed organisms (Ramesh *et al.*, 2011). Additionally, the intensification of extensive agricultural activities involved an increased concentration in the environment of pesticides (i.e., organochlorines compounds). The latter are commonly known for their risks of hormone-related cancers but also hypertension, cardiovascular diseases, and other human diseases (Kaur and Kaur, 2018).

Remediation of sediment contaminated with organics requires further research and development, especially for innovative ISC amendments, which have demonstrated to be effective and not invasive (Pagnozzi *et al.*, 2020). Laboratory investigations by Choi *et al.* (2013) showed the effectiveness of AC amendment for PAH sequestration from petroleum-contaminated sediments. An AC dose of 5 wt % tested after 1, 2, 8, and 12 months showed a total 99.5% reduction in PAH uptake in polyethylene (PE) 5 wt % samplers. In this case, the site-specific mass transfer parameters were evaluated. However, for the 75−300 μm particle size AC, the numerical modeling overestimated the capping performance and underestimated the relative uptakes in PE, particularly after 1 and 2 months of contact. The overestimation of AC performance especially in the beginning of AC−sediment contact (by the mass transfer model) has been found in previous studies with PCBs (Hale *et al.*, 2010) and DDTs (Hale *et al.*, 2009).

In presence of PCBs, Choi *et al.* (2018) demonstrated that AC facilitated the desorption of 2-chlorinated biphenyls and contributed to their complete sequestration. However, in the study by Cornelissen *et al.* (2012), AC showed to be less effective in field applications compared to laboratory tests, probably because of longer sediment-to-AC mass transfer times and field phenomena, such as bioturbation and new local deposition of contaminated sediment. Additionally, Pagnozzi *et al.* (2020) demonstrated that activated carbon enhanced microorganism bioactivity, biodegradation, catabolic gene abundance, and early biofilm formation. Microorganisms activity has often had a positive role on the degradation of pollutants, as shown in the study by Sun *et al.* (2010), where more than 90% of nitrobenzene (NB) and aniline (AN) were removed over 3 days, and over 67% of AN was degraded in the first day of treatment. Results by Wang *et al.* (2014) also demonstrated that the bio-reactive capping system effectively removed the 70% of NB from contaminated sediments by microbial degradation. SEM (Scanning Electron Microscopy) observations proved that all the caps consisted of a top layer and a porous sub-layer, blocking NB transport from sediment to water, by enhancing NB degradation. Adsorption tests showed that GAC performed better than polysulfone (PS) ISC system as confirmed by Freundlich adsorption isotherm parameters. Sarkar *et al.* (2012) described how a bioremediation approach integrating microorganisms with organoclays can degrade pollutants. They demonstrated that hexavalent chromium sorbed on organoclay was reduced to its trivalent state, which is considerably less toxic. Therefore, the integration between sorption by organoclays or activated carbon and biodegradation by microorganisms could be a promising, cost-effective, and environmentally friendly technique.

Biological-based amendments have been demonstrated to be effective in reducing contamination in the long-term by Atashgahi *et al.* (2014). Methanogenesis and short chain fatty acids decreased, and high dechlorination rates of cDCE and VC were recorded. The effectiveness of the capping materials was evaluated using a 1D numerical model. The latter consisted of a 10 cm layer of aquifer, 100 cm of sediment and a 40 cm thick capping layer. After an intensive observation period of the phenomena involved in the site, the model domain was set with a Darcy velocity of 2 cm/day and a first-order degradation rate for cDCE and VC in the aquifer. The modeling simulation indicated that a 40 cm-thick capping layer with hay, straw, tree bark or shrimp waste was effective to reduce VC concentration below the threshold limit. However, an important drawback in the short-term was the release of fermentation products and subsequent methane production.

Organoclay (OC)-based capping systems, obtained by the combination of clay minerals and surfactants (quaternary alkyl ammonium salts and others), have great sorption capacities (Knox *et al.,* 2012a; Reible and Lampert, 2014; Guégan, 2019). Hawkins *et al.* (2011) compared AC and three types of organoclay products (CETCO, Polymer Ventures, and Biomin, Inc.) for a lab-scale test using sediment from Cottonwood Bay in Grand Prairie, Texas, US. The overall adsorption of higher chlorinated PCB congeners was greater than that of lower chlorinated PCB congeners. With regards to PAHs, AC was found to be more efficient than CETCO OC. Furthermore, they found that for CETCO organoclay, preloading with humic acid improved PAH adsorption capacity, underscoring that if sorbing amendments are exposed to natural organics (at >1 g/L), their interactions can influence the performance of the sorbent. A similar comparison was carried out in the study by Bortone *et al.* (2020), where the effectiveness of OC RCM and Powdered Activated Carbon (PAC) in the AquaGate® technology was investigated in the presence of PAH-contaminated marine sediment. The best result, in terms of higher contamination reduction in the water column, was given by the AquaGate® + 5 % PAC capping system for the cases of benzo[a]pyrene and pyrene. They highlighted that numerical modeling uncertainties were mostly related to the short observation period (20 days) and to some chemical and geophysical site-specific parameters, crucial to the calibration of the numerical simulations. In agreement with Yan *et al.* (2019), they stressed the necessity to evaluate the influence of mass transfer coefficients on contaminant transport through the sediment and capping layers. The mass transfer coefficient for the sediment desorption process was calculated through the equation proposed by Minick and Anderson (2017).

Consolidation of the sediment layer can strongly influence the pollutant transport mainly in the short-term. In the bench-scale tests performed by Meric *et al.* (Meric *et al*., 2012; Meric *et al*., 2013; Meric *et al*., 2013), PAH and PCB-contaminated sediment was covered with a layer of OC RCM and one of clean sand with organic trout chow mix. A coupled large-strain consolidation was used for PCB numerical simulations (due to the pressure applied by the weight of the overlying armoring layer) considering solute transport through a deformable saturated porous media, reactive advective/dispersive transport through the OC RCM, and a non-reactive advective transport through the armoring layer. A substantial decrease of the bioavailable persistent HOCs was observed after the placement of the RCM. Naphthalene was used for PAH-simulations due to its higher solubility in water compared to other PAH compounds. However, it was highlighted how the Freundlich equation failed to simulate the phenomenon, when a full data set was considered. Therefore, the measured PAH-sorption isotherm was considered as a bilinear relationship with two different linear partitioning coefficients, in particular one for the equilibrium concentration (Ce) < 0.5 mg/L (9,180 L/kg) and one for Ce > 0.5 mg/L (40,350 L/kg).

An additional ISC material is zero-valent iron (ZVI), extensively investigated for its capacity to reduce organic pollutants to non-toxic or more easily degradable low-toxic forms, combined with the oxidation of Fe(0) to Fe(II) and Fe(III) (Yang *et al*., 2010). Sun *et al*. (2010) showed that Fe(0) effectively transformed NB to AN, which has higher biodegradability. With the integration of cinder, Fe(0) and microorganisms, the NB released from sediment to the upper-layer water was effectively blocked. However, it was more difficult to biodegrade NB than AN, as expected and in line with other studies (Zhao et al., 2018). The NB reduction kinetics fitted a pseudo-first order kinetic model, but they underlined the need to optimize the related parameters to achieve a better remediation efficiency. In recent times, the development of nanotechnology has offered more efficient and cost-effective solutions. Thanks to a larger specific surface area with a higher surface reactivity, the nanoscale zero-valent iron (nZVI) increases the contaminant removal compared to conventional ZVI (Jiang *et al*., 2018).

Another innovative amendment is biochar (BC), which is a porous carbonaceous material produced by the thermal processes (e.g., pyrolysis and gasification) of biomass under anoxic or limited oxygen conditions (Ahmad *et al.,* 2014). The principal BC effects are correlated to the phenomena of pollutant adsorption–desorption, microbial metabolism, and pollutant bio-chemical degradation (Luo and Gu, 2016). Silvani *et al*. (2017) tested a 3 cm (∼150 g) thick layer of biochar as ISC material for oil spill contaminated sediment in lab-scale experiments. After 1 month, anthracene porewater concentrations were reduced respectively by 69%, 56%, and 99%, for AC, OC, and BC. They considered the linear-equilibrium model to describe the instantaneous sorption as retardation factor in cap materials. Additionally, lateral concentration gradients were considered negligible because vertical gradients were much more detectable, justifying the 1D approach. Therefore, BC showed an extensive capping efficiency compared to conventional amendments. Additionally, it represents a cost-effective alternative. For a thin capping layer, AC materials cost about 10 USD/m2, whereas BC costs about 2 USD/m^2 (Zhu *et al*., 2019).

Tables 2 and **3** list the main points of ISC application for organic compounds with information about ISC design and material, target compounds investigated, experimental and modeling details and the main remarks of the study.

Table 2. Reactive amendments employed in ISC in case of dioxin/PCB-contaminated sediment.

Targeted compounds	**ISC material and design**	**Scale**	**Intervention duration and site**	**Phenomena investigated and/or modeled**	**Main remarks**	**Reference**
Polychlorinated dibenzodioxin and dibenzofuran (PCDD/F)	• Clay + AC 2.5 cm thick	Field scale (200 × 200 m, 40,000 m^2)	600 days, Grenlandsfjords, Norway	• Sorption • Diffusion	• Only 25% of AC was found inside the pilot fields. • Sediment-to-water PCDD/F fluxes in-situ diffusion chambers were lower in capping system than reference fields. • Reductions of contaminants were 60% for clay + AC. • Reductions in water PCDD/F concentrations were of 20–40%. • AC was more effective in laboratory than in field.	(Cornelissen *et al.*, 2012)
PCBs	• Activated carbon from coconut shell and coal based • Three types of organoclays (CETCO, Polymer Ventures and Biomin, Inc.)	Bench-scale (subsequently field-scale)	180 days, Cottonwood Bay in Grand Prairie, Texas, US	• Advection • Consolidation • Diffusion • Sorption	• In CETCO OC, adsorption of higher chlorinated PCB congeners was stronger than lower chlorinated PCB congeners. • Humic acid preloading effect was less significant for higher chlorinated congeners.	(Hawkins *et al.*, 2011)
	• OC RCM 1.25 cm thick	Bench-scale (ICSTAC device)	28 days, Neponset River, Milton, MA, US	• Biota-sediment accumulation	• Bioavailability was reduced up to 50% for PCBs (and also naphthalene). • 1D-numerical model was used with coupled large-strain consolidation and solute transport through deformable and saturated porous media, with reactive-advective and dispersive transport through the cap. • Worms exposed to contaminated sediment showed higher contaminant concentrations than those exposed to sand and trout chow mix.	(Meric *et al.*, 2012)
	• Pristine GAC • GAC impregnated with Fe/Pd where 0.6 g of AC is mixed with sediment	Bench-scale reactor	100 days, Waukegan Harbor, Illinois, US	• Sorption and dechlorination	• PCBs desorbed from sediment were absorbed by AC and subject to dechlorination. • PCBs in sediment exhibited a five-order of magnitude lower dechlorination yield than those in pure aqueous phase.	(Choi, 2018)

Table 3. Reactive amendments employed in ISC in case of PAH-contaminated sediment.

Targeted compounds	**ISC material and design**	**Scale**	**Intervention duration and site**	**Phenomena investigated and/or modeled**	**Main remarks**	**Reference**
Nitrobenzene, aniline	• Cinder 1-3 cm thick • Fe(0) + cinder 1 cm thick • Fe(0) + cinder + microorganisms 1-3 cm thick	Bench-scale (50 x 16 cm reactor)	40 days, Songhuajiang river in Jili, China	• Sorption • Degradation	• Fe(0) effectively transformed NB to AN, which has better biodegradability. • With cinder, Fe(0) and microorganisms, the released NB from sediment was effectively stopped.	(Sun *et al.*, 2010)
	• Polysulfone (PS)/GAC hybridmembrane • PS/GAC + microorganisms • Sand 1 cm thick (particle size 0.1 mm construction sand) • Sand + AC 0.5 cm thick (sand amended with 2% by weight AC)	Bench-scale (2 L becker)	40 days, Qinhuai River, China	• Sorption • Degradation	• About 21.7, 28.3 and 43.9% of NB in the sediment was removed for PS/GAC membrane, sand-alone and sand amended with AC, respectively. • More than 70% of NB was reduced with PS/GAC + microorganisms capping system.	(Wang *et al.*, 2014)
PAHs	• OC RCM 1.25-cm thick	Bench-scale (ICSTAC device)	28 days, Neponset River, Milton, MA, US	• Sorption • Consolidation	• The measured sorption isotherm was investigated as a bilinear relationship with two different linear partitioning coefficients. • RCM short-term efficiency to capture the contamination with nonlinear sorption behavior for OC and sorption capacity increasing with increasing naphthalene concentration.	(Meric *et al.*, 2013; Meric *et al.*, 2013)
	• AC at different doses (5 or 10 dry wt %) and particle sizes (<32, 32−75, 75−150, or 75−300 μm)	Bench-scale (0.04 L vial)	365 days, Petroleum-impacted site	• Desorption • Mass transfer model	• Reduction up to 99% and 98% for sediments contaminated with oil contents of 1% and 2%, respectively, by treatment with 5% AC.	(Choi *et al.*, 2013)
	• Sand mixed with 4% sorbent material (AC, OC or BC, w/w) 3 cm thick	Bench-scale (cylinder (20 × 7 cm)	180 days, Messina sea, Sicily, Italy	• Sorption • Diffusion	• Anthracene porewater concentrations in capping material were reduced by 69%, 56% and 99% for AC, OC, and BC, respectively (after 1 month).	(Silvani *et al.*, 2017)

	• OC RCM (CETCO PM100) • powdered activated carbon (PAC), particle size of 500> d >250 µm	Bench-scale (0.03 L vial)	Duration not specified, Midwestern, US	• Degradation	• AC performed better than OC and enhanced biodegradation, catabolic gene abundance, early biofilm formation, and a greater sorption capacity.	(Pagnozzi *et al.*, 2020)
	• Aquagate + 5% PAC 5 cm thick; • OC RCM 1 cm thick	Bench-scale (700 cm^2 x 150 cm column)	20 days, Mar Piccolo of Taranto, Italy	• Sorption/Desorption • Mass transfer model • Diffusion • Advection	• 1D numerical model was used with mass transfer parameters to simulate pollutant desorption from sediment. • A good fitting of both AG PAC and OC RCM modeling for PAH fate and transport over time was found. • The best material, in terms of higher contamination reduction in water, was AquaGate® + 5% PAC in presence of benzo[a]pyrene and pyrene.	(Bortone *et al.*, 2020)

4. ISC amendments for inorganic contamination

The main sources for heavy metal pollution can be identified in natural (e.g., volcanoes, rock weathering, biological decay, thermal springs, etc.) and anthropogenic origin such as industries, mining practices, vehicle emissions, farming and agricultural activities (Esmaeili *et al.*, 2014). The effectiveness of reactive materials on metal sequestration relies on several factors such as pH, redox potential, competing ions, and others. For instance, metal speciation and transport in aquatic environments is influenced by a dynamic equilibrium between oxidized and reduced forms of iron (Fe), manganese (Mn), and sulfur (S) (Simpson *et al.*, 2002). Furthermore, organisms can bioturbate and bioirrigate the sediment, influencing metal speciation and fluxes. There is a variety of sequestering agents, which can be used in reactive caps including mineral additives such as zeolites, apatite, fly ash, zero valent iron, goethite, hematite, ferrihydrite, and biopolymers. Specifically they act by reducing the availability and toxicity of metals by modifying their chemical speciation in sediments (Taneez *et al.*, 2018). Apatite, for instance, represents a readily available phosphate source able to retain lead (Pb), cadmium (Cd), nickel (Ni), and zinc (Zn) in subsurface and surface sediments, representing an economical and environmentally-friendly treatment for contaminated sediment (Knox *et al.*, 2007). However, apatites can naturally contain other impurities (i.e., arsenic, chromium, uranium) that may cause additional contamination when used for environmental purpose. Therefore, future studies should weigh pros and cons of using these materials as capping amendments and identifying new possible alternatives. From a circular economy perspective, fish bone (as waste from fish processing) represents a cheap source of biogenic apatite that could be used as an apatite source in capping amendments (Zhang et al., 2016).

Another material particularly effective to also remove metal(loids) is organoclay (OC), widely described in section 3, which consists of a bentonite layer modified with quaternary amines. Knox *et al.* (2014) evaluated the effects of apatite, OC, zeolite, and biopolymers (i.e., chitosan and xanthan) on metal speciation, mobility, and retention. Their main results revealed that amendments reduced chromium (Cr), cobalt (Co), Pb, and Ni concentrations in water in reduced conditions. A second experiment highlighted that the reactive materials limited Pb, Zn, Ni, Cr, and Cd potentially mobile fractions. These results were in agreement with the model simulations that showed that even sand can retain metals in sediment for one year in presence of only diffusion phenomenon (Dixon and Knox, 2012). However, in the long-term the metal binding capacity of reactive materials became crucial and they considerably retarded the release of pollutants compared with sand (Knox, Paller and Roberts, 2012b).

Xiong *et al*. (2018) showed that natural zeolite can have strong sorption capacities for different metal ions. The Langmuir and Freundlich models were used for isothermal adsorption kinetics fitting. The kinetic approach used to model the experimental results revealed that the adsorption capacity of natural zeolite was in order: $Pb^{2+} > NH^{+4} > Mn^{2+} > Cd^{2+} > Zn^{2+} > PO_4^{-3}$. In the study carried out by Ting *et al*. (2020), three caps with different AC and clay percentages were applied to Hg-contaminated sediment with an initial concentration of 76.0 ± 2.6 mg Hg/kg. The two caps with AC + bentonite and AC + kaolin effectively reduced both total mercury (THg) and methylmercury (MeHg) concentrations at the end of the observation period (75 days). In contrast, the capping system with AC + montmorillonite did not show a substantial reduction of the Hg-derived compounds in the overlying water, probably because of the unstable suspension of montmorillonite. Furthermore, under occasional turbation events, the caps exhibited higher resistance to Hg breakthrough. Limestone (LS), steel slag (SS) and AC were investigated in the experiments by Park *et al.* (2018), where a horizontal flow tank was filled with 1 cm of cap, together with contaminated sediments and seawater and monitored over 32 days. AC capping binded via electrostatic interaction with Cr, Cu, Ni, and Zn from the sediments stopping their spreading to the overlying water. In the case of SS, As, Cr, Cu, Ni, Pb, and Cd elution from the contaminated sediments were effectively reduced in the overlying marine water, mainly via sorption and surface precipitation mechanisms in the SS layer. For LS, instead, only Cu and Pb release appeared to be treated with high hydrolysis reaction constants. Sequential extraction results also showed that LS was effective to stabilize Zn, whereas AC was more suitable for Cd and Pb. In line with Park's study, Taneex *et al.* (2018) investigated low cost industrial by-products (i.e., bauxaline and SS) as capping amendments through a bench scale laboratory experiment. Capping with SS, bauxaline, and their mixture completely prevent Cd, Zn, and As propagation. However, in presence of Cr, only SS effectively blocked its release. Without the capping system, all the contaminants were released with different kinetics depending on mineral oxidation, dissolution and leaching. The kinetic approach used to simulate the experimental data for Cr and As highlighted that As and Cr characteristic times for release from the sediment were of the same order of magnitude. The Cr and As modeled release time was about 6 days; instead, in the scenario with capping, the Cr capture time was equal to 57 days for SS and 7 days for bauxaline. Therefore, SS performed better since bauxaline became a source of Cr, releasing it quickly (release time 1 day).

Many researchers have proven that biochar has a strong capacity to adsorb metals both from water and sediments, thanks to BC organic components and surface physical adsorption or precipitation (Tong *et al.*, 2011). Zhang *et al*. (2018) investigated the efficacy of *Phyllostachys pubescens* (PP) biochar with Cd, Cr, Cu, Ni, Pb, and Zn. Except for Cr, the results revealed that biochar considerably reduced the bioavailable fraction of metals through the measurement of the diffusive gradients in thin film in porewater. The case with sediment amended with 15% w/w biochar reduced the acid-soluble fraction for Cu, Pb, Ni, Zn, Cd, and Cr, of 79.71%, 73.20%, 54.86%, 49.75%, 31.16%, and 0.99%, respectively. Additionally, bioaccumulation tests demonstrated that, with the exception of Cr, metal accumulation in *Limnodrilus hoffmeisteri* was reduced by 18.45%-59.15% in presence of BC amended sediment. The lethality rate using biochar at 15% was reduced by 37.5%, 18.1% and 36.3% for *Chlorella vulgaris, Daphnia magna* and luminescent bacteria *Vibrio qinghaiensis*, respectively. The effectiveness of BC was also investigated by Wang *et al.* (2019) to control long-term Hg release in fluvial settings characterized by numerous changes in hydro-geochemical conditions. BC successfully minimized the release of dissolved Hg derived from riverbank sediments even in presence of rapid flow conditions. It is noteworthy that BC did not promote methylation reactions since moderately low concentrations of MeHg were observed in water and solid matrices at the end of the experiment.

As an inexpensive reducing agent, ZVI has been applied in lab-scale experiments to reduce inorganic metals like chromium (Gheju, 2011; Jamieson-Hanes *et al.*, 2014), arsenic (Sun *et al.*, 2006; Singh *et al.*, 2015), and others. However, the study by Todaro *et al.* (2021) assessed the sustainability of several ISC amendments employed in lab-scale experiments. Results demonstrated that ZVI has the highest environmental impacts, closely followed by AC and OC. The overall ratings of alternatives indicated that capping designs with AC appeared more sustainable.

Lastly, metals bound to amendments are sensitive to changes in pH and salinity, and they can be released when the pH changes (Park *et al.*, 2019). Therefore, site-specific conditions need to be taken into account carefully.

Table 4 highlights the main points of ISC application for inorganic compounds with information about ISC amendments and design, target compounds investigated, and the main remarks of the investigation.

Table 4. Reactive amendments employed in ISC in case of inorganic contamination.

Targeted compounds investigated	**ISC material and design**	**Scale**	**Intervention duration and site**	**Phenomena modeled and/or investigated**	**Main remarks**	**Reference**
Cr, Co, Ni, Pb	• Rock phosphate from North Carolina (NCA) • Organoclay from Biomin Inc. (OCB; San Antonio, TX) • Natural zeolite, clinoptilolite (ZC) • Sand coated with the biopolymer xanthan cross-linked with chitosan and calcium chloride (XCc)	Bench-scale (0.05 L tubes)	180 days, Anacostia River near Washington, DC, US	• Diffusion	• Long-term modeling revealed that metal release in water was much lower in all columns with capping layers. This did not happen in sandy cap (good performance only in the first year). • The caps prevented metal diffusive transport from contaminated sediment.	(Knox *et al.*, 2014)
Cd, Zn, As, Cr	• Bauxaline 1-1.5 cm thick • SS 1-1.5 cm thick • 50% bauxaline and 50% steel slag 1-1.5 cm thick	Bench-scale (40x20x25 cm aquarium)	90 days, Vallauris Golfe-Juan, French Riviera coast, France	• Sorption/Desorption	• A kinetic model to simulate As and Cr release, in presence and absence of capping. • Cr and As release times from the sediment were about 6 days. • With capping, Cr capture time was found equal to 57 days for SS, and 7 days for bauxaline. • Despite its high capture time, SS showed the best performance (only SS blocked Cr release).	(Taneez *et al.*, 2018)
Pb, Mn, Zn, Cd	• Zeolite 0.5-2 cm thick	Bench-scale (1250 cm^2 x 20 cm column)	30 days, Guangdong Province, China	• Sorption	• No noteworthy difference in the efficacy of heavy metals inhibition was recorded among the various capping thicknesses. • The highest efficiency was reached with 2 cm thickness, where Cd^{2+}, Pb^{2+}, Mn^{2+}, and Zn^{2+} inhibition rates were 35.7%, 85.7%, 65.6%, and 57.8%, respectively.	(Xiong *et al.*, 2018)
Cd, Cr, Cu, Ni. Pb, Zn	• Biochar	Bench-scale (1 L flask)	15 days, River of southern Hebei Province, China	• Sorption	• Biochar significantly reduced the bioavailable fraction of metals (with the exception of Cr) by diffusive gradients. • The reductions in the acid-soluble fraction for Cu, Pb, Ni, Zn, Cd, and Cr	(Zhang *et al.*, 2018)

					resulted in 79.71%, 73.20%, 54.86%, 49.75%, 31.16%, and 0.99%, respectively, for the amended sediment treated with 15% w/w biochar.	
As, Cd, Cr, Cu, Ni, Pb, Zn	• LS 1 cm thick • SS 1 cm thick • AC 1 cm thick	Bench-scale (flat flow tank)	32 days, a coastal dock in Inchon, Korea	• Sorption	• SS reduced As, Cr, Cu, Ni, Pb, and particularly Cd elutions from the contaminated sediments to the seawater. • LS was effectiveonly with Cu and Pb with high hydrolysis reaction constants. • AC interrupted Cr, Cu, Ni, and particularly Zn release from the contaminated sediments by binding with the metals via electrostatic interaction.	(Park *et al.*, 2018)
Hg	• Hardwood biochar	Bench-scale (acrylic column)	7 days, South River in the Shenandoah Valley in Virginia, US	• Sorption	• Great potential for controlling long-term Hg release and transport in fluvial environments characterized by frequent changes of hydro-geochemical conditions.	(Wang *et al.*, 2019)
Hg	• AC (3%) + bentonite (3%) 1.5 cm thick • AC (3%) + kaolin (3%) 1.5 cm thick • AC (3%) + montmorillonite (3%) 1.5 cm thick	Bench-scale (cell with a canal segment (50 x 10 x 10 cm; V = 5.0 ± 0.2 L) on top of a sediment tank segment (40 x 10 x 10 cm; V = 4.0 ± 0.1 L).	75 days, estuary pond within a former chloralkali plant, the An-Shun Plant of China Petrochemical Development Corp., Tainan, Taiwan	• Sorption • Advection • Bioturbation	• The caps with AC (3%) + bentonite (3%) and AC (3%) + kaolin (3%) were effective in reducing total mercury (THg) and methylmercury (MeHg) concentrations in water by 75–95% and 64–98%, respectively. • The AC (3%) + montmorillonite (3%) cap did not reduce significantly THg and MeHg in the overlying water, probably because of the unstable suspension property of montmorillonite.	(Ting *et al.*, 2020)
As, Cd, Cr, Cu, Hg, Ni, Pb, Zn	• AC RCM 3 kg/m^2 • OC RCM 3 kg/m^2 • OC+AC RCMs 3 kg/m^2 • Granular Provect-CH4 2.5% 3 cm thick and granular PAC 5% 5 cm thick	Bench-scale (700 cm^2 x 150 cm column)	20 days, Mar Piccolo of Taranto, Italy	• Sorption • Degradation • Advection	• Capping with AC and ZVI was the best solution for chemical isolation (efficiency of 95%) and degradation aid (efficiency of 40%).	(Todaro *et al.*, 2021)

	• Granular Provect-CH4 2.5% 4 cm thick and granular ZVI 5% 3 cm thick • Granular Provect-CH4 2.5% 3 cm thick, granular ZVI 5% 4 cm thick and granular PAC 5% 5 cm thick				• ZVI had the highest environmental impacts, closely followed by AC and OC. • The sustainability assessment highlighted that both the RCM with 3 kg/m^2 AC and the 5% AC granular amendment reduced the concentrations in water of Cu, Hg, Pb, Zn.	

5. Discussion

The processes involved in the ISC technology are strictly dependent on the type of amendment adopted and nature of contaminants present in the site. However, many more aspects need to be taken into account in a remediation project. Firstly, capping with clean sediment is expected to create a less prominent ecological impact, but this should be weighed against its partial efficiency in reducing contamination in a remediation project. A good balance among particle size, amendment type, benthic health, and global efficiency needs to be reached for an ecological and efficient remediation.

One of the main ISC drawbacks almost never investigated in literature is that the reactive layer risks to become saturated over time, which can be faster, if site-specific conditions vary. Once saturated, the capping system needs to be replaced, but this is not always a feasible activity in the field. The contaminated capping layer must be carefully dealt with to avoid that pollutants are desorbed into the environment (Zhang *et al.*, 2016). For this reason, numerical modeling can be essential to promptly predict cap replacement and ISC amendment effectiveness in the long-term. However, modeling is very sensitive and most studies in literature overly simplify real phenomena in the environment. For instance, the competition behavior among diverse pollutant sorption processes is rarely investigated in an ISC system (Bortone *et al.*, 2020). Additionally, the solid particle porosity and bulk density are usually assumed constant in the layer. However, in the study by Shen *et al.* (2018) they were calculated as linear sums of the single solid components properties weighted by the volumetric fraction. The latter can be considered as a constant parameter in the i-th layer except near the surface in the bioturbation zone, where it should be considered as a continuous temporal-spatial variable (Shen *et al.*, 2018).

Another aspect often neglected in the studies in literature is that contaminated sediment in reality is frequently underlain by uncontaminated sediment. The latter could become a sink of contaminants coming from the contaminated matrix and affect the contaminant transport process, misjudging the real transport rate and leading to incorrect design. Therefore, groundwater flow direction should be taken into consideration during cap design and impermeable caps should be employed to avoid downward flow through contaminated sediment (Calace et al., 2018).

It is necessary to investigate the economic aspects and possible alternatives to optimize the whole remediation performance (Cho *et al.*, 2012). Amendments can be contained in a mat or applied in bulk on the top of sediment surface or even mixed with the sediment (US EPA, 2013). Consequently, the equipment required for ISC installation can be different and potentially generating stressing events to benthic communities but also diverse handling costs for the cap placement (US EPA, 2013). The effectiveness of reactive materials such as AC and OC has been studied in a few field applications, while other amendments, such as ZVI, BC, and biopolymers are still in the bench-scale phase. Unfortunately, complete information about amendment potential side effects are still missing. With the exception of AC and ZVI, eco-toxicity data are rare or unavailable (Todaro, 2019). Caps may be colonized by microorganisms potentially becoming a zone of pollutant biotransformation with effects on benthic biogeochemical fluxes, microbial processes, and meiofauna abundances (Bonaglia et al., 2019). Therefore, exposure to contamination by benthos might still be significant (Zhang et al., 2016). Furthermore, there is still lack of knowledge of their efficiency and suitability in the long-term (>10 y). For instance, Bonaglia *et al.* (2019) developed a 4-week experiment investigating the ecological effects on benthic microbial activity and meiofauna communities (e.g., *Nematoda, Harpacticoida, Turbellaria, Rotifera*, etc.) of a bituminous coal-based (BCB) and a coconut shell-derived (CSD) powdered ACs and one control material installed on contaminated sediments. Their results showed that the total benthic metabolism assessed by sediment oxygen uptake was reduced by 30 and 43% in presence of BCB AC and CSD AC, respectively. Meiofauna decreased by about 60–62% during the AC treatments. This was likely due to an increase in porewater pH and to the sequestration of natural, sedimentary organic matter by AC particles. This was in line with the outcomes of Cornelissen *et al*. (2011).

This review highlighted a limited numbers of studies where experimental and modeling activities were developed synergistically. Most of the studies were only experimentally-based, without giving any information about the long-term effectiveness of the ISC intervention. At first, it is necessary to develop a Conceptual Site Model (CSM) to understand the type and the level of contamination in natural matrices, how contaminants can migrate from sediment and move through the cap, and what types of phenomena are involved in the system. Therefore, every lab-scale experimental activity should be followed by short and long-term numerical simulations able to predict pollutant fate and transport in the system and a subsequent field-scale application, as in the study by Hawkins *et al*. (2011).

Table 5 shows a comparison of the main studies using both experimental and modelling approaches in literature. It is noteworthy that all simulations considered a 1D system. Unfortunately, the goodness of fitting between the observed and the modeled values was not always computed. Bortone *et al.* (2020) chose the mean absolute errors

(MAE) as the best fitting descriptor; in addition, Silvani *et al.* (2017) evaluated the root mean squared error (RMSE) and the correlation coefficient (ρ). However, ISC-based numerical models in literature are often verified analytically (Lampert and Reible, 2009; Li and Cleall, 2010; Qiu *et al.*, 2021).

The ISC treatment efficiency in **Table 5** is higher than 90% in most of the literature cases analyzed, underlining the long-term effectiveness of the ISC system.

Table 5. Matrix of experimentations and modelling in literature.

N.	Modeling											Experimentation		Reference
	Advection	**Diffusion**	**Degradation**	**(De) Sorption**	**Bioturbation**	**Consolidation**	**1/2/3D**	**Capping mono/ multilayer**	**Modeling Time**	**Verification**	**% Treatment efficiency ***	**Contamination**	**Scale**	
1	X	X		X			1D	Mono	10,000 years	Experimental	99.9%	Mixed	Bench	(Knox *et al.*, 2007)
2	X	X	X	X	X		1D	Multi		Analytical			No	Lampert and Reible, 2009
3	X	X		X			1D	Mono	100 years	Experimental	99.9%	Mixed	Bench	(Yin *et al.*, 2010)
4	X	X		X		X	1D/2D	Multi		Experimenta	96.8%	Mixed	Pilot	Hawkins 2011
5		X		X			1D	Mono	10,000	Analytical	75%	Organic (trichloropropane)	No	Li and Cleall 20111
6	X	X		X		X	1D	Mono	20 years	Experimental	91.7%	Organic (PCBs)	Bench	(Meric *et al.*, 2013)
7		X		X			1D	Mixed	1 year	Experimental	98%	Organic (PAHs)	Bench	(Choi *et al.*, 2013)
8	X	X		X	X		1D	Multi	1,000 years	Experimental	95%	Organic (PCBs)	Bench	(Azhar, 2015)
9		X		X			1D	Mono	20 years	Experimental	69%, 56% and 99% (AC, OC, BC, respectively)	Organic (PAHs)	Bench	(Silvani *et al.*, 2017)
10	X	X		X			1D	Mono	1 year	Experimental	99%	Organic (PAHs)	Bench	(Bortone *et al.*, 2020)
11		X		X			1D	Mono	100	Analytical	91.1%	Inorganic (Pb)	No	(Qiu *et al.*, 2021)

* Highest value obtained calculated as: %Eff $= 1 - C_{with\ cap}/C_{no\ cap}$ (Eek *et al.*, 2007) with C = concentration in water.

6. Conclusions

This contribution presented a comprehensive review of ISC applications, by investigating key aspects such as reliability, applicability, long-term effectiveness, and numerical simulations applied to ISC technology, also comparing reactive and physical ISCs. Based on the current technologies, AC and OC resulted in the most investigated amendments for sediment contaminated with organics, whereas biochar, clay minerals, and industrial-by products were more employed in presence of sediment contaminated by metal(loids). However, there is no best ISC system in absolute terms, since its technological performance depends on the environment conditions, which can vary in pH, salinity, hydraulic currents, and other characteristics. A systematic review of the research findings showed that only a few experimental investigations included a long-term modeling phase to predict ISC long-term efficiency, which often was based on a simplified conceptual model of the real phenomena involved. Most of the numerical models included simplified transport equations often considering only diffusion and adsorption as the main phenomena. Apart from the type of materials potentially useful in reactive capping or mixing, little is known regarding their long-term application on field-scale and their ability to sequester contaminants over time in presence of site-specific tidal forces and currents. Additionally, a good fitting between experimental and modeled data was not always computed. Significant gaps still remain between the current understanding of the ISC technology and the level of engineering know-how necessary for real-scale implementations. As a direct consequence, costs are not always clear from the beginning as the full range of drawbacks. In order to provide a methodological contribution to the addressed topic, more effort needs to be directed to the design phase of an entire experiment. A standardized procedure can certainly help, involving experimental and modelling activities both at lab and field level, with a clear idea of the main parameters to be measured or assumed. Future research should therefore focus on the role of site-parameters evaluation in modeling hydrodynamic phenomena and solute dispersion, but also bioturbation, degradation processes, solute kinetics, and multiple contaminant competition, thus defining the boundary conditions of the investigated site, the starting point for proper experimentation at laboratory scale. Furthermore, new research at a lab scale should aim at studying the long-term effects of the capping investigated. When proposing new amendments, the use of preliminary life-cycle assessments can be helpful before setting up laboratory-scale research, providing information on the feasibility of the proposal.

CRediT author statement

Claudia Labianca: Data curation, Conceptualization, Investigation, Formal analysis, Methodology, Visualization, Writing - Original Draft; **Sabino De Gisi**: Methodology, Resources, Validation, Visualization, Writing - Original Draft, Writing - review & editing; **Francesco Todaro**: Resources, Writing - review & editing; **Michele Notarnicola**: Conceptualization, Project administration, Resources, Writing - review & editing; **Imma Bortone:** Conceptualization, Methodology, Project administration, Resources, Supervision, Validation, Writing - Review & Editing.

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