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Review article

A critical review of the remediation of PAH-polluted marine sediments: current knowledge and future perspectives

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ABSTRACT

PAHs are largely spread in the aquatic environment, and the drawbacks of conventional remediation techniques as well as the expenditures for alternative disposal of polluted sediments lead to seek more effective, environmentally-friendly and sustainable approaches. Therefore, the present review shows a critical overview of the literature evaluated with VOSviewer, focusing on the problem of PAH-contaminated marine sediments and the knowledge of available remediation processes to shed light on what research and technology lack. This review supplies specific information about the key factors affecting biological, physical-chemical and thermal remediation techniques, and carefully examines the drawbacks associated with their employment for remediating PAH-polluted marine sediments by showing adequate alternatives. The technologies thoroughly discussed here are biostimulation, bioaugmentation, sediment washing, carbonaceous adsorbent addition and thermal desorption. The environmental and economic impacts associated with the application of the mentioned remediation technologies have been also taken into account. Finally, this review examines new research directions by showing future recommendations.

Contents

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds consisting of at least two benzene rings settled in linear, angular, or cluster chains (Akinpelu et al., 2019). PAHs can exhibit toxic, genotoxic, teratogenic, mutagenic and carcinogenic effects, and, therefore, the United States Environmental Protection Agency (US EPA) identified 28 PAHs as priority pollutants in 2008 (Mihankhah et al., 2020).

PAHs are mainly produced by pyrogenic and petrogenic processes through natural and artificial sources (e.g. biomass heating systems, oil spills, forest fires) (Fig. 1) (Ferrara et al., 2020). Pyrogenic PAHs (e.g. pyrene, benzo[a]pyrene) are formed by incomplete combustion

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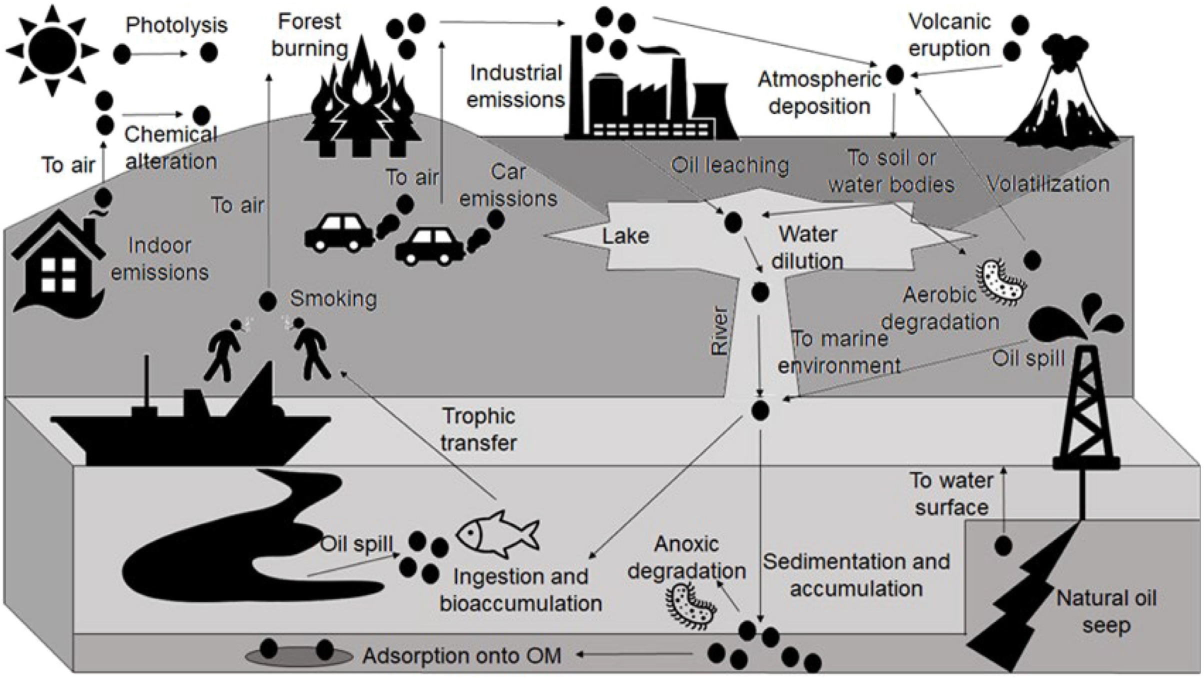

Fig. 1. The possible pathways of polycyclic aromatic hydrocarbons (•) in the environment. OM = organic matter.

or pyrolysis of the organic matter at temperatures ranging from 350 to 1200 °C (Hussain et al., 2018). Petrogenic PAHs (e.g. phenanthrene, anthracene) are generated by the decomposition of the organic matter at low temperatures (i.e. 100–150 °C) and geological formation times (Abdel-Shafy and Mansour, 2016). The process that generates a specific PAH consequently affects its physical-chemical properties.

In general, PAHs are characterized by low solubility in water (i.e. 0.001–30 mg L^{-1}), high hydrophobicity (i.e. logarithm of octanol-water constant of 3.37–6.50), high boiling (i.e. 100–500 °C) and melting points (i.e. 100–300 °C), and low vapor pressure (i.e. $6.4 \cdot 10^{-12}$ –0.085 mm Hg) and volatility (i.e. Henry's law constant of $7.30 \cdot 10^{-2}$–$3.00 \cdot 10^{-6}$) (Ghosal et al., 2016; Wu et al., 2019a). These properties are more pronounced with the increase of PAH molecular weight. Therefore, PAHs can be listed as low molecular weight (LMW) and high molecular weight (HMW) compounds when formed by 2 or 3 benzene rings (e.g. naphthalene, phenanthrene) and more than 3 benzene rings (e.g. benzo[a]pyrene) (Caniani et al., 2021), respectively. The abovementioned parameters are important for understanding the fate and distribution of PAHs in the environment (Fig. 1).

Indeed, after an intricate PAH transport due to physical-chemical characteristics (e.g. hydrophobicity) and environmental conditions (e.g. geomorphologic properties), naturally and artificially generated PAHs can be adsorbed onto the organic matter and subsequently combined with sediments (Fig. 1) (El-Shahawi et al., 2010). Afterwards, sediment-adsorbed PAHs can be driven by external factors such as water flow and bioaccumulation in fishes (Fig. 1), thus posing a potential risk to human health (Wu et al., 2019a,b).

In general, the concentration of $\sum$ 16 PAHs in sediments can vary greatly and be in a range from approximately 0.1 to 300 mg kg^{-1} (Wu et al., 2019a). However, according to Merhaby et al. (2019), Mediterranean sediments exhibit a higher PAH concentration (i.e. up to about 1700 mg kg^{-1}) in calm zones such as harbors compared to those shown in dynamic environments such as river bodies (i.e. up to about 600 mg kg^{-1}). Since the dredging of harbor areas is highly required every year (i.e. up to 200 Mm^3 in Europe, SedNet) to maintain a constant level of the seabed and allow navigation (Ferrans et al., 2021), subsequent issues for the sediment management (e.g. landfilling, coastal nourishment, reuse) are posed due to the PAH pollution of dredged sediments (Sprovieri et al., 2007). Therefore, the study of innovative and proper technologies for the remediation of PAH-contaminated marine sediments is of vital importance to the scientific community, industrial stakeholders and public authorities.

The academic community has recently focused its attention on the marine sediment issue (Fig. 2). Perelo (2010) first reviewed the bioremediation techniques for removing organic contaminants from aquatic sediments. Also, Hilber and Bucheli (2010) focused their review article on the addition of activated carbon to remediate polluted sediments and soils. Dell'Anno et al. (2018) reviewed the use of biosurfactants to remediate marine sediments contaminated by toxic metals and organic pollutants. Zhang et al. (2021) lately summarized ex-situ sediment remediation technologies aimed at the removal of heavy metals from polluted sediments. Dai et al. (2022) mainly overviewed the characteristics of PAHs and their distribution in soils and coastal sediments. Labianca et al. (2022) recently focused their review on the application of in-situ capping for the adsorption of both inorganic and organic contaminants in marine sediments. Notwithstanding, previous reviews have dedicated minor attention to the main remediation techniques for PAH-contaminated marine sediments (Fig. 2). Indeed, most of these studies only provided a comprehensive overview of a specific technology including soil remediation or removal of a group of contaminants (e.g. inorganic).

This review is aimed at investigating the current state of knowledge on PAH removal through the application of biological, physical-chemical and thermal remediation techniques in polluted sediments. The advantages and disadvantages of the main conventional remediation techniques have been carefully discussed. The effect of key operating parameters on the efficiency of various examined technologies has been emphasized. The emerging technologies as well as the lack in the present literature have been defined by proposing prospects for upcoming research.

2. Semiquantitative assessment

As shown in Fig. 2, more than 100 research papers about the remediation of PAH-contaminated marine sediments were published over the last 10 years. In this study, a semiquantitative assessment was performed using the VOSviewer (https://app.vosviewer.com/) software (Fig. 3). This software can create maps between related contents (Gao

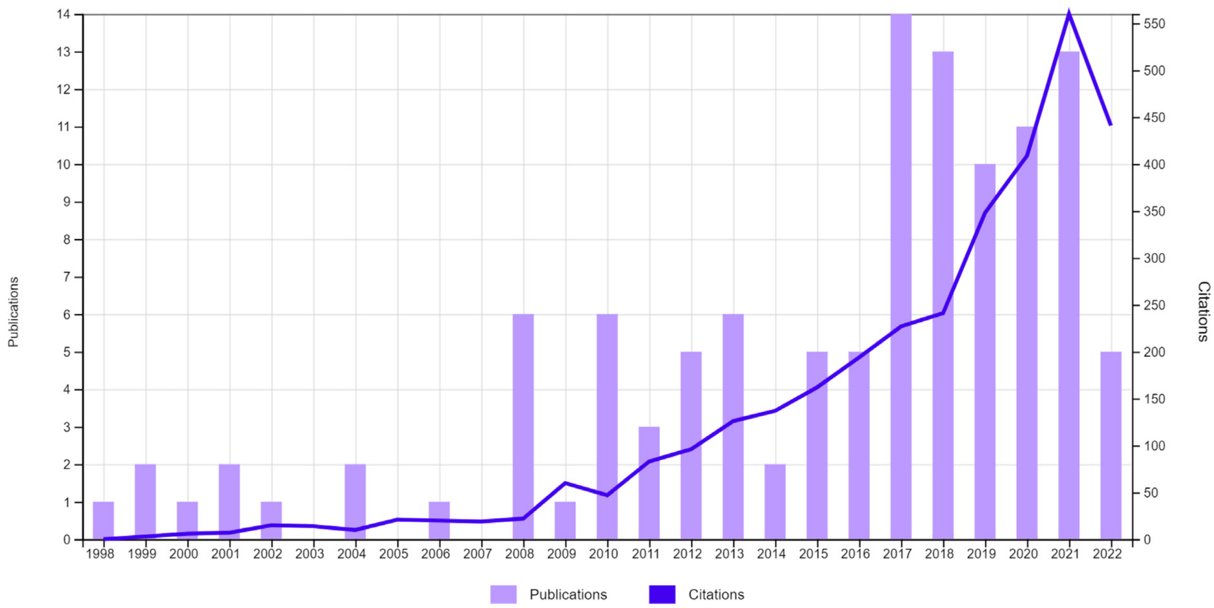

Fig. 2. Publications over time and times cited by analyzing the keywords "remediation", "polycyclic aromatic hydrocarbons" and "marine sediments" through the Web of Science database.

et al., 2022), which were taken from the Web of Science database (https://www.webofscience.com/). The keywords used in the Web of Science for the document search were "remediation" "polycyclic aromatic hydrocarbons" and "marine sediments". The relevant studies were obtained after that a manual selection was carried out to avoid inconsistent references, which can be automatically found after the document search. Afterwards, the documents were exported as a "plain text file" for being imported into VOSviewer, which was computed by selecting the keyword co-occurrence.

The generated map is presented in Fig. 3. The thickness of the lines (Fig. 3) increases with the strength of the correlation among the keywords. For example, the blue-colored lines (Fig. 3) include biodegradation, bacteria, biostimulation, bioaugmentation, crude oil, degradation, and PAHs, which suggest that they are characterized by a high correlation. Fig. 3 also shows that the studies on the remediation of PAH-contaminated marine sediments investigated the bioremediation (e.g. bioaugmentation, biostimulation, anaerobic digestion), physical-chemical techniques (e.g. adsorption, sediment washing), and thermal treatments (e.g. thermal desorption). Therefore, these remediation techniques are discussed in detail in the following sections.

3. Bioremediation

Bioremediation is an environmentally-friendly and inexpensive approach (i.e. 5–300 € m^{-3}, Table 4) and includes a pool of techniques in which microorganisms can be used for the removal of pollutants (e.g. PAHs) from an environmental matrix (Pal et al., 2020; Sinha et al., 2020). When plants are involved, the process is called phytoremediation, in which rhizosphere microorganisms can biodegrade PAHs near the root system (Gabriele et al., 2022). The microbial communities involved during the PAH biodegradation process can be composed of bacteria, fungi or algae, which can directly use PAHs as an energetic substrate or for cellular synthesis and biomass growth (McGenity et al., 2012). Thus, microorganisms can proceed through a PAH biomineralization into simpler compounds (e.g. catechol) by eventually transforming the pollutant into stable substances such as carbon dioxide and water under aerobic conditions, or methane under anaerobic conditions (Haritash and Kaushik, 2009). Microorganisms can also biodegrade a PAH by co-metabolism in which the simultaneous action of other similar compounds (e.g. pyrene and benzo[a]pyrene) or enzymes leads to the decrease of PAH concentrations (Haritash and Kaushik, 2009). When co-metabolism occurs, PAHs are biologically converted into lower molecular weight compounds, which can be subsequently used as a source of carbon and energy by microorganisms. However, the reaction intermediates can be toxic to the involved microbial community and even more reactive in the environment than the original pollutant. For instance, Zhao et al. (2008) reported a decreased biodegradation (i.e. from about 100 to 20%) after raising the initial phenanthrene concentration up to 1000 mg L^{-1} in a batch flask, which can be attributed to the increase of the concentration of decarboxylated metabolites (e.g. 2-naphthol) as intermediates (Mallick and Dutta, 2008).

As said, the biodegradation process can occur under either aerobic, anaerobic or anoxic conditions. The aerobic biodegradation of PAHs is well known in the literature and can present some limitations such as the difficulty to maintain the aerobic conditions due to high oxygen demand for the biological reactions (Lei et al., 2005). On the other hand, the anaerobic degradation of PAHs does not require oxygen as an electron acceptor but can be difficult to be performed due to a higher complexity of the ongoing reactions (Wartell et al., 2021). Otherwise, anoxic conditions can be exploited by using nitrate or sulfate as electron acceptors, being these oxyanions commonly present in PAH-contaminated marine sites (Zhang et al., 2010). The anoxic conditions also occur during phytoremediation (Liu et al., 2014; Verâne et al., 2020), whose application is still limited in marine sediments due to the necessity to employ halophytic species (Huesemann et al., 2009; Lama et al., 2022) or to dilute the salinity with salt-free soils (Paquin et al., 2002). In this sense, the adsorption capacity of *Salicornia fragilis* (*S. fragilis*) should be further evaluated (Meudec et al., 2006).

The efficiency of biodegradation is influenced by the PAH bioavailability towards the microorganisms. Bioavailability mainly depends on the physical-chemical properties of the involved PAH and sediment (e.g. organic substance) and, therefore, bioremediation can be enhanced by biostimulation, bioaugmentation or using surfactants (Perelo, 2010).

3.1. PAH biodegradation with oxygen as electron acceptor

Aerobic bioremediation is a degradation process in which microorganisms can degrade pollutants in the presence of oxygen. PAHs are oxygen-free compounds, and due to their low solubility in water inevitably require molecular oxygen as a terminal electron acceptor

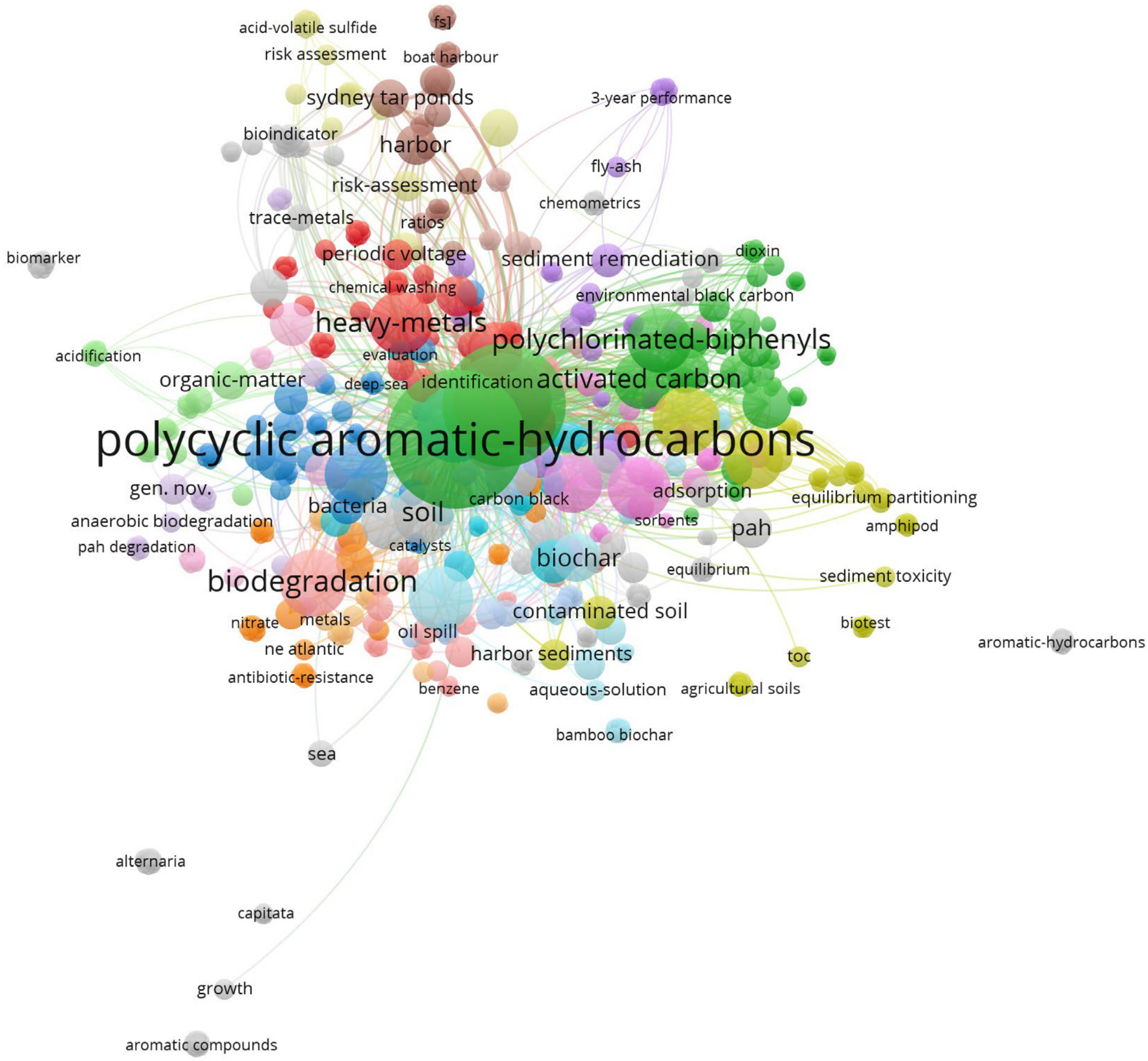

Fig. 3. Semiquantitative assessment of the data collected from the Web of Science database performed via the VOSviewer software. The line thicknesses refer to the correlation between the keywords. The line colors are generated according to the keyword colors. The color similarity of the keywords refers to their correlation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(TEA) to be metabolized by microorganisms (Parmar et al., 2014). Thus, aerobic bacterial strains through the oxygenase can degrade PAHs into simpler molecules using enzymes (e.g. monooxygenase and dioxygenase), which are formed by polypeptide chains and can catalyze the entrance of O_2 into PAHs (Tao et al., 2007).

The catabolic degradation of naphthalene, taking naphthalene as a LMW PAH to understand the degradation pathway that can occur via aerobic bacteria, is also a part of the degradation process of other PAHs such as phenanthrene. Several bacterial strains belonging to genera such as *Pseudomonas, Rhodococcus, Sphingomonas*, and *Streptomyces* can use naphthalene as an energy source (Seo et al., 2009). During the aerobic naphthalene degradation, the monooxygenase enzyme introduces only one oxygen atom into the organic pollutant, whereas the second oxygen atom is reduced to water via a reducing agent such as $NADH_2$ (Pérez-García et al., 2022). On the contrary, dioxygenase introduces two oxygen atoms in the hydroxyl form into the PAH molecule (Pérez-García et al., 2022). Therefore, the first step of the aerobic bacterial degradation of PAHs is the hydroxylation of an aromatic ring through the activation of dioxygenase enzymes by producing a cis-dihydrodiol compound (Ullrich and Hofrichter, 2007). Afterwards, a PAH is re-aromatized with the dehydrogenase enzyme and subsequently the aromatic ring is cleavaged by adding further O_2 with the dioxygenase (Cerniglia, 1992; Mallick et al., 2011). A catechol is finally produced by the cleavage of salicylate, which is obtained by the meta-cleavage of 1,2-dihydroxynaphthalene (Waigi et al., 2015) by leading to the pyruvate and acetyl CoA production (Shon et al., 2020).

For HMW compounds (e.g. benzo[a]pyrene, Fig. 4), the degradation pathway is more complex than that commonly reported for naphthalene. In brief, benzo[a]pyrene can be aerobically converted into benzo[a]pyrene-cis-9,10-dihydrodiol in the presence of *Beijerinckia* B-8362, as a result of dioxygenase at 9 and 10 positions (Nzila and Musa, 2020), which can lead to a substituted pyrene upon the ring cleavage (Fig. 4). Benzo[a]pyrene-cis-7,8-dihydrodiol can be alternatively produced by *Mycobacterium* RJGII-135(McLellan et al., 2002), implying the action of dioxygenase at 7 and 8 positions, which can be also coupled with the formation of a substituted pyrene (Fig. 4). In addition, benzo[a]pyrene-cis-4,5-, -11,12- and trans-11,12-dihydrodiol can be generated as benzo[a]pyrene metabolites by *Mycobacterium vanbaalenii* PYR-1(Moody et al., 2004). 4,5-dihydroxy-benzo[a]pyrene can be subsequently obtained from the hydrogenation at C4 and C5 of benzo[a]pyrene, which can lead to chrysene formation (Fig. 4) and follow the abovementioned naphthalene route due to chrysene hydrogenation (Nzila and Musa, 2020).

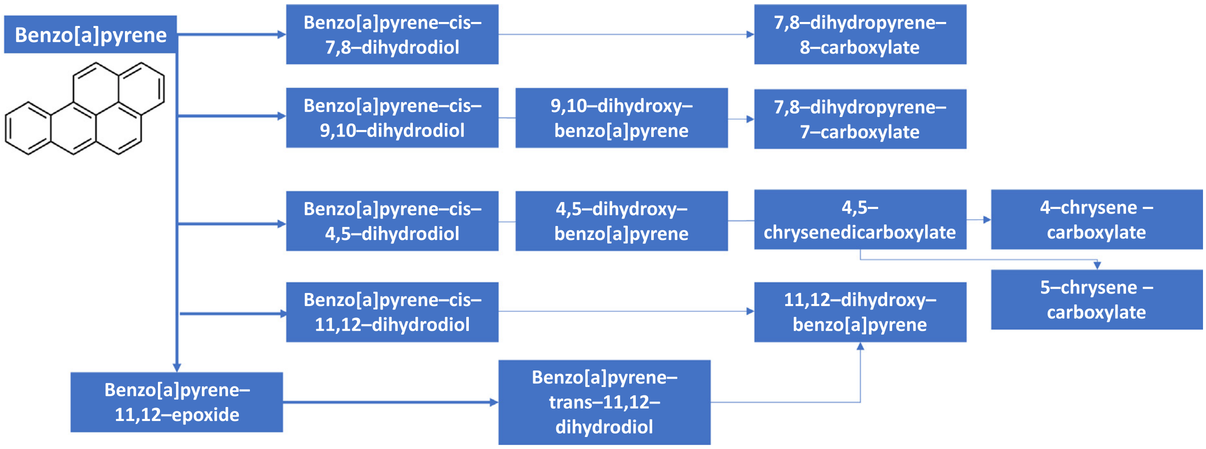

Fig. 4. The possible biodegradation routes (arrows) of benzo[a]pyrene under aerobic conditions.

Table 1
Summary of studies reporting the aerobic bioremediation of polycyclic aromatic hydrocarbon (PAH)-contaminated sediments. LMW = Low molecular weight; HMW = High molecular weight.

Aerobic bioremediation	Investigated PAHs	Removal	References
Composting	ΣPAHs	1%–60% after 120 d	Mamindy-Pajany et al. (2010)
Co-composting with green waste	ΣPAHs	26%–57% after 180 d	Mattei et al. (2016)
Landfarming	ΣPAHs	14%–16% after 30 d	Adams and Guzmán-Osorio (2008)
Landfarming of phytoremediated sediments	ΣPAHs	>99% after 90 d	Macci et al. (2021)
Biostimulation and bioaugmentation with bacterial consortium	Fluorene, phenanthrene and pyrene	97% with biostimulation after 30 d, bioaugmentation was not significantly effective	Yu et al. (2005)
Biostimulation with the addition of salt mineral medium	Phenanthrene, fluoranthene and pyrene	96% after 30 d	Louati et al. (2015)
Biostimulation with inorganic nutrients, bioaugmentation with the addition of fungi and microbial fuel cell-based strategy	ΣPAHs	At least 60% after 30 d, >90% with biostimulation	Dell'Anno et al. (2020)
Slurry bioreactor	Phenanthrene and fluoranthene	57%–63% after 42 d	Wang et al. (2019)
Biostimulated slurry bioreactor	ΣPAHs	>93% after 56 d	Chikere et al. (2012)

The main processes and results referring to aerobic bioremediation are summarized in Table 1. The bioremediation techniques generally employed for PAH-contaminated sediments are landfarming and composting (Tables 1 and 4), which are sustainable and economic technologies (Kumar et al., 2018). Landfarming (Table 1) is performed with periodic sediment turning over for the removal of PAHs, and the microbial activity can be also stimulated with the addition of extra substances such as urea and phosphate, plants or allochthones bacteria (Harmsen et al., 2007). For instance, Adams and Guzmán-Osorio (2008) reported a PAH degradation only of 14%–16% after 30 days of landfarming (Table 1), whereas Macci et al. (2021) achieved almost a complete PAH removal with landfarming of phytoremediated sediments (Table 1) due to an improved bioavailability of pollutants. Similarly, composting is performed with air injection to allow the degradation of $\sum$ PAHs with a reduction of 1%–60% after 6 months (Table 1) (Mamindy-Pajany et al., 2010; Mattei et al., 2016). Also in this case, when composting lacks a sufficient presence of nutrients, biostimulation can be employed to improve and speed up biodegradation (Kuppusamy et al., 2017). The use of slurry bioreactors for treating PAH-polluted sediments is still limited (Table 1) due to the high amount of water that should be introduced into the reactor (Wang et al., 2019) to reach similar efficiencies of the abovementioned techniques (i.e. about 60%, Table 1).

Thus, the bioaugmentation and biostimulation processes (Table 1 and S2) can represent a booster for the well-established bioremediation processes (Dell'Anno et al., 2020). Bioaugmentation (Table 1 and S2) is based on the addition of further microbial cultures, previously enriched in laboratory, with a high capability to biodegrade a specific compound (Herrero and Stuckey, 2015). This technique can be used when the indigenous microbial community is poor and cannot biodegrade PAHs. Although the biodegradation process can be improved, the inoculated microorganisms may compete with the existing microbial community and inevitably decrease the process efficiency (Ławniczak et al., 2020). Also, Wu et al. (2019b) showed that the introduction of an allochthonous species (i.e. *Pseudomonas*) can lead to a decrease in microbial biodiversity since the surviving inoculum can increase in number and become predominant compared to other microorganisms. Hence, better biodegradation results can be achieved with a heterogeneous microbial community (Wu et al., 2019b). Moreover, the expansion from lab-scale, to pilot- and full-scale should be gradual to avoid drawbacks (Ma et al., 2022).

A further technique to be considered is biostimulation (Table 1 and S2), in which nutrients or amendments are added to the sediment to stimulate the activity of the existing microbial community (Hamdan and Salam, 2020). In such way, the carbon to nitrogen to phosphorous (C:N:P) molar ratio is balanced (e.g. 100:10:1) by adding fertilizers or organic amendments rich in nitrogen such as sewage sludge (Ortega et al., 2018). A drawback of this process can be that the organic substrate is competitive compared to PAHs (Tyagi et al., 2011). In this case, biostimulation can be combined with bioaugmentation to further enhance biodegradation efficiency (Ławniczak et al., 2020). Haleyur et al. (2019) recently showed a high PAH removal (i.e. by approximately 94%) after 30 days of combined biostimulation and bioaugmentation in a soil initially contaminated by 1.5 g of PAHs kg^{-1} of total solids. On

Table 2
Summary of works reporting the anoxic and anaerobic degradation of polycyclic aromatic hydrocarbons (PAHs). OFMSW = organic fraction of municipal solid waste.

Conditions	Tested PAH	PAH degradation	References
Nitrate-reducing	Naphthalene	Up to 93% after 25 d	Dou et al. (2009)
	Phenanthrene	Up to 68% after 42 d using sewage sludge as inoculum	Bianco et al. (2020a)
	Phenanthrene, naphthalene	17%–96% after 30 d	Rockne and Strand (2001)
	Benzo[a]pyrene	84% after 10 d	Qin et al. (2017)
Metal-ion-reducing[a]	Phenanthrene, fluorene, fluoranthene and pyrene,	No significant effect	Li et al. (2010)
Sulfate-reducing	Fluorene and phenanthrene	65%–88% after 21 d	Tsai et al. (2009)
Methanogenesis	Naphthalene and benzo[a]pyrene	52%–85% after 90 d	Ferraro et al. (2021)
	Fluorene, Phenanthrene, Anthracene, Fluoranthene and Pyrene	31%–91% after 50 d	Sayara et al. (2011)
	Fluorene, phenanthrene and pyrene	>50% after 20 d in presence of glucose or acetate	Ambrosoli et al. (2005)
	Phenanthrene, Anthracene, Fluoranthene and Pyrene	Up to 55% after 120 d of biostimulation with digestate and OFMSW	Bianco et al. (2020b)
	Phenanthrene, anthracene, fluoranthene, pyrene and benzo[a]pyrene	Up to 85% after 126 d with HCO_3^- amendment	Mu et al. (2022)

[a]= includes Fe(III).

the contrary, other studies reported that bioaugmentation cannot significantly contribute to the biodegradation process, with biostimulation alone being sufficient to achieve a satisfying bioremediation efficiency (Haleyur et al., 2019; Sayara et al., 2011). Further research focused on the supplementation of external biosurfactants (e.g. lipopeptide) (Bezza and Nkhalambayausi Chirwa, 2016; Dell'Anno et al., 2018) to improve biostimulation (Table 1 and S2) by increasing PAH bioavailability (Lee et al., 2018). Notwithstanding, it should be considered that such substances can be toxic to microorganisms, become a preferential source of nutrition instead of PAHs, increase bacteriostatic properties and unintentionally mobilize the contaminants in the surrounding areas by enhancing their solubility (Gaur et al., 2021).

3.2. Other terminal electron acceptors

Anoxic and anaerobic biodegradation of PAHs can occur when freely dissolved oxygen is limited or absent (Li et al., 2010). Microorganisms can use various TEAs different from O_2, i.e. SO_4^{2-}, NO_3^-, Fe^{3+}, Mn^{4+} and CO_2 (Table S2) to enhance the conversion of PAHs into lower molecular weight compounds (Zhang et al., 2019). The breakdown of organic compounds releases electrons that can convert ADP to ATP and are accepted by the mentioned TEAs by obtaining water and other molecules (Table S2). Specifically, sulfate is biologically transformed to sulfide by sulfate-reducing bacteria, nitrate is converted to nitrogen gas by denitrifying bacteria, ferric iron and manganese oxides are reduced to Fe^{2+} and Mn^{+3} by metal-ion-reducing bacteria, and carbon dioxide can be reduced to biomethane by hydrogenotrophic methanogens (Table S2) (Dhar et al., 2019; Policastro et al., 2022). The ability to receive electrons is affected by the TEA redox potential, which value is higher for nitrate (i.e. +433 mV) compared to metal ions, sulfate and carbon dioxide (i.e. +200, −200 and −380 mV, respectively) (Table S2) (Nzila, 2018; Sikora et al., 2017). Therefore, the anaerobic biodegradation through the reduction of CO_2 to CH_4 can occur when the other TEAs are limited or absent due to the lowest redox potential (Bianco et al., 2021a; Mu et al., 2022).

The vast majority of PAHs cannot be solubilized in water and tends to accumulate in the sediment layers where anoxic and anaerobic conditions occur. Thus, the organic pollutants can be biodegraded through the ability of the abovementioned microorganisms, which swarm in these environments. This type of treatment is recommended in the presence of high PAH concentrations in order to limit aeration and, thus, to keep the remediation costs low (Gan et al., 2009).

The studies conducted on bioremediation under anoxic and anaerobic conditions are summarized in Table 2. PAH removal is enhanced under nitrate-reducing conditions (i.e. up to 96%, Table 2) than metal-ion- and sulfate-reducing, and methanogenic conditions (i.e. no effects, up to 88 and 91%, respectively). When metal ions are involved, no significant effects on PAH biodegradation can be achieved (Li et al., 2010) likely due to the fact that a high concentration of Fe^{3+} and Mn^{4+} as TEA can inhibit the anaerobic process by inducing toxic effects towards bacterial activity (Li et al., 2011). Likewise, sulfate reduction can exhibit toxicity towards microorganisms due to hydrogen sulfide production (Zhang and Lo, 2015). Therefore, the scientific community is moving forward to the employment of anaerobic digestion (AD) processes (See Section S1 in Supporting Materials) aimed at producing an energetically valuable product (i.e. biomethane) by simultaneously remediating the contaminated site (Bianco et al., 2020b; Oliva et al., 2022, 2021). Strategies to improve AD (Figure S1) can be the bioaugmentation and biostimulation (Table S1), as also reported for aerobic processes (see Section 3.1). However, these techniques have been barely used for the remediation of PAH-contaminated sediments, indicating the need of future studies to shed light on the employment of organic wastes for simultaneous PAH degradation and biogas production.

4. Physical-chemical treatments

4.1. Adsorption

Adsorption consists in the use of an adsorbent (e.g. AC, BC) to entrap an adsorbate (e.g. PAHs) within an intricate porous structure in order to remediate a contaminated matrix (e.g. sediment) (Han et al., 2019). The main advantage is represented by the feasibility of this physical remediation treatment either in-situ or ex-situ (i.e. capping, Table 4). For instance, sediment capping is frequently employed in aquatic environments as an in-situ treatment for decreasing pollutant mobilization (Bortone et al., 2020). In this context, carbonaceous adsorbents (CAs) were extensively used for the adsorption of PAHs due to a high specific surface area (i.e. up to 1300 $m^2\ g^{-1}$) and the presence of functional groups that lead to a great adsorption capacity (Tan et al., 2021). Various studies investigated environmentally-friendly and economic CAs such as AC and BC, which showed their effectiveness in order to decrease bioavailable PAHs (i.e. up to 99%) in polluted sediments (Bianco et al., 2021b; Maletić et al., 2019).

The reduction of pore water PAHs (i.e. bioavailability) can occur through several interaction mechanisms (Fig. 6) between the adsorbate and adsorbent that are generally identified as weak (e.g. hydrophobic interaction, Van der Waals forces) and strong interactions (e.g. π–π, electrostatic interaction) (Gusain et al., 2020). The involved adsorption

Table 3
Summary of sediment and soil washing treatments using organic solvents, surfactants and vegetable oil aimed at PAH desorption from contaminated matrices. TW80 = Tween® 80; TRX = Triton X−100; ΣPAHs = total polycyclic aromatic hydrocarbons.

Extracting agents	Compounds	S/L ratio (w/v)	Time [h]	Removal [%]	References
Mixture of water, acetone and ethyl acetate (10:40:50, v/v/v)	Naphthalene	1:8	1	87	Silva et al. (2005)
Mixture of water and ethanol (1:1, v/v)	Phenanthrene	1:3–1:10	1	>99	Bianco et al. (2022b, 2020a)
Non-ionic surfactant, i.e. TW80	Phenanthrene	1:4–1:20	24	91	Bianco et al. (2022c)
Non-ionic surfactants, i.e. Brij 30, TRX100, Tergitol NP−10, Igepal CA−720 (0.1, 1, 4, 6 and 10%, v/v)	Phenanthrene	2:1	1	61	Chang et al. (2000)
Cationic surfactant, i.e. Dodecylpyridinium bromide				45	
Anionic surfactant, i.e. sodium dodecyl benzenesulfonate	Phenanthrene and pyrene	1:13	24	71–95	Zhao et al. (2010)
Peanut oil	ΣPAHs	1:2	168	81–100	Pannu et al. (2004)
Sunflower oil	ΣPAHs	1:20–1:40	3	52–90	Gong et al. (2005)

interaction (Fig. 6) is affected by CA characteristics, and the occurrence of strong interactions (i.e. chemisorption) can be generally preferred to weak mechanisms (i.e. physisorption) since these adsorbents can show a certain hysteresis index by leading to the desorption of PAHs (Akinpelu et al., 2021). An appropriate pore size allocation can supply mass transfer routes for PAHs and considerable surface areas can provide more active sites for PAH adsorption, thus enhancing the adsorption capacity and rate (Li et al., 2017). Cheng et al. (2019) reported that CAs with micropores, mesopores, and macropores can show higher adsorption rates than CAs with only micropores at parity of adsorption equilibrium capacity. This can be attributed to the fact that a small pore size is characterized by tight passage and long crossing channels, hindering the effective mass transfer and diffusion of PAH molecules through CA (Sharp et al., 2021). Nevertheless, the functional groups of CA surface can fulfill critical roles in the remediation of specific PAHs (Bianco et al., 2022a). The lack of heteroatoms mainly including O, N, H, S, P, and halogens can affect the adsorbents' chemical properties by leading to low H/C and O/C ratios, which suggest high aromatization and carbonization (Stefaniuk and Oleszczuk, 2015). Therefore, proper modifications of the adsorbent (e.g. acidic and alkaline activation, or VFA addition) can be performed to either add or eliminate the surface functional groups (e.g. –OH, –COOH) and to improve PAH adsorption onto CAs (Bianco et al., 2022a; Cashin et al., 2018). In addition to CA properties, the decrease of PAHs in pore water can be affected by CA dosage; PAH properties, initial PAH concentration, pH and ionic strength, natural organic matter, remediation time and mixing (Table S4) (Li et al., 2020). The increase of CA dosage and ionic strength, the presence of LMW PAHs, a low amount of organic matter and pore water PAHs, a prolonged contact time and mechanical mixing (ex-situ) can enhance the adsorption efficiencies (Table S4). However, the employment of a high CA dosage can be costly and interfere with the habitat quality by reducing the dissolved organic carbon, thus affecting living organisms (Jonker et al., 2007). The sediment permeability, water retention capacity and nutrient content can be affected as well (Li et al., 2020). A further drawback can be associated with the recovery of CAs in order to avoid the release of PAHs after CA oxidation or CA swallowing by living organisms. Indeed, conventional separation methods such as sieving and centrifugation are not effective for CA recovery (Rakowska et al., 2014). Therefore, CA modification via magnetite has been recently proposed to improve CA recovery, taking advantage of the adsorbent magnetic properties (Hao et al., 2021). More attention should be paid to this aspect in future studies by evaluating the use of alginate hydrogel spheres for coating BC in marine sediments.

4.2. Sediment washing

Among the physical-chemical treatment, sediment washing (SW) can be used as an ex-situ method to remediate contaminated sediment after dredging operations (Peng et al., 2009). Natural extracting agents such as organic solvents (e.g. ethanol, humic acid, vegetable oil) and synthetic surfactants (e.g. cationic, non-ionic) can be employed during SW to remove the pollutant adhered to the solid matrix via a desorption mechanism (Table 3).

The type of extracting agent employed to increase the aqueous solubility of PAH mainly governs the removal mechanism. In the presence of synthetic surfactants (Fig. 5), several mechanisms can occur: the reduction of interfacial tension, the transfer of PAHs from sediment to the micellar pseudo-aqueous phase, and the solubilization of PAHs inside the hydrophobic core due to micelles (Trellu et al., 2016). Otherwise, PAH desorption from the polluted sediment is governed by the partition coefficient in the solution phase (Thiele-Bruhn and Brümmer, 2004). It is commonly assumed that desorption biphasic rates (i.e. fast and slow) are involved during SW by following kinetic models such as the intra-particle diffusion. Indeed, most of PAH adsorbed within sediment pores can desorb at a slower rate than the fraction adsorbed onto the outer surface (Kang et al., 2019).

This remediation technique can achieve high PAH removal efficiencies, limiting the deterioration of physical-chemical properties of sediment and microbial activity while leading to reduced operating costs (Lee and Hosomi, 2000; Trellu et al., 2016). However, the applicability and efficiency of SW can be affected by the organic fraction content in sediment (Mulligan et al., 2001) and pollutant properties (e.g. solubility). Also, SW efficiency can be influenced by the quantity and type of the extracting agent, the solid-to-liquid (S/L) ratio and the remediation time (Harati et al., 2021). Generally, removal efficiencies higher than 90% can be achieved by using an S/L ratio lower than 1:10, with a consequent increase of the process costs (i.e. 10–600 € m^{-3}, Table 4) and amounts of spent SW effluents requiring further treatment (Gan et al., 2009; Kuppusamy et al., 2017), which would make this technique invasive to the environment. On the other hand, a low S/L ratio can lead to a decrease of time for reaching the equilibrium, which does not exceed 72 h (Table 3) (Zou et al., 2009). With regard to the type of extracting agents (Table 3), the use of surfactants generally enhances the SW efficiency by increasing PAH solubility due to the presence of the hydrophobic interior structures (Trellu et al., 2016). However, PAH removal efficiency can be lower than 90% when the dosage of the extracting agent is below 2 g L^{-1}, especially in the presence of cationic surfactants (Von Lau et al., 2014). A non-toxic, biodegradable and economic alternative can be the use of vegetable oil (e.g. sunflower, peanut) (Table 3) as extracting solvent, which can also be combined with biological processes (Gan et al., 2009; Kuppusamy et al., 2017). Various studies obtained a PAH removal comprised between 52 and 100% (Table 3) by employing sunflower and peanut oils (Gong et al., 2005; Pannu et al., 2004), with the remaining vegetable oil in sediment subsequently turning into a growth medium for the microbial community. Also, the abovementioned limitations (i.e. low S/L ratio) can be overcome using a mixture of ethanol and water, which leads to the decrease of spent SW effluents to be treated due to a high S/L ratios used (i.e. 1:3) (Bianco et al., 2020a).

In each case, in order to reach a complete PAH degradation, SW can be combined with advanced oxidation processes (AOPs) in which the complete mineralization of the extracting agent and pollutant can occur (Huguenot et al., 2015; Muscetta et al., 2022). However, AOPs such as Fenton processes can show disadvantages such as the use of a high reagent dosage (i.e. in the order of grams per liter), the consumption

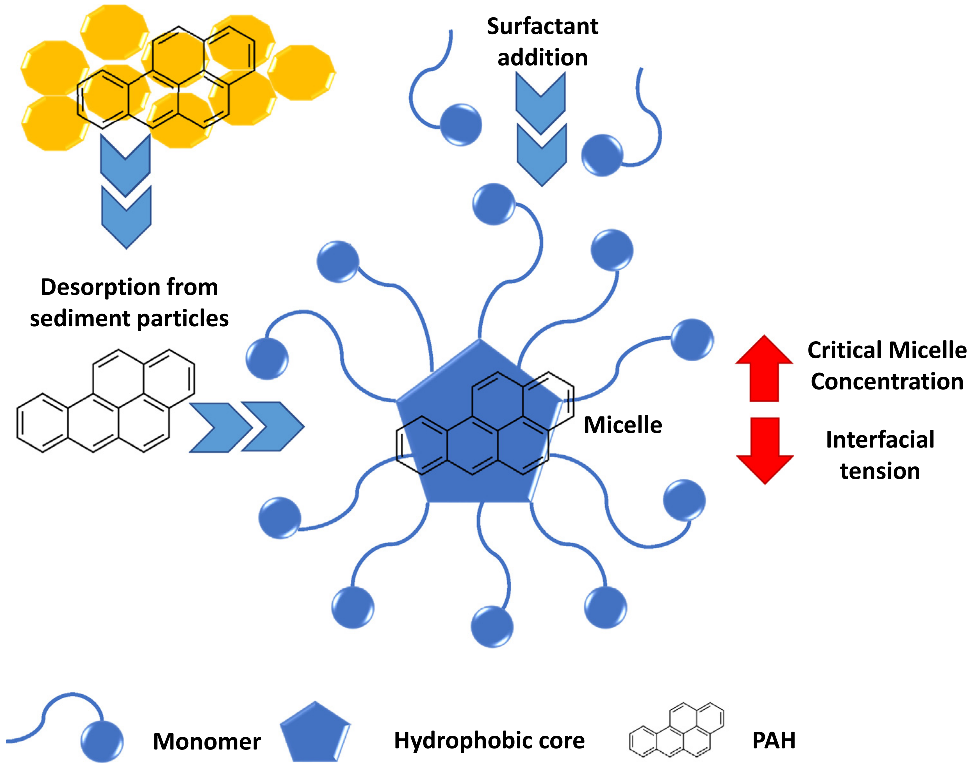

Fig. 5. Schematic representation of the sediment washing mechanism involved during polycyclic aromatic hydrocarbon (PAH) desorption from the polluted sediment in the presence of surfactants. A decrease in surface tension (↓) and a surfactant concentration above the critical micelle concentration (↑) can improve PAH solubility in the aqueous phase.

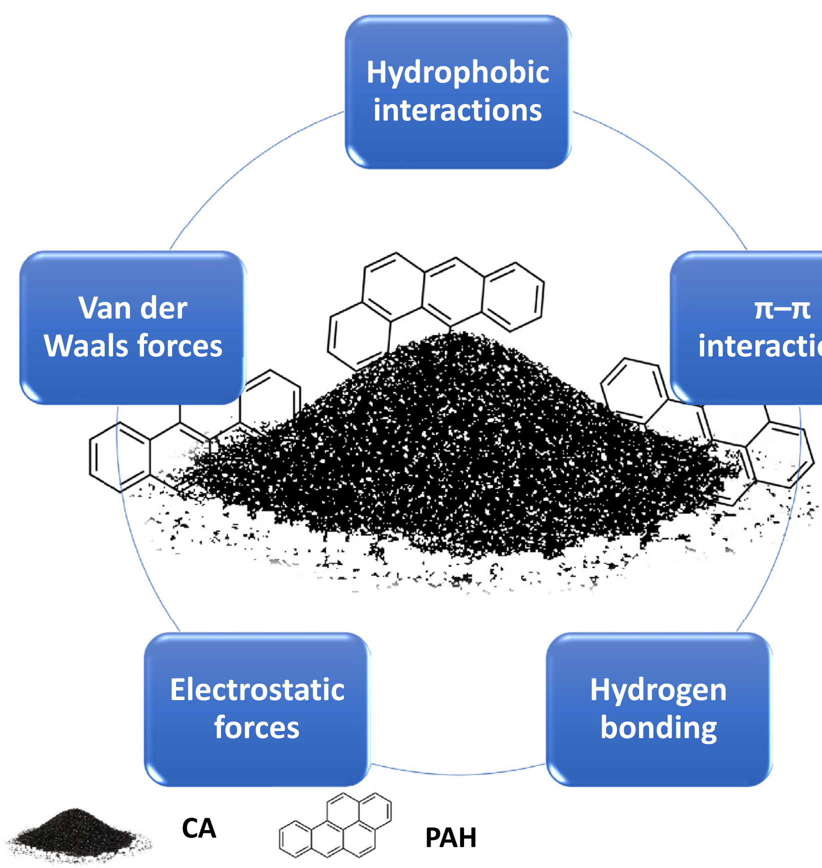

Fig. 6. Schematic representation of the occurrence of adsorption mechanisms after carbonaceous amendment (CA) addition to polycyclic aromatic hydrocarbon (PAH)-contaminated sediments.

of hydroxyl radicals due to parasitic reactions and the generation of an excessive amount of sludge as iron(III) oxide-hydroxide (Trellu et al., 2016). Therefore, the classic Fenton reaction can be combined with UV-A irradiation (i.e. photo-Fenton process) and the use of solid iron-containing catalysts (e.g. pyrite) to tackle the abovementioned issues (Ammar et al., 2015).

4.2.1. Treatment of spent sediment washing solution

The main disadvantage of the SW process is the generation of a large amount of PAH-containing effluents, which require proper treatment to remove pollutants and allow the recovery of the extracting agent for its reuse in a subsequent SW. Also, synthetic surfactants present different drawbacks such as low biodegradability, higher toxicity, and lower ecological compatibility, and should not be preferred to organic extracting agents (Trellu et al., 2021) to allow a safe release of the treated SW effluent. Otherwise, the SW effluent should be further treated (e.g. selective physical adsorption) after PAH removal prior to its discharge into the environment (Ahn et al., 2008).

Several technologies can be applied for the treatment of spent SW solutions such as AOPs (Figure S2), physical adsorption (e.g. active carbons), biological and integrated processes (Ahn et al., 2008; Gharibzadeh et al., 2016; Qu and Fan, 2010). During the choice of the mentioned alternatives, a major interest is focused on cost-saving, reduced use of chemicals, low energy input, sludge production, and effluent discharge after treatment (Trellu et al., 2016). The most effective processes for the removal of organic pollutants are AOPs (Figure S2) (Muscetta and Russo, 2021) such as electro-Fenton due to the generation of hydroxyl radicals ($OH^{\cdot}$) (Eq. (1)) enabling the oxidation of PAHs without the addition of H_2O_2 (Eq. (2)) (Huguenot et al., 2015; Satyro et al., 2014), as also shown for various soil washing experiments (Trellu et al., 2016).

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \tag{1}$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^{\cdot} \tag{2}$$

However, selective and complete mineralization of the target PAH cannot be achieved using AOPs (Figure S2) in order to allow the recovery and reuse of the extracting agent (Trellu et al., 2021). Also, the oxidation of PAHs can lead to the generation of oxygenated-PAHs, which are toxic for living organisms (Lamichhane et al., 2016). Finally, a further limitation of AOPs (Figure S2) could be the requirement of a high amount of chemicals and consumption of energy (Trellu et al., 2016).

A biological treatment can be alternatively employed as an eco-friendly and cost-saving alternative to AOPs. High PAH removal can be reached by optimizing the operating parameters such as pH, temperature and dissolved oxygen concentration in bioreactors where the available substrate is adsorbed onto biomass and subsequently biodegraded through engineered microorganisms (Sundaramurthy et al., 2011). Previous studies reported high PAH biodegradation (i.e. up to 97%) after several weeks in sequential batch, fed-batch and continuous-flow bioreactors (Bianco et al., 2022c,b; Hu et al., 2013). This is most likely due to the recalcitrance of PAHs, PAH concentration, type of the extracting agent and the specific effects towards the microbial community. Further studies should be addressed in the future by focusing on the biodegradation mechanisms and PAH removal kinetics mainly aimed at the application of continuous-flow systems (Trellu et al., 2016). In order to decrease the use of chemicals, improve the selective PAH degradation and increase the amount of extracting agent employed for SW, an initial physical separation step can be performed prior to the degradation phase (i.e. integrated processes) through PAH adsorption onto carbonaceous materials such as activated carbon (AC) and biochar (BC) (Feng and Zhu, 2018; Sayyahzadeh et al., 2016). Notwithstanding, PAH adsorbed phase onto AC can occlude the adsorbent micropores by limiting the removal efficiency (i.e. up to 57%) (Gan et al., 2009). On the contrary, AC can be subsequently regenerated via a thermal process for further PAH removal but, however, this solution is limited due to the high amount of energy needed (Marchal et al., 2013). Therefore, a good alternative can be the use of BC immobilized-cell reactors, in which PAHs are firstly adsorbed onto BC and subsequently biodegraded by bacteria (Bianco et al., 2022c; Lu et al., 2021).

5. Thermal remediation

Thermal processes can be reliable to carry out a rapid remediating intervention with high PAH removal efficiencies (i.e. >99%, Table S5) (O'Brien et al., 2018). Indeed, thermal techniques are based on the use of heat to mobilize PAHs into gaseous flow, such as thermal desorption (TD), break down the contaminants into simpler compounds (e.g. pyrolysis), destroy the pollutants (i.e. incineration), or immobilize them through stabilization or vitrification (Vidonish et al., 2016).

TD is an invasive ex-situ technique aimed at releasing PAH concentrations by heating the contaminated sediment with temperatures typically above 100 °C (Khan et al., 2004). TD can be classified into low-temperature (LTTD) and high-temperature (HTTD) when the heating is ranging between 100–300 and 300–600 °C (Bianco et al., 2020a), respectively. The involved temperature depends on the type of PAHs due to a different boiling point, which also affects the appropriate heating times. This remediation treatment should not be confused with incineration, being PAH desorption and volatilization from the polluted matrix the main mechanism involved (Figure S3) (Kuppusamy et al., 2016). For instance, PAH combustion can lead to the production of carbon dioxide and water by employing higher temperatures than TD (i.e. up to 1000 °C) (O'Brien et al., 2018). However, in real conditions, the heating temperature and the presence of oxygen in the atmosphere can be coupled with further removal mechanisms (e.g. pyrolysis, thermal oxidation) (Figure S3), and the occurrence of these reactions is enhanced with the increase of temperature and oxygen percentage (Zhao et al., 2019). Also, desorption/volatilization mechanisms that occurs during TD are coupled to subsequent treatment of gaseous flow (Figure S3) via destructive (e.g. thermal combustion, photocatalytic oxidation) or recovery techniques (e.g. membrane separator, solid adsorption) (Zhao et al., 2019).

Although thermal techniques can achieve high PAH removal efficiencies (Table S5), TD is not generally recommended due to a high energy demand that is reflected in the total treatment costs (i.e. up to 2000 € m^{-3}, Table 4). A solid-liquid separation can be performed prior to TD to remove the sediment moisture and reduce the energy input (O'Brien et al., 2017). A further drawback can be that TD can alter sediment the physical-chemical and biological sediment characteristics. Indeed, the desorption/volatilization of organic pollutant during TD is also coupled with the removal of SOM up to 85% after 1 h (O'Brien et al., 2016). Also, pH values can increase from about 6.9 up to 9.0 after TD (i.e. at 360 °C for 1 h) likely due to the removal of organic acids and release of cations from the SOM (Pape et al., 2015). Therefore, TD-remediated sediments can affect plants and microbial population by decreasing biomass, genetic diversity and enzymatic activity, probably due to the changes in SOM and nutrient availability (Yi et al., 2016).

6. General discussion

As in detail discussed in the previous sections, the remediation of PAH-polluted sediments can be achieved by employing a range of biological, physical-chemical and thermal technologies, each of them showing advantages and disadvantages, as summarized in Table 4.

Bioremediation can be used as an environmentally-friendly solution involving microorganisms to biologically degrade PAHs from polluted sediments. In particular, both biostimulation and bioaugmentation take place under aerobic conditions to overcome some limitations due to the lack of sufficient nutrients or when the indigenous microbial community is not adequate for remediating PAH-containing sediments

Table 4
The advantages and disadvantages of the main conventional remediation techniques for contaminated sediments. PAH = Polycyclic aromatic hydrocarbon; VOCs = volatile organic compounds.

	Bioremediation	Physical-chemical	Thermal
Technologies	Composting; Landfarming; Bioreactor; Bioaugmentation; Biostimulation; Biosparging; Phytoremediation	Sediment washing; Immobilization; Stabilization; Chemical oxidation; Photocatalytic degradation; Electrokinetic; Capping; Air sparging	Incineration; Thermal desorption; Vitrification
Advantages	Biotransformation of PAHs to a less toxic compound; simple equipment; high safety; low energy consumption; cost saving; can be applied both in-situ and ex-situ; can be performed in both aerobic and anaerobic environments	Effective for dissolved and adsorbed contaminants; high removal efficiencies; competitive costs; short treatment time; controlled production of VOCs; possible combination with biological processes	Low treatment time; the remediated sediment can be reused; reduced production of toxic substances; PAHs are destroyed; the polluted matrix can be used for energy production; high treatment efficiency; applicable in emergency situations due to accidental discharge of PAHs
Disadvantages	Limited to biodegradable compounds; high remediation time; slow microbial growth; low PAH bioavailability; superficial treatment; highly dependent on environmental and operational factors; can require an external energy source; inoculated microorganisms can compete with the indigenous microbial structure	Risk of PAH mobilization; large spent washing effluents; preferential routes of the contaminant; non-eco-friendly; possible dispersion of chemicals; reagent cost; acidic pH values (Fenton process)	High cost due to energy consumption; dredging required; can be applied only ex-situ; altered chemistry of sediment; treatment of off-gas; difficult to be implemented in industrialized and residential areas; a poor design of the intervention could create the migration of the pollutant; could impact on groundwater
Costs € m^{-3} of sediment	5–300	10–600	50–2000
PAH removal	Up to 97%	>99%	>99%

(Table 4) (Kuppusamy et al., 2017). However, PAH bioavailability still represents a limiting factor for sediment bioremediation. Physical-chemical technologies can cope with this issue by also acting on the non-bioavailable fraction of PAHs. Among the physical-chemical approaches, SW represents a well-established technique due to its operational simpleness, cheapness (i.e. 10–600 € m^{-3}) and high effectiveness (i.e. up to 100%) (Table 4), which has been proven with various extracting agents such as organic solvents and non-ionic surfactants (Bianco et al., 2022b). However, the generation of a considerable amount of spent SW effluents requires further treatment efforts (e.g. AOPs or biological process) and still limits the use of the SW process. The addition of CAs to PAH-polluted sediments (e.g. capping, Table 4) can tackle the drawbacks shown by other remediation techniques (Han et al., 2019). In particular, the efficiency (i.e. reduction of pore water PAHs up to 99%) obtained with the amendment of BC in PAH-contaminated sediments mainly encourages the adoption of this technique, even more considering that BC is thermochemically obtained from pyrolysis of substances regarded as wastes. Also, some limitations related to the recovery of CAs and desorption phenomena can be overcome with proper adsorbent modifications (e.g. magnetic BC, VFA-coated BC). Thermal processes such as TD are not affordable to perform sediment remediation due to the high costs and energy demand (Table 4) (Zhao et al., 2019). Plant growth and microbial population can be significantly affected in thermally-treated sediments compared to the untreated matrix (Yi et al., 2016).

Although the discussed technologies can show great removal efficiencies, high efforts for their implementation and management can be required. First of all, it is necessary to identify the most suitable remediation technology for a certain site taking into account all technical, environmental and economic aspects. Also, it is important to evaluate whether the remediation process can be performed in-situ or ex-situ, meaning to carry out the treatment directly in/on the polluted site with considerable cost savings or after sediment dredging elsewhere with an improved efficiency, respectively. The operation of an ex-situ treatment system may be a more expensive solution, but remediation is not affected by external environmental conditions and the optimal parameters for speeding up the PAH removal from the contaminated matrix can be maintained and monitored. Finally, the choice of the most suitable technique should be conducted only after a risk analysis as a function of a host of factors including PAH type, contamination size and distribution as well as location. This analysis strongly depends on the goals imposed by the specific national legislation.

7. Conclusions and future perspectives

This review is a comprehensive overview of the main remediation techniques employed for the removal of PAHs from marine sediments, by examining the key aspects such as effectiveness, drawbacks, operating parameters, economic and environmental aspects. All the revised remediation techniques can show specific advantages and drawbacks, which should be examined on the basis of each specific case. The major disadvantages of sediment washing and thermal remediation are their negative effect on the environment (e.g. spent washing solution, flora destruction) and the requirement for high investment (e.g. chemical cost, energy demand). Bioremediation can be a promising solution to tackle these disadvantages, but being a biological approach can be hard to be managed and also affected by long degradation times and low PAH removal efficiencies. The employment of CAs such as biochar can be also affordable due to their effectiveness, low costs and eco-friendliness for the remediation of PAH-polluted sediments. On the other hand, the adverse effects associated with CA recovery should be carefully assessed.

Future studies should be addressed to the use of biostimulation and bioaugmentation techniques under anaerobic conditions to investigate the role of key factors (e.g. bioreactor type, temperature, pH, C/N ratio, moisture content) and biogas production during bioremediation. AD should be also investigated for the treatment of PAH-polluted SW solutions in continuous-flow systems by shedding light on operating parameters such as hydraulic retention time (HRT), PAH loading rates. The use of immobilized-cell bioreactors can improve the microbial retention developing a biofilm onto supporting carriers, thus decreasing the HRT rather than in suspended-cell bioreactors. Therefore, the choice of the most appropriate carrier for cell immobilization would play a major role for a proper bioreactor functioning. In this sense, the combination of BC obtained by co-pyrolysis of several feedstocks (e.g. lignocellulosic materials, sewage sludge) and modified with alginate hydrogel spheres as well as fatty acids should be carefully assessed. Finally, more effort should be put into the study of halophyte species (e.g. S. *fragilis*) aimed at the phytoremediation of contaminated marine sediments, which can be enhanced with the employment of substances such as non-toxic surfactants aimed at improving PAH mobilization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.resenv.2022.100101.

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