



Recent advances in PCB removal from historically contaminated environmental matrices

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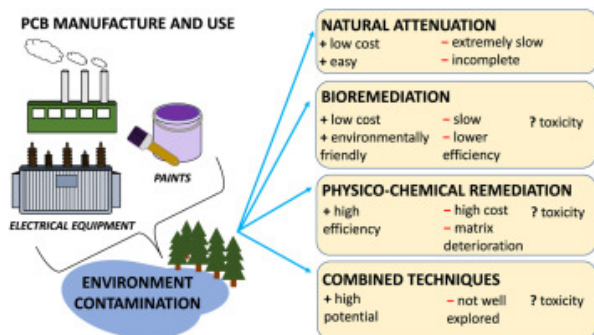
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Abstract

Despite being drastically restricted in the 1970s, polychlorinated biphenyls (PCBs) still belong among the most hazardous contaminants. The chemical stability and dielectric properties of PCBs made them suitable for a number of applications, which then lead to their ubiquitous presence in the environment. PCBs are highly bioaccumulative and persistent, and their teratogenic, carcinogenic, and endocrine-disrupting features have been widely reported in the literature. This review discusses recent advances in different techniques and approaches to remediate historically contaminated matrices, which are one of the most problematic in regard to decontamination feasibility and efficiency. The current knowledge published in the literature shows that PCBs are not sufficiently removed from the environment by natural processes, and thus, the suitability of some approaches (e.g., natural attenuation) is limited. Physicochemical processes are still the most effective; however, their extensive use is constrained by their high cost and often their destructiveness toward the matrices. Despite their limited reliability, biological methods and their application in combinations with other techniques could be promising. The literature reviewed in this paper documents that a combination of techniques differing in their principles should be a future research direction. Other aspects discussed in this work include the incompleteness of some studies. More attention should be given to the evaluation of toxicity during these processes, particularly in terms of monitoring different modes of toxic action. In addition, decomposition mechanisms and products need to be sufficiently clarified before combined, tailor-made approaches can be employed.

Graphical abstract



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Introduction

Polychlorinated biphenyls (PCBs) are organochlorine aromatic compounds classified as persistent organic pollutants that are subject to the restrictions imposed by the Stockholm Convention. Altogether, 209 PCB representatives (referred to as congeners) can exist. The congeners are numbered 1–209 and vary in the number and positions of 1–10 chlorine substituents on the biphenyl molecule. Individual congeners also differ in their physicochemical properties, behavior in the environment, susceptibility to different degradation mechanisms, and toxicity. Typically, highly chlorinated PCBs are less soluble in water, have lower vapor pressure and are more recalcitrant than less chlorinated congeners. PCBs have numerous adverse effects on living organisms. Moreover, PCBs have been classified as carcinogenic to humans (Group 1) by the International Agency for Research on Cancer (Lauby-Secretan et al., 2013).

PCBs are anthropogenic pollutants and lack natural sources of contamination. Owing to their advantageous physicochemical properties, which include high thermal stability, chemical resistance, high thermal conductivity, low flammability, and low dielectric constant, they were extensively used as dielectrics in capacitors and transformers, thermally conductive liquids, hydraulic oils, plasticizers, and additives in oil-based paints and carbonless copy paper. PCBs were manufactured as mixtures containing different amounts of the individual congeners, which were characterized by their average chlorine content. PCB production started in 1929 and peaked during the 1970s; at least 1.3 million tons were produced in total (Breivik et al., 2007). A substantial amount of PCBs has been released into the environment during their production, utilization, and disposal, whether from industrial accidents or inappropriate waste management.

Long-range transport has resulted in the ubiquitous presence of PCBs even in remote areas where they have never been used, such as polar regions (Cabrerizo et al., 2017; Combi et al., 2017; Ubl et al., 2012). As a result of their lipophilic nature, PCBs bioaccumulate in food chains and bind to organic matter in soil and sediments. The estimated environmental half-lives of PCB congeners generally increase with higher chlorine number and range from days (trichlorinated PCBs) to years (heptachlorinated PCBs) in the air, months (trichlorinated PCBs) to decades (heptachlorinated PCBs) in water and years (trichlorinated PCBs) to decades (heptachlorinated PCBs) in soil and sediments (Sinkkonen and Paasivirta, 2000). The global burden of soil matrices from PCBs was estimated at 21,000 tons in 2003 (Meijer et al., 2003). Overall, background PCB concentrations in the environment are on the decline, but in many cases, exceed recommended limit values and still pose a threat to wildlife and humans (Guo et al., 2017; Schnitzler et al., 2019). Moreover, many recent articles have still dealt with highly contaminated matrices (Monfort et al., 2019; Siracusa et al., 2017; Stella et al., 2015).

Apart from legacy spills, PCBs are, together with polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs), also unintentionally released from industrial activities, such as incineration, metallurgical processes, or cement production, through many mechanisms, including de novo synthesis, and pollute the environment in their vicinity (Collina et al., 2017; Dat et al., 2020; Fujimori et al., 2020; Gabryszewska and Gworek, 2020; Zou et al., 2018). Recently, numerous reports have been published regarding the contamination of electronic waste-recycling sites, predominantly in Asia and Africa (Chen et al., 2014; Liu et al., 2019b; Moeckel et al., 2020). This issue seems to be on the rise, and PCBs originating from discarded capacitors and transformers are one of the main pollutants monitored, together with polycyclic aromatic hydrocarbons (PAHs), brominated flame retardants, and heavy metals. In 2016, the UNEP estimated that 17 million tons of PCB-containing liquids and equipment (83% of the total amount) still needed to be dealt with to fulfill the goals of the Stockholm Convention of PCB elimination by 2028 (Robertson et al., 2018). Furthermore, large quantities of sediment are dredged annually as a part of waterway maintenance. Dredged sediment usually contains a mixture of organic and inorganic pollutants, prompting researchers to develop and improve methodologies for their decontamination or disposal (Benamar et al., 2020; Urbaniak et al., 2020a; Wang et al., 2018).

Dozens of methods, each with its own advantages and obstacles, have been developed over the decades of PCB research. While physicochemical approaches are usually characterized by high efficiency, they are also accompanied by high costs. Moreover, traditional methods such as incineration present risks in terms of toxic byproduct generation (PCDD/Fs) and deterioration of the treated matrices. It may be argued that in certain cases, the activities and risks associated with remediation, such as excavation, transport, and disposal, can bring about their own negative effects, and the process may not be universally beneficial (Kvasnicka et al., 2020). However, it is necessary to bear in mind that PCBs represent highly bioaccumulative compounds and that their presence in the environment can pose long-term risks. In contrast, bioremediation is generally characterized by significantly lower costs and can result in an overall improvement in soil quality. On the other hand, bioremediation is also often associated with lower efficiency and/or incompleteness, and the potential for the generation of toxic metabolites is also of concern.

One of the key factors influencing whether contaminants pose a threat to the environment and which remediation technologies can be used for their removal is bioavailability. Since PCBs are, for the most part, legacy contaminants persisting for decades in the environment, the biologically available portion has been largely reduced, and the congener patterns have been slowly shifting in favor of the more recalcitrant compounds. On the one hand, the decrease in bioavailability reduces imminent risks associated with contamination (Taylor et al., 2019); on the other hand, it can prevent bio-based methods from being successfully applied. Many research studies assessing the capability of biodegrading organisms have utilized PCB standards or artificially contaminated matrices, neither of which reflect realistic scenarios. While experiments performed under precisely defined conditions are invaluable for gaining insight into degradation pathways and the potential of the methods, discrepancies are often found when the same approaches are applied to aged contamination (Bako et al., 2021; Rybnikova et al., 2016). In an attempt to combat this, in some studies, following artificial spiking with PCBs, the material was left to equilibrate for a period of time (days to months) (Cao et al., 2018; Fan et al., 2016; Salimizadeh et al., 2019; Ye et al., 2020). Nevertheless, the years and decades that have resulted in the sequestration and degradation of PCBs cannot be easily simulated. Therefore, studies focusing on aged contamination provide more realistic insight into the possibilities and limitations of the methods.

The scale-up of newly developed treatments represents another challenge. For physicochemical methods, technical feasibility and improvements in terms of cost-effectiveness mainly need to be resolved. Regarding bioremediation, while many organisms have proven their biodegradation capabilities under laboratory

conditions, the cultivability of the species under real conditions and their ability to compete with indigenous biota as well as seasonal limitations complicate their large-scale and in situ application.

The aim of this review is to provide a synthetic and critical overview of recent studies describing both biological and physicochemical approaches to removing PCBs under more realistic conditions, i.e., from aged, historically contaminated matrices. The main results of the studies, as well as any encountered shortcomings, are listed. In addition, special attention was given to articles reporting on pilot-scale and in situ trials. Recent studies regarding pilot-scale and in situ remediation attempts and laboratory-scale remediation experiments are summarized in Table 1, Table 2, respectively. Specific conditions, outcomes, and other aspects are discussed below.

Section snippets

Natural attenuation

Provided that low immediate risks are associated with the contaminants, the simplest way to approach remediation is through natural attenuation. Here, careful evaluation of the potential of the ecosystem to mitigate pollution by natural processes is key. By definition, this approach is the least demanding in terms of costs and technical feasibility, relying mostly on prospecting and monitoring. Nevertheless, because of the recalcitrance of PCBs, natural attenuation requires decades or may remain ...

Biostimulation/bioaugmentation

To enhance biodegradation processes, autochthonous organisms capable of PCB degradation may be supported by supplementation of nutrients and providing suitable growing conditions (biostimulation), and/or the microbiota can be amended by species with degrading capabilities (bioaugmentation). Biostimulation can occur after simple addition of a growth substrate, as was demonstrated using pine needles (Lehtinen et al., 2014). In another example, Song et al. (2015) applied lactate to polluted soil...

Excavation and landfilling

One of the most straightforward and common solutions for dealing with PCB-impacted soil is to excavate the contaminated portion and then either dispose of it in a hazardous waste landfill or destroy the PCBs by incineration or other thermal methods (see Section 4.4). An example study of excavation and landfilling was provided by Sharov et al. (2019), who described a project during which 804m³ of sandy soil contaminated by aromatic hydrocarbons and PCBs was removed from the site. The soil was...

Combined approaches

By using a combination of the available methodologies and technologies, the drawbacks of individual methods can be overcome. After the application of most physicochemical methods, natural soil properties are compromised. This can be remedied with the amendment of plants, which also provides additional potential for biodegradation (Ye et al., 2014). Urbaniak et al. (2020b) studied how thermal treatment affected PCB levels, toxicity, and subsequent phytoremediation of Hudson River sediment by C...

Conclusions

PCBs are particularly difficult pollutants to manage as a result of their stability and persistence. It is not possible to recommend a single technology for all types of PCB-contaminated sites because of the vast spectrum of factors that determine their suitability. Many of the reviewed technologies have shown insufficient results or limitations when dealing with historically contaminated matrices, and it is unclear whether the available large-scale and in situ reports accurately reflect trends ...

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....

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...Polychlorinated biphenyls (PCBs) were widely used in industrial because of their chemical and physical stability (Kumari, Dhankhar, & Dalal, 2021; Šrédlová & Cajthaml, 2022)....

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