Is sorption technology fit for the removal of persistent and mobile organic contaminants from water?

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HIGHLIGHTS

- Many PMT substances have particular sorption behavior because they are ionizable.
- Sustainability and resource use are becoming more important in sorbent design.
- Sorbent type and regeneration approaches are evaluated in terms of sustainability.
- Coupling sorption to destruction or other technologies can be necessary for PMT.

GRAPHICAL ABSTRACT
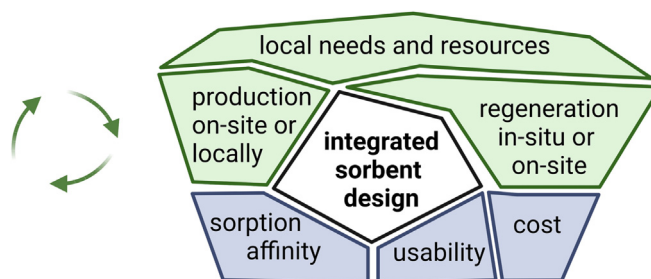
ABSTRACT

Persistent, Mobile, and Toxic (PMT) and very persistent and very mobile (vPvM) substances are a growing threat to water security and safety. Many of these substances are distinctively different from other more traditional contaminants in terms of their charge, polarity, and aromaticity. This results in distinctively different sorption affinities towards traditional sorbents such as activated carbon. Additionally, an increasing awareness on the environmental impact and carbon footprint of sorption technologies puts some of the more energy-intensive practices in water treatment into question. Commonly used approaches may thus need to be readjusted to become fit for purpose to remove some of the more challenging PMT and vPvM substances, including for example short chained *per-* and polyfluoroalkyl substances (PFAS). We here critically review the interactions that drive sorption of organic compounds to activated carbon and related sorbent materials and identify opportunities and limitations of tailoring activated carbon for PMT and vPvM removal. Other less traditional sorbent materials, including ion exchange resins, modified cyclodextrins, zeolites and metal-organic frameworks are then discussed for potential alternative or complementary use in water treatment scenarios. Sorbent regeneration approaches are evaluated in terms of their potential, considering reusability, potential for on-site regeneration, and potential for local production. In this context, we also discuss the benefits of coupling sorption to destructive technologies or to other separation technologies. Finally, we sketch out possible future trends in the evolution of sorption technologies for PMT and vPvM removal from water.

1. Introduction

The use of charcoal to bind unwanted odors and toxins historically goes back to the ancient Roman and Chinese empires. Since then, humanity has come a long way in developing ever more sophisticated ways to generate tailored sorbent materials for contaminant removal from air and water.

Following the first modern activated carbon (AC) developments in the early 20th century, carbonaceous sorbents (carbons) have become a cornerstone of water treatment and purification (Hagemann et al., 2018; Schanz and Parry, 1962). Decades of research and industrial development have optimized production conditions and the use of chemicals (e.g., H_2O, CO_2, H_3PO_4, and $ZnCl_2$) resulting in ACs specifically tailored for a variety of applications (Bhatnagar et al., 2013). Approaches for synthesizing even better-defined ordered porous carbons from templates (e.g., zeolites or silica nanoparticles) also exist (Lee et al., 2006), and complement the range of carbon nanomaterials (e.g., carbon nanotubes and fullerenes) which have structural similarities with AC. All carbons consist of highly aromatic graphene-like structures that form micropore (< 2 nm) and mesopore (< 50 nm) networks with high specific surface areas in the order of up to 2500 m^2/g (Borchardt et al., 2017).

Recently, a new class of contaminants that are very mobile and often difficult to remove from water captured the interest of researchers, water professionals and policymakers alike. This contaminant class is now known as Persistent, Mobile, and Toxic (PMT) and very persistent and very mobile (vPvM) substances. What characterizes these contaminants is their high stability and mobility in the aquatic environment which enable such compounds to reach groundwater bodies and withstand a number of existing water treatment technologies (Hale et al., 2022; Jin et al., 2020). Increasingly lower regulatory threshold concentrations for risk that can range from ng/L to μg/L depending on the specific substance, necessitate advanced water treatment at concentrations that previously were often neglected (Hale et al., 2022). Importantly, an overwhelming majority of PMT and vPvM substances identified by researchers are charged or ionizable compounds, which strongly affects their mobility and sorption behavior (Sigmund et al., 2022).

To efficiently remove PMT and vPvM substances from water while minimizing the environmental footprint of treatment technologies will be a challenge for the years to come. Therein, sorption-based approaches offer a number of advantages compared to other technologies, including simple process engineering and safe application. However, to harness the full potential of sorption technologies for PMT and vPvM removal from water, a number of issues need to be tackled. For instance, small negatively charged and highly mobile PMT vPvM substances such as trifluoroacetic acid (TFA) remain difficult to remove, and operating times of fixed-bed adsorbers need to be shortened, which necessitates strategies for on-site low-tech and low-cost sorbent regeneration. Considering the environmental footprint of sorbent materials also necessitates reconsideration of non-traditional feedstocks for AC production and further development of regeneration approaches with minimal energy and chemical inputs, which may involve improved on-site sorbent production and/or regeneration approaches (Fig. 1).

In the following sections, we critically evaluate the potential and limitations for applying carbons and complementary non-carbon sorbents to remove PMT and vPvM substances from water, highlight examples of promising applications from literature, and sketch out possible paths forward in improving performance and sustainability of sorbent-based PMT and vPvM removal in water treatment.

2. Interactions that determine sorption to carbons

The sorption of organic contaminants to carbons is driven by the compounds hydrophobicity (i.e. thermodynamic driving force to leave the water phase), the compounds aromaticity and polarizability, as well as the sorbent porosity and aromaticity (Kah et al., 2017; Pignatello et al., 2017). The high porosity and specific surface areas in carbons offer a large number of potential sorption sites, and small pore structures are available for condensation and pore filling (Nguyen et al., 2007). Steric hindrance can exclude compounds from accessing the pore networks. It has been suggested that for sorption to occur, pores need to be at least 1.3 to 1.8 times larger than the kinetic diameter of the target contaminant (Li et al., 2002). More precisely, the spatial conformation and "flexibility" of compounds affect their ability to reach sorption sites (McCleaf et al.,
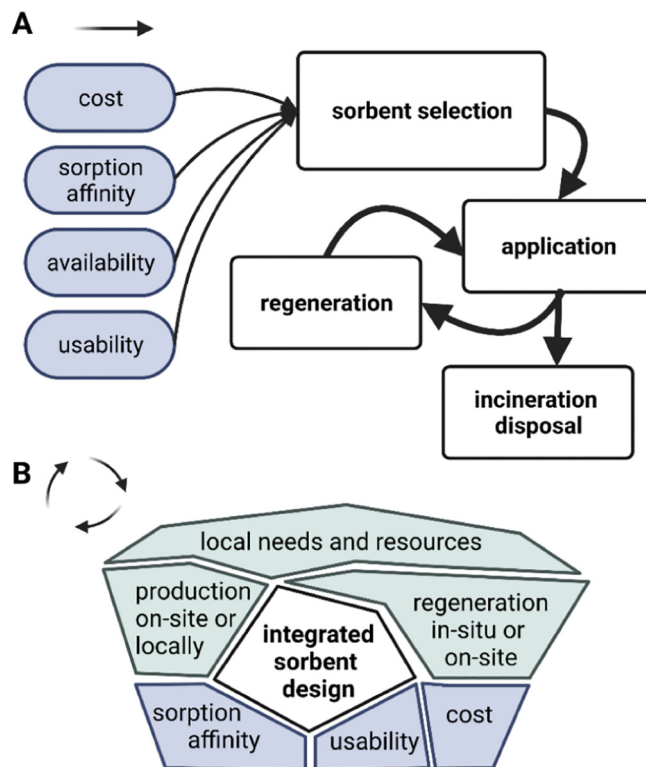

Fig. 1. The current sequential practice in sorbent selection and design with driving factors in blue and use-design factors in white (A) compared to a future integrated approach for sustainable sorbent selection and design with additional environmental footprint factors in green and the use-design factors integrated into one box in white (B).

2017). In many relevant applications, dissolved organic matter can reduce contaminant sorption by competing for sorption sites (Newcombe et al., 2002), and/or by blocking pores and thus reducing the accessible pore space for sorption (Pelekani and Snoeyink, 1999).

Graphene-like structures in carbons are highly polarizable and have a capacity to accept and donate π–electrons. These properties are directly linked to the degree of aromatic ring condensation within the material and are responsible for a particularly high sorption affinity for aromatic compounds which occurs via π—π electron donor-acceptor interactions (Wang et al., 2010; Yang et al., 2021a; Zhu and Pignatello, 2005). In contrast, purely aliphatic compounds are not capable of π– π electron donor-acceptor interactions. For instance, sorption of *per-* and polyfluoroalkyl substances (PFAS), an important group of PMT and vPvM substances, is determined mainly by hydrophobic effects and/or electrostatic interactions. As most carbons maintain a negative surface charge at relevant pH ranges (5–9), anion-exchange resins are often better sorbents than AC (in terms of performance) for such aliphatic anionic compounds, particularly for short-chain (C4-C6) and ultrashort-chain (<C4) PFAS with very low hydrophobicities (Gagliano et al., 2020; Saeidi et al., 2021a). When a positive charge is present in the target contaminant, electrostatic attraction towards negatively charged surface functional groups usually overshadows the importance of possible π—π electron donor-acceptor interactions (Sigmund et al., 2020).

Based on the existing knowledge and experience from practitioners, traditional carbons can be expected to be well-suited to remove a wide variety of aromatic compounds from water. Aliphatic compounds that are not capable of accepting or donating π–electrons can still be removed if they are somewhat hydrophobic, neutral or positively charged. Small aliphatic and/or negatively charged molecules, however, are the most challenging contaminants for traditional carbons as they show early breakthrough e.g. in AC fixed-bed adsorbers, and shorten their operation times (McCleaf et al., 2017).

3. Optimizing carbons for PMT and vPvM removal

Optimization of sorbents typically starts at the screening of available commercial AC products for a certain contaminant spectrum. In the past, the available specific surface area determined via N_2 physisorption BET analysis or approximated via the iodine number was arguably the central criterion used to approximate the materials' sorption potential. However, as discussed above, when considering ionic and ionizable contaminants additional characteristics need to be considered. For instance, the abundance and type of surface functional groups and related (localized) surface charges greatly affect the sorption of long- and short-chained PFAS anions, and can determine the vastly different performances reported for AC products and PFAS removal in literature (Saeidi et al., 2020). This is because charged molecules are subject to attractive and repulsive electrostatic interactions that do not occur for neutral compounds (Sigmund et al., 2020). To maintain phase neutrality, sorptive removal of any ionic compound from the water phase requires charge compensation either via charges on the solid surface or the formation of ion pairs with dissolved counterions, which can be associated with a free energy penalty (Saeidi et al., 2021a; Zhou et al., 2021).

The nature and density of acidic and basic sites on the AC surface is determined by the original feedstock and the production characteristics (pyrolysis conditions and activation steps), leading to AC types with vastly different properties (Hagemann et al., 2018). To ease selection of AC, product notes should be extended to include standardized parameters that characterize the surface chemistry and charge of the product at relevant pH ranges which are currently rarely reported. Therein, pH-dependent anion and cation exchange capacity (AEC and CEC) are well suitable as they are related to the density of positively and negatively charged sites on the material and can be related to sorption of ionic organic compounds (Sigmund et al., 2022). For example, perfluorooctanoic acid sorption strongly correlates with increasing AEC and decreasing CEC of AC (Saeidi et al., 2020, 2021a). Also, Boehm titration allows the pH-dependent charge characterization of the surface functional groups (Boehm, 2002). The point of zero charge is less suitable because it only gives a singular qualitative information on the net charge of the surface above or below this pH value. The elemental composition can be a good complement to AEC and CEC, providing insights on aromaticity and the presence of heteroatoms, but cannot replace charge-related parameters. To harmonize characterization protocols and develop strategies to account for assessing matrix effects such as ageing and fouling on surface chemistry and charge will be a seminal task for future research in the field.

ACs and related materials are generally negatively charged at circumneutral pH, which explains why cationic compounds tend to sorb well to ACs. Still, increasing CEC of AC by surface oxidation can be useful to increase sorption of cationic organic compounds such as aromatic and aliphatic tertiary amines (Zhou et al., 2021). The relative increase in sorption is however, expected to be moderate, as negative charges already exist on most AC surfaces. Cations whose charge is located within an aromatic ring or delocalized into the ring can also undergo a combination of cation-π and π-π electron donor-acceptor interactions (Kah et al., 2017; Vijay et al., 2008).

In contrast, anions generally do not show a high sorption affinity towards carbons. To overcome this limitation, the surface chemistry and charge of ACs can be optimized for the removal of specific contaminant classes, e.g. by introducing positive charges to sorb negatively charged compounds. Therein, two strategies for increasing the density of positive surface charges on AC can be distinguished: (i) increasing or modifying functional groups to enhance heteroatom-associated positive charges and (ii) defunctionalization to increase C-backbone-related basicity. Heteroatom-related basicity in AC is assigned to certain oxygen-containing basic functional groups (pyrone-type structures i.e. carbonylic and etheric oxygens in close vicinity) and N-containing groups which can be specifically introduced via reactions with nitrogen-containing reagents (such as NH_3, nitric acid, and amines) or activation with nitrogen-containing precursors under high temperature (Pittman et al., 1997). Surface defunctionalization is based on the phenomenon that highly delocalized electrons in condensed aromatic regions of the carbon basal plane and carbon edges readily attract protons from water which creates a positively charged site in a low polarity environment (Shafeeyan et al., 2010). Defunctionalization of conventional AC by high-temperature treatment in N_2/H_2 atmosphere can enhance sorption affinity for negatively charged compounds such as short- and long-chain PFAS (Saeidi et al., 2021a) while retaining a strong sorption affinity for neutral and cationic compounds (Zhou et al., 2021). Surface defunctionalization of a microporous AC improved PFAS-anion removal more than introducing N-containing groups, possibly because the carbon-backbone-related sites allow for a better combination of hydrophobic effects and electrostatic attraction compared to the sterically limited sites created by N-groups (Saeidi et al., 2021a). Thus, defunctionalization may offer some advantages over hetero-atom associated modifications for PMT and vPvM removal from water. Nevertheless, carbon-based sorbents are limited in their capacity to remove some important types of PMT and vPvM substances; most importantly, aliphatic, small, and negatively charged contaminants.

4. Complementary high-performance sorbents

A number of non-carbon-based sorbents exist that are capable to overcome some of the limitations of carbons for PMT and vPvM removal and will be discussed in the next paragraphs.

Amino-functionalized sorbents contain positively charged surface functional groups, mostly, quaternary amines with stabilized charge. These groups can be introduced not just to ACs but also to other materials (Ateia et al., 2019a). Amino-functionalized sorbents include carbon nanotubes (Zhang et al., 2016), graphene (Cai and Larese-Casanova, 2016), chitosan-based polymers (Long et al., 2019), poly(ethylenimine)-functionalized cellulose (Ateia et al., 2018), polymers that form hydrogels for rapid uptake (Ateia et al., 2019b), and amino-functionalized covalent organic frameworks (Ji et al., 2018). For example, amino-functionalized cellulose can effectively and rapidly (within 25 min) remove trace concentrations of perfluorooctanoic acid (PFOA) from water at pH from 4.5 to 10.9 and in the presence of competitive inorganic and organic compounds (Ateia et al., 2018). However, for many of these materials, regeneration remains challenging.

Ion exchange resins and notably weak anion (WAX) and weak cation exchange resins (WCX) are widely applied in water treatment, and typically used for salt removal or ion exchange. WAX can be effective to remove highly mobile negatively charged organic contaminants such as trifluoroacetic acid and other PFAS, during water treatment (Dixit et al., 2021). While anion exchange resins have advantages compared to AC in terms of removal performance of short-chain PFAS anions, knowledge gaps exist in the optimal selection of resins in terms of minimizing negative impact and fouling by natural organic matter as well as regeneration options (Edgar and Boyer, 2021; Levchuk et al., 2018). For contaminants and anion exchange resins which dominantly interact via electrostatic interactions, regeneration by inorganic salts and/or alkaline solutions is efficient and brine treatment can be done by electrooxidation and other advanced oxidation approaches. High-performance anion exchange resins that interact with organic anions such as perfluorooctanoic acid by a combination of hydrophobic effects and electrostatic interaction require organic solvents such as methanol or mixed solvent and salt solutions for desorption (Taniyasu et al., 2008). Riegel et al. (2023) could elute a wide range of PFAS from a loaded anion exchange resin using mixtures of ethanol and NaOH or HCl, reaching satisfactory desorption with only 3 to 10 bed volumes of regeneration solution. After only 2 to 5 regeneration cycles, ion exchange resin could outcompete single-use granular AC cost-wise (Riegel et al., 2023). However, due to the challenges in preventing solvent leaching during reuse and increased efforts for solvent/brine treatment such resins are mainly used in a single-use-and-dispose mode in water treatment so far (Dixit et al., 2021).

Modified cyclodextrins include mainly cross-linked beta-cyclodextrin polymers with well-defined structures. In a recent study, styrene groups were covalently attached to beta-cyclodextrin to form a discrete monomer

for radical polymerization with various comonomers(Wang et al., 2022). This allows controlling the composite structure and suppressing side reactions and substitutions, leading to a well-defined material. Copolymerization with methacrylate bearing a cationic tertiary amine group led to a composite with very high affinity towards charged contaminants such as PFAS compared to other cyclodextrin composites (Wang et al., 2022). Adsorption of anionic PFAS by this cyclodextrin composite is governed by an interplay of electrostatic interactions and hydrophobic effects. Hydrophobic effects gained relative importance with increasing PFAS chain length, whereas sorption of shorter-chain PFAS was suppressed in the presence of inorganic salts pointing to a mainly electrostatic interaction mechanism. Regeneration of these novel sorbents is possible by organic solvent extraction (Alsbaiee et al., 2016), but is still not done at a larger scale due to solvent leaching issues similar to those discussed for ion exchange resins above.

Synthetic high-surface area zeolites are aluminosilicates which can be tuned by varying the Si/Al ratio to modify their hydrophobicity (Jiang et al., 2018, 2020). High-silica zeolites can have a similar sorption capacity as AC (Gonzalez-Olmos et al., 2013; Jiang et al., 2018). The main driving forces for sorption are hydrophobic effects and Lifshitz-van der Waals interactions. In addition, specific interactions such as hydrogen bonding or ion exchange can occur between polar functionalized or even ionized compounds with oxygen sites (Lewis base) and Brønsted/Lewis acidic sites of the zeolite (Jiang et al., 2018). Hydrophobic effects and Lifshitz-van der Waals interactions are important for high-silica zeolites, whereas specific interactions such as hydrogen bonding or ion exchange represent the dominant sorption mechanisms for high-aluminum zeolites. Moreover, zeolites with their distinct pore size allow for selective removal of small (i.e., low molecular weight) compounds, while sterically excluding larger organic compounds. Thus, zeolite sorption tends to be less affected by competitive sorption of background organic matter compared to AC.

Additionally, as mineral sorbents they are rather inert towards strong oxidants and radicals. Zeolites can fix highly disperse redox-active transition metal ions such as $Fe^{2+/3+}$ on the ion exchange sites where they can act as heterogeneous Fenton-like catalysts in a wide pH-range, so that sorbed contaminants can be degraded by hydrogen peroxide addition (Thomas et al., 2021). Fe-zeolites are also active in photo-Fenton reactions (Gonzalez-Olmos et al., 2012) under UV or solar irradiation and able to degrade perfluorooctanoic acid (Qian et al., 2020) and perfluorooctanesulfonic acid (Qian et al., 2021) in the sorbed state. Industrial zeolites are available at large scale. However, their high price compared to AC (7 to 30 fold) and uncertainties on their long-term on-site regeneration using oxidants such as ozone, hydrogen peroxide or persulfate (Qian et al., 2022), are obstacles to their successful implementation by industry.

Further inorganic high-surface area materials suitable for certain adsorption tasks are the well-known pillared clays (Hacıosmanoğlu et al., 2022) and layered-double hydroxides (Chen et al., 2021; Santamaría et al., 2020) which received increasing interest in the last decade. Recently, novel inorganic 2D materials such as MXenes are also proposed for adsorption of organic contaminants from water; however, they are still in the lab-research stage (Dixit et al., 2022).

Metal-organic frameworks (MOF) are known for rapid removal of heavy metals (Sun et al., 2018), but also polar and charged organic contaminants (Yang et al., 2021b) from water. MOFs can have exceptionally high specific surface areas up to 3800 m^2/g (Hong et al., 2009) and specific reactive functional groups, mostly active metal sites. These functional groups determine their catalytic properties and drive contaminant sorption (Yang et al., 2021b).

To be used in water purification, MOFs need to be stable in water at least at moderate acidic or alkaline conditions. MOF stability is strongly dependent on their chemical composition and structure (Pirngruber et al., 2012). For example, Fe-linked MOFs used for PFOA removal, were stable at pH range 3.3 to 11 (Yang et al., 2020), whereas Zn-linked MOFs showed significant structural changes while applied for adsorption of PFOA at acidic pH (Chen et al., 2016). Thermal stability, chemical stability and conductivity of MOFs are important for their regeneration by thermal, solvent leaching and electrochemical techniques, respectively. Some MOFs showed unchanged porosity and surface area after exposure to steam at up to 200 °C in several cycles (Jeremias et al., 2012). However, chemical stability was not evaluated. Some MOFs showed high stability against raised temperature, without thermal decomposition up to 500 °C (Healy et al., 2020). Other MOFs remained chemically stable after desorption using 0.1 M HCl/methanol mixtures at 40 °C, and could be reused several times (Lin et al., 2018). Poor conductivity is an obstacle for electrochemical regeneration of most MOFs, which can be overcome to some degree by hybridization with other functional materials with high electrical conductivity, such as carbon materials, conducting polymers, and metals (Deng et al., 2018).

To make the performance of the sorbent types discussed above more tangible, a comparison of sorption of PFOA as an exemplary anionic PMT substance are shown in Table 1. Therein, commercial sorbents that are readily available are highlighted, as these can be considered mature sorbents.

5. Combination of individual sorption or separation technologies

As previously discussed, in many cases AC is not capable to efficiently remove all PMT and vPvM substances from water. For such scenarios, a treatment train combining sorption and other separation technologies is possible. Two-stage or mixed-bed adsorbers are frequently used. For example, the combination of AC with anion exchange resins can be used to firstly remove dissolved organic matter and a significant proportion of organic compounds so that the water reaching the resin in a second step will contain substantially less compounds competing for sorption sites with its actual target compounds (e.g., low-molecular weight hydrophilic acids). Such a serial configuration drastically increases the usability and lifetime of the anion exchange resin (Riegel, 2022). Sorbent design where AC is combined include BRITA® point-of-use filters consisting of AC and ion exchange resins (Alsbaiee et al., 2016), which ensures a wider range of removable contaminant groups in a single mixed-bed adsorber. Similarly, AC can be combined with reverse osmosis (RO). The combined use of RO or nanofiltration and AC has been suggested as reliable multibarrier approach for water purification (Albergamo et al., 2019; Franke et al., 2021; Schoonenberg Kegel et al., 2010). As discussed above, AC is not well suited to remove small, negatively charged compounds (Hale et al., 2022; Jamil et al., 2019), which are, however, removed well by RO (Albergamo et al., 2019). With the exception of a few very small, hydrophilic but non-charged molecules, a combined multibarrier approach is thus well equipped to treat a large variety of waters. In water reuse schemes, AC filtration is commonly employed for RO-concentrate treatment prior to concentrate discharge.

All separation technologies cause up-concentration of target contaminants left for further treatment or safe disposal which is mostly performed offline and off-site. This of course can be logistically challenging and energetically costly, as illustrated by the example of PFAS destruction via incineration at very high temperatures (mostly 1000 °C or above) in specialized facilities (Winchell et al., 2021). Thus, in the next two sections we explore promising developments in sorbent regeneration and possibilities to combine sorption with other approaches (notably reactive abatement / destructive technologies) to overcome those limitations.

6. Sorbent regeneration

The regeneration of sorbent materials is of increasing interest, as in addition to the economic motivation to re-use sorbent materials, sustainability aspects surrounding energy and material use are gaining importance in conceptualizing and designing water treatment technologies (Fig. 2).

Thermal reactivation encompasses the thermal desorption of the sorbed compounds, concurrent thermal decomposition of these compounds and a subsequent carbonization and reactivation using mild oxidants such as H_2O or CO_2 at temperatures up to 1000 °C in large-scale multiple hearth furnaces, rotary kilns, or fluidized-bed reactors. Thereby, desorbed contaminants in the gas phase are thermally destroyed and the gas is subsequently

Table 1

Exemplary PFOA single-point adsorption coefficients (K_d) for various adsorbents reported in literature. Log K_d was calculated at a certain aqueous concentration (C_{aq}) of PFOA from information reported in the respective studies, if several similar sorbents were reported on, the best performing sorbent was selected. If possible, C_{aq} was selected in the range of 10^{-1} to 10^{1} μg/L to maximize comparability. Commercial sorbents are bold and marked with a superscript letter for the producers: [a] Fisher Scientific, [b] Calgon Carbon, [c] NORIT® A ULTRA E 153, [d] ACTITEX WK L20 (Jacobi CARBONS), [e] Biochar Supreme Inc., [f] Purolite® A860, [g] Amberlite™ IRA67 (Octochem, Inc.), [h] DEXSORB + (Cyclopure Inc., Encinitas, CA), [i] Clariant Produkte GmbH.

Sorbent	log K_d [L/kg]	C_{aq} [μg/L]	pH	Reference
Granular AC [a]	5.11	30	7	(Zhang et al., 2021)
Granular AC [b]	4.49	1.3	7.5	(Sörengård et al., 2020)
Powdered AC [c]	3.75	6.7	7.5	(Sörengård et al., 2020)
Pristine AC felt [d]	4.11	20	7	(Saeidi et al., 2021a)
Defunctionalized AC felt	6.46	20	7	(Saeidi et al., 2021a)
Biomass derived carbon spheres	5.34	20	6.5	(Balda et al., 2023)
Wood based biochar [e]	4.91	40	7	(Zhang et al., 2021)
Sewage sludge based biochar	5.73	1	5.4	(Krahn et al., 2023)
Strong base anion exchange resin [f]	6.11	21	7	(Dixit et al., 2019)
Weak base anion exchange resin [g]	5.08	85	5	(Yao et al., 2014)
Porous β-Cyclodextrin polymer [h]	6.70	0.016	7	(Ching et al., 2020)
Porous β-Cyclodextrin polymer	6.28	0.1	5.5–6	(Wang et al., 2022)
Poly(ethylenimine)-functionalized cellulose	5.80	20	6.5	(Ateia et al., 2018)
BEA type zeolite [i]	4.49	64	7	(Qian et al., 2022)
Zi-based MOF	4.18	21	5–7	(Chen et al., 2016)
Fe-based MOF	3.67	15,000	3.3	(Yang et al., 2020)

cleaned in a flue gas cleanup step. Thus, also complex organic matrices from natural waters and waste waters can be effectively eliminated (Worch, 2021). This approach is commonly used at an industrial scale by AC producers. The cleaned up AC can somewhat differ from the original material in terms of pore size distribution and surface chemistry, and 3 to 12 % of carbon are lost in the process depending on the reactor type (Crittenden et al., 2012). Reactivation can cause a shift to larger pores which can, for example, decrease the adsorption of short-chain (C ≤ 6), hydrophilic PFAS and increase the adsorption of long-chain (C ≥ 7), hydrophobic PFAS (Cantoni et al., 2021). In addition, reactivation can add or remove oxygen-containing functional groups and carbon basicity depending on the reactivation atmosphere (Sonmez Baghirzade et al., 2021). These changes can shift sorption affinities, particularly for polar and charged compounds where both steric and electrostatic effects can determine sorption.

In the context of a future carbon-neutral society and the corresponding need for energy efficiency, the question arises whether thermal regeneration is fit for the future. Currently applied reactivation is a high-temperature process linked to combustion of fossil fuels and/or a high energy demand that cannot be easily electrified with renewable energy sources. Thus, alternative regeneration strategies are expected to become increasingly important and may include regeneration at milder temperatures or exploiting other driving forces than thermal desorption/degradation. Even though long-term application of AC might require 'deep cleaning' by high-temperature reactivation at longer intervals, intermittent regeneration to renew sorption performance for the target contaminants could be done with lower energy input. The following regeneration methods were developed for different use cases, however, in light of the above discussed issues they could inform alternative regeneration strategies for carbon materials used for water treatment.
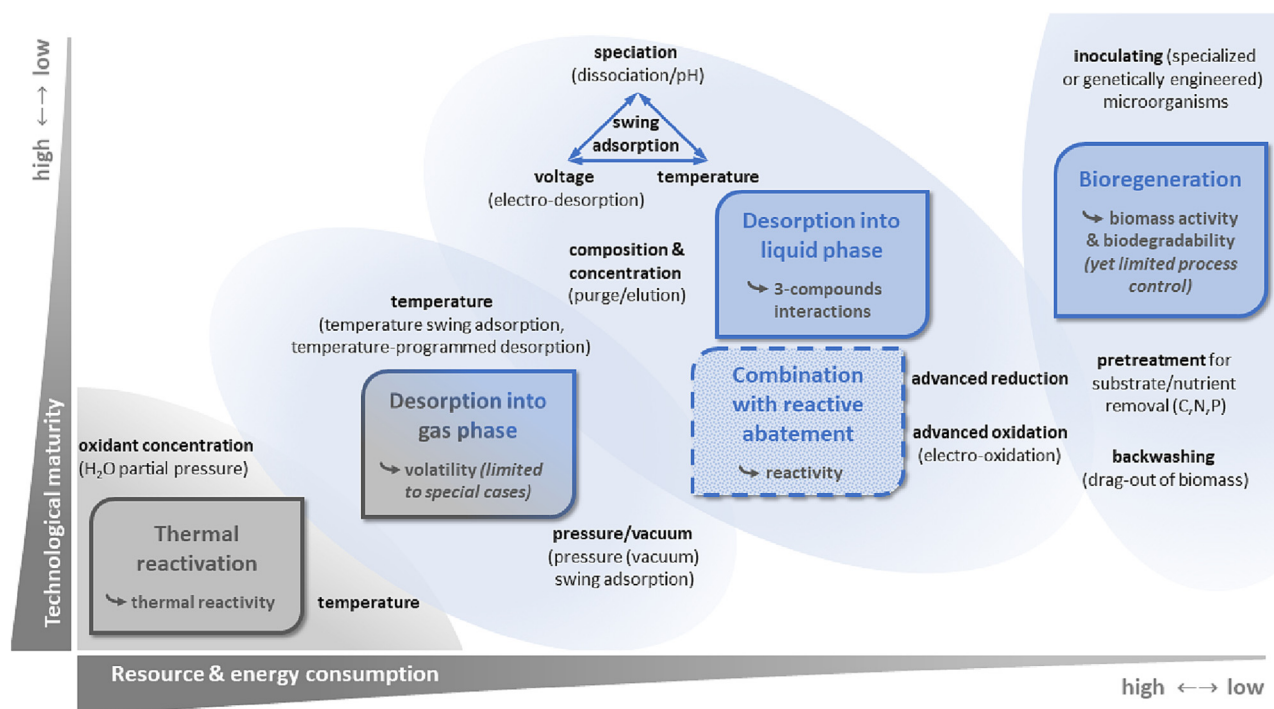

Fig. 2. Approaches for sorbent regeneration mapped with regard to the maturity of the technology and its resource and energy consumption. Boxes highlight underlying processes and fundamental application requirements; associated colored areas show process parameters and application variants.

Assisted thermal desorption into the gas phase aims at volatilization or evaporation of the sorbed compounds at lower temperatures compared to "pure" thermal reactivation (up to 400 °C). As with the thermal desorption described above, desorption kinetics can be limited by pore diffusion and/or by desorption from specific sorption sites. Vacuum or pressure swing adsorption is a common approach in gas-phase sorption (Hedin et al., 2013; Ntiamoah et al., 2016; Ohs et al., 2018) and only rarely applied in water treatment and sorbent regeneration, but may be considered more frequently if energy costs and sustainability considerations increase in weight. In general, desorption into the gas phase crucially depends on the volatility of the organic compounds or their susceptibility to thermal decomposition into volatile compounds which prevents it from wide application in water treatment. For example, the thermal desorption of PFAS follows their boiling point which increases with carbon chain length and is higher for sulfonic acids than carboxylic acids (Sonmez Baghirzade et al., 2021). Thermal desorption and reactivation at high temperatures can decompose the vast majority of PFAS, however, some fluorine traces can be found in reactivated carbons (Cantoni et al., 2021). Thermal desorption can also be assisted by feeding steam to separate volatile organics from another (Kim et al., 2001; Salvador et al., 2015), or effected by electrical current referred to as electrothermal swing adsorption (Grande and Rodrigues, 2008; Keller et al., 2019; Salvador et al., 2015), as well as by ultrasonication or microwave applications (Salvador et al., 2015).

Assisted desorption into the liquid phase can be facilitated by increasing the activity gradient between the sorbed and the freely dissolved compounds. This can be achieved by creating gradients in concentration or solution composition including solvent exchange (Purkait et al., 2007), altered ionic strength (Nur et al., 2014; Purkait et al., 2007), extraction with supercritical fluids (Madras et al., 1993; Tan and Liou, 1989), reactive removal by hydrolysis of previously sorbed compounds (Heilmann et al., 1996; Roth et al., 2016), and temperature.

Temperature swing adsorption or more generally, thermal parametric pumping does not require any dedicated regeneration solution and is based solely on the temperature-dependent sorption equilibrium. This regeneration method thereby resembles the coffee extraction in an Italian moka pot. Thermal parametric pumping has already been employed for several separation tasks, including the recovery of phenol, aniline, and their derivatives from process waste water (Otero et al., 2005b; Rivera-Utrilla et al., 2003), salicylic acid recovery (Otero et al., 2005a, p. 206), and amino acid recovery (Díez et al., 1998). In contrast to desorption into the gas phase, the desorption dynamics into the liquid phase differs from the relationships observed during initial sorption. Recently, this temperature swing adsorption approach was advanced to exploit mild conditions (p ~ p∞, T ≪ 300 °C), employed product water under hydrothermal conditions as sole regeneration agent (not relying on sorbate volatilization), and adapted to the needs of water supply (Aumeier et al., 2019, 2020). This temperature swing adsorption process was developed using the polar herbicide amitrole as model compound. Also others have employed subcritical water as regeneration agent, but used higher temperatures and pressures (Ledesma et al., 2014; Rivera-Utrilla et al., 2003; Salvador and Jiménez, 1996). As in gas-phase applications, temperature swing may be driven by heat exchange, electrical heating (see next paragraph) or microwave irradiation.

To desorb ionic and ionizable contaminants, changes in pH have a high potential in facilitating desorption by affecting the speciation of the sorbed compounds as well as the functional groups on the carbon surface. For example, for acidic contaminants, an increase in pH could cause an increase in negative charge both on the carbon surface as well as in the contaminant, causing electrostatic repulsion. Dissociation is also a temperature-dependent process. Aumeier et al. (2022) recently harnessed the relationship between temperature and solute dissociation to control sorption of ionizable organic contaminants such as benzotriazole to AC. Approaches combining small changes in pH and temperature to desorb compounds at relatively low temperatures may be a promising avenue to explore in the future.

Electro-desorption is based on the tuning of sorption and desorption at conductive porous carbon materials by applying electric potentials, which can facilitate on-site regeneration of sorbents (Saeidi et al., 2021b; Wang et al., 2018; Zhou et al., 2022b). Even though potential-induced sorption and desorption is to some extent compound-specific, its use may be crucial for the removal of ionic contaminants that cannot be well removed by conventional AC. This group of compounds includes for example short- and ultra-short-chained perfluoroalkyl acids. For instance, selective sorption and electro-assisted desorption with high recoveries of ≥95 % at low cell voltages (< 1.1 V) was obtained for trifluoroacetic acid in a small-scale continuous flow experiment with an optimized AC felt (Zhou et al., 2022a). The sorption coefficients (K_d) of short- and long-chain PFAS (perfluorobutanoic acid and perfluorooctanoic acid) on conductive AC felts changed drastically by a factor of 40 to 100 by applying mild anodic (sorption) and cathodic (desorption) potentials at environmentally relevant concentrations (Saeidi et al., 2021b).

All three swing adsorption approaches (temperature swing, pH swing and electro-desorption) are usually deployed in fixed-bed adsorbers. This offers the opportunity for on-site regeneration where desorption is carried out in counterflow mode yielding so-called elution curves. Besides the trigger itself (equilibrium shift), their profile depends on the desorption kinetics and flow velocity or residence time. With the given interdependencies, the optimal regeneration conditions can be found for each individual case from the trade-off between regeneration efficiency and throughput measured as water recovery.

Bioregeneration refers to biodegradation of sorbed organics in a biofilm which forms on the surface of the AC. Bioregeneration can take place simultaneously to sorption and thus does not require any downtime or material replacement. The underlying biological phenomena can be guided via changes in dissolved oxygen, pH and nutrients (Scholz and Martin, 1997; Xiaojian et al., 1991). For example, the bioregeneration of melamine loaded AC can be substantially increased by providing the degrading microorganisms with an additional carbon source such as methanol (Piai et al., 2022). Another study identified nitrogen species (NH_4-N and NO_2-N) as the limiting biomass growth factor under predominantly oxic conditions (Fundneider et al., 2021).

Bioregeneration is often employed downstream of ozonation which increases the biodegradability of organic matter (Reungoat et al., 2012). In contrast, if membrane separation is used prior to AC, biofilm growth can be delayed due to the removal of organic matter acting as substrate for the microbes (Edefell et al., 2022; Fundneider et al., 2021). Moreover, in biological AC filters whose adsorption capacity is at- or close to- saturation, biofilm activity can be fostered by low backwashing intervals in the order of weeks to months instead of hours to days and thus reduced biomass drag-out (Edefell et al., 2022; Fundneider et al., 2021). Moreover, increasing bed contact time can increase contaminant removal, as shown for gabapentin in a bioactive granular AC filter (Fundneider et al., 2021).

For a more targeted degradation of compounds of interest, the inoculation of the AC filter with specialized microorganisms has also been suggested (Aktaş and Çeçen, 2007) and may in the future also involve the use of genetically engineered microorganisms such as the ones suggested for groundwater bioremediation (Janssen and Stucki, 2020). How and to what extent bioregeneration can be utilized for the in-situ degradation of PMT and vPvM substances remains a question for future research that should be based on molecular level understanding using methods to differentiate sorption from (bio)degradation (Yuan et al., 2022).

7. Sorption coupled to destructive technologies

When dealing with highly persistent substances such as PFAS, there is broad consensus that complete mineralization can only be realized after pre-concentration, due to the high energy and/or chemical demand of destructive treatments. Examples of such two-step combinations are nanofiltration and electrooxidation for treatment of concentrates (Soriano et al., 2017). Also, combinations of foam fractionation and non-thermal plasma or electrooxidation treatment of the foam have been proposed (Smith et al., 2022). Concentration factors are typically up to 10-fold in nanofiltration or foam fractionation. In comparison, sorbents can up

concentrate compounds by a factor of 1000 or 10,000 in their pore volume. A range of combined interactions can be utilized for separation via sorption technologies, including hydrophobic effects, size selectivity as well as electrostatic and other specific interactions. Therefore, pre-concentration of contaminants via sorption technologies can be very selective, which can be utilized for subsequent more efficient and targeted destruction. These Thereafter a combination of desorption with effective mineralization is possible even for some of the most recalcitrant substances such as PFAS (Trang et al., 2022).

Carbon-based sorbents are generally not stable against strong oxidants and consume reactive species such as OH or sulfate radicals. Thus, they are not well suited for regeneration by advanced oxidation approaches due to their high oxidant consumption and decline in sorption performance in reuse. In order to exploit the broad-range effectiveness of advanced oxidation approaches in combination with sorption technologies, it is either necessary to desorb the contaminants by one of the above discussed approaches or to use non-carbon-based inorganic high-surface area sorbents which are stable against oxidants and highly reactive radicals produced from ozone, hydrogen peroxide or persulfate. Considering the fact that these oxidants can also be produced by electrochemical approaches and thus renewable energy, such combinations deserve deeper consideration in future.

In contrast to oxidation, certain hydrolysis and reductive approaches are very compatible with carbon-based materials such as AC which can accelerate and support reactions of the sorbed compounds (Mackenzie et al., 2005). For example, the combination of zero-valent iron and AC provides synergies as the carbon can shuttle electrons to sorbed target contaminants and thus increases the active reaction surface by orders of magnitude (Kopinke et al., 2020; Vogel et al., 2019). Similarly, carbon-based electrodes with embedded catalytically active Fe_3O_4 nanoparticles can serve as combined sorbent and electrode. Such a combination was shown to enable a heterogeneous electro-Fenton process at neutral pH without performance loss over 12 cycles (Mohseni et al., 2021).

Electrooxidation of organic compounds occurs by direct oxidation on the anode, indirect oxidation via electrogenerated oxidizing agents, such as hydroxyl radicals (OH•) (Santos et al., 2022), or both of them simultaneously (Radjenovic and Sedlak, 2015). In general, both processes can be combined in a sequence of (i) sorption → desorption → degradation or (ii) sorption → in-situ degradation at the sorbent electrode. The existing literature suggests that (i) and (ii) occur simultaneously if electrooxidation is induced directly in the reactor containing the sorbent electrodes (Chen et al., 2018; Duinslaeger and Radjenovic, 2022; Gineys et al., 2017; Kim et al., 2020; Ormeno-Cano and Radjenovic, 2022). However, the required cell voltages for electrooxidation (typically ≥ 3 V) can oxidize carbon and are thus detrimental for stability and cycleability of combined sorption-electrooxidation approaches processes. Realizing mechanism (i) in

sequential reactors is a promising alternative to overcome this limitation. Therein, the first step is the electrosorption/ − desorption using mild potentials for sorption-optimized electrodes and electro-assisted desorption for delivering a low volume concentrate for post treatment. In a second step, electrooxidation is used for treating desorption concentrate in a separate reactor with oxidation-optimized electrodes such as boron-doped diamond (BDD) or sub-stoichiometric titanium dioxide (TiO_x) with higher stability against harsh electrochemical conditions than conventional carbon electrodes (Radjenovic et al., 2020; Zhuo et al., 2012). Nevertheless, this approach relies on the effective potential-induced desorption of the concentrated contaminants from the sorbent. While this was demonstrated for some PFAS anions, future studies need to explore the applicability of such sequential processes for a wider range of contaminants.

Table 2 provides an overview of sorbents and their compatibility with the abovementioned destructive technologies as well as with on-site regeneration approaches.

8. Trends and implications of current developments

Sorption technologies will remain a valuable tool in future water treatment and will even become more important in view of the growing requirements for PMT and vPvM removal. Carbons such as AC have a long history of successful application with confirmed safety e.g. in drinking water production and certainly remain an indispensable part in future water treatment. Nevertheless, the current view that the combination of AC sorption and its incineration or thermal regeneration can solve virtually all organic contaminant removal tasks may prove to be outdated. Increasing awareness about the risks of PMT and vPvM substances which challenge ACs and the global need to implement sustainability issues in all fields of human activities are strong drivers for a diversification in water treatment technologies and materials. Sustainable and local solutions to water treatment as illustrated in Fig. 1 will be advanced by future research on solving the challenges listed below:

– Phase out of fossil fuels and replacement by decentralized and local renewable energy creates strong advantages for electricity-driven approaches in sorbent regeneration. As, current high-temperature regeneration of AC cannot be easily electrified, novel materials and alternative regeneration approaches in suitable applications are expected to receive substantial attention and be further developed in the years to come.

– In order to reduce the CO_2 footprint in AC production, more efforts towards phase out of bituminous coal-derived carbon and replacement by renewable, regional feedstocks such as agricultural waste, woody residues and sewage sludge should be taken. A number of researchers recently explored the use of such waste materials to produce AC suitable for PMT and

Table 2

Strengths, weaknesses and potential combinations of sorbent-based technologies. AC = activated carbon, fAC = functionalized activated carbon, dAC = defunctionalized activated carbon, resins = ion exchange resins, MOFs = metal organic frameworks.

Material	Removal of small anions	Thermal stability	Oxidative stability	Material cost	On-site regeneration
AC	no	up to 1000 °C	no	+	temperature swing, pH swing, electro-desorption, advanced oxidation or reduction; bioregeneration in fixed bed / if sorbent is retained
fAC	yes if nitrogen and positive charge no if oxygen and negative charge	depends on functionalization	no	+ + +	same as AC depending on functionalization
dAC	yes	no	no	+ +	same as AC but not advanced oxidation
Modified cyclodextrin	yes	some	some	+ + +	concentration purge (solvent extraction), temperature / pH swing to be proven, advanced oxidation / reduction to be proven; bioregeneration if sorbent is retained
Resins	yes	up to 60–140 °C	some	+ +	concentration purge, pH swing (only weak ion exchangers), advanced oxidation or reduction, bioregeneration
MOFs	yes	no but some up to 500 °C	yes	+ + + +	temperature swing (if stable under hydrothermal conditions), pH swing, electro-desorption (if conductive), advanced oxidation or reduction; bioregeneration if sorbent is retained
Zeolites	no except for some removal with high-silica zeolites	up to 800 °C depending on type	yes	+ + +	temperature swing, advanced oxidation/ reduction, bioregeneration if sorbent is retained

vPvM removal from waters (Hagemann et al., 2020; Krahn et al., 2023; Militao et al., 2021). These results indicate that suitable sorbent materials with somewhat lower or similar removal efficiencies compared to commercial AC can be achieved. For example, a steam AC produced from compost residual wood and waste timber could outperform commercial ACs in the removal of PMT substances from biologically treated wastewater (Hagemann et al., 2020). In another study, researchers found that pyrolyzed sewage sludge, which could be produced on-site at wastewater treatment plants, could remove PFAS from water to a similar extent as a commercial AC (Krahn et al., 2023). Open questions remain on the leachability of contaminants from such materials as well as their re-use and (re) generation. Addressing those open questions may enable the development of localized solutions for AC production and regeneration, substantially decreasing logistical and energy costs of AC use in water treatment, which in turn could make shorter cycling of a given sorbent more practicable.

– Adaptation to climate change includes establishing local water cycles for reuse in order to prepare for heavy rain and intense drought periods. For instance, in urban setting such sponge city concepts need to consider water quality issues and at the same time limit the production of waste streams. This is a clear driving force for decentralized solutions with a strong focus on reuse and regeneration that may include swing sorption and bioregeneration. At the same time, wherever possible local nature-based solutions such as advanced soil passage systems should be strengthened as they can additionally contribute to greening of the city (wetlands). More research is needed on how (carbonaceous) sorbents should be best designed to support biodegradation of urban contaminants e.g. by increasing retention times in blue-green infrastructures while maintaining bioavailability.

– Water treatment tasks with demanding and complex contaminant mixtures will increasingly be solved by multi-barrier approaches. Finding the most efficient solutions and combinations of technologies for complying with strict water quality criteria and coping with the abovementioned sustainability issues in water treatment, will be a challenging but also stimulating task for research and fruitful developments in the water treatment sector in coming years.

CRediT authorship contribution statement

Benedikt M. Aumeier: Writing – original draft, Writing – review & editing, Visualization. **Anett Georgi:** Writing – original draft, Writing – review & editing. **Navid Saeidi:** Writing – original draft, Writing – review & editing. **Gabriel Sigmund:** Conceptualization, Writing – original draft, Writing – review & editing, Visualization.

Data availability

No data was used for the research described in the article.

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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