Onafhankelijk
Papendrecht

Aan de Gemeenteraden van de gemeenten Papendrecht, Dordrecht, Molenlanden en Sliedrecht
Alsmede aan de Colleges van B&W van deze gemeenten
Verzonden via de griffies van deze gemeenten

Papendrecht maandag 12 augustus 2024
Onderwerp Saneringsplannen PFAS-schade / vervuiling door Chemours

Geachte dames en heren,

Er zijn plannen in de maak om samen met Chemours tot het saneren van de PFAS in onze regio te komen. Het probleem is echter dat deze plannen voornamelijk van cosmetische aard zijn. Voor het zorgvuldig saneren van met PFAS verontreinigde grond komt heel wat meer kijken.

→ Ten eerste: hoe diep zit de PFAS verontreiniging? Je moet, als je gaat afgraven in elk geval ruim onder de onderste significante PFAS concentraties zitten. Wij denken dan eerder aan **een meter (diepte)** dan aan 20/30/40 cm, maar het hangt af van de locatie;
→ Ten tweede: een laag verse grond van 10 of 20 cm heeft geen enkele zin vanwege de zogenaamde **bioturbatie**: wormpjes en andere kleine organismen woelen de bovenlaag altijd om met als gevolg weer opwelling van PFAS van lagen daaronder;
→ Ten derde: het is daarnaast vermoedelijk zeer verstandig om **een dichte laag** aan te brengen na het afgraven en voor er nieuwe grond op komt **om het grondwater te beschermen**.

In een aantal bij deze brief meegestuurde wetenschappelijke artikelen kunt u lezen dat er veel nadruk wordt gelegd op een zogenaamde **binder**: iets met veel koolstof erin zodat resterende PFAS zich daaraan kunnen binden en eraan gebonden blijven. Los hiervan zijn er meer mogelijkheden. Wat wij u meegeven met deze brief en de meegestuurde artikelen is dat we er als gemeenten niet mee wegkomen om alleen een paar cm af te graven en die te verversen. Overigens zouden wij ons als gemeenten onafhankelijk moeten laten adviseren door onafhankelijke wetenschappers zonder directe en/of indirecte belangen bij de overheid of het bedrijfsleven. Wij stellen voor een team van internationaal gerenommeerde wetenschappers die aan het woord kwamen bij de Zembla documentaires over de PFAS schade.

Met vriendelijke groet,

Ruud Lammers,
Fractievoorzitter Onafhankelijk Papendrecht.

Bijlagen: drie wetenschappelijke artikelen

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Comparison of PFAS soil remediation alternatives at a civilian airport using cost-benefit analysis

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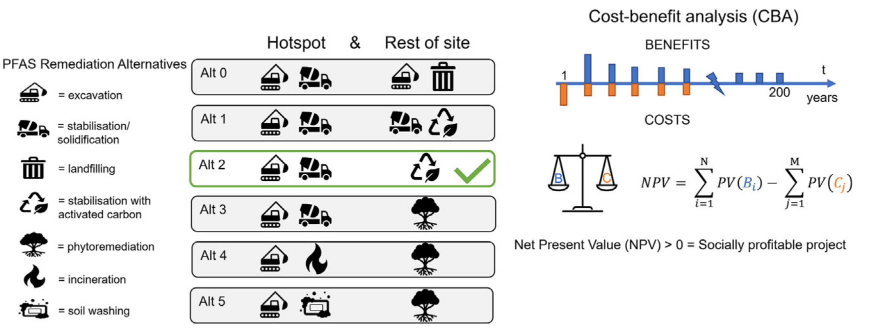
HIGHLIGHTS

- Comparison of remediation techniques for managing a PFAS contaminated site
- Probabilistic cost-benefit analysis to evaluate PFAS remediation alternatives
- PFAS in the environment is associated with high costs of inaction to society.
- Simulation of different annual avoided cost of inaction to find breakeven points
- Ex-situ S/S of hotspot and stabilization of rest of site highest ranked alternative

GRAPHICAL ABSTRACT



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ABSTRACT

Contamination of soil and water systems by per- and polyfluoroalkyl substances (PFAS) due to uncontrolled use of aqueous film-forming foams (AFFFs) at firefighting training sites at civilian and military airports is a universal issue and can lead to significant human health and environmental impacts. Remediation of these sites is often complex but necessary to alleviate the PFAS burden and minimise the risks of exposure by eliminating the hotspot/source from which the PFAS spreads. This study presents a probabilistic cost-benefit analysis (CBA) for evaluating PFAS remediation alternatives, which includes monetisation of both direct costs and benefits as well as externalities. The method is applied for a case study to compare five remediation alternatives for managing PFAS contaminated soil at Stockholm Arlanda Airport in Sweden. The social profitability, or the net present value (NPV), of each remediation alternative was calculated in comparison to two reference alternatives – 'total excavation' of the site (Alt 0) or 'do nothing'. Sensitivity analyses and model scenarios were tested to account for uncertainties, including small or large PFAS spreading and simulating different values for the magnitude of annual avoided cost of inaction (i.e., aggregate benefit) from PFAS remediation. In comparison to total excavation, four of the five studied remediation alternatives resulted in a positive mean NPV. Excavation and stabilization/solidification of the hotspot on-site combined with stabilization using activated carbon for the rest of site (Alt 2) had the highest NPV for both spreading scenarios, i.e., Alt 2 was the most socially profitable alternative. Simulations of the annual avoided cost of inaction enabled estimation of the breakeven point at which a remediation alternative becomes socially profitable (NPV > 0) compared to 'do nothing'. Alt 2 had the lowest breakeven point: 7.5 and 5.75 millions of SEK/year for large and small spreading, respectively.

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1. Introduction

Ubiquitous contamination of per- and polyfluoroalkyl substances (PFAS) in environmental media has become a critical global issue in recent years because of their persistent, bioaccumulative, highly mobile and toxic nature (ITRC, 2020a, 2020b; Wang et al., 2019). One major source of PFAS contamination is the legacy usage of PFAS-containing aqueous film-forming foams (AFFF) in firefighting operations, which has been used since at least the 1960s and gradually phased out by 2011 across Europe (Ahrens et al., 2015; Goldenman et al., 2019). The repeated use and uncontrolled release of AFFFs during firefighting has contaminated the local environment with PFAS chemicals, and firefighting training sites at airports, military facilities, etc. have been highlighted as major point sources (hotspots) of PFAS contamination (Ahrens et al., 2015; Cousins et al., 2016; Goldenman et al., 2019). AFFF sites have been linked to adverse health effects in local populations in recent epidemiological studies, e.g., (Xu et al., 2022).

Environmental fate and transport of PFAS can vary depending on their physicochemical properties as well as environmental conditions but, in general, short-chain PFAS are potentially more water soluble and mobile while long-chain PFAS tend to sorb more strongly to soil particles and can accumulate in the food chain (Ahrens et al., 2015; ITRC, 2020b). Primary exposure pathways relevant for human health risks from PFAS contamination are linked to intake of contaminated food or contaminated drinking water, which may result from elevated PFAS concentrations in groundwater (Cousins et al., 2016; Ojo et al., 2021). Treating PFAS contaminated drinking water is therefore a primary concern; however, the soil in hotspot areas such as at airports can retain PFAS and release it over a long period of time. This can result in subsequent leaching into groundwater, spreading to adjacent surface water and contamination of drinking water sources, which can cause significant harm to both human health and the environment (Cousins et al., 2016; Fenton et al., 2021; Gobelius et al., 2017; Rosenqvist et al., 2017; Sörengård et al., 2021). Indeed, the costs of inaction to society for not managing PFAS contamination in the environment are estimated to be as much as €2.1–2.4 billion annually in the Nordic countries alone, due to health impact-related costs from PFAS-contaminated drinking water (Goldenman et al., 2019). It is therefore important to also manage the hotspots of PFAS soil contamination effectively; for the longer the PFAS contamination remains in the environment without remediation, the wider it will spread and the greater the quantity of soil, groundwater and other drinking water sources that will need be treated (Goldenman et al., 2019).

PFAS exposure has been definitively linked to multiple detrimental health effects to humans, including different types of cancer, osteoporosis, liver damage, decreased fertility and increased risk of asthma, reduced immune response and endocrine disruption (ATSDR, 2021; Ojo et al., 2021; Wang et al., 2019; Xu et al., 2022). While there are still uncertainties due to a lack of data, PFAS exposure has also been shown to have toxic effects on aquatic animal species and is linked to endocrine disruption and impaired thyroid function, impaired immune responses, metabolism and reproduction disruption (Birgersson et al., 2021; Lee et al., 2020; Ojo et al., 2021; Wang et al., 2019), as well as a reduction in soil bacterial biodiversity (Cao et al., 2022). It is now clear that PFAS contamination is poorly reversible, ubiquitous in the environment, and the social costs of inaction are and will continue to be high (Goldenman et al., 2019). Remediation of PFAS contaminated sites, therefore, is crucial to mitigate the risks from PFAS exposure to both humans and ecosystems by managing the source of PFAS contamination in soil and/or spreading and exposure pathways. However, in some cases the site-specific costs of PFAS remediation may exceed the expected benefits gained from the remedial action at a particular site which warrants careful consideration when spending limited resources.

In addition, remediation is not inherently sustainable (Bardos et al., 2020; Rosén et al., 2015; Söderqvist et al., 2015) and selecting a remediation alternative to manage the risks posed by the PFAS contamination can be difficult for decision-makers due to the associated costs and e.g., potential impacts on provisioning of ecosystem services. Cost-benefit analysis (CBA) is a decision-support tool that relies on welfare economics for expressing positive (benefits) and negative (costs) effects on human well-being including both financial costs and benefits as well as positive and negative externalities (i.e., positive or negative effects on health and the environment in terms of provisioning of ecosystem services, carbon emissions, noise, traffic etc.). Using monetary units makes it possible to weigh the costs of a remedial action against associated benefits over a certain time horizon and in relation to a reference alternative. A positive net sum of discounted costs and benefits means that the remedial action entails a social profitability, whereas a negative net sum indicates social loss (Johansson and Kriström, 2018; Rosén et al., 2015; Söderqvist et al., 2015). CBA has been highlighted as a decision-support method with great potential for incorporating sustainability measures in an understandable, easy-to-use approach and account for the value of restoring or preserving soil functionality and ecosystem services (ES) (Onwubuya et al., 2009). Additionally, economic valuation of ES contributes to the decision-making process by integrating ES into decision-support and engaging potentially responsible parties to participate in both the remediation process and funding of risk mitigation measures (Harwell et al., 2021). The novelty of this study lies in its systematic approach to evaluating feasible techniques for remediating PFAS in soil, according to prevailing literature, and estimating the economic impacts of each alternative while also including impacts to the environment and taking uncertainties into account. To our knowledge, applying a probabilistic CBA methodology for evaluating and comparing PFAS remediation alternatives is unique. Given the scale of PFAS contamination and society's limited resources, demonstrating the use of CBA in a case study for a specific site also provides a valuable contribution by supporting decision-makers to cost-effectively and sustainably remediate a PFAS contaminated site.

The aim of this study is to further develop a methodology for performing a probabilistic CBA of remediation alternatives for managing PFAS contamination in soil. The CBA is illustrated through practical application to evaluate five remediation alternatives for managing the risks to human health and the environment posed by PFAS in the soil at the firefighting training site of Stockholm Arlanda Airport. The specific objectives are to: i) develop a probabilistic CBA model for five PFAS remediation alternatives, ii) estimate the costs and benefits of the remediation alternatives based on literature studies and personal communication with contractors, and by taking uncertainties in the input variables of the model into consideration; and iii) investigate the sensitivity of the CBA model regarding both parameter and model uncertainty. The CBA is carried out by means of Monte Carlo simulations and both parameter and model uncertainty are investigated. The model uncertainty is analysed by creating alternative scenarios for: a) choice of reference alternative, b) the social discount rate, c) two PFAS spreading scenarios, and d) the magnitude of avoided cost of inaction.

2. Site: Stockholm Arlanda Airport

2.1. Site description

The firefighting training site is situated at Stockholm Arlanda Airport outside of Stockholm, Sweden, where AFFF-containing PFAS was used until 2011 (Gobelius et al., 2017).

The geology consists primarily of surface layers of glacial clay underlain by sandy glacial till which varies between a depth of 1.5–8 m below the surface depending on the thickness of the clay (Rosenqvist et al., 2017). The glacial till is deposited on crystalline metamorphic rock of igneous origin. To the immediate north and northeast of Stockholm Arlanda Airport, layers of sand (glacio-fluvial or beach deposits with silt) have been deposited on top of the clay layer. The firefighting training site is located within one such area with a top layer of beach sand and silt, varying between 0.3 and 2 m in thickness, that thins out and disappears altogether closer to the landing strips southwest of the training site. Filling material of sand and gravel form the immediate surface layer of 0.5 m in the built area above the natural geological soil layers. Hydrogeological investigations have determined that there are two distinct aquifers: an unconfined aquifer in the upper layer of sand and silt above the clay and a confined aquifer in the sandy glacial till below. The upper aquifer is contaminated with PFAS and constitutes an important spreading pathway for PFAS off-site to nearby surface water systems. At the training site, the groundwater depth ranges between 1.1 and 1.8 m across the site, but the water table is at or near the surface layer in some areas. It has also been determined that the groundwater flows in a south-westerly direction, towards a nearby open ditch that is in hydraulic contact with nearby surface water but away from and not in contact with the glacio-fluvial sand deposits to the northeast of Stockholm Arlanda Airport with high hydraulic conductivity (Rosenqvist et al., 2017).

Sampling campaigns at the site have extensively investigated PFAS concentrations in soil, sediment, groundwater, surface water and aquatic organisms. Gobelius et al. (2017) took soil samples at three locations within 500 m of the site in the direction of groundwater flow to a depth 10 cm. The sum total of the 26 PFAS analysed in the soil samples ranged from 20 to 160 ng g^{-1} dry weight (dw). Groundwater samples at the same locations showed concentrations ranging from 1200 to 34,000 ng L^{-1}. A more extensive soil sampling campaign by Rosenqvist et al. (2017) analysed 40 soil samples and reported even higher maximum values with significant variation between the different types of PFAS compounds in concentration as well as spreading distance from the source (see Table S1 and Figs. S1 and S2 in Supplementary Material (SM)). The sum total of the 13 analysed PFAS compounds ranged from 0.63 ng g^{-1} to 2700 ng g^{-1} dw. They found that PFOS (a subset of PFAS compounds) made up 88 % of the PFAS compounds measured in soil with an average value of 234 ng g^{-1} dw across the site and PFHxS, PFHxA and PFOA were the next highest in concentration. An important note is the median value of 34 ng g^{-1} dw, indicating large differences in measured concentrations closer to the source (the training site hotspot) versus further downstream away from the immediate source. The depth to which the soil is contaminated with PFAS varies considerably between the immediate hotspot and soil layers throughout the rest of the site.

For comparison, preliminary guidelines have been established by the Swedish Geotechnical Institute (SGI) which provide a soil guideline value of 20 ng g^{-1} dw for PFOS for "less sensitive land use," e.g., industrial use, and 3 ng g^{-1} dw for "sensitive land use," e.g., residences or recreation, to protect human health and the environment (Pettersson et al., 2015). The guideline value for groundwater is 45 ng L^{-1}. The tested concentrations in both soil and groundwater greatly exceed the guideline values in many sampling locations.

2.2. Extent of PFAS contamination at the site

The size of the contaminated area is difficult to estimate, and no reliable figure could be found in existing reports. In fact, two firefighting training sites have been noted – a new and an old site – though only the newer one is included in this analysis (see the marked area H in Fig. 1). A rough approximation was made using an online mapping tool (Eniro) by delineating the square area to include the soil sampling points and the firefighting training site itself (Fig. 1). Remediation of the PFAS soil contamination for the firefighting training site at Stockholm Arlanda Airport has been separated into two components: remediation of the 'hotspot' and remediation of the 'rest of site'. There are low uncertainties with respect to the size of the hotspot area at the study site; however, contamination spreading in the rest of the area is highly uncertain since PFAS are persistent, mobile and spread widely in both soil and water systems. Therefore, two different PFAS contamination scenarios were evaluated in this study to account for a 'small' and 'large' contamination spreading for the rest of the site (Fig. 1).

3. Methods

3.1. Probabilistic CBA modelling

In a CBA, cost and benefit items of remediation alternatives are monetised in comparison with a reference alternative. The cost and benefit items are discounted over a time horizon of 120 years using a real social discount rate of 3.5 %, as recommended for CBA in Sweden (STA, 2020). Present values (*PV*) for each alternative and the net present value (*NPV*) are calculated using as follows (Eqs. (1), (2)) (Söderqvist et al., 2015):

$$NPV = \sum_{i=1}^{N} PV(B_i) - \sum_{j=1}^{M} PV(C_j), \tag{1}$$

$$PV(B_i) = \sum_{t=0}^{T} \frac{\mathbf{1}}{(\mathbf{1}+\boldsymbol{r})^t} B_{it} \text{ and} PV(C_j) = \sum_{j=0}^{T} \frac{\mathbf{1}}{(\mathbf{1}+\boldsymbol{r})^t} C_{jt}, \tag{2}$$

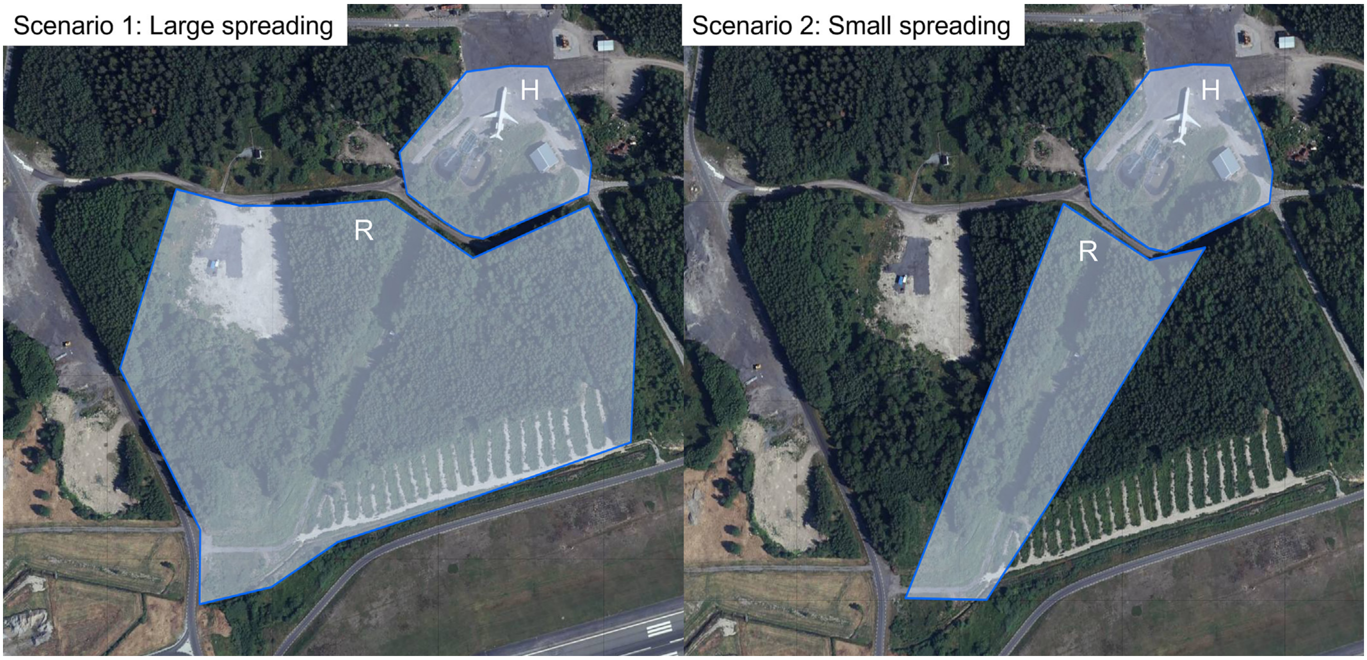

Fig. 1. Forecasted spreading of PFAS contamination in Scenario 1 and 2 based on site investigation (Rosenqvist et al., 2017). H is a hotspot area. R is the rest of the contaminated site.

where T is the time horizon, r is the social discount rate, and t is the time when benefits and costs occur for each benefit item (B_i, $i = 1 \ldots N$) and cost item (C_j, $j = 1 \ldots M$).

The most profitable remediation alternative for society is that with the highest positive *NPV*. If all the *NPV*s are <0, then the remediation alternative with the lowest negative *NPV* results in the least social loss in economic terms.

The CBA was carried out by adapting the method presented in Söderqvist et al. (2015) and Volchko et al. (2020), according to the following steps:

1. Identification of remediation alternatives, a reference alternative, the social discount rate and a relevant time horizon associated with the alternatives.
2. Identification of costs and benefits associated with each remediation alternative and defining scenarios to account for model uncertainties.
3. Quantification and monetization of costs and benefits by defining a minimum, maximum, and most likely value based on literature studies and personal contact with contractors and assigning probability distributions to input variables and cost and benefit items to represent the uncertainties in these input variables.
4. Calculating the *NPV* and associated uncertainties of each alternative by using Monte-Carlo simulations and discounting the cost and benefit items using a social discount rate and a relevant time horizon, simulating the CBA for the different defined scenarios, and investigating the results to evaluate the uncertainties in *NPV*s of the remediation alternatives and performing sensitivity analyses.
5. Concluding about the social profitability and ranking of remediation alternatives to provide recommendations as decision-support.

The cost and benefit items relevant for CBA in the remediation project and the methods used to quantify them are presented in the SM (Table S2).

The probabilistic CBA model was set up in MS Excel using the Palisade add-in software @Risk 8.2 for defining uncertainty distributions for input variables (Table S3). Monte Carlo simulations were run 10,000 times by repeatedly picking random values from the probability distributions of input variables to calculate the *NPV*s (Bedford and Cooke, 2001). The probability of each remediation alternative generating *NPV* > 0 is calculated using the RiskTarget feature of @Risk 8.2.

3.2. CBA model: base scenario

A 'base scenario' was defined as the default model settings with which to compare the remediation alternatives. The default parameters for the base scenario are a discount rate of 3.5 %, time horizon of 120 years (~4–6 generations), 7.5 MSEK for 'annual avoided cost of inaction' (in the CBA corresponding to the benefit items B2-B3, see Table S2 in SM), and 20 years for time of risk reduction by phytoextraction. Also, in the base scenario, the mean *NPV*s of Alt 1–5 (Table 2) were evaluated in comparison to the 'total excavation' remediation alternative as a reference, Alt 0, to demonstrate the positive or negative effects of each alternative compared to the conventional remediation technique, which is also the 'most likely' remediation alternative to be first considered for the site if remediation is mandated.

Alt 0 assumes total excavation of the entire site to a depth just above the clay layer (0–2.5 m), treatment of the hotspot (and backfilling) and disposal of the material from the rest of site (Table 2). Alt 0 is thus a reference situation entailing complete risk reduction, which from a duty-based ethical perspective could be argued as the most correct thing to do for achieving environmental protection targets and protecting future generations despite the potentially high costs, environmental impacts or other externalities. This reference can also represent a 'business-as-usual' case for remediation of contaminated soils by excavation and disposal, which is the most common remediation technique used in Sweden and many other countries, and is useful to evaluate against other remediation alternatives to determine whether they are more socially profitable in comparison. However, it was modified by employing on-site stabilization/solidification (S/S) ex-situ on-site (hotspot) and ex-situ off-site (rest of site), since disposing of PFAS-contaminated soil is currently not permitted at disposal sites in Sweden (SEPA, 2019).

3.3. Parameter and model uncertainties

For investigating the sensitivity of the input variables in the probabilistic CBA model, Spearman rank correlation coefficients were calculated by @Risk 8.2 (Palisade).

In addition, multiple model 'scenarios' were defined to test different model assumptions and sensitivity of model uncertainties. The following scenarios were defined and tested: a) choice of reference alternative, b) the social discount rate, c) two PFAS spreading scenarios, and d) the magnitude of annual avoided cost of inaction. The various scenarios and how they were simulated to account for uncertainties in the model are summarized in Table 1.

3.3.1. Reference alternative

Moral and legal obligations to remediate PFAS contamination suggest that a 'do nothing' option is untenable, but nevertheless is a common reference (Söderqvist et al., 2015; Volchko et al., 2020) for investigating when a project, in this case a remedial action, becomes socially profitable (*NPV* > 0). Therefore, leaving the site in its current state (i.e., 'do nothing') was considered as an alternative reference for comparing the mean *NPV*s of the remediation alternatives (including Alt 0) thus providing a different basis with which to consider the overall value of remediating a specific site.

3.3.2. Social discount rate

The sensitivity of the *NPV*s to changes in a social discount rate was tested for each studied remediation alternative Alt 1–5, compared to Alt 0. The following social discount rates were tested: 0, 1.7, 3.5, 5 and 7 %, with 3.5 % as the 'base scenario' as done in Söderqvist et al. (2015).

3.3.3. PFAS spreading

The size of the hotspot has remained constant for the remediation alternatives, however, the conceptual uncertainty in the extent of the PFAS contamination ('rest of site') has been accounted for by creating two separate scenarios – large or small spreading – as shown in Fig. 1.

3.3.4. Annual avoided cost of inaction (AACOI)

Annual avoided cost of inaction (AACOI) represents the aggregated benefit of avoided societal costs from PFAS contamination due to remediation (B2-B3, Table S2 in SM). These costs of inaction include at least costs due to negative impacts on human health (e.g., kidney cancer, all-cause mortality, increased infection risk, hypertension) and the (non-health) environment-related costs such as upgrading drinking water treatment plants, ongoing (bio)monitoring and remediation costs amongst others (Goldenman et al., 2019). For modelling purposes, a value of 7.5 MSEK was used as a fixed amount in the 'base scenario', which results in a present value of ca.

Table 1

The different scenarios for model parameters accounted for in the study. Total area sizes were considered for two studied scenarios measured using Eniro mapping tool. PERT is the PERT-beta distribution, Min is minimum, M-likely is the most likely value, Max is maximum, ha is hectares.

Scenario analysis	Parameters	Reference/comment
a) Reference alternative	i) Alt 0 – total excavation ii) 'Do nothing'	
b) Social discount rate (%)	0, 1.7, 3.5, 5 and 7 %	(STA, 2020)
c) Spreading scenarios (PERT dist.):		
- Large spreading – total area (ha)	Min: 11; Max: 15; M-likely: 13	(Gobelius et al., 2017; Rosenqvist et al., 2017)
- Small spreading – total area (ha)	Min: 3; Max: 7; M-likely: 5	
d) Annual avoided cost of inaction (MSEK)	6 simulations: 5, 7.5, 10, 12.5, 15, 25	

196.59 MSEK or €18.03 million (1€ is ca. 10.9 SEK as of December 2022). However, it is important to note that this is not an estimate of a site-specific avoided cost of inaction but rather as a tested value for PFAS remediation benefits to evaluate potential social profitability and compare alternatives. The site-specific AACOI is unknown because the reported costs of inaction are based on assumptions for large, aggregated sums for a country or the Nordic region, so a reliable value for how much of this damage is ascribed to a particular site is still prohibitively difficult to determine. Given this uncertainty, a scenario analysis was conducted to test the sensitivity of the model for different values for AACOI – 5, 7.5, 10, 12.5, 15, and 25 MSEK. The simulations were used to find the 'breakeven points' at which each remediation alternative becomes socially profitable (*NPV* > 0) in comparison to 'do nothing' that could provide valuable decision-support when evaluating potential remediation alternatives and more data is available to monetize the local impacts of PFAS contamination more accurately.

4. Results

4.1. PFAS remediation alternatives for the site

As emerging contaminants have gained widespread attention only in recent years, remediation technologies to immobilise, remove or destroy PFAS and its associate compounds are not yet well-established (Held and Reinhard, 2020; ITRC, 2018; Ok et al., 2020; Smith et al., 2016). Indeed, a combination of multiple technologies (i.e., treatment chains (Lu et al., 2020)) is often required to remediate a site effectively (ITRC, 2018; Merino et al., 2016). Gentle remediation options (GRO) – nature-based solutions using combinations of plant, bacteria, fungi and soil amendments – are considered for their potential to manage PFAS contamination as well as improve (or at least not reduce) soil functioning while producing useful biomass as part of a phytomanagement strategy (Cundy et al., 2016). Also, in defining remedial goals, it is important to consider which specific PFAS are considered to pose a risk according to the risk assessment and determine the specific 'risk driver' for the site by considering the source-pathway-receptor linkages ('contaminant linkages') and how best to manage them (Held and Reinhard, 2020; Ross et al., 2018). Many reviews on PFAS remediation options have been carried out which show that there are some promising technologies and strategies to manage PFAS contaminant linkages (Bolan et al., 2021; Held and Reinhard, 2020; ITRC, 2018; Mahinroosta and Senevirathna, 2020; Ok et al., 2020; Ross et al., 2018; Smith et al., 2016).

Five remediation alternatives were developed, where each alternative is a combination of several technologies for managing the risks posed by PFAS contamination in soils, summarized in Table 2 and described in detail in SM2.

4.2. CBA results: the base scenario

The simulated mean present values of cost and benefit items for a discount rate of 3.5 % and time horizon of 120 years are shown in Table 3. These values were used in the CBA to calculate *NPVs* for the respective PFAS remediation alternatives for both large and small spreading scenarios.

4.2.1. Net present values

The outcome of the probabilistic CBA model for the 'base scenario' is shown in Fig. 2. Alt 2 (excavation and S/S of the hotspot and stabilization of PFAS at the rest of the site with activated carbon) generates the greatest mean *NPV* for both the large and small spreading scenarios, 123 MSEK and 14.1 MSEK, respectively. The results indicate that all studied remediation alternatives except for Alt 4 are associated with remediation cost savings (Table S4 in SM) compared to Alt 0. This is valid for both spreading scenarios. The ranking of the other alternatives varies depending on the spreading scenario. For the small spreading scenario, Alt 1 and Alt 2 generate an almost equally positive mean *NPV*. Alt 3 and Alt 5 generate a slight negative mean *NPV* in the small spreading scenario but have the second highest mean *NPV* in the large spreading scenario. The mean *NPV* of Alt 4 is substantially negative in both spreading scenarios.

Table 2

Overview of the remediation alternatives for PFAS-contaminated soils at the Stockholm Arlanda Airport site. REF indicates the reference alternative used in the CBA.[a]

CBA of Alt 1–5 compared to Alt 0, i.e., 'total excavation' (base scenario)		REF	Remediation alternatives evaluated against Alt 0				
		Alt 0	Alt 1	Alt 2	Alt 3	Alt 4	Alt 5
CBA of Alt 0–5 compared to the 'do nothing' case	REF	Remediation alternatives evaluated against 'Do nothing'					
	Do nothing	Alt 0	Alt 1	Alt 2	Alt 3	Alt 4	Alt 5
Remedial actions at the hotspot							
Excavation (before treatment)		X	X	X	X	X	X
Ex-situ stabilization/solidification (S/S) with cement and activated carbon on-site		X	X	X	X		
Ex-situ thermal treatment off-site						X	
Ex-situ soil washing On-site							X
Backfilling with the treated masses		X	X	X	X		X
Backfilling with pristine soils						X	
Remedial actions at the rest of the site							
Excavation (before treatment or disposal)		X					
In-situ stabilization/solidification (S/S) with cement and activated carbon			X				
In-situ immobilisation/stabilization with activated carbon without cement				X			
Phytoremediation with birches and spruces					X	X	X
Landfilling at a disposal site		X					
Backfilling with pristine soils		X					
Achievement of risk reduction targets (years required to manage risks)							
Hotspot	–	2	2	2	2	2	2
Rest of site	–	2	2	2	20	20	20
Long-term project management and monitoring	–	0	0	20[b]	20[b]	20[b]	20[b]

[a] CBA: cost-benefit analysis.

[b] It is assumed that risk reduction can take a shorter time, but the site may not be left without monitoring and adaptive management when using gentle remediation options (Drenning et al., 2022).

4.2.2. Reduced negative externalities

An additional point of comparison for the remediation alternatives is the potential generation of reduced negative externalities (i.e., negative effects on health and the environment in terms of provisioning of ecosystem services, avoided carbon emissions, noise, traffic accidents etc.) as a result of the remedial action (Fig. 3). The reference alternative (Alt 0) generates substantial negative externalities due to the remedial action, and any alternative that generates reduced negative externalities will therefore result in reduced costs (shown as a 'negative cost' in Table S4). In comparison to Alt 0, all alternatives, except for Alt 4 in the small spreading scenario, are associated with reduced negative externalities during the remedial action compared to Alt 0. Alt 1 is just slightly better than the reference Alt 0 with respect to externalities during remedial action. Alt 4 is even worse than the reference alternative in the small spread scenario because of more extensive air emissions and noise from the ex-situ thermal treatment of the hotspot. However, the externalities are associated with large uncertainties (shown as error bars in Fig. 3) in the large spreading scenario in particular, and Alt 4 may generate even more negative externalities than Alt 0 in the large spread scenario too.

4.3. Uncertainty analysis

4.3.1. Parameter uncertainty

The calculated Spearman rank correlation coefficients for each studied remediation alternative are presented in Tornado charts (Figs. S5–S14, SM). Regarding uncertainties and sensitivity of input variables associated

Table 3
Summary of cost and benefit values used in the cost-benefit analysis to calculate net present values. Mean PV: the mean present value of cost and benefit items. L: Large spreading scenario. S: Small spreading scenario. The annual avoided cost of inaction (B2-B3) in Alt 0-Alt 5 is assumed to be 7.5 MSEK. The social discount rate is 3.5 %. The time horizon is 120 years.

Time horizon (years): 120 Discount rate: 3.5 %		Alt 0 S/S hotspot & disposal rest		Alt 1 S/S hotspot & S/S rest		Alt 2 S/S hotspot & Stabilization AC		Alt 3 S/S hotspot & Phytoremediation		Alt 4 T/T hotspot & Phytoremediation		Alt 5 SW hotspot & Phytoremediation	
Category	Item	Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)	
Benefit categories and items													
Spreading scenarios		L	S	L	S	L	S	L	S	L	S	L	S
B2-B3. Avoided cost of inaction	Improved health and increased provision of ecosystem services	197	197	197	197	197	197	177	177	177	177	177	177
Cost categories and items													
C1. Remediation costs	C1a-e.I. Short-term costs (total area)	109	60.0	80.4	52.1	67.5	48.6	129	75.9	322	268	129	75.3
	C1b.II,C1e.II. Long-term costs of management, monitoring (rest of the site)	0	0	0	0	7.39	7.39	7.39	7.39	7.39	7.39	7.39	7.39
	C1f. Project risks	32.1	17.3	23.4	14.9	19.6	13.9	16.8	13.1	74.4	70.8	16.6	12.9
	Total C1	141	77.3	104	67.0	94.5	69.8	154	96.4	404	346	153	95.6
C2. Impaired health due to remedial action	C2b. From transport activities and C2c. At a disposal site	0.709	0.201	0.0346	0.015-7	0.0197	0.0116	0.00854	0.00852	0.401	0.400	0	0
C3. Decreased provision of ecosystem services due to remedial action (C3a-C3c)		71.8	82.5	10.1	79.3	9.23	6.57	3.68	2.58	2.58	32.3	32.3	4.76

with the remediation alternatives in the base scenario, Alt 4 is shown to have the largest variability of the mean *NPV* (Fig. 2), where the size of the hotspot and the associated thermal treatment cost contribute most to the variability. Uncertainties regarding costs of phytoremediation contribute most to the variability of the mean *NPV* of Alt 3–5 (except for Alt 5 in the small spreading scenario) in both spreading scenarios. However, if PFAS spreading is small, hotspot size and the associated costs for soil washing of Alt 5 contribute most to the variability of the mean *NPV* of this alternative. Damage costs associated with tree clearing at the site for Alt 1 in the large spreading scenario is the input variable that contributes most to the variability in the mean *NPV* for this alternative. However, for the small spreading scenario, the size of the hotspot and the cost associated with S/S contribute most to the variability of the mean *NPV* of Alt 1.

4.3.2. Social discount rate

In the base scenario, regardless of which of the social discount rate levels is used, Alt 2 is highest ranked of the remediation alternatives with respect to *NPV* for both the large and small spreading scenarios (Table S5, SM). The next highest-ranking alternative differs in the large or small spreading scenario and varies between Alt 3 or Alt 1 being the second highest, respectively, followed closely by Alt 5. Alt 4 is the lowest ranked alternative for all the social discount rate levels and spreading extent of PFAS at the site.

4.3.3. 'Do nothing' as reference

When 'do nothing' is used as reference, all alternatives, including Alt 0 but excepting Alt 4, generate a positive mean *NPV* for the base scenario

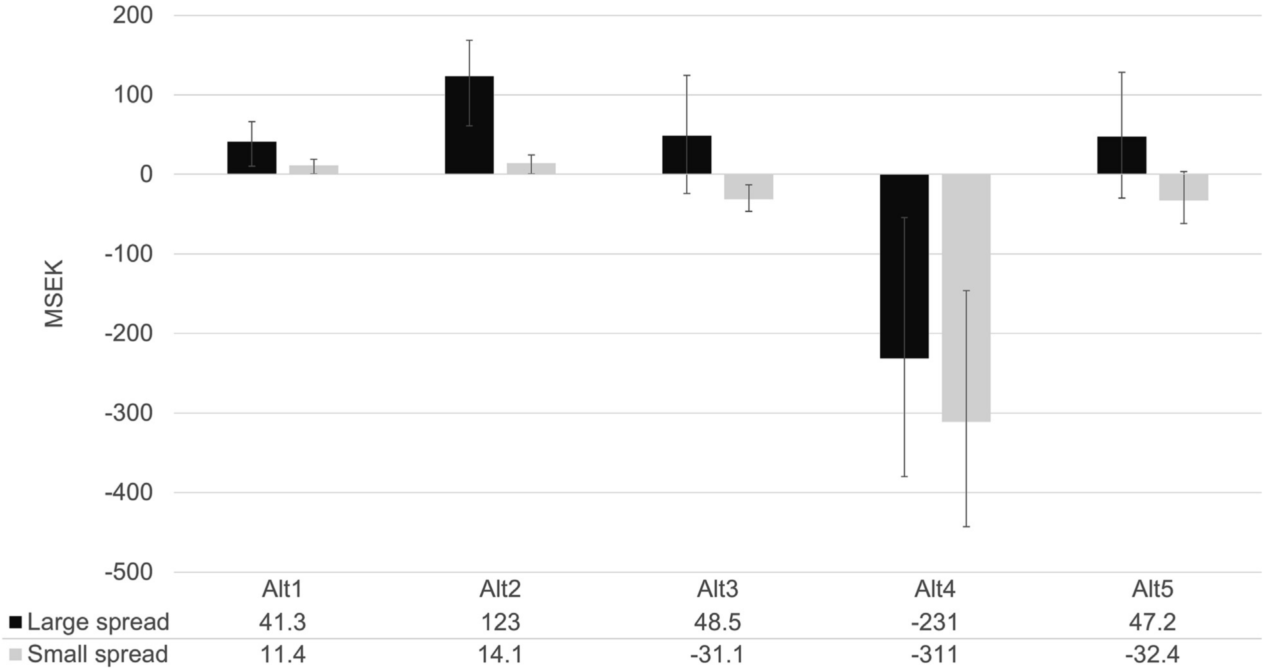

Fig. 2. The simulated mean of the net present values (NPV) for Alt 1–5 in comparison to Alt 0 as the reference alternative; the 5th and 95th percentiles are shown as error bars. The values in the data table below the chart area represent the simulated mean values of the NPV for each alternative and spreading scenario in millions of SEK (MSEK).

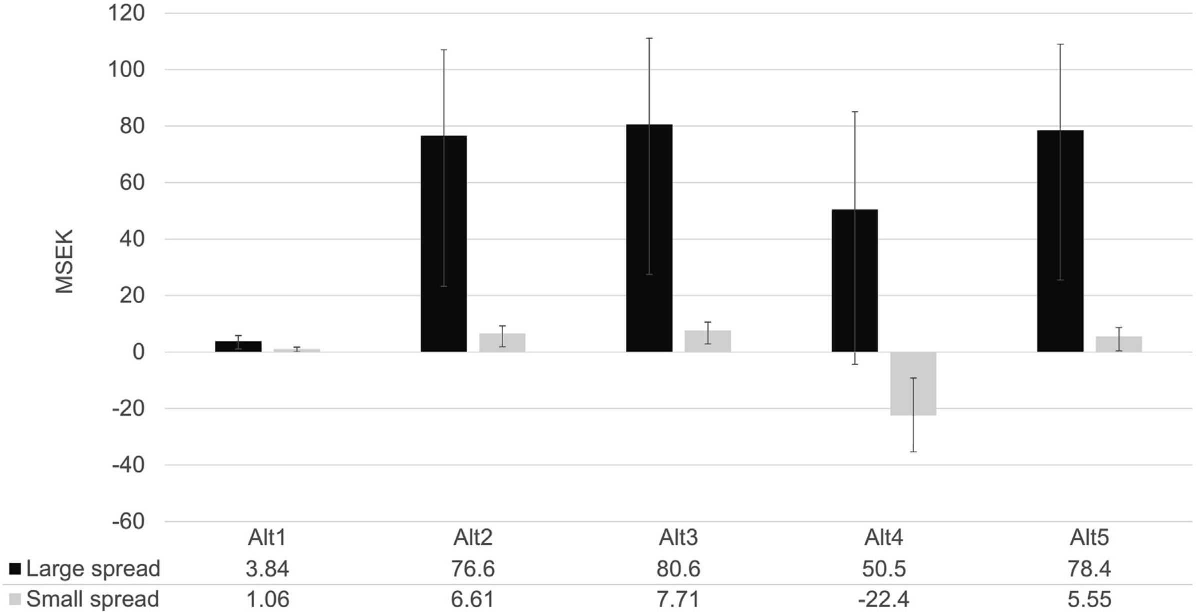

Fig. 3. Cost reductions in terms of reduced negative externalities for remediation alternatives Alt 1–5, in comparison to Alt 0. The 5th and 95th percentiles are shown as error bars. The values in the data table below the chart area represent the simulated mean values of the reduced negative externalities for each alternative and spreading scenario in millions of SEK (MSEK).

with an annual cost of inaction of 7.5 MSEK in the small spreading scenario, social discount rate of 3.5 %, and time horizon of 120 years (Fig. S13, SM). However, only Alt 2, Alt 3 and Alt 5 generate a positive mean *NPV* in the large spreading scenario, though with larger uncertainties for Alt 3 and Alt 5. The mean *NPV* for Alt 2 compared to the 'do nothing' alternative is the greatest for both spreading scenarios (95.4 MSEK and 123 MSEK for large and small spreading, respectively). All remediation alternatives generate negative externalities when compared to the 'Do nothing' reference alternative; however, the alternatives utilising gentle remediation options (GRO) without thermal treatment (Alt 2, Alt 3, Alt 5), incurred the least negative externalities (Fig. S14, SM). Tables compiling the present values (*PV*) of each cost and benefit item as well as resulting mean *NPV* for each alternative compared to the 'do nothing' reference alternative for both large and small spreading scenarios are available in the SM (Table S6).

4.3.4. Annual avoided cost of inaction

The sensitivity of the outcome of the CBA in relation to the 'annual avoided cost of inaction' (AACOI, i.e., the aggregated benefit of B2-B3) is investigated by identifying at which value of AACOI an alternative is socially profitable (*NPV* > 0) with at least 50 % probability. This value is referred to as the 'breakeven point' and is found at the cross-section with the red dashed line in Fig. 4 for the two modelled spreading scenarios (large and small spreading) compared to 'Do nothing'.

Alt 2 has the lowest value of the breakeven point: an AACOI of approximately 7.5 and 5.75 MSEK for large and small spreading of PFAS, respectively. The difference in breakeven points between alternatives is clearly distinguishable in the large PFAS spreading scenario. Alt 2 is socially profitable with a very high probability (>90 %) at an AACOI of ca. 9 MSEK, but the AACOI would have to be at least 12.5 MSEK/year to make Alt 1, Alt 3 and Alt 5 socially profitable with a probability >90 % or 20 MSEK/year for Alt 0. In the small PFAS spreading scenario, all alternatives, including Alt 0 but excepting Alt 4, have similar breakeven points of avoided cost of inaction (ca. 5.5–7 MSEK) for generating an *NPV* > 0 (for details see Table S7, SM). An avoided cost of inaction of at least 8 MSEK/year will generate a positive NPV for Alt 0, 1, 2, 3 and 5 with a probability of at least 85 % in the small spreading scenario.

5. Discussion

5.1. Ranking and associated impacts of the PFAS remediation alternatives

In this study, combinations of soil remediation technologies to manage both the hotspot and rest of site area were evaluated using CBA. Each alternative entails both distinct advantages and disadvantages, which is reflected in their resulting rankings for each modelled scenario (Table S8 in SM). An important note is that the relative rankings did not change significantly as a result of differing values simulated for AACOI, but the rankings change depending on whether the modelled PFAS spreading and resulting size of the 'rest of site' is large or small.

The intensive hotspot remediation over a short period of time (2 years) generates much of the direct health and environmental benefits from the PFAS remediation; however, the techniques differ with regard to total cost and externalities. Excavation of the hotspot followed by S/S on-site (Alt 0 and Alt 1–3) has the lowest remediation cost but requires an extensive use of cement, which is associated with large carbon emissions from production of the cement and transportation. For calculating the cost of carbon emissions, the cement is assumed to be produced in the EU and thus within the EU greenhouse gas emission trading system (see SM4.6 for calculation details). Thermal treatment (Alt 4) is both expensive and carbon intensive due primarily to transportation and energy requirement from operating the facility, which resulted in a large cost that caused this alternative to be ranked last in all tested scenarios. Soil washing (Alt 5) of the hotspot soil was estimated to be more expensive than excavation and S/S but would entail much lower negative externalities due to lower carbon emissions of the remedial action due to e.g., not requiring transportation of heavy trucks for remediation or backfilling since the treated masses are assumed to be reused on site (Table S4 in SM for respective cost and benefit estimates).

The remedial techniques considered for the 'rest of site' were more varied and the assumed time required for the remedial action ('time of risk reduction') differed between the alternatives. Alt 0–1 are assumed to achieve the risk reduction targets within the same 2-year timespan as the hotspot remediation, but the equivalent time for Alt 3–5 was estimated to be 20 years. Alt 2 uses an activated carbon stabilizing agent to achieve the rapid risk reduction but will require project management and monitoring costs for

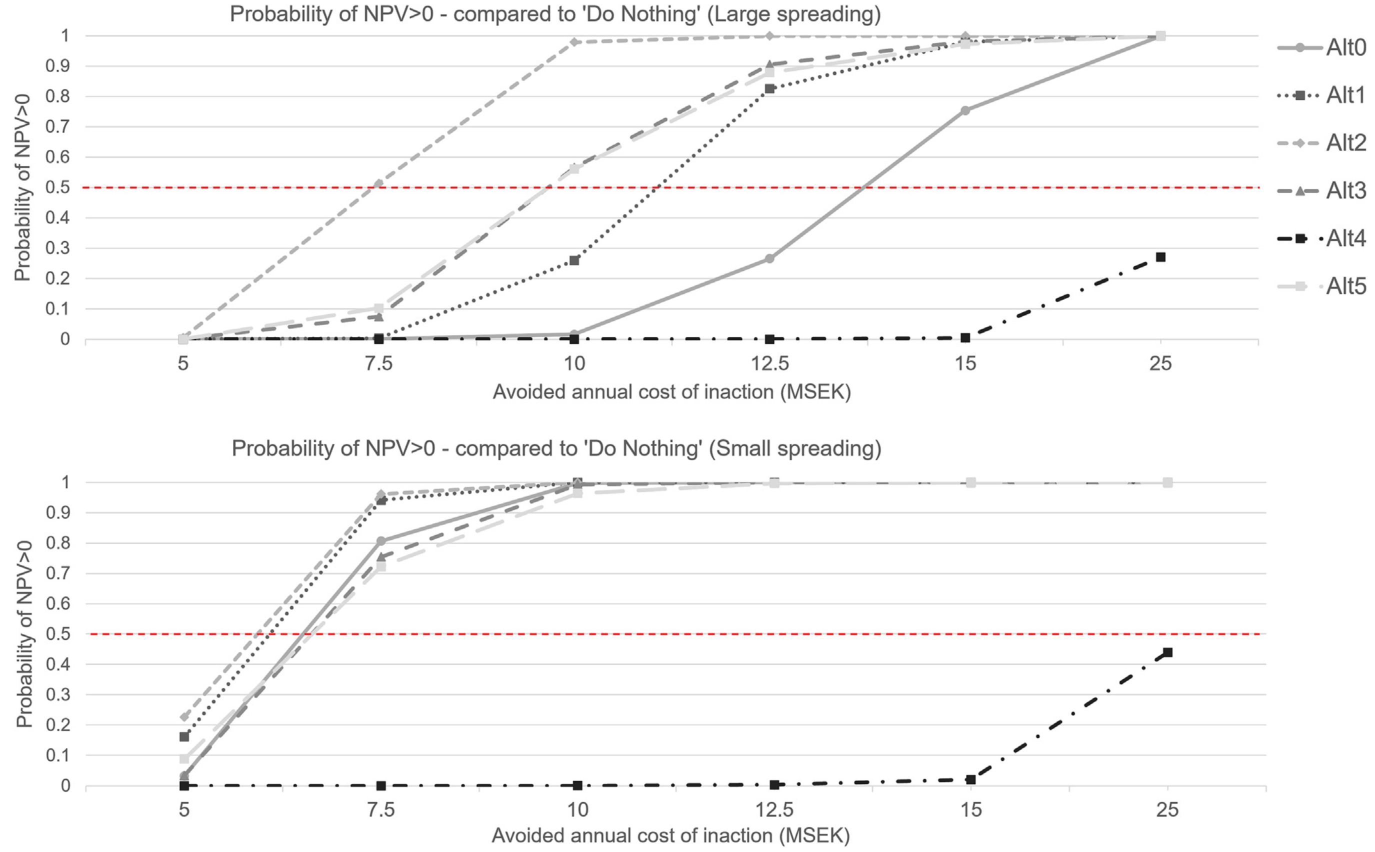

Fig. 4. Probability of NPV being positive (>0) for Alt 0–5 in comparison to 'Do nothing' for simulated values representing avoided annual cost of inaction, in millions of SEK (MSEK) given a social discount rate of 3.5 %. The red dashed line indicates the 'breakeven point' where the NPV has a >50 % probability of being >0. Note: the scale on the x-axis is not linear.

20 years. Phytoremediation (Alt 3–5) also includes these long-term costs, in addition to the long time required to achieve the risk reduction targets. Alt 0–2 were thus assumed to generate 100 % of the benefit within 2 years while Alt 3–5 generated 80 % from hotspot remediation and the remaining 20 % over the duration of the remediation time required for the rest of the site (Eq. (S1)). This partitioning favoured the faster remediation techniques, but Alt 3 and Alt 5 were still shown to consistently rank 2nd or 3rd highest in the different scenarios. Different values for the proportion of the total benefit gained from hotspot remediation were not tested but it could change with greater knowledge of the scale and severity of the PFAS spreading and impacts.

Importantly, the long time requirement for phytoremediation is an assumption for phytoextraction, which may be infeasible, but mitigation of spreading risks from short-chain PFAS through hydraulic control of groundwater via phytomanagement could be achieved in a shorter timeframe while also providing valuable ecosystem services (Evangelou and Robinson, 2022). A distinct advantage with phytomanagement is that the remedial costs are often lower than other remediation techniques and it could potentially generate a mean *NPV* closer to zero or positive if accounting for long-term additional benefits such as provision of ecosystem services and the potential production of valuable biomass, which are currently not included. For example, Wan et al. (2016) expected that the benefits of phytoremediation of a metal-contaminated soil (from e.g., cash crop production) would offset the project costs in less than seven years. The longer timeframe for GRO may not even be such a disadvantage in the Stockholm Arlanda Airport case if there are no plans for rapidly redeveloping the site for immediate profit, especially in the large PFAS spreading scenario which supports the view that GRO are well-suited for large areas where there are no time restrictions (Cundy et al., 2016). GRO may even be a profitable option for both the problem owner and society in the long-term if the present value of remediation cost savings (benefit) exceeds the present value of postponed increased property value (cost) resulting from capital costs and long-term monitoring and management activities necessary to carry out the remediation alternative (Bell, 1996). However, if there are plans for immediate development then the long timeframe of remediation alternatives that include gentle remediation via stabilization with active carbon or phytomanagement (Alt 2, 3, 5) may be a disadvantage when compared to Alt 0.

The effectiveness of each remediation alternative to manage the contamination risks is also an important aspect that must be directly addressed per alternative, especially considering the novelty of PFAS compounds. Typically, this would be considered during the selection of viable remediation alternatives to include in the CBA, which assumes that these meet the requirements with respect to their effectiveness. However, when including newer, more innovative techniques (e.g., phytomanagement) or if there are uncertainties due to the complexity of remediating PFAS, there is a risk of failure to meet the risk reduction targets. As previously noted, a primary shortcoming with phytoremediation is the long time required for phytoextraction, which may be effective only for short-chain PFAS with carbon chain length (<C6), though stabilization of longer-chain PFAS through accumulation in the roots can also help to mitigate risks (Bolan et al., 2021; Evangelou and Robinson, 2022; Gobelius et al., 2017; Huff et al., 2020). Conventional remediation techniques may also entail difficulties that limit their effectiveness when applied to remediate PFAS-contaminated soil. Immobilisation technologies like S/S have been shown to be effective in binding PFAS but a significant downside is that the contamination ultimately remains at the site, with short-chain PFAS potentially breaking through over time, and the long-term stability of amendments is still unknown which limits their use and application as a long-term solution (Bolan et al., 2021; Goldenman et al., 2019; Mahinroosta and Senevirathna, 2020; Ross et al., 2018; Sörengård et al., 2021). Thermal treatment of soil at high temperatures (>1000 °C) can destroy PFAS, but at lower temperatures

(500–600 °C) it may vaporize and generate PFAS transformation by-products that can be released into the air if not captured and treated (Held and Reinhard, 2020; Ok et al., 2020; Wang et al., 2015, 2022) Soil washing can likewise have high removal efficiencies but its effectiveness varies for individual PFAS and type of soil (Grimison et al., 2023).

One method to account for varying effectiveness is to include an 'efficiency surcharge' per alternative (Chen and Li, 2018), which could reflect the technical uncertainty by increasing the cost of the alternative by a factor (%) of failure. Similarly, the risk of failure is accounted for in this CBA including a probabilistic 'project risk' (C1f, Table S2 in SM) cost (i.e., the probability of failure multiplied by the costs of necessary additional remedial actions). The probabilistic project risk costs could thus be considered as a contingency plan in the case of the remedial action not meeting the remediation objectives in the estimated time and account for the relative uncertainty and effectiveness of the specific remediation alternatives used to manage the PFAS contamination. Linacre et al. (2005) emphasized that 'uncertainty in project success' (i.e., the possibility that complete remediation may not be realized) may significantly increase the perceived costs of phytoremediation operation for decision makers. However, the extra project risk cost due to probability of failure for phytoremediation was not shown to impact the resulting mean *NPV* or rankings of these alternatives (Alt 3–5).

5.2. Costs of inaction

A challenge in this study was to determine a reasonable value for the AACOI (B2-B3) for Stockholm Arlanda Airport. According to Goldenman et al. (2019, pg. 129), "The costs for remediating some cases of contamination run to many millions of EUR. Total costs at the European level are expected to be in the hundreds of millions of EUR as a minimum." The costs of remediation at PFAS contaminated sites will accordingly be large and weigh heavily on the resulting *NPV* for the remediation project. However, the 'costs of inaction' for not managing PFAS in the environment are estimated to be even larger – €2.1–2.4 billion annually in the Nordics from health impact-related costs due to contaminated drinking water alone (Goldenman et al., 2019)– and provide a substantial counterpoint to the high remediation costs that could even tilt the scales towards a positive *NPV* outcome for many of the remediation alternatives. Furthermore, continued inaction will lead to more sources of contamination, more people exposed, higher remediation costs, and ultimately will require more extensive remediation of soil and groundwater as PFAS spreads throughout the environment over time.

In the CBA, 7.5 MSEK was used as a base scenario for the AACOI (B2-B3), and the resulting *NPV*s in comparison to 'do nothing' for some alternatives indicate that they may indeed be socially profitable at a social discount rate of 3.5 %. In the case of negative *NPV*s, the results may imply that PFAS remediation in these cases is not socially profitable given this discount rate. A lower discount rate would result in lower breakeven points and a higher discount rate in higher breakeven points. However, remediation may still be motivated for other reasons and even be required by regulatory authorities since 'doing nothing' is not legally permissible. Also, due to the direct and indirect (e.g., inhibited recreation, contaminated fish) costs associated with PFAS contamination, the AACOI correlated with remediating Stockholm Arlanda Airport could potentially be higher.

In the Swedish context, Goldenman et al. (2019) estimate that 290,000 people in Sweden alone (ca. 47 % of the estimated total exposed Nordic population of 621,000) are exposed to PFAS above a statutory limit, which could then be roughly equated to ca. €1 billion per year of associated health-impact costs to represent 47 % of the total estimated €2.1–2.4 billion for the Nordics. The difficulty then is disaggregating this large lump sum of health impact-related costs, most of which are attributed to exposure via contaminated drinking water, and parsing the avoided costs to specific remedial actions taken at a particular PFAS contaminated site such as Stockholm Arlanda Airport. That is, what fraction of these avoided costs can be attributable to a remedial action taken at Stockholm Arlanda Airport?

The case could be made that remediation of the firefighting training site at Stockholm Arlanda Airport would result in a substantial amount of 'avoided' harm from PFAS contamination. As noted in Goldenman et al. (2019), much of the contribution of PFAS contamination to the Sweden is likely due to sites where AFFF were used, such as at civilian and military airports. Indeed, evidence from site investigations indicates that Stockholm Arlanda Airport is a significant source of PFAS spread to Lake Mälaren by as much as 2.4–5.3 kg of PFAS per year (Ahrens et al., 2015). Lake Mälaren is an important drinking water source for Stockholm and PFAS contamination has necessitated extensive investments in drinking water treatment plants in recent years to treat the PFAS contaminated drinking water with increasingly strict drinking water guidelines (Franke et al., 2021). Soil remediation, especially of the hotspot but also the rest of site, would undoubtedly mitigate these negative impacts but the fraction of the avoided health-impact costs that could be attributed to such a remedial action would require further investigation.

In comparison to the 'do nothing' reference, Alt 0, Alt 1, Alt 2, Alt 3 and Alt 5 have a high probability of generating $NPV > 0$ in the range of tested AACOI values from 5 to 15 MSEK/year, depending on the PFAS spreading scenario. The breakeven point provides an indication of when remediation may be justified from an economic standpoint and which alternative is most attractive (highest probability of $NPV > 0$) for lower expected values of avoided cost of inaction. However, given the severity of PFAS contamination and its expected impacts, the more intensive and faster remediation alternatives to rapidly mitigate risks would become more profitable in comparison to the other alternatives if the expected value of avoided cost of inaction is determined to be very high. At present, an accurate value for the cost of inaction attributable to Stockholm Arlanda Airport is unknown but this CBA provides a valuable piece of information to decision-makers by demonstrating the full range of costs and benefits and breakeven points for social profitability.

5.3. Impact of choices, assumptions and uncertainty

An early-stage CBA faces the challenge of using limited data to provide reliable decision-support. In this case, developing a probabilistic CBA for a novel application required making many assumptions due to the novelty of PFAS contamination and lack of both technical and economic data. Also, the CBA results is sensitive to choices and assumptions made in developing the model such as selection of the reference alternative and choice of discount rate.

In this case, two different reference alternatives were included in the model that serve different purposes. Alt 0 represents a modified 'business-as-usual' case entailing 'total excavation', which is a common remediation approach in Sweden and other countries. It is therefore useful as a comparison case for when remediation is mandated, and a conventional approach can be evaluated against alternatives to provide decision-support. 'Do nothing', on the other hand, is a helpful reference alternative in a CBA for obtaining indications on whether it is economically reasonable for society to spend scarce resources on remediating a particular site or rather use its resources for other purposes. It should be noted that even if it is not found to be economically reasonable to remediate a site (i.e., $NPV < 0$), remediation might still be motivated from legal and moral consideration. The choice of reference alternative in a CBA is thus context-dependent and should always be carefully considered and motivated.

The choice of discount rate is also important, especially in applications with long time horizons (Söderqvist et al., 2015). In essence, the choice of discount rate reflects the emphasis placed on future values: the higher the discount rate the lower the present value of the future benefits and costs, other things being equal (Johansson and Kriström, 2018); which is important when valuing, for example, the expected positive externalities (or avoided damage). Furthermore, the choice of discount rate can become an issue of inter-generational equity, particularly in the case of PFAS with its large current and expected future impacts, and where the expected value of some remediation projects is long into the future and can only be accurately reflected in a CBA with a suitably low, long-term discount rate, or even a declining discount rate over a long time horizon (Johansson and Kriström, 2018). The mean *NPV* for Alt 2–5, which employ GRO, vary significantly depending on the discount rate and the social profitability of each alternative can be much higher if the expected benefits from positive externalities are not heavily discounted over the remediation time period (Table S5). Still, Alt 2 is the most promising remediation option from an economic perspective, regardless of changes in the discount rate. It should be noted that a higher discount rate might be more appropriate for evaluating the profitability of these alternatives to a landowner who is more concerned with short-term impacts (Volchko et al., 2017).

5.4. Limitations of the study

A CBA is about investigating consequences for human well-being and, whenever possible, monetize them, including those caused by changes in the supply of ecosystem services. However, there might be other values that cannot be captured by a CBA, e.g., the intrinsic value of soil health or ecosystems, which suggests a need for complementary assessments in the decision-making process for making well-informed and sound decisions. Currently, the improved health and environmental benefits from the remediation alternatives are bundled into the lump sum value of avoided cost of inaction. There are, however, many wider benefits (or costs) that may result from remediation such as the loss or gain of soil functionality, which is difficult to account for in a CBA but is an important aspect of soil remediation (Chen and Li, 2018). The improvement of soil functionality and increased provision of ecosystem services could be an important benefit of using GRO that is currently neglected. Multi-criteria decision analysis is increasingly being used for evaluating positive and negative effects of remedial actions in the three domains of sustainability (environmental, social and economic) to support the decision on the most reasonable alternative taking into consideration other values which are not accounted for in a CBA (Bardos, 2014; Rosén et al., 2015; Söderqvist et al., 2015).

Assumptions were made regarding the technical applications of certain remediation technologies that impacted the CBA results. For soil washing, it was assumed that 100 % of the excavated soil was washed and reused on site, which may not be completely accurate but is favourable based on the site geology of primarily sand. Similarly, thermal treatment is assumed for the treatment of fine aggregates with the reuse of coarse aggregates on-site, but the particle size distribution in terms of proportion of fine aggregates is unknown. Also, the annual cost of phytoremediation was not possible to accurately quantify based on literature and could have affected the resulting *NPV* of Alt 3–5. The lump sum used as a present value of this cost includes total project costs (e.g., costs of establishment, operation and maintenance, biomass harvest and management) was consequently not sensitive to changes in the time required for risk reduction which could impact the present value of costs and benefits when discounted over a longer time. It is also a simplification to assume that full risk reduction will be achieved so quickly and in similar time for each hotspot remediation alternative. Other potentially viable PFAS remediation techniques such as engineered caps/covers were not included in this study but may be considered as similar to the considered containment techniques (i.e., S/S and stabilization with AC) and entail similar processes, costs, and limitations.

6. Conclusions

The following main conclusions can be drawn from this study:

- There are many uncertainties associated with PFAS contamination, including costs, benefits and effectiveness of PFAS remediation alternatives. Limitations in data and the novelty of PFAS remediation required making assumptions to compensate. Probabilistic CBA is demonstrated to be a robust method to account for uncertainties and parameter sensitivity and the model was further improved by creating multiple scenarios to test different model assumptions. The analysis provides valuable decision-support by evaluating the social profitability of different PFAS remediation alternatives.
- In general, excavation and stabilization/solidification of the hotspot on-site combined with stabilization of PFAS at the rest of the site with activated carbon (Alt 2) has the highest probability of being socially profitable (greatest mean *NPV*) and highest ranking in all scenarios. All other alternatives, except for thermal treatment of the hotspot (Alt 4), are socially profitable and entail reduced negative externalities to varying degrees compared to ‘total excavation’ of the entire site. The extent of PFAS spreading (large or small spreading) is shown to be the most sensitive variable in the CBA model and affect the ranking of subsequent remediation alternatives.
- Costs of inaction to society from PFAS contamination are high but associated with uncertainties, in particular how much avoided damage to human health and the environment is attributable to remediation at a particular site like Stockholm Arlanda Airport. Simulations of different values for annual avoided cost of inaction (AACOI) as an aggregated benefit to society are useful to compare breakeven points for when a remediation alternative becomes socially profitable.
- Two different reference alternatives – Alt 0 as a modified business-as-usual case entailing ‘total excavation’ and ‘do nothing’ as a common reference for economic analysis – were used for comparison to evaluate the remediation alternatives from different perspectives and to provide balanced support to decision-makers.

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CRediT authorship contribution statement

The authors contributed as follows: Conceptualization: P.D., Y.V., T.S., L.R., J.N.; Methodology: P.D., Y.V., T.S., L.R., J.N.; Validation: L.A.; Formal analysis: P.D., Y.V.; Investigation: P.D., Y.V.; Visualization: P.D., Y.V.; Writing - original draft preparation: P.D., Y.V., L.A., J.N.; Writing - review and editing: P.D., Y.V., L.A., T.S., L.R., J.N.; Supervision: Y.V., L.R., J.N.; Project administration: J.N.; Funding acquisition, Y.V., L.R., J.N.

All authors have read and agreed to the published version of the manuscript.

Data availability

Data will be made available on request.

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 data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.163664.

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Corrigendum

Corrigendum to “Comparison of PFAS soil remediation alternatives at a civilian airport using cost-benefit analysis” [Sci. Total Environ. 882 (2023) 163664]

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The authors regret that the printed version of the above article contained an error: the last row (C3) in Table 3 is incorrect. The correct and final version of Table 3 follows. The authors would like to apologise for any inconvenience caused.

Table 3
Summary of cost and benefit values used in the cost-benefit analysis to calculate net present values. Mean PV: the mean present value of cost and benefit items. L: Large spreading scenario. S: Small spreading scenario. The annual avoided cost of inaction (B2-B3) in Alt 0-Alt 5 is assumed to be 7.5 MSEK. The social discount rate is 3.5 %. The time horizon is 120 years.

Time horizon (years): 120 Discount rate: 3.5 %		Alt 0 S/S hotspot & disposal rest		Alt 1 S/S hotspot & S/S rest		Alt 2 S/S hotspot & stabilization AC		Alt 3 S/S hotspot & phytoremediation		Alt 4 T/T hotspot & phytoremediation		Alt 5 SW hotspot & phytoremediation	
Category	Item	Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)		Mean PV (MSEK)	
Spreading scenarios		L	S	L	S	L	S	L	S	L	S	L	S
Benefit categories and items													
B2-B3. Avoided cost of inaction	Improved health and increased provision of ecosystem services	197	197	197	197	197	197	177	177	177	177	177	177
Cost categories and items													
C1. Remediation costs	C1a-e.I. Short-term costs (total area)	109	60.0	80.4	52.1	67.5	48.6	129	75.9	322	268	129	75.3
	C1b.II, C1e.II. Long-term costs of management, monitoring (rest of the site)	0	0	0	0	7.39	7.39	7.39	7.39	7.39	7.39	7.39	7.39
	C1f. Project risks	32.1	17.3	23.4	14.9	19.6	13.9	16.8	13.1	74.4	70.8	16.6	12.9
	Total C1	141	77.3	104	67.0	94.5	69.8	154	96.4	404	346	153	95.6
C2. Impaired health due to remedial action	C2b. From transport activities and C2c. At a disposal site	0.709	0.201	0.0346	0.0157	0.0197	0.0116	0.00854	0.00852	0.401	0.400	0	0
C3. Decreased provision of ecosystem services due to remedial action (C3a-C3c)		82.5	10.1	79.3	9.23	6.57	3.68	2.58	2.58	32.3	32.3	4.76	4.76

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Review

Challenges in the PFAS Remediation of Soil and Landfill Leachate: A Review

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Abstract

The use of per-and poly-fluoroalkyl substances (PFAS) has resulted in the contamination of different environmental matrices. In EU countries, the sites contaminated with PFAS are usually remediated by excavating the soil and disposing of it in a landfill, as no in-situ or on-site techniques capable of treating large quantities of soil cost-effectively have been developed. Landfilling of PFAS-contaminated soil is one of the sources of PFAS in landfill leachate. In this paper, the physical and chemical treatment methods to remove PFAS from soils and landfill leachates are described. Among the challenges that may limit the remediation of contaminated sites, we highlight the lack of strict regulation of PFAS in soils, the cost, the ineffectiveness of some methods for the remediation of certain PFAS compounds, and the limitation of the environmental matrices.

Keywords
Contaminated soil; landfill; leachate; treatment techniques; sorption; stabilization

1. Introduction

Per-and poly-fluoroalkyl substances (PFAS) have been widely used for more than 50 years as surfactants, coatings, water repellents for leather and textiles, impregnating agents, as well, in metal plating and aqueous film-forming foams (AFFFs) [1-3]. Today, more than 4,730 compounds related to PFAS are registered, and some of them are very mobile in the environment [1, 4]. These substances have been detected in surface water [5-7], sediments downstream of the production facilities [8, 9], groundwater [10, 11], soil [12-16], wastewater treatment plant (WWTP) effluents [17, 18], and sewage sludge [19, 20]. PFAS contamination in aquatic environments has been linked to major production sources [21, 22]. Additionally, effluents from WWTPs are another major source from which PFAS are introduced into the aquatic environment [22].

In general, PFAS are fluorinated aliphatic compounds with strong carbon and fluorine (C-F) bonds. The hydrogen atoms on all the carbon atoms have been replaced with fluorine in the perfluoroalkyl compounds, whereas hydrogens on some but not all carbon atoms are replaced with fluorine in the polyfluoroalkyl compounds (which also contain a perfluoroalkyl moiety, C_nF_{2n+1}). Perfluoroalkyl sulfonic acids (PFSAs, $C_nF_{2n+1}SO_3H$), with 4-10 fully fluorinated carbon chains, and perfluoroalkyl carboxylic acids (PFCAs, $C_nF_{2n+1}COOH$), typically with 3-15 fully fluorinated carbon chains, are the most commonly reported subgroups of PFAS [23]. Long-chain PFAS with ≥8 carbon atoms are referred to as PFCAs, and those with ≥6 carbon atoms are referred to as PFSAs [24]. These long-chain compounds are characterized as non-biodegradable, highly persistent, potentially toxic, and bio-accumulative (in blood, liver, and kidneys) [25]. For these reasons, together with their widespread occurrence, PFAS have been highlighted as environmental and health hazards in recent years. Two compounds, in particular, perfluorooctanoic acid (PFOA, $C_7F_{15}COOH$) and perfluorooctane sulfonic acid (PFOS, $C_8F_{17}SO_3H$), have been identified as the most abundant and widespread species in soil and aquatic environments [7, 11].

Excavation and landfilling are two conventional methods for the proper management of contaminated soils, although these are becoming less popular nowadays due to their high cost. Landfilled soils and other waste materials (e.g., carpets, textiles, paper, and packaging) containing PFAS may release these compounds in the leachate [1, 26, 27], which can then migrate to the surrounding aquatic sources, particularly groundwater, due to the lack of barriers at old landfill sites or due to damage to the bottom liners [28]. Traditional facilities for leachate treatment at the landfills in Sweden (and worldwide) are inadequate in removing PFAS [1, 3, 29].

This review aims to evaluate the available techniques for PFAS treatment in soils before landfilling and the potential release of PFAS through landfill leachate, as well as to discuss the possible challenges of implementing these techniques for the remediation of contaminated sites. We have reviewed the current legal concentrations of PFAS in landfill soil and suggest revising the legislation on the landfilling of PFAS-containing materials. The reported concentrations of PFAS in the soil in contaminated areas were compared to assess the differences among contaminated sites and discuss the need for soil treatment. The chemical stability of the treated soil in landfills is also discussed and some knowledge gaps are identified. The concentrations of PFAS in landfill leachates in both new and old landfills are compiled to identify the differences among landfills globally and their environmental impacts.

2. PFAS Regulations

In 2004, the EU introduced Directive 850/2004 on Persistent Organic Pollutants (POPs), which restricted the marketing and use of PFOS [30]. The Directive applies to the substances and preparations of POPs with concentrations equal to or higher than 0.005% by mass (Table 1). A summary of the applicable regulations for PFAS in soil, waste, and groundwater is provided in Table 1. The Stockholm Convention includes PFOS and PFOA in the list of POPs, and these substances are restricted under the EU POP regulations [31]. PFHxS, which is currently being assessed under REACH for potential restriction, is also considered a candidate to be listed under the POPs by the Stockholm Convention [32]. No official guideline values have been published for the PFAS concentrations in soil that could pose a potential risk to human health or the environment, either at the EU level or the national level by the member states. The Swedish Geotechnical Institute (SGI) and the Swedish EPA have established preliminary guideline values for highly fluorinated substances in soil and groundwater, which recommend the maximum values of 0.003 mg/kg of PFOS in the soil for sensitive land use (e.g., residential land), 0.02 mg/kg of PFOS for less-sensitive land use (e.g., industrial land), and 0.045 µg/L of PFOS [33] in groundwater. In the US [34], different states have established different soil screening levels for groundwater protection which range from 0.00022 to 0.05 mg/kg of PFOS and 0.0015 to 19 mg/kg of PFOA, while the human health soil screening levels range from 0.5 to 6 mg/kg of PFOS and 0.33 to 16 mg/kg of PFOA (Table 1). In Canada, Health Canada has issued a direct contact residential soil screening value of 0.70 mg/kg for PFOA [35], and the Canadian Council of Ministers of the Environment has drafted guidelines for the protection of groundwater used for watering livestock and irrigation with the recommended PFOS limits of 12 mg/kg (coarse soil) and 9 mg/kg (fine soil) [36]. Additionally, the Canadian Federal Soil Quality Guidelines (FSQGs) have prescribed the maximum concentrations of 0.01 mg/kg of PFOS in agricultural, residential, or parkland soil, 0.14 mg/kg of PFOS in coarse soil, and 0.21 mg/kg of PFOS in fine soil at the industrial sites [37]. In Australia, several guidelines for human health soil screening criteria have been issued: 1) residential areas with garden/accessible soil: 0.009 mg/kg of PFOS and 0.1 mg/kg of PFOA; 2) residential areas with minimal opportunities for soil access: 2 mg/kg of PFOS and 20 mg/kg of PFOA; 3) public open spaces: 1 mg/kg of PFOS and 10 mg/kg of PFOA; and 4) industrial/commercial areas: 20 mg/kg of PFOS and 50 mg/kg of PFOA, which represent the values applied to PFOS as well as to the summed concentrations of PFHxS and PFOS [38]. Although several reference values are available for soil, further assessment, such as a leachability test, should be performed to evaluate the potential risk of PFAS leaching from the soil to the surrounding environment. Waste classification in the EU is regulated by the Waste Framework Directive 2008/98/EC (which categorizes waste into inert, non-hazardous, and hazardous groups) [39] and the list of waste substances in Decision 2000/532/EC [40]. The EU Regulation 850/2004 (Appendix 7) for the waste management of POPs stipulates that the waste containing more than 50 mg/kg of PFOS should be destroyed or converted irreversibly, although it does not specify where this waste should be disposed of [41]. This may cause extremely high concentrations of PFOS in the landfills, which are the sources of PFAS contamination in landfill leachate. Moreover, inappropriate landfilling (e.g., highly contaminated waste disposed at landfills for inert or non-hazardous waste) may pose a high risk of PFAS leakage into the environment. Legal concentrations of PFAS in waste need to be revised, and maximum limits of PFAS concentrations in treated leachate should be established to minimize the risk of contamination of the recipients.

Table 1 Summary of the available regulations for PFAS in soil, waste, and groundwater.

Country	Regulation	Subject	Limit values	Reference
EU	Directive 850/2004 on Persistent Organic Pollutants (POPs)	prohibiting, phasing out or restricting the production, placing on the market and use of substances subject to the Stockholm Convention on POPs	Waste containing more than 50 mgPFOS/kg TS should be destructed	[30, 41]
Sweden	Regulation under construction	Suggested values of PFOS in soil and ground water	PFOS<3 µg/kg in soil for sensitive land use PFOS<20 µg/kg in soil for less sensitive land use PFOS<0.045 µg/L in groundwater	[33]
USA	Regulations, Guidance, and Advisories for Per- and Polyfluoroalkyl Substances	levels of PFOS and PFOA in soil for ground water protection	PFOS in soil from 0.22 to 50 µg/kg PFOA in soil from 1.5 to 19000 µg/kg	[34]
		Levels of PFOS and PFOA in soil for human health protection	PFOS in soil from 0.5 to 6 mg/kg PFOA in soil from 0.33 to 16 mg/kg PFOA	[34]
Canada	Updates to Health Canada Soil Screening Values for PFAS & Canadian Environmental Protection Act	Concentrations of PFOA in residential and parkland soil, agricultural soil and soil at industrial sites	Residential, parkland and agricultural soil PFOA<0.70 mg/kg and PFOS<0.01 mg/kg Industrial sites PFOS<0.14 mg/kg in coarse soil and PFOS<0.21 mg/kg in fine soil	[35, 37]

		Concentrations of PFOS in water used for livestock watering and irrigation to protect groundwater	PFOS<12 mg/kg (coarse soil) PFOS<9 mg/kg (fine soil)	[36]
Australia	Commonwealth Environmental Management Guidance on PFOS and PFOA	Guidelines for human health soil screening criteria	Residential soil: PFOS = 9 µg/kg & PFOA = 100 µg/kg; Residential with minimal opportunities for soil access: PFOS = 2 mg/kg & PFOA = 20 mg/kg Public open space: PFOS = 1 mg/kg & PFOA = 10 mg/kg Industrial/Commercial: PFOS = 20 mg/kg & PFOA = 50 mg/kg	[38]

3. PFAS in Contaminated Soil

Fire-fighting training areas and manufacturing plants are the largest sources of PFAS in soil. PFOS are the most intensively studied PFAS contaminants in the soil at fire-fighting training facilities [9, 16, 17]. However, the contribution of PFOS to total PFAS at the sites may be over-estimated as there would be several other PFAS that were not previously included in the analysis. These overlooked PFAS might be the novel ones, such as chlorinated, dechlorinated, and ketone PFSA, perfluoroalkane sulphonamides (FASAs), 6:2 fluorotelomer sulphonamidoalkyl betaine, 6:2 and 8:2 fluorotelomer sulfonates, or short-chain perfluorocarboxylic acids [6, 42]. In a study published in 2014 on the chemical analysis of selected fire-fighting foams from the Swedish market, PFHxA and 6:2 FTSA were found in considerably high concentrations in the foams, with some 6:2 fluorotelomer-based products also identified in most of the foam samples [43]. The reported ranges of PFOS concentrations in soil were 6.4-2400 µg/kg at Oslo airport, Norway [16], 21-120 µg/kg at Kristianstad, Bergen, and Hastad airports in Norway [16], 2.2-8520 µg/kg at Tullinge, Sweden [9], and 21-2400 µg/kg at Ellsworth Air Force Base, USA [17]. Additionally, an extremely high concentration (36000 µg/kg) of PFOS was reportedly found near a burning pit at a former firefighter training area at the Ellsworth Air Force Base [44]. The PFAS concentrations and the number of contaminated sites across the world suggest that soil is a major environmental reservoir of PFAS [15].

Hale et al. [16] and Filipovic et al. [9] found the highest concentrations of PFOS at 1-2 m below the soil, suggesting that PFOS can gradually migrate down the layers of soil with the rainwater. This migration of PFAS through the soil is complex and depends on several factors, such as the properties of PFAS, the soil properties, and the climatic conditions. Depth-profile data and mathematical modeling have demonstrated that the PFAS in the soil can be retained in the vadose zone for decades, mostly due to the adsorption of PFAS at the air-water interface as well as on soil grain surfaces [14, 15].

The transport of PFAS through the soil may result in the contamination of groundwater, surface water, and drinking water wells, along with the contamination of biota. Filipovic et al. [9] reported large differences in the concentrations of PFOS between the surface water (1.1-79 ng/L) and groundwater (1-51000 ng/L) close to an old air-force base at Tullinge near Stockholm [8]. The contamination of groundwater may occur over decades or centuries since contaminated soil acts as a reservoir of PFAS and the transport of PFAS through soil is quite slow [14, 15]. Thus, it is necessary to remediate the contaminated areas to prevent contamination of watercourses. One of the most common techniques for the remediation of the hotspots contaminated with PFAS is “digging and dumping”. Although this strategy entails costly landfilling, sometimes without proper handling and disposal facilities, its application has been continued due to the deficiencies in and the uncertainties of in-situ (on-site) techniques for treating PFAS-containing soils. Disposal of contaminated soil in a landfill from a site without treatment is merely a transfer of the problem of one site to another. Thus, PFAS-contaminated soils need to be treated before disposal to minimize further environmental problems associated with the contamination of non-contaminated areas and drinking water sources.

4. Influence of the Landfill Environment on the Release of PFAS from Contaminated Soil

The solubility of PFAS in the contaminated soil in a landfill should be kept low to avoid contamination from leachates. However, many factors in a landfill may influence the release of PFAS from the soil. Infiltration of the precipitation into the system, the type of waste, the degradation of disposed waste, and the operating conditions (e.g., compaction of wastes, gas collection system, leachate recirculation) [45] are the factors that can affect the pH, redox conditions, temperatures, and stability of PFAS in landfills [26, 46-49]. The infiltration of precipitation, moisture content, leachate production, and release of PFAS from the soil in landfills are positively correlated [26, 27]. Thus, the installation of a landfill cover reduces the production of leachate and the release of PFAS. Moreover, the reduction of infiltration reduces the microbial activity (and gas production) in the landfills, which may, in turn, reduce the release of PFAS [50, 51].

Encapsulation of PFAS-contaminated soil (e.g., with a geomembrane or clay minerals) after disposal in a landfill (during active landfilling) may prevent or reduce the contact between PFAS and the leachate, and the subsequent release from the soil. However, this technique may not have long-term (several decades to centuries) efficacy due to the potential for damage to the geomembrane and percolation of the leachate through the layer of clay [52].

Degradation of organic waste occurs in several landfilling phases, including the aerobic, acidogenic, and methanogenic phases, in which the leachate has characteristic pH values and composition (both organic and inorganic) [53]. Alkaline soils may promote the leaching of PFAS from the soil since the sorption of PFAS to the soil has been shown to decrease with an increase in the pH of the leachate. This is due to a reduction in the protonation of the adsorbent surfaces and the associated reduction in the frequencies of positively-charged sites on the sorbent [54-56]. Thus, landfilling of PFAS-contaminated soil together with alkaline materials should be avoided.

The anaerobic microbial biotransformation of precursors (e.g., polyfluoroalkyl phosphate esters) may generate fluorotelomer alcohols, followed by perfluorinated/polyfluorinated acids but is probably not a major source of the PFCAs detected in anaerobic environmental matrices under methanogenic conditions [57]. This observation suggests that PFAS-contaminated soil in landfill cells, together with other biodegradable organic wastes, may reduce the degradation of PFAS precursors. However, a shift in conditions from reduction to oxidation can be expected as landfills age. Oxidation of precursors in a landfill may generate more contaminated leachate. For example, Houtz et al. [17] estimated that the oxidation of precursors accounted for 23% and 28% of total PFAS on average in groundwater and soil samples, respectively. Therefore, it is important to maintain a low redox potential in a landfill to reduce the mobility of PFAS.

Landfill leachate has a complex chemical composition, and the effects of its composition on the leaching of PFAS from the waste into the landfills are not well documented. For example, anions in leachates (e.g., Cl^-, SO_4^{2}, $Cr_2O_7^{2-}$) and dissolved organic content (DOC) may increase the mobility of negatively-charged PFAS as they compete for the same (positively charged) adsorption sites [47, 58]. This suggests that variation in leachate quality likely contributes to the variability and patterns of PFAS concentrations in leachates [28]. Operating temperatures in a landfill may also affect the mobility of PFAS. For example, Kim et al. reported enhanced release of PFAS (especially PFHxA and PFHpA) with an increase in the contact time and temperature [59], whereas Jia et al. found that sorption of PFOS on humic acids (the main components of organic matter in sediments) doubled when the temperature increased from 5 °C to 35 °C [49]. Thus, due to inconsistencies in the findings, more research is needed to clarify the effects of temperature on the release of PFAS from contaminated soils.

Some PFAS that leach from the soil are likely to be re-adsorbed as the leachate percolates through a landfill, for example, on carbonaceous surfaces or humic acids in the waste [49]. Such re-adsorption has not been addressed in any study, although analogous sorption of PFAS on the sediments near WWTPs (particularly long-chain PFAS) has been reported [21, 60]. Thus, waste in a landfill could potentially serve as a filter for the PFAS leached from contaminated soils, although further research is required to test this hypothesis.

5. PFAS in Landfill Leachate

Around 140-188 million tons of total annual municipal solid waste is disposed of in landfills in the USA and Europe [61, 62]. In Europe and Australia, waste containing 50 mg/kg of PFOS can be disposed of in landfills without any pre-treatment [41, 63]. In the USA, the calculated annual PFAS disposal rate is 1,250 kg/year for municipal solid waste and 470-590 kg/year for biosolids [64]. The waste containing PFAS spreads in landfill leachates. A total of 17 studies have reported the concentrations of PFAS in landfill leachates, including seven conducted in North America [27, 29,45, 51, 65-67], seven in Europe [1, 64, 68-72], two in China [3, 73], and one in Australia [74]. The concentrations of PFAS in the leachates from active landfills vary by several orders of magnitude. For example, total reported PFAS (∑PFAS) concentrations in the leachates from Canadian and Chinese landfills ranged from 27 to 21,300 ng/L and 7,280 to 290,000 ng/L, respectively [3, 29, 67, 75]. This variation is less pronounced in closed landfills than active landfills, as was reported in a study on Australian landfills (2016). Lang et al. [27] found that the concentrations of only six (PFNA, 8:2 FTCA, 5:3 FTCA, PFBS, MeFBSAA, and MeFOSAA) of the 70 PFAS analyzed were significantly higher in the younger US landfills than in the older ones. This could be due to reductions in the concentrations of these species with time or changes in the types of PFAS used in products in recent years (e.g., a shift to the use of PFBS-based and fluorotelomer-based products instead of PFOS and PFOA). The similarity in the concentrations of almost all the analyzed PFAS in the leachates from old and young landfills indicates that PFAS may be released for many years after the disposal of the soil.

Concentrations of PFAS in landfill leachates and volumes of accumulated leachates must be known when evaluating the hazards posed by landfills, particularly the risk of PFAS contamination to the aquatic environment. For example, Busch et al. [1] estimated that discharges of ∑PFAS from 22 landfills in Germany ranged between 0.08 and 956 mg/day (mean: 135 mg/day), and the total discharges from all landfill sites in Germany (ca. 1,700) amounted to approximately 240 g of total PFAS/day, on average. Fuertes et al. [71] estimated a 1,209 g/year mass flow of ∑PFAS from four studied landfill sites serving a population of about 1.8 million people in northern Spain. In the USA, landfills released 563-638 kg of ∑PFAS to WWTPs via leachates in 2013 [27]. The actual amount of PFAS in landfill leachate in the USA could be higher as the cited authors' analyses did not include total oxidizable precursor or total fluorine assays. Furthermore, Yan et al. [3] estimated that approximately 3,110 kg of PFAS is released in landfill leachates in China annually. Thus, landfill leachates can be a significant source of PFAS contamination in the environment. PFAS should, thus, be removed from leachates before discharging to recipients, such as WWTPs, rivers, or lakes [65].

6. Treatment of PFAS-Contaminated Soil before Landfilling

6.1 Removal of PFAS from Contaminated Soil through Washing

Soil washing involves the separation of PFAS-contaminated particles from the bulk of the soil, followed by the extraction or desorption of PFAS from those particles (Figure 1). Mechanical separation is suitable for coarse-grained soils (e.g., soils dominated by stones and gravel), but it is not economically viable for fine-grained soils (dominated by fine sand, silt, and clay) [16]. Chemical extraction of PFAS should be performed on fine-grained soil particles under saturated conditions to enhance the treatment and minimize the adsorption of PFAS in unsaturated soils (see Section 3). Extraction agents, such as methanol/sodium hydroxide (MeOH/NaOH), methanol/ammonium hydroxide (MeOH/NH_4OH) and acetonitrile (CH_3CN), and water, have been used to extract PFAS from contaminated soils in the laboratory under saturated conditions [16, 42, 54]. Hale et al. [16] found that in the determination of the total concentration of PFOS in the soil, water extracted more PFOS compared to either methanol or acetonitrile, indicating that water could serve as an excellent extraction agent. However, extraction efficiency depends not only on the type of extraction agent but also on the soil properties (e.g., organic matter and clay content) [16, 60]. Removing PFAS by washing may be feasible for soils with low DOC (0.7-6 mg/L) and soils with low clay content (e.g., soils predominantly composed of sand) at an alkaline pH [16, 60]. In addition to the treated soil, this technique generates large amounts of contaminated process water that needs further treatment. Soil washing has been used to remove PFAS commercially in-situ ("flushing") but not for the treatment of soil before landfilling due to the high costs involved [54].

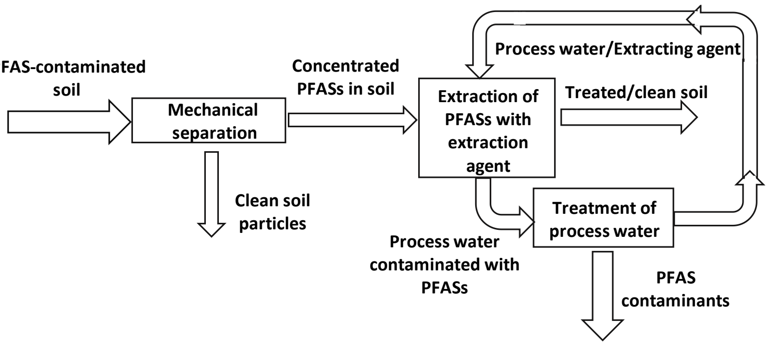

Figure 1 A combined treatment of PFAS-contaminated soil consisting of mechanical separation of the contaminated particles from the soil mass and the extraction of PFAS using an appropriate agent.

6.2 Stabilization and Solidification of PFAS in Contaminated Soil

Stabilization of PFAS in contaminated soil refers to the immobilization of PFAS by the addition of a substance that reduces mobility through leaching. Solidification of PFAS-contaminated soil involves the mixing of a cementitious binder and additives into the contaminated matrix to reduce hydraulic conductivity and PFAS exposure to leaching. Detailed knowledge of PFAS sorption behavior is essential for developing effective stabilization techniques to manage contaminated soils. The most important types of interactions involved in the sorption of PFAS to potential sorbents are electrostatic interactions, hydrophobic interactions, and ion exchange [56, 76, 77]. Electrostatic interactions between PFAS in solution and sorbents rely significantly on the pH [46-48, 78]. Anionic PFAS are adsorbed on the positively-charged surfaces of the sorbents [47-49, 79]. When the pH of a PFAS-contaminated solution becomes more acidic than the adsorbent's pHpzc (pH at the point of zero charge), the number of positively-charged sites on the sorbent increases (by surface protonation), and the adsorption of these PFAS is enhanced [76]. Hydrophobic interaction between PFAS and sorbents is the strongest when the sorbents are uncharged [60, 76]. Ion exchange is the dominant mechanism for the immobilization of anionic PFAS, especially for short-chain PFAS, when the anion-exchange resin is used [73]. The described sorption mechanisms of PFAS on sorbents are quite complex as the immobilization of PFAS in stabilized soil depends not only on the properties of the added sorbent but also on the soil characteristics (e.g., organic matter content) and leaching conditions (e.g., pH, redox potential, and temperature). Both inorganic and organic substances have been tested as possible PFAS adsorbents under experimental conditions, mostly in soil-free conditions. The inorganic substances include sand, high iron sand, kaolinite, Fe_2O_3 (hematite), alumina, zeolite, hydrotalcite, montmorillonite, FeO(OH) (goethite), and modified commercial clay [48, 73, 80, 81]. The organic substances include oil, black carbon, activated carbon (AC), biochar (BC), and ion-exchange resin [16, 82-85]. However, only four of the reviewed studies [16, 80, 84, 86] included tests evaluating the PFAS sorption efficiency in the presence of soil and only one published study described the solidification of PFAS-contaminated soil [86]. The stabilization efficiency of the substances should be verified in the presence of soil containing multiple contaminants collected from a field site.

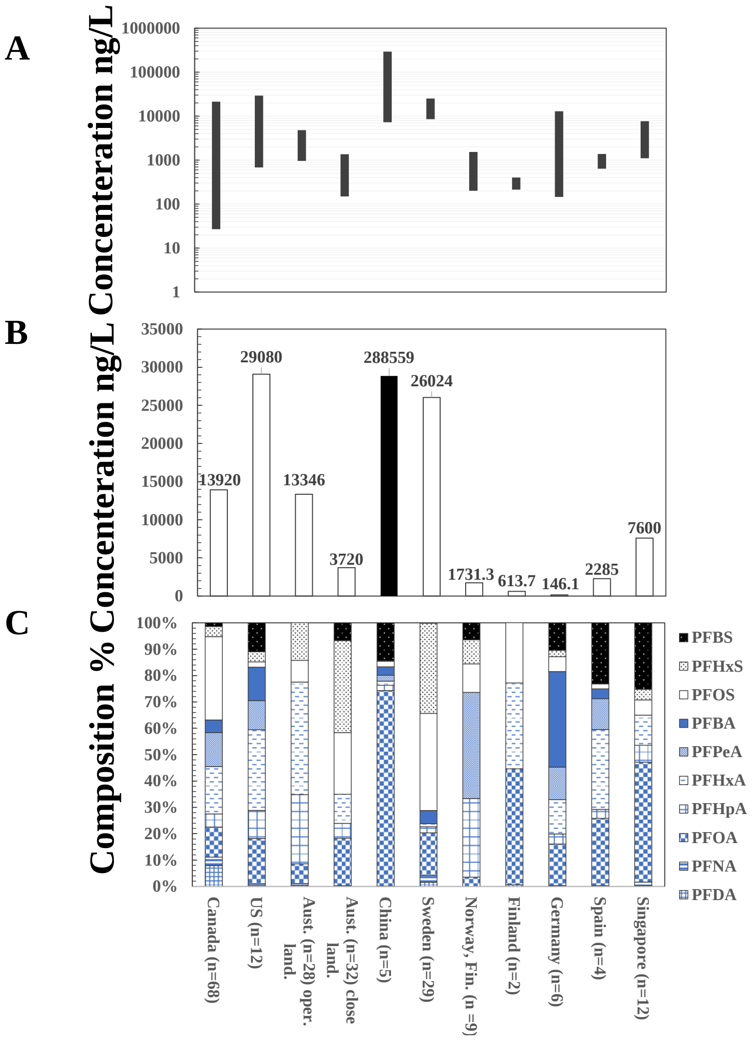

Figure 2 Graphs showing (A) Graphs showing minimum and maximum concentrations (ng/L) of ∑PFAS in raw leachate; (B) Graphs showing average concentrations (ng/L) of ∑PFAS in raw leachate. (The bar in black does not fit onin the y-axis.); and (C) Graphs showing the composition of PFAS (%) in the raw leachate. Ref. From earlier studies: Canada [29, 67, 75], the USA [45, 51], Australia [74], China [3], Sweden [64, 72], Norway and Finland [68], Finland [70], Germany [1], Spain [71], and Singapore [87].

6.2.1 Treatment of PFAS-Contaminated Soil Using Activated Carbon (AC) and Biochar (BC)

Biochar is a type of charcoal produced mainly from biomass (e.g., mixed wood, coconut husks, or bamboo) through pyrolysis (thermal decomposition of organic matter in the absence of oxygen). Granulated AC (GAC) and powdered AC (PAC) are produced similarly (using biomass, coal, and peat), in combination with physical activation (using hot gases) or chemical activation (with acid or base, such as H_3PO_4, KOH, and NaOH), to increase the pore volume and surface area. Both AC and BC treatment technologies have been used in the remediation of soil, groundwater, and drinking water contaminated with PFAS [16, 83, 84, 88]. AC is more suitable for remediation as its pore structure provides considerably large specific surface areas, pore volumes, and sorption capacities for organic pollutants (Table 2).

Table 2 Chemical composition, surface area (SA), pore volume (PV), soil-water distribution coefficient K_d (in L/kg), and organic carbon normalized sediment-water distribution coefficient logK_{OC} (estimated as K_d/f_{oc}) of soil, and the soil treated with powdered activated carbon (PAC), mixed wood biochar (MW BC), paper mill waste biochar (PMW BC) [8, 16, 76, 84, 89].

	Soil/sediment	PAC	MW BC	PMW BC
pH		9.6 [84]	9.6 [84]	9.5 [84]
Chemical composition, %				
C		81 [84]	53 [84]	19 [84]
H		0.03 [84]	0.8 [84]	0.5 [84]
N		0.2 [84]	0.3 [84]	0.1 [84]
O		8.4 [84]	2.5 [84]	9.4 [84]
Physical properties				
SA, m2/g		346-812[16, 76, 84]	404-520 [84]	13.2-48 [84]
PV, cm3/g		0.27-0.6 [84]	0.14-0.19 [84]	0.01-0.09 [84]
Soil-water distribution coefficient logKd (L/kg)				
PFBA (C4)	1.1-<2.3 [84]	3.01 [84]	1.1-<2.2 [84]	1-<2.2 [84]
PFHxS (C6)	1.2-1.8 [84]	3.5 [84]	1.4-1.8 [84]	1.3-1.8 [84]
PFOS (C8)	1.3-3.8[8, 16, 76, 89]	3.8-4.6[16, 76, 84]	1.8-3.1 [84]	1.8-3 [84]
PFOA (C8)	0.3-4 [76, 89]	3.3-4 [76, 89]	2.2-4 [84]	3-4 [84]
PFNA (C9)	2.3-<2.6 [84]	>3 [84]	2.2-<2.8 [84]	2.1-<2.8 [84]
Organic carbon normalized sediment-water distribution coefficient logKOC (L/kg)				
PFBA (C4)	1.8-<4.8 [84]	4 [84]	2.4-4.8 [84]	2.3-4.8 [84]
PFHxS (C6)	1.8-4 [84]	3.9-5.8 [84]	1.8-4[84]	1.8-4 [84]
PFOS (C8)	2.9-4.3 [76, 89]	4.3-7.2 [76, 89]	2.9-4.5[80]	3.1-4.4 [84]
PFOA (C8)	1.9-2.6 [8, 76, 89]	4.4-5.5 [76, 89]	2-4.7 [84]	2.1-4.6 [84]
PFNA (C9)	<2.8-<5.3[84]	>4 [84]	<2.9-<5 [84]	<3.1-<5.5 [84]

Hale et al. [16] observed that the addition of 3% PAC (in a specific area of 726 m^2/g) to contaminated soil in batch leaching tests reduced the leaching of PFOS by 94%-99.9%, from 6.4-54.5 µg/kg to 0.003-0.43 µg/kg. Modeling of these results using a one-dimensional box model indicated that this would reduce the concentrations of PFOS in pore water from 19.5 µg/L to 0.4 µg/L, just above the clean-up target of 0.3 µg/L. The same model indicated that PFOS would be released slowly from the untreated soils, taking more than 100 years to decrease the pore water concentrations of PFOS below the arbitrary target. This suggests that PAC might serve as an effective sorbent to diminish releases from contaminated soil to the surrounding environment, although more information is needed to confirm its efficacy. For example, further investigation is needed to determine the suitability of the method for field application, PAC's durability in soil, and the subsequent release of immobilized PFAS (i.e., the long-term treatment efficiency), as well as, the material costs and the wider environmental impacts of the process.

Biochar could immobilize PFAS in solution in sorption experiments (as shown by the Freundlich sorption coefficients presented in Table 2) but could not immobilize PFAS in soils in batch leaching tests [84]. This is probably due to the blockage of biochar pores with dissolved compounds and particles (e.g., humic substances and oil) in the soil [84]. Moreover, other soil components (e.g., DOC and ions) can compete with PFAS for the sorption sites [47, 90, 91]. The effectiveness of BC in immobilizing PFAS may be higher in the soils with lower organic content (e.g., total organic carbon (TOC)<3% in dry mass), indicating that for successful amendment, the amount of carbonaceous material added to the contaminated soil should exceed the initial carbon content of the soil [84, 92]. Thus, BC can be used for the sorption of PFAS in soils with low organic content.

The effectiveness of soil treatment with carbonaceous amendments (CAs), such as PAC and BC, depends not only on the CA but also on the chain-length of the PFAS. For example, the logarithm of the distribution coefficient ($\log K_d$) of PFBA in the soil stabilized with PAC is about 3.01 L/kg, whereas, the $\log K_d$ of PFOS in the same system varies between 3.8 and 4.8 L/kg [13, 28] (Table 3). These results show that PFAS with longer chains (e.g., PFOS and FOSA) bind more strongly to CA than those with shorter chains, indicating the importance of hydrophobic interactions in the sorption of PFAS (Higgins and Luthy, 2006). Moreover, PFAS with a sulfonate moiety have stronger adsorption than those with a carboxylate moiety (e.g., in the soil stabilized with PAC, the $\log K_d$ of PFOS reportedly varies between 3.8 and 4.6 L/kg, while the $\log K_d$ of PFNA is about 3; see Table 2). This is probably due to the slightly higher hydrophobicity of the slightly larger sulfonate moiety and/or differences in the specific electrostatic interactions of these moieties [60, 84]. Moreover, short-chain PFAS and fluorotelomers (which have a partially fluorinated carbon chain) can be more easily desorbed from the sediments with a low fraction of organic carbon (e.g., f_{oc} of 0.03%) in the aqueous phase than the species with longer chains or fully-fluorinated carbon chains (e.g., PFOS and FOSA) [84]. For example, Ahrens et al. [8] and Kupryianchyk et al. [84] calculated the carbon-normalized distribution coefficients and log (K_d/f_{oc}) values of 2.9-4.3 L/kg for PFOS and 2.1-2.6 L/kg for PFOA. Thus, it is important to add enough CA to PFAS-soil to ensure the stability of immobilized substances. Normally, PFAS-contaminated soils are remediated with 2%-4% CA (about 30-60 t/ha, assuming mixing to a depth of ca. 10 cm and soil density of ca. 1.5 kg/m^3) in-situ [11]. Whether this amount of CA is sufficient to stabilize soils in landfills remains unknown as no relevant information was found in the reviewed papers.

Table 3 Comparison of ∑PFAS in untreated and treated leachate with respective treatments [1, 87]. WAO-wet air oxidation, AC-activated carbon, CW-constructed wetlands.

Treatment method	∑PFAS in untreated leachate, ng/L	∑PFAS in treated leachate, ng/L	Landfill status	Reference
WAO	1889	1993	inactive	[1]
AC	31	9	inactive	[1]
AC	12819	4079	inactive	[1]
AC	8370	20	active	[1]
CW	814-4324	367-954	active	[87]

6.2.2 Treatment of PFAS-Contaminated Soil with Compost

Hale et al. [16] found that treating contaminated soil with 3% compost (composed of peat and cow manure with a high content of nitrogen, phosphate, and potassium) reduced PFOS leaching by 29%-34%, from 3.8 ± 2.4 to 2.54 ± 1.5 μg/L; the calculated distribution coefficient (K_d) of PFOS between compost and soil was 8.8 L/kg. Sorption of PFAS on compost occurs probably through direct polar interactions, hydrophobic interactions, ion-exchange, and addition to more specific sites of moieties with an affinity for PFAS present in the organic matter [16, 79]. More information is, nonetheless, needed on the sorption of PFAS using compost, the influence of co-contaminants, the stability of immobilized PFAS, and the efficacy of the treatment.

6.2.3 Treatment of PFAS-Contaminated Soil with Clay Minerals (Montmorillonite and Kaolinite)

Soil rich in clay minerals, such as montmorillonite and kaolinite, can be used to stabilize PFAS-contaminated soil. For example, Hale et al. [16] found that treating contaminated soil with 3% montmorillonite decreased the release of PFOS by 28%-40%, from 3.8 ± 2.4 μg/L to 2.7 ± 1.8 μg/L. The reported K_d values for the sorption of PFOS to montmorillonite were in the range of 5–10 L/kg [16, 93]. Hydrophobic and electrostatic interactions are the main mechanisms involved in the adsorption of PFOS to montmorillonite and kaolinite. More specifically, PFOS may be immobilized via the formation of outer-sphere surface complexes and specific interactions of the sulfonate group of PFOS with the hydroxyl groups on the surfaces of clay minerals [94]. However, dissolved humic substances in soil were shown to hinder the adsorption of PFOS on clay minerals via electrostatic repulsion and occupation of adsorption sites [93]. Thus, treatment with clay minerals can be effective for PFAS-contaminated soils with low organic matter, such as soils predominantly composed of stones and sand. Moreover, the addition of clay particles to such soil probably reduces its hydraulic conductivity, and hence, the contact between the contaminated soil and infiltrating water. Further research is needed on the sorption of PFAS in the presence of soil, the influence of co-contaminants, and the stability of immobilized PFAS.

6.2.4 Solidification of PFAS-Contaminated Soil

Sörengård et al. [86] tested the solidification of PFAS-contaminated soil with a combination of Portland cement (PC), fly ash (FA), and ground granulated blast furnace slag (GGBS) with a PC:FA:GGBS ratio of 1:1:2. This solidification was done together with chemical stabilization using several additives, such as PAC, Rembind® (AC-based and containing amorphous aluminum hydroxide and kaolin clay), pulverized zeolite, chitosan, hydrocalcite, and bentonite at a concentration of 0.2% (w/w) of dry PFAS-contaminated soil. The results showed that most PFAS (except the PFCAs with a perfluorocarbon chain length of less than five carbon atoms, e.g., PFBA and PFPeA) were successfully stabilized with CA in the solidified soil (e.g., $\log K_d$ increased from 1.5 L/kg for PFOS in the soil to about 3.5 L/kg treated with PAC) by hydrophobic sorption. Sörengård et al. recently demonstrated the long-term stabilization of PFAS in AFFF-contaminated soil, with a removal rate of up to 97% for the most important PFAS (PFOA and PFOS) [95].

The short-chains of perfluorocarbon could not be immobilized in significant amounts in stabilized/solidified soil (S/S-soil) under basic pH as they have a lower hydrophobic bond strength and faster diffusion in the water phase compared to the long-chains [86]. Hence, the S/S-soil, with large numbers of short perfluoroalkyl chains, needs pH adjustment to reduce leaching. Apart from the positive effects on the immobilization of PFAS, the addition of CA to soil exerts negative effects on solidification. For example, Sörengård et al. [86, 95] reported that unconfined compressive strength (as a measure of solidification) decreased by 38% and 14% in a carbon-poor sandy silt soil treated with 0.2% Rembind® and PAC, respectively. This means that the solidification of soils rich in organic carbon and clay through treatment with CA may entail high costs as these soils need significantly more binder in combination with PC and the addition of sodium silicate and/or lime compared to sandy soils [86]. Further laboratory studies should be performed before S/S-treatment is implemented in the field to optimize the S/S-binder methods for the specific contaminated soil, due to the complexity of the binding of soil to aggregates.

The stability of PFAS in S/S-soil over time is not quite clear. The carbonation of FA and PC may result in a decrease in the pH and improvement in the electrostatic interactions between PFAS and the treated soil. Contrary to the positive effects of carbonation, the aging of CA in S/S-soil may result in low sorption capacity for PFAS or even in the desorption of PFAS [58]. Thus, more research is needed to explore the stability of PFAS in S/S-soil over time.

6.3 Destruction of PFAS in Soil Using Thermal Treatment

Treatment of contaminated soil by exposure to high temperatures (850-1200 °C) may be an effective technique to mineralize PFAS [96]. For example, incineration in a fluidized bed incinerator at temperatures of up to 900 °C can reduce the PFOS levels to below the detection limits in the exhaust air (15 ng/m^3), while also reducing solid residues (10 µg/kg dry matter) and water discharges (25 ng/L) [96]. However, incineration of PFAS-contaminated soil together with other wastes may lead to the formation of highly volatile and mobile 1H-perfluoroheptane, dioxins, furans, tetrafluoromethane, and hexafluoroethane (C_2F_6) [97-99]. Thus, the addition of appropriate substances, such as $Ca(OH)_2$, may be required to reduce the emission of these harmful by-products [100]. More research is needed to better understand the effects of incineration on PFAS and the by-products formed. Thermal treatment is not suitable for large masses of soil contaminated with PFAS to be subsequently landfilled due to the high costs involved [9].

7. Treatment of Landfill Leachate Contaminated with PFAS

In modern landfills, leachate is collected and treated on-site at local treatment plants and then discharged into a recipient water body or pumped to WWTPs [65]. Particularly in Sweden, the landfill leachate must be treated before it is delivered to WWTPs to meet the quality requirements set by the WWTP operators. Most of the traditional treatment systems for landfill leachates are not designed to remove PFAS, and thus, the extent to which they reduce the PFAS levels is not well-known due to a lack of published data. The processes for removing PFAS from landfill leachate discussed in the literature include separation of PFAS from leachate by sorption, biological treatment, membrane-based treatments (such as nanofiltration and reverse osmosis), and compacted wetlands (Table 3). Destruction techniques used for removing PFAS from groundwater, such as electrochemical treatment, thermal treatment, wet air oxidation, and advanced oxidation processes (such as photolysis, photocatalysis, and activated persulfate oxidation), have also been discussed.

7.1 Separation of PFAS from Landfill Leachate

7.1.1 Sorption

The sorption of PFAS to GAC (e.g., Filtrasorb 300 and 600, or AquaCarb 1240) and anion exchange (AE) materials (such as Purolite® A600) has been tested for the treatment of landfill leachate, groundwater, and drinking water [1, 58, 101]. These sorbents are more effective for eliminating long-chain PFAS than short-chain PFAS (e.g., PFOS>PFBS, in terms of removal efficiency), but they have a low sorption capacity [47, 76, 85]. As shown in Table 3, Busch et al. [1] and Yu et al. [76] found that AC can adsorb 68.2% to 99.8% of PFAS. However, in these studies, ∑PFAS in the treated leachates remained high (e.g., 4,079 ng/L) due to the high concentrations in raw leachate (e.g., 12,819 ng/L) and the short residence times of the leachate through the AC filters, or high mass flows. Additionally, DOC in leachates may compete with PFAS for the adsorption sites. Short-chain PFAS, such as PFBA (C_4), PFPeA (C_5), and PFHxA (C_6), maybe inefficiently adsorbed because they are less hydrophobic compared to long-chain PFAS, such as PFNA (C_9), PFDA (C_{10}), and PFOS (C_8), and may, therefore, be displaced by long-chain PFAS and DOC [58]. Thus, the selective nature of PFAS removal and the associated desorption of short-chain PFCAs during the co-removal of multiple PFAS must be considered during the designing and operation of the adsorption treatment processes.

7.1.2 Membrane Technology

Reverse osmosis (RO) and nanofiltration (NF) involve the use of semi-permeable membranes (with 1-10 nm and <1 nm pores, respectively) to remove contaminants from leachates. These are physical separation technologies that transfer PFAS from the leachate to a secondary phase (rejected water or concentrate), which then needs further treatment. Appleman et al. [101] reported that NF removed PFOS, PFOA, PFDA, PFPeA, and PFBA to below-detection limits (with >97% removal efficiency) from artificial groundwater spiked with 1 µg/L of each PFAS; PFBS and PFHxS were present in extremely low concentrations (e.g., 10 and 20 ng/L, respectively) in the permeate. Soriano et al. found that the NF90 membranes with a smaller pore size (0.34 ± 0.02 nm) rejected over 99% of PFHxA at 100 mg/L from the industrial effluents [102]. Their results showed that size exclusion and electrostatic interaction occurred simultaneously and affected the selectivity of PFHxA separation. Reverse osmosis in WWTP was able to remove PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, and PFOS below the detection levels from their initial concentrations, which varied between 3 µg/L for PFNA and 39 µg/L for PFOS [103]. After membrane-treatment, PFAS were concentrated in the rejected water, which was about 15% of the treated water volume for RO [103]. Soriano et al. showed an efficient removal of PFHxA (over 99%) using XLE and BW30, two types of RO membranes [102]. These results indicate that both NF and RO can be effective in treating PFAS of a wide range of molecular weights. Franke et al. removed PFAS from drinking water using a combination of NF with AE or GAC adsorbents [104]. In the cited studies, water with no DOC was used, while the landfill leachates may contain significant amounts of DOC. Thus, more research is required on the membrane filtration treatments of PFAS in landfill leachate. Although RO and NF showed promising results in removing PFAS from contaminated water, Fuertes et al. [71] found a two-fold increase in the concentration of PFAS in the membrane bioreactors of the effluent treatment plants (using ultrafiltration treatment) compared to the raw leachate, indicating degradation of the precursor compounds, which increased the levels of some PFAS [71]. These results indicated that not all membrane methods are suitable for treating PFAS-contaminated water.

7.1.3 Biological Treatments

Biological treatments (BIO) applied by Busch et al. [1], Yan et al. [3], and Fuertes et al. [71] were found to be ineffective in removing PFAS from landfill leachate. Instead, the concentrations of PFCAs and PFSAs were increased with the use of these types of treatments (Table 3). This could have occurred due to the resistance of PFAS to biodegradation owing to their high-energy carbon-fluorine bonds [105], and the possible biodegradation of precursor compounds (such as N-ethyl perfluorooctane sulphonamidoethanol and fluorotelomer-based compounds, or other unidentified components) to PFCAs and PFSAs, as observed for the biological processes in WWTPs [7, 19, 106].

Constructed wetlands (CWs) include various kinds of aerated lagoons, sedimentation tanks, reed beds, and polishing ponds. Multiple remedial processes may be conducted simultaneously in these systems, including biodegradation (aerobic and anaerobic), phytoremediation (plant uptake and enhancement of biodegradation), and sorption on diverse substances [87]. During tests in a large tropical wetland, 61% of ∑PFAS and 50%-96% of individual PFAS were removed from landfill leachate. The most effective processes were sorption of long-chain PFAS to soils and sediments and uptake of short-chain PFAS by plants. These processes removed about 44% of ∑PFAS during the passage of the leachate through a reed bed. Additionally, an aeration lagoon removed 55%-73% of the PFAS precursors through biotransformation, but it was ineffective in the removal of long-chain PFAS. This approach may only be suitable as a pre-treatment for landfill leachate as the concentrations of ∑PFAS in the treated leachate were still high (Table 3). However, the use of young plants or species with a high affinity for PFAS, and the substrates with high PFAS sorption capacity can potentially improve the removal of PFAS from landfill leachates in CWs.

7.2 Techniques for the Destruction of PFAS in Landfill Leachate

7.2.1 Oxidation Processes

Chemical oxidation involves the use of oxidants, such as sodium permanganate ($NaMnO_4$), sodium persulfate ($Na_2S_2O_8$), hydrogen peroxide (H_2O_2), or ozone (O_3) to degrade organic pollutants and decontaminate water. For example, H_2O_2 mixed with water generates hydroxyl radical ($OH^•$), which is an extremely strong oxidant (E = 2.33 V) that reacts with organic molecules with the reaction rate-constants ranging between 10^6 and 10^{10} Ms^{-1} [107]. H_2O_2 is often used in conjunction with Fe^{2+} as a catalyst.

The efficiency with which the chemical oxidation processes degrade PFAS strongly depends on the pH, temperature, and initial concentration of the oxidant. Ozone does not degrade PFOA and PFOS significantly at pH 4-5 [108], but $NaMnO_4$ has been shown to degrade 47% of PFOS in an aqueous solution at pH 4.2 in 18 days. Increasing the pH to about 10-11 (by adding NaOH to the solution) decreases PFOS degradation to about 10% of the that achieved by treatment with $NaMnO_4$, while the addition of 8.7 g O_3/h (2.5 wt%) reduces the concentrations of PFOA and PFOS from 50 µg/L to about 5 µg/L and 7.5 µg/L, respectively, after 4 h [108, 109].

Ozonation is less efficient without pre-treatment; the results are best when O_3 is added at an acidic pH and the process is continued by adding O_3 at an alkaline pH [108]. At an acidic pH, O_3

decomposes slowly via reaction with hydroxide ions (as their concentrations are low), and dissolved O_3 accumulates in the solution. When the concentration of O_3 is sufficiently high in the solution (e.g., 3.5 mg/L), making the solution alkaline, it generates large amounts of $OH^\bullet$ that react rapidly with O_3 and form abundant reactive free radicals, including potent hydroxyl radicals that can efficiently degrade PFOA and PFAS. Accumulation of O_3 at an alkaline pH (without pre-treatment) in the solution is poor (e.g., about 0.3 mg/L at pH = 11) as the large amounts of $OH^\bullet$ continually react with O_3, causing the concentrations of reactive free radicals to be low throughout the process [108]. The process can be improved by increasing the dose of the oxidant and the temperature (e.g., the kinetic rate constant of PFOS increased from 0.46×10^{-2} L/d at 25 °C to 3.64×10^{-2} L/d at 65 °C) in the tests or when combined with other processes, such as photolysis [92]. A synergistic effect was observed when ozone was combined with UV or with air fractionation, whereby the removal of PFAS reached 73% and 95%, respectively [108].

Heterogeneous photocatalysis is a promising solution for the removal of PFAS from contaminated water. This technique directly uses photons (*hv*) from a VUV or UV to generate negatively-charged electrons (e^-) and positively-charged holes (h^+), which have high oxidation capacity [110]. The positively-charged holes react with water, yielding $OH^\bullet$ that can effectively degrade diverse organic compounds. The negatively-charged electrons migrate to the surface of the photocatalyst and react with the adsorbed water to form hydrated electrons (e^-_{aq}). PFAS sorbed on the catalyst's surface are attacked by these hydrated electrons and degraded into shorter-chain compounds [110].

Photocatalysis with TiO_2 may degrade PFCAs (e.g., PFOA, PFNA, and PFDA), but several hours are required for satisfactory results. For example, Panchangam et al. reported that TiO_2 irradiated with a 16 W (254 nm) low-pressure mercury lamp could degrade 99% and mineralize 38% of PFOA and PFDA in 7 h [111]. The treatment time could be reduced (several-fold) by adding acid to the treated solution, for example, adding 0.225 M perchloric acid ($HClO_4$) reduced the pH of the solution to 3 and cuts down the treatment time for PFOA from 24h to 7h (Ibid.). According to Panchangam et al., the presence of acid improves the ionization of PFCAs by increasing the chances of electron transfer from PFCAs to the photoholes of excited TiO_2, thereby prolonging the life of the photoholes [111]. The process can be further improved by the recovery of the acid.

Furthermore, a recent development on the photocatalytic degradation of PFOA using a TiO_2-rGO (0.1 g L^{-1}) composite catalyst was shown to degrade up to 93.7% of PFOA compared to the TiO_2 photocatalysis (24% ± 11%) and direct photolysis (58% ± 9%) [112]. The degradation efficiency was related to several parameters, which included the initial PFAS concentration, photocatalyst, UV lamp intensity, and media.

β-Ga_2O_3 is relatively more efficient as a photocatalyst than TiO_2 for PFCA degradation owing to the wider band-gap of the former (4.8 eV for βGa_2O_3 compared to 3.2 eV for TiO_2) and the higher position of its conduction band relative to the vacuum energy level (e.g., ECB values: - 2.95 eV and - 4.21 eV, respectively) [113]. For example, Shao et al. found that PFOA was completely degraded after 65 min of exposure to upgraded β-Ga_2O_3 along with VUV irradiation at 185 nm and pH 4.3 [114]. The catalytic β-Ga_2O_3 had a sheaf-like nanostructure consisting of plates elongated in the [1 0 0] direction, a specific surface area of 36.1 m^2/g, and high densities of the nanopores (2-4 nm and 8 nm). In contrast, commercial β-Ga_2O_3 degraded only 38% of the PFOA in 3 h (which was still two times higher than the rate obtained with P25 TiO_2) [114]. On the other hand, boron nitride (BN) was found to be quite an efficient catalyst, being 2 to 4 times more active than TiO_2 [115]. Duan et al.

[115] showed that low concentrations of PFOA were detected after 240 min of photolytic degradation using BN [115].

The removal of PFAS from landfill leachate through photocatalysis may be affected by the presence of dissolved organic matter (DOM), bicarbonate, and chlorides. The DOM and bicarbonate in leachates probably impair the photodegradation process by occupying the catalyst's surface, thereby reducing its adsorption capacity for PFAS [110]. Thus, applying photocatalysis as the final step may be an effective strategy in leachate treatment (when most of the DOM has been removed in the previous steps). Another option may be to add O_3 gas to promote the removal of long-chain PFAS (e.g., PFOA), i.e., by increasing the rate at, which photo-produced H^+ reacts with PFOA, rather than recombining with photo-produced e^- [112]. Additionally, the pH of the leachate should probably be adjusted to about 4 to promote high adsorption of PFAS and enhance the performance of e^- and H^+ pairs [110]. Moreover, future research should include the evaluation of the efficiency of photocatalysis in treating short-chain PFAS in landfill leachates.

In electrochemical treatment (ET), "non-active" anodes composed of materials such as Ti/RuO_2, SnO_2, PbO_2, and boron-doped diamond (BDD) are used for degrading and mineralizing non-biodegradable organic compounds, such as PFOA, PFBA, PFHxA, PFDA, PFBS, PFHxS, and PFOS [116-118].

The electrochemical technique is reported to efficiently remove PFAS from wastewater and groundwater, but the required treatment time might be quite long. For example, Gomez-Ruiz et al. reported that ET treatment with a BDD electrode removed 99.7% of diverse PFAS from wastewater [119], with their total concentration dropped from 1,652 μg/L to 4.2 μg/L during 10 h of treatment at a current density of 50 mA/cm^2 and a voltage between 13.9 and 15.3 V. Similarly, Zhuo et al. found that a current density of 0.59 mA/cm^2 and an oxidation potential greater than 2.76 V were adequate for removing about 97.5% of PFOA from spiked water (at the concentrations of 20, 30, and 50 mg/L) after 2 h of electrolysis [120]. Besides the electrochemical parameters (such as current density and voltage), the success of ET depends on the pH and the initial concentrations of PFAS. The degradation rate of PFOA is higher at an acidic pH than at an alkaline pH. At alkaline pH, OH^- migrates to the anode surface due to electrostatic attraction, which limits the sites available for degraded $CF_3(CF_2)_6COO^-$ anion [120]. The removal ratio was shown to increase with an increase in the initial PFOA concentration. Thus, ET may be an effective treatment for water with high levels of PFAS contamination at an acidic pH.

Regarding the treatment of landfill leachate, Witt et al. recently demonstrated that 98% of PFOA was removed electrochemically under a flow through a BDD cell in a 20 L pilot water treatment system [121]. However, this technology is not fully developed for commercial applications, partly due to high energy demands, which increases the costs of operation. For example, 99.7% removal of PFAS from contaminated water would require approximately 256 kWh/m^3 (corresponding to about 14 euro/m^3) [119]. Thus, large-scale implementation of ET would require pre-treatment of the PFAS-contaminated water (e.g., using membrane separation) and the prevention of its mixing with diluting water streams (to reduce the volumes requiring treatment).

7.2.2 Thermal Treatment

The thermal treatment methods for water include sonochemistry, sub-critical and supercritical water treatment, microwave-hydrothermal treatment, and wet air oxidation (WAO).

Sonochemical treatment refers to the use of ultrasonic waves at the frequencies of 20-1000 kHz to form cavitation bubbles in water, and hence, high vapor temperatures (near 4,000-5,000 K) in the vapor core. Thus, PFAS may be pyrolytically decomposed at bubble–water interfaces [122]. Decomposition of PFOA and PFOS results in the production of mineralized fluoride (F^-), sulfate (SO_4^{2-}), CO, and CO_2 [123]. Campbell and Hoffmann showed that the treatment's efficiency relied on the frequency of the ultrasonic radiation and the power density [124]. The measured rate constants of PFOS, PFOA, PFHxA, PFHxS, and PFBS showed that degradation increased linearly with the increase in power density (e.g., the rate constant for PFOS increased from 0.01 L/min at 100 W/L to 0.045 L/min at 330 W/L at the same frequency of 358 kHz). Similarly, the rate of PFAS degradation was shown to increase with an increase in the frequency (e.g., the rate constant for PFOS increased from 0.045 L/min at 358 kHz to 0.06 L/min at 610 kHz at the same power density of 330 W/L). Moreover, the degradation rates of PFOA and PFOS were enhanced by 12% and 23%, respectively, through the application of a dual-frequency (20 and 202 kHz). The increase in degradation with the application of dual-frequency may be attributed to a better overlap of acoustic waves (which enhances bubble expansion and shortens collapse time), thereby strengthening the cavitation effects and the induction of sonochemical reactions [125]. The treatment cost can be reduced by combining sonification with oxidation, for example, by using persulfate ($S_2O_8^{2-}$) oxidant to generate sulfate radicals ($SO_4^{\bullet-}$), which increases the efficiency of the process [126-128]. Nevertheless, the higher cost of the sonochemical treatment process compared to other destructive methods may limit its application in the treatment of PFAS [129].

According to Hori et al., pure subcritical water treatment cannot degrade PFOS, but it can degrade PFOS efficiently when used in combination with the addition of zero-valent metals (e.g., Al, Cu, Zn, Fe) to the reaction system [125]. Zero-valent iron, Fe (0), provided the best synergistic results for the degradation of PFOS, followed by Zn, Cu, and Al (in decreasing ability of degradation), in the tests performed by the researchers. For example, adding Fe(0) powder (>99.9%, <53 µm at 4.8-19 mmol concentrations) to subcritical water treatment mixtures reduced the concentrations of PFOS from 93-372 µM to <2.2 µM (at 350 °C, 22 MPa after 6 h), with the formation of F^- ions, 46%-51% and no PFCA formation (Ibid.). Additionally, shorter C-F chains, such as PFHxS, could be degraded from the initial concentrations of 741 µM to 97 µM using subcritical water treatment combined with the addition of 19.3 mmol Fe [130]. This process can be described as the adsorption of PFAS to the Fe surface and the decomposition of PFAS with rising temperature along with the release of F^- ions in the solution phase above 250 °C [125]. Thus, zero-valent Fe powder with a large specific surface area should be used in subcritical water treatment to improve the decomposition of PFAS.

Supercritical water treatment is more efficient than subcritical water treatment. For example, in other tests reported by Hori et al., these treatments resulted in the decomposition of 94.8% and 84% of PFHxA, respectively, using the same concentration of Fe (9.6 mmol) [130]. However, supercritical water treatment caused an 8-fold increase in fluoroform (CF_3H) creation compared to subcritical water treatment. Thus, supercritical water treatment of PFAS may be problematic from an environmental perspective as CF_3H is a greenhouse gas with high diffusivity, low viscosity, and an atmospheric lifetime of approximately 270 years [131]. Moreover, both subcritical and supercritical water treatments may encounter operational problems, such as severe reactor corrosion (caused by acids formed during the treatment, such as H_2SO_4) or serious plugging of the reactor caused by the salts precipitating at high temperatures and low densities. This technique is not suitable for treating PFAS in landfill leachate due to operational problems and extremely high costs [132].

Lee et al. found that microwave (MW) irradiation at 140 W and 130 °C for 8 h degraded only 3% of PFOA (with an initial concentration of 241 µM in the solution) [133]. In combination with 5 mM $S_2O_8^{2-}$, the method degraded 35% and 68% of PFOA at 60 °C and 90 °C, respectively, after 4 h. The process can be further improved by adding more $S_2O_8^{2-}$. For example, in the study, the decomposition rate constant increased from 0.74 to 0.88 L/h at 90 °C when the concentration of $S_2O_8^{2-}$ increased from 5 to 10 mM at acidic pH (e.g., 2.5) [133, 134]. The degradation rate of PFOA can be further increased by adding zero-valent iron; for example, Lee et al. recorded 58% degradation of PFOA in the presence of 3.6 mM Fe(0) and 5 mM $S_2O_8^{2-}$ after 1 h of MW irradiation at 90 °C [133]. Aerobic and anaerobic corrosion of Fe(0) generate Fe^{2+}, which accelerates the decomposition of $S_2O_8^{2-}$ by acting as a transitional metal activator [135]. It is important to determine the optimum concentration of Fe(0) as surplus Fe(0) may release large quantities of Fe^{2+} ions that consume $SO_4^{\bullet-}$, thereby reducing the decomposition efficiency of PFAS [133, 136]. Moreover, chloride ions may reduce the decomposition of PFAS as they react with free sulfate radicals that limit the oxidation efficiency [134]. Thus, degradation of PFAS by using MW irradiation in combination with oxidants may be an option for treating contaminated leachate with low buffer capacity (since pH should be adjusted to acidic values) and low concentrations of Cl^-.

Busch et al. [1] and Silva et al. [137] reported that PFAS treatment using wet air oxidation (WAO) was ineffective in removing PFAS from landfill leachate, most likely due to a short treatment time [1, 137] (Table 3). High concentrations of PFAS in leachates have been observed after treatment with WAO (Table 3), which can be explained by the degradation of fluorotelomer precursors to PFCAs [138].

8. Conclusions

The Registration, Evaluation, Authorization, and Restriction of Chemicals (REACH) should focus not only on long-chain but also on short-chain PFAS (such as PFBA and PFBS) because of their persistence, high mobility in water and soil, and difficulty in remediation and water purification. The use of PFAS in products should also be better regulated to reduce their release into the environment. Directives on the definition and classification of waste should be more specific about the concentrations of PFAS in wastes, such as contaminated soil. The Landfill Directive should include the maximal concentrations of soluble PFASs in waste that can be disposed of in landfills. The introduction of legal limits for soluble/mobile PFAS in contaminated soil would reduce the amount of PFAS in landfill leachate. Additional requirements to treat the soil before landfilling and improvements in the treatment techniques would further reduce the release of PFAS into the surrounding environment.

“Digging and dumping” continues to be the most widely used remediation technique at the sites contaminated with PFAS as the current in-situ and on-site remediation techniques are not reliable. Excavated soil may contain high amounts of PFAS and may comprise one of the major waste streams at landfills in the future. These soils have the potential to contaminate landfill leachate with PFAS, which places new demands on the leachate treatment systems at landfills. PFAS-contaminated soil, therefore, requires treatment before landfilling to reduce its potential to contaminate landfill leachate. The stabilization of PFAS-contaminated soil with AC, compost, Fe-oxides, or clay minerals may be an option to reduce the leaching of PFAS from landfilled soil. The potential to treat PFAS-contaminated soil with another waste (e.g., carbon-rich waste) should be studied as this may reduce treatment costs, thereby enhancing sustainability. More research on the longevity of stabilized contaminants in the soil at landfills is also needed.

The installation of a cover (or encapsulation) decreases the volume of leachate in contact with the PFAS-contaminated soil in a landfill. PFAS-contaminated soil should not be disposed of in a landfill together with alkaline materials to avoid increasing the mobility of PFAS. Waste in a landfill may act as a filter for leached PFAS. Furthermore, co-contaminants, including minerals and natural organic matter, can significantly affect the remediation. Thus, the transport of PFAS through landfills should be studied in more detail.

Biological methods are not suitable for the treatment of PFAS in landfill leachate as they are inadequate in the reduction of PFAS concentrations. Membrane-based treatments, in combination with sorption on AC, might be viable for treating PFAS in landfill leachate. Advanced oxidation and thermal treatments also show promising results for degrading PFAS. Any treatment process selected should be optimized for, inter alia, leachate flow, treatment time, and concentrations of PFAS in untreated leachate, as well as, the desired levels of PFAS in the treated leachate. Foam fractionation is another technique that is not well-documented in the scientific literature. Additional research is needed to study the transformation of PFAS from contaminated water to foam, the efficiency of the process, and the treatment costs.

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Author Contributions

Igor Travar: Conceptualization, writing, and reviewing. Jean Noel Uwayezu: writing and reviewing. Jurate Kumpiene: reviewing and editing. Leo W.Y. Yeung: reviewing and editing.

Competing Interests

The authors have declared that no competing interests exist.

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LAND POLLUTION (GM HETTIARACHCHI AND A JUHASZ, SECTION EDITORS)

A Review of Immobilisation-Based Remediation of Per- and Poly-Fluoroalkyl Substances (PFAS) in Soils

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Abstract

Purpose of Review This review provides an overview of the latest developments in immobilisation of per- and poly-fluoroalkyl substances (PFAS) for soil remediation. It examines the efficacy of a range of amendments, including those with binding agents, along with a discussion of immobilisation mechanisms and post-immobilisation assessment needs.

Recent Findings Researchers have recently applied a variety of soil amendments to soil for PFAS immobilisation. Efficacy of these has varied widely, both between amendment and soil types and for individual PFAS molecules present in contaminated soils. Activated carbon based amendments, including composite amendments exhibit the highest efficacies of the examined studies.

Summary Immobilisation of PFAS is complex, with efficacy of immobilisation varying with soil properties including pH, clay and organic matter content, amendment properties, and molecular properties of the individual PFAS. Optimal remediation strategies need to be adjusted accordingly to site specific soil properties and contamination profiles. Additionally, bioavailability testing needs to supplement standard leaching approaches to determine effectiveness of PFAS soil immobilisation strategies.

Keywords PFAS · Soil remediation · Immobilisation · Soil amendments · Leachability

Introduction

Per- and poly-fluoroalkyl substances (PFAS) are a group of man-made chemicals found in fire-fighting foams, water repellent coatings, and consumer products such as carpets, textiles and food packaging [1, 2]. In recent years, PFAS have emerged as contaminants of concern due to their potential for persistence, mobility and bioaccumulation in humans and the environment. PFAS contamination is complex, with PFAS consisting of thousands of different compounds (over 3,000 of these commercially available) [3] although only a limited number are routinely assessed in environmental samples. Of these, perfluorooctan`e sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and perfluorooctane sulfonyl fluoride (PFOSF) are listed in the Stockholm convention, and perfluorohexane sulfonic acid (PFHxS) is listed as potential compounds to be added [4]. Consisting of a fully or partially fluorinated carbon chain and a functional group, PFAS exhibit surfactant-like properties, are characterised by high octanol-water partitioning coefficients (e.g. log K_{OW}=3.94-6.56 for a range of PFAS [5]) but high aqueous solubility and are proteinophilic rather than lipophilic like other persistent organic pollutants. Exposure to PFAS has been shown to result in a number of adverse health effects [6] including hepatotoxicity [7], immunotoxicity [8], neurotoxicity [9], and developmental toxicity [10] although the molecular mechanisms influencing these effects are only beginning to be elucidated [11]. Environmental concentrations in some organisms have also been measured that exceed experimentally derived lowest observable adverse effect levels (LOAEL) [12, 13].

Over the past 5 years, there has been a substantial increase in research on PFAS[1], including the fate of PFAS in the environment, their bioaccumulation in aquatic and terrestrial food webs and approaches for remediation and risk management. While a large proportion of research has been undertaken on

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[1] Web of Science topic search results for “PFAS” 2016: 141, vs 2020: 561, accessed 15/06/2021.

PFAS impacted water, and associated technologies for its removal, less information is available on strategies for the treatment of contaminated soil. PFAS contamination of soil is prevalent at fluorochemical manufacturing sites [14], firefighting training grounds [15], landfill sites [16], airports and air-force bases [17] along with sites where PFAS contaminated biosolids [18], wastewater [19] or recycled water [20] have been utilised. Given the number of possible per and polyfluoroalkyl congeners in commercial products, PFAS contamination from different sources may vary widely in terms of concentration and composition. For example, Brusseau et al. [21] determined that the median background concentration of

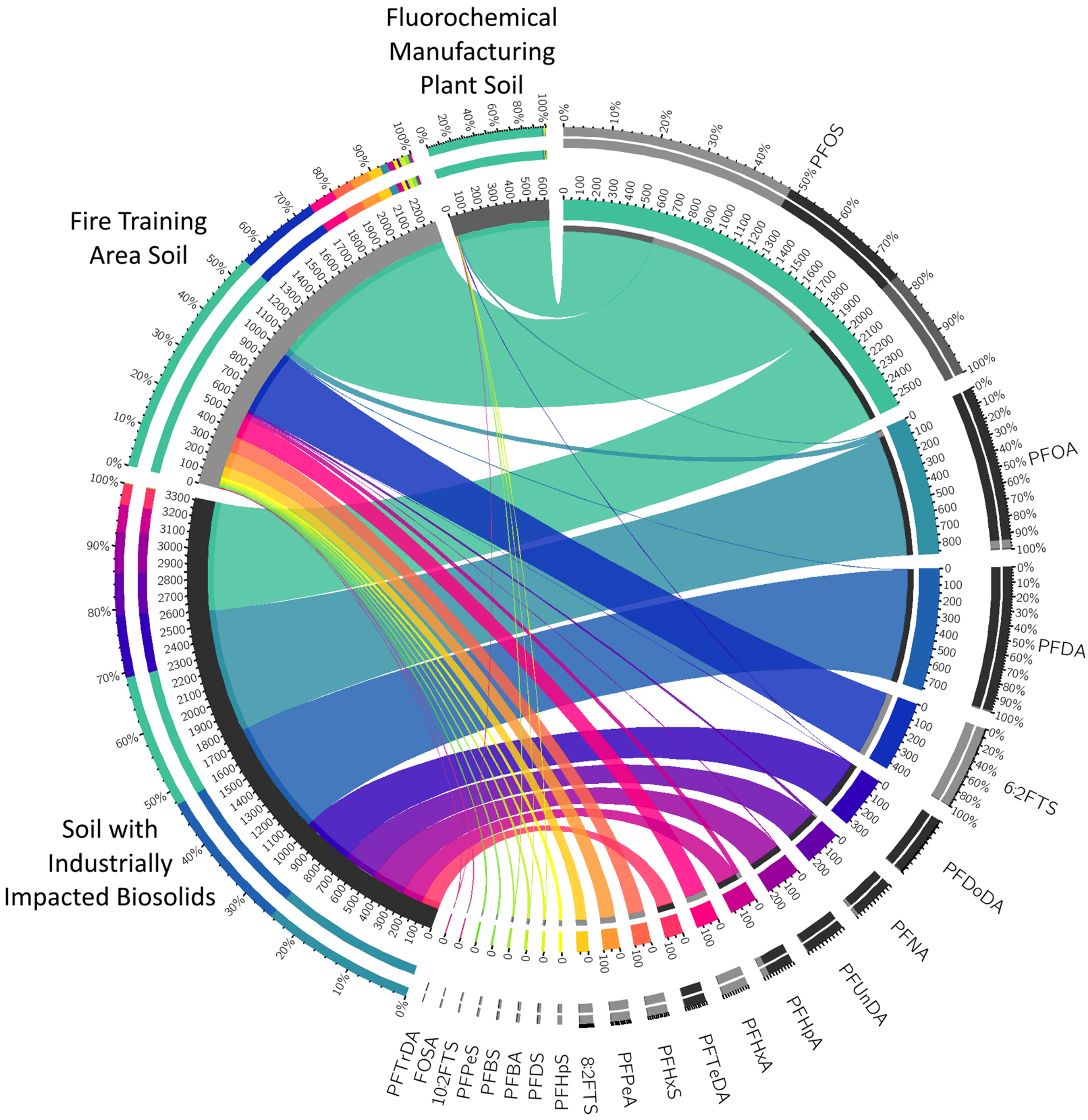

Fig. 1 Composition of PFAS contamination from three locations; soil from an AFFF fire-fighting training ground, a soil with industrially impacted biosolids [22], and a soil from a fluorochemical manufacturing plant [14]. On the right hand side of the circle, moving clockwise, individual PFAS are shown ordered by decreasing concentration (μg kg^{-1}). This is followed by location type, ordered by decreasing total PFAS concentration. The size of the link connecting an individual PFAS with a location type represents the concentration, sized according to the inner numbered scale (μg kg^{-1})

PFOS and PFOA in soil from a large range of international sites where neither primary nor secondary PFAS contamination occurred, was 2.7 μg kg^{-1} compared to up to 460 g kg^{-1} (PFOS) and 11 g kg^{-1} (PFOA) at contaminated sites. PFAS congeners in contaminated soil may vary considerably, as depicted in Fig. 1 where three example PFAS contamination profiles representing impacts from fluorochemical manufacturing, the use of aqueous film-forming foam (AFFF) at fire-fighting training grounds and biosolid amended soil are shown. PFAS profiles may range from being dominated by a single compound (e.g., perfluorooctane sulfonic acid, PFOS) in impacted soil from a manufacturing plant [14] to having a wide range of congeners present. A soil from a fire-fighting training area contained significant proportions of both PFOS and 6:2 fluorotelomer sulfonic acid (6:2 FTS), along with smaller amounts of perfluorohexanoic acid (PFHxA) and PFHxS among others. Due to wastewater treatment plants (WWTPs) having PFAS inputs from multiple sources, biosolids and biosolid-amended soil [22] may contain a range of PFAS often dominated by long-chain PFAS ($\geq C_7$) due to their preferential retention (in biosolids) during wastewater processing [23].

Irrespective of the activity leading to soil contamination, potential adverse environmental health effects associated with PFAS exposure has necessitated the development and application of remediation technologies. However, due to the unique physicochemical properties of PFAS, some traditional remediation strategies are ineffective for PFAS remediation. In particular, the strength of the C-F bond limits the application of chemical oxidation and bioremediation strategies and in some cases, oxidation of poly-fluoroalkyl and/or precursor compounds may increase the concentration of recalcitrant perfluorinated carboxylic acids (PFCAs) [24]. Soil washing results in secondary liquid waste that requires additional treatment, while phytoremediation is slow, treatment zones are dependent on plant root depth, and uptake may be limited by mass transfer effects. While thermal treatment technologies, such as incineration with excess oxygen, gasification and pyrolysis have the potential to destroy PFAS by breaking C-F bonds, temperatures in excess of 1000 °C are required to minimise the production of greenhouse gases [25, 26].

Immobilisation techniques, whilst not removing PFAS from the soil matrix, offer a risk management strategy through a reduction in leachability and/or bioavailability. The effectiveness of immobilisation strategies is influenced by a wide variety of parameters, including soil and amendment properties in addition to PFAS profiles in contaminated soil. While recent PFAS remediation reviews have provided a synopsis of the application of established technologies for PFAS contaminated soil and sediment [27–31] or focused on emerging treatment strategies [32], this paper provides a comprehensive overview of soil-based PFAS immobilisation approaches including amendment properties, treatment efficacy, immobilisation mechanisms, existing data gaps and future research priorities.

Immobilisation

Immobilisation is a strategy which reduces the mobility and bioavailability of contaminants through the addition of ‘sorptive’ soil amendments, thereby reducing exposure for human and ecological receptors. Amendments may be applied either in situ or ex situ and used with or without additional binding agents (e.g. Portland cement). Soil amendments offer the advantage of ease of application, cost-effectiveness and commercial viability, however, selecting suitable amendments that exhibit high sorptive capacity is critical for delivering sustainable immobilisation approaches. While immobilisation strategies are well developed for many legacy contaminants (e.g. metal(loid)s, PAHs), amendments and immobilisation approaches for PFAS have only recently gained research, development and commercial attention. Table 1 provides details of amendments that have been applied for PFAS immobilisation including activated carbon, biochar, clay, polymer and composite sorptive phases. A comparison of physicochemical properties which influence PFAS sorptive capacity and immobilisation efficacy, which in many cases, may be manipulated during amendment development; along with advantages and disadvantages of each amendment type are discussed in detail in the following sections.

Activated Carbon-Based Amendments

Activated carbon is a commonly used sorbent for the removal of contaminants from (waste)water due to its high surface area, high pore volume and surface functional groups. This sorbent has also been applied to contaminated soil for the immobilisation of a variety of organic and inorganic compounds including PFAS [43, 45, 46]. Activated carbon’s effectiveness for PFAS sorption is due its high surface area, size and number of pores. However, physicochemical properties and sorptive potential of activated carbon varies depending on the carbon source and temperature used for its production [47]. These parameters influence porosity, surface area and surface functional groups whereby increased production temperature decreases surface functional groups, and in doing so, increases surface charge [47]. Activated carbon has the ability to form both electrostatic and hydrophobic interactions with PFAS, and possibly hemi-micelle and micelle formation in pores [48]. Charge-assisted hydrogen bonding was also suggested from experimental spectroscopy of PFAS interacting with functional groups on the surface of activated carbon [49].

Table 1 Laboratory and field-based immobilisation studies detailing the impact of soil amendment on PFAS leachability

Amendment category	Amendment type(s)	Technique	Application rate (w/w)	Ageing period	Soil properties	Initial soil conc. (μg kg^{-1})	PFAS tested	Leaching method/details	Author, year and reference
Activated carbon	CB + GAC	Stabilisation and solidification	15% CB, 0.2% GAC	12 months	AFFF contaminated (sand 30%, clay 42%, silt 28% pH 8.6, OC 6%)	Not stated	C_{5-8} PFCAs, $C_{4,6,8}$ PFSAs, 6:2 FTS, PFOSA, 3:2 FTOH, HpF-PipB	EN 12457/in situ, 0.1-2.0 mm particles	Sörengård et al. (2021) [33]
Activated carbon	Colloidal activated carbon (PlumeStop®)	Sorbent amendment (injection)	1%	10 months	Spiked 0.6 mg kg^{-1} 10 soils with varying OC, clay	Σ_{18}=10,800	$C_{3-11,13,15,17}$ PFCAs, $C_{4,6,8}$ PFSAs, 6:2,8:2 FTS, PFOSA	1:7 liq:soil, 0.45 μm filter	Sörengård et al. (2019b) [34]
Biochar	Activated and unactivated waste timber biochar	Sorbent amendment (ex situ)	0-5%	Not stated	AFFF contaminated, 1 low, 1 high TOC	Σ_{21}=1200-3800	$C_{4,6,7,8,10}$ PFSAs, $C_{4-10,12-14,16}$ PFCAs, 4:2, 6:2, 8:2 FTS, HPFHpA, DMOA	Modified CEN EN 12457 <1mm, 1.2 μm filter	Sørmo et al. (2021) [35]
Biochar	Biochar (pine, 750 °C)	Sorbent amendment	0-5%	Not stated	Spiked sandy clay, loamy sand	Not stated*	$C_{6,8}$ PFCAs, $C_{6,8}$ PFSAs	0-96 hours, 0.22 μm	Askeland et al. (2020) [36]
Biochar	Activated and unactivated biochar	Sorbent amendment	0-20%	2 weeks	Industrially contaminated	Σ_{4}=1,125-3,671	$C_{6,8}$ PFCAs, $C_{6,8}$ PFSAs	EN 12457-2	Silvani et al. (2019) [37]
Composite	RemBind®, RemBind Plus®	Sorbent amendment	5-30%	2 months	AFFF contaminated	Σ_{12}=2,400-14,000	$C_{4,6,8}$ PFSAs, C_{4-10} PFCASs	24 h, 0.45 μm filter	Bräunig et al. (2021) [38]
Composite	Modified clay (MatCARE$^{\text{TM}}$)	Sorbent amendment	10%	1 week-1 year	AFFF impacted sandy, clay and sandy clay loams	Σ_{1}=9,262-74,379	PFOS	0.45 μm filter, 24 h	Das et al. (2013) [39]
Multiple	RemBind® PAC (FiltraSorb$^{\text{TM}}$ 400)	Sorbent amendment	5%	2 weeks	AFFF contaminated. loamy sand (1), sandy (1)	Σ_{21}=700-37,200	$C_{4-8,10}$ PFSAs, C_{4-13} PFCAs, PFOSA, 4:2, 6:2, 8:2, 10:2 FTS,	ASLP	Kabiri et al. (2021a,b) [40, 41]
Mulitple	CB + bentonite, CaCl, chitosan, hydrotalcite, PAC, pulverized zeolite, RemBind®	Stabilisation and solidification	10% CB 0.2% amend	4 months	Loamy sand (spiked)	Σ_{14}=6240-7310	C_{3-10} PFCAs, $C_{4,6,8}$ PFSA, 6:2, 8:2 FTS, PFOSA	0.1-2.0 mm particles, modified EN-12457-1	Sörengård et al. (2019a) [42]

Table 1 (continued)

Amendment category	Amendment type(s)	Technique	Application rate (w/w)	Ageing period	Soil properties	Initial soil conc. (μg kg^{-1})	PFAS tested	Leaching method/details	Author, year and reference
Mulitple	Activated carbon, compost soil, montmorillonite	Sorbent amendment	3%	Not stated	Sandy AFFF contaminated	Σ_{12}=6.4-2,510	C_{4-10} PFCAs, $C_{4,6,8}$ PFSAs, 6:2, 8:2 FTS	Modified EN 12457-2. 1:1 soil water, 8 days, 0.7 μm filter	Hale et al. (2017) [43]
Multiple	AC (coal-based), Biochar (mixed wood, paper-mill waste)	Sorbent amendment	4%	2 weeks (sorption)	Natively AFFF contaminated	Σ_{15}=780-3,600	PFOS, PFOA, PFHxS	47 μm filter	Kupryianchyk et al. (2016) [44]

AC activated carbon, *ASLP* Australian Standard Leaching Procedure, *CB* concrete binder, *DMOA* perfluoro-3,7-dimethyl octanoic acid, *FTOH* fluorotelomer alcohol, *FTS* fluorotelomer sulfonate, *GAC* granular activated carbon, *HPFHpA* 7H-dodecafluoro heptanoic acid, *HpFPipB* heptafluoropiperidin butane, *OC* organic carbon, *PAC* powdered activated carbon, *PFCAs* perfluoroalkyl carboxylic acids, *PFSAs* perfluoroalkyl sulfonic acids, *PFOSA* perfluorooctanesulfonamide, *TOC* total organic carbon

**Spiked with 5 μg L^{-1} per PFAS

A number of activated carbon products are available for PFAS sorption including powdered activated carbon (PAC), granular activated carbon (GAC) and colloidal/liquid activated carbon, such as PlumeStop®. Smaller particle size activated carbon has the advantage of higher surface area but is problematic to apply due to dust issues, however, colloidal/liquid activated carbon formulations have overcome these issues. Activated carbon amendments have been found to have the highest sorption capacity across a range of PFAS compared to other immobilisation amendments. In a PFAS sorption study using 44 adsorbents in water, Sörengård et al. [50] identified that activated carbon amendments (GAC, PAC, RemBind®) had the highest sorption capacity across most PFAS, with mean log K_d values of 3.5 for PFCAs and 4.2 for PFSAs, PFOSA and fluorotelomer sulfonates (FTSAs). Similarly, GAC was found to be superior at removal of PFAS compared to zeolite and sludge in another comparative study [51].

Biochar-Based Amendments

Biochar is used as an amendment due to its high cation exchange capacity (CEC), surface area and pore volume [52]. It is a carbonaceous material alternative to activated carbon, being produced from sustainably sourced materials [53] for carbon sequestration. As with activated carbon, biochar physico-chemical properties are influenced by carbon source material and pyrolysis temperature. Higher pyrolysis temperatures result in more organised carbon layers, with less functional groups, especially those containing O-H groups (a result of deoxygenation and dehydration) [52]. Higher temperatures also result in higher porosity, specific surface area, pH and ash content with a concomitant decrease in CEC and volatile matter [52]. Animal and solid-waste biochars tend to have lower surface area and volatile matter, but higher CEC than wood and crop-based biochars. The impact of these properties on PFAS sorption are complex, but generally result in biochars being less effective in sorption compared to their more homogenous activated carbon counterparts.

In an attempt to increase sorption capacity, research has been undertaken whereby surface functional groups are modified through biochar treatment. For example, biochar treated with $MgCl_2$ was found to have the highest mean log K_d for PFCAs (0.93) and PFSAs (0.87) of the non-activated carbon sorbents, while the untreated, MnO and $FeCl_3$ treated biochars were less effective at sorbing PFAS [50].

Clay-based Amendments

Of the non-carbon-based amendments, clays, such as zeolite, bentonite, hydrotalcite, montmorillonite and kaolinite have been utilised as a sorbent phase for PFAS immobilisation due to their large surface area and recognised potential to sorb organic contaminants (e.g. PAHs). Clay minerals are hydrophilic, chemically stable and have high cation exchange capacities [54]. The abovementioned clays differ in their constituent molecules, structures and interlayer spacing which drives sorptive capacity. For example, Zhang et al. [55] determined that kaolinite had a higher PFOS sorption capacity (77.6 μg g^{-1}) than montmorillonite (54.5 μg g^{-1}), however, PFOS was observed to sorb in the montmorillonite interlayer. A study of PFAS sorption on kaolinite in water [56] found that adsorption of ≤C_4 molecules was not thermodynamically favourable. Clay minerals may also be modified to increase their PFAS sorptive capacity [54]; some of these modified clays are discussed in the following section on composite amendments.

Composite and Other Amendments

To improve the efficacy of soil amendments for PFAS immobilisation, composite products have been developed that comprise a variety of sorptive phases in combination. The philosophy behind composite amendments is to utilise different components exhibiting different physicochemical properties to enable multiple sorptive mechanisms for PFAS retention. Composite amendments used for PFAS immobilisation include RemBind®, RemBind Plus® and MatCARE.

RemBind® is a composite amendment, consisting of aluminium hydroxide, kaolin, and activated carbon. It is proposed that the aluminium hydroxide component of RemBind® interacts via electrostatic forces with anionic PFAS, binding the functional group. The activated carbon and organic matter components of RemBind® bind to the PFAS ‘tail’ through hydrophobic and Van der Waals forces. This provides a twopronged reaction mechanism for binding PFAS anions. In addition, the kaolinite and organic matter component of RemBind® provides the ability to bind cationic compounds through electrostatic interactions, regardless of the length of the CF backbone. The activated carbon component also binds PFAS cations through hydrophobic interactions. Most long-chain PFAS (≥C_8) bind to activated carbon, however, smaller compounds (≤C_4) may not bind efficiently as there are limited contact points with the carbon surface and Van der Waals forces are weak. In RemBind®, short-chain compounds are bound through electrostatic interactions as detailed above. RemBind Plus® [38] contains a higher quality and amount of both activated carbon and aluminium oxyhydroxides.

MatCARE^{TM}, is another composite amendment that consists of clay (palygorskite-based, an Mg-Al phyllosillicate), which has been modified with an unsaturated fatty amine (oleylamine), and also contains quartz, kaolinite, dolomite, amorphous materials and traces of NaCl [39]. It was reported to have a PFOS adsorption capacity of 0.09 mmol g^{-1}, which was higher than the activated carbon product, Hydraffin (0.07 mmol g^{-1}) from the same study [39]; however, the sorptive performance for other PFAS and environmental implication related to the use of the oleylamine modification has not been reported.

Other amendments investigated for their PFAS sorptive capacities include polymers, carbon nanotubes and nanoparticles with ligands. In an aquifer solids sorption study, the cationic polymers polydiallyldimethyl ammonium chloride (polyDADMAC) and polyamine [57] were found to increase sorption of PFSAs and PFCAs by a factor ranging from 2.0 to 6.1. For example, K_D values for PFOS increased from 1.44 L kg^{-1} for the untreated soil, to 2.94L kg^{-1} and 8.75 L kg^{-1} for polyDADMAC and polyamine respectively. It was proposed that the increase in PFAS sorption was due to the increase in carbon content, but also due to enhanced exchange interactions between the cationic polymers and anionic PFAS [57]. Similarly, a study of two polyquaternium polymers, poly-(dimethylamine-co-epichlorohydrin) and poly-(diallyldimethylamonium) in water/soil suspensions, found that PFOS was bound, predominantly in soil-polymer-PFOS complexes, with respective affinities of 9.3×10^4 and 7.3×10^4 M^{-1} [58].

PFOS sorption onto Gold nanoparticles with different combinations of dual ligands has been demonstrated by Liu et al. [59], while the adsorption of PFOA onto multi-walled carbon nanotubes (MWCNT) and MWCNT with embedded Fe, Co, Al, Mg, Mn, Fe_2O_3, Cu and Zn has been demonstrated in several studies [60–62]. Carbon nanotubes are useful sorbents due to their large surface areas; however, the cost of MWCNT is generally prohibitive relative to GAC and PAC for large scale applications.

Amendment Additions and Binding Agents

The amount of amendment added to a contaminated soil will influence treatment costs, and may also influence whether the soil is able to be reused post-treatment. Application rates for immobilisation studies included in Table 1 range from 0.1% to 30%. Soil amendments may also be added to contaminated soil in conjunction with binding agents such as cement and lime. Binders are added to reduce the hydraulic conductivity and to increase or maintain unconfined compressive strength so that it is compatible with the intended site end use. The co-addition of amendments and binders provides a two-pronged approach for minimising PFAS leaching through adsorptive and encapsulation processes, as described in "Mechanisms of Immobilisation". Binding agents listed in Table 1 have typically been added at 10% w/w, and cured for a number of months.

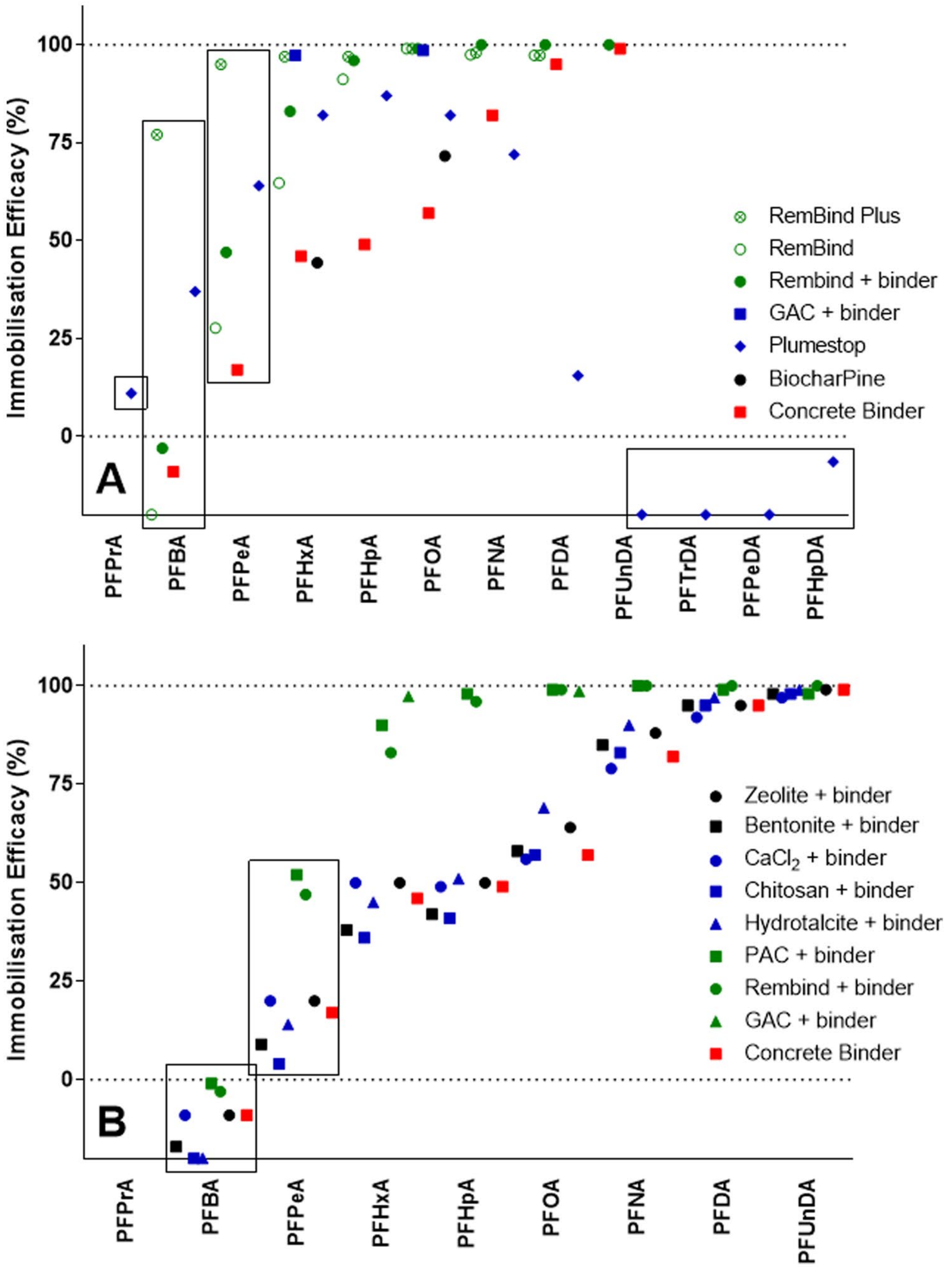

Fig. 2 Comparison of PFCA immobilisation efficacy when different amendment strategies were applied to contaminated soil. Data points may be cross-referenced with information in Table 1 to glean information on application rates, curing times and methods used for the assessment of PFAS leachability. PAC, Rembind® and RemBind Plus® [38], compost, montmorillonite [43], pine biochar [36], MatCARE^{TM} [39], PlumeStop® [34], GAC and CB [33], all other amendments with binder [42]. Points lower than -25 are shown at -25. Some results may be averages over several soil types, conditions and/or replicates, or the most realistic application rate. (**A**) Includes the activated carbon based amendments; (**B**) includes only studies involving a concrete binder

Remediation Effectiveness

A common strategy for determining immobilisation efficacy is the assessment of contaminant leaching, with a comparison between pre- and post-amendment values. Leaching assays are routinely undertaken to characterise the soil's mobile phase with data then utilised to estimate ground or surface water contamination potential. Leaching assays may also be utilised to classify waste for disposal and/or to demonstrate the effectiveness of soil amendment strategies. This may be achieved through the use of numerous leaching tests, including the Australian Standard Leaching Procedure (ASLP; AS4439-1997), Synthetic Precipitation Leaching Procedure (SPLP; USEPA method 1312), Toxicity Characteristic Leaching Procedure (TCLP; USEPA method 1311), TCLP variations (e.g. ASTM D3987-85), Multiple Extraction Procedure (MEP; USEPA method 1320) and Leaching Environmental Assessment Framework methods (LEAF; USEPA methods 1313-1316). The aforementioned leaching procedures vary in operational parameters including leaching fluid pH, buffering capacity, ionic strength, single versus multiple extractions, solid-to-solution ratio, contact time, agitation, maximum

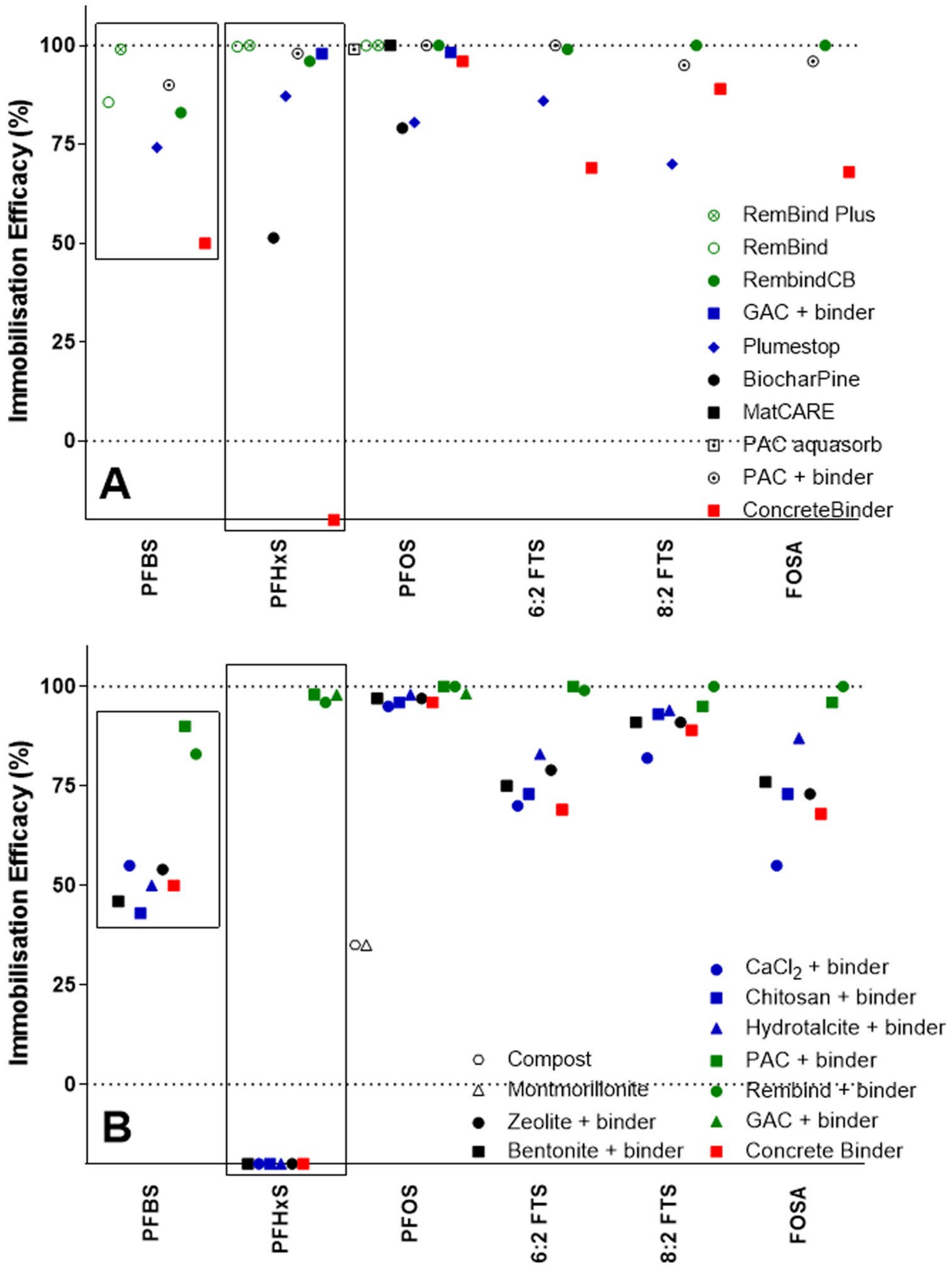

Fig. 3 Comparison of PFSA, FTS and FOSA immobilisation efficacy when different amendment strategies were applied to contaminated soil. (**A**) Includes the activated carbon based and composite amendments; (**B**) includes compost, montmorillonite and studies involving a concrete binder. Data point sources as described in Fig. 2 and Table 1

particle size, batch versus column tests; all of which will influence PFAS leachability outcomes. For example, differences in fluid pH (e.g. buffered TCLP at pH 4.93 versus unbuffered ASLP using deionised water) will influence leachability due to the impact of pH on sorption-desorption (as detailed in "Mechanisms of Immobilisation") while particle size differences (the maximum particle size for AS4439 is 2.4 mm versus 9.5 mm for TCLP) will also influence leaching outcomes due to disparities in reactive surface areas.

Notwithstanding the methodological differences between leaching assays, data from PFAS immobilisation studies (Table 1) are represented in Figs. 2 and 3 to provide an overview of immobilisation efficacy for different amendment strategies applied in both laboratory and field-based studies for PFAS varying in carbon chain length and functional group. Treatment efficacy was calculated, as in Sörengård et al. [34] by:

$$\Delta Re = \left(1 - \frac{c_{aq}^{t}}{c_{aq}^{u}}\right) \times 100 \qquad (1)$$

where c_{aq}^{t} is the PFAS concentration in the leachate of the treated soil and c_{aq}^{u} of the untreated soil leachate.

Values greater than zero indicate that the amendment strategy decreased PFAS leachability compared to unamended soil with increasing values up to a maximum of 100 indicating increasing treatment efficacy up to complete immobilisation. Negative values may also be calculated whereby the addition of the soil amendment results in enhanced PFAS leachability compared to unamended soil. Negative values may also result from analytical variability particularly when PFAS concentrations are being reported near the level of quantification. Conceivably precursor transformation may influence PFAS immobilisation efficacy; however, data illustrating this is lacking due to analytical

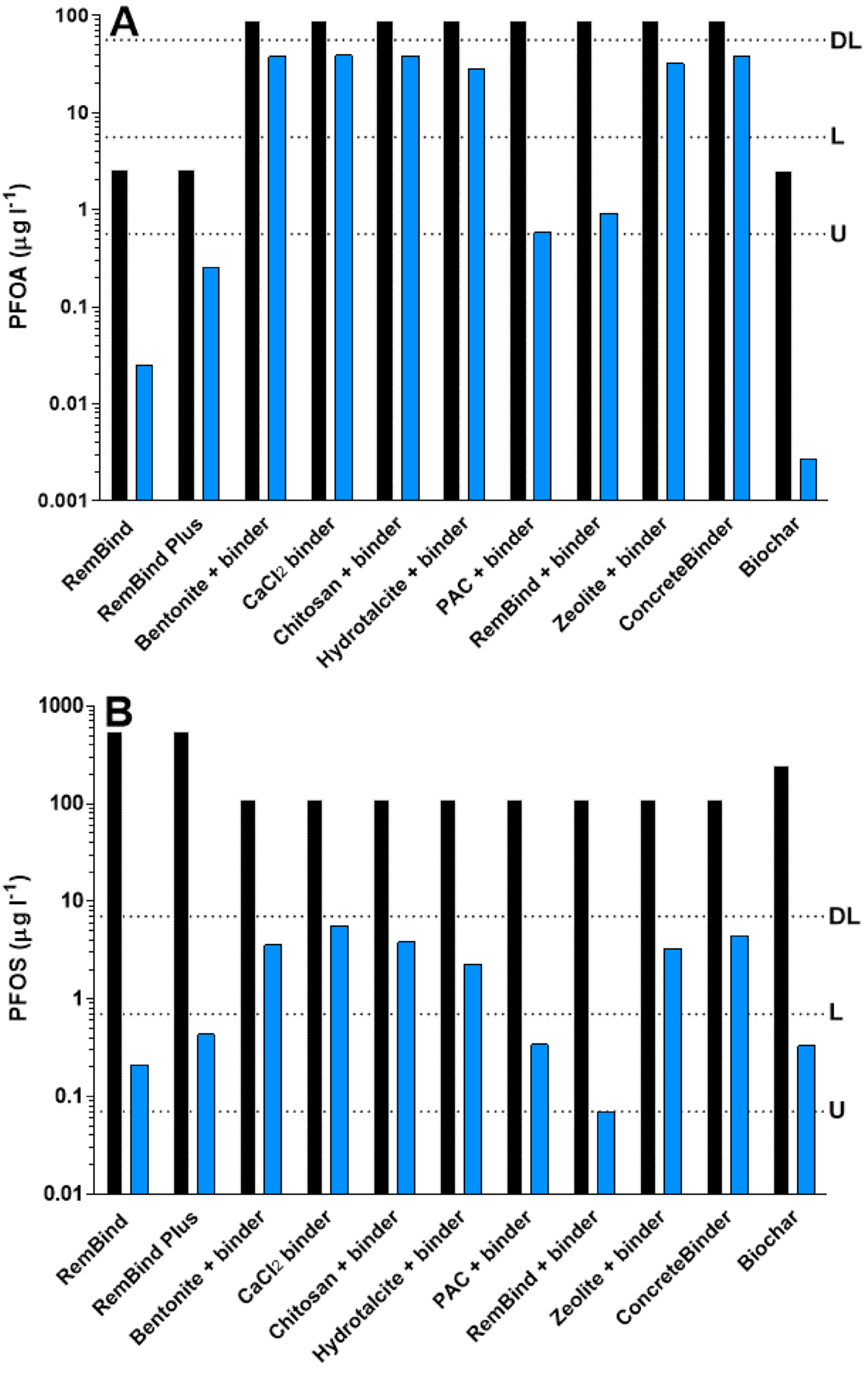

Fig. 4 Change in PFOA (**A**) and PFOS (**B**) leachability following treatment of soil with amendments (as detailed in Table 1). PFOA and PFOS leachability pre- (■) and post-amendment (🟦) is presented in the context of regulatory standards for unlined (**U**), lined (**L**) and double-lined (**DL**) landfills [63]. Amendments shown: RemBind [38], biochar [35], amendments with binder [42]

challenges associated with precursor identification and quantification. The plot of treatment efficacy, depicted in Figs. 2 and 3, provides a high-level approach for comparing data across amendment strategies. However, efficacy calculations do not reflect whether the extent of immobilisation was suitable against regulatory standards (i.e. landfill leaching criteria, water quality standards). As a result, Fig. 4a, b highlight PFOA and PFOS pre- and post-amendment leaching results in the context of regulatory standards.

As shown in Fig. 2, treatment efficacy trends were observed for PFCAs when different amendment strategies were utilised; PFAS immobilisation efficacy increased with increasing carbon chain length although some variability was observed for C_{10}-C_{14} compounds in the study of Sörengård et al. [34]. For $\leq C_6$ PFCAs, immobilisation efficacy for most treatments was <50% with the exception of three amendments incorporating activated carbon. Activated carbon amendments were highly effective at reducing leachability of $\geq C_8$ PFCAs in amended soil as exemplified by RemBind® and RemBind Plus® amendments [38]. At loadings of 25% and 15% w/w respectively, >97% reduction in leachability was observed for C_8-C_{10} PFCAs. For PlumeStop®, a decrease in immobilisation efficacy was observed for $>C_7$ PFCA possibly due to reduced hydrophobic interactions [34]. While only a limited number of studies have assessed the impact of biochar addition on PFCA leachability, Askeland et al. [36] observed increasing immobilisation efficacy with increasing carbon chain length. However, compared to activated carbon amendments, biochar appears to be less effective at reducing PFCA leachability, though further comparative studies with equivalent amendment loadings and leaching assessment methodologies would be beneficial. While the addition of bentonite, $CaCl_2$, chitosan, hydrotalcite and zeolite in conjunction with a concrete binder had little effect on treatment efficacy compared to the binder alone, the addition of activated carbon-based amendments with concrete binders enhanced immobilisation efficacy compared to binder alone, particularly for C_5-C_9 PFCAs. When immobilisation efficacy was compared between RemBind® amendments with and without the addition of concrete binders, C_5-C_7 PFCA treatment efficacy was enhanced by inclusion of the binding agent, however, this effect was not observed for longer chain PFCAs.

Immobilisation trends were less apparent for PFSAs compared to PFCAs (Fig. 3). For activated carbon-based amendments, higher immobilisation efficacy was observed for short-chain compounds (perfluorobutane sulfonic acid (PFBS) and PFHxS) compared to the equivalent carbon chain length carboxylic acids. Although some variability in PFSA immobilisation efficacy was observed for PlumeStop® [34], the remainder of the activated carbon-based amendments reduced PFSA leachability, irrespective of carbon chain length and degree of fluorination, by >80%. Limited data points are available for biochar amendments, however, similar trends were observed for PFSAs and PFCAs. Similarly, the addition of bentonite, $CaCl_2$, chitosan, hydrotalcite and zeolite in conjunction with a concrete binder had little effect on PFSA treatment efficacy compared to the binder alone [42].

Figure 4 shows PFOA and PFOS leachability pre- and post-amendment addition from selected studies, with reference to landfill regulatory guidelines [63]. In soil containing high PFOA leachability pre-treatment (~90 $\mu g\ L^{-1}$), irrespective of amendment type, leachability post-treatment was reduced to an acceptable level for soil disposal into a double-lined landfill, although for most treatments there was no advantage of adding an amendment (0.2% w/w) over the binder alone (10% w/w, Figure 4a). However, carbon-based amendments (PAC + binder, RemBind® + binder [42]) performed significantly better, reducing PFOA leachability to achieve the single-lined landfill criterion. For soil with low PFOA leachability pre-treatment (<3 $\mu g\ L^{-1}$), addition of RemBind® (25% w/w), RemBind Plus® (15% w/w) [38] and biochar (5% w/w) [35] reduced PFOA leachability to below the unlined landfill criterion. Although landfill acceptance criteria are lower for PFOS compared to PFOA, all treatments were able to reduce PFOS leachability to at least the double-lined landfill criterion (Fig. 4b). However, carbon-based amendments (RemBind® RemBind Plus® Biochar, PAC + binder, RemBind® + binder) achieved the single-lined landfill criterion at application rates ranging from 5-25% w/w.

Mechanisms of Immobilisation

A reduction in PFAS leachability occurs as a result of a number of sorption mechanisms. Sorption mechanisms are dependent on physico-chemical properties of PFAS and amendments and are influenced by the surrounding soil and environmental conditions. The following conditions will impact PFAS sorption:

1. C chain length and function group influence hydrophobic and electrostatic interactions respectively
2. Amendments may influence sorption through changes in soil pH (electrostatic interactions), by providing surfaces for hydrophobic interactions, and cation bridging.
3. PFAS adsorption studies performed on soils with varying characteristics (e.g. OC, clay, pH, anion exchange capacity (AEC) and protein) [64–70] have found that soil pH, organic carbon and clay content all play important roles in sorption through a combination of electrostatic and hydrophobic interactions [71].
4. Most PFAS have pKa<2 and thus are anions under most environmental circumstances. However, some PFAS (e.g. PFOSA, pKa 6.2-6.5), do not form anions under many circumstances [42, 72], which will impact sorption strategies based on direct electrostatic interactions.

PFAS have widely varying physico-chemical properties, and in particular, both the functional group and fluorinated C chain length affect sorption behaviour. PFAS may exist as uncharged, anionic, cationic, or zwitterionic molecules, although the most commonly observed compounds (PFOS and PFOA), both form anions. Charge differences affect sorption and desorption processes, and impacts the efficacy of soil amendments. The sulfonate functional group of PFSAs is considered to be a hard base, having higher sorption onto oxide surfaces than the soft base of the carboxylate group in PFCAs [73]. However, different amendments vary in their PFAS immobilisation efficacy due to the complex interplay of properties. Surface charge is a significant parameter, for example with anionic PFAS, sorption increases with increasingly positive surface charge [74].

Activated carbon and biochar have a number of different functional groups on the amendment surface, depending on the temperature of production, which will influence electrostatic interactions. More functional groups are present on the surface when production occurs at a lower temperature, therefore electrostatic interactions are likely to be higher; however, hydrophobic interactions may be the predominant PFAS sorption mechanism, as carbonaceous material produced at higher temperature have higher PFAS sorption capacities. pH influences will impact electrostatic interactions as an increase in pH will result in competition for sorption sites, between OH- groups and anionic PFAS [73].

PFAS octanol-water partitioning coefficients are influenced by C-chain length (e.g. PFHxA K_{OW}=3.12 versus PFOA K_{OW}=4.59 [75]), while log K_{OC} has been shown to increase by 0.6 and 0.83 with each additional fluorinated carbon for PFCAs and PFSAs respectively [67]. As such sorption efficacy of varying carbon chain length compounds to soil and soil amendments is influenced by K_{OW} through hydrophobic interactions [42, 66]. It has been reported that PFAS hemi-micelles and bilayers may form through self-aggregation on amendment surfaces [76] which is influenced by sorbent pore size, organic matter fraction, surface area and degree of carbonisation [36, 42].

In addition to electrostatic and hydrophobic interactions, PFAS may also form divalent cation bridges, with ions such as Ca^{2+} [77] and Mg^{2+}. Smaller PFAS are more likely to participate in divalent cation bridging, as observed by Campos Pereira et al. [67], potentially due to their shorter C-F tail, which makes them less hydrophobic, and subsequently less likely to sorb via hydrophobic interactions. Carbon nanotubes (CNT) with a variety of metal cations (Co, Mn, Mg and Al) on their outer and inner surface were found to enhance PFOA sorption by a factor of 6 compared to CNT containing inner surface Fe as a consequence of enhanced electrostatic interactions [60]. In addition, ligand exchange may occur between the carboxyl group of PFCAs and hydroxyl groups on metal oxides [76], forming inner sphere complexes via covalent metal-ligand bonds.

Compared to anionic PFAS, fewer sorption studies have been undertaken on cation, zwitterionic and neutral PFAS [42]. A study on cationic and zwitterionic PFAS sorption in soils [78], found that sorption of these compounds was non-linear, in contrast to anionic PFAS, however, electrostatic and hydrophobic interactions were the predominant mechanisms controlling PFAS sorption. Cationic PFAS sorption was correlated with soil organic matter content and was reversible. In contrast, sorption of zwitterionic

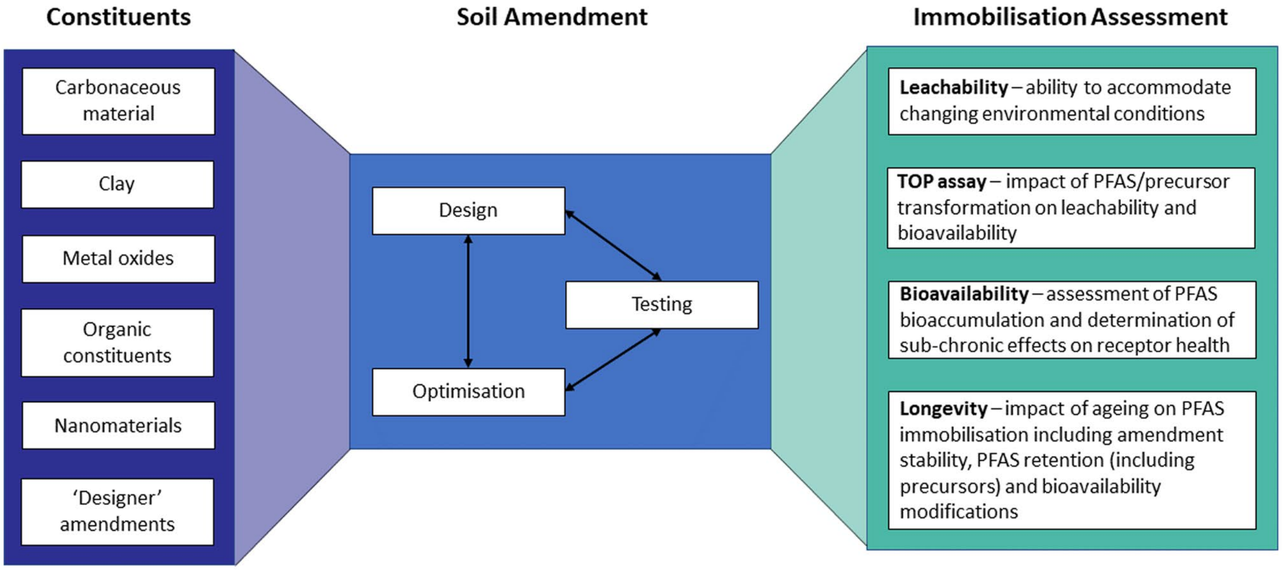

Fig. 5 Assessment framework for the design, optimisation and validation of PFAS immobilisation efficacy and longevity

PFAS was hysteretic rather than reversible and was hypothesised to be due to entrapment in porous structures present in inorganic soil components [78]. Differences in behaviour between cationic and zwitterionic PFAS highlight the necessity of further studies to assess the influence of soil amendments on PFAS fate and transport and immobilisation efficacy.

When binding agents are utilised, encapsulation also contributes to the decrease in PFAS leachability. Although the addition of cement binders may raise the pH to around 10 [42], thereby decreasing the impact of electrostatic interactions, the decrease in surface area and reduction in hydraulic conductivity of stabilised/solidified material results in reduced PFAS leachability.

Conclusions and Future Research Needs

As detailed in "Remediation Effectiveness", a common approach for assessing the efficacy of PFAS immobilisation is the use of leaching methodologies (e.g. SPLP, TCLP, ASLP). These approaches are simplistic, however, differences in operational parameters may restrict true comparison of immobilisation efficacy between treatments and studies. In addition, these approaches for the assessment of PFAS immobilisation utilise operationally defined procedures which may generate methodological artefacts, such as perceived enhanced contaminant mobility via colloidal generation [79–81]. Water-dispersible naturally occurring colloids are generated as a result of vigorous shaking during the batch extraction process or by leaching under elevated pH. Colloids exhibit high specific surface area and charge, which are effective sorbents for PFAS. Although an understanding of colloidal facilitated mobility of PFAS is limited, this transport mechanism has been shown to be significant for other contaminants of concern including metal(loid)s, mono-aromatic hydrocarbons, polycyclic aromatic hydrocarbons, polychlorinated biphenyls and organochlorine pesticides [82–87]. The use of the Leaching Environmental Assessment Framework (LEAF) [88] may overcome some the aforementioned limitations as the framework incorporates a number of standardized testing methods (including assays to assess pH dependency, liquid:solid dependency, percolation and mass transfer) and generic or application-specific release models. In addition, the LEAF overcomes single-point leaching tests that assess a specific environmental scenario and can accommodate defined particle sizes and monoliths (i.e. minimized colloidal generation) which are relevant for solidified materials.

Post immobilisation assessment should provide further consideration to the influence of aging and environmental factors on long-term immobilisation efficacy. Limited studies have assessed immobilisation stability over extended timeframes, however, PFAS immobilisation longevity was demonstrated in RemBind® amended soil following a 3-year aging period [38]. Although small increases in PFAS leachability (ASLP) were observed in aged samples when 2 month and 3-year leachability data was compared, PFAS leachability after 3-years was reduced by >99% compared to unamended soil. Additional studies like this are needed to elucidate the long-term effect of soil amendments on PFAS immobilisation. However, a limiting factor for ageing studies is the timeframe required to assess longevity. Conceivably, wetting and drying (at elevated temperature) cycles could be introduced into protocols as a means to accelerate ageing processes. This may provide indicative information regarding the impact of ageing on immobilisation stability although interpretation of data may be confounded by the difficulty in replicating natural ageing processes in the laboratory [89].

Transformation of PFAS and/or precursor compounds may potentially occur post-treatment via biotic and abiotic oxidation processes. Although little research has been undertaken to investigate the rate and extent of post-immobilisation transformation, the total oxidisable potential (TOP) assay [90] may provide an approach for quantifying transformation. However, as the name suggests, the assay quantifies the impact of oxidation on PFCA formation, although it fails to elucidate timeframes for such processes. Nevertheless, this information may provide a worst-case scenario for PFAS immobilisation stability. In the aforementioned 3-year RemBind ageing study [38], it was proposed that the minor increase in PFAS leachability in aged immobilised soil may have resulted from the generation of labile PFCAs through precursor compound transformation, however, it is conceivable that minor increases in PFAS leachability may have also resulted from immobilising agent breakdown. For some soil amendments with organic modifications (e.g. amine-modified clays), constituents may be utilised by the indigenous microflora as a carbon and energy source, thereby potentially impacting amendment integrity and PFAS sorption. The stability of these and other soil amendments has not been assessed in the context of PFAS sorption and retention; further assessment of this is required to ensure long-term stability of PFAS immobilised soil.

While leaching tests are utilised to understand the leaching profile of immobilised soil and/or determine the regulatory acceptability of stabilised material for disposal, data generated from these methods do not provide information regarding the influence of soil amendments on PFAS biological exposure and impact. If PFAS leachability can be reduced to below the criterion for unlined landfills, conceivably immobilised soil could remain on site; in such a case, other assessment methodologies (i.e. bioavailability assessment) would be required to ensure stabilised material was environmentally benign. Conceivably, leaching approaches may provide a conservative estimate of labile/exchangeable PFAS available to plants, however, other release mechanisms may be pertinent for ecological receptors facilitated by oxidative processes (of precursor compounds or immobilising agent) and/or pH changes (e.g. in gut environments). However, limited studies have been undertaken whereby changes in PFAS bioavailability (e.g. decrease in PFAS bioaccumulation) as a result of amendment application have been quantified. Bräunig et al. [38] reported that PFAS bioaccumulation in *E. fetida* decreased by 74-98% following immobilisation of a soil with 25% w/w RemBind®, although earthworm weight loss was reported, presumably due to decreased nutrient availability. These potential ecological impacts require further investigation in addition to the assessment of potential sub-chronic health effects including oxidative stress, DNA damage and reproductive issues.

Due to the variability in physicochemical properties of legacy and replacement PFAS (e.g. ADONA, GenX), a single agent may not be adequate/suitable to immobilise all compounds of concern in impacted soil. As such, soil amendments may need to be tailored to site-specific conditions. To drive future PFAS soil amendment research and development, a robust assessment framework is essential to elucidate not only immobilisation efficacy but longevity of immobilisation processes. The schematic framework detailed in Fig. 5 provides key assessment components required to test and validate the efficacy, longevity and biological impact of amendments for PFAS immobilisation in soil. This information is critical to provide a lines-of-evidence approach for the application of sustainable, cost-effective, immobilisation strategies to minimise the impact of PFAS on environmental health.

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Declarations

Conflict of Interest The authors declare that they have no conflict of interest.

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