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## Effect of physical, chemical, and biological aging on activated carbon and biochar properties and PFAS immobilisation in soils

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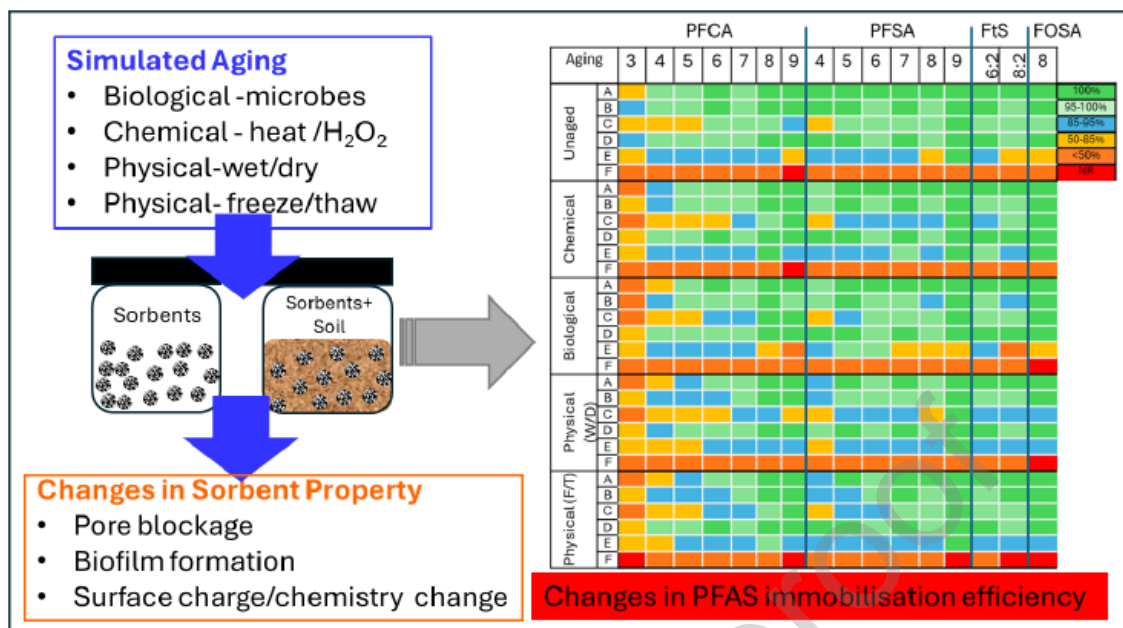
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### Abstract

An increasingly common technique for managing per- and polyfluoroalkyl substances (PFAS)-contaminated vadose soils is immobilisation using sorbents. However, uncertainty remains regarding the long-term stability of PFAS immobilisation. This study investigated the effects of different environmental aging processes, physical (wetting-drying cycles), chemical, and biological, on the performance of widely used sorbents for PFAS immobilisation in soil using accelerated aging to simulate long-term environmental conditions. Six sorbents were evaluated, including two granular activated carbons (GAC), two powdered activated carbons (PAC), colloidal activated carbon (CAC), and biochar. Simulated physical and biological aging caused the greatest reductions in sorption efficiency, likely due to pore blockage, as confirmed by material characterisation, including surface area measurement and scanning electron microscopic images. Sorbent type significantly influenced performance, with PAC being the least affected by aging and biochar the most susceptible. When soils and sorbents were aged together, sorbent responses differed between soil types, with sorption efficiency decreasing after chemical aging in sandy soil and after physical aging in high-clay soil compared with other aging treatments. Despite these deliberately harsh simulated conditions, activated carbon-based sorbents remained highly effective, maintaining over 85% PFAS immobilisation in most scenarios, including highly contaminated soils ( $\sum\text{PFAS} > 20 \text{ mg/kg}$ ). Overall, these findings demonstrate that although aging alters sorbent properties, activated carbon-based sorbents remain durable and effective for sustainable PFAS management under diverse environmental conditions.

### Graphical abstract



**Keywords:** PFAS, Immobilisation, Accelerated aging, Leaching

## Environmental Implications

Long-term stability is a critical uncertainty limiting the broader implementation of sorbent-based stabilisation strategies for PFAS-contaminated soils. This study provides, for the first time on evaluation of physical, chemical, and biological aging effects on multiple sorbents (GAC, PAC, CAC, and biochar), offering mechanistic insight into how environmental conditions influence long-term PFAS immobilisation. Across aging scenarios, performance declines were primarily governed by pore blockage and alterations in sorbent surface charge, which reduced accessible surface area and sorption affinity. These findings indicate that structural and interfacial transformations- in other word changes in the physical properties such as surface area and pore blockage are the dominant controls on aging-induced efficiency loss.

## 1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals developed in the 1930s and widely used since the 1950s in a range of consumer and industrial products (Benskin et al., 2010). One major application is in aqueous film-forming foam (AFFF) (Brusseu et al., 2020). Though now largely being phased out, AFFF has been extensively used at military bases, airports, and firefighting stations for training purposes, which has led to widespread soil contamination (Ross et al., 2018). Many PFAS associated with AFFF are

highly mobile in the environment and can leach through soil into groundwater or be transported via runoff to surface waters and nearby areas, especially during rainfall (Mahinroosta and Senevirathna, 2020). Because of the mobility of these PFAS, areas of contaminated soil may be spatially too large to practically remediate by removal of soil. Therefore, treating contaminated soil by immobilising PFAS can reduce risks by limiting the spread of PFAS in the environment.

Soil amendment with binding additives, commonly referred to as soil stabilisation or immobilisation, has been implemented at many contaminated sites. Various types of sorbents, such as granular activated carbon (GAC), powdered activated carbon (PAC), biochar, and mixtures of carbon-based materials with minerals (e.g., RemBind), have been applied for PFAS treatment in contaminated soils (Kabiri et al., 2021; McDonough et al., 2022; Navarro et al., 2023). These sorbents have shown varying degrees of effectiveness for PFAS immobilisation, with PAC generally outperforming GAC, and both performing better than biochar in stabilising PFAS in soil (Kabiri et al., 2023; Söregård et al., 2019). The physical and chemical properties of the sorbents, including surface area, particle size, surface charge, and elemental composition, have been shown to influence their effectiveness in removing PFAS from water or stabilising PFAS in soil (Cheng et al., 2010; Kabiri et al., 2023; Park et al., 2020). For instance, in a study comparing the performance of 28 carbon-based sorbents, surface area measured by methylene blue adsorption was a significant indicator of long-chain PFAS stabilisation, while surface charge correlated more strongly with the stabilisation of short-chain PFAS (Kabiri et al., 2023). However, since soil stabilisation does not remove PFAS but rather immobilises it, demonstrating the long-term effectiveness of this approach is crucial.

Several studies have examined the durability and longevity of various amendments to immobilise or stabilise PFAS in soils at both laboratory (Kabiri et al., 2021; Navarro et al., 2023) and field scales (McDonough et al., 2022). To evaluate sorbent performance, different standard batch leaching tests have been used to simulate worst-case scenarios, including the equivalent of 100 years of rainfall (Kabiri et al., 2021; Kabiri and McLaughlin, 2021). While these standard leachability tests provide useful insights, these methods do not account for potential changes in the sorbent's physico-chemical properties that could happen over time with aging. It has been shown that some sorbents work well in the short term, but start to lose effectiveness in the longer term, potentially because of aging (Navarro et al., 2023). They assessed the leachability of PFAS in soils treated with different carbon-based sorbents, and a biochar was initially effective but did not effectively reduce PFAS leachability from soils 4 years post-treatment (Navarro et al., 2023). However, conducting long-term studies on environmentally relevant timescales is not possible given that urgent remediation decisions are required. This underscores the value of adopting alternative strategies that evaluate treatment longevity and enable meaningful comparisons between sorbents.

When sorbents are in soil, their physical and chemical properties may change due to changes in soil conditions and microbial activity (Hale et al., 2017). This may affect the sorbents' properties in the long term and change their efficiency on PFAS immobilisation in soil. For example, some alteration in the soil hydrology due to changes in the water table level,

because of the rainfall, can change the physical properties of the sorbents (Spokas et al., 2014). Moreover, the abiotic and biotic processes can constantly generate different reactive oxygen species, such as superoxide ( $O_2^{\bullet-}$ ), hydrogen peroxide ( $H_2O_2$ ), and hydroxyl radical ( $\bullet OH$ ), which can interact with sorbents in the soil and change their properties (Yu and Kuzyakov, 2021). Exposure to temperature decline, including freezing conditions in some regions, can further alter sorbent properties; for example, freezing has been shown to change the sorption behaviour of biochar towards lead (Pb) (Tan, L. et al., 2020). Soil microorganisms can also affect the mineralisation of sorbents such as biochar (Liu and Chen, 2022). Qian et al. (2015) observed changes in the biochar C content and mineralisation after incubation of biochar with the microorganisms extracted from the soil. However, most studies to date have focused primarily on how aging alters the physical and chemical properties of sorbents, with limited investigation of their performance in removing contaminants (Hale et al., 2017; Jing et al., 2018; Nie et al., 2019; Tan, Lianshuai et al., 2020). A recent study by Jiang et al. (2025) reported that aging of colloidal activated carbon (CAC) reduced its surface area, decreased surface charge, and altered its anion-exchange capacity, collectively diminishing its PFAS sorption performance in water. Therefore, simulating these aging processes on sorbents or immobilised soil is essential for understanding the long-term stability, reliability, and performance of sorbents used for PFAS immobilisation in soil (Liu and Chen, 2022).

To date, aging processes have not been systematically applied to commonly used sorbents from the perspective of PFAS immobilisation in soils, nor has their post-aging sorption performance been adequately evaluated. While field-based studies using naturally aged materials offer realistic insights into long-term transformations, they are inherently time-intensive and impractical for assessing changes that may occur over decadal to centennial timescales, particularly given the extreme persistence of perfluoroalkyl acids. Therefore, in this study, we aimed to simulate these environmental conditions at laboratory scale and evaluate how different physical, chemical, and biological stressors (accelerated aging) influence sorbent performance during PFAS immobilisation in soil. To achieve this, we examined several commonly used sorbents for PFAS immobilisation, including PAC, GAC, CAC, and biochar. These materials were exposed to a range of environmentally relevant stress conditions both on the sorbents alone and when mixed with soil, in order to understand how aging affects their ability to immobilise PFAS. To simulate long-term, environmentally relevant harsh conditions, accelerated aging treatments were applied. The study compared the performance of aged and unaged sorbents and assessed how changes in sorbent properties influence PFAS retention across different soil types and contamination levels. The novelty of this work lies in (i) the comparative assessment of physical, chemical, and biological aging processes, which has not been comprehensively addressed in PFAS sorbent research; (ii) the direct comparison of different activated carbon forms and biochar under consistent experimental conditions, both in the presence and absence of soil; and (iii) the explicit focus on PFAS immobilisation in soil rather than aqueous-only adsorption. Collectively, these findings provide critical insights into the long-term stability, durability, and practical effectiveness of sorbents under conditions representative of realistic environmental stressors.

## 2. Materials and Methods

### 2.1. Contaminated soil characterisation

Soils were obtained from two Royal Australian Air Force bases. All soils were air-dried and sieved through 2 mm mesh. PFAS concentrations in the undiluted soils A and B were high (~20 mg/kg PFOS). To investigate the efficiency or changes of PFAS immobilisation by aged sorbents at highly contaminated ( $\sum\text{PFAS} > 20 \text{ mg/kg}$ ) and lower contaminated soil ( $\sum\text{PFAS} \sim 2\text{-}4 \text{ mg/kg}$ ), Soils A and B were diluted with a clean soil of similar texture and organic carbon content that contained no detectable PFAS. Soil properties and PFAS concentrations are in Tables S1-S2, SI. These concentrations were selected to enable a consistent comparison of the effects of different aging treatments on PFAS immobilisation. In the case of highly contaminated soil, immobilisation might not be an option, depending on the regulations in different countries. However, it enables us to properly monitor PFAS immobilisation for different PFAS compounds, as in diluted soil the concentration of some PFAS is very low. On the other hand, low-contaminated soil may be more representative of the type of soil where immobilisation could be used as an option.

### 2.2. Characterisation of unaged and aged sorbents

Six different sorbents were tested in this study; these are commonly used for PFAS immobilisation in soils, and their characterisation is summarised in Table 1. The sorbents differed not only in type but also in origin and physicochemical properties. Brunauer–Emmett–Teller surface area (BET-SA), average pore diameter, and pore volume were measured by nitrogen adsorption using a Micromeritics 3Flex Adsorption Analyser. Before conducting nitrogen adsorption isotherm measurements at  $-196 \text{ }^\circ\text{C}$  (77 K), all samples were activated at  $200 \text{ }^\circ\text{C}$  for 4 hours. Surface area was determined using BET analysis and Micromeritics' Density Functional Theory (DFT). The values obtained from BET analysis are presented in Table 1. Additionally, the surface area (MB-SA) of the sorbents was measured using the methylene blue (MB) adsorption method, following the procedure described in our previous study, and due to the correlation of PFAS immobilisation and MB-SA surface area (Kabiri et al., 2023). Briefly, 100 mg of sorbent was mixed with 50 mL of MB solution (300 mg/L) and shaken for 48 hours to achieve adsorption–desorption equilibrium. After centrifugation (30 min at 3042 RCF) to remove the sorbent, the MB concentration in the supernatant was measured using UV–Vis spectroscopy at 665 nm and compared to a calibration curve prepared from the initial MB solution. The surface area was calculated using the following equation:

$$SA = \frac{N_A \cdot A_{MB}}{M_{MB}} \times \frac{(C_i - C_e) \times V}{m_s} \quad (\text{equation 1})$$

where,  $N_A$  is Avogadro's number ( $6.02 \times 10^{23} \text{ mol}^{-1}$ ),  $A_{MB}$  is the surface area occupied by a single MB molecule (assumed to be  $1.35 \text{ nm}^2$ ),  $C_i$  and  $C_e$  are the initial and equilibrium concentrations of MB, respectively,  $V$  is the volume of the MB solution,  $M_{MB}$  is the molar mass of MB, and  $m_s$  is the mass of the sorbent.

The particle sizes of PAC and GAC were provided by the supplier. The biochar was ground to a particle size similar to GAC (500  $\mu\text{m}$  to 2 mm). The zeta potential of PAC, GAC, and biochar suspensions was measured across a range of pH values using a Malvern Zetasizer. A 0.1 M  $\text{CaCl}_2$  solution was used as the background electrolyte for all sorbents, to create a similar condition to the leaching experiments. The point of zero charge ( $\text{pH}_{\text{pzc}}$ ) was determined by plotting pH values against the corresponding zeta potential measurements. The  $\text{pH}_{\text{pzc}}$  of GAC was determined following the method described by Park et al. (2020). In brief, 100 mg of GAC was added to 20 mL of 0.1 M  $\text{CaCl}_2$  solution, with pH values adjusted between 3 and 10 using 0.5 M NaOH or HCl. The suspensions were shaken at 100 rpm for 48 hours at room temperature using a rotary shaker. The pH was measured before and after shaking. The  $\text{pH}_{\text{pzc}}$  was identified as the point at which no significant change in pH occurred over the 48-hour period. All measurements were conducted in triplicate.

Elemental composition of the sorbents was analysed using a Vario MACRO cube elemental analyser for carbon (C), hydrogen (H), and nitrogen (N), and a Rapid OXY cube for oxygen (O). The accuracy of CHNO analyses exceeded 95%, based on comparisons with certified reference materials. The morphology of selected sorbents before and after aging was characterised via SEM analysis (Model Quanta 450, FEI, USA) followed by EDS (Energy Dispersive Spectroscopy) analysis for detection of elemental composition. SEM was mostly performed in GAC sorbents as it was easier to distinguish some morphological changes and to separate them from soil, while separation of PAC and CAC was impossible from soil for the soil-sorbent aging studies. Furthermore, SEM imaging and morphological analysis were performed on several randomly selected granules. Any results included are based on observations from multiple granules, but ultimately, only the image of a single representative sample is presented.

Table 1. Characterisation of sorbents (A-F) used in this study.

Sorbent ID/label	Unit	A	B	C	D	E	F
Type		PAC	GAC	CAC	PAC	GAC	Biochar
Origin	-	Coal	Coal	-	Coconut	Peach	Almond
BET surface area	$\text{m}^2/\text{g}$	815	1158	1067	1134	561	6.0
MB surface area	$\text{m}^2/\text{g}$	588.1	408.6	262.5	716.2	177.0	70.0
PZC	-	+ all pH	9.0	- all pH	+ all pH	8.0	8.0
N	%	0.70	0.58	0.75	0.63	0.30	0.65
S	%	0.40	0.30	1.2	NA	NA	NA
O	%	5.2	4.23	22.3	5.9	4.2	31.4

## 2.3. Accelerated aging

### 2.3.1. Aging of sorbents

Sorbents (A-E) were subjected to different aging procedures following some of the published works and were used as received. Physical aging was undertaken by subjecting the samples to freeze/thaw ( $-20\text{ }^\circ\text{C}$  for 16 h and  $20\text{ }^\circ\text{C}$  for 8 h) or wetting and drying cycles (wetting to full saturation for 8 h and drying at  $70\text{ }^\circ\text{C}$  for 16 h) for 21 cycles for both methods. (Liu and

Chen, 2022) Chemical aging involved subjecting the samples to 15% H<sub>2</sub>O<sub>2</sub> solution at 80 °C for 6 h followed by drying at 70 °C (Liu and Chen, 2022).

Biological aging involved exposing the materials to a microbial inoculum, a nutrient solution (comprising 6.4 g L<sup>-1</sup> glucose, 40.6 g L<sup>-1</sup> NH<sub>4</sub>Cl, 4.7 g L<sup>-1</sup> KH<sub>2</sub>PO<sub>4</sub>, 10.0 mg L<sup>-1</sup> peptone, 24.0 mg L<sup>-1</sup> CaCl<sub>2</sub>, 4.0 mg L<sup>-1</sup> MnSO<sub>4</sub>, 4.00 mg L<sup>-1</sup> ZnCl<sub>2</sub>, 4.0 mg L<sup>-1</sup> CuSO<sub>4</sub>, and 16.0 mg L<sup>-1</sup> MgCl<sub>2</sub>), and a glucose supplement (40 µg mg<sup>-1</sup> BC). The inoculum was prepared by mixing the soil with Millipore water, rolling end-over-end at 11 rpm for 2 hours, and filtering the mixture through a 2.7 µm filter. The nutrient solution was added to prevent any environmental limitations, and glucose served as a readily degradable carbon source to support microbial activity and potentially enhance sorbent degradation.

To extract the inoculum, we mixed the field-moist soil samples with Millipore water in sterile 50 mL centrifuge tubes and vortexed the tubes for 2 hours at high speed. After vortexing, the tubes were allowed to stand for 5 minutes, and the soil suspensions were filtered through sterile 2.7 µm Whatman membrane filters into sterile centrifuge tubes. For the biological ageing process, a nutrient solution was prepared from autoclave-sterilised basal salt solution (500 mL/L final sorbent nutrient media), modified from Stevenson et al. (2004), filter-sterilised vitamin B12 solution (200 µL/L final sorbent nutrient media), filter-sterilised vitamin mixture (200 µL/L final sorbent nutrient media) and a filter-sterilised trace elements solution (1 mL/L final sorbent nutrient media). We combined 5 g of sorbent and glucose supplement (40 µg/mg sorbent carbon) with 250 mL ultrapure water and autoclave-sterilised the mixture. After autoclaving, the sorbent mixture was transferred to a quart-sized (946.35 mL) mason jar and combined with 250 mL of the nutrient solution. The pH of the basal salt solution, which is a part of the nutrient solution, was adjusted to 7 to obtain pH-neutral final biochar nutrient media (Zeba N et al., 2022). The aged sorbents were characterised after finishing the procedures.

### 2.3.2. Aging of sorbent-treated soils

Fifty grams of Soil A and B, and undiluted counterparts Soil A and B, were treated with either unaged sorbent (1% w/w), incubated for 1 month, then subjected to the aging procedures described in section 2.3.1. The treated soils were kept in a sealed jars while incubated for a month. Some sorbent material was also collected from the aged soils and characterised later. This was mostly performed on GAC, as the separation of PAC from soil was impossible.

### 2.4. Leaching assessment

Unaged or aged sorbents (chemical, biological, physical-wet/dry (W/D) and physical-freeze/thaw (F/T) sorbents at 1% w/w application rate were mixed with diluted or undiluted soil wetted to the 60% field water holding capacity and incubated for a month. One-month post-treatment, PFAS leachability was assessed. In a separate set of experiments, soils (both diluted and undiluted) treated with unaged sorbents were subjected to the same physical, chemical, and biological aging protocols at 1-month post-treatment. Leachability testing was

conducted again following the aging process to evaluate the long-term performance of the treatments under accelerated stress conditions.

ASLP (Australian Standard Leaching Procedure) simulates worst-case leaching conditions. (Kabiri et al., 2022) Briefly, samples equivalent to 2 g dry weight (untreated or treated) were weighed into a 50 mL PP tube, mixed with 40 mL milli-Q water to achieve a liquid to soil ratio (L/S) of 20 L/kg, then shaken end-over-end at 30 rpm. After  $18 \pm 2$  hours of shaking, samples were centrifuged @ 2500 g for 45 minutes to collect leachates for PFAS analysis (Kabiri et al., 2022). Leachates were analysed for PFAS, following the methods used previously (Cai et al., 2022).

## 2.5. PFAS analysis

Soils and leachates from this study were analysed for 28 target PFAS by liquid chromatography tandem mass spectrometry (LC-MS/MS). Concentrations of individual PFAS in the soils, as well as ASLP leachate samples, were analysed by the Australian Laboratory Services (soil samples) and in-house at CSIRO (leachates). Quantitation was based on recoveries of the isotopically labelled standards used as internal standards. PFAS analysis is comprehensively described in section S3 of the supporting information and Table S3, SI.

## 2.6. Calculations and statistical analysis

To evaluate and compare the relative performance of the different sorbents, for each treated soil, the efficiency of stabilisation was reported as a percentage reduction (%).

The percentage reduction was calculated relative to the concentration of PFAS leached from the untreated soil based on current analysis.

$$\% \text{ reduction} = \frac{C_{w, \text{untreated soil}} - C_{w, \text{treated soil}}}{C_{w, \text{untreated soil}}} * 100 \quad (\text{equation 2})$$

Where  $C_{w, \text{untreated soil}}$  is the concentration of an individual PFAS ( $\mu\text{g/L}$ ) measured in the leachate of the untreated soil, and  $C_{w, \text{treated soil}}$  is the concentration of an individual PFAS ( $\mu\text{g/L}$ ) measured in the leachate of the treated soil.

To assess the general behaviour of PFAS in untreated soils, PFAS leached from the samples were reported as a concentration ( $\mu\text{g/L}$ ) and as a percentage (%) leached.

The percentage of leached PFAS was calculated as follows:

$$\% \text{ of leached PFAS} = \frac{C_w}{C_{\text{total}}} * 100 \quad (\text{equation 3})$$

where  $C_{\text{total}}$  is the total concentration of an individual PFAS in the aged untreated soil ( $\text{mg/kg}$ ) prior to leaching, and  $C_w$  is the concentration of an individual PFAS measured in the leachates calculated according to:

$$C_w = \frac{C \cdot V}{m} \quad (\text{equation 4})$$

where  $C$  ( $\mu\text{g/L}$ ) is the concentration of an individual PFAS in the leachate,  $V$  (L) is the volume of leachate (varied depending on leaching procedure), and  $m$  (g) is the weight of untreated soil used in each leaching experiment. All values are reported as an average of 3 replicates  $\pm$  standard deviation, unless specified otherwise.

Statistical analyses were performed using IBM SPSS to evaluate differences in PFAS immobilisation among sorbents and aging conditions. One-way analysis of variance (ANOVA) was conducted to determine whether there were significant differences in PFAS leachability for specific compounds under different aging treatments compared with unaged conditions or comparing different aging treatments together for a specific sorbent. Additional analyses were performed within each aging group to identify differences in sorbent efficiency under a given aging condition when required. Post hoc tests were also conducted to determine pairwise differences where significant effects were observed.

### 3. Results and Discussion

#### 3.1. Sorbents' physical and chemical characteristics before and after aging

Physical aging, particularly wet/dry cycling, reduced the methylene blue surface area (MB-SA) of the sorbents between 8-32% (Figure 1a), while biological and chemical aging had a more mixed effect. The surface area of colloidal activated carbon (CAC) increased under all types of aging (3 to 19%), whereas biochar showed a consistent decrease in surface area (7 to 60%) following all aging treatments. For GAC and PAC, both forms of physical aging led to a reduction in surface area. In contrast, chemical and biological aging had mixed effects, with non-coal-based GAC and PAC showing an increase in surface area (Figure 1b). The increase in the surface area of the sorbents after some types of aging could be related to the exposure of pores that were initially covered or filled, and the creation of new pores (Liu et al., 2019) (Tan, L. et al., 2020). Alternatively, the increase of SA could result from the breakdown of granular particles, leading to a gradual reduction of particle size, which has been observed in some studies (Oleszczuk et al., 2016) (Ma et al., 2019). The opening of some pores, especially due to chemical aging, was observed in the SEM images of some sorbents, e.g., sorbents B, E (Figure S1, SI) and biochar. For some sorbents, chemical aging may cause an increase in pore opening and a decrease in microporosity, resulting in a reduction of surface area, as observed in some studies with biochar (Liu and Chen, 2022). Jiang et al. (2025) also observed the creation of more holes and some cracks after chemical aging in CAC. They also observed an increase in the BET SA after chemical aging and no changes in the BET SA after Phys (W/D) of CAC. We had the same results with  $\sim$ 15% increase in the SA of CAC after chemical aging (Figure 1b).

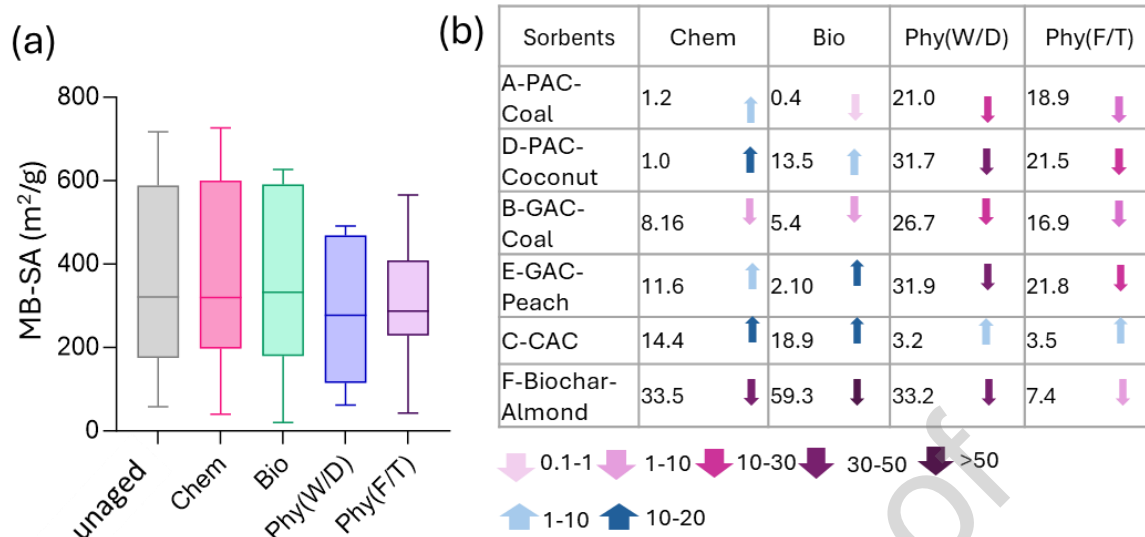


Figure 1. (a) Boxplots showing the surface area of sorbents measured using the methylene blue method for both unaged and aged samples, and (b) Changes in the surface area of individual sorbents subjected to different aging methods compared to the unaged sorbent. Blue arrows indicate the percentage increase in surface area, while pink arrows denote the percentage decrease in surface area. The colour gradient of the arrows represents the extent of increase or reduction in surface area, with the range indicated under the Figure b. The numbers shown are the average of three replicates ( $n=3$ ) for each sorbent.

The decrease in the SA measured by methylene blue after some of the aging was also observed, which could be related to the pore blockage resulting from reprecipitation of inorganic minerals, biofilm formation during the aging process, or formation of the oxygen-containing functional group at the pore entrance (Chang et al., 2018). Evidence of biofilm formation and pore blockage due to different aging processes, specifically biologically and physical aging (W/D) was also observed by SEM images of the sorbents (Figure 2, S1 and S2, SI). The EDS analysis of the surface of the aged sorbents after biological and physical aging (W/D) confirmed the biofilm formation by detecting the typical biofilm composition with the presence of Mg, Cl, Al, Si, Na, P, and Ca compounds contained within the activated carbon (Figure 2 and S3, SI) (Remoundaki et al., 2008) (Glamoclija et al., 2012) (Hu et al., 2013). The biofilm is mostly detected in the biologically and physically (W/D) aged samples, with more pore blockage observed in their SEM images. High-magnification SEM images of the aged GAC (both biologically and physically aged) show the presence of various round or filamentous morphologies (in GAC E), which are associated with different microbial species and have been reported in other studies (Glamoclija et al., 2012). Meanwhile, the EDS analysis of the chemically aged or unaged samples only showed the presence of C and O (Figure 2). For GAC B, the surface was covered with iron oxide particles, which appeared after biological and physical aging, while iron nano/micro particles were undetectable in the unaged GAC. These iron particles blocked some of the pores. The iron-based materials were used during the processing of these coal-based GAC (provided by the producer), which leached out during the long-term biological or wet/dry cycles. No significant or observable changes were noted in the surface morphology or EDS analysis of both unaged and aged

PAC, which implies a slight potential decrease in PFAS immobilisation on aged PAC compared to GAC.

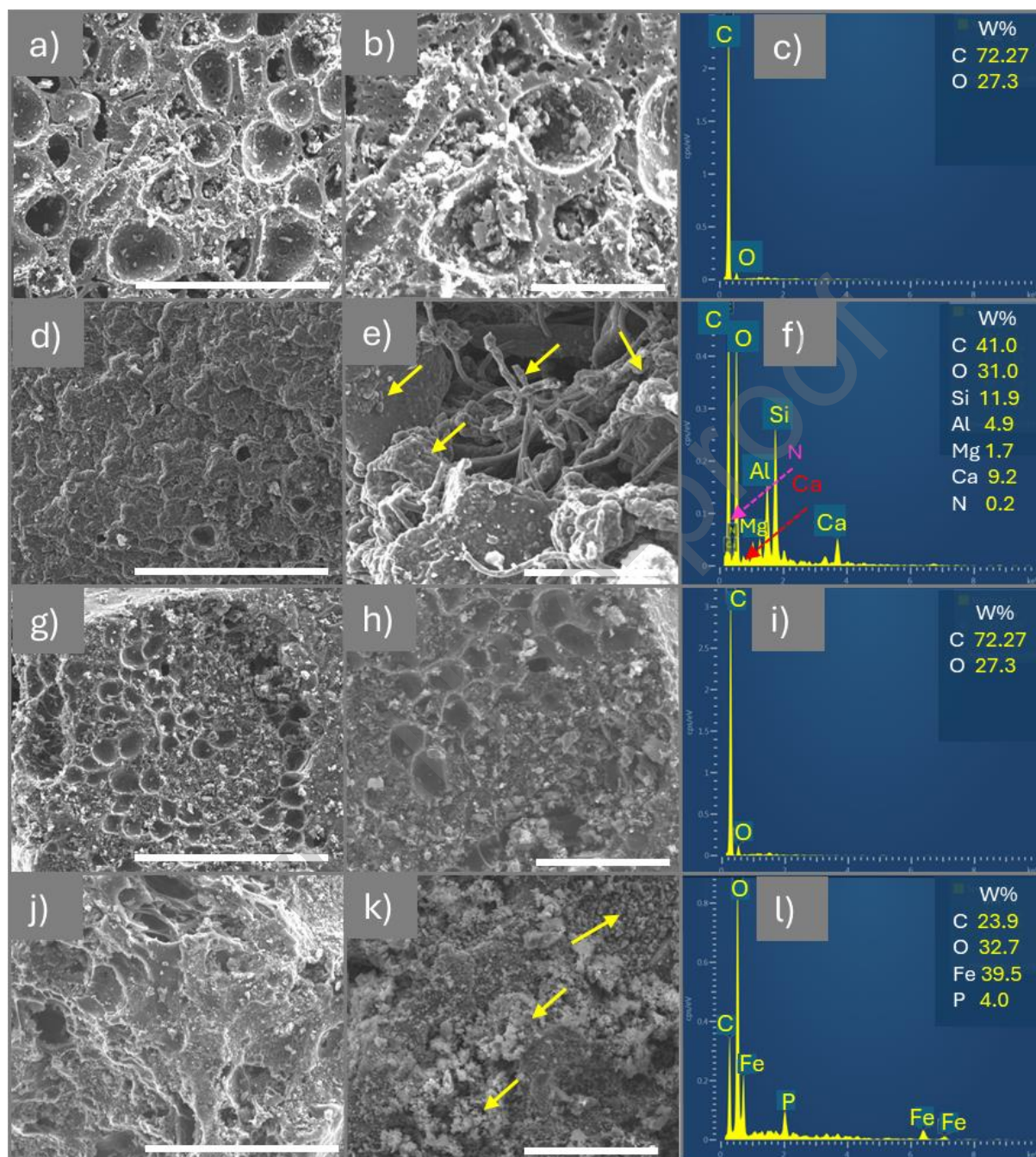


Figure 2. SEM images and EDS analysis of a-c) unaged GAC E, d-f) biologically aged GAC E, g-i) unaged GAC B and j-l) biologically aged GAC B at different magnifications, showing the pore blockage. The scale bar for the images on the left side is 100  $\mu\text{m}$  and for the middle images is 50  $\mu\text{m}$ . Arrows show the area where EDS analysis was performed in aged sorbents, but the results for one area are reported.

In average, the physical aging (both types) led to a decrease in carbon content of the sorbents, while chemical and biological aging perhaps led to an increase in the carbon content of the sorbents (Figure 3 and Table S4, SI). However, some of these changes were sorbent specific

and details of changes for each sorbet is provided in (Table S4, SI). These changes were more pronounced for biochar than for activated carbon. Similar trends have been reported in other studies where aging of biochar resulted in either carbon loss or increased carbon content (Liu and Chen, 2022). For biochar, carbon loss could be due to the leaching of dissolved organic carbon (DOC), (Mukherjee and Zimmerman, 2013) and mineralisation of organic carbon, which can occur mostly when the aging process occurs in the presence of soil. (Zimmerman, 2010)

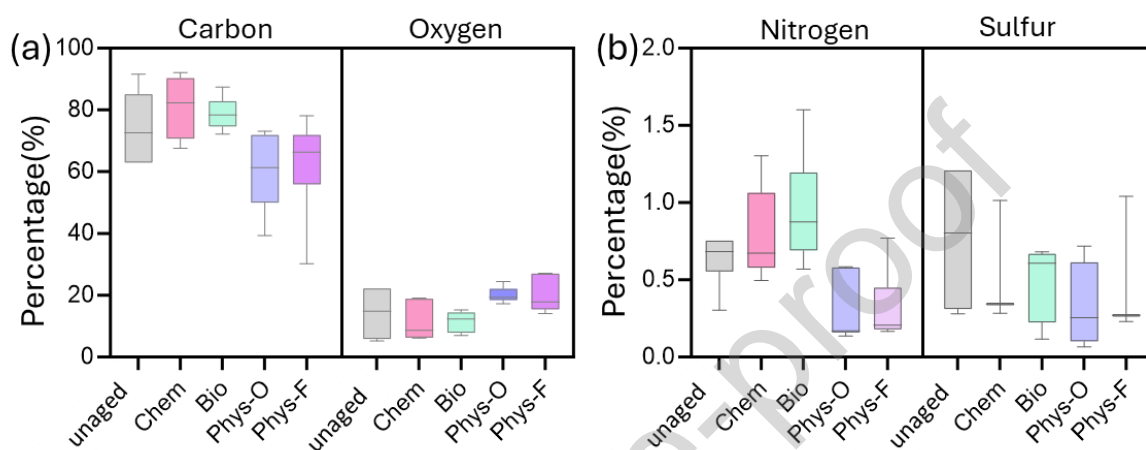


Figure 3. Boxplots showing (a) carbon and oxygen, and (b) nitrogen and sulfur contents of sorbents for the unaged and aged sorbents.

The O/C ratio is increased for most of the aged samples compared to unaged samples (Table S4, SI). This observation is consistent with previous studies reporting an increase in the O/C ratio following natural or artificial ageing of carbon-based materials such as biochar, through both abiotic and biotic processes (Cao et al., 2019; Zeba et al., 2021; Zimmerman, 2010). The increase in O content in aged sorbents is likely due to the formation of oxygen-containing functional groups, resulting from abiotic oxidation (in chemically and physically aged), (Cheng et al., 2006; Zeba et al., 2021) as well as microbially mediated oxidation (in biologically aged) (Quan et al., 2020). In some cases, colonised microorganisms (biofilm) were observed in aged sorbents, as evident from SEM images and EDS analysis (Figure 2 and S1-S3, SI), which may further influence the elemental composition of the sorbents (Gibert et al., 2013). Biofilm formation can increase the nitrogen content on activated carbon, as shown by others (Ramos et al., 2023). Therefore, the increase in N content of the sorbents (Figure 3) after biological aging could be another piece of evidence of biofilm formation on sorbents, which may consequently block the pores or change the surface chemistry of the sorbents and affect their PFAS removal efficiency. The N content was not significant in the EDS analysis, as this analysis was localised and the peak for N may have been integrated with C peaks, which had high content.

The changes in the functional groups of the sorbents after aging may influence their hydrophobicity and hydrophilicity. Several studies have shown that the oxygen-containing groups or heteroatoms such as S and N, (Ochedi et al., 2020) can affect the hydrophobicity/hydrophilicity of carbon-based sorbents, with the degree of hydrophilicity

being influenced specifically by the amount of polar O-groups (Petrovic et al., 2022). These changes in the surface chemistry may also affect the surface charge of the sorbents and, altogether, PFAS sorption.

Powdered sorbents were either positively or negatively charged across the measured pH and had no PZC (Figure 4, SI). Depending on the sorbent type, aging affected the surface charge of each sorbent differently, and some of these changes can be linked to the elemental composition. These changes are likely associated with the formation of oxygen-containing functional groups during the aging process (Cheng et al., 2008) (Liu et al., 2013).

Carboxylation and oxidation of biochar can promote the development of  $-OH$  and  $-C=O$  groups, thereby increasing the surface acidity of the sorbents (Hale et al., 2011). In addition, aged sorbents may absorb carbon dioxide from the environment, leading to the formation of carbonates and a subsequent reduction in alkalinity (Liu and Chen, 2022; Xu et al., 2016). Some studies have also reported that the organic carbon, particularly aromatic moieties in biochar, may degrade into low molecular weight organic acids, further contributing to the decrease in biochar alkalinity (Liu and Chen, 2022). There was no significant variation in the zero-point charge of the granular sorbents aged with different methods (Figure S4, SI). However, there was a shift toward a lower PZC, indicating that the aged sorbents became more negatively charged compared to the unaged materials. While this analysis provides an overview of changes in surface charge due to different aging processes, the pH of the leachates is also important.

### **3.2. Leaching from soils treated with pre-aged sorbents**

#### **3.2.1. PFAS leaching from highly contaminated (undiluted soils) treated with aged and unaged sorbents**

Biochar showed the lowest PFAS immobilisation efficiency compared to other unaged sorbents, followed by GAC-E derived from biomass (not coal-based), in undiluted (highly contaminated and having high clay content) Soil A (Figure 4, S5 and S6, SI). Similar results have been observed in our previous studies and others, which report lower efficiency of biochar for PFAS immobilisation in soils (Kabiri et al., 2023). Furthermore, the efficiency of the sorbents for PFAS immobilisation was lower for short-chain PFAS compared to long-chain PFAS, despite some long-chain compounds, such as PFOS, being present at the highest concentrations in the soil (accounting for more than 99% of total PFAS). In addition, PFAS immobilisation was lower for short-chain PFAS with carboxylic acid head groups compared to those with sulfonic acid head groups of the same  $CF_2$  chain length in unaged sorbents (Figure S6, SI). Similar results have been observed in our previous studies and others, showing lower efficiency of carbon-based sorbents for the immobilisation of short-chain PFAS compared to long-chain PFAS in soils (Kabiri et al., 2021; Kabiri and McLaughlin, 2021; Navarro et al., 2023; Söregård et al., 2019).

A reduction in PFAS immobilisation was observed for aged sorbents, with the extent of reduction depending on sorbent type, aging method, and PFAS chemistry. When comparing the reduction in  $\sum$ PFAS leaching (16 detected PFAS) from highly contaminated Soil A treated with two different PACs or GACs that are aged (Figure S6, SI), both biological and physical

(both wet/dry and freeze and thaw) aging significantly reduced the effectiveness of the sorbents for  $\Sigma$ PFAS immobilisation compared with unaged sorbents ( $p < 0.001 - 0.05$ ). In contrast, chemically aged sorbents did not differ significantly from unaged sorbents. A similar effect was observed by Hale et al. (2011) on the sorption of pyrene on AC and biochar after wetting and drying cycles (physical) and biological aging of the sorbents. Statistical analysis of the two PACs for individual PFAS immobilisation in undiluted Soil A showed that aging effects were more pronounced for short-chain PFCA's, including PFBA and PFPeA. All aged sorbents, regardless of aging type, exhibited lower immobilisation efficiency than unaged sorbents ( $p < 0.001 - 0.05$ ) for short-chain PFCA. Significant differences were also observed among aging treatments (except between the two physical aging methods, which did not differ significantly), indicating that aging altered sorbent efficiency for short-chain PFCA immobilisation, particularly PFBA.

Similar trends to those observed for PAC were also observed for the two GACs. When  $\Sigma$ PFAS leachability was compared between aged and unaged GACs, both biologically and physically aged GACs showed significantly higher leachability (i.e., lower immobilisation efficiency) than unaged GACs (Figure 4, S5 and S6, SI). Statistical analysis for individual compounds showed that a greater number of PFCAs (e.g., PFBA, PFPeA, PFHpA, and PFOA), as well as some PFSAs, including PFOS, exhibited significantly reduced immobilisation efficiency following aging, particularly under biological and physical aging conditions ( $p < 0.05 - 0.001$ ). For GAC-E (not coal-based), immobilisation efficiency differed significantly among aging treatments for most PFCAs and PFOS, and GAC-E consistently showed the lowest efficiency compared with the other activated carbon-based sorbents. These results highlight that the source of activated carbon may influence its long-term effectiveness for PFAS immobilisation. Others also reported lower efficiency of coconut - based GAC than coal based on PFAS removal in water.<sup>10</sup>

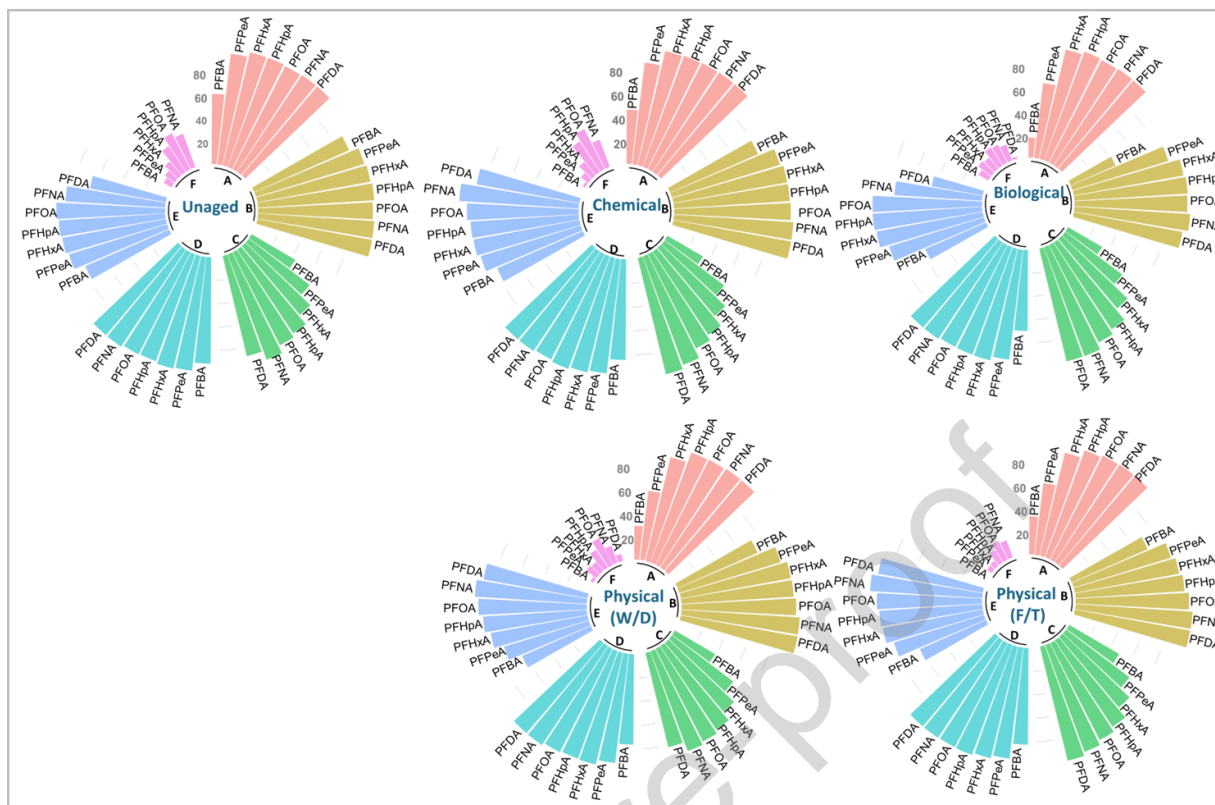


Figure 4. Circular bar chart showing PFCA immobilisation for undiluted ( $\Sigma$ PFAS~20mg/kg) Soil A treated with unaged and aged sorbents compared to untreated soil A ( $\Sigma$ PFAS~20mg/kg). A refers to PAC (coal), B to GAC (coal), C to CAC, D to PAC (coconut), E to GAC (peach) and F to biochar. Results are reported as the average of three replicates ( $n = 3$ ).

The reduction in PFAS immobilisation efficiency observed for PACs and GACs following biological and physical aging can be attributed to changes in both surface charge and chemistry, and surface area. Biologically aged sorbents, particularly PAC, exhibited the lowest surface charge across a range of pH values (Figure S4, SI), which may have reduced electrostatic interactions with short-chain PFAS, the key compounds with reduced immobilisation. Furthermore, the surface oxygen content as well as the O/C was significant compared to the unaged sorbent, making the surface less hydrophobic (Table S4, SI), which can affect the sorption of both short- and long-chain PFAS. Physically aged sorbents showed a significant reduction in surface area (SA) compared with unaged materials (Figure 1b), likely contributing to their decreased immobilisation efficiency. Overall, aging reduced the surface area of the PACs and GACs by approximately 20–32% (Figure 1b), indicating a loss of accessible sorption sites. Both physical and biological aging caused the most pronounced morphological changes, primarily through pore clogging, as evidenced by SEM images (Figures S1-S3, SI). Although some pores remained open in biologically aged sorbents, many were partially filled with residues (biofilms), whereas physical aging resulted in more extensive pore blockage, as evident by the SEM images. These changes are consistent with reduced accessibility of internal pore space, thereby limiting sorption capacity. In contrast, chemical aging led to pore rupture and, in some cases, the formation of new, smaller pores due to the oxidative effects of  $H_2O_2$  (Figure S1, SI). Similar changes in the sorbent

morphology were observed in the microscopic images of the sorbents in other studies (Tan, Lianshuai et al., 2020) (Nie et al., 2019). This could be the reason for no significance effect of chemically aged PAC and GAC efficiency on PFAS immobilisation compared to unaged sorbents. Changes in surface morphology and pore blockage were mostly observed for GAC, whereas these features were not as clearly visible in the SEM images of PAC. Another reason may be the much smaller particle size of PAC, which makes the detection of morphological changes less distinct in SEM analysis (Figure S7, SI). Physically and biologically aged PAC and GAC also showed a significant increase in O/C ratio compared to unaged or biologically and chemically aged sorbents (Table S4, SI), which could be one of the reasons for the significant decrease in PFOS immobilisation under biological and physical aging compared to unaged and chemically aged sorbents.

For CAC (Sorbent E), the immobilisation efficiency of all aging treatments differed significantly from that of the unaged sorbent, except for physical aging (wet–dry cycles). In addition, the efficiencies of the different aging types differed significantly from each other for  $\Sigma$ PFAS immobilisation (Figure S6 and S7, SI). No differences in immobilisation efficiency between wet–dry aged CAC and the unaged sorbent may be attributed to the absence of a significant change in specific surface area (SA) following aging (Figure 1b), as well as the fact that sorbents subjected to different aging treatments remained negatively charged across the entire pH range (Figure S4, SI). In addition, this sorbent may contain additives to maintain it in suspension, which could influence its properties and respond differently under various aging conditions. Consistent with our findings, Jiang et al. (2025) reported no significant change in CAC efficiency for PFAS removal from solution following wet–dry cycling aging.

With biochar, there were no significant differences in PFAS immobilisation between aged and unaged biochar used in this study (Figure 4 and S6, SI). This was due to biochar already having a very low removal efficiency for all PFAS. However, the significant difference was observed on immobilisation of some of the long-chain PAFS, such as PFHxS, PFHpS, PFNS, PFDA, FOSA and 6:2 FTS after biologically or physically (W/D) aged biochar with unaged biochar.

In undiluted Soil B (sandy soil) treated with unaged sorbents, PAC and GACs showed similar immobilisation efficiency, retaining more than 95% of most PFAS. In contrast, biochar again exhibited the lowest efficiency, followed by CAC (Figure S8, SI). For PAC and GAC, a decline in sorbent efficiency was observed only for PFBA, primarily under biological and physical aging conditions, consistent with trends observed in Soil A. Likely due to its sandy texture, lower mineral content, and low organic matter, the complexity of leachates (less DOC and suspended minerals) from Soil B was less, which resulted in only a minimal decline in sorbent efficiency. The presence of high OC and suspended minerals not only affects PFAS leachability but also the sorption by sorbents, as there would be a competition between these particles and PFAS for sorbent sorption sites (Kabiri and McLaughlin, 2021).

### 3.2.2. PFAS leaching from diluted soils treated with pre-aged and unaged sorbents

No significant differences were observed across the different aging treatments in PFAS leachability in diluted Soil A. Biochar showed the lowest immobilisation efficiency compared to the AC-based sorbents (Figure S8, SI) using the unaged (original) sorbents, similar to undiluted soil and findings from our previous studies (Kabiri et al., 2023; Navarro et al., 2023). Similar to undiluted soil, a significant increase in PFAS leaching was observed for short-chain PFAS with carboxylic acid functional groups (e.g., PFBA and PFPeA) in diluted Soil A (Figure S8, SI), with the most pronounced effect in biologically aged sorbents, likely due to decreased surface charge (Figure S4, SI) and biofilm formation. We and others observed that the surface charge can affect PFAS sorption and immobilisation, especially for short-chain PFAS, where electrostatic interaction plays a key role (Kabiri et al., 2023; Park et al., 2020). In diluted Soil B, most sorbents were able to immobilise PFAS completely in soil (Figure S8, SI), likely because diluted Soil B had a sandy texture with less organic carbon compared to the clayey Soil A. As a result, there was less competition between the PFAS and other compounds (e.g., DOC) on sorbent sorption sites. These results indicate that aging-induced changes in the physical and chemical properties of the sorbents did not significantly affect PFAS sorption in soils with  $\sum$ PFAS of 2–4 mg/kg. In highly contaminated soils ( $\sum$ PFAS  $\approx$  20 mg/kg), aging effects were primarily observed for short-chain PFAS, where a reduction in immobilisation efficiency was evident. In contrast, long-chain PFAS, which accounted for more than 98% of total soil PFAS, remained sorbed, and immobilisation efficiencies for long-chain PFAS remained between 85–99.9% for most activated-carbon-based sorbents, even after aging.

### **3.3. Leaching from sorbent-treated soils subjected to accelerated aging**

When soil and sorbent were aged together, the reduction in PFAS leaching from undiluted Soils A and B following treatment (different aging) was lower than that observed before aging (Figure 5). This trend was evident for both short-chain and long-chain PFAS with different functional head groups. In undiluted Soil A and B treated with pre-aged AC-based sorbents, PFAS leachability was comparatively less than when soil and sorbent were aged together. Hale et al. (2011) also observed a greater effect of aging on decreasing AC and biochar efficiency on pyrene sorption when the soil and sorbents were aged together. It was also evident that chemical aging had a more pronounced adverse effect on sorbent performance compared with the other aging treatments, with higher PFAS leachability observed for most PFAS (Figure 5). In chemically aged Soil B, below 50% to no PFAS immobilisation was observed for most PFCAs with  $\text{CF}_2$  chain lengths from C3 to C6 across all AC-based sorbents tested. However, when the sorbent aged chemically without soil, their efficiency did not differ from unaged sorbents (Figure 5 and S6, SI). Differences between the two soils were apparent following chemical aging, with the impact of chemical aging appearing to be greater in Soil B than in Soil A. In contrast, when soils were treated with pre-aged sorbents, changes in PFAS leachability were more evident in Soil A than in Soil B.

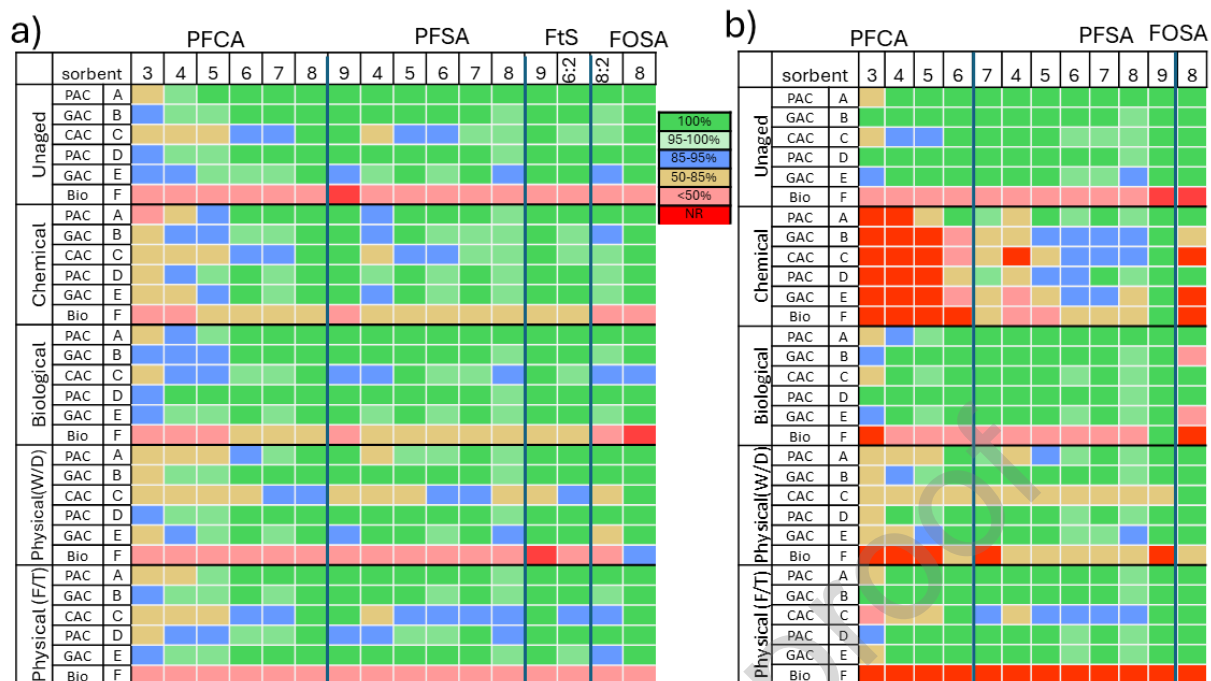


Figure 5. Heatmap of the percentage of reduction in PFAS leaching of (a) Undiluted Soil A ( $\Sigma$ PFAS~20mg/kg) treated with sorbents, followed by different aging and then leaching and (b) Undiluted Soil B ( $\Sigma$ PFAS~20mg/kg) treated with sorbents, followed by aging and then leaching. Unaged means that the treated soil with sorbent was not subjected to aging. The numbers in the top row refer to the CF<sub>2</sub> number of the PFAS. A refers to PAC (coal), B to GAC (coal), C to CAC, D to PAC (coconut), E to GAC (peach) and F to biochar. Results are reported as the average of three replicates ( $n = 3$ ).

SEM images of sorbents separated from the soil after the sorbents and soils were chemically aged together showed distinct differences in sorbent morphology between the two soils, as well as compared with sorbents that were chemically aged alone. In Soil A, chemical aging primarily led to pore blockage of GAC, with some pores being partially or completely blocked (Figure 6a, b, e, and f). In Soil B, pore blockage was also observed; however, more extensive structural damage to GAC was evident, including the formation of large cracks and erosion of the sorbent surface (Figure 6c and d). In contrast, when GAC-B or GAC-E was chemically aged alone, chemical aging (H<sub>2</sub>O<sub>2</sub> treatment at elevated temperature) resulted in the opening of some pores and disruption of the pore structure, including the merging of adjacent pores (Figure 6g and h). Despite these morphological changes, no differences in PFAS immobilisation were observed compared to the unaged sorbents.

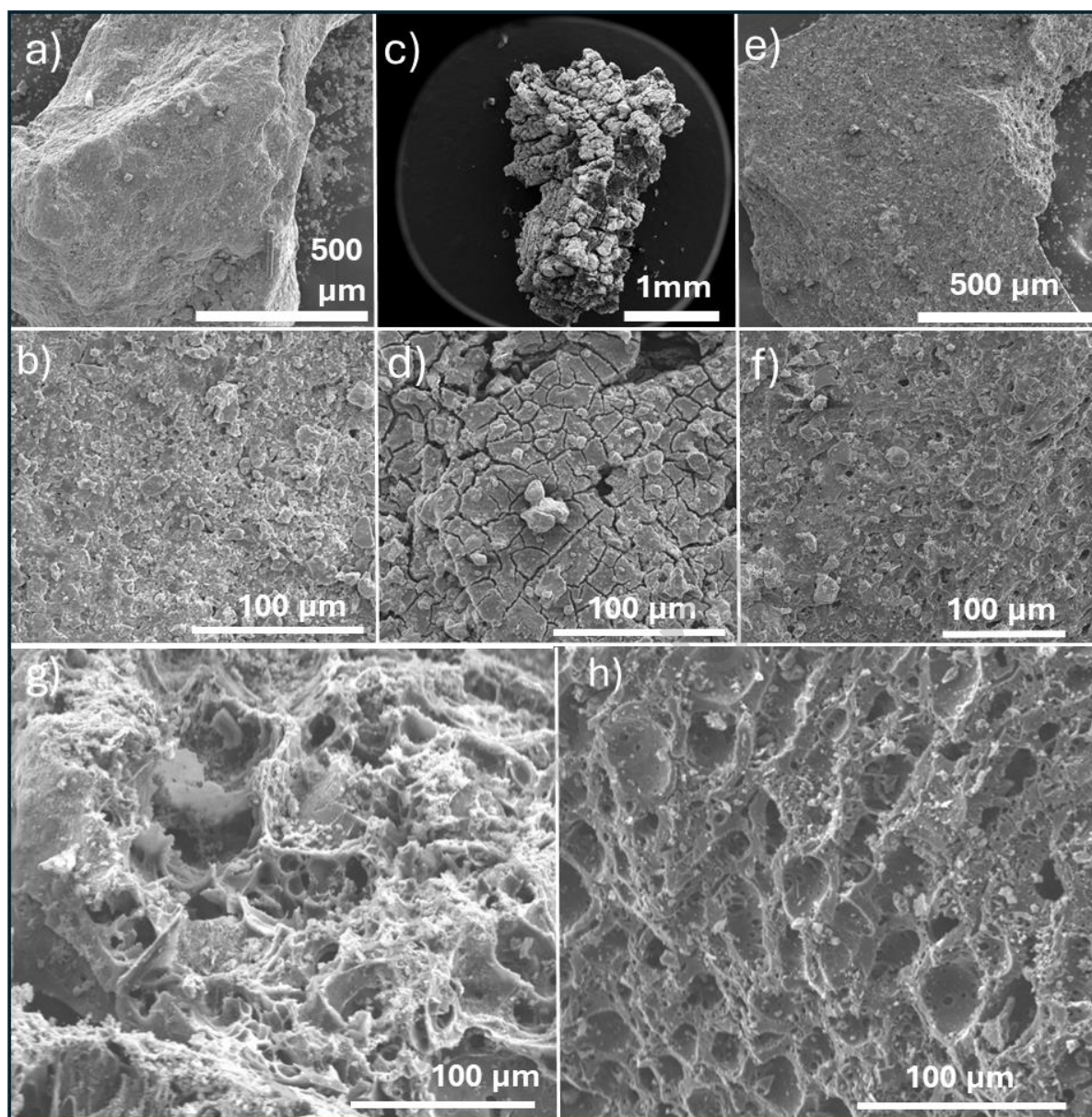


Figure 6. SEM images of (a and b) unaged sorbent B, (c and d) chemically aged sorbent B with Soil A, and (e and f) chemically aged sorbent B with Soil B, (g) chemically aged GAC B without soil and (h) chemically aged GAC E without soil.

The observed increase in PFAS leachability when soil and sorbent were chemically aged together may be related to a combination of mechanisms. Treatment of soils with  $H_2O_2$ , particularly at elevated temperature, can increase DOC concentrations, which may compete with PFAS for sorption sites on the sorbents. For example, Xu et al. (2022) reported that  $H_2O_2$  treatment increased soil DOC by approximately fivefold compared with pristine soil. In addition,  $H_2O_2$  exposure may alter sorbent surface chemistry by increasing oxygen-containing functional groups and reducing surface charge, potentially decreasing PFAS sorption, particularly for short-chain compounds (Liu et al., 2013; Xu et al., 2016). Increased oxygen content after chemical aging was confirmed with elemental analysis and EDS analysis (Table S4 and Figure S3, SI). Chemical aging with  $H_2O_2$  may also promote the degradation of PFAS precursors or long-chain PFCAs into shorter-chain PFAS under elevated temperatures and in

the presence of catalytic species such as iron nanoparticles or Fe(III), which are commonly present in soils (Mitchell et al., 2014). Therefore, higher concentrations of short-chain PFAS may be present in chemically aged soils compared with the original soils. As the original sorbents are not fully capable of immobilising short-chain PFAS, a further decline in PFAS sorption would be expected following chemical aging, particularly when combined with sorbent morphological changes and increased concentrations of short-chain PFAS.

Physical aging (wet/dry) was the second type of aging that led to a substantial increase in PFAS leachability compared to unaged AC-based sorbents in both soils (Figure 5), affecting both short- and long-chain PFAS. The increase in PFAS leachability (decrease in immobilisation) is likely related to pore closure and clogging during physical aging, as evidenced by low- and high-magnification SEM images (Figure S9, SI). This suggests that some soil pores may have been filled with suspended colloids (minerals or DOC) during wetting and drying cycles. Biological aging had the least effect on PFAS leachability, with no significant difference from the unaged conditions. Similar efficiency of the biological aging and unaged samples could be related to the aging conditions during biological aging (3 weeks), where the sorbents and soil were suspended in the liquid with a different cation (inoculum solutions) that can facilitate PFAS sorption through the cation bridging or complexation (Cai et al., 2022). SEM images of the GAC separated from the soil and sorbent aged biologically together showed less pore blockage compared to chemical and physical aging, especially in Soil B with sandy texture (Figure S10, SI). This again shows that the soil texture and composition are also important during the aging condition and the way they may affect the sorbents.

Biochar showed the lowest efficiency, followed by CAC, in PFAS immobilisation across different types of aging when soil and sorbents were aged together, compared to the other sorbents (Figure 5). The same mechanisms that contributed to the decrease in PFAS immobilisation efficiency in other sorbents likely apply to biochar and CAC. However, it should be noted that for chemically and biologically aged biochar in soil, its efficiency slightly increased compared to the unaged condition, where the SEM images showed that pores were still open in chemically aged biochar with Soil A and B (Figure S10, SI).

In diluted soil, the effects of aging differed slightly from those observed in highly contaminated soil. In Soil A, physical aging (wet/dry) resulted in the lowest PFAS immobilisation efficiency compared to unaged soil, whereas in Soil B, chemical aging exhibited the lowest efficiency, consistent with trends observed in undiluted soils (Figure S11, SI). Biochar again showed the lowest efficiency, followed by CAC on PFAS immobilisation in both undiluted soils.

It is important to note that a few weeks of accelerated laboratory aging cannot fully replicate natural environmental conditions. However, the elevated temperatures used during physical (W/D) or chemical aging in this study are likely to accelerate reactions that would otherwise occur slowly in the environment. For example, for reactions with an activation enthalpy of approximately 70 kJ/mol, a 10°C increase in temperature can double reaction rates (Hale et al., 2011). Given that the decomposition of organic matter in sediments typically has activation enthalpies ranging from 54 to 125 kJ/mol (mostly between 50–90 kJ/mol),

increasing the temperature to 70°C for about one month could accelerate this reaction (Middelburg et al., 1996) (Kelly and Chynoweth, 1981) (Therkildsen and Lomstein, 1993). Hale et al. (2011) suggested that if similar processes occur during the aging of AC and biochar, exposing the sorbents to 60 or 110 °C for two months during the physical aging (wet/dry) could represent approximately 100 to 20,000 years of aging at a constant temperature of 10 °C. In this study, with wetting followed by drying cycles at 70 °C for approximately a month, the physical aging conducted may correspond to approximately 70 years of natural aging at 10°C. While such estimates help contextualise accelerated aging experiments, laboratory aging methods still cannot capture the full complexity of field conditions, such as the influence of plants, macrofaunal activity, soil structural changes (e.g., shrink-swell dynamics), and other long-term weathering.

#### 4. Conclusions

This study, for the first time, investigated the effects of different environmental aging conditions (physical, chemical, and biological) on the efficiency of various sorbents—including GAC, PAC, CAC, and biochar—for PFAS immobilisation in soil. When aging was applied solely to the sorbents, physical aging (wetting and drying cycles) and biological aging caused a greater decline in sorbent performance than chemical aging, likely due to pore blockage and the associated reduction in specific surface area, as a result of biofilm formation. When soil and sorbents were aged together, chemical and physical aging (wetting/drying cycles) resulted in the most significant decrease in sorption efficiency, again due to the pore blockage and reduced surface area. However, the impact of aging varied with soil type; soils with different physical and chemical properties (e.g., texture) responded differently to aging. Therefore, future studies should consider the effect of soil properties on PFAS immobilisation following aging with systematic studies on understanding the reason behind it. Notably, the effect of changing in reducing the PFAS immobilisation was mostly related to the pore blockage and changes in the sorbent surface charges.

Among sorbents, PAC showed the greatest resistance to aging, whereas biochar was the most susceptible to efficiency loss. Feedstock origin also played a role, with PAC and GAC derived from coal sources being more stable than those derived from alternative feedstocks such as coconut and peach. Despite the reduced efficiency under harsh aging conditions, AC-based sorbents remained highly effective, retaining over 85% PFAS immobilisation capacity in soil. Furthermore, the aging impact was more pronounced in highly contaminated soils ( $\Sigma$ PFAS > 20 mg/kg), whereas in less contaminated soils ( $\Sigma$ PFAS 2–4 mg/kg), sorbents maintained comparable performance to unaged materials.

It should also be noted that the ASLP leaching test used in this study to assess PFAS immobilisation in treated soils is an aggressive batch leaching method involving vigorous shaking. Therefore, it does not represent point-in-time leaching under natural environmental conditions. These results demonstrate that, even under accelerated and harsh aging scenarios, AC-based sorbents generally remain effective for PFAS immobilisation, especially in moderately contaminated soils.

The accelerated aging approach used in this study provides a practical means of assessing the potential long-term treatment stability at bench scale, complementing field monitoring, where extended timelines are impractical. Future studies should investigate a broader range of sorbents across diverse soil types and contaminant profiles to better define the mechanisms governing performance changes under different aging regimes. This approach could also be valuable for researchers developing new engineered sorbents for soil treatment, as it enables early assessment of their durability and long-term performance under realistic environmental stressors. Such efforts will help develop predictive tools and strengthen confidence in the long-term effectiveness of sorbent-based treatments for PFAS-contaminated soils.

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## **6. Declaration of competing interest:**

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

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**Declaration of interests**

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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**Highlights**

- Physical and biological aging reduce sorbent efficiency due to pore blockage.
- PAC sorbents least affected, while biochar most susceptible to aging effects.
- Aging effect differed based on the soil type when soil/sorbent aged together.
- Activated carbon-based sorbents remain effective even after harsh aging conditions.