Review

Interactions of Per- and Polyfluoralkyl Substances (PFAS) with Landfill Liners

Will P Gates ^{1, *}, Alastair JN MacLeod ¹, Andras Fehervari ¹, Abdelmalek Bouazza ², Daniel Gibbs ³, Ryan Hackney ³, Damien L Callahan ⁴, Mathew Watts ⁵

1. Deakin University, Geelong, Australia. Institute for Frontier Materials (Burwood Campus), Victoria, Australia; E-Mails: will.gates@deakin.edu.au; alastair.macleod@research.deakin.edu.au; andras.fehervari@deakin.edu.au
2. Monash University, Department of Civil Engineering, Clayton, Victoria, Australia; E-Mail: malek.bouzza@monash.edu
3. Geofabrics Centre for Geosynthetic Research, Innovation and Development, Geofabrics Australasia, Molendinar, Queensland, Australia; E-Mails: d.gibbs@geofabrics.com.au; r.hackney@geofabrics.com.au
4. Deakin University, Geelong, Australia. School of Life and Environmental Sciences (Burwood Campus), Victoria, Australia; E-Mail: damien.callahan@deakin.edu.au
5. Environment Protection Authority Victoria, Macleod, Victoria, Australia; E-Mail: mathew.watts@epa.vic.gov.au

* **Correspondence:** Will P Gates; E-Mail: will.gates@deakin.edu.au

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Abstract

This review synthesises the available published research on interactions of per- and polyfluoroalkyl substances (PFAS) with landfill liners, with the view to inform on the expected behaviour of these persistent environmental pollutants in landfills. The review addresses the nature and significant types of PFAS compounds that are destined for landfills, as well as their by-product. It discusses the known and anticipated interactions with separate landfill liner

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components, namely geomembranes, geosynthetic clay liners and compacted clay liners. Various water-soluble PFAS are shown to advectively transport through geosynthetic clay liners (GCL) and showcase the limitations of relying on mineral liners alone to retain PFAS. Addition of activated carbon, while increasing saturated hydraulic conductivity, significantly increases PFAS retention by the GCL and reduced PFAS flux to manageable concentrations. An assessment of the relative risk for environmental exposure of different types of PFAS from landfills through interaction with those liner components is achieved with reference to published case studies of PFAS detection in and around landfills from Australia and around the World.

Keywords

Landfills; PFAS; environmental fate and migration; geomembranes; geosynthetic clay liners; compacted clay liners; hydraulic performance

1. Introduction

Waste is an industry that, in Australia alone, exceeds \$AUD 13.4 Billion per year (2016-2017 data) [1], with more than \$AUD 3 Billion associated with disposal in waste facilities. Waste containment facilities are critical infrastructure providing essential community services. Waste containment facilities such as municipal solid and hazardous waste landfills and repositories are indispensable for mitigating the long-term environmental and human health impact of contaminants [2]. Hydraulic barrier liner systems are required to minimise the escape of pollutants into soil and groundwater from these containment facilities and to achieve the aim of long-term and safe storage.

Recent attention has been paid to contaminants such as per- and polyfluoroalkyl substances (PFAS) due to growing concerns around their toxicology as well as increasing evidence for their long-term presence in aquatic, terrestrial and atmospheric ecosystems [3, 4]. PFAS in Australia are most notorious lately for their use in aqueous film-forming foams (AFFF) for firefighting [5-8]. PFAS comprise a highly diverse group of chemical compounds which have been manufactured and used in myriad industries worldwide for more than 60 years [3, 4], including metal coatings, electronics, automotive, medical, textiles and food. Since the 1960s, perfluorooctane sulfonate (PFOS) has been a critical ingredient in AFFF used to combat petroleum fires [9], and is the source of many legacy releases of PFAS into the environment. Although the predominant PFAS in AFFF is now in the form of precursor fluorotelomers, AFFF still constitute significant environmental releases of PFAS when used.

The chemical stability of PFAS compounds means that when released, they persist in, and transport between, the atmospheric [10-14], terrestrial (soil and sediments) [15-20] and aquatic environments (surface and ground waters) [19-26] together with the anthropic environment (agriculture, landfills, sewage, and water treatment systems) [27]. Possible transport pathways are illustrated in Figure 1. Within each of the spheres or systems, a range of conditions exist which determine the partitioning of PFAS [4, 15, 18] and thus their fate and behaviour. In soils and sediments, the mineralogy, organic matter content, surface charge, particle size, porosity, permeability, infiltration rate, pore water content and pore water pH all contribute to the high

mobility of PFAS with minimal breakdown [18]. In groundwater, the depth from surface, flow rate, infiltration rate and subsurface conditions such as reduction-oxidation (redox) potential, pH and salinity are important in determining the extent of transfer [22]. In contrast, an additional impact of atmospheric conditions (humidity, pressure, temperature) control volatilisation in surface waters [19]. Within the atmosphere itself, atmospheric conditions, wind direction and speed, air-borne particulate/colloid load and concentration of potential oxidants determine transfer and breakdown [12].

Figure 1 Pathways for PFAS compounds into atmospheric, terrestrial, aquatic and anthropic environments.

Characteristics of the soils and sediments strongly impact PFAS transfer into groundwater [20] or from surface water to groundwater [22] and from groundwater to the air. The environmental behaviour of perfluoroalkyl acids (PFAA) depends on their perfluoroalkyl chain length and functional group. Long carbon-chain PFAA ($C_nF_{2n+1}COOH$, where $n \geq 7$ and $C_nF_{2n+1}SO_3H$, where $n \geq 6$) show higher sorption to soils and sediments compared to short carbon-chain ($C_nF_{2n+1}COOH$, where $n < 7$ and $C_nF_{2n+1}SO_3H$, where $n < 6$) PFAA [28]. Hydrophobic interactions between PFAA and organic carbon, ligand binding through divalent cations, electrostatic interactions between the functional end groups of PFAA and mineral and organic phases and oxides in soils all play a role in the sorption process.

While most research worldwide has focussed on the partitioning of PFAS with, and between, the environments described in Figure 1, surprisingly limited research has been conducted on how partitioning influences the fate and behaviour of PFAS in engineered hydraulic barrier systems. This paper reviews the various sources of PFAS in wastes that generally are disposed of in landfills, the known make-up of PFAS currently existing in landfills and discusses aspects of PFAS partitioning considered to be important in controlling their fate and behaviour in landfill settings. Given the lack of specific information regarding PFAS behaviour in liner components of modern composite lining systems, a relevant discussion is presented to identify knowledge gaps and provide limited evidence from recent research attempting to address these gaps.

1.1 Overview of PFAS in Wastes

According to the Australian National Waste Report (2018) [29], in 2016-2017 (latest data available) of the 67 million tonnes (MT) of waste generated in Australia, 21.73 MT (32%) was disposed of in about 1,000 operating landfills. About 54 MT of the 67 MT generated in Australia was classed as core waste made up of 13.8 MT (25.5%) municipal solid wastes (MSW) from households, 20.4 MT (37.8%) each from commercial and industrial (C&I) wastes from businesses and construction and demolition (C&D) activities. Selected examples of PFAS-bearing materials from these waste streams are presented in Table 1.

Table 1 Major sources of PFAS contaminants destined for landfills.

Waste stream	Industry Application	Selected examples	Waste stream quantity destined for Australian landfill (approx.) ¹
Municipal solid waste	Cosmetics & personal care products	Cosmetics, shampoos	13.8 MT
	Food processing	PTFE liners (trays, ovens), food packaging	
	Household products	Non-stick coatings, surface treatments for textiles, upholstery, carpet and leather, floor polishes, cleaning agents, car waxes	
	Biocides (herbicides & pesticides)	EtFOSA in ant/termite baits, PFPAs and PFPiAs as anti-foaming agents	
Commercial & industrial	Aviation & aerospace	PTFE and PFA tubing, gaskets, cables	20.4 MT
	Automotive	Wiring and cabling, seals, gaskets, lubricants	
	Electronics	PVDF and PTFE as insulators	
	Energy	FEP, PVDF films covering photovoltaic panels	
	Medical products	Biocompatible implants and surgical patches	
	Paper & packaging	Oil, grease and water repellent (including food packaging), LDPE bags	
	Textiles (upholstery, carpets), leather & apparel	PTFE in consumer products (outdoor equipment and clothing, housewares), oil- and water-repellent coatings, PFOA-based chromium treatment for paper and leather.	

	Photo-lithography & semi-conductors	PFOS used in chip manufacture	
	Cable & wiring	PTFE, PVDF coatings and jackets	
	PFAS production	Processing aids	
Construction & demolition	Building & construction	Waste concrete and brick (contamination from AFFFs) Sealants, PTFE, PVDF coatings, adhesives, surface treatments	20.4 MT
Contaminated wastes & wastewaters	Metal plating	Wetting agent, mist suppressant, surfactants	
	Mining	Surfactants in ore mining	1.8 MT
	Firefighting & safety	AFFFs	

¹Waste stream data from [29]

Worldwide, municipal solid waste (MSW) is made up of household goods, organics, paper, plastic, glass, metals, dry goods, white goods, carpets, textiles including curtains and clothing, and other materials not separated for reuse, recycling, or energy production. The concentrations of PFAS compounds in these wastes are not well characterised; however, PFAS compounds are known to be present as constituents in many products disposed to landfill, and PFAS and their precursors are now recognised as being ubiquitous in most MSWs worldwide [30-34]. In addition to being a direct source of consumer exposure, single-use products such as cleaning products [34], take-away or fast-food packaging and household textiles (furniture fabrics, curtains and clothing) constitute a significant potential source of PFAS in MSW [35, 36].

Generation of core wastes in Australia increased by about 5.9 MT (12%) over the 11 years spanning 2006 to 2017. C&D activities drove most of this growth. A further 32.7 MT (for 2016-2017) of non-core wastes (e.g., agricultural organics, fly ash from coal power plants, red muds from aluminium processing) are landfilled in Australia [29]. While C&D wastes, and some industrial ash wastes pose continued risk for PFAS contamination and transfer to landfill [37], most mine wastes pose minimal risk.

Most di- and tri-substituted fluorotelomer alcohol (FTOH) phosphate surfactants (diPAPS, triPAPS) and di-substituted FTOH thioester phosphate surfactants (S-diPAPS) form stable fluorotelomeric carboxylic acids (FTCA) under anaerobic conditions [38]. Thus, while FTOH are unlikely to form PFCA in landfill, FTOH are known to metabolise to perfluoralkyl carbonic acids (PFCA) [39] under aerobic conditions [39], and thus serve as a potential source of PFCA in landfills. Various PFCA were found to be present in 5 of 14 food packaging products studied [35]. As much as 56% of sweet and bread wrappers, 38% of sandwich wrappers, and 20% of paperboard products (used with fried foods) from US fast food outlets exhibited detectable fluorine (predominantly from PFAS compounds) [36]. Samples with high total fluorine levels, but low measured PFAS values were considered to contain either (i) undetected volatile PFAS, (ii) polymeric PFAS, (iii) newer, undisclosed PFAS compounds, or (iv) other fluorinated compounds.

Another significant potential source of PFAS is from plastic and textile wastes, a large proportion coming from C&I wastes, primarily made up of plastics, metals, white goods, electronics, textiles (including carpets), and other materials not yet separated for reuse, recycling, or energy production [40, 41]. A range of PFCAs, and notably variously substituted FTOH, were found in the range 10 - 50 µg/kg (although concentrations as high as 4,000 µg/kg were observed) in 21 different industrial textile products (marquee awnings, vehicle seat covers and maritime use fabrics) where a need for stain and water resistance was required [42]. Overall, PFAS are highly prevalent in industrial products, and are most likely to have high concentrations in materials with requirements for stain, oil and water resistance.

C&D wastes make up a further 36% of the total core waste disposed of in landfills in Australia. Core wastes from C&D activities, particularly concrete from firefighting training grounds [43], is a potential source of PFAS contamination in landfills. C&D activities themselves can be a source of PFAS compound contamination of the sites where the C&D wastes are processed for recycling, and in the locations where C&D recycled material may be used (e.g., as functional fill and road base).

A recent study [41] of 126 building materials detected up to 32.9 µg/kg PFCAs in all processed timber flooring and building products analysed, including wood fibre insulation, and up to 24.5 µg/kg in construction sealants and façade materials. Significant levels of total PFAS in several building coatings and sealants - up to 4.3 g/L in one sampled coating - have been observed in a variety of building materials [42]. The high loading in the coatings was to achieve desired water, dirt, and oil-repelling properties upon application to building façades and surfaces.

Finally, biosolids recovered from wastewater treatment (and from the treatment of municipal water supplies in some Australian jurisdictions) can have elevated levels of PFAS [44]. PFCA and perfluoroalkyl sulfonic acids (PFSA) were detected in the influent of two undisclosed waste water treatment plants (WWTP) in Australia (mean total PFAA concentrations of 57 ± 3.3 to 94 ± 17 ng/L and 31 ± 6.1 to 142 ± 73 ng/L) [45]. Perfluorohexanoic acid (PFHxA) was observed in the highest mean concentration (20 ± 2 and 17 ± 13 ng/L, respectively) at the same two WWTP. Significantly, the precursor telomer, 6:2 FTOH, was detected during a 15-month testing campaign at concentrations greater than either perfluorooctanoic acid (PFOA) or PFOS [45], indicating the replacement of telomer-based products in recent years. In some Australian jurisdictions, biosolids are disposed of in landfills or stockpiled, and given their known concentrations of PFAS, can contribute significant amounts of PFAS compounds to landfill leachates.

2. PFAS in Landfills

Past research has focussed largely on understanding the hydraulic performance of the various components of composite liners for containing traditional contaminants over both the short-term and some aspects of the long-term. However, there has been no evaluation of the effectiveness of liner systems for contaminants of emerging concern such as PFAS. of significance is that existing liner systems have not been specifically designed to consider the fate (containment, transformation, or release) of PFAS.

2.1 PFAS Landfill Leachates

The presence of PFAS compounds in landfill leachate is well documented, and much of this information is now incorporated into geochemical indices for both monitoring and planning

purposes in Australia [46-48]. Recent comprehensive surveys on PFAS in Australian landfills [49-51] have found PFAS to be ubiquitous in the leachates emanating from them. Five different PFAS (Table 2) were universally present in 97 leachate samples, collected in mid-2013 to late-2014 from 27 landfills across Australia (accepting municipal, construction/demolition, contaminated or hazardous wastes), including three PFCAs (PFHxA, perfluoroheptanoic acid PFHpA, and PFOA) and two PFSA (perfluorohexanesulfonic acid PFHxS and PFOS) [49]. Notably, several trends should be noted:

- Average concentrations of eight different PFAS compounds were higher in leachates collected from operating landfills or landfill cells accepting primarily MSW than were observed from closed landfills.
- A significant negative correlation existed between leachate PFAS concentrations and landfill age - with younger landfills having greater PFAS concentrations, as well as greater concentrations of shorter perfluoroalkyl chain lengths, which reflects the increased prevalence of the use of shorter chain length PFAS as well as fluorotelomer and other precursor PFAS compounds.
- Leachates collected from landfills accepting C&D wastes had higher levels of PFAS than landfill accepting municipal wastes.
- Alkaline leachate pH and higher total organic carbon content were associated with elevated concentrations of several PFAS compounds.

Table 2 Mean (ng/L) concentrations (standard deviations) of PFASs in landfills/landfill cells grouped by operational status and dominant waste type accepted [49]. Data reported to 2 significant figures.

PFAS compound detected	Mean Concentration (ng/L)		
	Operating landfills (> 50% MSW) n = 12	Operating landfills (> 50% C&D) n = 7	Closed landfills (> 50% MSW) n = 7
PFHxA	1300 (1700)	5000 (8100)	660 (300)
PFHxS	940 (1000)	3700 (5100)	740 (490)
PFOA	510 (410)	1400 (1200)	390 (170)
PFHpA	360 (360)	760 (760)	220 (110)
PFOS	300 (330)	1100 (910)	180 (250)
PFNA	29 (24)	98 (110)	13 (6.8)
PFDA	22 (36)	46 (83)	11 (14)
PFUdA	3.0 (4.7)	4.6 (3.4)	2.2 (1.9)
PFDoDa	1.8 (3.5)	1.1 (1.6)	2.7 (3.3)

In a pilot study conducted in Victoria, Australia, PFHxS (0.76 - 5.3 µg/L), PFOA (0.49 - 1.6 µg/L), PFOS (0.4 - 1.2 µg/L) and 6:2 FTOH (0.062 - 0.87 µg/L) were found to be present in leachates from stockpiled solid inert wastes [51]. Similar levels of PFOA (0.09 - 3.12 µg/L), PFOS (0.02 - 1.2 µg/L) and PFHxS (0.03 - 2.41 µg/L), but also PFHxA (0.11 - 7.74 µg/L), perfluorobutansulfonic acid (PFBS) (0.04 - 23 µg/L), perfluoropentanoic acid (PFPeA, undetected - 2.07 µg/L) and PFHpA (0.02 - 1.6 µg/L) were also observed in leachates from landfilled wastes. When grouped by waste type, landfills that accept higher proportions of C&D wastes have, on average, higher concentrations of PFAS compounds in their leachates, apart from perfluorododecanoic acid (PFDoDa). Two sites exhibited higher levels of total PFAS, mostly as PFBS and PFHxA, compared to the other 17 sites studied, and both landfills received waste products from end-of-life vehicle interiors and upholstery, known to contain PFAS compounds, and/or paper manufacture wastes and/or sludge from wastewater treatment plants. Significantly, both sites were the only sites identified to recirculate leachate [51]. An important observation is that about a third of Australian landfills discharge leachate directly to WWTP [49] and this is expected to result in re-distribution and concentration of PFAS to biosolid phases.

Similar findings have been reported in Canada [52], China [30, 53], Ireland [54, 55], Spain [56] and the United States [32, 33, 37, 57, 58], at generally similar concentrations, with one measurement near manufacturing activities at concentrations exceeding 290,000 ng/L (290 mg/kg) [53]. Carpet waste and clothing were determined [33] to be sources of biotransformation precursors of PFAS compounds in the aqueous phases of anaerobic landfill model reactors. The biotransformation products - 5:3 FTCA (3.9 nmol/L, $\approx$ 1.3 µg/L), PFHxA (2.9 nmol/L, $\approx$ 0.9 µg/L) and PFOA (2.6 and 6.3 nmol/L, $\approx$ 1.1 to 2.6 µg/L) - were responsible for most of the observed 8.5 nmol/L PFAS. Sample heterogeneity was considered the main reason for the large range in PFOA release. Because most of the release of measured PFAS compounds into the aqueous phases occurred after 100 days reaction, unbound PFAS was considered unlikely to be the main source of biotransformation.

The concentrations of 70 PFAS compounds in 95 leachate samples taken from 18 US landfills in different climates and waste ages [32] indicated that of an estimated 600 kg PFAS released into leachates per year (from 2013 data), the biotransformation precursor 5:3 FTCA was the largest contributor, amounting to about 30%, or $\approx$ 190 kg/y. As for the Australian studies [49-51], PFAS compounds were found to be in higher concentrations in leachates of younger (< 10 years of operation) landfills, indicative of increased use and disposal of PFAS-containing consumer products, but also the tendency for PFAS to disperse rapidly from landfills. It was also shown that short-chain PFASs tend to dominate over long-chain PFASs in most landfill leachates [59].

2.2 PFAS Volatiles in Landfill Gaseous Emissions

Common volatile forms of PFAS are the FTOHs, generally in the hundreds of pg/m³ -range in outdoor air. Volatile biotransformation products such as PFBA and PFHxA have been observed in biogas [60]. Based on data collected from eight biogas plants [61], PFOS was the principal compound present in digestates, typically in the range of 1 - 170 µg/kg dry material. Various FTOH were also observed to have concentrations ranging between 8.3 and 16 pg/m³ in air, as the sum of volatiles and particulates.

One potential concern for biogas production from purpose-built waste-to-energy landfills is the co-production and concentration of the more volatile PFAS and precursors. Various studies have investigated the impact of PFAS on the production of biogas, with diverging observations. For example, methane production was observed to be enhanced in the presence of PFOS [62], and results suggested that PFOS may act as metabolic decouplers. Conversely, PFAS-containing AFFFs have been reported to inhibit both methane production and co-contaminant degradation in an anaerobic microbial community [63]. This remains an area of intense research activity because the potential presence of PFAS volatiles in biogas raises concerns regarding the further spread of PFAS via atmospheric routes [64]. If PFAS compounds are not destroyed during the utilisation of biogas, then they pose a long-term and more widespread environmental exposure threat than previously considered. Research should also focus on minimising PFAS exposures from biogas production through improved separations [60].

3. Modern Landfill Barriers - Can They Cope with PFAS?

The purpose of engineered hydraulic barriers in many modern lining systems is to provide multiple levels of protection, including composite liners composed of a geomembrane (GMB) combined with a geosynthetic clay liner (GCL) or a compacted clay liner (CCL) as shown in Figure 2. This approach is generally adopted in the event of failure of the geomembrane, to reduce the hydraulic conductivity of fluids and to limit (retard or attenuate) discharge into the surrounding environment. The presence of a CCL or GCL below the geomembrane GMB will hinder both the advective transport of liquids and diffusion of ions and gases due to their low permeability.

Figure 2 Sectional views of modern composite hydraulic barrier systems used in landfills where a geomembrane lies directly on top of a geosynthetic clay liner (a and b) or above a compacted clay liner (c). Other options (not shown) exist including double liner systems, where the geomembrane primary liner is separated from the geosynthetic clay liner or compacted clay liner secondary liner by a drainage layer with a leachate collection system.

3.1 Compacted Clay Liners

Due to the paucity of available data, interactions of PFAS compounds with engineered CCL must be primarily inferred from interactions with soil materials augmented by findings from specific studies. CCL are generally expected to operate as effective hydraulic barriers under a wide range of field moisture and temperature conditions. In Australia, and the State of Victoria, in particular, the use of CCL as primary capping liners is being mostly phased out for municipal landfills. However, they are still widely used in landfills, mining (as tailings dams), agriculture (stock dams), and urban (aesthetic ponds and stormwater management) settings as both primary and secondary base and side wall liners. As discussed above, the presence of PFAS compounds in each of these environments cannot be neglected.

Critical properties of CCL (or any soil material) expected to influence the fate and behaviour of PFAS compounds include the following:

- i. Mineralogy, or the composition of the mineral phases present in the CCL,
- ii. Hydration (moisture content) status, and
- iii. Thickness, density, and porosity of the compacted material,

These properties affect the chemical compatibility of the CCL with any particular leachate and have a direct impact on specific surface properties, such as reactive surface area, surface charge and surface acidity, which alter the retention of PFAS within the CCL.

3.1.1 Mineralogy

The essential mineral phases present in the CCL are the clay minerals (kaolinite, montmorillonite, etc.) and clays (e.g. allophane, imogolite, etc.), due to their small particle size (i.e. clay-sized, < 2 μm), high available surface area and reactivity with constituents in the gaseous and aqueous phases. As a 'master variable', the mineralogy of a CCL will influence nearly all physical, chemical and geotechnical properties of the barrier itself [65]. Other mineral phases may prove to be more highly effective for the retention of PFAS, including those with positively charged surfaces, for example, layer double hydroxides, or pH-dependent layer charges such as allophane or hydroxyoxides of iron and aluminium. CCL enriched with these mineral phases will express different surface properties than a CCL enriched in montmorillonite, with a usually high and (mostly) pH-independent negative layer charge.

As a natural material sourced from predominantly local areas, CCL material is inherently inhomogeneous, because it is often a blend of soils. Thus, the exact mineralogy will differ for each CCL, and within a given CCL, differences in mineralogy will exist that will impact on its functionality. Some CCL may have elevated or heterogeneous levels of associated organic matter, even in the same liner system. High concentrations of organic material in CCL would be expected to increase bulk partitioning (K_d) or soil partitioning (K_{soil}) coefficients [15, 16, 18, 19, 66] through increased retention by the organic matter and thus could retard PFAS compound migration.

Recent studies [67-70] have highlighted the importance of organic carbon on adsorption and retardation of PFAS through clay soils. Soil clay content was observed to be important in retarding PFAS transfer in high permeability soils where air-water interfacial partitioning dominated PFAS behaviour [23].

3.1.2 Hydration Status

It is well known that clay materials undergo swelling and shrinkage when hydrating or desiccating, respectively. For some clay materials - particularly those mostly composed of montmorillonite - that experience extensive swelling, the porosity and density will change commensurate with hydration status. Some CCL express this property more so than others, governed by their mineralogy. As a porous material, CCL will have proportionate changes in the amount of water- or air-filled pores as a function of water content, which can be quantified by their water retention characteristics.

Volatile PFAS compounds can be expected to partition to, and migrate more readily in, the gaseous phase at lower water contents than at higher water contents. The heat generated in landfills [71] can result in elevated gaseous flux through even robust bentonite liners [72]. PFAS with increased water solubility (e.g., PFNA, PFBA, PFDA, PFBS, and some precursor FTCAs and FTCSSs) will more likely partition into the pore waters of wetter clays and thus be more susceptible to hydraulic transport via advection and diffusion than less soluble PFAS (e.g., PFHxA, PFHxS, PFOA, PFHpA and PFOS). The ability of the CCL to maintain a low saturated hydraulic conductivity (k_{sat}) would be expected to at least retard soluble PFAS transfer by enabling time for sorption to mineral surfaces to occur (if this will happen) and thus potentially causing the partitioning of some PFAS from the liquid phase.

These same PFAS may also partition to the clay surfaces in drier conditions, and thus their overall transport could be retarded. It must be stressed, however, that the available evidence for existing landfills in Australia [32, 33, 49-51] does not indicate PFAS migration through CCL base liners. However, PFAS migration from unlined landfill was reported to occur within a few years [46-48]. Thus, a significant gap exists in understanding PFAS migration through CCL.

3.1.3 Thickness, Density, Porosity

Most jurisdictions follow national and international specifications covering minimum compacted liner thickness and optimum moisture content and compaction density (thus porosity) required for adequate performance of CCL under general conditions [73]. However, CCL can be notoriously heterogeneous with respect to density and porosity [74, 75].

Any porous material in a wet condition appears to be highly amenable for penetration by PFAS. This is the case, for example, with the known penetration of PFAS compounds into concrete materials associated with firefighting training grounds with AFFFs [43]. A higher density (lower porosity) CCL would be expected to, at the least, impede the transport of PFAS, but be unlikely to halt that transport over long periods. Thus, low porosity and small pore size do not preclude their transmission (all else considered equal). As indicated above, in the dry condition, the same material may allow the more volatile PFAS compounds to migrate in gaseous form. Shorter chain PFAS like PFNA, PFBA, PFDA or PFBS can be expected to penetrate more effectively than longer chain PFAS like PFHxA, PFHxS, PFOA, PFHpA or PFOS. Limited experimental or field evidence that exists supports this conclusion, pointing to relatively high transmission rates [15, 21, 76]. As will be detailed in Section 3.2.4, under standard test conditions for the saturated hydraulic conductivity of geosynthetic clay liners, many of these PFAS compounds do breakthrough under environmental concentrations.

Assuming a well-designed and constructed CCL with an ideal k_{sat} of 1.0×10^{-9} m/s for water [73], and assuming PFAS compounds will not diffuse or advect faster than this rate, then ≈ 31 mm of wetting front will pass through the liner per year providing about ≈ 15.8 years for migration through 500 mm depth. While sorption to clay and organic carbon surfaces may retain and retard the transfer of a portion of the PFAS, the capacity may be limited, particularly given the inhomogeneity in the CCL as illustrated in Figure 3. Thus k_{sat} , and the resulting migration rates of non-retarded PFAS could be as much as 100-times greater (10^{-7} m/s), taking just under two months to progress through 500 mm depth. Natural soils and sediments have much higher k_{sat} (5×10^{-5} for a typical fine sand), and thus would penetrate this same distance in a matter of days, or even hours.

3.1.4 Surface Area, Surface Charge, Acidity

The high reactive surface area of clays - that is, the proportion of surface available for interaction with contaminants in air or water - is a major (albeit largely unknown until the modern era [77]) reason CCL have been used for many centuries as hydraulic barriers. The solubility of any given PFAS will control solid-phase partitioning with the various minerals present in the CCL, provided inhibiting processes such as electrostatic repulsion or surface hydration can be overcome.

The charge of particles - negative for most clays and clay minerals in natural environments - expressed at the surface influences how closely dissolved solutes can approach and undergo sorption, transformation, or degradation reactions. Variable (pH-dependent) charge minerals such as kaolin, allophane or the oxyhydroxides of iron could partake in direct electrostatic attraction with PFAS anions if the pH of the leachate is below the point of zero charge (PZC) of the mineral surface and above the acid dissociation value (pK_a) for the given PFAS. Theoretically, such a condition could be expected for most PFAS in acidic ($pH < 4.5$) conditions, where other variable charge minerals, like kaolinite, will begin to express a significant proportion of their surface charge as positive. At more typical pH conditions, however, these reactions may be restricted to specific mineral phases, such as the oxyhydroxides goethite (PZC at $pH \approx 7$), ferrihydrite (PZC at $pH \approx 8.5$) or possibly allophane (PZC at $pH \approx 6$), the latter of which is relatively uncommon in Australia.

Very little mineral-specific information on PFAS partitioning exists, and instead, most of the information is related to determinations of bulk K_d or K_{soil} , or bulk retardation, R_d [67, 68, 73, 77-82]. Mineral surface charge (e.g., zeta potential) and specific surface areas appear to be the main factors impacting PFAS retention [83] with less negative zeta potential and larger specific surface areas promoting sorption of PFOA. PFOA, PFOS PFHxA and PFNA were all shown to weakly and reversibly adsorb to positively charged alumina (Al_2O_3), with the extent of adsorption being inversely proportional to solubility [84]. Significantly, the same PFAS were observed to not adsorb to negatively charged silica (SiO_2). Short-chain PFAS are little affected by rate-limiting sorption in soils with low organic carbon contents [85], and thus bulk transfer was essentially unaffected by mineralogy. Only where the organic carbon fraction was around 4.5% was PFAS transport found to be largely dependent on heterogeneous subsoil mineralogy. It needs to be emphasised that the unique interface-attracted behaviour of PFAS compounds will enable them to interact with mineral phases even when conditions would be suspected to inhibit such interactions [7, 31, 47, 81]. The interaction may be both favourable and unfavourable in terms of transmission of the PFAS compounds.

3.2 Composite Liner Systems

Modern base and side-wall liner systems designed to contain MSW often incorporate composite liner systems, consisting of a polymeric GMB overlying a GCL or a CCL (Figure 2). A GCL is a manufactured liner consisting of a thin layer of bentonite (5-10 mm) contained between two polymeric geotextiles through needle punching [75]. In Australia alone, an estimated 6 Mm² of GMB and GCL are deployed each year in composite lining systems, and their use is expected to increase over the next few decades. Not all components in composite liners, as depicted in Figure 2, are used, because landfill design philosophy (regulation-driven versus performance-driven) differs between jurisdictions.

GCL are an increasingly important component in composite lining systems in MSW landfills [75, 86, 87], typically in the base and side-wall liners or in capping systems. In these situations, liner components are subjected to a myriad of conditions often not considered in the landfill design [88]. In base liner systems, liner components may be exposed to long-term elevated temperature and/or hydration conditions coupled with exposure to harsh permeants from underlying human-made or natural formations under considerable confining stresses [71, 89]. Where composite systems are deployed as side-wall barriers, components are often exposed to the elements for months under low confining stress conditions during which they undergo diurnal thermal cycling that only eases with landfilling which increases confinement [90].

In capping systems, which in general provide long-term very low confining stress conditions, components can be exposed to extreme weather conditions that cause inundation or desiccation, or exposure to inputs from surrounding human-made or natural environments [91].

In each of these uses, a given GCL is expected to perform to required specifications for many decades, although inappropriate installation or inadequate design considerations can significantly reduce their lifespan. In some jurisdictions and for some applications, GCL is the primary environmental liner deployed. Increasingly, GCL are installed in conditions outside of the scope for which they were designed, and are currently marketed, for example, in energy exploration operations where temporary storage of saline pump water is required [87, 88], or where they are exposed to extremes of wind, rain, cold, solar radiation, salinity and acidity or alkalinity [87-109] for considerable periods.

Since PFAS compounds can pose undesired known (as well as unexpected or unknown) risks to liner system effectiveness, and given their presence in landfill leachates [49, 50, 59] and known breakdown mechanisms [110], it can be expected that PFAS concentrations in landfill leachates and emissions will be a significant issue for many decades to come. Given that PFOA, PFOS and other PFAS have been identified in Australian (e.g. [51]) landfill leachates at concentrations exceeding the lifetime drinking water health advisory levels (0.07 and 0.56 µg/L, respectively) reported in the National Environmental Management Plan [111], a critical need exists to assess the performance of modern landfill lining systems with respect to minimising environmental exposure of PFAS compounds.

3.2.1 Geosynthetic Clay Liners

Geosynthetic clay liners (GCL) are a manufactured composite material of bentonite contained between a cover and a carrier geotextile through needle-punching and are often thermally treated

[75]. The geotextiles (GTX, discussed further below) are either nonwoven or woven, and combinations are often used (e.g. nonwoven scrim-reinforced woven geotextile). Most GTX used in GCL are made from various grades of polypropylene (PP), but polyesters (PE) are also used. During manufacture, generally 4 - 5 kg/m² of sodium bentonite, in either powder or granular form, is spread to a thickness of 6 - 10 mm between the two geotextiles. Due to different GTX available and whether stitching, needle-punching or adhesion bonding of the components is employed, GCL have a k_{sat} between 2×10^{-12} to 2×10^{-10} m/s depending on their void ratio [75].

Much regarding the expected interactions of PFAS with GCL can be inferred from known interactions with CCL (discussed above in Section 3.1) and with polymer materials (discussed below). Three main PFAS-dependent mechanisms can be considered strongly associated with their fate and behaviour in landfills deploying GCL as part of composite barrier systems: partitioning, including sorption; transformation, including biotic and abiotic processes; and volatilisation. The following sections synthesise the available information on expected interactions of PFAS compounds with components in GCL.

3.2.2 Bentonite

The main component responsible for the impervious nature of GCL is sodium bentonite; thus, factors that impact on bentonite performance with respect to known contaminants will likely also influence their performance as hydraulic barriers to the migration of PFAS compounds. Much of the information assessed for CCL (above) is also relevant for GCL, with the caveat that most bentonites used in GCL are mainly composed of sodium montmorillonite [74]. Due to the relatively strict standards associated with GCL manufacture, they contain a somewhat limited range for minimum montmorillonite content, and also a limited range of surface or layer charge (related to the cation exchange capacity), which in general optimises the swelling in low ionic strength water.

Suitable sodium bentonites for GCL manufacture are unique as a natural material in that a given source deposit generally has relatively consistent attributes, such as predictable cation and metals retention, large surface area, relatively high liquid adsorption capacity, and when allowed to swell under confinement, have very low hydraulic conductivity to water and other low ionic-strength (generally equivalent to < 0.1 M) permeants. Despite these beneficial properties, each bentonite has different compositional mineralogy and chemistry, and thus expresses its macroscopic behaviour - the uptake of water, swelling, inhibition of advective and diffusive transport of aqueous leachates etc. - differently. Strict standards and guidelines for these properties apply for bentonite. For example, Na-Mt content needs to be at least 70 wt.%, swell index > 24 mL/2g and k_{sat} should be $< 1 \times 10^{-11}$ m/s, corresponding to $< \approx 1.3$ mm/year (refer to Figure 3).

Figure 3 Calculated transport times (in years) for water over a range of k_{sat} values through clay-based liners of varying thicknesses. Line labels indicate the thickness of the CCL or GCL layer (mm). Shaded regions denote the expected k_{sat} range for CCL and GCL. A hydraulic gradient to 150 was assumed in the calculations.

The sodium bentonite component of the GCL must perform over a wide range of field conditions, but the hydration of sodium bentonite used in GCL in Australia has been the focus of intense local [72, 88, 89, 92-94, 96-101, 112-120] and international [65, 87, 90, 104, 106, 108, 109, 121-125] research. It is well established that pre-hydration and maintenance of a high hydration state of the sodium bentonite are necessary for optimal attenuation of both liquid and gaseous diffusion [72, 112, 126] and therefore the presence of an effective barrier against PFAS compounds (Figure 4). A variety of polymer-enhanced sodium bentonites are currently available on the worldwide market, including in Australia [127-130]. While they have been promoted for their ability to enhance swelling and to provide some resistance to increased ionic strength, there remains some possibility that those polymers of low ionicity, or even the non-ionic forms, could additionally be useful for enhancing partitioning of PFAS compounds to the bentonite.

Figure 4 Gas permeability of a granular GCL as a function of hydration under two nominal applied stresses associated with cover and side-slope liner systems [112].

The amount of confinement to which the GCL is subjected while hydrated (or hydrating) is an important consideration (Figure 4), especially when used as a cover where it is expected to have low permeability to gaseous emissions. Lower gas permeability is generally observed for a given water content (above a threshold value ≈ 0.3 vol.% or $\approx 100\%$ gravimetric water content, GWC) with increasing confinement. However, even high water content cannot provide acceptably low gas permeability if the GCL is under very low confining stress [112]. The amount of hydration is also dependent on the form of the bentonite in the GCL, with granular bentonite usually requiring a higher water content (and more time to equilibrate at that water content) than powder bentonite [112, 126, 131]. Hydration to a minimum of 100% GWC decreases the gaseous diffusion coefficient k_{gas} to $< 10^{-9}$ m²/s (Figure 5). However, a GWC of about $\approx 160\%$ (or about 80% of saturation) is required to cause gas to be fully limited by diffusive (instead of advective) transport. These diffusivities equate to a 5 order-of-magnitude change in gas permeability rate (from 10^{-7} m/s for < 100 wt.% GWC to 10^{-13} m/s at 160 wt.% GWC) [112, 126].

Figure 5 Gas diffusion (a) and permeability (b) as a function of GWC (wt.%) and degree of saturation (Sr%) for a granular GCL under a nominal 20 kPa applied stress associated with a cover liner system. In (b), a powder GCL is shown for comparison [126].

One significant factor that affects the overall performance of bentonite in GCL, whether deployed as cover, side-wall or base liners, is its subsequent desiccation, mainly when deployed on side-slopes or in capping liners where overburden confinement is limited (Table 3). Desiccation induced cracking can cause significant increased over-all gaseous flux in cap liners as well as increased leaching until cracks have re-closed on rehydration. While sodium bentonites can “self-heal” during re-hydration of desiccation cracks, cyclical desiccation and re-hydration will eventually result in increases in leachate flux through the GCL [105, 107]. When accompanied by the exchange of calcium (Ca^{2+}) for sodium (Na^{+}) ions, cyclical desiccation will lead to severe losses of hydraulic barrier function in bentonite in high rainfall areas when used in capping scenarios [91, 132, 133]. If the bentonite is hydrated to >150 wt.% GWC and remains hydrated during cation exchange reactions, little change in the overall k_{sat} may occur, mainly where adequate vertical stress (confining) conditions exist, as in a base liner. For base liners, confinement stress can be considerably larger compared to e.g., side-wall liners; stresses in both situations change with time during filling of the landfill.

Table 3 Factors impacting bentonite performance in GCL deployed in capping, side-wall and base liner systems.

Hydration (GWC)	Condition of bentonite in GCL		
	Ca ²⁺ for Na ⁺ exchange No desiccation	No exchange Cyclical desiccation	Ca ²⁺ for Na ⁺ exchange Cyclical desiccation
Capping			
< 80%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑	k _{gas} or D _{gas} : ↑
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{major}	k _{liquid} : ↑ _{major}
	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely
> 80% <150%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑	k _{gas} or D _{gas} : ↑
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑	k _{liquid} : ↑
	Loss _{bentonite} : unlikely	Loss _{bentonite} : possible	Loss _{bentonite} : possible
> 150%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑	k _{gas} or D _{gas} : ↑
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{minor}
	Loss _{bentonite} : possible	Loss _{bentonite} : possible	Loss _{bentonite} : possible
Side-wall liners			
< 80%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{major}	k _{liquid} : ↑ _{major}
	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely	Loss _{bentonite} : possible
> 80% <150%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : -
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑	k _{liquid} : ↑
	Loss _{bentonite} : unlikely	Loss _{bentonite} : possible	Loss _{bentonite} : possible
> 150%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : -
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{minor}
	Loss _{bentonite} : unlikely	Loss _{bentonite} : possible	Loss _{bentonite} : possible
Base liners			
< 80%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑	k _{gas} or D _{gas} : ↑
	k _{liquid} : ↑	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{minor}
	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely
> 80% <150%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑	k _{gas} or D _{gas} : ↑
	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑
	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely	Loss _{bentonite} : possible
> 150%	k _{gas} or D _{gas} : -	k _{gas} or D _{gas} : ↑	k _{gas} or D _{gas} : ↑
	k _{liquid} : -	k _{liquid} : ↑ _{minor}	k _{liquid} : ↑
	Loss _{bentonite} : unlikely	Loss _{bentonite} : unlikely	Loss _{bentonite} : possible
Neutral change: - ↑: increase, GWC: gravimetric water content			

Possible interactions of water-soluble and volatile PFAS with a base liner GCL undergoing no desiccation are provided in Tables 4 and Tables 5 with a base liner GCL undergoing cyclic hydration and desiccation with exchange histories depicted in Table 3. The likelihood of various partitioning reactions of water-soluble and volatile PFAS compounds are provided under the same conditions described for Table 3. The likelihoods are colour-coded to indicate probable negative impact (red),

the potential for negative impact (yellow), and improbable negative impact potential (green). It must be stressed that these possible ageing scenarios are mostly untested, and the impact on GCL performance as a base liner is presented in general terms only and are thus hypothetical likelihoods based on what current literature indicates.

Table 4 Possible interactions of water soluble and volatile PFAS compounds with GCL undergoing no desiccation. Colour coding denotes relative benefit to retarding PFAS transport.

Constant hydration state		Condition of bentonite in GCL					
		< 80% GWC		> 80% < 150% GWC		> 150% GWC	
		<i>H₂O</i> soluble	Volatile	<i>H₂O</i> soluble	Volatile	<i>H₂O</i> soluble	Volatile
No exchange	air→solid	Possible	Improbable	Possible	Improbable	Improbable	Improbable
	air→liquid	Possible	Improbable	Probable	Possible	Probable	Possible
	liquid→air	Improbable	Probable	Improbable	Probable	Improbable	Possible
	liquid→solid	Possible	Improbable	Possible	Possible	Possible	Improbable
	solid→air	Improbable	Probable	Improbable	Possible	Improbable	Possible
	solid→liquid	Possible	Improbable	Probable	Improbable	Probable	Possible
Ca ²⁺ for Na ⁺ exchange	air→solid	Possible	Improbable	Possible	Improbable	Improbable	Improbable
	air→liquid	Probable	Improbable	Probable	Possible	Probable	Possible
	liquid→air	Improbable	Possible	Improbable	Probable	Improbable	Possible
	liquid→solid	Possible	Possible	Possible	Possible	Possible	Possible
	solid→air	Improbable	Probable	Improbable	Probable	Improbable	Possible
	solid→liquid	Possible	Improbable	Probable	Possible	Probable	Possible

H₂O soluble: most PFCAs, PFSAs, FTSAs, FTCAs

Volatile: PFBA, PFBS, PFPeA, PFHxA, 4:2, 6:2 FTOH, other volatile PFAS replacement precursors

Table 5 Possible interactions of water soluble and volatile PFAS compounds with GCL undergoing cyclic hydration with the exchange histories depicted in Table 3. Colour coding denotes relative benefit to retarding PFAS transport.

Cyclic hydration		Condition of bentonite in GCL					
		< 80% GWC		> 80% < 150% GWC		> 150% GWC	
		<i>H₂O soluble</i>	<i>Volatile</i>	<i>H₂O soluble</i>	<i>Volatile</i>	<i>H₂O soluble</i>	<i>Volatile</i>
No exchange	air→solid	Probable	Possible	Probable	Possible	Improbable	Improbable
	air→liquid	Possible	Possible	Probable	Possible	Possible	Possible
	liquid→air	Improbable	Possible	Improbable	Probable	Improbable	Probable
	liquid→solid	Possible	Possible	Possible	Possible	Possible	Possible
	solid→air	Improbable	Probable	Improbable	Probable	Improbable	Probable
	solid→liquid	Possible	Improbable	Probable	Possible	Probable	Possible
Ca ²⁺ for Na ⁺ exchange	air→solid	Probable	Possible	Probable	Possible	Improbable	Improbable
	air→liquid	Possible	Possible	Probable	Possible	Possible	Possible
	liquid→air	Improbable	Possible	Improbable	Probable	Improbable	Probable
	liquid→solid	Possible	Possible	Possible	Possible	Possible	Possible
	solid→air	Improbable	Probable	Improbable	Probable	Improbable	Probable
	solid→liquid	Possible	Improbable	Probable	Possible	Probable	Possible

H₂O soluble: most PFCAs, PFSA, FTSAs, FTCAs

Volatile: PFBA, PFBS, PFPeA, PFHxA, 4:2 and 6:2 FTOH, other volatile PFAS replacement precursors

For interpretation of these tables note that both 'Probable' and 'Improbable' tags may be both red and green in the same column. For example, for water-soluble PFAS, 'Probable' liquid → solid partitioning would be desirable (green), as it would induce retardation or partitioning away from the leachate in which it is more mobile. For volatile PFAS compounds, 'Probable' air → solid partitioning would be desirable because it retards the movement in the primary medium, but 'Improbable' air → water partitioning could be considered unfavourable as it provides an alternative route for transmission. The various partitions considered are air → solid and air → liquid interfaces, liquid → air and liquid → solid interfaces, and solid → air and solid → liquid interfaces.

Four scenarios are considered in terms of the known impact they would have on the bentonite as a hydraulic barrier and its expected effect on the fate of water-soluble and volatile PFAS. These are:

- i Hydration of the bentonite to three different water contents and no desiccation.
- ii Hydration to three different water contents with Ca^{2+} for Na^+ exchange occurring and no desiccation.
- iii Cyclical hydration to three different water contents followed by desiccation.
- iv Cyclical hydration to three different water contents with Ca^{2+} for Na^+ exchange occurring followed by desiccation.

The scenarios represent increasing changes to the microstructure and/or the development of desiccation cracking, which are known to deteriorate the performance of bentonites in GCL deployed within cover systems [91, 105, 107, 132]. The level of functional impairment of the bentonite as a barrier to fluids generally increases from (i) to (iv) above, and the issues are amplified under low confinement conditions, as found in capping and side-wall liners. However, exactly how these levels of impairment may influence GCL performance with respect to PFAS is practically unknown. Thus, any predictions are hypothetical and currently largely unsupported by experimental evidence.

The low pH of aqueous solutions interacting with bentonite can have a similar effect to that of Ca^{2+} for Na^+ exchange, and results in altered permeability of the bentonite [88]. Conversely, high pH conditions would unlikely be detrimental in most lining situations, unless accompanied by elevated salinity, under which circumstances the permeability would increase.

3.2.3 Preliminary Results for Performance of GCL to PFAS

At the time of writing, only two research papers have studied the interaction of PFAS leachates with sodium bentonite [59, 134]. The study reported in [134] observed that aqueous phase PFAS compounds were only partially retained by fully hydrated compacted mixtures of sand-sodium bentonite in batch testing. Column testing showed that the spiked concentrations of PFAS had no impact on the hydraulic conductivity of the barrier. However, the ionic strength of landfill leachate caused a mild increase in k_{sat} . Thus, while the interactions and fate of PFAS with GCL remain mostly unknown, this solitary study highlights the limitations of relying upon mineral-based barrier systems alone.

Preliminary laboratory trials [59] indicated that standard GCL, containing sodium bentonite, had minimal impact on most PFAS from landfill leachate, at least up to 6 pore volumes of flow (PVF)¹, or 50 days of permeation under standard laboratory testing conditions². The standard GCL, containing $\approx 4 \text{ kg/m}^2$ bentonite, returned k_{sat} values of $2.8 \times 10^{-11} \text{ m/s}$ in deionised water, but $5.9 \times 10^{-11} \text{ m/s}$ in the landfill leachate (containing a total of about $35 \text{ }\mu\text{g/L}$ total PFAS) after 50 days (Figure 6). A higher grade GCL (termed pH+) had k_{sat} values of 4.2×10^{-11} after 5 PVF (50 days) permeation. Another GCL

¹ A pore volume of flow is the volume of liquid that saturates the pore spaces of the GCL and is thus inversely related to the GCL porosity. For this calculation, a hydrated density of 1.9 g/cm^3 was used for the GCL, equating to a hydrated pore volume of $\approx 45 \text{ cm}^3$ in the test specimen.

² Hydraulic conductivity tests were conducted on non-prehydrated thermally-locked, needle punched GCL samples (100 mm dia. disks). Paired GCL samples, with and without activated carbon powder intermixed with the bentonite, were subjected to either deionised water or landfill leachate as the hydrating and permeating medium. Testing was conducted following both scenarios outlined in ASTM D6766 [135].

product, modified with activated carbon powder (AC GCL in Figure 6) returned k_{sat} of 6.6×10^{-11} m/s in deionised water after 6 PVF and 50 days permeation. When permeated with landfill leachate containing about 35 $\mu\text{g/L}$ total PFAS, the k_{sat} for this same product has stabilised about 40% higher near 1.0×10^{-10} m/s over the last 15 PVF (100 days).

Figure 6 Saturated hydraulic conductivity as a function of (A) test duration and (B) pore volumes of flow of PFAS-containing landfill leachate in GCL specimens. A total of 272 days was required to attain 30 pore volumes of flow. Lines indicate best logarithmic fits to the data.

While the addition of the activated carbon, impacts moderately on the hydraulic performance of the GCL, it does have an effect on the PFAS concentrations permeating through [59]. PFAS concentrations in the effluent (after passing through the GCL) were quantified periodically at a NATA certified laboratory³. The latest results for the AC GCL to 30 PVF are depicted in Figure 7. Except for one measurement (10.9 PVF), total PFAS dropped exponentially from an initial concentration of ≈ 35 $\mu\text{g/L}$ to the limit of reporting (LOR) within 13 PVF. Total PFAS increased to 25.5 $\mu\text{g/L}$ in the effluent by ≈ 30 PVF (average of nearly 15 $\mu\text{g/L}$ above 16.5 PVF). These results highlight the need to conduct such trials for sufficiently long times to attain more than 20 PVF.

³ A suite of 27 PFAS compounds were quantified by HPLC-MS at ALS Environmental Testing (Brisbane, Australia). ¹³C-labelled PFOS (at C4) and PFOA (at C8) served as internal standards during analysis.

Figure 7 Landfill leachate PFAS concentrations in effluent after permeation through a modified GCL product containing powdered activated carbon as an additive. Initial PFAS concentrations are depicted at the “0” pore volume. The limit of reporting (LOR) is 100 ng/L (0.1 µg/L).

The total PFAS measured in both the initial landfill leachate and in the effluent after passing through the activated carbon modified GCL, was dominated by two short-chained PFAS analytes, PFBA and PFBS, both of which behaved classically relating to exceeding adsorptive capacity of the reactive medium. PFBS concentrations initially decreased exponentially to LOR within 5.3 PVF, but then increased exponentially in concentration from 21.6 PVF. PFBA concentrations also decreased exponentially to LOR by 5.2 PVF but revealed a more sporadic albeit higher concentration from 16.5 PVF. Significantly for PFBA, the concentrations in the effluent from 16.5 PVF onward exceeded the initial PFBA concentrations in the landfill leachate, indicating that perhaps some degradation reactions have occurred or, more likely, that PFBA that was attenuated during initial penetration has broken through.

PFHxA, PFPeA and PFHxA largely dropped to LOR (an exception being a single anomalous result for PFHxS after 5.2 PVF, then decreasing to its LOR up to 30 PVF). PFPeA evidenced breakthrough by 16.4 PVF and increased to over its initial values by 28 PVF, returning to around a third of its initial value by 30 PVF. Levels of PFOA, PFOS, were below the LOR of 0.1 µg/L up to 30 PVF except for a single anomalous result for PFOS at 5.2 PVF, then also returning to LOR up to 30 PVF. Concentrations of the three currently regulated PFAS (PFOS, PFOA and PFHxS) were reduced to LOR (Figure 8) with the AC GCL, but only PFOS was reduced to LOR by standard and pH+ GCL products. Given that these three PFAS largely remain at LOR after 30 PVF, the AC GCL appears to provide a good degree of resistance to their migration. Note, however, that due to the method of direct injection and single dilutions conducted at many commercial labs, the LOR does change with time, particularly for PFOS and PFOA. In addition, LOR also depends upon instrumentation used. Nonetheless, these ongoing results highlight the fact that a few of the shorter carbon-length PFAS - more commonly present in modern landfill leachate - are highly mobile.

Figure 8 Residual concentrations of PFOS, PFOA and PFHxS after permeation of 2 pore volumes of flow in different GCL products. The limit of reporting (LOR) is 100 ng/L (0.1 µg/L).

Further, while the preliminary results perhaps unsurprisingly, show that a GCL modified with activated carbon may only provide temporary respite against some shorter carbon-chain length PFAS depending on the level of exposure (e.g., volume, hydraulic head and initial concentration) to the liquid, they do indicate that significant retention can occur, particularly for the regulated PFAS (PFOA, PFOS and PFHxS). If built to Best Practice Environmental Management type 2 guidelines [64] in which landfill seepage rates are < 10 L/ha/day, then the retention of PFBA, PFBS, PFHxA and PFPeA below LOR by the AC GCL is equivalent to at least 250 years attenuation, while that for PFOA, PFOS and PFHxS is equivalent to at least 450 years.

Of concern is the rather high temporal variability in the PFAS analyses. For example, PFOS was measured as high as 4000 ng/L at 5.2 pore volumes, more than 200-times its concentration in the original leachate. Hypothetically, this could be a result of analyte concentrating associated with its breakthrough when sorptive capacity within the modified GCL is attained. However, this is unlikely given the (i) number of PVF and estimated saturated pore volume of the specimen, (ii) return in measured values to LOR and (iii) variability in total PFAS analysis between 7.7 and 13 PVF. Instead, this is most likely associated with an as-yet unidentified experimental error, particularly with the samples taken at 5.3 PVF. Further work on this and other systems is required to fully understand the efficacy by which GCL-based liner components can mitigate PFAS mobility.

3.3 Geotextiles and Geomembranes

3.3.1 Geotextiles

Geotextiles (GTX) are used in several parts of composite liner systems in modern landfills (see Figure 2) and can be made from both polymeric fibres as well as natural fibres. GTX are used in GCL to hold the bentonite component in place and are either woven, non-woven or a mixture, and are often reinforced by needle-punching. GTX are also essential components of modern composite lining systems due to their unique properties, functioning as protective layers (specifically to protect

the geomembrane from overlaying drainage aggregates and punctures during installation), and for filtration above drainage layers (to minimise silting) and for stabilising side-slopes.

Essentially no data exists on how GTX components may impact the fate and behaviour of PFAS compounds. Thus, in the absence of experimental evidence, any predictions are largely speculative at this time. Nonetheless, some inference can be made from studies on the adsorption of poorly miscible (very low solubility) volatile organic (hydro)carbons (VOCs) by the GTX components of GCL [108, 136]. For example, the woven and non-woven GTXs taken from different GCL were found to adsorb up to 0.6 µg/kg of ortho-chlorine substituted cresol and 5 µg/kg of bisphenol A [136]. For the several chlorinated phenolic compounds studied (di- and tri-chlorophenols), sorption onto the GTX increased with increasing amounts of Cl in the phenol, with greater sorption into non-woven than woven GTXs. Thus, non-polar, poorly miscible VOC contaminants interact with the polymer components of GTX, and these interactions can be both favourable and unfavourable. For example, the ability of PFAS compounds to sorb to PP may enhance retardation of their transmission, but may also remove antioxidants from the GTX, thereby diminishing the overall longevity of the GCL.

One other consideration with respect to GTX in GCL is that the needle-punched fibre bundles, while serving to hold the GCL together, can act as conduits for hydration of the bentonite within the interior of the GCL [99]. The bundles are made up of several individual polymer threads, and through the displacement of the bentonite during needle punching, impart a greater local porosity. It follows that water-soluble PFAS would exploit these regions through GCL. Further research is required to substantiate whether this is the case and to determine means to minimise the effect if real.

3.3.2 Geomembranes

Geomembranes (GMBs) are thin (2-5 mm thick) continuous polymeric sheets that are now considered the main functional primary liner for MSW landfills. They feature extensively in composite liner systems worldwide due to their high degree of chemical resistance to many solvents, and resistance to thermal and oxidative degradation. The most common polymers used in the production of GMBs are high-density polyethylene (HDPE), but low-density and linear low-density polyethylene (LDPE and LLDPE), polyvinyl chloride (PVC) and PP, among many others are also used.

Because polyethylene has limited functionality to nonpolar volatile organic compounds (VOCs), coextruded GMBs have been recognised as better barriers to petrochemicals [137]. These are multi-layered GMBs which include different inner functional cores. The co-extruded cores have traditionally been polyamide (Nylon VPB15) [138], or ethylene vinyl alcohol (EVOH), both of which can substantially reduce the diffusive transmission of aqueous and gaseous phase nonpolar organic compounds, as well as have enhanced partitioning and retention with the core phase of the GMB. For example, permeation coefficients were observed [137] to be as low as 2×10^{-12} m²/s for poorly miscible aqueous phase benzene, toluene, ethylbenzene and xylenes (BTEXs) in EVOH coextruded GMB, which were $\approx$ 3-times lower than those of VPB15 coextruded GMB, $\approx$ 15-times lower than HDPE, but more than 100-times lower than that of LLDPE GMB. The coextruded GMB also showed a lower diffusion to the volatile fractions of these same BTEX compounds.

Many polymer- and GMB-specific factors are expected to influence the potential for PFAS interactions with, and migration through, GMBs [59]. These include the chemical composition, crystallinity and degree of cross-linking of the polymers that make up the GMB, along with the density, weight and thickness of the GMB, quantity and type of antioxidant and other additives (e.g.

inert fillers, colour agents, UV stabilisers) used, amongst many others [139]. Together, these factors impact the direction of the vapour and solvent transmission of the GMB. Many of the mechanical properties of GMBs (tensile strength, tear, impact and puncture resistance, interface shear and anchorage strengths) influence the physical durability of the GMB. In contrast, stress cracking (pre- and post-exposure) directly influences the propensity for fluids (gas and liquid) to penetrate with age. As for GTX, the interaction of many organic species can also remove antioxidants for the GMB, and thus shorten its effective operational life.

HDPE is a mixture of crystalline (typically 60%) and amorphous polymer oligomers with less cross-linking than lower density versions (LDPE). Thus, it is generally stronger, harder and able to withstand higher temperatures (up to 100°C) without severe loss of functionality [140, 141]. The amount of crystallinity in an HDPE GMB is essential because it controls the relative degree of permeability of the polymer to non-polar substances [59]. The amorphous or poorly crystalline portions of the GMB are more mobile and thus can enable the penetration on non-polar chemicals more readily than in crystalline regions. Greater crystallinity decreases the free volume through which non-polar substances can penetrate.

Surface fluorination of the GMB reduces both diffusion and permeation of non-polar volatile hydrocarbons common in landfill leachates (PFAS compounds were not evaluated) by as much as 5-fold [142]. Reduction in free volume, but also decreased surface wettability and increased cross-linking are thought to be responsible for improved resistance of surface fluorinated GMB (SF-GMB) to non-polar molecules. The partitioning of the hydrocarbons to the GMB was essentially unaffected in this study [142]. SF-GMB also display increased oxidative induction time - a measure of the thermal stabilisation of the polymer within the GMB - so are thus more durable than traditional HDPE GMBs.

Other issues affecting the durability and functionality of HDPE GMB are related to installation. They include (i) punctures during placement, (ii) integrity of welds, and (iii) the length of time the GMB is exposed to solar radiation, which can lead to a depletion of antioxidants [139] and shifting and wrinkling of the GMB, resulting in poor interface with, for example, an underlying GCL [143]. Additive depletion (consumption of antioxidants), bond dissociation (breaking) are the leading chemical causes of embrittlement, and eventual GMB failure [140]. However, prior to physical failure under applied stresses, the GMBs are susceptible to crazing and cracking, and can become quite porous to gases and solvents [144]. Thicker GMBs are more resistant to these processes [145]. Excellent barrier integrity and long functional lifetimes can be expected if the installation process is conducted according to recommended guidelines and specifications.

The most recent results on PFAS in GMB [146] examined the diffusion of PFOA and PFOS through LLDPE and LLDPE-EVOH co-extruded GMB. However, care is required in interpretation of these results because the diffusion tests were conducted on very thin GMB (<0.8mm) which are not used - particularly LLDPE - in landfill liners. The initial concentrations used were 19.8 mg/L for PFOA and 22.7 mg/L for PFOS, but initial concentrations at ≈ 1.1 mg/L were also used to ensure no influence of hemi-micelle formation on the diffusion results. They reported estimated octanol-water partitioning (K_{ow}) coefficients for these materials were essentially controlled by the polyethylene and were 4.81 (LLDPE) and 4.49 (LLDPE-EVOH coextruded). Measured diffusion coefficients (D_g) for LLDPE ranged from $< 10 \times 10^{-16}$ m²/s (PFOA) to $< 6.7 \times 10^{-16}$ m²/s (PFOS). Diffusion was observed to increase with increasing temperature from 23°C to 50°C. Diffusion through LLDPE-EVOH co-extruded GMB was not reported (measurements are on-going), but estimated permeation (P_g) was

observed to be about an order of magnitude lower in the LLDPE-EVOH coextruded compared with LLDPE. Ranges for the same temperatures were: P_{gLLDPE} $9-19 \times 10^{-16} \text{ m}^2/\text{s}$ (PFOA), $19-52 \times 10^{-16} \text{ m}^2/\text{s}$ (PFOS); P_{gEVOH} $8.6-10 \times 10^{-16} \text{ m}^2/\text{s}$ (PFOA), $6.8-8.2 \times 10^{-16} \text{ m}^2/\text{s}$ (PFOS). Observed PFOA and PFOS concentrations in receptor leachates associated with LLDPE-EVOH co-extruded GMB were $< 1 \text{ ng/L}$ (PFOA) and $< 5 \text{ ng/L}$ (PFOS) after 500 days elution of leachate.

The current results for PFAS retention by GMBs [146] need to be viewed with some caution especially with regard to LLDPE GMBs as they are not used in landfill base or side-wall liners and the thicknesses considered (0.1 mm and 0.75 mm) are not relevant to practice. Also, LLDPE has a higher amorphous content than HDPE, and it is expected that an HDPE based GMB (typically 1.5 to 2mm thick) will have lower diffusion properties.

The potential for migration of PFAS compounds through HDPE GMBs will be related to the polymer-specific and GMB aging factors as described above, but also PFAS-specific factors, such as whether the substance is polar (i.e. pH above pKa), volatile, or water-soluble. Table 6 provides some untested and hypothetical performance expectations of GMB with regards to ageing. Actual performance would be subject to different factors in the field.

Table 7 and Table 8 project how possible ageing scenarios to different GMB types (traditional HDPE and coextruded) may impact their integrity as liners with respect to interaction with non-polar, polar, water-soluble and volatile PFAS. Exactly how these scenarios may influence GMB performance with respect to PFAS is unknown, and thus any predictions remain hypothetical in the absence of experimental evidence. Note that both 'Probable' and 'Improbable' tags may be both red and green in the same column. For example, for water-soluble PFAS compounds, 'Improbable' gaseous diffusion would be desirable (green), as it would imply an adequate performance of the GMB. For volatile PFAS compounds, 'Probable' partitioning to the solid would be desirable because it retards the movement in the primary medium. The various situations considered are how different exposures to solar radiation - no exposure (i.e. ideal installation), < 3 months, many months - and their effect on gas diffusion, liquid permeation and partitioning to the polymer. Partitioning to the polymers of the GTX component in GCL would be expected to mostly follow that indicated for an ideal situation in Table 6.

Table 6 Chemical and physical factors impacting performance of GMBs deployed in capping, side-wall and base liner systems.

Ideal installation	Condition of GMB	
	Exposed for < 3 months	Exposed for many years
Oxidative loss: -	Oxidative loss: $\uparrow_{\text{minor}}$	Oxidative loss: $\uparrow$
Embrittlement: -	Embrittlement: $\uparrow_{\text{minor}}$	Embrittlement: $\uparrow_{\text{minor}}$
Punctures: -	Punctures: $\uparrow_{\text{potential}}$	Punctures: $\uparrow_{\text{potential}}$
Weld integrity: -	Weld integrity: -	Weld integrity: $\downarrow$
Interface with GCL: -	Interface with GCL: -	Interface with GCL: $\downarrow_{\text{minor}}$
$k_{\text{gas}}/D_{\text{gas}}$: -	$k_{\text{gas}}/D_{\text{gas}}$: $\uparrow_{\text{minor}}$	$k_{\text{gas}}/D_{\text{gas}}$: $\uparrow$
k_{liquid} : -	k_{liquid} : $\uparrow_{\text{minor}}$	k_{liquid} : $\uparrow$
Neutral change: - $\uparrow$: increase $\downarrow$: decrease		

Table 7 Possible interactions of non-polar, polar, water soluble and volatile PFAS compounds with LLDPE and HDPE type GMBs under different exposure conditions. Colour coding denotes relative benefit to retarding PFAS transport.

Condition of LLDPE and HDPE GMB				
PFAS interactions		<i>Ideal installation</i>	<i>Exposed for < 3 months</i>	<i>Exposed for many months</i>
Non-polar, H₂O soluble	Gaseous diffusion	Possible	Possible	Probable
	Liquid permeation	Possible	Probable	Probable
	Partitioning to polymer	Possible	Possible	Possible
Non-polar, volatile	Gaseous diffusion	Probable	Probable	Probable
	Liquid permeation	Possible	Possible	Probable
	Partitioning to polymer	Possible	Possible	Possible
Polar, H₂O soluble	Gaseous diffusion	Improbable	Possible	Probable
	Liquid permeation	Improbable	Possible	Probable
	Partitioning to polymer	Possible	Possible	Probable
Polar, volatile	Gaseous diffusion	Improbable	Possible	Probable
	Liquid permeation	Improbable	Improbable	Probable
	Partitioning to polymer	Possible	Possible	Probable

H₂O soluble: most PFCAs, PFSA, FTSAs, FTCAs

Volatile: PFBA, PFBS, PFPeA, PFHxA, 4:2 and 6:2 FTOH, other volatile PFAS replacement precursors

Table 8 Possible interactions of non-polar, polar, water soluble and volatile PFAS compounds with internal section of coextruded Nylon VPB15 and EVOH type GMBs under different exposure conditions. Colour coding denotes relative benefit to retarding PFAS transport.

Condition of Nylon VPB15 and EVOH GMB			
PFAS interactions	<i>Ideal installation</i>	<i>Exposed for < 3 months</i>	<i>Exposed for many months</i>
Gaseous diffusion	Improbable	Unknown	Unknown

Non-polar, H₂O soluble	Liquid permeation	Improbable	Unknown	Unknown
	Partitioning to polymer	Probable	Unknown	Unknown
Non-polar, volatile	Gaseous diffusion	Improbable	Unknown	Unknown
	Liquid permeation	Improbable	Unknown	Unknown
	Partitioning to polymer	Probable	Unknown	Unknown
Polar, H₂O soluble	Gaseous diffusion	Improbable	Unknown	Unknown
	Liquid permeation	Improbable	Unknown	Unknown
	Partitioning to polymer	Probable	Unknown	Unknown
Polar, volatile	Gaseous diffusion	Improbable	Unknown	Unknown
	Liquid permeation	Improbable	Unknown	Unknown
	Partitioning to polymer	Probable	Unknown	Unknown

H₂O soluble: most PFCAs, PFSAs, FTSAs, FTCAs

Volatile: PFBA, PFBS, PFPeA, PFHxA, 4:2 and 6:2 FTOH, other volatile PFAS replacement precursors

It must be emphasised that these hypothetical ageing scenarios are largely untested, and the impact on HDPE is presented in general terms only. However, exposure of HDPE GMBs to many months of solar radiation can have serious detrimental effects on performance (e.g. [90, 147, 148]). The same ageing depicted in Table 6 may have similar impacts upon the LLDPE/HDPE shells of coextruded GMBs. Again, it must be stressed that there is currently no evidence available on how aging influences their performance, nor is there any data specific to their actual application as barriers to PFAS compounds.

4. Next Generation of Modern Composite Lining Systems

Modern composite liner systems - even today - have not been designed with the collection, retention and isolation of PFAS compounds in mind. Thus, with the increased awareness of the ubiquitous presence of PFAS compounds in atmospheric, aquatic, terrestrial and anthropic environments, it is imperative to consider whether or not shortfalls exist in the current design and installation of modern composite lining systems for municipal landfill wastes, which are the destination for a significant proportion of PFAS-laden waste materials.

Despite the current lack of knowledge specific to PFAS compounds, well-supported predictions can still be made regarding the next generation of composite liners designed to mitigate the current problems with PFAS compounds. Based on the latest data to date (as reported herein and in [59, 146]) contemporary design appears to have primarily addressed the retention of PFOA and PFOS. However, shorter carbon chain PFAS can still be mobile. Thus, considerable research - in terms of

case studies and product development is required to address the mobility of short-chain PFAS, in particular PFBA and PFBS. Several aspects of landfill liner design and installation need to be reassessed to minimise further egress of PFAS compounds into the environment. These include the choice of lining component materials:

- LLDPE vs HDPE vs SF HDPE vs Nylon VPB15 vs EVOH as geomembrane-based primary liners in caps, base and side-walls,
- Higher bentonite mass or the use of bentonites modified with activated carbon and/or other minerals and specialty polymers in geosynthetic clay liner secondary liners in double liner systems, and
- Stricter control of clay materials used in compacted clay liners in composite liner systems.

Continued research is also required that addresses the gaps in knowledge regarding biotic and abiotic transformations of PFAS in landfill environments, particularly the conditions in the lining systems where leachate accumulates. Additionally, research is needed that focusses on interactions of PFAS with specific landfill liner components, including the assessment of the performance of current, modified and new liner components for their potential to retain PFAS and to mitigate the risk of environmental exposure or release.

In addition, designs should bring the most advanced aspects to bear, including the incorporation of geotextile (modified or otherwise) protection layers, drainage layers, as well as both leachate and gaseous collection systems. Better design cannot work if installations are inadequate (e.g. poor welding of GMB), outside of specification (e.g. low bentonite mass in GCL), or when normal practices continue that are known to impair liner integrity (e.g. prolonged environmental exposure of composite liner on side slopes). Ultimately, however, more laboratory experimental and field case studies are required, specifically regarding the fate and behaviour of PFAS compounds - or at least classes of PFAS compounds - with the lining components.

5. Conclusions and Summary

This review intended to bring to the attention of researchers, asset managers and policy makers the challenges surrounding PFAS containment in engineered landfill liner systems. It also focussed on the current best practice in design and management of landfill and temporary containment facilities, and ways to mitigate further transmission of this class of persistent, emerging contaminant into the terrestrial, aquatic, atmospheric and anthropic environments. In particular, the review highlighted potential roles composite lining systems may play in PFAS containment and to cast into sharp relief the many and significant knowledge gaps within the academic literature regarding the complete understanding of the fate and behaviour of PFAS compounds within landfill systems.

Recent literature shows that PFAS contamination of landfill sites is omnipresent, increasing with the worldwide increase in consumer products and is becoming an ever-growing concern, due to both the ubiquity of PFAS, and the substitution of longer-chain compounds for more mobile shorter carbon-chain length PFAS and volatile precursors, such as FTOHs.

While little evidence exists as to the performance of compacted clay liners per se on PFAS retention, as with soils, in general one can expect that efficacy as a hydraulic barrier is wholly dependent on how soundly the liner has been constructed. Given the above discussion on the generally low level of PFAS retention in soils [15, 25, 84], coupled with generally large bulk transfer rates, particularly of short-chain PFAS [85], and long-known issues associated with quality assurance

and control of compacted clay liner construction both with depth and over large areas [149-151], it is recommended that lining systems for PFAS containment utilise CCL only in combination with geomembranes. CCL should only serve as part of composite liners in designs of landfills for PFAS containment and not as the primary or only liner in modern landfills for PFAS containment.

Recently published results [59], along with those provided herein (Section 3.2.4) highlight that the bentonite component by itself is not able to diminish total PFAS concentrations in the effluent, but can impact on longer carbon chain-length and currently regulated PFOS, PFOA and PFHxS attenuation through the bentonite layer. Other PFAS, including the shorter carbon chain-length PFBA, PFBS, PFHpA and PFHxA appear to remain mostly mobile after a short-duration attenuation, even in activated carbon modified GCL products. These findings highlight that GCL alone should not be relied on solely for PFAS containment and should be used in combination with GMB products (as is generally recommended for waste containment applications).

This literature review has shown that there currently exists a complete paucity of research on how PFAS interact with geotextiles and considerable effort should focus on PFAS sorption and retention, but also how GXT, for example in GCL, may serve as flow channels for some PFAS. The limited research on how geomembranes can contain PFAS indicate that co-extruded GMB [137, 146] may provide increased performance over conventional commodity GMB. Surface fluorinated forms of GMB should also be considered given that they likely impart other desired attributes, including increased heat resistance.

Continued research is needed to clarify important gaps relating to the fate of PFAS compounds within landfills. Further study is also required to address the gaps in knowledge regarding biotic and abiotic transformations of PFAS in landfill environments, particularly the conditions in the lining systems where leachate accumulates. Additionally, research is needed that focusses on interactions of PFAS with specific landfill liner components, including the assessment of the performance of current, modified and new liner components for their potential to retain PFAS and to mitigate the risk of environmental exposure or release.

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Author Contributions

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Competing Interests

The authors have declared that no competing interests exist.

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