Proceedings of the 9ICEG
9th International Congress on Environmental Geotechnics
25-28 June, 2023 | Chania, Greece

https://doi.org/10.53243/ICEG2023-73

Hydraulic Conductivity of Compacted Clay Liners to Landfill Leachates Containing PFAS

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are ubiquitous and persistent organic contaminants in the environment that are toxic at very low concentrations. PFAS concentrations in municipal solid waste (MSW) landfill leachates greater than 1000 ng/L have been reported, and concern has been raised regarding the effectiveness of landfill liners to contain PFAS. Moreover, no studies have been conducted to evaluate the impact of PFAS in leachate on the hydraulic conductivity of compacted clay liners (CCLs). In this study, hydraulic conductivity tests were conducted on three compacted clay liner materials using synthetic MSW leachate or deionized water (DIW) spiked with PFAS as the permeant solution. A low plasticity clay (CL), a high plasticity clay (CH), and a moderately plastic organic clay (OH) were used for testing. The permeant solutions contained perfluorohexanoic acid (PFHxA, a common short-chain PFAS) or perfluorooctane sulfonic acid (PFOS, a common long-chain PFAS) at a concentration of 1000 ng/L. Tests were also conducted with synthetic MSW leachate or DIW without PFAS. Hydraulic conductivity of the compacted clays to the synthetic MSW leachate was modestly higher (up to 4 times) than to DI water. Hydraulic conductivity of the compacted clays to MSW or DIW spiked with PFAS was comparable to the hydraulic conductivity to MSW leachate or DIW alone, indicating that PFAS in landfill leachates does not have significant impact on the hydraulic conductivity of CCLs.

Keywords: Per- and polyfluoroalkyl substances (PFAS), municipal solid waste, landfill, leachate, compacted clay liner, hydraulic conductivity

1 INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are a group of fluorinated organic chemicals having a hydrophobic and fluorine-saturated carbon chain and a lipophobic functional group (Remucal 2019). The unique chemical characteristics of PFAS have led to their extensive use in industrial and consumer applications such as fast-food containers, water-repellent clothing, firefighting foams, and products that resist grease, water, and oil (Glüge et al. 2020, Evich et al. 2022). The strong carbon-fluorine bond renders PFAS highly resistant to degradation, resulting in persistence in the environment. PFAS also bioaccumulates in humans and are thought to have health impacts at very low concentrations (Domingo and Nadal 2019, Ng et al. 2021). Because of their potential toxicity, some regulatory agencies have proposed maximum acceptable PFAS concentrations in groundwater as low as 4 ng/L.

PFAS are ubiquitous in municipal solid waste (MSW) and landfills serve as long-term repositories for PFAS. PFAS in the waste is released into landfill leachate, providing a pathway for release into the environment (Allred et al. 2015, Lang et al. 2017). PFAS concentrations in landfill leachates have been reported to be more than several thousand ng/L (Bouazza 2021, Liu et al. 2022). While the impact of other organic compounds dissolved in MSW leachate has been shown to have negligible impact on landfill liner materials (Bowders and Daniel 1987, Yanful et al. 1990, Shackelford 1994, Kim et al. 2001, Edil 2003, Park et al. 2012), little is known regarding how PFAS compounds may affect landfill liners (Di Battista et al. 2020, Gates et al. 2020, Rowe and Barakat 2021, Tan and Benson 2023).

In this study, the impact of PFAS on the hydraulic conductivity of clays used for compacted clay liners (CCLs) was evaluated. Hydraulic conductivity tests were conducted on three clays typical of those used for compacted clay liners in MSW landfills: a low plasticity clay, a high plasticity clay, and a moderately plastic clay with elevated organic carbon content (0.46%) that potentially could attenuate PFAS. Deionized water (DIW) and a synthetic MSW leachate spiked with a long-chain PFAS (perfluorooctane sulfonic acid, PFOS) or a short-chain PFAS (perfluorohexanoic acid, PFHxA) were used as permeant liquids. PFHxA and PFOS are common PFAS that have been detected in MSW landfill leachates (Allred et al. 2015, Lang et al. 2017). Control tests were also conducted on the three soils using DIW or synthetic MSW leachate without PFAS as the permeant liquid.

2 MATERIALS AND METHODS

2.1 Compacted clay liners

The clays used in this study were selected from the University of Wisconsin-Madison (UW) Soil Bank (Benson and Gurdal 2013) to represent three soils typically used for compacted clay liners in landfills. Compaction and index properties of the soils are summarized in Table 1, which were measured following the methods described in ASTM D698 (ASTM 2021), D4318 (ASTM 2017), D422 (ASTM 2007), and D854 (ASTM 2014). In the Unified Soil Classification System (USCS), the soils classify as low plasticity clay (CL), high plasticity clay (CH), and moderately plasticity organic clay (OL-OH). The low plasticity and high plasticity clays, designated herein as CL and CH, are ASTM International standard soils (Benson and Yesiller 2016). The organic clay, designated OH herein, is a moderately plastic organic clay from Dane County, Wisconsin, USA that is known as Kamm clay (Park et al. 2012). The OH clay has an organic matter content of 0.46% (Park et al. 2012).

Table 1. *Compaction, particle size fractions, Atterberg limits, and specific gravity of solids of soils.*

Soil	Standard Proctor Compaction		Atterberg Limits		Particle Size Fractions (%)				Specific Gravity (G_s)
	w_{opt} (%)	$\gamma_{d,max}$ (kN/m^3)	Liquid Limit	Plasticity Index	Gravel	Sand	Fines	Clay	
CL	17.6	17.1	32	12	0	11	89	34	2.68
CH	23.6	15.3	55	33	0	2	98	47	2.72
OH	19.3	17.4	48	27	0	11	89	30	2.72

Note: Clay defined as particles finer than 2 μm (Benson et al. 2018).

2.2 Permeant liquids

The typical synthetic MSW leachate described in Bradshaw and Benson (2014) was used to represent MSW leachate in this study. Bradshaw and Benson (2014) identified the typical MSW leachate chemistry based on an extensive review of MSW leachates in the United States. The ionic strength of the synthetic MSW leachate is 60 mM and the relative abundance of monovalent and polyvalent cations (RMD) is 0.36 $M^{0.5}$. The synthetic MSW leachate was prepared by dissolving reagent-grade salts (1.70 g/L sodium chloride, 0.41 g/L calcium chloride, 0.49 g/L magnesium chloride, and 0.33 g/L potassium chloride) in DIW following the procedures in Jo et al. (2001) and Benson et al. (2022). The synthetic MSW leachate had an electrical conductivity (EC) of 0.56 S/m and pH 7.4.

The synthetic MSW leachate was spiked with PFOS (8 carbons), a typical long-chain PFAS in landfill leachate, or PFHxA (6 carbons), a typical short-chain PFAS in landfill leachate. Permeant solutions were also prepared with DIW spiked with PFOS or PFHxA to study the effects of PFAS on the hydraulic conductivity without interactions from the other constituents in the MSW leachate. All permeant solutions had a PFOS or PFHxA concentration of 1000 ng/L to represent a typical PFAS concentration in MSW leachate (Bouazza 2021, Zhang et al. 2023). The PFOS and PFHxA were obtained in concentrated solutions from Wellington Laboratories (Guelph, Ontario, Canada).

2.3 Specimen preparation

Test specimens of compacted clay were prepared using the procedures in Benson and Yesiller (2016), which have been demonstrated to create highly reproducible specimens for hydraulic conductivity testing. The soil was aired dried and crushed to pass a No. 4 sieve (4.75 mm) per ASTM D698 (ASTM 2021) and then moistened with tap water (pH = 8.1, EC = 0.08 S/m at 25 °C) using a spray bottle. The target water content was 1% wet of optimum water content per standard Proctor, which is common for construction of CCLs (Benson et al. 1994, Benson and Trast 1995). The moistened soil was thoroughly mixed using a trowel and subsequently sealed in a plastic bag to equilibrate for at least 24 h. Test specimens were prepared by compacting the moistened soil in a short steel compaction mold (51 mm height, 152 mm diameter) using a standard Proctor hammer with the number of blows adjusted to achieve the dry unit weight corresponding to standard Proctor effort. Compacted specimens were extracted from the mold using a hydraulic jack, sealed in plastic zip-top bags, and stored in a 100% humidity room prior to hydraulic conductivity testing. All specimens were set up in a permeameter within 24 h of compaction.

2.4 Hydraulic conductivity testing

The clay test specimens are being permeated in flexible-wall permeameters using the falling headwater-constant tailwater method in ASTM D7100 (ASTM 2016). None of the tests had met the termination criteria when this paper was prepared. The most recent data available at the time of publication are reported herein.

All tests are being conducted using an average effective stress of 20 kPa and an average hydraulic gradient of 25. No backpressure was applied to minimize alterations in geochemistry that would not be observed in the field (e.g., Le Chatelier's principle). Specimens were initially equilibrated with the solution in the permeameter by applying the headwater and cell pressure for at least 48 h with the effluent line closed (i.e., no flow). Tests are being conducted with the synthetic MSW leachate and the DIW spiked with PFAS and with synthetic MSW leachate and DIW without PFAS to evaluate the impact of PFAS on hydraulic conductivity. High-density polyethylene (HDPE) tubes are used in the permeameters to create a PFAS-free environment for testing and to minimize adsorption of PFAS to the tubes during long-term hydraulic conductivity testing.

The tests are to be conducted until the termination criteria in ASTM D7100 are satisfied. The hydraulic conductivity is required to be steady, the ratio of incremental outflow to inflow is required to be steady and fall between 0.8 to 1.2, the pH and EC of the effluent are required to be within 20% of those of the influent, and the PFAS concentration in the effluent is required to be at least 80% of the concentration in the influent. Permeation is being continued until at least two pore volumes of flow pass through the specimen and both hydraulic and chemical equilibrium are achieved.

3 RESULTS

Hydraulic conductivity of the compacted clays to DIW, synthetic MSW leachate, and the DIW and MSW leachate spiked with PFAS are summarized in Table 2. The specimens had been permeated for at least 79 d at the time this paper was prepared. All had reached hydraulic equilibrium, but none had reached chemical equilibrium, with PFAS concentrations in the effluent considerably lower (<20%) than in the influent. All of the hydraulic conductivities reported in Table 1 are less than 1×10^{-9} m/s, the common requirement for CCLs (Benson et al. 1999, 2018).

Hydraulic conductivities of the compacted clays to permeant solutions spiked with PFAS are compared to hydraulic conductivities to permeant solutions without PFAS in Fig. 1. Hydraulic conductivities to permeant solutions containing PFHxA are shown as solid symbols, whereas those to PFOS solutions are shown with open symbols. The black dashed lines in Fig. 1 represent a two-fold difference between the hydraulic conductivities to solutions with or without PFAS, and the blue dashed lines correspond to a four-fold difference. These lines represent the reproducibility of hydraulic conductivity measurements on more permeable and less permeable compacted clays measured in flexible-wall permeameters, as defined in the ASTM Interlaboratory Study (ILS) conducted by Benson and Yesiller (2016).

Hydraulic conductivities of the CL and CH clays permeated with DIW spiked with PFAS are within the two-fold band, and the hydraulic conductivity of the OH clay permeated with DIW spiked with PFAS is within the four-fold band (Fig. 1a). None of the hydraulic conductivities obtained with DIW and PFAS fall outside the four-fold band. Similar results have been obtained with synthetic MSW leachate (Fig. 1b). Hydraulic conductivities of all the three compacted clays permeated with synthetic MSW leachate spiked with PFAS are within the four-fold band of the hydraulic conductivity to synthetic MSW leachate alone. Moreover, there is no systematic difference between hydraulic conductivities of the compacted clays permeated with solutions containing PFHxA relative to solutions containing with PFOS. These finding are consistent with conclusions reported by Bowders and Daniel (1987), Broderick and Daniel (1990), and Shackelford (1994). They report that aqueous solutions containing dissolved organic compounds have little impact on the hydraulic conductivity of compacted clays.

Table 2. *Summary of hydraulic conductivities and other test conditions when paper prepared.*

Soil	Permeant Liquid		Hydraulic Conductivity (m/s)	Pore Volumes of Flow (-)	Cumulative Test Duration (d)
	Matrix	PFAS			
CL	DIW	NA	2.0×10^{-10}	2.87	263.0
	MSW	NA	4.5×10^{-10}	3.68	265.4
	DIW	PFHxA	2.4×10^{-10}	1.67	160.1
	MSW	PFHxA	2.2×10^{-10}	1.60	160.1
	DIW	PFOS	1.4×10^{-10}	0.53	88.9
	MSW	PFOS	1.4×10^{-10}	0.60	88.9
CH	DIW	NA	2.2×10^{-11}	0.55	263.0
	MSW	NA	6.7×10^{-11}	1.95	263.0
	DIW	PFHxA	3.1×10^{-11}	0.49	165.5
	MSW	PFHxA	2.4×10^{-11}	0.32	165.5
	DIW	PFOS	2.4×10^{-11}	0.27	95.5
	MSW	PFOS	3.8×10^{-11}	0.41	95.5
OH	DIW	NA	1.2×10^{-11}	0.38	263.0
	MSW	NA	1.6×10^{-11}	0.50	263.0
	DIW	PFHxA	2.5×10^{-11}	0.38	172.2
	MSW	PFHxA	2.1×10^{-11}	0.61	172.2
	DIW	PFOS	2.9×10^{-11}	0.48	79.5
	MSW	PFOS	2.4×10^{-11}	0.37	79.5

Notes: “NA” indicates no PFAS in permeant solution, MSW = synthetic MSW leachate, DIW = deionized water.

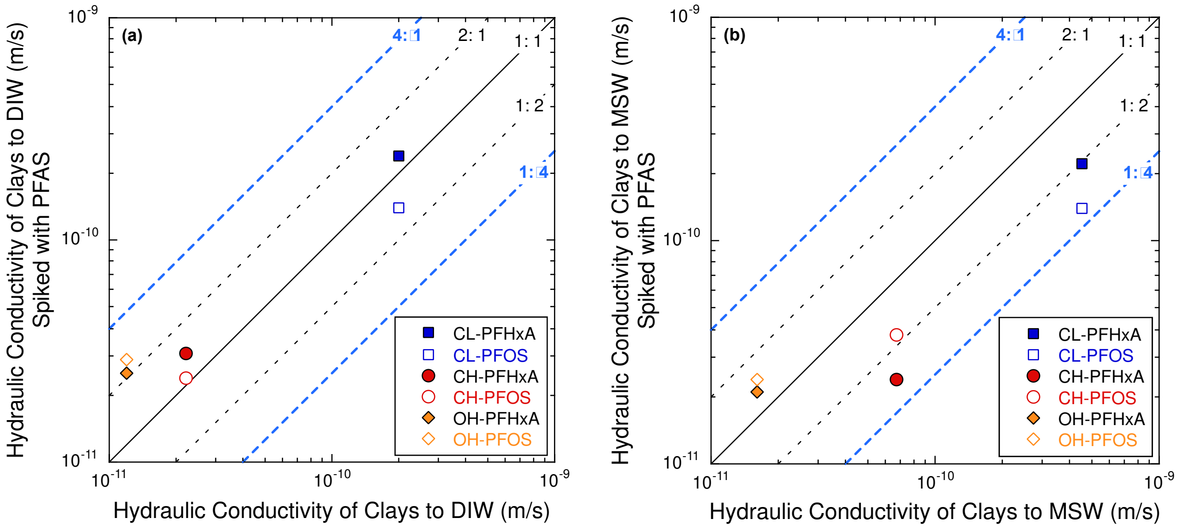

Figure 1. *Hydraulic conductivity of compacted clays to DIW with PFAS vs. hydraulic conductivity of to DIW (a) and hydraulic conductivity to synthetic MSW leachate with PFAS vs. hydraulic conductivity to synthetic MSW leachate alone (b).*

Hydraulic conductivities of the compacted clays permeated with synthetic MSW leachate are compared to the hydraulic conductivities to DIW in Fig. 2. The data shown in Fig. 2 correspond to pairs of tests spiked with the same PFAS (e.g., MSW leachate spiked with PFOS compared to DIW spiked with PFOS) or without PFAS. Bands corresponding to the 2-fold and 4-fold reproducibility limits for hydraulic conductivity testing are also shown in Fig. 2. All of the hydraulic conductivities fall within the 4-fold bound, and most within the 2-fold bound, indicating that the hydraulic conductivities obtained with MSW leachate are comparable to those obtained with DIW, regardless of whether the permeant solutions contain PFAS. However, the hydraulic conductivities to MSW leachate tend to be modestly higher relative to those obtained with DIW.

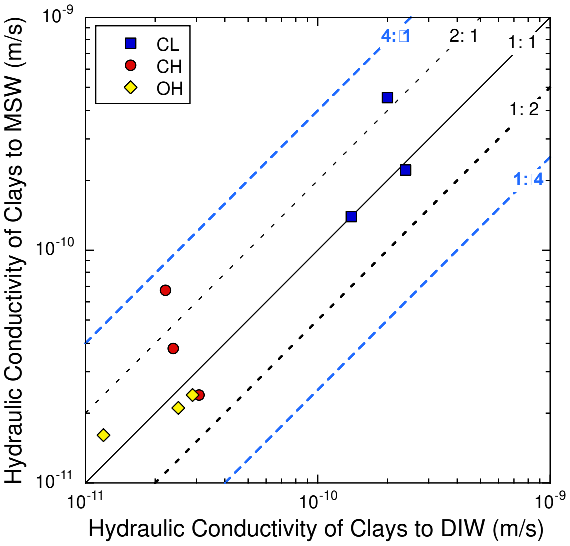

Figure 2. *Hydraulic conductivity of compacted clays to synthetic MSW leachate relative to DIW.*

Hydraulic conductivities of the clays obtained with each of the permeant solutions are shown as a function of plasticity index and void ratio in Fig. 3. Composition of the clays has a much larger impact on hydraulic conductivity than the permeant solution, with no distinguishable effect associated with PFAS in the solution. The lowest hydraulic conductivities correspond to the moderately plastic organic clay (OH) and the highest hydraulic conductivities correspond to the low plasticity clay (CL). These findings are consistent with those reported by Benson et al. (1994), Benson and Trast (1995), and Tan et al. (2023), which show that the hydraulic conductivity of compacted clays to dilute solutions is affected predominantly by composition and compaction condition rather than characteristics of the permeant solution.

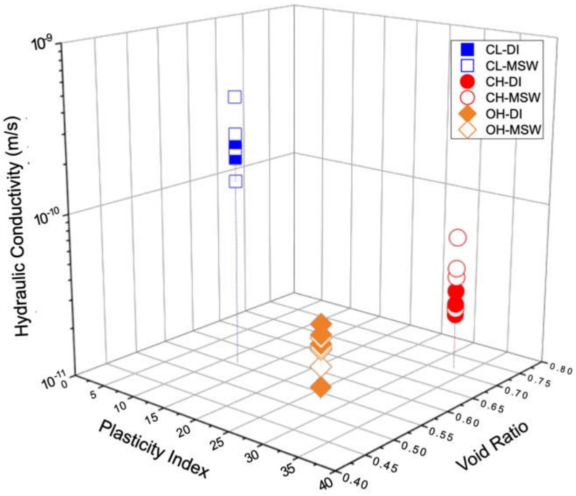

Figure 3. *Hydraulic conductivity of CCLs to DIW or synthetic MSW leachate with or without PFAS as a function of plasticity index and void ratio.*

4 CONCLUSIONS

Tests were conducted on three clay soils with USCS classifications of CL, CH, and OL-OH to evaluate how PFAS may affect the hydraulic conductivity of typical CCLs in MSW landfills. Hydraulic conductivity tests were conducted on the three soils using DIW and synthetic MSW leachate spiked with a short-chain (PFHxA) or a long-chain (PFOS) PFAS at 1000 ng/L. Hydraulic conductivity of all three compacted clays to the permeant solutions with PFAS were comparable to hydraulic conductivities obtained with the same solutions without PFAS, falling within established bounds of reproducibility for hydraulic conductivity testing of clays. Hydraulic conductivity of the clays to the synthetic MSW leachate was comparable, but modestly higher than the hydraulic conductivity to DI water. These findings suggest that PFAS in landfill leachates will not adversely affect the hydraulic conductivity of CCLs, and confirms that typical MSW leachates do not adversely affect the hydraulic conductivity of CCLs.

5 ACKNOWLEDGEMENTS

Financial support for this study was provided by the Environmental Research and Education Foundation (EREF), the US Department of Energy's Consortium for Risk Evaluation with Stakeholder Participation (CRESP) III through Cooperative Agreement No. DE-FC01-06EW07053, and through an EREF fellowship to the first author (Y. Tan).

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The paper was published in the proceedings of the 9th International Congress on Environmental Geotechnics (9ICEG), Volume 1, and was edited by Tugce Baser, Arvin Farid, Xunchang Fei and Dimitrios Zekkos. The conference was held from June 25th to June 28th 2023 in Chania, Crete, Greece.