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Impact of PFOS concentrations on the contaminating lifespan of landfills with a single composite liner

B. Enes Taskesti[1] and Abdelmalek Bouazza[2]

[1]PhD Student, Monash University, Melbourne, Australia, email: bilal.taskesti@monash.edu

[2]Professor, Monash University, Melbourne, Australia, email: malek.bouazza@monash.edu

ABSTRACT

This study investigates the migration of perfluorooctane sulfonic acid (PFOS) through a composite liner consisting of a geomembrane, a geosynthetic clay liner (GCL), and a 2 m thick attenuation layer located above an aquifer. The transport of PFOS is analysed using models for pure diffusion and advective transport through holed wrinkles in the liner. The findings indicate that (a) when there is no leakage and only pure diffusive transport occurs, the regulatory threshold based on the recent US EPA interim guidelines for allowable PFAS concentration in drinking water is not fully satisfied, and (b) if there exist wrinkles in the geomembrane that have holes, the degree of impact depends heavily on the number of the holed wrinkles, the interface transmissivity between the geomembrane and GCL, and the hydraulic conductivity of the GCL. The analysis reveals that in all scenarios considered, the highest concentrations of PFOS detected in the aquifer surpass the thresholds established by the relevant authorities, suggesting that relying solely on a composite liner may not be adequate to prevent PFOS from reaching unacceptable levels. The study indicates that realistic evaluations of the contaminant impact be carried out for each specific scenario to ensure appropriate containment.

Keywords: Contaminant transport, PFOS, diffusion, geosynthetic clay liner, geomembrane

1 INTRODUCTION

Per and polyfluoroalkyl substances (PFASs) are a group of synthetic chemicals that are used in a wide range of industrial and consumer products because of their unique properties (Lyu et al., 2018; Di Battista et al., 2020; Brusseau et al., 2020; Bouazza, 2021; Kabiri & McLaughlin, 2021; Van Glubt et al., 2021; Wang et al., 2021; Cai et al., 2022; Cousins et al., 2022). They are known for being hydrophilic, hydrophobic and oleophobic; thus able to repel stains, grease, and water (Brusseau, 2018; Hamid et al., 2018; Li et al., 2019). These properties make them useful in various applications, including non-stick coatings for cookware, waterproof clothing, food packaging, cosmetics and firefighting foams, among many others (Buck et al., 2011). These unique properties are attributed to strong bond forces, in their chemical structures, between carbon and fluorine atoms which are replaced with hydrogen atoms (Buck et al., 2011). They are currently classified as emerging contaminants of concern. PFASs are known for being persistent in the environment as they don't break down easily and can bioaccumulate over time. For example, perfluorooctane sulfonic acid (PFOS), one of the most commonly detected PFAS types, has been found to be toxic and can cause health problems, including cancer, liver damage, and developmental problems (Shearer et al., 2021) and has been detected in human blood (Lindh et al., 2012; Zeng et al., 2015).

Leachates from municipal solid waste landfills have been shown to contain various PFAS compounds with an extensive range of concentrations, typically well above permissible concentrations (Bouazza, 2021). Although PFAS have known toxicity, there are currently no established regulations for their disposal, nor are landfill liner design guidelines addressing PFAS migration. Modern waste containment facilities commonly have composite liners consisting of a geosynthetic clay liner or compacted clay liner and a geomembrane layer to provide hydraulic containment (Bouazza, 2002; Rowe, 2005, 2012, 2020; Rentz et al., 2016; Scalia et al., 2018; Mukunoki et al., 2019; Yu & El-Zein, 2019; Rowe & Abdelrazek, 2019; Yu et al., 2020; Rowe & Barakat, 2021). A liner is engineered to function as a hydraulic barrier to minimise contaminant migration into ground and surface waters. In an ideal scenario, the geomembrane would be devoid of defects resulting in diffusion becoming the primary contaminants transport mechanism through the composite liner (Sangam & Rowe, 2001; Joo et al., 2005; McWatters & Rowe, 2009, 2010; Jones & Rowe, 2016). However, a geomembrane free of defects is unlikely to be encountered on-site; thus, standard practice is to assume the existence of holes in the geomembrane. The range of defects/holes is a function of the on-site construction quality assurance (CQA). For example, if reasonable construction quality assurance (CQA) applies, the expected number of holes can vary between 2 to 5 holes/ha (Giroud & Bonaparte, 1989; Giroud, 2016). In the case of poor CQA, 20 or more holes/ha may be envisaged (Giroud & Bonaparte, 1989; Giroud, 2016). Furthermore, holes may be present in wrinkles which exacerbate the situation further. The existence of holes means that advection becomes the primary contaminant transport mechanism through the composite liner.

This paper investigates the hydraulic efficiency of a composite liner constituted of a geomembrane (with and without defects) and a geosynthetic clay liner to contain PFOS in the light of the recent release by the United States Environmental Protection Agency (US EPA) (2022) of an interim guideline on PFOS allowable concentration in drinking water which was lowered from 70 ng/L to 0.02 ng/L.

2 METHOD

Using contaminant migration analysis software (POLLUTE v8), a composite liner was modelled to estimate contaminant concentrations at specific times based on Rowe & Abdelrazek's (2019) approach. The landfill size was taken as 400 m long and 100 m wide. The GCL, GMB and attenuation layer thicknesses were taken as 0.007 m, 0.002 m and 2 m, respectively, based on landfill design requirements of EPA Victoria (2015). The hydraulic conductivity of the attenuation layer was 1.0×10^{-7} m/s. The effective diffusion (D_e) and partitioning coefficient (S_{gf}) of PFOS through geomembrane were assumed as 1.5×10^{-16} m^2/s and 4.0, respectively (Rowe & Barakat, 2021). The diffusion coefficient of PFOS through GCL was assumed to be 3.8×10^{-11} m^2/s based on the typical diffusion coefficient range of GCLs (between 1×10^{-10} and 1×10^{-12}). Two hydraulic conductivity values of GCL were used to mimic the stress-free zone beneath a wrinkle (k_b) and in the zones where there is good contact between GCL and the geomembrane away from the wrinkle (k_a). The PFOS distribution (or partition) coefficient (Kd) of the GCL was taken as 220.5 mL/g based on laboratory measurement. The aquifer thickness was considered as 3 m. The dispersivity of the attenuation layer and aquifer were considered as 0.1 and 3 m, respectively. The leachate height acting on top of the geomembrane was 0.3 m. The groundwater level was 1.26 m above the aquifer surface. The head loss acting on the composite liner was 2.31 m. Based on Rowe & Barakat (2021), the inflow Darcy velocity of the aquifer was taken as 1 m/a. The infiltration through the soil cover was assumed to be 0.15 m/a, and the waste mass per unit area was 25000 kg/m^2. The vertical stress acting on the composite liner was about 250 kPa. Based on the literature review, the PFOS ratio in the waste and initial concentration of PFOS was assumed to be 1.2×10^{-3} mg/kg and 4800 ng/L (Rowe & Barakat, 2021).

Two cases were considered in the current investigation. The first case considered the case of an intact geomembrane having intimate contact with the GCL. While no leakage occurs in this case, diffusion would govern the PFOS migration. It is assumed that the leachate collection system removes the entire infiltration; thus, there is no Darcy flux through the composite liner. Pure diffusion is not an expected case in real conditions; however, it was modelled as a reference baseline and comparative purposes. Under site conditions, holes may occur (1) during the installation of the geomembrane, (2) during the covering of the geomembrane, or (3) during operations. The development of wrinkles on the geomembrane is inevitable; thus, a worst-case scenario of having a hole in the wrinkle was assumed in the current investigation. Wrinkle length and width were taken as 100 m and 0.1 m, respectively (Rowe & Barakat, 2021). The number of holes on the wrinkle ranged from 2 to 8 to examine the effect of holed wrinkle length on leakage and contaminant concentration in the aquifer. The leakage rates were calculated based on Rowe (2005, 2012, 2020) and Rowe & Abdelrazek (2019).

POLLUTE v8 allows the modelling of contaminant transport through composite liners. Advection and diffusion-based contaminant migration were modelled owing to the relatively low diffusion coefficient of PFOS through geomembrane (Di Battista et al., 2020). However, the dominant transport type was advection below the area impacted by wrinkles with a hole. To simulate PFOS in the landfill, finite mass was applied considering the initial contaminant concentration (c_0) and reference height of leachate (H_r).

3 RESULTS

If no defects exist in the geomembrane, the contaminant transport will be diffusion driven only. Based on the analysis conducted in the current investigation, the peak PFOS concentration in the aquifer was 0.13 ng/L and was reached at 570 years. This concentration level was much higher than the allowable PFOS concentration level proposed by US EPA (2022), 0.02 ng/L, which was reached at 174 years.

Rowe & Jabin (2021) conducted a study demonstrating that the interface transmissivity in a composite liner and hydraulic conductivity of GCL change depending on the vertical stress applied to the liner and type of geomembrane and GCL. Rowe & Jabin (2021) also reported that the interface transmissivity values ranged from 0.5 to 0.0002 m^2/a. To simulate a situation where holes will be located on a wrinkle, three scenarios were considered in the current investigation to examine the effect of transmissivity at the GMB-GCL interface and GCL hydraulic conductivity. The case ID for each scenario was denoted considering interface transmissivity values and hydraulic conductivity of GCL. Accordingly, low, moderate, and high transmissivity situations were denoted as L-8, M-8 and H-8 (see Table 1). The number next to the letters symbolises the number of holes on the wrinkle. The values of the interface transmissivity used in the current investigation are based on Rowe & Barakat (2021); thus, the value for Case L-8 was 0.001 m^2/a, indicating reasonable construction quality assurance (CQA) during the geomembrane installation. In the case of M-8, the interface transmissivity value was 0.01 m^2/a. In the case H-8, the interface transmissivity value was 0.1 m^2/a, the highest, indicating poor CQA. Also, Case L-8, M-8 and H-8 represent landfill liners with a hydraulic conductivity of $k_a=4\times10^{-11}$ and $k_b=2\times10^{-10}$ m/s. Figure 1 shows the change in PFOS concentration in the aquifer as a function of time for the three cases considered. Together with a decrease in interface transmissivity and hydraulic conductivity, peak PFOS concentration values decreased considerably. However, all cases considered have higher peak PFOS concentrations than allowable PFOS concentration values proposed in the US EPA interim guidelines (i.e. 0.02 ng/L).

Table 1 summarises the results obtained from the three cases. Interestingly, the leakage rates tend to increase as the transmissivity rate at the GMB-GCL interface increases concurrently with the increase of the wetted area under the holed wrinkle. The range of leakage rates (48 to 179 litre per hectare per day (lphd)) is well above the leakage rate permitted by EPA Victoria, which is 10 lphd for municipal solid waste (MSW) landfills. Furthermore, the time for PFOS to reach the aquifer ranges from 8 years to 74 years, depending on the interface transmissivity rate. These durations need to be longer in the context of MSW landfills in any jurisdiction. The above observations point to the need to adopt a double composite liner to meet the specifications set by the regulators.

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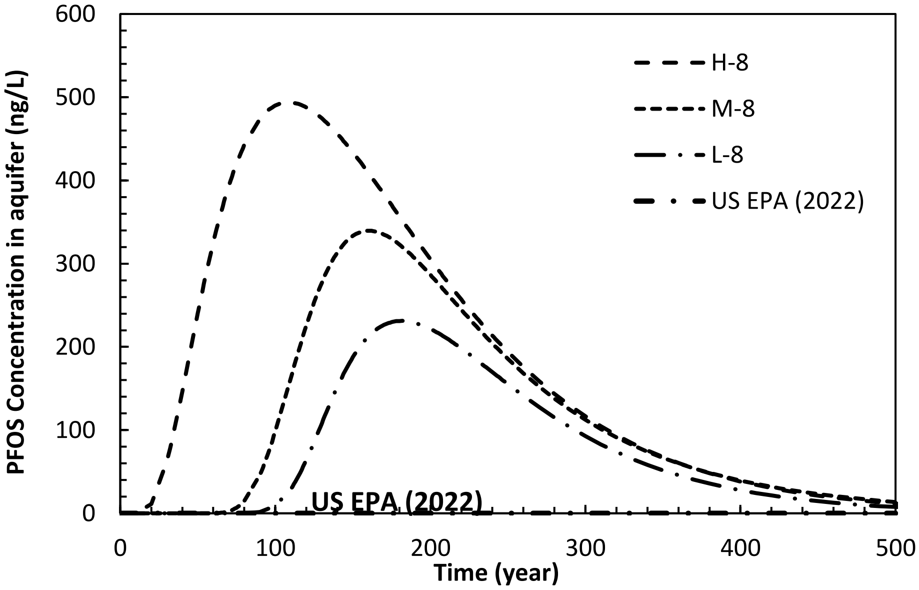

Figure 1. Change in PFOS concentration in the aquifer as a function of time for different leakage scenarios

Table 1. *Results obtained for a range of interface transmissivities, θ (m^2/a) and GCL hydraulic conductivities k_a, in contact with the geomembrane, and k_b, below the wrinkle (q_o = 0.15 ($m^3/m^2/a$ = m/a), c_o = 4800 ng/L, p = 1.2 × 10^{-3} mg/kg, based on Rowe & Barakat (2021).*

Case ID	**θ (m^2/a)**	**k_a (m/s)**	**k_b (m/s)**	**Q (lphd)**	**US EPA[a] t_{allow} (year)**
L-8	0.001	4×10^{-11}	2×10^{-10}	48	74
M-8	0.01	4×10^{-11}	2×10^{-10}	79	54
H-8	0.1	4×10^{-11}	2×10^{-10}	179	8

[a] Allowable PFOS concentration in drinking water in US=0.02 ng/L, transmissivity values based on Rowe & Barakat (2021)

The best case (L-8) from Table 1 was selected to examine the effect of the number of holed wrinkles on the leakage rate and PFOS contamination of the aquifer. Figure 2 depicts the change in PFOS concentration in the aquifer function of time for each number of holed wrinkles considered in the current investigation. The peak PFOS concentrations in the aquifer exceeded permissible PFOS concentration levels allowed by US EPA in all cases.

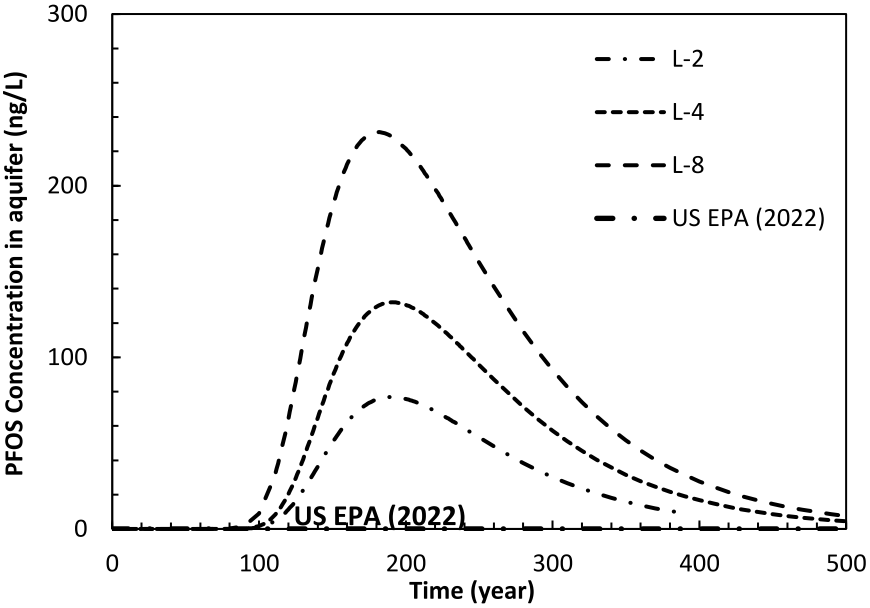

Figure 2. *Change in PFOS concentration in the aquifer as a function of time for different leakage scenarios with different numbers of holed wrinkles*

Table 2 shows results from three cases that examined the effect of the number of holed wrinkles on the leakage rate and PFOS contamination in the aquifer. One can observe that the leakage rate increased as the number of holed wrinkles increased, and the time for PFOS to reach the allowable concentration in the aquifer was shorter as it decreased from 90 years to 74 years.

Table 2. *Results obtained for different number of holed wrinkles (q_o = 0.15 ($m^3/m^2/a$ = m/a), c_o = 4800 ng/L, p = 1.2 × 10^{-3} mg/kg, values from Rowe & Barakat (2021).*

Case ID	**θ (m^2/a)**	**k_a (m/s)**	**k_b (m/s)**	**Q (lphd)**	**US EPA[a] t_{allow} (year)**
L-2	0.001	4×10^{-11}	2×10^{-10}	12	90
L-4	0.001	4×10^{-11}	2×10^{-10}	24	81
L-8	0.001	4×10^{-11}	2×10^{-10}	48	74

[a] Allowable PFOS concentration in drinking water in US=0.02 ng/L

Figure 3 shows that the probability of exceeding regulatory limits set by different jurisdictions and the conditions modelled is 99% for US EPA, UK and Denmark, 96% for Italy, 94% for Japan, 91% for Australia, 87% for Germany and Sweden, and 72% for the Netherlands. This indicates that a single composite liner will likely not meet the specifications set in the US EPA interim guidelines released in June 2022 for allowable PFOS concentration levels in drinking water. As suggested above, a double composite liner system may be required to achieve a very low permissible level set by US EPA (i.e. 0.02

ng/L). However, European countries (Denmark, Italy, Germany, Sweden, and Netherlands) generally rely on a thicker attenuation layer, so the probability is likely lower than reported here.

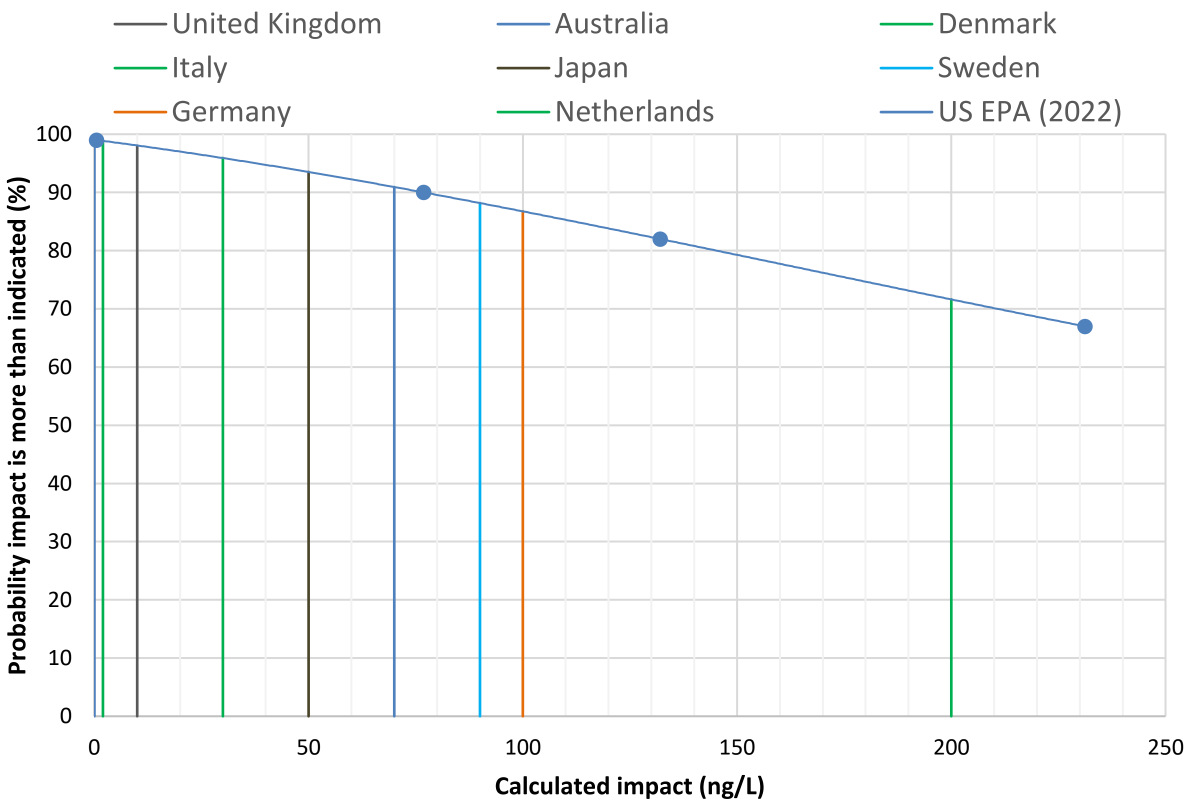

Figure 3. *Calculated probability of exceeding a specific PFOS concentration in the aquifer for the conditions examined based on the probability of leakage deduced from* Gilson-Beck (2019) *and Rowe & Barakat (2021)*

4 CONCLUSIONS

This study investigated the effect of PFOS concentration on the contaminating lifespan of a landfill cell where the bottom liner was composed of a GMB-GCL composite liner. The salient conclusions that can be drawn from this investigation are:

1. A single composite liner, consisting of a 2 mm thick HDPE geomembrane devoid of defects and a GCL, was found to be an effective barrier against PFOS diffusion. However, it would only meet regulatory limits in terms of allowable concentration levels set by the new US EPA interim guidelines for 174 years (i.e. short contaminating lifespan). Also, the peak PFOS concentration in the aquifer would extensively exceed the allowable PFOS concentration level.

2. The allowable PFOS concentration limits set in the recent US EPA interim guidelines are so stringent that all the examined leakage scenarios through a GMB-GCL composite liner exceeded the concentration threshold when considering holed wrinkles. Thus, a double composite liner would be necessary to contain PFOS.

Overall, the results obtained from this study showed that the contaminating lifespan of landfills with a single composite liner consisting of a GCL and a geomembrane is very short, even under the more stringent CQA conditions when the US EPA PFOS concentration limit was considered. Therefore, the performance of a single composite liner needs to be improved to meet the PFOS concentration recommended by the US EPA. This may be accomplished using a sorptive GCL or equivalent in single composite liner systems. In past studies, in cases where the transport of contaminants cannot be managed appropriately, it was found that GCL modification helped increase the contaminating lifespan of landfills with single composite liner systems (Lake & Rowe, 2005). Further research is needed to understand the impact of sorptive materials on the contaminating lifespan of landfill with single composite liners. Otherwise, switching to a double composite liner system will be inevitable to manage US EPA interim PFOS concentration limit.

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