

EMERGING CONTAMINANT ARTICLE

TOPICAL COLLECTION ON PFAS ANALYTICS AND TREATMENT

Foam fractionation removal of multiple per- and polyfluoroalkyl substances from landfill leachate

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are a common contaminant in municipal landfill leachate and are recognized as a pollutant on global scale. The present work examined foam fractionation (FF) in batch and continuous modes as an appropriate treatment technique for PFAS removal for the landfill leachate and found stable removal efficiency of greater than 90% for PFOA (C₇), PFOS (C₈), PFHxS (C₆), and PFHpA (C₆) and 6:2 FTSA (C₆). For other PFAS such as PFNA (C₈), PFPeS (C₄), PFHxA (C₅), PFHpS (C₇), and PFBS (C₄), a less stable removal between 80% and 50% was achieved while between 50% and 20% removal was observed for EtFOSAA (C₈), PFBA (C₃), PFDA (C₉), FOSA (C₈), PFPeA (C₄), and MeFOSAA (C₈). Increased air flowrate, addition of iron (III) oxide (Fe⁺³) coagulant, conductivity, and greater untreated leachate PFAS concentration were factors resulting in increased removal efficiency for the majority of PFAS.

KEYWORDS

foam fractionation, landfill leachate, PFAS, removal, surface active foam fractionation, treatment techniques

1 | INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are a common contaminant in municipal wastewater, municipal solid waste (MSW) landfill leachate, and the groundwater underlying MSW landfills (Ahrens, 2011; Ahrens et al., 2015; Ahrens & Bundschuh, 2014; Busch et al., 2010; Moody et al., 2003; Oliaei et al., 2012; Pan et al., 2016; Schultz et al., 2006; Solo-Gabriele et al., 2020). PFAS can be categorized by functional group perfluoroalkyl carboxylates (PFCA) or perfluoroalkyl sulfonates (PFSA) and by their carbon chain length (Buck et al., 2011). Typical concentrations

in landfill leachate and underlying groundwater range from the 100's to the 1000's ng L⁻¹ (Benskin et al., 2012; Hamid et al., 2018; Huset et al., 2011; Lang et al., 2017; Yan et al., 2015) but may occur at levels greater than 100,000 ng L⁻¹ (Oliaei, 2006) and are typically dominated by C₄-C₇ PFCA (Hamid et al., 2018). Perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) are recognized as persistent organic pollutants by Stockholm Convention (Stockholm Convention, 2009) and the recent revision in 2021 of the European drinking water quality standard EU-Directive 98/83/EC (EU, 2020) expanded the number of regulated PFAS setting a limit of 100 ng L⁻¹ for the sum of a

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select 20 PFAS (Table S1) and a total limit of 500 ng L⁻¹ for total organic fluorine. The United States Environmental Protection Agency (USEPA) has issued drinking water Lifetime Health Advisories for PFOS and PFOA (USEPA, 2016a, 2016b) and in February 2021 stated that PFOS and PFOA will be included in the preliminary determination procedure for promulgation of new drinking standards while hazard assessments are being prepared for additional PFAS (USEPA, 2021). Especially relevant for leachate and wastewater, the Swedish Agency for Marine and Water Management promulgated in accordance with EU-Directive 2013/39/EU (EU, 2013) an environmental quality standard of 0.65 ng L⁻¹ for the average annual PFOS concentration in inland surface waters (HVMFS, 2019) which has made PFAS removal a priority for Swedish municipalities which discharge landfill leachate or wastewater into the environment.

Proven methods for PFAS removal include adsorption via granular active carbon (GAC), powdered activated carbon (PAC), and anion exchange (AIX) or via size exclusion using reverse osmosis (RO) or nanofiltration (NF) (Appleman et al., 2013; Appleman et al., 2014; Belkouteb et al., 2020; Crone et al., 2019; Du et al., 2014; Franke et al., 2019; Franke et al., 2021; Kucharzyk et al., 2017; McNamara et al., 2018; Ochoa-Herrera & Sierra-Alvarez, 2008; Rahman et al., 2014; Senevirathna et al., 2010; Woodard et al., 2017; Zaggia et al., 2016; Zeng et al., 2020) and have been utilized in full-scale predominantly for drinking water and groundwater treatment. For GAC and AIX adsorption processes, PFAS removal efficiency has been shown to be dependent on the PFAS chain length, functional group, pH, and is typically negatively affected by the presence of dissolved organic matter (DOC) (Dixit et al., 2019; Gagliano et al., 2020; Kothawala et al., 2017; McCleaf et al., 2017; Schuricht et al., 2017; Siriwardena et al., 2019) and other negatively charged ions. Full-scale implementation of GAC, AIX, or RO processes for treatment of municipal landfill leachate or wastewater is problematic due to the challenging water matrix, which compared to drinking water or groundwater, typically has higher levels of DOC, suspended solids (SS), salts, nutrients (i.e. nitrate, ammonia, phosphate), heavy metals, phenolics, hydrocarbons, pesticides, phthalates, and pharmaceuticals (Wei et al., 2019). Thus, GAC, AIX, RO/NF treatment techniques typically are not suitable for direct application on leachate or wastewater without extensive pretreatment to prevent hydraulic clogging by SS, DOC, biofouling, and fouling due to iron or carbonate precipitation (Appleman et al., 2014). An alternative PFAS removal method not similarly adversely affected by SS, DOC, nutrient concentrations, or other components in landfill leachate is foam fractionation (FF).

FF can be used in continuous or batch mode, wherein gas bubbles are introduced into the liquid to be treated.

Article Impact Statement

Efficient PFAS removal from landfill leachate using surface-active foam fractionation treatment with examination of key process parameters.

The surface-active molecules in the liquid attach to the gas-liquid interface of the bubbles and form a foam on the liquid surface (Burghoff, 2012; Lemlich, 1968; Leonard & Lemlich, 1965). The foam once removed from the retentate and collapsed is referred to as foamate and the treated liquid as retentate. The optimization of FF has been investigated for the effect of pH, gas flow rate, temperature, bubble size, and additives (Merz et al., 2009, 2011). Typically an increase in gas flow rate increases recovery since more hydrophobic bubble surface area is present but decreases the foamate concentration since there is greater entrainment of interstitial liquid in the foam (Brown et al., 1999; Burghoff, 2012; Lemlich, 1968; Merz et al., 2011; Shea et al., 2009). Greater recoveries can be attained by creating greater bubble surface area, i.e. by using smaller gas bubbles, and lower gas flow rates can provide longer residence time for bubbles in the liquid allowing more time for adsorption to the bubble surface (Chai et al., 1998; Merz et al., 2009).

Application of FF for PFAS removal has been implemented in laboratory scale using metallic activators such as iron (III) chloride (FeCl₃) for batch removal (Lee et al., 2017) from PFOS and PFOA spiked deionized water at pHs greater than seven with up to 90% and 91% removal, respectively. Meng et al. (2018) reported removal efficiencies of 93%, 50%, and 30% for PFOS, perfluorohexane sulfonate (PFHxS), and perfluorobutane sulfonate (PFBS), respectively, from AFFF-spiked wastewater after 2 h of aeration (Meng et al., 2018). Dai et al. (2019) using AFFF-spiked tap water reported 81% removal of total PFAS consisting of PFHxS, perfluoroheptane sulfonate (PFHpS), PFOS, perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), and PFOA in a bench scale continuous process with 20 min. residence time and observed greater removal efficiency for PFSA than PFCA. Ebersbach et al. (2016) found over 99% removal of PFOS, PFOA, and 6:2 fluorotelomer sulfonate (FTSA) for electroplating process wastewater (Ebersbach et al., 2016) and Robey et al. (2020) found selective removal and an average removal efficiency of 69% for native and spiked PFAS for 10 replicate lab-scale foaming experiments from a 20 L sample of landfill leachate (Robey et al., 2020).

FF has not been implemented at full-scale for municipal landfill leachate or wastewater treatment and the inclusion of FF in the overall treatment train has yet to be investigated. FF-treated retentate would most likely require additional treatment stages for removal of organics, nutrients,

metals, or polishing for additional removal of PFAS, thus FF might be best utilized early in the overall treatment train in order to minimize PFAS concentrations in sludge or other residuals created by downstream processes, for example, an activated sludge process. In order to obtain complete PFAS removal from the environment, PFAS laden foamate can be fractionated in series (primary, secondary, tertiary, etc.) in order to increase enrichment and decrease foamate volume with the aim to apply advanced destruction methods such as chemical oxidation, chemical reduction, electrochemical, and sonochemical methods or incineration (Kucharzyk et al., 2017; Lu et al., 2020).

The present study investigated the use of FF as a method for PFAS removal from MSW landfill leachate by performing 13 batch column tests and 5 continuous flow column tests using air bubbles for removal of native PFAS present in the leachate with the goal to provide information for aiding in up-scaling of the process. The efficiency and selectivity of FF for removal of 16 PFAS of five different classes was studied along with the effect of salt content, iron chloride coagulant addition, initial PFAS concentration and operational parameters such as residence time, water depth and airflow. The present study is unique in that removal of native PFAS was investigated for an existing MSW site where the PFAS concentrations in the leachate are variable. Both batch and continuous mode FF were utilized in order to compare both modes of operation and to determine design parameters for a future full-scale FF process at Uppsala Water and Waste's landfill facility at Hovgården, Sweden.

2 | EXPERIMENTAL SETUP

2.1 | Leachate collection

Leachate water was collected for the experiments at Hovgården landfill at the inlet chamber to the existing leachate treatment plant receiving the combined flow from the landfill's two leachate pump stations using a manual action suction pump and stored in 25-liter polyethylene high-density (PE-HD) plastic containers and kept in the dark at room temperature, approximately 18°C. Leachate collection was performed on April 28, May 11 and 18, and June 3, 2020, and the storage time before individual batch tests was from 1 to 10 days while for the individual continuous tests from 1 to 2 days. The containers were thoroughly mixed for approximately 5 min before the start of each column experiment.

2.2 | Column experiments

The FF column experiments were performed in batch mode and continuous flow mode, the batch mode setup is shown in Figure 1 and the continuous mode setup in Figure S1 in the SI. Both systems introduced oil-free air into the bottom of the column (acrylic plastic, inner diameter 27 mm, height = 2 m) through a membrane (Xylem Silver Series II®). Airflow was measured using a rotameter (ZYIA instrument company, FL3-1) with air bubbles moving upward through the water column with

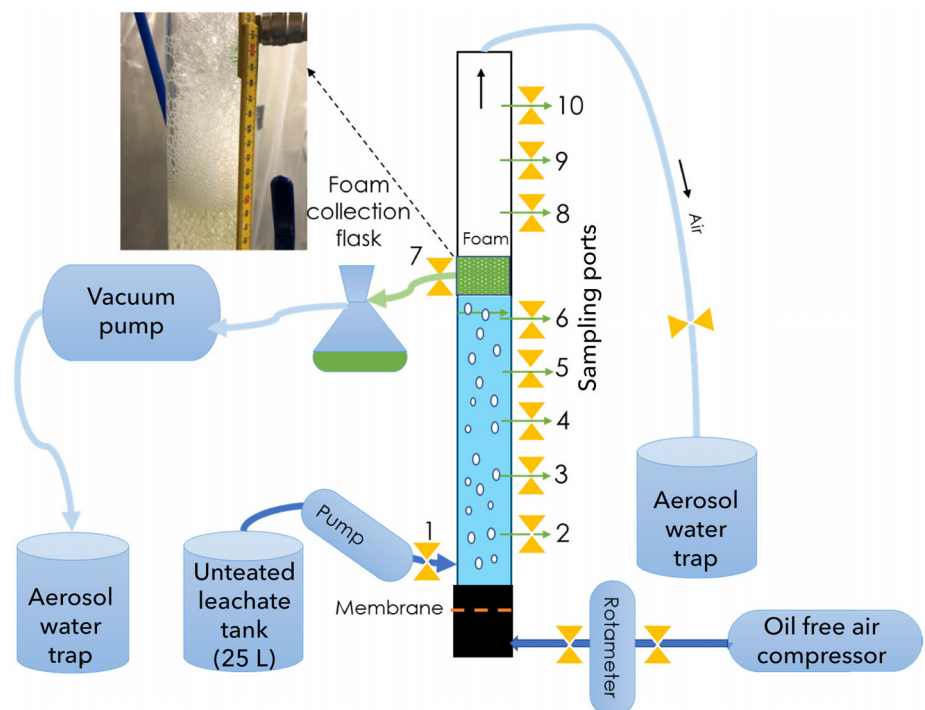


FIGURE 1 Experimental setup for foam fractionation batch mode

air exiting the column through an aerosol water trap. Foam forming on the water surface was removed from the column using a vacuum pump (GAST, Model DOA-P504-BN) with silicone suction tube inserted through one of the sampling ports 2 through 10. All leachate and foamate samples were weighed using a Mettler Toledo® scale (Model PB602-S/FACT) and untreated and treated leachate were measured for pH and temperature (Knick Portavo 902 PH) and conductivity and temperature (WTW Cond 340i). A Watson-Marlow Pumps Group peristaltic pump (323E/D 400RPM) was used for filling the column for batch tests and pumping water through the column for continuous tests.

2.3 | Batch mode column experiments

For the 13 batch mode tests, the column was filled with leachate water using the peristaltic pump through port 1 (see Figure 1) to the desired column height of 0.5 m (1.2 L volume) or 1.0 m (2.4 L volume). Sampling of the treated column water was performed by removing 5 ml at sampling port two into a 250 ml PE-HD bottle, which was weighed, then the sample was diluted using Milli-Q® water and weighed again. Foam was harvested continuously between each sampling time step in a polypropylene (PP) foam collection flask, transferred to a 250 ml PE-HD bottle, weighed, then diluted using Milli-Q® water and weighed again. Sampling time steps for foam and column water were 5, 10, 20, 40, and 60 min from the start of air inflow. A 250 ml untreated water sample was taken from sampling port two before the start of air inflow for the initial PFAS concentration.

For the batch mode, the parameters tested were air flow rate, water column height, iron chloride coagulant addition, and effect of salinity; details are listed in Table 1. For the leachate concentration tests, the leachate

was diluted with Milli-Q® water to achieve 50% and 25% leachate solutions. The FeCl₃ coagulant used was Kemira Pix 311®, 37%–44% FeCl₃ and for ionic strength tests the table salt was used (sodium chloride [NaCl] with iodine 0.005% and lump inhibitor E535) to increase the conductivity by 50% and 100%. Batch tests were conducted for a minimum of 60 min.

2.4 | Continuous mode column experiments

For the five continuous mode tests, the column was operated with air flow counter-current to water flow wherein the peristaltic pump supplied water continuously to the column at a flowrate required to provide the desired contact time of 5, 10, and 20 min for each water column height tested as summarized in Table 2. About 10 ml retentate samples were collected from the exit tube and untreated leachate samples were taken from sample port two at the start of the experiment in the same manner as for the batch experiment including measurement of pH, temperature, and conductivity. Foam was harvested between each sampling interval in the same manner as the batch experiment from port seven or eight depending on the height of the foam.

2.5 | Target compounds

The 16 target analytes included C₃–C₉ PFCA (C_nF_{n+1}COO⁻, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA), C₄–C₈ PFSA (C_nF_{2n+1}SO₃⁻, PFBS, PFPeS, PFHxS, PFHpS, PFOS), 6:2 FTSA (C₆F₁₃CH₂CH₂SO₃⁻), perfluorooctanesulfonamide (C₈F₁₇SO₂NH₂, FOSA) and methyl and ethyl perfluorooctane sulfonamidoacetic acids (C₈F₁₇SO₂N[C_nH_{2n+1}]CH₂CO₂H, MeFOSAA, EtFOSAA).

2.6 | Sampling and analysis

Samples of the raw leachate entering the FF column, retentate leaving the column, as well as fractionated foam

TABLE 1 Batch test parameters

Test parameter	Value	Air flowrate (L min ⁻¹ m ⁻²) ^a
Water column height (m)	0.5	3500, 7000
Water column height (m)	1.0	3500, 7000, 10,500
Iron chloride coagulant g Fe ⁺³ L ⁻¹	0.025, 0.050, 0.100	3500
Ionic strength conductivity (mS cm ⁻¹)	5.0, 7.7, 10.0	3500, 7000

^aAir flowrate expressed as air flow per area of the water column (L min⁻¹ m⁻²).

TABLE 2 Continuous test parameter conditions

Test parameter	Test parameter variation (min)	Water column height (m)	Air flowrate (L min ⁻¹ m ⁻²)
Contact time (min)	5, 10, 20	1.0	7000

was sampled for every batch test and at specific intervals for the continuous testing. Samples were stored at 4 °C in the dark and shipped to ALS Scandinavian located in Stockholm, Sweden, for PFAS analysis (ALS) using liquid chromatography - tandem mass spectrometry (LC-MS/MS). For quality assurance, one blank sample, one spiked sample, and one duplicate sample was analyzed for every 20 samples. Thirty-two PFAS compounds were included in the laboratory analysis as listed in Table S1 in SI. On three occasions, duplicates of the untreated leachate were taken; the average variation in the PFAS concentrations was 7%. For the determination of PFAS removal calculations and graphing, the PFAS results with concentrations lower than the limit of detection (LOD) were treated as having the value of half the LOD which is recognized to introduce bias into calculated removal efficiencies but still indicates the effectiveness of the removal process. For some treated water sample concentrations, i.e. for leachate exiting the columns, the reported PFBA, MeFOSAA, and EtFOSAA concentration was found to exceed the concentration for the leachate entering the column and was assumed to be equal to the concentration of the untreated leachate entering the column for that run since PFAS was assumed not to be generated within the column. This anomaly was probably due to the low levels of PFBA, MeFOSAA, and EtFOSAA in the untreated leachate and was presented as a treatment removal efficiency of zero in the results. General chemical analysis (Table S3) was performed by Uppsala Water and Waste AB's accredited laboratory, Uppsala, Sweden.

The removal efficiency was calculated as shown in Equation (1).

$$\text{Removal efficiency (\%)} = 100 - \frac{C}{C_0}, \quad (1)$$

where C is the PFAS concentration in the batch column water or the continuous exit water (ng L^{-1}) and C_0 is the untreated concentration (ng L^{-1}) before the start of the batch test or the water entering the continuous process.

The foamate to feed water ratio was (FWR) calculated for volume of foamate captured compared to the volume of leachate treated as shown in Equation (2).

$$\text{FWR (\%)} = \frac{V_{\text{foamate}}}{V_{\text{leachate initial}}}, \quad (2)$$

where V_{foamate} is the volume of foamate harvested (ml) and $V_{\text{leachate initial}}$ is the initial volume of leachate treated (ml).

A foamate enrichment factor was calculated as shown in Equation (3).

$$E (\%) = \frac{C_{\text{foamate}}}{C_0}, \quad (3)$$

where C_{foamate} is the concentration in the foamate (ng L^{-1}) and C_0 is the initial PFAS concentration (ng L^{-1}) in the untreated leachate.

The PFAS recovery (R_{PFAS}) for the mass of PFAS in the foamate compared to the mass removed from the water column as shown in Equation (4).

$$R_{\text{PFAS}} (\%) = \frac{\text{Conc. PFAS foamate} \times V_{\text{foamate}}}{\text{Conc. PFAS leachate initial} \times V_{\text{leachate initial}}}, \quad (4)$$

where $C_{\text{PFAS foamate}}$ is the PFAS concentration in the harvested foam (ng L^{-1}), V_{foamate} is the volume of the foam (L), $C_{\text{PFAS initial PFAS}}$ is the initial PFAS concentration in the leachate (ng L^{-1}), and V_{initial} the initial volume of the leachate in the column.

Contact time (C_t) for the water in the continuous column tests was calculated using Equation (5).

$$C_t = \frac{\text{Volume}}{\text{Flowrate}}, \quad (5)$$

where C_t is the contact time (min), volume is the volume of water in the column (L) and the flowrate (L min^{-1}).

Airflow to feed ratio (ASFR) for the continuous tests was calculated using Equation (6).

$$\text{AFR} = \frac{Q_{\text{air}}}{Q_{\text{water}}}, \quad (6)$$

where air AFR is the air to feed ratio ($L_{\text{air}}/L_{\text{leachate}}$), Q_{air} is the air flowrate (L min^{-1}), and Q_{water} is the feedwater flowrate (L min^{-1}).

3 | RESULTS AND DISCUSSION

3.1 | Untreated leachate

The average total PFAS concentration of 6200 ng L^{-1} for the untreated leachate for both the batch and continuous tests consisted of predominantly PFOS followed by PFOA, average concentrations 2500 and 1800 ng L^{-1} , respectively, and which made up 40% and 29%, respectively, of the total measured PFAS (Table S2). The proportion of PFCA, PFSA, and PFAS precursors were 48%, 49%, and 3%, respectively.

As shown in Table S2, PFAS concentrations of the untreated leachate varied over the course of the experiment

as indicated by the standard deviations since fresh leachate was collected as needed from the Hovgården Landfill in order to avoid degradation of the leachate and to validate the variability of PFAS concentrations at the landfill leachate site. The high standard deviations for the untreated water concentrations makes specific comparison between individual tests uncertain due to different initial PFAS concentrations, however, the quantification of the PFAS removal for each run can be accurately quantified and indicates the relative efficiency for the target PFAS. The general water chemistry is given in Table S3 and reflects the challenging water quality matrix of the leachate wherein the average total organic concentration was 42 mg L^{-1} , conductivity 570 mS cm^{-1} , ammonia 51 mg L^{-1} , total iron 3.9 mg L^{-1} , and bicarbonate 1200 mg L^{-1} , and which is typical for Hovgården Landfill's leachate.

3.2 | Batch mode PFAS removal

For the eight batch experiments without salt or FeCl_3 additives, the total PFAS removal efficiency ranged from 72% up to 86% which indicates the reliability of the removal process despite the varying test conditions listed in the Figure 2's sidebar. PFAS removal efficiencies for each batch experiment for each time-step including temperature, pH, and initial conductivity are given in Table S4 together with results from all batch tests performed. As depicted in Figure 2, the total PFAS removal efficiency reached a minimum of 70% within the first 10 min. of the start of airflow; correspondingly an

average of 67% of the total mass of PFAS was captured in the foam during the first 0–5 min as shown in Figure S2 and corresponds to the observations made by Lee et al., 2017. The PFAS removal as a function of chain length is provided in Figure S3.

For the batch experiments, the foamate to feed water ratio averaged 21% as shown in Table S6 and is similar to the 22% value reported by Robey et al. (2020), however the average enrichment factor of 2.9 for all PFAS analyzed was less than the 3.7 estimated from Robey et al. (2020) and significantly less than the factor of 8400 for PFOS reported by Meng et al. (2018) who used a non-ionic hydrocarbon surfactant to enhance removal. Similarly, this work's 21% is less than the enrichment factor of 45 up to 1300 reported by Ebersbach et al. (2016) for 6:2 FTSA enrichment in aerosols which may be attributed to the higher PFAS concentrations and greater ionic strength of the wastewater.

The average enrichment factor of 2.9 was less than the theoretical value of 3.7 calculated assuming all the PFAS removed from the column water was captured in the foamate. Similarly, the PFAS mass balance between treated leachate and foamate averaged 0.72 in Table S6 and indicates that on average approximately 30% less PFAS was measured in the foamate than was removed from the water column. This discrepancy may be due to PFAS migrating to the top of the water column without being captured in the foam while retentate sampling from port two located near the bottom of the column did not include any PFAS in the water column directly under the water surface. This phenomenon may be due to weak

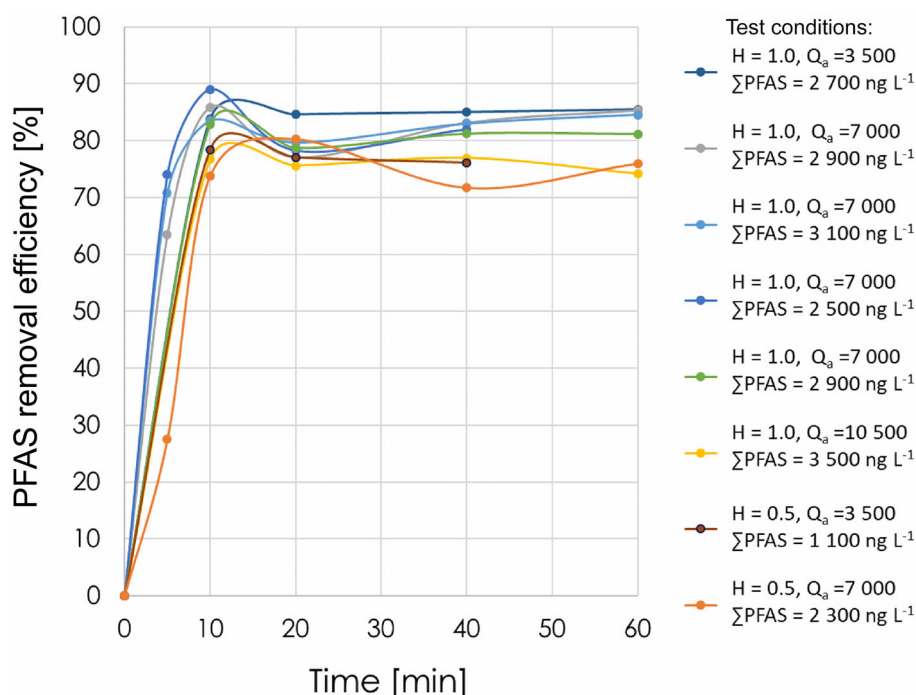


FIGURE 2 PFAS removal efficiency for batch experiments ($n = 8$, 34 samples) with no additives, column water height (H) 0.5 and 1.0 m and air flowrates (Q_a) from 3500–10,500 $\text{L min}^{-1} \text{ m}^{-2}$, initial conductivity 1.6–5.7 mS cm^{-1} , avg. temp. 18 °C, avg. pH 7.8, initial ΣPFAS 1100–3500 ng L^{-1}

foam creation and poor bubble stability due to the low surfactant concentration intrinsic in the landfill leachate as compared to solutions spiked with AFFF or other surfactants.

3.3 | Batch mode effect of water column depth and air flowrate

As shown in Figure 3, the average removal efficiency increases with increasing air flowrate for both the 0.5 and 1.0 meter columns, with the maximum average removal efficiency of 83% for both column heights for the air flowrate of $7500 \text{ L min}^{-1} \text{ m}^{-2}$, an increase of 6% as compared to the efficiency of 76% with $3500 \text{ L min}^{-1} \text{ m}^{-2}$ air flowrate and corresponded to the observation reported by Meng et al. (2018) and Dai et al. (2019), that removal efficiency increased with increasing air flowrate. No increase in efficiency was observed for the 1.0 m column for the air flowrate increase from 7500 and $10,500 \text{ L min}^{-1} \text{ m}^{-2}$ which may indicate an optimum design air flow between these two airflows for this water matrix and FF system. The difference in removal efficiency for water depth 0.5 and 1.0 m for the same airflow rate was less than 1%, thus water depth did not appear to affect removal efficiency.

3.4 | Effect of conductivity

As shown in Figure 4, the average removal efficiency increased with increasing conductivity or ionic strength from approximately 75% at conductivity of 5.7 mS cm^{-1} to approximately 93% for both conductivities of 8.1 and 11 mS cm^{-1} for the $3500 \text{ L min}^{-1} \text{ m}^{-2}$ flow rate, an

increase of 24%. For the higher air flow rate of $7000 \text{ L min}^{-1} \text{ m}^{-2}$ removal efficiency increased from 78% to 85%, respectively, for the increase in conductivity from 1.6 to 3.0 mS cm^{-1} . Interestingly, removal efficiency did not increase for the greatest conductivities for both air flow rates which may indicate a limit or plateau to improved efficiency coupled to increasing conductivity for this water matrix and FF system. The apparent increase in removal efficiency confers with Meng et al. (2018) and Ebersbach et al. (2016) who also reported improved removal efficiency for increasing ionic strength. The PFAS removal as a function of chain length for the two batches with greatest conductivity is provided in Figure S4.

Improved PFAS removal with increasing ionic strength may be due to the resulting increased bubble surface area due to increased solution viscosity as well as due to lowering the critical micelle concentration (CMC) and providing a more stable foam. CMC is the critical concentration above which aggregations of surfactant molecules or micelles spontaneously form. Increased viscosity in this case is due to the addition of sodium salt whereby the inorganic Na^+ ions of high surface charge density may be capable of holding enough water molecules around them to increase the overall amount of structured water, even reducing the electrostatic repulsion between PFAS molecules leading to a decrease in CMC and improved foam formation (Michaux et al., 2018). The increased viscosity also results in decreased bubble size which would increase the total bubble surface area available (Fanaie & Khiadani, 2020) for PFAS attachment for a constant air flowrate. In addition to conductivity, the increase in removal efficiency may also have been affected by the differing initial PFAS concentrations, especially for the two runs with the highest

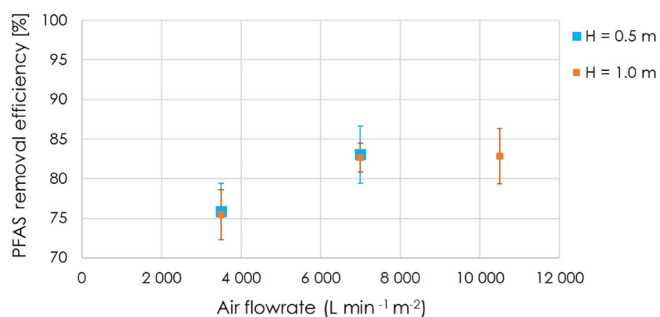


FIGURE 3 Average PFAS removal efficiency for batch experiments ($n = 5$, 19 samples) with varying water height (H) and air flowrate ($\text{L min}^{-1} \text{ m}^{-2}$), avg. initial conductivity $5.6\text{--}5.7 \text{ mS cm}^{-1}$, avg. temp. 17°C , avg. pH 7.8, initial $\sum \text{PFAS}$ $2300\text{--}3100 \text{ ng L}^{-1}$

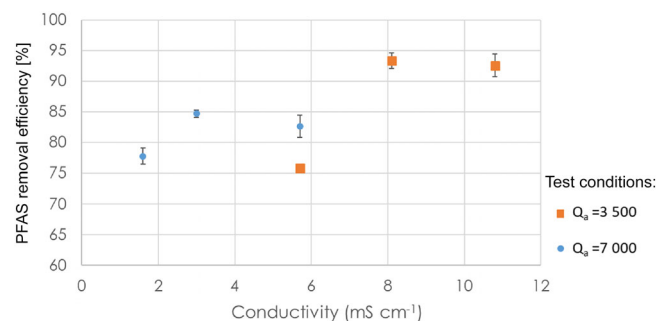


FIGURE 4 Average PFAS removal efficiency for batch experiments ($n = 6$, 21 samples) with varying conductivities, water height 1.0 m, air flowrate (Q_a) 3500 and $7000 \text{ L min}^{-1} \text{ m}^{-2}$, avg. temp. 18°C , avg. pH 7.7, initial $\sum \text{PFAS}$ $1100\text{--}15,300 \text{ ng L}^{-1}$. Error bar is standard deviation all experimental datapoints

removal efficiency which had the greatest total PFAS concentrations.

3.5 | Effect of iron chloride coagulant

The effect of addition of Fe^{+3} coagulant chemical is shown in Figure 5 in SI and as a function of chain-length in Figure S5 and indicates an increase of PFAS removal efficiency from 75% for a no addition of FeCl_3 coagulant up to 88% removal efficiency for doses of 0.05 and 0.1 $\text{g Fe}^{+3} \text{ L}^{-1}$ leachate, an increase of approximately 13%. Comparison of the average removal efficiency for Figure S5 with batch tests without Fe^{+3} addition, Figure S3, shows the greatest increase in efficiency 65% for FOSA (C_8), 62% for PFDA (C_9) followed by 45% for EtFOSAA, (C_8), 36% for PFHpS (C_7), and 28% for MeFOSAA (C_8).

Increased removal efficiency with Fe^{+3} was also reported by Lee et al. (2017) and may be due the trivalent iron neutralization of the negatively charged functional group of the PFAS thereby increasing hydrophobicity of the PFAS compounds (Campos-Pereira et al., 2020) and the Fe^{+3} neutralization of negatively charged organic carbon particles in the water column resulting in incorporation of these into the foam via a floatation process (Crittenden et al., 2012; Valade et al., 1996). The end result being a more stable attachment of PFAS to the bubble surface and a foam with greater organic carbon content which provides an additional mechanistic PFAS attachment site via absorption to organic carbon (Campos Pereira et al., 2018; Higgins & Luthy, 2006).

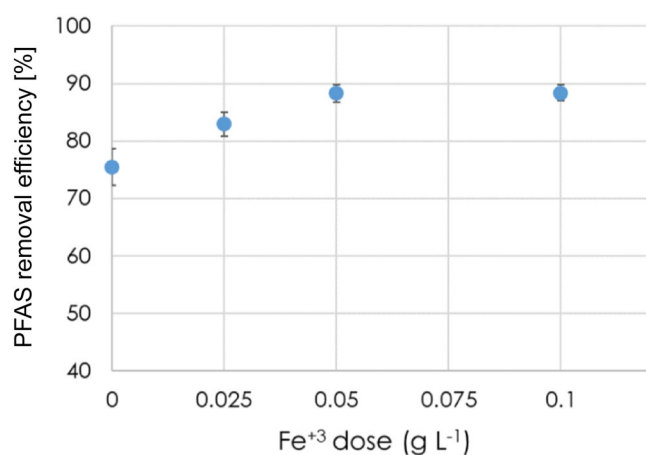


FIGURE 5 Average PFAS removal efficiency for batch experiments ($n = 4$, 13 samples) with varying Fe^{+3} coagulant dose, water height (H) 1.0 m, air flowrate (Q_a) $3500 \text{ L min}^{-1} \text{ m}^{-2}$, initial conductivity 5.7 mS cm^{-1} , temp. $18 \text{ }^\circ\text{C}$, avg. pH 7.8, initial $\sum\text{PFAS}$ $2400\text{--}5600 \text{ ng L}^{-1}$. Error bar is standard deviation all experimental datapoints

3.6 | Continuous flow mode PFAS removal

Continuous flow experiments shown in Figure 6 indicate a stable removal of PFAS from the feed water from after the time the test's CT is reached with removal efficiencies from 73% up to 97% with volumetric ratio of leachate feed flow to air flow rates (AFR) of 4, 8, and $16 \text{ L}_{\text{air}} \text{ L}_{\text{leachate}}^{-1}$ and contact times (CTs) from 5 up to 20 min, respectively. This is similar to the result of 81% total PFAS removal (i.e. PFHxS (80%), PFHpS (100%), PFOS (87%), PFHxA (17%), PFHpA (50%), and PFOA (91%)) reported by Dai et al. (2019) for a continuous process with 20 min CT and airflow to feed ratio of 3. The total PFAS removal efficiency appears to be dependent on the PFAS makeup of the untreated leachate, with greater total PFAS removal if untreated leachate contains PFAS with relatively high removal efficiencies such as of PFHpA, PFOA, PFHxS, PFOS, and 6:2 FTSA. Increasing contact time did not result in a greater removal efficiency and indicates that other parameters such as initial PFAS concentration, metal content, conductivity, DOC, or even suspended solids may play a greater role than CT in providing robust foam formation and greater PFAS removal efficiency. The PFAS removal efficiencies for each continuous mode experiment for each time-step including initial temperature, pH, and conductivity are given in Table S5 and results as a function of chain length in Figure S6.

3.7 | Overall PFAS removal efficiency, chain-length, and air-water partitioning coefficient

As summarized in Table 3, the average removal was 83% for all PFAS which is comparable to the 81% removal reported by Dai et al. (2019), while removal efficiency for PFSA was greater than for PFCA, 94% versus 77%, respectively, which also agrees with Dai et al. (2019). PFOS removal efficiency was greater than 98% and PFOA 99% which is similar to the 99% for PFOS removal reported by Meng et al. (2018) for PFOS in wastewater and 99% removal for PFOS and PFOA reported by Lee et al., 2017 using trivalent iron as a removal aid. The variability of the results expressed as relative standard deviation (RSD) in Table 3 indicates that removal efficiency of PFHpA, PFOA, PFHxS, PFOS, and 6:2 FTSA with $\text{RSD} \leq 10\%$ was consistently achieved. PFAS such as PFBA, PFPeA, PFDA, FOSA, MeFOSAA, and EtFOSAA had RSDs greater than 50% and the removal efficiencies reported in Table 3 for these PFAS should be considered much less certain. Note that of PFHpA,

FIGURE 6 PFAS removal efficiency for continuous flow experiments ($n = 5$, with 20 samples), no additives, water height 1.0 m, air flowrate $3500 \text{ L min}^{-1} \text{ m}^{-2}$, initial conductivity $5.3\text{--}5.4 \text{ mS cm}^{-1}$, avg. temp. 19°C , pH 7.7, initial ΣPFAS $3200\text{--}25,200 \text{ ng L}^{-1}$

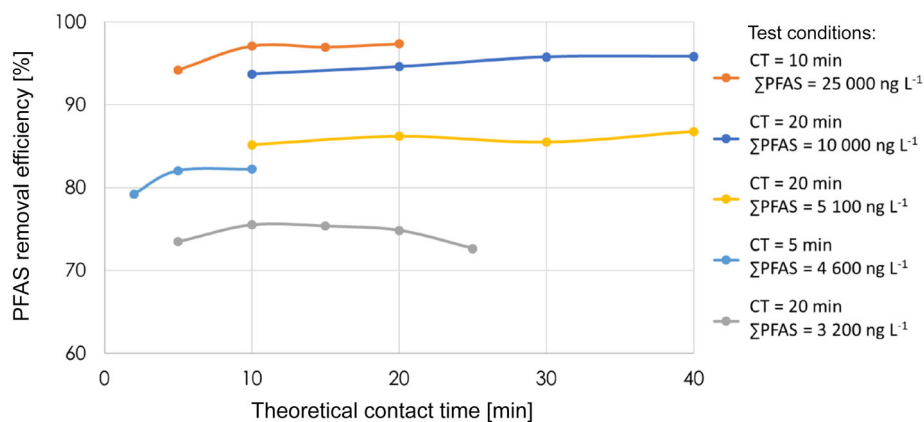


TABLE 3 Average removal efficiency versus perfluoroalkyl chain length based on eight batch mode and five continuous mode tests without additives ($n = 44$ samples), avg. initial conductivity 5.0 mS cm^{-1} , avg. temp. 18°C , pH 7.7, initial ΣPFAS $1100\text{--}25,200 \text{ ng L}^{-1}$

PFAS compound	Perfluoroalkyl chain length	Avg. %	SD	RSD
PFBA	C ₃	38	31	82
PFPeA	C ₄	30	20	67
PFHxA	C ₅	66	24	36
PFHpA	C ₆	93	7	8
PFOA	C ₇	99	1	1
PFNA	C ₈	76	17	22
PFDA	C ₉	39	38	97
<i>Sum PFCA</i>		77	8	10
PFBS	C ₄	60	26	43
PFPeS	C ₅	73	27	37
PFHxS	C ₆	97	2	2
PFHpS	C ₇	64	29	45
PFOS	C ₈	98	2	2
<i>Sum PFSA</i>		94	4	4
6:2 FTSA	C ₆	90	9	10
FOSA	C ₈	32	41	128
MeFOSAA	C ₈	11	24	218
EtFOSAA	C ₈	40	34	85
<i>Sum precursors</i>		68	20	29
<i>Total PFAS</i>		83	7	8

Abbreviations: Avg., average removal efficiency; RSD, relative standard deviation; SD, standard deviation.

PFOA, PFHxS, PFOS, and 6:2 FTSA made up approximately 80% of the total PFAS in the untreated leachate (Table S1) at the site.

Neither chain-length as shown in Figure 7 or air-partitioning coefficient (K_{aw}) (Figure S7) appears predictive of PFAS removal; however, for design purposes, PFAS may be divided into three groups as shown in Figure 7 in when considering FF treatment of the leachate at the site.

Specifically, the greatest average removal efficiency 80–98% are indicated for PFOA (C₇), PFOS (C₈), PFHxS (C₆), and PFHpA (C₆) and 6:2 FTSA (C₆), 80–50% for

PFNA (C₈), PFPeS (C₄), PFHxA (C₅), PFHpS (C₇), and PFBS (C₄), and finally 50–20% for EtFOSAA (C₈), PFBA (C₃), PFDA (C₉), FOSA (C₈), PFPeA (C₄), and MeFOSAA (C₈). Similar results were reported for spiked leachate by Robey et al. (2020), specifically, greater than 90% removal for PFOA, PFOS, PFHxS, and PFHpA, while PFHpS had lower removal efficiency than that of PFOS or PFHxS, and less than 10% removal efficiency for PFBA, PFPeA. Ebersbach et al., 2016 also reported similar results including over 90% removal for 6:2 FTSA.

For planning of leachate treatment using FF, removal efficiency appears dependent on raw leachate PFAS

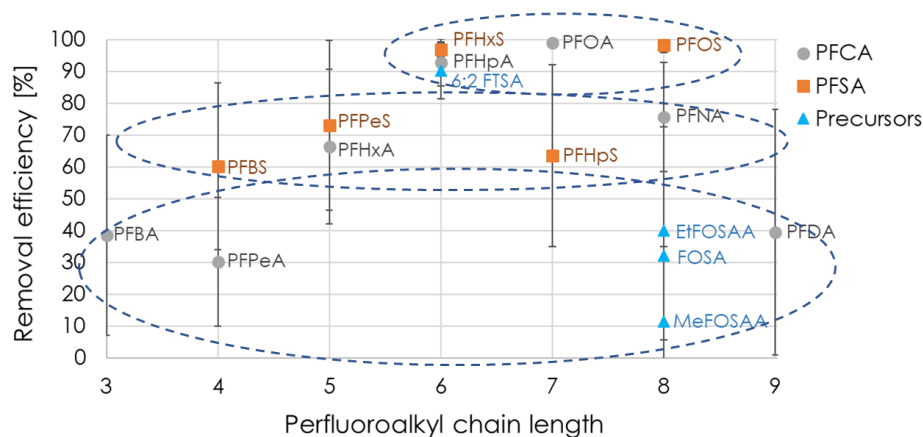


FIGURE 7 Average Removal efficiency as a function of PFAS perfluoroalkyl chain-length, based on eight batch mode and five continuous mode tests without additives ($n = 44$ samples), avg. initial conductivity 5.0 mS cm^{-1} , avg. temp. 18°C , avg. pH 7.7, initial $\sum \text{PFAS}$ 1100–25,200 ng L^{-1} . Error bar is standard deviation all experimental datapoints

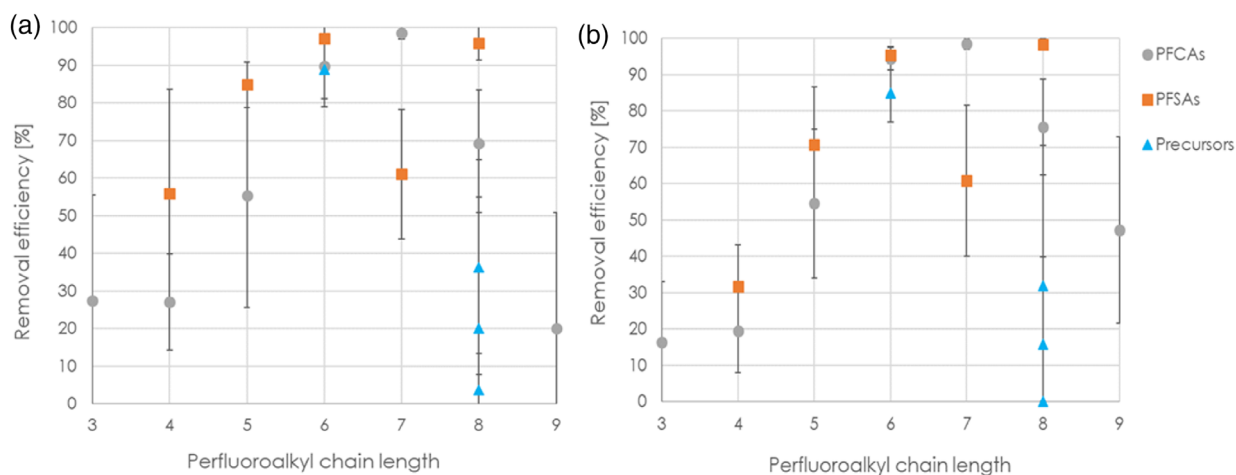


FIGURE 8 Average PFAS removal efficiency as a function of perfluoroalkyl chain length for experiments with no additives for (a) two batch experiments ($n = 8$) with no additives, $H = 1.0 \text{ m}$, $Q_a = 3500 \text{ L min}^{-1} \text{ m}^{-2}$, cond. 5.7 mS cm^{-1} , temp. 18°C , pH 7.8, initial $\sum \text{PFAS}$ 2300–3000 ng L^{-1} , and (b) two continuous flow experiments, $CT = 20 \text{ min}$ (eight effluent samples) with no additives, $H = 1.0 \text{ m}$, $Q_a = 3500 \text{ L min}^{-1} \text{ m}^{-2}$, avg. cond. 5.5 mS cm^{-1} , temp. 19°C , pH 7.7, initial $\sum \text{PFAS}$ 3300–5100 ng L^{-1} . Error bar is standard deviation all experimental datapoints

concentration with greater overall PFAS removal efficiencies for batches with greatest initial raw leachate PFAS concentrations as shown in Figure S8. Total PFAS removal efficiencies greater than 90% were achieved for some runs since the increase in raw leachate PFAS concentration was primarily due to increases in PFOA (C_7), PFOS (C_8), PFHxS (C_6), and PFHpA (C_6) which have the greatest removal efficiencies. This illustrates the impact of the composition of total PFAS in the feed water on overall removal efficiency. PFAS such as PFDA, PFNA, PFHpS, FOSA, 6:2 FTSA, and MeFOSAA, also appear to have a removal efficiency dependent on their respective feed concentration as shown in Figure S9 with removal efficiency of greater than 90% achieved after a concentration of greater than 50 ng L^{-1} is reached for each respective compound, but which may also be an artifact from the laboratory analysis and detection limits.

3.8 | Batch versus continuous mode removal efficiency

The removal efficiencies for the PFSA, PFCA, and PFAS precursor are shown in Figure 8a for two batch mode and Figure 8b for two continuous mode experiments with similar conditions, i.e. conductivity, pH, air-flowrate, column depth, and initial PFAS concentration. For both modes, the greatest removal efficiency was achieved for PFOA (C_7), PFOS (C_8), PFHxS (C_6), and PFHpA (C_6) with the average of both modes being a minimum of 98%, 96%, 95%, and 90% as shown in Table S7. For the batch mode the overall PFAS removal average was 76% while the continuous mode was 81% which indicates continuous mode can provide similar removal efficiency as the batch process. With the batch mode greater removal efficiency was achieved for the C_3 and C_4 PFAS and a C_3 ,

C₄ PFSA, lesser improvement for the C₈ precursors appears for the batch system which may indicate the batch system is more effective at removing these PFAS.

4 | CONCLUSIONS AND FUTURE CHALLENGES

Based on results of the present work, FF appears to be an effective PFAS removal technique providing greater than 90% removal for PFOA (C₇), PFOS (C₈), PFHxS (C₆), and PFHpA (C₆) and 6:2 FTSA (C₆). Other PFAS such as PFNA (C₈), PFPeS (C₅), PFHxA (C₅), PFHpS (C₇), and PFBS (C₄) are also removed but with efficiencies that are more variable and between approximately 50–80%. The PFAS EtFOSAA (C₈), PFBA (C₃), PFDA (C₉), FOSA (C₈), PFPeA (C₄), and MeFOSAA (C₈) also displayed variable removal efficiency between approximately 0–50%. Removal efficiency appears not to be correlated to chain-length or air-water partitioning coefficient, but to the magnitude of the untreated PFAS concentration, thus identification of predominant PFAS compounds, their concentrations and variability is important before considering implementation of FF for a specific site. The PFAS removal for both batch and continuous mode using FF occurs rapidly with the majority of PFAS removed within 10 min of introduction of air bubbles which allows intermittent operation without loss of removal efficiency. FF process removal efficiency increases with increased air-flow, addition of Fe⁺² coagulant, and increase in conductivity until a limit or plateau is reached which may indicate design limitations for these parameters when considering up-scaling of the process. Also important for FF upscaling, there was no significant difference between removal efficiency for a water column depth of 0.5 or 1.0 m or between continuous tests with longer contact times. Utilization of continuous mode FF appears to be almost equally efficient as the batch mode, 76% versus 81% total PFAS removal efficiency, respectively, with the same PFAS constituents i.e. PFOA (C₇), PFOS (C₈), PFHxS (C₆), and PFHpA (C₆) achieving the greatest removal efficiencies.

Based on the present work, FF appears to provide a viable PFAS removal process to deal with difficult water quality matrices such as MSW landfill leachate, however, future work on optimization of the process should focus on improvement of removal efficiency for EtFOSAA (C₈), PFBA (C₃), PFDA (C₉), FOSA (C₈), PFPeA (C₄), and MeFOSAA (C₈), investigation of the impact of bubble size, addition of surfactants to improve foam formation as well as improved collection of the fractionated foam. The variability in the removal efficiency of some PFAS may be due to weak foam creation or poor bubble stability and

resulting congregation of some PFAS directly under the retentate surface without incorporation into the foam. This phenomenon should be examined in order to determine if harvesting of foam along with the top layer of the water column would provide improved PFAS removal.

Although not addressed in the present work, the results point to potential application of FF for PFAS removal for existing treatment trains such as the activated sludge process and reverse osmosis (RO)/nanofiltration (NF) membrane concentrate treatment. Successful upscaling of continuous FF for municipal leachate and wastewater plants appears promising since existing technology for bubble aeration for waste-activated sludge full-scale processes is very similar, however, efficient methods of foam capture have yet to be developed for large-scale application. Application of the FF process in series should be examined, wherein the foamate volume could be reduced so as to make utilization of advanced oxidation or destruction processes economical and avoid PFAS discharge into the environment. Additionally, the total operational cost should be determined in order to make cost comparisons to conventional PFAS removal techniques such as GAC and AIX.

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CONFLICT OF INTEREST

The authors have no conflict of interest to declare.

DATA AVAILABILITY STATEMENT

Data available on request from the authors

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SUPPORTING INFORMATION

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