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Technical Memorandum

Prepared for:	Maine Department of Environmental Protection, Bureau of Remediation and Waste Management
Project Title:	PFAS Treatment of Dairy Farm Waste-Treatment Decision Support
Project No.:	157360

Technical Memorandum

Subject:	Evaluation of Available Information and Recommended Path Forward
Date:	September 30, 2021
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Executive Summary

This Technical Memorandum (TM) provides a review of proposals offered by two suppliers (ACV Enviro (ACV) and ECT2) to manage a combined manure/dairy waste that contains elevated concentrations of per- and polyfluoroalkyl substances (PFAS). The overall objective is to remove solids and aqueous waste from a contaminated manure pit such that the pit is available for use by the dairy farm. In addition to reviewing the proposals, this TM describes activities to date by the two proposers as well as a recommended path forward based on Brown and Caldwell's (BC) dairy waste and PFAS experience. Key findings are:

1. The proposed approach by ACV had not adequately considered the characteristics of the contaminated manure pit. Specifically, the characteristics of the solids as a result of on-going digestion of the combined manure/milk waste resulted in solids that would not separate via gravity with the addition of a polymer. Testing to determine the most suitable polymer (or coagulant) was not performed before large-scale application. Additionally, the use of granular activated carbon (GAC) for removal of PFAS was unlikely to be successful given the high concentrations of competing material (e.g., biochemical oxygen demand/ chemical oxidation demand (BOD/COD)) in the manure/milk waste blend for adsorption sites on the GAC.
2. The ECT2 proposal consisted of treating the PFAS waste through a local publicly owned treatment works (POTW) and installing anion exchange resin (AER) columns to treat the POTW effluent for PFAS. This concept is technically feasible although it would result in PFAS-contaminated sludge from the POTW for a period of at least three (3) sludge ages. Additionally, the resin system size would be much larger than for source treatment due to the higher flow at the POTW. Lastly, the efficiency of the AER would likely be lower as compared to source treatment since the PFAS concentrations in the POTW effluent would be lower due to dilution from domestic sewage resulting in a lower driving force for adsorption.
3. BC suggests using a pre-coat plate and frame filter press to separate/dewater solids from the aqueous phase. The aqueous phase (filtrate) would be processed through bag filters to remove remaining solids. We suggest that anion exchange resins be considered for PFAS removal rather than GAC due to the high concentrations of competing contaminants (e.g., soluble BOD/COD) in the aqueous waste material that will rapidly consume GAC adsorption capacity. If this is also a concern with AER, then GAC could be used for pretreatment prior to the AER. The dewatered solids would be disposed in a landfill. The solids could be solidified to encapsulate and sequester PFAS if needed for landfill acceptance.
4. An alternative approach (in the event that a precoat filter is not available or successful based on off-site, bench-scale testing) would consist of solids separation/dewatering by a centrifuge. Centrate (with higher solids content than from a precoat filter press) would be treated through a dissolved air flotation (DAF) unit to remove remaining solids. PFAS would also be removed in the DAF via coincident foam fractionation partitioning. Additives can be applied to enhance PFAS removal in the DAF. The solids and recovered DAF float (containing the bulk of the PFAS) could be solidified to encapsulate and sequester PFAS if needed for landfill acceptance.
5. BC recommends conducting concurrent bench-scale solids separation/dewatering tests with pre-coat filter and centrifuge technologies as an initial step. Determination of rental equipment availability by ACV/BC would occur in parallel. ECT2 would confirm viability of AER for PFAS removal from the filtrate. BC will work to identify waste disposal (e.g., landfill) options and will work with ACV/ECT2 to design and implement the field treatment system.

Section 1: Project Background

The State of Maine Department of Environmental Protection and Department of Agriculture have been assisting a dairy farm in central Maine to address per- and polyfluoroalkyl substances (PFAS) contamination. The farms have used their manure pits to store manure as well as contaminated milk and wash water. The material in the pits — which is likely high in BOD, nitrogen, and phosphorus — needs to be removed to provide additional storage. The material must be treated to reduce PFAS prior to disposal. The State has indicated that reductions in BOD and nutrients are not required for this particular application although this may not be the case at other sites and may also be required for efficient PFAS removal. The State of Maine has received two proposed approaches to address the issues and has requested BC to review the approaches and provide feedback on the efficacy, potential risks, and associated costs.

Section 2: Treatment Alternatives

2.1 Dairy Wastewater Treatment

The site in Maine contains tainted milk that has been discharged to a manure pond. The manure pond provides solids settling. The sediment layer in this pond undergoes anaerobic degradation. This degradation will convert organic nitrogen to ammonia-nitrogen and organic phosphorus to orthophosphate to the degree to which anaerobic degradation is completed. Under fermentation (partial degradation) caused by a lack of pH maintenance, organics will be degraded to volatile fatty acids. As the volatile fatty acids accumulate, the pH in the pond will stabilize at less than 5. The rate at which fermentation drives the pH to less than 5 has been accelerated by addition of the tainted milk. At pH 5 or less, negligible conversion of volatile fatty acids to carbon dioxide and methane will be observed.

As the contents of the pond cools below the melting point of milkfat (92° F), the milkfat will come out of solution and a floating layer will form on the surface of the pond. The surface layer will remain there until the milkfat (like other organics) can be converted to volatile fatty acids and ultimately to carbon dioxide and methane.

The best pretreatment (for removal of conventional pollutants such as BOD, total suspended solids (TSS) and nutrients (nitrogen and phosphorus) for this wastewater depends upon the degree of biological treatment that has already experienced. On fresh manure pond contents and fresh milk[1] , DAF, or suspended air flotation (SAF) with a cationic coagulant would be most appropriate to achieve 95 percent TSS and turbidity removal, percent total nitrogen and total phosphorus removal, 70 percent fats, oils, and grease removal (FOG), and 50 percent BOD removal. For largely degraded wastewater, the most appropriate treatment would be to complete biological treatment. If such treatment needs to occur in the winter, aerobic biological treatment would provide the most expedient and thorough treatment. Work by BC[2] indicates within a 3-day solids retention time, aerobic treatment can provide 90 percent BOD removal. Further work by BC indicates that greater than 90 percent total nitrogen, total phosphorus, and FOG removal can be achieved with this wastewater in less than a 20-day solids retention time in cold weather climates.[3]

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[1] Part 637 Environmental Engineering National Engineering Handbook, Chapter 4 Solid-Liquid Separation Alternatives for Manure Handling and Treatment, United State Department of Agriculture, Natural Resources Conservation Services, Table 4B-1.

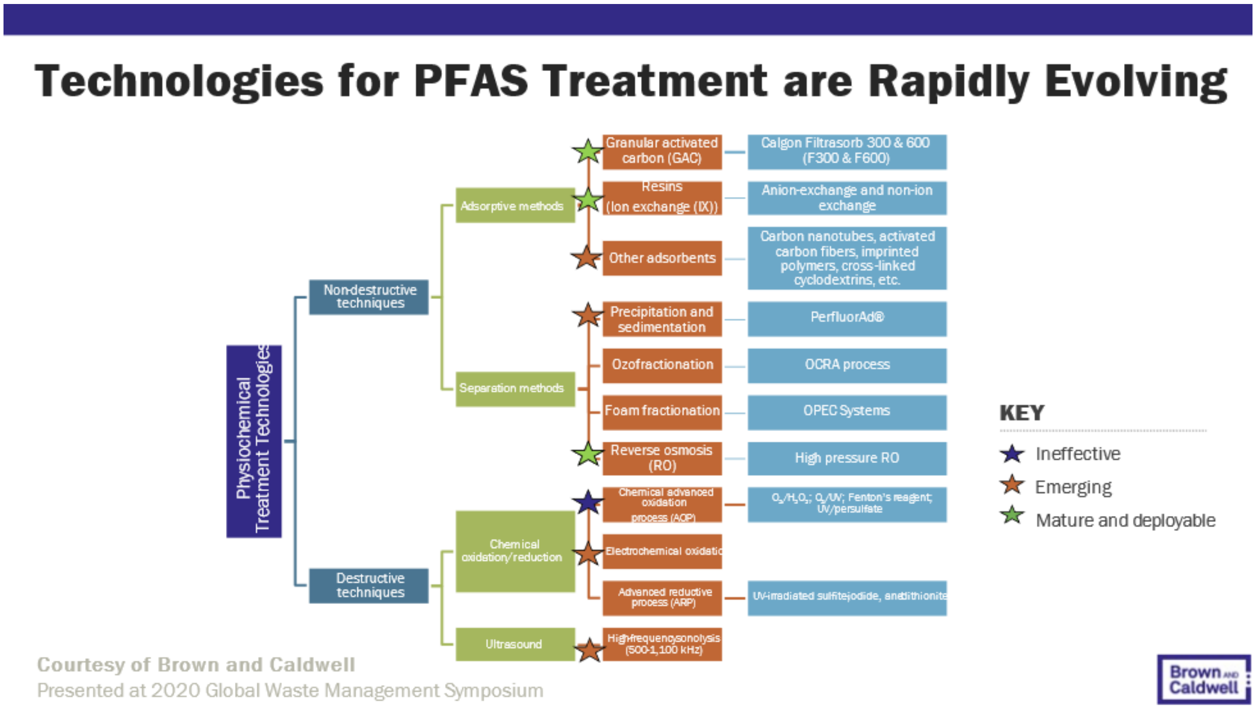
[2] Modifying Equalization to Provide Pretreatment of High Strength Wastewaters," with D.A. Moye, 19th Annual North Carolina AWWA/WEF Conference Proceedings, Winston-Salem, North Carolina, November 2002.

[3] Performance of DFA plants in Garden City, Kansas and Cass City, Michigan.

The sludge in this pit would be expected to dewater poorly. Milkfat is curdled out of solution at less than pH 3.5 and is emulsified at alkaline pH. The solids in the pit have already undergone partial degradation resulting in a dispersed floc. Dewatering may require pH adjustment and/or coagulant addition to break the emulsion and flocculant addition to promote large floc formation. A belt filter press or centrifuge could provide required dewatering while producing a likely solids capture of 95 percent (1800 mg/L TSS in filtrate or centrate when feed solids are 3.5 percent TS) and approximately a 16 percent TS cake. A precoat plate and frame filter press for the same chemical addition could produce a drier sludge (approximately a 26 percent TS cake) and up to 99 percent solids capture (350 mg/L TSS filtrate).

2.2 PFAS Removal within a Wastewater Matrix

There is a broad universe of technologies available for wastewater treatment in the marketplace. Within this universe there is a subset of technologies that are effective with PFAS. Within this subset there are only a few that are commercially available and that are demonstrated to be effective at removing PFAS from a wastewater matrix. Technologies are rapidly evolving and most PFAS treatment technology developed to date has been focused on relatively clean matrices such as contaminated groundwater or drinking water.



Technologies for PFAS Treatment are Rapidly Evolving

Courtesy of Brown and Caldwell
Presented at 2020 Global Waste Management Symposium

Figure 1: Technologies for PFAS Treatment are Rapidly Evolving

Wastewater treatment technologies generally fall into two broad categories:

1. Concentrating Technologies: These technologies remove constituents from the liquid stream via adsorption, filtration, or partitioning (e.g., activated carbon, ion exchange resins, membranes, electrocoagulation (EC), precipitation, foam fractionation, evaporator, electrodialysis reversal (EDR)). A solid or liquid residual of wastewater concentrate that contains the contaminants of interest (e.g., PFAS) is produced that requires final disposal. The leachate constituents are concentrated and not destructed.
2. Destruction Technologies: These technologies remove constituents from the liquid stream via biological, chemical, or thermal reactions that alter the molecular structures of the constituent of interest. Example technologies include aerobic or anaerobic biological treatment, advanced

oxidation processes (AOP), electrochemical oxidation (EO), or incineration. Note that biological processes are not effective for PFAS destruction but are effective for removal of other wastewater constituents such as BOD, COD ammonia and sus-pended solids.

Due to the diverse suite of constituents found in this wastewater, multiple technologies may be required to achieve a particular set of treatment requirements (depending on concentration targets) for conventional contaminants as well as emerging contaminants such as PFAS.

As with typical wastewater constituents, technologies that are applicable for removal of emerging contaminants such as PFAS are either separation (concentrating) or destructive in their mechanism for removal. The primary focus for research conducted on PFAS removal has been on comparatively clean liquid matrices (compared to wastewater) such as drinking water and groundwater due to their potential for human contact and ingestion. There is limited information available regarding treatment of PFAS in a complex matrix such as combined milk and manure. Many of the advanced technologies that have been applied, or are in the research stage, for treatment of PFAS in groundwater or drinking water are not directly applicable to this wastewater due to the presence of interfering constituents (e.g., solids, COD, salts, metals) that are not present in a clean matrix. Pretreatment of the wastewater can be applied to improve the efficacy of technologies targeting PFAS.

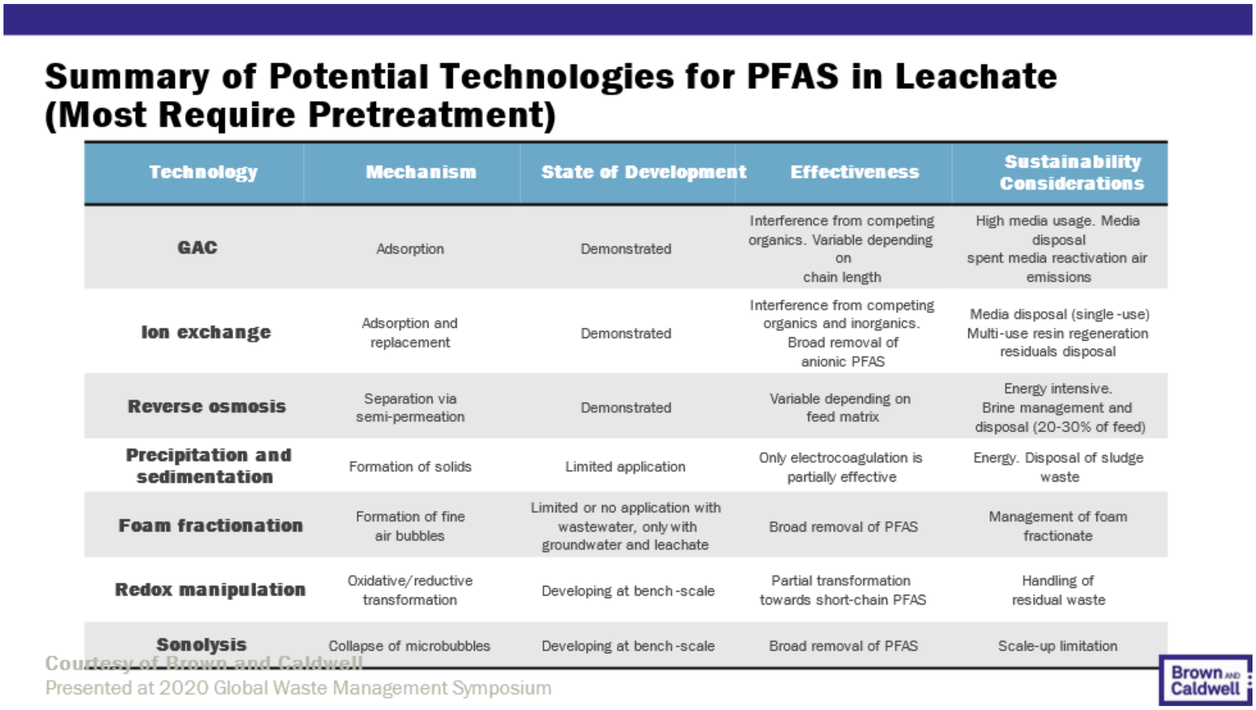
Given the objective of identifying treatment approaches that can comply with anticipated disposal option treatment requirements, the universe of available technologies for removal of typical dairy wastewater constituents as well as emerging contaminants can be reduced to those that have been proven for treatment of dairy wastewater and those that have been proven for emerging contaminant (particularly PFAS) removal and that are compatible with either raw or pretreated leachate and are at a maturity level for full-scale implementation. Examples of technologies that have been identified for potential removal of PFAS that were not considered for this application due to the level of technology maturity or incompatibility with dairy wastewater are shown in Table 1.

Table 1: Technologies That Are Not Applicable

Technology	Reasoning
Immobilized fungi degradation of PFAS	Not demonstrated with wastewater or PFAS treatment to low ppt concentrations
Direct treatment with GAC	GAC fouling and premature breakthrough
Biochar adsorption	Less effective than GAC or resins, non-regenerable
Direct treatment with IX resins	Resin fouling and premature breakthrough
Boron-Doped Diamond (BDD) anode EO	Not demonstrated with wastewater nor at full scale; Anode production challenges
Titanium Dioxide anode EO	Not demonstrated with wastewater nor at full-scale; Anode production challenges
Electrocoagulation (EC)	PFAS removal not demonstrated with wastewater
Sonolysis	Not demonstrated with leachate nor at full-scale
Advanced Oxidation Processes (AOP)	Not applicable due to high concentrations of competing oxidizable organic material in dairy wastewater coupled with limited proven effectiveness on PFAS compounds; By-product formation such as perchlorate and bromate.
Reductive defluorination	Not applicable due to high concentrations of competing organic material in leachate coupled with limited proven effectiveness on PFAS compounds
Anaerobic defluorination	Not demonstrated with wastewater or proven to be effective with PFAS compounds
Carbon nanotubes	Not proven with wastewater and not commercially available
Thermal distillation	Not proven with wastewater or for PFAS removal
Plasma arc thermal destruction	High energy, not suitable for high volumes, not commercially proven or available

Table 1: Technologies That Are Not Applicable	
Technology	**Reasoning**
Incineration	Not feasible for significant volumes of wastewater. Air emission issues; Hydrofluoric acid and other by-products formation in emissions; Requires >1,000 degrees Celsius for destruction.
Electrodialysis	Not demonstrated with wastewater for PFAS removal

This section summarizes the list of treatment technologies that have been identified as applicable for leachate treatment which has some similar properties as the dairy waste. Figure 2 presents a comparison of the leachate treatment technologies.



Summary of Potential Technologies for PFAS in Leachate (Most Require Pretreatment)

Technology	Mechanism	State of Development	Effectiveness	Sustainability Considerations
GAC	Adsorption	Demonstrated	Interference from competing organics. Variable depending on chain length	High media usage. Media disposal spent media reactivation air emissions
Ion exchange	Adsorption and replacement	Demonstrated	Interference from competing organics and inorganics. Broad removal of anionic PFAS	Media disposal (single-use) Multi-use resin regeneration residuals disposal
Reverse osmosis	Separation via semi-permeation	Demonstrated	Variable depending on feed matrix	Energy intensive. Brine management and disposal (20-30% of feed)
Precipitation and sedimentation	Formation of solids	Limited application	Only electrocoagulation is partially effective	Energy. Disposal of sludge waste
Foam fractionation	Formation of fine air bubbles	Limited or no application with wastewater, only with groundwater and leachate	Broad removal of PFAS	Management of foam fractionate
Redox manipulation	Oxidative/reductive transformation	Developing at bench-scale	Partial transformation towards short-chain PFAS	Handling of residual waste
Sonolysis	Collapse of microbubbles	Developing at bench-scale	Broad removal of PFAS	Scale-up limitation

Courtesy of Brown and Caldwell
Presented at 2020 Global Waste Management Symposium

Figure 2: Comparison of Leachate Treatment Technologies.

2.2.1 Granular Activated Carbon

GAC is a proven technology for removal of organic compounds in wastewater. Note that some inorganics, such as heavy metals, may also be removed incidentally. GAC removes constituents through adsorption and is not selective on the constituents removed. GAC is established as an effective technology for removal of PFAS (particularly long-chain compounds such as perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS)). GAC is less effective for short-chain PFAS compounds (e.g., less than six carbon atoms). PFAS removal efficiency and the rate of carbon usage are impacted by other constituents (e.g., COD) that compete for adsorption sites. GAC is not suitable for direct treatment of some wastes due to the high concentration of organic material present that would reduce adsorption efficiency of PFAS and other emerging constituents that are present at comparatively low concentrations. Additionally, the high organic content of raw dairy wastewater will quickly exhaust available adsorption capacity resulting in frequent carbon replacement. GAC is generally suitable only as a polishing step after pretreatment for removal of organic material. The GAC must be replaced when capacity is exhausted. Batch isotherm testing was conducted on POTW effluents to estimate the carbon usage rate and costs for GAC polishing of POTW effluent for PFAS removal. The estimated carbon usage rate (CUR) for treated POTW effluent with an initial PFAS concentration of 36 ng/L was 0.00036 lb. GAC/gallon. The CUR would be higher with increased PFAS and competing organic concentrations. The exhausted GAC is typically sent off-site or reactivated off-site. The thermal regeneration process results in PFAS destruction if conduct-ed at temperatures of approximately 1,000 degrees Celsius or higher (1,300 degrees Celsius has been reported).

2.2.2 Anion Exchange Resin (AER)

AER function by preferentially exchanging anions or cations (depending on the resin) for target compounds. Resins tailored to PFAS removal are anion exchange type. Resin adsorption capacity for PFAS can be reduced by other competing anions such as chloride and sulfate that are found in some wastes at elevated concentrations. Depending on the resin, the exhausted media may be regenerated, or the exhausted resin may be disposed. Single use resin is applicable for this situation. Single use resins are disposed and replaced after capacity is exhausted. Resin disposal may consist of incineration or stabilization and placement in a landfill.

2.2.3 Reverse Osmosis (RO)

RO is a separation (filtration) process that uses a membrane to separate and concentrate constituents at the molecular level from water via high pressure. RO is effective at removing PFAS and most other wastewater constituents by concentrating contaminants into a side-stream that must be subsequently managed (e.g., via destruction or sequestration of contaminants). RO has reduced efficacy with lower molecular weight compounds such as VOCs and short chain PFAS. In most cases, RO requires a very clean influent to prevent membrane fouling. In a wastewater application, conventional RO membranes (e.g., spiral wound) require prefiltration by ultrafiltration (UF), and perhaps nanofiltration (NF), as well as removal of most organic material to reduce/prevent fouling. As such, conventional membranes are suitable as a final polishing step only when coupled with other technologies for pretreatment. Conventional RO has been shown to remove PFAS to below about 5 ng/L and in some cases below detection limits (less than 2 ng/L), but effluent (permeate) concentrations vary depending on the specific compound and overall liquid matrix characteristics.

A different RO configuration is offered by Rochem Americas (Rochem) that has been demonstrated to operate effectively on raw wastewater such as leachate (with preliminary filtration at 10 microns [µm]). For the dairy wastewater of interest, more significant pretreatment such as dissolved air flotation for removal of solids and fats, oil and grease would be required. The Rochem system uses an alternative membrane construction with a high crossflow velocity to reduce fouling coupled with higher operating pressures than conventional membranes (up to about 1,800 pounds per square inch). Rochem reports achieving PFAS concentrations for a broad list of PFAS compounds, including those of interest at in the dairy wastewater to below detection limits (less than 2 ng/L) in raw leachate applications using a two-stage system (>99.9% removal). Removals to about 5 ng/L can be achieved with a single stage system. Rochem also offers a three-stage system that improves permeate recovery, thus potentially reducing leachate concentrate to approximately 12-15 percent of RO feed volume.

2.2.4 Electrochemical Oxidation (EO)

EO utilizes an electric charge distributed to the leachate via submerged anodes and cathodes and addition of oxidizing agents or catalysts to produce hydroxyl radicals, and to change the chemical structure of constituents (loss of electrons in one chemical, to create a gain of electrons in another), ozone, and hydrogen peroxide. These oxidizing species mineralize organic and some inorganic (e.g., ammonia) species. The process may serve as part of an overall treatment train for pretreatment prior to advanced processes targeting PFAS or other emerging contaminants. EO has generally not been demonstrated to provide significant and sustainable removal of PFAS in a complex matrix at large scale. Complex anode formulations (e.g., Boron Doped Diamond electrodes (BDD)) have shown promise at bench scale but are not demonstrated or available at a commercial level. One supplier (Xogen) claims to achieve high removal of organics (e.g., COD) and ammonia in leachate. The EO process may be applicable as a pretreatment process upstream of the technologies targeting PFAS.

2.2.5 Electrocoagulation (EC)

EC is similar to EO in that it uses an electric charge to change the charge properties of dissolved and suspended material to allow for agglomeration (coagulation) into solids that can be removed from the liquid. An anode and cathode are typically used, and the anode materials can be customized depending on the target constituents. EC has not been demonstrated for PFAS removal in wastewater as a stand-alone process. No oxidizing agents are typically added. EC may precede other PFAS removal technologies.

2.2.6 Foam Fractionation

Foam Fractionation is a process that can concentrate PFAS compounds, which have surfactant characteristics. Foam is formed on the water surface by applying aeration and captured.

The PFAS concentration by air-water interface through foam generation was used by OPEC Systems which includes multiple process steps that remove PFAS from contaminated water and then concentrate PFAS into low volumes of final PFAS concentrate. This technology is called Surface Active Foam Fractionation (SAFF). SAFF technology has been successfully used for leachate treatment in Sweden.

The surfactant properties are not identical among all species of PFAS. Longer-chain PFAS generally have good surfactant properties and can fractionate into bubbles effectively. Shorter-chain PFAS have higher water solubility and lower surfactant properties, and therefore, generally have poorer removal efficiency via SAFF compared to longer-chain PFAS. Longer aeration time may enhance short-chain PFAS uptake by bubbles. While water with very low total organic carbon (TOC) can limit foam generation, water with higher TOC and protein content and higher ionic strength, which is similar to typical dairy waste characteristics, would be easier to generate foams and improve the PFAS uptake by bubbles/foam. We anticipate a DAF would provide adequate PFAS removal for this application.

2.2.7 Mineral-Based Sorption

FLUORO-SORB® is a mineral-based sorbent media. The manufacturer (CETCO) claims that the sorbent media is selective to the entire PFAS spectrum and higher sorbent capacity. FLUORO-SORB® media is commercially available but there are limited demonstration studies and performance data available.

Similar to the GAC and AER, FLUORO-SORB® media is suitable only as a polishing step after pretreatment for removal of organic material and solids. The FLUORO-SORB® media must be replaced when capacity is exhausted. The exhausted FLUORO-SORB® media can be disposed, typically off site. The media is available in powder or pellet form.

2.2.8 Sequestration/Solidification

Sequestration/solidification is a process to solidify liquid and solids to avoid leaching of the contaminants from to the environment by adding special additives/mixtures. Many landfills are no longer accepting waste that is known to have elevated concentrations of PFAS since these wastes will contribute to PFAS in leachate that is typically disposed at POTWs. Solidification effectively encapsulates and sequesters the PFAS so that it does not contribute to leachate PFAS. Solidification of residuals may allow for either on or off-site disposal options not available to un-solidified waste.

2.3 Treatment Required at This Site

As a minimum, sludge dewatering is required at the site to allow treatment of aqueous phase PFAS. Samples of the waste should be subjected to bench-scale testing to evaluate the relative feasibility of belt press, centrifuge, and precoat filter press technologies. Based on these results, selection will be made based on bench-scale test performance and equipment availability.

The best treatment following sludge dewatering is strictly contingent upon the degree of pretreatment required before the PFAS removal step. The pretreatment requirements before ion exchange, mineral-based sorbent, and dirty water reverse osmosis can be determined through a review of waste characterization with suppliers. Based on this review and availability of equipment and materials, a process selection would be made for implementation. The likely treatment processes would be:

- Sludge dewatering (belt filter press or centrifuge) followed by bag filtration followed by dirty water reverse osmosis and followed by solidification.
- Sludge dewatering (belt filter press or centrifuge) followed by DAF or SAF followed by bag filtration and followed by mineral based sorption or AER.
- Sludge dewatering (belt filter press or centrifuge) followed by DAF or SAF followed by bag filtration followed by ion exchange.
- Sludge dewatering (precoat filter press) followed by bag filtration and followed by mineral based sorption or AER.

Section 3: Evaluation of Proposals and Recent Site Work

The proposed approach by ACV had not adequately considered the characteristics of the contaminated manure pit. Specifically, the characteristics of the solids as a result of on-going digestion of the combined manure/milk waste resulted in solids that would not separate via gravity with the addition of a polymer. Polymer testing to determine the most suitable polymer (or coagulant) was not performed before large-scale application. Additionally, the use of GAC for removal of PFAS was unlikely to be successful given the high concentrations of competing material (e.g., BOD/COD) in the manure/milk waste blend for adsorption sites on the GAC.

The ECT2 proposal consisted of treating the PFAS waste through a local POTW and installing AER columns to treat the POTW effluent for PFAS. This concept is technically feasible although it would result in PFAS contaminated sludge from the POTW for a period of at least three (3) sludge ages. Additionally, the resin system size would be much larger than for source treatment due to the higher flow at the POTW. Lastly, the efficiency of the AER would likely be lower as compared to source treatment since the PFAS concentrations in the POTW effluent would be lower due to dilution from domestic sewage resulting in a lower driving force for adsorption.

Recent work has identified that the material is difficult to dewater using 25-micron bag filters (geotubes). Proper chemical conditioning of the wastes prior to dewatering has been determined to be the most difficult component encountered thus far. The need for acquiring rental equipment in a timely manner is realized if this project is to be completed before winter. All of these observations align well with BC’s anticipated challenges for this project.

Section 4: Risks to Consider

An all-hands-on-deck approach is needed if this site is to be remediated before the pit freezes. The time remaining will require:

- only demonstrated technologies to be used for PFAS removal (reverse osmosis, ion exchange, and mineral-based sorption).
- pretreatment requirements to be defined through conversations and abbreviated (if any) testing.
- process selection to be based on equipment availability.

The final disposition of dewatered solids needs to be determined. These solids may require onsite storage under cover until a final decision is made.

Section 5: Next Steps

The next steps that are needed are as follows:

1. Conduct off-site tests to select sludge conditioning and dewatering technology.
2. Mobilize most effective and available sludge dewatering equipment.
3. Determine final disposition of dewatered solids.
4. Conduct off-site solidification testing if needed.
5. Meet with stakeholders, review, document and agree upon most proven and available PFAS treatment processes to pursue (presumably ion exchange, mineral-based sorption, and reverse osmosis).
6. Determine minimum pretreatment requirements for each process.
7. Select and mobilize the most effective and available process.
8. Initial onsite dewatering.
9. Initiate onsite PFAS treatment.
10. Mobilize and initiate onsite solidification if needed