PFAS RO AND NF REJECTION REVIEW

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Introduction to the PFAS Pollution Problem

PFAS is a classification of 5,000 to over 10,000 or more poly-or-perfluoroalkyl chemicals which have strong carbon-fluorine bonds that repel oil and water and thermal resistance. (1) These PFAS compounds are found everywhere around us and unfortunately in us. Invented in the 1930's and commercialized after World War 2 these compounds are water soluble and have been dubbed "forever chemicals" and are extensively used by industries of all sorts. They were used to make familiar products like Scotchgard™ that repelled water, stains and oils on our fabrics and rugs, non-stick Teflon™ used for cooking, water-proof clothing, pizza box coatings to resist grease and heat, microwave popcorn containers, hamburger wrappers, lubricants, surfactants, cosmetics, and in AFFF foam used to put out the hottest of fires that no other compound could smother and extinguish.

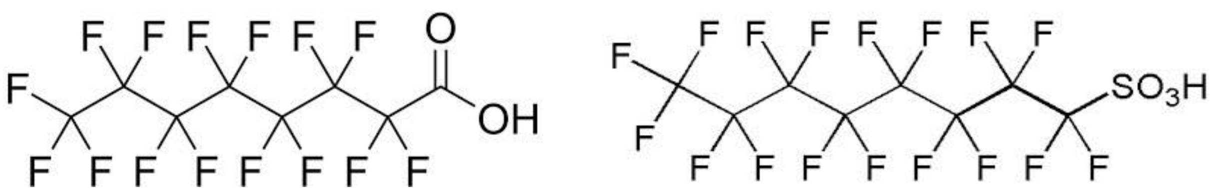
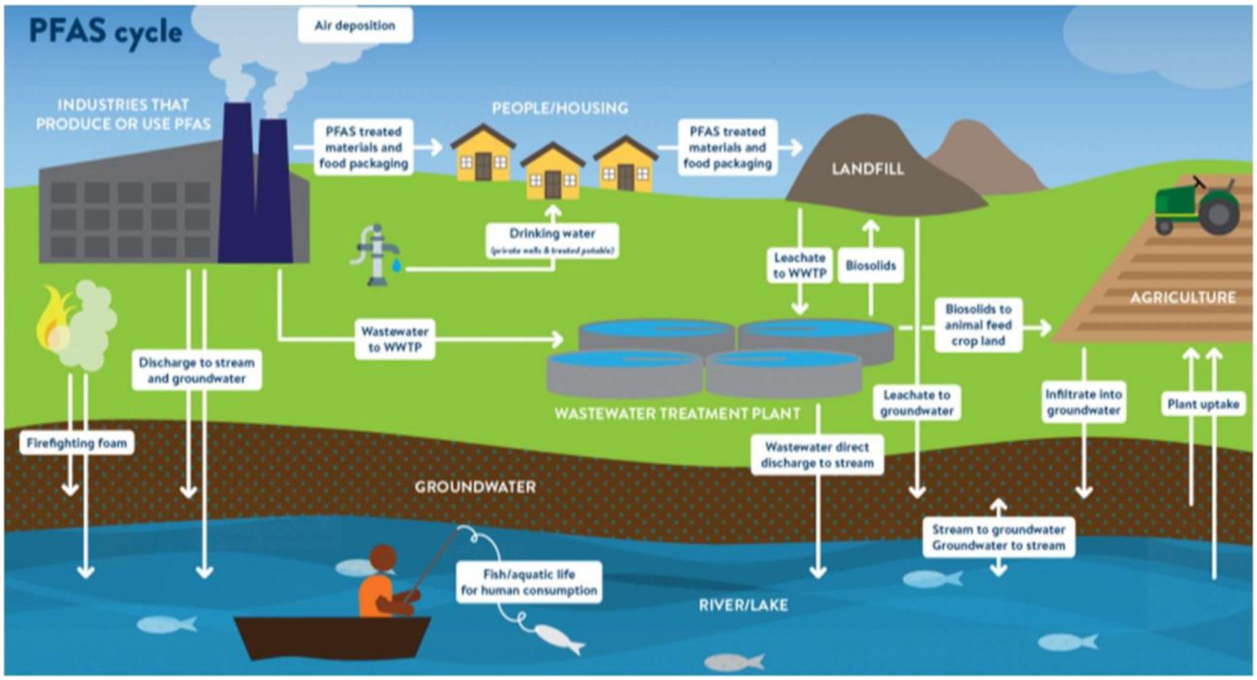


Diagram 1: Eight Carbon Chain PFOA and PFOS

The world is slowly finding out these wonderful PFAS products that improved our lives had a "dark" side and finding "safe" and suitable replacements is a very real challenge. They are not readily degraded by natural processes and have a concerning tendency to bioaccumulate in animals and humans and in waters, soils, and the air. Exposure to PFAS can lead to adverse health effects resulting in local, state, and federal governments around the world promulgating regulations in the PPT (parts per trillion PPT or ng/L) levels to protect life and the environment. Researchers trying to determine how prevalent PFAS in humans around the world had to use archived blood from before the Korean War to find samples without PFAS. (2)

Exposure through water has become an increasing concern due to the tendency of PFASs to accumulate in ground water. The US EPA in 2017 had a HAL (health advisory limit) of 70 ppt for PFOS and PFOA combined for drinking water. (3) On June 15, 2022 the EPA established new HALs for PFBS at 2,000 ppt and GenX at 10 ppt and revised PFOA to 0.004 ppt and PFOS at 0.02 ppt which are below the detection limits by our most sophisticated labs. (4)

The PFAS recycle graphic illustrates the insidious nature of PFAS in our environment. PFAS, a man-made contaminant, is introduced into our ground water or surface water from our wastewater and essentially is not destroyed.



Graphic 1: Courtesy of the Minnesota Pollution Control Agency

Regulations

Perhaps one of the most frustrating parts of the water treatment industry is tracking what state and federal regulations have to be met and when. The US EPA has not issued a national primary drinking regulatory limit yet for any particular PFAS compound as of December 2022. In 2012 public water systems were asked to monitor under the 3rd UCMR "Unregulated Contaminant Monitoring Rule" six PFAS compounds noted in Table 1. The EPA in 2019 also added two other short chain PFAS compounds to monitor, GenX and PFBA. These short chained PFAS were supposed to have shorter half-lives and be less hazardous substitutes for the longer chain PFAS they replaced. Now there is a concern that short chained PFAS have an increased potential to bioaccumulate in plants and they are not as well removed by conventional water treatment technology as compared to long-chain PFAS.

The EPA issued the 5th UCMR in December 2021 to monitor a total of 29 PFAS. Table 1 shows the PFAS components sorted by increasing number of carbons and then by molecular weight.

Table 1: PFAS Monitored up to EPA 5th UCMR of December 2021 (5)

PFAS	MW	# of C	UCMR
PFBA	214	4	5
PFMPA	230	4	5
PFBS	**300**	**4**	**3 & 5**
PFEESA	316	4	5
PFPeA	264	5	5
PFMBA	280	5	5
NFDHA	296	5	5
PFPeS	349	5	5
PFHxA	314	6	5
4:2 FTS	328	6	5
HFPO-DA (Gen-X)	330	6	5
PFHxS	**400**	**6**	**3 & 5**
PFHpA	**364**	**7**	**3 & 5**
ADONA	377	7	5
PFHpS	488	7	5
PFOA	**414**	**8**	**3 & 5**
6:2 FTS	450	8	5
PFOS	**500**	**8**	**3 & 5**
9Cl-PF3ONS	571	8	5
PFNA	**464**	**9**	**3 & 5**
PFDA	514	10	5
N-EtFOSA (N-EtFOSAA)	527	10	5
8:2 FTS	550	10	5
11Cl-PF3OUdS	632	10	5
PFUnDA or PFUnA	564	11	5
N-MeFOSAA	571	11	5
PFDoA	614	12	5
PFTrDA	664	13	5
PFTA	714	14	5

It may be useful to understand the PFAS drinking water regulatory categories:

- **Guidance:** The government has established a PFAS limit, but no notification or other action is required.
- **Notification:** The government has to be notified that a PFAS is above the drinking water limit but no remedial action is required.

- **HAL:** Health Advisory Level is a state or US EPA predecessor to enforceable level.
- **MCL:** The state or US EPA establishes a PFAS limit and remedial action will be required.

In addition to increased regulations on potable drinking water, the EPA has also announced on September 8, 2021 plans for new key industrial wastewater regulations which include discharge limits for PFAS. This program is called "Preliminary Effluent Guidelines Program Plan 15". (6) This program may force industries to treat PFAS at the plant with severe restrictions on what levels of PFAS can be discharged from the plant.

To further complicate PFAS standards and limits can vary drastically based on the feed water source and usage and are they state or federal:

- Potable drinking water
- Ground waters e.g. Wells
- Surface waters e.g. Lake or Rivers
- Municipal Wastewater Discharges and where they are going
- Industrial Wastewater Discharges and where they are going
- Landfill Leachates

Water Treatment Strategy

The good news we have the technology to remove PFAS from water. The bad news is what do we do with PFAS once we have corralled and concentrated the toxic compounds. The 1st part of this paper will address different methods of purifying water to potable and/or wastewater discharge standards. The last part of this paper will summarize various PFAS destruct technologies used to treat concentrated PFAS streams or detoxify the media or solid PFAS compounds.

There is no one perfect technology for the removal of PFAS. Key first questions to ask are:

- Does it remove PFAS compounds of varying carbon chain lengths and sizes sufficiently to meet quality goals?
- Is this a short-term or long-term fix?
- What is my capital cost?
- What is my operating cost?
- What do I do with the PFAS waste?

There are a number of treatment options and the short list of those which may be viable is listed in Table 2. Various processes such as NF/RO membranes, GAC, IX, and certain novel adsorptive media have varying levels of effectiveness in removing short and long chain carbon PFAS compounds. All of these processes result in the removed PFAS being concentrated into either a smaller waste streams or onto a solid media. Currently, most PFAS contaminated media are encapsulated and put in landfills or incinerated but this is not a good long-term answer.

Table 2: Some Industrial PFAS Removal Options

RO/NF membranes	Effective at removing short and long C chains. Cons are high OPEX and CAPEX costs and concentrate disposal.
IX anion exchange	Effective at removing long C chains, but suspect for short chains. Cons are dealing with regenerant waste and spent resin disposal.
GAC granular activated carbon	Effective at removing long C chains, but suspect for short chains. Cons are dealing with feed organic exhaustion and with revivification waste and spent carbon disposal.
Novel Adsorbents	Base material can be corn, clay, polymeric, etc. Effectiveness have shown promise but need further studies.
Electrocoagulation/Foam Fractionation	Removal of PFAS as a surfactant in the air-water foam interface shows promise but further studies are needed.

This paper will also report on PFAS rejection studies by progressively tighter NF and RO membranes. 3 different water sources were evaluated ranging from a high 534 ppm TOC Ohio landfill leachate with 8481 ppt PFAS, a 10 ppm TOC California potable drinking water RO plant concentrate stream with 100 ppt PFAS, and a low TOC lab generated feed spiked with PFAS at 1109 ppt. It will discuss that the rejection of specific PFAS compounds can vary by carbon chain length, the acid or salt form, pH, and may vary from site-to-site dependent on organic and inorganic variations in the feed water source.

Table 3: PFAS % Removal Comparison by Technology

Based on EPA Drinking Water Treatability Data Base as reported by AWWA Sept. 21 2020. (7)

PFAS	MWCO	# of Carbons	GAC Potential % Removal	IX Anion Potential % Removal
PFBA	214	4	99%	97%
PFBS	300	4	98%	98%
PFHxS	400	6	90%	99%
PFHpA	364	6	90%	94%
PFOA	414	8	40-99%	77-97%
PFOS	500	8	18-98%	90-99%
PFNA	464	9	93%	98%

PFAS and GAC Granular Activated Carbon Summary: GAC is granular activated carbon and uses pressure vessels on site or trailers. PFAS as an organic is absorbed into the pores and onto the surface. Popular when a quick answer is needed. Highest removal rates are for sulfonates and longer chain PFAS carbons. Carbon has a finite capacity of about 0.2-0.3 pounds per cubic foot of GAC for all organic matter in the feed. So carbon not only removes PFAS but all other TOC and can quickly exhaust. For example, at 100 gpm and 10 ppm TOC, you can remove 12 pounds of TOC exhausting 40-60 cubic feet of GAC daily. An important issue is there is no inline automatic measurement monitors to alarm for PFAS break through. One has to rely on volume. Once exhausted the carbon must be revivified offsite using high temperature steam which could cause a release of PFAS into the atmosphere or disposed of at landfills

PFAS and Ion Exchange IX Summary (8): Custom strong base anion resin is used that favors removal of PFAS over inorganic anions. The resin is designed to attract the negative anionic head of the PFAS on its positive charged sites and adsorb the hydrophobic non-ionic tail onto the non-charged polystyrene or "plastic" part of the resin bead. Works best on low TOC feed. Pretreatment for organics and other foulants that plug up or blind off the resin is required. Like GAC, there is no inline automatic measurement for PFAS and relies on volume to predict exhaustion. Regeneration is proprietary process but may use NaCl to remove the ionic PFAS and either an organic solvent or a surfactant on the hydrophobic part of PFAS. Regenerant waste would have to be concentrated by another on-site system, with the super-concentrated waste hauled to a hazardous waste landfill or incinerate

NF and RO Membrane Treatment for PFAS

Landfill Leachate Membrane Study

A Nitto lab study was performed on landfill leachate obtained at the Chester County Pennsylvania landfill with NF and RO membranes in June 2020. The NF membrane was a Hydranautics model PRO-XS2 and would be rated for 11,000 gpd at 110 psi and 99.7% rejection of a 2,000 ppm MgSO4 test solution. The RO membrane is a tighter membrane and would be an ESPA2-LD rated for 10,000 gpd at 150 psi and 99.6% rejection of a 1,500 ppm NaCl test solution. The landfill leachate had a high TDS level of 12,100 ppm and a high TOC level at 534 ppm. The TOC rejection of the NF was > 95% and for the RO was > 99%.

Table 4 below shows six PFAS compounds on the original 2012 USA EPA UCMR3 Unregulated Contaminant Monitoring Rule list. It is theorized, and still needs to be proven that the improved PFAS rejection for the Chester County landfill leachate relative to the Nitto spiked lab samples made up of distilled organic-free water, are the result of an irreversible organic absorption onto the membrane surface which makes the membranes tighter. The theory of organic absorption making a membrane tighter has been observed on other high organic waters at municipal wastewater plants with TOC > 5 ppm and at brackish plants in Florida with TOC up to 20 ppm. One can observe that the longer the carbon chain the better the PFAS rejection, though this can vary based on the type of functional group associated with compound e.g. carboxylate, sulfonate, etc

Table 4: PFAS Rejection Comparison of Landfill Leachate vs Nitto Spiked Lab Water UCMR 3

				Chester	**Chester**		**Nitto**	**Nitto**	**Nitto**
				10.3 gfd	**10.0 gfd**		**20 gfd**	**20 gfd**	**20 gfd**
				66% Rec	**62% Rec**		**50% Rec**	**50% Rec**	**50% Rec**
				PRO-XS2	**ESPA2**		**PRO-XS2**	**ESPA2**	**CPA7**
PFAS	**MWCO**	**# of C**		**% Rej**	**% Rej**		**% Rej**	**% Rej**	**% Rej**
PFBS	**300**	**4**		**93.6%**	**99.7%**		**71%**	**82%**	**82%**
PFHxS	**400**	**6**		**97.2%**	**99.2%**		**65%**	**87%**	**88%**
PFHpA	**364**	**7**		**98.0%**	**99.4%**		**75%**	**87%**	**87%**
PFOA	**414**	**8**		**98.6%**	**99.5%**		**78%**	**93%**	**90%**
PFOS	**500**	**8**		**98.7%**	**98.5%**		**87%**	**97%**	**95%**
PFNA	**464**	**9**		**98.3%**	**96.4%**		**86%**	**96%**	**95%**

NF and RO PFAS Rejection of UCMR5 Listed Compounds

An internal Nitto study was also performed using PFAS Nitto spiked lab water samples for PROXS2 NF, ESPA2-LD RO and CPA7-LD RO. Flux was 20 gfd and recovery was 50%.

Table 5: PFAS Rejection of Spiked Lab Water UCMR 5

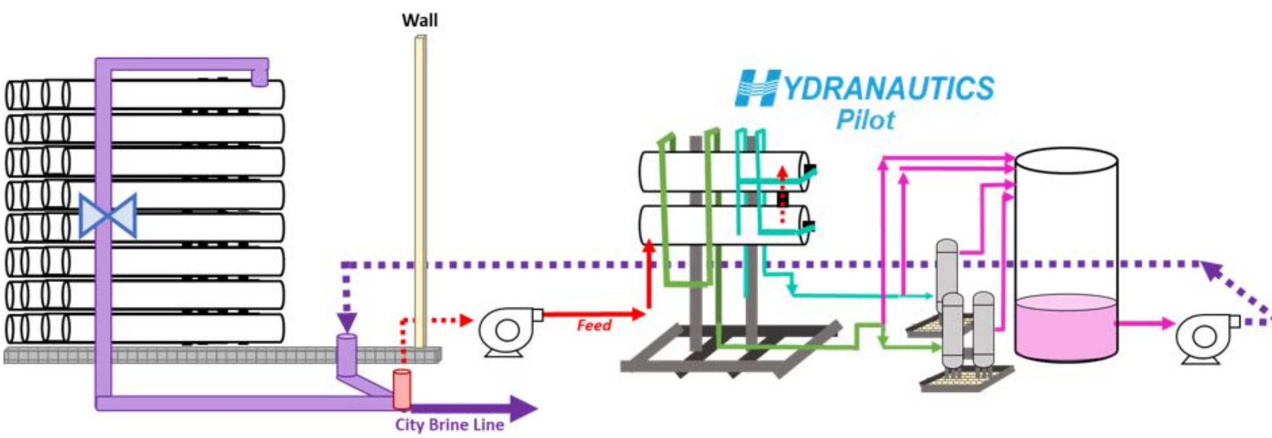
PFAS			PRO-XS2 NF	ESPA2 RO	CPA7 RO
Note: On UCMR 5 list	MWCO	# of C	% Rej	% Rej	% Rej
PFBA	214	4	71%	83%	81%
PFPeA	264	5	69%	82%	80%
PFPeS	349	5	72%	86%	84%
PFHxA	314	6	77%	87%	85%
HFPO-DA (Gen-X)	330	6	71%	83%	81%
4:2 FTS	350	6	70%	81%	83%
ADONA	377	7	77%	89%	87%
PFHpS	488	7	75%	94%	94%
6:2 FTS	450	8	79%	90%	90%
9Cl-PF3ONS	571	8	89%	100%	100%
PFDA	514	10	87%	97%	96%
N-EtFOSA (N-EtFOSAA)	527	10	100%	100%	100%
8:2 FTS	550	10	90%	98%	100%
11Cl-PF3OUdS	632	10	100%	100%	100%
PFUnDA or PFUnA	564	11	95%	97%	100%
PFDoA	614	12	100%	100%	100%
PFTrDA	664	13	100%	100%	100%
PFTA	714	14	100%	100%	100%
Note: Below is not on UCMR 5 list					
PFOSA	499	8	97%	98%	100%
PFNS	567	9	92%	100%	100%
PFDS	600	10	100%	100%	100%
N-MeFOSA	557	11	100%	100%	100%
N-MeFOSE	564	11	100%	100%	100%
MeFOSAA	571	11	94%	100%	100%
N-EtFOSE	580	12	100%	100%	100%
EtFOSAA	590	12	100%	100%	100%
PFDoS	699	12	100%	100%	100%

The results shown above in Table 5 are in order of number of carbon chains first followed by the molecular weight. In almost all cases rejection for a membrane is dependent on this pattern. Also, when you start getting larger than 8 carbon chain rejection becomes very good.

Well Water PFAS NF/RO Pilot

This pilot was operated in 2022 to treat the concentrate from the current RO system at a municipal well water treatment facility in Oceanside California located just south of the Camp Pendleton military base. The objective was to further concentrate the current RO waste stream shown as Train A in Graphic 2. The current RO treats ground water at 21.3 C and operates at 85% recovery to produce a concentrate that is send to the pilot. The pilot treats the current RO concentrate with a TDS of 7147 ppm, TOC of 12.3 ppm, and 100-115 ppb of PFAS. The pilot has a 1x1-1M array and was operated at about 17 gfd, 17% recovery, 7.2 gpm of permeate and 36 gpm of concentrate. Seven different brackish water membranes were tested, 3 NF and 3 RO and one membrane which has the feed pressure of a NF and the rejection of a RO. Each membrane was operated for one to two weeks. A novel corn-based adsorbent was tested on both the pilot permeate and concentrate streams with mixed results due to operational issues. Absorbent results will not be reported in this paper.

Graphic 2: The Municipal Well Water Treatment Facility Flow Diagram



Pilot Conditions: PFAS NF-RO Pilot

Feed TDS	Feed TOC	Feed Total PFAS	Temperature	Flux	Recovery %
9,000 ppm	10 ppm	100 to 115 ppt	21.3 °C	17 GFD	17 %

Table 6 is summary of the 10 measurable PFAS compounds in the pilot feed, which add up to about 100 ppb, and the potential sources.

Table 6: Well Water Pilot PFAS Summary

	MW	# C	UCMR	PPT	
PFBS	300	4	3 & 5	17.5	Industrial surfactant. Water and stain resistant coating in fabrics, carpets, paper.
PFPeA	264	5	5	5.1	Leached from fluorinated HDPE containers used to store pesticides, etc
PFPeS	349	5	5	7.1	Solvay. Oil/water resistance for large number of materials like textile, paint, stains, etc
PFHxA	314	6	5	6.7	Leached from fluorinated HDPE containers used to store pesticides, etc. Breakdown chemical in grease-proofing food contact, stain resistance.
PFHxS	400	6	3 & 5	28.6	Fire fighting foam. Stain resistant fabric. Food packaging. 3M phased out in 2002.
PFHpA	364	7	3 & 5	3.1	Leached from fluorinated HDPE containers used to store pesticides, etc.
PFHpS	488	7	5	1	Not sure. May be associated with GenX.
PFOA	414	8	3 & 5	10.1	Leached from fluorinated HDPE containers used to store pesticides, etc. Dupont C8 and Teflon.
PFECHS	461	8	no	5.2	Aircraft hydraulic fluid since 1940's. Replacement of 3M PFOS.
PFOS	500	8	3 & 5	14.3	Stain resistant 3M Scotchgard, fire fighting foam after USS Forrestal, food packaging.
PFNA	464	9	3 & 5	1.1	Leached from fluorinated HDPE containers used to store pesticides, etc
Total PFAS in Feed:				99.8	

Table 7 is a summary of the operational feed pressure, % salt passage, % TOC passage and % Total PFAS passage for each of the membranes tested.

Table 7: Well Water Pilot Membrane Results

Membrane	Type	Feed PSI	% Salt Passage	% TOC Passage	% PFAS Passage
PRO-XS2	NF	122	35.00%	3.50%	2.20%
ESNA1-LF2-LD	NF	112	8.80%	0.80%	0.50%
ESNA1-LF-LD	NF	120	5.30%	0.50%	0.50%
ESPA4-LD	RO-NF	135	1.10%	< 0.4%	0.50%
ESPA2-LD MAX	RO	151	0.40%	< 0.4%	0.60%
PRO-XR1	RO	203	0.20%	< 0.4%	0.60%
CPA7-LD	RO	208	0.20%	< 0.4%	0.60%

Table 8 is the PFAS data which shows the pilot RO feed and permeate and concentrate concentrations for the four RO membranes. Please note that the minimum detection level for the permeate is 0.05 ppt. When the permeate reading was ND (Not Detectable) then the % Passage was highlighted in red indicating that the % passage is a value less than reported. The % passage of a PFAS compound is the permeate level divided by the arithmetic average of the feed-concentrate.

It should be noted that the results for the PRO-XS2 and ESNA1-LF-LD are not reported in Table 8 below. These were the 1st 2 elements tested and the permeate detection limits were 0.7 ppt and 0.2 ppt respectively which essentially made all the results ND (Not Detectable) and calculation for % Passage not meaningful. The last 5 elements tested permeate levels were ran at 0.05 ppt for more meaningful data.

Table 8: Well Water Pilot PFAS Data

ESNA1-LF-LD	Concentration (ppt)											
	PFBS	PFPeS	PFHxS	PFHpS	PFOS	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFECHS	Total
Feed	16.9	5.7	33.0	0.7	13.3	6.0	7.4	3.3	9.6	1.1	3.5	101
Permeate	0.08	0.05	0.11	0.05	0.13	0.08	0.06	0.05	0.08	0.05	0.05	0.79
Concentrate	20.6	6.9	40.5	0.7	16.1	7.2	8.3	4.1	11.7	1.4	4.7	122
% passsage	**0.4**	**0.8**	**0.3**	**7.1**	**0.9**	**1.2**	**0.8**	**1.4**	**0.8**	**3.9**	**1.2**	**0.7**

ESPA4-LD	Concentration (ppt)											
	PFBS	PFPeS	PFHxS	PFHpS	PFOS	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFECHS	Total
Feed	16.5	6.9	32.8	0.8	13.8	6.5	7.5	3.5	12.2	1.2	4.7	106
Permeate	0.05	0.05	0.05	0.05	0.06	0.06	0.06	0.07	0.08	0.05	0.05	0.63
Concentrate	20.7	8.6	39.8	1.1	16.8	7.5	8.9	4.8	14.8	1.4	5.6	130
% passsage	**0.3**	**0.6**	**0.1**	**5.2**	**0.4**	**0.9**	**0.7**	**1.6**	**0.6**	**3.8**	**1.0**	**0.5**

ESPA2-LD MAX	Concentration (ppt)											
	PFBS	PFPeS	PFHxS	PFHpS	PFOS	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFECHS	Total
Feed	16.6	7.0	30.9	0.9	15.2	6.4	7.4	4.8	11.7	1.0	5.5	107
Permeate	0.05	0.05	0.05	0.05	0.16	0.06	0.05	0.07	0.08	0.05	0.05	0.71
Concentrate	20.4	8.9	40.5	1.0	16.7	7.6	9.0	5.5	14.5	1.3	5.5	131
% passsage	**0.3**	**0.6**	**0.1**	**5.4**	**1.0**	**0.8**	**0.6**	**1.3**	**0.6**	**4.3**	**0.9**	**0.6**

PRO-XR1	Concentration (ppt)											
	PFBS	PFPeS	PFHxS	PFHpS	PFOS	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFECHS	Total
Feed	16.5	6.8	32.9	1.0	18.1	6.5	7.6	4.3	13.0	1.4	6.4	114
Permeate	0.05	0.05	0.05	0.05	0.09	0.05	0.06	0.11	0.09	0.05	0.05	0.69
Concentrate	20.1	8.1	37.8	1.0	15.7	7.7	9.3	4.8	14.2	1.4	5.0	125
% passsage	**0.3**	**0.7**	**0.1**	**5.0**	**0.5**	**0.7**	**0.7**	**2.4**	**0.7**	**3.7**	**0.9**	**0.6**

CPA7-LD	Concentration (ppt)											
	PFBS	PFPeS	PFHxS	PFHpS	PFOS	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFECHS	Total
Feed	17.3	6.9	32.0	0.8	13.5	6.8	7.5	3.3	12.2	1.1	4.5	106
Permeate	0.05	0.05	0.05	0.05	0.05	0.12	0.05	0.05	0.11	0.05	0.05	0.68
Concentrate	20.4	8.6	38.4	1.0	16.6	8.0	8.9	4.0	13.9	1.4	5.3	127
% passsage	**0.3**	**0.6**	**0.1**	**5.7**	**0.3**	**1.6**	**0.6**	**1.4**	**0.8**	**3.9**	**1.0**	**0.6**

Cradle to Grave: PFAS Detoxify or Destruct Options

The holy grail for addressing the PFAS problem is once we have purified a stream and have concentrated the PFAS, then what do we do with the concentrated PFAS? There are a number of technologies being pursued, but all need to be proven practical and effective. A short list include:

- Mechanochemical Degradation MCD: MCD describes the mechanism of solid PFAS salt destruction using a high-energy ball-milling device. (9) This method does not require high temperature incineration to detoxify the PFAS salts.
- Supercritical Water Oxidation SCWO: Concentrated PFAS is fully broken down to form water, nitrogen gas, and carbon dioxide as the water reaches it supercritical state above 705 F and 218 atmospheres. (10)
- The Elemental™ is a proprietary photochemical process by Claros that claims 97-100% destruction of all PFAS compounds within 1-3 hours at ambient temperature and pressure conditions. By-products free fluoride and carbon dioxide. (11)
- Incineration has been proposed but has raised many questions about its safety. Incomplete incineration could create toxic carbon-fluoride gases in its emissions. Illinois and New York have banned the use.
- Landfill
- Deep well injection
- Ozonation
- Biological
- Plasma
- Electrochemical oxidation
- Sonochemical treatment
- Electron beam

Summary

This paper reviewed the seriousness and pervasiveness of PFAS in our environment and what regulations are being introduced to protect us. There is no one perfect water treatment strategy to purify our water resources though purification is very achievable. GAC and IX technologies are good intermediate answers. RO and NF are very effective at meeting water purification requirements, but they will need to be coupled with a good PFAS destruct and detoxification technology which still has not been established.

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