Recommendation on Perfluorinated Compound Treatment Options for Drinking Water

New Jersey Drinking Water Quality Institute
Treatment Subcommittee
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Background

At the request of the Commissioner of the New Jersey Department of Environmental Protection the Drinking Water Quality Institute (DWQI) is working to develop recommended Maximum Contaminant Levels (MCL) for three long-chain perfluorinated compounds (PFC): Perfluorononanoic acid (PFNA), Perfluorooctanoic acid (PFOA) and Perfluorooctanesulfonic acid (PFOS).

The Treatment Subcommittee of the Drinking Water Quality Institute is responsible for identifying available treatment technologies or methods for removal of hazardous contaminants from drinking water. The subcommittee has met several times over the last year beginning in July 2014 to discuss and investigate best available treatment options for the long-chain (8 – 9 carbon) PFCs identified above. The subcommittee decided to research and report on treatment options for all three compounds, as the treatment options are not expected to differ from compound to compound due to their similar properties (e.g. persistence, water solubility, similar structure, strong carbon-fluorine bonds, and high polarity). This approach contrasts with the other two subcommittees which will address the three compounds separately. The subcommittee has gathered and reviewed data from several sources in order to identify widely-accepted and well-performing strategies for removal of long-chain PFCs, including use of alternate sources. This report is intended to present the subcommittee's findings.

At this time, there are no Federal drinking water standards for PFNA, PFOA or PFOS; however in 2009 the United State Environmental Protection Agency (USEPA, 2009) established a Provisional Health Advisory (PHA) level of 0.4 µg/L for PFOA and 0.2 µg/L PHA for PFOS for short-term exposure. New Jersey released a recommended long-term exposure guidance level of 0.04 µg/L for PFOA for a specific water system in 2007 (NJDEP, 2007). In addition, there are various examples of recommended guidance levels for PFOA and PFOS worldwide that are of similar magnitude (e.g. Maine, Minnesota, North Carolina, United Kingdom and Germany). As such, there are many examples of systems globally, that are endeavoring to remove PFCs from drinking and wastewater.

According to published literature long-chain PFCs, such as PFNA, PFOA and PFOS can be successfully removed from water using treatment techniques discussed in the section below. It should be noted that the techniques outlined below may not be effective in removing all PFCs; for example short-chain PFCs are not effectively removed by some of the techniques listed. Removal ratios vary depending on a number of factors (e.g. initial concentration and presence of other contaminants) but can exceed 90% removal and result in finished water with non-detectable levels (Rahman et al., 2014 and Water Research Foundation [WRF], 2014). Given this information, the ability of several treatment options to remove these contaminants is not anticipated to be a limiting factor in the development of a recommended Maximum Contaminant Level (MCL) for PFNA, PFOA or PFOS.

Treatment Options

Water systems facing PFNA, PFOA or PFOS contamination should thoroughly evaluate all possible approaches, including the use of alternate sources that are not contaminated as a replacement or in combination with existing sources. While engineering a solution that includes blending is not "treatment," and may not be the preferred solution, it may be a cost-effective and viable option for some water systems. The cost of this option will vary depending on the availability and proximity of alternate sources. In order to make an informed and cost effective decision, the use of an alternate source should be evaluated in addition to all available, effective treatment options.

At the present time the members of the treatment subcommittee recommend that the use of granulated activated carbon (GAC) or an equally efficient technology should be considered for treatment of PFNA, PFOA and PFOS detected above the DWQI recommended MCL subject to the on-site pilot testing performance results.

When selecting an appropriate treatment option(s) there are many factors to be considered. According the USEPA's *Emerging Contaminant Fact Sheet – PFOS and PFOA*, these include: initial concentration of PFCs, the background organic and metal concentration, (e.g. competition for active sites on sorptive media), available detention time and other site conditions. Additional considerations include operation and maintenance costs, the ability to address more than one contaminant with one treatment option, and waste disposal. While technology continues to evolve and new methods continue to emerge this report lists technologies demonstrated in the literature and in practice to be effective for removal of PFNA, PFOS and/or PFOA.

While the following discussion of treatment options evaluates the advantages and limitations as well as cost considerations for each method, the information is general. Selection of the most cost effective treatment process requires case-by-case evaluation (i.e. bench and/or pilot-scale studies) and may result in the use of more than one of the identified options in a treatment train. To that end, bench and/or pilot studies should be designed to aid in the establishment of the required design parameters specific to the treatment processes being evaluated. Conceptual level design should then be used to develop reasonable cost estimates for a full life-cycle cost analysis to include capital, operation and maintenance costs. The full life-cycle cost analysis can be utilized to define the best option specific to an individual water system.

The treatment options identified in the literature are summarized in Table 1 below and are described in greater detail in the section that follows.

TABLE 1. **Summary of Treatment Options for Removal of PFNA, PFOA and PFOS**

Treatment Option	**Notes**	**Removal Rates[1]**		
		PFNA	**PFOA**	**PFOS**
Activated Carbon	**Granulated** - GAC is the most common treatment method for long-chain PFC removal. Competition for adsorption with other contaminants can reduce effectiveness. Thermal regeneration of GAC is effective. **Powdered** -High concentrations of PAC are necessary. PAC may be useful in responding to spills but the required high concentrations may make this an infeasible option. PAC combined with waste residuals may create a challenge for disposal of waste products.	>90%	>90%	>90%
Membrane Filtration	Multi-contaminant removal. Rejection rate can be high. Waste/ byproducts must be managed. Mineral addition may be necessary.	>90%	>90%	>90%
Anion Exchange	Single-use systems do not produce contaminant-containing brine but required replacement and proper disposal. Regenerable systems produce brine that must be disposed of responsibly; such systems are automated, have small footprints and high regeneration efficiencies. Competition with common ions for binding sites on resins can impact effectiveness. Organics, total dissolved solids, minerals can clog resins and reduce efficiency.	>67%	10-90%	>90%
Advanced Oxidation	Low removal rate. Can destroy pollutants to produce less complex compounds. Other organic contaminants will compete for hydroxyl radicals and reduce efficiency.	<10%	<10%	<10 - 50%

Although four treatment techniques are described below, the subcommittee found that Granulated Activated Carbon was by far the most common treatment for long-chain PFC removal. Accordingly, more information (i.e. case studies and published literature) was available for Granulated Activated Carbon than for the others.

Activated Carbon

Activated carbon is commonly used to adsorb contaminants found in water. It is used to remove synthetic organic chemicals, natural organic compounds, and other compounds affecting taste and odor in drinking water treatment. Adsorption is a physical and chemical process of accumulating a substance at the interface between liquid and solids phases (USEPA, Treatability Database). Activated carbon, which is used in a granulated or powdered form, is an effective

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[1] Note that the removal rates are referenced and cited in the sections below.

adsorbent because it is highly porous and provides a large surface area on which contaminants may adsorb. Activated carbon is made from organic materials (e.g. coconut shell, coal, wood) that contain high amounts of carbon.

Granulated Activated Carbon

A review of the literature and several case studies indicated that Granulated Activated Carbon (GAC) is a common and effective (>90% removal) treatment for long-chain PFC contamination (WRF, 2014 and Eschauzier et al., 2012). For example, GAC was found to be highly effective for PFOA removal at two public water systems, one in Ohio and one in West Virginia with the use of dual filter design, careful monitoring for breakthrough, and frequent filter changes.

The efficiency of this method varies based on several factors including: target effluent contaminant concentration, pH, water temperature, contact time, the properties of the selected carbon, concentration of inorganic substances in the water, ambient natural organic matter and the presence or absence of chlorine (Black & Veatch, 2008). For example, the amount of natural organic material present will reduce the ability of carbon to remove PFCs. While the positively charged surface of the carbon is limited, there is evidence that the absorptive capacity can be increased by coating the surface (Interstate Technology & Regulatory Council [ITRC], 2008). Although they are not the subject of this report, it should also be noted that this method is less effective at removing shorter chain PFCs (Appleman et al., 2014). This is a consideration if the intent is to remove both long and short-chain PFCs.

Design of a GAC treatment facility, like other treatment options, requires pre-design bench and/or pilot-scale studies. These studies will aid in defining the most efficient GAC product and defining the empty-bed contact time to remove the contaminants of concern based on source water quality, source water contaminant concentration and target effluent concentration. The studies would be used to establish the required configuration necessary for the most efficient carbon usage rate, ability for continuous operation during change out, and options for disposal/regeneration of the GAC. These studies would aid in the development of treatment costs which will most likely be driven by the carbon usage rate. A conceptual design project would be required to develop reasonable cost estimates for construction and long-term operation and maintenance costs.

Costs will vary depending on the level of contaminant in the source water as well as the presence and concentration of other contaminants that compete for carbon surface area. See the case studies below for more examples of costs. In addition to capital costs (e.g. labor, replacement, operation, maintenance, equipment, and transport), disposal of exhausted carbon is also a cost consideration. Treatment of spent carbon may be necessary prior to disposal (ITRC, 2008).

Case Studies

Little Hocking, Ohio –The public water supply (groundwater) in Little Hocking, OH was contaminated with high concentrations of PFOA (1.9 - 8.5 µg/L) as a result of discharges from a local manufacturing plant that date back to the 1950's (Rahman et al). Ohio does not have a regulatory or guidance level for PFOA, however, in a 2007 Water Quality Report, the Little Hocking Water Association indicated that they maintain that there should be "no detectable level" of PFOA and related compounds in the water. To that end, according to the Little Hocking Water Association website, a GAC treatment plant opened in November 2007. As a result of the high concentration of PFOA, the carbon is changed approximately every three months, for a total of 29 changes since the plant began operating. In 2008, sampling results indicate that water entering the plant contained 3.3 µg/L, while water exiting the first GAC bed and second GAC bed then entering the distribution system contained no detectable amount of PFOA (Little Hocking, 2008).

Oakdale, Minnesota – After a pilot study was conducted by 3M to establish the most effective means to remove PFOA, PFOS, perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFeA), perfluorohexanoic acid (PFHxA), perfluorobutane sulfonate (PFBS) and perfluorohexane sulfonate (PFHxS) in 2006 Oakdale began operating a plant with ten GAC filters containing a total of 100,000 lbs. of GAC and that has a capacity of 2,000 gallons per minute (Minnesota Department of Health [MDH], 2010 and Bachmeier, 2015). The Oakdale plant monitors for breakthrough of PFOA and PFOS at minimum detection levels of 14 ng/L and 8 ng/L, respectively. Carbon is changed after PFOA concentrations from the lead filters reaches levels that are half the levels in the raw water (approximately every 12 – 24 months). At all times Minnesota's Health Risk Limit of 0.3 µg/L for both PFOS and PFOA is maintained or exceeded in the finished water. PFOS levels are consistently maintained below the minimum detection level (Martin, 2015). The facility cost approximately $3,000,000 to construct. The annual operational costs are about $25,000 (e.g. power, daily inspections). The cost of carbon is approximately $250,000 every 18 months (Bachmeier, 2015).

New Jersey American Water – Penns Grove – Elevated PFOA levels were detected in New Jersey American Water's (NJAW) Penns Grove water system, which supplies water approximately 10,900 people in Salem County, NJ. NJAW evaluated several treatment options to address the PFOA contamination as well as sodium levels elevated above recommended limits. A comprehensive alternatives analysis of all factors, including PFOA treatment, resulted in the selection of a brand new treatment plant that included GAC treatment at an estimated cost of $12.2 million and with an annual operating and maintenance cost of $80,000 (New Jersey American Water, 2010). The new treatment plant combined designed capacity is 3MGD to achieve removal PFC removal below the NJDEP guidance level of 0.04 µg/L using 80,000 lbs. of carbon. The cost of carbon was estimated at $1.50 per pound (New Jersey American Water, 2010).

After an accelerated column test (ACT) to evaluate two types (i.e. Calgon F-400 and F-600) of carbon, the F-400, which demonstrated a longer run volume was selected with an empty-bed contact time of 10 minutes. Based on this test, it was recommended that the lead bed effluent concentration be monitored on a monthly basis and that plans to change out the carbon be initiated when effluent concentrations approach 50% of the inlet concentration. For PFOA, the ACT showed that F-400 did not reach 50% breakthrough even after treating more than 231,666 Kgal.

New Jersey American Water – Logan System Birch Creek - According to a study of the occurrence of PFCs in Logan System Birch Creek had detections of PFNA (18 – 72 ng/L) and of PFOA (33 – 60 ng/L), in addition to three other PFCs, all of which were removed below the reporting level of 5 ng/L with the installation of granulated activated carbon (Table 2). In this full scale installation, the GAC adsorbers were operated with an empty-bed contact time of approximately 15 minutes. Additional rapid small-scale carbon tests were used to design this process where Calgon F-400 performed significantly better than Calgon F-600 and where Calgon F-400 was installed at the full-scale (Engineering Performance Solutions, 2010).

TABLE 2. **New Jersey American Water – Logan System Birch Creek PFNA Pre- and Post-Treatment Data**

Notes: Reporting Limit = 5 ng/L; GAC treatment operable as of 5/25/12

Date	Raw Water (ng/L)	GAC Treated Water (ng/L)
9/27/2011	60	N/A
2/14/2012	50	N/A
6/14/2012	72	<5
8/1/2012		<5
11/27/2012	46	<5
1/18/2013		<5
2/28/2013	54	<5
5/30/2013	50	<5
4/24/2014	70	<5
9/10/14	18	<5

Source: NJDEP Drinking Water PFC database, accessed 5/28/2015

Amsterdam, Netherlands

A study of the removal of a number of PFCs from raw water in Amsterdam at different steps in the treatment process showed that GAC was able to reduce both PFOS and PFNA measured in the raw samples at values of 6.7 to 10 ng/L and 0.5 to 0.8 ng/L respectively were removed through the final GAC adsorber to levels measured below the limits of quantitation (0.23 ng/L and 0.24 ng/L, respectively) (Eschauzier, 2012). PFOA concentrations in the influent ranged between 3.8 to 5.1 ng/L and in the final GAC adsorber ranged between 3.6 to 6.7 ng/L. GAC adsorption for this study was done in two stages with adsorbers operated in series, each with a 20 minute empty bed contact time. The GAC in the lag adsorber is placed in the lead position after 15 months of operation and replaced with fresh GAC. The GAC used in this study was Norit ROW 0.8S.

Powdered Activated Carbon

Powdered activated carbon (PAC), a smaller diameter form of activated carbon than GAC, can be added as a dry powder or as slurry during the coagulation treatment process. Contaminants adsorb to PAC in the same manner as they do to GAC. According to the USEPA Treatability Database, PAC is removed by filter beds during backwashing or by sedimentation.

The subcommittee reviewed a recent study that found that a combination of adsorption by powdered activated carbon and coagulation was an effective (>90% removal) method of removal of both PFOS and PFOA from water (Bao et al., 2014). It should be noted that coagulation alone is not an effective means of removal for long-chain PFCs, such as, PFNA, PFOS and PFOA (Rahman et al., 2014 and Appleman et al., 2014). According to the Bao et al. study, the removal was "achieved by adsorption onto the surface of coagulants and flocs via electrostatic interaction." Removal of PFOS and PFOA by coagulation works by adsorption of the contaminants onto the surface of the coagulants. Anions absorb onto the positive surface of coagulants and flocs and are then removed with sedimentation and filtration. The Bao et al. study (2014) showed that PAC has a significantly higher absorption rate and capacity than GAC, and higher absorption efficiency for PFOA than PFOS. The study showed that removal ratios for PAC increase with decreasing pH and with increasing coagulant dose, which is consistent with the results of the Water Research Foundation's Project # 4344. This project also evaluated PAC and found that >90% removal of PFNA and PFOS was possible but only with unreasonably high adsorbent dosages unless contact times could be extended to approach adsorption equilibrium. Likewise, Rahman et al suggest that "PAC may be a more appropriate choice for removing PFASs[2] in situations that require a prompt short-term response (e.g. spills)."

Membrane Filtration

Reverse Osmosis

Reverse Osmosis (RO) can remove many types of molecules and ions from solutions, including bacteria, and is used in both industrial processes and the production of potable water. For RO treatment of perfluorinated compounds in drinking water, the result is that the solute (perfluorinated compound) is retained on the pressurized side of the membrane and the pure solvent (i.e. water) is allowed to pass to the other side. RO can serve as a pre- or stand-alone treatment (ITRC, 2008).

Reverse Osmosis is commonly used around the world for household drinking water purification systems, in the production of bottled mineral water, in branches of the United States armed forces as self-contained water purification units, and industrial applications (e.g. water supply to cooling towers, boilers, deionized water). The largest application of Reverse Osmosis is in desalination. In fact, RO is an increasingly common method of desalination for potable water, because of its relatively low energy consumption. According to the International Desalination Association, for 2011, reverse osmosis was used in 66% of installed desalination capacity and nearly all new plants; the other plants mainly use thermal distillation methods, which are energy intensive.

Pretreatment is important when working with reverse osmosis membranes due to the nature of their spiral-wound design. Since accumulated material cannot be removed from the membrane surface systems, they are highly susceptible to fouling (loss of production capacity). Therefore, effective pretreatment is a necessity for any reverse osmosis system. The use of RO to remove PFOS from wastewater was studied by Tang et al. and showed 90% removal and a study

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[2] PFASs are perfluoroalkyl substances, which include PFNA and PFOA.

published in 2013 showed ≥99% removal of PFOS and PFOA in drinking water (Flores et al., 2013).

Nanofiltration

Nanofiltration (NF) is another form of membrane technology that is also pressure-driven and shown to be effective in removal of PFCs (Tang et al., 2007). This method of filtration provides high water flux at low operating pressure (Izadpanah & Javidnia, 2012). Like reverse osmosis, this method is easy to operate and reliable for removal of pollutants. Tang et al. reported 90% removal of PFOS using nanofiltration.

There are many advantages associated with RO with respect to the removal of the PFCs being discussed. PFNA, PFOA and PFOS are relatively high molecular weight compounds that could be removed effectively by RO membranes. Related to PFOA and PFOS, the Octanol-water partition coefficient is non-measurable; thus, indicating uncertainty of effective and preferably irreversible adsorption onto activated carbon. Conversely, RO removes all contaminants, including shorter chain PFCs (Appleman et al., 2014). Thus, RO may be a good choice if the intention is to remove multiple contaminants. . Chemical oxidation treatment options (e.g. hydrogen peroxide, ozone, etc.) can result into partial oxidation of these compounds, which could result in various byproducts that could have different properties/effect on human health, RO membranes will filter out the precursory material providing disinfection byproduct free potable water.

Although, recovery rates were a concern in the past, high recovery RO systems are available. It could be possible to reduce the volume of reject to 5% of the treated water. Reject handling/management options are to be considered prior to selecting high recovery options. Finally, it should be noted that RO/NF are equally applicable to other high molecular weight perfluoro-organics also.

Based on experience with such types of chemicals, it is expected that RO is more preferable than NF in providing higher rejection of the contaminants and in reducing the volume of the reject stream.

As with all other treatment options, bench scale and pilot scale testing will be required to understand the field applicability, establish essential detail design criteria such as pre-treatment needs, and cost effectiveness of the above options.

RO does have limitations that must be considered. A typical RO system will generate about 20 to 25% of reject streams. Rejected water from NF/RO must be managed properly to avoid further contamination of surrounding water and in accordance with applicable regulations. Reducing the quantity of rejected water will determine the selection of either RO or NF. Further reduction of reject to approximately 5% could be achieved through High Recovery RO systems. Systems must account for the disposal of the waste by-products, one of which is a high level of total dissolved solids.

Finally, due to its fine membrane construction, reverse osmosis not only removes harmful contaminants present in the water, but it also may strip many of the good, healthy minerals from the water. The addition of such minerals is a common practice in the industry.

According to the ITRC report on perchlorate (2008), the costs of RO are dependent on "power needs, water chemistry (pre-treatment, post-treatment and pH adjustment) and labor," although labor costs can be low due to automation of the equipment. The cost of proper disposal of brine can present challenges and should also be considered. The cost of RO systems could be reasonable for groundwater systems since the expected TDS in groundwater would be <500 mg/L. Low pressure RO could be applied (operating at <250 psi) for treatment. The expected cost of producing treated water can range from $0.70 to $1.00 per cubic meter of treated water (i.e. approx. range from $2.65 to $3.80 per thousand gallons) (Sentilnathan, 2014). The use of centralized reject processing/management facilities to serve several local satellite water treatment plants could be considered to minimize capital and operating costs.

Anion Exchange

Ion exchange involves the use of resins (i.e. very small plastic porous beads with a fixed charge) that are used to exchange undesirable ions with hydrogen or hydroxyl, to produce potable water. The rate of removal is dependent on initial concentration of the contaminant, the concentration of competing ions, treatment design (i.e. flow rate, size of resin beads) and the nature of the ions within the beads (ITRC, 2008). Anion exchange has been studied for its ability to remove PFOA, PFNA and PFOS. One publication reported on removal of PFOA, and PFOS at a New Jersey drinking water treatment plant using "porous anion exchange resin impregnated with iron oxide" to treat arsenic of 76% and >90%, respectively (Rahman et al., 2014). In one study, two anion exchange treatments, not designed for PFC removal, demonstrated the effective removal of PFOA (74%), PFNA (>67%) and PFOS (>92%) (Appleman et al., 2014). The same study noted that shorter chain PFCs, which are not the subject of this report, were not removed. Both studies note that further research is needed to identify the most suitable resins for removal, understand the necessary frequency of resin changes, and its ability to be regenerated. Such research is ongoing with the Water Research Foundation's Project #4322, which should be published later this year (WRF, 2014). The management of both the resin (e.g. incineration, landfill or regeneration) and of brine (e.g. chemical or biological processes or disposal) must also be considered (ITRC, 2008).

Costs vary depending on the type of resin and treatment system. Capital and operating costs for single-use tend to be lower than for regenerable he concentration of competing anions in the water, which affects efficiency of resins to remove anions, and required disposal of brine or resins (ITRC, 2008). With respect to disposal, the ITRC study noted that there is new technology that allows regeneration of single-use resin and another that resulted in the complete destruction of perchlorate without altering the properties of the regenerant solution such that it can be used over and over again with no waste regenerant being produced (ITRC, 2008). Although this study investigated perchlorate removal, due to their similar structure (i.e. one polar end, one non-polar end) it is expected that long-chain PFCs to be adsorbed by ion exchange mechanism similar to perchlorate.

Advanced Oxidation

Conventional oxidation is not an effective treatment for PFCs due to the resistance of the fluorine bond to oxidation (Appleman et al., 2014). However, advanced oxidation processes, which rely on highly reactive hydroxyl radicals (-OH) to remove contaminants have shown some removal (Ribeiro and WRF, 2014). The Water Research Foundation reported removal rates of less than 10%, while another study showed removal of PFOS between 10 – 50%, depending on the process used (Riberio et al., 2015). The oxidation process can destroy contaminants and change them into less complex compounds (Ribeiro et al., 2015). However, if the contaminant is not entirely destroyed, pre-treatment for wastes may be necessary. Research on these processes continues, but at this time the effectiveness appears to be limited when compared to the other techniques described herein.

Point-of-Use Treatment

Although the DWQI is charged with making recommendations as they related to the Safe Drinking Water Act, which regulates public water systems, it is worth noting that the efficiency and effectiveness of point-of- use treatment systems was evaluated in a document prepared for the Minnesota Department of Health. The final report entitled, Performance Evaluation Removal of Perfluorochemicals (PFC's) with Point-of-Use (POU) Water Treatment Devices Final Report (Olsen & Paulson, 2008). This document is available at http://www.health.state.mn.us/divs/eh/wells/waterquality/poudevicefinal.pdf . It evaluated devices in two categories: those using GAC and those use a multiple methods of removal in combination. Eleven devices were found to remove PFCs in field tests below the employed detection limits (50 ng/L).

Waste Management

The waste stream produced by a particular treatment is only one consideration when selecting an appropriate treatment method, however, because improper waste disposal has the potential to re-create the very problem that treatment is seeking to address, it warrants extra emphasis. The method and cost for disposal of brine, reject water, resins or spent media waste products should be researched and considered during the selection process. Disposal methods should ensure that contaminants are not simply released back into the environment. USEPA notes that "incineration of the concentrated wastes would be needed for the complete destruction of PFCs" (2014).

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