

Review of New Jersey Second Addendum to Appendix C: Recommendation on Perfluorinated Compound Treatment Options

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Executive Summary

The treatment system must be capable of removing both PFOA and PFOS, as well as providing best treatment technology available to remove other perfluorinated compounds, such as PFBA, that pose some toxicity. GAC alone does not remove low carbon number perfluorinated carboxylic acids (e.g. PFBA) and, at times, low carbon number perfluorinated sulfonic acids (e.g. PFBS). There are differential removal efficiencies among perfluorinated compounds through GAC systems. As described in the full-scale GAC operating systems and research discussed herein, PFOA is often marginally removed by GAC alone. Granular activated carbon (GAC) followed by reverse osmosis technology is needed at public water treatment systems to assure removal of all perfluorinated compounds. Further testing of nanofiltration, as discussed in the Subcommittee's Recommendation on Perfluorinated Compound Treatment Options (2015), may demonstrate adequacy to remove PFOA and low carbon number perfluorinated compounds, as a substitute for reverse osmosis.

Prevalence of PFCs in New Jersey Drinking Water

PFOS and PFOA are found in New Jersey water supplies at relatively high concentrations, in some instances at levels greater than the existing 14 ng/l MCL for PFOA and the proposed 13 ng/l MCL for PFOS. We expect that in excess of one million people are ingesting PFOS and PFOA at these levels. PFOS levels in some New Jersey PWS (public water supplies) are such that ingestion of this contaminated water further increases residents' PFOS blood serum levels beyond those already found in epidemiologic studies in the U.S. population to be associated with adverse health effects (immunotoxicity).

Following discovery of PFOA levels in PWS (public water systems) at levels up to 190 ng/l in groundwater and 64 ng/l in tap or finished drinking water, New Jersey Department of Environmental Protection completed two studies, in 2006 and 2009-2010, to test for PFOS, PFOA, and other perfluorinated compounds in 53 PWS (NJDWQI 2017).

PFOS was found in the 2006 study in 30% of 23 PWS tested at or above the minimum reporting limit (MRL) of 4 ng/L (NJDWQI 2017), with the highest PFOS level at 19 ng/L (NJDWQI 2017). In the 2009-2010 study raw water was tested in 30 PWS in 19 of New Jersey's 21 counties. PFOS was found in 8 of 29 PWS sampled at levels up to 12 ng/L in 5 PWS using groundwater, and up

to 43 ng/L in 3 PWS using surface water (NJDWQI 2017). Finished drinking water in these PWS would be expected to contain the same concentrations, since minimal to no removals of perfluorinated compounds are achieved through conventional water treatment technologies.

Testing of 175 New Jersey PWS, including 165 large community systems and 10 small community systems, completed under the UCMR3 (Third Unregulated Contaminant Monitoring Rule) found 6 PWS with PFOS levels exceeding 40 ng/L, and 18 PWS with PFOA levels exceeding 20 ng/L (UCMR3 2017, NJDWQI 2017). Based on the New Jersey DEP database we note that 10 of 76 PWS tested were found to contain PFOS \geq 20 ng/L, with 7 PWS containing PFOS at levels exceeding 40 ng/L.

The 80 PWS tested represent about 14% of the total community water supplies in New Jersey. In 2016 New Jersey had 581 community water systems (NJ DEP Division of Water Supply and Geoscience 2017), serving about 91% of the total population, of which 42% were medium to large systems, and 58% were small systems.

State-wide studies of PFOA and PFOS in private wells have not been conducted in New Jersey. About 12% of New Jersey's population obtains drinking water from private wells (NJDEP Division of Water Supply and Geoscience 2017). Although it is likely the majority of these wells are not contaminated, groundwater at locations proximate to industrial activities using perfluorinated compounds or where AFFF (aqueous fire fighting foam) has been used may exhibit levels of PFOS and PFOA. PFOA has been found at levels exceeding 40 ng/L (maximum >400 ng/L), in 59 private wells within 2 miles of a New Jersey industrial source (NJDWQI 2016 Report, DuPont, 2009). PFOS was found in private wells at levels above the USEPA advisory of 70 ng/L (PFOS + PFOA) and the proposed MCL of 13 ng/L near sites contaminated with fire fighting foam (NJDWQI 2017).

Treatment Technologies and Capabilities, Chemistry of PFSA's versus PFCAs

The NJDWQI Treatment Subcommittee states in its 2015 report, Appendix C: Recommendation on Perfluorinated Compound Treatment Options for Drinking Water: "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)". The NJDWQI subcommittee does not diverge from this position in the Appendices to the 2015 Report. We disagree with this position. Peer-reviewed studies show that treatment options differ in removal capability among perfluorinated compounds.

Although perfluorinated compounds have somewhat similar structure, polarity, and solubility, there are differences in structural chemistry that affect removal among treatment options. Specifically, the charged functional group, carboxylic or sulfonic acid, affects the adsorption capability of activated carbon. PFSA's (perfluoroalkyl sulfonic acids, e.g. PFOS) are stronger acids and more hydrophobic compared to PFCAs (perfluoroalkyl carboxylic acids, e.g. PFOA, PFBA). Therefore, their tendency to adsorb onto activated carbon is greater.

In a review of adsorption behavior of perfluorinated compounds (mostly PFOS and PFOA) by Du

et al. (2014) sorption capacities of PFSA (e.g. PFOS) onto activated carbon or other adsorbents were observed to be higher than PFCAs (PFOA) with the same carbon numbers due to greater hydrophobicity of PFSA versus PFCAs (Du et al. 2014).

Activated column experiments by Ostlund (2015) found higher removal efficiency of PFSA than PFCAs, comparing the same number of carbons in the perfluorocarbon chain length, indicating that that functional group affects removal efficiencies of PFASs; "sulfonic group resulted in higher removal efficiency compared to carboxylic group" (Ostlund 2015). This study also found that branched isomers (for PFOS) were less efficiently removed by GAC (granular activated carbon) compared to linear PFOS isomers. We note this finding could, in part, account for differences in PFOS removal among locations using GAC.

The Water Research Foundation study of 15 full-scale water treatment systems in the U.S., including two potable reuse treatment systems, found that full-scale anion exchange and GAC column treatments were more effective at removing long-chain perfluorinated compounds and PFSA (e.g. PFOS) versus PFCAs (e.g. PFOA, PFBA) (Water Research Foundation 2016). Full-scale reverse osmosis systems demonstrated significant removal for all perfluorinated compounds, including the smallest, perfluorobutanoic acid (PFBA).

The WRF (2016) study further evaluated nanofiltration (NF) for removal of a suite of PFCAs and PFSA and notes that NF "has been deemed potentially effective (> 95%) in bench-scale experiments using NF270 membranes" (WRF 2016; Steinle-Darling and Reinhard 2008). WRF (2016) indicated that NF may be as capable of rejecting (treating) perfluorinated compounds as reverse osmosis at lower cost.

A study of sorption onto GAC, zeolite, and sludge found that PFOS is strongly adsorbed by GAC; PFOA and PFBS were also removed by GAC but to a lesser extent. The authors noted "that the length of the fluorocarbon chain and the nature of the functional group influenced sorption of the anionic surfactants" (Ochoa-Herrera and Reyes-Sierra 2008).

Perfluorinated compound removal was studied at two water reclamation plants (treating domestic effluents as influent) in Southeast Queensland, Australia. In the treatment plant using reverse osmosis, PFOA was removed to less than reporting level to 1.4 ng/L, from influent levels ranging from 15 to 27 ng/L, and PFOS was removed to less than reporting level with influent PFOS levels ranging from 23 to 39 ng/L. In the treatment plant using biologically activated carbon, PFOA and PFOS were ineffectively removed, although lack of removal may have been due to the age of the carbon or short contact times (Thompson et al., 2011).

In a study at a water treatment plant in Amsterdam using GAC, PFOA was not effectively removed, with a final (treated) mean PFOA concentration of 5.3 ng/L (range 0.8 ng/L - 9.4 ng/L) versus a mean influent (raw) PFOA concentration of 4.4 ng/L (range 3.8 ng/L - 5.2 ng/L). The authors found greater removals of PFOS and PFNA with a mean final (treated) water level of <0.23 ng/L and <0.24 ng/L, respectively, versus influent (raw) levels of 6.7 to 10 ng/L for PFOS and 0.5 to 0.8 ng/L for PFNA (Eschauzier et al. 2012). This study also found that PFBA, PFPeA,

PFHxA, PFOA, and PFBS were not well removed by the (operating) GAC filtration. In general, the authors found that PFOA decreased by only 50% using GAC.

A study in Spain suggests that although GAC alone was reasonably effective to remove PFOS, reverse osmosis was needed to achieve efficient PFOA removal. In this study 2 separate stages following conventional water treatment (GAC, or Ultrafiltration followed by Reverse Osmosis) were evaluated. The system treats 100 million gallons per day of surface river water to supply over 1 million inhabitants. The authors found that ultrafiltration/reverse osmosis removed PFOS and PFOA by $\geq 99\%$, but GAC alone removed PFOS and PFOA by only $64 \pm 11\%$ and $45 \pm 19\%$, respectively (Flores et al. 2013).

In a study monitoring drinking water treatment facilities across the U.S., a utility that used microfiltration and reverse osmosis for indirect potable reuse in wastewater treatment reduced total perfluorinated compound influent levels of 80 ng/L and influent PFOS of 41 ± 18 ng/L to no reportable levels. Minimum reporting levels were 1.0 ng/L for all perfluorinated compounds monitored except PFOA, where the minimum reporting level was 5 ng/L (Quinones and Snyder 2009).

Granular Activated Carbon Treatment Systems – Removal Efficiency

NJDWQI subcommittee describes 2 facilities in New Jersey, one in Pennsylvania, and one in Minnesota where GAC is used to treat perfluorinated compounds in public water supplies.

In the New Jersey Penns Grove GAC treatment system PFOS was reported at levels lower than the reporting limit of 5 ng/l in finished water, although the highest PFOS level in the raw water was 13 ng/L. At the New Jersey Logan System Birch Creek GAC system PFOS was reported at levels lower than the reporting limit of 5 ng/l in finished water, although the average PFOS raw water concentration was only slightly above the reporting limit, at 7 ng/L. We believe that PFOS and PFOA removal performance may be much different at other locations, where PFOS and PFOA are present in raw water at higher levels and/or where source water contains greater natural organic matter.

The Subcommittee notes the Horsham Water and Sewer Authority (HWSA) in Horsham, Pennsylvania, where GAC was recently installed to treat well water contaminated with perfluorinated compounds. Wells are believed contaminated from fire fighting foam used at the nearby Naval Air Station Joint Reserve Base - Willow Grove. NJDWQI subcommittee states that HSWA raw (well) water samples collected between January and March 2017 “show a range of PFOS concentrations from 230 - 1297 ng/L and an average of 629.3 ng/L”, and indicates PFOS was not detected in finished water. We characterize HSWA well data differently, based on our review of HWSA active well data (HWSA, PFOS PFOA Active Source Monitoring Results with Charts, 2017), as summarized in Table 1 below.

We observed that, during limited periods, post-GAC treatment in 4 HSWA contaminated wells

did not remove PFOS or PFOA to levels below the reporting limit of 5 ng/L. This appeared to occur just prior to carbon change-out, as shown in the HSWA data. However, it should be noted that GAC treated water is also blended with other HSWA wells and water sources, as shown in HSWA system schematics (HSWA June 2016), to assure that levels are below applicable standards and protective. As of December 2017 the HSWA indicates “the combined concentration of PFOS/PFOA from all sources currently supplying the public system is approximately 4 ppt (ng/l)” (HSWA Dec 2017 Update). This is below the USEPA health advisory level applied of 70 ppt (ng/l) for combined concentration (PFOA + PFOS).

Based on HSWA data as of December 19, 2017, the following table summarizes results for active HSWA wells (HWSA, PFOS PFOA Active Source Monitoring Results with Charts, 2017).

HSWA well number	Period	PFOS lowest	PFOS highest	PFOA lowest	PFOA highest
7	May 5, 2016 - Dec 19, 2017	ND	11	3.1	11
10*	Jan 19, 2016 - April 5, 2017	16	76	12	48
17**	Jan 19, 2016 - Dec 22, 2016	50	110	20	37
21***	Jan 2016 - Jan 11, 2017	5.1	14	8.4	13
26****	Jan 25, 2016 - March 8, 2017	340	1297	640	1765
40*****	Jan 11, 2017 - March 24, 2017	230	1203	33	88

Table 1. Summary Table of HSWA Raw Water PFOS and PFOA Concentrations (ng/L)

ND – non detect at reporting limit 5 ng/L

*After April 5, well 10 was treated through GAC system with N.D results.

** After December 22, 2016 well 17 was treated with GAC. Results post GAC treatment in 2017 mostly ND (reporting levels 5 ng/L), except for 6 positive PFOS values ranging from 3.1 to 159 ng/L, and 5 positive PFOA values ranging from 13 ng/L to 29 ng/L.

*** After January 2017 well 21 was treated with GAC. Results post GAC treatment ND (reporting levels 5 ng/L), except one PFOS value of 4.2 ng/L, and 5 PFOA values ranging from 3.3 to 7.8 ng/L.

**** After March 8, 2017 well #26 was treated with GAC. Results post GAC treatment ND, except for one PFOS value of 5 ng/l and one value of 5 ng/L for PFOA

***** After March 24, 2017 well #40 treated with GAC. Results post GAC treatment ND, except 4 PFOS

values ranging from 2.5 ng/L – 131 ng/L and 3 PFOA values ranging from 3.1 ng/L to 5.3 ng/L

The Second Addendum discusses the city of Oakdale, Minnesota GAC system used to remove perfluorinated compounds, including PFOS, from well (drinking) water. NJDWQI subcommittee notes that the Oakdale PFOS method detection limit is 0.5 ng/L and the Minimum Reporting Limit (MRL) is 5 ng/L. However, these limits were only recently put into effect by the Minnesota Department of Health (Rinker communication, Jan 5, 2018). (A MRL for PFOA of 5 ng/L was also recently put into effect.) Prior RLs (reporting limits) for PFOS and PFOA at Oakdale were 25 ng/L and 35 ng/L, respectively. The Subcommittee states that “samples taken after GAC treatment show no detection of PFOS”. However, these non-detects are based on the prior RLs (25 ng/L and 35 ng/L), not 5 ng/L. It is yet unknown whether the Oakdale GAC system will remove PFOS or PFOA to ≤ 5 ng/L.

Based on Oakdale’s perfluorinated compound data (Bachmeier 2017), PFOS was non-detect in finished drinking water for the period November 2015 through November 2017, at a RL of 50 ng/L for the period November 2015 through July 2016, and a RL of 25 ng/L for the period August 2016 to present. PFOA was non-detect for all samples at a RL of 50 ng/L for the period November 2015 through July 2016, and a RL of 35 ng/L for the period August 2016 to present. We observed that PFBA (perfluorobutanoic acid) remains at relatively high levels in Oakdale’s drinking water, typically at around 1400 ng/L, consistent with other installations and research showing poor or no removal by GAC of low carbon number PFCAs. Generally PFBA passes through unchanged in concentration through the Oakdale GAC system. However, PFBA levels in Oakdale’s finished water are well below the Minnesota Department of Health HRL (health risk limit) for PFBA of 7000 ng/L.

The 3M Cottage Grove manufacturing plant in Cottage Grove, Minnesota operates a GAC system to treat wastewater discharged to the Mississippi River, installed in 2004 pursuant to requirement by the NPDES (National Pollutant Discharge Elimination System) permit. 3M was the primary global producer of PFOS-related perfluorinated compounds, and PFOA, and manufactured these chemicals at its two U.S. plants in Decatur, Alabama and Cottage Grove, Minnesota, and in Europe at its plant in Antwerp, Belgium. 3M perfluorochemical production began at the Minnesota plant around 1950 (Oliaei et al. 2006).

Based on one sampling event in 2006 by Minnesota Pollution Control Agency staff, the 3M GAC treatment facility removed PFOS by 95% and PFOA by 79%. The GAC treatment plant was less effective at removal of carboxylic perfluorinated compounds (PFCAs). In the 2006 sampling of post-GAC treated wastewater perfluorinated compound concentrations were very high: PFOA 1670 ng/L, PFOS 1330 ng/L, PFBS 169,000 ng/L, and PFBA 58,100 ng/L (Oliaei et al. 2006).

Since 2006 levels of perfluorinated compounds in the 3M discharge (post-GAC treated wastewater) are considerably lower, but remain elevated, as summarized in the following table for the period October 2015 through November 2017 (NPDES 3M data provided by Marco Graziani, Minnesota Pollution Control Agency, January 2018).

Period	PFOA ng/L	PFBA ng/L	PFNA ng/L	PFOS ng/L	PFBS ng/L
Oct-15	126	13100	<25	<46.4	94.7
Nov-15	<48	102000	<25	<23.2	1170
Dec-15	61.9	8470	<25	<23.2	1410
Jan-16	38.2	36500	<25	<46.4	1660
Feb-16	166	27500	<25	<23.2	210
Mar-16	329	15400	<25	27.8	217
Apr-16	584	55700	<25	58.1	434
May-16	<24	28100	<25	<23.2	353
Jun-16	75.9	6040	<25	<23.2	120
Jul-16	193	83700	<25	<23.3	817
Aug-16	116	380000	<25	<46.4	644
Sep-16	81.6	21200	<25	<23.2	1240
Oct-16	90.3	13400	<25	<23.3	469
Nov-16	194	18200	<25	43.3	848
Dec-16	370	19200	<25	69.3	286
Jan-17	77.2	7470	<25	25.4	195
Feb-17	108	6870	<25	26.6	201
Mar-17	157	12800	<25	37.5	6240
Apr-17	265	10700	<25	44.9	676
May-17	165	11000	<25	38.6	465
Jun-17	113	14100	<25	71.4	1340
Jul-17	170	21700	<25	28.7	3740
Aug-17	328	13900	<25	60.3	2580
Sep-17	50.6	4090	<25	<23.2	1780
Oct-17	129	11600	<25	29.4	18700
Nov-17	133	7680	<25	<46.4	6920
~ mean	172	36555	<25	43	2031

Table 2. 3M Perfluorinated Compound Discharge (SD001) Post-GAC Treatment ng/L

Mean concentrations in Table 2 are means of positive values, excluding non-detects. The post-GAC discharge perfluorinated compound discharge concentrations for October 2015 to present remain high: mean PFOA 172 ng/L, mean PFBA 36,555 ng/L, mean PFOS 43 ng/L, and mean PFBS 2031 ng/L. However, these concentrations are much lower than those discharged during the period January 2007 through July 2010: mean PFOA 2989 ng/L, mean PFBA 54,098 ng/L, mean PFOS 595 ng/L, and mean PFBS 18,673 ng/L. The 3M data indicate that low carbon perfluoroalkyl compounds (such as PFBS and PFBA) and PFCAs (e.g. PFBA, PFOA) are not removed by this GAC system to low levels.

Recommended Treatment - GAC Followed by Reverse Osmosis

GAC alone has not been shown in most cases to consistently remove PFOA to low ng/L levels. Therefore, GAC followed by reverse osmosis (RO) is required to remove PFOA and, in some cases PFOS, to assure consistent removal. GAC followed by RO will also enable removal of low carbon number perfluorinated compounds such as PFBA. PFBA has been shown in animal studies to cause toxicity. The Minnesota Department of Health identified critical toxic effects of “liver weight changes, morphological changes in liver and thyroid gland, decreased TT4, decreased red blood cells, decreased hematocrit and hemoglobin, and subcritical toxic effects of increased relative thyroid weight, decreased serum TT4 and dFT4, decreased cholesterol, and delayed eye opening (rat)”, in PFBA animal studies as the basis to derive a HRL (health risk limit) for PFBA (MDH 2017 Perfluorobutyrate).

A consideration using GAC/RO in large municipal treatment systems is disposal of the RO reject. Technologies to treat RO reject are generally limited to evaporative technologies applied to high salt concentrating RO systems, to eliminate the reject discharge. Evaporative systems require excessive energy input and are often prohibitively expensive. RO reject evaporative systems may be relatively cost effective, however, in arid climate locations to allow lined evaporation ponds, or where untreated reject water does not pose a concern or environmental impact (such as RO systems used for drinking water treatment in coastal areas, where the salt RO reject is discharged to the ocean). Such is not the case for the temperate geographic region under consideration in New Jersey. Thus, RO rejects at large GAC/RO plants would likely require direct discharge to a receiving water.

The GAC system, however, will enable removal of a significant mass of PFOS, and to a lesser extent, PFOA. A primary concern with waters receiving perfluorinated compounds is uptake of PFOS in fish, which bioaccumulates, and subsequent consumption of PFOS-contaminated recreationally caught fish. PFOA does not bioaccumulate. In any case, the mass of perfluorinated compounds, including PFOS and PFOA, discharged to a receiving water in a GAC followed by reverse osmosis system (reject) would not be greater than the mass discharged by a GAC system alone.

NJDWQI briefly discusses Point of Use (POU) drinking water systems, for use in individual homeowners on private wells, and certification by NSF (National Sanitation Foundation). NSF developed protocol NSF P473 to evaluate drinking water treatment device capability to reduce PFOA and PFOS in drinking water. The NSF certified a number of systems using GAC or GAC/RO that meet the EPA “standard” (combined PFOS and PFOA) of 70 ng/L. The NSF protocol included “challenge” of influent level of 1500 ng/L [5 parts PFOA and 10 parts PFOS by weight] to the GAC and GAC/RO systems. Studies by NSF showed good removal performance by GAC with highest performance in POU systems using GAC followed by reverse osmosis (NSF personal communication, E. Valentine, Jan 4, 2018)

We concur that GAC/RO POU systems offer a treatment solution to homeowners on private wells. Perfluorinated compounds do not volatilize and therefore inhalation via showering and bathing do not pose an exposure pathway, versus other compounds such as DBPs (disinfection byproducts) where inhalation in showering may comprise a significant portion of total

exposure. In addition, perfluorinated compounds do not cross the dermal barrier. However, further testing is needed to assure that POU GAC/RO systems remove PFOS, PFOA, and PFNA to low ppt levels. Use of POU GAC/RO systems is somewhat complicated by the requirement of homeowner management of carbon change-out and proper operation.

Conclusion

The proposed New Jersey MCL of 13 ng/L, our recommended MCL in this review of 5 ng/L, the Minnesota limit (HRL) of 27 ng/L, and the Vermont drinking water advisory of 20 ng/L (combined) for PFOS are within the same very small “ballpark”. Changing an uncertainty factor or exposure value used in these derivations obscures the difference in values. Accordingly, further emphasis should be placed on treatment and removal.

In addition to shorter chain perfluorinated compounds such as PFBS and PFBA, PFOA usually co-exists with PFOS in water supplies. Although the shorter chain perfluorinated compounds (e.g. PFBA, PFBS) are less toxic and excreted faster than longer chain perfluorinated compounds (e.g. PFOS, PFOA), they remain persistent, as demonstrated in the Oakdale, MN drinking water supply. The toxicity of shorter chain perfluorinated compounds is not fully understood, although toxicity of PFBA has been shown in animal testing and, in the case of Minnesota, a health risk limit (limitation) has been derived. There are differential removal efficiencies among perfluorinated compounds in GAC systems. PFOA is often poorly removed by GAC alone, as described in the full-scale GAC operating systems and research discussed above. Short chain perfluorinated carboxylic acids (e.g. PFBA) generally remain unchanged (not removed) through GAC systems.

The treatment system chosen for removal of perfluorinated compounds must be capable of removing both PFOA and PFOS, as well as providing best treatment technology available to remove other perfluorinated compounds, such as PFBA, that pose some toxicity. Removal of shorter chain perfluorinated compounds requires reverse osmosis in addition to GAC. Granular activated carbon (GAC) followed by reverse osmosis, or nanofiltration if pilot-scale studies demonstrate efficiency, is needed as a combined option to adequately remove both PFOS and PFOA, as well as other perfluorinated compounds that may be present.

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