Estimated Greenhouse Gas Emissions from PFAS Treatment of Maine Drinking Water—Appendix

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METHODS

The sustainability field distinguishes between life cycle analysis and environmental footprint analysis. Life cycle analysis is a complex modeling exercise that develops impact tallies across many factors, whereas environmental footprint analysis uses limited inventories and can focus on specific environmental effects (Favara et al. 2011). Given the need for an expeditious estimate to inform the rulemaking process, we conducted the latter, focusing on GHG emissions using a literature review for emissions factors of key components of water treatment to remove PFAS. Rather than being sufficient for design of any particular water treatment system, the estimates are meant as a point of reference for policymakers considering potential drinking water standards for this class of contaminants.

For the private residential water scenario, the untreated groundwater concentration was obtained by first downloading the full set of groundwater sampling results from data made available to the public by DEP on the web (https://www.maine.gov/dep/spills/topics/pfas/; March 7, 2022 dataset). These available data are based on site prioritization by DEP with expected PFAS presence (e.g., sludge spreading), so may not be indicative of conditions statewide. Still, the data provided a useful starting point for the current evaluation. Sites with an exceedance of the interim drinking water sum-of-six standard (0.020 micrograms per liter [µg/L]) were established as a subset of the DEP database. Then the 75th percentile concentration for each of the six PFAS in this subset was calculated and used as the private residential water initial value (Table 1). Municipal water supply information for PFAS in Maine were generally not available at the time of writing, as the sampling deadline is not until the end of 2022 (SP 64 – LD 129). The Kennebunk River Well was sampled in November, 2014, and had a PFOS detection of 0.050 µg/L (PFOA not detected). For the purposes of the current demonstration and to allow consideration of variable treatment depending on PFAS constituent, this 0.050 µg/L concentration was applied to all six of the regulated constituents.

Water treatment was simulated using granular activated carbon because this is a widely demonstrated and applied technology and sets the benchmark for removal of PFAS from drinking water (ITRC 2020). The method involves passing contaminated water through a column of the adsorbent, where PFAS and other nontarget compounds (e.g., dissolved natural and synthetic organic matter [Siriwardena et al. 2019]) partition to the adsorbent surface. This technology is expected to be the primary method as treatment needs expand across the state of Maine.

Adsorption calculations are often modeled using a Freundlich isotherm that has two model parameters: K_F and 1/n. The K_F value indicates capacity under pseudo-equilibrium and 1/n describes the uniformity/heterogeneity of adsorption with changing target water concentration:

$$q_e = K_F C_e^{1/n}$$

where q_e is equilibrium solid phase concentration (µg PFAS / g GAC) and C_e is the equilibrium aqueous phase or

TABLE 1: **Untreated Water Concentrations for Treatment Simulations**

Constituent	Residential well (µg/L)	Hypothetical municipal well (µg/L)
PFOA	0.175	0.050
PFOS	0.087	0.050
PFNA	0.011	0.050
PFHxS	0.020	0.050
PFHpA	0.067	0.050
PFDA	0.005	0.050
Sum of six	0.365	0.300
Data sources	75th percentile for sites in ME DEP database (March 7, 2022 download) with at least one exceedance of the sum-of-six interim standard.	0.05 µg/L PFOS value for Kennebunk River Well November 25, 2014 sample, applied across the other 5 PFAS (hypothetical only)

treated water concentration (μg/L). The equation above was of particular use for the current evaluation because the treated water concentration can be varied to assess the adsorption performance of different potential PFAS cleanup levels. However, it is noted that the "e" subscripts in the equation indicate equilibrium conditions, which are achieved when water is allowed to contact the adsorbent for long periods of time. Actual treatment systems that run in continuous mode for shorter contact times may not reach equilibrium. We conducted a separate, comparative analysis to confirm that activated carbon usage rates by our approach were consistent with those reported in a full-scale application (Belkouteb et al. 2020).

Adsorption efficiency to granular activated carbon has been evaluated widely for PFOA and PFOS and less so for other PFAS (Crone et al. 2019). For those two compounds, the K_F values have been observed to span large ranges, affected by both water quality factors and characteristics of the activated carbon. For the current evaluation, we used the Freundlich isotherm to model adsorption, using a study that included all six PFAS compounds currently regulated in the state of Maine (Burkhardt et al. 2022). The study had additional advantages including (1) it used raw water that had undergone one step of biological filtration, which would be expected for typical future applications (especially municipal) that seek to optimize activated carbon efficiency for PFAS removal, and (2) it tested activated carbon sourced from both a fossil source (i.e., coal by Calgon Filtrasorb 400) and a biomass/green source (i.e., coconut by Evoqua 1230AWC). One disadvantage of the dataset was that it treated river water, which may have a different mixture of nontarget compounds (e.g., dissolved organic matter) than groundwater. Still, it provided a data set to run a full set of simulations. Specific isotherm values are presented in Table 2.

The first adsorption simulation set a target or post-treatment sum-of-six PFAS concentration to a value of 0.020 μg/L. This is the current interim standard for the state of Maine. It also set four additional target concentrations: 0.1 times the standard (0.002 μg/L), 0.5 times the standard (0.010 μg/L), 2 times the standard (0.040 μg/L), and 10 times the standard (0.200 μg/L). This allowed for systematic comparison of adsorption and related GHG emissions based on level of treatment. The sum-of-six target concentration was divided equally between the individual PFAS. For instance, a 0.020 μg/L sum-of-six target concentration was partitioned to 0.0033 μg/L for each of PFOS, PFOA, PFNA, PFxHS, PFHpA, and PFDA such that all six PFAS were treated. An actual PFAS treatment system may achieve the sum-of-six standard by reaching unequal concentrations of individual compounds, so the equal partitioning used here can be considered a simplification.

Water volumes requiring treatment were set to an annual value. For private residential water treatment, the 2015 Maine self-supplied water use per capita value of 49 gallons per day (Dieter et al. 2015) was multiplied by 4, for a household total of 70,000 gallons per year. For the municipal water treatment scenario, the Kennebunk River Well flowrate f

TABLE 2. **Activated Carbon K_F and 1/n Values for Adsorption Simulations**

Constituent	Calgon Filtrasorb 400 (coal-sourced) K_F [(μg/g)(L/μg)$^{-1/n}$]	Calgon Filtrasorb 400 (coal-sourced) 1/n	Evoqua 1230AWC (coconut-sourced) K_F [(μg/g)(L/μg)$^{-1/n}$]	Evoqua 1230AWC (coconut-sourced) 1/n
PFOA	8.95	0.7	3.96	0.51
PFOS	79.3	1.00	4.54	0.4
PFNA	9.43	0.70	9.91	0.72
PFxHS	21.6	0.85	24.3	0.88
PFHpA	1.8	0.3	1.85	0.31
PFDA	3.9	0.51	4.56	0.54
Data sources	Burkhardt et al. (2022) bituminous coal activated carbon. Water is Cape Fear River following biological treatment.		Burkhardt et al. (2022) coconut activated carbon. Water is Cape Fear River following biological treatment.	

700 gallons per minute (https://kkw.org/kennebunk-river-well-pfas-information/), or 367 million gallons per year, was used. For a Maine public-supplied per capita value of 52 gallons per day (Dieter et al. 2015), this corresponds to water supply for approximately 19,000 people.

With an initial water concentration for each type of PFAS (Table 1), Freundlich adsorption coefficients (Table 2), target water concentrations, and treated water volumes, it was possible to calculate a mass of granular activated carbon required per year for each treatment scenario. This activated carbon quantity for each scenario was then multiplied by emissions factors for various components of the treatment process to generate an annual GHG emissions footprint. Specific components of the treatment process included in the emissions estimates are described below and detailed in Table 3.

Adsorption treatment for water typically involves passing water through lead and lag vessels that contain granular activated carbon. Two 48-inch tall fiberglass treatment vessels were incorporated for the private residential water treatment scenario. A fiberglass (E-glass) GHG emissions factor was obtained from (Dai et al. 2015). For the municipal

TABLE 3: **Emissions Factors for Greenhouse Gas Emissions Estimates**

Description	Value	Units	Source	Notes
Activated Carbon Generation				
Coal-based activated carbon generation	18.28	kg CO_2 eq / kg AC	Gu et al. (2018)	
Woody biomass-based activated carbon generation	8.6	kg CO_2 eq / kg AC	Gu et al. (2018)	Used as value for coconut-based AC
Activated Carbon Transportation from Vendor				
Coal-based activated carbon transport to or from Kennebunk, ME	0.95	kg CO_2 eq / kg AC	SiteWise	AC obtained from Calgon, Moon Township, PA. Assumes spent AC is picked up in Kennebunk when new AC is delivered.
Coal-based activated carbon transport to or from Fairfield, ME	1.13	kg CO_2 eq / kg AC	SiteWise	AC obtained from Calgon, Moon Township, PA. Assumes spent AC is picked up in Fairfield when new AC is delivered.
Coconut-based activated carbon transport to or from Kennebunk, ME	0.92	kg CO_2 eq / kg AC	SiteWise	AC obtained from Pittsburgh, PA. Assumes spent AC is picked up in Kennebunk when new AC is delivered.
Coconut-based activated carbon transport to or from Fairfield, ME	1.0	kg CO_2 eq / kg AC	SiteWise	AC obtained from Pittsburgh, PA. Assumes spent AC is picked up in Fairfield when new AC is delivered.
Activated Carbon Regeneration				
Activated carbon regeneration	0.7	kg CO_2 eq / kg AC	He (2012)	
AC Vessel Installation				
Steel	1.77	kg CO_2 eq / kg steel	SiteWise	
AC vessel, large	682	kg steel	Recofiltration: https://recofiltration.com/liquid-scrubbers	Vessel holds 2,000 lbs AC
AC vessel, large	1,207	kg CO_2 eq / AC vessel, large (no shipping)	Calculated	
AC vessel shipping, large	5,438	kg CO_2 eq / AC vessel, large (shipping only)	SiteWise	Tank obtained from Recofiltration in Houston, TX. Assumes one way with vessel, second way without vessel in cargo.

TABLE 3: **Continued**

Description	Value	Units	Source	Notes
Fiberglass (E-glass)	0.158	kg CO_2 eq / kg fiberglass	Dai et al. (2015)	
AC vessel, small	23	kg fiberglass	General Carbon Corporation interview of staff	48 inch tall by 11 inch diameter tank
AC vessel, small	3.6	kg CO_2 eq / AC vessel, small (no shipping)	Calculated	
AC vessel shipping, small	1,130	kg CO_2 eq / AC vessel, small (shipping only)	SiteWise	Tank obtained from General Carbon Corporation, Paterson, NJ. Assumes one way with vessel, second way without vessel in cargo.

water treatment scenario, two steel vessels were incorporated, each sized to hold 2,000 pounds of activated carbon. A steel GHG emissions factor was obtained from the SiteWise™ Tool for Green and Sustainable Remediation (SiteWise; Battelle, Columbus, Ohio). For both scenarios, the emissions associated with the activated carbon vessels were divided by 5 to indicate a 5-year amortization. Actual steel vessel lifespan is expected to be greater than five years, but amortization in this case is based on the possibility that treatment technologies may change in this timeframe.

New (i.e., "virgin") activated carbon was used for the simulations to align with common practice. Processes to generate virgin activated carbon vary depending on the feedstock. In the case of coal-sourced activated carbon, mining processes generate GHG emissions, but the coal is readily amenable to activation. For contemporary biomass sources, the extraction process is less intensive, but carbonation (high temperature heating) to generate biochar is required before activation. One literature reference (Gu et al. 2018) was used for emissions factors of both coal- and coconut-sourced activated carbon. This reference reports emissions factors for coal and woody biomass activated carbon. Coconut shells likely have a similar emissions factor to this woody biomass and were used here. An alternative literature source specific to coconut shells (Arena et al. 2016) was available, but the results were presented as output from an LCA software as person-equivalents with insufficient information to convert to a CO_2 emissions rate.

Activated carbon transportation was included both as shipment of virgin material from the vendor and for shipment from the water treatment location back to the vendor for recycling. Mileages were estimated separately for coal-based activated carbon (Calgon, Moon Township, Pennsylvania) and coconut-based activated carbon (Evoqua, Pittsburgh, Pennsylvania). SiteWise™ software was used to convert mileages to GHG emissions rates based on diesel-powered road shipment.

Recycling (i.e., regeneration) of activated carbon was included to account for potential desorption of PFAS from the treatment media. An alternative scenario of simply disposing of activated carbon to a landfill was not considered, as this would result in leachate that requires treatment. At a minimum, recycling allows for concentration of PFAS for additional treatment. Further, the recycling process could potentially destroy some or all of the target PFAS compounds (Sonmez Baghirzade et al. 2021). For the purposes of an initial estimate, one general activated carbon emissions factor was used (He 2012). The simulations assumed that the recycled activated carbon is used for some other purpose than for additional PFAS treatment. This likely best simulates current practice for activated carbon recycling. As developed in the discussion, use of recycled activated carbon could provide a reduced GHG emissions footprint.

Finally, PFAS disposal following desorption was not included in the analysis. This part of the remediation field is still under significant technology development and emissions factors are not available at this time. A theoretical estimate for PFAS destruction could potentially be generated based on the energy required to break each of the carbon-fluorine bonds in the PFAS molecules. However, this would be an underestimate of a practical PFAS destruction energy, and

currently it is not possible to estimate the level of energy efficiency destruction technologies will eventually reach. Further, it is possible that PFAS destruction may be so energy-intensive that long term stabilization and disposal of PFAS may be the preferred approach. This is a large, developing topic and was not considered further for this analysis based on current practices.

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