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LIFE CYCLE SUSTAINABILITY ASSESSMENT

Evaluation of treatment options for well water contaminated with perfluorinated alkyl substances using life cycle assessment

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Abstract

Purpose As knowledge grows of the potentially harmful effects of chemicals in widespread use, emerging contaminants have become a major source of concern and uncertainty for public health officials and water quality managers. Perfluorinated alkyl substances, often referred to as perfluorinated compounds, have come under recent scrutiny and are present in groundwater at many sites across the USA. We examine the life cycle impacts of treating drinking water at one such site.

Methods We assembled life cycle models for groundwater treatment and bottled water delivery to residents of Wright-Patterson Air Force Base, Ohio, where wells were recently taken out of service due to concerns related to perfluoroalkyl and polyfluoroalkyl substance (PFAS) contamination. Two treatment methods, granular activated carbon filtration and ion-exchange columns, were modeled under a range of contaminant concentrations covering three orders of magnitude: 0.7, 7.0, and 70 μg/L PFAS. On-site infrastructure, operations, and adsorbent cycling were included in models. Impacts of bottled water production and supply were assessed using two data sets reflecting a range of production and supply chain assumptions. Uncertainty in input data was captured using Monte Carlo simulations.

Results and discussion Results show that for contaminant concentrations below 70 μg/L, the dominant contributor to life cycle impacts is electricity use at the treatment facility. Production, reactivation, and disposal of treatment media become major sources of impact only at very high PFAS concentrations. Though the life cycle impacts of bottled water are up to three orders of magnitude higher than remediated groundwater on a volumetric basis, supplementing a contaminated water supply with bottled drinking water may result in lower life cycle human health impacts when only a small proportion of the total population is vulnerable.

Conclusions These results provide quantitative data and proposed scenarios for water quality managers and risk management officials in developing plans to address PFAS contamination and emerging contaminants in general. However, more information on the direct human health effects of these poorly understood pollutants is needed before the trade-offs in life cycle health impacts can be comprehensively assessed.

Keywords Bottled water · Emerging contaminants · Life cycle assessment · PFAS · Water treatment

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1 Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFAS) have recently attracted attention as emerging contaminants of drinking water. Some PFAS have been associated with reproductive and developmental toxicity, cancer, endocrine disruption, and immunotoxicity (Grandjean and Clapp 2015; Merino et al. 2016; Schaider et al. 2017). Two PFAS compounds in particular, perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), are often singled out by regulators due to their solubility and bioaccumulative, toxic, and transport potential in the environment (Hale et al. 2017). The US EPA recently issued a health advisory limiting the combined concentration of PFOA and PFOS to 70 parts per trillion in drinking water, a concentration expected to protect vulnerable populations such as young children, pregnant, and nursing women (USEPA 2016a). Though these health advisories are non-enforceable and non-regulatory, many federal, state, and private organizations follow them voluntarily, including the US Department of Defense.

Use of material containing PFAS is ubiquitous due to the compounds' surfactant and anti-adhesive properties. PFAS are in fire- and stain-resistant household materials, food packaging, non-stick cookware, paints, adhesives, emulsifiers, and products containing Teflon (Schaider et al. 2017; USEPA 2016a). Sampling efforts across Europe, Australia, and the USA have reported PFOA and PFOS at or above 100 ng/L (nanograms per liter) in samples from multiple locations including rivers and wastewater treatment systems (Huset et al. 2008; Lang et al. 2017; Pistocchi and Loos 2009; USEPA 2016a). PFOS has been a principle active fluorinated surfactant in aqueous film forming foams (AFFF) used by the aviation community, the US Department of Defense (DOD), and other international defense agencies (Filipovic et al. 2015; Lloyd-Smith and Senjen 2015; Moody and Field 2000; Place and Field 2012). Fire training exercises, test and calibration duties, accidental releases from aircraft hangar fire suppression systems, and real-world emergencies have led to PFAS releases to the environment (Anderson et al. 2016) and contributed to the discovery of about 40 classes of PFAS in groundwater at many DOD installations (Barzen-Hanson et al. 2017; USEPA 2016b, 2016c).

PFAS are persistent synthetic organic compounds, recalcitrant to microbial degradation and various physical-chemical processes used in water treatment. Conventional treatment techniques such as coagulation, micro- or ultra-filtration, aeration, oxidation (e.g., permanganate, ultraviolet/hydrogen peroxide), low-pressure membranes, and disinfection have been mostly ineffective (Appleman et al. 2013; Rahman et al. 2014). Several studies have identified granular activated carbon (GAC) as a promising treatment technology, particularly for longer-chained PFAS including PFOA and PFOS (Appleman et al. 2013, 2014; Dickenson and Higgins 2016). Ion exchange (IEX) resins have also been successful at removing PFAS at various bench-scale studies and pilot system tests, achieving up to 99% removal of both PFOS and PFOA (Appleman et al. 2014; Dickenson and Higgins 2016). Concerns remain for scaling up ion exchange resins from pilot scale, such as media interactions with dissolved organic matter or other competing compounds, and the effectiveness and efficiency of media regeneration.

Some institutions have responded to the EPA health advisory by supplying bottled water to populations whose water may exceed the recommended maximum concentrations for PFOA and PFOS as either a temporary or permanent solution. Though bottled water has been shown to have relatively high life cycle impacts relative to tap water (Fantin et al. 2014), differences in the quantity of water supplied through drinking water distribution and municipal water systems could affect the net impacts of bottled water and groundwater treatment scenarios. Environmental implications of bottled water vary depending on the size of the bottle, transportation distance from the bottler to consumer, and particular processing, labeling, refrigeration, and bottle disposal methods (Amec Foster Wheeler 2017). For water packaged near to the consumer, the energy requirements are dominated by the energy used to produce the plastic bottles, while bottled water requiring for long-distance shipment, energy costs for transportation can be comparable to or even larger than energy used for plastic bottle manufacture (Gleick and Cooley 2009).

In this study we examine, using life cycle assessment (LCA), two methods of high-throughput groundwater treatment that have been shown to be effective in eliminating PFAS (GAC and IEX). LCA is a widely recognized method to evaluate "cradle-to-grave" environmental impacts from a product or a process. According to ISO 14040 series of standards, it involves a goal and scope definition, inventory analysis, impact assessment, and interpretation (Finkbeiner et al. 2006). For this study, we assessed the life cycle impacts to climate, human health, and ecosystem health of operating these systems at PFAS concentrations representative of some contaminated sites in the USA (e.g., CDPH 2016; USEPA 2016a, b, c, d). We compare these results to life cycle analyses of bottled water provisioning as a substitute for PFAS-contaminated drinking water. A contaminated aquifer at Wright-Patterson Air Force Base (OH, USA) served as case study for the analysis.

2 Methods

We assembled life cycle models for clean water supply to residents of Wright-Patterson Air Force Base near Dayton, OH, where two groundwater production wells were recently taken out of service due to concerns related to PFAS contamination. Water supply scenarios include groundwater treatment and bottled water delivery. Two treatment methods, granular activated carbon (GAC) filtration and ion-exchange columns (IEX), are examined under a range of contaminant concentrations covering three orders of magnitude. Bottled water models are based on two major data sources, a prior life cycle study of beverage production with detailed process data for bottle manufacturing (Quantis 2010) and a broader review of the scientific literature. Figure 1 shows the processes modeled in each scenario. Our analyses are based on a functional unit of 1 m^3 of safe drinking water delivered to a residential customer at Wright-Patterson Air Force Base.

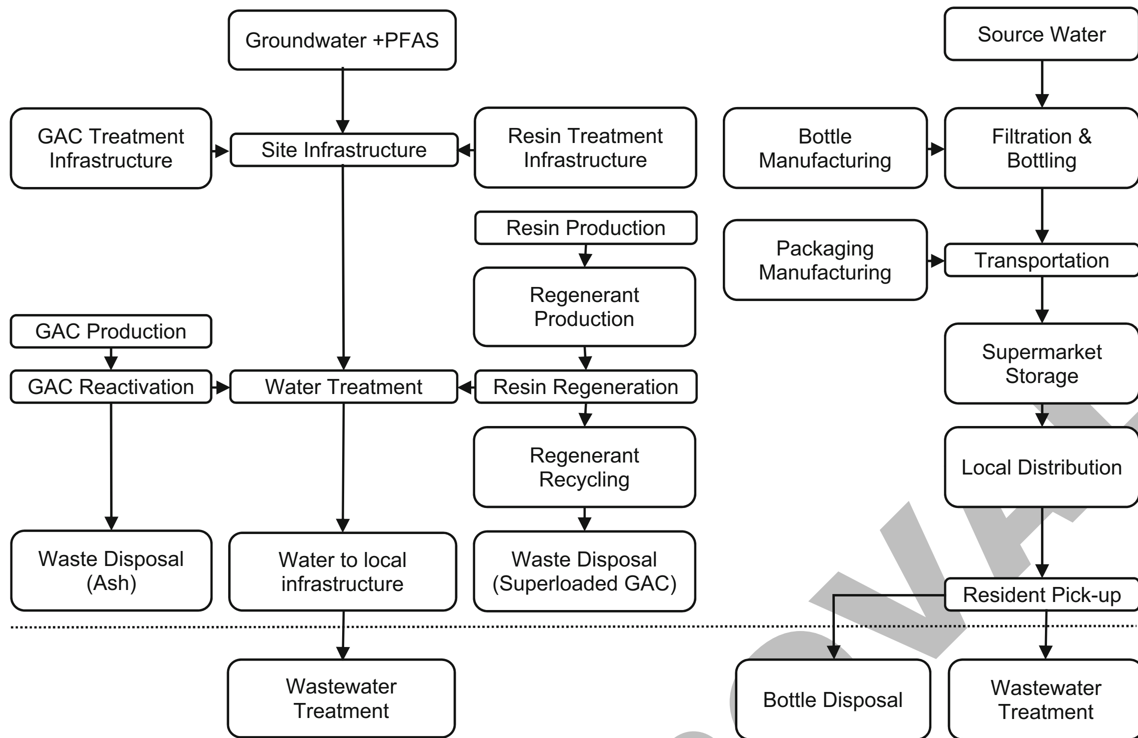

Fig. 1 System diagram of stages included in life cycle assessment of water supply scenarios. Stages below the dashed line are not included in the analysis

2.1 Site data and infrastructure

Site data and infrastructure are based on designs for a groundwater treatment system at Wright-Patterson Air Force Base. The planned facility will treat 3800 m^3 of groundwater per day from one contaminated well. Treated water enters a system supplying 1290 permanent residents and an average weekday population of 16,550. The treatment equipment will be housed in a 280-m^2 facility designed and built for this purpose. Site infrastructure for the treatment site is included based on architectural designs supplied by Legacy Building Solutions, Inc., and industry standards for construction materials. Infrastructure for the treatment system, including contactors, pipe, fittings, and corrosion-resistant coatings of internal surfaces, are included in our model (Table S1, Electronic Supplementary Material). GAC use rates and IEX column regeneration frequency were calculated for treatment of groundwater at 0.7, 7.0, and 70 μg/L (micrograms per liter) combined PFOA and PFOS concentrations, reflecting the range of contaminant concentrations at several USAF installations (Anderson et al. 2016).

2.2 GAC-based treatment system

Model data GAC treatment of PFAS-contaminated water is based in large part on a report of energy consumption and GAC use rates at a similar facility currently in operation (personal communication, Ryan Morrish, 11 November 2016). The model system pumps contaminated groundwater through two contactors in series each containing 9100 kg GAC. When breakthrough is detected in the primary contactor, the used GAC is removed and replaced. Facility energy requirements (including HVAC and office needs) have all been included in the model. Activated carbon production from bituminous coal was modeled primarily based on previous studies (Bayer and Finkel 2006; Jeswani et al. 2015) supplemented with energy use data from Calgon Carbon, Inc. (personal communication, Kendra Ryan, 7 December 2016) and waste ash production estimate from Isla-cabaraban et al. (2016). We model GAC use rates based upon an adsorption capacity of 0.11 g PFCs per kg GAC at anticipated system operating conditions (Amec Foster Wheeler 2017).

A key component of our treatment system is the reactivation of used GAC. Used GAC is transported by truck to a central reactivation facility where application of heat and steam degrades and removes contaminants, restoring most of the treatment potential of the carbon. Our model includes complete reactivation of used carbon. Ten percent of carbon mass are lost during reactivation, which is compensated by addition of fresh GAC on an equivalent mass basis. Carbon mass loss during thermal regeneration was reported by previous studies (e.g., Martin and Ng 1991). Water and energy use during reactivation are from the available literature (Hutchinson 1975; Isla-cabaraban et al. 2016; Jeswani et al. 2015) and local experts (personal communication, William Scoville). Transportation of carbon to and from the treatment site is based on distances from Wright-Patterson Air Force Base to a Calgon Carbon, Inc. facility (Huntington, WV). Impacts from on-site GAC handling (such as loading and unloading into the GAC tanks) are expected to be insignificant compared to long-distance GAC transport. Parameters used in life cycle modeling of GAC-based groundwater treatment are provided in Table S2, Electronic Supplementary Material.

2.3 IEX-based treatment system

A treatment system using ion exchange columns was modeled based on pilot-scale tests conducted at a DoD facility. The modeled system uses three IEX columns in series with a contact times of 2.5 min each and an adsorption capacity of 1.3 g combined PFOA and PFOS per kg resin (Amec Foster Wheeler 2017). Once saturated, the ion-exchange columns are regenerated by washing with solvent and brine solutions to remove PFAS and any other contaminants. Based on protocols developed at the pilot scale, we assume that washing with five bed volumes of regenerant, five bed volumes of brine, and one bed volume of water is sufficient to completely restore the column's adsorptive capacity (Amec Foster Wheeler 2017).

The large volumes of waste produced during resin regeneration are mitigated by recycling the regenerant solutions. Used regenerant is distilled to produce a "clean" methanol fraction and concentrated brine. Methanol demand in subsequent regeneration cycles is supplemented with 5% new methanol by volume. Contaminants in the concentrated brine are removed in a GAC superloader. Based on pilot-scale trial data, treated brine is expected to perform equivalently to fresh brine in IEX column regeneration. Waste GAC from the superloader is assumed to be incinerated as hazardous waste (Amec Foster Wheeler 2017).

Contactors and regenerant storage tank materials were estimated based on the necessary capacity of contactors to process well water at the desired flow rate and residence times. Piping and other infrastructure requirements were conservatively assumed to be double that of the GAC system due to the increased complexity of the resin treatment and regeneration system. Parameters used in life cycle modeling of IEX-based groundwater treatment are provided in Table S3, Electronic Supplementary Material.

2.4 Bottled water supply

After the release of the US EPA health advisory for PFCs in May of 2016, the USAF Mission Support Group made bottled water available to residents of affected areas at WPAFB. Data for brand 1 bottles in this analysis come primarily from a report on Nestle manufacturing and supply chain life cycle impacts, which included life cycle (upstream) energy and materials use through delivery and storage at a local supermarket (Quantis 2010). This was supplemented with distribution transportation distances from regional Nestle facilities (Greenwood, IN) to WPAFB (OH). Data for an alternative bottled water scenario, referred to as brand 2, were sourced from the literature, supplemented by Quantis (2010) where necessary. Bottle manufacturing and spring water sourcing and facility energy use were supplemented with distribution transportation distances from regional Crystal Geyser company facilities (Benton, TN) to WPAFB (OH). Bottles were purchased as packages of 28 0.5-L bottles, and we include the additional packaging (cardboard tray and low-density polypropylene wrap) in our analysis. Parameters used in life cycle modeling of bottled water are provided in Table S4, Electronic Supplementary Material (brand 1) and Table S5, Electronic Supplementary Material (brand 2).

We developed two scenarios for local bottled water distribution at WPAFB based on actual on-site procedures. Under both scenarios, bottled water is picked up from a local supermarket and delivered by gasoline-fueled light truck to distribution centers at WPAFB. These distribution centers include sites with potentially sensitive populations (such as a childcare center). This local distribution stage includes round-trip transportation between the distribution site and supermarket, with impacts distributed among the 1–4 m^3 of water transported per trip. The second distribution scenario includes pick-up from the distribution sites by residents. Driving distances were determined from several factors, including proximity of working and living facilities to the pick-up location (0.25–7.0 km), attributable share of the trip to water pick-up (10–100%), and volume of water acquired per trip (one to four cases, or 0.013–0.052 m^3). This resulted in a best estimate of 29 km/m^3 with a range of 0.48–538 km/m^3.

Based on the anticipated health effects of PFAS-contaminated groundwater, we assessed two levels of bottled water supply to WPAFB. Both are based on a substitution of the drinking water fraction of total water use, assumed to be 5 L per person per day (USEPA 2011). The provisioning of tap water to homes and workplaces is not included in these scenarios. The sensitive resident (SR) scenario includes only bottled water delivery to locations serving EPA-designated sensitive populations to the effects of PFOA and PFOS (pregnant and nursing mothers, infants, and young children), who are assumed to make up 5% of the population. The resident and sensitive non-resident (RSN) scenario includes bottled water delivery to locations serving sensitive populations, assumed to make up 5% of the non-resident population, as well as individual bottled water pick-up by the non-sensitive full-time resident population.

Post-consumer treatment of plastic bottles is not included due to a lack of reliable data on crucial parameters such as local recycling rates, waste transportation distances, and recycling process energy use and end products. The potential influence of recycling on bottled water supply as an alternative to treatment of contaminated groundwater was examined in a sensitivity analysis based on default Ecoinvent 3 recycling parameters for plastic and cardboard.

2.5 Life cycle impact modeling and Monte Carlo simulation

SimaPro software was used to implement the life cycle models (v.8.3; Pré Consultants, Amersfoort, the Netherlands). Database entries designed for North America, the USA, or the region containing the study area (such as North American Electric Reliability Corporation regions) were preferred when assembling the life cycle models. However, relatively few local database entries were available. Most entries were sourced from the Ecoinvent 3 database, using global or rest-of-world (non-European) values (Tables S2–S5, Electronic Supplementary Material). Impact calculations were performed using TRACI 2.1 v1.04 US 2008 indicator methods. TRACI is widely regarded as the most accurate midpoint impact assessment model for the US region, particularly for the categories of primary interest in this study (climate change and human health) (Hauschild et al. 2013).

Likelihood distributions were introduced for most model parameters. Where available, operations data from full-scale or pilot-scale facilities or industry reports were used to develop distributions based upon best estimates, maximum, and minimum values. For most parameters, a pedigree matrix approach was used to introduce uncertainty around a best estimate based upon the degree of confidence in the accuracy of the chosen value. The pedigree matrix implementation in SimaPro with Ecoinvent database version 3 adjusts parameter value according to a lognormal distribution parameterized based on a matrix of six data quality categories each ranked from 1 (best) to 5 (worst): data type, reliability, completeness, temporal correlation, geographical correlation, and technological correlation (Ciroth et al. 2012). Likelihood distribution parameters based on pedigree functions are shown in Tables S2–S5, Electronic Supplementary Material. Stochastic analysis was done with SimaPro using built-in Monte Carlo simulation capabilities with 5000 iterations per model.

To clarify differences between models with similar impacts, we ran additional simulations of the difference in impacts between the GAC and IEX water treatment systems at the baseline PFAS concentration of 0.7 μg/L and the maximum concentration of 70 μg/L, and between the two bottled water systems (brand 1 and brand 2). Unfortunately, because SimaPro can only conduct stochastic comparison modeling between systems on an equivalent mass basis, a meaningful comparison of groundwater treatment systems and bottled water provisioning according to our parameters for supply of bottled water (only for drinking) for sensitive individuals could not be assessed.

2.6 Aggregated human health impacts

Life cycle human health damage due to treatment operations and bottled water provisioning are calculated by SimaPro in comparative toxic units (CTUs), which indicate the expected incident rate of cancer (c) and non-cancer (nc) health effects. We convert CTUs to disability-adjusted life-years (DALY) using conversion factors developed by Huijbregts et al. (11.5 DALY per cancer CTU and 2.7 DALY per non-cancer CTU) (Huijbregts et al. 2005). Economic endpoints are presented by using the US EPA guidelines on value of a statistical life (VSL), \$10.7 million in 2017\$ (USEPA 2016d) and average US life expectancy of 78.8 years (USCDC 2016). The economic value of a DALY (VDALY) is estimated as \$136,000 based upon the ratio of VSL to life expectancy.

3 Results

3.1 Groundwater treatment

Life cycle impacts of groundwater treatment at the baseline concentration of 0.7 μg/L PFAS are dominated by electricity use at the treatment facility, which is responsible for 90 and 95% of GWP impacts in GAC- and IEX-based treatment systems, respectively. Remaining sources of GWP are GAC supply and reactivation and on-site infrastructure and equipment supplying (7 and 3%, respectively, in the GAC model) and on-site IEX infrastructure and equipment (4%). Other life cycle impacts are highly correlated with energy use, and so human health, ecotoxicity, smog formation, and other impacts are similar between the GAC and IEX treatment methods (Table 1). Ozone depletion, impacted by the production of ion-exchange resins, is higher in the IEX treatment scenarios.

At higher contaminant concentrations (7.0 and 70 μg/L PFAS), greenhouse gas impacts of GAC-based treatment increase substantially to 0.54 and 2.7 $kgCO_2eq/m^3$ H_2O, respectively. GAC production and reactivation leads GAC-based treatment impacts to outpace those from IEX-based treatment at higher contaminant concentrations (Table 1). At the highest contaminant concentrations, electricity consumption represents only 15% of climate change impacts from GAC-based treatment, but 63% of impacts from IEX-based treatment. Higher overall energy consumption throughout the GAC production and reactivation process leads to higher energy-related life cycle impacts across most categories, including fossil fuel depletion, respiratory effects, and smog formation (Table 1).

Human health effects appear less sensitive to changes in GAC use rate. At 70 μg/L PFAS, electricity production continues to dominate human health and ecotoxicity impacts of groundwater treatment with over 75 to 90% of health and ecotoxicity impacts from both GAC and IEX systems, with approximately 50% of life cycle health and ecotoxicity

Table 1 Deterministic model results for supply of 1 m^3 treated groundwater water or bottled water

	Scenario	PFOA + PFAS combined concentration (μg/L)						Local distribution		Resident pick-up	
		0.7		7.0		70					
		GAC	IEX	GAC	IEX	GAC	IEX	Brand 1	Brand 2	Brand 1	Brand 2
GWP	kgCO_2eq	0.33	0.32	0.54	0.33	2.7	0.50	139	241	144	246
Toxicity (cancer)	CTU(h,c)	2.8E−08	3.1E−08	2.9E−08	3.1E−08	3.9E−08	3.6E−08	6.6E−06	8.2E−06	6.7E−06	8.3E−06
Toxicity (non-cancer)	CTU(h,nc)	9.3E−08	9.6E−08	9.9E−08	9.7E−08	1.5E−07	1.1E−07	3.2E−05	4.4E−05	3.3E−05	4.5E−05
Ozone depletion	kg CFC-11	2.7E−08	1.7E−07	2.8E−08	1.7E−07	3.6E−08	1.7E−07	7.3E−06	8.0E−06	7.3E−06	8.0E−06
Smog	kg O_3 eq	8.2E−03	8.3E−03	0.011	8.6E−03	0.035	0.011	9.7	20	10	21
Acidification	kg SO_2 eq	1.2E−03	1.2E−03	1.7E−03	1.3E−03	5.7E−03	2.0E−03	0.72	1.4	0.74	1.4
Eutrophication	kg N eq	2.8E−03	2.8E−03	2.8E−03	2.8E−03	3.7E−03	3.0E−03	0.38	0.38	0.38	0.38
Respiratory effects	kg $PM_{2.5}$ eq	1.1E−03	1.1E−03	1.2E−03	1.1E−03	1.7E−03	1.4E−05	0.14	0.16	0.14	0.16
Ecotoxicity	CTUe	3.0	3.1	4.6	3.1	4.6	3.4	821	1089	836	1103
Fossil fuel depletion	MJ	0.16	0.16	0.28	0.18	1.5	0.38	345	629	356	640

impacts from coal-fired electric generation. Life cycle electricity consumption increases from 1.5 to 1.9 (GAC) and 1.5–1.6 (IEX) MJ/m^3 remediated water from the baseline (0.7 μg/L PFAS) and 70-μg/L PFAS scenarios.

Stochastic analyses generally mirror deterministic scenario results, with average climate change and human health impacts increasing substantially only at the highest contaminant level (70 μg/L PFAS; Figs. 2a and 3a). Mean and median values for climate change impacts of groundwater treatment trend higher than the deterministic model results, particularly at higher contaminant concentrations and for IEX-based treatment (up to 400–600% of the deterministic model). Stochastic assessment of the difference between GAC and IEX treatments at the baseline PFAS concentration supports deterministic results that impacts are similar between treatment methods. Across most impact categories, GAC results in higher impacts in roughly 50–60% of model runs (Fig. S1, Electronic Supplementary Material). At the highest contaminant level, stochastic assessment of the difference between treatments continues to show higher average climate change impacts of GAC treatment, though not at 95% confidence (Fig. S2B, Electronic Supplementary Material).

Average human health impacts are similar in stochastic and static models (± 20% of the deterministic model), except at 70 μg/L PFAS, where stochastic averages are 180–230% of the deterministic model for GAC and IEX, respectively. Though GAC treatment generates higher average impacts than IEX-based treatment, confidence intervals for each pair of treatment scenarios largely overlap. Uncertainty in both climate change and aggregated human health impacts for both systems cover an order of magnitude (1–13 kgCO_2eq (Fig. 2); $0.5–4.5 \times 10^{-6}$ DALY (Fig. 3)). Though a stochastic assessment of the difference between GAC and IEX treatments shows higher human health impacts of IEX in a majority of cases (Fig. S1, Electronic Supplementary Material), differences between the two treatment methods are near zero at both baseline PFAS concentration and 70 μg/L (Fig. S2D,E, Electronic Supplementary Material).

3.2 Bottled water

Production and delivery of bottled water generates life cycle impacts orders of magnitude larger than remediated groundwater on a common volume basis (Table 1). Differences between the two model data sets (brand 1 and brand 2) stem primarily from the type and quantity of energy used in PET bottle manufacturing. Production of brand 1 bottles generates 75 kgCO_2eq/m^3 bottled water, relative to 135 kgCO_2e/m^3 for brand 2 bottles. Life cycle GWP impacts for the brand 1 system are 18 kgCO_2eq for electricity and 9.6 kgCO_2eq for natural gas. In contrast, life cycle GWP impacts of brand 2 are 5.8 kgCO_2eq for electricity and 42 kgCO_2eq for natural gas. Regional transportation impacts from the bottling facility to a local supermarket also differ between models, contributing 29 kgCO_2eq for brand 1 and 80 kgCO_2eq for brand 2. Life cycle human health impacts are similar between both bottled water models (Table 1).

Local distribution and consumer pick-up of bottles were modeled identically for both models. Local distribution contributes 0.6 kgCO_2eq (0.6 and 0.4% of total GWP for brands 1 and 2, respectively), 0.2–0.4% of total human health impacts, and 0.2–0.3% of total ecotoxicity. Pick-up of bottled water by consumers from a local distribution site contributes 5.3 kgCO_2eq (4 and 2% of total GWP for brands 1 and 2, respectively), 1–2% of total human health impacts, and 1–2% of total ecotoxicity.

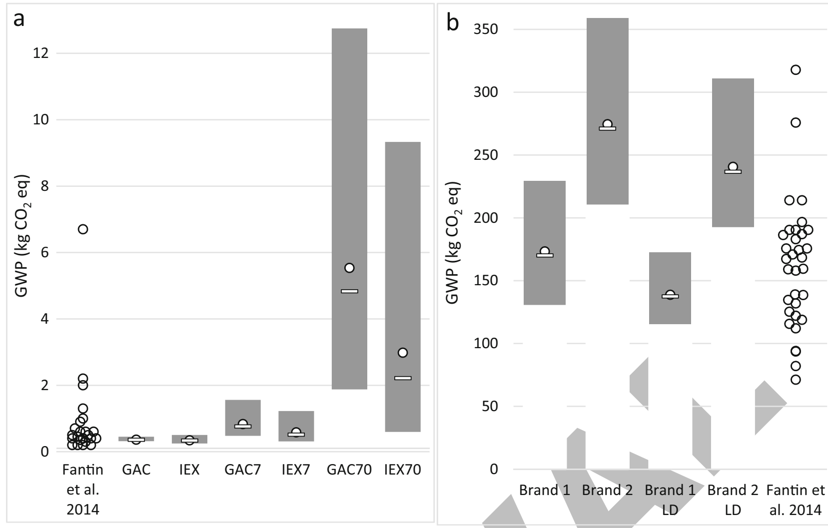

Fig. 2 **a** Greenhouse gas impacts of GAC and IEX remediation methods from baseline (0.7 μg/L), 7.0, and 70 μg/L combined PFOA and PFOS concentrations and **b** bottled water with resident pick-up or local delivery (LD). Bars indicate 95% confidence intervals (2.5 to 97.5 percentiles). Lines and circles indicate median and mean values, respectively. Studies (*n* = 24 (**a**), *n* = 33 (**b**)) reviewed by Fantin et al. (2014) are displayed in both figures for comparison purposes

Stochastic simulation of bottled water production and delivery provides expected values similar to deterministic results for climate change and human toxicity impact factors, with median climate change, human health, and ecotoxicity impacts within 20% of deterministic values. Ninety-five-percent confidence intervals for climate change impacts range from 110 to 170 and 190–310 kgCO_2eq with local distribution to 130–230 and 210–360 kgCO_2eq with local distribution and consumer pick-up (brands 1 and 2, respectively) (Fig. 2b). Life cycle human health impacts are similar across brands and delivery scenarios (Fig. 3b). Stochastic assessment of the difference between bottled water brands provide evidence for higher human health impacts in the brand 2 system, though not at 95% confidence (Fig. S2F, Electronic Supplementary Material).

3.3 Scenario results

Over the 20-year lifetime of a groundwater treatment system, life cycle climate change impacts may range from 6000 to 14,000 tCO_2eq at 0.7 μg/L PFAS, using GAC or IEX-based treatment methods. At higher contaminant concentrations, we estimate median lifetime system climate change impacts of 21,000 or 130,000 tCO_2eq (GAC) and 14,000 or 62,000 tCO_2eq (IEX) treating 7.0 or 70 μg/L PFAS, respectively (Fig. 4a). Supplying bottled drinking water as a supplement to PFAS-contaminated drinking water, in lieu of a treatment system, results in 270–420 tCO_2eq (brand 1) or 810–1100 tCO_2eq (brand 2) when bottled water is supplied only to the sensitive fraction of permanent residents on-site. When bottled water is supplied to all residents and also to the

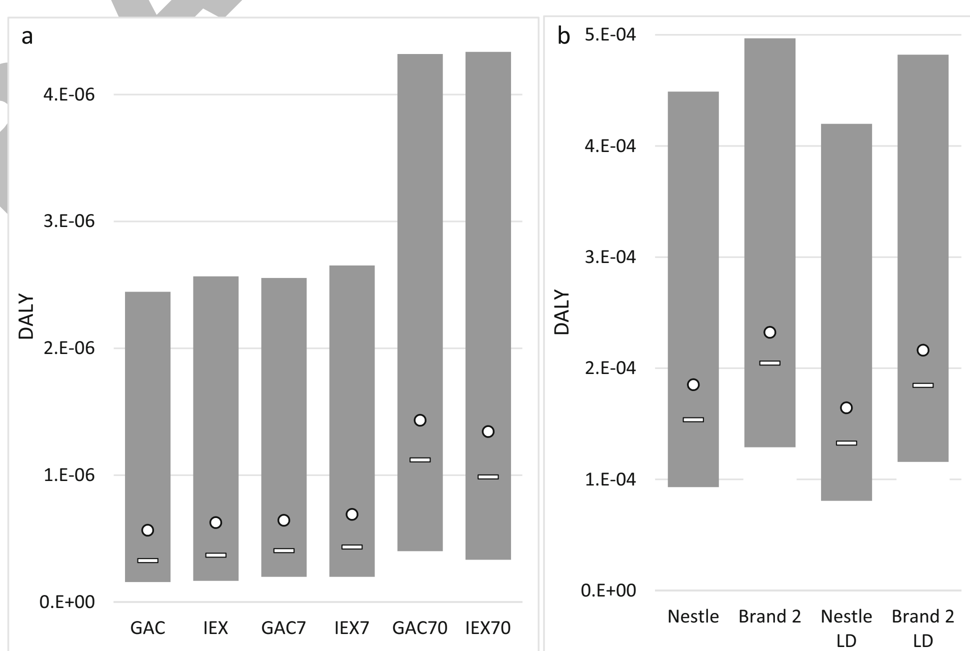

Fig. 3 **a** Human health impacts of clean water supply via remediated groundwater from baseline (0.7 μg/L), 7.0, and 70 μg/L contaminant concentrations and **b** bottled water with resident pick-up or local delivery (LD). Bars indicate 95% confidence intervals (2.5 to 97.5 percentiles). Lines and circles indicate median and mean values, respectively

sensitive fraction of non-residents, we estimate climate change impacts of 8100–14,000 tCO_2eq (brand 1) or 23,000–33,000 tCO_2eq (brand 2) over a 20-year period (Fig. 4a).

Model results for system lifetime impacts on human health indicate that, at low to moderate contaminant concentrations, a groundwater treatment system may result in 4–74 DALYs across the full system life cycle. These impacts may increase to 10–120 DALYs at higher contaminant concentrations due to increased cycling of the remediating material (GAC or ion exchange resin) (Fig. 4b). Economic valuation of these health impacts results in likely total costs of $1.2 M or $1.4 M for GAC and IEX, respectively, at 0.7 μg/L PFAS, up to $4.2 M or $3.7 M for GAC and IEX at 70 μg/L PFAS.

Supplemental drinking water scenarios may result in 0.2–1.2 DALYs over 20 years when bottled water is supplied only to the sensitive fraction of permanent residents on-site. When bottled water is supplied to all residents and also to the sensitive fraction of non-residents, life cycle human health impacts may rise to 6 to 33 DALYs. Economic impacts of these losses to human health may range from $42,000 (brand 1) or $69,000 (brand 2) in the sensitive resident scenario to $1.3 M (brand 1) or $2.0 M (brand 2) in the residents and sensitive non-resident scenario.

4 Discussion

GAC- and IEX-based groundwater treatment methods appear to perform similarly on a life cycle basis. The dominance of on-site energy use, which is used primarily for pumping water through the contactors and into the distribution system, helps explain the similarities in climate change impacts between our scenarios and a review of tap water life cycle assessments (Fig. 2a) (Fantin et al. 2014). Only when GAC use and IEX column regeneration rates increase at higher contaminant concentrations do impacts exceed the range of expected results for tap water provisioning.

Our results for climate change impacts of bottled water supply fall within the range of bottled water life cycle assessments harmonized by Fantin et al. (2014) (Fig. 2b). The similarity between deterministic and stochastic results for our brand 1 scenarios for local delivery and resident pick-up and the majority of studies reviewed by Fantin et al. (2014) supports the validity of the Quantis (2010) data set. Unfortunately, insufficient data on the human health or ecotoxicity impacts of bottled water production is available in published life cycle assessments to make substantive comparisons with the literature.

4.1 Role of energy use in groundwater treatment

The primary source of life cycle climate change, human health, acidification, and ecotoxicity impacts from both systems is electricity use at the treatment facility. In particular, electricity generation from coal represents 78 and 82% of total life cycle greenhouse gas emissions from GAC- and IEX-based treatment methods, respectively. At higher PFAS concentrations, the recycling of treatment components makes up a larger fraction of life cycle impacts and the contributions of coal-fired power plants drop to 49 and 13% (GAC) and 78 and 54% (IEX) for groundwater with 7 and 70 μg/L PFAS, respectively.

Electric generation offers an easy target for impact reduction. Substitution of low-impact electricity sources such as wind or solar could reduce the human health costs of groundwater treatment by 76 to 96% (Hertwich et al. 2014), and reduce global warming potential by 92 to 99% (Asdrubali et al. 2015; Hertwich et al. 2014). When treatment systems treat groundwater with higher concentrations of contaminants, the influence of GAC reactivation and IEX regenerant recycling increase, powered in large part by natural gas combustion for heat and power, and coal extraction (in the case of GAC). At 70 μg/L PFAS, GAC reactivation and IEX regenerant recycling produce 30 to 40 and 10 to 15% of human health and ecotoxicity impacts, respectively.

4.2 Logistical considerations

At higher contaminant concentrations at or above those modeled in this study, logistical considerations could outweigh environmental costs and benefits in system design decision-making. Under the current system design, at 700 μg/L, GAC may need to be replaced every 0.5–4 days, or IEX columns regenerated every 5 days. Even at 70 μg/L, GAC may need to be replaced every 1–3 weeks at the flow rate and configuration modeled in this study. Ion exchange resins could have the added benefit of greater adsorption capacity, and therefore slower column saturation cycles. On-site recycling of resin columns and regenerant solution could also provide benefits, reducing the logistical and financial burden of scheduling GAC replacement.

4.3 Comparing groundwater treatment and bottled water scenarios

On a functional unit basis, life cycle impacts of bottled water far surpass those of groundwater treatment. However, only 2% of the water supplied by the contaminated well are expected to be consumed as drinking water (the primary mode of exposure to PFAS considered by the EPA). The remaining water is used for a wide range of services, such as washing clothes, bathing, flushing toilets, watering plants, and aircraft maintenance. Though it is possible that some of these activities could result in additional exposure (wearing clothes washed with contaminated water, walking across a lawn watered with contaminated water, etc.), we expect the majority of impacts to human

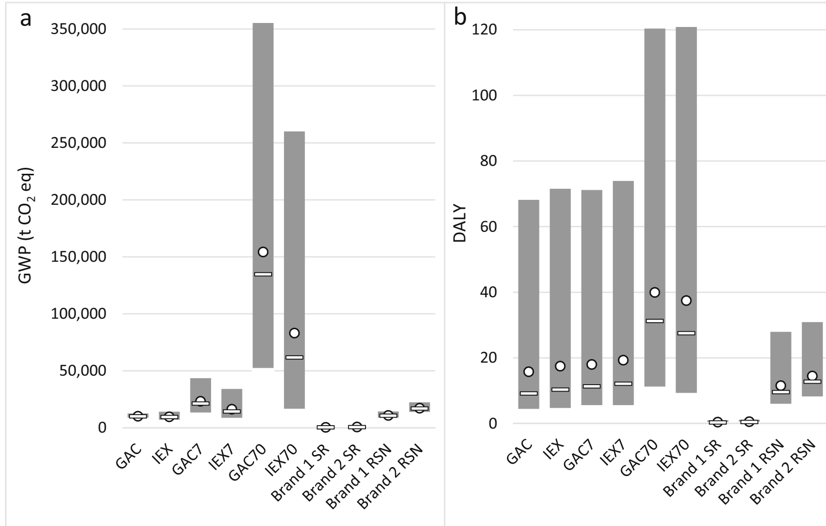

Fig. 4 Life cycle impacts of clean water supply over 20 years to **a** human health and **b** climate change by groundwater remediation (GAC, IEX) and bottled water (brand 1, brand 2). Bottled water scenarios include supply only to sensitive resident populations (SR) or to all residents and sensitive non-residents (RSN). Bars indicate 95% confidence intervals (2.5 to 97.5 percentiles). Lines and circles indicate median and mean values, respectively

health to occur via direct consumption. Supplying drinking water to the subsets of the total site population considered in our scenarios reduces the fraction of contaminated water replaced by bottled water to 0.008% (sensitive resident scenario) or 0.2% (residents and sensitive non-resident scenario). Providing these smaller quantities of bottled water to sensitive fractions of the site population could result in similar or reduced life cycle impacts compared to full treatment of groundwater at the well pump (Fig. 4).

Supplying bottled water to sensitive populations (infants and nursing and pregnant mothers) in lieu of implementing full-scale groundwater treatment can be examined from a risk analysis perspective on the basis of expected damage to the population exposed to contaminated groundwater. Our results show that average life cycle human health impacts are higher from GAC-based treatment than from supplying bottled water to the sensitive fraction of permanent residents at WPAFB. This accounting does not take into account the potential health impacts of consuming PFAS-contaminated water, which are not fully understood and thus difficult to quantify. In our sensitive resident scenario, implementing GAC-based groundwater treatment would reduce net health impacts if the expected incidence rate of health effects from PFAS-contaminated groundwater is higher than 2–3 cases per 1000 people. Though the actual incidence rate of PFAS-related health impacts is unknown, current evidence suggests that at the baseline concentration in this study (0.7 μg/L), incidence of adverse health effects is far below 3 cases per 1000 among the non-sensitive population (USEPA 2016a).

4.4 Sensitivity assessments

Alternate criteria for bottled water supply rates, or alternate demand for bottled water by the affected population, could lead to variability in the impacts associated with a bottled water supply program. Disaster preparedness documents cite a broad range of possible supply rates for clean water, depending on the population affected, environmental conditions, expected uses of the water, and the duration of the supply (USEPA 2011). A plausible high-end estimate of 15 L per person per day would roughly triple the life cycle impacts of bottled water supply. However, with the difference between scenarios and confidence intervals for results spanning orders of magnitude, this would have limited effect on the conclusions of this study. At 15 L per person per day, providing bottled water to sensitive residents would still have lower life cycle impacts than any groundwater treatment scenario, and impacts of bottled water supply to all residents and sensitive non-residents would still be within the 90% confidence intervals of impacts from groundwater treatment at higher contaminant concentrations.

We found that, at the rate of 5 L per person per day, supplying bottled drinking water to between 8 and 16% of the total population would have equivalent human health impacts compared to a treatment system treating 100% of water extracted from the ground. Similarly, climate change impacts of bottled water supply and groundwater treatment are equivalent when drinking water is supplied for 7–12% of the population (a total of 5.5–10 m^3 per day). As previously stated, the sensitive portion of the population is comprised of nursing mothers, pregnant females, and young children. This latter segment of the sensitive population is unlikely to consume 5 L, much less 15 L, of water per day.

Though we do not consider the disposal of water bottles in our analysis, a sensitivity analysis suggests that including curbside pick-up and disposal in a municipal landfill could increase the climate change impacts from bottled water by 22 $kgCO_2e/m^3$, or 9–16%. Recycling of some portion of waste bottles could mitigate that impact. Garfi et al. found that life cycle greenhouse gas emissions of bottled water, including the

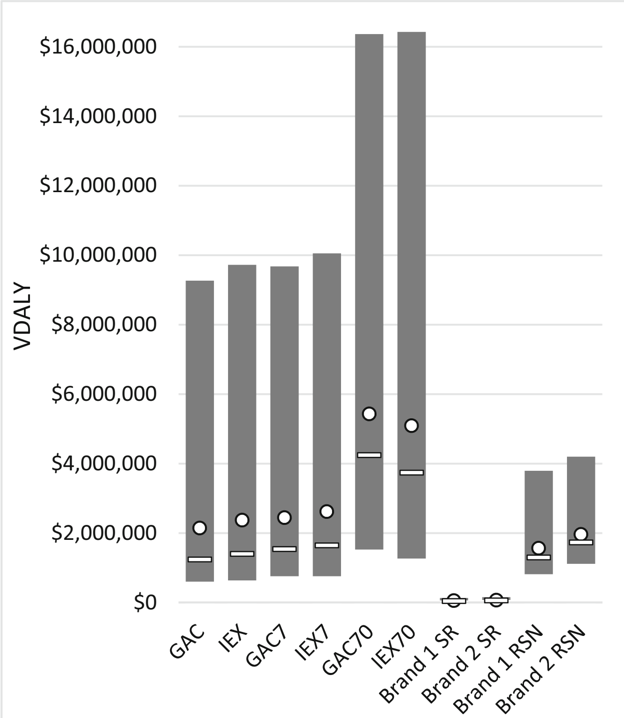

Fig. 5 Economic costs from life cycle human health impacts of clean water supply by groundwater remediation (GAC, IEX) and bottled water (brand 1, brand 2) over 20 years. Bottled water scenarios include supply only to sensitive resident populations (SR) or to all residents and sensitive non-residents (RSN). Bars indicate 95% confidence intervals (2.5 to 97.5 percentiles). Lines and circles indicate median and mean values, respectively

disposal phase, decreased from 79 to 71 $kgCO_2e/m^3$ when the modeled recycling rate increased from 25 to 75% (Garfí et al. 2016). Unfortunately, like many prior life cycle assessment studies of bottled water, Garfi et al. did not report outcomes for human health impacts. Our sensitivity analysis suggests that the disposal phase could contribute up to 1.6×10^{-4} $DALY/m^3$, potentially doubling the human health impacts of bottled water supply.

PFCs have been identified as potent greenhouse gases with substantial contributions to life cycle climate change impacts semiconductor manufacturing and aluminum production (Krishnan et al. 2008; USEPA 2017). However, we expect volatilization rates of PFOA and PFOS to be very low during treatment of contaminated groundwater. Even if 1% of total combined PFOA and PFOS were volatized during treatment, this would contribute only 0.3–1.4% of total climate change impacts of GAC- or IEX-based treatment, respectively.

4.5 Economic valuation

The economic burden of health-related externalities are similar to, or exceed, the costs associated with provision of potable water. Based on the actual costs of supplying bottled drinking water on an on-demand basis at WPAFB during the 6 months following the announcement of the EPA health advisory for PFOA and PFOS, the purchase cost of bottled water may total \$140,000 over 20 years. Costs associated with treatment of PFAS-impacted water are higher; over a 20-year system lifetime, not considering infrastructure costs, a groundwater treatment facility may pay \$0.35 M for GAC (at 0.7 μg/L combined PFOA + PFOS) and \$1.7 M for electricity. These costs are comparable to the economic burden of health-related externalities from the various scenarios explored in this work (Fig. 5). Over a 20-year system lifetime, externalized health costs range from a low of \$42,000 to \$59,000 (brand 1 and brand 2 bottled water, sensitive resident scenario) to a high of \$3.7 M to \$4.2 M (IEX and GAC treatment at 70 μg/L PFAS). The total cost of health externalities for other six scenarios in Fig. 5 were between \$1.2 M and \$1.7 M. The life cycle analysis demonstrates the significant cost contribution from health-related externalities over the expected duration of the remedial decision.

5 Conclusions

This study examined the life cycle impacts to climate and human health from treatment of perfluorinated alkyl substances using case studies based on approaches developed for the US Air Force. We found that, at concentrations ten times higher than the EPA health advisory for PFOA and PFOS, the climate impacts of treatment using GAC and IEX are similar to those for conventional tap water supply and human health impacts are negligible. Impacts of both treatment technologies were highly dependent on energy use at the treatment site, making renewable energy sources a key target for reducing climate, human health, and ecotoxicity impacts.

The ranges of likely impacts from GAC- and IEX-based treatment are similar across most scenarios. At PFOA and PFOS concentrations above 70 μg/L, 1000 times the US EPA health advisory, reactivation of carbon and recycling of ion-exchange regenerant become primary impacts, resulting in higher climate and human health damages.

Supplying bottled drinking water to sensitive populations could have lower life cycle impacts than full-scale groundwater treatment. When contaminant concentrations are low enough to pose a negligible risk to the general population, supplementing tap water with bottled drinking water for up to 5% of the most sensitive individuals within a population could be a low-impact alternative both environmentally and economically. Better understanding of the health impacts of PFAS-contaminated groundwater is necessary to make informed decisions on the risk management of water contamination and treatment. The knowledge from this study should help managers consider trade-offs between toxicity risks of PFAS-contaminated water and the LCA impacts of cleaning it. Although this study used WPAFB contaminated site as a case study, the LCA presented here can apply to any study comparing treatment options for water impacted by emerging contaminants.

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