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RESEARCH ARTICLE

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A sustainability assessment of an in situ ultrasonic reactor for remediation of PFAS-contaminated groundwater

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Abstract

The environmental remediation industry has increasingly considered the sustainability impacts of remediation systems. The ideal remediation system has a greater environmental benefit than detriment. While sustainable systems are increasingly available for many contaminants, per- and polyfluoroalkyl substances have presented new challenges for remediation practitioners seeking effective and sustainable systems. Here the sustainability and cost impacts of an ultrasonic reactor installed in a horizontal well (referred to as the In situ reactor technology [inSRT] system) at a hypothetical site are quantified and compared to hypothetical pump-and-treat (PT) system impacts. Direct costs of emissions from remediation were compared using levelized cost of energy data. The indirect impacts from pollution were quantified based on literature values. The results showed that the InSRT system had lower lifecycle impacts and costs compared to the PT system when each was considered under individually optimal conditions. InSRT was found to meet sustainability goals when used in a low hydraulic conductivity source zone and the PT system lifecycle impacts were reduced when the system was used in a high-hydraulic conductivity area.

1 | INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are a group of over 3,000 fluorinated organic compounds shown to be harmful to human health and resistant to conventional water treatment methods. Numerous remediation methods have been evaluated for PFAS but, due to the strength of the carbon–fluorine bonds, many conventional remediation methods have not been viable for PFAS remediation (Horst et al., 2019). The present paper addresses a novel destructive treatment concept using theoretical site models followed by a sustainability evaluation of the technology. Remediation methods that are effective for PFAS degradation can be adapted for sustainable implementation, as discussed below. It is first necessary to define sustainability, including in the context of environmental remediation.

1.1 | Sustainable remediation

Sustainability is frequently used to describe the intersection of socially useful, environmentally appropriate, and economically feasible approaches or concepts (Brundtland, 1987; Costanza & Patten, 1995; Goldenman et al., 2019; Vos, 2007). Sustainable remediation has become a widely held concept in the remediation industry where systems have increasingly been designed to meet remediation goals while minimizing impacts on the environment.

To quantify and assess the impacts of remediation (and other) systems a number of tools have been developed (Battelle & NAVFAC, 2013; Beames et al., 2014). For example, SiteWise™ is a quantitative spreadsheet-based tool that calculates the environmental impacts from inputs related to numerous parts of a system lifecycle (Battelle & NAVFAC, 2013). Several studies have used SiteWise with or without other tools to fully assess remediation alternatives available (Reddy & Chirakkara, 2013; Søndergaard et al., 2018).

1.2 | Review of active and passive remedial options

Active technologies are those that do require energy input during the operating phase of the lifecycle. Passive technologies are those

that require no additional energy inputs other than during installation (Bayer & Finkel, 2006). A brief review of active and passive remediation methods is included below.

Pump-and-treat (PT) systems refer to pumping water into and through an ex situ water treatment system using one or more unit treatment methods. Regardless of the method used, PT systems are considered active because removal of water, via pumps, from the aquifer or water body precedes treatment. If any remediation system has a component for the operation that is considered active, the system overall will be considered active. Most current remediation of PFAS-contaminated groundwater has occurred via PT systems using sorbents (Interstate Technology & Regulatory Council [ITRC], 2020). Typically, PT for PFAS has been either ion exchange resin or activated carbon, which have been implemented in full-scale treatment systems (Appleman et al., 2014; Ross et al., 2018).

In addition to sorptive technologies, PT can include advanced oxidation processes, reduction processes, and others. Emerging technologies including electrochemistry, plasma, sonolysis, and others have been evaluated for ex situ PFAS treatment with varying success and energy demand for operations (Schaefer et al., 2019; Stratton et al., 2017; Vecitis et al., 2008).

Permeable reactive barriers (PRBs) involve an in situ treatment zone situated perpendicular to groundwater flow (Bayer & Finkel, 2006). The technology is considered passive and meets many sustainability goals due to low operation and maintenance impacts compared to many active ex situ technologies. In conventional remediation practice, PRBs use a variety of reactive media, including, notably, zero-valent iron (ZVI) within the barrier, to degrade or destroy the contaminants. A literature search indicates PRBs have not been applied at scale for PFAS remediation. This is most likely because laboratory and ex situ studies of media effectiveness are ongoing. For example, one study found ZVI to be effective at 350°C and under pressure for perfluorooctanesulfonate [PFOS] (2% or less remaining after 6 h) in the laboratory (Hori et al., 2006). While these results were promising, heating groundwater and ZVI within the PRB would be technically challenging and, because the increased temperature (energy) would be needed, not considered a passive method. The results indicate the media options should be further developed to make PRBs effective for PFAS remediation.

Horizontal wells for groundwater remediation have been used for several years. Horizontal wells offer the advantage of passive water capture (not requiring pumps or other equipment), making them a potentially sustainable option (Divine et al., 2013; Plummer et al., 1997; Steward & Jin, 2001). Water is passively captured because the well provides a channel through the aquifer when the hydraulic conductivity is greater than in the aquifer (Horst et al., 2019). If hydraulic conductivity in the well is less than in the aquifer, the horizontal well will not capture water. Capture is enhanced when the difference between well and aquifer hydraulic conductivity is greater. The optimal conditions for horizontal well capture are in contrast to those for a vertical extraction well where lower aquifer hydraulic conductivity limits the pumping rate. One recent adaptation is the Horizontal Reactive Media Well (HRX Well®) which uses reactive or sorptive media to treat contaminants in situ (Divine, Wright, et al., 2018). The HRX Well is the first implementation to specifically be packed with reactive media and has the potential to house other media options. In addition, media implementation and removal in the HRX Well is simplified compared to PRBs (Divine, Wright, et al., 2018). Horizontal wells in general offer an opportunity to implement other technologies in situ, including sonolysis.

1.3 | Sonolysis

Sonolysis is a term describing the collapse of ultrasonically induced cavitation which occurs when sound waves are applied to a liquid. When the sound waves pass through the liquid, changes in pressure create microscopic cavities, collapsing when a critical radius is reached. The collapse generates local temperatures up to 5,000 K (Niemczewski, 2007; Suslick et al., 1999). Sonolysis has been used to treat PFAS-spiked samples, aqueous film-forming foams formulations, and landfill groundwater under varied conditions at scales ranging from less than 1–91 L (Gole et al., 2018; Moriwaki et al., 2005; Rodriguez-Freire et al., 2016; Vecitis et al., 2008). Each study reported varying removal of PFAS, dependent upon initial concentration, frequency, and other co-contaminants. PFAS chain-length also plays a role in degradation outcomes when using ultrasound. Partitioning favors long-chain compounds resulting in competition for the cavity interface. Similar partitioning behavior has been observed for granular activated carbon (GAC) and other media and should be accounted for in sonolysis system designs (Campbell et al., 2009). Vecitis et al. (2008) proposed that the PFAS molecules orient at the cavity with the fluorinated tail toward the cavity interior and the head group toward the bulk fluid outside the cavity. When multiple chain lengths are present the longer chain compounds will have a greater affinity for the interface. At higher frequencies (500 kHz and above), cavities decrease in size but increase in number (Suslick et al., 1999). The higher frequencies may be used to mitigate the chain-length dependence issue by providing sufficient total surface area in the system such that more PFAS may be treated in a given time period. If the PFAS makeup at a given remediation site is known, the frequency could be adjusted to optimize treatment for the PFAS present. Though there are several iterations of ultrasonic technology described in the literature subsurface operation to treat PFAS-contaminated groundwater has not been validated in the field.

1.4 | Levelized cost of energy

Levelized cost of energy (LCOE) compares the costs to establish energy infrastructure (e.g., solar and wind) to the electricity produced by each system (Pawel, 2014). The lower the LCOE value, when comparing these systems, the more favorable it is because of lower equalized cost per kilowatt hour of electricity produced. The concept of LCOE can also be applied to remediation alternatives to describe the direct costs of electricity consumed and CO_2 emissions produced. A similar metric was described by Turchi et al. (1992) where treatment costs per 1,000 gallons treated were compared

between systems. Their approach is valid for systems operating on comparable time and volume scale, for example, comparing two or more ex situ technologies operating at high-flow rates, relative to ambient groundwater flow. Comparison of a low flow rate in situ system to the high-flow rate ex situ scenario does not sufficiently consider the distinct operational differences, in addition to their intended applications. For example, systems used for direct drinking water treatment versus mass flux control. A method is needed to compare technologies on dissimilar time or volume scales on the basis of the ability to achieve remediation goals and risk reduction.

The work presented herein considers the potential sustainability impacts of in situ remediation using the In situ reactor technology (inSRT) system against a PT system. to be considered a sustainable option, the technology benefits must outweigh the costs. Further, when comparing InSRT and PT the method with the lowest lifecycle emissions should be selected. An optimal (low cost, practical, minimal emissions) scenario was selected for InSRT and for PT but the respective scenarios did not necessarily represent the same hypothetical site. InSRT operation in a low to medium aquifer hydraulic conductivity setting is preferred while PT is best under higher aquifer hydraulic conductivity conditions. To have a fair comparison of the technologies, each was theoretically applied under their optimum aquifer hydraulic conductivity ranges. The emissions profile and direct and indirect costs were determined for each and the results were used to explore if and when InSRT should be applied based on the cost of pollutant emissions versus remediation benefits.

2 | METHODS

2.1 | General approach

A horizontal well with sonolytic reactors and a PT system with GAC were compared in the context of PFAS remediation. The former is anticipated to be applied for source zone treatment while the latter may be applied to address the greater extent of the plume. The application of the technologies is described in Figure 1.

The technologies were first compared on the basis of energy demand and resulting emissions. A general system boundary was established for the lifecycle analysis and direct and indirect pollutant emissions impacts were categorized and costs determined. Direct impacts are those resulting from the operation of the remediation system, and indirect impacts are impacts from activities peripheral to system operation. For example, a direct cost is the electricity consumed by the remediation system. An example of an indirect cost is the cost of pollutant emissions from electricity generation. Literature values for the indirect cost of carbon emissions were used to estimate indirect costs of remediation and these were compared to the direct costs.

2.2 | InSRT reactor scenarios

InSRT is an ultrasonic reactor operating within a horizontal well and has been conceptualized to work best in a source zone as a plume cutoff method. InSRT targets a smaller area with higher contaminant concentrations, as compared to the rest of the contaminant plume, thereby controlling PFAS mass flux. The horizontal well operates passively with the well-oriented parallel to the flow path and flow depends on ambient groundwater velocity which is slow relative to those in active ex situ systems. Several of the analyses presented in this paper were based on the rate constants in Table 1. The values were obtained from a previous study in which PFAS-contaminated site groundwater was treated using InSRT (Laramay, 2020).

Plume dimensions were determined to be 15-ft thick 350-ft wide, and 1,000 ft in length. A series of preliminary scenarios were established from Table 1. First, the hydraulic retention time was

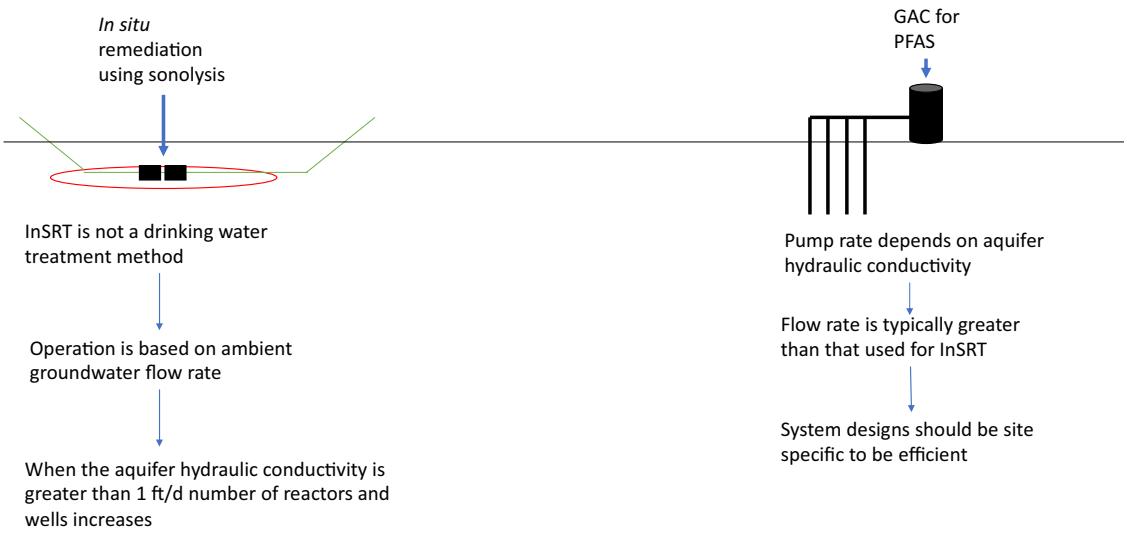

FIGURE 1 General schematic of in situ remediation (left) versus ex situ pump-and-treat (right) installations illustrating the difference in the application of each technology. GAC, granular activated carbon; InSRT, In situ reactor technology; PFAS, per- and polyfluoroalkyl substances [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 1 Rate constants from a PFAS contaminated site groundwater treated in the reactor

PFAS	S1 (h^{-1})	S2 (h^{-1})	S3 (h^{-1})	S4 (h^{-1})
4:2 FTS	0.001		0.06	0.02
6:2 FTS	0.13		0.14	−0.01
8:2 FTS	0.002		0.07	0.06
FOSA-1		0.09	0.18	0.01
PFBA	0.01	−0.01	0.03	−0.02
PFBS	0.06	−0.06	0.08	0.03
PFPeA	0.05	0.04	0.21	0.10
PFPeS			0.12	0.04
PFHxA	0.22	0.11	0.29	0.15
PFHxS	0.17	0.08	0.15	0.06
PFHpA			0.18	0.06
PFHpS	0.07		0.12	0.05
PFOA	0.22	0.09	0.21	0.11
PFOS	0.15	0.13	0.13	0.07

Abbreviations: 4:2 FTS, 4:2 fluorotelomer sulfonate; 6:2 FTS, 6:2 fluorotelomer sulfonate; 8:2 FTS, 8:2 fluorotelomer sulfonate; FOSA, perfluorooctane sulfonamide; PFBA, perfluorobutanoic acid; PFBS, perfluorobutanesulfonate; PFPeA, perfluoropentanoic acid; PFPeS, perfluoropentansulfonate; PFHxA, perfluorohexanoic acid; PFHxS, perfluorohexanesulfonate; PFHpA, perfluoroheptanoic acid; PFHpS, perfluoroheptanesulfonate; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonate.

determined for a remediation goal of 90% reduction of each known PFAS concentration. Each InSRT reactor has an active area 1.5-ft long and 0.55-ft wide. The length was used to calculate the velocity in 1, 2, 3, 4, or 5 reactors needed to achieve the desired retention time and the area was used to calculate flow rate (Q). As the number of reactors increases, the allowed velocity can also increase. Then, Equation (1) can be used to calculate capture width (Divine, Wright, et al., 2018):

$$W = \frac{Q}{K_A b_A i_A}, \tag{1}$$

where W is capture width, K_A is aquifer hydraulic conductivity, b_A is aquifer thickness, and i_A is aquifer hydraulic gradient. According to Equation (1) as Q increases capture width also increases. Additionally, as K_A, b_A, and i_A decrease W will also increase. K_A will have a greater influence on W than b_A or i_A, and according to Horst et al. (2019), under low permeability conditions (1 ft/day or less) certain technologies (e.g., an HRX Well) are particularly viable. To evaluate InSRT sustainability, a range of hydraulic conductivity values was used (ft/day): 0.02, 0.1, 2, 20, 50, and 100. The capture width for each K_A value and 1–5 reactors was calculated. The number of wells needed can be determined from the target treatment area where the target width is divided by W. As capture width increases the number of wells needed decreases. In this study, fractions of the total PFAS plume were considered (e.g., 25% and 5%) to determine where the InSRT reactor could be implemented for a reasonable cost. An optimal scenario was selected for additional analysis. Optimal was defined as the smallest number of wells and reactors required to treat the compound which had the longest required retention time.

2.3 | PT scenarios

Several PT scenarios were initially established to determine the ideal site conditions for implementation. The same series of K_A values described above were used in the equation given below (U.S. EPA, 2008):

$$Q = K_A \times (b \times w) \times i, \tag{2}$$

where Q is the flow rate, K_A is aquifer hydraulic conductivity, b is aquifer thickness, w is plume width, and i is aquifer hydraulic gradient. Then, using Equation (3), the radius of influence was calculated for each well according to the calculated flow rate.

$$Q = (2 \times \pi \times b \times r) \times K_A \times \frac{dh}{dr}. \tag{3}$$

A ratio was estimated for the change in depth to change in radius of influence (dh and dr, respectively) based on the progressively greater radius of influence expected from increasing hydraulic conductivity values. The plume width and length determined the area where pumps should be installed.

The mass of GAC in the PT scenarios was 9,000 kg based on a cost report for the Former Pease Air Force Base (Weston & Sampson, 2017). GAC capacity was determined from literature data, including Woodard et al. (2017) and Siriwardena et al. (2019). The literature values were used with concentration data in Table 1 to determine loading in the equation given below:

$$q = C_i \times Q, \tag{4}$$

where q is loading (mg/day), C_i is initial PFAS concentration (mg/L), and Q is the flow rate (L/day). q Was then used to calculate the number of changeouts per year in the following equation:

$$y = \frac{\left(\frac{c \times M}{q}\right)}{365 \text{ days/year}}, \tag{5}$$

where y is changeout frequency (years), c is capacity (mg/kg), and M is the mass of GAC per changeout (kg). The number of changeouts depended on the lifecycle duration considered, between 1 and 30 years.

2.4 | Analysis of InSRT and PT using SiteWise

An alternatives analysis was completed to understand how lifecycle phase impacts varied between InSRT and PT using the optimized

scenarios for InSRT and PT established using the methods described above. A general diagram of system boundaries for the LCA is provided in Figure 2, including the location of direct and indirect impacts relative to the system boundary. The definitions of direct and indirect impacts are described in Sections 1 and 2.1.

Each of the remediation alternatives divided into four lifecycle phases (Phase 1: travel, Phase 2: materials manufacture, Phase 3: installation, and Phase 4: operation) as shown in Table 2. A specific lifecycle duration was not assigned to either system because monitoring data would be used to determine when remediation goals were met. Instead, energy and materials consumption and resulting pollutant emissions were determined for the optimal scenarios for 1, 5, 10, 20, and 30 years to demonstrate the change in impacts over time.

A similar approach was taken in other studies (Favara et al., 2011; Sparrevik et al., 2011). In this study, an end-of-life phase for each system was not considered because preliminary research had shown

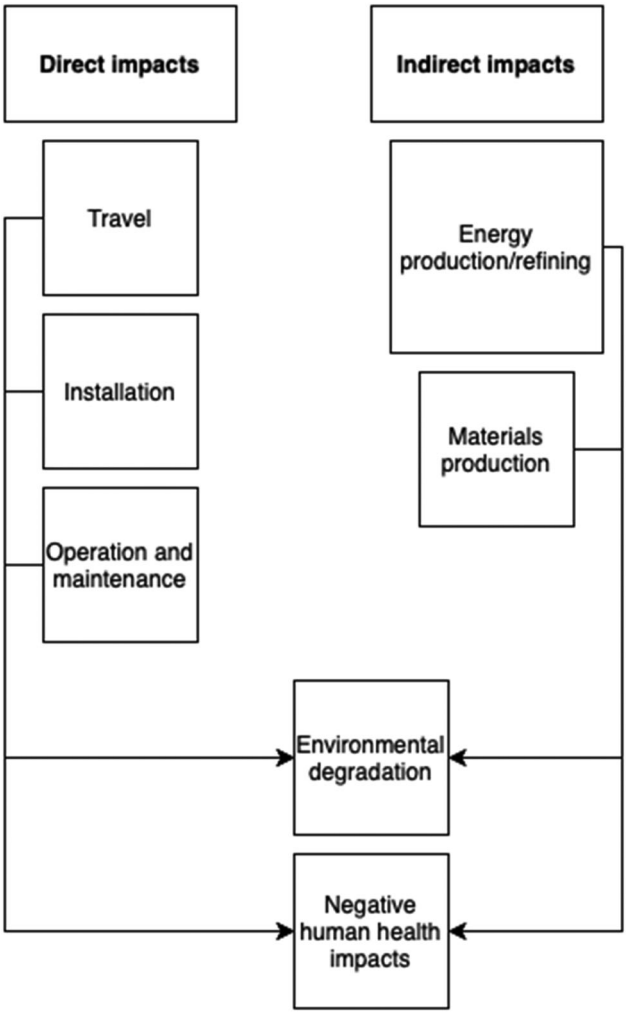

FIGURE 2 Travel, operation, maintenance, and installation activities were referred to as direct impacts. All energy production (e.g., fossil fuels capture and processing) and materials production impacts were considered indirect with regard to on-site activities

TABLE 2 Lifecycle phases used in the remediation alternatives analysis

Phase	Description
Phase 1	Personnel and materials transport. Only finished materials transport to the worksite was included
Phase 2	Materials manufacturing impacts
Phase 3	Technology installation includes fuel impacts
Phase 4	Lifecycle operation includes recurring impacts (e.g., equipment operation, granular activated carbon replacements)

individual GAC incineration events and well-end-of-life impacts were minimal. Incineration to destroy PFAS occurs at between 800°C and 1100°C, for approximately one ton of GAC at a time, depending on the incinerator capacity (Minimata Mercury Convention, 2013). The typical residence time was reported as less than 1 min (Yang et al., 2000). Incineration is often considered to have significant emissions impacts but, per batch of GAC incinerated, no clear evidence was found that the impacts were significant. Regenerated GAC was also not considered as a lifecycle endpoint because of dissimilar temperatures for GAC regeneration versus PFAS destruction (Horst et al., 2020). The differences in temperature (and the residence time at each temperature) and corresponding environmental impacts require additional research, particularly in a PFAS context.

SiteWise quantifies the impacts from the production of a product (materials, electricity), as greenhouse gas emissions (as CO_2 equivalents [CO_2 e]), energy used (MMBTu), electricity used (MWh), nitrous oxide (NO_X), sulfur dioxide (SO_2), and particulate emissions (10 μm or smaller in diameter [PM_{10}]) for each remedial alternative considered. The input values for individual phases were entered into SiteWise and the summation of lifecycle impacts from each phase was used.

2.5 | Direct cost calculation method

As presented in Section 1, LCOE represents the cost to produce a kilowatt hour of electricity, per Equation (6). The costs included can vary with data availability; in this study capital and operational expenses related to electricity use were included.

$$\text{LCOE} = \frac{\text{Expenses}(\$)}{\text{kWh produced}}. \quad (6)$$

Equation (6) gives the ratio of expenses in U.S. dollars to the kilowatt hour produced. However, electricity production is a cost–benefit analysis whereas CO_2 emissions resulting from electricity generation, are detriments. In this study, the cost of CO_2 was calculated for InSRT and PT, rather than LCOE as in Equation (6). To produce the electricity used, some mass of CO_2 was emitted (estimated using SiteWise), described as metric tons of CO_2 per kilowatt hour. Therefore, the dollars spent on electricity consumed were also

spent to produce the mass of CO_2. By implementing each system under optimal site conditions, appropriate estimates of costs, electricity consumption, and CO_2 emissions can be made. Equation (6) can be rewritten as the ratio of the sum of capital and lifecycle electricity costs to the CO_2 emissions produced during electricity generation, as in the equation given below:

$$\text{Cost of } CO_2 = \frac{\text{Expenses}(\$)}{\text{Mass of } CO_2}, \quad (7)$$

where expenses are capital costs and lifecycle electricity costs and mass of CO_2 is the mass produced when electricity is generated. The results can be used to determine which system has higher direct environmental costs, and to compare direct and indirect costs from remediation to the cost of *not* remediating. After the capital costs were calculated, a 3% discount rate was used to calculate the present value at intervals over 30 years. The discount rate was determined from the literature where values ranged from 1% to 7% and 3% was often selected as an intermediate value (Moore et al., 2004; Moore & Vining, 2018; U.S. EPA, 2010). Table 3 provides the assumption details used to calculate the cost of direct CO_2 emissions for different scenarios.

While several documents were used to inform site design and the assumptions used to calculate the cost of direct CO_2 emissions, one report was used to estimate costs. A cost report for either renovating or replacing an existing water treatment plant and adding GAC beds at the Former Pease Air Force Base is publicly available (Weston & Sampson, 2017). The cost of new pumps and capital costs overall included in the report were used to estimate the PT capital costs. These estimates were used rather than attempting to produce an exact bill of materials for the hypothetical case.

PFAS remediation is intended to prevent further harm to human health, but there are costs associated with pollution. These costs are considered indirect in this study (as indicated by exclusion outside the system boundary) and result in an additional cost per mass of CO_2 for InSRT and PT. The net cost of remediation is then comprised of the difference between the sum of capital, electricity (operations), social and environmental damages, and the social and environmental benefits of remediation. Literature references were used to determine the indirect cost of emissions. Samadi (2017) examined electricity production at the plant level (where the electricity was produced), the system level (how the electricity was distributed to consumers), and the external level (indirect impacts). Using data from the United States and Europe, the authors determined the average cost to society (which they refer to as social cost) of indirect CO_2 emissions was (in 2015 Euros) 37 euros/ton CO_2 emitted at a 3% social discount rate. The social cost was defined as the sum of private (costs included in the cost of electricity by the producer) and external (costs not included in the electricity price). The external costs are those referred to in this paper as indirect costs, for example, the cost of illness arising from increased PM_{10} exposure. The social cost data were used to estimate the social cost of CO_2 emissions from electricity generation. Assuming that the system meets remediation goals, and if indirect and direct costs were less than avoided mortalities, benefits of remediation could be argued to outweigh the negatives (i.e., pollution and its effects).

3 | RESULTS

3.1 | Sustainability analysis of two groundwater remediation alternatives

Previous site groundwater data were used as the basis to evaluate InSRT's sustainability. The calculated flow rates were used to determine capture width when 1–5 reactors were used. These values were then used to determine the number of wells to address the entire plume (Table 4).

As hydraulic conductivity increased, the number of horizontal wells needed to be increased; and when K_A was greater than 0.02 ft/day, the number of wells was too high to be practical. To have sufficient capture width by horizontal wells, there must be sufficient difference between the aquifer and well hydraulic conductivity values. As the aquifer and well hydraulic conductivity values increase toward each other the less influence the well value has on capture width, which can also be seen in Equation (1). When the contrast between the two values decreases, the capture width also begins to decrease. The number of wells increases when well flow rate and capture width decrease as a product of increased aquifer hydraulic conductivity and or increased hydraulic retention time. Costs per well beyond 2 or 3 wells were outside suggested reasonable cost ranges (Divine, Wright, et al., 2018) and

TABLE 3 Assumptions and data sources used to calculate the cost of direct CO_2 emissions

Input type	Assumptions and references
MWh for pumping per day	Estimated MWh from SiteWise™
Approximate PT capital cost	Pease cost report (U.S. EPA, 2001; Weston & Sampson 2017)
Annual electricity cost	$0.12/kWh
Cost of reactors	Assumes $30,000 cost per reactor
Annual kWh	Used 60% of 2.4 kW rating, multiplied by 2 reactors (2.8 kW per reactor pair), each reactor operates 12 h/day

Abbreviations: kWh, kilowatt hours; MWh, megawatt hours; PT, pump and treat.

TABLE 4 Number of wells required to remediate FOSA, PFBA, PFOA, and PFOS using 1–5 reactors

PFAS	K_A (ft/day)	1 Reactor	2 Reactors	3 Reactors	4 Reactors	5 Reactors
PFBA	0.02	8	4	2	2	2
	2	814	407	271	204	163
	20[a]	8140	4070	2713	2035	1628
FOSA	0.02	1	1	1	1	1
	2	135	68	45	34	27
	20	1356	678	452	339	271
PFOA	0.02	1	1	1	1	1
	2	116	58	39	29	23
	20	1163	581	387	291	233
PFOS	0.02	2	1	1	1	1
	2	185	93	62	46	37
	20	1850	925	617	463	370

Note: Successful remediation was defined as decreasing the respective concentrations 90% from the initial value. From left to right number of wells based on K_A (ft/day): 0.02, 2, and 20.

Abbreviations: FOSA, perfluorooctane sulfonamide; PFBA, perfluorobutanoic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonate.

[a]In all cases, 20 ft/day is the maximum aquifer hydraulic conductivity for horizontal well installations and demonstrates the impractical number of wells required when using InSRT.

the lifecycle emissions impacts may exceed the benefits of remediation. The number of wells required decreased with an increasing number of reactors but the impacts of five reactors per well would quickly become substantial. It can be noted here that including PFBA in the remediation goals causes an eightfold increase in the number of wells required. In situ pretreatment methods are being explored to address inefficiencies related to the degradation of PFBA and some other PFAS but are outside the scope of this paper. By increasing reactor efficiency (e.g., faster treatment) the number of reactors can be reduced and flow through the well increased, potentially offering an opportunity to increase capture width.

The feasible range of K_A values was restricted to 0.1 ft/day or less for additional analysis for InSRT. Using the lower end of the K_A range minimizes the number of reactors used and increases capture by maximizing the contrast in well and aquifer hydraulic conductivities. The application and effectiveness of horizontal wells in low permeability settings are in contrast to PT. PT extraction rates are maximized under high permeability conditions. In addition to the influence of hydraulic conductivity, the plume dimensions are also an important consideration. It was hypothesized when the InSRT reactor was proposed that the most efficient implementation would be in a source zone configuration where a smaller area of higher concentration occurs. Table 4 provides the number of wells required when the entire 350-ft wide plume was included in the remediation goals. To test the hypothesis of source zone size, the number of wells was recalculated for 25% and 5% of the PFAS plume width (Table 5).

In both cases, the number of wells required decreased as the number of reactors increased. Additionally, PFBA required more wells and reactors when either source zone was considered. When 5% of the source zone was used, two reactors and one well were sufficient to address PFBA. In the subsequent InSRT analysis, one well and two reactors were used, recognizing that the analysis would show an increase in the number of wells with increasing K_A and a decrease in the number of wells for PFOA, PFOS, and FOSA as those compounds are more readily degraded than PFBA. While scenarios requiring more materials or electricity can be considered, it is

TABLE 5 The number of InSRT wells required for K_A = 0.1 ft/day and 25% or 5% of the plume width (85.5 and 17.5 ft, respectively)

PFAS	Percent of plume	1 Reactor	2 Reactors	3 Reactors	4 Reactors	5 Reactors
PFOA	25%	1	0.7	0.5	0.4	0.3
	5%	0.3	0.1	0.1	0.1	0.1
PFOS	25%	2	1	0.8	0.6	0.5
	5%	0.5	0.2	0.2	0.1	0.1
PFBA	25%	10	5	3	3	2
	5%	2	1	0.7	0.5	0.4
FOSA	25%	2	0.8	0.6	0.4	0.3
	5%	0.3	0.2	0.1	0.1	0.1

Abbreviations: FOSA, perfluorooctane sulfonamide; PFBA, perfluorobutanoic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonate.

TABLE 6 Number of groundwater extraction wells required to remediate the target PFAS plume area (350-ft wide and 1,000-ft long) for different hydraulic conductivity values

Hydraulic conductivity (K_A) (ft/day)	0.02	0.1	2	20	50	100
Number of wells and pumps	17,546	1,082	168	40	9	1

Abbreviation: PFAS, per- and polyfluoroalkyl substances.

important to differentiate when InSRT will be operating efficiently to discourage inappropriate implementation.

Potential PT systems were established using the aquifer hydraulic conductivity values, as described in Section 2. It was found that as K_A increased, the total mass of well materials used at the site decreased along with the greenhouse gas emissions from materials and well construction (Tables 6 and 7).

When K_A was 0.1 ft/day, 1082 extraction wells were required, each with its own in-well pump operating at 0.1 gpm. As K_A increased, the number of wells and pumps decreased because potential flow rates increased thereby increasing the radius of influence (Fetter, 2000). When K_A was 50 ft/day, nine wells and nine pumps (54 gpm each) would be needed. In addition, the total mass of well materials will also increase. As aquifer hydraulic conductivity increases, PT becomes increasingly viable as noted by Horst et al. (2019). Low permeability sites are challenging for groundwater extraction but are optimal for some systems, including horizontal wells (Horst et al., 2019). The acceptable hydraulic conductivity range for PT was restricted hereafter to a minimum of 2 ft/day for additional analysis.

3.2 | Lifecycle environmental impacts

The ideal implementation scenario for InSRT and PT reduces the site footprint and lifecycle energy consumption of either system. Phases 1 (travel), 2 (materials manufacture), and 3 (well installation) were previously described in Table 2 and the impacts of each are discussed below. The Phases 1, 2, and 3 impacts were estimated for InSRT and PT for the optimal configurations determined above. First, energy consumption was determined for Phases 1–3 (Figure 3).

The results show that travel impacts were very similar, however, travel impacts were specific to initial installation, not recurring impacts because the frequency and distance of site-related travel may be difficult to predict. In Phase 2, the energy used for materials production was greater for PT than for InSRT because the total horizontal well length and mass of the reactors used was greater than for the PT extraction well. Phase 3 (installation) energy consumption was much less than the energy consumption in Phases 1 or 2. Consumption for InSRT installation was 1.2 MMBTu while PT was 0.9 MMBTu, or about 25% less.

TABLE 7 Direct and indirect costs for InSRT and PT for 30 years

Cost type	InSRT	PT
Capital cost (USD)	266,000	4,980,000
Lifecycle electricity cost (USD)	18,160	236,000
Direct cost of CO_2 (USD)	284,000	3,360,000
Indirect cost of CO_2 (USD)	15,000	126,000

Abbreviations: InSRT, In situ reactor technology; PT, pump-and-treat.

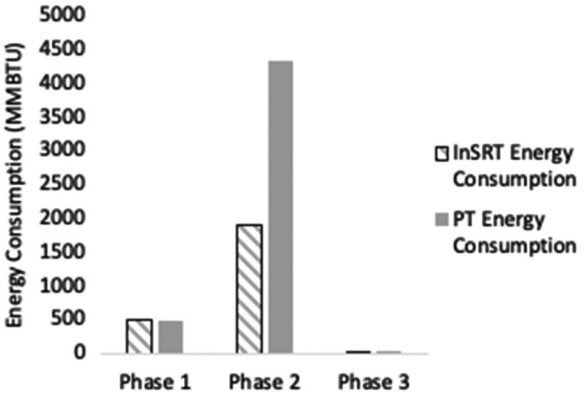

FIGURE 3 Energy consumption (in millions of BTu, MMBTu) in Phases 1 (travel), 2 (materials manufacture), and 3 (well installation) for InSRT (gray-striped bars) and PT (gray solid bars). The InSRT system was one well with two reactors operating at 0.1 ft/day and the PT system was nine extraction pumps each operating at 54 gpm with a radius of influence of 89 ft. InSRT, In situ reactor technology; PT, pump-and-treat

The Phases 1–3 emissions were also quantified using SiteWise. The results for CO_2 emissions equivalents are shown in Figure 4. Phase 1 impacts were again similar given the type of travel considered. Phase 2 impacts were greater for PT than for InSRT because of emissions from virgin GAC production.

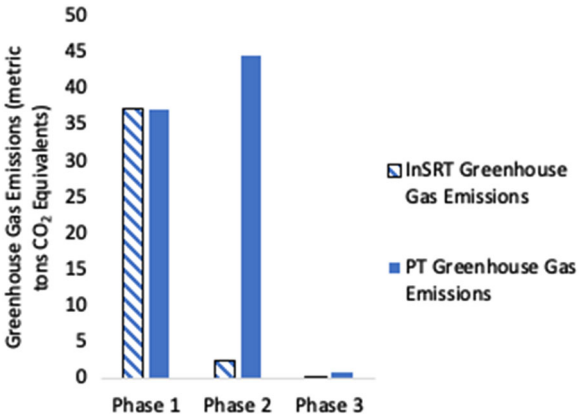

FIGURE 4 Greenhouse gas emissions as CO_2 equivalents for Phases 1 (travel), 2 (materials manufacture), and 3 (well installation) for InSRT (blue striped bars) and PT (blue solid bars). The InSRT system was one well with two reactors operating at 0.1 ft/day and the PT system was nine extraction pumps each operating at 54 gpm with a radius of influence of 89 ft. InSRT, In situ reactor technology; PT, pump-and-treat [Color figure can be viewed at wileyonlinelibrary.com]

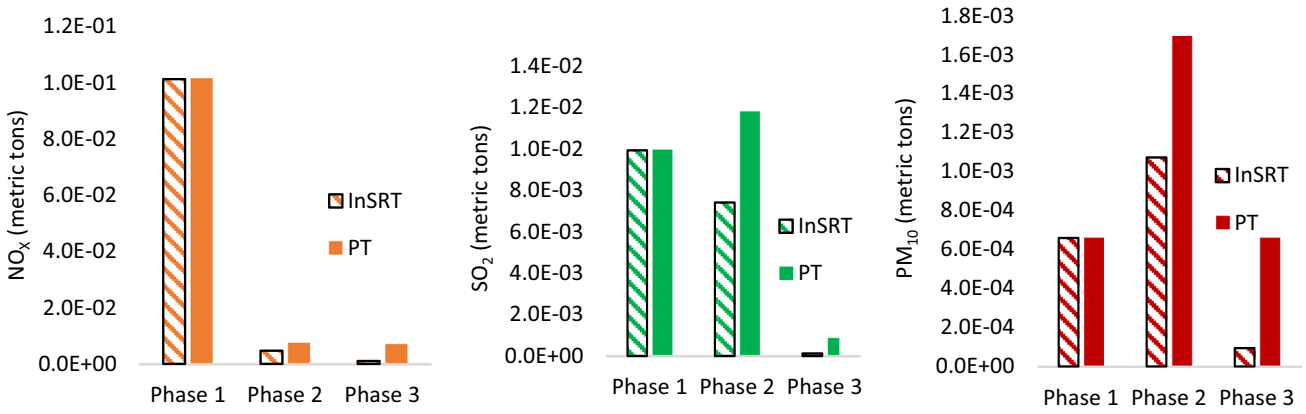

FIGURE 5 From left to right: nitrogen oxide (NO_X, orange), sulfur dioxide (SO_2, green), particulate matter smaller than 10 μm in diameter (PM_{10}, red) emissions. For Phases 1 (travel), 2 (materials manufacture), and 3 (well installation) for InSRT (blue striped bars) and PT (blue solid bars). The InSRT system was one well with two reactors operating at 0.1 ft/day and the PT system was nine extraction pumps each operating at 54 gpm with a radius of influence of 89 ft. InSRT, In situ reactor technology; PT, pump-and-treat [Color figure can be viewed at wileyonlinelibrary.com]

In Figure 5 the metric tons of NO_X, SO_2, and PM_{10} emissions are given for InSRT and PT in Phases 1, 2, and 3.

The NO_X emissions were greater in Phase 1 due to travel impacts, particularly air travel, and were greater in Phase 2 for PT because of the mass of well materials. The same was true for SO_2 and PM_{10} emissions.

These results do not account for recurring lifecycle emissions impacts for InSRT or PT which include electricity for both systems, and GAC production in the case of PT. Specific contributions to these impacts are covered in Section 4.

The lifecycle duration will be determined using data from monitoring systems until remediation goals are achieved, which could vary site to site. A specific lifecycle duration cannot proactively be established for either system but impacts over time can be estimated for operation from 1 to 30 years. The scenario with the smallest number of wells was selected for InSRT (one well, two reactors) and PT (nine wells, one pump, 54 gpm per well). Therefore, Phase 4 (operations) electricity consumption was estimated separately over 30 years (Figure 6).

Figure 6 indicates electricity consumption was less for InSRT than for PT over the lifecycle duration. For a given period of time, when each system is installed under optimal conditions, electricity consumption and associated impacts will be greater for PT than for InSRT. After 30 years, for example, the greenhouse gas emissions for reactor and pump operation were 236 and 666 metric tons, respectively.

The recurring impact of GAC replacements was also considered for the pump and treat scenarios. For example, at a system flow rate of 54 gpm, using a sorption capacity of 0.4 mg/g determined for influent water with a mixture of PFAS (Woodard et al., 2017) replacements would occur every 2 months to 1.5 years, depending on the target PFAS. Replacing GAC every 2 months may be considered too frequent, depending on budget and logistical considerations and a higher capacity media may be required. It is important to note that the actual capacity will depend on the influent water characteristics, including the PFAS present, and would be determined from site-specific testing. Application of GAC at different sites has been summarized and replacements reported to occur from 2 months to more than 1.5 years (Reade et al., 2019; Weston & Sampson, 2017). Expanding on the above example using the 0.4 mg/g value, GAC was assumed to be replaced once per year for up to 30 years. The CO_2 equivalent emissions from the GAC replacements was compared to the CO_2 emissions from InSRT and PT electricity (Figure 7).

The CO_2 emissions equivalents were greatest for electricity-related emissions from pumping then GAC replacements, followed by InSRT operation emissions. Of course, a lower-impact sorbent material or reduced replacement frequency would decrease media-related emissions impacts. As shown in Figure 6, in terms of electricity consumed, the PT pump operation resulted in more CO_2 equivalent emissions than InSRT operation as a product of lower electricity consumption under optimal conditions.

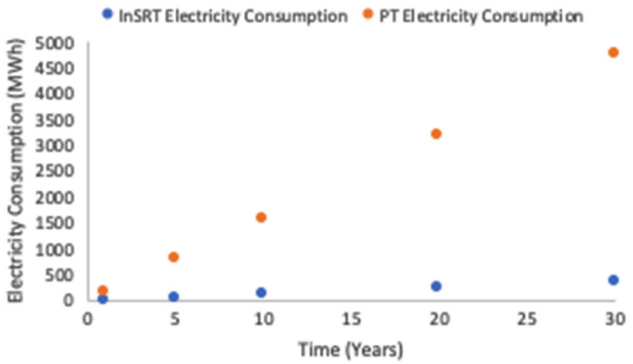

FIGURE 6 Electricity consumption (in MWh) from operation of InSRT (blue circles) and PT (orange circles) over 30 years. The actual lifecycle duration would depend on the rate of PFAS mass reduction in practice and the time to reach remediation goals based on data from sampling events. InSRT, In situ reactor technology; PFAS, per- and polyfluoroalkyl substances; PT, pump-and-treat [Color figure can be viewed at wileyonlinelibrary.com]

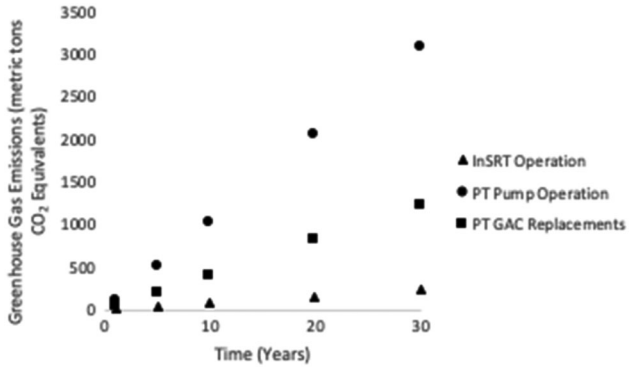

FIGURE 7 Greenhouse gas emissions (as CO_2 equivalents) from Phase 4 for both InSRT (electricity) and PT (electricity and GAC). InSRT emissions are indicated by black triangles, PT pump emissions by black circles, and PT GAC-related emissions by black squares. GAC, granular activated carbon; InSRT, In situ reactor technology; PT, pump-and-treat

If InSRT or PT are implemented at sites with nonideal conditions, the energy demand and pollutant emissions values will increase. Given the multitude of InSRT scenarios observed when analyzing the data and the wide range of environmental impacts possible, the InSRT reactor should only be used under optimal conditions, that is, the InSRT reactor has not been found to be appropriate for all sites, particularly where aquifer hydraulic conductivity values above 0.2 ft/day exist.

3.3 | Direct and indirect costs of CO_2 emissions

The optimized scenarios above were used to calculate the direct and indirect costs and compare sustainability performance of each system. In each case, PFBA was used because it had the slowest rate constant in Table 3 and created a worst-case scenario for InSRT hydraulic retention time. Moreover, short-chain PFAS have been found to limit GAC lifetimes compared to long-chain PFAS (Westreich et al., 2018). The same analysis was performed for PFOA (not shown) and it was found that impacts were less for InSRT and PT because both systems performed better for PFOA degradation or removal, respectively. Table 7 shows the costs calculated for InSRT and PT for a 30-year lifecycle, as an example. If one or both lifecycles were less than 30 years, the costs would also be less.

The capital costs were greater for PT than InSRT because PT systems described in cost documents required more infrastructure than has been estimated for InSRT (U.S. EPA, 2009; Weston & Sampson, 2017). The lifecycle electricity costs for PT were greater than for InSRT (Table 7). When the Phase 4 CO_2 emissions for 30 years were considered, the direct cost of the emissions was an order of magnitude greater for PT than for InSRT. The higher direct cost for PT was the product of increased electricity consumption. The direct cost results again reflect the importance of optimizing lifecycle operating time for InSRT and PT to conserve energy and limit the CO_2 emissions while achieving PFAS remediation goals. The indirect costs were calculated as described in Section 2 at a cost of $40.85 per metric ton of CO_2 emissions. The indirect costs of CO_2 emissions from InSRT were $15,000 and from PT were $126,000 for a 30-year lifecycle. The indirect data are not comprehensive but do demonstrate that the negative costs (i.e., pollution potentially harming human health) is less for InSRT than PT, assuming InSRT operation can be optimized to less than a 10-year lifecycle.

4 | DISCUSSION

4.1 | Costs and benefits of the remedial alternatives

The CO_2 emissions produced (Figures 4 and 7) contribute to the greenhouse effect which has successive impacts on environmental and human health, quantified by the indirect costs in Table 7. NO_X, SO_2, and PM_{10} all contribute to acid rain which damages vegetation and acidifies surface waters. In this context, InSRT will contribute less to global warming, acid rain, or health effects from PM_{10} emissions than PT will when operating under their respective scenarios. These results demonstrate, at least initially, that remediation using InSRT will have the greatest net benefit to the environment and society by minimizing emissions. The following discussion evaluates greenhouse gas emissions that occur when producing the electricity consumed by the reactor or different PT scenarios.

CO_2 emissions equivalents were greater for GAC than InSRT or PT electricity consumption. The environmental impacts of GAC have been evaluated in the literature. For example, Bayer and Finkel (2006) reported that during the activation step, 60% of the mass of coal used is typically lost, such that 3 tons of hard coal are required to produce 1 ton of GAC (Bayer & Finkel, 2006). Regenerated GAC production was found to have lower pollutant emissions compared to virgin GAC (1.17 kg CO_2/kg GAC vs. 11 kg CO_2/kg GAC; Bayer & Finkel, 2006). Reducing electricity-related emissions would decrease the environmental impacts from each remediation system. It is well understood that the electricity mix dictates total pollutant emissions from electricity production. When a larger percentage of nonrenewable energy sources are used to produce electricity, the resulting pollutant emissions are greater. Florida was used as the electricity mix in SiteWise with the greatest contribution from natural gas (12,400 kWh), and lesser contributions from nuclear (2224 kWh), and coal (1947 kWh; U.S. Energy Information Administration, 2019). Increasing the contribution of renewable energy to electricity production would reduce pollutant emissions. The U.S. EPA (2009) has provided guidance to reduce the environmental footprint of PT systems with particular emphasis on completing thorough site investigations. Detailed site assessments may show optimal well placement allowing pump rates to be minimized saving electricity over the system lifetime.

Another important benefit of the horizontal well and in situ remediation in general, is the reduced risk of exposure. In ex situ systems both personnel on-site and the public have an increased risk of exposure to contaminants from accidental release. Moreover, any

contaminated material removed will also need to be treated or securely stored adding additional costs to the project. Though not quantified, horizontal remediation wells were also previously noted to have a smaller footprint at a site compared to other technologies thereby limiting habitat disturbance as well (Lubrecht, 2012).

4.2 | Direct cost comparisons

The direct cost data indicated that InSRT had lower associated cost per kilowatt hour compared to PT. As noted above, the direct costs were found to vary considerably with changes in aquifer hydraulic conductivity. The results show that, in terms of direct financial considerations, InSRT can be the lower cost option but the selected system should first and foremost be viable for the given site.

Direct (and indirect) cost of emissions can be useful to assess environmental and social impacts for a remediation system. However, it is important to note that these costs may only be considered important if sustainability goals have been set and are generally a concern for the project.

For difficult-to-treat contaminants, such as PFAS, fewer treatment options may exist and higher sustainability costs may be unavoidable to meet remediation goals. Willingness to pay higher direct capital and operations costs is likely to be different (if remediation goals are met) than willingness to incorporate indirect costs into the total. Indirect impacts and associated costs are much less tangible than direct impacts and they may not occur immediately. Much like direct impacts, considering whether indirect impacts allow a remediation system to be considered sustainable depends on the importance stakeholders place on sustainability. If they are concerned about impacts outside the immediate area, then indirect costs are more likely to be weighed with direct costs. Otherwise, it may be easy to neglect the indirect costs.

Regardless of who pays, and even if they are willing to pay a relatively large sum, the ideal scenario still minimizes costs. According to Smith (2019), historically, when human health is concerned, public pressure has often meant no remediation system can be implemented quickly enough, even if the system is not fully optimized for the site or has environmental impacts greater than those it relieves. For emerging contaminants with high levels of public scrutiny these situations are more apt to occur. If the direct costs were within the stakeholder's cost boundary the direct emissions costs would be less likely to be considered. Nonetheless, one or more populations will carry the indirect cost burden. Considering the indirect costs will require a balance of meeting public and government demand for rapid remediation and meeting the sustainability goals. To better identify if goals were met, the indirect impacts need to be quantified, as presented in Section 3.

4.3 | Comparison of InSRT and PT inclusive of indirect costs

In Figure 2 the indirect impacts were outside the system boundary indicating that these impacts were not included in the direct costs because they did not occur from operation. Materials and energy production were assumed to occur offsite, possibly hundreds of miles away. The impacts from those activities may be realized in the immediate vicinity of the production area, or at a greater distance from production. The impacts from production activities may not immediately be apparent. For example, negative health outcomes may not be observed until one or more generations have been exposed. The indirect costs were included (Table 7) and were greater for PT as a result of greater electricity demand. The total indirect costs for InSRT were less than those for PT, so assuming both systems achieve the remediation goals, the system with lower indirect costs would be selected.

The value of remediation, and acceptance of the indirect costs, can be associated with costs to human health. A recent report estimated the costs from kidney cancer mortalities as a result of not addressing PFAS contamination. Kidney cancer is a probable endpoint from occupational PFAS exposure, and the report estimated for a European population (335,000 individuals), 83,627 would have an elevated risk of developing kidney cancer and from those individuals, 3.6 additional deaths would occur. The total value of these lives lost would be approximately 12.7 million Euros (Goldenman et al., 2019). These calculations used value of a statistical life (VSL) which is essentially the monetary benefit of avoiding a fatality. The U.S. EPA VSL is $7.4 million (in 2006 dollars; U.S. EPA, 2010). Using the data (above) from Goldenman et al. (2019) and kidney cancer as an endpoint, the number of additional deaths was estimated for a population of 12,000 individuals assumed to be affected by contaminated water in the case study. It is important to note that the estimate represents a simplified estimate of one mortality source to highlight the importance of considering health-related costs. The estimate does not represent a thorough risk assessment for the population. The result of the estimate was less than one additional death from kidney cancer would occur, therefore, pursuing remediation for the sake of protecting human health will depend on the risk the population considers acceptable. The perceived need for remediation would also depend on the potential exposure levels. However, kidney cancer is not the only possible endpoint, and the number of fatalities is uncertain, meaning that more or fewer fatalities could result and influence willingness to pay. U.S. EPA VSL for a single life is greater than the total remediation costs for InSRT in Table 7 but less than those for PT. As risk increases, from a cost perspective, it would be less expensive to prevent the potential deaths. Alternatively, if the risk is considered acceptable and the value of remediation is determined to be low (and remediation is not mandated by a governing body), it is unlikely to be completed voluntarily. The indirect costs were calculated (Table 7) to account for costs of negative health outcomes from exposure to emissions from electricity generation. If there is elevated risk of mortality from exposure to pollutant emissions, these same VSL-based costs should be included and considered. If those costs do not balance (i.e., there is a greater risk of mortality related to electricity generation than PFAS exposure) the system cannot be considered sustainable according to the definitions discussed in Sections 1 and 2. If PFAS-related

mortalities are prevented and the value is considered significantly greater than the negative indirect costs, the emissions impacts may be considered acceptable. The indirect costs then must be considered on a site-specific basis, not only for the PFAS site, but for the electricity and materials production sites as well. While indirect costs could be predicted in a more general case, risk-based values would add useful detail to compare direct and indirect costs.

5 | CONCLUSION

The study was intended to evaluate if the InSRT system could be considered sustainable in comparison to an alternative system (PT) that could also be used for PFAS remediation. It was found that InSRT is not the efficient option to use in high hydraulic conductivity settings and the PT system may be more viable. However, under lower hydraulic conductivity conditions, InSRT is more efficient than PT. Compared to a PT system using GAC, InSRT used less energy and produced fewer pollutant emissions. Moreover, the reactor has been shown to mineralize PFAS as compared to GAC, which captures but does not degrade PFAS. Incorporating direct and indirect costs of pollutant emissions showed that InSRT was a more sustainable option compared to PT under the given operating conditions.

ACKNOWLEDGMENTS

The authors declare the following financial interests: We have co-founded RemWell, LLC a company which intends to market the device described in this article pending complete field validation. However, we recognize the need for transparency regarding all methods and results. We also do not promote the reactor as a solution for all sites.

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