In situ thermal remediation for source areas: Technology advances and a review of the market from 1988-2020

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In situ thermal remediation (ISTR) is perhaps the only *in situ* technology that is capable of complete removal of non-aqueous phase liquids (NAPLs) from contaminant source zones within time frames of less than a year. Other in situ technologies that may be capable of similar treatment times (chemical oxidation applied via soil mixing, in situ soil stabilization, surfactant-enhanced remediation, etc.) are often plagued by limitations to either their implementability or their effectiveness. Most often, this relates to the nature of the geology impeding sufficient contact with the NAPL or non-target compounds resulting in partial treatment of the targeted contaminant mass.

Excavation is arguably the only comparable technology to ISTR for reliability and effectiveness of achieving expedited source zone mass removal. Both have their place and must navigate site features such as geology, depth, surface topography and proximity to infrastructure (roads, buildings, and utilities). However, excavation is inherently more intrusive and requires more complex material handling logistics to consider, including both disposition of the excavated material and its backfill.

As we discuss later, ISTR has the ability to achieve complete removal of NAPL (and other volatile contaminant types) across a variety of complex site settings, subsurface environments, or compounds of interest and has driven a steady use of the technology through the present time. As stated by Kira Lynch, Superfund and Technology Liaison for the United States Environmental Protection Agency (USEPA), Region 10, "…in situ thermal treatment is really one of the most effective ways to deal with significant source areas in a reasonable timeframe" (Lynch et al. 2020).

In this column, we have endeavored to pick up from the comprehensive review of ISTR completed in the late 2000s by Jennifer Triplett Kingston during her PhD work at Arizona State University (Triplett Kingston 2008). We have included a review of vendor evolution, the growth of the market for the technology, related application trends (contaminant applications, geography), advancements to the technology, and thoughts on where the market for ISTR may be headed in the next 10 years. This includes a fresh look at the sustainability and resilience profile for a technology that was once considered to fall outside of the sustainable remediation toolkit.

ISTR History, Development, and Growth

The concept of using thermal technologies for in situ remediation purposes began in the oil and gas sector, where the heating of oil reservoirs has been performed since the 1950s to enhance recovery of petroleum by decreasing viscosity of heavy oils and increasing volatility of lighter fractions. From here, the following have developed:

- Steam enhanced extraction (SEE) was the first thermal method used for environmental remediation, with technology development during the 1980s in both the Netherlands (Hilberts et al. 1986) and at the University of California Berkeley (Udell et al. 1991).
- Electrical resistance heating (ERH), which had been studied since the 1970s for bitumen recovery in the Alberta oil sands (Vermeulen et al. 1979), was first applied for environmental remediation at the Lawrence Livermore National Laboratory (LLNL) in 1992 by combining ERH with SEE in a process termed dynamic underground stripping (DUS) (Daily et al. 1995; Newmark and Aines 1995) and at the Savannah River Site in 1993 by the Battelle Memorial Institute in a six-phase configuration (Gauglitz et al. 1994).

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- In 1989, the first patents for thermal conductive heating (TCH) use for remediation were filed by Shell Oil Company (Vinegar and Stegemeier 1991; Vinegar et al. 1993), which were later donated to the University of Texas and then licensed in 2000 for commercial application.
- The first trial of radio frequency heating (RFH), where radio waves are applied at authorized frequencies of 6.78, 13.56, 27.12, or 40.68 megahertz (MHz) (Price et al. 1999), occurred in the late 1980s at the Volk Air National Guard Base (U.S. Army Corps of Engineers [USACE] 2009).
- The concepts of using smoldering combustion reactions for remediation also began in academia at the University of Edinburgh in 2005. The first smoldering field projects were initiated in the late 2000s.

Detailed descriptions of SEE, ERH, and TCH can be found in Johnson et al. (2009), Davis (1998), USEPA (2004), USACE (2009), and others. In situ smoldering combustion (or STAR technology) is described in Grant et al. (2016) and RFH is described in Price et al. (1999). In our discussion, any low-frequency electrothermal processes (i.e., ET-DSP™, OptiFlux™) are referred to as ERH technologies, while any in situ thermal desorption (ISTD) processes are referred to as TCH technologies.

As previously mentioned, in the late 2000s a comprehensive review of ISTR technologies was completed by Jennifer Triplett Kingston during her PhD work at Arizona State University (Triplett Kingston 2008). This work was also published elsewhere (Johnson et al. 2009; Triplett Kingston et al. 2010; Kueper et al. 2014) and involved research and interviews with the major thermal vendors at the time (as well as other key regulators and practitioners), ultimately identifying 182 pilot and full-scale projects completed between 1998 and 2007. Data collected (where provided) included design parameters (area, depth to treatment zone, thickness of treatment zone, target chemicals of concern, treatment area and depth, density of energy delivery, spacing of delivery points); operating parameters (target temperature, temperature achieved, heating duration); and performance measures (pre-and post implementation monitoring data). This work was timely as the thermal market was in a rapid growth mode and provided mixed viewpoints on applicability and success criteria for the sites of interest. Observations made by the Triplett Kingston work included:

- The absence or misinterpretation of pre-design investigation data resulted in poorly constructed conceptual site models and undersized treatment zones.
- ISTR was most often applied in lower-permeability or heterogeneous settings with few applications in weathered or fractured bedrock.
- Achieving the target temperature in situ, in and of itself, did not appear to correlate well with desired treatment performance. Performance was more likely dependent on a combination of heating duration, treatment zone size, and achieving desired temperature.
- Most project durations were implemented over a time frame of less than six months, with little documentation as to the rationale used for determining the duration during the design phase.
- ERH sites dominated the total number of projects at that time, with three times as many ERH projects as all other technologies between 2000 and 2007. TCH sites were trending upward as SEE sites were trending downward.

With respect to the first bullet, and to put it simply, it was recognized that a solid handle on the mass distribution and treatment volume was critical to the success of the application, especially in light of the frequent applications in low-permeability or heterogeneous settings (second bullet) where contaminant distribution is even more challenging. In the time frame of the former study, investigation tools and techniques mostly relied on visual observations of staining or measurable product in monitoring wells to determine extent of source areas, or soil and groundwater samples used to determine saturation or solubility thresholds for compounds. Coupled with unrealistic performance objectives, ISTR treatment can have minimal benefit or improvements in groundwater quality/site progress to closure when targeting only part of the source zone. Today, we have better investigative tools in our toolkit to provide more accurate definition of a source zone and minimize data gaps. Those tools are discussed in sections below.

With respect to the third bullet, it seems evident that achieving the target temperature is not only a threshold consideration, but one that is spatially and temporally variable. For instance, if steam distillation of a chlorinated solvent is expected to occur at a certain temperature, locations anticipated to heat the slowest (i.e., farthest from the energy delivery points) must achieve that temperature to accelerate removal at these locations. Sustained energy input is then necessary for boiling mechanisms to drive mass transport towards capture points until mass or concentration reduction objectives are met. Lastly, as indicated by the first bullet, the treatment volume must be properly delineated so that the entirety of the source zone is heated to above threshold temperatures.

The fourth bullet is difficult to comment on, as time frame for operation is not only dependent on achieving temperature and holding it steady, but also partly on the design philosophies of heating and treatment. To complete a project properly in a shorter time, the spacing between energy delivery points would need to be minimized and a larger utility service drop would be necessary. To handle the same mass removed in a shorter time would also require a larger treatment system. Project economics likely rule the balance among spacing, system size, and duration of heating.

Related to market trends (fifth bullet), 13 years have passed since the accumulation and reporting of the Triplett Kingston compilation. Therefore, we thought it would be of interest to examine how the technology has evolved over that time and have undertaken an effort to provide an update on projects completed. The data compiled in this column was obtained through interviews of current ISTR vendors, including Deep Green of Duferco (Tubize, Belgium), GEO (Orange, California), Haemers (Brussels, Belgium), JR Technologies (South Egremont, Massachusetts), McMillan-McGee (Calgary, Alberta), Savron (Guelph, Ontario), TerraTherm (Cascade Thermal) (Gardner, Massachusetts), and TRS Group (Longview, Washington).

Each vendor provided its respective database of sites current at the time of this column for creation of a comprehensive list of ISTR projects that represent and were used to evaluate the current state of practice. Select key site parameters were also provided where possible, including technology used, location, contaminants targeted, treatment volume, days of operation, and total mass removed. Vendor interviews included a retrospective look at the evolution of the marketplace, specific technology developments, changes in cost, and projections on the future of the ISTR market over the next 10 years. The cumulative database is representative of most projects completed to date but is certainly not all-inclusive. Projects that were self-performed by consultants or industry clients have not been included, nor have any projects with strict confidentiality requirements. Projects using SEE, for which the patent expired in 2009, are likely underrepresented, as are lower-temperature groundwater recirculation and hot air injection applications. Also, while some vendors provided data on ex situ heating projects, those were not included in the analysis to limit the topic of discussion to in situ treatment of source zones. As part of the discussion, the vendors provided information on their business history, which shows a tremendous amount of start-up and consolidation activity.

Figure 1 presents a summary of the information collected, depicting a timeline of notable events for each of the vendors that participated in this study, including when they began ISTR, and activities such as mergers, acquisitions, or joint ventures (JVs). The JVs primarily establish relationships with other companies that are used to implement the vendor's technology in other areas of the world. The first JV documented was in 2006 and established a presence in Europe. Currently, several of the thermal vendors interviewed have established relationships on other continents (Europe, Asia, South America, and Australia), the majority of which were established in the 2010s. The number of vendors with established JVs in other continents speaks to the growth of ISTR outside of North America over the past decade.

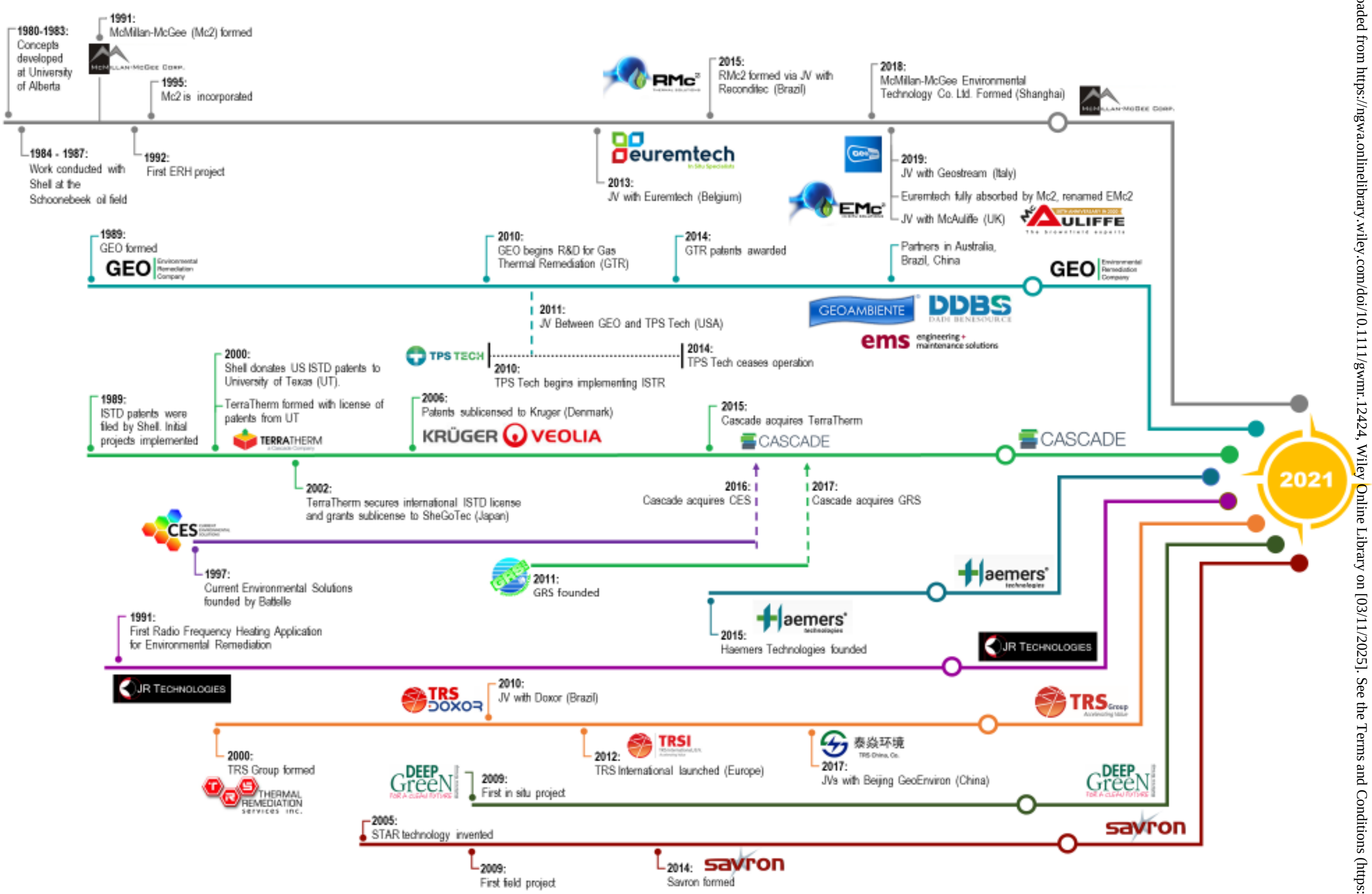

Figure 1. In situ thermal remediation vendor history.

The cumulative data set provided by the vendors included 643 records (inclusive of the original 182 from the Triplett Kingston work). Of those records, including both pilot and full-scale, 623 reported the type of technology used, 610 included the location, 613 included the date of implementation, 531 included the volume treated, 536 included type of contaminant(s), and 169 reported an estimate of total mass removed. Figure 2 shows the distribution of projects by (a) technology and (b) contaminant type. Chart (a) accounts for each technology independently in cases where technologies were combined on a project site (e.g., where a project used a combination of both TCH and SEE, both technologies are counted independently). The dominance of project technology by ERH has continued since the early 2000s, with more than half the number of projects (57.2%) involving ERH, followed by TCH at 29.5%. SEE, STAR, and RFH make up the remainder of the reported technologies used (8%, 2%, and 1%, respectively). This data covers the entire study period beginning as early as 1988 and any projects operating or planned for operation in 2021. Projects labeled as 'Other' capture soil mixing with steam, hot water recirculation, and possibly other technologies that were recorded in the earlier portion of the data set (Triplett Kingston 2008). Chart (b) shows contaminant types generalized into a few key categories. Chlorinated solvents have been the major contaminant class where ISTR has been applied, with 53% of projects involving chlorinated solvents as the only contaminant, and an additional 7% involving both chlorinated solvents and petroleum hydrocarbons. Projects targeting only petroleum hydrocarbons make up an additional 13% of projects, and then creosote and coal tar sites at 4%. Sixteen percent (16%) of projects did not include data on contaminant. While used in only a small percentage of projects, ISTR has been used for a broader list of compounds, including 1,4-dioxane, pesticides, herbicides, dioxins, furans, explosives, dyes, and perchlorate, as discussed in detail later.

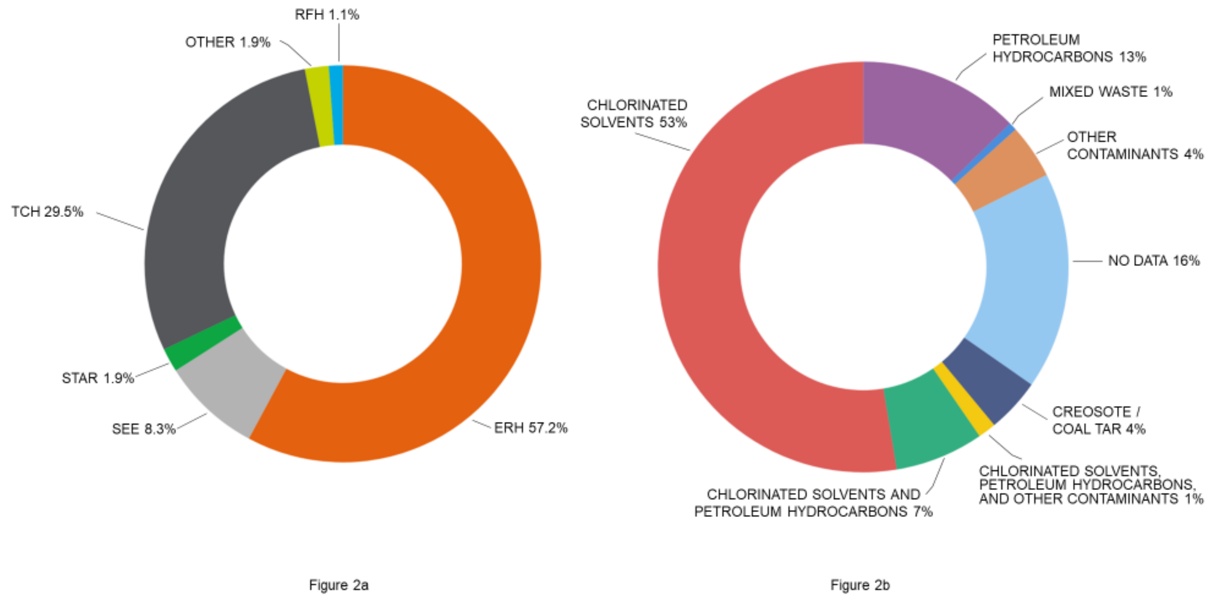

Figure 2. a) Distribution of technologies used and b) distribution of contaminants targeted (1988-2021).

Separately, the growth of the ISTR market was visualized by tallying the number of projects completed each year, as shown on Figure **3**. The stacked bar portion of the graph shows the total number of technology implementations (inclusive of projects with combined technologies) per year (where records provided both implementation date and technology). The graph indicates a steady growth between 1988 and 2007 and a notable reduction in projects between 2008 and 2010, which could be due to several factors. The dip in projects correlates with the Great Recession (2007-2009), which affected countries globally and may have caused several projects to be delayed based on market conditions. Also, the lower number of projects during that time correlates to the end of the prior period of study by Triplett Kingston et al. (2008) and the beginning of this data set, which may reflect differences in the sources of data used. However, the pace of projects completed appears to be fairly linear since approximately the year 2000, with an average of 24 projects completed per year.

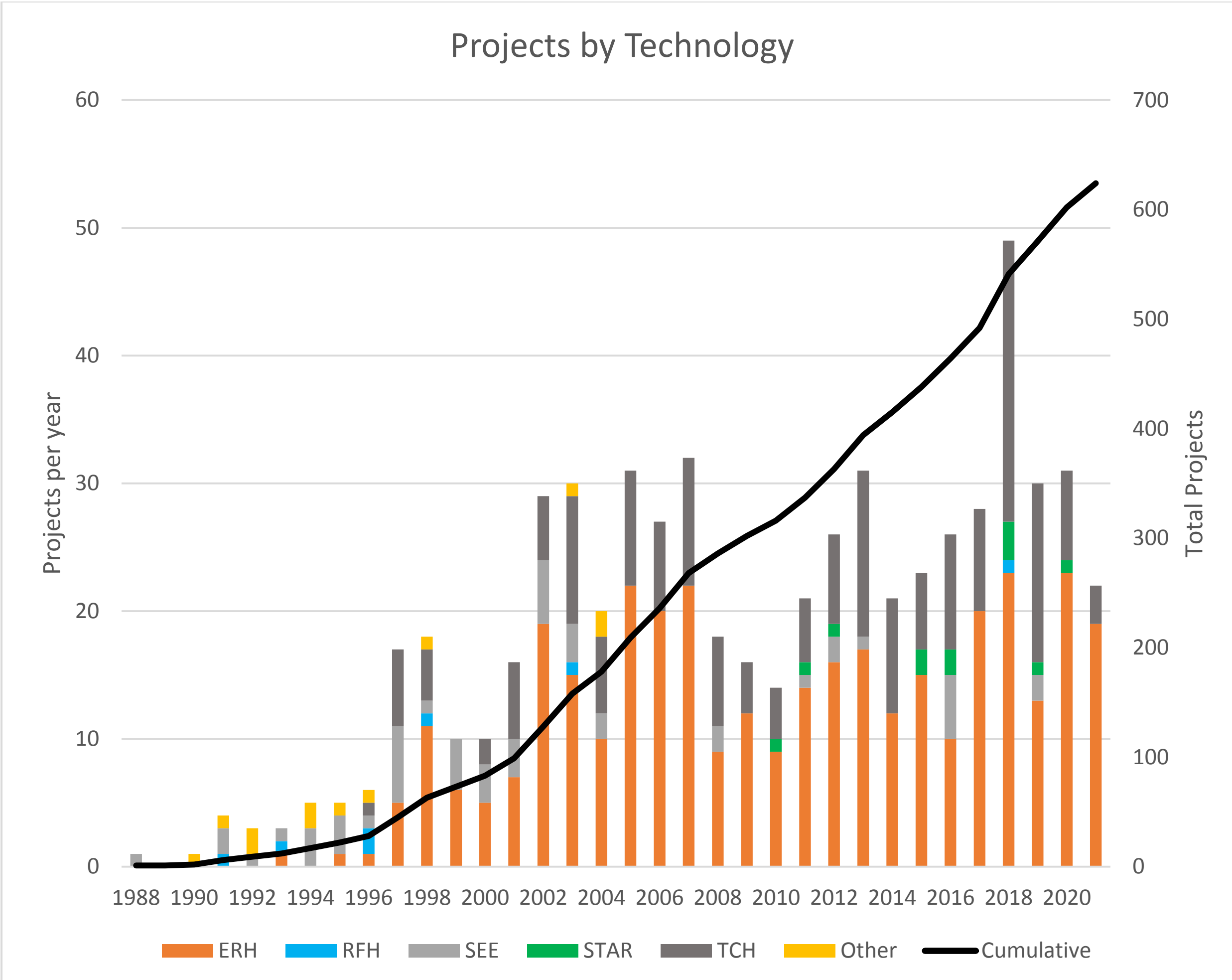

Figure 3. Number of ISTR projects each year by technology.

Considering project size, the map shown on Figure 4 plots each of the projects for which the location was provided (610 of 643 records). The diameter of the circle corresponds to the in-place volume treated as reported by the vendor. Where no volume was reported, the circle is set to the minimum diameter (531 of 643 records reported a volume). The largest project reported is the Visalia Poleyard Superfund Site in California, completed in 1997 using SEE with a total volume of 629,630 cubic yards. In terms of mass removed, the largest reported project included a calculated 2,648,320 pounds of petroleum hydrocarbons from the Williams Air Force Base in Mesa, Arizona (2016).

Figure 4. Global distribution of ISTR projects and relative size (volume of soil treated).

When analyzed by decade (Figure 5), the expansion of the thermal market outside of North America to other continents is evident. In the 1990s, ISTR development and project locations were predominantly in North America, with few exceptions. In the 2000s, a significant number of projects were completed in Europe, consistent with the business timeline shown on Figure 1. European companies that started in the ex situ thermal business (i.e., rotary kilns, ex situ thermal desorption) began implementing in situ projects, and many U.S.-based companies established JVs for implementation in Europe. Significant expansion into South America and Asia is also evident in the 2010s, with additional JVs created to gain access to emerging global markets.

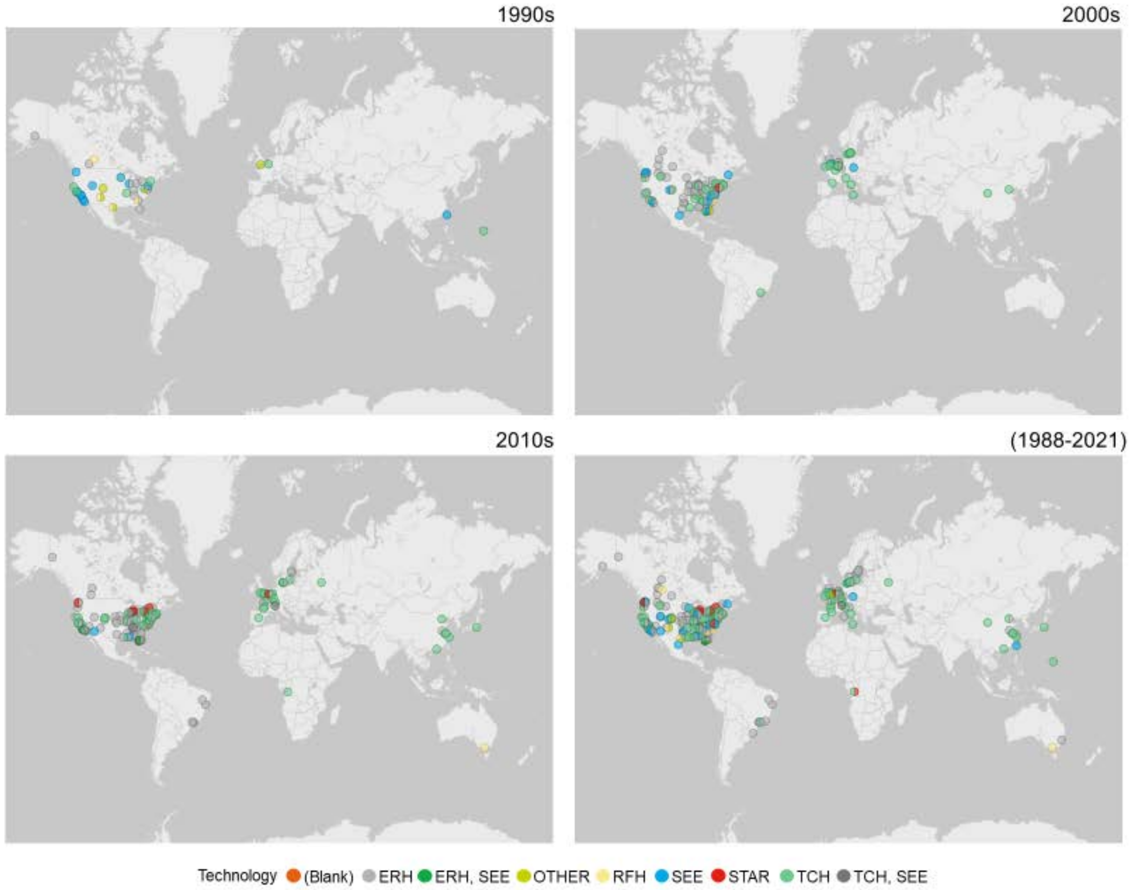

Figure 5. Global distribution of ISTR projects by decade (1990s, 2000s, 2010s) and all records (1988-2021).

Pricing and Reliability

Vendors reported that after a period of initial commercialization of SEE, TCH, and ERH in the 1990s to 2000s, and subsequently STAR in the 2010s, efforts shifted towards reducing costs and allowing for implementation in increasingly complicated or challenging settings. Early developments in the 2000s included value engineering to reduce the costs of TCH, which was initially envisioned by Shell Oil Company for higher-temperature applications (above 100 degrees Celsius [°C]), so that it became competitive with ERH for treating chlorinated solvent and volatile hydrocarbon source zones. Since then, ISTR vendors have incrementally reduced implementation costs by using less expensive materials or reusing materials from project to project where possible, capitalizing their equipment to reduce rental rates, modularizing designs, automating control systems, recovering waste heat for pre-heating fluid process streams, and eliminating redundant data collection when it provides little value, among other developments. Practitioners also recognized that by increasing the spacing between energy delivery points and running for longer periods, smaller ex situ treatment process equipment could be used, leading to overall costs savings. For instance, full-scale operations reportedly took an average of 166 days between 1998 and 2007, while the average length of operations from 2008 to present was 236 days.

For the present study, costs were not provided consistently across the vendors to be able to draw conclusions of how they have varied over time, as no context was provided as to cost breakdown (e.g., whether the vendors' costs included civil work, utilities, ex situ treatment, etc.). However, most reported that unit costs have remained relatively flat or increased slightly as reliability in performance has increased and projects have become more complicated. For example, angled or horizontal drilling is now regularly performed to install electrodes or heaters underneath buildings, roadways, railroads, or other surface obstructions; cold-water recirculation or quenching systems are often installed to protect critical utilities that cannot be rerouted; surface completions are being buried or placed in false floors to provide existing building tenants with unencumbered access above the thermal footprint; and ISTR has been implemented at sites with dozens or hundreds of identified compounds, each of which requires consideration in the design of ex situ treatment and permitting processes (e.g., Bowerman et al. 2018). These cost trends are consistent with data presented by Kinney and Blundy (2020), which reported average unit costs of approximately $142 and $259 per cubic yard from 2012 to 2019 for treatment volumes greater than or less than 10,000 cubic yards, respectively, with costs having risen about $8 to $10 per cubic yard annually. A typical cost breakdown is approximately 30% drilling, civil, electrical, and other subcontractors; 20% labor; 15% materials; 15% treatment and support equipment; 10% energy; 5% heating equipment; and 5% other direct costs (LaChance 2020). Costs are often driven by economies of scale and risk-sharing among the executing parties; thus, savings can be realized for larger, lump-sum projects that are tied to clear and achievable performance goals, allowing for more flexible implementation.

Technology Developments

Several TCH technologies have been commercialized since the publication of Triplett Kingston et al. (2008). In the early 2010s, gas-fired TCH burners (e.g., GTR®, Smart Burners) established a place in the market as a modular technology that did not necessarily require upgrades to the onsite electrical service. Gas-fired TCH has also been optimized to beneficially reuse the extracted vapors as a supplemental fuel source, virtually eliminating the need to dispose of contaminants that have a suitable BTU value (Geckeler 2019). Use of more sustainable biofuels has also been introduced. More recently, TCH technologies using (i) downhole induction IT-DSP™ heaters that induce eddy currents onto direct-buried casings and eliminate the radiant heat transfer step (Delos Reyes et al. 2018) and (ii) helical resistive Flexheaters® that modify power input along the length of the wellbore by varying coil densities (Oberle et al. 2019) have been commercialized and deployed at sites in the United States, Brazil, Sweden, and China. As demand for treating higher boiling-point and/or emerging contaminants continues to grow, we foresee the market share of TCH projects increasing in the coming years.

STAR has also recently established a position in the market with an entirely different strategy for treatment of high-BTU value, low-volatility compounds such as coal tar, creosote, and heavy fuel oils in situ at concentrations of 3,000 to 5,000 milligrams per kilogram (mg/kg) or higher (Pironi et al. 2011; Savron 2018). This technology is based on engineered smoldering combustion reactions, where the contaminant (i.e., the fuel source) is ignited subsurface and supplied oxygen in the form of injected air that sustains the combustion propagation. The first pilot test was reported to have been completed in approximately 2010, while the first full-scale project was completed from 2015 to 2019. To date, the most important advancement in reducing the cost of STAR has been the development of a smouldering ignition method that uses convection of heater air to ignite NAPL immediately adjacent to the wellbore (Scholes et al. 2015).

Design and Performance Monitoring

The thermophysical properties that drive the volatilization, distillation, stripping, desorption, dissolution, and/or viscous mobilization of organic contaminants at elevated temperature have been well-documented in the literature (e.g., USACE 2009; Kueper et al. 2014). How they are considered in ISTR design, however, has continued to evolve. Rather than design from first principles, practitioners often rely on empirical performance criteria derived from previous projects, which have become increasingly refined with experience, where a specific energy density (e.g., 200 to 350 kilowatt-hours per cubic yard), temperature target held for a length of time (e.g., 100°C at 90% of sensors for 30 days), or percent boil-off of the interstitial water (e.g., 30 to 50%) achieved a specific remedial objective for the same or similar contaminant. Heat and mass transfer calculations are then performed to predict how to achieve these performance criteria. Calculation methods may include energy balances to predict average temperatures within the major strata (USACE 2009), single-well analytical methods to model radial heat injection (e.g., Marx and Langenheim 1959; Killough and Gonzalez 1986), or non-isothermal multiphase flow simulators to model three-dimensional (3D) temperature distributions within elements discretized as finely as 1 cubic yard (e.g., Vinsome et al. 1994; Pruess and Battistelli 2002). Though approaches vary between practitioners, most report that they have incrementally improved their workflows and can predict energy budgets to within approximately 10% of the actual energy used on a project, as long as pertinent field parameters, including groundwater fluxes, electrical conductivities, and intrinsic permeabilities, are adequately characterized.

Bench-scale treatability studies are sometimes performed to substantiate the performance criteria on which ISTR designs are based, especially for novel contaminants or multicomponent systems. Most measure concentration reductions as increasing volumes of steam are generated within or flushed through a sample (USACE 2009). Results from these studies should be interpreted with caution, however, when mass balances are neglected, length scales for fluid transport are unrealistic, or heat losses are substantial. Another robust approach is to perform coupled phase partitioning or flash calculations as heat is injected into the subsurface (e.g., Falta et al. 1992; Elliott et al. 2003; Molnar et al. 2019). Equations-of-state within such models can be parameterized using experimental data (e.g., Chen et al. 2012), chemical databases (e.g., Reid et al. 1987), or group-contribution methods (e.g., Joback and Reid 1987), and can be useful to estimate mass loading for sizing the ex situ treatment or predicting treatment durations. For multi-component systems, experimentally derived steam distillation curves can be used to establish representative pseudo-components for design. Characterization of degradation by-products, corrosion rates, and evaluations of how microbial populations respond to heating are also sometimes performed. However, laboratory testing is most common to measure soil electrical properties for ERH design and to assess whether STAR can self-sustain smoldering combustion.

Pilot-scale ERH and TCH projects are not performed as often as they once were, reflecting the increased predictability and reliability of these technologies over time. For example, in the 1990s, more pilot tests (39) than full-scale ERH or TCH projects (31) were completed. From 2000 to 2009, the trend was reversed, with more than twice as many full-scale projects reported (108) than pilot tests (45). This continued over the next decade (2010 to 2019), with 135 full-scale projects and only 38 pilot tests reported. Although ERH and TCH pilot tests are no longer common, they may still be desired for higher-risk projects with challenging geologic conditions or complex contaminant mixtures. In contrast, for SEE projects, single-well injection tests are still the preferred method for many designers to assess site-specific injection pressures, flow rates, thermal efficiencies, steam override, and impacts of heterogeneity on temperature response. Additionally, 85% of the STAR projects identified on Figure 3 were performed as pilot tests. As a newer technology that utilizes air injection to control the combustion front, pilot testing will likely continue for STAR designs to establish site-specific radius of influence, rates of combustion front propagation, mass destruction, and volatile mass loading to the ex situ treatment system (e.g., Grant et al. 2016; Savron 2018).

Since their infancy, ISTR technologies have incorporated live updating dashboards, remote-access programmable logic controllers, and other digital tools to automate their operation and/or data collection for performance monitoring (e.g., Stegemeier and Vinegar 2001). Given the cost sensitivity of ISTR projects to extended operation periods, real-time data accessibility and automated interpretation are important to quickly identify upset conditions, minimize downtime, recommend adjustments, and predict future performance. As the demand for data analytics and visualization tools has increased, ISTR vendors have improved their own systems to increase automation and improve transparency to enable faster decision-making. Recent research has focused on how to interpret this data to identify asymptotic trends in mass recovery and inform shutdown decisions (e.g., Parker et al. 2017). Coupling ISTR operations with autonomous third-party systems, such as distributed fiber-optic temperature sensors

(Alemohammad et al. 2017) or vapor intrusion monitoring systems (Kram et al. 2019), has also become commonplace, allowing practitioners to improve the efficacy of their own technologies.

Extraction Approaches

For ISTR technologies that rely on contaminant recovery, extraction designs have varied over time and among vendors. Methods have included (i) pressure relief points co-located or adjacent to the energy delivery wells, (ii) soil vapor or multiphase extraction wells at the centroid of the nearest energy delivery points in a modified four-spot pattern, and/or (iii) horizontal vapor recovery wells in the vadose zone. It is recognized that in dense non-aqueous phase liquid (DNAPL) water systems, gas first develops in the saturated zone at immiscible fluid interfaces, and vapor ganglia that mobilize towards capture points are influenced by subsurface heterogeneity and local thermodynamic condition; as a result, contamination can temporarily redistribute within the treatment volume as heating progresses (Hegele and Mumford 2014; Munholland et al. 2016). Additionally, once the DNAPL is depleted, some vapors may become trapped until steam is generated at a critical saturation sufficient to reinitiate mobilization (Hegele and Mumford 2014). To overcome these challenges, practitioners have learned to use permeable filter packs at the energy delivery points and/or install multiphase extraction wells that penetrate the entire depth interval of treatment, on a tight enough grid spacing to intercept gas that may pool beneath capillary barriers (Martin and Kueper 2011), while ensuring that the treatment volume is completely heated. Some also advocate using a hot floor or bottom-up approach for heating if DNAPL is anticipated to develop vertically along condensation banks, although the co-injecting air has also been shown to be effective for SEE (Kaslusky and Udell 2005). Multiphase extraction is also often used when NAPL is mobile or may become mobile during heating, although it is advisable to minimize excessive groundwater production, which given its relatively high heat capacity, can lead to energy penalties.

Design rates to provide overall pneumatic and hydraulic control are often based on previous projects in similar lithologies and may target a specific ratio of condensable and non-condensable vapors (e.g., 50:50), a range of vadose zone pore volume exchanges (e.g., 5 to 15 per day), a minimum ratio of groundwater extracted versus water dripped into electrodes or injected through steam spears (e.g., above unity), or flow rates necessary to achieve inward pneumatic and hydraulic pressure gradients. Some practitioners advocate installing an engineered permeable vapor collection layer at ground surface to provide low-vacuum, high flow rate conditions for sweeping vapors into horizontal wells, while others aim to achieve a higher vacuum or radius of influence around vertically screened extraction points. Both approaches are aided in lower-permeability media by pneumatic fracturing caused by in situ steam generation, which accelerates vapor recovery and drops interstitial fluid pressures so that boiling is not suppressed (Liu et al. 2014). Because of these mechanisms, it is difficult to pilot test extraction designs. Instead, pneumatic and hydraulic control are often verified during operations by measuring pressure gradients or soil vapor concentrations at monitoring points installed within and around the treatment volume, recognizing that pressures may develop locally near energy delivery points. Thermally insulating vapor caps have also seen widespread adoption by most vendors over the past 10 years, as they provide a no-flow boundary condition to minimize short-circuiting or fugitive emissions, limit infiltration, and improve thermal efficiencies (Nielsen et al. 2014). Additionally, pulsed operations and pressure cycling continue to be regularly employed during the later stages of operations to redistribute stagnation points and accelerate vapor phase partitioning (e.g., Davis 1998).

Fractured Rock Applications

Of the five generalized geologic scenarios identified in Triplett Kingston et al. (2008), ISTR projects in the 2000s were largely implemented in lower-permeability or heterogeneous surficial sediments, and only a handful of pilot tests were attempted in weathered or fractured bedrock. Although early SEE projects in fractured rock at Edwards Air Force Base Site 61 and Loring Quarry recovered appreciable quantities of contaminant mass, electrical resistance tomography data collected during operations confirmed that steam propagated only along discrete fractures and was insufficient for heating rock matrices with low permeability or fracture interconnectivity (Davis et al. 2005). TCH was subsequently demonstrated in saprolite and fractured gneiss in Heron et al. (2008) and successfully recovered approximately 12,000 pounds of trichloroethene while reducing 95% upper confidence limit concentrations to 17 micrograms per kilogram (μg/kg). Further studies characterized several fundamental processes, such as cooling through transmissive fractures and percolator-like steam drive effects at the wellbore (Lebrón et al. 2012), or boiling suppression in the primary porosity (Baston et al. 2010), that were later used to optimize designs. Today, TCH has been successfully implemented at several crystalline bedrock sites, with unit costs of $175, $210, and $230 per cubic yard in three recent applications larger than 15,000 cubic yards (LaChance 2020). The deepest reported TCH project to date treated chlorinated impacts in fractured granite up to 150 feet below grade in Varberg, Sweden, though we believe there to be no practical depth limitation, as downhole electric heaters thousands of feet long have been used in the oil and gas industry (e.g., Penny et al. 2019). ERH has also been effectively used for heating sedimentary bedrock at more than 10 sites (e.g., CDM Smith 2018), including applications in limestone epikarst (Beyke et al. 2014) and sandstone up to 300 feet below grade (McGee 2013). However, ERH is not as effective for heating igneous or metamorphic rocks, as their bulk resistivities are often too high to induce resistance heating within the formation. In contrast, dielectric heating mechanisms can prevail during RFH and may allow electromagnetic fields to propagate in bedrock with little water content (Vermeulen et al. 1979; Vermeulen and McGee 2000). In the current data set, at least two bedrock RFH projects were identified, which both targeted temperatures of approximately 60 to 80°C to accelerate degradation via hydrolysis.

Combined Thermal Technologies

Combined ERH/SEE remedies to simultaneously heat interbedded strata of lower and higher permeability were first performed at the DUS demonstration at LLNL (Newmark and Aines 1995) and subsequently at the Young-Rainey STAR Center (Heron et al. 2005). In the present review, combined ERH/SEE and TCH/SEE remedies were identified in at least 22 projects. Most used SEE to deliver power more rapidly into permeable layers or to manage convective cooling associated with rapid groundwater fluxes, which can be detrimental to ERH or TCH performance (Hegele and McGee 2017). Steam spears can be installed relatively easily after start-up to deliver energy into recalcitrant areas, especially if a steam generator is already onsite for activated carbon regeneration. At some sites, practitioners prepared contingency steam spears by nesting them with energy delivery points or finishing them as temperature monitoring points. In addition to these steam-supported implementations, one ERH/TCH project was identified in the data set. As most vendors now offer multiple ISTR technologies, we foresee the increased use of combined thermal remedies in the future.

Multi-Component Remedial Strategies

With at least 643 installations over the past 31 years, ISTR has consistently proven itself to be a reliable and aggressive source zone treatment technology that is capable of successfully achieving a high degree of mass removal in a relatively short duration (e.g., Triplett Kingston et al. 2010; Kueper et al. 2014; Baker et al. 2016). ISTR has proven to be effective, and in many cases preferred, for surgically treating high-mass NAPL source zones in heterogeneous and lower-permeability settings as the subsurface properties that govern heat propagation are more uniform than the intrinsic permeabilities controlling the effectiveness of many other extraction or fluid-delivery based technologies (USEPA 2014). As mentioned previously, technological advances in energy delivery and extraction methods, along with an array of engineering and institutional controls during operations, have made ISTR a flexible and adaptable approach that can effectively navigate a variety of site conditions (e.g., below the water table, beneath occupied buildings, at substantial depth) and be readily scaled to match the extents of the source zone that is being targeted.

ISTR is most effective when it is not considered as a standalone remedy to accomplish the entire spectrum of potential remedial objectives, but rather as an important piece of a holistic remedial approach or long-term risk management strategy. Combined or multi-component remedial approaches, especially in lower-permeability or heterogeneous settings where ISTR is often implemented, should be focused on risk reduction and control, and inherently be flux-focused. With a clear understanding of mass flux emanating from a particular source zone, the connection of performance objectives to specific remedial components will provide an appropriate level of remedy selection and help to establish the aggressiveness, duration, sequencing, and endpoints of each part of the treatment train. For the ISTR portion, these performance objectives narrate ISTR shutdown criteria and should involve clear targets, such as NAPL removal, mass reduction targets, or particular reductions in soil and/or groundwater concentrations, prior to implementation. Once these shutdown criteria are met, the resulting mass flux discharging to the surrounding plume should be at a decreased rate, such that other remedial approaches like directed groundwater recirculation (DGR), in situ chemical oxidation (ISCO), enhanced in situ bioremediation (EISB), or monitored natural attenuation (MNA), are more cost-effective and feasible for implementation. Even without the addition of amendments, ISTR is so adept at driving contamination out of lower-permeability strata and mitigating back-diffusion that concentrations in the downgradient plume have repeatedly been observed to decrease substantially after thermal operations are suspended (e.g., Baker et al. 2016; Heron et al. 2016). For instance, Baker et al. (2016) presented data from 10 separate DNAPL source areas at five sites where post-ISTR groundwater concentrations were reduced by 2 to 4 orders of magnitude and downgradient concentrations achieved drinking water standards in approximately 5 to 10 years, allowing the existing pump-and-treat systems to be shut down early. Additionally, most of the vendors interviewed for this column reported similar observations across multiple sites over the past decade.

Accelerated rates of biotic and abiotic degradation mechanisms, including hydrolysis and oxidation, have also long been recognized as valuable by-products of subsurface heating (e.g., Dablow et al. 1995; Powell et al. 2007; Truex et al. 2007; Suthersan et al. 2012), and are increasingly being leveraged as part of combined source zone or heat-enhanced plume remedies. Elevated temperatures may persist up to two years after thermal operations are terminated (Krauter et al. 1996), and in addition to accelerating dissolution and reaction kinetics in the source zone, may also increase the release of electron donors via the hydrolysis of humic and fluvic acids (CDM Smith 2018) and/or downgradient microbial populations capable of degrading contaminants (e.g., Thompson et al. 2018). For instance, sustained temperature increases of 6 to 10°C were recently observed two years after TCH operations were suspended and 225,000 kilograms of volatile organic compounds (VOCs) were removed at the SRSNE Superfund Site in Southington, CT, and associated qPCR analysis demonstrated a greater diversity and abundance of chlorinated and benzene, toluene, ethylbenzene, and total xylene (BTEX) degraders downgradient (Thompson et al. 2018). For design, practitioners can perform heat transfer calculations to identify regions of the subsurface susceptible to accelerated reaction kinetics, and couple the results to literature, laboratory-derived, and/or field-measured degradation rates. “Low-temperature” ERH and TCH designs, on wider grid spacings, have also been used to accelerate EISB, zero-valent iron (ZVI), and hydrolysis reactions without active extraction (e.g., Macbeth et al. 2012; Singer 2014; Smith et al. 2017). However, given the similar power delivery infrastructure and longer time frames necessary to evenly heat the subsurface on a wider grid spacing, the costs of heating all the way to boiling may be only incrementally higher. In contrast, renewable or self-sustaining low-temperature technologies (e.g., Horst et al. 2018) may be a competitive option for long-term heating with minimal operating costs. Either way, these heat-enhanced approaches can accelerate time frames for remediation and are best incorporated in the plume management approach upfront when initially evaluating ISTR design and performance criteria. A depiction of a combined approach as such is presented on Figure 6.

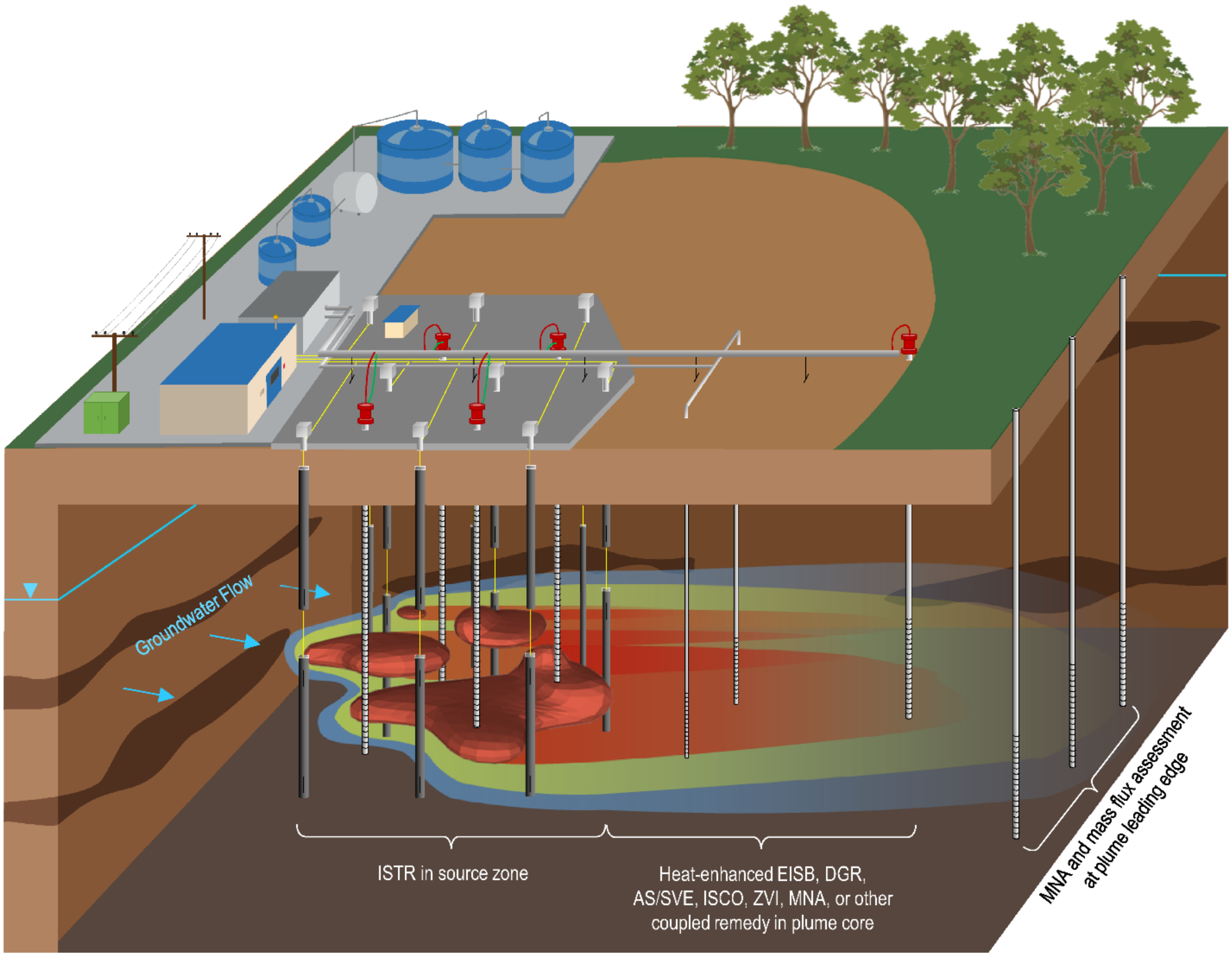

Figure 6. Combining ISTR in a DNAPL source zone with a heat-enhanced remedy in the plume core.

Leveraging High-Resolution Site Characterization

At sites with groundwater plume restoration objectives, failure to adequately treat the source(s) will hamper or frustrate achievement of groundwater-specific remedial objectives. Previous authors have concluded that poorly defined source zones can lead to ineffective thermal remedies, where the heated volume did not adequately address regions of high mass (Johnson et al. 2009) or untreated contamination upgradient re-contaminated the ISTR volume (USEPA 2014). This may have partly been a consequence of the characterization tools available at the time, which primarily consisted of visual observations or measurements at soil borings or monitoring wells; inferences from soil, groundwater, and soil vapor concentrations at partitioning thresholds; or hydrophobic dye testing. These methods can be favorable for delineating light non-aqueous phase liquid (LNAPL) smear zones, but may not be as effective for fully capturing the complex architecture of DNAPL source zones. When designing a multi-component remedial strategy, it is imperative that source zone, plume core, and plume edges are clearly defined. In the context of ISTR, NAPL source zone geometries are often used to establish the boundaries of the ISTR treatment footprint, and when combined with corresponding mass estimates and remedial objectives, are usually the primary driver for implementation costs. As a result, pre-design investments that further refine the spatial distribution of NAPL will attain a higher level of certainty that ISTR will achieve remedial objectives. High-resolution site characterization (HRSC) tools, including direct-push hydraulic profiling tools (HPTs), optical screening tools such as the traditional or dye-enhanced laser induced fluorescence (LIF) systems (e.g., Einarson et al. 2018), membrane interface probe (MIP) systems, and other geophysical tools are now widely available for improving source zone delineation.

These systems allow for 3D modeling and visualization of the geology and NAPL distribution, which provides increased confidence that the ISTR footprint encompasses the entire NAPL source zone and will not leave behind areas with significant mass that may continue to diffuse to the surrounding plume. Additionally, by defining a clear boundary between the source zone and plume core, reasonable ISTR performance objectives centered around mass removal or soil concentration metrics can be established, such that low levels of contaminant mass being pulled into the ISTR system from outside its footprint do not unnecessarily prolong thermal operations.

HRSC tools can also identify heterogeneities in subsurface permeabilities, and more importantly, zones of relatively higher groundwater fluxes, much more discretely than traditional methods. From an ISTR design standpoint, being able to confirm depth intervals where higher groundwater fluxes may be present can be useful for evaluating whether engineering controls or a steam-supported approach is warranted (e.g., Hegele and McGee 2017). Additionally, the depth-discrete characterization of intrinsic permeabilities, in combination with the use of HRSC screening tools, can provide an in-depth understanding of mass flux zones across a site. This enables post-ISTR monitoring programs to better monitor mass discharge and be specifically aligned with flux-focused remedial goals (e.g., Horneman et al. 2017). Although many new flux-focused characterization technologies have been developed in recent years, remediation system effectiveness continues to be assessed primarily based on contaminant concentrations measured at conventional monitoring wells. With a higher degree of source zone removal certainty achieved by leveraging HRSC to increase the certainty of ISTR performance, regulatory viewpoints may shift towards mass flux reduction targets within a multi-component remedial strategy. This in turn will lead to a greater client willingness to invest upfront capital for reduced risk and to potential return-on-investment opportunities in the long term.

Sustainability and Resilience

Sustainable remediation is typically evaluated based on the environmental, social, and economic impacts associated with the lifecycle of a remedy. ISTR as a standalone application is often not considered a sustainable remediation treatment technology due to material consumption for construction and the large energy demand required for operation. However, comparing the full lifecycle of the project entirety, combining ISTR with other technologies may minimize material input and significantly reduce energy consumption. For instance, coupling various technologies can balance applications with large environmental impacts in areas where the benefit outweighs the use (i.e., within source zones) with less impactful (passive) technologies for plume treatment. Additionally, combined treatment can leverage the symbiotic conditions created from ISTR to enhance the benefits from hydrolysis reactions, biological degradation, or even use of residual heat to benefit reactions induced through additional amendment delivery (e.g., ISCO or EISB). Finally, the social (reduction in treatment time frame, restoration to beneficial reuse, stakeholder concerns) or economic impacts of implementing ISTR often outweigh the environmental impact considerations. We refer the reader to Vidonish et al. (2016) and Ding et al. (2019) for further discussion of how the physiochemical properties of soil respond to heating and the opportunities for sustainable land reuse following thermal treatment.

Several options for reducing environmental impacts and improving sustainability associated with ISTR are available through Sustainable Best Management Practices (SBMPs). Guidance for implementation of SBMPs is publicly available from USEPA, the Interstate Technology and Regulaotry Council (ITRC), and Naval Facilities Engineering Systems Command (NAVFAC). Additional standard guidelines have been published by ASTM International on Green Remediation (E2893) and Sustainable Remediation (E2876). Following the guidelines to implement a sustainable project, early management can benefit the project with engagement from stakeholders and identification of project risks and costs. Many of the in situ thermal remediation SBMPs coincide with optimization considerations (e.g., Lemming et al. 2013). There are a few SBMPs that should be considered based on site-specific conditions because, although they might result in a smaller environmental footprint, they might also affect the performance of the remedy. An example is the USEPA SBMP that states "Consider co-locating electrodes and recovery wells in the same borehole, particularly in the saturated zone, to minimize land disturbance." Vendors often use co-located wells for pressure relief and to reduce implementation costs; however, some site conditions may warrant the installation of dedicated recovery wells at the centroids between energy delivery points to drive the sweeping of fluids throughout the treatment volume. Another example is the NAVFAC SBMP that stipulates "Use of a phased approach that sequentially heats large sites, to reduce energy and equipment use." Perimeter heat losses play an important role in thermal efficiency, and if a large source zone has been delineated, it may be best to heat the entire volume at once, as long as the utility service can satisfy the power demand. Also, some SBMPs such as the NAVFAC SBMP "Evaluate and compare the estimated carbon footprints when selecting the heating technology" may lead to development of technology innovations for improved energy efficiency. These carbon footprint evaluations can be completed with publicly available tools such as SiteWise (NAVFAC) or Spreadsheets for Environmental Footprint Analysis (SEFA, USEPA) and provide insight to the heating strategy selection. When performing these evaluations, it is important to include any planned SBMPs, such as the use of gas-fired TCH burners with biofuels or extracted hydrocarbons as supplemental fuel source, or the recycling and pre-heating of treated groundwater for electrode reinjection water during ERH. Additionally, improving delineation to reduce ISTR treatment footprints and using lower-temperature approaches in the plume core may allow supplemental wind and solar energy-based or waste heat recycling technologies to be used. Smoldering-based technologies such as STAR can also provide energy-efficient solutions for constituents with a suitable BTU value. For example, Scholes et al. (2015) showed the energy required for ignition was approximately 1.1 kilojoule per kilogram of soil remediated, while maintaining removal efficiencies above 97% (Ding et al. 2019). Understanding the implications of implementing SBMPs should be gained by continuing engagement of stakeholders and confirmation of alignment with establish performance metrics for the site.

Finally, with the increased importance of resiliency in remediation, ISTR provides the complete destruction of constituents of concern, limiting the potential for future concerns related to physical or chemical changes over time, such as flooding in coastal areas or geochemical shifts over time that may result in changed/worsened conditions or lowered rates of attenuation. Similarly, climatological changes over decades can drastically change the storage and velocity of groundwater in an aquifer, thereby altering the groundwater plume in ways that would not be expected based on a present-day evaluation of flux. Another benefit is with an alternative that has a short implementation window, the treatment time can be targeted during low-risk periods for extreme weather and the results of extreme events such as power loss, extreme winds, fire, or flooding. Even in a combined remedy situation, the technical team could easily implement a resilient solution with a short window of "high-risk" activities that involve equipment and infrastructure above ground, moving to more passive "low-risk" activities involving either infrequent injections with equipment mobilized for the event or passive monitoring for natural attenuation.

Emerging Contaminants

Although early thermal projects were successful in remediating coal tar, creosote, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs) (Vinegar et al. 1997; Stegemeier and Vinegar 2001), ISTR initially solidified its place in the market as a resilient and cost-effective approach for remediating high-mass chlorinated solvent and petroleum source zones where excavation was impractical or expensive. As ISTR technologies have matured and their use has propagated globally, however, the diversity of contaminants for which they are being used to remediate has also grown. In recent years, ISTR has experienced a resurgence at manufactured gas plant and wood treating facilities, and has been used to target plasticizers, pesticides, dioxins, furans, chlorophenols, nitrosamines, chloroalkyl ethers, methacrylates, energetic compounds, dyes, and perchlorates, among others. Treatment of mercury-related compounds has also been demonstrated using TCH both at the laboratory scale with average reductions of 99.8% to a concentration of 22.4 mg/kg (Kunkel et al. 2006), and at the field scale with average reductions of 99.6% to 48 mg/kg (Hilbert et al. 2019).

ISTR can also be effective for treating 1,4-dioxane impacts in soil and groundwater, though given its relatively low Henry's law constant, a substantial amount of groundwater must be vaporized to effectively reduce its aqueous-phase concentrations in the saturated zone. For instance, Oberle et al. (2015) showed greater than 88% reductions of 1,4-dioxane concentrations in soil by boiling off roughly 45% of the interstitial water. By comparison, a 30% boil-off target is more typical for reducing chlorinated solvent DNAPL source zones to below 1 mg/kg (Heron et al. 2013). For feasibility and design assessments, boil-off targets necessary to achieve specific concentration reductions can be modeled with the equations of Udell (1996). The need for specialized liquid treatment equipment to manage 1,4-dioxane during ISTR can also be reduced or mitigated by using insulated well field piping with duct heaters to minimize condensate production. Similarly, vadose zone applications using hot air injection have also shown promise, with Hinchee et al. (2018) reporting 94% reductions in 1,4-dioxane following 14 months of operation in a recent field study.

Thermal treatment of per- and polyfluoroalkyl substances (PFAS)-impacted soils has also been proposed via direct volatilization (Crownover et al. 2019) and smouldering combustion with a surrogate fuel (Duchesne et al. 2020). Preliminary laboratory studies have measured reductions of PFAS soil concentrations of 99.91% and 99.998% via volatilization when heated to 350°C and 400°C, respectively (Crownover et al. 2019). Continued research is ongoing at the pilot-scale to demonstrate both remediation performance in the vadose zone (Irey 2020) and the effective PFAS mineralization and/or management of any potential fluorinated by-products (e.g., Horst et al. 2020). The cost-effectiveness of using ISTR for treating PFAS source zones, which may be relatively dilute due to their mobility in the environment, will become clearer as the broader market for PFAS remediation technologies develops.

Summary and Outlook

In the 13-year period that has followed the original survey by Triplett Kingston (2008), it is safe to say that ISTR has become a standard option in the remediation toolkit, and has continued to evolve in many ways:

- Implementation is more flexible and adaptable, with the supporting science and vendor experience having been enhanced to provide a higher degree of certainty regarding treatability.
- Project implementation risks have been reduced through vendor experience, real-time data acquisition and interpretation, and tools available to ensure treatment of the correct volume.
- An array of new technologies associated with ISTR are available, creating more options for remediation practitioners and owners, including additional thermal technologies such as STAR, gas-fired burners, induction heaters, and helical heating coils, as well as monitoring technologies such as temperature monitoring with fiber optic cables.
- ISTR has more often become one part of a holistic multi-component strategy where source removal creates other opportunities for treating mass outside of the source.
- The above combined with the expanded definition of sustainability and the importance of remedy resilience has increased the likelihood that ISTR can be leveraged in combination with other technologies to create a remedy with an ideal sustainability profile. This is an important evolution for a technology that originally was thought to be a non-sustainable option.

One consideration that was true in 2008 that remains true today is that the characterization of the thermal treatment volume must have the right level of accuracy for the technology to achieve desirable endpoints. Most of the treatment failures are related to incomplete or insufficient characterization relative to the contaminant distribution, geologic or hydrogeologic settings, or thermodynamic characteristics of the contaminants.

Though unit costs have remained relatively similar over the years, the reliability of the technology has improved and the projects for which it is being targeted have become more complicated, with implementations at higher mass sites, in more complex geologies, at greater depths, beneath surface obstructions, or near active utilities. That said, practitioners and vendors are more inclined to divide work scope with regards to implementation and operation activities, which has ultimately provided increased opportunities for risk sharing and allowed for more efficient implementation.

In the future, as project sizes and complexity increase and higher value is placed on physically distanced remediation methods, we expect ISTR will continue to increase in prominence. This line of technologies is one that should be at the forefront and that every remediation practitioner should be aware of as a tool to support the clients and communities they serve.

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