Integrated Systems-Based Approach for Reaching Acceptable End Points for Groundwater - 13629

M. Hope Lee[1], Dawn Wellman[1], Mike Truex[1], Kent S. Sorenson, Jr.[2], Ryan Wymore[2], and
Mark D. Freshley[1]
[1]Pacific Northwest National Laboratory, 902 Battelle Blvd, Richland, WA, 99352, USA
[2]CDM Smith, Camp Dresser & McKee Inc., 555 17th Street, Denver, CO 80202, USA

ABSTRACT

The sheer mass and nature of contaminated materials at DOE and DoD sites, makes it impractical to completely restore these sites to predisposal conditions. DOE faces long-term challenges, particularly with developing monitoring and end state approaches for clean-up that are protective of the environment, technically based and documented, sustainable, and most importantly cost effective. Integrated systems-based monitoring approaches (e.g., tools for characterization and monitoring, multi-component strategies, geophysical modeling) could provide novel approaches and a framework to (a) define risk-informed endpoints and/or conditions that constitute completion of cleanup and (b) provide the understanding for implementation of advanced scientific approaches to meet cleanup goals.

Multi-component strategies which combine site conceptual models, biological, chemical, and physical remediation strategies, as well as iterative review and optimization have proven successful at several DOE sites. Novel tools such as enzyme probes and quantitative PCR for DNA and RNA, and innovative modeling approaches for complex subsurface environments, have been successful at facilitating the reduced operation or shutdown of pump and treat facilities and transition of clean-up activities into monitored natural attenuation remedies. Integrating novel tools with site conceptual models and other lines of evidence to characterize, optimize, and monitor long term remedial approaches for complex contaminant plumes are critical for transitioning active remediation into cost effective, yet technically defensible endpoint strategies.

INTRODUCTION

The U.S. Department of Energy (DOE) oversees some of the largest environmental cleanup operations in the world. For more than 50 years the United States created a vast network of facilities for research and development, manufacturing, and testing of nuclear materials, leaving an enduring legacy of over 6 billion cubic meters of contaminated soil and groundwater in 29 states [1]. Subsurface contamination is present at more than 7,000 known sites and over 100 facilities across the nation with more than half of these containing metals and radionuclides and many with chlorinated hydrocarbons [1]. In addition to these known wastes, there are unknown quantities of waste buried across the nation. Innovative solutions, based on scientific understanding of subsurface processes, are needed to remediate, manage and monitor these various contaminated sites [2]. These cleanup efforts present an enormous technical, scientific, and financial challenge for the U.S. Department of Energy Office of Environmental Management (EM), the U.S. Department of Defense (DoD), and the nation as a whole. While technologies exist for dismantling/decommissioning surface structures, contaminants that have entered the subsurface are difficult to remove and/or remediate, especially for those contaminants whose toxicity and persistence require removal to very low levels. The anticipated cost to complete soil and groundwater remediation across the DOE complex ranges from \$17.3 billion to \$20.9 billion [3] and the estimate for DoD is \$33B [4].

Chlorinated solvents are a large contributor to the subsurface contamination problem facing DOE and DoD. Eight of the top 20 contaminants detected at hazardous waste sites on the National Priorities List are chlorinated solvents, including trichloroethene (TCE) and tetrachloroethene (PCE), which are first and third, respectively. DoD alone has approximately 3,000 sites contaminated with chlorinated solvents [5]. Many of the remaining DoD and DOE sites which are contaminated with chlorinated solvents are complex (range of conditions: deep fractured rock, vadose zones, large, oligotrophic, dilute with respect to concentration of contaminants ($< 1000\ \mu g\ L^{-1}$)), and in many cases, comingled with other contaminants (e.g. 1,1,1-trichloroethane (TCA), 1,4 dioxane, and methyl-tert butyl ether (MTBE), metals, andradionuclides). These complex plumes are notoriously difficult and costly to treat by an active remediation method such as pump and treat (PNT). Chlorinated solvents in complex subsurface environments, specifically TCE as a dense nonaqueous phase liquid (DNAPL) in deep fractured rock, is recognized to be one of the most difficult challenges in groundwater remediation [6].

Once organic contaminants, such as chlorinated solvents, have migrated into the subsurface, one of the most cost- and timely ways to remove them is through a strategy known as bioremediation [7-10]. Bioremediation is based on the exploitation of *in situ* metabolic potential of subsurface microbes to attenuate the toxic effects of contaminants by transforming them to lesser toxic products, completely mineralizing, or immobilizing them [11-13]. Most living organisms possess some ability to detoxify contaminants, however microbes have shown the greatest potential [14, 15] to be manipulated, directly or indirectly, and to provide potential cost-effective strategies. In many instances, biological degradation of contaminants offers solutions in previously intractable cases where there was “no solution at any price”. In general, the overall success of these strategies relies heavily on the relative abundance, structure, catabolic versatility, and biotic/abiotic interactions of the microbial communities that are present at the site, whether indigenously, stimulated, or augmented [16-19]. Bioremediation strategies and methods targeting the characterization of degrading populations have been implemented at hundreds of contaminated sites across the country and internationally (DoD, DOE, industry, private; [20-24]). Currently there is a diversity of molecular methods available for identifying and characterizing the microbial community in any given groundwater, surface water, sediment or soil sample. These tools have significantly increased the efficiency and success of the remediation processes and have significantly decreased the overall cost of treatment/removal both short- and long-term when compared with more aggressive treatment technologies (removal, pump-and-treat, thermal, chemical) [25-28]. While it is generally accepted that bioremediation is an effective strategy, monitoring performance and assessing long-term potential or continued removal/degradation, is an essential metric for both site owners (DOE, DoD) and regulators, particularly when these approaches have been instrumental in achieving an alternative endpoint.

The challenging nature of remediating complex sites makes approaches involving combinations of different remediation strategies attractive including those that rely on natural attenuation. The underlying technical limitations as well as evaluation of costs and benefits of removing residual contamination drive remediation decisions toward consideration of alternative endpoints [29]. The approach described herein presents a framework to achieve risk-informed endpoints for remediation, an approach that is being considered by DOE in collaboration with DoD and the Environmental Protection Agency (EPA) through participation in the Federal Remediation Technologies Roundtable. The concept of endpoints will be described and an example where a multicomponent remediation strategy including alternate endpoints has been successfully implemented at the Idaho National Laboratory (INL) will be described.

DISCUSSION

Alternate Endpoints

An **end state** is a standards-based cleanup objective associated with closure of a waste site and/or long-term management that is permitted by regulation and is protective of human health and the environment. It is the final product of a remediation or management scenario. A familiar example of an end state is a condition where contaminants at a site are at or below the maximum concentration limits (MCL) established by regulation for contaminants in drinking water. An **alternate endpoint** is a risk-informed remediation goal permitted by regulations that is protective of human health and the environment. The concept of an **alternate endpoint** enables establishing a path for cleanup that may include intermediate remedial milestones and transition points and/or regulatory alternatives to standards-based remediation. Alternate endpoints can be used to determine technology development needs as described in Dettmers et al. [30] for a complex site at INL.

Current end states and requirements for site remediation and closure are generally standards-based. This approach leads to remediation goals that often are overly conservative, costly—and in some cases—technically impractical to achieve. There is growing recognition that there are a number of complex sites where active remedies will not be successful and alternate endpoints will be required [31]. There are multiple currently acceptable alternate endpoints that apply to groundwater [29] including attenuation approaches, adaptive site management, groundwater reclassification, alternate concentrations, and Applicable or Relevant and Appropriate Requirements (ARAR) waivers. Attenuation approaches include monitored natural attenuation (MNA guidance; [32]) and enhanced attenuation (EA; [33]) that are implemented based on robust conceptual models with adequate site characterization, long-term monitoring, and limited active remedies. Attenuation is important to consider in most remedial strategies for distal portions of a plume or remnant contaminants from an active remedy. EA involves either source reduction or actions to enhance the attenuation rate to stabilize or shrink a contaminant plume. Adaptive site management involves an iterative approach, with actions implemented over time in response to site conditions. Groundwater reclassification involves regulatory changes so that groundwater at a site is no longer designated as drinking water. Alternate concentration limits replace or modify cleanup standards; for example, where contaminated groundwater discharges to surface water. ARAR waivers are used where compliance with a regulatory limit is technically impractical.

The process of defining and implementing alternate endpoints is risk informed. This decision process is based on analysis of the potential for a contaminant to cause immediate and/or long-term harm to a receptor resulting from exposure and the likelihood of this occurrence. Comparable to end states, alternate endpoints must be scientifically and technically defensible and based on systematic, objective understanding of the contamination issue and impact of proposed solutions to provide justification for the site remediation decisions.

Alternate Endpoints Framework

Fig. 1 presents a systems-based framework for implementing remediation at a site where an alternate endpoint is expected. The framework provides a means to define the nature and extent of the problem to determine which risks are most critical and establish alternative endpoint cleanup decisions. The framework is based on a rigorous site conceptual model in conjunction with assessing risks and potential endpoints as part of a systems-based assessment that integrates site data with scientific understanding of processes that control the distribution and transport of contaminants in the subsurface and pathways to receptors. This systems-based assessment and subsequent implementation of the remediation strategy with appropriate monitoring are targeted at providing a holistic approach to addressing risks to human health and the environment. Goals of the framework are to provide the following:

- Appropriate and necessary insight into the *important* remedial/transport processes
- Platform for integrating new knowledge into flux-based conceptual site models that are significantly more predictive to provide defensible criteria/data for making long-term decisions
- Holistic assessment of risk to human health and the environment
- Flexible approach for application to a range of sites, from simple to complex
- Appropriate path for transitioning to long-term monitoring and stewardship.

Implementation of this framework and alternative approaches provides opportunities for novel remediation approaches such as in situ bioremediation and alternative endpoints such as MNA.

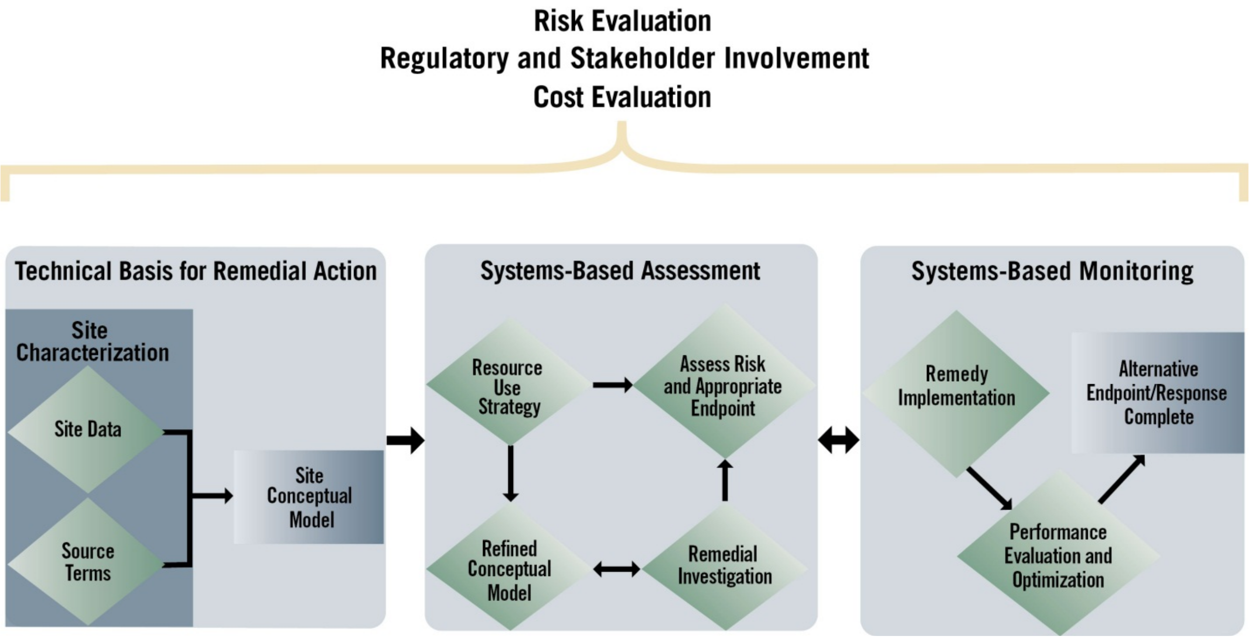


Fig. 1. Systems-based framework for endpoints evaluation

The described framework requires that from a data perspective: (a) source terms and site data are integrated across spatial and temporal scales into a systems-based site conceptual model, (b) iterative evaluations allow for new technologies or approaches to be assessed and incorporated into a revised site conceptual model, (c) risks are holistically evaluated (resource use, groundwater, surface water, future casting for receptors), (d) remedial strategies or interim remedies have measurable goals, and (e) the entire process is optimized based on performance and cost such that an endpoint can be achieved and maintained that is accepted upon by ALL the parties involved.

Test Area North, Idaho National Laboratory

The Test Area North (TAN) site of the INL is the location of a contaminated groundwater site characterized in the early to mid-1990s. TAN brought together several agencies to determine a remediation path forward for chlorinated solvent contaminated site; an agreement was made, as documented in a Record of Decision (ROD), and signed in 1995 and identified pump and treat as the remedial technology. However these agencies recognized that new technologies and approaches would likely allow for a better understanding of the location of sources, the behavior of the contaminants moving through the subsurface, and likely afford better means for remediation and as such documented in the ROD the need for future studies to be conducted to evaluate alternatives.

The TAN site is at the north end of INL (Fig. 2) and was developed for nuclear fuel operations and heavy metal manufacturing. From 1953 to 1972, liquid wastes and sludge from experimental facilities were disposed in an injection well at the site. The subsurface hydrogeology at the site is both deep and complex, consisting of fractured basalt. The wastes were primarily industrial and sanitary waste water, but also included organic, inorganic and low-level radioactive constituents. The historical records provide uncertain estimates on the organic wastes (TCE) that were disposed to groundwater, ranging from as little as 1,325 L to 132,500L [30]. In 1987, TCE and PCE were detected in wells used to supply drinking water to workers at TAN and the groundwater contamination was traced to the injection well. In 1989, INL was included on the National Priorities List, resulting in a Federal Facility Agreement and Consent Order (FFA/CO) between EPA, DOE, and the Idaho Department of Environmental Quality (IDEQ), initiating the Comprehensive Environmental Response and Liability Act (CERCLA) process. A remedial investigation/feasibility study was completed in 1995, which identified the following contaminants of concern: TCE (12,000 – 32,000 ppb), PCE (110 ppb), *trans*-1,2-dichloroethene (*trans*-DCE, 1,300 – 9,000 ppb), and *cis*-1,2-dichloroethene (*cis*-DCE, 3,200 – 7,500 ppb), as well as radionuclides including H-3 (14,900 – 15,200 pCi/L), Sr-90 (530 – 1,880 pCi/L), Cs-137 (1,600 – 2,150 pCi/L), and U-234 (5.2 – 7.7 pCi/L) [30]. The sludge material within the injection well was nearly 3% by weight TCE, serving as a source of contamination to groundwater. The resulting groundwater plume was nearly 3 km long and 0.8 km wide (Fig. 3).

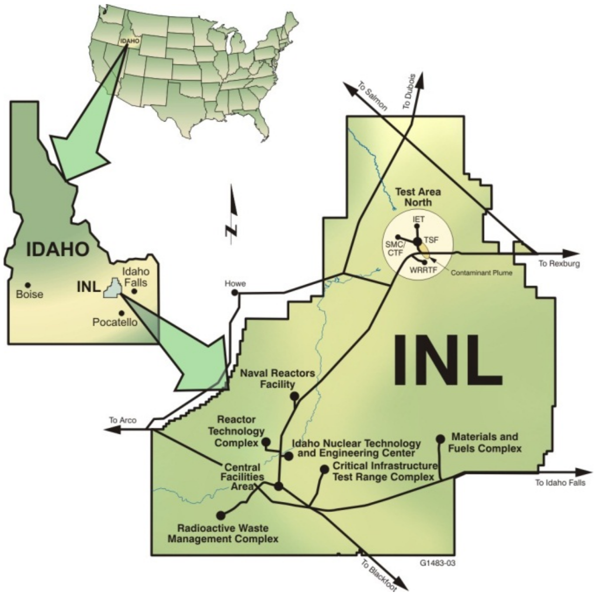


Fig. 2. Location of TAN.

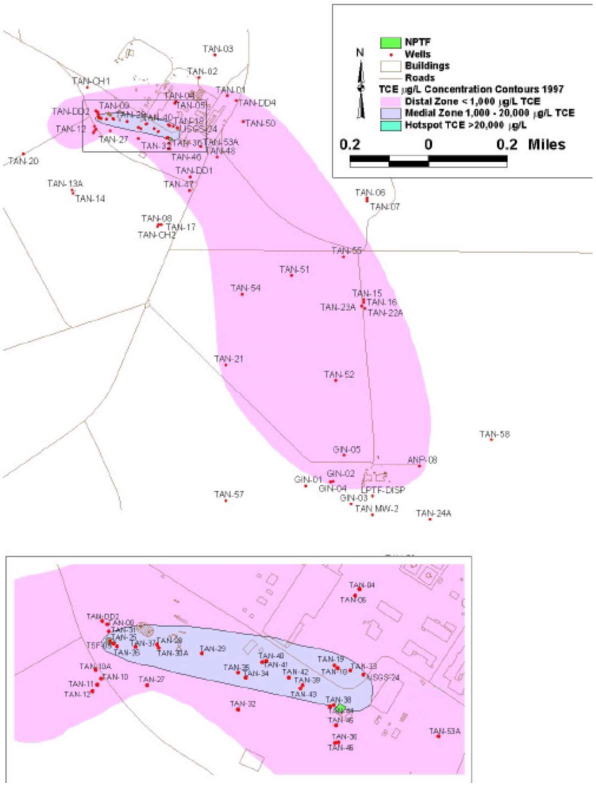


Fig. 3. TCE plume at TAN measured in 1997.

The initial ROD for remediating the TCE plume implemented groundwater pump and treat, with an estimate of 100 years to reach cleanup standards. However, the ROD allowed other treatability studies to be completed for evaluation of alternative remediation technologies and approaches.

Three-Component Remedy

The agencies' agreement to remediate the plume, as documented in a ROD signed in August 1995, identified pump and treat as the remedial technology to be used for restoration of the entire plume, but allowed for treatability studies to be conducted to evaluate alternative remedial technologies. In November 1997, the agencies published an Explanation of Significant Differences [34]. The remediation strategy identified three separate contamination zones and different remediation approaches for each zone [30]. The contamination zone included 1) the source (hot spot) with initial TCE concentrations greater than 20,000 ppb, 2) a medial zone of groundwater contamination with TCE concentrations between 20,000 to 1,000 ppb extending downgradient from the source zone, and 3) a distal portion of the TCE plume with concentrations less than 1,000 ppb. Treatability studies demonstrated that a holistic systems-based approach consisting of three components was most effective: 1) in situ bioremediation (ISB) for the source zone, 2) continued groundwater PNT for the medial zone, and 3) MNA for the distal zone. The three-component remedy was incorporated into a ROD Amendment signed in September 2001 [35].

One critical component of the remedial strategy was the development of a comprehensive site conceptual model (SCM).The key components of the SCM included assessment of the historical activities that resulted in the contamination, the nature and extent of contamination, and the hydrogeologic, geochemical, and microbiological framework that governed contaminant fate and transport [36].

The SCM for TAN was developed through an iterative process of identifying the data requirements needed to understand the parameters described above, identifying the analyses that could provide the required data, and evaluating the quality of the data generated, and identifying data gaps [37, 38].

The source zone was first treated by sludge removal actions, between 1990 and 1998, beginning with the removal of 55 feet of sludge material from the 12-inch diameter casing of monitoring well TSF-05. Following , and as described in the *Field Demonstration Report, Test Area North Final Groundwater Remediation, Operable Unit 1-07B* [39], ISB was determined to be the most likely remedial option to achieve remedial goals and at a cost savings over pump and treat. Source zone treatment of biological degradation of TCE was investigated in a field-scale pilot study providing nutrients to the contaminated aquifer to stimulate biological growth and activity. TCE is susceptible to microbial degradation to ethene under conditions where it serves as a growth-linked electron acceptor under strictly anaerobic conditions, a process known as anaerobic reductive dechlorination (ARD) [30]. A high-concentration electron donor solution consisting of sodium lactate was injected into the source well. During the injections increases in total molar concentrations of contaminants at well locations impacted by the electron donor injections suggested that enhanced mass transfer of TCE from the residual source was occurring as a direct result of the injections [38, 40, 41]. Most important, this newly mobilized TCE was efficiently biodegraded to ethane, as measured in analyses of groundwater from monitoring wells impacted by the injections as well as downgradient. The use of high-concentration electron donor solutions to enhance mass transfer of contaminants into the aqueous phase to facilitate rapid reductive dechlorination and residual source depletion is referred to as Bioavailability Enhancement Technology (B.E.T.™, United States Patent 6,783,678). The use of B.E.T.™ was critical for demonstration that enhanced ISB was a viable option for remediation of the chlorinated solvent residual source area because accelerated mass transfer of contaminants from the residual phase to the aqueous phase makes the contaminants available for biological degradation and significantly shortens the overall remedial time frame [38]. Additional studies have shown that the enhanced ISB strategy also enhanced downgradient biological processes through the addition of bioavailable carbon into the system as well as an additional carbon source, methane, which resulted from ISB operations [42]. The continued optimization of operations, such as evaluation of available electron donors, injection strategies, and advanced monitoring techniques, can lead to substantial life-cycle cost savings. For instance, the switch from sodium lactate to whey powder at TAN is estimated to save over $100,000 annually, and the increased residual source destruction will likely reduce the remedial time frame and increase savings further [36, 38].

The medial zone has predominantly been treated by an air stripper pump and treat system. This zone historically was too large for bioremediation strategies to be effective, and the agencies wanted to ensure protection of human health and the environment. Uncertainties remained with respect to flux of contaminants from the source area, naturally or as a result of treatment, so the pump and treat was the primary means to ensure treatment in this zone. The system includes three extraction wells, an air stripper treatment train, and reinjection into a downgradient well [43]. The water is treated to below regulatory limits for VOCs with the air stripper system. The system was operated from 2001 to 2005, resulting in decrease of TCE concentrations to approximately 100 ppb. A rebound test was conducted from 2005 to 2007 and evaluation of the data from the test suggested that operating the system on a pulsed-pumping strategy through a cycle of operation and standby modes would produce optimal reduction of TCE concentrations [43]. Several rebound tests were conducted over the last 7 years, and the unit has operated primarily in pulsed-pumping operations. The system has shown to decrease contaminant concentrations efficiently; therefore, the unit is operated when breakthrough or other metrics are exceeded. The pump and treat air stripper unit was placed in cold standby on July 28, 2011 [44]. The new pump and treat facility is currently operated several days a week to process purge water and to maintain concentrations below breakthrough (pers. communication).

In addition to the operation of the pump and treat in the medial zone, aerobic biological degradation was shown to be enhanced as a result of the bioavailable carbon in the system and higher than normal concentrations of methane, both resulting from source area strategies [42, 45].

The distal portion of the plume, which included concentrations of up to 800-900 µg L^{-1} TCE, was evaluated for implementation of MNA beginning in the early 2000s. Initially, the attenuation processes were believed to be the result of ARD and dispersion [30]. However Sorenson et al., [46], using the tracer corrected method, determined that TCE was being degraded under aerobic conditions relative to co-contaminants that served as internal plume tracers. The method showed TCE attenuation relative to the co-contaminants, and estimated the aerobic degradation half-life for TCE was between 13 and 21 years [46]. Success of MNA depends on monitoring and observations of a stable or shrinking plume as detailed in the EPAs guidance [32]. Although the plume was not expanding at a significant rate, groundwater analyses showed that conditions in the downgradient portion of the plume were aerobic, suggesting that a mechanisms other than ARD was contributing to attenuation. Lee and colleagues [47, 48] evaluated biological attenuation mechanisms using a novel suite of assays, including DNA, enrichment cultures, and enzyme activity probes, to reveal that TCE was being cometabolized by indigenous microorganisms and significantly contributing to the attenuation of the contaminant. Numerous studies over the past 10 years have supported these early findings and documented the activity and rate of degradation of the microbes in the distal portion of the plume [42, 45, 47-49]. A study by Lee et al., [49] determined a degradation rate and corresponding half-life for TCE that aligned well with other lines of evidence including the tracer-corrected method and contaminant trend data. The distal plume has expanded an insignificant amount in the last 11 years (Fig. 4) but well within the range predicted, and concentrations remain well below limits set in the FFA/CO.

Together these data were used to support the MNA remedy selected for the distal plume as stated in a ROD Amendment signed in September 2001 [35]. The MNA remedy provides an alternative endpoint for the largest portion of the contaminant plume. The cooperation of all of the parties involved coupled with innovative thinking, the ability to iteratively evaluate and modify the conceptual model, optimization of the source area treatment and the pump and treat, and most importantly the evaluation of the site from a holistic perspective allowed the site contractors to successfully define, optimize, and ultimately defend the alternative endpoint strategies.

Overall, the three component remediation approach has effectively reduced concentrations in all three zones of the TCE plume (Fig. 5). Long-term monitoring is continuing but the number of wells has been reduced over the timeframe that the multicomponent remedy has been implemented [43]. The multicomponent approach has resulted in a significant reduction of remediation duration and costs associated with remediation, compared with that projected for the original pump and treat approach, without increasing risks to human health or the environment.

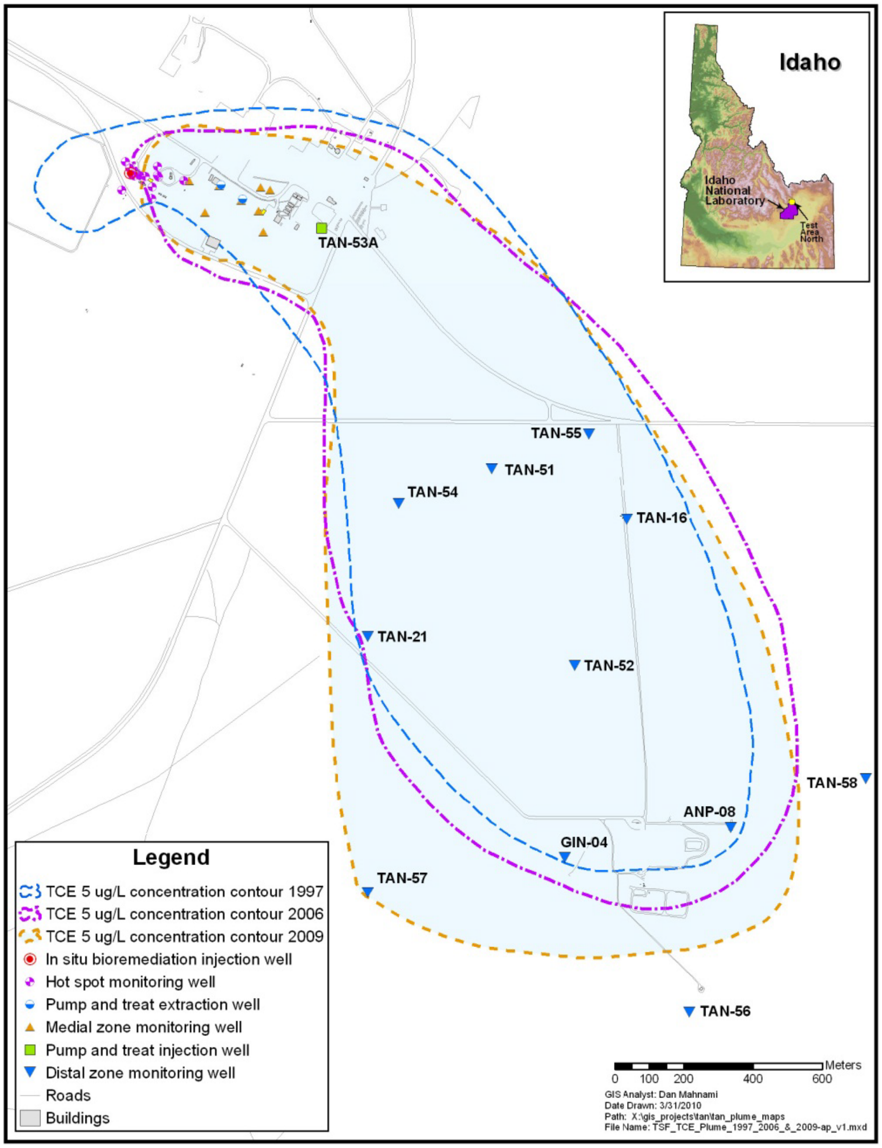


Fig. 4. Comparison of the May 2009, 2006, and 1997 TCE plume (the plume shape is based on the 5-µg/L contour line).

CONCLUSIONS

The current approach of active engineered remediation using a single technology works at “simple” sites to achieve remediation and closure goals, but has proven ineffective at complex sites such as TAN at INL. A systems-based approach of combing innovative remedial technologies and approaches was required at TAN in order to avoid solely relying on the pump and treat strategy which is costly and estimated to take 100 years to receive regulatory goals. The systems-based approach was implemented through cooperation between DOE, the site steward, the site contractor (North Wind and INL), and regulatory agencies (EPA and IDEQ). Novel approaches and collaboration from the onset led to achieving risk-informed end states for the site and ultimately significant cost savings.

Novel science and technology on both enhanced bioremediation in the source zone and natural degradation processes in the distal plume were required. A first of its kind combination of direct and indirect evidence [30, 47, 48], resulted in acceptance of MNA (an alternate endpoint) as a major component of the TAN remedy. Continued performance monitoring and optimization of the pump and treat facility led to several rebound demonstrations, and with the reduction of the source term through ISB, the system was placed into cold standby. This alone saved millions over the proposed lifetime of the system, 100 years. The remaining challenges facing EM are complex and require holistic systems-based approaches that integrate research and understanding between technical areas, and take into account the entire system. The approach used for remediation at TAN provides a template for establishing alternate endpoints for remediation of EM sites.

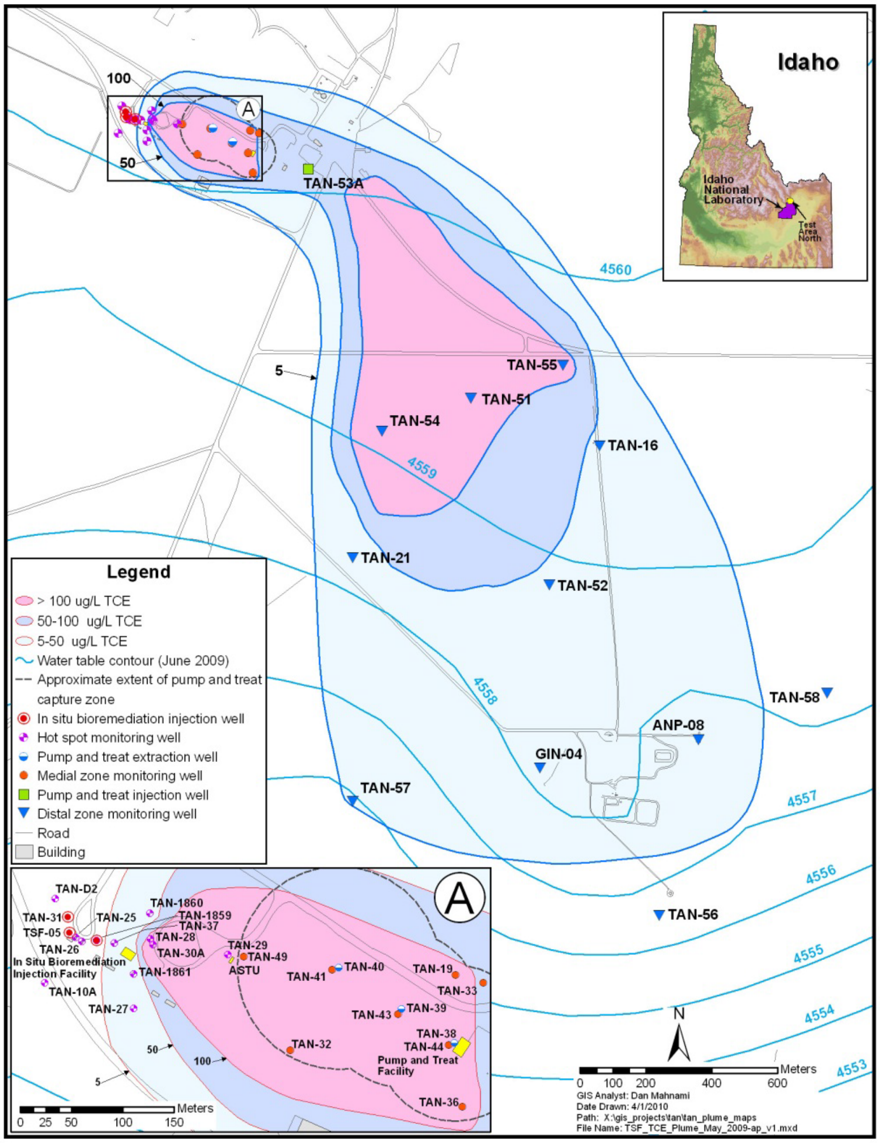
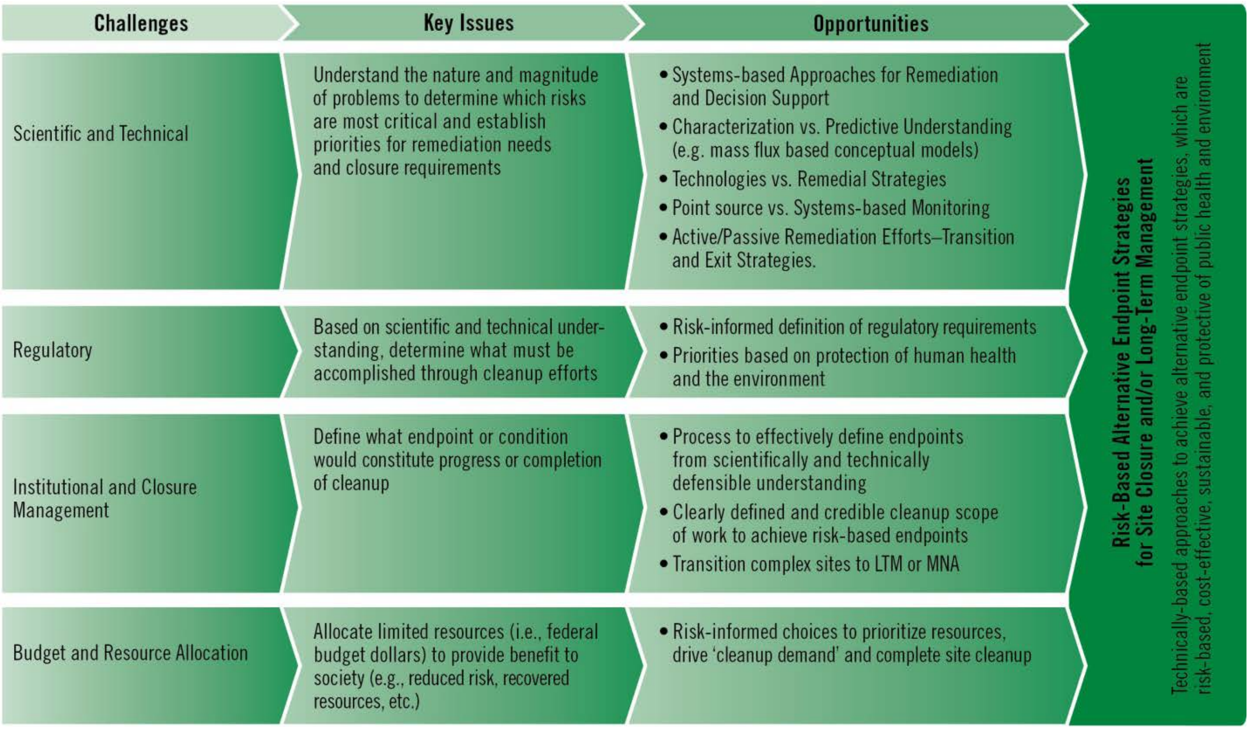


Fig. 5. Distribution of TCE in groundwater at TAN, May 2009 (this plume representation uses data from in situ bioremediation, medial zone, and MNA sampling).

Challenges and Opportunities

There are a number of challenges associated with establishing alternate endpoints such as those implemented at TAN (Fig. 6). Categories of these challenges include scientific and technical, regulatory, institutional, and budget and resource allocation issues. Opportunities exist for developing and implementing systems-based approaches for determining remediation approaches and enabling implementation of alternate endpoints. Characterization, monitoring, predictive modeling, and risk assessments are critical components of the implementation framework. Technology development and evaluation, as well as attenuation-based approaches, are foundational elements supporting the ability to achieve remediation goals and close waste sites using alternate endpoints. Communication with all parties involved is critical for implementation of alternate endpoint approaches. The transition of sites to long-term monitoring and stewardship [50] is also a key component of an alternate endpoint approach. While some development and policy efforts are needed to enable broad implementation of alternate endpoints for EM, the alternate endpoint approach has the potential to expedite cleanup and reduce cost through understanding what should be accomplished through cleanup efforts, what endpoint(s) or condition(s) constitute progress or completion of cleanup, and schedule commitments with defensible and credible technical scopes of work, including clear requirements to achieve risk-informed endpoints.



Challenges	Key Issues	Opportunities
Scientific and Technical	Understand the nature and magnitude of problems to determine which risks are most critical and establish priorities for remediation needs and closure requirements	• Systems-based Approaches for Remediation and Decision Support • Characterization vs. Predictive Understanding (e.g. mass flux based conceptual models) • Technologies vs. Remedial Strategies • Point source vs. Systems-based Monitoring • Active/Passive Remediation Efforts–Transition and Exit Strategies.
Regulatory	Based on scientific and technical understanding, determine what must be accomplished through cleanup efforts	• Risk-informed definition of regulatory requirements • Priorities based on protection of human health and the environment
Institutional and Closure Management	Define what endpoint or condition would constitute progress or completion of cleanup	• Process to effectively define endpoints from scientifically and technically defensible understanding • Clearly defined and credible cleanup scope of work to achieve risk-based endpoints • Transition complex sites to LTM or MNA
Budget and Resource Allocation	Allocate limited resources (i.e., federal budget dollars) to provide benefit to society (e.g., reduced risk, recovered resources, etc.)	• Risk-informed choices to prioritize resources, drive 'cleanup demand' and complete site cleanup

Risk-Based Alternative Endpoint Strategies for Site Closure and/or Long-Term Management

Technically-based approaches to achieve alternative endpoint strategies, which are risk-based, cost-effective, sustainable, and protective of public health and environment

Fig. 6. Challenges, issues, and opportunities associated with risk-based alternate endpoint strategy.

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