advances in remediation
Our industry-leading scientists and engineers are rethinking the future of site evaluation and restoration and redefining what is possible.
Introduction

The remediation industry is constantly changing, and new technology is leading to breakthroughs that are changing the way we approach complex environmental issues. Emerging contaminants, new treatment techniques and state-of-the-art tools are helping us to rethink the future of site investigation and remediation.

In 2016, Arcadis published 12 articles in our first Advances in Remediation book, sharing insights from our industry-leading experts on a wide range of environmental topics. Our scientists and engineers never rest in their pursuit to develop the next-generation technologies and redefine what’s possible, and, in this updated version of Advances in Remediation you will hear about Arcadis’ newest findings on technologies and achievements that are shaping the industry today.

Our focus on increasing the certainty of outcomes, attaining sustainability and achieving business objectives is driving innovation in this industry. It requires a new way of thinking to break through the conventional mindset. And these are the discoveries that we’d like to share with you.
Our focus on increasing the certainty of outcomes, attaining sustainability and achieving business objectives is driving innovation in this industry.
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We are on the cusp of major breakthroughs that are not only significant to the remediation industry of tomorrow, but are also applicable to your remediation projects today.
Leading the Way: Transforming Remediation through Arcadis Innovation

New remediation technologies that support the destruction, stabilization, sequestration and recovery of contaminants are being developed every year. However, they are still reliant on our abilities to navigate complexities in the subsurface and appropriately interpret data. Over the next decade, we can expect some of most impactful technological advancements in remediation to fall into two primary categories: (1) innovations that increase the resolution at which we can see and interpret the overlay of contaminant mass and hydrogeology; and (2) digital innovations that will support the collection and analysis of dramatically larger data sets with very little labor.

Arcadis is constantly pushing the envelope in both of these directions and can share how new technology is advancing both fronts and passing on their related benefits. We invest in hundreds of new concepts each year through our global innovation program to bring fresh ideas to the remediation industry — and the best examples are funded for industry release.

This process led to some of the first patents for in situ anaerobic bioremediation for metals, chlorinated solvents and nitrates in groundwater. We have pioneered site closure risk management through guaranteed outcome project delivery, leading to the closure of thousands of sites across North America and saving clients more than $1 billion in remediation costs. We are now using advances in materials and technology to improve environmental cleanup decision making through highly accurate subsurface characterization, enhanced in situ remediation alternatives and advanced data analysis platforms.

While investing in innovation is critical to delivering positive outcomes, not all innovation is created equal. Linus Pauling stated it best in that “the best way to have a good idea is to have a lot of ideas.” Significant advances have driven improvements in the remediation industry over the past four decades, and some technologies have come and gone. That’s why we are firm believers in technical knowledge — not technology — as the key element to our clients' success.
As part of a current pipeline of more than 40 new innovation concepts, Arcadis is demonstrating new ways to map chlorinated solvent dense non-aqueous phase liquid (DNAPL), rapidly assess concentrations of emerging contaminants, and drive innovation in regulatory management strategies. We recognize that every company is now transforming into a digital company, and we’ve developed new tools that apply cutting-edge digital innovations, including holographic displays in mixed and augmented reality, unmanned aerial vehicles for data collection and the monitoring well network of the future — the Internet of Wells. Several of these are highlighted below, with others featured elsewhere within this document (see inset).

**DyeLIF: A NEW WEAPON FOR CONQUERING CHLORINATED DNAPL SOURCE ZONES**

DNAPL source zones represent one of the most significant challenges in the remediation industry. When present, they create contaminant plumes in groundwater that can persist for hundreds of years unless they are effectively removed or controlled. Unfortunately, the nature and extent of DNAPL distribution is often extremely complicated and difficult to accurately characterize.

Arcadis partnered with Dakota Technologies to deliver a new characterization tool called DyeLIF™ (Figure 1), and has completed the greatest number of industry applications of this tool. Used extensively for petroleum characterization, DyeLIF

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**Figure 1:** Arcadis partnered with Dakota Technologies to deliver a new characterization tool called DyeLIF™. Used extensively for petroleum characterization, LIF characterization techniques have been unavailable for DNAPLs — which don’t naturally fluoresce.
Characterization techniques have been unavailable for DNAPLs—which don’t naturally fluoresce. The DyeLIF technique relies on the application of indicator dyes that can partition into the DNAPL and impart the ability to fluoresce, and therefore provides high-resolution characterization of both DNAPLs and aquifer permeability. Outputs from these studies can be viewed in a DNAPL dashboard to drive better data interpretations and more-accurate DNAPL conceptual site models.

The DyeLIF technology is an example of a new class of data-rich, real-time characterization and monitoring techniques that are supporting the implementation of more accurate, efficient and timely remediation efforts. Techniques like this are not only easy to operate but also gather thousands of data points in a single mobilization. Employing adaptive decision making, these approaches can achieve characterization objectives in a single mobilization of several weeks to months, versus the many years and multiple mobilizations that conventional approaches often take.

REAL-TIME ANALYSIS OF PFASs

The emergence of poly- and perfluoroalkyl substances (PFASs) as contaminants in surface- and groundwater resources has driven significant scrutiny and demand for rapid assessment and characterization of potentially impacted water supplies. Fixed-based lab methods are both high in cost and sample process time, which can delay characterization activities and remedial response. The introduction of a mobile laboratory capable of producing reliable PFAS results for soil and groundwater in hours instead of weeks has enabled us to conduct adaptive characterization at PFAS sites for the first time. When combined with high-resolution permeability logging methods like the hydraulic profiling tool (HPT), the Stratigraphic Flux approach can map PFAS impacts within a mass flux framework to identify key transport pathways that help focus monitoring strategies and remedial development.

Utilization of real-time sample collection and in-field monitoring allows an adaptive characterization approach for PFAS sites that streamlines site evaluation and reduces the expensive step-wise process of planning, investigating and reporting. Deployment of the PFAS mobile laboratory has yielded excellent results, with strong correlation between mobile lab and fixed laboratory data. Where applied, the mobile lab has been used to process up to 20 groundwater samples per day and guide two drilling rigs, while eliminating wasted borings and samples. The ability to evaluate PFAS sites in real time and at high resolution has greatly enhanced our ability to efficiently characterize these sites; allow stakeholders to quickly understand potential risk; and identify focused, efficient remedies.

MIXED REALITY: HOLOGRAMS AND THE FUTURE OF COMPUTING

Mixed reality (MR) is a technology that transforms your field of view into a computing surface and merges the physical and digital worlds through holograms displayed in your environment.
The data, applications and contacts that were previously available only on your laptop or mobile phone can now be accessed anywhere, anytime.

Arcadis has implemented MR technology using the Microsoft HoloLens and mobile devices. The Microsoft HoloLens is a portable headset that can project holographic content into your view of the real world in front of you. The HoloLens gives users an unmatched 3D experience of the model in the real environment. The option to view MR content on a mobile device means specialized hardware is not required for users to take advantage of what the technology offers.

Arcadis used a MR application to support the closure of a 1-mile-long TCE plume with no active remediation and no long-term monitoring. MR was a critical tool for communicating the complex subsurface conditions to a range of stakeholders.

MR also can instantly connect users in the field with remote experts, providing the experts a real-time view of the user’s environment, as well as the ability to digitally mark up the user’s environment by adding holograms to walk them through complex tasks.

**UNMANNED AERIAL VEHICLES**

Today more than 200 million unmanned aerial vehicles (UAVs) are deployed, and they are capable of surveying 40,000 square miles of terrain per day. Advances in technology and the simplification of the regulatory landscape have created an environment where UAV use has proliferated — there are now more registered UAVs in the United States than registered airplanes. With a nearly unlimited capacity for data collection, rapid deployment capabilities, low “per datum” cost, and the large geographic footprints that can be evaluated in a short period of time, numerous UAV-based systems are already replacing traditional information collection techniques.

The use of UAVs allows for the rapid collection of very large amounts of georeferenced data in a cost-effective
manner. But perhaps even more important are the ways in which UAVs are changing the way data is not being collected. The deployment of UAVs into hazardous or unknown environments before human responders reduces or eliminates risk to responders and allows for more-informed decision making as to when and how to send people into dangerous environments.

UAV usage in the environmental industry will be limited only by the availability of deployable sensor technology and the imagination of the end user. UAVs have a bright future in our industry, as they cost less than traditional methods, are quick to deploy and reduce staff exposure to risks.

BRINGING GROUNDWATER MONITORING INTO THE DIGITAL AGE: THE INTERNET OF WELLS

Routine monitoring of groundwater contaminant concentrations and water levels is one of the largest costs associated with a remediation project, with hundreds of millions of dollars spent each year. Current monitoring costs are high because monitoring requires multiple steps: travel to a remote site, labor to conduct an event, laboratory costs of analyzing groundwater data and reporting of results. Arcadis is bringing groundwater monitoring into the digital age by developing a smart monitoring well that will significantly reduce the cost and effort associated with monitoring.

The smart monitoring well integrates the best of recent innovations, with solar-powered down-well sensors providing continuous monitoring of geochemical parameters (e.g., dissolved oxygen, pH and conductivity); groundwater elevation; and site-specific (or well-specific) groundwater and contaminant flux. Transmitted via Bluetooth or wireless connection, sensor results are intended to make data available on a web-based dashboard to enable viewing of the results in real time. Real-time data would provide instantaneous review of plume change and variability, and yield an entirely new dimension of plume behavior assessment related to seasonal variabilities, remediation performance or changes in potential exposure risk.

REGULATORY INNOVATION – RCRA FIRST AND LEAN SIGMA SUPERFUND DASHBOARDS

Innovation isn’t always about technology. In a rapidly changing regulatory era, leveraging a 360-degree perspective of the restoration process has provided a platform from which significant value can be created for all process stakeholders. Integration of strategic advice from former executive management regulators, future trend analysis related
to emerging issues and major policy/regulatory changes and knowledge of Environmental Protection Agency (EPA) and Environmental Council of States’ (ECOS) strategic plans has opened several unique opportunities for collaboration that allow achievement of responsible party business objectives and regulatory milestone achievement. Several primary areas where Arcadis has driven regulatory innovation are the development of the RCRA First program, development of state-led land stewardship services for legacy sites and development of the Superfund Remediation Executive Dashboard.

Working closely with EPA, Arcadis leveraged the Lean Sigma process to eliminate unnecessary activities in key Resource Conservation and Recovery Act (RCRA) cleanup process steps as part of the RCRA First pilot initiative. These pilot projects were successful in streamlining the process for facility investigations from an average of 10-20 years to <5.1 years, and the remedy selection from >6 years to 1-2 years. Based on the pilot program success, the RCRA First Toolbox was identified by the Trump Administration for expansion across the regions as part of its 2018 Appropriations Justification.

In West Virginia, Arcadis served as technical advisor for the governor’s initiative to develop the West Virginia Land Stewardship Corporation (WVLSC) Act and establish a nonprofit corporation with a primary mission to provide land stewardship services for protection of federal and state risk-based cleanups with engineering and institutional controls (IECs).
Under the Act, the nonprofit accepts legacy sites from the business sector into its land bank, provides long-term stewardship services, and works with state and local economic development agencies to deploy legacy sites back into productive job-creating ventures. As a result, Arcadis’ clients have eliminated closed legacy assets in West Virginia while leveraging the WVLSC to sustain IECs as these properties are transferred and redeveloped.

Another innovation opportunity is our concept of the Superfund Remediation Executive Dashboard (Figure 3), where value can be created for all involved stakeholders. Those familiar with the Superfund process understand there are many inefficiencies and delays in remediating these complex sites to closure. Administrator Pruitt has called for a Superfund Priorities List to identify Superfund sites to prompt robust dialogue with stakeholders to promote expeditious remediation.

Common EPA limitations for these cleanups include delays related to short-staffing, overall workload allocation, lack of internal technical resources or multi-stakeholder project planning. The dashboard is therefore an excellent means for EPA, states and remediating parties to track remedy implementation and completion progress in real time with current EPA digital data collection and analytics tools.

Key performance indicators can include:

1. Remediation milestone stage
2. Regulatory submittal dates for submittals awaiting approval
3. Dates of pending key events (major milestone decision documents/field work implementation)
4. Regulatory or technical challenges
5. Stakeholder interests (state interests, potential redevelopment) and other information to accelerate decision making and cleanups

Many clients have fully funded plans to remediate Superfund sites to regulatory closure. Without this information and other data sets, EPA executive management is unaware there is low-hanging fruit available for significant Superfund accomplishments.

The dashboard can assist all stakeholders with a clear understanding of the issues, decision points and steps needed to make progress. The dashboard’s data collection of key information in real time can dramatically remove barriers and increase transparency, predictability and collaboration while reducing costs and years of time.

The dashboard also will assist EPA and ECOS as they recast their interaction under Cooperative Federalism 2.0 to reduce operating costs through a more-efficient division of best practices and resources while eliminating redundancies. This collectively allows a more-matrixed management of Superfund sites with appropriate technical and project management from EPA and state team members.
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The Secret to Better, Faster and Cheaper: "Smart" Characterization

"Smart" Characterization is an innovative approach to site investigation that lowers costs and helps solve the unsolvable problems. The results speak for themselves — the closure of a mile-long trichloroethylene (TCE) plume (Figure 1) is a prime example.

The site went from characterization to closure in less than 10 years, was characterized without installing a single well and is being closed with no active remediation, which would have taken decades and cost tens of millions of dollars. Arcadis’ investigation of the site started with a smarter approach to characterization, identifying the predominant contaminant transport pathways, and allowed better decision making between risk, liability and business objectives.

THE PROBLEM WITH THE STATUS QUO

Monitoring wells — the traditional approach to characterization — give a fuzzy picture of conditions in an aquifer because they smooth out variability. A sharper focus reveals the significant variability that has been missed (Figure 2). While traditional approaches using monitoring wells for site characterization provide reproducible groundwater data, wells often give a misleading picture of plume magnitude and migration. Plumes that may appear thick have been exaggerated by the monitoring well network. In reality, the plume often consists of a few “ribbons” of contaminants that can be treated faster and more cost effectively, as shown on Figure 3. But, how are these

For the vast majority of sites, more than 80 percent of the groundwater plume mass moves in less than 20 percent of the aquifer volume.

Figure 1: A mile-long TCE plume characterized with high-resolution groundwater sampling.
ribbons located? Moreover, how do we characterize the source zone that produces the ribbons in the first place? Enter Smart Characterization.

THE SMART CHARACTERIZATION APPROACH

Smart Characterization is a way of collecting high-resolution data, translating that data to plume behavior, and then relating that behavior to remedy performance. High-resolution characterization (HRSC) and the importance of mass flux are frequently cited in the site investigation field.

At Arcadis, we recognize that success depends not only on the tool, but also on how we interpret the data to create remediation success. HRSC is not just about collecting a lot of data. If those data are not collected in the proper geologic context, interpreted in three dimensions by an experienced practitioner and then leveraged to enhance remedy execution, it creates a misleading conceptual site model (CSM). Worse, if the data are not collected efficiently and adaptively, the HRSC effort is needlessly expensive. The Smart approach reimagines the HRSC process:

• First, it identifies applicable high-resolution methods to meet the goals of the investigation — source evaluation versus downgradient risk, mass storage versus mass flux.
• Evaluation of source areas is focused on mass distribution within

Figure 2: Monitoring wells present a blurred picture of conditions in an aquifer; high resolution methods bring the right details into focus.

Figure 3: A plume characterized with monitoring wells vs. the same plume characterized with high-resolution sampling.
transport and storage zones to better inform source remedy evaluation. Downgradient tools are selected to develop a mass flux framework at the site: tools that characterize concentration and permeability.

• The resulting 3D heat map of mass flux or source distribution is constrained by geologic interpretation, an approach we call “Stratigraphic Flux,” which can then be used to focus and prioritize remediation efforts (Figures 4 and 5).

• This investment in Smart Characterization leads to better, more-focused remedies. It also pays dividends, a return on investigation (ROI), by avoiding remedy misfires and supporting more-focused remedies that limit the total life-cycle cost of remediation.

The key is to use tools that enable adaptive, high-resolution site characterization methods for permeability and contaminant mapping, which are then combined with geologic interpretation and 3D visualization. The result is a 3D heat map that illustrates the transport zones where more than 90 percent of mass flux occurs, and differentiates flux from the immobile (and low-risk) mass located in low-

**Figure 4:** Characterizing stratigraphic flux allows for focused, efficient remedy options. On this transect, 90 percent of the mass flux occurs within just 10 percent of the plume area.

**Figure 5:** TCE source area beneath a building. An adaptive grid and mobile lab were used to complete the investigation in seven days. The majority of the source mass resides in low-permeability storage. An evaluation of the data determined that the source, although concentrated, posed little risk to the structure and underlying aquifer, thus avoiding a needless remedy.
permeability storage zones. With this data in hand, source areas can be surgically targeted, or streamlined management strategies can be pursued to better meet business objectives. This enables focused remedies, reduction of life-cycle costs and improved reliability. The Smart Characterization approach uses HRSC tools and a technical and economic decision-making framework to:

- Reduce the total cost of remediation
- Better define uncertainty and risk
- Establish achievable endpoints before remediation begins

THE NEW ROI: RETURN ON INVESTIGATION

This approach leads to what we call return on investigation, or ROI. To obtain a ROI, we use a decision process that aligns your investment in site evaluation with your business objectives. For example, our experience with Stratigraphic Flux shows that, for the majority of sites, more than 90 percent of the groundwater plume mass moves in less than 10 percent of the aquifer volume. This presents a missed opportunity at many sites. If most of the plume is in a small portion of the aquifer, then treatment can be focused there and tailored based on the plume maturity.

Understanding mass flux enables you to tailor the remedy to balance risk and cost to match project business objectives. For example, we recognize that aggressive source remediation isn’t always the best approach. Sometimes it makes more sense to balance capital expenditures with long-term management strategies and focus on optimizing an existing remedy and reducing annualized project expense. We developed the Smart Characterization program to help focus existing remedies on the mass that matters most in order to develop the best total cost solution to match the unique client business model.

Now you can surgically target the sources that matter most or pursue streamlined management strategies that meet your business objectives.
Smart characterization tools provide quantitative mapping so you can find the flux and focus on the mass that matters.

**Figure 6**: Graphical method for comparing the ROI achieved using a Smart Characterization approach. The first graph illustrates the best-case scenario, ROI added when a project starts with Smart Characterization. The second graph shows the ROI when Smart Characterization is applied as a pre-design step prior to remedy construction. The third graph illustrates the potential ROI gained when using Smart Characterization to optimize an existing remedy. In all cases, the key is to predetermine how much to spend on characterization so that the ROI matches your business objectives.
Arcadis designs our *Smart* Characterization programs with exit ramps, or decision points, where the cost benefit of the approach is evaluated against stakeholder objectives regarding incremental improvement in the CSM and potential reduction in life-cycle cost through remedy optimization. This approach enables right-sizing of the *Smart* Characterization investment based on anticipated improvements in existing remedy performance and design support for anticipated remedial alternatives that can provide substantial reductions in life-cycle costs, cleanup duration and risk/liabilities. As illustrated on Figure 6, there are multiple entry points that can achieve differing levels of ROI.

*Smart* Characterization methods are transforming the way we investigate and remediate contaminated sites. The new ROI demonstrates that the biggest impact on remedy performance and total cost is driven not by remedial technology, but by narrowing the focus for remediation.

The bottom line is this: *Smart* Characterization is one of the most powerful tools available to improve the reliability and cost effectiveness of site remediation.

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Arcadis recently completed a demonstration for the US Air Force Civil Engineering Center (BAA ID 967) to show how CSMs can be improved using the Stratigraphic Flux approach. The field demonstration was completed at a former waste disposal pit (CRP3). The figure illustrates the hydrofacies (transport, slow-advection and storage zones) below the water table along each flux transect and the results of the stratigraphic flux evaluation. At each transect, the relative flux is truncated to the top two orders of magnitude to illustrate region where >90 percent of mass flux occurs. The relative flux results are colored on a consistent scale to show variability with distance from the CRP3 source area.
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Connecting the Dots for Large Plume Restoration: The Three-Compartment Model, Smart Characterization and Dynamic Groundwater Recirculation

Contaminant transport in the subsurface is controlled by hydrological, microbiological and geochemical processes that occur over scales ranging from the microscopic (sub-nanometer) to macroscopic (kilometer). As a remediation industry, we have historically focused on the macroscopic impacts of these processes and, at the same time, overlooked more-finite details that are essentially to reducing treatment time, achieving remedy objectives and reducing cost.

Successful remediation of an impacted aquifer requires a firm understanding of each of these processes, and the ability to characterize the spatial variability of complex subsurface properties that control groundwater flow and contaminant transport. This knowledge, along with predictive models that can assimilate the relevant processes over a range of scales, is needed to forecast the potential effectiveness of various remedial strategies. While this sounds intuitive, it has been a great challenge for remediation professionals — the ability to accurately predict finite remedial timeframes.

Historically, subsurface behavior has typically been investigated using a reductionist approach, where the

Figure 1: The scale of site specific investigations has decreased with evolving advanced technologies and at the same time the scale of the plumes we focus on has increased.
details of small-scale processes (e.g., soil gradation or site depositional features in a soil boring) are generalized and then scaled up using simplifying assumptions to match field-scale behaviors (i.e., prediction scale). These simplified, lumped parameters can be conveniently incorporated into predictive tools (e.g., groundwater models) originally developed for regional potable water supply aquifer systems.

While this approach has had limited success, it lacks the nuance necessary to reliably simulate contaminant transport and predicting treatment response. Until recently, it has been impractical to obtain the multi-scale characterization data needed to adequately and objectively represent the subsurface environment to enhance this approach.

The advent of high resolution site characterization (HRSC), along with advancement of advective transport and storage zone concepts, has provided a more-realistic and detailed approach to mapping a contaminant’s fate and transport in the subsurface and, more importantly, to increasing the efficiency and reliability of its remedy.

**THE THREE-COMPARTMENT MODEL: A NEW FRAMEWORK TO FOCUS REMEDIATION**

Over the last 40 years, the scale of contaminant plumes has been increasing, while the scale at which we collect our subsurface data has been decreasing (Figure 1). We have evolved from applying simplified “bulk averages” and steady-state assumptions to where we now collect high-resolution data and consider transient behavior.

We recognized that our conceptual site models (CSMs) and predictive tools needed to be improved to better understand and explain the nuances and increased level of detail in the data being collected. More importantly, we realized that, to develop appropriate remedial strategies and accurately predict their associated cleanup times, we needed to do a better job conceptualizing our sites and modeling solute transport behavior.

*Figure 2: (A) Three-dimensional depiction of variations in hydraulic conductivity and heterogeneous conditions within an alluvial aquifer. Blues and whites represent sands and gravels, and greens represent silts and clays. (B) Highlights primary groundwater flow pathways (blue and white zones) and primary storage zones (brown and green zones) through the aquifer.*

Connecting the Dots for Large Plume Restoration: The Three-Compartment Model, Smart Characterization and Dynamic Groundwater Recirculation
Before the start of the new millennium, practitioners believed that, by reducing the complexity observed in soil borings into a simplified, but equivalent, homogenized model (representative elemental volume [REV]), we could adequately describe groundwater flow dynamics and make accurate predictions. Limitations with the REV model were observed in countless groundwater extraction remedies, where projected treatment times were exceeded by decades — coupled with ongoing annualized cost.

The solution to the REV model was initially revealed in the early 2000s by advances in site characterization methods (e.g., tracer studies and high-resolution aquifer profiling), which found that: (1) plumes were moving much faster than we initially thought; and (2) plumes do not become homogenized across the aquifer with distance. Instead, we found that plumes move through networks of small-scale pathways of higher permeability consistent with the geologic processes that created the aquifer (Figure 2). It was obvious that the subsurface needed to be assessed at a scale smaller than the REV to incorporate these fine-scale geologic heterogeneities that control groundwater flow and contaminant transport.

The dual-domain model (DDM) — originally developed in the early 1960s but not fully embraced until the early 2000s — advanced our ability to represent heterogeneities in transport models by dividing the aquifer matrix into two compartments: one immobile and the other mobile with mass exchanged between the two via a transfer coefficient. DDM significantly improved our ability to match observed breakthrough data by representing the fast- and slow-moving flow and transport processes, and created a clearer distinction between the “bulk average” concepts of the REV and site-specific contaminant transport. While an improvement over the REV, it still simplified subsurface heterogeneity by lumping together a broad range of aquifer permeabilities.

This simplification becomes even more evident when we consider the findings of high-resolution aquifer studies that have repeatedly shown that much of the transport occurs in only a small fraction of the cross-sectional area. Typically, we find that more than 80 percent of the
contaminant flux occurs in less than 20 percent of the cross section. If the two-compartment model held true, then upwards of 20 percent of the contaminant remains in storage (i.e., low-permeability sediments, which are commonly silts and clays) accessible only via matrix diffusion occurring over a very long time or by implementing costly “brute force” measures to recover or destroy mass.

This thinking, however, is an artifact of how we have viewed the subsurface as a binary system of either transport or storage, and does not reflect the insights gained from HRSC that have repeatedly shown that the true range of permeability in the subsurface cannot be adequately represented by a two-compartment model (Figure 3).

While high-permeability sands and gravels acting as pure transport zones and low-permeability silts and clays acting as pure storage zones are consistent with the two-compartment DDM model, we also know that intermediate permeability zones (interbedded sands/silts/clays) are prevalent in natural depositions. While some portions of these intermediate zones are static, most of the groundwater is slowly moving, but at rates far faster than the velocity of diffusion, and these are, therefore, not pure storage zones. This small but important distinction offers an improved framework for developing more-effective remedial systems and for making meaningful progress at sites where it was previously not thought possible.

These concepts form the foundation of our three-compartment model: Compartment 1 (C1) is where

Figure 4: Separation of the aquifer depicted on Figure 2 demonstrating the three-compartment model of the subsurface aquifer architecture and representing a realistic conceptual model of solute transport based on order-of-magnitude contrasts in groundwater flux: (C1) pure advective/transport zones [sands and gravels] (C2) slow advective/storage zones [sands mixed with silts and clays], and (C3) pure storage zones [silts and clays].
advection is the predominant transport mechanism; Compartment 2 (C2) is where slow advection is dominant, but diffusion has observable effects; and Compartment 3 (C3) is dominated by both diffusion and storage (Figure 4).

**DGR™: A MORE FOCUSED REMEDIAL STRATEGY**

The three-compartment model helps explain what a monitoring well observes during remediation (Figure 5): initial improvements in water quality are due to flushing of C1 (pure transport), while the tail is mostly slow-advection rather than diffusion from C2. This tells us that if we can increase the flow through C2 (i.e., enhance flushing of slow-advection zones), we can reduce the time to access impacted groundwater and achieve shorter cleanup times. But how can we leverage this knowledge?

One solution has been found in looking at conventional pump and treat (P&T) systems. P&T is arguably the original groundwater remediation technology and continues to be widely used. It is a strategy that works primarily to control rather than restore. The central weakness of conventional P&T is that it tends to create fixed hydraulic conditions, which leads to the development of stagnation zones and limits flushing to the primary transport zones (C1) while isolating other zones containing contaminant mass (C2).

To successfully restore an aquifer, we believe it is necessary to create dynamic conditions that mimic, even exaggerate, the natural variability that initially created the plume. Groundwater levels rise and fall, transient shifts in flow patterns occur, and groundwater demands vary over time, which work to collectively spread contaminants within an aquifer.

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The DGR approach leads to a more-focused remedial strategy and, subsequently, a significant reduction in lifecycle remedial costs.
By strategically manipulating a combination of injection and extraction volumes and patterns in the subsurface, we can induce dynamic conditions and create differential gradients between transport zones that will flush contaminant mass from the adjacent slow-advection zones to where it can then be easily recovered (Figure 4). This is the basis for the enhanced flushing technology known as dynamic groundwater recirculation (DGR™).

The underlying concept is relatively simple: accelerate the influx of clean groundwater to enhance hydraulic and concentration gradients that, in turn, drive contaminant mass out of the aquifer via all advective pathways and diffusive gradients.

Faster cleanup times can be achieved by strategically moving more pore volumes and manipulating gradients to increase mass flux/advective transport through C1 and C2 (transport and slow zones, respectively). Clean water within these two compartments works to enhance the mass transfer/diffusive transport of stored massed across C3 (storage zones), providing a means to overcome aquifer heterogeneities and the effects of matrix-controlled back diffusion.

To that end, the primary distinction between DGR and conventional P&T is the difference in how site data are leveraged to develop an appropriate flushing framework, a dynamic operation plan and an approach for continuous adaption based on remedial performance. The goal of system design and operation is to maximize contaminant mass removal by extracting within the plume core while injecting clean water at strategic locations to enhance flushing and drive contaminants toward extraction wells (Figure 6).

The key to success is adopting a dynamic and adaptive management approach to system operation. Frequent system optimization by varying pumping/injection rates and locations in response to changes in performance data accelerates the removal of contaminant mass while maintaining hydraulic control of the plume.

When properly designed and operated, DGR can be a highly effective remedial technology that significantly advances conventional P&T applications of the past — in some cases, existing P&T systems can even be re-engineered to a more effective DGR remedy. It provides an efficient way to manage treated water while maintaining water levels; reduces time required for remediation through enhanced flushing; and, most importantly, can achieve endpoints that were previously considered virtually impossible to reach — particularly for large, diffuse plumes.
Large plume is a term that covers a wide range of contaminated aquifer scenarios, but, in general, a large plume can be characterized as one that:

- Occurs in productive aquifers with the potential to transport dissolved contaminants over large distances;
- Consists of contaminants that do not sorb to the aquifer matrix and are not quickly degraded (chemically or biologically) under natural aquifer conditions; and
- Develops from a large-enough source mass (or multiple sources) to generate a large volume of groundwater that exceeds regulatory criteria.

While there has been steady progress moving small plume sites to closure through improved technology (e.g., transition to in situ reagent-injection programs) and institution of risk-based regulatory standards over the last 20 years, there has been little advancement for large plumes, and the total inventory has increased with establishment of regulatory guidance for emerging contaminants. The historical remedial approach to large plumes has been conventional P&T and, as we have outlined in this article, this strategy can contain large plumes — often in costly, extended ways.

In the late 1990s, as part of a performance-based contracting program, we committed to achieving closure for several large plume sites, and we recognized early on that, to meet these commitments, the traditional approach to large plumes had to be abandoned.

First, we re-examined our understanding of contaminant transport and storage in aquifers. Next, we needed to develop and apply cost-effective characterization strategies and tools to support the improved science of remediation hydrogeology (i.e., introduction of HRSC). Finally, we needed to create and test technologies that could remediate large plumes cost effectively.

Through this process, we learned that, because of the wide range of soil permeabilities found in even the simplest geologic settings, groundwater flow (and contaminant flux) occurs in a very small fraction of the total aquifer volume (Figure 4). By improving our understanding of and ability to map the relevant contaminant transport zones, we can reduce the scope of remedial action to a fraction of what is needed for a classic remedial method. We have also translated this new interpretation of contaminant flow pathways and aquifer matrix storage processes into more-effective remedial technologies, such as DGR.

The enhanced flushing approach employed by DGR has been and is currently being applied to numerous contaminated sites in various settings with great results. While this strategy may not work for every project site, the primary elements of large plume CSM and performance optimization allow realization of project cost savings regardless of whether the goal is to improve annual remedy efficiency or to advance plume cleanup to clean closure.

These flux-based and data-driven operational elements are also especially favorable for successful application of DGR, allowing achievement of what was previously unthinkable for many large and/or complex contaminant plumes — within relatively shorter time frames and with reduced costs.
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Marc Killingstad, PE, is a Principal Water Resources Engineer/ Groundwater Hydrologist for Arcadis and serves as the Director of Hydrogeology and Remediation Hydraulics within our Technical Knowledge and Innovation Group. He has 20 years of experience developing and applying groundwater models to support site investigation and remedial design in a variety of settings throughout North America, Europe and Australia. Killingstad also provides technical leadership in the areas of remediation well design, installation, development, hydraulic testing/assessment and rehabilitation.
Surface water sheens develop when non-aqueous phase liquid (NAPL) seeps from soil and sediment to the surface of a water body. Sheen mitigation is regulated under the Clean Water Act of 1972 (the “Sheen Rule”), and public perception can also be a key driver to developing engineered solutions for elimination and control of anthropogenic sheens.

Development of these mitigation solutions has also been historically challenging due to the poor predictability of sheen occurrence under variable environmental conditions. Increased awareness of NAPL mobility and the governing processes for sheen formation, however, have served to significantly improve our awareness of NAPL seepage and sheen formation — enabling significant improvements in sheen mitigation and management solutions.

The ability to predict the potential for NAPL seepage and resulting sheens, as well as their potential magnitude, requires a characterization of NAPL quantity, mobility, natural depletion, hydrogeology and groundwater-surface water hydrology.

This information can be used to predict the existing and future potential for mobile NAPL seepage to generate sheens, thereby supporting scaled mitigation approaches that are appropriate to site conditions. This is a considerable improvement on overly conservative conventional approaches, which often assumed that all NAPL, both near the

To reduce the generation of sheens, the focus must be on the NAPL at the GSI.

Figure 1: NAPL hydraulic gradient NAPL transport mechanism
groundwater-surface water interface (GSI) and in the upland, will contribute to the generation of sheens.

This article discusses several key elements to the identification of a state-of-the-art NAPL seepage mitigation strategy, including an understanding of NAPL mobility, an assessment of an aquifer’s NAPL assimilative capacity, and integration of these data into NAPL seepage conceptual site models (CSMs).

HOW DOES NAPL GET TO THE GSI?

For subsurface NAPL to reach an adjacent surface body, it must be both mobile and driven by sufficient hydraulic gradient. NAPL may be present in soils, but at volumes below the residual saturation threshold, and is therefore not mobile. NAPL volumes may be high and expected to be mobile, but there may be insufficient hydraulic gradient to induce flow. In both these cases, NAPL will not migrate to the surface water body and cause sheen. NAPL mobility, like the potential productivity of a groundwater aquifer, can be quantified by evaluating transmissivity, a parameter useful in characterizing NAPL properties, soil properties, and site-specific NAPL saturation and spatial distribution.

NAPL migration toward the GSI is most generally controlled by the NAPL transmissivity and NAPL gradient, but other mechanisms can contribute to NAPL migration. The assimilative capacity of the subsurface naturally mitigates NAPL migration and NAPL seepage. Storage plays a role as an assimilative factor because releases will spread only while both sufficient volume and hydraulic gradient are present to overcome soil capillary forces and allow NAPL flow. If sufficient aquifer storage capacity exists, or if volumes and gradients decline, NAPL will no longer migrate. Natural source zone depletion (NSZD) is another key assimilative mechanism that continuously acts to naturally reduce the mass of NAPL in the subsurface through dissolution, volatilization and biodegradation.

WHAT ARE THE MECHANISMS FOR NAPL SEEPAGE AT THE GSI?

Contrary to previous perceptions, the generation of sheens is not directly connected to the discharge of groundwater at the GSI. Dissolved chemicals that are discharged with groundwater, even if they originate from NAPL dissolution, cannot recondense into NAPL. There must be a NAPL hydraulic gradient toward the water body, and there must be flow of NAPL across the GSI for a sheen to be generated (Figure 1). The discharge of groundwater at a GSI, which is due to groundwater mobility...
and a driving gradient, does not imply that there is NAPL mobility and a driving NAPL gradient to induce NAPL seepage.

Water-table variation can control NAPL mobility. When a rising water table submerges NAPL, it can lose larger-scale continuity and therefore mobility. When the water table subsequently drops, water drains from the largest pores. NAPL can then flow through those air-filled pores to the surface water and cause sheen (Figure 2).

NAPL wicking occurs when NAPL propagates along the top of the groundwater capillary fringe by the same mechanism that causes sheens to spread on a water surface (Figure 3).

Ebullition can drive NAPL seepage because the NAPL forms an intermediate wetting phase between air and water. The NSZD process of methanogenic degradation can produce methane in excess of its solubility so that it forms gas bubbles. These gas bubbles provide an air-water interface that accumulates a film of NAPL, and that NAPL is carried to the water surface by the bubbles’ buoyancy. When a gas bubble breaks at the water’s surface, the NAPL on the bubble forms sheen on the water surface (Figure 4).

Finally, erosion of soil or sediment that contains NAPL can also cause sheen. As the sediment particles disaggregate during erosion, the NAPL may be released to the surface-water body.
Performance monitoring of the initial remedy at the Pine Street Canal Superfund Site revealed sheens on the surface water and NAPL on the surface of a 3-foot-thick sand cap in the canal intended to isolate NAPL in the sediment. Arcadis executed a re-design investigation to fill data gaps and develop a CSM that included NAPL mobility within a peat layer. Conducted over three seasons, the investigation included borings, TarGOST™, diver surveys of NAPL seepage and surveying ebullition (Figure 5). NAPL residual saturation testing and bench-scale column testing were conducted to confirm the basis of design. Arcadis concluded that hydraulic gradient and ebullition were causing NAPL seepage into the canal and causing sheens. A reactive cap made of organoclay reactive core mat was placed in areas of NAPL seepage and ebullition to sequester NAPL. The organoclay reactive core mat is effectively controlling NAPL seepage.

HOW LONG WILL UPLAND NAPL FLOW RECHARGE NAPL AT THE GSI?

Understanding the potential contribution of upland NAPL to long-term NAPL seepage and sheen generation potential at the GSI is critical to selection of a long-term mitigation technology. Mobile NAPL and a NAPL gradient are necessary for ongoing NAPL seepage at the GSI. NSZD reduces the mobile NAPL and the NAPL hydraulic gradient and prevents the spread of the NAPL body.

Even if there is NAPL flow from the upland toward NAPL seepage at the GSI, the flow may be attenuated by NSZD before it results in seepage. NAPL smearing will also reduce NAPL migration (Figure 5). Capillary forces reduce the NAPL gradient by limiting the intrusion of NAPL into pore space not already occupied by NAPL. In this way, the rate of NAPL seepage and resulting rate of sheen generation can be substantially less than what would be inferred from upland NAPL flow.

Site-specific characterization of NAPL migration can elucidate whether reduction of NAPL mass in the upland is needed to mitigate NAPL seepage and sheen generation. To reduce the generation of sheens, the focus must be on the NAPL at the GSI.

Sheen generation processes are complex but can be understood and mitigated when a robust CSM exists.

Understanding NAPL Seepage: Leveraging Robust CSMs to Achieve Cost-Effective Sheen Mitigation
NAPL SEEPAGE CSM

The NAPL seepage CSM includes delineation of mobile NAPL and identification of relevant NAPL transport mechanisms. A robust NAPL seepage CSM will allow selection of the appropriate remedial technology — the design of which is based on the amount of mobile NAPL, seep mechanism(s) and the mass flux of NAPL.

The appropriate measures for effective, long-term mitigation of NAPL seeps and resulting sheens should be informed by this CSM, which effectively connects sheen observation to a NAPL seepage mechanism and the potential for NAPL recharge from the upland.

The CSM is developed by understanding the nature and extent of NAPL in the upland and the sediment. As discussed above, not all NAPL is mobile. The CSM should include the following key aspects:

- Identification (and quantification) of mobile versus residual NAPL;
- The primary NAPL seepage mechanisms that are active at the site;
- Estimates of NAPL mass flux to the surface water; and
- Evaluation and quantification of natural NAPL loss rates (i.e., NSZD) to evaluate long-term flux behavior.

Development of a robust NAPL seepage CSM is critical to identifying and operating the appropriate sheen remedial technology. The CSM connects the observed sheen to a NAPL seepage mechanism and to the potential for NAPL recharge from the upland and, more importantly, is then validated following implementation of mitigation steps via operational data collection.

NAPL SEEPAGE AND SHEEN GENERATION MITIGATION MEASURES

Remedial technologies to control NAPL seepage can be categorized based on how they prevent NAPL seepage and the resulting sheen. Understanding the mechanisms driving NAPL seepage and developing a clear CSM enable identification of a fine-tuned remedial technology or management approach for the most-relevant aspects to seepage. Remedy selection frequently entails a combination of either passive or active steps and leverages NSZD to balance business objectives and control NAPL seepage.

Using the NAPL seepage CSM helps determine whether measures such as NAPL recovery in the upland, upland

Figure 6: Oleophilic bio barrier sediment cap
barriers that prohibit NAPL migration to the water body, or upland excavation of NAPL would be worth the cost. NAPL recovery can reduce the source but only to a point. Once recovery rates become asymptotic and NAPL transmissivity drops below 0.1 square foot per day (ft$^2$/day) to 0.8 ft$^2$/day, additional recovery effort does little to reduce source longevity. This is due in part to the fractional reduction of NAPL mass that can be accomplished by hydraulic recovery when NAPL transmissivity is low.

More importantly for mitigation of upland-fed NAPL seepage at the GSI, low NAPL transmissivity implies low NAPL flow that is likely insufficient to exceed losses driven by NSZD. If NSZD losses are greater than the NAPL transmissivity, then NAPL mass near GSI will not be supplemented by NAPL migration from the upland (Figure 5).

Upland barriers, while they may be a measure that disconnects upland NAPL from the near GSI NAPL, should therefore only be necessary where there is demonstrated NAPL flow greater than the rate of NSZD. Similarly, complete removal (excavation) of upland NAPL is a measure that is rarely, if ever, necessary on the sole basis of the risk of sheens.

Finally, it should be acknowledged that the thin wedge of NAPL-affected soil between the excavation and the body of water can still generate sheens, despite the complete removal of upland NAPL.

Sediment caps, including sediment reactive caps, may be sufficient to control sheens if the upland source is controlled. Sediment caps are materials placed on the surface of the sediment to isolate the contaminated sediment from the overlying water and receptors.

Sediment caps for NAPL operate by reducing NAPL flux, sorbing NAPL, and/or promoting biodegradation of the NAPL (Figure 6). The appropriate capping technology will depend on the NAPL seepage mechanisms and the mass flux of NAPL.

The CSM should carefully evaluate NAPL mass near the GSI that is directly connected to NAPL seepage and sheen generation. Through characterization of NAPL seepage mechanisms and rates, and NAPL mass, informed decisions can be made about the use of sediment cap media and the amount of capacity to design into these measures.

Collectively, the above examples demonstrate how multiple techniques can be combined to mitigate sheening.
Sheen generation processes are complex, but can be understood and mitigated with a robust CSM.
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1,4-Dioxane: Increasing Concern Requires Risk Management and Innovative Treatment

1,4-Dioxane is an emerging contaminant that is a growing concern. It is present in a substantial number of water supply sources and has entered groundwater aquifers. Concern over the presence of 1,4-dioxane in drinking water has led to increased scrutiny, regulation and legal action.

Historically, advanced oxidation processes (AOPs) have been the presumptive ex situ treatment strategy, but they come with significant capital and operational costs. Bioremediation is quickly becoming an attractive, sustainable and economical alternative.

Arcadis is leading the industry in attaining site closure under natural attenuation strategies, applying advanced analytical tools to demonstrate 1,4-dioxane treatment, implementing in situ bioremediation approaches and innovating with ex situ bioreactors that are expected to become attractive retrofit options for existing water treatment systems. A changing regulatory landscape and liability uncertainties have prompted many Arcadis clients to consider assessing 1,4-dioxane risk across their site portfolios — a proactive management strategy that saves both money and corporate reputation.

1,4-DIOXANE OCCURRENCE AND REGULATION

Most 1,4-dioxane has historically been used as a stabilizer in chlorinated solvents. Increased awareness has revealed that 1,4-dioxane is also a byproduct of many manufacturing processes and is present in a variety of personal care products, detergents and other commercial items.

Its prevalence in industry and consumer products — and release into the environment — has led to its presence in nearly 20 percent of the large-scale public water-supply systems tested by the United States Environmental Protection Agency. While a federal
maximum contaminant level has not been established for 1,4-dioxane, more than 30 states have developed drinking-water or groundwater guidelines and promulgated standards (Figure 1).

Unfortunately, these standards vary by more than three orders of magnitude, complicating remediation and liability management decisions. Nevertheless, increased focus has resulted in regulatory mandates for investigation, remediation or legal action against potentially responsible parties.

Increasing legal and regulatory drivers for 1,4-dioxane treatment have also been amplified by public concern, sometimes resulting in both fiscal and reputational consequences for responsible parties. These can be mitigated by combining an awareness of the regulatory climate, familiarity with potential liabilities within a portfolio, and adoption of a proactive management strategy.

STRATEGIES FOR ASSESSING 1,4-DIOXANE RISK

Given the growing concern around 1,4-dioxane, and the expense and potential challenges associated with treatment, it’s becoming increasingly important for potentially responsible parties to know what their 1,4-dioxane risk may be.

There are two major management strategies (Figure 2A): a reactive management strategy (waiting for regulatory agencies to require 1,4-dioxane testing before taking action), or a proactive management strategy (evaluating the potential for liabilities associated with 1,4-dioxane in advance of regulatory and/or legal action).
A proactive management strategy allows implementers to mitigate reputational risk and reduce costs by allocating resources effectively. In many cases where 1,4-dioxane liability risk is high, proactive clients are taking actions to begin internal data collection and weigh treatment alternatives ahead of regulatory and legal drivers.

A proactive management strategy doesn’t necessarily require a large investment. A step-wise portfolio evaluation (Figure 2B) allows for an increasing level of effort as needed or desired.

The first step includes initial liability ranking of sites within a portfolio based on a high-level review of project details. The second step delves deeper into the site to generate a more-rigorous risk evaluation and quantify the potential financial impact. The third step is development of a full portfolio management strategy to allow for effective allocation of resources.

Understanding the potential liability across a portfolio of projects and treatment alternatives ultimately makes for better-informed decision making around management and treatment.

1,4-DIOXANE TREATMENT OPTIONS

Conventional drinking-water and wastewater treatment processes often remove only a portion of the 1,4-dioxane present — typically less than 50 percent. Likewise, traditional groundwater treatment systems historically designed to remove chlorinated solvents via sorptive processes (e.g., granular activated carbon) and stripping processes (e.g., air strippers) are generally not effective at also removing 1,4-dioxane.

The most dependable, and widely used, technology for 1,4-dioxane treatment has been AOPs, which entails a combination of powerful chemical oxidants and/or ultraviolet light that results in complete destruction of 1,4-dioxane. AOP implementation requires harsh chemicals and significant energy input, and can come with substantial capital and annual operational costs. While AOP has been the historical gold standard, other treatment methods are emerging — with bioremediation serving as a reliable, safer, sustainable and economical alternative to AOPs.

Biodegradation of 1,4-dioxane occurs naturally and can be enhanced in engineered systems. 1,4-Dioxane biodegradation occurs metabolically and cometabolically. Understanding which mechanism is most applicable to a given site setting is critical to understanding how natural attenuation may be occurring and how best to deploy an engineered application.

Fundamentally, successful natural and enhanced bioremediation requires that microorganisms have what they need to mediate the biodegradation reactions of interest and that the co-residence times of the microorganisms, their substrates, nutrients and the targeted contaminants are balanced.

This may occur naturally in some subsurface environments, and natural attenuation has been approved or utilized to achieve site closure at multiple...
**Figure 2A: Environmental liability management strategies**

**Proactive Management**
- Evaluate and manage risks ahead of inevitable pressures
- Effective for clients managing a portfolio of sites in several locations, those with a low threshold for reputational risk, and projects that need to meet/maintain closure
- Allows for effective allocation of resources, timely decision making, and optimized exit strategies

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**Reactive Management**
- Reacting to regulatory pressure, potential health risk, and/or reputational risk
- Appropriate when no regulations are in place and there is no human exposure risk
- When action is needed, it may be urgent, expensive, and include limited options

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**Figure 2B: Options for proactive portfolio management**

- **Mitigate Reputational Risk**
- **Understand Potential Liability**
- **Allocate Resources Effectively**

**Step 1: Precursory Heat Map**
- High-level questionnaire
- Identify potential regulatory and human health risk
- Result: Initial ranking and heat map

**Step 2: Risk Ranking and Quantification**
- Full portfolio or focused project reviews
- Rigorous evaluation of financial, operational, and reputational risk
- Result: Quantification of risk in portfolio (e.g., financial impact)

**Step 3: Portfolio Management**
- Detailed portfolio review
- Client-specific risk profile
- Result: Development of management plan

1,4-Dioxane: Increasing Concern Requires Risk Management and Innovative Treatment
sites. At some of these sites, a proactive management strategy provided enough historical data to demonstrate stable and decreasing 1,4-dioxane trends in groundwater and achieve closure. Emergent molecular diagnostic techniques now provide stakeholders with low-cost sampling capability to provide supplemental lines of evidence and boost the case for natural attenuation. These sampling programs also include target geochemical data to elucidate which destructive mechanism is most relevant.

Where conditions are not favorable for natural attenuation of 1,4-dioxane, innovative in situ and ex situ engineered bioremediation systems may be cost-effective alternatives to conventional AOP.

For in situ systems, the necessary components may be delivered directly to the subsurface (Figure 3A), or groundwater may be extracted and necessary components added and then reinjected (Figure 3B). Because one (oxygen) or more (light hydrocarbon gasses for cometabolism) of the necessary substrates for 1,4-dioxane biodegradation is a gas, the most effective mechanism for in situ delivery must carefully consider site hydraulics and facility constraints.

At a site in California, Arcadis took the direct-delivery approach and sparged air, propane, nutrients and a bioaugmentation culture into the subsurface treatment area to facilitate biodegradation of 1,4-dioxane, as illustrated on Figure 3A. For that project, Arcadis used stable isotope probing to confirm that biodegradation was a dominant treatment mechanism for 1,4-dioxane concentration decreases.

At another direct-delivery site in Michigan, Arcadis is treating 1,4-dioxane in a weathered, interbedded bedrock aquifer with biosparging points installed in emplaced sand lenses within bedrock fractures.

An example of the groundwater extraction, amendment and reinjection approach (Figure 3B) is a site in Colorado where Arcadis extracted groundwater from several wells in the treatment area, infused air and a light hydrocarbon gas into the water stream while it was above ground, and reinjected the water to distribute the substrates into the subsurface. This treatment strategy was initially focused on cometabolism of trichloroethene, but also led to remediation of 1,4-dioxane.

At many chlorinated solvent sites, ex situ treatment systems are already actively treating contamination that was historically identified, but often these systems were not designed with 1,4-dioxane in mind. Retrofitting existing ex-situ treatment systems with AOPs is a typical reactionary response at sites where 1,4-dioxane was recently identified after years of system operation.

Other end-of-pipe treatment options include bioreactors, which may prove more cost effective than AOPs. Additionally, conventional pump and treat systems can be reconfigured to become dynamic groundwater recirculation systems and effectively shorten remedial timeframes.
Figure 3A: Biosparge with bioaugmentation

Figure 3B: Recirculation with gas infusion

1,4-Dioxane: Increasing Concern Requires Risk Management and Innovative Treatment
While there are fewer examples of ex situ bioreactors than in situ treatment systems, it is a technology that is rapidly developing. The longest-running 1,4-dioxane cometabolic bioreactor is located at the Lowry Landfill in Colorado and has been successfully operating since 2003.

In 2017, Arcadis pilot tested two bioreactor configurations at a site in Michigan (Figures 4A and 4B): one that relied on metabolic biodegradation of 1,4-dioxane, and one that relied on propane-mediated cometabolic biodegradation of 1,4-dioxane — both the first of their kind in the industry.

CONCLUDING THOUGHTS

1,4-Dioxane is a growing concern for clients across multiple market sectors. Anywhere chlorinated solvents were historically used may have an uncharacterized 1,4-dioxane problem — but potential liability risk cannot be assessed based solely on sites where chlorinated solvents were used.

The widespread presence of 1,4-dioxane in a variety of products and commercial applications has created an abundance of mechanisms through which 1,4-dioxane can be released to the environment. Its detection in municipal water supplies has also increased public scrutiny and concern, increasing attention and demand for management and remediation solutions.

Proactive management steps can be taken to understand applicable regulatory requirements and mitigate risk. Where necessary, proactive strategies also enable selection of more-innovative techniques to support natural attenuation demonstration, cost-effective remedy implementation, or system retrofitting and optimization to offset long-term cost.

Demand for Remediation Requires Proactive Management
1,4-Dioxane: Increasing Concern Requires Risk Management and Innovative Treatment
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Dr. Monica Heintz has more than 10 years of academic and environmental consulting experience. She works at the nexus of groundwater hydrology, geochemistry and microbiology to understand, manage and mitigate environmental impacts. She specializes in application of environmental molecular diagnostic and statistical tools to understand, describe and predict contaminant fate and distribution. She excels at conceptual site model and remediation strategy development for sites with complex constituent mixtures. Dr. Heintz currently leads 1,4-dioxane biodegradation research and development efforts for Arcadis North America.
Flux-Based Monitoring Programs:
If Your Monitoring Well Isn’t Talking about Flux, It’s Lying to You

It is increasingly recognized that, at many sites, the complete restoration of all contaminated zones to drinking water quality standards may be impractical and very costly due to a variety of rate-limiting mechanisms, including dissolution, desorption and diffusion from mass stored in lower-permeability zones.

The spatial distribution of contaminant mass flux (the rate of mass flow per unit aquifer cross-sectional area) at even the geologically simplest of sites is highly variable, and high-resolution site characterization data support that the majority of contaminant mass transport occurs in a small fraction of the aquifer that is associated with the most conductive zone(s).

This mass flux is a measure of plume strength and associated risk to downgradient receptors. Accordingly, high-resolution characterization tools are now widely incorporated into flexible and adaptive investigations to map contaminant mass flux and support the design of efficient flux-focused remediation systems.

In response to the evolving mass flux prioritization, some regulatory programs are beginning to support remedial performance objectives that prioritize reducing contaminant concentrations in the most highly permeable/flux zones to achieve risk-based goals.

These risk-based remedial goals conceptually acknowledge the primary importance of reducing the mass discharge, defined as the integrated mass flux, and recognize that groundwater quality standards may not need to be universally achieved at some sites with a significant amount of mass in low-permeability (i.e., low mass flux) zone(s).

Despite the continuing development of many new flux-focused investigation technologies, the assessment of...
remediation performance continues to rely primarily on reducing contaminant concentrations at permanent monitoring wells. These wells are generally selected without consideration for the permeability of the zones they monitor, and they often have long screen lengths. Consequently, most monitoring programs are not designed to measure changes in mass flux but are primarily intended to monitor changes in the plume’s spatial footprint. Furthermore, they often focus on wells with high concentrations without specific consideration as to whether these wells are the best indicators of significant reductions in mass flux or risk to receptors.

While orders-of-magnitude differences exist in mass flux across most sites, data from monitoring wells screened in higher hydraulic conductivity media or highly fractured rock are inappropriately weighted equal to those screened within low hydraulic conductivity zones. This is unfortunate, as higher-conductivity

**Figure 1:** Contaminant concentrations in monitoring wells during the remediation of a source zone. The estimated pump-and-treat mass recovery (volume of water recovered multiplied by concentration, which can be seen as proportional to mass discharge) declined approximately four orders of magnitude. However, concentrations at three wells declined two to four order of magnitude (flux zone), while concentrations at a well screened in low-conductivity (K) zone declined only about half an order of magnitude.
Figure 2: Distribution of permeability from high-resolution HPT profiling data represented as overall distribution of aquifer soil profile (A) Raw data output from borehole hydraulic conductivity logging using a Geoprobe® HPT; (B) Raw data sorted from highest to lowest permeability (note that the y axis is reversed, decreasing upward); (C) CDF distribution represented as an overall fraction of boring thickness, 0.90, 0.99 and 1.0 benchmarks represent 90, 9 and 1 percent of permeability distribution; (D) 0.90, 0.99 and 1.0 benchmarks from (A) are represented as hydraulic conductivity from HPT profiling data.
media are more likely to contribute to contaminant transport, receptor risk, or serve as a water source.

Figure 1 provides an example where multiple orders-of-magnitude reduction in mass recovery from groundwater extraction was realized without corresponding concentration reductions at monitoring wells, particularly those screened in low-permeability (i.e., storage) zones.

Preferably, performance monitoring wells should more appropriately be defined using flux-based criteria. Simply put, at sites where the most-appropriate remedial goals are flux-focused, wells screened across very low-permeability zones should be eliminated from remedial performance monitoring programs because they produce unrepresentative and potentially misleading data. Fortunately, at most recently investigated sites, most monitoring wells already target higher-flux zones. However, for legacy sites where the monitoring network consists of a mix of monitoring wells screened within both high and low hydraulic conductivity zones, we support and encourage discussions between regulators and stakeholders to

**Figure 3:** Tracer washout data from a series of simple single-well tracer tests that measure local groundwater flux. These results quantitatively show that groundwater flux values in wells screened in unweathered rock were more than two orders of magnitude lower than wells screened in partially weathered rock (PWR) wells. These results provided the basis for an approved performance monitoring program that focused only on the PWR wells and excluded bedrock monitoring wells.
shift the focus of remedial performance monitoring to wells screened within zones that contribute most to the contaminant mass discharge and potential risk. This includes weighting both contaminant concentrations and groundwater flux at the individual monitoring wells. This could include a statistical approach that assigns weight to both contaminant concentrations and hydraulic conductivity of the individual monitoring wells.

As an example, based on high-resolution characterization data, the cumulative distribution function (CDF) sheds light on what fraction of an aquifer sequence is responsible for the variable groundwater flux for that sequence (Figure 2). More specifically, the CDF screening can identify where the first 90 percent, the next 9 percent and the last 1 percent of the groundwater flux moves through the vertical profile and thereby allows practitioners to set appropriate site-specific permeability targets for performance monitoring wells.

For legacy sites not investigated by high-resolution approaches, hydraulic tests or single-well tracer tests (Figure 3) can provide compelling quantitative data to support this type of assessment.

Monitoring programs are in place to document remedial progress towards cleanup goals and demonstrate when receptor risk has been eliminated. While the methodology has held consistent for decades, these programs need to align with our current understanding of transport, which will require current programs for many sites to undergo significant modification. Only wells that monitor significant mass flux zones should be included; wells that monitor very low-permeability zones should not be included, even if current contaminant concentrations are relatively high. In some cases, these wells could be abandoned or used for purposes not directly related to the remedial performance monitoring program.

Utilization of a monitoring network that adequately tracks progress related to not just cleanup goals, but also receptor risk is the true objective of any performance monitoring program. A flux-based well network is the most appropriate way to adequately guide remedial performance and demonstrate when performance objectives have been met.
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Thermal In Situ Sustainable Remediation (TISR™): Linking Renewable Energy to Sustainable Site Restoration

The effects of temperature on contaminant viscosity, dissolution and degradation are well documented for multiple chemical constituents, highlighting an important option to improve site restoration timeframes. As an example, comparison of petroleum hydrocarbons biodegradation rates in soils at 10 and 20 degrees Celsius shows a three-fold increase in degradation for benzene, toluene, ethylbenzene and xylene (BTEX). Numerous studies evaluating biodegradation rates at temperatures between 5 and 50 degrees Celsius have shown peak degradation rates at 30 and 40 degrees Celsius.

At many sites, these changes in temperature profile can be achieved via low-cost energy inputs and an efficient method to improve remedy performance. In addition to elevated rates of biodegradation, select chlorinated volatile organic compounds (VOCs), specifically chlorinated alkanes such as 1,1,1-trichloroethane, 1,2-dichloroethane and carbon tetrachloride, readily undergo hydrolysis reactions under elevated temperatures (60-80 degrees Celsius) — in situ temperatures that can also be achieved via economic infrastructure and reduced power utilization.

THE SUSTAINABILITY EVOLUTION

To strike a balance between high costs associated with high-temperature (HT) remediation and remedial timeframe, low-temperature (LT) thermal remediation
has been driven by sustainability solutions and incentivized by corporate financial pressures. Initial success stories around LT started to emerge after HT remediation operation would cease. These observations were due to the resurgence of microbial communities as temperatures cooled within or near HT treatment zones but remained in excess of background temperatures.

At the periphery of successfully completed HT remedial areas, residual low-level groundwater impacts were polished by these resurgent communities under thermophilic (40-80 degrees Celsius) or mesophilic (20-40 degrees Celsius) conditions that prevailed for months following subgrade heating discontinuation — and provided the basis for LT thermal enhancement of naturally occurring biodegradation. Consequently, practitioners started to design dialed-down versions of traditional HT thermal treatments. The key advantage is that while below-ground infrastructure (to deliver LT heat) is similar to the methodology of HT approaches, it requires significantly less energy input and considerably lower operational costs.

LT versions of electrical resistive heating (ERH) and thermal conduction heating (TCH) have now emerged as common applications by thermal vendors and some consultancies. While this is a great start, there are several examples of how LT thermal remediation has become even more sustainable, including the use of solar power and wind power to run an LT-ERH application; successful implementation of ex situ soil treatment via solar reflection, hot-water injection to facilitate hydrolysis of chlorinated alkanes and manipulation of geothermal heat storage to enhance contaminant attenuation. Arcadis’ Thermal In Situ Sustainable Remediation (TISR™) patent-pending technology allows focused subsurface delivery of solar and/or waste heat capture to achieve low-cost LT treatment.
TISR — SEEING THE EXTRAORDINARY IN THE ORDINARY

TISR is an inexpensive, practical and environmentally sustainable method for heating target subsurface contaminant treatment zones. The combination of thermal conduction and advection allows temperature propagation to enhance contaminant physical, biological and chemical attenuation processes.

TISR provides a system and method that collects solar heat or waste heat and transfers that heat via a closed-loop and a set of borehole heat exchangers (BHE) to subsurface soil in the proximity of the target contaminant treatment zones (Figure 1).

The target treatment zone may be composed of contaminated soil, contaminated groundwater in an aquifer, or industrial wastewater and/or solids. Either solar collectors or heat exchangers that collect waste heat from industrial processes may be used as the heat source.

The TISR process can elevate the ambient groundwater temperature to between 20 and 60 degrees Celsius using renewable energy, thereby enhancing hydrolysis or enhanced biodegradation rates by at least three times (depending on contaminant). Therefore, faster cleanup times, lower operational maintenance and 50 percent life-cycle cost reduction compared to traditional physical-extraction-based remedial alternatives are predicted.

Various other solar applications, if designed correctly for the available solar radiation (Figure 2), have been successful in colder, higher latitudes such as Canada and Germany. Current TISR projects exist at various stages of progression in New York, Colorado, New Jersey, Las Vegas, Brazil and Mexico.

The subsurface conduction of heat in TISR applications allows the technology to address many of the key challenges inherent to remediation, including high capital costs, slower degradation rates or asymptotic mass recovery due to low permeability zones, and complex hydrogeology. The utilization of solar power also offsets capital costs, conventional energy resources, fossil fuel use and consequent greenhouse gas emissions associated with other contemporary LT remediation methods.

CLOSE THE LOOP

The closed-loop heat transfer system operates in the absence of groundwater pumping or direct circulation of fluids through an aquifer. Energy delivery and heating of the subsurface occurs via thermal conduction due to relatively uniform thermal conductivity of soils and advection due to passive groundwater flow. The entire targeted treatment zone can be heated to the desired treatment temperature via a uniform and controlled process.

In 2014, based on FlexPDE® 2D modeling results, TISR was predicted to increase subsurface temperatures to 40-70 degrees Celsius. Since then, using empirical data, the model has been refined (2016) and applied to other sites to design for geographical and geological variations. Refinement of these model outputs based on emerging field pilot test data will continue as more data are collected.
PROGRESS THUS FAR

A two-year pilot test was concluded in 2017 at a site in New York (Site #1). Groundwater impacts were primarily composed of BTEX, 1,2,4-trimethylbenzene and 1,3,5-trimethylbenzenes. Baseline total VOC concentrations near the BHE points ranged from 17 parts per million (ppm) to 3 ppm (average 12 ppm). Concentrations during the last two sampling events (October 2017 and January 2018) have been below detection limits — after approximately 18 months of heat application. The system was shut down during most of 2017 to evaluate post-treatment rebound, during which an average VOC concentration of 0.001 ppm was reported in the test area.

TISR was able to reduce the VOC concentration by 99.9 percent in the two-year period (Figure 3). An estimated 330 kilograms of dissolved-phase VOCs were treated within an area approximately 300 square feet in size with a 10-foot vertical thickness. The treatment infrastructure is currently being moved and expanded to address a different location at the same site (Animation).

During late 2017 and early 2018, additional TISR operations were initiated at multiple sites. These include one chlorinated solvent site in Colorado where enhanced reductive dichlorination was successfully implemented over the course of the past 10 years. This TISR program is being implemented to further accelerate groundwater polishing to

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**Figure 3:** Total volatile organic compounds (BTEX and Trimethylbenzenes) groundwater concentration trends
offset approximately $500,000 to $1 million in estimated costs. This technology is also being implemented to address BTEX in New Jersey and Sao Paulo, Brazil, and to facilitate 1,2-dichloroethane and 1,2-dichloropropane hydrolysis at a site in Mexico.

**TISR ENHANCES OTHER REMEDIAL TECHNOLOGIES**

Physical extraction systems (e.g., ex situ groundwater treatment, air sparge/soil vapor extraction) remain conventional solutions to address non-aqueous phase liquid (NAPL) source zones. Increased temperatures have a positive influence on expediting source mass removal with select target contaminants exhibiting increased solubility and volatility at higher temperatures, more readily from the NAPL and viscosity decreases.

These properties all enhance the recovery of contaminant mass in the dissolved and vapor phases, thus significantly improving the treatment efficiency of a physical extraction system. Subsequently, remediation time and life-cycle costs of conventional physical extraction systems can be significantly reduced if implemented in conjunction with TISR.

More significantly, TISR offers significant benefit as an enhancement to biodegradation-based remedies, particularly for petroleum NAPL sites. An improved understanding of natural degradation processes and better NAPL conceptual site models have demonstrated the significance of natural source zone depletion (NSZD) in making progress toward remedial goals, and we now realize that NSZD can play a primary role in petroleum degradation. NSZD processes deplete mass both above and below the water table and target residual NAPL in addition to recoverable or mobile fractions. TISR implementation relies on leveraging these same processes to accelerate mass removal.

Enhanced bioremediation remedies also leverage NSZD and utilize terminal electron acceptor delivery (e.g., oxygen, nitrate, or sulfate) to increase treatment rates. These technologies (e.g., biosparging, bioventing, sulfate injection) all rely on electron acceptor injection to zones where LNAPL mass is present, and success is highly dependent on soil permeability.

In low-permeability or heterogeneous aquifers, LT thermal enhancement offers a unique advantage, as methanogenesis does not require external influx of electron acceptors, and the heat delivery mechanism relies on soils’ thermal properties, rather than permeability. This difference allows TISR to reach mass that permeability-based technologies cannot readily access — filling a key niche among remedial options (Figure 4).

By providing a sustainable, low-cost enhancement to NSZD, TISR can also help shift timeframes for attainment of remedial objectives into zones that are acceptable to stakeholders. For example, NSZD rate estimates may suggest that several decades to hundreds of years may be required to reach stringent remedial goals; however, incorporating TISR can potentially shift that timeframe into shorter ranges that are consistent with regulatory policies and site owner priorities.
Conventional Physical Extraction or Reagent Injection – Soil Permeability a Challenge

During Remediation
“Access” to lower-permeability zones is very limited and these zones serve as preferred storage areas for contaminant mass.

After remediation
Residual mass remains in storage areas and can lead to persistent dissolved-phase or vapor-phase exceedances.

TISR™ Fills a Key Niche – Soil Permeability Less Relevant

During Remediation
Heat transfer is not limited by soil permeability. TISR™ enhances biological, physical and chemical attenuation processes and NSZD in low-permeability areas as well.

After remediation
Residual mass is no longer present in storage areas to feed long-term dissolved-phase or vapor-phase issues.

**Figure 4:** TISR™ utilizes soil thermal properties to expand effective remediation into lesser permeable zones
By providing a sustainable, low-cost enhancement to NSZD, TISR can also help shift timeframes for attainment of remedial objectives into zones that are acceptable to stakeholders.
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The group of chemicals known as poly- and perfluoroalkyl substances (PFASs) has come under increasing scientific and regulatory scrutiny in recent years as more is understood about their toxicity, environmental persistence, aquifer mobility and potential to bioaccumulate. PFASs are used in a wide range of industrial applications and commercial products due to their unique surface tension and levelling properties. These applications include stain repellents for textiles and carpeting, grease-proof paper, water- and oil-resistant coatings, and mist suppressants used in metal plating. PFASs are also components of the Class B (flammable liquid) firefighting foams known as Aqueous Film Forming Foam (AFFF), Film Forming Fluoroprotein Foams (FFFP) and Fluoroprotein Foams (FP).

PFASs are a very diverse class of xenobiotic, “manmade” chemicals, united by the common structural element of a fully fluorinated alkyl chain, known as the perfluoroalkyl group (typically two to 18 carbon atoms in length). The whole PFAS molecule may be either fully (per-) or partly (poly-) fluorinated, but each compound always contains a perfluoroalkyl group.

The physicochemical properties of PFASs conferred by their high degree of fluorination and the strength of the carbon-fluorine bond lead to unique partitioning behavior (i.e., both hydrophobic and oleophobic properties) and thermal stability. These same molecular features also result in extreme recalcitrance and resistance to chemical attack, making PFASs extremely difficult to remove and destroy using conventional water or soil treatment technologies.

Perfluoroalkyl substances can also be referred to as perfluorinated compounds (PFCs), but are now more commonly termed perfluoroalkyl acids (PFAAs). They contain a fully fluorinated carbon chain with no additional carbon-to-hydrogen bonds in the molecule. The two primary classes of PFAAs are the perfluoroalkyl...
carboxylic acids (PFCAs) such as perfluorooctanoic acid (PFOA), and the perfluoroalkyl sulfonic acids (PFSAs) such as perfluorooctane sulfonic acid (PFOS). Less commonly assessed PFAAs include perfluoroalkyl phosphinic acids, perfluoroalkyl phosphonic acids, and the recently developed perfluoroalkyl ethers.

Polyfluoroalkyl substances comprise compounds that are susceptible to abiotic and biological transformation, forming PFAAs as terminal products (Figure 1). As a result, these compounds are often called PFAA precursors. The polyfluorinated compounds represent a much larger group of chemicals than the PFAAs, and most of them cannot currently be directly measured by conventional laboratory analytical methods that quantify PFAAs.

Since the issuance of the U.S. EPA public health advisories for PFOS and PFOA in drinking water in 2016, many sites have been under consideration for preliminary assessment, site investigation, and water treatment and environmental remediation of PFOS, PFOA and other PFASs.

PFOS and PFOA are typically the initial focus of attention from environmental regulations in many countries. In several locations, environmental regulations or advisory values also encompass some of the shorter-chain PFAAs, such as in Texas, Minnesota, Germany, Denmark, Switzerland, Italy, Canada and Sweden. Regulations considering PFASs beyond PFOS and PFOA are evolving quickly. In some jurisdictions, the use of PFAS-containing Class B foams such as AFFF has already been restricted significantly, with a recent ban on fluorosurfactant-based foams announced in South Australia in February 2018 [1, 2] and similar restrictions progressing in Washington State [3].

The U.S. EPA's long-term health advisory level recommending no more than 70 nanograms per liter (ng/L) or parts per trillion (ppt) for the sum of PFOS and PFOA in drinking water is an extremely low concentration, but it is similar to standards considered acceptable in drinking water in other countries and individual U.S. states. In New Jersey, enforceable maximum concentration limits (MCLs) of 14 ng/L PFOA and 13 ng/L PFOS have been recommended for drinking water [4] [5].

The number of analytes to include in site assessment and the use of advanced tools to comprehensively characterize PFAS mass require consideration of the phase of investigation or remediation, the potential for additional compounds to be regulated in the future, and the future risks associated with screening and treating for a subset of PFASs.
Arcadis has incorporated real-time analysis of PFASs and the use of advanced PFAS characterization tools into multiple site investigations and developed and tested new water treatment and remedial strategies for PFASs. This column presents laboratory and field-based updates from our 2016 article on advancements in site investigation and remediation for PFASs that Arcadis has conducted over the past several years.

**PFAS CHARACTERIZATION TOOLS**

In 2017, the U.S. Department of Defense released the Quality Systems Manual (QSM) 5.1, which contained guidelines standardizing the analysis of PFASs in soil and groundwater. While it is not as prescriptive as a U.S. EPA method, it has helped to standardize the accurate quantitation of a larger list of PFASs than can be measured using U.S. EPA Method 537 (i.e., measurement of 14 PFASs in drinking water). Multiple fixed laboratories are now accredited to conduct analysis of PFASs in soil and groundwater under QSM 5.1, including a mobile laboratory offered by Cascade. The mobile laboratory operates similarly to a fixed laboratory but can be brought onsite for real-time analysis.

Arcadis has now used Cascade’s mobile laboratory at multiple U.S. sites to enable real-time analysis of PFASs in groundwater and soil. Splits of whole groundwater samples and sample extracts (i.e., samples processed through a solid-phase extraction step) were analyzed at both the mobile laboratory (QSM 5.1-accredited) and a fixed laboratory (QSM 5.0-accredited) for PFOS and PFOA (Figure 2a-b). Sample extract concentrations agreed very well for both PFOS and PFOA, suggesting that the QSM 5.1-approved procedures followed through the extraction step by the mobile laboratory led to consistent analytical results between a fixed laboratory and the mobile laboratory.

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**Figure 2**: Comparison of PFOS (a) and PFOA (b) concentrations in five groundwater samples analyzed by a QSM 5.1-accredited mobile laboratory and a QSM 5.0-accredited fixed laboratory.
For the whole sample analysis, analysis of four of five samples agreed well for PFOS, although a significantly different PFOS concentration was measured in two co-collected groundwater samples, possibly due to actual sample variability. The fixed laboratory under quantified PFOA in whole-sample analysis compared to the mobile laboratory, which may have been the result of procedural differences between the fixed laboratory’s procedures and requirements under QSM 5.1. Overall, comparability was seen to be reasonable, and the accreditation of Cascade’s mobile laboratory provided confidence in the groundwater analytical results. The mobile laboratory offers significant advantages when mapping a PFAS plume and source area and planning strategic groundwater extraction. Arcadis intends to use the laboratory for future projects requiring detailed site characterization.

To measure the total mass of PFASs, including the PFAA precursors, Arcadis has been applying the Total Oxidizable Precursor (TOP) Assay [6] for analysis of soil and groundwater at multiple sites. Using this technique, samples are oxidized with hydroxyl radicals using thermal activation of persulfate under highly alkaline conditions. Precursors are converted to a mixture of PFAAs after treatment with TOP Assay. Prepared samples are measured by liquid chromatography-tandem mass spectrometry (LC-MS/MS) before and after oxidation. The amount of PFAAs produced is roughly equivalent to the total concentration of PFAA precursors in the sample. Detection limits are in the low range. The information that TOP Assay can provide about PFAS levels in samples is demonstrated with five groundwater samples collected from a fire training area (FTA; Figure 3). TOP Assay is useful both for confirming the
Overall scale of PFAS contamination present and confirming complete PFAS or PFAA precursor absence.

Measurement of PFSAs, PFCAs, a constrained list of PFAA precursors and PFAAs evolved from the TOP Assay allowed more-comprehensive assessment of PFASs in soil and groundwater at an FTA at different subsurface depths. The site is characterized by low-permeability soils and up to 2 meters (m) of annual variation in groundwater levels. From assessment of the types and relative levels of PFASs measured (Figure 4a-b), it was concluded that a foam made with a characteristic electrochemical-based chemistry (i.e., PFOS dominant), vs. a fluorotelomer-based chemistry, was predominantly used at the site. Precursors at the site made up a higher percentage of PFASs in groundwater (Figure 4a; 25 percent to 40 percent, 2.3 m to 7.7 m) than in soil (Figure 4b; 12 percent to 25 percent, 0 m to 1.8 m), and generally declined as a proportion of total PFASs with increasing depth, suggesting they are less mobile than the PFAAs at this site. Nearly 98 percent of total PFAS mass in soils was estimated to occur within the top 1.8 m of unsaturated soils. The use of comprehensive analysis tools provided confidence that the location of the PFASs was correctly mapped. Thus, remediation can be targeted to the most-impacted zones.

Tools such as TOP Assay are well suited to conduct detailed site characterizations. They may be particularly important in the context of releases that are largely fluorotelomer, where many of the PFASs released may not be directly quantifiable.

Figure 4: Groundwater (a) and soil (b) profiles of PFASs measured at a fire training area at different depths
In addition, when developing remedial strategies, assessing the treatment design considering a more-comprehensive suite of PFASs facilities a more future-proofed approach. The efficacy of a treatment’s performance for the less-sorptive short-chain PFAAs and PFAA precursors may need to be considered if they pose a potential risk upon discharge.

CONCEPTUAL SITE MODEL DEVELOPMENT USING ADVANCED PFAS CHARACTERIZATION TOOLS

Class B firefighting foam formulations are composed of many PFASs that are PFAA precursors. Unlike the PFAAs, these species are not strictly anionic, as some contain multiple types of charges (zwitterionic), and some are positively charged (cationic). The zwitterionic and cationic PFAA precursors are largely undetected by conventional methods, including enhanced PFAS methods governed by QSM 5.1. A significant mass of PFAA precursors in addition to the PFAAs have been detected in both AFFF-impacted groundwater and soil (Figure 4a-b). A conceptual site model (CSM) describing PFAS fate and transport at an FTA is hypothesized and presented on Figure 5, and a description follows.

Cationic PFAA precursors (and some zwitterions) will be retained in the soils at the source zone via strongly binding ion exchange processes. The source zones are anticipated to be strongly reducing and anaerobic as a result of the presence of residual hydrocarbons used in firefighter training. Under these redox conditions, cationic precursors will strongly sorb and slowly biotransform to simpler anionic PFAA precursors and PFAAs. Anionic PFAAs and PFAA precursors will migrate away from the source and, as they enter a more-aerobic geochemistry, the hidden anionic PFAA precursors will be more readily biotransformed to PFAAs, resulting in in situ generation of detectable PFAAs. PFAAs will not break down further and will continue to migrate as a plume with shorter-chain PFAAs generally migrating farther.

Figure 5: Diagrammatic CSM for a Class B firefighting foam impacted FTA

Defining the “risk driver” PFASs allows transparent decision-making to define which PFASs are in need of treatment at each site and, thus, focuses the remedial objectives.
SOLUTIONS FOR PFAS-IMPACTED SITES

The current state of the practice of remediation of PFASs is a treatment-train concept, primarily focused around reducing the treatment volume and concentrating the PFASs to be destroyed. In the absence of any biodegradation, and with significant challenges to practical field-scale chemical degradation, the viability of destruction-based treatment for PFASs increases when applied to lower volumes and higher concentrations of PFASs. For water treatment, PFASs may be concentrated through adsorption or separation-based technologies: those technologies that exploit electrostatic and/or hydrophobic adsorption or partitioning to the gas-liquid interface. For soil treatment, PFASs may be concentrated through temperature-induced volatilization or through soil washing. Due to the extreme recalcitrance and mobility of the PFAAs, typical PFAS groundwater remediation entails transitioning from large-volume and low-concentration PFASs to more concentrated forms. This summary will explore optimization measures for adsorption-based remediation, highlight a case study using the emerging technology of ozofractionation to enhance the separation of PFASs from water, and lastly discuss promising destruction-based technologies for the residual waste stream. The focus of this discussion will be on water; however, several developing strategies for source-zone soil and regenerating adsorbents that have reached their sorption capacity exist. Relevant treatment technologies for soil and groundwater are presented on Figures 6A and 6B, respectively.

Adsorption-based removal of PFASs from water is currently being implemented using granular activated carbon (GAC) or ion exchange resins (AX), where GAC represents primarily hydrophobic adsorption, and AX can represent both hydrophobic and electrostatic adsorption. These conventional adsorption-based technologies are moderately effective at removing PFOA and PFOS from water. However, when it comes to the broader class of PFASs (including short-chain PFAAs and variably charged polyfluorinated precursors), the effectiveness of these adsorption technologies is either reduced
or largely unknown. For example, Dickenson and Higgins demonstrated that perfluorobutanoic acid (PFBA) was minimally adsorbed by GAC, with nearly immediate breakthrough [7], while Xiao et al. postulated marginal effectiveness of GAC to remove PFAA precursors [8]. AX has demonstrated some affinity for short-chain PFAAs [9], but cationic and zwitterionic polyfluorinated precursor removal is largely unstudied in the literature. In typical anion exchange, anionic compounds are removed from solution through electrostatic adsorption, which is applicable to cations or cation-dominated zwitterions. Aside from the challenges and unknowns of adsorption-based removal of short-chain PFAAs and PFAA precursors, the efficiency associated with PFOA and PFOS removal using GAC and AX typically results in less than favorable operation and maintenance (O&M) costs. The low to moderate affinity of PFOA and PFOS for the adsorbents and ppt removal targets can lead to unacceptable PFOA and/or PFOS breakthrough. Therefore, a focus on optimizing PFOA and PFOS removal using these conventional adsorbents is relevant. For example, natural organic matter (NOM) — often measured as total organic carbon (TOC) — may lead to faster breakthrough of PFOA and/or PFOS. In rapid small-scale column testing (RSSCT) conducted by Arcadis, an order-of-magnitude increase in TOC from 0.3 to 3 milligrams per liter (mg/L) resulted in a 75 percent reduction in throughput before breakthrough. Pre-treatment to remove NOM or change its interaction with GAC may improve PFOA and PFOS removal. For example, slight alkaline adjustments to the influent pH will deprotonate common organic acids while having no effect on the affinity of PFOA or PFOS for GAC. This imparts polarity on the organic acid, decreasing its affinity for the GAC.

**Figure 7:** Ten-year life-cycle costs comparing 2,000-, 10,000- and 20,000-pound (lb) GAC vessels (two vessels each, installed lead-lag). Although 10,000-lb vessels have a higher capital cost, there is a comparable O&M cost with far fewer GAC changeouts than the 2,000-lb vessels that may be advantageous in remote locations or where GAC changeouts cause disruptions to site activities. In addition, the influence of TOC removal on the O&M costs is clear, with considerable reductions in O&M costs for lower TOC concentrations.
Other optimizations for GAC include appropriately sized GAC vessels for a given application, less-dense and equal-or better-performing forms of GAC (e.g., subbituminous coal versus bituminous coal), and increased mesoporosity (e.g., bituminous coal and over-activated coconut shell). Often, the capital cost of a GAC setup may influence the selection of the vessel size without consideration of more-frequent changeouts. Less-dense GAC that performs slightly better or equivalently to denser GAC can decrease the cost of changeouts because GAC is typically charged on a per-weight basis. RSSCT data generated by Arcadis and commercial laboratories suggests that a greater percentage of mesopores enhance PFOA and PFOS removal when compared to GAC with a greater percentage of microporosity. A comparison of capital expenditure and associated O&M costs considering the influence of variable concentrations of TOC is presented on Figure 7 and suggests that, over a 10-year operation period, 10,000-pound vessels have a comparable O&M cost and considerably fewer changeout disruptions than the 2,000-pound vessels. Although adsorption-based remedies are deployed rapidly, easily understood, and readily available, they come with a difficult-to-predict continuous O&M cost that needs to be considered and optimized.

An emerging technology, termed ozofractionation, has demonstrated the ability to remove >99.99 percent of total PFASs in a recent field-scale implementation in Australia. Ozofractionation exploits the physicochemical tendency of PFASs to partition to the gas-liquid interface, concentrating them in a resultant foam. The concentrated foam is separated from the treated water, achieving a reduction in the contaminated volume. Ozofractionation represents the state-of-the-practice treatment-train concept for PFAS removal from water by combining the foam fractionation step with an adsorbent or filtration polish with the intention of reducing the resultant volume for destruction and greatly increasing the concentration of the waste foam. A photo of the ozofractionation system from the field-scale implementation in Australia is presented on Figure 8.

At the subject field site, a large-volume release to a sewer of proprietary, C6-dominated AFFF occurred. To manage the release, the sewer lines were flushed and 4 million gallons of high-chemical-oxygen-demand, high-PFAS-concentration wastewater were isolated and stored for treatment of PFASs. The staged water requiring treatment contained a maximum total PFAS concentration measured post-TOP Assay of 3.95 million ppt; the targeted discharge limit was less than 250-ppt total PFAS, also measured post-TOP Assay. Because the proprietary AFFF included numerous polyfluorinated precursors, analyzing the wastewater without TOP Assay would have resulted in a significant underreporting of the total PFAS concentration. For example, in the abbreviated data set presented in Table 1, the total PFAS concentration measured pre-TOP Assay was 7,480 ppt, while the total PFAS concentration measured post-TOP Assay was 28,800 ppt.

Targeting remedial technologies for PFASs means exploiting their physicochemical properties to optimize their removal and destruction. PFASs tend to migrate to a gas:liquid interface so using this property enables comprehensive removal via ozofractionation.
The results summarized in Table 1 demonstrate >99.99 percent removal of PFASs using ozofractionation and nanofiltration. This demonstration shows that ozofractionation can be a power treatment technology for the removal of PFASs from water.

After either adsorption-based or separation-based removal technologies have created a smaller volume, higher-concentration PFAS waste stream, the next step in the treatment process is destroying the concentrated PFAS waste. Although no destruction-based technology is presently commercially viable, there are several in the developmental phase, each with its own set of operational challenges. Electrochemical treatment involves the chemical reduction and defluorination of PFASs via direct electron transfer to PFASs that have affixed themselves to an anode in an electrochemical cell. Some challenges associated with electrochemical treatment include high energy demand; long treatment residence times; secondary water-quality concerns such as the formation of perchlorate, hexavalent chromium and bromate, acid-based adsorption of PFAAs to the anode masking true destruction; and reduced effectiveness on partially defluorinated daughter products.

Sonolysis is another form of destruction-based treatment. Sonolysis, as it relates to PFAS treatment, is the creation of microbubbles generated through ultrasound application to water. The successful rarefaction and compression of the microbubbles leads to cavitation, which can facilitate generation of point sources of plasma at temperatures over 5,000 degrees Kelvin, as the bubbles collapse. The heat generated is not transmitted efficiently to the aqueous matrix, so the system can be maintained at 35 degrees Celsius, with little energy needed for cooling. PFASs partition to the gas-liquid interface of the microbubbles and, upon the plasma-associated bubble collapse, are destroyed through pyrolysis. Some challenges associated with sonolysis include high energy demand, long treatment

Degradation of PFASs using biological and conventional chemical treatments is very challenging due to the strength of C–F bonds.
residence times, interferences from co-contaminants and dissolved ions, and the complexities of reactor scale-up using multiple transducers. While destruction-based technologies exist for handling the low-volume concentrated PFAS wastes, it is clear their field-scale practicality is still under development.

Assessing and remediating PFASs present many challenges. First, there are the typical challenges associated with remediating any site, such as correctly identifying the source zone and the hydrostratigraphy that drives plume distribution. Second, measuring PFASs is more challenging than measuring other contaminants, such as 1,4-dioxane and trichloroethene, because the commercial methods usually applied measure only a small portion of the PFASs that are likely to be present. A significant portion of PFAS mass will be overlooked if only the PFAAs are measured. The TOP Assay is a pragmatic and comprehensive assessment tool for total PFAS measurement that has recently become commercially available.

Finally, the options for remediation are limited compared to many other common contaminants due to the general lack of volatility of most PFASs and the total lack of biodegradation of the whole contaminant class. Groundwater extraction and ex situ treatment with GAC or AX is the current state of the practice for treating PFAS-impacted groundwater, but significant amounts of GAC are needed to treat PFOA and/or PFOS, and GAC’s efficacy is reduced for short-chain PFAAs and potentially many PFAA precursors. The evolution of technologies to remove total PFASs, such as the recent deployment of ozofractionation, underscores how treatment technologies are developing quickly to comprehensively and cost effectively treat total PFASs in aqueous waste streams. Excavation of source soils and subsequent landfilling and incineration is extremely costly. Although other options for soil treatment exist, they are largely unproven and conceptual, so there are significant opportunities to develop soil treatment processes for PFASs.

Table 1: Full-scale ozofractionation system performance data
References


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Ian Ross, PhD, is a biochemist and remediation technical expert with 23 years of experience. In 2011, he won a Brownfield Briefing award in the UK for designing and implementing the world’s first in situ remediation of carbon disulfide using activated persulfate. In 2012, he won another Brownfield Briefing award for designing a combined soil washing and chemical oxidation project for a contaminated landfill. Ross’ recent focus has been on developing in situ remedial solutions for poly- and perfluorinated alkyl substances (PFAS). He was involved in the development of the CONCAWE PFAS guidance document published in 2015.

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