A Fellow Speaks: Perspectives Gained Across Boundaries

Industry Experience Driving Fundamental Research in Hydrogeology

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It is an honor to be selected as an AGU Fellow and join such an esteemed group of colleagues and I appreciate the nomination and support. I feel privileged to have been taught and mentored by so many smart and capable people at all ages and stages of my life. This is a great opportunity to reflect and review some highlights in my past that most strongly impacted where I am today.

If I trace my career path to its headwaters, it takes me back to Allegheny College, a small liberal arts college in northwestern Pennsylvania. I liked the sciences and math, but picking a degree program overwhelmed me until I stumbled into an evening lecture about two novel interdisciplinary environmental science programs: one was natural science-based and the other a mix of natural and social sciences. Environmental and interdisciplinary programs are common today, but not so in the 1970's, when I went to college. An environmental science program focused on water resonated with me given that I grew up on a dairy and cash-crop farm where we were always mindful of water and soil quality for growing crops and milking cows. As kids on the farm, we were aware of the weather, the amount of water in the cistern or the position of the water table in our well and cognizant of the operation of our septic system and waste streams. My advisor and mentor, Dr. Samuel Harrison, was a hydrogeologist, and I was hooked on groundwater after I took his geomorphology course. He took us out to the field to pound piezometers along a point bar in a local creek so we could understand hydraulic head and how ground- and surface waters are linked. I learned the importance of field-based teaching and appreciated different perspectives coming from varied disciplines like biology versus chemistry, geology, or economics. I also learned the benefits of interdisciplinary degree programs that provide distinct perspectives and insights that come from integrating these fields. Another critical piece of advice from Dr. Harrison was to "take as much math as I could possibly stand!" My minor in math most likely opened the door for me to get a master's degree in Engineering, which I pursued at Duke University immediately after getting my undergraduate degree.

My supervisor at Duke University, the late Dr. Aarne Vesilind, was also a pioneer in his field. He was an expert in water and wastewater treatment and promoted a modern view of environmental engineering as a distinct discipline from other classical engineering programs and wrote several textbooks in this diverse field. Dr. Vesilind and his colleagues cared little whether you were coming from a natural science or engineering undergraduate program because they viewed the environmental engineering profession as necessarily interdisciplinary. I benefitted from this enlightened view, as did other students in the program. We took core undergraduate engineering courses upon our arrival, but were also expected to learn the engineering principles and terminology along with the advanced subject material in our graduate courses and research. As many experience in graduate school, my student cohort became my most important teachers as we bounced ideas off of one another to solve problems with experimental design or data analysis. We were a small cohort of 12 environmental engineering graduate students from different regions of the country, different size universities and various undergraduate disciplines, which greatly enhanced our abilities as a team. After this experience, although excited to pursue a PhD, I found my interests still too numerous and decided to obtain some 'real world' work experience to gain insights and a personal perspective.

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Most jobs in the environmental profession are with the consulting service industry or within regulatory agencies. I did discover, however, that large corporations often manage their own waste streams and employed scientists and engineers within their corporate structure. This was the nature of my first permanent job in the Health, Safety and Environmental Division at Eastman Kodak Company from 1985 to 1991. Eastman Kodak was a chemical-based manufacturing company in operation since 1878. Compliance with the then new environmental regulations such as RCRA (1976) and Superfund (1980) were beginning to influence the amount of work focused on groundwater monitoring, which was the area that interested me most.

During my first week at work I met the consultant advising the company on groundwater monitoring and he wondered if I had heard the term "DNAPL" before? I had not! He explained that many of the chemicals used at Kodak were Dense, Non-Aqueous Phase Liquids, requiring wells to be drilled deeper than the water table and might change the size of the contaminant plumes currently mapped at the site. This was presented to my management and changed the nature of my job overnight. The groundwater contamination issues at the > 2,000 acre facility were complex because these chemicals could sink deep below the water table through fractures in the sedimentary rock.

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Over the more than 110 years of plant operations, there were many areas on-site, adjacent to residential neighborhoods within an urban area, where inadvertent leaks and spills created contamination requiring investigation and remediation. It was a time when the hydrogeology profession had little experience with these contaminants, especially in fractured rock. The regulations were new and not tailored for the nature of the contamination issues being discovered. At Kodak, I was mentored by Dr. Richard Poduska, who was committed to identifying the nature of the problem and developing logical and reasonable solutions that were science-based. This was a time of extreme uncertainty and we all learned fast, working across various disciplines from manufacturing to environmental compliance, legal to corporate communications and community relations. I was now working with an even more diverse, multi-generational team including folks with different amounts of experience, which sometimes helped and other times hindered our progress.

As an environmentalist, I was empowered by the corporate commitment to do the job properly, seeking both short term and long-term solutions that were not readily available. My job was focused on the science and I hired advisors that were the best experts in the profession. Dr. Robert Mutch introduced me to the importance of chemical diffusion as contaminants travel through fractures in clays and rocks presented in the modern textbook Groundwater by Freeze & Cherry (1979), although with no mention of chlorinated solvents. We installed wells to multiple depths in rock, innovated sampling methods to show these organic solvent contaminants were stored in the low-permeability rock matrix, and worked with the in-house environmental chemistry lab to develop better analytical procedures for the site-specific contaminants; all to try and understand the evolution of concentrations in wells after drilling or reported spills. We learned many things but still had more questions than answers. Eventually, I left Kodak to pursue my PhD at the University of Waterloo, motivated to find answers to numerous questions about organic solvent contaminant behavior, noting that some of the complexities were due in part to the characteristics of fractured rock.

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At Waterloo, I was encouraged by my supervisors, Drs. John Cherry and Robert Gillham to work in clays as an analog to sedimentary rock. As I prepared for the field release experiments in fractured clay, my diffusion calculations showed that the typical-sized fractures in clays would dissolve the DNAPL volume quickly, causing its disappearance, consistent with observations at Kodak. The implications were substantial and led to new insights about the future of these contaminated zones. We set out to show mathematically and prove with field evidence, starting with controlled release experiments, how diffusive transport controls contaminant behavior. We then sought to address this as evidence for flux as well as transport,

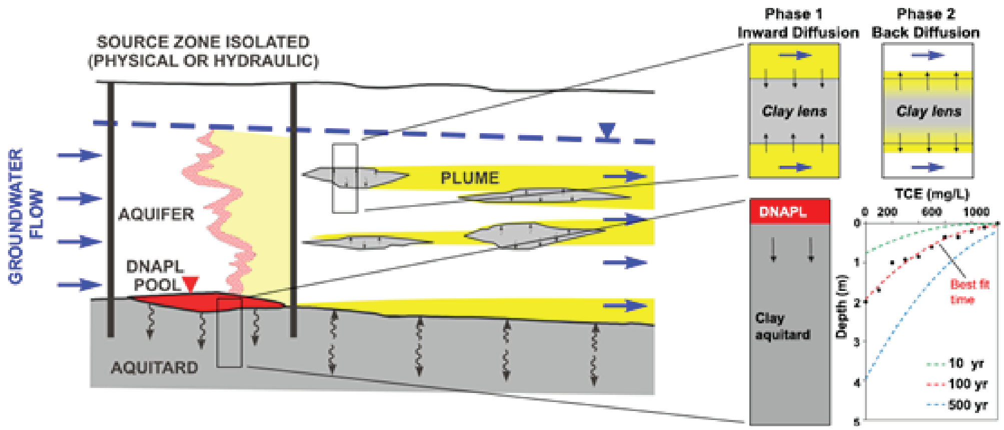

Figure 1. Persistent plume after complete source zone isolation due to back diffusion from clay lenses and aquitard. Profiles from cores inform source zone age and diffusion rates.

combining the engineering and hydrogeology frameworks. This insight became evident when the groundwater remediation industry was just getting used to the idea that DNAPL was going to persist forever and most of the remediation technologies were geared for DNAPL removal. The concept of source zone evolution was published in 1994 and 1997 as part of my PhD research, along with evidence for diffusion in water-saturated, low permeability natural clays (Figure 1). Diffusion haloes in clays were measurable over a few to 10's of centimeters to accurately measure mass transfer over periods of weeks to years. Collection of diffusion dominated profiles allowed quantitation of the mass transport by diffusion and both mass storage and flux enhanced by sorption, and were used forensically to identify DNAPL migration pathways in the Borden aquitard (Parker, 1996) and in a large column experiment with fractured clay (O'Hara et al., 2000).

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My next 12 years at the University of Waterloo as a research faculty member provided opportunities for me and my students to adapt these ideas to real-field sites with scenarios similar to those depicted in **Figure 1**, seeking direct field evidence of contaminant distributions in both low and high permeability zones and using diffusion haloes forensically to inform timescales since contamination began (Parker and Cherry, 1995; Parker et al., 2004) and quantitation of processes for predicting remediation time-scales prolonged by back diffusion (Parker and McWhorter, 1994; Chapman and Parker, 2005; Parker et al., 2008). Furthermore, I also recognized that chlorinated solvents are ideal tracers of the flow system and contaminant migration pathways where the combined effects of specific processes provide fundamental insights. This is because they are mobile, relatively persistent, can be analyzed over six or more orders of magnitude, have distinct and known decay products, entered the groundwater system at many sites decades ago, and traveled under natural-gradient conditions.

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What was essential in this approach was the importance of selecting industrial sites with both the appropriate hydrogeologic and contaminant conditions and sufficient simplicity to allow understanding of processes quantitatively; for example, sharp interfaces between lithologies with higher and lower permeabilities like the clayey aquitard and overlying sandy aquifer with DNAPL accumulation shown in Figure 1. Continuous cores with good recovery and retention of pore fluids were essential for measuring contaminant profiles over the diffusion transport distances (Parker, 1996; Parker et al., 2004) and identified the critical processes controlling contaminated site evolution, where it was evident that diffusion in and out of the low permeability zones was important over multiple decade time-scales (Chapman and Parker, 2005; Parker et al., 2008, 2010). Not all field sites are well suited for advancing fundamental concepts; sites were carefully selected, often complementing each other with distinct geologic and contaminant conditions (e.g. Parker et al., 2003; Guilbeault et al., 2005; Parker et al., 2008; Meyer et al., 2014).

I also found ways to transfer my experience with chlorinated solvent behavior in fractured clay to sedimentary rock, as answers to my questions from my Kodak

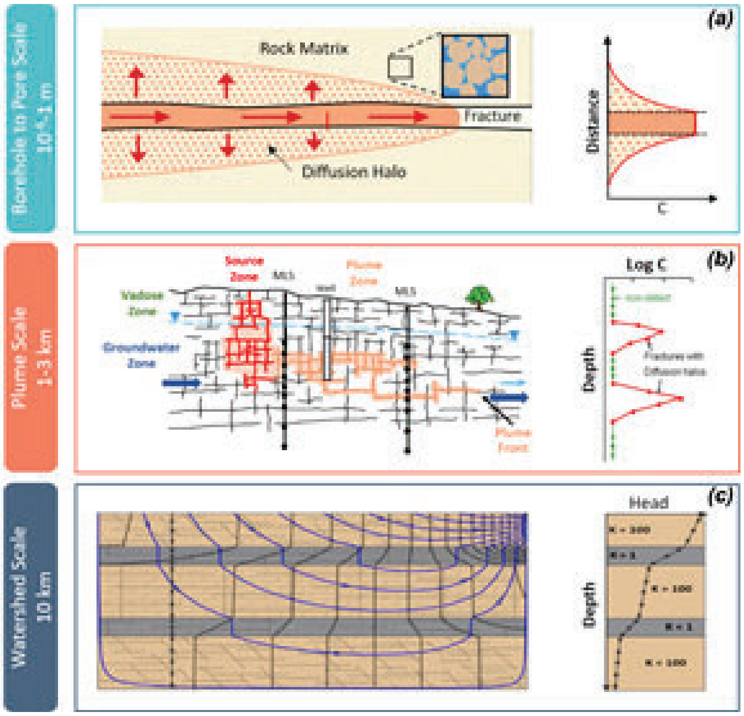

Figure 2 Site complexity requires field data at multiple scales to inform processes and constrain uncertainty.

days remained elusive and approaches for resolution of these questions more expensive. Given that controlled release experiments in bedrock were unlikely to be approved by any site manager or jurisdiction, I continued my pursuit for industrial sites with a certain style of contaminant and hydrogeologic condition to be suitable as 'field laboratories'. I adapted the use of contaminant concentration profiles to identify diffusion haloes in clays to the scale appropriate for haloes in fractured sedimentary rock, complemented with depth-discrete hydraulic data informing the flow system (Figure 2).

The rock matrix porewater method we used at Kodak to show diffusion was too tedious to use at the resolution needed to assess the occurrence of haloes away from many visible fractures in core. I worked collaboratively with my chemistry colleague Dr. Tadeusz Gorecki to advance the extraction of aqueous and sorbed mass from low-permeability clay and rock to be efficient and robust (Dincutoiu et al., 2003, 2006) to identify preferential advection pathways by their diffusion haloes into the lower permeability matrix used by O'Hara et al. (2001) in a fractured clay column experiment and Sterling et al. (2005) for TCE in sedimentary rock. It became readily apparent from these rock core TCE concentration profiles (**Figure 2b**) that there were many more hydraulically active fractures participating in the flow of DNAPL and transport of contaminants than deduced from open hole flow meter measurements of fractures (Sterling et al., 2005; Goldstein et al., 2004; Parker et al., 2018) and how vertical flow typical in open bedrock boreholes skew our measurements of hydraulic head and concentrations, leading to poor interpretations due to cross-connection (Sterling et al., 2005; Pehme et al., 2013; Meyer et al., 2014).

This showed me the need for better field methods. I had begun working with Carl Keller with Water FLUTeTM multilevel systems in the late 1990's. Conversations with Carl also lead to sealing boreholes with blank liners (water FLUTes without ports) in 2000 at one of my research sites, and many important permutations of ideas and data sets from our collaborations occurred (e.g. Cherry et al., 2007; Keller et al., 2014; Keller, 2017). Temporary borehole seals lead to temperature logging in sealed boreholes with and without added heat (Pehme et al., 2007; 2010; 2013) which provides insights about the position of hydraulically active fractures under natural gradient flow conditions. The insights form these high-resolution profile data sets complemented the rock core information from the same boreholes. Options for measurements in these boreholes continued to expand with temporary deployment of sensors and samplers behind liners (Pehme et al., 2014) and deployment of fiber optic cables in bedrock boreholes for active distributed temperature sensing for identifying flow and quantifying groundwater fluxes (Coleman et al., 2015; Maldaner et al., 2019) and improved coupling for distributed acoustic sensing vertical seismic profiles (Munn et al., 2017).

Field measurements at select contaminated industrial sites provided opportunity for fundamental research by pursuing multiple depth-discrete, high-resolution sampling methods. These were initially focused on continuous cores (**Figure 2**), where the creation of measurable diffusion-haloes in sedimentary rock (**Figure 2a**) show contaminant storage and identify, which fractures are hydraulically connected and important in the system for migration or remediation. This sampling strategy shows mass distributions along the full length of the borehole that can be aligned with fractures and lithology (**Figure 2b**). Multiple lines of evidence also bring transmissivity and groundwater flow profiles from various field methods discussed previously together to determine advection versus diffusion controls on migration rates, including DNA extractions on rock core samples (Lima et al., 2012), hydrochemistry, and isotopes through collaborations with Dr. Ramon Aravena providing supporting evidence for attenuating reactions (Pierce et al., 2018). These interpretations, however, are highly dependent on the direction and magnitude of groundwater flow, which can be complex. Ultimately, the flow system must be understood at a scale larger than the contaminated zone to produce reliable predictions of transport and fate. High resolution depth-discrete hydraulic head data provide 1-D profiles showing head loss over lower vertical hydraulic conductivity zones (Figure 2c) identifying the position, thickness and lateral continuity of aquitards that inform the 3-D groundwater flow system. Monitoring wells can then be constructed within this hydraulically-calibrated geologic framework with less cross-connection. These concepts of vertical head profiles as a direct measure of aquitard positions in sedimentary rocks was advanced by Meyer et al. (2008, 2014, 2016), with insights regarding thin aquitard layers or surfaces due to fracture terminations influencing flow system conditions. Once a geologic system has been informed hydraulically, the newly informed hydrogeologic framework and calibrated tools (e.g. geophysical datasets) can help to design future monitoring systems across the 3-D system with better precision, accuracy and lower cost.

Studying these sedimentary rock systems for a sufficient amount of time allowed me to observe at many sites, the complete (or nearly complete) DNAPL dissolution in the subsurface source zone (Figure 3, stages 1 through 4), with diffusion having caused substantial mass transfer from the many well-connected fractures into the lower permeability rock matrix between fractures (where the mass resides in the dissolved and sorbed phases) over a few decades. Moreover, each of the plumes has evolved to a near-stationary position ("quasi-steady state" plumes) shown in stage 4 (Figure 3), primarily due to the combined influences of advection and diffusion enhanced by sorption throughout the source zone and plume. In advanced stages 5 and 6 (Figure 3), these plumes are shown to retreat toward the source zones, rather than flushing toward the receptor, based on evidence for very slow abiotic and biological degradation rates in the matrix. These similarities, found at all sites despite their different hydrogeologic, climatic, and contaminant conditions, form the basis of the general conceptual model (GCM) for chlorinated solvent source zones and their plumes in fractured sedimentary rock, as they evolve over a few

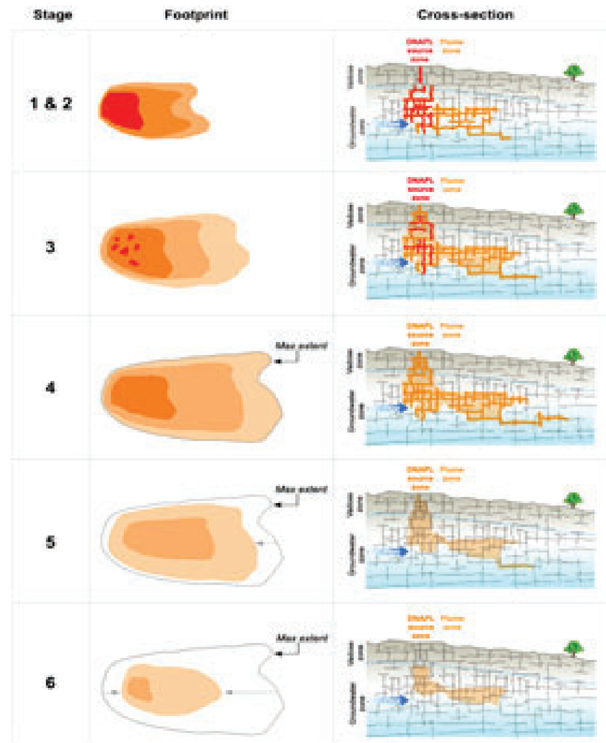

Figure 3 General conceptual model of source zone and plume evolution in fractured sedimentary rock. Stage 1-2: DNAPL quickly migrates through interconnected fractures, partially dissolves and is transported by advection in fractures and diffusion into the matrix. Stage 3: DNAPL in fractures has partially dissolved and contaminants diffuse into the rock matrix and the plume advances in direction of groundwater flow. Stage 4: DNAPL phase has completely dissolved away and plume front reaches the maximum downgradient extent. Stage 5: Diffusion with slow degradation causes plume front to retreat back toward the source. Stage 6: Diffusion with established degradation causes plume front to retreat

to several decades. The GCM summarized in Figure 3 represents typical plume characteristics and behavior for most sedimentary rock sites where initial DNAPL conditions have caused persistent bedrock contamination. This GCM and a field-based methodology for site characterization, referred to as the Discrete Fracture Network – Matrix (DFN-M) Approach, provides a fundamental framework to inform conceptual site models used to forecast future conditions and inform site remediation decisions (Parker et al., 2012).

In summary, my quest to better understand organic contaminant behaviour in various types of geology, I have used large columns of fractured natural clay for lab experiments and performed controlled DNAPL release experiments in field settings, but the essence of my work has been the study of organic contaminant distributions at actual contaminated industrial sites where the contaminants have been in the groundwater zone for decades. Time is a key factor that cannot be practically represented in the laboratory for the long periods that are relevant. I have found the key to progress is using high resolution measurement methods to discern diffusion patterns archived in the low permeability parts of whatever system is being studied. Fick's law of diffusion, like Darcy's law for flow, has been known since the mid-1800's. Like flow, diffusion manifests in complex groundwater systems with many facets. What I find so intriguing is the many relevant and important ways these two laws combine to govern the transport and fate of contaminants beyond what our initial intuition suggests.

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I was trained to pursue excellence and this was honed by both industry and academia. This allowed me to pursue fundamental research at industrial sites where the aged contamination in relevant hydrogeologic settings was not contrived. The contaminant mass distributions were controlled by natural processes over the time and distance scales of relevance. I only needed to convince a few site owners (manufacturing companies) that the research might provide useful insights that would help them manage their subsurface contamination more cost-effectively. The idea was to pursue fundamental research that was also practically relevant. The stage was set with robust regulations for mandatory cleanup with a "one size fits all" approach, written before the professional experience was advanced. Industry was caught in a bind, and they needed the effective solutions our profession could not deliver without experimentation.

I have been able to balance perspectives between industry and academia, providing fundamental process understanding that altered the professional practice along the way. My field research at these sites with complex hydrogeologic conditions would not have been possible without being extremely relevant to the contaminated site owners with obligations to stringent environmental regulations making this fundamental research highly relevant, justifying a significant funding commitment. I am grateful for the corporate donations that generated federal matching funds from the Government of Canada. The conceptual model for source zone evolution of chlorinated solvents in fractured porous clays and rock proposed in 1994 as part of my PhD has advanced to a six-stage model for the source and down-gradient plume to achieve a maximum extent within a few decades before attenuating and retreating back toward the source zone position (Figure 3), which is now being validated at several sites around the globe in different hydrologic systems.

The past 35 years has been an exciting era, with an interdisciplinary and highly collaborative approach to field-focused research aimed at testing conceptual models that engages students in hands-on training. I continue to pursue this work with numerous collaborative partners, colleagues and students of all ages, across many disciplines and cultures, combining insights from working with both industry and academia. The long-term nature of the problem has also spanned a few generations, and this has also enriched the experience, speaking to another element of team diversity. However, you will notice my career lacked many professional women mentors, a condition I hope changes in the future. **I would be remiss if I didn't acknowledge that there were numerous women role-models that I emulated regardless of their presence inside or outside my professional world; and many others, including my family, not mentioned by name.**

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