Complex Challenges at Light Non-Aqueous Phase Liquid Sites

Introduction

There are a variety of site-specific characteristics that can increase the complexity of managing a light non-aqueous phase liquid (LNAPL)-contaminated site. These site conditions may result in the need for specialized techniques to characterize and remediate the LNAPL in order to achieve remedial action objectives (RAOs) in a reasonable timeframe and cost.

A NAVFAC Remedial Project Manager (RPM) survey indicated that the top three challenges that add complexity to their LNAPL sites are: 1) the presence of co-contaminants, 2) highly-heterogeneous conditions, and 3) the presence of fractured bedrock (including karst). In addition, RPMs have requested assistance on strategies for addressing LNAPL sites within arctic regions and sites contaminated with long-chained hydrocarbons such as Navy Special Fuel Oil (NSFO), Bunker C, and heating oils.

This fact sheet presents an overview of these important challenges to support the Navy's approach in managing complex LNAPL sites and identifies conceptual site model (CSM) elements to consider for an improved understanding of these challenges. For each challenge, the current understanding of the effects it has on LNAPL distribution are discussed to aid in CSM development.

In addition, site characterization methods and considerations for remedial technology selection and design are presented to better address these site complexities, risks associated with LNAPL components, and the potential for LNAPL migration.

Complex Challenges at LNAPL Sites

Background

Challenge #1: LNAPL Sites with Co-Contaminants

Challenge #2: LNAPL Sites with Highly-Heterogeneous Lithology

Challenge #3: LNAPL Sites with Fractured Bedrock

Challenge #4: LNAPL Sites in Arctic Regions

Challenge #5: Long-Chained Hydrocarbon Impacts

Background

LNAPLs are among the most commonly encountered organic contaminants in the subsurface environment due to their pervasive use, historic disposal practices, and accidental releases during storage and handling causing potential risks to human health and the environment. Background information on addressing LNAPL sites is available in NAVFAC (2010) and Interstate Technology Regulatory Council (ITRC) (2009) including a summary of mass recovery, mass control, and phase change technologies. This fact sheet is focused on developing CSMs and LNAPL site management strategies for the complex site conditions noted above. There are two main drivers in LNAPL site management that should be addressed in the CSM:

- the potential risks posed to human and ecological receptors by mobile LNAPL (saturation-based risks), and the migration of any associated dissolved-phase and vapor-phase plumes (composition-based risks); and

- constraints on remediation selection, design, and operation, which often requires targeted investigation and pilot testing of the system (Contaminated Land: Applications in Real Environments [CL:AIRE], 2014).

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For each challenging site condition noted above (No. 1 to 5), additional insights are provided below related to how the CSM is impacted and how remediation may be constrained by these challenging site conditions.

LNAPL RISK DEFINITIONS

Two risk classifications have been designated for LNAPL source areas:

Composition-based risks = The risks related to the toxicity of the chemicals that diffuse and dissolve from the LNAPL;

Saturation-based risks = The potential for a LNAPL to spread within the pore space or within the transmissive fractures of bedrock (ITRC, 2009).

Challenge #1: LNAPL Sites with Co-Contaminants

LNAPL sites that contain co-contaminants present unique challenges for remedy selection and treatment train design, as certain technologies effective for treating LNAPLs might not be effective for treating a given co-contaminant. In addition, co-contaminants may pose a greater risk to human health and the environment than the petroleum constituents in the LNAPL. The remedies selected must be compatible with site conditions and RAOs for all contaminants of concern (COCs).

The nature and extent of the contamination may be different for different COCs, and must be accounted for when building the CSM and defining the target treatment zones. Common co-contaminants at LNAPL sites include chlorinated volatile organic compounds (CVOCs) and polychlorinated biphenyls (PCBs) that may be commingled with LNAPL from historic disposal or operational practices. In addition, physical and chemical changes due to the presence of LNAPL may alter aquifer geochemistry and facilitate dissolution of naturally occurring metals. The primary factor responsible for these changes is the biodegradation of petroleum hydrocarbons, which occur by a variety of microbial-mediated reaction pathways. These reactions typically cause more reducing conditions to develop within the aquifer and can also lower the pH and mobilize metals in soils.

Table 1 describes how these organic and inorganic co-contaminants may affect the LNAPL CSM and provides an overview of potential remedial management options.

Challenge #2: LNAPL Sites with Highly-Heterogeneous Lithology

The migration of LNAPL in the subsurface is controlled by several factors including the heterogeneity of the soils within the unsaturated (i.e., vadose) and saturated zones. LNAPL is typically distributed across the water table (or capillary fringe) interface due to its buoyancy and the nature of this distribution is greatly influenced by the lithology as described below.

CSM Considerations for Highly-Heterogeneous LNAPL Sites

Within a homogeneous aquifer, the LNAPL saturation distribution can be approximated by considering the capillary pressures of the various liquid phases and properties that relate fluid contents of the porous media to capillary pressures. Using this data, the LNAPL saturation profile at the water table interface can be modeled within a homogeneous isotropic unconfined aquifer under equilibrium conditions. This idealized LNAPL saturation profile is presented in Figure 1a for a homogenous aquifer (which assumes a shape similar to that of a "bell curve" or "shark fin"). The LNAPL saturation profile is quite different in a heterogeneous aquifer and highly dependent on the grain size of the layers in the formation. This concept is illustrated in Figure 1b where the numbered layers are meant to represent differing soil types with fine-grained layers shaded in grey and coarser-grained layers in white (CL:AIRE, 2014).
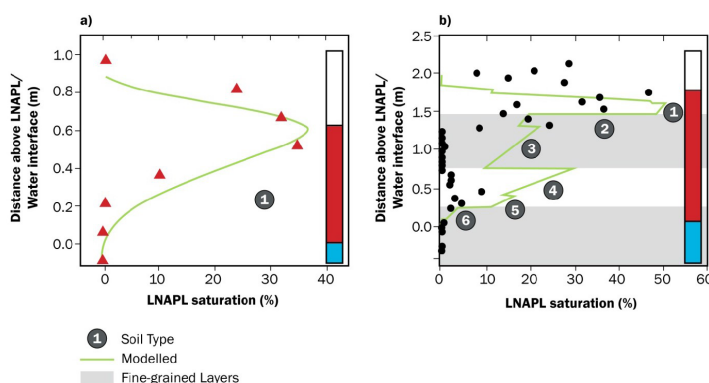

Figure 1. LNAPL Saturation near the Water Table

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Note: a) homogeneous case and b) heterogeneous case with finer grained layers (grey) and coarser grained layers (white)

Table 1. Co-Contaminant Considerations for LNAPL Sites

Constituent	CSM Considerations	Remedial Considerations
ORGANIC CO-CONTAMINANTS		
CVOCs	When CVOCs are mixed with LNAPL, its density may increase and viscosity may decrease. The increase in density limits the lateral spread of the LNAPL because it will be heavier with respect to balancing of the buoyancy forces. However, reduced viscosity potentially can render the LNAPL more mobile and enhance lateral spread. With respect to the vapor intrusion pathway, the plume cannot be managed as a petroleum-only source from a human health risk perspective as the LNAPL contains CVOCs.	Benzene, toluene, ethylbenzene and total xylenes [BTEX] can serve as electron donors that support reductive dechlorination. Newer releases of LNAPL that contain more soluble fractions of CVOCs, as well as lighter fractions of LNAPL, can support dechlorination. Older, weathered LNAPLs that have leached much of their soluble fractions, or longer-chained LNAPLs are less bioavailable and thus poorer electron donors. Toluene can also be used as a primary substrate in the aerobic cometabolic bioremediation of TCE. If a CVOC plume is co-located with LNAPL, then air sparging/soil vapor extraction can also be used to remove the CVOCs through phase change.
PCBs	PCBs may co-occur within LNAPL at some sites due to historic disposal of transformer oils or past waste disposal practices that caused commingling within waste lagoons. Alternatively, soils contaminated with PCBs may be flushed by a spilled LNAPL, causing a commingling of PCBs and residual LNAPL within the aquifer.	Total fluids pumping is the primary remedial approach for LNAPL contaminated with PCBs. Once removed, the LNAPL/PCBs will typically have to be managed under Toxic Substances Control Act (TSCA). Other alternatives include in situ smoldering, thermal conduction heating, and electrical resistance heating. In situ stabilization/solidification could be used as a mass containment approach for impacted soils and/or the LNAPL body.
INORGANIC CO-CONTAMINANTS		
Hexavalent Chromium, Cr(VI)	Hexavalent chromium [Cr(VI)] may be encountered as a co-contaminant at former industrial sites. Cr(VI) is acutely toxic and very mobile in groundwater. At LNAPL sites, Cr(VI) should generally be reduced to CrIII(OH)3(s) due to the reducing geochemical conditions induced by biodegradation of the LNAPL.	If dissolved phase Cr(VI) is prevalent within or adjacent to an LNAPL source, there are a variety of in situ technologies that can be used to contain or treat it including: in situ chemical reduction, anaerobic enhanced in situ bioremediation, permeable reactive barriers (PRBs), natural attenuation, and phytoremediation.
Arsenic (As)	In shallow aquifers, the background redox conditions are typically aerobic. Prior to LNAPL release, arsenic is likely present as As(V) (arsenate) which is strongly adsorbed to ferric iron oxyhydroxides (API, 2011). However, upon the onset of reducing conditions, naturally-occurring arsenic can be released from soils in the more mobile As(III) form. This is an issue where arsenic is bound to iron and manganese oxide coatings on the aquifer grains (such as glacial outwash sand and gravels).	A main concern with arsenic is containing the plume within the site boundaries. This can be accomplished either through hydraulic containment, aerobic remedies such as air sparging to oxygenate the aquifer to precipitate the arsenic, or permeable reactive barriers. Another in situ strategy includes precipitating arsenic as stable metal sulfides. Elevated concentrations of arsenic may also naturally attenuate through transformation of As(III) to As(V) and adsorption onto iron minerals downgradient of the LNAPL plume as redox conditions return to their background state.

In the case of highly heterogenous sites, high resolution site characterization (HRSC) tools can be used to refine the CSM and better understand the LNAPL distribution and the variability in the soil and bedrock structure. Tools that are useful to understand highly heterogeneous sites include the use of cone penetrometer testing (CPT) or hydraulic profiling tools (HPT) to understand the lithology. When used along with laser induced florescence (LIF) or membrane interface probe (MIP), these technologies will aid in understanding the variability of the LNAPL source term and how to best apply the remedial approach. For highly-heterogeneous sites, an improved understanding of the LNAPL distribution can increase the likelihood of remedy success. A summary of the various tools available to characterize unconsolidated media is provided in ITRC (2009) and CL:AIRE (2014).

Remedial Considerations for Highly-Heterogeneous LNAPL Sites

At highly heterogeneous sites, it is important to understand the heterogeneities with a reasonable degree of accuracy in order to support selection and design of effective remedial systems. Recovery and treatment by excavation and in situ mixing and stabilization is not impacted by heterogeneous conditions. Heterogeneity also has minimal impact on trenches and barriers. However, it is important that the trenches and barriers intersect the LNAPL plume in all lithologic units and all depths and are keyed into an aquitard. In situ technologies for plume treatment including chemical oxidation, bioventing, air sparging, as well as recovery via multi-phase extraction are impacted by heterogeneities to various degrees. For example, preferential flow pathways will be prevalent when using any technology that relies on the introduction or removal of a fluid and must be accounted for in the design. Low permeability semi-confining units can also impact the introduction and removal of fluids. Soil heterogeneities influence natural source zone depletion as LNAPL will have varying attenuation rates in lithologic units having differing permeability, hydraulic conductivity, grain size, and organic carbon content.

Challenge #3: Sites with Fractured Bedrock

LNAPL behavior and multiphase flow in fractured media is complex. The success of LNAPL remediation in fractured media depends highly on understanding how LNAPL moves and behaves in the subsurface.

CSM Considerations for Fractured Bedrock

The behavior of LNAPL in fractured bedrock is not necessarily intuitive and is not easily described using the principles and techniques adopted for more homogeneous porous media. Significant differences have been noted for how LNAPL behaves in fractured bedrock formations as follows:

- Fracture size, orientation, and connectivity typically dominate LNAPL migration and provide preferential flow pathways;
- Dipping fracture planes can increase initial LNAPL gradients and result in increased initial migration and greater overall lateral spread of LNAPL; and
- Initial LNAPL head pressures can propagate LNAPL deep into the saturated zone via vertical and/or sub-vertical fractures.

Fluctuations in groundwater elevation can be significant and may result in further expansion or lateral spread of LNAPL over longer time frames than for unconsolidated materials. The type of primary porosity will also greatly affect the LNAPL distribution. If a formation has a high primary porosity, then the LNAPL will behave similarly to the unconsolidated overbearing material; whereas, the less permeable the primary porosity, the more the LNAPL distribution is dominated by the secondary porosity of the fracture network. Table 2 provides examples of low and high matrix porosity bedrock environments.

As summarized in CL:AIRE (2014), the matrix rock may or may not have significant porosity and potentially some permeability; however, its fracture system will typically exert the dominant control on LNAPL migration. Within the partially saturated zone, LNAPL will move downwards through vertical and sub-vertical fractures under the influence of gravity. The steeper the fracture dip, the greater the influence of gravity and the lower the LNAPL retention (residual saturation). LNAPL will be immobilized in fully horizontal fractures. LNAPL flow also increases as fluid viscosity decreases and as fracture aperture increases. With greater LNAPL-water density contrast, vertical driving forces are increasingly resisted by LNAPL buoyancy that promotes lateral spread of the LNAPL at the water table and through water table fluctuations. Significant lateral LNAPL migration may also occur along dipping fractures in the partially saturated zone. Once at the water table, the LNAPL's accumulated weight will begin to depress the LNAPL-water interface within the fracture. In a water-wet system, LNAPL will enter a fracture only if the LNAPL-water capillary pressure at the fracture entrance is greater than the fracture entry pressure (CL:AIRE, 2014).

Table 2. Low and High Matrix Porosity Bedrock Formations

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Formation Characteristic	Flow Characteristics	Example
Bedrock – Low Matrix Porosity	Small aperture fractures	Granite/Igneous rock
	Large aperture fractures	Karst limestone
	Fracture and matrix	Cemented sandstone/gritstone
Bedrock – High Matrix Porosity	Large aperture fractures	Chalk
	Small aperture fractures	Shale/mudstone
	Fracture and matrix	Sandstone

As shown in Figure 2, water table fluctuations within fractured rock may significantly influence LNAPL entrapment and migration. A fluctuating groundwater surface can essentially "pump" LNAPL laterally, with LNAPL entering new fractures during each cycle of rise and fall leading to a more dispersed LNAPL source zone. LNAPL typically will follow a declining water table almost immediately through larger vertical aperture fractures, with potentially lateral migration of LNAPL into newly-unsaturated, less steeply dipping fractures. As the water table rises, LNAPL will spread laterally into less steeply dipping fractures via buoyancy, as well as migrate within the steeply dipping fractures (CL:AIRE, 2014).
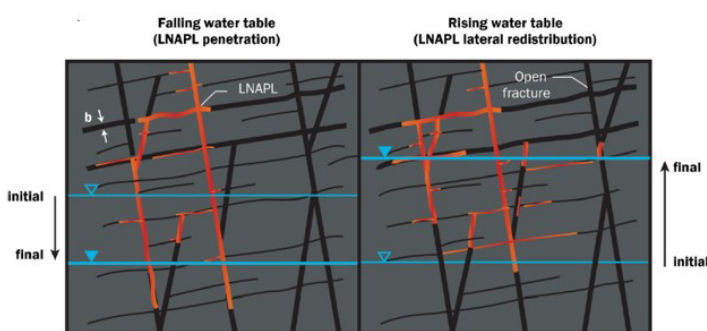

Figure 2. LNAPL Entrapment by a Fluctuating Groundwater Surface

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To adequately define the remedial footprint, it is necessary to understand the CSM and how the fracture rock network is interconnecting, and which fractures are dominant in the distribution and spread of the released LNAPL. One method for developing an accurate CSM involves use of geophysical logging methods to evaluate locations and dip angles of fractures and using transducers in nearby bedrock wells while drilling to understand fracture connectivity. A summary

of various tools available to characterize fractured systems is provided in Hardisty et al. (2003), Davis et al. (2006), and CL:AIRE (2014).

Remedial Considerations for Fractured Bedrock

Depending on the rock type, the bedrock may behave as an equivalent porous media and can be remediated in the same manner as an unconsolidated formation (such as a highly-fractured sandstone). Knowledge of the fracture locations, interconnectivity, and flow gradients will allow better placement of injection/extraction wells or points to facilitate introduction of amendments and/or removal of fluids. Preferential flow pathways through larger and more permeable fractures must be considered and addressed in the design. Technologies such as multi-phase extraction tend to be less applicable to unconsolidated formations since a large portion of residual LNAPL may be confined in narrow fractures minimally impacted by hydraulic recovery. Targeted extraction using packers and/or discrete points can facilitate recovery in some cases.

Challenge #4: LNAPL Sites in Arctic Regions

LNAPL releases and their management within arctic regions is complex due to the impact of frozen conditions on LNAPL migration and on mass recovery, mass control, and phase change remedies.

CSM Considerations in Arctic Regions

An ongoing research need is to achieve a better understanding of how petroleum moves through seasonal freezing soils (active layer) and frozen soil (permafrost). Permafrost is defined as soils where soil temperature remains below 0 °C for at least two years. The upper layer of the permafrost undergoes a cyclic temperature change during

the year from frozen in the winter to thawed in the summer. This is called the active layer or the seasonally thawed layer. The active layer can extend up to 2 m (6 feet) based on climate, soil texture, and organic content above mineral soil.

LNAPL migration through the active layer and permafrost soils is influenced by the formation and the presence of ice. The frozen surfaces can reduce the vertical migration of spilled LNAPL, thereby increasing lateral migration. Ice in pore spaces will either interrupt downward migration causing petroleum to spread laterally, or impede petroleum movement altogether due to the lack of open pore space. Segregated ice formation in the active layer can generate fissures that will enhance petroleum movement when the soil is thawed. Discontinuous and continuous permafrost will slow, redirect, or impede contaminant migration. The freeze-thaw cycle may also cause the detachment of mobile LNAPL from larger ganglia during freezing and vertical upward transport above or with the freezing front. Further fragmentation of LNAPL ganglia can occur due to shedding and entrapment of mobile LNAPL in the ice, potentially causing remobilization of residual LNAPL upon thawing.

Redistribution of contamination can occur by the formation of ice in pore space and fluctuations in the seasonal groundwater level during the freeze and thaw cycles. LNAPL can penetrate into frozen soils through the cracks and fissures formed during freezing of the soil; whereas a summer spill may allow LNAPL to migrate downwards through the soil. Spatial variability in soil properties and capillary forces can also enable lateral movement of LNAPL during downward migration. In general, the contribution of diffusion to the overall contaminant movement in permafrost is minimal (Filler et al., 2008).

Remedial Considerations for Arctic Regions

Ice present in active layer soil will influence the movement of petroleum and dissolved compounds, thereby impacting the design of monitoring systems to track natural attenuation. Moreover, cold soil temperatures will slow the dissolution, volatilization, and biodegradation of compounds in the subsurface, requiring additional time for technologies such as bioremediation or monitored natural attenuation (MNA) to achieve remedial goals. Further, physical characteristics including density and viscosity of the LNAPL will change due to the temperature. Viscosity will increase in arctic conditions and will slow lateral migration, and greatly impede hydraulic recovery of LNAPL using technologies such as multi-phase

extraction and skimmer pumps. Density will increase, which can facilitate downward vertical migration.

Ex situ management of waste is complicated using technologies that involve water or vapor transport including hydraulic containment, multiphase extraction, dual phase extraction, and even air sparging and soil vapor extraction. For these technologies heating lines and heated buildings are a necessity to continue with mass recovery efforts during the colder winter months (Filler et al., 2008). Special considerations for applying various remedial options within the arctic region are presented in Barnes et al. (2002) and Filler et al. (2008) including: excavation, thermal desorption, soil washing, composting, landfarming, soil vapor extraction, barometric pumping, bioremediation, and bioventing.

Challenge #5: Long-Chained Hydrocarbon Impacts

RPM survey results indicate that sites contaminated with petroleum products containing long-chain hydrocarbons such as NSFO, Bunker C, and heating oil are far less prevalent than sites contaminated with gasoline, diesel, and JP-5. However, sites that are contaminated with these long-chain hydrocarbon products result in a high cost to the Navy to remediate and close these sites due to their unique physical properties (i.e., high viscosity, specific gravity near that of water, and low volatility).

CSM Considerations for Long-Chained Hydrocarbons

Long-chain LNAPLs such as Bunker C, heavy crude oil, fuel oils, and NSFO are highly viscous and move very slowly through the subsurface. In general, hydrocarbons having longer carbon chains and/or a greater number of branches on the chain will have a higher viscosity, higher specific gravity, and lower volatility. Longer-chain hydrocarbons are also less bioavailable. The main concern with long-chain LNAPLs is not composition-based risk, but rather the removal or containment of LNAPL mass for saturation-based risks. Long-chained LNAPLs typically are poorly transmissive; thus, the use of transmissivity testing can be used to confirm the stability of the LNAPL body. In addition, the higher specific gravity of these compounds should be taken into account when measuring LNAPL thickness and groundwater elevations. The weight of the hydrocarbon depresses the groundwater elevation in the well requiring a correction to obtain the total hydraulic head.

Remedial Considerations for Long-Chained Hydrocarbons

The characteristics described above make it difficult to recover the free-phase liquid using conventional LNAPL recovery technologies. Because of their molecular size, low volatility, low aqueous solubility, and poor bioavailability, long-chain hydrocarbons are difficult to biodegrade and therefore not well suited to conventional vadose zone treatment technologies such as bioventing and soil vapor extraction. Applicable technologies are generally limited to excavation and thermal treatment. Excavation can be a useful and practical solution when the LNAPL is present near the ground surface. Thermal technologies are energy intensive, but may be able to achieve higher mass removal rates compared to conventional recovery processes.

Several recent technological innovations have been proposed and demonstrated to treat long-chain hydrocarbons. In situ combustion/smoldering is an emerging technology to destroy the long-chained hydrocarbons in place. Ex situ enhanced bioremediation using fungal enzymes to breakdown the heavier total petroleum hydrocarbon (TPH) components is another emerging technology. Lastly, natural source zone depletion is rapidly gaining acceptance to address sites containing long-chain hydrocarbons because their high viscosities and low volatilities result in limited mobility and minimal risk to human health and the environment. It is important to consider the environmental footprint of the technology and the overall life cycle of the project on a site-specific basis during selection and design of conventional or innovative approaches to address these sites.

Conclusions

This fact sheet provides an overview of challenging site conditions that can impact LNAPL CSMs and suggestions for managing these complex LNAPL sites. Additional technical resources are provided for NAVFAC RPMs to further understand best practices for managing saturation- and composition-based risks at these complex LNAPL sites.

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