

New Developments in LNAPL Site Management

Introduction

The ways in which owners manage industrial sites containing light non-aqueous phase liquids (LNAPL) is changing rapidly due to new conceptual models, new technologies, and new tools. This fact sheet summarizes new developments in the area of natural source zone depletion (NSZD) and reviews key tools for evaluating the practicability of Total Petroleum Hydrocarbons (TPH) and LNAPL recovery. It extends the discussion about LNAPL started in the 2015 NAVFAC LNAPL fact sheet.

More Natural Source Zone Depletion Than Originally Thought

NSZD is the natural loss of LNAPL due to volatilization, dissolution, and most importantly, biodegradation. Because of new understanding about LNAPL biodegradation and new ways to measure NSZD, there has been a movement by the remediation industry to rely on NSZD at many sites to manage LNAPL.

The original conceptual model of how LNAPL biodegrades has significantly changed over the past several years. Originally, it was thought that LNAPL biodegradation was controlled by the movement of groundwater through LNAPL sources in the saturated zone that delivered electron acceptors. These electron acceptors were then used by the naturally occurring bacteria to biodegrade the dissolution products from the LNAPL (primarily benzene, ethylbenzene, toluene, xylenes, or "BTEX"). However, a series of LNAPL research projects determined that anaerobic methane-generating biodegradation processes in both the vadose and saturated zones are more important and act on more of the LNAPL components than first thought. These key studies include:

California Oil Field Study: Researchers studying an oil field site compared the natural depletion caused by groundwater electron acceptor delivery and more importantly, measured the oxygen consumption in the vadose zone by measuring the vertical concentration gradient of these gases (Johnson et al., 2006; Lundegard and Johnson, 2006). They determined that the amount of biodegradation that was expressed

by the vertical gas exchange was almost 100 times the biodegradation from the electron acceptors (Figure 1).

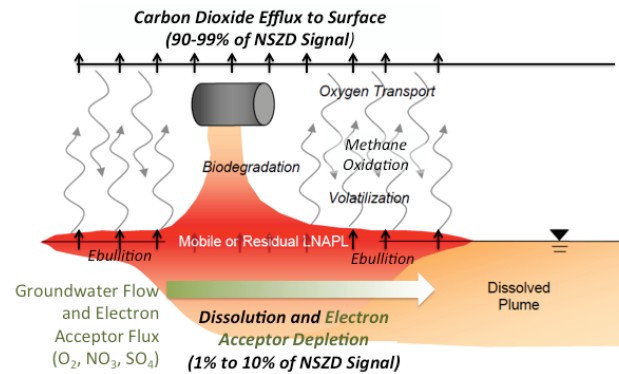
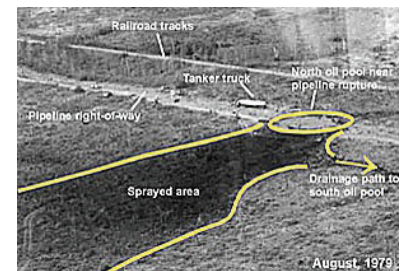


Figure 1. Key Natural Source Zone Depletion Processes (adapted from ITRC, 2009 and Suthersan et al., 2015).

Crude Oil Spill Research Site: University of British Columbia and U.S. Geological Survey researchers performed detailed measurements of both dissolved gases in groundwater and gases in the unsaturated zone at a 1979 crude oil pipeline release in Bemidji, Minnesota in a series of projects from 1983 to 2015.

They identified the importance of methanogenesis (degradation of LNAPL compounds by methane-producing bacteria) in the saturated zone, which can result in degassing (formation of bubbles with methane and other gases), followed by ebullition (the transport of these bubbles to the vadose zone).



Aerial View of Bemidji, MN Pipeline Break (Graphic: USGS.gov)

Overall, these processes indicate that LNAPL biodegradation rates are higher than first thought, and can target more compounds (such as alkanes) than just the BTEX compounds. NSZD rates measured by the site researchers are shown below (Sihota and Mayer, 2012):

Method	NSZD Rate (g TPH/m ² /day)
Oxygen Gradient	3.3
Dynamic Chamber	1.1 – 3.6

One other implication of this evolving NSZD conceptual model is that methane accumulation under impervious structures such as parking lots or buildings may be a concern. However, Ma et al. (2014) concluded “The U.S. EPA’s guidance document for petroleum vapor intrusion (USEPA, 2013) is based on field measurement data at retail service station sites including sites for which E10 (10% ethanol fuel) would have been used for decades. According to this document, regular gasoline or E10 releases are unlikely to cause a flammability hazard, unless the gasoline is in the building or in direct contact with the foundation.” However, these researchers have hypothesized there is an increased potential for methane-induced explosive conditions for releases of ethanol rich fuels (comprised of 20% ethanol or more) (Ma et al., 2014).

Carbon Trap Studies at Refineries and Terminals: Researchers at Colorado State University developed a carbon dioxide trap that could be deployed (typically for two weeks) and capture any carbon dioxide being emitted to the surface (McCoy et al., 2014;). When applied to a number of different LNAPL sites around the country they typically show NSZD rates at LNAPL sites in the range of 100s to 1,000s of gallons LNAPL biodegraded per acre per year.

Rates from Key Studies: There is a trend by site owners to report LNAPL NSZD rates in units of gallons per acre per year, and NSZD rates from key studies converted to this unit are reported below. For comparison, 1,000 gallons per acre per year is the equivalent of reducing the LNAPL saturation (the percent of pore space occupied by LNAPL) by ~1% over a 10-year period.

NSZD Study	Site-Wide NSZD Rate (gallons per acre per year)
Six Refinery Terminal Sites (McCoy, 2012)	2,100 – 7,700
1979 Crude Oil Spill (Sihota et al., 2011)	1,600
Refinery/Terminal Sites in Los Angeles (LA LNAPL Wkgrp, 2015)	1,100 – 1,700
Five Fuel/Diesel/Gasoline Sites (Piontek et al, 2014)	300 - 3,100
Eleven Diverse Petroleum Sites (Palaia, 2016)	300 – 5,600

Four Ways to Measure NSZD: Four methods for measuring NSZD rates have been developed:

1. *Carbon Traps:* carbon dioxide is captured at the surface using a receptacle with CO₂ adsorbent. The amount of CO₂ in the trap can then be measured and converted to an NSZD rate (McCoy et al., 2014). Currently, it is the most common method to obtain NSZD rates.



Deployed carbon trap (Source E-Flux)

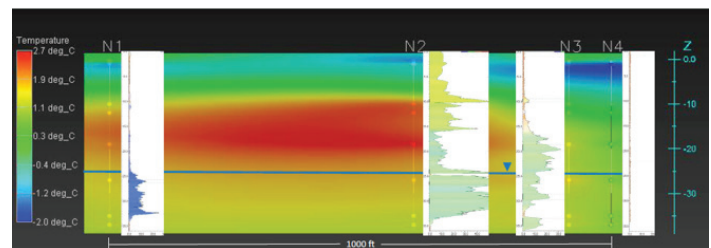
2. *Dynamic Closed Chamber Method:* captures CO₂ in a closed chamber at the surface. The resulting CO₂ flux can then be converted to an NSZD rate (Sihota et al, 2011). This method is more commonly used for research compared to actual NSZD projects at LNAPL sites.



Deployed automated soil flux chamber (Source: LI-COR Biosciences)

3. *Gradient Method:* concentration gradients of gas phase constituents in the vadose zone are calculated using soil vapor tubes installed vertically and then used to determine NSZD rates by applying Fick’s Law (Lundegard and Johnson, 2006; Sihota et al., 2011). This method is useful if existing vertical probes are already installed, but it can be difficult and costly to get diffusion coefficients. This method also assumes homogeneous soil conditions in the vadose zone.

4. *Temperature-Based Method:* developed at Colorado State University (Stockwell, 2015) where the heat generated by biodegradation processes is measured using a vertical series of thermocouples and then converted to a biodegradation rate. The red area in the figure below shows the net heat (additional heat compared to background location) generated by LNAPL degradation from four stations, each with eight thermocouples distributed vertically to 30 feet below ground surface.



Source: Stockwell, 2015

How NSZD Rates Are Used: NSZD rates can be used in several ways:

- To confirm that LNAPL (particularly the more volatile and leachable components) is biodegrading and that the LNAPL is not an unchanging, permanent mass in the subsurface.

- As a benchmark to evaluate the usefulness of continuing active LNAPL recovery and/or in-situ remediation. For instance, if NSZD rates are 1,000 gallons per acre per year, and an active LNAPL skimming system is only removing 10 gallons per acre per year, then a case could be made for discontinuing the active treatment.

Frequently Asked NSZD Questions (LA LNAPL Workgroup, 2015).

- **How do you know the CO₂ is coming from LNAPL, and not from soil or plants?** A background correction is required to exclude the CO₂ produced from plant respiration: i) one can take the CO₂ flux measurements in a clean area and subtract from the LNAPL-impacted area (Sihota et al., 2011) or ii) one can use 14C isotope analysis to determine contribution of CO₂ from hydrocarbons (Sihota and Mayer, 2012).
- **Do the carbon efflux methods measure NSZD from the vadose zone, saturated zone, or both?** They represent the contribution from any LNAPL in the vadose zone and any LNAPL in the saturated zone.
- **Why can it be difficult to compare the different methods?** The different methods measure CO₂ flux over different time periods and some are snapshot measurements (Gradient Method, Dynamic Closed Chamber short-term) as compared to an integrated average over an extended time period (Dynamic Closed Chamber long-term, Carbon Traps, Temperature Method).

TPHCWG Method for Risk Assessment

Gasoline, diesel, and other petroleum mixtures contain hundreds or thousands of different individual petroleum hydrocarbons. Total petroleum hydrocarbon (TPH) analytical methods measure the TPH in a sample without quantifying the concentration of each individual petroleum constituent (e.g., benzene, hexane, etc.).

Although bulk TPH methods are useful for measuring total petroleum concentrations, TPH measurements are not an indicator of health risks. The TPH analytical method serves to indicate the amount of petroleum (or similar non-petroleum compounds) that may be present in a sample, but the test results provide no direct indication of risk to human health or the environment (ATSDR, 1999; API, 2001). Petroleum mixtures are composed of thousands of different chemicals, many of which pose little or no toxicity to humans. Consequently, TPH tests that indicate concentrations within certain carbon ranges (but not the chemical composition of that petroleum) cannot be used alone to determine the health risk associated with that particular TPH value. For example, two soil samples impacted by equal concentrations of baby oil vs. gasoline would have virtually identical TPH values, but very different toxicities.

To solve this problem, the TPH Criteria Working Group (TPHCWG) (a group comprised of environmental experts representing both industrial and federal site owners including the Department of Defense, and regulatory agencies) developed the TPH Fraction Method to calculate risk associated with petroleum hydrocarbon mixtures.

With this TPHCWG method, TPH is measured using an analytical method that provides somewhat more detail than total TPH concerning the composition of the petroleum mixture. These analytical methods include **TX1006** (based on the TPHCWG Method) or **Massachusetts EPH/VPH**. These methods provide concentration results for six (Massachusetts EPH/VPH) or 13 (TX1006) different TPH fractions separated into different classes by compound type (aliphatic vs. aromatics) and by carbon number (see figure below). These fraction results can be used in risk assessments by assigning conservative toxicity values and fate and transport characteristics to each different fraction. The toxicity and fate and transport values can be obtained from guidance documents that describe the application of the TPH fraction approach for risk assessment (e.g., MassDep, 2002; TPHCWG, 1998). While typically used to demonstrate that soils containing petroleum hydrocarbons have heavier, low-risk fractions, the method can also be applied to groundwater samples.

Aliphatics	Aromatics
nC ₆	≥nC ₇ -nC ₈ (<i>Toluene</i>)
>nC ₆ -nC ₈	≥nC ₈ -nC ₁₀
≥nC ₈ -nC ₁₀	≥nC ₁₀ -nC ₁₂
≥nC ₁₀ -nC ₁₂	≥nC ₁₂ -nC ₁₆
≥nC ₁₂ -nC ₁₆	≥nC ₁₆ -nC ₂₁
≥nC ₁₆ -nC ₂₁	≥nC ₂₁ -nC ₃₅
≥nC ₂₁ -nC ₃₅	

Adapted from Rhodes, 2006

In summary, to get detailed risk assessment information from the TPHCWG method, analyze your soil or groundwater samples with one of the two analytical methods above and then apply risk calculations. More information on these example risk calculation methods can be found on state websites such as:

<https://www.tceq.texas.gov/remediation/trrp/trrppcls.html>
(click on "TPH spreadsheet")

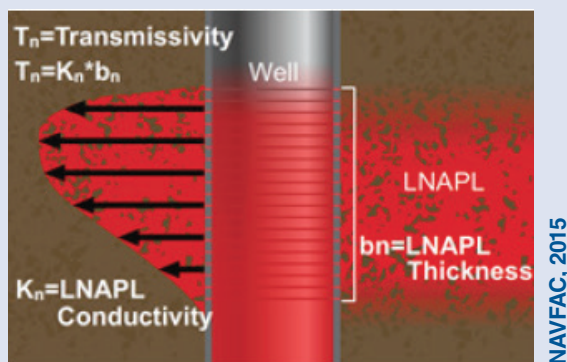
<http://www.mass.gov/eea/agencies/massdep/toxics/sources/riskasmt-htm.html#7>
(see "Shortforms" for risk calculation spreadsheet)

What is the Importance of LNAPL Transmissivity?

(Excerpted from NAVFAC, 2015).

An important new development in LNAPL site management is the use of transmissivity to evaluate LNAPL recovery. LNAPL thickness in site wells has historically been used to estimate its recoverability and mobility. The old “pancake” saturation and distribution model assumed that LNAPL thickness in the formation was similar to that measured in site wells, which led to overestimates of LNAPL volume, recoverability, and risk. More recently, LNAPL transmissivity has been defined as the volume of LNAPL through a unit width of aquifer per unit time per unit drawdown (see Figure below). It has units of length²/time (L²/T) and is being used as a line of evidence to predict LNAPL recoverability. Unlike product thickness in a well, transmissivity is dependent on soil type and properties (e.g., porosity, conductivity), chemical and physical properties of the LNAPL (e.g., density, viscosity, composition), LNAPL saturation in the formation, as well as the thickness of the mobile LNAPL (Interstate Technology & Regulatory Council [ITRC], 2009). This makes it a useful metric for estimating recovery.

LNAPL transmissivity is an important part of any LNAPL conceptual site model (LCSM). To determine transmissivity, soil lithology across the zone where LNAPL is present and the hydraulic conductivity within each interval must be known. Equilibrium fluid levels in wells and well construction details must also be known to establish baseline conditions. Calculation tools are available for estimating LNAPL transmissivity.



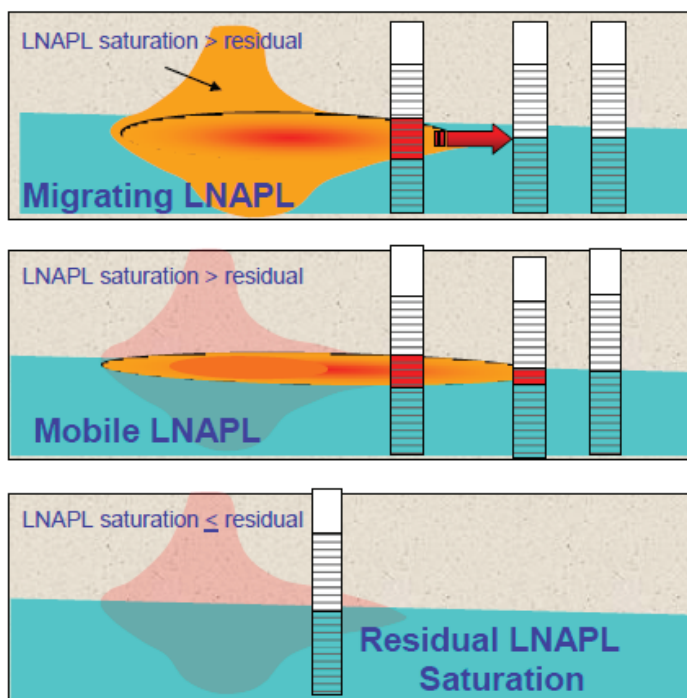
LNAPL Recovery Limits

The U.S. EPA regulations issued in 1998 state that Leaking Underground Storage Tank (LUST) owners/operators must “remove free product (LNAPL) to the maximum extent practicable as determined by the implementing agency (State/local regulator)”. Historically, common interpretation by state programs was that “LNAPL present in a well at a thickness greater than

0.01 foot was practicably recoverable and must be addressed in an active manner” (Suthersan et al., 2015). Over time, different states developed their own criteria for meeting “to the extent practicable”. Table 1 highlights site closure practices for LNAPL sites that some state programs have used from time to time.

Since then, developments in the understanding of LNAPL transport, geology, and the practical limitations of LNAPL recovery have enabled state agencies to redefine “maximum extent practicable” and in some cases move to more realistic hydraulic metrics such as

LNAPL Transmissivity (see text box) or interpreting it as eliminating migrating LNAPL (see Figure 2). While some states are sticking to their original criteria listed in Table 1, other states appear to be amenable to these new approaches. The 2009 ITRC LNAPL guidance document captures much of the new thinking about “to the extent practicable” and LNAPL remediation.



ITRC, 2014

Figure 2. Three LNAPL Conditions and Associated Remedial Drivers. The top panel shows a condition where the LNAPL body is still migrating due to LNAPL head and high LNAPL saturations in the soil. The middle panel shows a condition somewhat later where there is LNAPL present in monitoring wells, but the LNAPL is no longer migrating. The bottom panel shows a condition where there is no longer LNAPL present in monitoring wells. The LNAPL in soil is at saturations less than “residual saturation.”

There are several ways to determine LNAPL transmissivity (ASTM, 2013):

Short-Term Baildown Tests: A baildown test involves rapidly removing LNAPL from the well while minimizing recovery or disturbance of the groundwater. It is often difficult to only remove the LNAPL with an actual bailer, so a vacuum truck is sometimes used. During the test, both the elevation of the water-LNAPL and LNAPL-air interfaces are monitored over time. The API LNAPL Transmissivity spreadsheet can be used to convert the field data to an actual transmissivity value at that location.

Manual Skimming Methods. This method involves removing LNAPL from a well on a repeated basis without allowing for more than 25% of the recharge to occur between skimming events. To calculate LNAPL transmissivity, use this formula (ASTM, 2013):

$$T_n = \frac{Q_n \ln\left(\frac{R_{oi}}{r_w}\right)}{2\pi s_n}$$

where T_n = LNAPL Transmissivity; Q_n = measured LNAPL recovery rate; R_{oi} = radius of influence of LNAPL skimming; r_w = well radius; s_n = LNAPL drawdown is the geometric mean of the starting and ending drawdowns. Note: $\ln\left(\frac{R_{oi}}{r_w}\right)$ can be assumed to be equal to 4.6 without introducing significant error).

Long-Term Recovery Based Methods. LNAPL recovery rate vs. drawdown data are collected and then applied to a transmissivity formula such as the one used for manual skimming methods above. However, the LNAPL drawdown represents the steady state drawdown from continued pumping at rate Q_n .

Tracer Based Method. This method involves injecting a hydrophobic fluorescent tracer into the LNAPL accumulation, which is flushed away from a well that is screened in a zone with moving LNAPL. By taking periodic measurements over several months of the tracer concentration in the well (using a fiber optic cable introduced into the well and a photo spectrometer on the surface), the LNAPL flux in units of length per time can be determined and then used to estimate the LNAPL transmissivity (Sale et al., 2007). Mahler et al., (2012) report on application of the tracer method on 50 LNAPL wells at seven different sites.

Comparison: Short-Term Baildown methods can be performed quickly and at relatively low cost but are likely subject to larger uncertainties. The Skimming methods are dependent on achieving a > 75% of the maximum skimming drawdown which must be consistently induced. The Long-Term Recovery methods are best suited for existing LNAPL recovery systems. The tracer test is an emerging method that is relatively easy and quick, but requires constant fluid levels over the testing period, which can extend over several weeks.

Some states are using these concepts and the ITRC LNAPL guidance to determine “maximum extent practicable”. For example, the Michigan Department of Environmental Quality (MDEQ) states in its recently issued petroleum cleanup guidance that if LNAPL remaining at a site has a transmissivity greater than 0.5 ft²/day, it is likely that additional recovery would be beneficial and that the LNAPL may be recovered in a cost-effective and efficient manner (MDEQ, 2014). Similarly, the Massachusetts Department of Environmental Protection (MassDep) deems LNAPL recovery to be infeasible when LNAPL transmissivity in all recovery wells is less than 0.8 ft²/day or the total volume of LNAPL recovered at a site is less than 1 gallon in any 3-month period (MassDep, 2002).

One caution is LNAPL transmissivity is dependent on several parameters that vary widely in the subsurface and can change over time. Beckett et al., (2015) provided this analysis:

LNAPL transmissivity (T_n) is a potentially useful, but complex parameter related to the bulk ability of LNAPL to move through porous media under mixed saturation conditions. A key complexity is that LNAPL transmissivity is not a constant of the formation, but rather varies with test conditions, time, and changing piezometric conditions.

Overall they conclude that LNAPL Transmissivity “is a useful parameter and we support its use, but that must be considered in context with a broader range of multiphase mechanics to produce a robust and protective set of site specific management actions” and provide several recommendations on how to make better T_n estimates.

Site Closure: California’s Approach

In 2012 California promulgated their low-threat closure guidance for underground storage tank (UST) release sites (CSWRCB, 2012). This innovative guidance combines information about free product, location of receptors, and other factors to evaluate if a UST site can be closed.

In practice, UST sites can close if they meet some general criteria and the specific risk-based criteria for each risk pathway. For example, a site meets the groundwater-specific criteria for closure if it meets all the requirements in one of the five classes of sites:

- Class 1: a. The contaminant plume that exceeds water quality objectives is less than 100 feet in length.
b. There is no free product.
c. The nearest existing water supply well or surface water body is greater than 250 feet from the defined plume boundary.

Class 2: a. The contaminant plume that exceeds water quality

objectives is less than 250 feet in length.

- b. There is no free product.
- c. The nearest existing water supply well or surface water body is greater than 1,000 feet from the defined plume boundary.
- d. The dissolved concentration of benzene is less than 3,000 micrograms per liter (µg/l), and the dissolved concentration of MTBE is less than 1,000 µg/l.

- Class 3:
- a. The contaminant plume that exceeds water quality objectives is less than 250 feet in length.
 - b. Free product has been removed to the maximum extent practicable, may still be present below the site where the release originated, but does not extend off-site.
 - c. The plume has been stable or decreasing for a minimum of five years.
 - d. The nearest existing water supply well or surface water body is greater than 1,000 feet from the defined plume boundary.
 - e. The property owner is willing to accept a land use restriction if the regulatory agency requires a land use restriction as a condition of closure.

- Class 4:
- a. The contaminant plume that exceeds water quality objectives is less than 1,000 feet in length.
 - b. There is no free product.
 - c. The nearest existing water supply well or surface water body is greater than 1,000 feet from the defined plume boundary.
 - d. The dissolved concentration of benzene is less than 1,000 µg/l, and the dissolved concentration of MTBE is less than 1,000 µg/l.

- Class 5:
- a. The regulatory agency determines, based on an analysis of site-specific conditions that under current and reasonably anticipated near-term future scenarios, the contaminant plume poses a low threat to human health and safety and to the environment and water quality objectives will be achieved within a reasonable time frame.

The California low-threat closure policy is an important development for risk-based LNAPL site management. For example, “free product” can still be present at a site as long as it has been removed to the maximum extent practicable but does not extend off site and it meets a 1,000 foot or more requirement to water supply wells or surface water; and the property owner is willing to accept a land use restriction (see Class 3 above). According to the California’s Low-Threat LUFT Site Closure Policy Looking Forward publication (Hadley et al., 2015):

California’s regulatory agencies have historically been at the forefront of national efforts to address environmental concerns. In 2012, California’s agency for addressing leaking underground fuel tanks (LUFTs) adopted a policy that identifies low-threat conditions warranting closure of an LUFT case. That development clearly fulfills the role of risk management in the risk assessment–risk management paradigm inherent in environmental remediation. It also encourages identification of additional categories of sites and other circumstances that are “low threat” to develop similar guidance on closure to apply to those sites.

As can be seen in the first four classes of sites in the California Low-Threat guidance, short plume lengths and plume stability serve as the foundations for defining certain sites as “Low Threat.” The values used in the Low Threat guidance were based on extensive scientific research on the hydrocarbon plume characteristics; for a summary of some of the key research see the API’s “Characteristics of Dissolved Hydrocarbon Plumes: Results from Four Studies” (Newell and Connor, 1998). Key statistics from this document are shown below.

Data from 604 Hydrocarbon Sites	BTEX Plume Length (feet)
Maximum	3020
90Th Percentile	319
75Th Percentile	203
Median	132
25Th Percentile	80
Minimum	8

Table 1. Summary of LNAPL Recovery Limits for Site Closure for Key Navy States

State	Measurable Level that Permits Closure	Closed Sites with LNAPL Greater Than Measurable Level? If So, Additional Criteria Used?	Source
CA	Removed to extent practicable	Yes. "LUST sites can be closed if the required level of water quality will be attained within a reasonable period of time. California has closed several sites with LNAPL."	Menatti, 2010; NEIWPC, 2006; Lund et al., 2014
FL	0.01 ft	Yes. "A site with residual soil contamination or groundwater contamination above cleanup target levels can only receive No Further Action if there are institutional controls (deed restrictions) on the property."	NEIWPC, 2006
HI		Yes. "Only if BTEX and PAHs are ND or well below action levels and no other receptors."	NEIWPC, 2006
IA	<0.01 ft	Yes. "Following NAPL recovery activities, a site may be closed if product does not return in a monitoring well in excess of 0.02 ft for a period of one year."	NEIWPC, 2006
IL		Yes. Obtained closures in Illinois with notable NAPL thicknesses. Site-specific basis, essentially along the same lines as Massachusetts.	Payne, 2016
MA		Yes. "Non-Stable NAPL is not present under current site conditions and for the foreseeable future, and all NAPL with Micro-Scale Mobility is removed if and to the extent feasible based upon consideration of CSM principles."(*)	Marra, 2014
MI		Yes. "Recovery of all LNAPL with a transmissivity greater than 0.5 ft ² /day and that can be recovered in a cost-effective and efficient manner."	Lund et al., 2014
MO	Maximum extent practicable	Yes. "Site-specific criteria."	NEIWPC, 2006
NC	< 0.01 ft	No. "No receptors and removal is technically and economically infeasible."	NEIWPC, 2006; Lund et al., 2014
NV	0.5 inch	Yes. "If a risk-based analysis was performed showing no vulnerable receptors and a fate and transport analysis performed showing that there was little potential for migration."	NEIWPC, 2006
RI		No	Lund et al., 2014
TX	Unrecoverable or impractical	Yes "Closure can be granted when recoverable NAPL is still present if there are no receptors and the plume is stable."	NEIWPC, 2006
UT	1/8-inch	Yes. "RBCA-based approach considered on site-specific basis."	Menatti, 2010
VA	<0.01 ft	Yes. "The data should support the claim that the technologies used and/or evaluated are no longer effective and that additional recovery is not practicable. If >0.01 ft exists also have to show: a) Remaining LNAPL and dissolved-phase constituents are not a risk to human health or the environment, and b) NSZD of the LNAPL body and natural attenuation of the dissolved-phase plume are documented as occurring at the site and are expected to further mitigate risk from the release, and c) The areal extent of the free phase plume at the site is shown to be stable or decreasing"	NEIWPC, 2006; Steers, J., 2012; Lund et al., 2014
WA	<0.01 ft	No. "Ecology won't close LUST sites with measurable free product"	NEIWPC, 2006

Notes: *Non-Stable NAPL: a NAPL with a footprint that is expanding laterally or vertically by: (a) migrating along or within a preferred flow path; (b) discharging or periodically discharging to a building, utility, drinking water supply well, or surface water body; or (c) spreading as a bulk fluid through or from subsurface strata; and NAPL with Micro-Scale Mobility: a NAPL with a footprint that is not expanding, but which is visibility present in the subsurface in sufficient quantities to migrate or potentially migrate as a separate phase over a short distance and visibility impact an excavation, boring, or monitoring well. (Marra, 2014).

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