Estimating NAPL Effective Hydraulic Conductivity and Potential Velocity in the Field Based on Laboratory Pore-Fluid Mobility Test Results

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ABSTRACT: Nonaqueous phase liquid (NAPL) mobility is important in NAPL site risk characterization, remedy selection and remedial design. Laboratory centrifuge or "water-drive" tests, which are commonly used for evaluating NAPL mobility in sediment or soil core samples, usually use hydraulic gradients orders of magnitude stronger than those in the field. If no NAPL is produced from a given sample in these tests, the NAPL in the sample is residual (i.e., immobile) under laboratory and field conditions. Cases when these tests produce NAPL indicate that the NAPL saturation was greater than residual saturation under the aggressive lab test conditions; however, the NAPL may still have been less than residual saturation under field conditions. In these cases, the distinction between residual and "potentially mobile" is inconclusive. However, the test data can be used to estimate the NAPL effective hydraulic conductivity (K_n) and NAPL velocity under field conditions assuming a hypothetical worst-case scenario in which the NAPL is migrating. The method presented herein is applicable for any NAPL mobility assessment. Results for NAPLs rich in polycyclic aromatic hydrocarbons (PAHs), such as coal tar, oil tar, and creosote consistently indicate K_n values in the range of 10^{-6} centimeters per second (cm/sec) or less, and estimated NAPL velocities in the field typically a few centimeters per year or less (if migrating).

INTRODUCTION

Problem Statement. When laboratory NAPL mobility tests are performed with driving forces orders of magnitude greater than those that typically exist in the field, what can one conclude from the test results? These tests have historically been conducted to force as much NAPL as possible out of the sample, such that the NAPL saturation at the end of the test can be considered an estimate for the residual saturation, albeit a conservatively small estimate. Under such test conditions, the only test results that can be taken at face value are those that produce no NAPL; in these cases, the conclusion is that the NAPL in the test sample is immobile (i.e., at or less than the residual saturation) under the aggressive laboratory test conditions, as well as under field conditions where the driving forces are much weaker.

However, when an aggressive laboratory test does produce NAPL movement, the results are ambiguous and can be misinterpreted. Although the final NAPL saturation could be construed as a "residual saturation," that conclusion is only accurate for the specific test conditions. The residual saturation in the field, where the driving forces are orders of magnitude weaker, would be greater. In samples that produce NAPL movement during aggressive lab tests, it is possible that the NAPL is at or less than the residual saturation in the field, and NAPL movement only occurred because the lab testing method imposed driving forces orders of magnitude stronger than those that existed in the field. That is, the NAPL may not have sufficient capillary pressure to exceed the pore entry pressure required for actual NAPL migration in the field. However, demonstrating this would require additional data collection, quantitative evaluation, and, in some cases, multiphase flow modeling. Ultimately, to be protective and conservative, the overseeing regulatory agency may still default to the assumption that the NAPL is capable of moving in the field.

This paper presents a methodology to develop an additional line of evidence to support the NAPL conceptual site model under the conservative, hypothetical assumption that the NAPL may migrate under field conditions.

BACKGROUND ON LABORATORY NAPL MOBILITY TESTS

First, this paper provides some background about the most common types of laboratory NAPL mobility tests. In general, laboratory NAPL mobility tests fit into two categories: centrifuge-based methods and water-drive (also known as water-flood) methods.

Centrifuge-Based Tests. Centrifuge-based NAPL-mobility tests apply a centrifugal force to the sample (e.g., Brady and Kunkel, 2005; PTS Laboratories, 2012). A common approach to these tests is to insert undisturbed (native-state) samples into centrifuge cups and then centrifuge for 1 hour at 1000 times the force of gravity (1000G) at a controlled temperature of 20 ± 1°C. Fluids produced during centrifuging are collected and volumes measured. The remaining fluid saturations are determined by Dean-Stark extraction and sediment or soil properties are determined at the completion of the centrifuge run. Initial NAPL saturations can be calculated using mass balance equations (PTS Laboratories, 2012). The samples can be run "under water," in which case the liquids that drain from the sample are replaced by water. However, centrifuge tests are more commonly run "under air," in which case the draining liquids are replaced with air. Centrifuge tests can be run with any selected driving force and for any duration within equipment limitations.

The goal of centrifuging at 1000G is to drive all drainable NAPL from the sample, leaving behind only the residual saturation. However, the force of 1000G is equal to a hydraulic gradient of 1000, which is thousands of times stronger than hydraulic driving forces in the field; therefore, the displacement of NAPL from the sample under this extreme test condition does not indicate whether the NAPL saturation is greater than the residual saturation under field conditions. Brady and Kunkel (2005) reported NAPL displacement at 1000G with initial NAPL saturations as small as 3.1% of pore volume. With 1000G centrifugation, the authors of this paper have seen displacement with initial NAPL saturations as low as 2.5%. These extremely low NAPL saturations are significantly less than expected residual saturation values (Cohen and Mercer, 1993; Hugaboom and Powers, 2002; U.S. EPA, 2004).

Centrifuge-based NAPL mobility tests directly control the driving force applied to the sample by adjusting the centrifuge spin rate. A common modification to the method described by PTS Laboratories (2012) is to use a "multi-step" approach with three test steps—10G, 100G, and 1000G—for 1 hour each. However, driving forces of 10G and 100G are still orders of magnitude stronger than those in the field. The authors of this paper observed NAPL displacement at 100G with initial NAPL saturations as small as 3.7% of pore volume, and at 10G with initial NAPL saturations as small as 7.0% of pore volume. Both are still less than typical residual saturation values under field conditions.

Another alternative is to inject water to displace NAPL from the sample, as described in the next subsection.

Water-Drive Tests. Water-drive tests (PTS Laboratories, 2010), also known as water-flood tests (Niemet et al., 2015), involve pumping water through a sample to mobilize drainable NAPL. The NAPL produced is collected and its volume is measured. Final fluid saturations are determined by Dean-Stark extraction based on porous sample physical properties. Initial saturations can be calculated using mass balance equations (PTS Laboratories, 2010). During water-drive tests, the water-injection rate is controlled, and the injection pressure is measured. The injection rate is often relatively small at first and then increased during the test. The goal is to force multiple pore-volumes of water through the sample. Achieving this goal within a reasonable timeframe often requires hydraulic gradients that are much greater than those in the field. For example, data reported by Niemet et al. (2015) indicate test hydraulic gradients ranging from approximately 5 to 200, with an average of approximately 50. Based on injection pressures described by PTS Laboratories (2010) and a typical test plug length of approximately 5 cm, water-drive tests may subject samples to hydraulic gradients between approximately 70 and 110. Thus, water-drive tests can also be extremely aggressive compared to field conditions.

MATERIALS AND METHODS

Although the typical laboratory-imposed driving forces are orders of magnitude stronger than those in the field, laboratory tests that produce NAPL provide a unique opportunity to calculate the effective NAPL hydraulic conductivity (K_n). Each centrifuge or water-drive test step is a constant-gradient test. To be conservative, test "plugs" are typically collected from portions of the core sample with the greatest apparent NAPL saturations. The change in NAPL saturation during a given test, or test step, can be converted to an average NAPL volumetric flow rate for the test conditions (Figure 1). Then the NAPL flow rate, hydraulic gradient, and test sample geometry can be used to calculate a K_n value for each test sample.

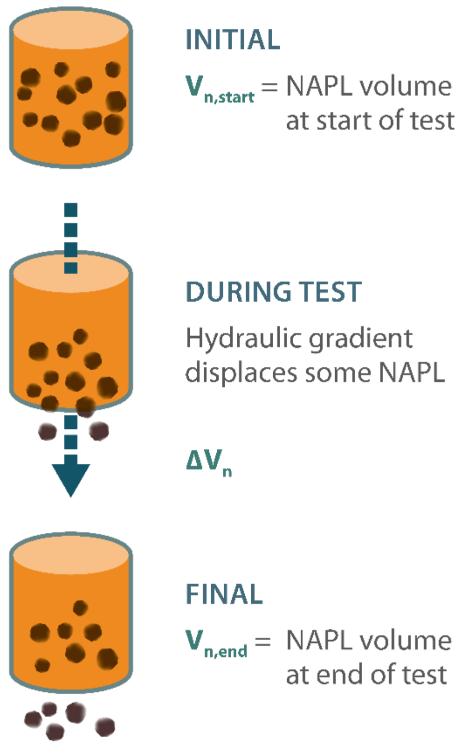

FIGURE 1. Schematic depiction of NAPL volume change in a laboratory test sample of soil or sediment.

Based on the test plug geometry and the applied hydraulic gradient, K_n is calculated using Darcy's Law as follows:

$$K_n = Q_n / A\ i \qquad (1)$$

where Q_n is the average NAPL flow rate from the test sample during the test, A is the cross-sectional area of the discharge end of the cylindrical test sample, and i is the hydraulic gradient imposed during the test. Q_n can be calculated as follows:

$$Q_n = \Delta V_n / t \qquad (2)$$

where ΔV_n is the change in NAPL volume in the sample during the test and t is the test duration. Testing laboratories typically report the initial and final NAPL saturation values and the sample porosity. The volume of NAPL either before or after the test can be calculated as follows:

$$V_n = V_t\ n\ S \qquad (3)$$

where V_t is test plug total volume, n is porosity, and S is the reported NAPL saturation (initial or final). Samples that indicate no change in NAPL saturation have Q_n and K_n values of 0 under the test conditions. It is reasonable to assume that a sample with a K_n value of 0 under aggressive laboratory test conditions also has a K_n value of 0 under field conditions. In other words, the NAPL in the sample is at or less than the residual saturation and immobile under field conditions.

The K_n results also can be used in screening-level calculations to estimate the effective NAPL Darcy flux (V_d) and pore-scale NAPL velocity (V_n) in the field using the following equations (based on Brooks and Corey, 1966):

$$V_d = K_n i_f \quad (4)$$

$$V_n = K_n i_f / (nS) \quad (5)$$

where i_f is the hydraulic gradient in the field.

Laboratory NAPL mobility tests can be run in either a horizontal or vertical direction relative to the original core orientation. A test with flow parallel to the original core length is a “vertical” test. If the direction of interest for field NAPL mobility in the field differs from the direction of laboratory testing, then the laboratory-measured K_n value can be adjusted proportional to the estimated or measured anisotropy ratio of the porous medium. In addition, to assess the potential velocity of vertical NAPL movement, the hydraulic gradient in the field should be corrected to account for the “hydraulic gradient due to gravity”, which depends on the NAPL and water density values (Cohen and Mercer, 1993).

RESULTS AND DISCUSSION

Example calculations based on tests conducted for PAH-rich NAPL samples (similar to creosote, coal tar, and oil tar) are presented in Tables 1 and 2. Table 1 shows example calculations for samples subject to multi-step centrifuge testing at 10G, 100G, and 1000G test steps for 1 hour each. These test samples were 2 inches (5 cm) long and 1.5 inch (3.8 cm) in diameter. The centrifugal force was applied parallel to the axis of the cylindrical test plug, which is perpendicular to the original core length (i.e., these are “horizontal” NAPL mobility tests). Example 1 illustrates a test result in which NAPL movement was produced at all three test steps. Of the three test steps, the results at 10G are considered the best approximation of K_n under field conditions, because the driving force is closest to field conditions and the initial NAPL saturation in the test plug is equal to the NAPL saturation in the field. The calculated K_n value is 9.3×10^{-7} cm/s. This K_n value can be used to perform a screening-level calculation of the NAPL velocity in the field. Assuming a field horizontal hydraulic gradient of 0.005, the NAPL velocity in the field (V_n) is calculated as 0.6 cm/year. Thus, although NAPL movement was observed in the lab at 10G, its velocity under field conditions would be extremely small if it is moving. That is, even if it is migrating (which may not be the case), it would still be practically immobile. The sample in Example 2 showed no NAPL movement at 10G; therefore, the calculated K_n value is 0 and the NAPL in the sample is interpreted as residual (i.e., immobile) under field conditions. The test results at 100G and 1000G are not used, but the calculated K_n values are so small that the NAPL is confirmed as practically immobile.

Table 2 shows two example calculations for single-step centrifuge tests at 1000G. Example 3 shows a sample that produced NAPL movement, with a calculated K_n value of 9.8×10^{-8} cm/s. Although the test results do not provide a basis to verify that the NAPL is capable of migrating under field conditions, if it is moving, the calculated K_n value suggests that the rate of NAPL migration would be extremely small. Assuming a field horizontal hydraulic gradient of 0.005, the calculated NAPL velocity in the field (V_n) would be 0.1 cm/year. Thus, although NAPL movement was observed under this extreme test driving force, the NAPL can be considered practically immobile under field conditions (if it is migrating at all). The sample in Example 4 indicated no NAPL movement at 1000G; thus, the NAPL in this sample is interpreted as residual and immobile under field conditions.

TABLE 1. Example results for multi-step centrifuge tests with 10G, 100G and 1000G spin for 1 hour each.

Parameter	Example 1	Example 2
porosity, n	0.66	0.46
NAPL saturation, S initial (%)	37.7	19.8
NAPL saturation, S after 10G (%)	36.7	19.8
NAPL saturation, S after 100G (%)	28.3	19.34
NAPL saturation, S after 1000G (%)	10.1	12.7
10G Kn (cm/s)	9.3E-07	0.0E+00
100G K_n (cm/s)	7.8E-07	2.9E-08
1000G K_n (cm/s)	1.7E-07	4.3E-08
Interpreted NAPL Mobility Condition	**Potentially Mobile**	**Residual**
Field Hydraulic Gradient	0.005	0.005
Field NAPL Velocity Calculation [$V_n = K_n i_f / (nS)$]		
Field NAPL Velocity (If Mobile) (cm/s)	1.9E-08	0.0E+00
Field NAPL Velocity (If Mobile) (cm/year)	**0.6**	**0**

TABLE 2. Example results for single-step centrifuge tests with 1000G spin for 1 hour.

Parameter	Example 3	Example 4
porosity, n	0.46	0.53
NAPL saturation, S initial (%)	29.2	9.3
NAPL saturation, S after 1000G (%)	14.2	9.3
1000G K_n (cm/s)	9.8E-08	0.0E+00
Interpreted NAPL Mobility Condition	**Potentially Mobile**	**Residual**
Field Hydraulic Gradient	0.005	0.005
Field NAPL Velocity Calculation [$V_n = K_n i_f / (nS)$]		
Field NAPL Velocity (If Mobile) (cm/s)	3.6E-09	0.0E+00
Field NAPL Velocity (If Mobile) (cm/year)	**0.1**	**0**

Based on evaluation of 19 multi-step centrifuge test results (10G, 100G, and 1000G), we have found that the conclusions from the 10G step are usually comparable to literature-reported NAPL residual saturation values. Specifically, only 1 of 12 samples that had an initial NAPL saturation less than 20% indicated NAPL mobility at 10G, but 5 out of 7 samples that had initial NAPL saturation greater than 20% did indicate NAPL mobility at 10G. For the one sample with an initial NAPL saturation less than 20% that indicated NAPL mobility at 10G, that driving force may have been sufficient to mobilize even residual NAPL based on site-specific soil permeability and NAPL-water interfacial tension values (Cohen and Mercer, 1993).

K_n values calculated based on 10G, 100G, and 1000G test steps are generally similar and sometimes show a sequential decrease from step to step, as seen in Example 1 (Table 1). In these cases, the result from the first step (10G) is considered the most representative of field conditions. The sequential decline in K_n values may relate to the reduction in NAPL relative permeability due to declining NAPL saturation during each successive phase of the test.

Based on similar calculations from dozens of NAPL mobility test results, K_n values for PAH-rich NAPL were consistently in the range of 10^{-6} cm/sec or less. Screening-level calculations of NAPL velocities in the field - estimated using calculated K_n, porosity, NAPL saturation, and field hydraulic gradient - were typically in the range of a few centimeters per year or less (if mobile).

CONCLUSIONS

Laboratory tests of NAPL mobility in soil and sediment samples often impose driving forces that are unrealistically strong to allow test completion within a reasonable period of time. However, the extreme test conditions can mobilize NAPL that is extremely unlikely to be mobile under field conditions. Centrifuge forces of 100G and 1000G were found to produce NAPL movement in some samples with initial NAPL saturations as small as 3.7% and 2.5%, respectively, which are significantly less than residual saturations typically reported in the literature. In addition, water-drive hydraulic gradients are often extremely strong compared to field conditions. The authors of this paper interpret that NAPL movement under these extreme test conditions does not inform whether the initial NAPL saturation is potentially mobile or residual in the field. However, the absence of NAPL movement provides a high level of confidence that the NAPL in a given sample is immobile under field conditions.

Test results with a centrifuge force of 10G are comparable to literature-reported NAPL residual saturation values, with few indications of NAPL movement at less than 20% initial NAPL saturation and relatively consistent NAPL movement at greater than 20% initial NAPL saturation.

K_n values calculated based on 10G, 100G, and 1000G test steps are generally similar and sometimes show a sequential decrease in calculated K_n values from step to step. In these cases, the result from the first step (10G) is considered the most representative of actual field conditions.

Calculated K_n values from 10G and single-step 1000G tests were almost exclusively in the range of 10^{-6} cm/sec or less. NAPL field velocities were estimated at a screening level using calculated K_n, porosity, NAPL saturation, and field hydraulic gradient, and the resulting NAPL velocity estimates were typically in the range of a few centimeters per year or less.

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