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Towards characterizing LNAPL remediation endpoints

Kaveh Sookhak Lari[a,b,c,*], John L. Rayner[a,b], Greg B. Davis[a,b,d]

[a] CSIRO Land and Water, Private Bag No. 5, Wembley, WA, 6913, Australia
[b] Cooperative Research Centre for Contamination Assessment and Remediation of the Environment (CRC CARE), Australia
[c] School of Engineering, Edith Cowan University, 270 Joondalup Drive, Joondalup, WA, 6027, Australia
[d] School of Earth Sciences, The University of Western Australia, 35 Stirling Highway, Crawley, WA, 6009, Australia

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ABSTRACT

Remediating sites contaminated with light non-aqueous phase liquids (LNAPLs) is a demanding and often prolonged task. It is vital to determine when it is appropriate to cease engineered remedial efforts based on the long-term effectiveness of remediation technology options. For the first time, the long term effectiveness of a range of LNAPL remediation approaches including skimming and vacuum-enhanced skimming each with and without water table drawdown was simulated through a multi-phase and multi-component approach. LNAPL components of gasoline were simulated to show how component changes affect the LNAPL's multi-phase behaviour and to inform the risk profile of the LNAPL. The four remediation approaches along with five types of soils, two states of the LNAPL specific mass and finite and infinite LNAPL plumes resulted in 80 simulation scenarios. Effective conservative mass removal endpoints for all the simulations were determined. As a key driver of risk, the persistence and mass removal of benzene was investigated across the scenarios. The time to effectively achieve a technology endpoint varied from 2 to 6 years. The recovered LNAPL in the liquid phase varied from 5% to 53% of the initial mass. The recovered LNAPL mass as extracted vapour was also quantified. Additional mass loss through induced biodegradation was not determined. Across numerous field conditions and release incidents, graphical outcomes provide conservative (i.e. more prolonged or greater mass recovery potential) LNAPL remediation endpoints for use in discussing the halting or continuance of engineered remedial efforts.

1. Introduction

The release of hazardous organic chemicals including light non-aqueous phase liquids (LNAPLs, such as petroleum hydrocarbons) into the vadose zone and groundwater is a significant environmental concern due to its potential adverse effects (Davis et al., 2009). LNAPLs form an immiscible liquid plume in the vadose zone and across the capillary fringe (Lenhard et al., 2018). This induces the partitioning of compounds into gaseous and aqueous phase exposure pathways (Lang et al., 2009; Rivett, 2014; Davis et al., 2005). An initial critical step for remediation of an impacted site is to recover LNAPL through appropriate remediation techniques. To enhance removal of LNAPL mass, various forms of recovery methods may be applicable. These include air, water or solvent flushing or single, dual and multi-phase purging of LNAPL, soil gas and water (Khan et al., 2004; Davis et al., 2013) (Johnston and Trefry, 2009). However, as the dynamics of LNAPL in the subsurface is a function of different parameters (including geo-physical properties of the porous media and the distribution and composition of the LNAPL itself), the feasibility and effectiveness of each (or any combination) of the aforementioned techniques for a particular site is a question to be answered prior to any remedial effort.

After early and primary stages of pumping LNAPL out of an aquifer, a considerable amount of LNAPL may still remain in the subsurface (Lenhard et al., 2018). This is mostly due to the dominance of capillary forces and therefore, a secondary or tertiary recovery effort may be required to remove less-mobile LNAPL (Hernández-Espriú et al., 2012). These may include application of dual and multi-phase recovery techniques. However, information and data in the literature with respect to how to best operate recovery methods to gain an optimum long-term LNAPL recovery are not extensive (Jeong and Charbeneau, 2014; Johnston and Trefry, 2009).

Analytical models have been used to estimate long-term LNAPL mass recovered (in the liquid phase) through multi-phase recovery techniques (Charbeneau et al., 2000). A number of studies have explored the level of LNAPL mass removal required to significantly reduce the net flux dissolved in groundwater (Huntley and Beckett, 2002;

* Corresponding author.CSIRO Land and Water, Private Bag No. 5, Wembley, WA, 6913, Australia.
E-mail address: kaveh.sookhaklari@csiro.au (K. Sookhak Lari).

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DiFilippo and Brusseau, 2008; Johnston et al., 2013). None of these studies indicate the extent of mass removal that may be achievable. Also, the rate of mass removal during active remediation is likely to decrease over time and may become comparable or less than that due to natural mass loss and biodegradation processes – referred to as LNAPL natural source zone depletion (NSZD) (Garg et al., 2017; Chaplin et al., 2002; Johnson et al., 2006). Such an occurrence would be a trigger to consider NSZD as an ongoing management option, compared to continued engineered mass removal efforts. Rates for NSZD via subsurface partitioning and biodegradation are increasingly reported in the literature (Barry et al., 2002; Mulligan and Yong, 2004; Prommer et al., 2000). To enable the comparison with NSZD mass removal rates, rates of mass LNAPL recovery via engineering approaches need to be better estimated over prolonged periods to yield feasible endpoints.

Numerical studies have partially addressed the quantitative endpoint problem. Gabr et al. (2013) applied the Bioslurp model and determined 4.5 years as an endpoint to a multi-phase recovery plan. Skinner (2013) conducted a similar numerical study to predict the endpoint of a skimming approach. Hernández-Espriú et al. (2012) used the API model (a one-dimensional quasi-analytical single component LNAPL model) to investigate the long term (3 year) performance of several multi-phase recovery approaches. Jeong and Charbeneau (2014) presented another analytical model (named LDRM) to study certain types of LNAPL recovery methods. However, no extensive investigation of the effective LNAPL mass removal endpoint has been undertaken taking account of compositional changes within a multi-phase simulation strategy (Sookhak Lari et al., 2018). Such an approach is required to best capture the physics of three-phase (NAPL, water, and gas) subsurface transport and the partitioning and fate of LNAPL components. The components have different risk profiles, and depletion of components alters the physical properties of the LNAPL itself.

A list of recent multi-phase and multi-component simulation studies using multi-phase numerical codes can be found in Sookhak Lari et al. (2016a). The serial-processing code TMVOC (which is a member of the TOUGH2 family of simulators (Pruess and Battistelli, 2002)) is able to represent key features of multi-component LNAPL transport and partitioning in porous media. This code has also been presented in a parallel version (TMVOC-MP) to cope with more complicated problems in terms of geometry, mesh resolution and the number of partitioning compounds in LNAPL (Zhang et al., 2007). However, multi-phase numerical simulators (including TMVOC) have not been used with component partitioning to determine an effective endpoint to field-scale LNAPL remedial approaches (Sookhak Lari et al., 2018).

Recently TMVOC-MP was verified on a CRAY supercomputer through a three-dimensional multi-phase and multi-component simulation of various multi-phase LNAPL remediation approaches applied sequentially over a 78-day period (Sookhak Lari et al., 2018). Here we use the same modelling framework to numerically assess the long term performance of various LNAPL remediation approaches including skimming alone and skimming with vacuum enhanced extraction, and with both these methods applied with and without water table drawdown. We consider five types of soils including sand, loamy sand, loam, silt and silty clay - spanning over three orders of magnitude in permeability with significantly different soil moisture characteristic curves over this range. Both finite and infinitely extended LNAPL plumes were assumed and for each, a high and a low initial LNAPL specific mass is considered. The superposition and performance of more than one recovery wells are not addressed here. Biodegradation is excluded and therefore, the results represent a conservative (upper limit) endpoint, since by excluding biodegradation processes, greater LNAPL mass will be preserved in the subsurface, creating predictions of longer time periods of potentially greater mass recovery to achieve an asymptotic LNAPL recovery endpoint for the cases studied here.

This is the first time an approach combining multiphase partitioning and phase mobility has been investigated to determine the physical (conservative) endpoints for LNAPL remedial techniques. This has been established on a supercomputer to address 80 scenarios of LNAPL recoverability to provide simulations in a new nomograph style for adoption by industry and regulators. Overall, these novel outcomes allow quantitative consideration of upper-limit endpoints as criteria by which remedial efforts might be halted or continued at LNAPL impacted sites.

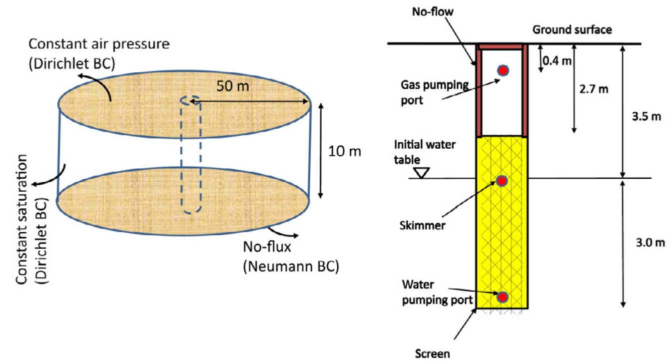

Fig. 1. The simulation domain and the boundary conditions (left); The recovery well configuration (right).

2. Modelling scenarios

2.1. Site layout and soil/aquifer properties

We consider an area 100 m in diameter and 10 m in depth (Fig. 1 left) with an initial water table elevation 3.5 m below the surface. A multi-phase recovery well is located at the centre (Fig. 1 right). We consider five types of soils introduced in Table 1 (Carsel and Parrish, 1988). They include sand, loamy sand, loam, silt and silty clay and span over three orders of magnitude with respect to the soil permeability. Also the van Genuchten parameters for the soils are adopted from Carsel and Parrish (1988) which shows the diversity in their soil moisture characteristic curves.

2.2. Initial LNAPL characteristics and distribution

LNAPL gasoline was considered, due to the global abundance of its release incidents and also since it includes a wide range of compounds with very different partitioning attributes and risk profiles (Lekmine et al., 2017; Vasudevan et al., 2016; Sookhak Lari et al., 2016b). Several studies have reported the subsurface composition of gasoline (GSI Environmental Inc., 2012; Kaplan et al., 1997; Lekmine et al., 2017). Here we use the reported composition for a weathered gasoline in Sookhak Lari et al. (2016b). Major components in the gasoline were bundled into 7 representative groups, as introduced in Table 2 (Lekmine et al., 2017). The composition and thermo-physical properties of these groups are reported in Table 3.

We consider two types of architecture for the LNAPL plume; the Finite case (FIN) and the Infinite case (INF). For the FIN cases, 4945 kg and 24728 kg of mass released over a circle around the well with a

Table 1
Soils used for the simulations and their hydraulic properties; k is the permeability, K is the hydraulic conductivity and n and α are the van Genuchten water retention parameters (Carsel and Parrish, 1988).

ID	Soil	k (m^2)	K (cm/s)	n	α (1/m)
S1	Sand	8.25×10^{-12}	8.25×10^{-3}	2.68	14.5
S2	Loamy sand	4.05×10^{-12}	4.05×10^{-3}	2.28	12.4
S3	Loam	2.89×10^{-13}	2.89×10^{-4}	1.56	3.6
S4	Silt	6.94×10^{-14}	6.94×10^{-5}	1.37	1.6
S5	Silty clay	5.56×10^{-15}	5.56×10^{-6}	1.09	0.5

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