THE MIGRATION OF VISCOUS NAPLS IN ALLUVIUM: IMPLICATIONS FOR BROWNFIELDS REDEVELOPMENT AND SURFACE-WATER CONTAMINATION

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ABSTRACT

Coal tar, creosote and similar viscous NAPLs behave in alluvial soils in a manner significantly different from less viscous NAPLs such as gasoline and chlorinated solvents. Their unique behavior is due to the interaction of their physical-chemical parameters: a density often greater than water, a viscosity significantly greater than water, and an interfacial tension that yields a positive initial spreading coefficient at air/water/NAPL interfaces. This results in slow, creeping flow that causes long-term contamination at former manufactured gas plants and wood-preserving sites and of their adjacent surface waters, e.g., Sydney Tar Ponds, Nova Scotia and sites along the lower Fraser River in British Columbia. Multiphase simulations of this creeping flow are shown for a site near Vancouver, B.C. and the long term consequences of the migration of viscous NAPLs in alluvium are discussed from the perspective of site characterization and brownfields redevelopment.

RÉSUMÉ

La créosote, le goudron et autres liquides visqueux (NAPL en anglais) se comportent dans les sols alluvionnaires de façon bien différente de ceux moins visqueux, par exemple, l'essence et les dissolvants chlorés. Leur comportement unique se doit à l'interaction de leurs parameters chimio-physiques: une densité souvent plus importante que celle de l'eau, une viscosité bien plus élevée que l'eau et une tension interfaciale qui produit un coefficient initial d'extension positif aux interfaces de l'air, de l'eau et de l'huile. Il s'ensuit que ces huiles visqueuses se répandent lentement et entraînent la contamination à longue terme de certains sites qui ont utilisé ces liquides immiscibles, et des cours d'eau adjacents. Nous présentons des simulations numériques de la migration de ces huiles visqueuses en sol alluvionnaire dans un site en Colombie Britannique. Nos résultats indiquent que cette huile visqueuse peut se répandre pendant plusieurs années avant de déboucher dans des fleuves à des niveaux plus bas.

1. INTRODUCTION

The re-development of former industrial sites – "brownfields" – poses some unique problems in geo-environmental engineering. One that is receiving increased attention arises when the sites being re-developed contain significant amounts of remaining non-aqueous phase liquid (NAPL) still in place. These NAPLs may be fuel hydrocarbons, chlorinated solvents or viscous oils. It is this latter group of contaminants, particularly coal tars and creosote, which is the subject of this presentation to the Canadian Geotechnical Society and the International Association of Hydrogeology.

Viscous NAPLs have long been associated with geo-environmental issues in Canada because of the widespread generation of coal tar in manufactured gas plants (MGP) in Canadian cities and the treatment of wood products with creosote at sites across Canada. Perhaps the first published case of NAPL contamination in Canada was the migration and attempted control of creosote into Thunder Bay presented at the 1978 Ontario Industrial Waste Conference (Thompson et al., 1978). Since that time, numerous problems of viscous NAPL migration have been described along the Fraser River (Patrick and Anthony, 1998), the Bow River in Calgary (Reitman et al., 1992) and at the Sydney Tar Ponds in Nova Scotia. Numerous other sites exist across Canada and will require various degrees of remediation, e.g., steelworks, shipyards, oil storage facilities. Measured physical-chemical properties of some viscous NAPLs collected from MGP and wood-preserving sites in North America are summarized in Table 1. Over time many of these viscous NAPLs have become more viscous through the dissolution of their low molecular weight components, i.e., they have 'weathered' in-situ.

When site re-development occurs, three issues of geotechnical and geoenvironmental consequence have been suggested:

1. the lowering of the ground-water table either by reduction in the available infiltration surface from the construction of a new building or by de-watering (or both) will promote NAPL drainage from the newly-unsaturated soils;

2. the potential settlement of a new structure built over compressible soils due to ground-water extraction to contain pre-existing contamination or facilitate construction and the potential for extrusion of NAPL during soil consolidation; and

3. the migration of NAPL due to its accumulation in the subsurface under conditions that may be enhanced by water-table lowering.

The intent of this paper is to consider the potential for each of these processes to affect the migration of viscous NAPLs, in particular creosote and coal tar, at sites that are underlain by unrecovered or even undiscovered NAPL.

Table 1. Physical-chemical properties of viscous NAPLs recovered from the field.

NAPL & Origin	Density [kg/L]	Viscosity [cP]	Interfacial Tension [dynes/cm]	Spreading Coefficient [dynes/cm]
Coal Tar[1] Stroudsburg, Pennsylvania	1.017	19 (7ºC)	22	+ ve[5]
Coal Tar Ottawa, Ontario	1.15- 1.20	160-750 (24ºC)	n/m[4]	--
Creosote[2] Burnaby, British Columbia	1.06- 1.07	20 – 21 (10ºC)	3-6	+ ve[5]
Creosote Cape Fear, North Carolina	1.11- 1.12	41-67 (13ºC)	17	17
Creosote[3] Laramie, Wyoming	1.03	54	n/m	--
Navy Special Fuel Pearl Harbor, Hawaii	0.91	~1000 (23ºC)	n/m	--
Diesel Fuel Washington State	1.08	12,000 (12ºC)	n/m	--
Weathered jet fuel Hill AFB OU1, Utah	0.88	36	8	36
Weathered gasoline former refinery site, Ohio	0.88	6.9	12	32

[1] from Villeaume (1985)
[2] from Golder Associates Ltd., 1999. Report on 8335 Meadow Avenue, Burnaby BC, Volume III, Appendix VI.
[3] Sale and Applegate (1996)
[4] n/m indicates not measured, therefore a spreading coefficient could not be computed
[5] Deduced on the basis of the very low reported NAPL-water IFT

2. GRAVITY DRAINAGE OF NAPL DUE TO WATER-TABLE LOWERING

While two-phase systems of NAPL and ground water are of great interest to geoenvironmental community, little attention has been paid to the effect of air entry into two-phase systems. However, the resulting three-phase systems (air-water-NAPL) have significantly lower residual NAPL saturations than the two-phase system of water and the same NAPL and may promote the migration of viscous NAPLs. These lower three-phase residual saturations for the same aquifer material under otherwise identical conditions (i.e., except for air entry) were documented over ten years ago in the experimental studies of a lighter-than-water NAPL (LNAPL) by Wilson et al. (1990) and of a denser-than-water NAPL (DNAPL) by Schwille (1989). Mercer and Cohen (1990) summarized this significant difference in residual saturations for NAPL as follows: the range in the vadose (or three-phase) zone is 0.10 to 0.20 whereas the range in the ground-water (or two-phase) zone is 0.15 to 0.50. Table 2 shows the data of Wilson et al. (1990) and Schwille (1989) for two- and three-phase conditions of residual saturation.

Table 2. Residual saturations in 2-phase and 3-phase systems with clean sand of hydraulic conductivity = 10^{-2} cm/s (CHC = chlorinated hydrocarbon DNAPL; Soltrol = Soltrol-130 LNAPL).

System	Condition	Saturation (%) or Retention Capacity (L/m^3)	Reference
Air + Soltrol + water	3-phase	9.1 ± 2.2 %	Wilson et al. (1990)
Soltrol + water	2-phase	27.1 ±1.7 %	Wilson et al. (1990)
Air + CHC + water	3-phase	30 L/m^3	Schwille (1989)
CHC + water	2-phase	50 L/m^3	Schwille (1989)

Kemblowski and Chiang (1990) pointed out the difference in these ranges and how they affect the thickness of LNAPL measured in monitoring wells. They also observed and quantified the accumulation of free-phase LNAPL on a declining water table that is a direct result of the conversion of a large part of the LNAPL zone from two- to three-phase conditions. The specific volume of NAPL remobilized (H_m) by a decline in the water table (Δz) is simply:

$$H_m = \phi\left(S_{ors} - S_{orv}\right)\Delta z \qquad [1]$$

where ϕ is porosity, S_{ors} is the two-phase or ground-water zone residual NAPL saturation and S_{orv} is the three-phase or vadose zone residual NAPL saturation. Figure 1 shows how much NAPL drains downward following a one-meter drop for given two- and three-phase residual saturations. Using Wilson et al.'s (1990) values S_{ors} = 27%, S_{orv} = 9%, and ϕ = 35%, Equation (1) yields a specific volume of 0.35(0.27-0.09)1 = 0.063 m^3 or 63 L of NAPL per square meter of aquifer area mobilized downward.

The potential for downward migration of the NAPL in the ground-water zone is given by the Bond number, N_B, (Delshad et al., 1996, p. 320):

$$N_B = kg\left(\rho_n - \rho_w\right)/\,\sigma \qquad [2]$$

where k is the geosystem permeability, g is the gravitational constant, ρ_n and ρ_w are the densities of the NAPL and water, respectively, and σ is the NAPL-water interfacial tension. Once dewatering occurs, NAPL will primarily be displacing air in which case the density difference will be far greater and the vertical migration of the NAPL will be facilitated. The presence of films of water remaining on the soil grains of the unsaturated zone acts to promote long-term drainage of DNAPL or LNAPL to very low saturations in the case of NAPL-water systems with positive spreading coefficients, which appear to be the majority of cases of field NAPLs (Dwarakanath et al., 2002, Table 1).

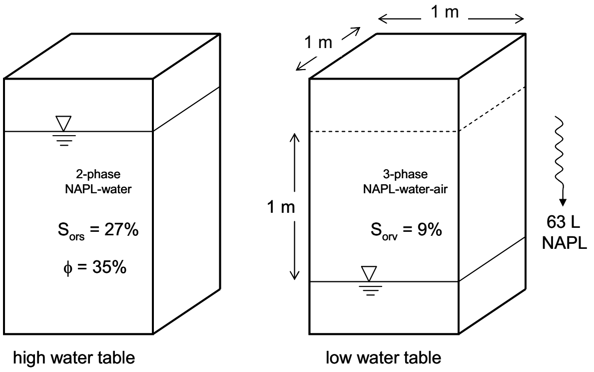

Figure 1. Schematic of drainage of NAPL due to water table lowering.

In two-phase, water-wet systems, residual NAPL exists primarily as discontinuous blobs within a single pore or several adjacent pore bodies (Chatzis et al., 1983; Schwille, 1988; Conrad et al., 1992; Powers et al., 1992). Generally, the residual NAPL will reside in the larger pores where capillary forces are smallest. Wilson et al. (1990) observed residual NAPL saturations ranging from 14% to 30% in unconsolidated sands and possibly higher in heterogeneous sand packs. Indeed, quantitative measurements of localized fluid saturations using dual gamma spectroscopy have revealed macroscopic entrapment of NAPL at saturations well above 30% in heterogeneous sand packs (Illangasekare et al., 1995).

In three-phase systems, the presence of air can alter the distribution of the NAPL dramatically. A schematic of the air-water-NAPL interface is depicted in Figure 2. The spreading tendency can be evaluated by performing a force balance on the interfacial tensions shown in Figure 2. If the air-water interfacial tension (σ_{aw}) is greater than the vector sum of the NAPL-water interfacial tension (σ_{nw}) and the contribution of the air-NAPL interfacial tension (σ_{an}), then the NAPL will tend to spread on the air-water interface:

$$\sigma_{aw}\cos\alpha - \sigma_{nw}\cos\varepsilon - \sigma_{an}\cos\beta > 0\,, \quad \text{NAPL spreading} \qquad [3]$$

Where α, β and ε are the wetting angles as shown in Figure 2. This relationship is often simplified for the case of α = β = ε = 0°, and the condition for oil spreading is defined in terms of the initial spreading coefficient (S) as follows (e.g., Miller and Neogi, 1985; Charbeneau, 2000, p. 473):

$$S \equiv \sigma_{aw} - \sigma_{nw} - \sigma_{an} \qquad [4]$$

When the spreading coefficient is positive, the NAPL will spontaneously spread in the form of a thin film between the air and water phases. Under these conditions, the residual NAPL saturation tends to be much lower than that for a two-phase system (Wilson et al., 1990). Measured interfacial tensions for several NAPL-water systems are summarized in Table 1 along with the associated spreading coefficients. These data indicate the common occurrence of both spreading viscous NAPLs under field conditions. Laboratory centrifuge experiments using a spreading weathered gasoline and diesel LNAPL and soil from an Ohio aquifer indicate a local residual LNAPL (weathered gasoline) saturation of approximately 1% under three-phase conditions.

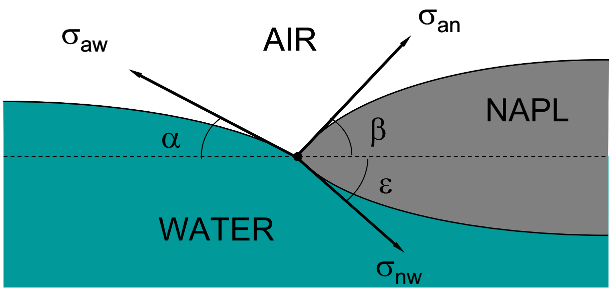

Figure 2. Schematic of air-NAPL-water interface.

Thus, gravity drainage and interfacial spreading may mobilize significant volumes of viscous NAPLs to the water table until it reaches a capillary barrier or is trapped as two-phase residual (Figure 3). In the unsaturated zone, only the relatively low, three-phase residual NAPL will be left behind. In the case of a DNAPL such as creosote or coal tar, if the DNAPL is already or should become pooled on a capillary barrier beneath the water table (e.g. bedrock or clay) and further DNAPL is mobilized downward as a result of dewatering, the pool thickness will increase (Figure 4). This, in turn, will lead to increased capillary pressure at the capillary barrier interface. The capillary pressure (p_c) at the bottom of the DNAPL pool can be calculated as:

$$p_c = (\rho_n - \rho_w)Hg \qquad [5]$$

where H is the height of the DNAPL pool as measured in a monitoring well. If this capillary pressure exceeds the entry pressure of the capillary barrier below it, the DNAPL may migrate deeper into previously uncontaminated regions. In such a case, the integrity of the capillary barrier will be of critical importance. Feenstra and Cherry (1996, p. 445) show this effect in their photograph of the migration of creosote through a fracture in lacustrine clay near Winnipeg, Manitoba.

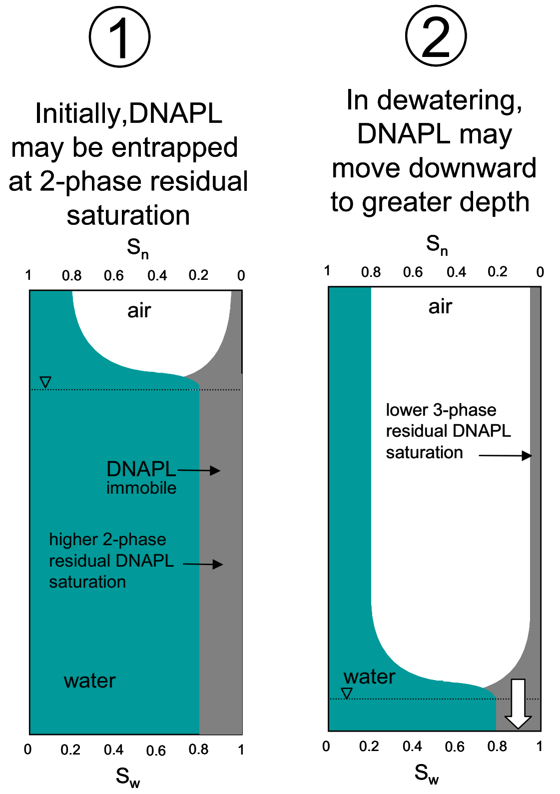

Figure 3. Concept model of dewatering a residual DNAPL source zone.

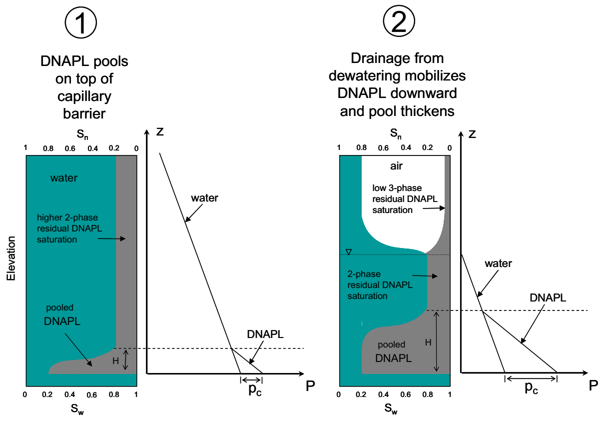

Figure 4. Conceptual model of DNAPL pool atop capillary barrier before and after dewatering.

3. SETTLEMENT, CONSOLIDATION & NAPL EXTRUSION

In soft soils, such as those of the Fraser River Delta in British Columbia, the extraction of ground water from beneath a brownfields site – either due to its contamination or for maintaining a low water table to prevent ingress – may cause consolidation in fine-grained soils and subsequent building settlement. Therefore, it is critical that site investigation produce reliable information on the spatial distribution and volume of viscous NAPL that might be present at a brownfields site.

For the wood-preserving site near Vancouver BC that is considered in this paper, it was unfortunate that the site investigation that was undertaken in the early 1980s failed to reveal the extent of contamination present and so the construction of a warehouse proceeded on the site. Subsequently, continued creosote migration into the Fraser River for 20 years following site de-commissioning lead to regulatory actions initiating ground-water extraction, subsequent building settlement, and finally litigation over the recovery of costs for the $30 million remediation program now completed. The site is typical of those along the Fraser River Delta (Patrick and Anthony, 1998). It is underlain by a thick sequence of fluvial sands and overbank silts that are shown in Figure 5 as a fence diagram based upon historical records. The hydrogeology and hydrostratigraphy of the Fraser River Delta have been described in detail by Neilson-Welch and Smith (2001) and Monahan et al. (1994).

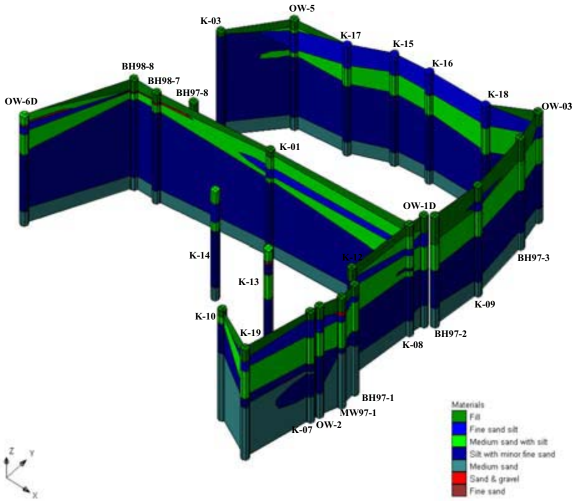

Figure 5. 3-D fence diagram of stratigraphy developed based on well log information.

One issue that arose during litigation was the potential for extrusion of NAPL from the soft soils during preloading the site prior to construction. Most of the soil materials beneath the site are silts and sands, although some clay has been reported. Investigations in the Fraser River delta of soil consolidation reach back to the 1960s and the nature of preloading-induced consolidation over time is well known, although not fully understood (Crawford & Morrison, 1996). However, NAPL extrusion could not be expected if the compressible soils present a capillary barrier to NAPL penetration. Thus, the issue that arises is ‘has the creosote invaded compressible soils’? Here it is possible to turn to empirical equations that define the entry or penetration pressure for a particular NAPL into such soils (McWhorter and Kueper, 1996).

The following parameter values may be considered (see Table 1): 1.1 g/cm^3 for the density of creosote, 4 dynes/cm for the creosote/water interfacial tension, and 0.3 for the porosity of the silty clay, and a hydraulic conductivity of the silty clay of 10^{-9} m/s. In such a geosystem, the creosote would have to be present as a continuous vertical column of more than 20 meters in height for it to penetrate the silty clay that was collected and tested from 8 meters depth below ground surface at the site. Even if such entry pressure was achieved, the low permeability of the silty clay would inhibit significant volumes of creosote from entering the clay. But, more importantly, as such a vertical column might build up at the contact between the sand and the silty clay beneath it, there would be a pronounced lateral migration of the NAPL along the contact as the relative permeability of the NAPL increased in the sand with respect to the ground water. This lateral displacement due to a head buildup is a common feature in spills of DNAPL into sand-tank models (e.g., Kueper et al., 1989). Thus, penetration and subsequent extrusion do not appear to be a particular concern that might arise from consolidation; building settlement would remain as a significant hazard.

4. NAPL MIGRATION

Numerical multi-phase simulations of viscous NAPL migration (e.g., Reitman et al., 1992; Wu et al., 2000) owe their reliability to the development of petroleum reservoir simulators such as UTCHEM, the University of Texas Chemical Flood Simulator (Delshad et al., 1996; Pope et al., 1999). The permeability distribution in this cross section is shown in Figure 6. The Upper Sand aquifer is given a permeability of 21 darcies, for which the equivalent hydraulic conductivity is 2.1 x 10^{-4} m/s. The silty aquitards have an assigned permeability of 100 millidarcies, for which the equivalent hydraulic conductivity is 1 x 10^{-6} m/s. This permeability contrast is sufficient to cause the creosote NAPL to migrate laterally through the geosystem.

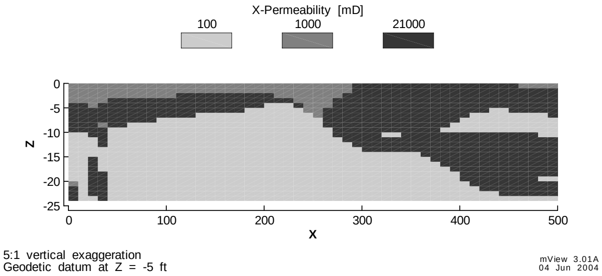

Figure 6. Permeability distribution in the geosystem.

The initial NAPL saturation is presented in Figure 7 and represents the zone of high creosote contamination believed to have been caused by the discharge of waste-water and creosote condensate from the operations prior to 1983. This model depicts the 'loading' of the model with the historical creosote that existed northwest of K-1. Figure 7 also shows that, with the exception of three cells that are part of the Upper Silt, the cells from x=100 ft to x=310 ft are 'loaded' with creosote at a saturation of 40%, i.e., 40% of the pore space is occupied by creosote, the remainder by ground water. The porosity of these cells, which are part of the fill material and Upper Sand aquifer, is 30%. These cells occupy the first five layers of the model in depth and thus represent a total creosote volume of 40 ft x 210 ft x 5 ft x 0.4 x 0.3 (minus the three silt cells) = 4,970 cubic ft or 141 cubic metres and represent the source zone along the axis of a culvert that connected a waste-water lagoon to the Fraser River.

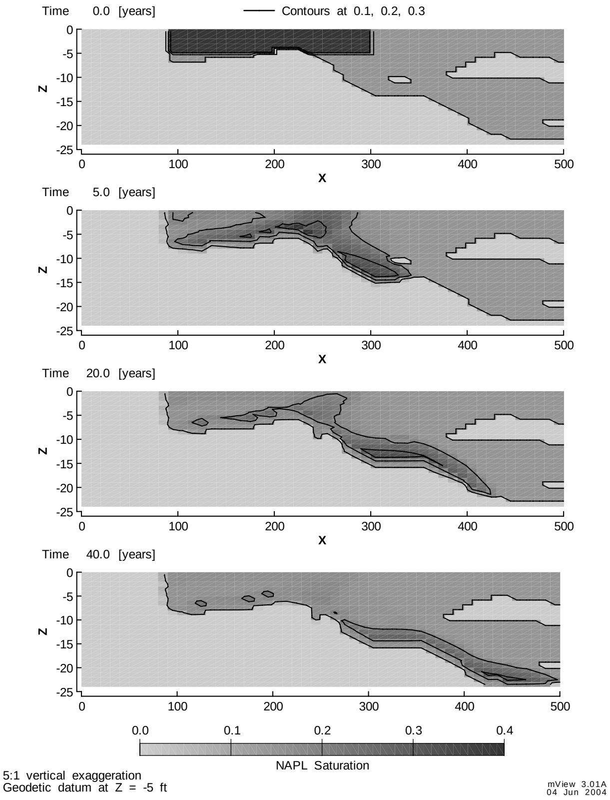

Figure 7. NAPL saturations.

Patrick and Anthony (1998) estimated the NAPL saturation as varying between 0 and 20% in the foreshore sediments and free-phase NAPL (i.e., that which is mobile due to it being at saturations greater than residual) was observed up to depths of 20 m below the ground surface (bgs). The saturation of 40% the source zone was chosen on the basis of experience with coal-tar contamination, a similarly viscous NAPL, which has been observed in sand and gravel aquifers by Young et al., 2002. The residual saturation for this model is set at 15%, which means that for any cells with NAPL saturations in excess of 15% there will be flow of mobile NAPL along the NAPL gradient until the cell's NAPL saturation is depleted to residual saturation.

The cells in the sand aquifer in Figure 7 are at residual saturation and therefore will transmit NAPL without a net change in saturation. The volume of the residual NAPL in these cells is approximately 6462 cubic feet or 183 cubic metres of creosote. Therefore the summed volume of creosote NAPL in the line-source model is approximately 324 cubic metres, which is equivalent to 2011 35-gallon (Imperial) drums of creosote or an implied spill rate from 1931 to 1982 of 39 drums per year. The actual spill rate, of course, is unknown, although annual usage appears to have been about one million gallons; a loss of less than one drum per week over 50 years is conservative and does not prejudice the simulations.

The cells at residual saturation in the fill and in the sand aquifer will not release creosote because of the capillary forces retaining it. However, the UTCHEM simulator will allow the creosote in those cells set at 40% saturation to release their NAPL until their saturations fall to residual (15%). Thus, approximately 63% ((0.40-0.15)/0.40) of the NAPL in the heavily contaminated cells in the line-source will become mobile once the simulator begins to run. This constitutes a volume of 89 cubic metres or 550 drums of creosote. The cells initially set at residual saturation are assumed to have been contaminated to that level by previous spills that were absorbed by the capillary forces in the geosystem without having released NAPL to the River. Thus, the model simulates creosote migration under idealized but plausible conditions from the far end of the site to the River and offers insight into whether it is reasonable to expect creosote seepage into the Fraser River tens of years after plant closure.

The initial state of the geosystem at time zero is shown in the top cross section in Figure 7, indicating the large zone of potentially mobile NAPL in the permeable zones that contain NAPL at residual saturation (i.e., non-mobile NAPL). The silts are considered free of NAPL. The progress of the viscous NAPL over a period of 40 years in the same slice of the model is shown in the lower cross-sections in Figure 7. During the 40 years simulated, the NAPL drains and migrates through the geosystem to discharge at the base of the sand aquifer into the Fraser River bottom sediments. Even at 40 years, the simulation indicates that, for the conditions simulated, mobile NAPL remains in the aquifer and continues to migrate. The results of this simulation indicate that mobile NAPL from the interior of the Site takes 30 years to reach the seepage face before it migrating into the Fraser River bottom sediments, which are beyond the boundary of the model domain. The long-term drainage of creosote at the Site may be described as the ‘creep’ of a viscous NAPL.

It may be argued that maintaining NAPL in the source zone at 40% saturation is unreasonable because the residual saturation is only 15%. However, when the warehouse was built in 1983/4, the water table would have declined because of a reduction in the area of rainfall infiltration, and large volumes of NAPL would have been released because the residual saturation of an air-water-NAPL geosystem is very much lower (i.e., 1-2%) than that of the ground-water-NAPL geosystem shown here (i.e., 15%). Thus, the slow release of NAPL due to the lowering of the water table would produce the same effect as is shown in Figures 8 through 10. This same effect of a lowered water table is seen at sites contaminated with gasoline at the end of a dry summer when free-phase gasoline becomes evident in monitoring wells. That is, the physics of drainage are identical, only the viscosities are significantly different and consequently gasoline becomes evident more quickly because of its low viscosity.

5. CONCLUSIONS

Viscous NAPLs, such as coal tar, creosote and heavy fuel hydrocarbons, pose long-term problems at brownfield sites undergoing re-development. Their high viscosity and moderate density (relative to ground water) result in their slow migration through the subsurface that is not readily observed. This creeping flow may cause viscous NAPLs to discharge from brownfields sites to adjacent surface waters for tens of years after their release. It is also argued that, in the case of compressible soils such as may be found in the Fraser River Delta and other alluvial floodplains throughout Canada, the extrusion of NAPL from soils undergoing consolidation is unlikely to be a serious problem. However, the lowering of the local water table, whether by ground-water extraction because of on-site contamination or because of the construction of impermeable surfaces on those which were formally open to infiltration, is a significant and latent problem. The drainage of water-saturated, NAPL-contaminated soils leads also to NAPL drainage. The volume of NAPL that percolates downward may be substantial and may initiate lateral migration towards adjacent surface waters.

Therefore, it is critical that hydrogeologists and geotechnical engineers ensure careful and complete site characterization prior to brownfields re-development. In soft soils this is readily accomplished by means of cone-penetrometer based tools and cohesionless soil samplers. In fractured tills and rock, intensive multi-level sampling should be undertaken to ensure that all NAPL source zones are spatially identified. Long-term pumping tests (i.e., > 24-hours duration) are critical irrespective of the foundation materials in that extraction will establish the potential for subsequent viscous NAPL migration in geosystems that may appear to have no mobile NAPL. These principles of site characterization will minimize the likelihood of costly post-redevelopment remediation.

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