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Multimedia environmental analysis of PCBs fate and transport mechanism through a case study of transformer oil leakage

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Abstract Transformer oil leakage having large quantity of PCBs is one of the most deplorable incidents resulting in huge negative impacts on the environment. In this paper, two different models, i.e. CHEMCAN and the hydrocarbon spill screening models, were applied to the case study adapted from a real case of PCBs in transformer oil spill in Montreal. The oil migration was examined in three dimensions including site-specific data, soil characteristics and hydrogeological properties. This study aims to investigate the fate, transport and transformation rates of PCB in the soil, the unsaturated and the saturated zones. The proposed modelling concept helps to define the processes to be used in characterization of soil and subsurface environment as a receptor of transformer oil spill. This study includes the simulation techniques for assessing the effect of PCBs on groundwater and soil. Results from multimedia environmental model and HSSM model verify with each other, and both show that 92.7 % of PCBs were found in soil as compared to groundwater. Considering decay and sorption processes at spill location, concentration of PCBs in soil was >50 mg/kg. This study can serve as a basis for further analysis and assist in selection of remediation technique.

Keywords Hydrocarbon spill screening model · Multimedia environmental model · CHEMCAN model · Transformer oil spill

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Introduction

Polychlorinated biphenyls (PCBs) have been previously used as a coolant fluids, for example, in electric motors, capacitors and electric transformers. Due to environmental toxicity and consideration as a persistent organic pollutant, PCBs usage was banned in 1979 by the United States Congress (Robertson and Hansen 2001) that announced to eliminate the production by the Stockholm convention on persistent organic pollutant held in 2001 (Porta and Zumeta 2002). According to the US Environmental Protection Agency (EPA), PCBs have been known to cause cancer in animals, as well as in human beings. PCBs are corrosive for the skin, eyes and respiratory tract. It can directly affect nervous and immune system. It can also cause many of the diseases related to skin and consider as carcinogenic compound (Lauby-Secretan et al. 2013; Imran and Hassan 2004). PCB-containing oils can easily catch fire. In such incidents, the oil would decompose and emit certain substances in the smoke that would cause irritation. Moreover, such a fire could lead to deposit particles containing furans and dioxins in the vicinity of affected site (Mddefp 2013). In this research paper, assessment of PCB-transformer oil leakage through the subsurface soil was studied based on the principle of multiphase flow. PCBs in transformer oil is categorized as non-aqueous phase liquid (NAPL) (Environmental Agency 2003), which is long-term sources of subsurface contamination and difficult to clean up (Youdeowei 2012).

When transformer oil is spilled, it flows through the subsurface of the soil to groundwater. On its travel, the transformer oil encounters with air and water phases present in the subsurface. This phenomenon is called multiphase flow, which is also defined as the relative movement of two or more immiscible phases such as gas, water and



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non-aqueous phase liquid (NAPL). Components of the NAPL may exist in the subsurface as four different phases: immiscible liquid, volatile gas phase, dissolved aqueous phase and adsorbed to soil particles. After release, the NAPL moves downwards in the vadose zone due to gravity and capillary forces. Vapour from the NAPL in the vadose zone can move significant distances through the air voids. If a sufficient volume is released, the NAPL may reach the saturated zone. In this instance, a NAPL will float on the water and spread across the capillary fringe. The distribution is then a function of NAPL, air and water pressures and pore-size distribution (Daubermann 2002). In the saturated zone, soluble components of the NAPL may dissolve in groundwater and move as a plume with local flow.

There are many environmental multimedia and numerical transport models that considered the transport mechanism and partitioning behaviour of PCBs in vadose zone as well as in groundwater (Boulding and Ginn 2004; Fagbote and Olanipekun 2010; Wang and Chen 2013). On the other hand, very limited studies were found on modelling of NAPL transport mechanism in porous media (Culliga 1998; Aggelopoulos et al. 2015). However, there are few practical screening models that deal with PCBs and NAPLs together in subsurface soil media. For example, Daubermann (2002) studied NAPL movement from transformer leakages in Cottesloe sand using numerical multiphase model. The study shows difficulties in parameter inputs and computation results in terms of PCBs. Scott (2012) developed a new numerical computer-coded model by coupling transport model and analytic multicomponent NAPL dissolution model to study PAH, phenolic compounds as well as PCBs. Other than complexity in coding, the results were based on steady condition. In contrast to complex numerical screening modelling, simple screening models offer minimum data requirement and provide an alternative approach to study physical behaviour of NAPLs releases through simplifications. An interpretation of hydrological data based on certain assumptions including homogenous soil conditions and uniform aquifer is used in screening models to get the analytical solutions. The hydrocarbon spill screening model (HSSM) is one of the screening models which simulates subsurface leakages of oil with the intent of assessing migration of potential contaminant in the groundwater (Hongkyu et al. 2009). The HSSM consists of three modules: the kinematic oily pollutant transport (KOPT) that treats transport of NAPL through the vadose zone to capillary fringe, OILENS module that simulates the formation and spreading of an oil lens in the capillary fringe and transient source gaussian plume (TSGPLUME) that simulates transport of soluble constituents of the NAPL (light) in the aquifer to receptor locations. This model addresses the questions that how far an oil release might go into the soil and how soon it might

join the groundwater (Weaver et al. 1994). Thus, HSSM was selected as a screening tool in this paper as it deals with multiphase NAPL along with subsurface contaminants and simple to use as well.

There are numerous factors influencing the migration, transformation and degradation of NAPL when it is released into the environment. In order to deal with these factors, several environmental multimedia models exist which attempt to study and predict the partitioning behaviour of contaminant in the environment. They include CEMC Level III (Mackay 2001), CALTOX (McKone and Enoch 2002), MEPAS model which can be used for sites that release radionuclide or toxic chemicals from different sources in landfill and ponds (Strenge and Smith 2006). Other representatives models are HWIR (USEPA 1999), MULTI-MEDIA 2.0 (Solhotra et al. 1995), 3MRA (Babendreier and Castleton 2005), and the one studied in this paper was known as CHEMCAN which is developed by Mackay and his colleague (Mackay et al. 1991). CHEMCAN is one of the notable examples of regional scale models, parameterized for 24 regions of the Canada based on the concept of fugacity. The primary objective of this research paper was to study three-dimensional multiphase flow of PCBs in transformer oil through vadose zone to groundwater using HSSM model. Particularly, formation and spreading of oil lens in the capillary fringes were also studied. This paper also focused on mass transfer of PCBs from NAPL lens to groundwater. Additionally, comparison was made between HSSM and CHEMCAN models in order to check validation of analysis.

Materials and methods

Hydrocarbon spill screening model (HSSM)

The model is intended to address the problem of NAPL flow and migration from the subsurface to a watertable aquifer. The importance of the model is to determine the NAPL lens size and the mass flux of contaminants into the groundwater. These key factors define conditions of the source and must be based upon multiphase flow phenomena in the unsaturated zone. The first two modules of HSSM, i.e. KOPT and OILENS, address the vadose zone flow of the NAPL and its migration. These two modules are combined into one computer code. HSSM-KO, which provides a time-variable source condition for the aquifer model, has separate computer code (Weaver et al. 1994).

$$\eta R \frac{\partial C}{\partial t} = \nabla \cdot D \nabla c - q \cdot \nabla c - \lambda \eta R c + J(t)$$
(1)

where η is the porosity, *R* is the retardation factor, *c* (mg/m³) is the contaminant concentration in the groundwater, *q* (m/s) is Darcy velocity, *D* (m) is the dispersion constant, λ (s⁻¹) is a

first-order decay constant, J(t) (mg/m³ s) is the amount of mass per unit volume of aquifer added per unit time.

A chemical pollutant dissolved in both the NAPL and water phase is tracked by KOPT and OILENS. Once the chemical pollutant reaches the water table, it contaminates the aquifer by contact and by dissolution from the NAPL oil lens. Thus, the third part of the model transported pollutant through the NAPL to aquifer. Notably, the mass flux from OILENS is time dependent, so that the aquifer model must be capable of simulating by varying a time with conditions of the source. In keeping with the level of approximation used in KOPT and OILENS, one suitable choice is the transient source gaussian plume (TSGPLUME) model, which uses different numerical techniques than KOPT and OILENS; so it is not incorporated within HSSM-KO, but rather it is implemented in the computer code HSSM-T (Hongkyu et al. 2009).

The main parameters of HSSM are (1) parameters specifying the magnitude and extent of NAPL, (2) residual oil contents in vadose and groundwater zones, (3) water content in oil lens, (4) transport properties of NAPL and water, i.e. velocity, density, viscosity and surface tension, (4) soil–water retention characteristics, i.e. hydraulic conductivity, porosity, pore-size distribution and water content, (5) characteristics of dissolved residual constituent, i.e. aqueous solubility, soil–water and oil–water partition coefficients, (6) aquifer transport characteristics, i.e. hydraulic gradient, dispersivity values and half-life of constituent within the aquifer.

CHEMCAN model

CHEMCAN is a multimedia model based on fugacity approach designed to estimate the distribution of single chemical pollutant in multiple media. It was developed for 24 different regions of Canada. It permits temporary and permanent additions or changes of environmental properties to a simulation. It estimates average concentration of contaminant in air, soil, sediments and water, etc. It is intended to help in human exposure assessment. In fact, this model can be used to check the validation of modelling results obtained through other screening models for case study of Canada region based on certain conditions (Webster et al. 2004). These conditions are related to the particular characteristics of the selected region including environmental temperature (an annual average/seasonal temperature is assumed), total surface area, precipitation rate, soil-water runoff, leaching from soil, diffusion to the atmosphere and advective flux rates of water and air. There are some characteristics which considered as constants include volume fractions for water and air particles. In addition, there are average depths of water, soil, air, the fraction of organic content of water, suspended particles, solids in the soil and mass transfer coefficient (MTC) and aerosol deposition.

Modelling framework

As inputs, the model required the physical and chemical properties of the pollutant (PCB in this case study) such as water solubility, Henry's law constant, vapour pressure, the octanol–water partitioning coefficient, reaction half-lives in soil, air and water and advective inflow concentrations in water and air. The model then used mass balance equations for multimedia zones taking into account emissions, intermedia-transfer processes, advection and diffusion flux. The mass balance equations were then solved, and mass balance fluxes of chemical input and output rates are calculated as shown in Fig. 1.

Case study

The reliance power equipment was storing polychlorinated biphenyl (PCB)-contaminated materials at their facility since 1998. The plant was located on Hymus Boulevard in the industrial park of Pointe-Claire across the street from residential area and various schools in neighbourhood as shown in Fig. 2. In Canada, the import and manufacturing of PCB were made illegal in 1977. On 26 March 2013, the leak of 800–1200 l of oil containing large concentrations of PCBs had spilled in Reliance Company's warehouse. This incident would result from the opening of the valve of an outside reservoir containing some oil and oily water (Mddefp Report and Press Release 2013; CTV Montreal News 2013).

The analysis of samples taken from the site had confirmed that the oil poured out in 86, Hymus Boulevard contained PCB in concentration of 430 mg/kg. Two days later, PCB-laced oil was found in St.Louis Lake; the results of its sample had concentration of 445 mg/kg (Mddefp Report 2013). It is important to know site characteristics as they play key role in migration of pollutant through subsurface soil to groundwater. Table 1 describes some of the important characteristics of the incident site and all other input parameters required for respective models (IRDA 2013).

Results and discussion

HSSM modelling

The oil migration profile (Fig. 3) depicted the configuration of the oil profile at the selected time. The vertical axis illustrated the configuration of the contaminant in the vicinity of the vadose zone before reaching the water table.





Fig. 1 Flow chart depicting conceptualization and implementation of the proposed framework



Fig. 2 Pointe-Claire, Montreal; the study area (plant location: Hymus Boulevard; area: industrial park; surroundings: schools and residential area)

The spread of oil in radius (m) of vadose zone was indicated by the horizontal axis. The horizontal axis started from the source out to some distance covered by transformer oil. The colours of circles denoted the concentration of PCBs at different depth and distance. PCBs in transformer oil contaminated the vadose zone vertically up to 10 m after 200 days. Whereas, the oil spread horizontally and increased in radius from 0 to 120 m in 200 days. It could be assumed that the PCB reached the water table after 200 days. Rather than accepting the results of one simulation, several times simulations should be run in order to get some feel for the effects of parameter variability. If the hydraulic conductivity was twice greater than the average value of 0.22 m/day, the transformer oil would flow deeper into the subsurface earlier than 200 days.

The oil lens in groundwater, i.e. NAPL, spread laterally in radius as a function of time in saturated zone as shown in Fig. 4. The vertical axis illustrated the configuration of the contaminant in the vicinity of the saturated zone with respect to depth. The spread of oil in radius (m) of



Table 1 Input parameters

Parameters	Values	Parameters	Values
NAPL density (g/cm ³)	0.885	Porosity θ	0.33
NAPL dynamic viscosity (cp)	2.0	Hydraulic conductivity (m/day)	.22
NAPL flux (m/day)	.452	Depth to water table (m)	10.0
Bulk density of unsaturated zone (kg/m ³)	1012	Aquifer saturated thickness (m)	15
Soil-water partitioning coefficient Kd (L/kg)	3.8	PCB in NAPL (ppm)	473
NAPL/water partition coefficient	251	Average infiltration rate (m/day)	$.365 \times 10^{-2}$
NAPL surface tension (dyne/cm) @20 °C	25	Longitudinal dispersivity (m)	10.00
Transverse dispersivity (m)	1.00	Vertical dispersivity (m)	.100



Fig. 3 Transformer oil (PCBs) migration profile in vadose zone within 200 days

groundwater is indicated by the horizontal axis. The lens radius increased rapidly as the oil entered the lens. Later on, the lens tended towards limiting the radius as concentration of contaminant, i.e. PCBs, reduced.

The oil lens contaminant mass balance Fig. 5 illustrated the mass of PCB contained within the oil lens as a function of time in years. Figure 6 indicates the cumulative mass of contaminant, i.e. PCB, which had been dissolved into the groundwater from the oil lens. As the PCB mass contained within the lens declined, the proportionality of cumulative amount of PCB dissolved in groundwater increased.

The concentration of PCB in the soil was more than 92 % based on the modelling result. One of the reasons is PCBs are lipophilic compounds with low solubility in water and have a greater tendency to bind with soil organic matter. In addition, PCBs are slowly degradable and persistent in soils under natural conditions (Pierzynski et al. 2000). In this case study, soil was composed of clay and

well-sorted gravels that impeded flow of transformer oil due to its low hydraulic conductivity, i.e. 0.22 m day^{-1} , as ability to transmit NAPL through vadose zone to ground-water depends upon hydraulic conductivity (Deurer and Bachmann 2007).

PCBs are often blended with carrier fluids such as mineral oil and chlorobenzene. In this case study, the carrier fluid was transformer oil which is classified as light NAPLs (LNAPLs) as it has a density less than water (Daubermann 2002). Depending upon the type of carrier fluid, the density of PCB oils encounters in range from 1100 to 1500 (kg m⁻³), while viscosity is from 10 to 50 cp (Environmental Agency 2003). The relative high density and viscosity indicate that PCBs as a part of dense NAPLs (DNAPLs) may be still migrating at the site where they were introduced through subsurface in the past decade. It means density and viscosity along with other site-specific factors are important for timescale of migration of NAPLs.









Fig. 5 Radius history by NAPL and PCB



Fig. 6 Contaminant mass in oil lens

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Although PCB is considered as persistent organic chemical, due to natural attenuation as the time passes concentration lessen. PCBs, particularly the highly chlorinated congeners, adsorb strongly to soil and sediment where they tend to persist with half-lives for longer period (Kumar et al. 2014).

CHEMCAN modelling

PCB was transferred from NAPL to water, soil and air phases in the subsurface soil. According to Table 2, CHEMCAN multimedia modelling results showed that 92.7 % of PCB sorbed into the soil, while rest of PCBs became the part of groundwater. Advection and diffusion were two main transport mechanism process or the dominant processes in migration of PCB from NAPL to water media and soil media in subsurface environment. The total residence time of contaminant was 1700 days. The value of fugacity for soil was 3.43×10^{-08} Pa (Pascal) and for groundwater was 2.52×10^{-10} Pa.

Comparison of HSSM and CHEMCAN model

The computed results from HSSM model were carefully analysed with respect to physical and dynamics of pollutant, i.e. PCB, in subsurface environment. Then, the simulated results from HSSM model were compared to those calculated from the similar case study by using regional multimedia model CHEMCAN to verify the performance of model. According to

Compartment	Z value (mol/m ³ Pa)	Fugacity (Pa)	Amount ^a (%)	Concentrations (g m ⁻³)
Groundwater	0.268	4.98E-10	4.21	1.27E-05
Soil	1.60E+02	3.43E-08	92.7	1.03E-03

^a Rest of percentage was divided among air voids, water present in vadose zone

Environment	Average predicted PCB concentration ^a (g m ⁻³)		Advection flux (g $m^{-2} day^{-1}$)		Diffusion flux (g $m^{-2} day^{-1}$)	
	HSSM	CHEMCAN	HSSM	CHEMCAN	HSSM	CHEMCAN
Soil	4.67E-03	1.03E-03	1.78E-03	2.33 E-03	3.65E-01	1.43E-01
Groundwater	1.61E-03	1.27E-05	8.60E-03	4.8E-02	1.80E-04	6.45E-03

^a Concentrations were based on average of first year of incident

HSSM, concentration of PCB in soil was 4.67×10^{-3} g m⁻³ after 1788 days (approximately more than 4 years), whereas by using CHEMCAN multimedia model PCB in soil was 1.03×10^{-3} g m⁻³ for the same timescale (see Table 3). Both models predicted almost the same value of PCBs in soil. Most of the time migration of pollutant depended upon the porosity and permeability of the soil.

The concentration of PCB in groundwater after a year using HSSM model was 1.61×10^{-3} g m⁻³. This concentration was gradually increased with time as mass of contaminant in oil lens decreased. Similarly, PCB concentration using CHEMCAN was 1.27×10^{-5} g m⁻³. This concentration was less as compared to HSSM model as in CHEMCAN NAPL phase was not kept into consideration. The average PCB dissolution flux rate was 0.1779×10^{-3} kg day⁻¹ and NAPL flux rate was 0.375×10^{-1} kg day⁻¹ using HSSM model, whereas PCB flux rate to groundwater using CHEMCAN model was 5.3×10^{-4} kg day⁻¹ (see Table 3).

The advection and diffusion flux for soil obtained by CHEMCAN model were 2.33×10^{-3} and 0.1425 g m⁻² - day⁻¹, respectively, whereas advection and diffusion flux for soil obtained by HSSM model were 1.78×10^{-3} and 0.365 g m⁻² day⁻¹, respectively. In addition to this, advection and diffusion flux for groundwater obtained by CHEMCAN model were 0.048 and 6.45×10^{-3} g m⁻² - day⁻¹, respectively, while advection and diffusion flux for groundwater obtained by HSSM model were 8.6×10^{-3} and 1.80×10^{-4} g m⁻² day⁻¹, respectively. Hence, diffusion was the dominant transport mechanism in soil media reported by the two models (see Table 3).

Concentration of PCBs at spill location with decay and sorption

The values of concentration varied if sorption and decay process was considered in model. In Table 4, it is shown that when decay process was considered in model, values were smaller as compared to concentration at spill location. Biodegradation plays an important role in the fate of oil and other petroleum products. Many components of oil are readily degraded by subsurface micro-organisms (Nilanjana and Preethy 2011). In addition, only aerobic bacteria can degrade PCBs oxidatively by attacking the carbon ring and destroying the compounds, whereas anaerobic bacteria can only remove the chlorine and leave the biphenyl rings (Abramowicz 1990). It was also observed from following Table 4 that when sorption process was considered in the model, values of PCB got higher than its concentration at spill location. The reason behind this is that sorption to natural solids is an underlying process affecting the transport mechanism and biological activity of organic compounds in the environment. In fact, sorption may require weeks to many months to reach equilibrium state (Pignatello and Baoshan 1996). In vadose zone, greater amounts of organic matter and metal oxides were present than in the saturated zone. Contaminants could absorb onto these metals and lessen their rate of movement as compared to saturated zone. Materials adhering to these absorbents could serve as a source of contaminants to the saturated zone even after remediation (Youdeowei 2012). In this case study, samples from soil were collected after 4 months showing concentration of PCBs greater than 50 mg/kg (Mddefp Report 2013). This reflects that sorption should be considered while assessing the risk in order to avoid underestimation of the impact.

The Canadian Environmental Protection Act (CEPA) regulates the complete removal and destruction of PCB-contaminated material if concentration is greater than 50 mg/kg (Environment Canada 2001). In this case study, soils containing over 50 mg/kg PCBs were referred to as CEPA soils. All contaminated area should be excavated with diligence to ensure the removal of all contaminated soils (Kalinovicha et al. 2008). High-temperature



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Time (days)	Concentration ^a at spill location (mg/kg)	Concentration with decay (mg/kg)	Concentration with sorption (mg/kg)	Field analysis (mg/kg)
1	480	415	5671	
7	230	215	4382	
14	115.3	100.2	2191	
20	80.7	66	1533	
30	53	40	1022	
60	26.9	15.1	511	
90	17.9	7.3	340	
120	13.4	5.01	255	>50
200	8.07	2.03	153	
365	4.43	0.12	84	

Table 4 Concentration at spill location with decay and sorption

Vertical mixing was considered negligible and assume homogenous soil

^a Concentration at which x = 0 and y = 0 included advection and diffusion

incineration and landfilling are also two traditional methods for remediation of PCB-contaminated soils and sediments (Rahuman et al. 2000). In addition to these, bioremediation is also viable and cost-effective options for remediating soil contaminated by organic and inorganic compounds (Perelo 2010). Surfactants are one of the most popular extracting agents for PCB-contaminated soils. Since surfactant molecules can deal with both hydrophobic and hydrophilic portion, PCBs can be solubilized in the hydrophobic phase in aqueous solution (Billingsley et al. 2002; Chu and Kwan 2003).

The HSSM model uses semi-analytical solutions of the transport equations which include many of the important physico-chemical processes. Based on this approach, it can incorporate three-dimensional effects easily as compared to CHEMCAN. Moreover, HSSM is designed for LNAPLs. It is not suitable for denser than water NAPLs (DNAPLs) as the NAPL is assumed to float over the water table. The vadose zone module of HSSM could, however, be used for a DNAPL, as the qualitative behaviour of that module is not affected by density of fluid (Weaver et al. 1994).

CHEMCAN is one of the multimedia models which is based on regional characteristics and offers great simplicity and minimum data requirements for input of parameters (Kawamoto et al. 2001). As compared to HSSM, this model does not account for partitioning behaviour of PCB in NAPL. Although this model is suitable for large scale, spreading of contaminant plume in radius with respect to time is not considered in CHEMCAN model. According to CHEMCAN, advection and diffusion were primary mechanism for migration of transformer oil which is also true for all NAPLs (Russell and Jon 2004). However, there are certain limitations in HSSM model as it does not include all processes which may be important, and because it is based on semi-analytical solutions, it does not account directly for heterogeneity of the soil. In addition, a 3D multiphase numerical models should be further studied to address the limitations of HSSM model especially heterogeneity of soil as these models are employed to stimulate the migration of NAPLs (Borden 2000).

Conclusion

According to the study made in this paper, the PCBs released into the environment as a part of transformer oil spill was concentrated in two major compartments: (a) soil and (b) groundwater. Based on the results analysis of screening models, i.e. HSSM and CHEMCAN, the occurrence of PCB concentration in soil was 92.7 % as compared to other media of environment. Particularly, more oil was trapped in vadose zone and less was available to migrate to aquifer depending upon porosity of soil. Transfer of PCBs from transformer oil spill to the environment was expected to take place by the two main processes: (a) advection and (b) diffusion. Virtually no evidence was available on volatilization of PCBs or any other transport mechanism. As the time passed, PCB concentration get lessen. The values of PCB concentration found by considering sorption were higher as compared to other processes as it is slow process to attain equilibrium, while decaying of pollutant get smaller value as compared to maximum concentrations. Ignoring sorption and decay process can lead to an underestimation of the true extent of equilibrium, false predictions about the mobility and fate of contaminants, and perhaps the wrong choice of clean-up technology.

The study also illustrates that HSSM model is one of the screening models which has advantage to assess the PCB constituent dissolved in both NAPL and water phase. The predicted concentrations of PCB in subsurface soil as well as in groundwater were obtained by using HSSM model.



The oil lens profiles as a function of time were also generated that can assist in remediation technologies by considering where the contaminants reside in the subsurface soil. CHEMCAN model was also studied to evaluate the partitioning behaviour of oil laced with PCBs in subsurface soil environment. A comparison showed that the CHEM-CAN model responded to analysis similar to HSSM model. However, both the models have their own limitations when applied for NAPL.

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