SIMULATION OF LNAPL MIGRATION AND REMEDITION AT A PETROLEUM REFINERY SITE

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Dedicated to my beloved family

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ABSTRACT

This study presents the results of simulation for the migration of light non-aqueous phase liquid (LNAPL) contamination at Port Dickson petroleum refinery and a remediation by steam injection using T2VOC numerical model. A kerosene leak of 20 years period from the distributing pipelines has caused a serious LNAPL contamination in the subsurface environments. Site investigations were conducted to identify the source of leak and to delineate the contaminations plumes. The concern raised was whether the LNAPL has travelled offsite and has reached the subsurface environments of the down-gradient residential areas. The support tools considered in this study for delineation and remedial solution are based on theexisting site investigations and the application of the numerical model. Numerical simulation will investigate the distribution of LNAPL and remediation within economical and shorter timeline, in response to management decision. In this study, T2VOC is used to simulate a ‘three-phase non-isothermal single component flow’ in a partially saturated homogeneous media for the injection of 876.5 kg of o-xylene (component of kerosene) in a two-dimensionalmodel. The model generated a distribution of LNAPLat a distance of 10 m for 5 years of injection period, while site investigation showed a migration plume of 100 m for 20 years of leak period. Verification with the formulation computed a plume length of 19.9 m. The numerical simulation results are underestimated compared to the site investigation data, although the distribution showed the same behavior for both of methods with time.Based on the results of the numerical simulation and validation with site investigation data, the LNAPL distribution in the subsurface of the refinery was unlikely to migrate offsite to the residential area. While, the simulation of remediation with steam injection showed that 90% of the LNAPL saturation was removed in 20 days of treatment. The result has therefore demonstrated the effectiveness of steam injection process for this study.

ABSTRAK

Kajian ini membentangkan hasil simulasi pencemaran minyak atau cecair bukan akues ringan (*LNAPL*) di kilang penapisan petroleum Port Dickson dan kaedah pemulihan pencemaran melalui suntikan stim menggunakan model T2VOC. Kebocoran saluran paip penghantaran minyak tanah (*kerosene*) yang berlaku selama 20 tahun telah menyebabkan pencemaran yang serius kepada tanah dan air tanah. Siasatan persekitarantelah dijalankan untuk mengenalpasti punca kebocoran dan menyiasat pergerakan pencemaran air tanah. Persoalan adalah sama ada *LNAPL*dalam air tanah telah mengalirdi bawah kawasan perumahan bersebelahan kilang. Dalam kajian ini, model T2VOC beserta kerja siasatan persekitaran digunakan sebagai alat sokongan untuk memilih kaedah pemulihan yang sesuai bagi kawasan yang tercemar. Kajian simulasi menyiasat pengaliran *LNAPL* dan pemulihan dalam tempoh lebih pendek dan jimat berbanding kerja siasatan lanjutan. Model simulasi dua-dimensi menggunakan T2VOC telahmenghasilkan pergerakan minyak dalam sistem ‘satu komponen tiga-fasa tanpa sesuhu’bagi permukaan tanah sejenis bagi suntikan 876.5 kg o-xylene (komponen minyak tanah). Pergerakan *LNAPL* telah dilihat pada jarak 10 m selama 5 tahun tempoh suntikan, manakala siasatan persekitaran sedia ada menunjukkan pergerakan *LNAPL* pada jarak 100 m selama 20 tahun tempoh kebocoran. Pengiraan formula menunjukkan *LNAPL* mengalir pada jarak 19.9 m. Keputusan simulasi adalah kurang berbanding dengan data penyiasatan sedia ada. Walau bagimanapun, pengerakan *LNAPL*adalah dalam tingkah yang sama untuk kedua-dua kaedah. Berdasarkan keputusan simulasi dansiasatan persekitaran, *LNAPL* di dalam tanah dan air tanahtidak mengalir keluar dari kilang ke kawasan perumahan. Simulasi pemulihan dengan suntikan stim menunjukkan 90% daripada*LNAPL* telah dirawat dalam tempoh 20 hari. Ini telah menunjukkan keberkesanan proses suntikan stim dalam kajian ini.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

BTEX	–	Benzene, Toluene, Ethylbenzene, Xylenes
DNAPL	–	Dense Non-Aqueous Phase Liquid
LNAPL	–	Light Non-Aqueous Phase Liquid
NAPL	–	Non-Aqueous Phase Liquid
PAH	–	Polycyclic Aromatic Hydrocarbon
PCE	–	Perchloroethylene
ppm	–	part per million
SVE	–	Soil Vapor Extraction
TCE	–	Trichloroethylene
TPH	–	Total Petroleum Hydrocarbon
USEPA	–	United States Environmental Protection Agency
VOC	–	Volatile Organic Compound

LIST OF SYMBOLS

%	–	Percentage
ρ_b	–	Bulk density
ϕ	–	Porosity
μ	–	Viscosity
β	–	Phase (air, water, chemical)
κ	–	Component (air, water, chemical)
ρ	–	Phase density
F	–	Mass flux of component
f_{oc}	–	Fraction of organic carbon
g	–	Gavitational acceleration vector
K	–	Hydraulic conductivity
k_d	–	Soil distribution coefficient
K_{oc}	–	Organic Carbon/Water Partition Coefficient.
kg	–	Kilogram
L	–	Liter
M	–	Mass of component
m	–	Meter
P	–	Pressure
R	–	Retardation factor
S	–	Saturation (pore volume fraction)

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 General

Subsurface contamination by hydrocarbon fuels is common throughout the industry. According to Naidu (2013), there are more than 3,000,000 potentially contaminated sites whichbesides posing risks to the health and well-being of humans and the environment, also represent a large lost economic opportunity. Contamination is the results of industrialization, inadequate environmental laws and inconsistent and lacking enforcement.. Although site contamination has been recognised since the 1960s, less than a tenth of potentially contaminated sites globally have been remediated due to the complex and challenging nature of both surface and subsurface contamination. These challenges are further aggravated by the cost and technical difficulty of dealing with contaminant mixtures, complex subsurface environments as well as recalcitrant and persistent pollutants. Common contaminants include petroleum hydrocarbons, chlorinatedsolvents, persistent organic pollutants, pesticides, inorganics,heavy metals and radioactive constituents. Thesecontaminants can be found in a variety of sites such asoil and gas operations, service stations, mines, industrialcomplexes, landfills, waterways, harbours and even inrunoff from urban and residential settings (Naidu, 2013).The constituent contaminants of concern from the petroleum release have been the relatively soluble aromatic hydrocarbons such as benzene, toluene, ethylbenzene, and xylenes, known collectively as BTEX which are also volatile organic compound (VOC).

Many of the pollutants are essentially immiscible with water and thus exist as non-aqueous phase liquids (NAPLs). The two types of NAPLs are light non-aqueous phase liquid (LNAPL) and dense non-aqueous phase liquid (DNAPL). LNAPL has liquid density less than water, called the 'floater', while DNAPL has liquid density more than water, or called the 'sinker'. This study focuses on the LNAPL contaminations in the subsurface environment resulted from the petroleum refinery operation. The LNAPL is hydrocarbon fluid such as gasoline, kerosene, diesel fuel or jet fuel. In this paper, the term "LNAPL" is used interchangeably with "free product" and is of kerosene origin from the leak event that is going to be discussed.Petroleum kerosene, a product of crude oil refining, used in jet fuel, solvent or home heating, contains a mixture of aliphatic, aromatic and a variety of other branched saturated and unsaturated hydrocarbons. Kerosene are the lighter end of a group of petroleum substances known as middle distillates, the heavier end being gas oils. Kerosene product contains aromatic hydrocarbon portion such as o-xylene (part of BTEX group) in a range of 0.1% to 3% by weight (Dunlap et al., 1988) depending on the manufacturers. Multiple acute toxicity studies have been reported on a variety of kerosene streams and fuels demonstrating low acute toxicity but with the potential to cause skin irritation (API, 2010). The BTEX components represent a threat to human health and ecosystems because of their toxicity (ATSDR, 2005).

When LNAPL leaks in the soil, it migrates downward through the porous media under the influence of gravity until it reaches the water table, where it forms a lense and spreads laterally. As the LNAPL spreads, some portions are left behind as residuals and other volatile compounds may move into vapor phase and spread in the vadose zone. Soluble constituents from the LNAPL lens may dissolve into the groundwater causing groundwater contamination. Continuous fluctuation of the water table will result in a smear zone of LNAPL directly above the floating oil. The LNAPL contamination causes concern because of their persistence in the subsurface and their ability to contaminate large volumes of soil and groundwater. LNAPL properties such as density, viscosity, interfacial tensions, solubility and vapour pressure are important in understanding LNAPL transport and in predicting subsurface contamination (Jeongkon, 2002). More than 99% of spilled fuel in the contaminated environments remains as trapped immobile or mobile free product in both the vadose and saturated zones of the subsurface (Weiner, 2000). Figure 1.1 shows the schematic illustration of LNAPL migration and contamination in the subsurface environments.

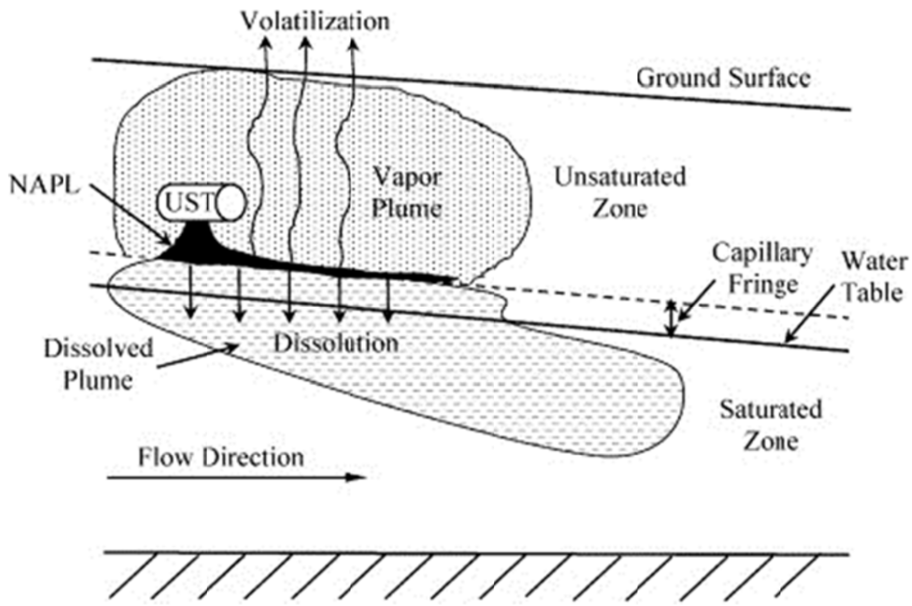

Figure 1.1: LNAPL Migration and Contamination in the Subsurface Environment (Jeongkon, 2002).

Estimating the amount of free product remaining and the extend of migration in the subsurface media are crucial to an effective remedial strategy. The goals of remediation plan include identifying the most practicable, cost-effective solutions, creating a decision-making framework for achieving cleanup goals, developing a procedure for cleaning up, and developing a better understanding of aggressive contaminants removal technologies (USEPA, 1988). When the needs of remediation work arise for a site, it is important to carefully select the best support tools that will be used to fully characterize the sites so it is aligned with the remediation goals. Good data and good judgment lead to good site decisions. The support tools that will be employed for this study are based on the existing site investigation work and the application of numerical model, T2VOC. Numerical simulation study will further investigate the distribution of LNAPL and selection of a remediation method within economical and shorter timeline. Numerical model can provide prediction of the fate and transportand the system behavior under natural conditions or in response to management decision. Numerical model may be used as a conceptual tool or, under specific assumptions and associated with site investigation data for further validation and verification.

1.2 Statement of the Problem

A petroleum refinery in Port Dickson had experienced a continuous kerosene product leak from its distributing pipelines for a period of 20 years (1973 – 1993). Multiple environmental consultants have been engaged to by the refinery operator to investigation the LNAPL contaminations in the subsurface environment of the refinery since 1992. Figure 1.2 shows the kerosene leak area at the refinery which is located in Zone 3. The leak point is located approximately 200 m from the western boundary of the refinery. Residential areas are located directly on the western boundary of leak area (Zone 3).

In response to this leak event, the operator of the refinery has appointed multiple environmental consultants to conduct site investigation studies since 1992. The objectives of the studies were to identify the source of leak and to delineate the LNAPL and dissolved plumes detected in the soil and groundwater environments. Intrusive site investigations conducted at the refinery comprised the advancement of soil bores, excavation of soil pits and trenches, installations of 51 onsite groundwater monitoring wells, ten offsite groundwater monitoring wells, and soil vapor surveys to study the hydrogeological conditions and to collect the subsurface samples for laboratory analysis.

Figure 1.2: Layout of the Refinery and Kerosene Leak Area (Source: Google Map)

Based on the environmental report in 2001, the consultants indicate that the volume of LNAPL sitting in the soil and groundwater was approximately 37,400 liters (L). The LNAPL was gauged in ten onsite monitoring wells, while no LNAPL was detected in the offsite monitoring wells. The site investigation reports indicated that the LNAPL was observed to have traveled approximately 100 m from the leak area. From the laboratory analytical data and the risk-based analysis, the investigations concluded that the serious LNAPL contaminations on the subsurface environments at the refinery can becategorized as unsafe for human exposure or consumption or other usages such as irrigations or industrial. The findings of the site investigation in 2001had raised a further health, safety and environment (HSE) concern for the immediate down-gradient receptors on the western boundary of Zone 3 such as residential and industrial workers whether there's a likelihood of migrationof LNAPL to the offsite.

Continuous intrusive subsurface investigations for typically large sites such petroleum refineries could lead to higher cost expenditure and is more time consumingdue to the complexity of the site. Expenditures are contributed by the extensive used of mechanical machines such us excavators, drilling machines, monitoring equipment, relocation of existing structures, cost for laboratory analysis and equipment as well as the professional fees for geologist, scientist and environmental engineers involved in the projects. According to USEPA (2004), the investigation and remediation at many manufactured gas properties or refineries are complicated by:

i. Nature and variety of contaminant constituents and media, including the fact that the constituents of concern tend to interact differently in different media. The factors influencing contaminant plume persistence for a given site is uncertainty in site properties (e.g., geologic, hydrologic) and conditions (e.g., nature and distribution of contamination).
ii. Location of the sites (i.e near waterways or in the heart of residential neighborhood).
iii. Condition of the sites (i.e. infrastructure at the sites, lack of surface features and mixeddebris is subsurface).

These are the same case for the Port Dickson refinery where it has a complexity of the refinery structures and the location of the refinery near the residential area. If the soil and groundwater contaminations are proven to pose risk to the human receptors at the refinery or at the boundary, remedial action should be planned accordingly. The remediation technology has to be selected carefully to address these complications and also at the reasonable cost. It is reported in the USEPA report (2004), kerosene refiners with an area of one to two acres cost around one to five million dollars to remediate. The remediation technology used were vapor extraction, thermal treatment, bioremediation (in-situ and ex-situ), air sparging, treatment wall, pump-and-treat, air stripping, soil flushing and surfactant flushing among others. For understand the scenario of the remedial plan and to close data gaps for the site investigation study, this numerical study is proposed as an additional support tool on top of the existing site investigations, to further investigate the LNAPL migration at the refinery and to measure the success of the remediation process of choice which is steam injection, as well as to validate the previous site investigation data. The numerical model could be used to provide prediction of the system behavior under natural conditions. However, the simulation model should not be seen as a prediction of the duration of the remediation in Port Dickson refinery, but rather as a qualitative study to compare different remediation strategies in the future. If successful, the model can be used in planning a full-scale treatment system for the leak area in the refinery.

1.3 Aim and Objectives of the Study

The aim of this study is to investigate the distribution behavior of LNAPL in the porous media, as well as identifying the remedial success through steam injection.The objectives of this study include:

i. To simulate the migration of LNAPL leak in the porous media using T2VOC model.
ii. To simulate the remediation of LNAPL by steam injectionusing T2VOC model.
iii. To validate and verify the length of LNAPL plume migration simulated by T2VOC simulation model, and estimated from site investigation data and formulation (Charbeneau, 2000).

1.4 Scope of the Study

There are three scopes of this study which include the following:

i. Collection of Site Investigation Data

Following the leak event at the Port Dickson refinery, a series of site investigation works have been conducted at the refinery for the objectives stated. The data collected from the site investigations include:

a. Hydrogeology data of the site and its vicinity such as geological formation, groundwater hydrology and aquifer properties.
b. Soil and groundwater contaminants data such as as constituent contaminants properties.

ii. Simulation of Migration and Remediation of LNAPL using T2VOC Model

T2VOC is a numerical simulator for three-phase, three-component, non-isothermal flow of water, air, and a volatile organic compound (VOC) in multidimensional heterogeneous porous media (Falta et al., 1995). The program was developed by KarstenPruess, a Senior Scientist in the Earth Sciences Division of the Lawrence Berkeley at early1980s. T2VOC is an extension of the TOUGH2 general-purpose simulation program which is defined as a transport of unsaturated groundwater and heat. T2VOC stands for TOUGH2 with volatile organic compound. T2VOC is a special version of TOUGH2 developed for simulation model of soil and groundwater contamination that will include the full heat transfer and thermodynamics. T2VOC has the ability to simulate migration of total mass, as well as constituent migration and reactions, in all combinations of the three phases. T2VOC also has the ability to simulate the remediation process such as vapor extraction, injection air for volatile removal, direct pumping and steam injection. As with TOUGH2, T2VOC has been used by many researchers to investigate the migration and remediation of LNAPL.Examples of LNAPL studies that have used T2VOC numerical model include Fagerlund et al. (2003, 2005, 2006 and 2007), MDH (2005), Mortensen et al., (2000) and (Kling et al., 2004)(refer to Chapter 2 - Literature Review).

This study presents the application and simulation of T2VOC to subsurface contamination problems involving LNAPL originated from a kerosene leak and subsequently a simulation of remediation using steam injection. There are four steps involve in the simulation that will be discussed in the methodology section (refer to Chapter 3). The technical descriptions of the T2VOC code are extracted from the User Guide of T2VOC(Falta et al., 1995) including a discussion of the physical processes modeledand the governing equations. Detailed instructions for preparing input data are presented in Chapter 3 and attached in the appendices along with several illustrative results in Chapter 4.

iii. Validation and Verification of Simulation Results

The results of the study will be validated with the site investigation data while verification of the plume length will be based on the plume length formula by Charbeneau (2000). This will be discussed in Chapter 4.

1.5 Significance of the Study

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The results of this study would serve as the support tool during decision-making process for selecting the decisive remediation plan for the site in the future. The study is significant as:

i. This study employs numerical modeling as the support tool and the best method to visualize LNAPL migration within economical and shorter timeline as opposed to extending the intrusive site investigation studies.
ii. The simulation helps to validate whether the LNAPL plume has migrated offsite the refinery to the residential area.
iii. This study measures the efficiency of the selected remediation technology i.e. steam injection.

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