RESEARCH ARTICLE OPEN ACCESS

Modelling the Transport of Crude Oil in Sandy Soil

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ABSTRACT
The transport and retention of crude oil in sandy soil was studied. A full factorial experimental design with three independent variables, namely soil depth, contaminant volume and rainfall intensity, was used. The soil was artificially contaminated with varying crude oil volumes and rainfall simulation was performed in a mesocosm. The results obtained showed variations in the transport and retention of crude oil with the independent variables. The leached total petroleum hydrocarbons (TPH) concentrations decreased with increasing soil depth but increased with increasing contaminant volume and rainfall intensity. Contrarily, the retained TPH concentrations increased with increasing soil depth and contaminant volume but decreased with increasing rainfall intensity. Transport and retention models were developed and used to fit the transport and retention of crude oil in sandy soil after accidental release with correlation coefficient of 0.9407 and 0.9446 for the transport and retention model, respectively.
Keywords: Modelling, Transport, Retention, Crude oil, Contaminant, Hydrocarbons, Soil, Unsaturated zone, Response surface method, Contamination risk

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Date Of Submission: 25-07-2019 Date Of Acceptance: 06-08-2019

I. INTRODUCTION

Crude oil has become one of the most important commodities and an exceptionally attractive product in the world market today (Aroh et al., 2010). The Niger Delta regionis rich in petroleum resources which play an important role in the nation economy by generating over 90% of the nation’s revenue (Odjuvwuederhie et al., 2006). Accordingly, the exploration and production of oil have concentrated in the Niger Delta region, with over 1000 oil wells and over 47,000 km of oil flow lines (Ngobiri et al., 2007). The development of the oil industry has become a threat to groundwater quality because of the pollution caused by petroleum hydrocarbons from petroleum spills experienced during the use, transportation and storage (Liu et al., 2007).

Crude oil is a naturally occurring compound, composed of a mixture of hydrocarbon and non-hydrocarbon compounds (Speight, 2002). Hydrocarbons are molecules that contain only hydrogen and carbon, they exist as a diverse mixture of linear, branched, cyclic and aromatic compounds. Non-hydrocarbons are fractions known as resins and asphaltenes containing elements such as nitrogen, sulphur and oxygen (Mullins, 2007). Crude oil varies in their properties following the different geographical location they are found, these properties includes density, specific gravity, American Petroleum Institute (API) gravity, viscosity, pour point, carbon residue, salt content and sulphur content.

Petroleum hydrocarbons have become of exceptional interest considering the effects on soil and groundwater after contamination from spills or accidental leaks associated with exploration, production and distribution of crude oil. These releases may result in contaminated sites that pose an unacceptable risk to human health, ecology, water resources and rendering the area unproductive. The negative effects of petroleum hydrocarbons to all aspect of life have prompted quite several researches on the environmental fate and transport of hydrocarbons (Verbruggen, 2004).

Most cases of oil spill can be traced to oil tankers involved in accidents, vandalism of pipelines used for distribution and oil storage facilities. Such accidents can be as a result of human mistakes, deliberate actions or natural disaster such as earthquakes (Abii and Nwosu, 2009). There has been a massive increase of oil spill through acts of sabotage or pipeline vandalism by unemployed youths in Nigeria in the past decades. The poor implementation of memorandum of understanding (MoU) between the oil companies and host communities, leaving the people unemployed has been blamed for this drift.

Uchegbu (1998) observed that oil pollution is a challenge experienced in the world that has no conclusive solution despite efforts to safeguard or counter measure it and the separation of oil spill incidents from oil exploration and exploitation is extremely difficult despite all control measures put in place. Atlas and Bartha (1987) reported that crude oil pollution is gradually creating an imbalance in the biotic and abiotic regimes. Okafor (1985) recounted that whenever there is incident of oil spill, it usually causes serious economic problem, contaminate drinking water and cause general discomfort of normal life. For minimum reduction or elimination of these effects, it is important that clean-up of these contaminants from the affected area be carried out by applying the appropriate remediation techniques (Ellis et al., 1990). Selection of the appropriate remediation techniques depends on understanding the transport and retention of target contaminants in the affected soil.

The transport of crude oil in the environment is essential for estimation and determination of the risk human and ecological receptors can be posed to (McMillen et al., 2000). The rate and extent of transport of crude oil will be based on the properties of soil andcrude oil as well as the volume of the spilled crude oil(API, 1972; Brost and DeVaull, 2000; Keller and Simmons, 2005).In the event of spill, crude oil can exist in the soil surface, dissolve in the groundwater or even occupy pore spaces in the unsaturated zone. Understanding the process of transport in soil-water environment gives better insight to predict contamination risk extent. Therefore, this work aims to develop models using response surface method to describe the transport and retention of petroleum hydrocarbons in the unsaturated zone following the contamination of sandy soil with crude oil.

II. MATERIALS AND METHODS

2.1. Crude oil

The crude oil used in this study was obtained from Aminamkpono Offshore in Rivers State. The crude oil has the following properties: density (0.8 kg/m^3), specific gravity (0.8014), API gravity (45.07), flash point (40-83^{o}C) reidvapour pressure (5 psi) and sulphur content (0.01-0.1%). The crude oil sample was analyzed in the laboratory using standard methods to obtain the baseline total petroleum hydrocarbons (TPH).

2.2. Soil characterisation

The soil used in this study was sandy soil obtained from the Centre for Occupational Health, Safety and Environment. The soil sample (1000 kg) was air dried and sieved using a 2 mm sieve to obtain a uniform particle size and classification. The soil has the following properties: particle size distribution (0.1 - 0.2 mm), particle density (2.286 g/cm^3), bulk density (1.861 g/cm^3) and porosity (0.1859). The soil was analysed for TPH using standard procedure.

2.3. Response surface method (RSM)

XLSAT software using RSM method of a full factorial design with 3 levels was used to generate the number of observations/ experimental runs required to develop transport and retention models. Theindependent variables were soil height ranging from 10 cm to 50 cm, contaminant concentration ranging from 1 mg/l to 5 mg/l and rainfall intensity ranging from 5 mm/hr to 10 mm/hr. The dependent variables were leached TPH and retained TPH. After the input of the independent variables, 27 observations /experimental runs were generated to conduct thestudy (Table 1).

Table 1: Experimental design

Observations	Soil height (cm)	Contaminant conc. (mg/l)	Rainfall intensity (mm/hr)
Obs1	10	1	5
Obs2	30	1	5
Obs3	50	1	5
Obs4	10	3	5
Obs5	30	3	5
Obs6	50	3	5
Obs7	10	5	5
Obs8	30	5	5
Obs9	50	5	5
Obs10	10	1	7.5
Obs11	30	1	7.5

Obs12	50	1	7.5
Obs13	10	3	7.5
Obs14	30	3	7.5
Obs15	50	3	7.5
Obs16	10	5	7.5
Obs17	30	5	7.5
Obs18	50	5	7.5
Obs19	10	1	10
Obs20	30	1	10
Obs21	50	1	10
Obs22	10	3	10
Obs23	30	3	10
Obs24	50	3	10
Obs25	10	5	10
Obs26	30	5	10
Obs27	50	5	10

2.4. Experimental setup

A simple laboratory experiment was performed to investigate the transport and retention of crude oil compounds in sandy soil. The experimental system used was designed and reported by Ugwoha et al. (2016) but with few modifications in the areas of material and soil height. A 4-inch (100 mm) polyvinyl chloride (PVC) pipe of height 70 cm was used. The soil was placed and well compacted in the PVC pipe to the heights of 10, 30 and 50 cm, representing different depths of unsaturated zone in Port Harcourt. The remaining void in the PVC pipe prevented the overflow of accumulated simulated rainfall water. The weights of the 10, 30 and 50 cm heights of the soil were 1524, 4572 and 7620g, respectively.

After the placing of the soil into the PVC pipe, water was released on the soil via the rainfall simulator until the soil became saturated. The 10, 30 and 50 cmsoil heights were saturated with 0.5, 1.2 and2.0 litres, respectively. The wetted soils were allowed for 2 days to attain water content equivalent to field capacity hence imitating soil in a natural environment.Thereafter, the soil was contaminated with varying volumes of crude oil and subjected to varying volumes of rainfall simulations. The number of experimental runs and the corresponding crude oil concentration, rainfall intensity and soil height were as shown in Table 1 in Section 2.3

The rainfall intensities (5 mm/hr to 10 mm/hr) used in this study were deduce from the average monthly rainfall of 203.03 mm/hr reported for Port Harcourt (Uko and Tamunobereton-Ari, 2013).

2.5. Sample collection

The soil and water used in the experiment was first analysed before contamination for background TPH. After the contamination of the soil and the rainfall simulation, the effluent and soil samples were collected and analysed for leached and retained TPH concentration, respectively. The soil was thoroughly mixed together before a portion was collected.

2.6. Sample analysis

TPH of the water sample was analysedusinggas chromatography with flame ionization detector (GC-FID). Exactly 250 ml of the effluent sample was measured into a separating funnel rinsed with dichloromethane. Exactly 25 ml of dichloromethane was added to the effluent sample and shook vigorously to extract all the available organic material. The organic extract was passed through a column containing cotton wool, silica-gel and anhydrous sodium sulphate to remove debris and moisture from the extract. Exactly 1 µl of the concentrated organic extract was injected into the GC-FIDby means of a hypodermic syringe through a rubber septum into the column. The various fractions of the hydrocarbons were automatically detected as they emerged from the column by the detector.From the detected hydrocarbons, the TPH expressed in mg/l was calculated.

The TPH of the soil samplewas also analysed using GC-FID after extraction and clean-up/separation.

Extraction of organic material

Exactly 2 g of soil sample was weighed into a clean extraction container. Exactly 10 ml of extraction solvent (pentane) was added into the container and mixed thoroughly and allowed to settle. The mixture was carefully filtered into a clean solvent rinsed extraction bottle using filter paper fitted into a

Buchner funnel. The extract was concentrated to 2 ml and then transferred for clean-up/separation.

Clean-up/Separation

Exactly 1 cm of moderately packed glass wool was placed at the bottom of a 10 mm ID x 250 mm long chromatographic column. Slurry of 2 g activated silica in 10 ml methylene chloride was prepared and placed into the chromatographic column. To the top of the column was added 0.5 cm of sodium sulphate. The column was rinsed with additional 10 ml of methylene chloride. The column was pre-eluted with 20 ml of pentane and allowed to flow through the column until the liquid in the column was just above the sulphate layer. Immediately 1 ml of the extracted sample was transferred into the column. The extraction bottle was rinsed with 1 ml of pentane and added to the column as well. The stop-clock of the column was opened, and the eluent was collected with a 10 ml graduated cylinder. Just prior to exposure of the sodium sulphate layer to air, pentane was added to the column in 2 ml increment. Exactly 10 ml of the eluent was collected and was labelled aliphatic.

GC Analysis

The concentrated aliphatic fraction was transferred into a labelled glass vial with Teflon rubber crimp caps for GC analysis. Exactly 1µlof the concentrated sample was injected into the GC. The TPH was calculated as described earlier.

2.7. Development of transport and retention models

The transport and retention models were developed from the input variables. With the use of RSM, a predicted concept of the transport and retention models were proposed as:

$$LC = a^0 + a^1SH + a^2C + a^3RI + a^4SH^2 + a^5C^2 + a^6RI^2 + a^7SH*C + a^8SH*RI + a^9C*RI \quad (1)$$

$$RC = a^0 + a^1SH + a^2C + a^3RI + a^4SH^2 + a^5C^2 + a^6RI^2 + a^7SH*C + a^8SH*RI + a^9C*RI \quad (2)$$

where LC is the leached concentration (mg/l), RC is the retained concentration (mg/kg), $a_1, a_2, a_3, a_4, a_5, a_6, a_7, a_8, a_9$ are coefficients, SH is the soil height (cm), C is the contaminant volume (ml) and RI is the rainfall intensity (mm/hr).

III. RESULTS AND DISCUSSION

3.1. Baseline concentrations of total petroleum hydrocarbons (TPH)

The baseline concentrations of TPH for the crude oil, water and soil samples used are shown in Table 4. While the TPH was as high as 23094.6 mg/l for the crude oil, no trace of it was found inthe water and soil samples.

Table 4: Total petroleum hydrocarbons in crude oil, water and soil samples.

Sample	TPH
Crude oil (mg/l)	23094.6
Water (mg/l)	0.0
Soil (mg/kg)	0.0

3.2. LeachedTPH concentrations

The leachedTPH concentrations through the soil depth at varying rainfall intensities and contaminant volumes is presented in Figure 1. Generally, the leached TPH concentrations decreased with increasing soil depth but increased with increasing contaminant volume and rainfall intensity.

The leached TPH concentrations ranged from 0.99 mg/l (for soil depth of 50 cm with contaminant volume of 1 mg/l at rainfall intensity of 5 mm/hr) to 7.39 mg/l (for soil depth of 10 cm with contaminant volume of 5 mg/l at rainfall intensity of 10 mm/hr). This implies that more groundwater contamination will occur with a thin unsaturated zone, higher volume of contaminant spill and higher rainfall intensity than with thick unsaturated zone, lower volume of contaminant spill and lower rainfall intensity.

3.2.1. Relationship between leached TPH concentrations and independent variables

The relationship between the leached TPH concentrations and the three independent variables are presented in Figures 2 - 4. Generally, the leached TPH concentration is inversely related to the soil depth but directly related to the contaminant volume and rainfall intensity.

Generally, the lowest leached TPH concentrations were obtained with the highest soil depth and lowest contaminant volume and rainfall intensity.

3.2.2. Leached TPH concentration model

The leached TPH concentration model generated from the model concept proposed in Equation (1) is presented in Equation (3). From the leached TPH concentration model it can be observed that rainfall intensity had the highest coefficient compared to the soil depth and contaminant volume. More so, the correlation matrix between soil depth, contaminant volume, rainfall intensity and the leached TPH concentration shows that rainfall intensity has a strong correlation (0.7369) with the leached TPH concentration while contaminant volume and soil depth have weak (0.2750) and inverse (-0.3840) correlation respectively with the leached TPH concentration. This implies that a change in rainfall intensity will have a higher impact on groundwater contamination than a change in contaminant volume or soil depth.

$$LC = 1.84148 + 0.69111 * SH + 0.49500 * C + 1.32611 * RI + 0.17222 * SH^2 + 0.08389 * C^2 + 0.81056 * RI^2 + 0.26167 * SH * C + 0.53667 * SH * RI + 0.37917 * C * RI \quad (3)$$

3.2.3. Leached TPH model validation

The relationship between the measured and predicted leached TPH concentration is shown in Figure 5. It can be observed that there is a good relationship between the measured and predicted leached TPH concentration. Also, the leached TPH concentrations clustered around the trendline giving a line of best fit and a very high correlation coefficient (R^2) of 0.9407 and predictive relevance (Q^2) of 0.8273.

The validation of the model using the relationship between the measured and predicted leached TPH concentration is presented in Figure 6. It can be observed that there is a very good agreement between the measured and predicted TPH concentration and this can be corroborated with high R^2 and Q^2 values, implying that the developed model can adequately predict the transport of crude oil through a sandy unsaturated zone to the groundwater after release.

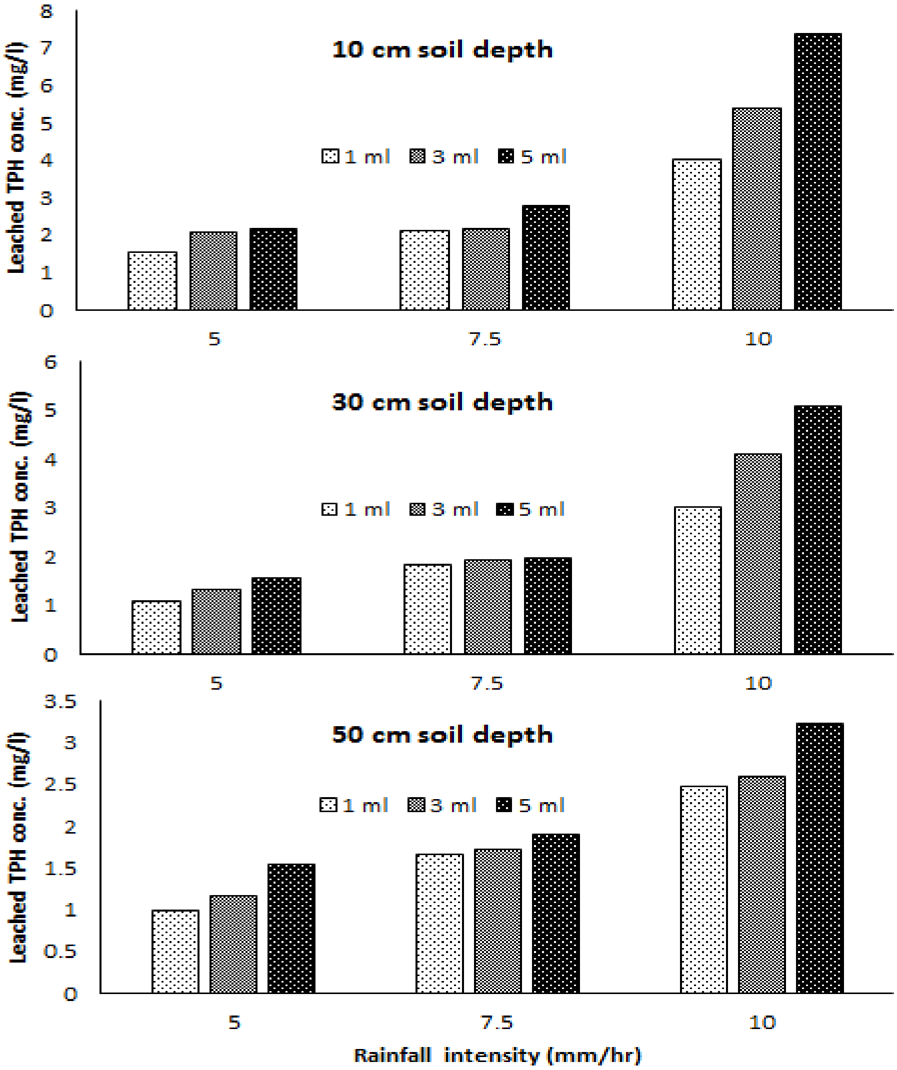

Figure 1: Leached TPH concentrations for various soil depths and rainfall intensities

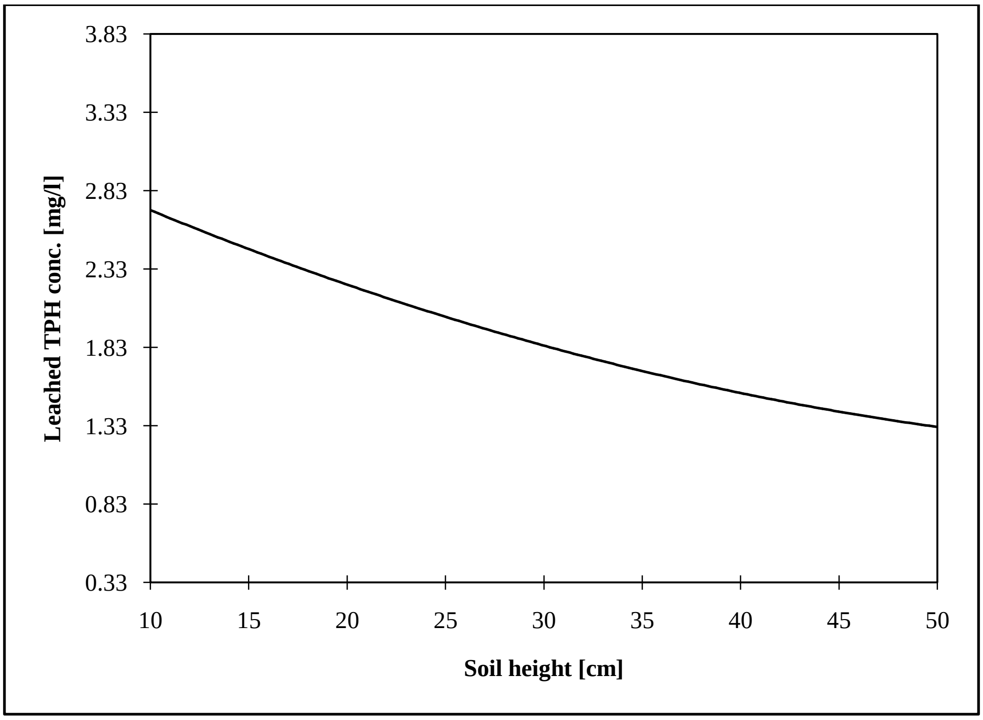

Figure 2: Leached TPH concentrations through varying soil depths.

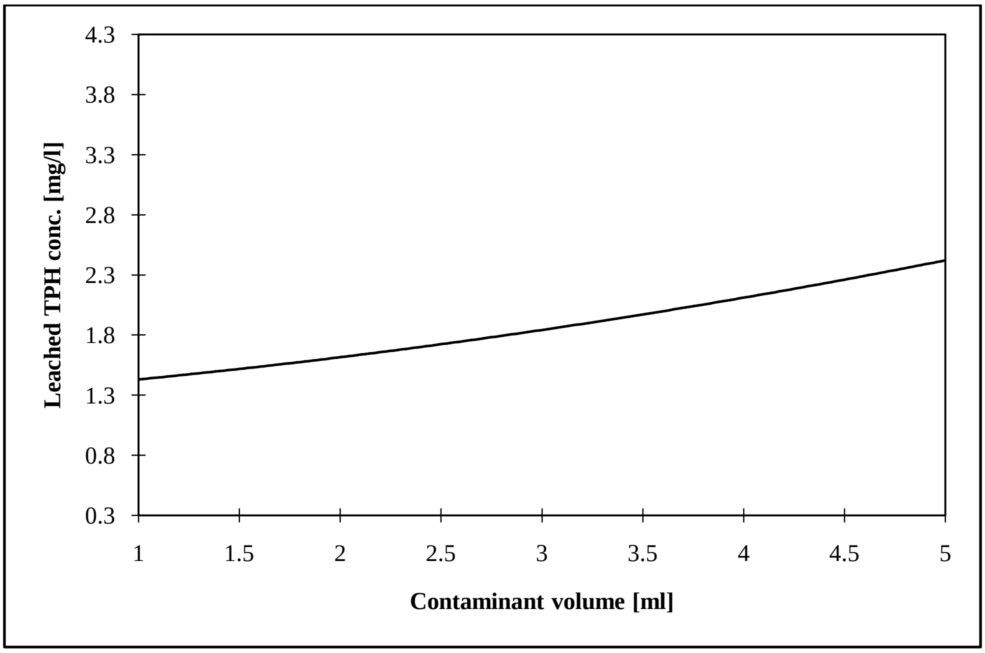

Figure 3:Leached TPH concentrations through varying contaminant concentrations.

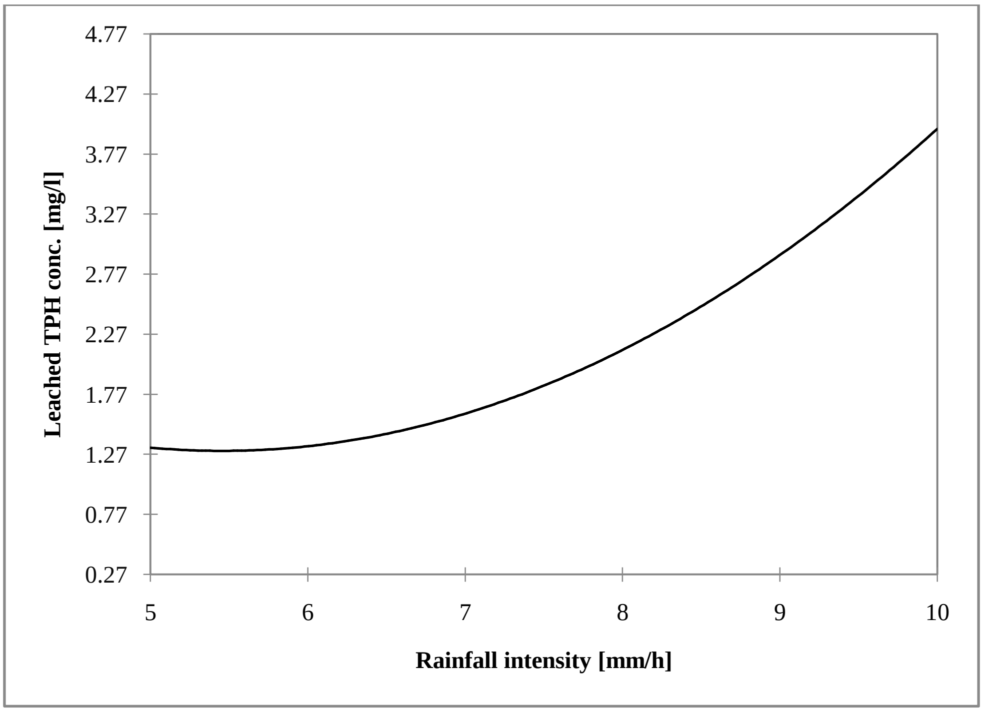

Figure 4:Leached TPH concentration through varying rainfall intensities.

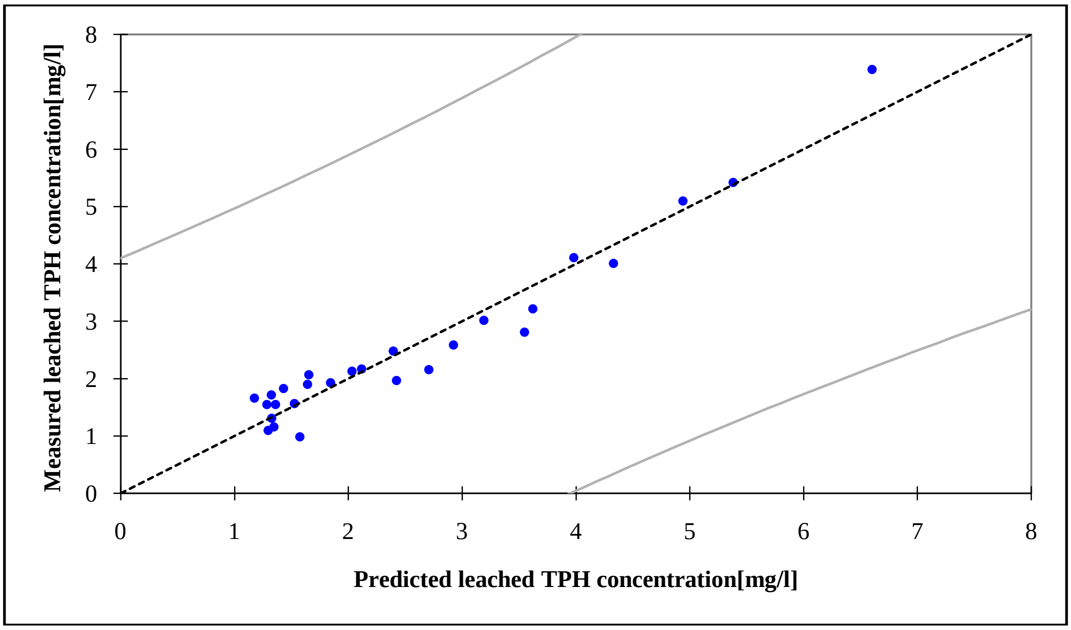

Figure 5: Relationship between actual and predicted leached TPH concentrations

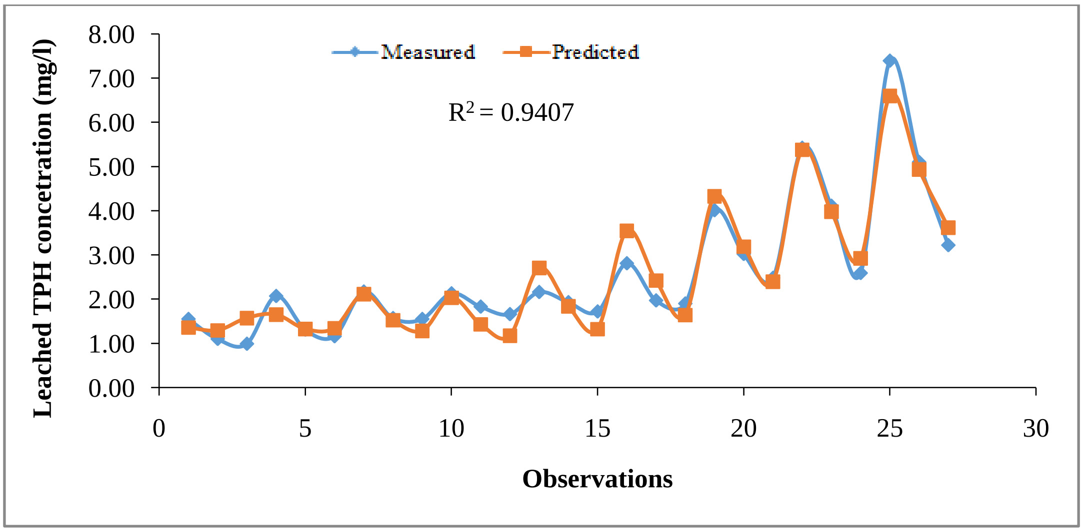

Figure 6: Comparison of measured and predicted leached TPH concentrations with different observation points.

3.3. Retained TPH concentrations

The retained TPH concentrations in the unsaturated zone of varying soil depths at varying rainfall intensities and contaminant volumes is presented in Figure 7. Generally, the retained TPH concentrations increased with increasing soil depth and contaminant volumebut decreased with increasing rainfall intensity.

The retained TPH concentrations ranged from 16.13 mg/kg (for soil depth of 30 cm with contaminant volume of 1 mg/l at rainfall intensity of 10 mm/hr) to 133.18 mg/kg (for soil depth of 50 cm with contaminant volume of 5 mg/l at rainfall intensity of 5 mm/hr). This implies that less groundwater contamination will occur with a thick unsaturated zone, higher volume of contaminant spill and lower rainfall intensity than with thin unsaturated zone, lower volume of contaminant spill and higher rainfall intensity.

3.3.1. Relationship between retained TPH concentrations and independent variables

The relationship between the retained TPH concentrations and the three independent variables are presented in Figures 8-10. Generally, the retained TPH concentration is directly related to the soil depth and contaminant volume but inversely related to the rainfall intensity.

3.3.2. Retained TPH concentration model

The retained TPH concentration model generated from the model concept proposed in Equation (2) is presented in Equation (4). From the retained TPH concentration model, it can be observed that contaminant volume had the highest coefficient compared to the soil depth and rainfall intensity. More so, the correlation matrix between soil depth, contaminant volume, rainfall intensity and the retained TPH concentration shows that contaminant volume has a higher correlation (0.6122) with the retained TPH concentration while soil depth and rainfall intensity have lower (0.5175) and inverse (-0.3083) correlation respectively with retained TPH concentration. This implies that a change in volume of contaminant will have a higher impact in the amount of contaminant retained than a change in rainfall intensity and soil depth.

$$RC = 31.07519 + 16.86278*SH + 19.94833*C + 10.04667*RI + 10.66278*SH^2 + 1.72278*C^2 + 2.89444*RI^2 + 13.16917*SH*C + 4.80917*SH*RI + 8.41333*C*RI \quad (4)$$

3.3.3. Retained TPH model validation

The relationship between the measured and predicted retained TPH concentration is shown in Figure 11. It can be observed that there is a good relationship between the measured and predicted retained TPH concentration. Also, the retained TPH concentrations clustered around the trendline giving a line of best fit and a very high correlation coefficient (R^2) of 0.9446 and predictive relevance (Q^2) of 0.8273.

The validation of the model using the relationship between the measured and predicted retained TPH concentration is presented in Figure 12. It can be observed that there is a very good agreement between the measured and predicted retained TPH concentration and this can be corroborated with high R^2 and Q^2 values, implying that the developed retention model can adequately predict the retention of crude oil in a sandy unsaturated zone following release.

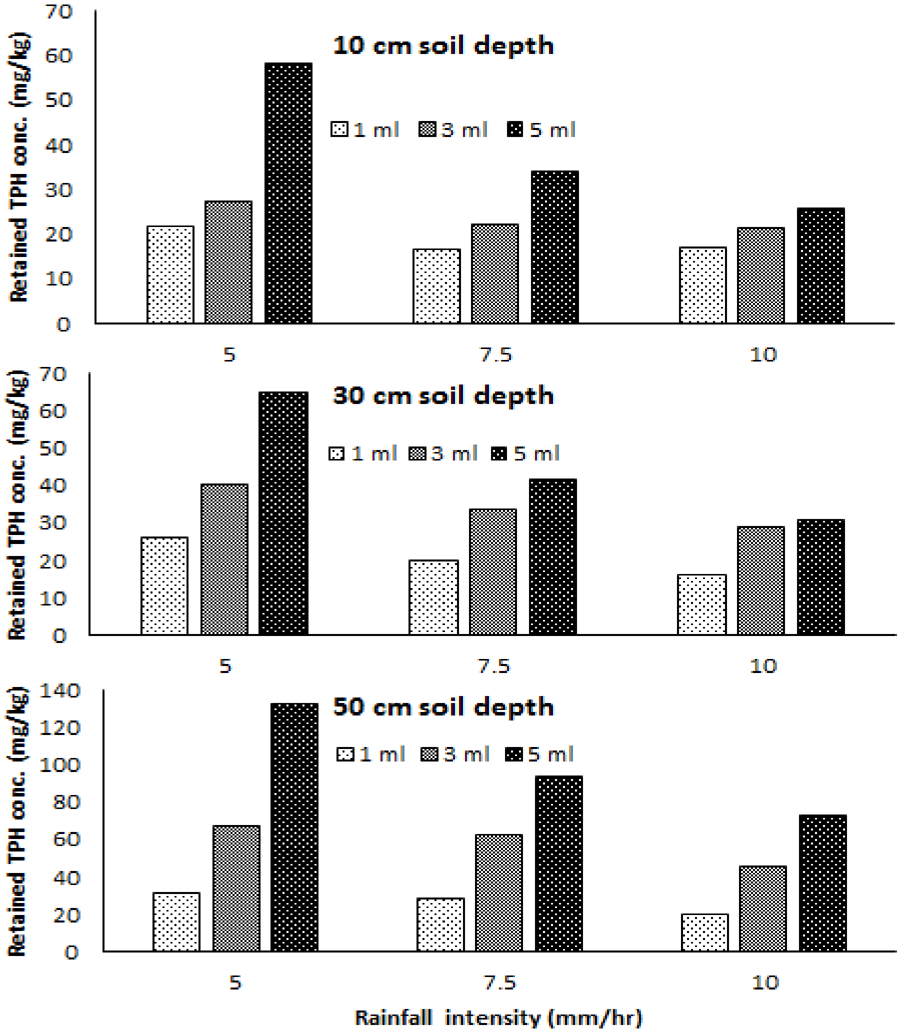

Figure 7: Retained TPH concentrations for various soil depths and rainfall intensities

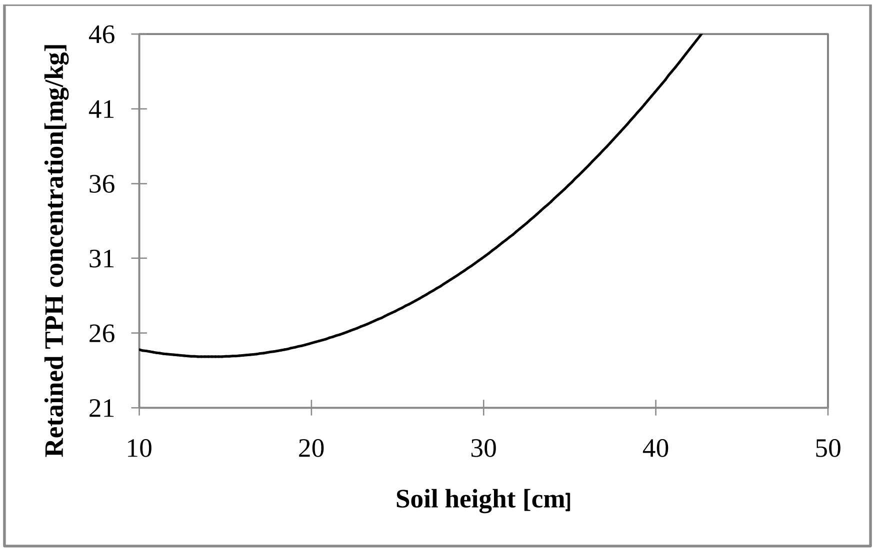

Figure 8: Retained TPH concentrations in varying soil depths.

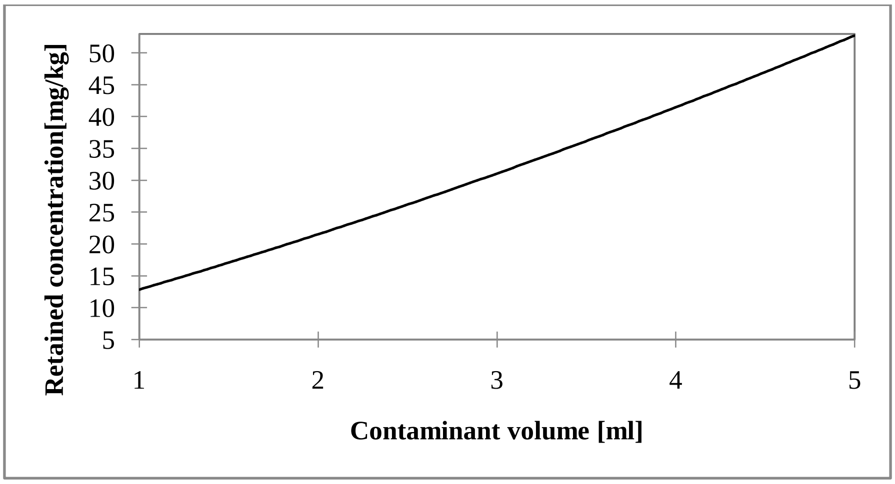

Figure 9: Retained TPH concentrations for varying contaminant volumes.

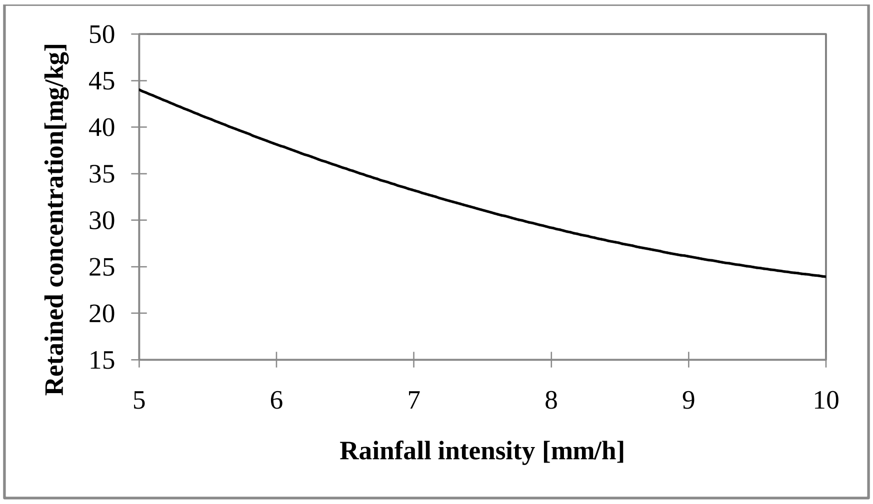

Figure 10: Retained TPH concentration for varying rainfall intensities.

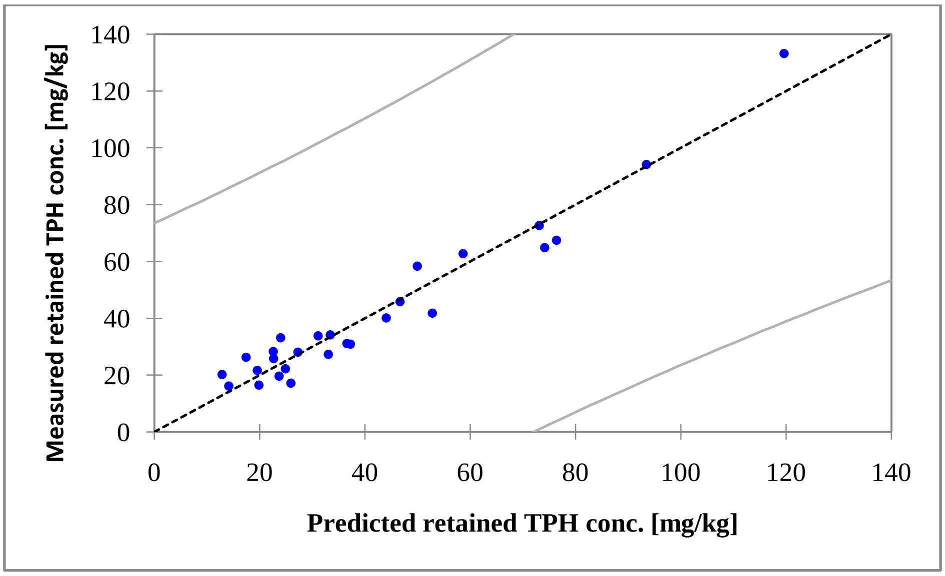

Figure 11: Relationship between measured and predicted retained TPH concentrations

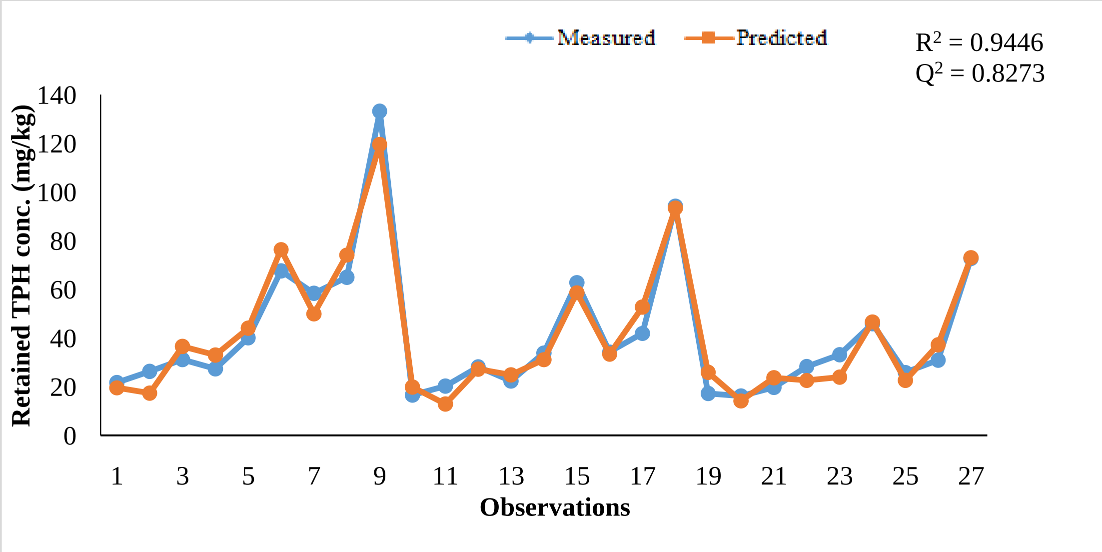

Figure 12: Comparison of measured and predicted retained TPH concentrations with different observation points.

IV. CONCLUSION

The transport and retention of crude oil in sandy soil was studied by varying three independent variables, namely soil depth, contaminant volume and rainfall intensity, in a mesocosm. The results obtained revealed that the leached total petroleum hydrocarbons (TPH) concentrations decreased with increasing soil depth but increased with increasing contaminant volume and rainfall intensity, implying that more groundwater contamination will occur with a thin unsaturated zone and higher volume of contaminant spill and rainfall intensity than with thick unsaturated zone and lower volume of contaminant spill and rainfall intensity.On the other hand, the retained TPH concentrations increased with increasing soil depth and contaminant volume but decreased with increasing rainfall intensity, implying that less groundwater contamination will occur with a thick unsaturated zone, higher volume of contaminant spill and lower rainfall intensity than with thin unsaturated zone, lower volume of contaminant spill and higher rainfall intensity.

From the measured leached and retained TPH concentrations, transport and retention models were developed. The models were used to fit the transport and retention of crude oil in sandy soil after release with correlation coefficient of 0.9407 and 0.9446 and predictive relevance of 0.8273 and 0.8422 for the transport model and retention model respectively, indicating that the developed models can adequately predict the transport and retention of crude oil in sandy soil following release.

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Ejikeme Ugwoha" Modelling the Transport of Crude Oil in Sandy Soil" International Journal of Engineering Research and Applications (IJERA), Vol. 09, No.08, 2019, pp. 01-12