IJISET - International Journal of Innovative Science, Engineering & Technology, Vol. 6 Issue 7, July 2019
ISSN (Online) 2348 – 7968
www.ijiset.com

Laboratory Assessment on the Rate of Diffusion of Spilled Oil in a Marine Environment

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ABSTRACT

This study was carried out using certain physio-chemical parameters to determine the Diffusion of Spilled Crude Petroleum in Marine Sediment. The work involved the collection of crude oil and sediment samples from five (5) Local Government Areas of Bayelsa State, in Niger Delta region of Nigeria. The investigation was carried out through a laboratory analyses between March and November, 2017. The sediment samples were charged into a fabricated tubular reactors and the three different crude oil samples were introduced through the sediments. Contaminated sediment samples in the tubular reactor were collected every 30 days at a depth of 15 meters and analyzed for TPH, THC, PAH and BTEX concentrations and other metallic elements using Gas Chromatograph-Flame Inoization Detector (GC-FID). After the analyses of the field samples for twenty two (22) parameters, four of the components of crude petroleum sample, namely, Total Hydrocarbon Content (THC), Total Petroleum Hydrocarbon (TPH), Poly Aromatic Hydrocarbon (PAH), and Benzene Toluene Ethylene Xylene (BTEX) were used to investigate the characteristics of crude oil diffusion in Marine Sediment. The transport process modeling tools includes: Analysis of Variance (ANOVA) and Exponential Smoothing equations. The study on examining the petroleum components and exchangeable cations parameters on the flow dynamics in the sediment confirmed that Spilled Crude Petroleum decreases as it diffuses into the Sediment within the Marine Environment. Results obtained from ANOVA revealed the following for TPH, $Y_A = -13.92x + 7171$ with correlation coefficient (r) of 0.9920; $Y_B = -18.72x + 4038$ with correlation coefficient (r) of 0.9494; $Y_C = -219.50x + 6165$ with correlation coefficient (r) = 0.9796. Similar equations were derived for THC, PAH and BTEX. Thus, it is possible to predict the depth of crude oil diffusion in event of subterranean emergency.

1.0 Introduction

Most nations round the world depend on oil and gas for filling of cars, generation of electricity and other domestic purposes. In view of the increases in the world population, the demand for oil and gas has also increases. The exploration for oil in Nigeria began in 1937 (Adeyinka, et al, 2004; Akpofure, et al, 2000). In order to meet the pressing need for oil and gas, oil exploration and exploitation is increasing at alarming rates. Oil exploration and exploitation activities have significant environmental consequences. With increasing production of crude oil and discovery of major oil reserves, more effort has been made to exploit this resource whose operations include oil exploration, drilling, production, transportation, processing, and storage.

Crude oil operations, refining and other allied industrial activities in the Niger Delta, have led to wide scale contamination of most of its farmlands, creeks, swamps and rivers with hydro-carbons and dispersant products (Abowei, and Sikoki (2005); Nelson and Egil, 1998). The contamination of the natural environment by crude oil constitutes public health and socio-economic hazards (Etukudo and Simeon, 2018).

The impact of oil spill in sediment can be related to short-and long-term toxicities. Various methods of protecting the environment against problems created by oil contamination include use of sorbent materials; chemical dispersants and pit incineration of oily waste within a firebreak zone, most of these methods are undesirable due to ecological effects both on the crops and the environment (Beckley and Anoliefo, 2012).

In the freshwater swamp and inland rain forests, crude oil pollution continues indefinitely as petroleum and gas exploitation corridors are continuously being opened to the detriment of biodiversity of all the agents of forest depletion and degradation in the Niger Delta. Petroleum resources development industries appear to be the most devastating unit as almost all their operations are within the Marine environment.

In the process of petroleum industries developments, vast areas of mangrove lands are polluted and left unproductive, oil spills occur destroying the vegetation and the entire ecosystem.

One of the major environmental problems associated with oil exploitation is crude oil spillages. Oil spills may occur for numerous reasons, such as equipment failure (Manufactures or Material error), corrosion (aging of pipes), operational (human) error, sabotage, blow out, mystery spill (spill of unknown origin) and disasters (Siddique and Adams, 2002). There were about 16,334 reported cases of crude oil spillages between 1976 and 2017, with over 5.8m barrels of crude oil released into the environment (Odiete, 1999).

Awobajo, (1981) reported that about 400, 000 barrels of crude oil was released into the sea of Bayelsa State from the Texaco's Funiwa well 5 blow-out in 1980. About 40, 000 barrels of crude oil was released into the Niger Delta environment on January 12, 1998 from Mobil producing Nigeria Unlimited'sIdoho oil spillage, which occurred off Akwa-Ibom State due to a burst on corroded oil pipeline conveying crude oil from Idoho oil field to Qua Iboe Terminal at Mkpanak. When crude oil is spilled on land, the greasy fractions permeate slowly into the soil and are slowly biodegraded, while the light hydrocarbon fractions evaporate. Some that could not permeate the soil become thicker like tar and impact on the surface of the incident area. Although, most crude oil have toxic ingredients, among the most troublesome are the high molecular weight compounds, especially the polycyclic aromatic hydrocarbons (PAH's) that include many known carcinogens, which can combine with environmental materials to form other carcinogens. Vandermulen, (1995)

The release of crude oil into the environment may therefore, result in habitat fragmentation, destruction and disruption of ecosystems, interference in natural biogeochemical cycles and the loss of plants and animals. Amajor (2002) reported the oil spill incident of 1970 at Ejamah-Ebubu near Elem, Rivers State rendered farmlands and streams devastated. The NNPC spillage of 1982 at Abudu and Owa in Delta State also rendered the soil totally oil logged, whereas economic crops withered to death. The pollutants and toxicants in crude oil may cause deaths of plants and animals, disrupt biochemical pathways, metabolites and enzyme systems in all the cells and tissues of organisms.

For sometimes now, Niger Delta region of Nigeria has provided the natural resources to support an extensive and successful petroleum industry. But with continued growth within the industry, there must be an increase in awareness and studies on how to protect the natural resources. Increase in petroleum exploration and production increases the potential hazards to ecologically sensitive areas, such as wet lands and forests. This region contains more than 40% of the nation's wetland. Oil production and exploration take place in this environment, and as a result, they are continually at risk to petroleum contamination caused by accidental spills, leaks, or discharges.

In such occurrences, most farmers especially fishermen and other land users are always left in confusion and rendered helpless choosing the best way of restoring the potentials of such lands. Indeed, much have been done and still being done so far on the remediation or techniques of curbing oil pollution on polluted soils; but a little has been done on the remediation response on marine environment.

Hitherto, the prevalent method adopted in curbing oil-spill on land is by scooping off the polluted top soil, which does not require much technological known-how. Whereas, the approach adopted for remediation of polluted marine environment requires high technological know-how, which is labour-intensive and highly expensive. In view of the above concerns, it has become imperative for researchers to develop interest for empirical evidence on the best possible approach to handle crude oil incident within marine environment.

Furthermore, the major connection in the hydrological cycle is the vertical (downward) and horizontal (sideway) penetration of superficial water via porous sediment down to the groundwater table and storage. The essential component of the water progression in the sediment can be exploited using the non-aqueous phase liquid (NAPL) contaminants. An example of an insoluble, light NAPL which are denser than water is Petroleum hydrocarbon. A mathematical model forecasting the diffusion spread of spilled crude petroleum in different directions especially along the vertical and horizontal will assist the spillers in their oil spill contingency planning. It is necessary to mention that the veracity of the model is very important to ascertain the accuracy and reliability. Hence, this work aimed at developing a diffusion model, which could be used in an event of crude petroleum spillage to determine the effectiveness of forecasting the status of spill incidents.

The Transport of spilled petroleum in soils is governed to a great extent by the difference in density of petroleum with respect to water (Watson et al., 2002a). Another factor that influences the transport of free spilled petroleum in the subsurface is the quantity of entrapment of the petroleum in the soil as a result of phase distribution between air, water and petroleum phases in the media (Grifoll& Cohen, 1994). The relative competition among these three can be understood in terms of interfacial tension between the phases, wettability of the soil by these phases and the capillary principle (Kemper &Rosenau, 1998; Zaher, 2001). In addition to the role played by the petroleum physical properties, soil characteristics also influence the transport of spilled petroleum in soil medium. It is therefore, imperative to start this review with a brief discussion of the general composition of soils i.e. soil formation and phase composition.

Classification of Oil Spill

Oil spillage is classified into four groups: minor, medium, major and disaster.

Table 1: Classification of Spill

S/N	Spill Location/Class	Land	Inland water	Coastal and Offshore
1.	Minor (level 1)	Any spill less than 250 bbls	Any spill less than 25 bbls	Any spill less than 250 bbls
2.	Medium (level 2)	Any spill between 250 – 2500 bbls	Any spill between 25 – 250 bbls	Any spill between 250 – 2500 bbls
3.	Major (level 3)	Any spill more than 2500 bbls	Any spill more than 250 bbls	Any spill more than 2500 bbls

Source: National Oil Spill Detection and Response Agent (NOSDRA) 2013 Oil Spill Contingency Plan (OSCP).

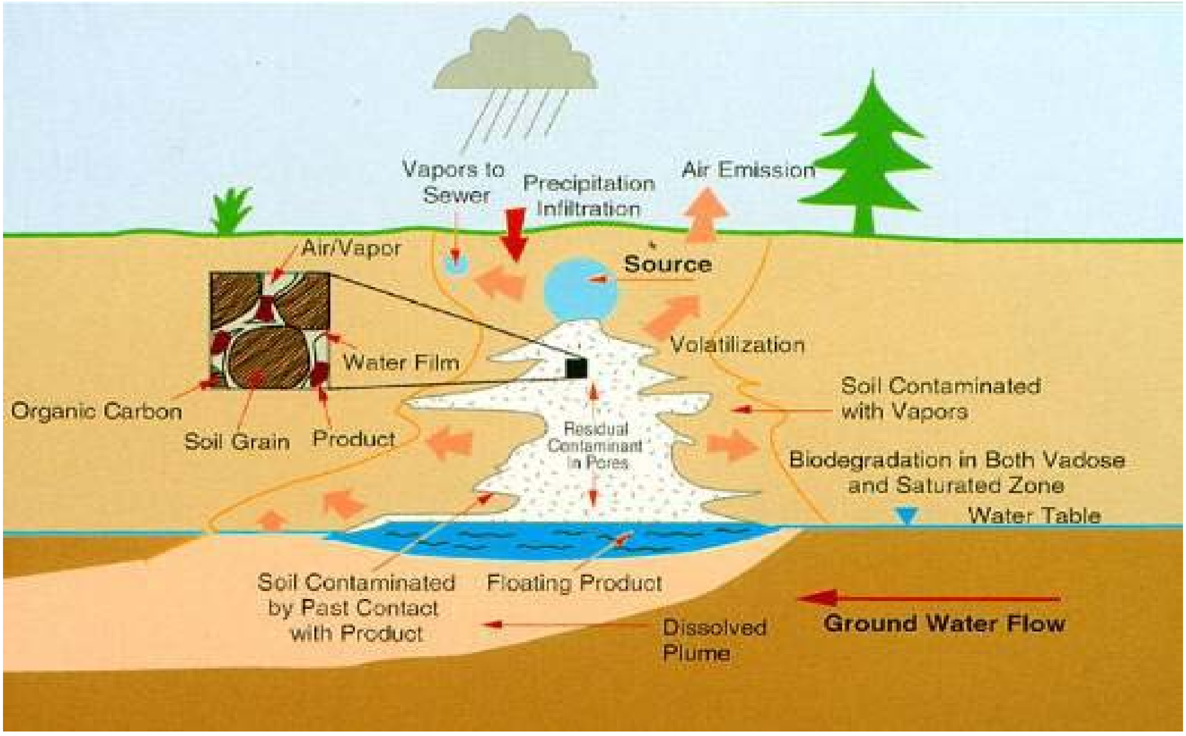


Figure 1: Fate of Oil Spilledand Transport Phenomena of Crude Oil in a Marine Environment **Source:** Gallagher, D. 1997.

Marine sediments are the products of a limited number of physical, biological, and chemical processes. The nature of the resultant sediments is determined by the relative rates of input of material supplied by these processes (Anyakora, et al, 2005). In this study, we focus on rivers sediments especially the shallow water deposits.

Classifications of Marine Sediments

There are two main classifications of marine sediments namely terrigenous and pelagic. Terrigenous sediments are derived from land and found near shore. They are normally delivered by rivers to coastal regions. Pelagic sediments settle slowly out of the water column and are deposited all over the river. According to Carsel& Parrish, (1988), pelagic sediments are deposited at such low rates that they tend to be overwhelmed near shore by terrigenous deposits from land. So, pelagic sediments are normally associated with deep water regions.

2.0 Materials and Methods

2.1 The study Area

The study area is within the Niger Delta area of Nigeria. Sediment samples werecollected from Southern Ijaw, Ekeremor and Ogbia Local Government Areas (LGA) of Bayelsa State (Fig. 2). These LGAs have communities hosting major oil producing companies and are located along the coastal region of Niger Delta in Nigeria. The project area is within the humid tropical zone with defined dry (November – March) and rainy (April - October) seasons. The rainy season is brought about by the Southwest trade wind blowing across the Atlantic Ocean. The dry, dusty and often cold Northeast trade wind blowing across the Sahara desert dominates the dry season and brings a short spell of harmattan (Oguntoyinbo& Hayward, 1987). The relative humidity of the area is high with values ranging from 70% in January to 80% in July. Previous study by Gobo &Ayotamuno, (2004) of the area reveals the average atmospheric temperature to be 25.5^{o}C in the rainy season and 30^{o}C in the dry season months (19.8 - 50.1mm).

The area is located in the geomorphic zone of the Actuate Delta. This area is described as having tidal Deltas with inertial shores that are commonly deflected in a downdraft direction by the dominant long shore current in the area. Thus, the area is water logged throughout the year. The other areas are also located within the Delta zone but the rain forest area.

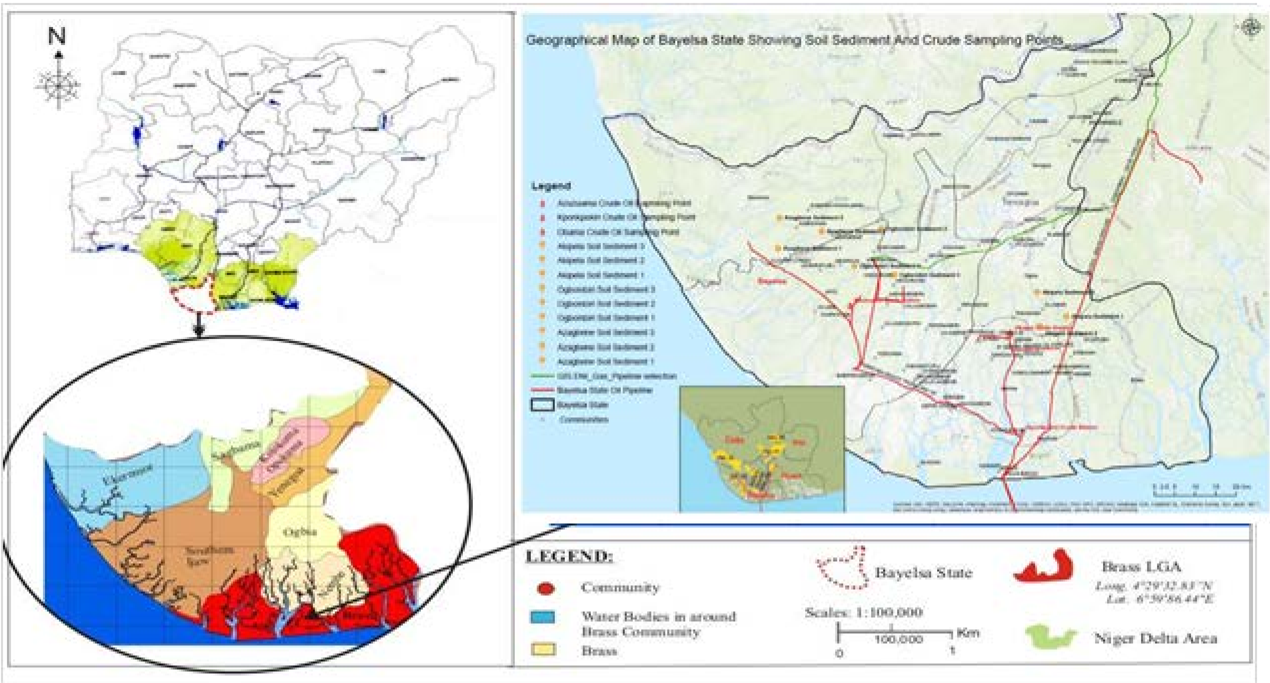


Fig 2: showing maps of Nigeria, Bayelsa State and the sampling areas.

2.2 Methodology

Sediment samples collection

Sediment samples were collected at 3 different stations from each of the three Local Government Areas from which a composite sample representative of the area was made. At each station, a portion of the top 1-2cm, middle 2-5cm and bottom 5-10cm were collected based on the sediment strata and habitants for periwinkle, muse Keeper, water snail and Crab/Cat fish respectively. The composite portion of the sediments was preserved for analysis in labeled polythene bags and stored in an iced cooler.

Table 2: Location; Time, Date and Coordinates of the Sediment Sampling

S/NO	Location	LGA	Date	Time (Hour)	Coordinates	Sediment Acronym
	Azagbene	Ekeremor	04/01/17	1615	(i)N04° 47' 49.3" E005° 44' 39.8" (ii) N04° 47' 40.2" E005° 44' 28.5" (iii) N04° 47' 32.7" E005° 44' 37.0"	SED A
	Ogboinbiri	Southern Ijaw	05/01/17	1435	(i) N04o 49' 03.6" E005o 58' 46.9" (ii) N04° 49' 11.2" E005° 58' 20.0" (iii) N04° 49' 28.3" E005° 58' 34.6"	SED B

	Akipela	Ogbia	06/01/17	1705	(i) N04^{o} 41' 58.3" E005^{o} 55' 10.5" (ii) N04^{o} 41' 40.2" E005^{o} 55' 30.1" (iii) N04^{o} 41' 35.0" E005^{o} 55' 40.7"	SED C

Crude Oil Samples Collection

Also, three different crude oil samples were collected using plastic sampling containers. The containers were washed with 0.1M dilute hydrochloric acid and dried in the sun. During the sampling, each crude type was used to pre-treat the plastic container first before sampling. The initial crude oil was allowed to flow out before collecting mid-stream.

Table 3: Location; Time, Date and Coordinates of the Crude Collection

S/NO	Location	LGA	Date	Time (Hour)	Coordinates	Crude Oil Acronym
	Obama	Nembe	07/01/17	1040	N04^{0} 37' 49.6" E006^{0} 15' 55.5"	CRU A
	Kponkpokiri	Brass	08/01/17	1315	N04^{0} 24' 07.8" E006^{0} 16' 57.2"	CRU B
	Azuzuama	Southern Ijaw	09/01/17	0955	N04^{0} 41' 58.3" E005^{0} 55' 10.5"	CRU C

Crude Oil Dispersion Experiment

In this section, the fabricated columns as shown in Figure 3 used to set up the experiment at Devine Concept Laboratory were thoroughly cleaned, brushed, washed with water and sun-dried before being put on stream after 2 days. The bolts and seals used for water tight connections were loosen, and the sediment were carefully charged from a distance of about 10cm above the bottom section of the column. The fully charged columns with sediment were covered with a rubber seal and the metal cover before tightening it with bolts and nuts. The setups were allowed to stand vertically for 6 hours to check for leakages. Thereafter, about 1200ml of the oil samples were introduced into the column via the top section with a funnel. The complete setups were assembled into a rectangular plastic container to avoid secondary oil spill and held vertically firm with retort stands.

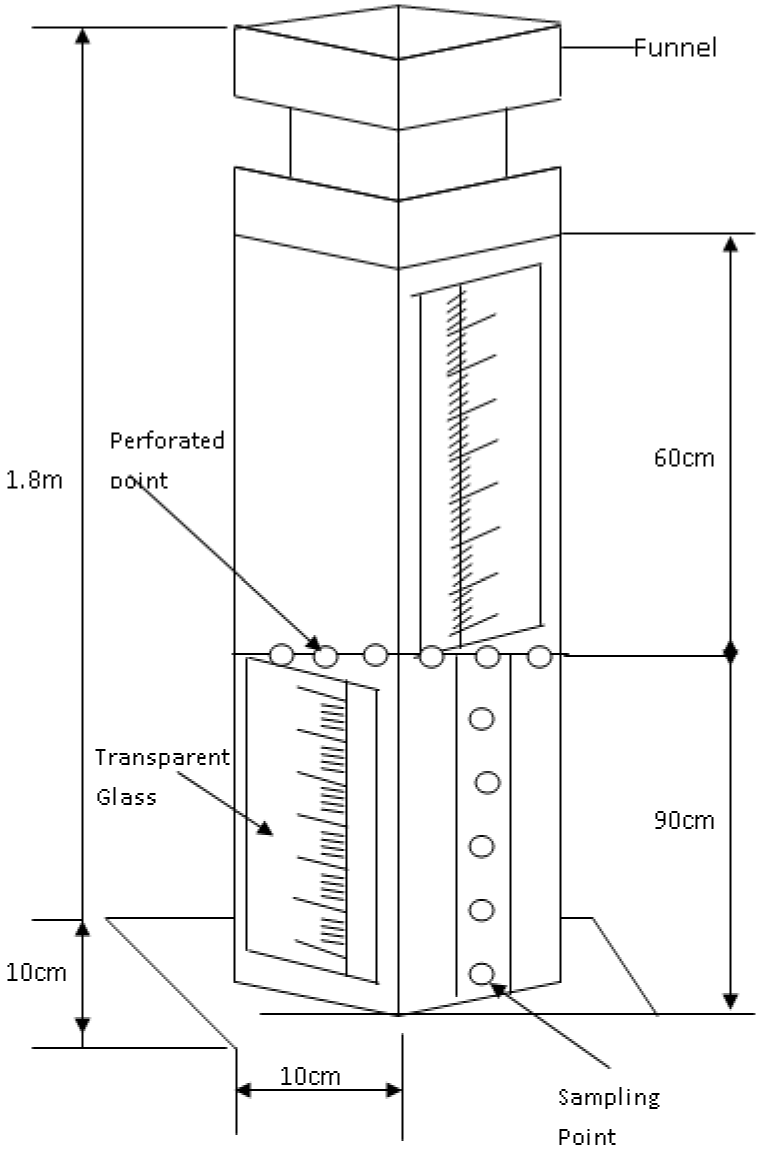


Figure 3: Schematic Diagram of the fabricated Reactor Column.

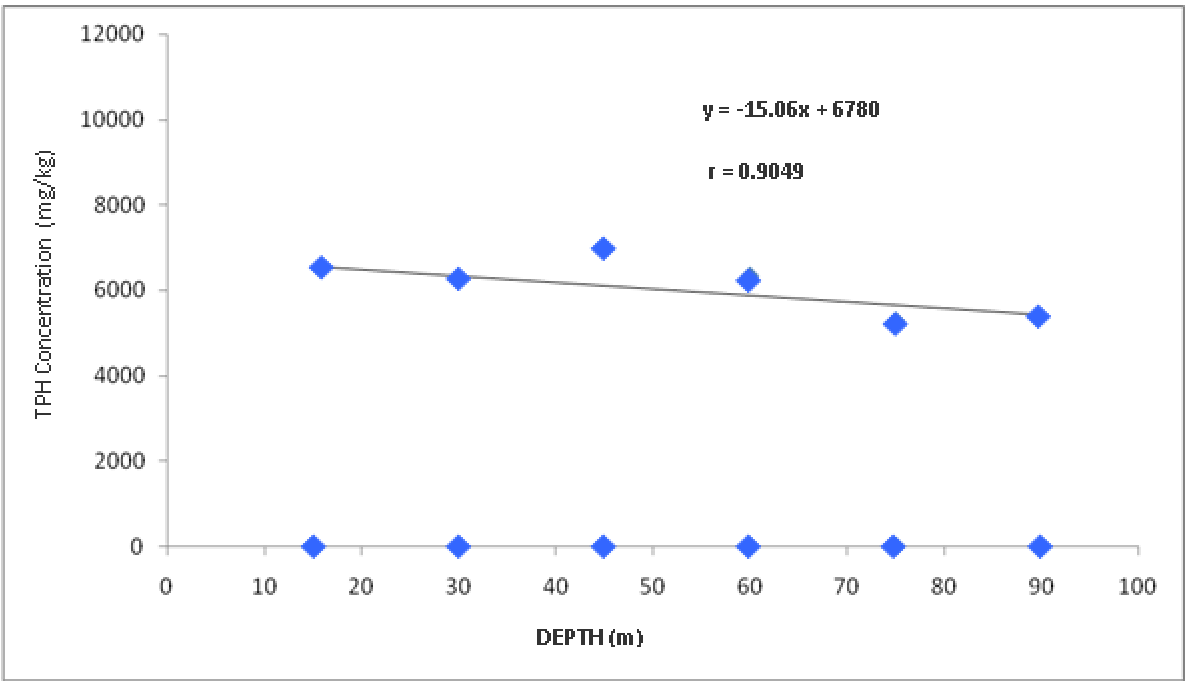
For analysis, sediment samples were collected on monthly bases from each of the perforated points besides the columns generally graduated about 15cm apart. The volume consumed or taken by the sediments were recorded and the samples taken to laboratory for final analysis and results. .Each stand was duly labeled as shown in Table 3.5 below. Also setup in the laboratory was the control where distilled water was used in place of crude oil as solvent.
At 30 days intervals for a period of 180 days, samples of the crude transported into sediment in the column were collected with the aid of spatula and poured into a test tube for collaborating analysis. The samples were collected from the sampling points at 15cm, 30cm, 45cm, 60cm, 75cm and 90cm and taken to the Laboratory for analysis.
At each of the identified sampling sites, sediment samples was carried out using a stainless steel Eckman grab to a depth of about 10cm. The sediment samples were immediately transferred in plastic containers and frozen at -4^{0}C for transport to Devine Concept Laboratory for samples preparation and analysis. At the laboratory, coarse materials (>2mm) like pebbles and plant debris were removed from the sediment samples and analysed for the

following parameters; silt, sand, clay, pH, temperature, redox potential (Eh), electrical conductivity (EC), total hydrocarbon content (THC), Zinc (Zn), Lead (Pb), Copper (Cu), Cadmium (Cd), Nickel (Ni), Manganese (Mn), Iron (Fe), and Chromium (Cr). pH, temperature, redox potential (Eh), and electrical conductivity (EC) were measured *in-situ* using a multi-parameter water quality monitor (Orion Model, 1260). At the determination of any of the parameter, the equipment was properly checked and calibrated before the after use.Samples for metal analysis were sieved at 200µm and stored in a dry atmosphere before analysis. The particle size distribution (PSD) was determined using the hydrometer method followed by sieving the sand fraction (>63µm) from silt (<63 and 2µm) and clay (<2µm). Levels of heavy metals in the sediment samples were measured using ATI Unicam Atomic Absorption Spectrophotometer, Model 939. All methods of sample preparation and analysis are consistent with standard methods (van Valine and Morse, 1982; Rauret, 1998; Tessier et al., 1979)

3. 0 Results and Discussion

Total hydrocarbon content (THC)

Graphic Representations of Correlation and Regression Using Analysis of Variance (ANOVA) tests



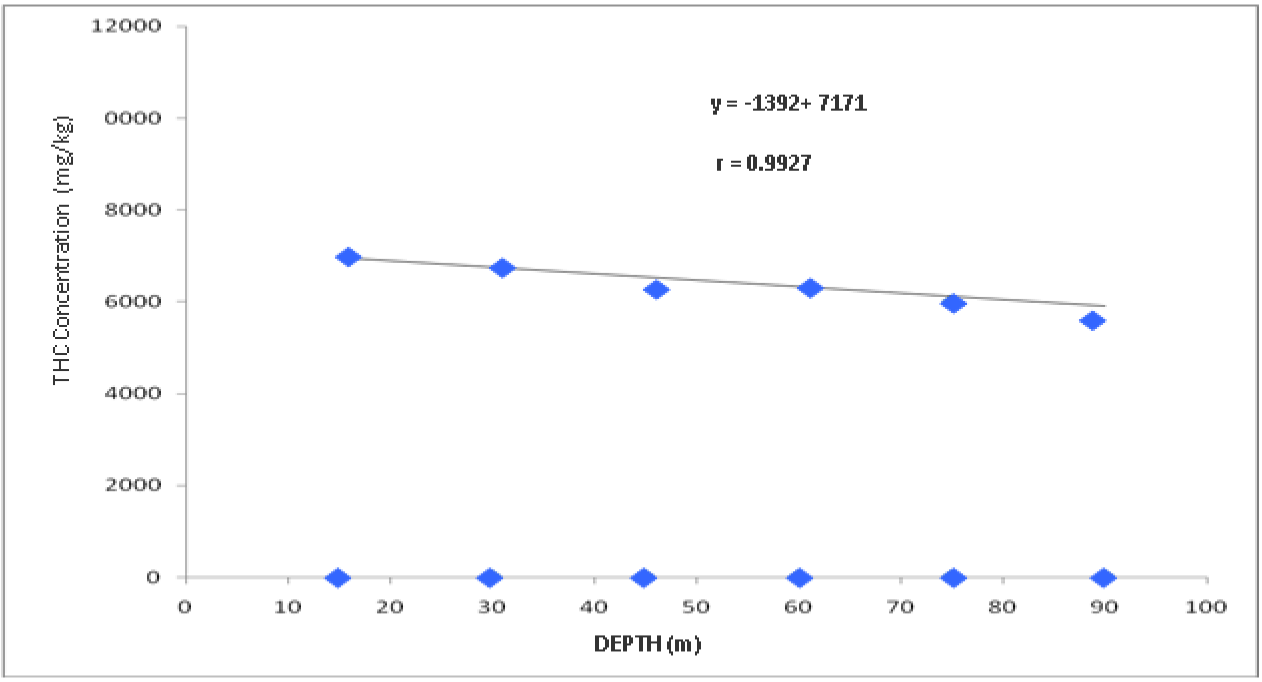
Plot of Correlation and Regression for TPH Concentration against Depth in Sediment A

The graph above illustrate the physic-chemical parameters including Total Hydrocarbon Content (THC) concentrations in the various crude and sediment types, The values ranged from 83.33± 641.89mg/kg and 908.27± 379mg/kg with a mean and control values of 439.07± 296mg/kg and 212.34± 562mg/kg respectively. The higher values in thistable is an indication of the presence of organic materials undergoing biological degradation in the sediment within the period under review. This results and graphical illustrations between the concentration and depth are consistent with the findings in Subramanian et al, 2010. The crude oil sample tend to increase in values as the diffusion process progresses during the laboratory setup. The work concentrated more on this component. Thus, the graphical representation of the experimental and model values were duly compared and their fittings were very closer and proportional with percentage accuracy of about 96.2% as shown in Appendix H-V. The statistical analysis of variance (ANOVA) using a straight line graph gave a correlation and regression equations as follows:

Y_A = -13.92X + 7171 and correlation coefficient (r) of 0.992 for sediment A,
Y_B = -18.72X + 4038 and r = 0.9494 for sediment B and
Y_C = -219.5X + 6165 and r = 0.9796 for sediment C.

In all the sediment types, the concentration of THC increases with depth but the regression is not significant. The graphical representations are presented in Figures 4.12. While the appraisal of the experimental and forecast results as illustrated in Figure 4.39 a remarkable degree of proportionality.

Total petroleum Hydrocarbon (THC)

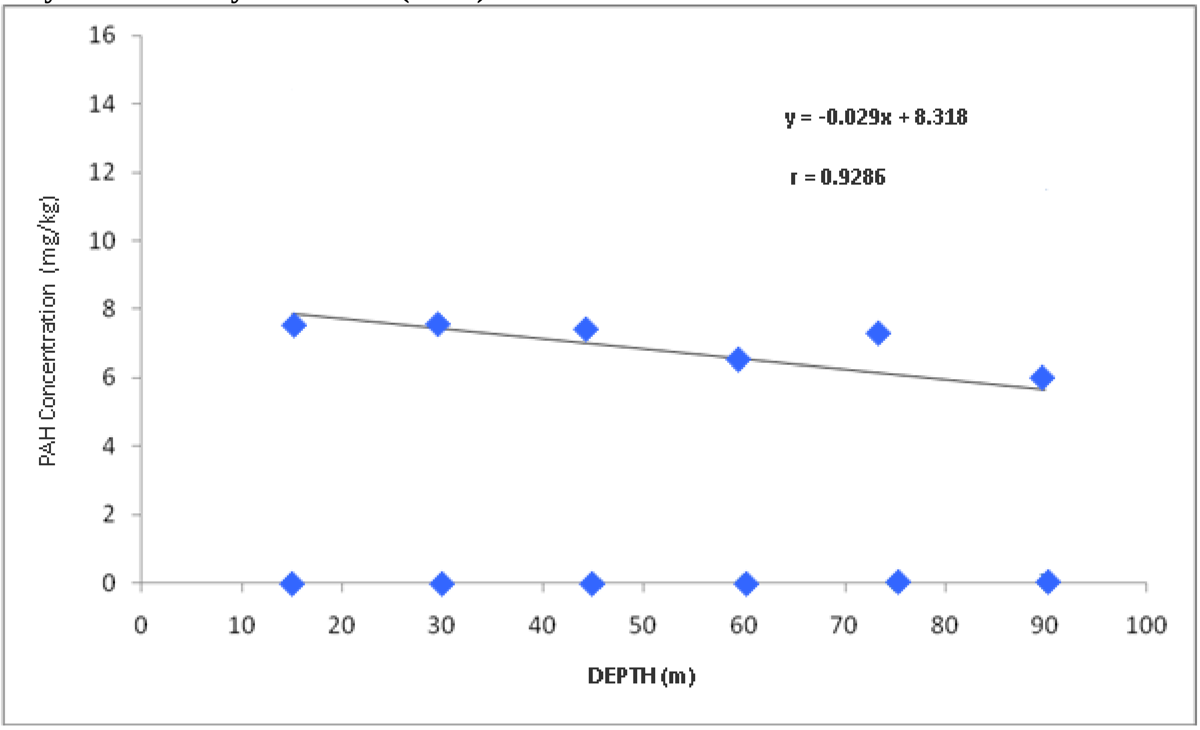


Plot of Correlation and Regression for THC Concentration against Depth in Sediment A

Total Petroleum Hydrocarbon values were high. The values ranged between 360.87± 267.38 mg/kg 860.32± 386.79mg/kg while the mean and control values are 5018.13mg/kg and 27.47± 1.672mg/kg respectively. The TPH values decrease down the systems as the resultant toxicity reduces with time due to contact with the organic matters and the chemical dispersion that occurs in course of the laboratory exercise (Abowei,. and Sikoki, 2005). Meanwhile, the values tend to increase at the bottom part of all the setup as the crude accumulates at the bottom part of the system due to the restricted flow caused by the seal at the end of the columns. The work concentrated more on this component. Thus, the graphical representations of the experimental and model values were duly compared and their fittings were very closer and proportional with percentage of about 96.2%. Also, the work concentrated more on this component. Thus, the graphical representations of the experimental and model values were

duly compared and their fittings were proportional with percentage error of about 93.75%. The statistical analysis using a straight line graph gave regression equations of:
Y_A= -15.06X + 6780 and correlation coefficient (r) of 0.9049 for sediment A,
Y_B = -134.6X + 4239 and r = 0.9971 for sediment B and
Y_C = -180X + 5137 and r = 0.9447 for sediment C.
In all the sediment types, the concentrations of TPH reduce with depth but the regression is not significant. See Appendix, C.

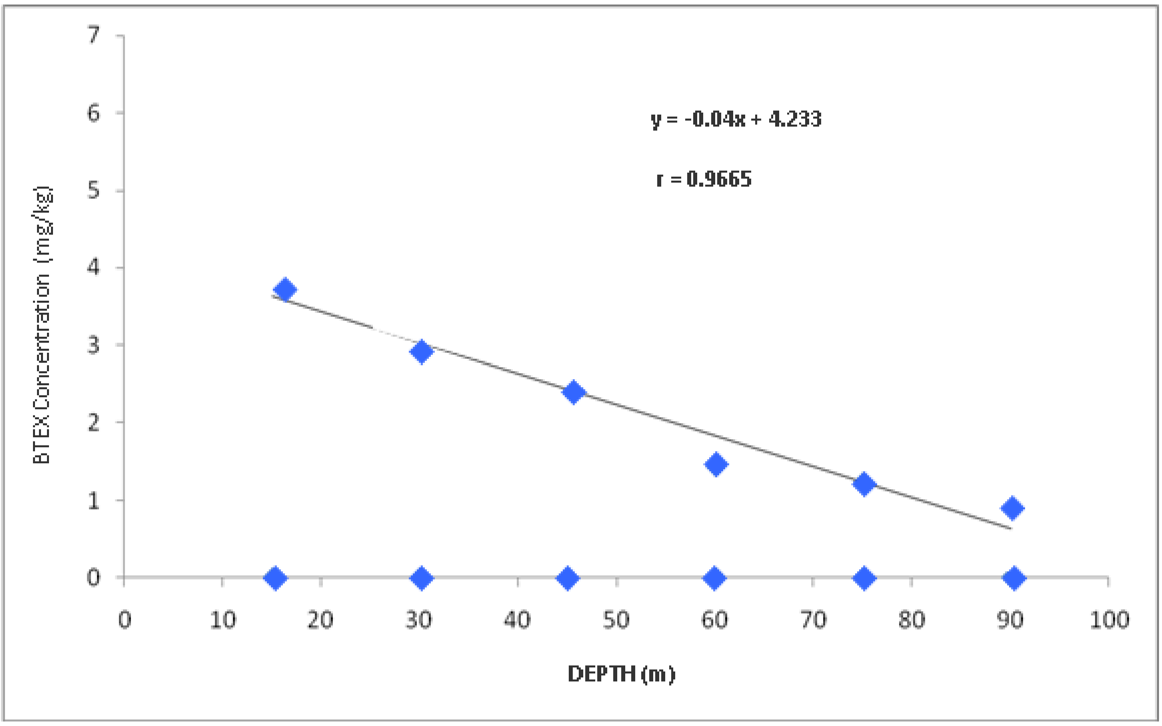
Poly Aromatic Hydrocarbon (PAH)



Plot of Correlation and Regression for PAH Concentration against Depth in Sediment A

PAH values ranged between 2.119± 0.585mg/kg and 12.35± 0.589mg/kg while the mean and control values of 7.399mg/kg and 0.01± 0.00mg/kg respectively. The differential diffusion values decrease down the system with respect to time and distance due to the volatility nature of the component (Abowei, et al, 2008). Though, the value was highest on the sixth month due to the accumulation of crude oil at the bottom section of the column as a result of the restriction caused by the partially sealed end section of the column. Furthermore, on the relationship between the experimental and model values, the graphical representations were closer and proportional with percentage of about 87.5%.accuracy. From the statistical analysis, the following regression equations were obtained,
Y_A = - 0.029X + 8.318 and correlation coefficient (r) of 0.9286 for sediment A,
Y_B = - 0.018X + 6.130 and r = 0.9473 for sediment B and
Y_C = - 0.290X + 6.195 and r = 0.9377 for sediment C.
In all the sediment types, the concentrations of TPH reduce with depth but the regression is not significant. See Appendix, C.

BTEX



Plot of Correlation and Regression for BTEX Concentration against Depth in Sediment A

The BTEX concentration values ranged between 2.090± 0. 5.25mg/kg with a mean value of 2.35mg/kg. The concentration decreases with time due to the volatility of the component. (Ndiokwere, 1984). On the relationship between the experimental and model values, the graphical representations were closer and proportional with percentage error of 100%.accuracy. From the statistical analysis, the following regression equations were obtained,

Y_A = - 0.04X + 4.233 and correlation coefficient (r) of 0.9665 for sediment A,

Y_B = - 0.219X + 5.216 and r = 0.9465 for sediment B and

Y_C = - 0.118X + 3.291 and r = 0.9301 for sediment C.

In all the sediment types, the concentrations of TPH reduce with depth but the regression is not significant. See Appendix, C.

4.0 Conclusion

This study was carried out using certain physio-chemical parameters to determine the Diffusion of Spilled Crude Petroleum in Marine Sediment. The work involved the collection of crude oil and sediment samples from five (5) Local Government Areas of Bayelsa State, in Niger Delta region of Nigeria. The investigation was carried out through a laboratory analyses between March and November, 2017. The sediment samples were charged into a fabricated tubular reactors and the three different crude oil samples were introduced through the sediments. Contaminated sediment samples in the tubular reactor were collected every 30 days at a depth of 15 meters and analyzed for TPH, THC, PAH and BTEX concentrations and other metallic elements using Gas Chromatograph-Flame Inoization Detector (GC-FID). After the analyses of the field samples for twenty two (22) parameters, four of the components of crude petroleum sample, namely, Total Hydrocarbon Content (THC), Total Petroleum Hydrocarbon (TPH), Poly Aromatic Hydrocarbon (PAH), and Benzene Toluene Ethylene Xylene (BTEX) were used to investigate the characteristics of crude oil diffusion in

Marine Sediment. A finite element and analytical models were used to analyze the crude oil diffusion within the study sediment samples. The transport process modeling tools includes: Model Development, Analysis of Variance (ANOVA) and Exponential Smoothing equations. The study on examining the petroleum components and exchangeable cations parameters on the flow dynamics in the sediment confirmed that Spilled Crude Petroleum decreases as it diffuses into the Sediment within the Marine Environment. From the model developed using second order partial differential equation in one flow-direction, the results obtained when graphically compared by plotting concentration against depth, showed a high degree of fitness. While the equations obtained from ANOVA revealed the following for TPH, $Y_A = -13.92x + 7171$ with correlation coefficient (r) of 0.9920; $Y_B = -18.72x + 4038$ with correlation coefficient (r) of 0.9494; $Y_C = -219.50x + 6165$ with correlation coefficient (r) = 0.9796. Similar equations were derived for THC, PAH and BTEX. Thus, it is possible to use any of the equations to predict the depth of crude oil diffusion in event of subterranean emergency.

LIST OF ACRONYMS

API	American Petroleum Institute
ESI	Environmental Sensitivity Index
GC	Gas Chromatograph
FID	Flame Ionization Detector
TPH	Total Petroleum Hydrocarbon
THC	Total Hydrocarbon Content
OSA	Oil Suspended Sediment Aggregates
DPR	Department of Petroleum Resources
PSD	Particle Size Distribution
Cr	Chromium
Fe	Iron
Mn	Manganese
Ni	Nickel
Pb	Lead
Cu	Copper
Zn	Zinc
Cd	Cadmium
Eh	Redox Potential
EC	Electrical Conductivity
Mg/Kg	Miligram/Kilogram
%	Percentage
µS/cm	Micro second per centimeter
G/cm^3	Gram per centimetre cubic
HA	Humic Acid
WHOI	Woods Hole Oceanagraphic Institute
BD	Bulk Density

NOMENCLATURE

Symbol	Definition	Unit
TPH	Total Petroleum Hydrocarbon	*ppm*
THC	Total Hydrocarbon Content	*PPM*
PAH	Poly Aromatic Hydrocarbon	Kg/m^3
BTEX	Benzene, Toluene Ethylene Xylene	Kg/m^3
D_{AB}	Diffusion Coefficient of Crude oil in Sediment	m^2s^{-1}

ISSN (Online) 2348 – 7968
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P	Density	kgm^{-3}
T	Temperature	^0C
M	Mass	kg
V	Volume	m^3
x	Distance (x Direction)	cm
y	Distance (y Direction)	cm
z	Distance (z Direction)	cm
t	Time	s
C_A	Crude Concentration	mg/l
A	Area	m^2

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