Current Science Perspectives 2(3) (2016) 69-77

International Scientific Organization
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Characteristics concept for the examination of total hydrocarbon content and total organic carbon in contaminated soil zone

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A R T I C L E   I N F O

Article type:
Research article
Article history:
Received December 2015
Accepted March 2016
July 2016 Issue
Keywords:
Characteristics
Concept
Examination
THC
TOC
Contaminated
Soil zone

A B S T R A C T

In the investigation to examine the rate of diffusion of Total Hydrocarbon Content (THC) and Total Organic Carbon (TOC) upon continuous discharge of effluent into the soil zone in Niger Delta area of Nigeria was studied using the concept of empirical model as well as experimental approach. The model obtained was simulated using MathCAD 2000 professional software. The experimental and model result obtained from the simulation of the developed equation was compared numerically and graphically. A reasonable level of agreement was obtained between the two results. The study of the fate of chemical pollutant in soil environment is vital, for sustaining agricultural productivity and land utility which are directly related to human safety. It's imperative to know, that whatever degrades, damages, or destroy the soil environment ultimately have an impact on human life and may threaten our very ability to survive. Hence, this paper has provided an in-depth understanding of the causes, distribution processes and effects of contaminants discharged in the environment.

Capsule Summary: The research work illustrates the relationship between experimental and developed model results obtained from the investigation for total hydrocarbon content and total organic carbon characteristics upon the influence of diffusion of pollutant in soil environment.

Cite This Article As: C. P. Ukpaka and Itabeni Collins. Characteristics concept for the examination of total hydrocarbon content and total organic carbon in contaminated soil zone. Current Science Perspectives 2(3) (2016) 69-77

INTRODUCTION

A number of other bonding mechanisms exist by which organic compounds are absorbed to soil surfaces. For any given compound (organic or inorganic), it is likely that a combination of mechanisms is responsible for sorption onto soil. Whatever the mechanism is, soil organic matter is the principal sorbent for many nonionic organic chemicals. It is important to know a particular chemical's attraction to organic matter, and the amount of organic matter available in a particular soil (Allan and Elnajjar, 2012; Allan et al., 2009; Amadi and Ukpaka, 2007; Amadi et al., 2007abc; Daichao and David, 1999; Ewing et al., 1999; Jacques et al., 2008; Looney and Falta, 2000; Maddalena et al., 2007; Mckone and Bennet, 2003; Mirbagheri, 2004; Muibat and Jimoh, 2013; Neeka et al., 2006; Njobuenwu et al., 2005; Ogoni et al., 2004; Parlenge and Hill, 1995; Prommer et al., 1999; Rushdi and Mohammed, 2010; Schmidt and Gier, 1990; Schwarz et al., 2009; Simnek et al., 2001; Simunek et al., 2013; Singh et al., 2007; Stagnitti et al., 1995; Ukpaka et al., 2005; Ukpaka et al., 2008; Ukpaka et al., 2009; Ukpaka, 2005ab; Ukpaka, 2006abcd; Ukpaka, 2007ab; Ukpaka, 2008; Ukpaka, 2012; Watson et al., 1998).
One interesting aspect of chemical transport involves whether chemicals are more or less dense than water. This demonstration shows a chemical developing finger because it

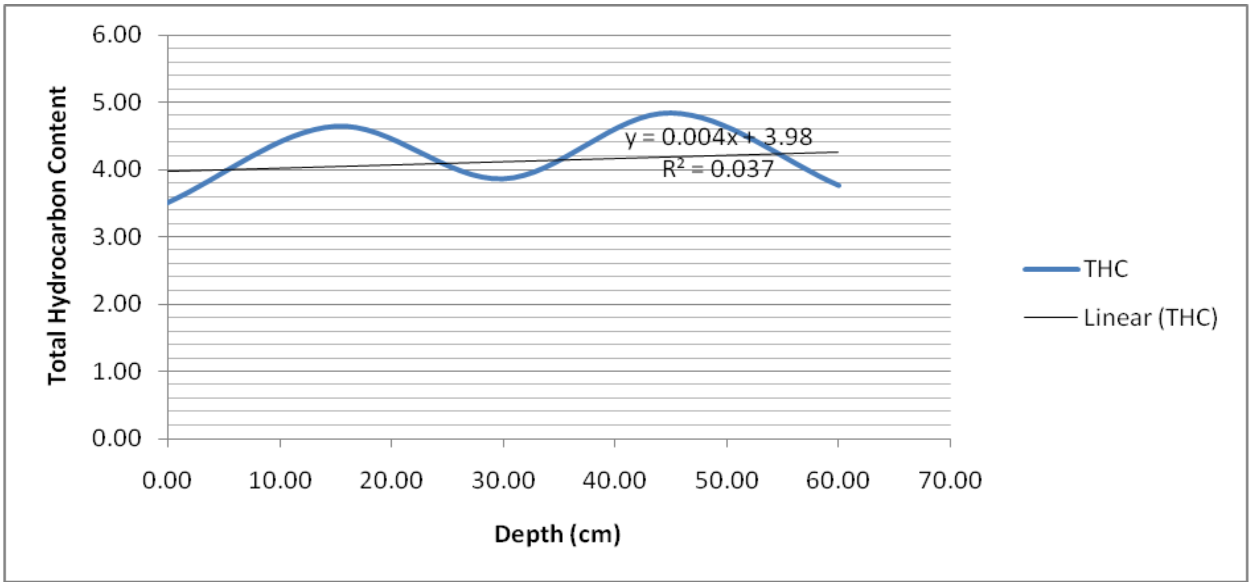

Fig. 1: Graph of Experimental Total Hydrocarbon Contentvalues against Depth.

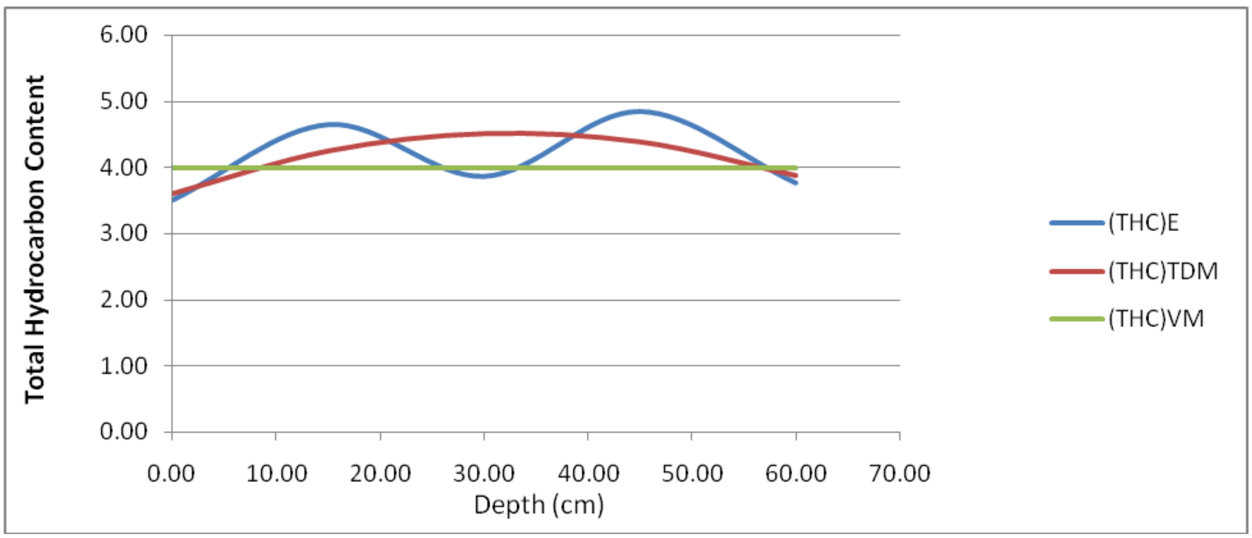

Fig. 2: Comparison of Experimental, Theoretical model and Validated model Total Hydrocarbon Content values against Depth.

Table 1: Results of Experimental Determination of Physico-chemical Parameters

Parameter	Unit	Depth (cm) (Dp)				
		0.0-5.0	15.0	30.0	45.0	60.0
Total Hydrocarbon	ppm	3.50	4.65	3.86	4.85	3.76
Organic Carbon	mg/kg	8.75	7.20	7.60	6.86	6.38

is denser than water. These fingers of concentrated chemical sink to the bottom of the water column before they appreciably mix with the water. Spilled chemicals that are denser than water will tend to sink to the lower depths of a ground water aquifer. Chemicals that are less dense than water (for example, gasoline) will tend to float near the top of a ground water aquifer. Without significant mixing due to groundwater movement, chemicals that are approximately the same density as water tend to remain near the top of a groundwater aquifer.

So, chemical sorption to soil particles, chemical solubility and chemical density all affect the rate of chemical transport.

Transformation

Chemicals undergo numerous transformations in both soil and water (Ukpaka et al., 2005). Hydrolysis, photolysis, oxidation, and reduction are some of the most common transformations.

Hydrolysis

Table 2: Theoretical Computation of Total Hydrocarbon (THC) values Investigated

Depth (cm) (Dp)	Total Hydrocarbon (ppm) $(THC)_{EV}$	Dp*THC	Dp^2	Dp^2*THC	Dp^3	Dp^4
0.00-5.00	3.50	0.00	0.00	0.00	0.00	0.00
15.00	4.65	69.75	225.00	1046.25	3375.00	50625.00
30.00	3.86	115.80	900.00	3474.00	27000.00	810000.00
45.00	4.85	218.25	2025.00	9821.25	91125.00	4100625.00
60.00	3.76	225.60	3600.00	13536.00	216000.00	12960000.00
∑ Dp	$\sum(THC)_{EV}$	∑ Dp*THC	$\sum Dp^2$	$\sum Dp^2$*THC	$\sum Dp^3$	$\sum Dp^4$
=150.00	=20.62	=629.40	=6750.00	=27877.50	=337500.00	=17921250.00

Table 3: Comparison of Experimental, Theoretical and validated values for Total Hydrocarbon (THC) investigated.

Depth (cm) (Dp)	Total Hydrocarbon (ppm) $(THC)_{EV}$	Total Hydrocarbon (ppm) $(THC)_{TDM}$	Total Hydrocarbon (ppm) $(THC)_{VM}$
0.00-5.00	3.50	3.59	3.98
15.00	4.65	4.24	4.04
30.00	3.86	4.50	4.10
45.00	4.85	4.38	4.16
60.00	3.76	3.87	4.22

Table 4: Theoretical Computation of Organic Carbon (OC) values Investigated

Depth (cm) (Dp)	Organic Carbon $(OC)_{EV}$	Dp*OC	Dp^2	Dp^2*OC	Dp^3	Dp^4
0.00-5.00	8.75	0.00	0.00	0.00	0.00	0.00
15.00	7.20	108.00	225.00	1620.00	3375.00	50625.00
30.00	7.60	228.00	900.00	6840.00	27000.00	810000.00
45.00	6.86	308.70	2025.00	13891.50	91125.00	4100625.00
60.00	6.38	382.80	3600.00	22968.00	216000.00	12960000.00
∑ Dp	$\sum (OC)_{EV}$	∑ Dp*OC	$\sum Dp^2$	$\sum Dp^2$*OC	$\sum Dp^3$	$\sum Dp^4$
150.00	36.79	=1027.50	=6750.00	=45319.50	=337500.00	=17921250.00

Is the cleavage of molecules by water, and is one of the most important reactions in breaking down pesticides. Hydrolysis can occur in the soil with or without microorganisms (Ukpaka and Nnadi, 2008. Ukpaka, 2006; Ukpaka and Oboho, 2006; Ukpaka et al., 2006; Ukpaka and Pele, 2012).

Photolysis

It's the process where ultraviolet or visible light supplies the energy for decomposition of chemical compounds. Photolysis can be a very important chemical transformation process (Ukpaka, 2004ab; Ukpaka, 2005; Ukpaka et al., 2007; Ukpaka, 2009).

Oxidation

Oxidation is the process where a chemical loses electrons, such as rust forming on iron. Reduction is the process where a chemical gains electrons. Reduction can be a non-biological process, or a biological process as in anaerobic sewage treatment (Ukpaka et al., 2004ab; Ukpaka et al., 2005ab; Ukpaka et al., 2007a).

Maddalena et al. (2007) in their work on contaminant flow processes showed that Biogeochemical and Physical processes in the unsaturated zone (vadose zone) control the contaminant fate and transport of diffuse pollution through the soil environment and to other compartments. Complexity of the zone flow processes arise from several sources first of

Table 5: Comparison of Experimental, Theoretical and validated values for Organic Carbon (OC) investigated

Depth (cm)	OC (mg/kg) $(OC)_{EV}$	OC (mg/kg) $(OC)_{TDM}$	OC (mg/kg) $(OC)_{VM}$
0.00-5.00	8.75	8.52	8.374
15.00	7.20	7.79	7.879
30.00	7.60	7.21	7.384
45.00	6.86	6.77	6.889
60.00	6.38	6.48	6.394

OC = organic carbon

all, the soil itself is a complicated disperse system made up of microscopically heterogeneous mixtures of solid, liquid and gaseous phases. The flow of contaminant is not limited to the dissolved phase. Contaminants may be attached to colloidal or suspended particles. Once mobilized, these particles can carry even strongly sorbing organic and inorganic contaminants. Contaminants migrating through the soil are reacting with the soil constituent and undergo complex physical, chemical and biological transformation. Most contaminant transport mechanism in soil are mediated or at least strongly affected by the presence and movement of soil water. Thus deep knowledge of the physics and chemistry of soil water movement in heterogeneous and dynamic system is an essential prerequisite for a valid and reliable estimation of the contaminant fate. Structural and Functional Biodiversity of soil is an essential aspect to consider on the fate of contaminants in soil environment.

Ewing (1999) studied the groundwater flow driven by different pressure under saturated and unsaturated conditions. They presented numerical techniques for modeling multi component gas flow in porous media. They utilized the mixed finite element method over quadrilaterals as a solver to the Non-Darcy flow equation and a conservative Godunov-type scheme for the mass balance equations. Schwarz et al. (2009), in their work they assets the risk on environment and heath resulted from the leakage of CO_2 in air near the ground, by having an exact solution of the advection–diffusion equation. Their results show a good agreements with numerical simulation performed for similar case with different boundary conditions. Maddalena et al. (2007) utilized a relatively simple transport equation modeling to give a reliable forecast of the pollutants spreading in ground water after a mismatch, and to determine the level of possible damage to the environment. The solution of the approximated transport equations was used to describe the concentration of pollutants. Knowledge of the concentration level is a prerequisite for finding efficient ways to prevent the further spreading and the removal of pollutants from the groundwater.

Pommer et al. (1999), in their work, numerically used an operator-splitting method to couple advection-dispersive transport of organic and inorganic solute with geochemical equilibrium package; One-dimensional multi-component model accounting for transport inorganic equilibrium chemistry. The work focus on validation of the experimental data to presented model. The model was modified to address the three dimensional case in order to compare to realistic field scale.

The convection –dispersion transport model was used (Ukpaka and Nnadi, 2008. Ukpaka, 2006; Ukpaka and Oboho, 2006; Ukpaka et al., 2006; Ukpaka and Pele, 2012; Ukpaka et al., 2004abcd; Ukpaka et al., 2005abc; Ukpaka et al., 2007; Ukpaka et al., 2007b; Van Genuchten and Alves, 1982; Watterson and Nicholson, 1996) in their study to model the concentration distribution of pollutant in ground water due to malfunction of the water proofing system in waste material sewage compositing plant. The model solves the transport equation for a simplified case where all covering parameters were treated as constants. The source of pollutant is considered as a constant mass flow flux supplied along the span of the waste well. The model predicted the development of the concentration distribution for different position and times.

Muibat and Jimoh (2013), In their work on the Mathematical Modeling and Simulation of Mobility of Heavy Metals in soil Contaminated with sewage Sludge, developed a model equation 1, from the convective-dispersive equation, that describes the flow of heavy metals material through soil.

$$c = e^{\frac{1}{\left(1+\frac{Kd.\rho b}{\theta}\right)}t} - e^{\frac{v}{D}x} \qquad (1)$$

Several works have been done on the mathematical modeling of the spread, movement, transport or fate of contaminants in the soil environment almost all the approaches were based on Theoretical (white box) models; using the physical and chemical laws of conservation such as mass balance, component balance and energy balance and correlation data from different sources. The Empirical (black box) models which are obtained by fitting experimental data have not been in use all that to the evaluation of the degree of contaminant transport in soil. This research work is based on Empirical a quadratic equation was obtained from the convective-dispersive equation that relates the Diffusivity (D), the water velocity (v) and the irreversible decay rate (f). The Experimental data obtained from the analysis of the Physio-Chemical parameters of soil samples obtained from the vicinity of the Asphalt Plant Company will be fitted into the Quadratic Equation derived from the previously derived theoretical models. The model his then validated using the best fit model from the experimental data.

MATERIALS AND METHODS

Sample collection

Soil samples were collected within the vicinity of the H & H Asphalt Plant Company Located at Enito 3, a village in Ahoada West Local Government Area, River State, Nigeria.

Soil samples were obtained from the surface (top soil) to a depth of 60cm. A total of five (5) soil samples were collected within the interval of 15cm depth, top soil 0-0.5cm, 15cm, 30cm, 45cm and 60cm. The samples were transported

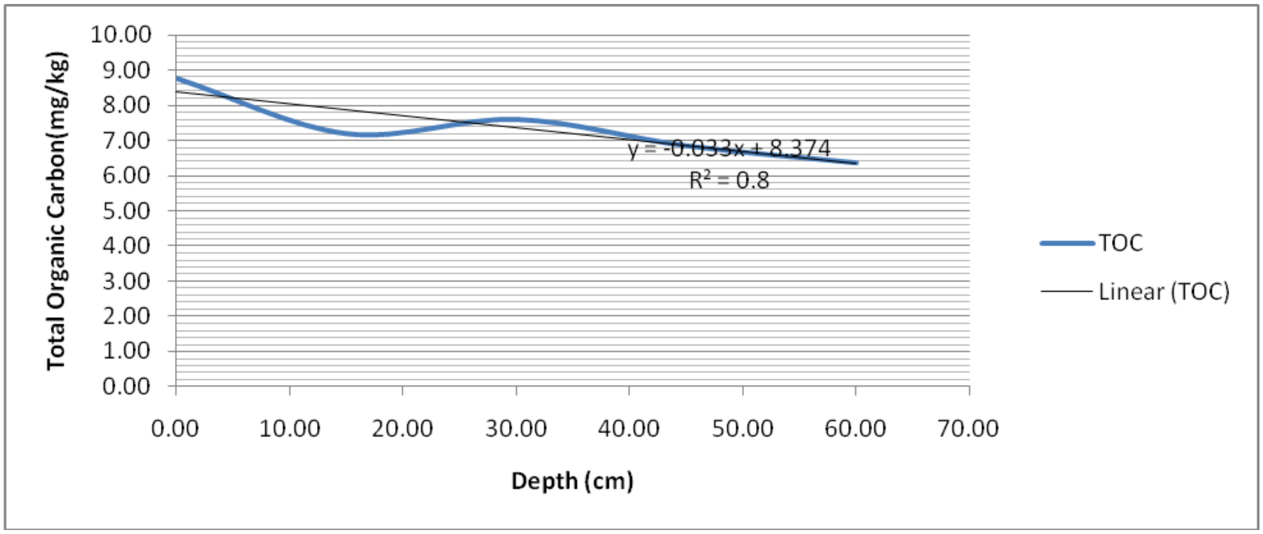

Fig. 3: Graph of Experimental Total Organic Carbon values against Depth.

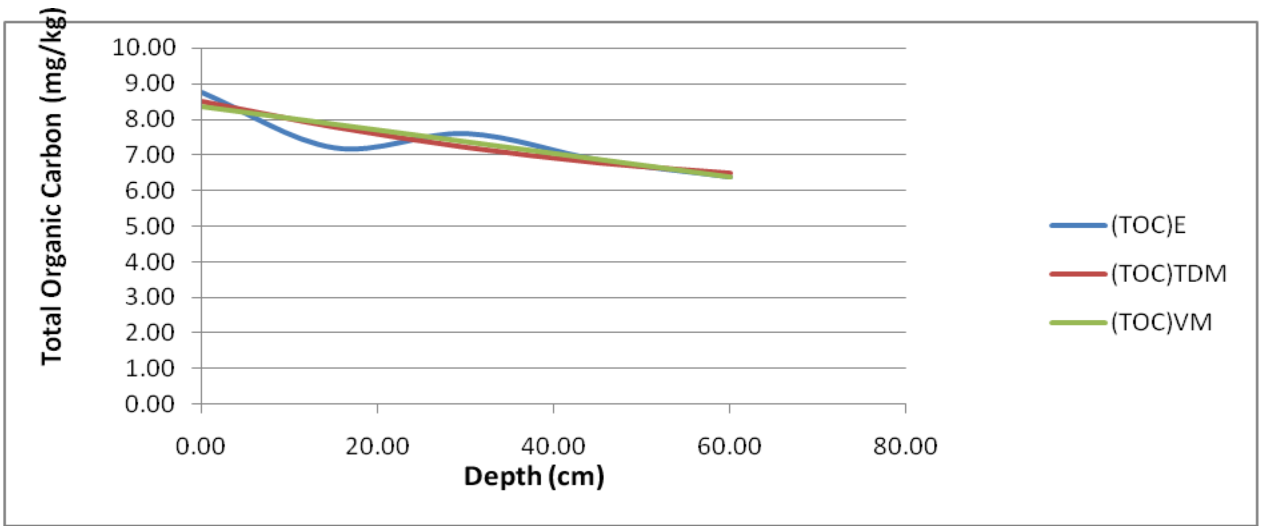

Fig. 4: Comparison of Experimental, Theoretical model and Validated model Total Organic Carbon values against Depth.

to the Chemical Sciences Research Laboratory of the Niger Delta University for analysis, for the determination of the Physio-chemical Parameters.

The total Hydrocarbon Content was determined by spectrophotometric method. 10g of soil was weighed into an extract in flask and 10ml of n-Hexane added; the suspension was shaken for 30minutes and filtered. The spectrophotometer was set at 420nm, absorbance of the filtrate was prepared with Bonny light crude oil. The amount of Total Hydrocarbon Content in the soil was determined using this graph by extrapolation.

Total organic carbon (TOC)

10ml of 1.0M K_2CrO_7 was added to a flask containing 1gm of soil and then swirled gently to disperse the soil. 20ml of concentrated H_2SO_4 was added rapidly using a burette, the flask was swirled more vigorously for 1min and allowed to stand foe 30minutes on a white tile. At the end of this period, 100ml of distilled water was added. 3-4 drops of Ferroin indicator (0.025M) were added and titrated with 0.5M Ferrous Sulphate ($FeSO_4$). The amount of organic carbon was calculated from the titer values (Ukpaka et al., 2004a)

Contaminants transport model in soil environment

The spread of pollutants in soil is controlled by the flow of fluid in the soil environment (Ukpaka et al., 2004a). Model describing pollutant fate in the soil environment is discussed here by the convective-dispersive equation which describes the physical phenomena of mass transfer where particles are transported in a media due to convection, diffusion chemical reactions and biological transformation. The convective-

$$\frac{\partial C}{\partial t} = \nabla.(D\nabla C - vC) - R - f \quad (1b)$$

$$\frac{\partial C}{\partial t} = \nabla.(D\nabla C) - \nabla.(vC) - \frac{\partial Ca}{\partial t} - f(2a)$$

Where, **C** is the concentration, **v** is the average water velocity, **D** is the dispersion coefficient (diffusivity), **Ca** is the concentration of the adsorbed chemical contaminant and **f** is the irreversible reaction decay rate.

By expanding the equation (2) into three coordinates

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) - v\frac{\partial C}{\partial x} - \frac{\partial Ca}{\partial t} - f + \frac{\partial}{\partial y}\left(D\frac{\partial C}{\partial y}\right) - v\frac{\partial C}{\partial y} - \frac{\partial Ca}{\partial t} - f + \frac{\partial}{\partial z}\left(D\frac{\partial C}{\partial z}\right) - v\frac{\partial C}{\partial z} - \frac{\partial Ca}{\partial t} - f \quad (2b)$$

1. Assuming one-dimensional flow. Then, the y- and z- coordinates are neglected and assumed to be zero, the equation becomes:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) - v\frac{\partial C}{\partial x} - \frac{\partial Ca}{\partial t} - f \quad (3$$

The equation 1 can be rearranged as

$$\partial C/\partial t - \partial/\partial x\,(D\,\partial C/\partial x) + v\,\partial C/\partial x = -\partial Ca/\partial t - f \quad (4$$

2. Assuming that contaminant are transported at a steady state within the soil environment.

Then, equation 2 reduces to a second order differential equation

$$-D\frac{d^2C}{dx^2} + v\frac{dC}{dx} + f = 0 \quad (5a$$

Since diffusion is taking place the negative sign is insignificant to the system.

$$D\frac{d^2C}{dx^2} + v\frac{dC}{dx} + f = 0 \quad (5b$$

Equation (5b) is expressed as a quadratic equation of the form

$DX^2+vX+f=0$ (6)

Where $X^2 = \frac{d^2C}{dx^2}$ and $X = \frac{dC}{dx}$,The variable X is the depth.

The concentration is expressed as a function of depth

$C(X) = DX^2+vX+f$ (7)

dispersive equation can be derived in a straight forward way from the continuity equation which states that the rate of change of a scalar quantity in a differential control volume is given by flow and diffusion into and out of the system along with any generation or consumption inside the control volume. The Figure 2: Shows a control volume in a soil environment describing the behavior of contaminants in the soil environment. The model was developed by considering the illustration as shown in figure 1. Then, the Concentration term can be expressed by the following differential equation. The material balance expression for the system is as shown in equation (1a), Where, CAR = contaminant accumulation rate, D = diffusion, C(A) = convection (advection), AR = adsorption rate and IDR = irreversible decay rate.

$$CAR = D - C(A) - AR - IDR \quad (1a)$$

The mathematical representation of equation (1a) is given in Eqs. 1b-7.

Empirical model approach

Empirical modeling depends on the availability of representative data for model development and validation of already existing models. Its function includes approximation of unknown functional relationship by some mathematical functions based on the experimentally gather data from the physical system.

The resultant quadratic equation relates the Diffusivity (D), water Velocity (v) and the irreversible decay rate (f).

Using the least square method the experimental data are fitted to the equation (7) for all the parameters that were analyzed as:

$$C(X) = DX^2+vX+f \qquad (8)$$

$$TOC(X) = DX^2+vX+f \qquad (9)$$

$$THC(X) = DX^2+vX+f \qquad (10)$$

RESULTS AND DISCUSSION

The results obtained from the research work are presented in Tables and Figures for the various investigations conducted on the empirical model approach to the evaluation for the degree of pollutant deposition and diffusionin soil environment.

The result in Figure 1 describes the relationship between the experimental Total Hydrocarbon Content Concentration and depth. The graph shows sinusoidal relationships due to the fluctuating experimental values. The polynomial of the curve is given as $(THC)_{VM} = 0.004Dp+3.98$ and the coefficient of determination is given as $R^2= 0.037$. The theoretical developed model is given as $(THC)_{TDM} = -0.000857Dp^2+0.056Dp+3.594$.

Figure 2 illustrate the relationship between the experimental, theoretical and validated values. The relationship obtained show that at point there's interception between these values it therefore indicates that the model can be used to predict the Total Hydrocarbon Content of the soil environment at various depths.

From figure 3 the relationship between the Total Hydrocarbon Content and the Depth shows that as the increases the Total Hydrocarbon content decreases. The polynomial for this relationship was also established as $(TOC)_{VM} = -0.033Dp+8.374$ and the coefficient of determination is given as $R^2=0.8$. The theoretical developed model is given as $(TOC)_{TDM} =0.000317Dp^2-0.053Dp+8.517$.

The result presented in Figure 4 show an excellent match, indicating that the model developed is reliable. From 40cm depth it is seen that the three values are on point.

The total hydrocarbon content at the surface was low which means that diffusion is already taking place. From the result the concentration varies at different depth.

CONCLUSIONS

From the comparison of the results shown there's a reasonable level of agreement between the experimented theoretical and validated model, indicating the viability of the developed model. The concentration of the parameters analyzed can be predicted and various depths, the coefficient of diffusivity (D), the water velocity (v) and the irreversible decay rate (f) can as well be obtained from the equation developed.

The Total Carbon Content at the surface is high compared to the subsurface as well with increase in depth. Soil organic carbon is important for the function of ecosystems and agro-ecosystems having major influence on the physical structure of the soil, the soil's ability to store water and the soil's ability to form complexes with metal ions and supply nutrients. Loss of Total Carbon Content or Soil Organic Carbon can, therefore lead to a reduction in soil fertility, land degradation and even desertification. For conversion of Total Organic Carbon to Organic matter. Organic matter (%) = Total Organic Carbon Content * 1.72. this means that reduction in organic carbon result to the depletion of organic matter in soil.

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