Research Article

Modeling of the transport and diffusion phenomena for petroleum aromatic hydrocarbons from spilled crude oil in a stagnant mangrove water body

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ARTICLE INFO	ABSTRACT
	In water, oil spill constitutes serious environmental concern. When impacted water is flowing, it can be self-cleansed by waves. Turbulent water bodies have attracted huge research attention with many models available. However, it becomes different with non-turbulent and stagnant water as in the case with mangrove swamps. This paper reports the development of mathematical models for monitoring transport and diffusion phenomena of aromatics through water column using equations of transport and mass transfer. The models were validated using results from a simulated stagnant water polluted with crude oil. From the result, aromatics transported through water at 0.25 m and 1 m for month 1 is 12.988 µg/l and 0.081 µg/l for experiment and 9.215 µg/l, and 0.0781 µg/l for model, respectively. While month 5, was 48.982 µg/l and 1.890 µg/l for experiment and 39.850 µg/l and 1.650 µg/l for model, respectively. These and others reported show that the models can adequately predict the transport of aromatics through such water body. **Keywords:** crude-oil, spill, mangrove-swamp, stagnant-water, aromatics, modeling

INTRODUCTION

Over the years, there have been focused efforts on in-depth reviews as well as mathematical model development that have now improved the understanding of the transport and trajectory forecasting of oil spill in water. These intellectual strides have enhanced the proper evaluation of oil spill management practice as well as the identification of the existing knowledge gaps (Chao et al., 2001; French-McCay, 2004; Garcia-Martinez & Flores-Tover, 1999; Giwa & Jimoh, 2010; Jimoh & Alhassan, 2006; Keramea et al., 2021; Kvočka et al., 2021; Li et al., 2018; Lonin, 1999; Njobuenwu, 2014; Njobuenwu & Abowei, 2008; Olugbenga et al., 2020; Susu et al., 1997; Tkalich, 2006; Wang et al., 2005). However, the existing models did not cover oil spill pollution in all types of marine environment particularly stagnant water bodies. Also, most of the existing models are based on whole oil (not dissolved components) in rivers and seas that are characterized by high turbulence. These existing models have the capacity to better predict horizontal transport of spilled oil, weathering and the general fate of oil spill, which are dependent on the effect of waves and wind (Röhrs et al., 2018). Even in cases where these models have vertical transport trajectories components incorporated, the basis of their conception and development are mostly particulate transport occasioned by the breaking of spilled oil slick into oil particles by waves (Röhrs et al., 2019). These particles are conceptualized to be stabilized by the presence of suspended sediments, which depending on density difference with the water, begin to sink and even re-suspend (Boufadel et al., 2020; Bragg & Owens, 1995; Le Floch et al., 2002; Owens & Lee, 2003; Owens et al., 2003; Page et al., 2000).

However, when there is an oil spill in some sections of a mangrove swamp, it is expected that the transport and general fate of an oil spill will differ significantly from when such a spill happens in water with waves and wind as there are no provisions (winds and waves) for formation of oil particles because these mangrove sections are not turbulent and lack the energy usually provided by waves as in turbulent water bodies. In this case, the spill transport will be governed by the simple principle of partitioning of oil components in water instead of depending on wave and wind action to decide its fate. Therefore, existing models will be un-amenable to cases of relatively static water bodies in mangrove swamps (Odisu, 2020; Odisu et al., 2021). Unfortunately, there exist very limited literatures and research effort in this direction.

In view of the above, Imanian et al. (2017) and Odisu et al. (2021) had establish that there exists a significant gap in available information and mathematical models for oil spill in non-flowing (stagnant water in mangrove swamps) environment with marshes and relatively stagnant water bodies (saddled with a high level of suspended sediments) against the case of turbulent water bodies where much information can be easily accessed. Of note is the fact that some of these mangrove swamps house oil facilities like pipeline network and surface handling facilities and are thus prone to oil spill incidences and their attendant negative effects. The above identified knowledge gap with some degree of adaptation appropriately calls for research action.

In line with the above, Odisu et al. (2021) considered the application of the fundamental governing equations of transport and mass transfer in an hydrostatic model for the evaluation of aliphatic hydrocarbon concentration at different strata (gradient) of a stagnant water body with consideration to dissolution from the oil slick into the water-oil interface, diffusion of dissolved components through water, and adsorption of a fraction of the diffusing components by suspended sediments, which may lead to sinking and sedimentation or trapping within the water column depending on density difference. This paper focuses on the development and application of similar models using same basis for determination of aromatic hydrocarbons under same water conditions.

The aim of this research was to develop a mathematical model for the prediction of aromatic hydrocarbon concentrations at varied strata (depth) of non-flowing, non-turbulent mangrove swamp water bodies using equations of mass transfer and transport under isothermal condition. This model unlike most existing models, does not consider the spilled oil being transported to be in the particulate form arising from the effect of waves on the oil slick but as dissolving and diffusing components transported vertically. Other considerations include the effect of suspended sediments as possible carriers and support for the transport process. Thus, the process is viewed as being controlled by dissolution and diffusion of the individual hydrocarbon components due to the absence of waves leading to increased residence time and subsequent vertical movement of all dissolved and diffusing components. This concept that has enabled the prediction via mathematical modeling of petroleum aromatic hydrocarbon concentrations transport down the depth of a non-flowing water is novel in the field of oil spill modeling. Researchers will no doubt be interested in further investigation and possible refinement of the developed model using other solution pathways that could possibly yield finer and more precise results. This is because around the world, there are several of this kind of non-flowing water bodies polluted or at the risk of being polluted and some that have been polluted are in the process of being remediated like the case of the Ogoni pollution in Nigeria. Therefore, having tools like this model will surely enhance the management of such spill incidence and add value to such operations.

The reliability of the model developed was verified using a simulated oil spill site typical of a mangrove swamp with static water and monitored for 10 months. Results of aromatic hydrocarbons obtained are, as shown in the result section.

MODEL DEVELOPMENT

The general conservation equation served as the basis for the mathematical description of the transport of the aromatic hydrocarbon component from the oil bulk (slick) into the oil water interface and then into the water column. Here, the mass flux was identified as a summation of both the convective and diffusive fluxes as expressed in Eq. (1) below:

Rate of accumulation of aromatic hydrocarbons in the water column=Net inflow rate of aromatics by convection+Net inflow rate of aromatics by diffusion+Net source of aromatics by dissolution from oil bulk+Net source of aromatics by biodegradation. (1)

Upon expressing the convective and diffusive transport terms as a volume integral using Gauss' theorem, the equation can then be assembled in a single volume integral, as in Eq. (2):

$$\frac{\partial(\rho\psi)}{\delta t} + \nabla.\left((\rho v\Psi) + \nabla.J - \Phi\right) = 0, \tag{2}$$

where $\frac{\partial(\rho\psi)}{\delta t}$ represents the transient term, $\nabla.(\rho v\Psi)$ is the convective transport term, $\nabla.J$ is the diffusive transport term, and Φ is the source term.

For the sake of this work, the following assumptions were made: The pond is static (no flow induced by wave or wind action), temperature is constant (isothermal condition), there is sufficient amount of suspended particles, the diffusion coefficient is independent of concentration due to the relatively low concentration of oil components (aromatics) in the water, the diffusion constant is assumed to be independent of concentration, the concentration of the aromatic hydrocarbons is continuous along the oil-water interphase, emulsification of the oil slick is ignored due to the fact that emulsions begin to form when the required chemical conditions are met and there is sufficient energy/agitation to cause entrainment of water in oil, which is not the case in this instance (Fingas et al., 2004), mass transfer of contaminants across the water column in the stationary pond is due to unidirectional molecular diffusion in z-direction, there is biodegradation of solute contaminants in the body of the water. Rate of evaporation is also assumed to be negligible since the mangrove swamp is shielded from effect of direct sunlight by vegetation and the core focus of the research is actually petroleum aromatic hydrocarbons that will succeed in migrating vertically through the water column and not what is trapped or otherwise in the atmosphere and as such, volatile components were not considered.

Also, we assumed that the convective transport term or convective flux reduces to zero. This implies that there is no transport due to convection or bulk flow because the water body under consideration is considered to be relatively stagnant. Eq. (2) will therefore reduce to the form in Eq. (3):

$$\frac{\partial(\rho\psi)}{\delta t} + \nabla(\nabla.J - \Phi) = 0. \tag{3}$$

The source term Φ is a contribution of the dissolution of aromatics from the bulk (oil slick) and the biodegradation of the aromatic hydrocarbons by microbial activities. Also worthy of note is that a proportion of the diffusing aromatic hydrocarbons could be adsorbed by suspended sediments that are available all around the water column. Thus, the diffusive

flux will be of two parts: the diffusion of aromatics through water and the transport of aromatics by adsorption onto available suspended sediments. On incorporation of the representative mathematical expressions for these components into Eq. (3), it becomes Eq. (4):

$$\frac{\partial(\rho\psi)}{dt} = -(\nabla(\nabla.J)\text{water} + \nabla(\nabla.J)\text{S}) + (\frac{dm}{dt} - r_{aro}), \quad (4)$$

where S means **sediment**. Eq. (4) can then be expanded upon proper representation as Eq. (5):

$$\frac{\partial C_{aro}}{\partial t} = D\frac{\partial^2 C_{aro}}{\partial z^2} + k_{ad}(q^* - q_t)_{aro} + k_{dis}(c_s - c_t)_{aro} - (k_{bio-w}\,c + k_{bio-sed})_{aro}, \quad (5)$$

where $D\frac{\partial^2 C_{aro}}{\partial z^2}$ represents the diffusion through water, $k_{ad}(q^* - q_t)_{aro}$ represents the transport by sediment adsorption, $k_{dis}(c_s - c_t)_{aro}$ represents the dissolution mass transfer term, and $(k_{bio-w}\,c + k_{bio-sed})_{aro}$ represents the biodegradation term.

In accounting for the biodegradation of dissolved aromatic hydrocarbons with different biodegradability, the process was represented by a multi-substrate Monod model where multiple growth substrates are available to oil degrading microbes.

However, Javier and Susan (2013) in attempt to formulate the theoretical work first tried both multi-substrate and sole-substrate models for the degradation of soluble oil. They reported that the outcome for both sole and multiple substrate models were very close. Therefore, because the kinetic coefficients for sole-substrate models are typically more available from total petroleum hydrocarbons biodegradation measurements or oxygen uptake measurements than those for multi-substrate models, for simplicity, the sole-substrate model is used for all aromatics as a pseudo-component as shown in Eq. (6), Eq. (7), and Eq. (8):

$$r_{b=}\mu_{max}\frac{C_{aro}}{k_s+C_{aro}}B \quad (6)$$

and

$$r_{aro} = \frac{1}{Y_{aro}}r_b. \quad (7)$$

Therefore,

$$r_{aro} = \frac{1}{Y_{aro}}\mu_{max}\frac{C_{aro}}{k_s+C_{aro}}B, \quad (8)$$

where r_b is the rate of microbial growth (cells/L-h), r_{ali} and r_{O2} are the rates of degradation of aromatics (mg-oil/L-h) and oxygen consumption (mg-O$_2$/L-h), respectively, μ_{max} is the maximum rate coefficient (h^{-1}), C_{aro} is the total aromatic hydrocarbon concentration (mg/L), Ks is the half saturation constant (mg/L), B is the concentration of microbes (cells/L) in the bulk fluid, and Y_{aro} is the microbial biomass produced per mass of aromatic hydrocarbons biodegraded.

A distinguishing feature in this model will be the application of species transport. Since it is the individual aromatic hydrocarbons that diffuse through the water body, it will be good that the model is further broken down to account for the transport of each aromatic hydrocarbon species mass as a contribution to the bulk mass transport equation above.

Eq. (8) represent the final mathematical expression that was applied in the calculation of the aromatic hydrocarbon concentration down the water depth for the various time intervals.

METHOD OF SOLUTION

The general model consisting of the diffusive flux (diffusion and adsorptive mass transfer obtained) is given by Eq. (8) above, where the source term is represented by dissolution and biodegradation. To effectively solve the equation, a set of partial differential equations to represent each of these model components developed by Yin et al. (2011) was used, as shown by Eq. (9), Eq. (10), Eq. (11), and Eq. (12):

$$\frac{\partial^2 C_{aro}}{\partial z^2} = (\frac{\partial Ac}{\partial t} + \frac{\partial c}{\partial z})_{aro} = \frac{\partial}{\partial z}\left(AD_z\frac{\partial c}{\partial z}\right)_{aro} \quad (9)$$

and Eq. (10) is

$$k_{ad}(q^* - q_t)_{aro} = (\frac{\partial AC_s}{\partial t} + \frac{\partial C_s}{\partial z} + P_w\frac{w_s}{h}AC_s - P_r\frac{\in}{h}AC_s = \frac{\partial}{\partial z}\left(AD_z\frac{\partial C_s}{\partial z}\right) + K_sK_pA\Theta C_d - K_pA\Theta C_d - K_bAC_s)_{aro}. \quad (10)$$

Similarly, Eq. (11) is

$$\boldsymbol{k_{dis}(c_s - c_t)_{aro}} = (\frac{\partial AC_d}{\partial t} + \frac{\partial C_d}{\partial z} = \frac{\partial}{\partial z}\left(AD_z\frac{\partial C_d}{\partial z}\right) + K_{dis}K_PAC_{dis} - K_{bio}AC_d)_{aro}. \quad (11)$$

Recall that

$$C_{ZT} = C_w + C_{sed}, \quad (12)$$

where C_{ZT} represents the total aromatic hydrocarbon components dissolved and diffusing in water column, C_w represents the aromatic hydrocarbon components in water alone, and C_{sed} represents the aromatic hydrocarbon components in sediments alone.

With Eq. (12) representing C_{ZT}, C_w can thus be represented as follows:

$$C_{w_{aro}} = (D\frac{\partial^2 c}{\partial z^2} - k_s(q^* - q_t) + k_d(c_s - c_t) - k_{bio}c)_{aro} \quad (13)$$

and

$$C_{sed} = C_{ZT} - C_w. \quad (14)$$

Finite element method with the aid of a computational software (Matlab 2015a 8.5.0 version) was then used to solve the above set of models [Eq. (9), Eq. (10), Eq. (11), and Eq. (14)] to get the responses in concentration of aromatic hydrocarbons. Concentrations of aromatic hydrocarbons were then obtained at different depth and time. These were thereafter represented with graphs that show the model trend. The trend obtained was then compared with those from experimental data. To achieve this, parameters such as diffusivities of aromatics, average pore- size of sediments, initial concentration of the aromatics in the crude oil used for spill as determined from GC analysis, depth of spill site, concentration of suspended sediments, and volume of water from size of spill site were either measured or sourced from literatures.

The boundary condition used are in Eq. (15), Eq. (16), and Eq. (17). Initial and boundary conditions used are, as follows:

$$c_t(z, 0) = c_t\ 0 \le z \le L, \quad (15)$$

and

$$c_t(0, t) = 0\ 0 < t \le T, \quad (16)$$

Table 1. Model parameters

S/N	Parameter	Value	Source
1	Diffusivity of aromatics	5.0×10^{-6} cm²/s	https://www.itrcweb.org/petroleumvi-guidance/Content/Appendix%20C.%20 Chemistry%20of%20Petroleum.htm (default physicochemical constants for BTEXN and TPH carbon ranges)
2	Average pore-size of sediments (m×10⁻⁹)	18.40	Measured using Quantachrome's patented NOVA 4200e series
3	Initial concentration of aromatics	38,380 µg/l	Obtained from GC-MS analysis
4	Concentration of suspended sediments	0.25 g/l	Measured in laboratory by collecting water samples, filtering out, & weighing sediments
5	Volume of water from size of spill site	60 m³	Calculated volume of water using size of simulation site
6	Other constants used like $P_r \frac{\epsilon}{h}$, $K_s K_p$, etc. as found in Eq. (10) & Eq. (11)	Obtained from literature	Yin et al. (2011)

$$\frac{\partial c_t(L,T)}{\partial t} = 0 \quad 0 < t \leq T. \quad (17)$$

Table 1 is a summary of the parameters used in obtaining solution for the model. It is to be noted that a major input assumption is that the convective mass transfer term (all velocity terms) is completely reduced to zero.

RESULTS AND DISCUSSION

Eq. (5), which describes the transport and diffusion of aromatic hydrocarbons in non-turbulent/stagnant water bodies was developed and its validity (response pattern) was tested using aromatic concentration data obtained from a simulated crude oil spill. The spill site was simulated in Ugbomoro, near Warri, Nigeria by securing an area (8 m long, 5 m wide, and 1.5 m deep) containing water, shaded with mangrove trees, with high suspended sediment load from decaying organic matter and vegetation. This site is typical of a non-turbulent water body section of a mangrove swamp as found in many parts of the World because of its non-flowing nature, presence of high suspended sediments load and vegetation in and around the water body. 60 liters of crude oil obtained from a flow station near Warri, Nigeria was used to pollute the site. This volume was applied to ensure that the slick covers the entire water surface after a short period of time and to achieve slick height of between 0.1-0.2 cm.

An improvised water depth sampler was used in the collection of water and sediment samples at different depth of the water column. The sampler has a pole as its stem, which also serves as support, a sampling cup with a firm lid, a spring connecting the lid to its base and a line (trigger) connecting the lid to the head of the pole along a calibrated wooden frame (Odisu, 2020; Odisu et al., 2021). In carrying out the sampling, the improvised sampler was lowered into the water and with a firm grip on the line, it was pulled upward once the desired depth was reached and sufficient amount of water has entered the sampling can. The trigger was then released to seal the sampler before it was pulled out. The collected samples were then separated into water and sediment fractions using a vacuum filter and the samples were kept in a refrigerator prior to analysis using a gas chromatography with flame ionization detector (GC-FID). The samples were collected once every 30 days at depths of 0.25 m, 0.5 m, 0,75 m, and 1.0 m (Odisu, 2020; Odisu et al., 2021).

Figure 1, Figure 2, Figure 3, Figure 4, and Figure 5 show the experimental result matched with the model result for the transport of aromatics in water without sediment adsorption for the first five months.
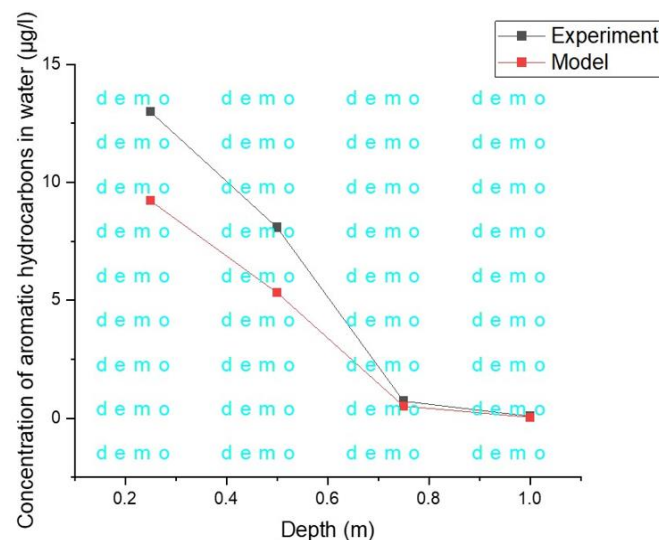

Figure 1. Aromatic hydrocarbon transport for varying depth and fixed time of one month for experiment and model (Source: Authors' own elaboration)
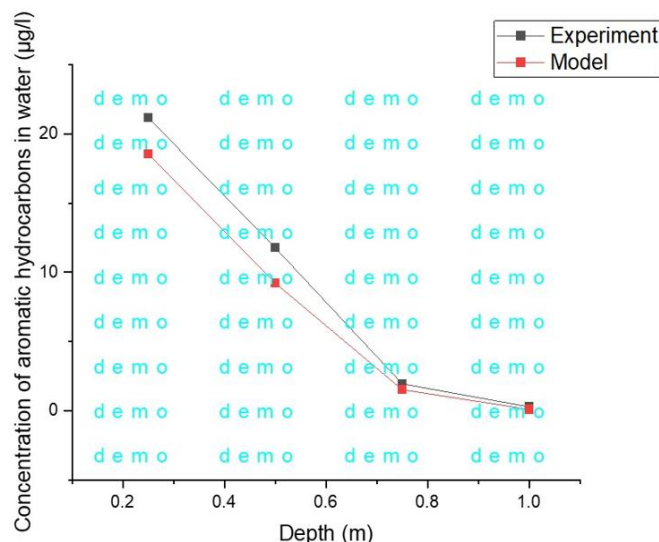

Figure 2. Aromatic hydrocarbon transport for varying depth and fixed time of two months for experiment and model (Source: Authors' own elaboration)
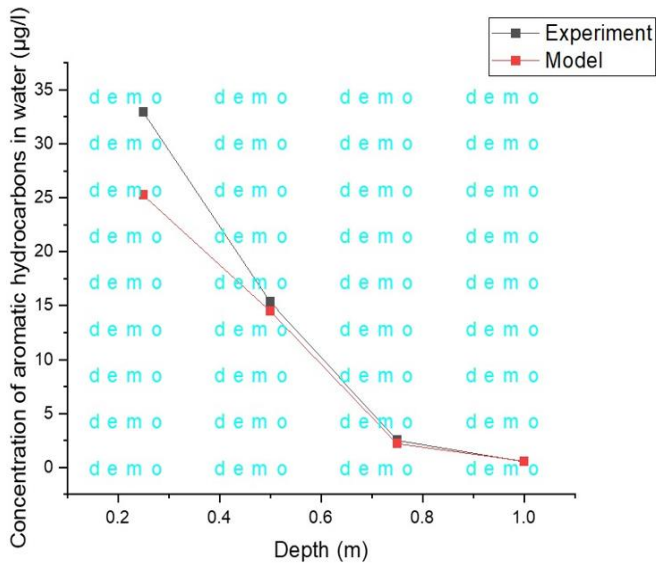
Figure 3. Aromatic hydrocarbon transport for varying depth and fixed time of three months for experiment and model (Source: Authors' own elaboration)
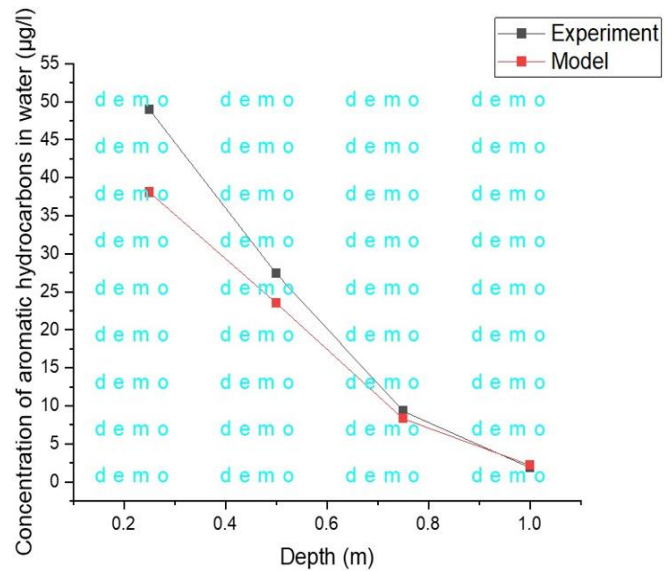

Figure 5. Aromatic hydrocarbon transport for varying depth and fixed time of five months for experiment and model (Source: Authors' own elaboration)
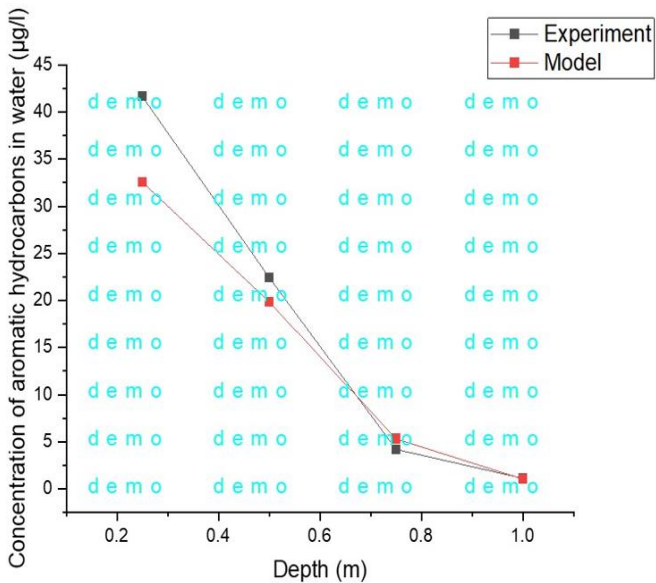

Figure 4. Aromatic hydrocarbon transport for varying depth and fixed time of four months for experiment and model (Source: Authors' own elaboration)

Figure 1, **Figure 2**, **Figure 3**, **Figure 4**, and **Figure 5** show the result of total aromatic hydrocarbon transport through the water column obtained from experiment and matched with that from model prediction at depths of 0.25 m to 1.0 m for one to five months (graphs of month 6-10 not shown). At month one and depth of 0.25 m, the values of total aromatic hydrocarbon for the predicted (model) and experiment came out relatively close. This trend follows for all other depths for month one and subsequent months where the predicted values show sufficient closeness to the experimental values. This suggests that the model developed is adequate and has the capacity to adequately predict the aromatic hydrocarbon concentration from depth to depth at different time or age of the spill down the water column.

The experimental result show that the dissolution of aromatics from the bulk did not limit the aromatic transport process as the result shows a relatively smooth dissolution process evident in a steady and observable aromatic hydrocarbon build-up at depth of 0.25 m, which is the closest depth strata to the source. However, the dissolution rate within depth of 0.25 m from the first to tenth month of study (12.988 µg/l, 21.15244 µg/l, 32.9135 µg/l, 41.6456 µg/l, 48.98 µg/l, 61.62411 µg/l, 73.399 µg/l, 89.446 µg/l, 101.918 µg/l, and 112.354 µg/l) seems relatively low when compared with that of the aliphatic hydrocarbons as reported by Odisu et al. (2021) against the hydrophilic nature of aromatics as reported by Brewer et al. (2013) and MADEP (2002b). The lower dissolution rate may be due partly to the relative availability of aromatic hydrocarbon (abundance) in the original spilled oil sample (38,380 µg/l) compared to 538,380 µg/l for aliphatics (almost fifteen times more in concentration) and the ease of dissolution controlled by the coefficient of dissolution as well as the hydrophilic nature of the aromatics (Dorothea et al., 2014). These three factors will and did determine to a large extent the concentration of aromatics available for diffusive transport down the column of water.

At subsequent depths of 0.5 m to 1.0 m, there is observed and appreciable build-up of aromatic hydrocarbon as evident from result. This suggests that for high residence time provided for by the absence of convective flow or turbulence, there is the possibility of aromatic hydrocarbon accumulation and downward transport to the waterbed (Polak & Lu,1973). Transport by diffusion obviously led to the presence of aromatic hydrocarbons down to depth of 1.0 m and most likely below.

These values are above the permissible limit of 0.2 µg/l in line with the Occupational Safety and Health Administration and the National Institute for Occupational Safety and Health standards (ATSDR, 1996). This is of serious concern because aromatic hydrocarbon availability in water bodies pose serious danger to the life therein and thus the entire interdependencies or life chain because of their very serious

toxic nature. This phenomenon makes a mangrove swamp housing non-turbulent water body and oil production facilities an endangered environment.

CONCLUSION

The relatively low research attention accorded to stagnant water bodies of the mangrove swamps have led to poor understanding of spilled oil transport phenomena and dynamics, with resultant poor management of oil spill cases in this kind of water environment. This led to the development of a predictive mathematical model (in this work) using basic transport and mass transfer equations for quantifying petroleum hydrocarbons pseudo components and here applied to aromatics hydrocarbons transported through water from depth to depth down the mangrove swamp water column. The model developed was then evaluated for fitness using data collected from a simulated mangrove spill site. The result shows that the models are adequate and sufficiently robust to predict the aromatic hydrocarbon concentration in water from depth to depth of a stagnant water column in a mangrove swamp.

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