

Groundwater Contamination and Environmental Risk Assessment of a Hydrocarbon Contaminated Site in Eastern Niger Delta, Nigeria.

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Abstract

This work evaluated existing and potential adverse impacts in a remediated hydrocarbon spill site impacted by an unquantified volume of petroleum hydrocarbon where depth to groundwater level may be less than one metre during peak rainy season. Pre-remediation concentration of Total Petroleum Hydrocarbon in soils was 30,000mg/kg to 160,000mg/kg while post remediation values were below the intervention values of 5,000mg/kg, but still higher than the target values of 50mg/kg specified for the Petroleum Industry in Nigeria. Similarly, BTEX levels reduced from 5mg/kg to less than 1mg/kg in soil and from 400µg/l to <3µg/l in groundwater. Post-remediation Primary Aromatic Hydrocarbon concentrations reduced from 700mg/kg to 5mg/kg in soil and from 10µg/l to <3µg/l in groundwater. Seasonal fluctuations of the groundwater level imprinted a hydrocarbon smear of about 3-6m within the silty and sandy soil profile. The presence of the post-remediation residual contaminants at these concentrations constitutes substantial risk to the environment.

Keywords: Contaminants, Bioscreen, BTEX, Hydrocarbon, Remediation, TPH, Niger Delta

1. INTRODUCTION

The hydrocarbon contaminated site discussed in this paper is a legacy site located in Ejama-Ebubu, Eleme in Eastern Niger Delta, Nigeria (Fig. 1). It has been studied by several authors including Amajor (1984), Mmom and Deckor (2010), Adoki (2012), Tse and Nwankwo (2013), Giadom et al (2015). The original spill occurred along a 24 inch crude oil delivery line in 1969 during the Nigerian civil war. It was accompanied by a fire which burnt for several months following the incident and it affected 255 hectares of land and the Ejama-Ebubu community. The terrain of the site is not uniform as it had several depressions of various sizes. Pockets of oil were held in these depressions and gradually percolated through the sand, silt and clay layers to the groundwater table which ranges from 6m to 9m in the source areas of the spill and intersects the ground surface on the surface as pond in the impacted site. The source of the spill source was deeply impacted to depths of about 9m to 10m. Vertical infiltration of the contaminant and the depth of impact reduced eastwards towards the clayey swampy terrain. Several remediation works mainly land farming have been attempted previously to reduce or reduce the concentration of the contaminants in the soil. During the remediation exercise of 2013, about 500bbls of free phase petroleum hydrocarbon, mainly crude oil were recovered from the site during the remediation process. Petroleum Hydrocarbons, being Light Non Aqueous Phase Liquids (LNAPLs) with specific gravities less than 1, float on the water table forming a depression. They are not entirely immiscible and thus dissolve in groundwater forming a plume of dissolved phase that migrates with groundwater to the nearest discharge point which is the pond through base flow. Seasonal fluctuation of the water table leaves a smear range of about 3-6m within the soil profile and this constitutes secondary sources of contamination. Geoenvironmental risk assessment integrating potential source hazards, migratory pathways and receptors (targets at risk) was attempted in this work to determine the significant risk concentrations of residual contaminants and the impact on human health and ecology, with emphasis on groundwater. This is because soils are limited in their ability to absorb, degrade or attenuate the effects of contaminants (Bierrens and Geerts, 2014). When this capacity is exceeded, pollutants may enter into surface or groundwater. The land use and groundwater conditions at the site determine the migratory paths of the contaminants and the eventual receptors. Knowledge of the sources of the contaminants, the pathways of migration and potential receptors is important for risk based corrective action with the overall objective to minimize associated risks when there is possibility of groundwater pollution.

2. MATERIALS AND METHODS

Remedial works carried out at the site included excavation of impacted soils to depths ranging from 2-10m below ground level at different locations within the site. The excavated soils were treated in engineered biocells to remove and dilute the contaminants and then used as backfill materials for the excavations. Pre remediation (2007), mid remediation (2012) and post remediation (2013) soil samples were obtained from augered and drilled holes situated in a gridded pattern at the site. The samples were collected at depth intervals of 1m and analysed for Total Petroleum Hydrocarbon (TPH), Polycyclic Aromatic Hydrocarbons (PAHs), Benzene-Toluene-Ethylbenzene-Xylene (BTEX). The lithologic logs of the wells were used to construct a conceptual site model. Post remediation measurements of TPH concentrations in soil samples collected at distances of 100, 150

and 200m away from the spill source in 2007, 2012 and 2013 were used to evaluate the post remediation risks subsisting at the site. Also Bioscreen software was used to produce a 3D image of petroleum hydrocarbons present at the site from the point of spill.

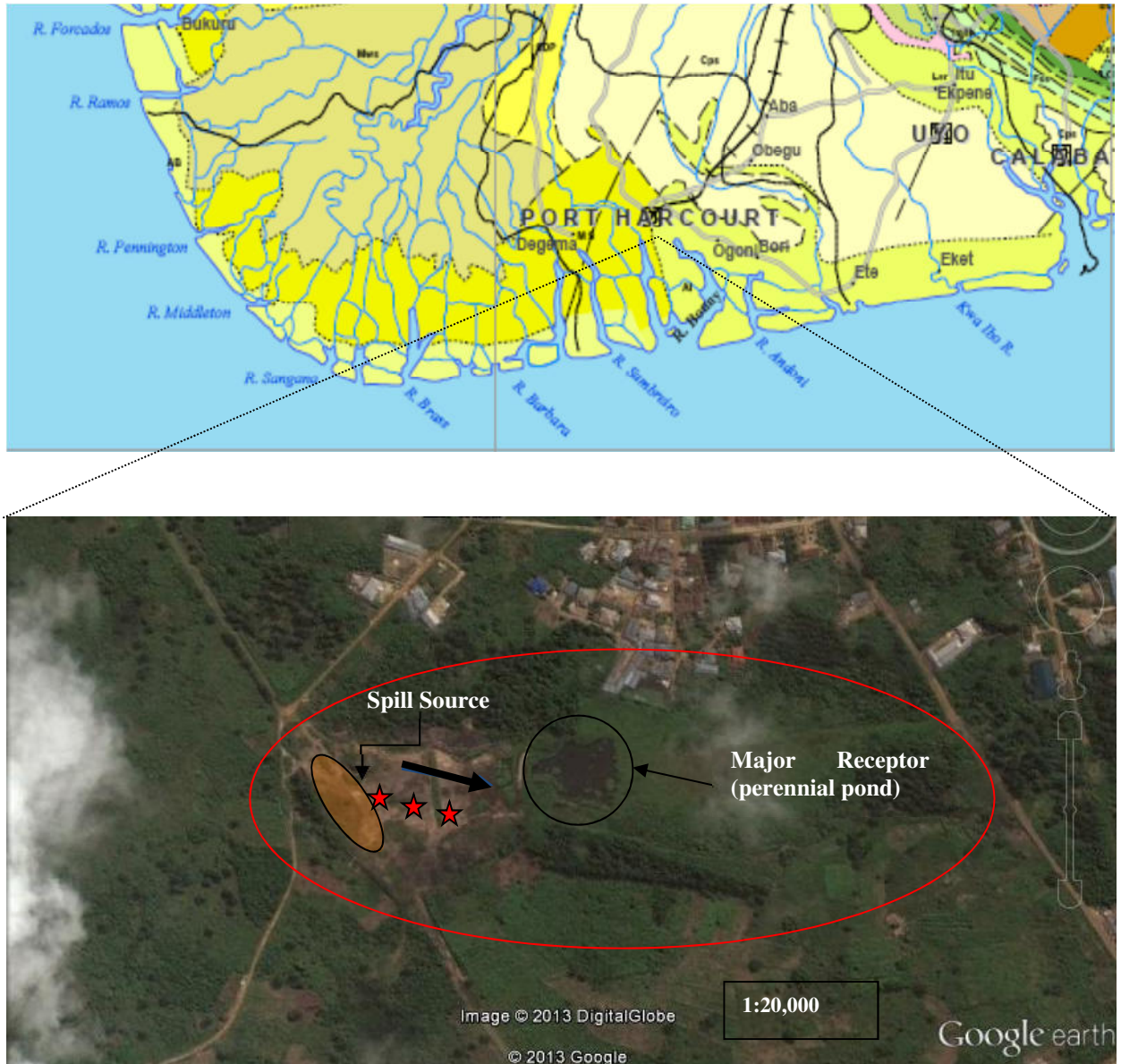


Figure 1: Geologic map of Southern Nigeria and Google imagery showing probable sphere of influence (red ellipse) of the spill at the site. Black arrow is direction of groundwater flow; stars show centerline of migrating plume.

3 RESULTS AND DISCUSSION

The conceptual site model showing lithology and stratification of soils at the site is presented in Fig. 2. Table 1 shows averages TPH concentration in soil samples collected from 0 to 4m below ground level at 3 points located midline along the contaminant plume within the sphere of influence shown in Fig. 1. There is progressive attenuation and decay of the contaminants from 2007 to 2013 shown by decreasing average TPH concentration against the distances of 100m, 150m and 250m from the spill point along a the centerline of the pipeline from which the migrating plume originates. DPR (2002) specifies that in an oil spill incidence, TPH concentration vales of 5,000mg/kg and above require intervention to reduce them to target values of 50mg/kg and below. However the present results indicate that post remediation values still exceed or are nearly equal to the intervention values, and in no case has the target values been attained. This implies presence of risks and associated hazards by exposure to theses residual contaminants.

3.1 Advection and dispersion of contaminants

The lithostratigraphy of the site shown in the conceptual site model consists of successions of silty sand layers and clay layers. This implies the hydraulic conductivity values are not uniform across the site. However, velocity of groundwater flow determined in the area is $1.403 \times 10^{-3} \text{ms}^{-1}$ or 121m/day and the dispersivity of soils in the area which increases logarithmically with increasing distances from the source of spill is about 17m (Giadom, 2013). The presence of clay in the soil profile has the capacity to limit or slow the migration of petroleum hydrocarbons farther away from the source of the spill.

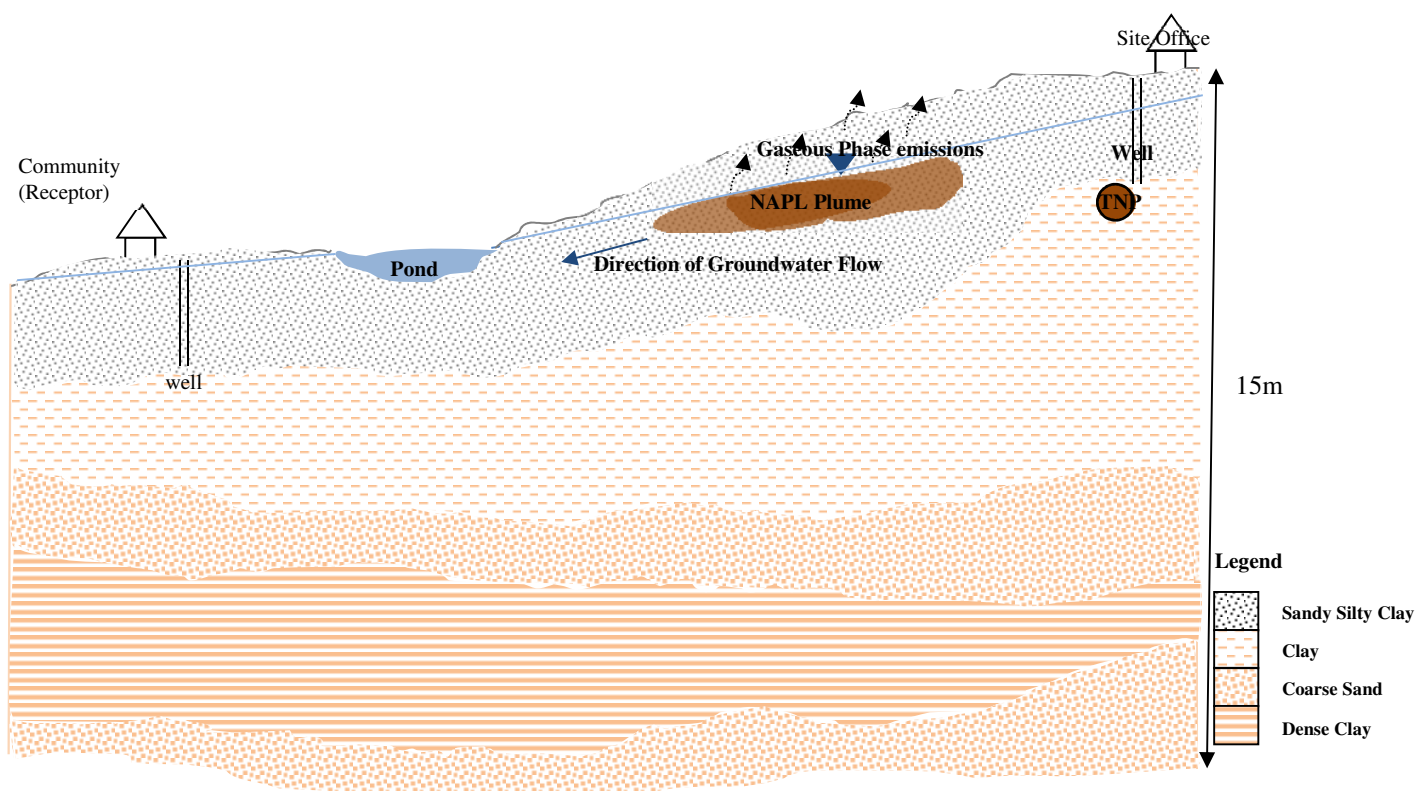


Figure 2 A conceptual site model of the spill site

Table 1 Variation by year in average TPH concentration

Sampling point	Distance from spill source	TPH concentration in soils (mg/kg)			DPR (2002) requirements	
		Year 2007	Year 2012	Year 2013	Intervention value	Target values
1	100m (328 ft)	60,000	4,000	5,000	5,000 mg/kg	50mg/kg
2	150m (492 ft)	40,000	10,000	2,000	5,000 mg/kg	50mg/kg
3	250m (820 ft)	95,000	8,000	4,000	5,000 mg/kg	50mg/kg

3.2 Advection and Dispersion on site:

The lithostratigraphy of the site shown in the conceptual site model consists of successions of silty sand layers and clay layers. This implies the hydraulic conductivity values are not uniform across the site. However, velocity of groundwater flow determined in the area is $1.403 \times 10^{-3} \text{ms}^{-1}$ or 121m/day and the dispersivity of soils in the area which increases logarithmically with increasing distances from the source of spill is about 17m (Giadom, 2013). The presence of clay in the soil profile has the capacity to limit or slow the migration of petroleum hydrocarbons farther away from the source of the spill.

3.3 Migration of contaminants on-site/off-site:

Advection is the principal process that drives the migration of crude from the contaminant source to other parts of the site. The direction of groundwater flow is south easterly. The soils in the primary source which is the spill point along the 24" pipeline and secondary sources (the smear range within the vadose zone of the soil) have been removed, treated and replaced. However residual contaminants still present in the soil are dispersed mainly through advection, and the permeability of the soils imply that these contaminants can still migrate through the soils with relative ease. Other potential exposure pathways include petroleum hydrocarbon vapour phase transport into the air surrounding the neighbourhood of the site.

3.4 Spilled oil Transport Model:

Oil phase immiscible with water will float on water. Following the rise and fall of water table in the rainy and dry seasons, the impacted water table will fall to lowest levels during peak dry seasons (about 7-9m below ground surface). This creates a smear zone, 3-6m thick. This smear zone becomes the secondary source of contamination that constantly impacts groundwater as its level fluctuates in response to seasonal regimes. The oil phase migrates by advection and dispersion in the prevailing direction of groundwater flow, and at velocities significantly lower than groundwater velocity due to its viscosity, oil-water interfacial angle, sorption, and other forces acting between the migrating oil phase, soil matrix and water surface. The dissolved phase will move downwards towards the bottom of the aquifer and flow towards the nearest discharge point at groundwater velocity. In the study area, the discharge point is the pond located to the east of the spill source.

Environmental Risk Analysis: Source-Pathway-Receptor Model:

The primary source of contamination on site was the point of spill along the 24 inch pipeline right of way. Since this has been dispersed during remediation by land farming, the entire plume of contaminants within the subsoil becomes the secondary sources of contamination. The migration pathways now are the interstitial pore spaces within the shallow aquifer, through which groundwater migrates by base flow towards the discharge point. Receptors at the site include all biota that are in contact with water from the

pond including the human community, thus creating ingestion and dermal contact exposure pathways. Fugitive emissions in the form of petroleum hydrocarbon vapours emanate from the soil which constitutes the source and escape into the air (inhalation exposure pathway), posing a risk to all biota including the human population (receptors) surrounding the site.

3.5 Transport via Groundwater and Offsite Receptors:

The site is located within the dry deltaic plains of the Niger Delta where the Benin Formation is the main aquifer. The deltaic plain is a hydrometeorological unit described by Fubara *et al* (1988), Akpokodje (1979) and Teme (2002), to be characterised by fresh water rivers, creeks and ephemeral depressions. Boreholes drilled in this zone encounter soil profiles comprising of a top lateritic clay layer which is succeeded by poorly graded sand and gravel that are of medium to high permeability and contains high yielding aquifers and layers of aquicludes. Rainfall intensity ranges between 200 and 600mm. The soils in the zone are poorly drained and are subject to flooding by high rainfall or high water stages. The Benin Formation comprises of a stacked aquifer system. The first aquifer can be encountered from 0-60m from the ground surface, while the 2nd, 3rd, 4th and 5th aquifers occur at depth ranges of 60-130m, 130-290m, 290-470m and 470m+ respectively (Etu-Efeotor and Akpokodje, 1990). Most households surrounding this site abstract water from the first aquifer which is contaminated by the spill. Abstraction of water from this aquifer creates hydraulic gradients that induce a dynamic flux of groundwater flow which influences and enhances contaminant migration onsite/offsite. This could present exposure pathway by ingestion for humans in the vicinity who consume the groundwater. It has been shown that dissolved plume of petroleum hydrocarbons can migrate 10 times the distance of the wetting front of the oil phase (Giadom *et al.* 2015, Domenico and Schwartz 1990). Moreso, fugitive hydrocarbon vapours and contaminated soil particles as aerosols can be transported by air to these off site households.

3.6 Exposure Assessment:

Risk from environmental contamination is largely determined by actual or potential exposure to contaminants that can occur through a variety of exposure or migration pathways. The potential for exposure is controlled by the physical, chemical properties and behaviour of the contaminants under the hydrogeological and geochemical conditions at a site. One critical aspect of assessing the toxic effects of TPH is the measurement of the concentration of the compounds which requires knowledge of the origin of the various fractions of TPH. Transport fractions are determined by several chemical and physical properties (i.e. solubility, vapor pressure, and propensity to bind with soil and organic particles, adsorption). These properties are the indices of measure of leachability and volatility of individual hydrocarbons and transport fractions. Although the remedial works have considerably reduced the concentration of TPH below the DPR (202) statutory intervention values of 5000mg/kg, they still exist in relatively higher concentrations above target values of 50mg/kg. Community households are less than 20m to the site and newer settlements are being developed due to population growth. These, in addition to farm lands surrounding the site increase the flux of human presence within the sphere of influence of the contaminants. The pathways of exposure include ingestion by drinking water from shallow wells, inhalation of fugitive emissions resulting from the vapour phase of the petroleum hydrocarbons and direct dermal contact with the soil. All biota within the area are thus at risk due to bio-concentration of these contaminants within their tissues. There is the

possibility that the bio-accumulated toxins will be passed from one trophic level to the next within and along the food chain.

3.7 Toxicity Assessment

Crude oil is composed of nearly 500 compounds, comprising of numerous hazardous substances (Thiergartner and Holtzmann, 2011) which are commonly found as environmental contaminants. They are nearly insoluble in water and coat soil grains asphyxiating essential soil micro-organisms leading to a cessation of soil functions and also the death of higher forms of animal life (Zabbey, 2004). Some components of the spilled oil released into the environment at the site have undergone changes during the course of over 40 years since release, but this does not eliminate their toxic effects. The chemistry of the released crude oil is altered by contaminant fate and transport processes, such as volatilization, leaching and biodegradation. Hydrocarbon compounds are broadly classified as aliphatic, alicyclic or aromatic compounds. The toxicity of aliphatic hydrocarbon compounds is a function of their carbon number/molecular weights (Deidre, 2002). The determination of non-cancer toxicity of aliphatic compounds requires the classification of the carbon number of these compounds present at the study site with a view to assigning toxicity values to each range (Table 2). The DPR (2002) maximum concentration values of 5000mg/kg of TPH stipulated for intervention in remediated sites is insufficient to assess this chemically complex contamination. It is most probable that risk to human health

and the ecosystem still persist in the remediated environment with the presence of these petroleum hydrocarbons in concentrations that fall below the intervention values but still significantly higher than the target values of 50mg/kg. Target and intervention values are not the same for all petroleum hydrocarbon contaminants therefore compounds such as BTEX which are more ecologically hazardous require more rigorous containment strategies.

Table 2: Toxicological approach for non-cancer health effects

Hydrocarbon Fraction	Reference dose (mg/kg/day)
C5-C8 Aliphatic Hydrocarbons	0.04
C9-C18 Aliphatic Hydrocarbons	0.1
C19-C36 Aliphatic Hydrocarbons	2.0
C9-C22 Aromatic Hydrocarbons	0.03

Source: www.mass.gov/dep/cleanup/laws/02-411.pdf

3.8 Risk Characterization

The characterization of risks at the site requires critical assessment of all possible toxic substances associated with petroleum hydrocarbons released into the environment. The assessment requires short and long term pathways and interactions of these substances and their chemical complexes within different components of the environment, their bioavailability, assimilation in organisms as well as the biological responses of these organisms, and damage mechanisms (endocrine disruption effects), and on their subsequent fate in the environment, food chain as well as in humans.

3.9 Specific Risk Characterization

There is still detectable smell of hydrocarbon odours at the site. This could be attributable to fugitive hydrocarbon vapours from soil gas or from hydrocarbon contaminant hotspots still present on site, and this constitute persistent onsite risk. Hydrocarbon odours are an indication that hydrocarbon compounds are present in another medium (air) other than soil or groundwater although lack of odours does not indicate

their absence. Such odours could constitute significant risk to human health, and/or a nuisance that may be considered a significant risk to public welfare. Soil gas monitoring probes would address the question of onsite hydrocarbon odours more specifically.

3.10 Exposure Point Concentration

The nature of the contaminants of concern and the sensitivity of the ecosystem in question has necessitated our consideration of specific points of exposure and pathways within the various media. In place of averaging concentrations of contaminants obtained from various points within the site, each sampling/monitoring point is treated as a separate point of exposure with its attendant risks. Hence, TPH values of more than 5000mg/kg obtained post remediation still constitute significant contaminant hotspots and pose significant risks to ecological receptors.

3.11 Modeling Dissolved Hydrocarbon Plumes at the site

Modeling dissolved hydrocarbon plumes at the site drew upon the solutions of the advection-dispersion equation. Advection describes the unidirectional displacement of contaminant masses in the prevailing direction of groundwater flow and velocity. Dispersion describes the spreading of the contaminant mass as a result of the spatial variability in aquifer properties. Advective transport is predominant on the site and this explains the migration of the plume towards the pond. However, base flow towards the pond is not unidirectional, which is evident from the spread of the plume beyond the pond towards the streamline (Fig. 3). In rationalizing the spread of contaminants, the site was discretized into cells/grids and the residual concentration of TPH was taken into consideration. Model parameters included TPH concentration over time shown in Table 1, hydraulic conductivity, hydraulic gradient, longitudinal and transverse dispersivities and porosity. The modeled area covers 1,800ft x 400ft at the site and it is assumed that biodegradation reactions (aerobic and anaerobic) occur almost instantaneously relative to the hydraulic residence time in the source area and in the plume over a time period of 6 years. To account for the effects of advective transport, three dimensional dispersion, adsorption and first order decay, the Bioscreen three dimensional analytical solute transport models was used. The model obtained (Fig.4) shows the spatial distribution of contaminants from the centerline of the plume. Elevated TPH concentration at a distance 360 feet die out to background levels at 720 feet from the source of contamination. Transverse spread of contaminants is confined to a distance of 100 feet from the centerline of the contaminant plume.

3.12 Factors Considered for the Definition of Risks at the Site.

Our definition of the risks at the study location is influenced by the potential exposure scenarios given its contiguity to human habitations and the utility of the surrounding arable lands. Other factors include climatic conditions, the ecological sensitivity of the site, local hydrological conditions, geology/hydrogeology of the subsurface with respect to the dynamics of contaminant migration. The human population transiting through, living or working within the sphere of influence of the site interact with potential health exposure pathways from the various sources of risk (which are residual petroleum hydrocarbons within the soils, groundwater and surface water), the transport mechanisms, the exposure media and pathways and the potential receptors. Contaminants are transported in a variety of ways: as wind dispersed particulates, as fugitive gases escaping as volatiles from the soil and as leachates infiltrate through the subsoil to groundwater and surface water

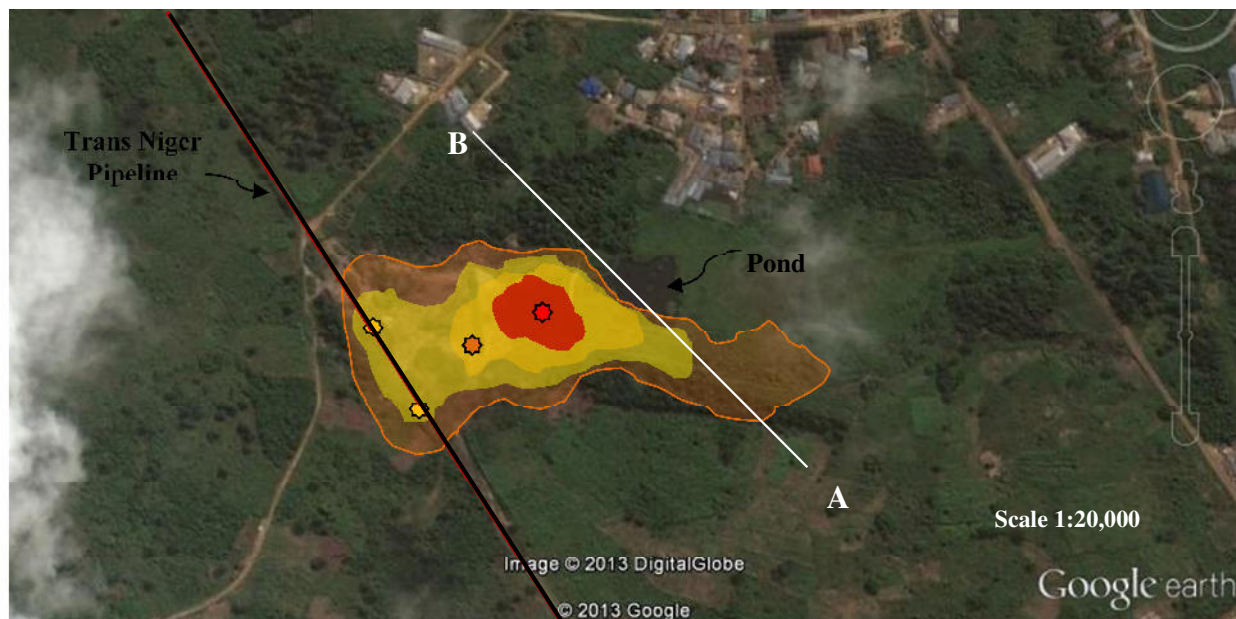


Figure 3: Configuration of contaminant plumes, the highest TPH concentration (>3000mg/kg) shown in red circle

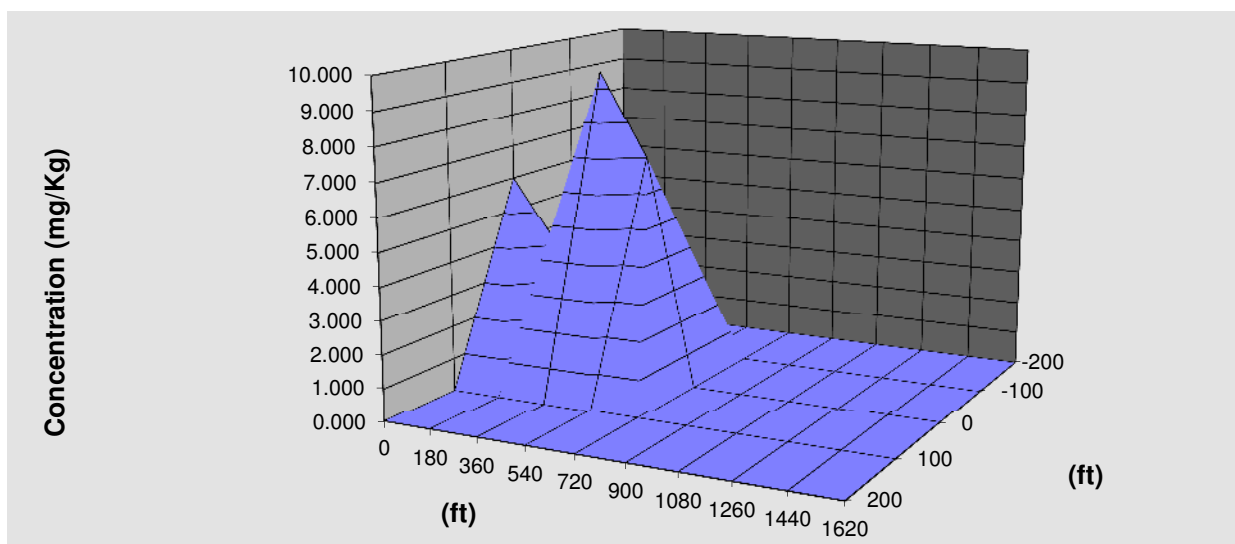


Figure 4: Distribution of TPH away from the spill source.

which flows to recharge the streams in the area. The exposure mechanism includes ingestion of contaminants bio-accumulation in plant and animal tissues (fish caught from the pond and surrounding streams), as biomagnifications along the food chain in the locality and inhalation of outdoor air. The Total Petroleum Hydrocarbon Criteria Working Group (TPHCWG) studied various samples of Nigerian crude oil and different spill sites and has established the potential human health risks associated with soils containing Nigeria crude oil. The TPHCWG used the following receptor specific parameters: lifetime, body weight, exposure duration, exposure frequency, soil ingestion rate, inhalation rate, soil to skin adherence factor and skin surface area exposed to soil. Other media and source geometric parameters used for the evaluation included lower depth of surficial soil zone, fraction of organic carbon in soil, particulate emission rate, wind speed above ground surface in outdoor air mixing zone, width of source area in major direction of wind, outdoor air mixing zone height, volumetric air content in vadose zone soils, total soil porosity, volumetric water content in vadose zone soils, soil

bulk density and averaging time for vapour flux. The receptors were onsite workers and residents close to the spill sites. The exposure pathways considered for the workers are incidental ingestion of soil, dermal contact with soil, and inhalation of soil vapour and particulate emissions. While on-duty, the workers wear full protective clothing and it is not likely that incidental ingestion and dermal contact would occur. These exposure pathways were included to take into consideration the times when workers may be off-duty and indulging in miscellaneous recreational activities unrelated to work like playing football. The soil ingestion rate was assumed to be 100mg/day, while skin surface area was 7,000 cm² with the assumption that the worker wears shorts and a short sleeved shirt when off duty but still on site. The inhalation rate of 22m³ was adopted as recommended by the World Health Organisation (McMillen *et al.* 2001). The resident is assumed to live at the site's boundary. Vapours and soil particulates are carried by the wind to habitations, therefore exposure pathways considered for the resident will be inhalation of vapour and dust emissions. When calculating risk from non-carcinogenic chemicals, the degree of risk is calculated by comparing the estimated average daily dose with a reference acceptable dose. For residential exposures to non-carcinogenic chemicals, it is standard risk assessment practice to assume that the receptor is a child. Children aged between 1-6 years typically have high inhalation and ingestion rates relative to their body weight and therefore they are usually the receptors with the greatest potential risk because they can experience the highest average daily dose (McMillen *et al.* 2001). There are some hotspots with TPH concentrations above the EGASPIN intervention values still present at the site as shown in Fig.3. These hotspots still constitute grave hazards to health and the environment and should be monitored continuously and efforts be made to further remediate them to acceptable levels. In addition, groundwater quality should be continuously monitored to ensure its continuous availability and usability.

4. Conclusion.

The characterization of the nature, magnitude and likelihood of adverse effects on human health and ecosystems from exposure to one or more residual contaminants after remediation attempts of a hydrocarbon spill site has been the focus of this work. Although the remedial works have considerably reduced the concentration of Total Petroleum Hydrocarbons below the EGASPIN statutory intervention values (5000mg/kg), there are hotspots where TPH concentrations still exist above the intervention values. The shallow aquifers at the site imply that there is possibility of groundwater contamination. Community households are contiguous (less than 20m) to the site and residents, especially children, are at risk to the hazards. The exposure pathways include dermal contact, inhalations and ingestions. It is imperative to further carry out remediation to remove or reduce the residual contaminants and restore the site for residential, agricultural and recreational uses, in addition to continuous monitoring of the various media at the site.

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