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Human health risk assessment of heavy metals from a crude oil polluted agricultural soil in Ogoniland, Nigeria

Chukwuemeka C. Chukwuma*, Chigozie Linda Onuah and Michael Okechukwu Monanu

Department of Biochemistry. Faculty of Science. University of Port Harcourt. PMB 5239 Choba. Rivers State. Nigeria. *Email: matrix4war2001@yahoo.com.

Abstract. Heavy metal contamination of agricultural soils can instigate risk to human health via oral ingestion, particle inhalation, and dermal contact. The study evaluates the concentrations, distribution, and human health risk of various heavy metals in soil samples from crude oil polluted agricultural soil of Ogoniland, Nigeria. Soil samples were collected and analyzed for cadmium, lead, chromium, copper, iron, manganese, zinc, arsenic and mercury using AAS (SensAA). Measured concentrations of these heavy metals were employed to calculate the health risk for children and adults using Hazard Index (HI). For the children and adult population, the HI value for oral ingestion to lead in the polluted site was greater than one (HI > 1), and hence non-carcinogenic effects is considered as significant for human health. It is therefore of the essence to consider taking risk management measures in order to reduce the risk of human health from lead.

Keywords: Risk assessment; Heavy metals; Polluted soil; Hazard quotient; Hazard index; Average daily intake; Carcinogenic risk; Non-carcinogenic risk.

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ORCID

0000-0002-3803-1203
Chukwuemeka C. Chukwuma

0000-0003-3373-8645
Chigozie Linda Onuah

0000-0002-8171-8895
Michael Okechukwu Monanu

Introduction

Heavy metals are elements of biological toxicity that arise naturally, and encompass high atomic weight and density, about 5 times exceeding water (Su et al., 2014). They naturally arise via weathering processes at levels < 1,000 mg/kg and could also arise from anthropogenic activities such as mining activities, pesticides usage, phosphate fertilizers, photographic materials, printing pigments, sewage irrigation, smelting, steel and electroplating industries, textiles, and dye and wood preservation (Wuana and Okieimen, 2011).They include Arsenic (As), Mercury (Hg), Lead (Pb), Cadmium (Cd), Chromium (Cr) etc. Heavy metals are deemed as trace and consequently rarely toxic and are not readily available to living organisms due to their vast adsorption capacity on soil (Ayangbenro and Babalola, 2017). Heavy metals are appraised as systemic toxicants with prospective to incite multiple organ damage even at minimal exposure levels and are classified as known carcinogens (IARC, 1993; USEPA, 2006).

There also exist other heavy metals of definite biological toxicity such as Zinc (Zn), Copper (Cu), Nickel (Ni), Selenium (Sn) and Vanadium (V) (Su et al., 2014). Heavy metals such as Cobalt (Co), Copper (Cu), Chromium (Cr), Iron (Fe), Manganese (Mn), Molybdenum (Mo), Nickel (Ni), Selenium (Se) and Zinc (Zn) have been appraised as essential nutrients requisite for several physiological and biological functions whose inadequate provision can be consequential in an array of deficiency syndromes or diseases. Other metals such as Aluminum (Al), Antimony (Sb), Barium (Ba), Beryllium (Be), Bismuth (Bi), Gallium (Ga), Germanium (Ge), Gold (Au), Indium (In), Lithium (Li), Platinum (Pt), Silver (Ag), Strontium (Sr), Tellurium (Te), Thallium (Tl), Tin (Sn), Titanium, Vanadium (V) and Uranium (U) have no entrenched biological function and consequently evaluated as non-essential (Tchounwou et al., 2012). Heavy metal contamination is hence defined as immoderate deposition of toxic heavy metals in the soil due to human activities (Su et al., 2014).

The heavy metals type, level and form depend on the sort of action that takes place at the site. New Jersey Department of Environmental Protection (NJDEP, 1996) and Department of Petroleum Resources (DPR, 2002) listed some ranges and guidelines for some heavy metals (Table 1). The DPR-EGASPIN has recommended guidelines on remediation of contaminated site based on intervention values and target values. The intervention values symbolize the quality at which the proper functionality of the soil for living organisms is threatened (Table 2). Values higher than the intervention values designate serious contaminations (Table 3), while the target values designate the required soil quality for sustainability (DPR, 2002).

Table 1. Soil concentration ranges and regulatory guideline for some heavy metals.

Metal	Soil concentration range ($mg.kg^{-1}$)*	Regulatory limits ($mg.kg^{-1}$)**
Pb	1.00-69,000	600
Cd	0.10-345	100
Cr	0.05-3,950	100
Hg	<0.01-1,800	270
Zn	150-5,000	1,500

Sources: Riley et al. (1992)*; NJDEP (1996)**.

Table 2. Target and intervention values for some metals for a standard soil.

Metal	Target value (mg.kg^{-1})	Intervention value (mg.kg^{-1})
Ni	140	720
Cu	0.3	10
Zn	n.a	n.a
Cd	100	380
Pb	35	210
As	200	625
Cr	20	240
Hg	85	530

Source: DPR (2002). n.a.: not available.

Table 3. Maximum allowable limit of concentrations of some heavy metals in soil (mg.kg^{-1}) for Different Countries.

Country	As	Pb	Hg	Cd	Cr	Cu	Zn	Co	Ni
Germany	50	70.0	0.5	1.0	60.0	40.0	150.0	n.a.	50.0
Poland	n.a.	100	n.a.	3	100	100	300	50	100
UK	32	450	10	10	130	n.a.	n.a.	n.a.	130
Australia	20	300	1	3	50	100	200	n.a.	60
Taiwan	60	300	2	5	250	200	600	n.a.	200
Bulgaria	10	26	0.03	0.4	65	34	88	20	46
Canada	20	200	0.8	3	250	150	500	n.a.	100
China	30	80	0.7	0.5	200	100	250	n.a.	50
Tanzania	1	200	2	1	100	200	150	n.a.	100
FAO/WHO Guidelines	20	100	n.a.	3	100	100	300	50	50
EU Guidelines	n.a.	300	n.a.	3	150	140	300	n.a.	75
South Africa	5.8	20	0.93	7.5	6.5	16	240	300	91

Source: Kamunda et al. (2016). n.a.: not available.

Materials and methods

Study area

The study area is a crude oil polluted agricultural farmland located in Bodo community of Ogoniland,Rivers State, Nigeria. Ogoniland has a terrific account of crude oil pollution which is mostly caused as a result of anthropogenic activities. Ogoniland covers about 1000 km^2 in South-East of Niger Delta and with populace close to 832,000 (Chukwuma et al., 2018).

Sampling and sample preparation

Soil samples were randomly collected from the polluted and unpolluted sites with steel auger at a depth of 0-15 cm. At each sampling location, five replicate samples were collected, homogenized, out of which 1kg was packaged in polyethylene bags. The collected samples were marked for easy identification and taken to the laboratory for further processing and analysis. At the laboratory, the soil samples were first spread out on a plastic sheet and allowed to air dry for 2-3 days. The samples were thereafter sieved through a 2 mm nylon mesh to obtain a homogenized sample matrix. Attention was paid to every sample to avoid cross-contamination (Kamunda et al., 2016 a,b).

Soil sample analysis

Heavy metals analyses were conducted at 60 °C by microwave digestion method as adapted by

Mwegoha and Kihampa (2010) and Rashid et al. (2016). Two and half grams of fine powdered soil was transferred to crucible and mixed with 10 mL of aqua regia comprising of HCl and HNO_3 (3:1), and further digested at 95 °C for 1 h. This was diluted to 50 mL using dH_20 after cooling and left to settle overnight, and thereafter filtered. The concentrations of Pb, Cd, Cr, Zn, Mn, Cu, Fe, As, and Hg were determined by atomic absorption spectrometry (SensAA).

Health risk evaluation

Theory of Risk Assessment. Human health risk assessment is a process used to estimate the health effects resulting from exposure to carcinogenic and non-carcinogenic chemicals (Kamunda, Mathuthu and Madhuku, 2016). The risk assessment process is made up of four basic steps: hazard identification, exposure assessment, toxicity (dose-response) assessment, and risk characterization (USEPA, 2001).

Hazard Identification basically aims to investigate chemicals that are present at any given location, their concentrations, and spatial distribution. The purpose of exposure assessment is to measure or estimate the intensity, frequency, and duration of human exposures to an environmental contaminant (Šukalić et al., 2018). Exposure assessment can be carried out by measuring the average daily intake (ADI) of heavy metals through ingestion, inhalation and dermal contact by adults and children (Kamunda et al., 2016). Adults and children are separated because of their behavioural and physiological differences (Wang, Sato and Xing, 2005).

Dose-response assessment estimates the toxicity due to exposure levels of chemicals (Table 4). The cancer slope factor (*CSF*, a carcinogen potency factor) and the reference dose (*RfD*, a non-carcinogenic threshold) are two important toxicity indices used. *RfD* values are derived from animal studies using the "No observable effect level" principle. For humans, *RfD* values are multiplied 10-fold to account for uncertainties (USEPA, 1989).

Table 4. Exposure parameters used for the health risk assessment for standard residential exposure scenario through different exposure pathways.

Parameter	Unit	Child	Adult
Body weight (*BW*)	kg	15	70
Exposure frequency (*EF*)	days/year	350	350
Exposure duration (*ED*)	years	6	30
Ingestion rate (*IR*)	mg/day	200	100
Inhalation rate (*IRair*)	m^3/day	10	20
Skin surface area (*SA*)	cm^2	2,100	5,800
Soil adherence factor (*AF*)	mg/cm^2	0.2	0.07
Dermal Absorption factor (*ABS*)	None	0.1	0.1
Dermal exposure ratio (*FE*)	None	0.61	0.61
Particulate emission factor (*PEF*)	m^3/kg	1.3×10^9	1.3×10^9
Conversion factor (*CF*)	kg/mg	10^{-6}	10^{-6}
Average time (*AT*)			
For carcinogens	days	365×70	365×70
For non-carcinogens		$365 \times$ ED	$365 \times$ ED

Source: Kamunda et al. (2016).

Risk characterization predicts the potential cancerous and non-cancerous health risk of children and adults in the study area by integrating all the information gathered to arrive at quantitative estimates of cancer risk and hazard indices (USEPA, 2004).

The potential exposure pathways for heavy metals in contaminated soils are calculated based on recommendations by several American publications. *ADI* (mg/kg-day) for the different pathways are calculated using the following exposure Equations (1) to Equations (3) as prescribed by USEPA (1989).

Ingestion of heavy metals through soil

$$ADI_{ing} = \frac{C \times IR \times EF \times ED \times CF}{BW \times AT} \qquad \text{Equation 1}$$

Where: ADI_{ing} is the average daily intake of heavy metals ingested from soil in mg/kg-day, C = concentration of heavy metal in mg/kg for soil. *IR* in mg/day is the ingestion rate, *EF* in days/year is the exposure frequency, *ED* is the exposure duration in years, *BW* is the body weight of the exposed individual in kg, *AT* is the time period over which the dose is averaged in days. *CF* is the conversion factor in kg/mg.

Inhalation of heavy metals via soil particulates

$$ADI_{inh} = \frac{C x IRair \times EF \times ED}{PEF \times BW \times AT} \qquad \text{Equation 2}$$

Where: ADI_{inh} is the average daily intake of heavy metals inhaled from soil in mg/kg-day, C_S is the concentration of heavy metal in soil in mg/kg, IR_{air} is the inhalation rate in m^3/day, *PEF*, is the particulate emission factor in m^3/kg. *EF*, *ED*, *BW* and *AT* are as defined earlier in Equation (1).

Dermal contact with soil

$$ADI_{derm} = \frac{C \times SA \times FE \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \qquad \text{Equation 3}$$

Where: ADI_{dems} is the exposure dose via dermal contact in mg/kg/day. C_S is the concentration of heavy metal in soil in mg/kg, *SA* is exposed skin area in cm^2, *FE* is the fraction of the dermal exposure ratio to soil, *AF* is the soil adherence factor in mg/cm^2, *ABS* is the fraction of the applied dose absorbed across the skin. *EF*, *ED*, *BW*, *CF* and *AT* are as defined earlier in Equation (1).

Non carcinogenic risk assessment

Non-carcinogenic hazards are characterized by a term called hazard quotient (*HQ*). HQ is the statistical term of the ratio of two variables that expresses the likelihood of an adverse effect on an individual (Šukalić et al., 2018). It is a unitless number that is expressed as the probability of an individual suffering an adverse effect. It is defined as the quotient of *ADI* or dose divided by the toxicity threshold value, which is referred to as the chronic reference dose (*RfD*) in mg/kg-day of a specific heavy metal as shown in Equation (4) (USEPA, 2004).

$$HQ = \frac{ADI}{RfD}$$ Equation 4

For *n* number of heavy metals, the non-carcinogenic effect to the population is as a result of the summation of all the *HQs* due to individual heavy metals. This is considered to be another term called the Hazard Index (*HI*) as described by USEPA document USEPA (U.S. Environmental Protection Agency, 2004). Equation (5) shows the mathematical representation of this parameter:

$$HI = \sum_{k=1}^{n} HQk = \sum_{k=1}^{n} \frac{ADIk}{RfDk}$$ Equation 5

where HQ_k, ADI_k and RfD_k are values of heavy metal *k.* If the *HI* value is less than one, the exposed population is unlikely to experience adverse health effects. If the *HI* value exceeds one, then there may be concern for potential non-carcinogenic effects (U.S. Environmental Protection Agency, 2004).

Carcinogenic risk assessment

For carcinogens, the risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen (Kamunda et al., 2016). The equation for calculating the excess lifetime cancer risk is:

$$Riskpathway = \sum_{k=1}^{n} ADIk . CSFk$$ Equation 6

where Risk is a unitless probability of an individual developing cancer over a lifetime. ADI*k* (mg/kg/day) and CSF*k* (mg/kg/day)$^{-1}$ are the average daily intake and the cancer slope factor, respectively for the kth heavy metal, for n number of heavy metals. The slope factor converts the estimated daily intake of the heavy metal averaged over a lifetime of exposure directly to incremental risk of an individual developing cancer (U.S. Environmental Protection Agency, 2004).

The total excess lifetime cancer risk for an individual is finally calculated from the average contribution of the individual heavy metals for all the pathways using the following equation:

$$Risk_{(total)} = Risk_{(ing)} + Risk_{(inh)} + Risk_{(dermal)}$$ Equation 7

where $Risk_{(ing)}$, $Risk_{(inh)}$, and $Risk_{(dermal)}$ are risks contributions through ingestion, inhalation and dermal pathways.

Both non-carcinogenic and carcinogenic risk assessment of heavy metals are calculated using *RfD* and *CSF* values derived largely from the Department of Environmental Affairs (South Africa) and USEPA as shown in Table 5.

Table 5. Reference doses (*RfD*) in (mg/kg-day) and Cancer Slope Factors (*CSF*) for the different heavy metals.

Heavy Metal	Oral *RfD*Dermal	*Dermal RfD*	Inhalation *RfD*	Oral *CSF* Dermal	*Dermal CSF*	Inhalation *CSF*
As	3.00E−04	3.00E−04	3.00E−04	1.50E+00	1.50E+00	1.50E+00
Pb	3.60E−03	-	-	8.50E−03	-	4.20E+02
Hg	3.00E−04	3.00E−04	8.60E−05	-	-	-
Cd	5.00E−04	5.00E−04	5.70E−05	-	-	6.30E+00
Cr (VI)	3.00E−03	-	3.00E−05	5.00E−01	-	4.10E+01
Co	2.00E−02	5.70E−06	5.70E−06	-	-	9.80E+00
Ni	2.00E−02	5.60E−03	-	-	-	-
Cu	3.70E−02	2.40E−02	-	-	-	-
Zn	3.00E−01	7.50E−02	-	-	-	-

Source: Kamunda et al. (2016).

Results and discussion

Concentrations of heavy metals in soils from the polluted and unpolluted site

The result presented in Table 6 showed that the mean triplicate determination of the concentration of the heavy metals from the polluted and unpolluted soil areas. For the polluted soil, the heavy metals concentrations were as follows: cadmium (6.27 mg/kg), lead (390.37 mg/kg), chromium (143.66 mg/kg), copper (12.81 mg/kg), iron (820.60 mg/kg), manganese (820.78 mg/kg), zinc (158.94 mg/kg), arsenic (3.16 mg/kg) and mercury (0.00 kg/mg); while for the unpolluted soil, the heavy metals concentrations were as follows; cadmium (3.21 mg/kg), lead (104.45 mg/kg), chromium (71.50 mg/kg), copper (1.48 mg/kg), iron (436.01 mg/kg), manganese (43.31 mg/kg), zinc (55.85 mg/kg), arsenic (0.00 mg/kg) and mercury (0.00 kg/mg). These concentrations were used to calculate average daily intake for non-carcinogenic and carcinogenic risk assessment.

Table 6. Mean Heavy metal concentration by the soil groups (mg/kg).

	Cadmium	Lead	Chromium	Copper	Iron	Manganese	Zinc	As	Hg
Polluted	6.27	390.37	143.66	12.81	820.60	820.78	158.94	3.16	0.00
Unpolluted	3.21	104.45	71.50	1.48	436.01	43.31	55.85	0.00	0.00

Non carcinogenic risk of heavy metals for children and adults

Non carcinogenic risk of adults and children were calculated based on RFD values as presented in Table 5 and ADI values in Tables 7 to 10. These results for ingestion, inhalation and dermal pathways are all presented in terms of HQs as shown in Tables 11 to 14.

Table 7. Average Daily Intake (ADI) values in mg/kg/day for adults and children in soil from the polluted site for non-carcinogenic risk assessment.

Receptor pathway		Average Daily Intake (ADI) values for heavy metals in mg/kg/day									Total
		Cd	Pb	Cr	Cu	Fe	Mn	Zn	As	Hg	
Adults	Ingestion	8.59E-06	5.35E-04	1.97E-04	1.75E-05	1.12E-03	1.11E-03	2.18E-04	4.33E-06	0.00	3.23E-03
	Inhalation	1.32E-09	8.23E-08	3.03E-08	2.70E-09	1.73E-07	1.73E-07	3.35E-08	6.66E-10	0.00	4.97E-07
	Dermal	2.13E-06	1.32E-04	4.87E-05	4.35E-06	2.78E-04	2.78E-4	5.39E-05	1.07E-06	0.00	7.99E-04
	Total	1.07E-05	6.67E-04	2.46E-04	2.19E-05	1.40E-03	1.40E-03	2.72E-04	5.40E-06	0.00	4.02E-03
Children	Ingestion	8.02E-05	5.00E-03	1.84E-03	1.63E-04	1.05E-02	1.05E-02	2.03E-03	4.04E-05	0.00	3.01E-02
	Inhalation	3.08E-09	1.92E-07	7.06E-08	6.30E-09	4.04E-07	4.04E-07	7.82E-08	1.55E-09	0.00	1.16E-06
	Dermal	1.03E-05	6.40E-04	2.35E-04	2.10E-05	1.34E-03	1.34E-03	2.60E-04	5.18E-06	0.00	3.85E-03
	Total	9.04E-05	5.63E-03	2.07E-03	1.85E-04	1.18E-02	1.18E-02	2.29E-03	4.56E-05	0.00	3.39E-02

Table 8. Average Daily Intake (ADI) values in mg/kg/day for adults and children in soil from the unpolluted site for non-carcinogenic risk assessment.

Receptor pathway		Average Daily Intake (ADI) values for heavy metals in mg/kg/day									Total
		Cd	Pb	Cr	Cu	Fe	Mn	Zn	As	Hg	
Adults	Ingestion	4.4E-06	1.43E-04	9.79E-05	2.03E-06	5.97E-04	5.93E-05	7.65E-05	0.00	0.00	9.80E-04
	Inhalation	6.77E-10	2.2E-08	1.51E-08	3.12E-10	9.19E-08	9.13E-09	1.18E-08	0.00	0.00	1.51E-07
	Dermal	1.09E-06	3.54E-05	2.43E-05	5.02E-07	1.48E-04	1.47E-05	1.89E-05	0.00	0.00	2.43E-04
	Total	5.49E-06	1.78E-04	1.22E-04	2.53E-06	7.45E-04	7.40E-05	9.54E-05	0.00	0.00	1.22E-03
Children	Ingestion	4.1E-05	1.34E-04	9.14E-04	1.89E-05	5.58E-03	5.54E-04	7.14E-04	0.00	0.00	9.15E-03
	Inhalation	1.58E-09	5.14E-08	3.52E-08	7.28E-10	2.14E-07	2.13E-08	2.75E-08	0.00	0.00	3.52E-07
	Dermal	5.26E-06	1.710E-04	1.17E-04	2.42E-06	7.14E-04	7.09E-05	9.15E-05	0.00	0.00	1.17E-03
	Total	4.63E-05	1.51E-03	1.03E-03	2.13E-05	6.29E-03	6.25E-04	8.06E-04	0.00	0.00	1.03E-02

Table 9. Average Daily Intake (ADI) values in mg/kg/day for adults and children in soil from the polluted site for carcinogenic risk assessment.

Receptor pathway		Average Daily Intake (ADI) values for heavy metals in mg/kg/day									Total
		Cd	Pb	Cr	Cu	Fe	Mn	Zn	As	Hg	
Adults	Ingestion	3.68E-05	2.29E-03	8.43E-04	7.52E-05	4.82E-03	4.82E-03	9.33E-04	1.86E-05	0.00	1.38E-02
	Inhalation	5.66E-10	3.53E-08	1.30E-08	1.16E-09	7.41E-08	7.41E-08	1.44E-08	2.85E-10	0.00	2.13E-07
	Dermal	9.12E-07	5.68E-05	2.09E-05	1.86E-06	1.19E-04	1.19E-04	2.31E-05	4.59E-07	0.00	3.42E-04
	Total	3.77E-05	2.35E-03	8.64E-04	7.71E-05	4.94E-03	4.94E-03	9.56E-04	1.91E-05	0.00	1.42E-02
Children	Ingestion	6.87E-06	4.28E-04	1.57E-04	1.40E-05	9.00E-04	9.00E-04	1.74E-04	3.46E-06	0.00	2.58E-03
	Inhalation	2.64E-10	1.65E-08	6.06E-09	5.40E-10	3.46E-08	3.46E-08	6.70E-09	1.33E-10	0.00	9.92E-08
	Dermal	8.80E-07	5.48E-05	2.02E-05	1.80E-06	1.15E-04	1.15E-04	2.23E-05	4.44E-07	0.00	3.30E-04
	Total	7.75E-06	4.83E-04	1.78E-04	1.58E-05	1.01E-03	1.01E-03	1.97E-04	3.90E-06	0.00	2.91E-03

Table 10. Average Daily Intake (ADI) values in mg/kg/day for adults and children in soil from the unpolluted site for carcinogenic risk assessment.

Receptor pathway		Average Daily Intake (ADI) values for Heavy Metals in mg/kg/day									Total
		Cd	Pb	Cr	Cu	Fe	Mn	Zn	As	Hg	
Adults	Ingestion	1.88E-05	6.13E-04	4.20E-04	8.69E-06	2.56E-03	2.54E-04	3.28E-04	0.00	0.00	4.20E-03
	Inhalation	2.9E-10	9.43E-09	6.46E-09	1.34E-10	3.94E-08	3.91E-09	5.04E-09	0.00	0.00	6.47E-08
	Dermal	4.67E-07	1.52E-05	1.04E-05	2.15E-07	6.34E-05	6.3E-06	8.12E-06	0.00	0.00	1.04E-04
	Total	1.93E-05	6.28E-04	4.30E-04	8.91E-06	2.62E-03	2.60E-04	3.36E-04	0.00	0.00	4.31E-03
Children	Ingestion	3.52E-06	1.14E-04	7.84E-05	1.62E-06	4.78E-04	4.75E-05	6.12E-05	0.00	0.00	7.84E-04
	Inhalation	1.35E-10	4.4E-09	3.01E-09	6.24E-11	1.84E-08	1.83E-09	2.35E-09	0.00	0.00	3.02E-08
	Dermal	4.51E-07	1.47E-05	1.00E-05	2.08E-07	6.12E-05	6.08E-06	7.84E-06	0.00	0.00	1.00E-04
	Total	3.97E-06	1.29E-04	8.84E-05	1.83E-06	5.39E-04	5.36E-05	6.90E-05	0.00	0.00	8.85E-04

Table 11. Hazard quotients for the ingestion pathway of children and adults population in soil from the polluted and unpolluted site for non-carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	**Unpolluted**	**Polluted**	**Unpolluted**
Cadmium	1.60E-01	8.21E-02	1.17E-02	8.80E-03
Lead	1.39E+00	3.71E-01	1.49E-01	3.97E-02
Chromium	6.12E-01	3.05E-01	6.56E-02	3.26E-02
Copper	4.44E-03	5.11E-04	4.47E-04	5.48E-05
Iron	n.a	n.a	n.a	n.a
Manganese	n.a	n.a	n.a	n.a
Zinc	6.77E-03	2.38E-03	7.26E-04	2.55E-04
Arsenic	1.35E-01	0.00	1.44E-02	0.00
Mercury	0.00	0.00	0.00	0.00

n.a = not available.

Table 12. Hazard quotients for the inhalation pathway of children and adults population in soil from the polluted and unpolluted site for non-carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	**Unpolluted**	**Polluted**	**Unpolluted**
Cadmium	5.41E-05	2.77E-05	2.32E-05	1.19E-05
Lead	n.a	n.a	n.a	n.a
Chromium	2.35E-03	1.17E-03	1.01E-03	5.02E-04
Copper	n.a	n.a	n.a	n.a
Iron	n.a	n.a	n.a	n.a
Manganese	n.a	n.a	n.a	n.a
Zinc	n.a	n.a	n.a	n.a
Arsenic	5.18E-06	0.00	2.22E-06	0.00
Mercury	0.00	0.00	0.00	0.00

n.a = not available.

Table 13. Hazard quotients for the dermal pathway of children and adults population in soil from the polluted and unpolluted site for non-carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	**Unpolluted**	**Polluted**	**Unpolluted**
Cadmium	2.05E-03	1.05E-03	4.25E-04	2.18E-04
Lead	n.a	n.a	n.a	n.a
Chromium	n.a	n.a	n.a	n.a
Copper	8.74E-04	1.01E-04	1.81E-04	2.09E-05
Iron	n.a	n.a	n.a	n.a
Manganese	n.a	n.a	n.a	n.a
Zinc	3.47E-04	1.22E-03	7.19E--04	2.53E-04
Arsenic	3.45E-03	0.00	3.57E-03	0.00
Mercury	0.00	0.00	0.00	0.00

n.a = not available.

Table 14. Hazard Index for the Children and Adults Population in soil from the polluted and unpolluted site for non-carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	Unpolluted	Polluted	Unpolluted
Cadmium	1.62E-01	8.32E-02	1.76E-02	9.02E-03
Lead	1.39E+00	3.71E-01	1.49E-01	3.97E-02
Chromium	6.15E-01	3.06E-01	6.67E-02	3.32E-02
Copper	6.13E-01	3.05E-01	6.58E-02	3.27E-02
Iron	n.a	n.a	n.a	n.a
Manganese	n.a	n.a	n.a	n.a
Zinc	1.02E-02	3.60E-03	1.44E-03	5.08E-04
Arsenic	1.52E-01	0.00	1.800E-02	0.00
Mercury	0.00	0.00	0.00	0.00

n.a = not available.

When HQ and HI values are less than 1, the population will not have any obvious risk, but if the values exceed one, this may lead to concern for potential non carcinogenic effects (USEPA, 1989; Kamunda et al., 2016; Šukalić et al., 2018). For both the children and adult population, calculated values of HQ were less than one in injection, inhalation and dermal pathways with the exception of lead which had HQ value of 1.39 for the ingestion pathway of children population exposed to the polluted soil site. The HI values for adults and children population exposed to the polluted and unpolluted sites were below one, indicating no risk to human health due to oral, Inhaled and dermal exposures investigated with heavy metals from the soil, except for Pb for children exposed to the polluted sites with HI value > 1. This measured HI value > 1 for Pb in children for oral intake shows a potential risk to human health. Luo et al. (2012) expressed concern for the non-cancerous risk of oral Pb for children; albeit the value of HI was lower than 1. The potential non-cancerous risk for adults' and children's health was explored by Oluseye et al. (2013) due to exposure to heavy metals in Nigeria in 2013. The risk assessment results indicated that the greatest risks to adults' and children's health is mainly related to Pb. Likewise, for the children population, the total HI from the polluted site was 1.55 while for the unpolluted, the total HI was 6.97×10^{-1}. For the adult population, the total HI for the polluted site was 1.69×10^{-1} while for the unpolluted site, the total HI was 7.54×10^{-2}. Overall, the total HI for both adults and children exposed to the polluted site was 1.72 while for the unpolluted, the total HI for both adult and children population exposed to the unpolluted site was 7.77×10^{-1}.

This high value expunged in the polluted soil site indicated heavy metal pollution that may pose a very high non-cancer health risk to children living around the area. The results also indicated that, in both adults and children exposed to the polluted and unpolluted sites, the oral pathway contributes the greatest to non-carcinogenic risk followed by the inhalation pathway. Dermal is the least contributor to the risk.

Carcinogenic risk assessment of heavy metals for adults and children

The excess lifetime cancer risks for adults and children are calculated separately from the average contribution of the individual heavy metals in soil for all the pathways using Equations (6) and (7). Based on the carcinogenic risk values of the calculated *ADI* values presented in Tables 9 and 10, the results of the excess lifetime cancer risks are presented in Tables 15 and 18.

Table 15. Risk Pathway for the ingestion pathway of children and adults population in soil from the polluted and unpolluted site for carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	Unpolluted	Polluted	Unpolluted
Cadmium	n.a	n.a	n.a	n.a
Lead	3.64E-06	9.73E-07	1.95E-05	5.21E-06
Chromium	7.87E-05	3.92E-05	4.22E-04	2.10E-04
Arsenic	2.31E-06	0.00	1.24E-05	0.00

n.a = not available.

Table 16. Risk pathway for the inhalation pathway of children and adults population in soil from the polluted and unpolluted site for carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	Unpolluted	Polluted	Unpolluted
Cadmium	1.66E-09	8.52E-10	3.57E-09	1.83E-09
Lead	6.91E-06	1.85E-06	1.48E-05	3.96E-06
Chromium	2.48E-07	1.24E-07	5.32E-07	2.65E-07
Arsenic	8.88E-11	0.00	1.90E-10	0.00

n.a = not available.

Table 17. Risk pathway for the dermal pathway of children and adults population in soil from the polluted and unpolluted site for carcinogenic risk assessment.

Metals/Groups	children		adults	
	polluted	unpolluted	polluted	unpolluted
Cadmium	n.a	n.a	n.a	n.a
Lead	n.a	n.a	n.a	n.a
Chromium	n.a	n.a	n.a	n.a
Arsenic	2.96E-07	0.00	3.06E-07	0.00

n.a = not available.

Table 18. Risk total for the children and adults population in soil from the polluted and unpolluted site for carcinogenic risk assessment.

Metals/Groups	Children		Adults	
	Polluted	Unpolluted	Polluted	Unpolluted
Cadmium	1.66E-09	8.52E-10	3.57E-09	1.83E-09
Lead	1.05E-05	2.82E-06	3.43E-05	9.17E-06
Chromium	7.90E-05	3.93E-05	4.22E-04	2.10E-04
Arsenic	2.60E-06	0.00	1.27E-05	0.00

n.a = not available.

The carcinogenic risk was calculated based on Pb, Cd Cr and As. The US Environmental Protection Agency considers acceptable for regulatory purposes a cancer risk in the range of 1×10^{-6} to 1×10^{-4} (USEPA, 2004). On the other hand, South Africa, considers the Individual cancer risk limit to be 5×10^{-6} (GSA, 2006). The cancer risk for adults and children in the polluted area was found to be 4.69×10^{-4} and 9.21×10^{-5}, respectively while the cancer risk for adults in the unpolluted area was found to be 2.19×10^{-4} and 4.21×10^{-5} for children. Overall, the total risk for both adults and children exposed to the polluted site was 5.61×10^{-4} while for the unpolluted, the total risk for both adult and children population exposed to the unpolluted site was 2.61×10^{-4}. In the study area, adults are therefore more at risk than children. The ingestion route seems to be the major contributor to excess lifetime cancer risk followed by the dermal pathway.

Conclusion

Based on the results obtained in the soil from the crude oil polluted sites, the heavy metals varied significantly and decreased in the order of Mn > Fe > Pb > Zn > Cr > Cu > Cd > As > Hg. Compared with recommended maximum permissible limits from FAO/WHO, EU and South Africa (SA), concentration of Pb in the polluted site was found to be the higher while concentrations of Cu and Zn were found to be lower. Cr was found to be greater than the limit depicted by FAO/WHO and SA but lower than the EU limit. Likewise, Cd content was above FAO/WHO and EU limits but lower than SA limit As and Hg concentrations were lower compared with all the three recommended permissible limits. The results also indicated that, in both adults and children, the oral pathway was the greatest contributor to carcinogenic and non-carcinogenic risk followed by the inhalation pathway. The dermal pathway was the least contributor to cancer and non-cancer risk. Based on the results obtained in this study, it can be concluded that soils in the polluted site are seriously polluted by heavy metals. There is critical need to put in place regulations to protect residents from heavy metal pollution in the environment and subsequent remediation of the polluted soil.

Conflict of interest

The authors declare that they have no conflict of interest.

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