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Human Soil Ingestion Exposure Assessment of Selected Toxic Metals for Soils Contaminated By Auto-Mechanic Spills, Crude Oil and Mining Wastes

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***Abstract**: Heavy metals constitute part the contaminants released to soils during auto-mechanic service activities, crude oil exploration, production and transportation, and lead mining. Communities hosting industries usually highlight the companies operating around them as the ones associated with highest risk to the local population. This study was designed to compare the risks resulting from soils contaminated by toxic metals from crude oil, auto-mechanic spills and lead mining. Nine (9) archive composite surface soil samples previously collected from cities and towns in Rivers, Bayelsa and Ebonyi state impacted by crude oil, auto-mechanic spills and lead mining wastes were used for the investigation. Dried soil samples (<250um) were digested with aqua regia and the resulting digests analysed for As, Cd, Mn, Pb and Zn. Human soil ingestion exposure assessment was conducted using pseudo-total concentrations of the individual metals. The pseudo-total concentration ranges for As, Cd, Mn, Pb and Zn in all three different contaminated matrices were 24.9 – 55.7 mg/kg, 7.4 – 18.4 mg/kg, 400 – 4938 mg/kg, 77 – 7451 mg/kg, and 109 – 2220 mg/kg, respectively. Among the three different contaminated matrices investigated the mine wastes contaminated surface soils indicated the highest mean pseudo-total concentrations for all five toxic metals of interest in this study. Calculation of potential daily intake doses of As and Pb from topsoils around mining sites indicated levels above TDI values set for the toxic metals. This study highlights relatively low levels of toxic metals in surface soils contaminated by crude oil. Data obtained from this study suggest that of the three industries investigated Pb mining indicated the highest concentration of all the toxic metals investigated in surrounding surface soils. This study has also highlighted auto-mechanic service operations as a potential point source of toxic metals, especially Pb.*

***Keywords**: Crude oil, auto-mechanic spill, lead mining, Toxic metals*

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I. Introduction

Governments all of over the world requires robust economies to meet the needs of their population. Nations are presently engaged in varying industrialization schemes to drive economic growth. Most of the industrial schemes have been reported to impact negatively on surrounding environments. Examples of such industrial schemes include the exploration for crude oil and mining of precious metals like lead. Also populations of nations with good economies use modern tools like motor vehicles, and these motor vehicles are serviced in developing countries without standard operating procedures that can guarantee minimal contamination of the environment. Heavy metals constitute part the contaminants released to soils during auto-mechanic service activities, crude oil exploration, production and transportation, and lead mining. Soils at different industrial sites may have distinct group of heavy metals contaminants due to their respective raw materials and products (Alloway, 2012). Several opinions have been aired and published by environmental experts and activists highlighting either lead mining and crude oil polluted soils as indicating higher risk to human subjects and the ecosystem. Risks associated with soils contaminated by auto-mechanic spills are presently grossly under reported. This study was designed to compare the risks resulting from soils contaminated by crude oil, auto-mechanic spills and lead mining.

More than 50% of environmental pollutions in Niger Delta region of Nigeria are from oil spillage (Baird, 2010). An approximate of 2300m^3 of oil are spill in more than 300 separate incidents in Niger Delta region of Nigeria annually (Ikporukpo and Omoboriowo 2015). Nigeria which happens to be one of the major exporters of crude oil has experienced several oil spills. Detrimental effects of oil spillage to soil have been reported by several authors who have adverse effect on agricultural activities and soil dependant organisms Rowell, (2014) and Baker (1970). Environmental problems resulting from mining activities have been highlighted in several scientific publications (Hobbs et al., 2008; Boisa et al 2013). The mining of ore bodies for pure metals has brought metals into close contact with human subjects. Likely sources of environmental redistribution of metals from the mining include mine water, particulate emissions, and mine tailings (Nriagu and Pacyna, 1988; Senesi *et al*., 1999; Boisa et al 2013). Surrounding soils serves as final rest place for the released metals (Nygard *et al*., 2012). This study looked at the distribution of metals near a mine site in Nigeria.

Presently, the impact of the auto-mechanic activities is usually not investigated and remediated; consequently people in contact with sites previously used for auto-mechanic services may be exposed to surface soil-associated potentially toxic elements. Disposal of spent oil into the surrounding may induce changes in geochemistry of soils because of additives and metal wear contaminants in used engine (Cachada et al., 2013).

Human exposure to crude oil, motor garage chemicals and mine tailing wastes contaminated soils have been suggested to induce deleterious effects (Oktem et al., 2004; 2011; Weuve et al., 2013;). However, a knowledge gap presently exist concerning the human health risk ranking of sites impacted by different industrial activities like crude oil exploration and production, metals mining and motor mechanic servicing. The aim of this study was to compare the distribution of As, Cd, Pb Mn and Zn in soils impacted by crude oil, motor garage chemicals and mine tailing wastes and their associated human health risk.

II. Materials And Methods

Soil sampling sites

Nine (9) composite soil samples were collected from cities and towns in Rivers, Bayelsa and Ebonyi state Nigeria. Three (3) soil samples were obtained from each of the location. Crude oil polluted soil samples were collected from sites at Mbikiba (MSS) and Onyeregbene (OSS), Bayelsa and Ererekiri Andoni (ESS), Rivers state. Soil samples from auto-mechanic workshop were also collected from three different locations around Port-Harcourt. These locations include Metals scrap at Oyigbo (MSO), Battery workshop near Oyigbo (AAS) and auto-Mechanic workshop near Community Secondary School, Ikoku Orubum (CSS); all in Port-Harcourt, Rivers State. Soil samples from lead mining site were also collected from three different locations around the artisanal mine site at Ishiagu village in Ebonyi State. The locations include the house of local dealer (HLD), roadside close to Mine Site (RCM) and from Mine Site(MS).

Samples collection

Soil samples were collected at each site at depth of 0-15cm. At least three composite soil samples were taken from each of the locations using stainless trowel and kept in a well labeled polyethylene bag. All the samples were transported to the Chemistry Department laboratory, Rivers State University of Science and Technology Port-Harcourt.

Heavy metal analysis

The soil samples were on a clean plastic sheet kept on a flat table and air-dried for some days in the laboratory under room condition. The dried soil samples were later sieved using <250um stainless mesh screen. Sieve samples were kept in a well clean glass bottles. 0.5g samples were taken from sieve soils into burning test tubes 20ml of hydrochloric acid and nitric acid (aqua riga) ratio 3:1 were added to each samples. The samples were digested for 1 hour at 100^0C by making use of hot plate and heated intermittently to ensure steady temperature. The digestate were then remove from the fume cupboard and allowed to cool to room temperature. The cooled digested were then filtered into the 100ml standard flasks and were made up to the mark with deionized water. Arsenic, cadmium, iron, manganese and zinc were analysed using Atomic Absorption Spectrophotometer (AAS) after calibration with standard reference solutions. Soil sample pH was determined with an electronic glass electrode pH meter.

Human Soil Ingestion Exposure Assessment

The samples investigated in this study irrespective of their matrix were all from residential areas or locations very close to human residence. Therefore, the estimation of exposure assessment doses is relevant. With total concentrations, the potential human exposure risk resulting from the ingestion soil-associated toxic metals from the different matrices can be quantified. The method employed here is based on calculation of the toxic metal daily intake (TM DI) from incidental ingestion of topsoils expressed on the basis of mechanic spills, crude oil and mining wastes. The TM DI is calculated for children between 2-6 years given their known hand-to-mouth habits and sensitivity to metal uptake:

TM DI = (EC x SIR x ED)/BW

Where: EC is the TM concentration (mg/kg), SIR is the soil ingestion rate (mg/day), set at 100 mg/day for 2-6 year old children (USEPA, 2011), ED is the exposure duration (set at 1, since it is assumed that children are likely to spend the whole day at home for full year), and BW is the body weight (set at 17.8 kg for age group of interest in this study).

Experimental quality Control

Analytical accuracy of aqua regia extractions were assessed against a reference material, BCR 143R (aqua regia certified Sewage sludge amended soil). All digestions for quality assurances was repeated five times. Good results were obtained for total Cd, Pb and Zn. Cd concentration measured was 69.0 ± 1.9 mg/kg compared to the certified value of 72.0 ± 1.8 mg/kg. Pb concentration measured was 187 ± 9.0 mg/kg compared to the certified value of 174 ± 5.0 mg/kg. Mn concentration measured was 834 ± 18 mg/kg compared to the certified value of 858 ± 11 mg/kg. Zn concentration measured was 1048 ± 39 mg/kg compared to the certified value of 1063 ± 16 mg/kg. There was no listed value for As. AAS was calibrated using at least 6 Pb standards (AccuTrace Reference Standard – AA29N-1) obtained from AccuStandard, Inc, USA. The instrument was re-calibrated after not more than 10 samples.

III. Results And Discussion

Aqua Regia-Soluble Concentrations of Selected Metals

The results for aqua-regia soluble As, Cd, Mn, Pb and Zn investigated are provided in Table 1. The mean pseudo-total concentration range for As, Cd, Mn, Pb and Zn in all three different contaminated matrices were 24.9 – 55.7 mg/kg, 7.4 – 18.4 mg/kg, 400 – 4938 mg/kg, 77 – 7451 mg/kg, and 109 – 2220 mg/kg, respectively (Table 1). The order of maximum pseudo-total concentration is Cd < As < Zn < Mn < Pb.

Auto-mechanic spill contaminated surface soils

The mean concentrations for As, Cd, Mn, Pb and Zn in the three composite samples collected from auto-mechanic service sites were 25.0 mg/kg, 7.7 mg/kg, 401 mg/kg, 398 mg/kg and 1983 mg/kg, respectively. For the auto-mechanic spill contaminated soils Zn mean concentration (1883 mg/kg) exceeded the Dutch intervention value (720 mg/kg) and the Italian soil intervention value (150 mg/kg) for residential land use scenarios (Carlon, 2007). However, only one of the samples (MSO) collected from a site used for car body works indicated a concentration (5278 mg/kg) that exceeded the intervention values. The very large concentration of Zn from the car body works site may have resulted from fine metal dust generated during grinding and polishing processes. The mean concentration (398 mg/kg) for Pb is below the UK soil guide value (450 mg/kg) (Environment Agency, 2002), but two of the composite samples (MSO and AAS) concentrations (507 mg/kg and 473 mg/kg) were above UK soil guideline value. The MSO sample was collected from a site used for car engine servicing and car body works associated with large metal scrape dump, while the AAS sample was collected from a site used for car engine servicing and car battery servicing.

Crude oil contaminated surface soils

The mean concentrations for As, Cd, Mn, Pb and Zn in the three composite samples were 37.8 mg/kg, 7.4 mg/kg, 404 mg/kg, 77 mg/kg and 109 mg/kg, respectively. For the crude oil contaminated soils only As mean concentration (37.8 mg/kg) exceeded the UK soil guideline value (32 mg/kg dry weight) (Environment Agency, 2009b) for residential land use scenario. Duker et al 2005 have suggested alluvial and deltaic environments similar the ones were we obtained these samples favour the accumulation of As. The relatively low concentrations of Cd, Mn, Pb and Zn observed in this study are consistent concentrations of metals reported by Gondal et al., (2006) for Arabian crude residue. The presence of As, Cd, Mn, Pb and Zn in surface soils/sediments may have been sourced from the crude oil because these metals have previously been identified and quantified in crude oil samples by Pereira et al., (2010) though at low concentrations. Pacya and Pacya (2001) from their study of global emission of cadmium to the atmosphere estimated about 23 % contribution from fossil fuel combustion.

Mine wastes contaminated surface soils

The mean concentrations for As, Cd, Mn, Pb and Zn in the three composite samples collected from mine wastes contaminated sites were 55.7 mg/kg, 18.4 mg/kg, 4938 mg/kg, 7384 mg/kg and 2220 mg/kg, respectively. The mean concentrations of As, exceed the UK soil guideline value (32 mg/kg dry weight) (Environment Agency, 2009b) for residential land use scenario. The mean concentrations of Cd exceed the UK soil guideline value (10 mg/kg dry weight) (Environment Agency, 2009b) for residential land use scenario. Pacya and Pacya (2001) from their study of global emission of cadmium to the atmosphere estimated about 73 % contribution from non-ferrous metal production. ATSDR Mn criteria value (3,000 mg/kg) (Kabata-Pendias, 2000) set for children is exceeded by the mean value (4938 mg/kg) obtained in this study. The mean concentration (7384 mg/kg) for Pb is above the UK soil guide value (450 mg/kg) (Environment Agency, 2002) and the Dutch intervention value (530 mg/kg) (Carlon, 2007). The mine wastes contaminated soils indicated Zn mean concentration (2220 mg/kg) that exceeded the Dutch intervention value (720 mg/kg) and the Italian soil intervention value (150 mg/kg) for residential land use scenarios (Carlon, 2007). The concentrations obtained for Cd (18.4 mg/kg) and Zn (2220 mg/kg) are consistent with those reported by Rieuwerts et al., (2000) for Cd (11.0 mg/kg) and Zn (1790 mg/kg) for a mining community in Pribam, the Czech Republic.

Table 1: Aqua-regia soluble concentrations of As, Cd, Pb, Mn and Zn in the three different contaminated soil matrices

Sample Code	As	Cd	Pb	Mn	Zn
Crude Oil Contaminated (mg/kg)					
OSS	28.7 ± 7.6	6.8 ± 1.7	60 ± 10	493 ± 29	102 ± 3
MSS	49.8± 5.3	8.7 ± 0	87 ± 15	332 ± 26	99 ± 7
ESS	35.0 ± 3.7	6.8 ± 1.7	83 ± 21	387 ± 23	127 ±3
Mean	37.8	7.4	77	404	109
Auto-mechanic Spill Contaminated (mg/kg)					
MSO	39.2 ± 5.3	5.8 ± 0	507 ± 0.4	574 ± 6	5278 ± 96
AAS	18.6± 2.9	8.5 ± 1.7	473 ± 26.6	353 ± 17	250 ± 7
CSS	17 ± 5.0	8.7 ± 0	213 ± 41.6	276 ± 48	423 ±21
MEAN	25.0	7.7	398	401	1983.50
Mine Wastes Contaminated (mg/kg)					
HLD	62.2 ± 8.1	15.5 ± 1.7	10133 ± 577	3518± 802	3333 ± 167
RCM	51.6± 3.1	22.2 ± 1.7	1687 ± 127	2130 ± 321	2778 ± 167
MS	53.0± 3.1	17.4 ± 0	10333 ± 127	9167 ± 833	548 ±15
MEAN	55.7	18.4	7384	4938	2220

The impact of the three different industries on the distribution of toxic metals in surface soils

The pseudo-total concentration ranges of the toxic metals investigated were diverse in the three different contaminated soil matrices. Among the three different contaminated matrices investigated the mine wastes contaminated soils indicated the highest mean pseudo-total concentrations for all five toxic metals of interest in this study. The elevated concentration of As, Cd, Mn, Pb and Zn in the mine wastes contaminated surfaces soils relative to the crude oil and auto-mechanic spills contaminated ones may be because the listed toxic metals have been reported (e.g. Boisa et al,. 2013) as components of Pb mineral assemblages, even at sampled location in Nigeria (e.g. Olubambi et al., 2008). Contrary to opinions held by environmental activists from most petroleum exploiting and producing communities globally, data generated from this study suggest that crude oil may not be a significant source of toxic metals in the environment. Results from this study have highlighted the relevance of auto-mechanic artisan activities on the distribution of toxic metals in soil environment.

Human Soil Ingestion Exposure Assessment when in contact with three different matrices

Using pseudo-total concentrations the potential human exposure risk associated with ingestion of the three soil matrices were estimated. The mean and range data obtained are presented in Table 2. The data suggest that for As; 0.20 µgkg^{-1} BW d^{-1}, 0.14 µgkg^{-1} BW d^{-1} and 0.31 µgkg^{-1} BW d^{-1} may be ingested from contact with crude oil, auto-mechanic spills and mine wastes contaminated surface soils, respectively (Table 2). For Cd; 0.04 µgkg^{-1} BW d^{-1}, 0.04 µgkg^{-1} BW d^{-1} and 0.10 µgkg^{-1} BW d^{-1} of the soils may be ingested from contact with crude oil, auto-mechanic spills and mine wastes contaminated surface soils, respectively (Table 2). The assessment suggest that for Pb; 0.43 µgkg^{-1} BW d^{-1}, 2.23 µgkg^{-1} BW d^{-1} and 41.90 µgkg^{-1} BW d^{-1} may be ingested from contact with crude oil, auto-mechanic spills and mine wastes contaminated surface soils, respectively (Table 2). The mean ingestible daily doses of Mn were 2.27 µgkg^{-1} BW d^{-1}, 2.25 µgkg^{-1} BW d^{-1} and 27.7 µgkg^{-1} BW d^{-1}, for the crude oil, the auto-mechanic spills and the mine wastes contaminated surface soils, respectively (Table 2). Also the mean ingestible daily doses of Zn were 0.61 µgkg^{-1} BW d^{-1}, 11.1 µgkg^{-1} BW d^{-1} and 12.5 µgkg^{-1} BW d^{-1}, for the crude oil, the auto-mechanic spills and the mine wastes contaminated surface soils, respectively (Table 2). Calculation of a potential daily intake doses of As and Pb from topsoils around mining sites indicated levels above TDI values set for the toxic metals. Similar exposure risks have been highlighted in published literature for mining communities (e.g. Boisa et al., 2013). Unacceptable daily intake doses were also obtained for Pb for inhabitants occupying land areas contaminated by auto-mechanic spill.

Table 2: Mean and range (in parentheses) TM daily intake (TM DI) orally ingested from the three different contaminated topsoil matrices, tolerable daily intake (TDIoral)

	TM DI (µg kg^{-1} BW d^{-1}) based on 100 mg d^{-1} ingestion			**TDIoral (µg kg^{-1} BW d^{-1})**
	Crude Oil	**Auto-Mechanic Spills**	**Mining Wastes**	
As	0.2 (0.16-0.28)	0.14 (0.011-0.22)	0.**31** (0.29-**0.35**)	0.3[a]
Cd	0.04 (0.04-0.05)	0.04 (0.03-0.05)	0.1 (0.09-0.12)	0.36[b]
Pb	0.43 (0.34-0.49)	**2.23** (1.20-**2.85**)	**41.9** (**9.5-58.1**)	1.9[c]
Mn	2.27 (1.89-2.77)	2.25 (1.55-3.22)	27.7 (12.0-51.5)	140[d]
Zn	0.61 (0.56-0.71)	11.1 (1.40-29.7)	12.5 (3.08-18.7)	600[e]

Data in in parentheses represents range
Numbers in bold indicate a TM intake greater than the TDI

a- Environmental Agency (2009)
b- JECFA (2012)
c- USEPA (2007)
d- Nathanail et al., (2009)

IV. Conclusion

This study has highlighted the relative concentrations of As, Cd, Mn, Pb and Zn in surface soils contaminated by crude oil, auto-mechanic spill and mine wastes. Whilst it is typical of most crude oil exploring and producing communities all over the world to proclaim unacceptable levels of chemical xenobiotics in their environment, this study highlights relatively low levels of toxic metals in surface soils contaminated by crude oil. Contrary to the activism and awareness expressed in developing countries toward pollutions resulting from petroleum based operations, little for now is said of soils contaminated by activities of artisans involved in auto-mechanic repair services. Data obtained from this study suggest that of the three industries investigated Pb mining indicated the highest concentration of all the toxic metals investigated in surrounding surface soils. This study has also highlighted auto-mechanic service operations as point source of toxic metals, especially Pb. This study in addition has highlighted the possible ingestion of doses of As and Pb above tolerable daily intake value from surface soils contaminated by auto-mechanic service spills and mine wastes. From our data there is a likelihood of children taking up toxic metals from three different matrices of surface soil.

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