



Lead Concentrations and Risk Exposure Assessment in Surface Soils at Residential Lands Previously Used for Auto-Mechanic and Auto-Welding Activities in Port Harcourt, Nigeria

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ABSTRACT: This study investigated lead concentrations in < 250 μm and < 75 μm of deposited dust and < 2000 μm , < 250 μm , and < 75 μm of surface soils at undeveloped residential lands leased to auto-mechanic artisans for a minimum of ten years and estimated exposure risk for children that will reside on the polluted lands after the lease periods. Soil-Pb levels ranges obtained were between 40.0 - 411 mg/kg, 62.0 - 435 mg/kg, and 61.0 - 491 mg/kg for < 2000 μm , < 250 μm and < 75 μm fractions, respectively. Dust-Pb levels ranges were between 138 - 1819 mg/kg, and 128 - 1584 mg/kg for < 250 μm and < 75 μm fractions, respectively. The estimated daily intake were in the range of 0.17 - 1.22 $\mu\text{g Pb Kg}^{-1}$ body weight [BW] d^{-1} and 0.39 - 5.11 $\mu\text{g Pb Kg}^{-1}$ BW d^{-1} for soil and deposited dust, respectively. This study has highlighted possible risks to occupants of lands previously used for auto-mechanic and auto-welding activities. © JASEM

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Lead is included in the formulation of several motor oils. Leaded gasoline has been phased out, but is still used in parts of Africa (Tuakuila *et al.*, 2010). Auto-mechanic yards may still be point sources of lead pollution because unacceptable lead concentrations in the range of 1500-9300 mg/kg have been reported for gear and motor oils by Clausen and Rastogi, (1977). The legacy of the auto-mechanic activities is usually not investigated and remediated; consequently people that develop their residential homes at sites previously used for auto-mechanic services may be exposed to surface soil-lead through the inhalation of re-suspended dust and ingestion of garden food (e.g. Boisa *et al.*, 2013; Laidlaw *et al* 2014; Mackay *et al* 2013; Zahran *et al.*, 2013; Boisa *et al.*, 2014). Exposure could also occur via borehole ground water, since Government treated pipe borne water is hardly available in cities of most developing countries including Nigeria.

Human exposure to motor garage chemicals has been suggested to induce genotoxic effects (Oktem *et al.*, 2004; Shastri and Plant, 2011; ; Eum *et al* 2013; Pottier *et al.*, 2013; Weuve *et al.*, 2013; Hong *et al.*, 2015). Results of previous epidemiological studies conducted on adolescent workers indicated elevated blood lead levels for auto- mechanics and garage owners (Enander *et al.*, 2004; Oktem *et al.*, 2004; Tuakuila *et al.*, 2010; Adela *et al.*, 2012). Previous published studies (e.g. Adelekan and Abegunde, 2011) at mechanic garages in Nigeria investigated contaminant levels without considering exposure risk to the artisans on site or risk to population that may

reside permanently at the emerging Brownfield areas. The aim of this study is to investigate lead concentrations in surface soils at residential lands leased to auto-mechanic and auto-welding artisans and estimate exposure risk to both artisans and people who will reside permanently on such lands.

MATERIAL AND METHODS

Study sites: Port Harcourt has large number of auto-mechanic service stations that are situated at private lands and roadsides. The study was conducted at selected roadsides (RS) and residential yards (YD) used as auto-mobile mechanic service stations within Port Harcourt City, Nigeria.



Fig 1: Map showing different sample sites

The criteria employed for the qualification of any of the sites was proximity to nursery and primary schools. Other factors considered were closeness to roadside commercial barbeque snacks points (e.g. for roasted plantain), children playgrounds and uncontaminated land (C). The zones of Port Harcourt

captured in this study were Town (RS1), Borokiri (RS2, RS3 and YD1), Ikoku (YD2 and RS4), Rivers State University -RSU (RSC and YDC) and Oyibo (YD3, YD4, YD5, YD6 and YD7) (Figure 1). Details

about the type of auto-mechanic services provided, public institutions and nature of surfaces at each of the study sites are listed in Table 1.

Table 1: Study sites and sampling details

Sample Code	Description of sample point	Port Harcourt Zone	Mech. activity	Vegetation
RS1	Roadside	Town	Car engine service	Exposed topsoil
RS2	Roadside	Borokiri	Car engine service and body refinishing	Exposed topsoil
RS3	Roadside	Borokiri	Car engine service	Exposed topsoil
YD1	Fenced Yard	Borokiri	Metal scrap dump	Exposed topsoil
YD2	Fenced Yard	Ikoku, Diobu	Car engine service and body refinishing	Exposed topsoil
RS4	Roadside	Ikoku, Diobu	None	Concrete floor
YD3	Fenced Yard	Oyigbo	Car engine service and body refinishing	Exposed topsoil
YD4	Fenced Yard	Oyigbo	Car engine service	Exposed topsoil
YD5	Fenced Yard	Oyigbo	diesel engine and battery services	Exposed topsoil
YD6	Fenced Yard	Oyigbo	Metal scrap dump	Exposed topsoil
YD7	Fenced Yard	Oyigbo	Car engine service	Exposed topsoil
RSC	Roadside	RSUST	None	Exposed topsoil
YDC	Fenced Yard	RSUST	None	Grass herbs

Sampling and Treatment: Surface soil (0-10 cm depth) samples were collected from all sites excluding the RS4 (concrete surface) with stainless steel trowel. Deposited dust samples were collected from pavements and concrete floors of adjoining schools and residential homes. Each soil sample comprised 4 – 6 closely spaced subsamples which were homogenized composite samples of about 1000 g (wet weight). Typical investigations involving contaminated surface soils in developing countries are based only on the < 2000 μm fraction. In order to estimate human exposure risk at the study sites, relevant particles size fractions for the three main exposure pathways (i.e. dermal, ingestion and inhalation) (Environment Agency, 2009; Siciliano et al., 2009) were considered. Following collection, soil samples were air dried, disaggregated and sieved through 2000 μm , 250 μm , and 75 μm meshes. The dust samples were also air dried and sieved through 250 and 75 μm .

Particle size analysis

The < 2000 μm fraction (51 g) of the different samples were quantitatively transferred into screw cap plastic bottles and organic matter was destroyed by adding hydrogen peroxide and sodium hexametaphosphate and the resulting soil analysed by hydrometer method (Rowell, 1994; Keller and Gee, 2006).

Organic matter: About 5 .00 g of the < 2000 μm fraction of soils were quantitatively transferred into previously weighed clean porcelain crucibles and heated in a muffle furnace at 400 $^{\circ}\text{C}$ for 3 hours. The resulting crucibles and their contents were allowed to cool in desiccators and weighed to determine the loss

on ignition as a measure of the organic matter (Rowel, 1994; Schumacher, 2002).

Pseudo Total Analysis: Aqua regia digestion (HCl : HNO₃ in the ratio 3:1 v/v) were performed on 0.5 g subsamples and an aqua regia certified reference material (BCR 143R Sewage sludge amended soil), using hotplate for 2 hours. For each digestion reagent blanks were also prepared. The resulting filtrates were transferred into plastic sample bottles and subsequently analyzed for their Pb content by Flame (air-C₂H₂) Atomic Absorption Spectrophotometer (AAS-Buck Scientific).

Experimental Quality Control: The accuracy of the aqua regia digestion method was monitored using BCR 143R (aqua regia certified Sewage sludge amended soil). Excellent results were obtained for total Pb (171 \pm 9 mg/kg) determined compared to certificate value (174 \pm 5). All test samples were extracted in triplicate. For the determination of Pb, 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. Two quality control standards (high and low range) were analysed after no more than 10 unknown samples. All reported measurements are based on the average of three replicate analyses.

RESULTS AND DISCUSSION

Particle size analysis and soil organic matter: The contents of sand, silt and clay for test samples are in the range of 2.8 – 80.0 %, 18.8 – 95.1 % and 0.6 – 2.1 %, respectively (Table 2). The lowest silt content (YD7) observed in this study is above the contents indicated in the control samples (RSC and YDC)

(Table 2). The higher content of silt obtained in mechanic workshops compared to the controls is expected since some of the specialized operations at the workshops includes metal grinding and automobile body refinishing. Sample site, YD1; an open air mechanic workshop with metal sheet scrap dumpsite indicated the highest silt content. At YD1 sample site large metal sheet are cut into smaller sizes before their movement to other locations for reuse. The very high silt content at the YD1 may have resulted from the cutting process. The relatively lower contents of sand and clay in the test samples compared to the vegetated control site (Table 1) may be due dilution of their original geochemistry by silts

generated during repair works. Samples, RS1, YD1, YD4, YD5 and YD6 indicated higher contents of silt-clay compared to the control, RSC and YDC. Excluding RS1 (a site previously used as open dumpsite for domestic waste), all test samples indicated lower soil organic matter than the controls (Table 2). The highest soil organic matter observed at the former domestic waste dumpsite is expected because of food and other human wastes that had interacted with the original surface soil. The lower soil organic matter observed for the other surface soil from mechanic workshops are expected since most waste generated are not organic and biodegradable in nature.

Table 2: Particle size analysis and soil organic matter results

Location	Sand %	Silt %	Clay %	Silt & Clay %	Organic Matter %
RS1	50.8	46.8	2.4	49.2	15.4
RS2	77.4	22.0	0.6	22.6	5.8
RS3	78.8	20.0	1.2	21.3	5.0
YD1	2.8	95.1	2.1	97.2	3.4
YD2	77.4	21.4	1.2	22.6	2.6
YD3	78.7	20.1	1.2	21.3	4.6
YD4	55.4	42.5	2.1	44.6	3.6
YD5	60.0	38.0	1.8	40.0	4.0
YD6	26.8	71.1	2.1	73.2	5.4
YD7	80.0	18.8	1.2	20.0	5.8
RSC	91.0	6.0	3.0	9.0	8.1
YDC	77.0	16.0	7.0	23.0	6.5

Lead concentration in surface soils: Lead levels measured in the < 2000 μm fraction of the surface soil samples were found to range between 40 and 411 mg/kg, and the range was above the concentrations 24 and 29 mg/kg found in control samples (YDC and RSC, respectively) (Table 3). The maximum concentration (411 mg/kg) indicated in this study is above that (298 mg/kg) reported by Ipeaiyeda et al., (2007) for a newer city, Iwo in Nigeria (Table 4). The maximum concentration 411 mg/kg observed in this study for the < 2000 μm fraction is below, 703, 703 and 2880 mg/kg reported for similar studies conducted by Iwegbue et al., (2006), Abidemi, (2011), and Adedemi and Abegunde, (2011), respectively (Table 4). The large difference between the maximum observed in this study to that reported for Ibadan, Nigeria by Adedemi and Abegunde, (2011) may be due the fact that Ibadan is a older city than Port Harcourt and the age of the automobile repair shop at Ibadan may be higher. Most of the concentrations are below the soil guideline value of 200 mg/kg set by the Rivers State Ministry of Environment (RSMenv). Only samples from Ikoku and Oyibo indicated concentrations of 208 and 411 mg/kg, respectively that are above the local regulatory value. Lead concentrations obtained for < 250 μm size fraction were in the range of 62 – 435

mg/kg (Table 3). The lead concentration range (62 – 435 mg/kg) observed in this study is consistent with the range (68.7 – 396 mg/kg) previously reported by Khan and Kathi, (2014) for surface soil sample investigated at automobile garages in India. Only concentrations of samples from Ikoku (YD2) (241 mg/kg), Oyibo (YD5) (435 mg/kg) and (YD6) (388 mg/kg) were above the local regulatory value (200 mg/kg). The concentrations obtained for the < 75 μm fractions were in the range of 61 – 491 mg/kg (Table 3). For the inhalable fraction (< 75 μm) about 50 % of the samples indication concentrations that were above the local acceptable limit of 200 mg/kg. The lead concentrations in soil samples were more in the finer fractions. The order of lead loadings in soil samples is; 75 μm > 250 μm > 2000 μm .

Lead concentration in deposited dust: The concentrations of lead in the < 250 fractions were in the range of 138 – 1819 mg/kg (Table3). Excluding sample points RS1 and YD7, all other sample points indicated concentration levels above the locally acceptable limit of 200 mg/kg. All the < 250 μm dust sample concentrations are two-fold or more those of the < 2000 μm soil size fractions. Presently most surface soil contamination studies conducted in developing countries are based on the < 2000 μm size

fraction (Table 4) (e.g. Adedemi and Abegunde, 2011). This observation suggest that published previous studies conducted at automobile repair shops may have under reported risk levels because they were based on the < 2000 μm fraction only. The < 75 μm fraction concentrations were in the range of 128 – 1584 mg/kg (Table 3).

Table 3: Pb concentrations (mg/kg) in soils; < 2000 μm , 250 μm and 75 μm , and dust; < 250 μm and 75 μm fractions

location	Soil < 75 μm	Dust < 75 μm	Soil < 250 μm	Dust < 250 μm	Soil < 2000 μm
RS1	61	243	62	138	69
RS2	122	128	64	214	72
RS3	375	524	140	502	75
YD1	310	350	181	217	108
YD2	376	398	241	726	208
RS4	154	367	124	191	98
YD3	152	106	133	205	81
YD4	287	767	107	587	59
YD5	583	1584	435	1819	411
YD6	491	565	388	420	166
YD7	126	153	73	188	40
RSC	69	77	50	62	30
YDC	54	48	36	45	24

The concentrations observed for the re-suspendable size fraction were also higher than those for the < 2000 μm fraction. Excluding sample points YD2 and

YD3 all other sample points indicated concentrations that were at least two times higher than those for the < 2000 μm . The finer fractions are usually expected to have higher loadings of contaminants, but results obtained in this study indicate some exceptions; samples obtained from RS2, YD2, YD3 and YD5 indicated higher Pb loading for the < 250 μm than the < 75 μm size fraction. The elevated lead concentrations observed for the more coarse size fraction samples (RS2, YD2 and YD3) may be due the fact that automobile body refinishing jobs were taking place alongside the car engine servicing. Since one of the activities at automobile body workshops involve scrapping of old automobile body surfaces the generated paint dust and metal fillings may have contributed the large lead loadings.

Soil ingestion exposure assessment at automobile mechanic yards: In developed countries, currently human health risk assessment studies from soil are based on ingestible (< 250 μm) and fine particles of soil (< 100 μm) fractions (e.g. Boisa et al., 2013; Boisa et al., 2014). Using the < 250 μm size fraction Pb concentrations in surface soil and deposited dust, it was possible to estimate the potential the potential human exposure risk associated with the ingestion of any of the matrices. The method used here is based on calculating the Pb daily intake (Pb DI) from incidental ingestion.

Table 4: Soil and dust Pb concentrations in automobile service garages

Pb Conc. Range (mg/kg)	Study Location	Particle size (μm)	Authors
40.0 – 411	Port Harcourt, Nigeria	< 2000	This study
62.0 – 435	Port Harcourt, Nigeria	< 250	This Study
61.0 – 583	Port Harcourt, Nigeria	< 75	This study
246 – 703*	Port Harcourt, Nigeria	Not indicated	Iwegbue et al., 2006
68.7 – 396	Puducherry, India	< 250	Khan and Kathi, 2014
106 – 298	Iwo, Nigeria	Not indicated	Ipeaiyeda et al., 2007
11.7 = 2880	Ibadan, Nigeria	< 2000	Adedemi and Abegunde, 2011
16.0 – 703	Osogbo, Nigeria	Not indicated	Abidemi, 2011

*- auto-mechanic waste dump sites

Pb DI ($\mu\text{g Pb Kg}^{-1}$ body weight [BW] d^{-1}) has been calculated for children (2-6 years) that may later occupy these emerging brownfields and pupils presently studying in schools situated within or around mechanic garages based on their pica habits as:

$$Pb.DI = \frac{(EC \times SIR \times ED)}{BW}$$

Where; EC is aqua regia Pb concentration, $\mu\text{g/g}$; SIR is soil ingestion rate, mg/d (given as 100 mg/d for the age group of interest) (US EPA, 2011); ED is exposure duration, to be 0.5 as exposure could be 12

hours per day, 365 days a year; BW is body weight, 17.8 kg (US EPA, 2011) for the age group of interest.

The automobile artisans workshops investigated in this study are adjoining either primary or nursery school. Some of the shops had additional businesses like food canteens and in few cases wives of artisans and their below school age children stay at the shops to assist. The soils and deposited dust sampled in this study are in contact with the population. However, because of the vulnerability of children this study sought to quantify the exposure to children in the age range of 2-6 years. Exposure estimate were

calculated for topsoil and deposited dust samples using the concentration data (Table 3). The data for test samples (Table 5) suggest the daily intake is in the ranges of 0.17 – 1.22 $\mu\text{g Pb Kg}^{-1}$ body weight $[\text{BW}]\text{d}^{-1}$ and 0.39 – 5.11 $\mu\text{g Pb Kg}^{-1}$ BWd^{-1} for soil and deposited dust, respectively. All soil samples indicated lower Pb DI values than the TDI (1.9 $\mu\text{g Pb Kg}^{-1}$ body weight $[\text{BW}]\text{d}^{-1}$), however, they are above the Pb DI values for the controls (0.10 and 0.14 $\mu\text{g Pb Kg}^{-1}\text{BWd}^{-1}$). Two Pb DI values, 2.04 and 5.11 $\mu\text{g Pb Kg}^{-1}$ BWd^{-1} obtained for YD2 and YD5, respectively, are higher than the recommended TDI for Pb (1.9 $\mu\text{g Pb Kg}^{-1}\text{BWd}^{-1}$). The YD2 site is employed for both car engine servicing and automobile body refinishing works (Table 1). The YD5 site is engaged in diesel engine truck and battery services (Table 1). The elevated Pb Di obtained for YD5 yard may be due the fact that the high pressure-resistant lubricants employed for diesel engines contains lead Naphthenate (Clausen and Rastogi, 1977). Our data highlights that a possibility of harm exist via exposure to the deposited dusts. Implications for occupiers of residence sited on Brownfields resulting from mechanic activities

Table 5: Lead daily intake (Pb DI) orally ingested from topsoil and deposited dust, tolerable daily intake (TDI_{oral}) based on sample aqua regia concentration. Number in bold indicate a Pb intake greater than the TDI

Location	Pb DI (μgkg^{-1} BWd^{-1})		TDI (μgkg^{-1} BWd^{-1})
	soil	Dust	1.9*
RS1	0.17	0.39	
RS2	0.18	0.60	
RS3	0.39	1.40	
YD1	0.51	0.61	
YD2	0.68	2.04	
RS4	0.35	0.54	
YD3	0.37	0.57	
YD4	0.30	1.65	
YD5	1.22	5.11	
YD6	1.09	1.17	
YD7	0.20	0.52	
RSC	0.14	0.17	
YDC	0.10	0.13	

*- JEFCA (2012)

Existing environmental risk assessment allows for the comparison of soil aqua regia extractable total of potentially harmful elements with environmental quality guidelines. This study has shown the possibility of link between Pb concentrations in surface soils and deposited dusts within automobile mechanic yards. Whilst direct soil ingested can be monitored in children, the deposition of suspended surface dust on children hands and subsequent ingestion might be difficult to monitor. All dust samples indicated higher Pb DI than the soil samples. Permanent residency at such land areas may expose

inhabitants to unacceptable Pb intake. In order to minimize Pb intake risk management strategies such as soil encasement, re-vegetation and soil replacement will be required. Whilst attempts are made to have better understanding of sources of Pb at mechanic yards and effect contaminants management, it is essential that the danger of leasing landed properties for mechanic services be highlighted.

Conclusion: This study investigated the levels of Pb in surface soils and dusts obtained from eleven (11) land areas previously used for automobile mechanic and other associated services and two (2) control sites. Our data indicated elevated soil- Pb levels for the automobile service sites. Whilst published similar studies in developing countries were principally based only on the < 2000 μm particle size fraction of soils, this study in addition of the classical < 2000 μm size fraction of soil also considered two other size fractions (< 250 and < 75 μm) and dust matrix.

The higher Pb levels observed in the dusts samples highlights the relevance of this matrix. Risk assessment for accidental ingestion of soil and dust based on the < 250 μm fraction indicated more daily intake doses of Pb from dust than soil. Two sites;(1). a site used for diesel engine and battery services, and (2) a site used for car engine service and automobile body refinishing work indicated daily Pb intake doses that exceeded current tolerable daily intake value. The utilization of dust data for the estimation of children exposure doses may however be over-protective of children's health.

Most of the sites investigated were engaged in multiple automobile mechanic and welding services; therefore it was difficult to account for the specific activities responsible for high Pb loadings. There is therefore need to conduct a source apportionment studies of potentially harmful elements at automobile yards and sites. Data from such studies may assist in identifying services associated with higher lead loadings.

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