

1 **Article Title:** MOBILITY, SPATIAL VARIATION AND HUMAN HEALTH RISK ASSESSMENT OF
2 MERCURY IN SOIL FROM AN INFORMAL E-WASTE RECYCLING SITE, LAGOS, NIGERIA

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4 **Authors:**

5 Mrs Anselm Oluwaseun H.^{1,3,4}

6 Dr. Cavoura Olga^{2*}, ORCID iD 0000-0002-8556-9225

7 Dr. Davidson Christine M.³ ORCID iD 0000-0002-8045-3530

8 Dr. Oluseyi, Temilola O.¹ ORCID iD 0000-0002-5722-3403

9 Dr. Oyeyiola Aderonke O.¹ ORCID iD 0000-0003-2395-3528

10 Mr. Toggias, Konstantinos⁵

11 *Corresponding author okavoura@uniwa.gr

12

13 ¹Department of Chemistry, University of Lagos, Akoka, Lagos, Nigeria.

14 ²Department of Public Health Policy, School of Public Health, University of West Attica, Greece

15 ³WestCHEM, Department of Pure and Applied Chemistry, University of Strathclyde, Glasgow, UK

16 ⁴Department of Chemical Sciences, Tai Solarin University of Education, Ijagun, Ogun State, Nigeria.

17 ⁵Athens, Greece.

18

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22

23 **Abstract**

24

25 Spatial variations and mobility of mercury (Hg), and Hg associations with other potentially toxic elements (PTEs)
26 were studied in soil samples from Alaba, the largest e-waste recycling site in Nigeria and West Africa. Total Hg
27 concentration was determined in surface soils samples from various locations using cold vapour atomic absorption
28 spectrometry (CVAAS) following microwave assisted acid extraction, while sequential extraction was used to
29 determine operationally defined mobility. The concentrations of the PTEs arsenic (As), cadmium (Cd) chromium
30 (Cr), copper (Cu), manganese (Mn), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) metals were determined
31 using inductively coupled plasma mass spectrometry (ICP-MS) following microwave assisted digestion with *aqua*
32 *regia*. Total Hg concentration ranged from < 0.07 to 624 mg/kg and was largely dependent on the nature and
33 intensity of e-waste recycling activities carried out. Mobile forms of Hg, which may be HgO (a known component
34 of some forms of e-waste) accounted for between 3.2 and 23% of the total Hg concentration, and were observed
35 to decrease with increasing organic matter (OM). Non-mobile forms accounted for > 74% of the total Hg content.
36 In the main recycling area, soil concentrations of Cd, Cd, Cu, Hg, Mn, Ni, Pb and Zn were above soil guideline
37 values (Environment Agency 2009; Kamunda et al., 2016). Strong associations were observed between Hg and
38 other PTEs (except for Fe and Zn) with the correlational coefficient ranging from 0.731 with Cr to 0.990 with As
39 in April but these correlations decreased in June except for Fe. Hazard quotient values > 1 at two locations suggest
40 that Hg may pose health threats to people working at the e-waste recycling site. It is therefore recommended that
41 workers should be investigated for symptoms of Hg exposure.

42

43 **Keywords**

44

45 e-waste recycling; mercury; potentially toxic elements; mobility; risk assessment

46

47

48 **Introduction**

49

50 The increase in global demand for information technology has led to the increased production of electronic gadgets
51 with new features, rendering old products quickly obsolete (Schluep et al. 2012). This has resulted in the
52 generation of a high volume of electronic waste (e-waste). Globally, approximately 20 to 50 million metric tons
53 are produced annually, with China and Nigeria being the major countries importing e-waste. Nigeria being the
54 second largest receiving country in world, has an annual inflow of over four million computers as well as many

55 other types of electrical and electronic equipment (EEE) (Adediran and Abdulkarim 2012; Ibrahim et al. 2013).
56 These e-wastes contain a wide range of organic and inorganic pollutants such as polybrominated diphenyl ethers,
57 polychlorinated biphenyls, and potentially toxic elements (PTEs) including arsenic (As), cadmium (Cd) chromium
58 (Cr), copper (Cu), manganese (Mn), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn). These arise from waste
59 components such as thermostats and batteries, cathode ray tubes, (Adekola and Dosumu 2019) and fluorescent
60 lamps that are used in the liquid crystal display (LCD) of screens and monitors (DGUV, 2016). When these are
61 disposed of improperly, Hg and other PTEs can be released into the environment. It is estimated (US EPA, 1997)
62 for example that broken fluorescent bulbs release 6% of their total Hg content to the atmosphere in a form that is
63 easily vaporized, such as elemental Hg, whilst the remaining Hg content is converted into solid compounds,
64 principally HgO, the fate of which is not well documented (NJDEP, 2004). Classified as a toxic element
65 (Cheremisinoff 2016), Hg and its compounds are capable of causing harm to human health and the environment.
66 Toxic effects in human health include motor, visual, and/or cognitive dysfunctions, cytogenetic damage such as
67 mitotic index decrease and increase in polypoid cells and kidney damage (Amorim et al. 2000; Passos and Mergler
68 2008). Recent studies have highlighted the potential risk of exposure to Hg from e-waste recycling sites. For
69 instance, children in e-waste recycling sites surrounding Jakarta (Indonesia) showed increased levels of Hg in
70 their hair samples compared to their counterparts living in areas where e-waste recycling was not carried out
71 (Soetrisno and Delgado-Saborit 2020). Similarly, 62% of 118 children living near e-waste recycling sites in Guiyu,
72 China, were found to have Hg blood level above 10 µg/dL, where blood Hg levels above 5.8 µg/dL have been
73 reported to cause harm in children (Xu et al. 2018). As with all metals, Hg is persistent in the environment. It can
74 be transformed as a result of biological and chemical processes that are affected by soil pH, temperature and
75 organic matter (OM) content (Biester et al. 2002). It is well established that the toxicity of Hg in the environment
76 depends on its speciation (Chen et al. 2009; Li 2011). Insoluble Hg species such as mercuric sulfide (HgS) are
77 considerably less mobile and therefore less bioavailable, while alkylmercury and soluble inorganic Hg²⁺ species
78 are mobile and account for a major part of Hg toxicity in soil (Han et al. 2003). Although soil contains several
79 inorganic forms of Hg, only mobile, dissolved Hg²⁺ can be methylated (Issaro et al. 2009). Methylmercury (Me-
80 Hg), formed from the action of methylating bacteria on the mobile or available Hg species, is one of the most
81 harmful Hg species due to its bioaccumulation and biomagnification potential (Leopold et al. 2010; Li et al. 2006a)
82 in the environment. Due to the different toxicities exhibited by various Hg species, information on the total Hg
83 concentration in soil is not sufficient to understand the potential methylating capacity and toxicity of Hg, hence
84 the need for speciation studies.

85 Sequential extraction procedures or column leaching tests, involving the extraction of species with different
86 chemical reagents of varying strengths can be used to study the speciation of Hg in soil (Han et al. 2003; Santos-

87 Francés et al. 2011). In these processes, Hg content in the soil sample is subdivided into operationally defined
 88 groups of more, or less, soluble species according to the compartments with which Hg is nominally associated
 89 (for example, the exchangeable, carbonate, metal oxide, OM and residual fractions). Species that are weakly
 90 bound to the solid matrix are extracted first during speciation protocols (Cavoura et al. 2019; Reis et al. 2010).
 91 Reagents used for the extraction of Hg from exchangeable and mobile fractions include ammonium chloride,
 92 ammonium acetate, ethanoic acid, sodium nitrate and calcium chloride, due to their capacity to release Hg by ion-
 93 exchange (Issaro et al. 2009). Reagents such as sodium hydroxide, potassium hydroxide and hydrogen peroxide
 94 are used to extract the fraction bound to OM through oxidation of the phase. The USEPA (2004) recommend 4 M
 95 and 5.33 M HNO₃ for the determination of the mobile and semimobile fractions respectively for Hg. The
 96 concentration of potentially toxic elements in topsoil and dust samples from e-waste recycling sites in Lagos has
 97 been previously studied (Ohajinwa et al. 2018) and contamination factors for Hg were found to exceed values
 98 from a nearby control site by a magnitude of 100s to 1000s. However, there is no information on Hg species,
 99 hence mobility and risk to human health. This research therefore aims to assess the spatial and temporal variation
 100 of total Hg concentration and its association with other PTEs in the topsoil of an informal e-waste recycling site
 101 in Nigeria, to determine the mobility of Hg species, and finally to assess the risk to workers and inhabitants in the
 102 area.

103

104 **Methodology**

105

106 Study Site Description

107

108 Alaba International Market, located in Ojo Local Government Area, Lagos, Nigeria, is the largest and oldest
 109 market for used and new EEE in West Africa. Founded in 1978, it is one of the most prominent and oldest business
 110 clusters for the refurbishing, collection, and recycling of used and end-of-life electronic products (Manhart et al.
 111 2011). Approximately ten to fifteen shipping containers arrive daily from Europe and Asia, each containing about
 112 400,000 second-hand units of EEE (Ohajinwa et al. 2018). Within the market is an e-waste recycling site which
 113 became fully functional for primitive recycling activities in 2010 and has rapidly grown into a wide expanse of
 114 land including shelters for the e-waste recyclers. Some of the activities carried out in this area include open burning
 115 of e-waste to recover precious metals and manual removal of important elements such as copper using bare hands,
 116 with occasional use of hammers, chisels and screwdrivers for materials separation as shown in Figures 1 a and b.

117

118 **Fig 1 a and b** Nature of e-waste recycling activities in Alaba e-waste recycling site, Nigeria.

119

120 Sampling

121

122 Like other cities in Nigeria, Lagos has two seasons namely the dry and rainy seasons. The rainy season begins in
123 April and spans to October with a break in August while the dry season begins in November and ends in March.
124 The highest rainfall is mostly in the month of June (Nnaji et al. 2016; Okorie et al. 2019). From a thirty-year
125 record (1980-2010), the mean rainfall in April was 137 ± 80.2 mm while in June the mean rainfall was 297 ± 112
126 mm in Lagos (Adepitan et al. 2017). Based on an initial spatial survey the e-waste recycling site was categorised
127 into five areas according to the different activities carried out. (Figure 2):

128 • A: main recycling area and some make-shift houses for e-waste recyclers,

129 • B: moderate recycling activities and resting areas for the recyclers,

130 • C: minimal recycling areas and some make-shift houses for the recyclers,

131 • D: a playground often used for football and some residential houses

132 • E: an old e-waste recycling site and now a dumpsite for municipal and unrecyclable e-wastes.

133 A total of seventeen samples were collected over the five areas. Each sample was collected in a star-like pattern,
134 and samples from the same area were combined to give a composite sample. The sampling areas were
135 georeferenced using a hand-held global positioning system (GPS) instrument. The topsoil samples were collected
136 using plastic shovels at depth 0-5 cm in April 2016 and June 2016 to represent varied wet periods in the area. The
137 collected samples were transported to the laboratory where they were homogenised, air dried for two weeks,
138 sieved to 100 mm and stored appropriately prior to digestion.

139

140 **Fig 2** Map of Alaba International showing the sample locations at e-waste recycling site

141

142 Determination of pH and loss on ignition

143

144 Soil pH was determined (FiveEasy FE 20 Mettler Toledo pH meter) using a soil to deionised water ratio of 1:5
145 (BSI 2005; Standard 2005). Soil OM content was estimated by loss on ignition where 1 g of the composite was
146 ignited at 550 °C for 8 hrs (Schumacher 2002).

147

148 Determination of total mercury concentration

149 Soil samples (0.5 g each) were digested with 10 mL HNO₃ using microwave assisted digestion, (Berghoff
150 Speedwave MWS-2 system). Following sample reduction with 3% NaBH₄, determination of total Hg

151 concentration was performed (PE 2006) using cold vapour atomic absorption spectrometry (CVAAS), (MHS-10
152 Hg/Hydride system, operated in CV mode, Perkin Elmer, MA, USA)

153

154 Determination of mercury mobility

155

156 Mobility of Hg species was determined using extractants of increasing acid concentration (USEPA 2014). Briefly,
157 mobile species such as HgCl_2 , $\text{Hg}(\text{OH})_2$, $\text{Hg}(\text{NO}_3)_2$, HgSO_4 and HgO were determined by microwave assisted
158 extraction with 4 M HNO_3 at 100 °C. The extraction for the determination of the semimobile species including
159 Hg^0 , Hg^0 -metal amalgam and Hg^{2+} complexes was achieved using 5.33 M HNO_3 as extractant with a combination
160 of vortex, heating and centrifugation. Nonmobile species, such as HgS and HgSe , were determined from the
161 difference between total Hg concentration and the sum of the concentrations of mobile and semimobile species of
162 Hg.

163

164 Determination of other potentially toxic elements

165

166 Concentrations of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn were determined following microwave assisted digestion
167 (MarsXpress, CEM, Buckingham, UK) of 1.0 g of each sample with 20 mL freshly prepared *aqua regia*
168 (microwave programme: 800 W at 160 °C with ramp time of 30 min and holding time of 20 min.). The digests
169 were analysed using ICP-MS (Agilent Model 7700x). All reagents used were of analytical grade or higher
170 purchased from Sigma-Aldrich and Fisher Chemical Company, UK.

171

172 Quality control and data analysis

173

174 All glassware used was soaked in 10 % v/v HNO_3 overnight and rinsed with deionised water prior to use. Standard
175 solutions and reagents were stored in glass containers. Analytical performance was checked using certified
176 reference material (CRM) BCR 320R (Channel Sediment) obtained from the Institute for Reference Materials and
177 Measurements, Geel, Belgium for Hg, and URBSOIL, an in-house reference material (Davidson et al., 2007) for
178 the remaining PTEs. All digestions, analysis and measurements were done in triplicate after instrument calibration
179 with appropriate standard solutions. Data analysis was performed using Minitab 19 statistical package.

180

181 Human health risk of exposure to mercury in the area

182

- 213 OSIR is the oral ingestion rate (100 mg/day for ISLU; 200 mg/day for SLU)
- 214 SAF Soil allocation factor of reference dose for potentially toxic elements (0.2)
- 215 VF is the Hg volatile factors ($32675.6 \text{ m}^3/\text{kg}^{-1}$)
- 216 SAE is the exposed skin area for adults, (5074.893 cm^2 for ISLU; 2447.557 cm^2 for SLU)
- 217 SSAR is the skin surface adhesion rate of soil on an adult body ($0.07 \text{ mg}/\text{cm}^2$ for ISLU; $0.2 \text{ mg}/\text{cm}^2$ for SLU)
- 218 DAIR is the daily air inhalation volume ($14.5 \text{ m}^3/\text{day}$ for ISLU; $7.5 \text{ m}^3/\text{day}$ for SLU)
- 219 EFO is the outdoor exposure frequency (62.5 day/yr for ISLU; 87.5 day/yr for SLU)
- 220 EFI is the indoor exposure frequency (187.5 day/yr for ISLU; 262.5 day/yr for SLU)
- 221 Ev is the frequency of daily event for skin contact with soil (1 day^{-1})
- 222 PM_{10} is the concentration of inhalable particulate matter in air ($0.15 \text{ mg}/\text{m}^3$)
- 223 PLAF is the retention ratio of soil particles in human body through inhalation (0.75)
- 224 fspo is the fraction of soil particles in outdoor air (0.5)
- 225 fspi is the fraction of soil particles in indoor (0.8)
- 226 RfD_{ing} is the reference dose via ingestion ($3 * 10^{-4} \text{ mg}/\text{kg}.\text{day}$)
- 227 RfD_{der} is the reference dose via dermal absorption ($2.1 * 10^{-5} \text{ mg}/\text{kg}.\text{day}$)
- 228 RfD_{inh} is the reference dose via inhalation ($7.66 * 10^{-5} \text{ mg}/\text{kg}.\text{day}$).

229

230

231 **Results and Discussion**

232

233 Limit of detection and recovery

234

235 The limit of detection (LOD) for Hg was $0.75 \text{ }\mu\text{g}/\text{L}$ corresponding to a soil concentration of $0.07 \text{ mg}/\text{kg}$ for 0.5 g
236 sample weight. Recovery of Hg from CRM BCR 320R, containing $0.85 \pm 0.09 \text{ mg}/\text{kg}$ Hg was $108 \pm 15.1 \%$ (n =
237 3). Limit of detection was 0.0238, 0.00175, 0.00863, 0.0124, 1.29, 0.0077, 0.00280, 0.0016 and $0.0755 \text{ }\mu\text{g}/\text{L}$ for
238 As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn respectively corresponding to 0.0119, 0.000875, 0.00432, 0.0062, 0.645,
239 0.00385, 0.0014, 0.0008 and $0.0378 \text{ mg}/\text{kg}$ for 1 g of sample and recovery of the elements ranged from 94 to
240 120%.

241

242 Physicochemical characteristics of the soil

243

244 Site locations, pH, OM and total Hg content are given in Table 1. Soil pH ranged from 6.86 to 8.97 and was
245 slightly alkaline at four locations, in agreement with previous studies on the Alaba e-waste recycling site
246 (Isimekhai 2017). The lower pH (6.86) observed in Area E may be as a result of the degradation of OM in
247 composts from the municipal dumpsite that can release organic and inorganic acids, and the production of H⁺
248 from the mineralization of nitrogen in the OM (Achiba et al. 2010), which can reduce the pH of calcareous soil
249 over time (Achiba et al. 2009; Hamidpour et al. 2012; Mohammad and Athamneh 2004). The pH increased in
250 June relative to April in two areas (C and E) while it was lower in Area A, B and D, however, the differences were
251 not significant (t-test, $p > 0.05$).

252 The OM content ranged from 1.35 to 29.4%. Area E had the highest OM content, possibly a consequence of
253 composted municipal waste that might have increased the soil OM in that area (Achiba et al. 2010; WalidBen et
254 al. 2009). Soil OM content was higher in June than in April, with the exception of area C where it decreased,
255 however, as in the case of pH, there was no significant difference ($p > 0.05$). While there was no correlation
256 between OM content and pH in June ($r = -0.356$), there was slight negative correlation between the two
257 physicochemical parameters in April ($r = -0.531$).

258

259

260 Total mercury concentration and influence of soil characteristics

261

262 The concentration of Hg in the site ranged from < 0.07 to 624 mg/kg (Table 1). Hg concentration was highest in
263 area A, the main e-waste recycling area, with mean values of 624 ± 84 mg/kg and 443 ± 53 mg/kg in April and
264 June respectively. The Hg concentration in area B ranked second highest with averages of 15.2 ± 3.8 and $13.1 \pm$
265 1.7 mg/kg in April and June respectively, probably because a lesser quantity of e-wastes are sorted and recycled
266 in B compared to A. Total Hg concentration was below LOD in areas D both in April and June while it was below
267 LOD in areas C and E in April only. In June the average concentrations were 1.65 ± 0.20 in C and 1.99 ± 0.25
268 mg/kg in F. The soil concentration of Hg in areas A and B exceeded the Hg level of 3.86 mg/kg found in
269 uncontaminated soil in Lagos (Bakare et al. 2004). While there are no soil guideline values for Hg in Nigeria, in
270 area A the Hg content was above other national guideline values for residential soil (170 mg/kg; Environment
271 Agency 2009) in both periods, and was above concentrations in gold mining areas where an Hg concentration in
272 top soil of 390 mg/kg was reported (Sakakibara and Sera 2020). Concentrations at the other locations with minimal
273 e-waste activities were below the soil guideline values indicating, as expected, that e-waste recycling activity is
274 the major source of Hg in the topsoil in Alaba e-waste recycling sites. In areas A and B of the site Hg
275 concentrations exceeding concentrations reported in previous studies (Ohajinwa et al. 2018) that investigated Hg

276 soil contamination from e-waste recycling areas in countries including China, Ghana and India (Fosu-Mensah et
277 al. 2017; Han et al. 2018; Pradhan and Kumar 2014; Wang et al. 2015; Zhang et al. 2014; Zhang et al. 2012; Zhao
278 et al. 2015) as shown in Table 2. This may partly be as a result of the use of different sampling depths. In most
279 literature studies, sampling was done at 0-20 cm while 0-5 cm depth was used in the current work. While Hg
280 concentration in soil can decrease down to a depth of 3 m (Blackwell et al. 2014; Ozunu et al. 2009), sampling
281 between 0-5 cm depth, where interaction with soil is greatest, is recommended for the determination of the effect
282 of human or animal exposure to top soil contaminants (Buczko and Kuchenbuch 2007). In addition to sampling
283 depth, land use within the recycling site and distance from main recycling activities also affect soil Hg content.
284 The concentration measured in this study was found to be far higher than that reported by Wang et al. (2015)
285 (Table 2) who sampled at the same depth (0-5 cm) in Longtang, Guangdong province, China, but at a greater
286 distance from the e-waste recycling site than in this study. Although pH and OM are important factors that affect
287 the retention of Hg in soil (Jing et al. 2007; Yang et al. 2007), there was no significant relationship between pH,
288 OM and total Hg content in April or June ($0.0296 < r < 0.307$, $p > 0.05$) in the current study.

289

290 Concentration of potentially toxic elements and correlations with Hg

291

292 Average concentrations of other PTEs from the sampling sites are given in Table 3 for April 2016 and Table 4 for
293 June 2016. Site A had the highest concentrations of PTEs, and levels exceeded the soil guideline values of 3, 100,
294 100, 50, 100 and 300 mg/kg for Cd, Cr, Cu, Ni, Pb and Zn respectively. Concentrations of Cd, Cr, Cu, Fe, Mn, Ni
295 and Pb were lower farther from the major recycling site, indicating that informal e-waste recycling activities
296 contributed significantly to high concentrations of PTEs in the area. Concentrations of As were below the
297 guidelines value of 20 mg/kg (Kamunda et al., 2016) at all sites.

298

299 In April, when rainfall was lower, there were high correlations between Hg and all other PTEs, except Fe and Zn,
300 with the correlational coefficient ranging from 0.731 with Cr to 0.990 with As (Table 5). However, in June, when
301 there was higher rainfall, Hg had high correlation with As, Cu, Fe, Mn, Ni and Pb ($r = 0.759$ to 0.995), slight strong
302 correlations with Cd ($r = 0.619$) and no correlation with Cr ($r = 0.445$) or Zn ($r = -0.173$) (Table 6). The correlations
303 between Hg concentration and concentrations of other PTEs were generally less strong in June than in April,
304 except for Fe. These changes might have been influenced by the higher mobility of Hg with increased rainfall.
305 Numerous studies relating to the relationship of Hg with other PTEs in soil have confirmed strong associations in
306 small particle size of soil from mining site for example Bueno, Bellido, Rubí, & Ballesta (2009).

307

308 The effect of different rainfall patterns on total mercury concentration

309

310 Climatic conditions are known to influence Hg concentrations and speciation in soil (Blackwell et al. 2014). Since
311 June signifies a period with heavy rainfall and April a period of lighter rainfall in Nigeria, the lower Hg
312 concentration observed in June in area A and B (Table 1) might be a result of the leaching of more mobile forms
313 of Hg from the soil or the washing away of the topsoil itself during episodes of heavy rainfall (Graydon et al.
314 2008; Zuo et al. 2013). However, in areas C and E higher Hg content is observed in June relative to April.

315

316 Mobility of mercury in the area

317

318 The sequential extraction performed on soil samples from areas A and B indicated that the proportion of
319 mobile forms of Hg ranged from 3.23% (14.3 mg/kg) to 23% (3.54 mg/kg) of the total content (Table 7,
320 Figure 3). This fraction could contain Hg in the form of HgO, as found in fluorescent bulbs in e-waste, since
321 this species would be removed in this extraction step. In the mobile form Hg can potentially be released into
322 other environmental compartments such as ground water. Subsequent extraction steps indicated that 74.1%
323 (corresponding to 9.7 mg/kg) to 89.4 % (corresponding to 558 mg/kg) of the total Hg content was tightly
324 bound in non-mobile forms. It is common for Hg to be tightly bound to soil especially to OM which in this
325 case constituted from 12.2 to 16.8 % of the samples. It has been established that Hg has a high affinity for
326 OM and becomes less mobile through Hg-OM bonding (Kwaansa-Ansah et al. 2012; Yang et al. 2007).

327

328 In June, there was an increase in soil OM and a decrease in soil pH in both areas (A and B, see Table 1).
329 There was also an increase in the percentage of semimobile forms of Hg in both areas in June. Semimobile
330 forms of Hg include Hg⁰, amalgams of Hg with other metals, Hg²⁺ complexes and Hg₂Cl₂. Although this
331 fraction is not as readily available as the mobile fraction, its species can readily be converted into mobile
332 forms, especially Hg²⁺ complexes that can be also found in the mobile fraction. The increased percentage of
333 semimobile Hg in both areas may be as a result of higher deposition of atmospheric Hg during rainfall
334 (Hintelmann et al. 2002). However, the semimobile fraction of Hg is often adsorbed by crystalline and
335 amorphous aluminium oxides in soil that can be mobilised with increased acidity (Reis et al. 2010).

336

337 Human health risk due to mercury exposure in Alaba e-waste recycling site

338

339 The dosage received via the principal exposure routes - oral ingestion, inhalation of particulate Hg, dermal
340 absorption and inhalation of Hg vapour - and the subsequent toxicity calculated as hazard indices are shown
341 in Online Resource 1 and Table 8, respectively. Increasing order of Hg exposure was vapour inhalation <
342 inhalation < dermal absorption < oral ingestion for ISLU and vapour inhalation < dermal absorption <
343 inhalation < oral ingestion for SLU. This indicated that, irrespective of the land use considered, exposure to
344 Hg was lowest through inhalation of Hg vapour while oral ingestion was the major route. Oral ingestion is
345 typically the major source of human exposure to Hg in contaminated environments (Rodrigues et al. 2014)
346 through the unintentional ingestion of fine particles of soil (Li et al. 2006b). These fine particles may also
347 be deposited on food that is eventually consumed. In soil, Hg may be absorbed by plants (Li 2013). As
348 expected, regarding the nature of land use, the amount of exposure to Hg as well as the HQs were higher in
349 sensitive land use than insensitive land use by all four exposure routes.

350

351

352 **Fig 3** Effect of rainfall pattern on the mobility of Hg in areas A and B of Alaba e-waste recycling site, Nigeria

353

354 Mercury poses non-carcinogenic risk when HI value is below 1, however risk of exposure to Hg will increase
355 as HI value increases. Apart from areas in which the Hg concentration was below the detection limit, the HI
356 ranged from 0.0369 (area C in June for ISLU) to 132 (area A in April for SLU). As expected, the HI was
357 highest in area A followed by area B and least in area E. The HI in A (both for ISLU and SLU) and B (for
358 SLU only) were above 1 while other areas had HI below 1. This indicates that Hg may pose toxic effect to
359 humans especially in area A which is also used for residential purposes. Although only 10% of ingested
360 inorganic Hg in soil can be absorbed (WHO 1991), the high concentration of Hg in area A may pose a threat
361 to human health. Hence, it is concluded that area A is unsuitable for either sensitive or insensitive land use,
362 and consideration should be given to either soil remediation or the cessation of activities in this area and
363 relocation of the workers. Studies have shown that people working in e-waste recycling sites had elevated
364 levels of Hg in their blood, urine and hair corresponding to e-waste recycling activities (Ni et al. 2014;
365 Srigboh et al. 2016). Thus, people working in Alaba e-waste recycling site should be examined for symptoms
366 of Hg toxicity.

367

368 **Conclusions**

369

370 Although primitive e-waste recycling activities are a major source of anthropogenic soil contamination of
 371 global concern, the mobility of Hg is not often studied. This study showed that Hg concentration and
 372 concentrations of other PTEs in topsoil of the Alaba e-waste recycling site was dependent on the type and
 373 intensity of activities carried out in different locations, with particularly high Hg concentrations (exceeding
 374 600 mg/kg) in the area where the majority of the recycling was carried out. A decrease in Hg soil concentration
 375 at more contaminated locations and less strong associations with other PTEs was observed in June when
 376 rainfall was heavier compared to April, possibly due to the release of more mobile Hg species from the soil
 377 matrix. These more mobile forms of Hg could contain HgO, the most prominent form of Hg found in disused
 378 fluorescent lamps. While Hg was primarily strongly bound to the soil, the HI indicated that Hg may pose a
 379 health threat to workers in the main recycling site, irrespective of what the land is used for. Hence Hg exposure
 380 assessment of the e-waste recyclers are recommended.

381

382 **Declarations**

383 **Funding** O.A. received a Commonwealth Scholarship Commission for award of a Split-site Scholarship

384 **Conflicts of interest/Competing interests** Not applicable

385 **Availability of data and material** Supplementary information is provided as online resource

386 **Code availability** Not applicable

387

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537 **Fig 1 a and b** Nature of e-waste recycling activities in Alaba e-waste recycling site, Nigeria.



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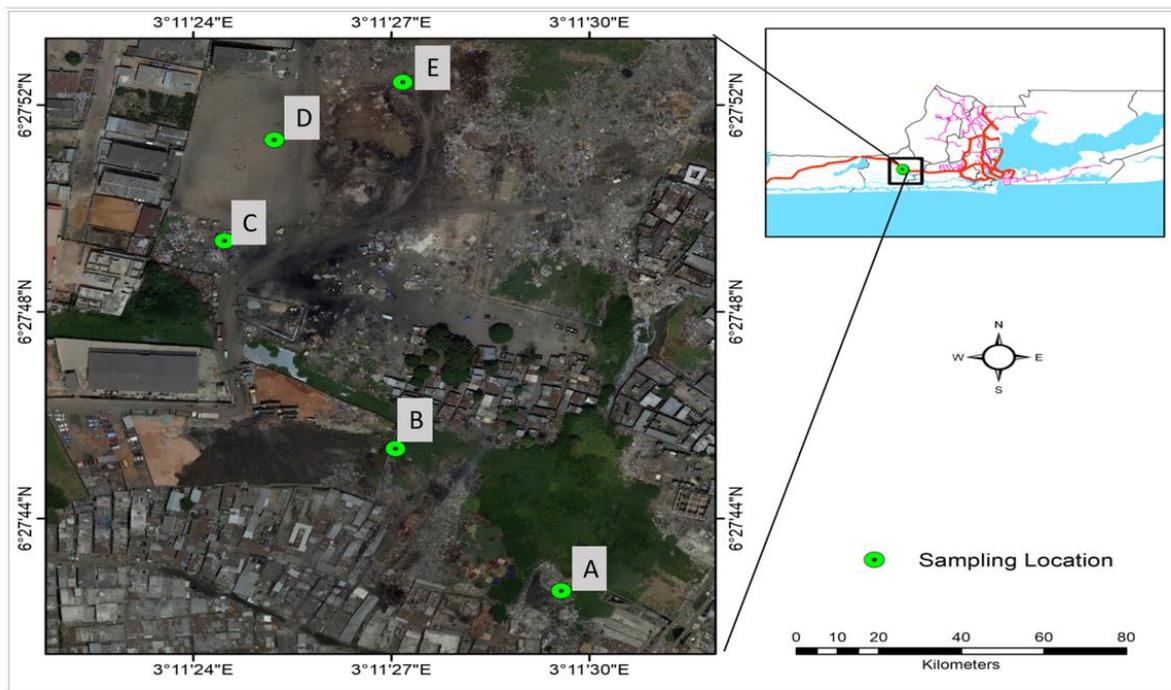
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543 **Fig 2** Map of Alaba International showing the sample locations at e-waste recycling site

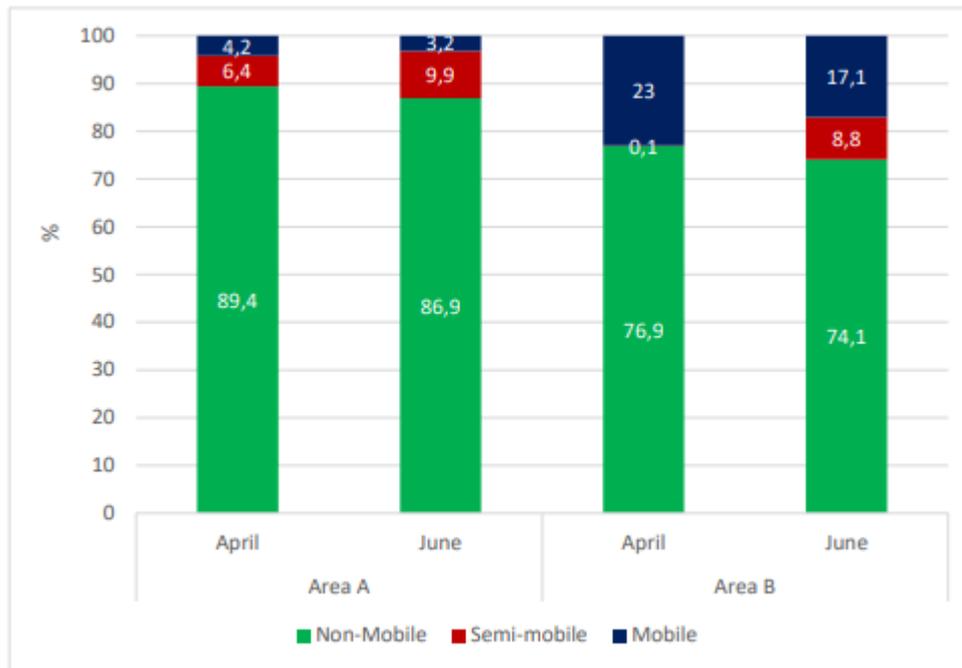
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546 **Fig 3** Effect of rainfall pattern on the mobility of Hg in areas A and B of Alaba e-waste recycling site, Nigeria

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549 **Table 1** Physicochemical characteristics and total mercury concentrations in soil samples from Alaba e-waste
 550 recycling site, Nigeria.

Site	Location		pH		OM (%)		Total Hg content (mg/kg)	
	Long.	Lat.	April	June	April	June	April	June
A	3.191550	6.461833	8.18	8.03	12.9	13.6	624 ± 84	443 ± 53
B	3.190853	6.462597	8.97	7.50	12.2	16.8	15.2 ± 3.8	13.1 ± 1.7
C	3.190883	6.464567	8.03	8.61	5.21	4.65	< 0.07	1.65 ± 0.20
D	3.190133	6.463714	8.36	7.09	1.35	1.40	< 0.07	< 0.07
E	3.190342	6.464258	6.86	7.15	19.6	29.4	< 0.07	1.99 ± 0.25

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553 **Table 2:** Comparison between Hg concentrations in soil samples from other e-waste recycling sites
 554 and in soil from Alaba e-waste recycling site, Nigeria

Location description	Soil depth (cm)	Range or mean concentration of Hg (mg/kg)	Reference
Ibadan, Lagos and Aba Nigeria	0-10	0.5-10.2	(Ohajinwa et al. 2018)
Bangalore, India	-	0.09-59	(Zhang et al. 2012)
Delhi, India	0-15	N.D-0.08	(Pradhan and Kumar 2014)
Korle Lagoon area, Accra, Ghana	0-2	0.67	(Fosu-Mensah et al. 2017)
Guiyu, China	0-20	0.052-0.27	(Li et al. 2011)
Guiyu, China	0-20	1.18	(Zhao et al. 2015)
Taizhou, China	0-15	3.1- 4.9	(Zhang et al. 2014)
Tianjin, China	-	N.D -1.31	(Han et al. 2018)
Longtang, Guangdong Province, China	0-5	0.11-3.42	(Wang et al. 2015)
Alaba, Lagos, Nigeria	0-5	< 0.07-624	This study

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Table 3 Concentrations of potentially toxic elements in soil samples from Alaba e-waste recycling site, Nigeria in April 2016 (expressed as % for Fe; mg/kg for all other elements)

Site	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
A	5.48 ± 0.38	34.9 ± 1.4	106 ± 4.2	13300 ± 195	8.80 ± 0.27	974 ± 38	240 ± 15	1690 ± 56	6600 ± 243
B	0.891 ± 0.092	23.5 ± 0.86	87.3 ± 5.6	1160 ± 125	6.58 ± 0.28	468 ± 18	74 ± 4.1	695 ± 34	3000 ± 180
C	< 0.0119	4.06 ± 0.20	22.7 ± 1.6	735 ± 100	1.57 ± 0.04	262 ± 13	20.9 ± 1.2	245 ± 7.2	1250 ± 17
D	< 0.0119	< 0.000875	6.53 ± 0.33	23.6 ± 1.8	0.27 ± 0.01	48.2 ± 5.1	2.93 ± 0.21	16.1 ± 0.60	98.2 ± 2.7
E	< 0.0119	9.54 ± 1.0	27.7 ± 3.1	2960 ± 368	4.04 ± 0.64	310 ± 35	28.5 ± 2.5	844 ± 103	29800 ± 4100

565 Results are mean ± 1 standard deviation (n=3)

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Table 4 Concentrations of potentially toxic elements in soil samples from Alaba e-waste recycling site, Nigeria in June 2016 (expressed as % for Fe; mg/kg for all other elements)

Site	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
A	6.98 ± 0.40	42.5 ± 4.7	102 ± 11	13000 ± 529	8.78 ± 0.07	1300 ± 39	230 ± 4.1	1680 ± 24	8140 ± 163
B	3.34 ± 0.48	43.4 ± 0.88	136 ± 1.9	1330 ± 33	6.34 ± 0.06	760 ± 3.1	90.1 ± 2.2	1160 ± 15	4060 ± 17
C	< 0.0119	3.75 ± 0.12	22.5 ± 0.82	587 ± 38	1.58 ± 0.10	324 ± 42	23.9 ± 2.7	256 ± 16	1310 ± 79
D	< 0.0119	< 0.000875	6.74 ± 0.64	10.6 ± 2.7	0.25 ± 0.02	29.1 ± 2.0	3.01 ± 0.32	7.74 ± 0.59	39.1 ± 3.8
E	1.79 ± 0.37	6.20 ± 0.11	31.5 ± 2.3	1540 ± 65	3.9 ± 0.20	358 ± 20	44.8 ± 2.8	551 ± 29	76400 ± 3200

574 Results are mean ± 1 standard deviation (n=3)

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579 **Table 5:** Correlational analysis of mercury with other potentially toxic elements in soil samples from the main recycling area in Alaba e-waste recycling site, Nigeria
 580 in April 2016

	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Hg	pH
Cd	0.872										
Cr	0.815	0.987									
Cu	0.967	0.816	0.726								
Fe	-0.253	0.252	0.327	-0.278							
Mn	0.943	0.957	0.915	0.927	0.031						
Ni	0.991	0.925	0.876	0.959	-0.129	0.978					
Pb	0.881	0.908	0.831	0.930	0.081	0.953	0.920				
Zn	-0.111	0.005	-0.103	0.118	0.304	0.035	-0.064	0.321			
Hg	0.990	0.804	0.731	0.980	-0.364	0.910	0.968	0.862	-0.076		
pH	0.183	0.281	0.410	-0.075	0.126	0.127	0.184	-0.125	-0.869	0.089	
OM	0.239	0.501	0.431	0.390	0.572	0.468	0.332	0.669	0.838	0.213	-0.531

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592 **Table 6:** Correlational analysis of mercury with other potentially toxic elements in soil samples from the main recycling area in Alaba e-waste recycling site,
 593 Nigeria, in June 2016

	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Hg	pH
Cd	0.870										
Cr	0.765	0.978									
Cu	0.915	0.643	0.477								
Fe	0.966	0.921	0.863	0.803							
Mn	0.971	0.899	0.808	0.888	0.967						
Ni	0.985	0.826	0.701	0.960	0.930	0.977					
Pb	0.975	0.940	0.874	0.828	0.994	0.984	0.950				
Zn	-0.028	-0.253	-0.211	-0.093	0.049	-0.130	-0.114	-0.055			
Hg	0.888	0.619	0.445	0.995	0.759	0.861	0.943	0.792	-0.173		
pH	0.126	0.136	0.088	0.292	0.143	0.336	0.269	0.191	-0.437	0.307	
OM	0.344	0.254	0.314	0.126	0.481	0.278	0.228	0.385	0.860	0.030	-0.356

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Table 7: Mobility of mercury in soils from areas A and B of Alaba e-waste recycling Site, Nigeria

Sample	A		B	
	April	June	April	June
Hg (mg/kg)				
Total	624 ± 84	443 ± 53	15.2 ± 3.8	13.1 ± 1.7
Non-Mobile	558	385	11.7	9.70
Semi-mobile	39.8 ± 5.9	43.7 ± 2.8	<0.03	1.15 ± 0.28
Mobile	26.7 ± 4.1	14.3 ± 2.8	3.54 ± 0.57	2.24 ± 0.75

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600 **Table 8** Hazard indices of mercury in the e-waste recycling areas depending on the land use

Area	Month	HI for ISLU	HI for SLU
A	April	13.9	132
	June	9.88	93.7
B	April	0.339	3.22
	June	0.292	2.77
D	April	ND	ND
	June	0.0368	0.349
E	April	ND	ND
	June	ND	ND
F	April	ND	ND
	June	0.0444	0.421

601 ND- not determined because mercury concentrations was below detection limit

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