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Leaching of trace metals from mixed electronic waste using four extraction methods

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ABSTRACT

Rapid developments in technology in recent times, coupled with increasing demands for electronic products, have led to a rapid increase in electronic waste (e-waste) in the environment. The leaching of metals from e-wastes into the environment is a potential health and environmental hazard. The present study investigates the leaching potential of fourteen metals (Ag, Al, As, Ca, Cd, Cr, Cu, Fe, K, Mg, Na, Ni, Pb and Zn) in mixed waste printed circuit boards (PCBs) and plastic housing (PH) from recycling/dump sites. The assessment was carried out using rain water, deionized water, synthetic precipitation leaching procedure (SPLP) solution and seawater over a period of 192 hours at regular intervals. The data obtained was compared to TCLP and WET limits. The toxicity characteristic potential of the elements extracted by these solvents were in the order deionized water>rainwater>SPLP>seawater. The levels of the 14 elements studied were generally low and in varying concentrations, with Ca, K, Mg and Na being relatively higher in all the leaching fluids. The results further proved that leaching from PCBs and PH was a significant source of trace metals from recycling/dump sites. Therefore, proper handling and disposal of these waste materials should be encouraged to lessen their impact on the environment.

Keywords: E-waste, Plastics, Printed circuit boards, Metals, Leaching fluids, Toxicity, Dump site

Introduction

Rapid technological advancement, coupled with the proliferation of electrical and electronic equipment (EEE), has made life easier for humankind. Nonetheless, the increasing demand for EEE has resulted in the generation of unwanted, discarded or malfunctioning electrical and electronic appliances (Michael and Sugumar, 2013). Newer EEE are brought into the market, rendering older versions obsolete. This pattern contributes to the soaring quantities of e-waste globally.

Currently e-wastes are known to present potential threats to the environment and human health as a result of growing volumes of the waste stream and the presence of toxic elements. It is estimated that about 20 to 50 million tons of e-waste are generated worldwide each year; less than 5% of all municipal solid waste (Keith *et al.*, 2008). The number of mobile phone subscribers worldwide is reckoned to reach a global population mark of 6.8 billion which will translate into more e-waste components in the years to come. This phenomenal growth is primarily

driven by developing countries, which accounted for over 80% of all new subscriptions as at 2011 (Yadav and Yadav, 2014). This suggests that developing countries, especially Sub-Saharan African countries (including Ghana), will become the hub of e-wastes as EEE approach their end of life (EoL), besides the already existing volumes of e-waste. Other electronic devices also contribute significantly to the waste disposal problem; for instance, about 100 million obsolete computers and televisions are disposed of annually in the United States alone (Hileman, 2006).

Great concerns have been raised over the management practices for waste electrical and electronic equipment (WEEE) in developing countries, where potentially toxic materials are discarded with municipal solid waste at open dump sites and even in surface waters, the sea (Osibanjo and Nnorom, 2007) and on tracts of land (Michael and Sugumar, 2013). A large portion of the e-waste is also recycled. Most e-waste contains toxic inorganic [lead (Pb), cadmium (Cd), mercury (Hg)] and organic [polybrominated diphenyl ethers (PBDEs) and dioxins] substances. The improper disposal of WEEE can pose environmental problems like leaching of heavy metals if not suitably managed (Zhou *et al.*, 2013). Subsequently, the e-waste components corrode and the heavy metals become mobile and travel with the leachate, eventually entering the environment. Thus leaching studies of toxic substances from e-waste in landfill scenarios is very necessary for an assessment of their potential environmental impact in the long run.

Accordingly, several nations have carried out comprehensive studies on environmental contamination resulting from leachability of heavy metals in e-waste (Keith *et al.*, 2008; Kiddee *et al.*, 2013; Oliverira *et al.*, 2012; Yadav *et al.*, 2014) and e-waste recycling activities (Osibanjo and Nnorom, 2007). Findings from these works have enabled policy makers in developed nations to establish legal directives to effectively manage e-waste. However, this is not the case with most third world nations.

In Ghana, there is no report on heavy metal leachability from e-waste materials. This is in addition to the paucity of data on e-waste composition and management. However, the few studies so far conducted have dwelt on e-waste contamination of soils in sites and health hazards among the workers in this industry within the Agbogbloshie vicinity (e.g. Amankwaa, 2014; Nukpezah *et al.*, 2014; Amfu-Otu, 2013; Asante *et al.*, 2012; Atiemo *et al.*, 2012). In the present work, the toxicity potential of metal leaching from mixed e-waste (e.g. PCBs and PH) of WEEE from e-waste recycling/dump sites has been assessed using rain water, deionized water, SPLP and sea water over a period of 192 hours. The data are compared with the prescribed toxicity characteristic leaching procedure (TCLP) limit for classifying the e-waste as hazardous (or not). This work also provides a simulation of the metal toxicity potential of the e-waste discarded at open dump or landfill sites and the effectiveness of the four leaching fluids from WEEE. The study additionally gives information on the metal leaching from mixed e-waste in natural leaching solution (e.g. rainwater, sea water) without using any chemical or buffering agent.

Methods

Sample Collection and Preparation

E-waste materials litter the environment in heaps in many locations across the country, particularly the regional cities. In this study the e-waste materials were randomly collected from two large e-waste recycling centres/dump sites at Agbogbloshie and Ashaiman motorway (Fig 1). The components consisted mainly of discarded plastic housing and printed circuit boards from different types of e-waste products (e.g. mobile phones, printers, computers, televisions, etc.). The components were reduced in size (about 1cm) by crushing them with hammer before subjecting them to the leaching test.

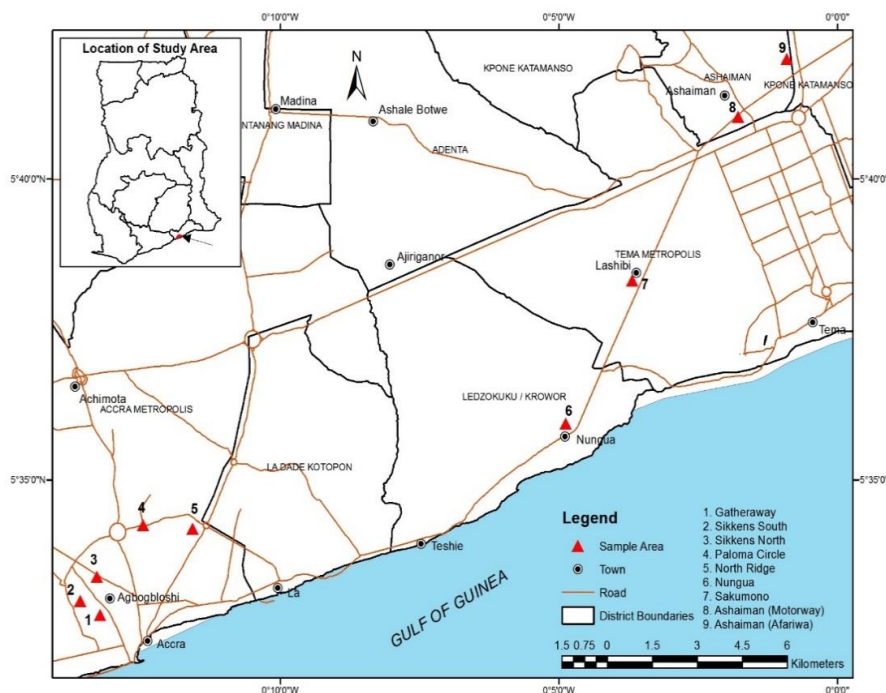


Fig. 1: Map of study area

Leaching Solution

Synthetic precipitation leaching procedure (SPLP) extraction fluid, a mixture of sulphuric (H_2SO_4) and nitric (HNO_3) acids (3:2), was added to water until pH reached 4.2 ± 0.1 . Sea water samples were collected from the Nungua beach in Accra Metropolis along with rain water in the month of June 2014 using acid cleaned polypropylene bottles. De-ionized water used was obtained from the Department of Chemistry, University of Ghana, Legon, Ghana.

Leaching Test

This laboratory experiment was designed to mimic the attitude of many Ghanaians in disposing of wastewater or sachet water on heaped piles of wastes (e.g. garbage, e-waste, etc.) besides other environmental conditions (rainfall and sea sprays). Based on this, four (4) different solvents (de-ionized water, rain water, sea water and SPLP) were chosen for the leaching test. Fifty grams (50

g) of sample (a mixture of plastic housing and circuit boards) was used for each solvent in a liquid to solid ratio of 4:1 in eight batches corresponding to 24, 48, 72, 96, 120, 144, 168 and 192 hours. The leaching experiment was carried out in a column, with gentle stirring. The leachates were drained according to the defined time periods for each extractant. The modified synthetic precipitation leaching procedure (SPLP) was also adopted to evaluate the leachability of contaminants from soils and waste samples under acid precipitation conditions (Li *et al.*, 2009). The leachates were acid digested, cooled, diluted and finally filtered through a filter paper (0.45 μ m pore size) using the pressure filtration pump, then analyzed with ICP-OES for 14 selected trace metals (silver (Ag), aluminum (Al), arsenic (As), calcium (Ca), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), potassium (K), magnesium (Mg), sodium (Na), nickel (Ni), lead (Pb), and zinc (Zn)).

Quality Control and Assurance

For quality control and assurance purposes, the leaching tests were performed in triplicates to ensure accuracy and reproducibility. A procedural blank and solvent blank were run for each batch of samples. Recoveries for surrogate standard used were in the range of 86-105 %. All glass and plastic wares used were cleaned with 5% nitric acid (HNO_3) overnight and rinsed thoroughly with de-ionized water before being dried and stored. Analytical grade reagents were used.

Data Analysis

The experimental data obtained were evaluated by descriptive statistics using Microsoft Excel.

Results and discussion

The concentrations of 14 trace metals detected in four leaching fluids (rain water, deionized water synthetic precipitated leaching procedure-SPLP solution and sea water) over 24, 48, 72, 96, 120, 144, 168 and 192 hours are presented in Tables 1-5. The studied elements were present in varying concentrations in the 4 different extraction fluids. Averagely, the macro elements (Na, Ca, K, Mg), and Fe and Zn concentrations in general were extremely high, particularly in de-ionized and rain water, except the SPLP solution. However, the mean concentrations of As, Cd, Cr, and Pb were the lowest for all the solvents. Overall, the toxicity characteristic potential of the elements extracted by these solvents were in the order deionized water>rainwater>SPLP>seawater.

Table 1: Average concentrations (mg/L) of trace metals in the four leaching solutions

Sample	Rain water	Sea water	SPLP solution	De-ionized water
Ag	0.031	0.027	0.099	0.065
Al	12.31	3.86	9.72	11.59
As	0.024	0.014	0.021	0.068
Ca	1083	157	7.39	1267
Cd	0.02	0.005	0.021	0.690
Cr	0.002	0.213	0.035	0.159
Cu	14.15	0.145	0.282	15.23
Fe	72.83	2.95	5.33	48.20
K	238	38.94	49.98	295
Mg	105	27.75	0.645	123
Na	494	10.88	24.29	614
Ni	3.68	1.03	1.56	30.20
Pb	23.79	0.001	0.004	0.002
Zn	42.85	0.813	1.43	68.47

Rain water

Table 2: Mean concentrations (mg/L) of extractable heavy metals in e-waste using rain water.

Sample	24 hrs	48 hrs	72 hrs	96 hrs	120 hrs	144 hrs	168 hrs	192 hrs	Average	Std	Min	Max	TCLP Limit
Ag	0.002	0.019	0.011	0.019	0.025	0.045	0.049	0.079	0.031	0.025	0.002	0.079	5
Al	2.12	1.58	2.20	3.30	3.41	9.66	10.25	66.00	12.31	21.96	1.58	66.00	-
As	nd	nd	nd	0.019	0.029	nd	nd	nd	0.024	0.007	0.019	0.029	5
Ca	617	866	861	940	1001	946	1185	2249	1083	497	617	2249	-
Cd	0.011	0.012	0.012	0.013	0.014	0.016	0.022	0.057	0.02	0.016	0.011	0.057	1
Cr	0.001	0.002	0.002	0.002	0.002	0.002	0.002	0.003	0.002	0.001	0.001	0.003	5
Cu	2.86	7.95	4.76	9.64	9.70	14.46	14.97	48.83	14.15	14.63	2.86	48.83	-
Fe	11.67	62.50	53.00	31.69	80.50	97.25	113	133	72.83	41.10	11.67	133	-
K	167	190	196	216.87	229	255	228	418	238	77.94	167	418	-
Mg	69.24	80.95	85.42	90.98	96.96	96.39	215	102	105	45.84	69.24	215	-
Na	369	398	424	560	392	655	539	613	494	111	369	655	-
Ni	1.34	2.04	2.61	3.80	4.85	4.30	4.21	6.32	3.68	1.62	1.34	6.32	-
Pb	19.63	30.62	56.54	63.18	1.53	3.26	7.31	8.24	23.79	24.26	1.53	63.18	5
Zn	22.54	29.42	34.78	34.23	34.53	41.45	52.02	93.86	42.85	22.33	22.54	93.86	-

hrs: hours, std: standard deviation, min: minimum value, max: maximum value, nd: not detected

Table 2 shows the distribution and levels of the leachable metals in rain water for the entire test period of 192 hours. The 14 different metals were detected in varying concentrations, with many of them having low concentrations except the elements Na, K, Fe, Mg, Ca, and Zn which were relatively higher, with mean values 494 (Na), 238 (K), 72.83 (Fe), 105 (Mg), 1083 (Ca), and 42.85 (Zn) mg/L respectively. This depicts the different solubility properties of the metals; thus Ca had the highest concentration among all elements in the rain water. The levels of Zn in the leachate were quite high and its presence is attributed to its use as a coating for ferrous metals. This may be likened to the report by other researchers (e.g. Yadav and Yadav, 2014; Li *et al.*, 2009) who demonstrated that Zn as an external layer dissolves first; hence being high with almost uniform concentrations. Fe also had a relatively high overall concentration, with the observed pattern indicating an increase in concentration with time. Furthermore, the presence of Al and Cu is explained by their use as small connecting wires embedded in the plastics and as a major component of most EEEs, especially the hard disc drives of computers, respectively (Li *et al.*, 2009; Yadav *et al.*, 2014). The presence of Ni with an average concentration

of 3.68mg/L is due to its application as alloy or the metal parts on the printed wire board (PWB) (Vuorinen *et al.*, 2006; Yadav *et al.*, 2014).

By contrast, the concentrations of well-known toxic heavy metals in e-waste, such as As, Cr and Cd, were very low. The behavior of As could not be explained. It was only detected in the 4th and 5th days averaging 0.24mg/L whereas Cr and Cd leached in minimal quantities with an average accumulation of 0.02 and 0.002mg/L respectively. Their presence is attributed to their addition as plastic polymers (pigments), UV stabilizers, etc. (Piorek, 2004; Nnorom and Osibanjo, 2009).

Pb concentration ranged from 1.53 to 63.18 mg/L with an average of 23.79 mg/L which exceeded the TCLP limit of 5 mg/L (Table 2). The concentration of Pb leached in rainwater was much higher than the other three leaching fluids (Fig 2). However, these values are lower compared to those from previous studies. Lincoln *et al.* (2007) reported 87.4 mg/L Pb and Keith *et al.* (2008) reported 83.3 mg/L Pb in PWBs. The major source of Pb is Pb-containing solders on motherboards. Overall, rainwater is slightly acidic and very low in dissolved minerals; as such, it is relatively aggressive during leaching.

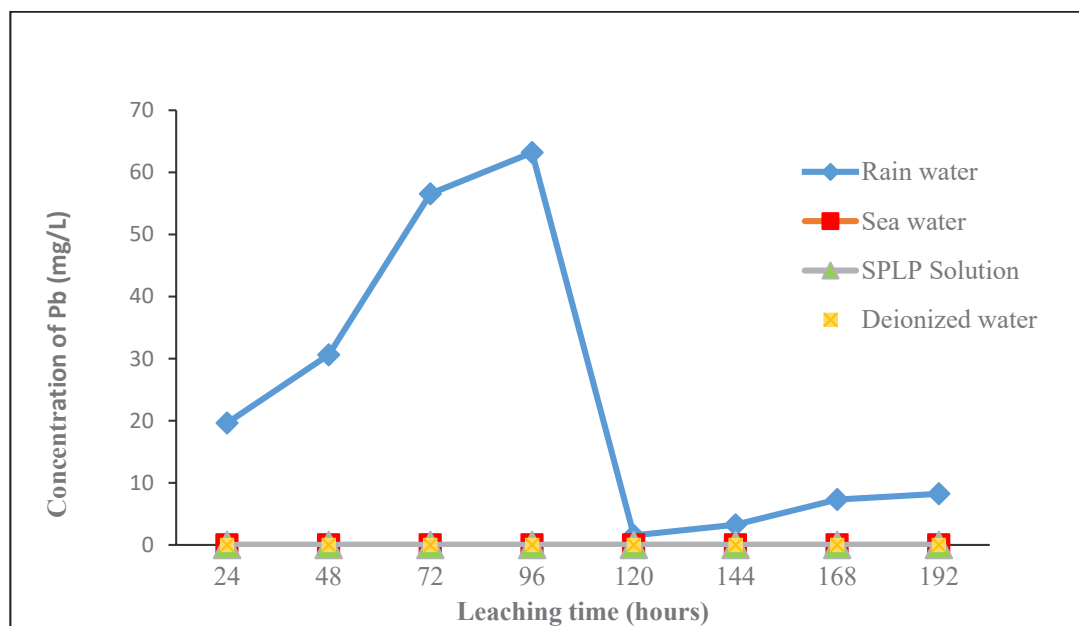


Fig 2: Leaching pattern of Lead (Pb) in four leaching fluids

Sea water

Leaching results from 14 elements using sea water are presented in Table 3. Among the leaching solutions used, sea water had the lowest concentration of metals. However, similar to rainwater, K, Ca, Na, and Mg were relatively higher compared to other elements present in the sea water leachate. According to Keith *et al.* (2008), the amount of elements leached is usually dependent on the type of extraction fluid. Therefore, the low levels of metals in this extraction fluid might be due to the saturated nature of the sea water.

Among the eight heavy metals on the US-EPA D list of toxic metals, Ag, As, Cd, Cr and Pb did not exceed their TCLP threshold limits with average concentrations of 0.027, 0.014, 0.005, 0.213 and 0.001 mg/L respectively (Table 3). Pb had concentrations of 0.001 mg/L throughout the leaching period except the first 24 hours. The toxicity characteristic potential of the elements in sea water was almost non-existent as elements detected were very low.

Table 3: Mean concentrations (mg/L) of extractable heavy metals in e-waste using sea water.

Sample	24 hrs	48 hrs	72 hrs	96 hrs	120 hrs	144 hrs	168 hrs	192 hrs	Average	Std	Min	Max	TCLP Limit
Ag	0.018	0.022	0.024	0.023	0.028	0.035	0.026	0.038	0.027	0.007	0.018	0.038	5
Al	1.12	1.57	2.19	3.83	3.20	7.15	5.42	6.40	3.86	2.26	1.12	7.15	-
As	nd	nd	nd	0.01	0.012	0.015	nd	0.017	0.014	0.003	0.01	0.017	5
Ca	139	147	151	157	165	166	167	166	157	10.62	139	167	-
Cd	0.001	0.003	0.004	0.005	0.006	0.01	0.007	0.005	0.005	0.003	0.001	0.01	1
Cr	0.053	0.105	0.174	0.225	0.312	0.438	0.367	0.032	0.213	0.149	0.032	0.438	5
Cu	0.263	0.131	0.119	nd	0.006	nd	nd	0.206	0.145	0.097	0.006	0.263	-
Fe	0.725	1.49	4.01	2.94	4.08	5.42	2.98	1.96	2.95	1.54	0.725	5.42	-
K	33.52	35.09	35.00	37.40	37.75	40.94	49.13	42.66	38.94	5.15	33.52	49.13	-
Mg	27.66	27.66	27.69	27.71	27.73	27.78	27.90	27.88	27.75	0.095	27.66	27.90	-
Na	6.58	8.50	9.63	11.21	11.32	12.48	14.41	12.93	10.88	2.54	6.58	14.41	-
Ni	0.603	0.660	0.840	0.965	1.10	1.23	1.52	1.34	1.03	0.325	0.603	1.52	-
Pb	nd	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0	0.001	0.001	5
Zn	0.650	1.28	0.856	0.831	0.769	0.500	0.944	0.675	0.813	0.232	0.500	1.28	-

hrs: hours, std: standard deviation, min: minimum value, max: maximum value, nd: not detected

SPLP

Table 4 shows the concentrations of trace metals from the SPLP solution. Generally, metals leached minimally over the entire period. Except for K and Na, the remaining metals recorded mean levels below 10 mg/L in the entire leaching cycle. Concentrations of K and Na ranged from 40.93 to 62.20 mg/L and from 14.05 to 36.15 mg/L respectively. The leaching pattern for the SPLP extraction followed the same order as the other

extraction fluids. The pattern was a little more consistent for concentrations of leached metals throughout the cycles at regular intervals. Compared to previous studies (e.g. Li *et al.*, 2009; Zhou *et al.*, 2013), SPLP leached minimally in this work, which could be a result of the differences in environmental conditions as well as the technique and method employed.

Table 4: Mean concentrations (mg/L) of extractable heavy metals in e-waste using SPLP solution.

Sample	24 hrs	48 hrs	72 hrs	96 hrs	120 hrs	144 hrs	168 hrs	192 hrs	Average	Std	Min	Max	TCLP Limit
Ag	0.021	0.034	0.044	0.055	0.066	0.085	0.299	0.184	0.099	0.095	0.021	0.299	5
Al	2.99	6.30	7.73	9.67	13.61	12.47	13.13	11.85	9.72	3.78	2.99	13.61	-
As	0.017	0.018	Nd	0.019	0.021	0.023	0.024	0.022	0.021	0.003	0.017	0.024	5
Ca	6.76	6.03	7.74	6.23	6.76	6.91	10.52	8.14	7.39	1.45	6.03	10.52	-
Cd	0.008	0.023	0.037	0.045	0.006	0.007	0.023	0.016	0.021	0.014	0.006	0.045	1
Cr	0.006	0.013	0.028	0.042	0.053	0.085	0.03	0.021	0.035	0.025	0.006	0.085	5
Cu	0.069	0.119	0.238	0.263	0.331	0.356	0.438	0.438	0.282	0.137	0.069	0.438	-
Fe	1.84	3.37	4.53	5.21	5.73	6.47	7.85	7.68	5.33	2.07	1.84	7.85	-
K	40.93	43.66	45.97	44.52	49.34	53.83	62.20	59.38	49.98	7.75	40.93	62.20	-
Mg	0.463	0.506	0.588	0.613	0.638	0.675	0.981	0.694	0.645	0.157	0.463	0.981	-
Na	14.06	16.51	16.87	22.17	23.12	32.17	36.15	33.29	24.29	8.54	14.06	36.15	-
Ni	1.13	1.25	1.34	1.50	1.59	1.75	2.09	1.87	1.56	0.33	1.13	2.09	-
Pb	0.003	0.003	0.004	0.004	0.004	0.004	0.004	0.004	0.004	0.000	0.003	0.004	5
Zn	0.075	0.619	0.850	0.888	1.29	1.42	3.83	2.47	1.43	1.19	0.075	3.83	-

hrs: hours, std: standard deviation, min: minimum value, max: maximum

De-ionized water

Table 5 displays the concentrations of elements from the e-waste pile in de-ionized water. In all, the amount of metals leached from deionized water was higher than the other three leaching solutions but followed a similar trend to that of the rainwater. Comparing this to sea water which had the least leaching, it is clear that deionized water is devoid of ions and species that did not enhance the leaching of elements. Ca had the highest concentration of the leached metals ranging from 2695 to 2694 mg/L. Additionally, Na, K and Mg had high concentrations with the respective averages of 614, 295, and 123 mg/L. Fe, Zn, and Ni, considered as having potential toxicity characteristics, were relatively high and leached in a gradual manner to the end (Fig. 3). Another

pattern typified by Cr, Cd, and Ag is shown in Fig 4. Here, the metal levels increased gradually and then sharply towards the end of the leaching cycle. On the other hand, Ni concentrations exceeded the waste extraction test (WET) regulatory limit of 20 mg/L (CDTSC, 2006).

Of the toxic elements, As, Pb and Cd, Cd was the highest with an accumulated concentration of 3.28 mg/L over the entire period and an average value of 0.69 mg/L. This concentration of Cd exceeded the TCLP and WET regulatory limits of 1.0 mg/L. Pb was almost constant in concentration in the hours found, averaging 0.002 mg/L while As averaged 0.068 mg/L. Thus As, Cd and Pb concentrations were within TCLP limits.

Table 5: Mean concentrations (mg/L) of extractable heavy metals in piled e-waste material using de-ionized water.

Sample	24 hrs	48 hrs	72 hrs	96 hrs	120 hrs	144 hrs	168 hrs	192 hrs	Average	Std	Min	Max	TCLP Limit
Ag	0.148	0.105	0.018	0.018	0.057	0.061	0.037	0.079	0.065	0.045	0.018	0.079	5
Al	27.25	15.50	32.65	2.45	3.37	3.42	2.55	5.53	11.59	12.19	2.45	5.53	-
As	0.069	0.016	0.223	nd	0.018	0.016	nd	nd	0.068	0.089	0.016	0.233	5
Ca	738	858	1081	1177	1169	1177	1245	2695	1267	603	738	2695	-
Cd	0.093	0.148	0.184	0.243	0.277	0.308	0.993	3.28	0.690	1.08	0.093	3.28	1
Cr	0.090	0.108	0.129	0.141	0.141	0.183	0.201	0.276	0.159	0.06	0.090	0.276	5
Cu	6.33	6.93	7.73	8.07	9.65	11.20	17.74	54.17	15.23	16.15	6.33	54.17	-
Fe	9.60	39.16	41.02	41.68	46.20	54.70	63.65	89.59	48.20	22.88	9.60	89.59	-
K	183	193	232	290	291	321	313	539	295	112	183	539	-
Mg	79.80	82.10	101	107	111	128	134	244	123	52.18	79.80	244	-
Na	398	485	549	588	652	678	719	842	614	140	398	842	-
Ni	13.89	18.73	29.82	29.22	29.24	33.26	36.32	51.13	30.20	11.24	13.89	51.13	-
Pb	nd	nd	0.001	0.001	0.001	0.001	0.003	0.003	0.002	0.001	0.001	0.003	5
Zn	17.59	26.40	58.57	59.75	82.74	82.59	95.33	125	68.47	35.51	17.59	125	-

hrs: hours, std: standard deviation, min: minimum value, max: maximum value

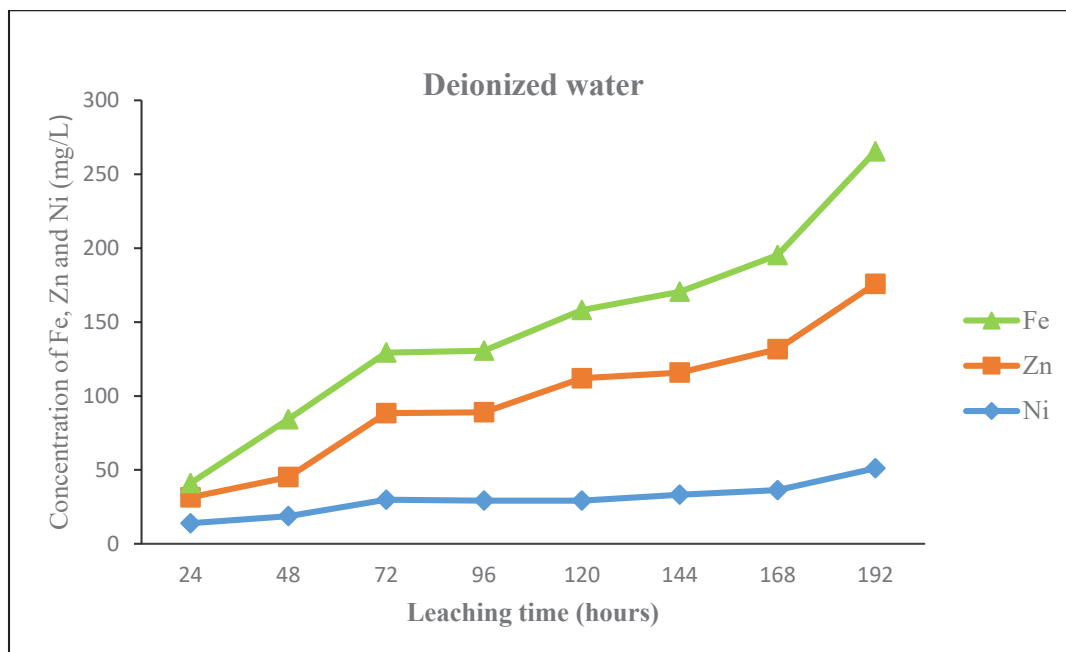


Fig. 3.: Leaching pattern of Fe, Zn and Ni in deionized water

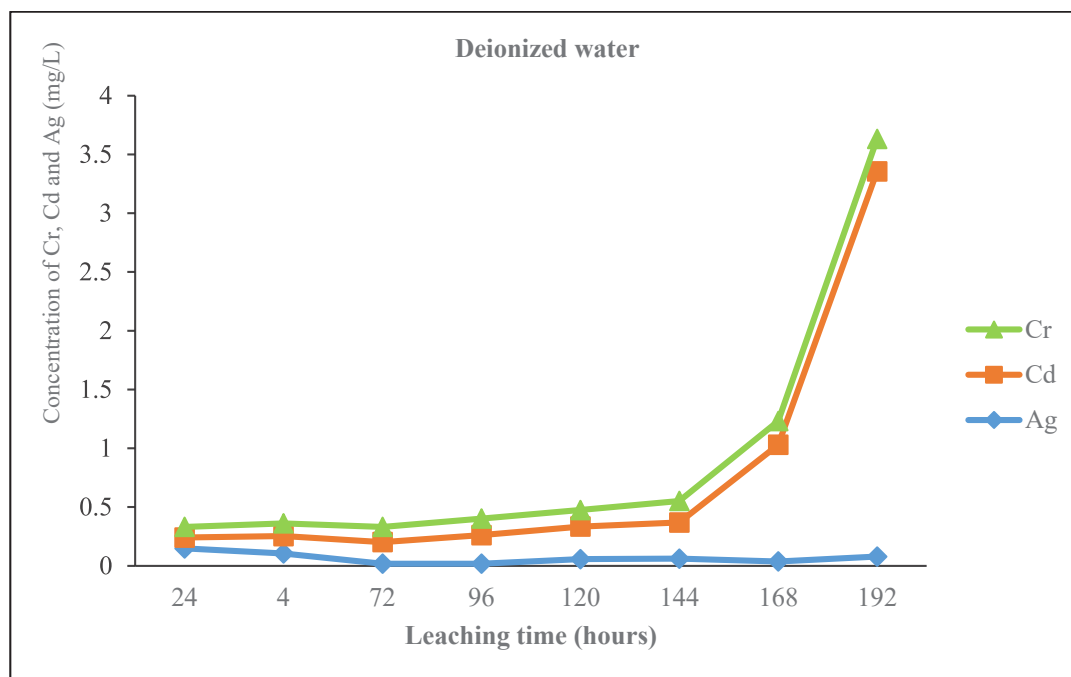


Fig. 4: Leaching pattern of Cr, Cd and Ag in deionized water

Conclusion

The leachability of 14 elements from four extraction fluids, notably rain water, sea water, deionized water and SPLP solution was studied. De-ionized water was the most predominant extraction fluid from which significant elements could leach in relatively higher concentrations while sea water extraction fluid showed the least leaching. Ca, K, Mg and Na were relatively higher in all the leaching fluids.

Comparing results obtained in this study for rainwater with the TCLP limit, concentrations of Pb were about 4-12 times higher than the TCLP limit of 5mg/L.

The results further confirmed that these trace elements do leach out into the environment, which could pose a significant danger to human health and the environment due to the poor management practices of these waste materials. Therefore, proper handling and disposal of these waste materials can be embarked on to mitigate the environmental impact. Further studies could be carried

out, investigating several types of e-waste components such as steel housing, batteries, and CRTs.

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