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Environmental impact assessment of waste electronic and electric equipment (WEEE) management practices in developing countries through leaching tests

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Increasing quantities of domestic and imported waste electronic and electric equipment (WEEE) stimulates a rapid development of informal recycling activities in developing countries. This economically driven recycling, oftentimes based on open burning or acid-base treatment, followed by open dumping of the residues, does not usually fit environmental and public health requirements. For the underlying research, the hypothesis was formulated that the specific conditions of dumping of residues from informal recycling activities are enhancing the leaching of hazardous substances into the ground- or surface water. Hence, this paper aims at identifying major influencing factors that affect leaching behaviour of heavy metals from WEEE components and assess environmental impact of informal WEEE recycling and dumping practice. A set of leaching tests were carried out with a homogenized sample of printed circuit boards from personal computers. The results of the study show that the concentrations of Pb, Zn, Cd, Ni and Cu leached from the test material, is higher than the Dutch standard of heavy metals reference value in soil. In addition, under the toxicity characteristics leaching procedure (TCLP) extraction test, 6879 ± 1342 mg/kg dissolved organic carbon (DOC) was measured. It was also confirmed that the leachability of Pb and Cd exceeds the regulatory limit of TCLP. Lead was the predominant heavy metal to leach from the waste material and 5617±739 mg/kg Pb was released by the standard pH-controlled leaching test. The column leaching test results suggested that high amount concentrations of heavy metals released were washed out before a liquid to solid (L/S) ratios of 5. Similarly, from the column leaching trend 99.3% of DOC was leached in the lower L/S ratios of (upto L/S ratio of 2). The standard pH controlled test results indicated that heavy metals were highly mobile in the acidic pH ranges.

Key words: Printed circuit boards (PCBs), e-waste, leaching tests, heavy metals, dissolved organic carbon (DOC).

INTRODUCTION

Due to decreasing costs and increasing availability, the lifespan of some of electronic equipment is very short. For example, computers in the early 1980s were used on average of about ten years but their lifespan has since

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For example, computers in the early 1980s were used on average of about ten years but their lifespan has since reduced to an average of about three years. This is due to the rapid and continual improvements in technology, which quickly outdates older models (Brigden et al., 2005). As a consequence of the increasing market expansion and short lifespan in electrical and electronic goods, the waste stream of these products is fast growing and much of this electronic waste goes into the normal municipal waste stream to be landfilled or incineration. Waste electronic and electrical equipments (WEEE) comprises more than 5% of all municipal solid waste by weight, nearly the same amount as all plastic packaging, and WEEE is growing steadily (UNEP, *2005*). In rich countries, WEEE may constitute 8% by volume of municipal waste (Widmer et al., 2005).

WEEE can contain over one thousand different substances, many of which are toxic and some of which have a relatively high market value when extracted. Inadequate disposal and poor recycling practices to recover metals such as gold, copper and silver contribute to potential harmful impacts on the environment and pose health risks to exposed individuals. The WEEE stream is thus important not only in terms of quantity but also in terms of its toxicity (Hicks et al., 2005). According to UNEP (2005) report, the vast majority of WEEE ends up in the landfills or incinerators and about 70% of the heavy metals (including mercury and cadmium) found in landfills come from discarded electronic equipments (Grossman, 2006).

As compared to waste from other manufactured products that is used for recycling (cans, bottles and paper), WEEE is a particularly complex kind of waste stuff. For the most part, these substances do not pose health hazards while the equipment is intact. However, when electronics are physically damaged, dismantled or improperly disposed of, their toxins emerge (Grossman, 2006). If the WEEE components corrode and the heavy metals become mobile, the metals will travel with leachate and eventually enter into the environment (Li et al., 2009).

Printed circuit boards (PCBs) are the foundation for almost all electronic products in the world and waste PCBs include the types which generate from retired electronic products as well as from the PCBs production process (Xiang et al., 2007). In composition, printed circuit boards contain metals, ceramics and polymers. Heavy metals such as lead and copper make printed circuit boards interesting raw materials both from the environmental and economical point of view (Cui and Forssberg, 2007).

Developing countries such as India, China, Thailand, Ghana and Nigeria are host of large and cheap informal recycling as well as recovery of WEEE (Visvanathan and Yin, 2009). In these workshops, obsolete electronic chips are removed from circuit boards and dissolved in strong acidic solutions to recover gold, silver, palladium and platinum (SVTC and BAN, 2002). The recycling activities in these informal sectors are conducted by a range of legal, unregistered and publicly accepted businesses who give little concern to illegal processes which have consequences of great concern to the environment and human health. According to review reports, open burning of printed circuit boards (PCBs) for component separation or for solder recovery, burning cables (wires) to recover copper, gold recovery from PCBs with cyanide salt leaching or nitric acid and mercury amalgamation are part of the recycling techniques that facilitates the release of the toxic substances to the environment (Sepulveda et al., 2009).

To estimate the environmental threat, leaching tests have proven to be indispensable characterization tools. These leaching tests yield more relevant information than for instance a component composition: they yield information about the leaching behaviour of materials in natural conditions and outdoor situations (Hage and Mulder, 2004).

Based on this, Li et al. (2006) performed heavy metal leaching tests on obsolete personal computer components using the toxicity characteristics leaching procedure (TCLP) (USEPA, 1996). The report indicates that lead was the predominant species which exceeded the toxicity characteristic (TC) limit. The lead concentration in the TCLP extract of printed circuit boards was found ranging from 150 to 500 mg/l which is 30 to 100 times the regulatory level of 5 mg/l.

This paper therefore, aims at identifying major influencing factors for the leaching behaviour of heavy metals from waste printed circuit boards of obsolete personal computers; providing quantitative data for the leaching potential for several dumping scenarios and assess environmental impact of informal WEEE recycling and dumping practice using various leaching test procedures: column leaching (PrEN 14405), batch leaching (EN 12457-4), pH controlled leaching (CEN/TS 14429) and toxicity characteristics leaching procedure (TCLP) extraction tests (SW-846, method 1311) under different environmental conditions.

MATERIALS AND METHODS

Sampling and sample treatment

Obsolete personal computers were disassembled and their printed circuit boards (PCBs) were separated from a solid waste collecting and sorting site. The dismantled PCBs containing resistors, chips and capacitators were crushed or shredded into pieces. The crushed test samples were sieved in three particle size ranges; the first sample portion contains particles that can pass through 10 mm sieve size, the second sample portion contains particles that can pass through a sieve size of 5.6 mm and the third portion contains particles that can pass through a sieve size of 1.4 mm.

Experiments

To characterize the waste materials, four standard leaching test procedures were conducted.

Experiment 1

Standard batch leaching test (EN 12457-4): In this compliance test, the sample material, after pre-treatment, has a particle size below 10 mm, which was brought into contact with de-mineralized water at a liquid to solid ratio of 10 and agitated for 24 h using an end-over-end rotary tumbler at 10 rpm (CEN, 2002).

Experiment 2

Standard toxicity characteristics leaching procedures (TCLP) extraction test, SW- 846, method 1311 (USEPA, 1996): In this test, the sample material, after pre-treatment, with a particle size below 10 mm, is brought into contact with extraction fluid prepared from glacial acetic acid, sodium hydroxide and de-mineralized water at a liquid to solid ratio of 20 and agitated for 20 h using an end-over-end tumbler at 15 rpm. The TCLP recommends the rotation of the samples at 30 ± 2 rpm. However, reports indicate that there is no significance difference between results of samples rotated at 28 rpm and samples rotated at 13 rpm (Vann et al., 2006b).

Experiment 3

Standard pH controlled leaching test, CEN/TS 14429 (CEN, 2005): This test separated test portions contact with leachant containing pre-selected amounts of acid or base at L/S ratio of 10 and extracted for 48 h using an end-over-end rotary tumbler at 10 rpm. Each leachant is added in three steps in the beginning of the test. The pH-values ranged from pH 3 to 12 and the amounts of acid or base needed to cover the pH ranges was determined by preliminary titration.

Experiment 4

A standard column (up-flow percolation) test, PrEN 14405 (CEN, 2004): A known amount of the test portion was packed in three different columns and leachant (de-mineralized water) was percolating up-flow through the columns using pumps at an average flow rate of 13.5 ml/h. The eluate was collected in seven separate fractions at a cumulative L/S ratio of 0.1, 0.2, 0.5, 1, 3, 5 and 10 from the top section of the columns.

Analytical work

All the above experiments were performed in triplicate and blanks were included for each leaching experiment. pH, redox-potential (E_h) and electrical conductivity (Ec) were measured immediately after the collection of each eluate fractions. All the measurements of pH, Ec and Eh were made using the WTW model 340i mobile pH meter, WTW model 330i Ec meter and platinum triode Eh meter. The pH meter was calibrated regularly with standard buffer solutions. After measuring E_h, E_c and pH, the eluate samples were filtered, then each filtrate was divided for heavy metals and dissolved carbon analyses and preserved below 4°C. Except for the eluate, fractions for heavy metals analysis were also acidified with 1 M HNO_3 (EN –ISO 5667-3).

All the heavy metals except for arsenic in the eluate samples were analyzed using inductively coupled plasma-optical emission spectrometer (ICP-OES). Screening of arsenic from the selected eluate samples was made using atomic absorption spectrometer (AAS). DOC was analyzed using TOC analyzer. All the elute samples analyzed for DOC were passed through a 0.45 µm diameter membrane filter paper pre-treated with milli-Q water to get rid-off its organic content. Moreover, all the dilutions of eluate samples for DOC analyses were also done using milli-Q water, which contains very low DOC in it.

RESULTS

Standard column leaching test (PrEN 14405)

The column leaching test results showed that zinc, lead, cadmium, manganese, nickel, copper and chromium were washed out from the waste material. The highest concentrations were generally released in the lowest liquid to solid (L/S) ratios. The released concentrations went decreasing while the L/S ratios increased (Figure 1). The concentration of chromium leached was below the detection limits (0.02 mg/l).

The cumulative released values of the metals indicated that manganese is more solubility controlled than zinc, lead, nickel, copper and cadmium. Chromium is the least soluble heavy metal by the column leaching test (Figure 2).

Standard pH controlled leaching test

The pH controlled leaching test results indicated that the mobility of metals is strongly pH dependent. The highest concentrations were generally leached under acidic conditions. Similar leaching trends were observed for Pb, Zn, Ni and Mn. The release concentrations of these metals increased with decreasing pH; however, considerable amounts were also recorded under alkaline pH ranges. On the other hand, the leaching pattern of Cu showed a relatively v-shaped leaching trend. The leaching pattern of Cd seems pH independent (Figure 3).

Batch leaching and toxicity characteristics leaching procedures (TCLP) extraction tests

High concentrations of Pb, Zn, Cr, Cd, Mn and Ni were leached by the TCLP test method result. Cu was not released from the test material under this test method. Particularly, the highest released concentrations of Zn, Cr, Cd and Mn were recorded by the TCLP test when compared with the other three test methods. For instance, 67.7 ± 6.0 mg/kg of Cd was released under the TCLP extraction test, however the maximum released concentration of the metal under pH controlled (at pH 6), column leaching and batch leaching tests was 0.21 ± 0.08, 0.08 ± 0.02 and 0.12 ± 0.006 mg/kg, respectively.

Maximum leachability of metals was observed under

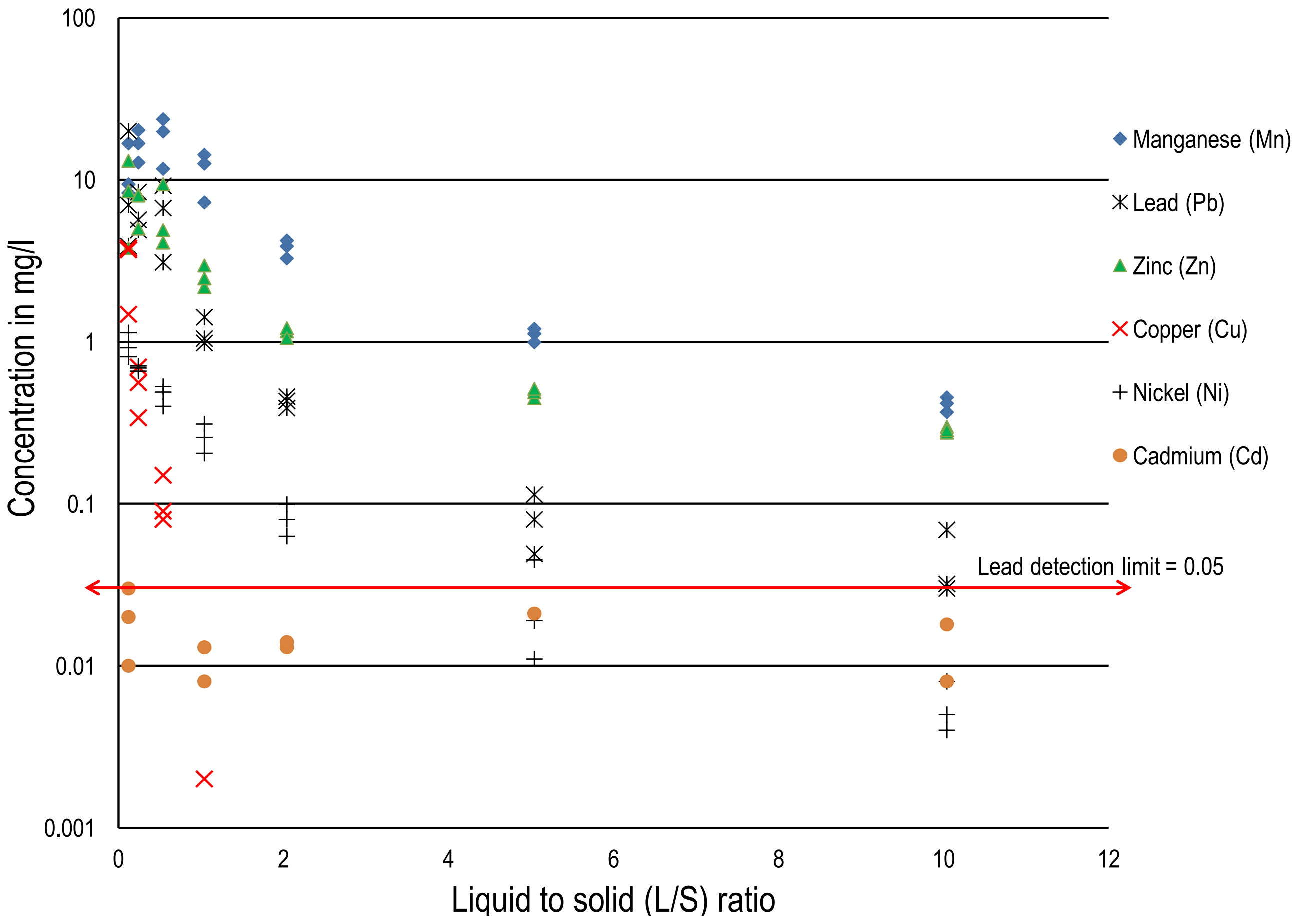


Figure 1. The solubility trend of Mn, Zn, Pb, Cu, Ni and Cd under the standard column leaching test conditions (tests were done in triplicate).

the pH controlled (at pH 3) and TCLP leaching tests. Based on this, Pb was detected as the highest leached heavy metal under the standard pH controlled leaching test and the leachability pattern of the other heavy metals was in the order of Cd < Zn < Cr < Ni < Mn < Cu regardless of the two test methods conditions. Maximum leachability of the metals in percentage is presented in Table 1.

Figure 4 shows that maximum concentrations of lead, copper and nickel (5617 ± 739, 38 ± 8.4 and 29.1 ± 6 respectively) was determined by the pH controlled (at pH 3) leaching method (orange bars) than the other test methods. Similarly, concentrations of zinc (547 ± 3.0), cadmium (68 ± 6), manganese (102 ± 7) and chromium (39.8 ± 2) obtained by the TCLP method (purple bars) were rather higher than concentrations obtained by the other three test methods. On the other hand, relatively minimum leachability of zinc, manganese, copper and chromium was observed under the batch leaching test (green bars). Nickel and copper were not leached under the batch (green bars) and TCLP (purple bars) leaching methods, respectively.

In this study, the oxidation-reduction potential (Eh) of the metals were also measured. Regardless of the four test methods, the Eh measurements of the metals ranged from 244 to 17.8 milli Volt (mV). Maximum Eh value was recorded under the standard column leaching test (233 ± 11 mV) whereas the minimum value (23.4 ± 4.9 mV) was measured under the standard pH controlled leaching test (at pH 12). Leachability of lead and zinc versus their oxidation reduction potential was presented as an example (Figure 5).

Dissolved organic carbon (DOC) content of the test material was also assesed by using the four test methods. High released concentration of DOC was measured in the lower liquid to solid ratio by the standard column leaching test. It was also observed that the leaching trend of DOC relatively increased after the first liquid to solid ratio (L/S of 0.1) and then it went decreasing.

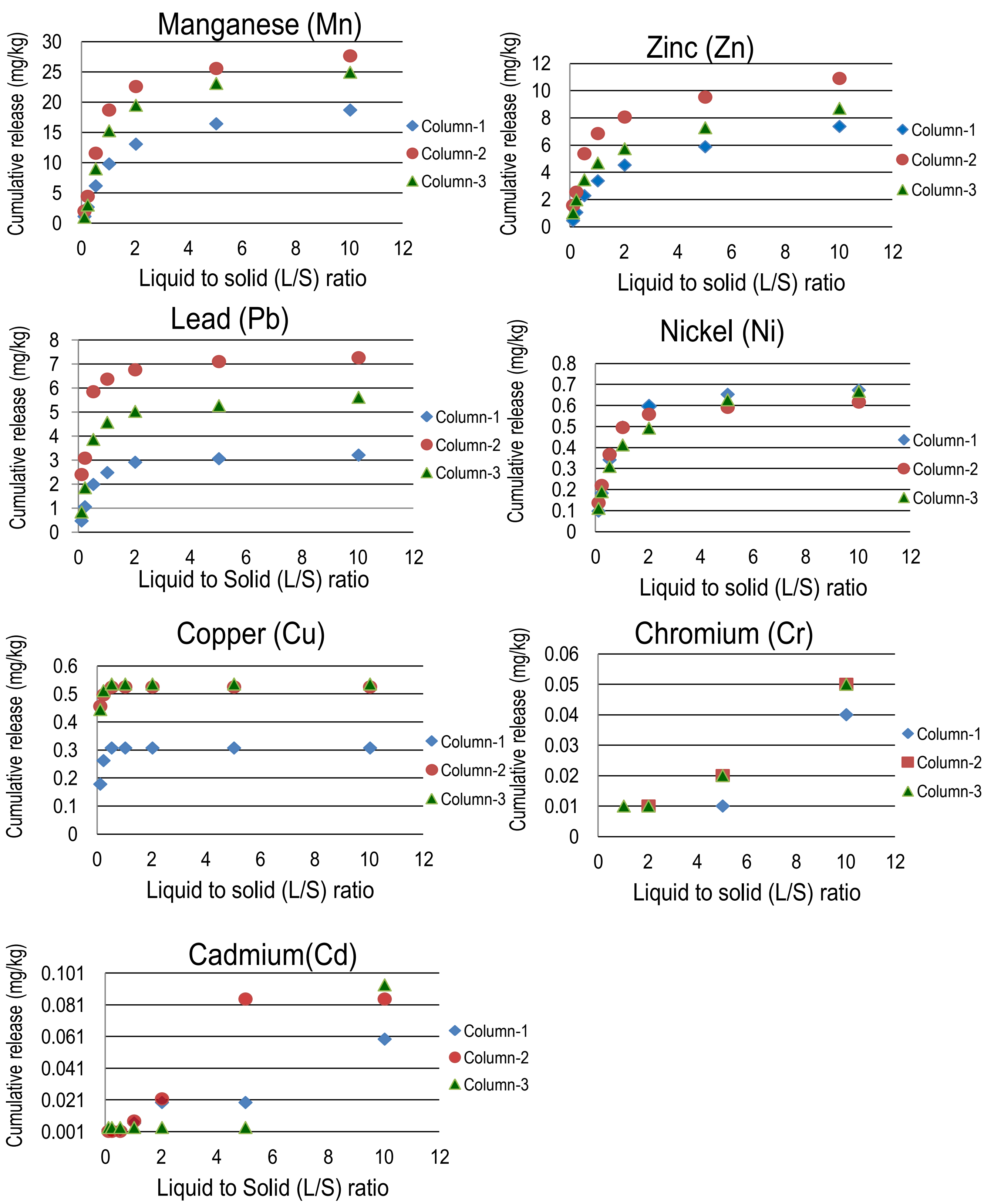


Figure 2. Cumulative release values of Mn, Zn, Pb, Ni, Cu, Cd and Cr from the three columns under the standard column leaching test conditions.

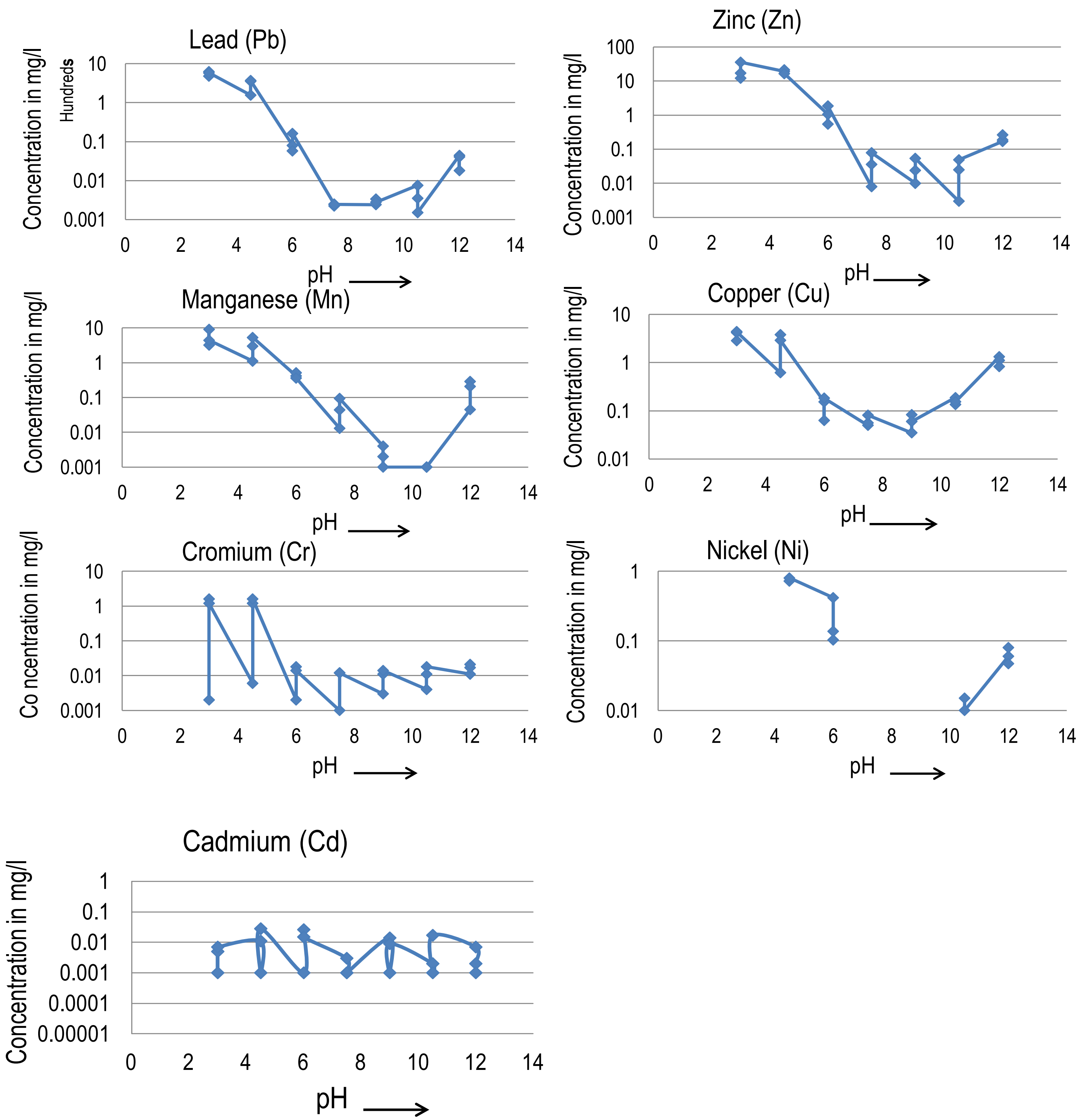


Figure 3. Leaching behavior of Pb, Zn, Mn, Cu, Ni, Cd and Cr as a function of pH (tests were done in triplicate).

Based on the test methods, maximum leachability (6879 ± 1342 mg/kg) of DOC was recorded by the TCLP extraction test which is followed by released concentration from the pH controlled (at pH 4.5) leaching test (2513 ± 140 mg/kg). Minimum leachability of DOC (1032 ± 115 mg/kg) was measured by the batch leaching test. However, under pH controlled leaching test, the leaching trend of DOC seems pH independent (Figure 6a, b

Table 1. Maximum potential leaching rate (%) of heavy metals under pH-controlled and TCLP leaching conditions.

Heavy metal	Max. leaching rate (%) from pH (pH = 3) controlled test	Max. Leaching rate (%) from TCLP extraction
Pb	22.2	8.95
Zn	1.5	3.77
Cr	1.58	3.71
Mn	0.28	0.44
Ni	2.38	0.16
Cd	0.01	17.15
Cu	0.1	0

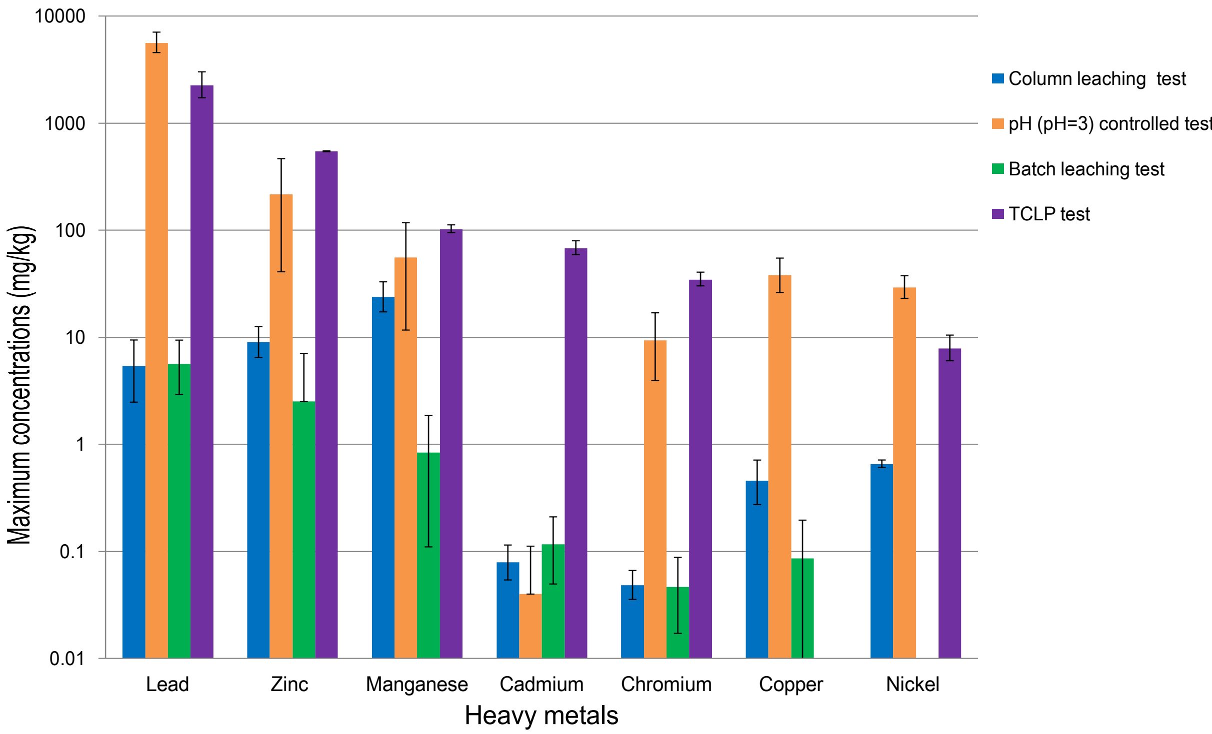


Figure 4. Maximum leachability (in mg/kg) of metals versus leaching test methods (all tests were in triplicate).

and c).

DISCUSSION

In this study, a maximum of 563 ± 74 mg/l Pb was leached from waste printed circuit boards (PCBs) by the pH controlled test method (Figure 4). 150 - 500 mg/l Pb was reported to leached from the same waste materials under TCLP extraction test elsewhere (Li et al., 2006). Furthermore, Yang (1993) and Vann et al. (2006a) reported 100.8 and 151 mg/l Pb leachability from PCBs under TCLP extraction test, respectively. Most commonly the leachability of Pb by the TCLP test method is in the range of 100 to 200 mg/l (Jang and Townsend, 2003). Moreover, the review work of Vann et al. (2006a) indicates that leached concentrations of Pb by the TCLP test method ranges from 56 to 1350 mg/l. In this study, 113.7 mg/l Pb was determined by the TCLP test method (Figure 4). Similarly, 3.4 ± 0.3 mg/l Cd was leached from the test material which is a bit less than a finding (5.8 mg/l) reported by Yang (1993).

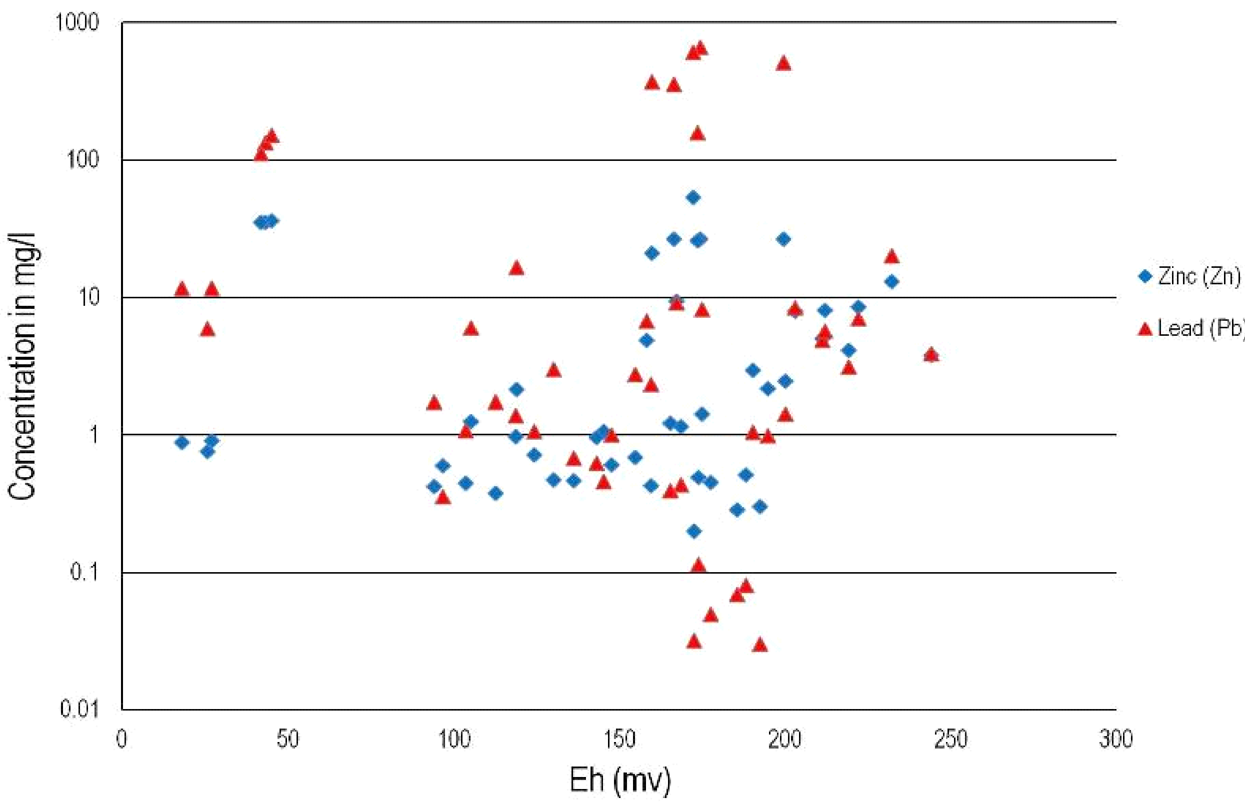


Figure 5. Leachability distribution of metals (Zn and Pb) as a function of oxidation reduction potential (E_h) values regardless of leaching test methods (all tests were done in triplicate).

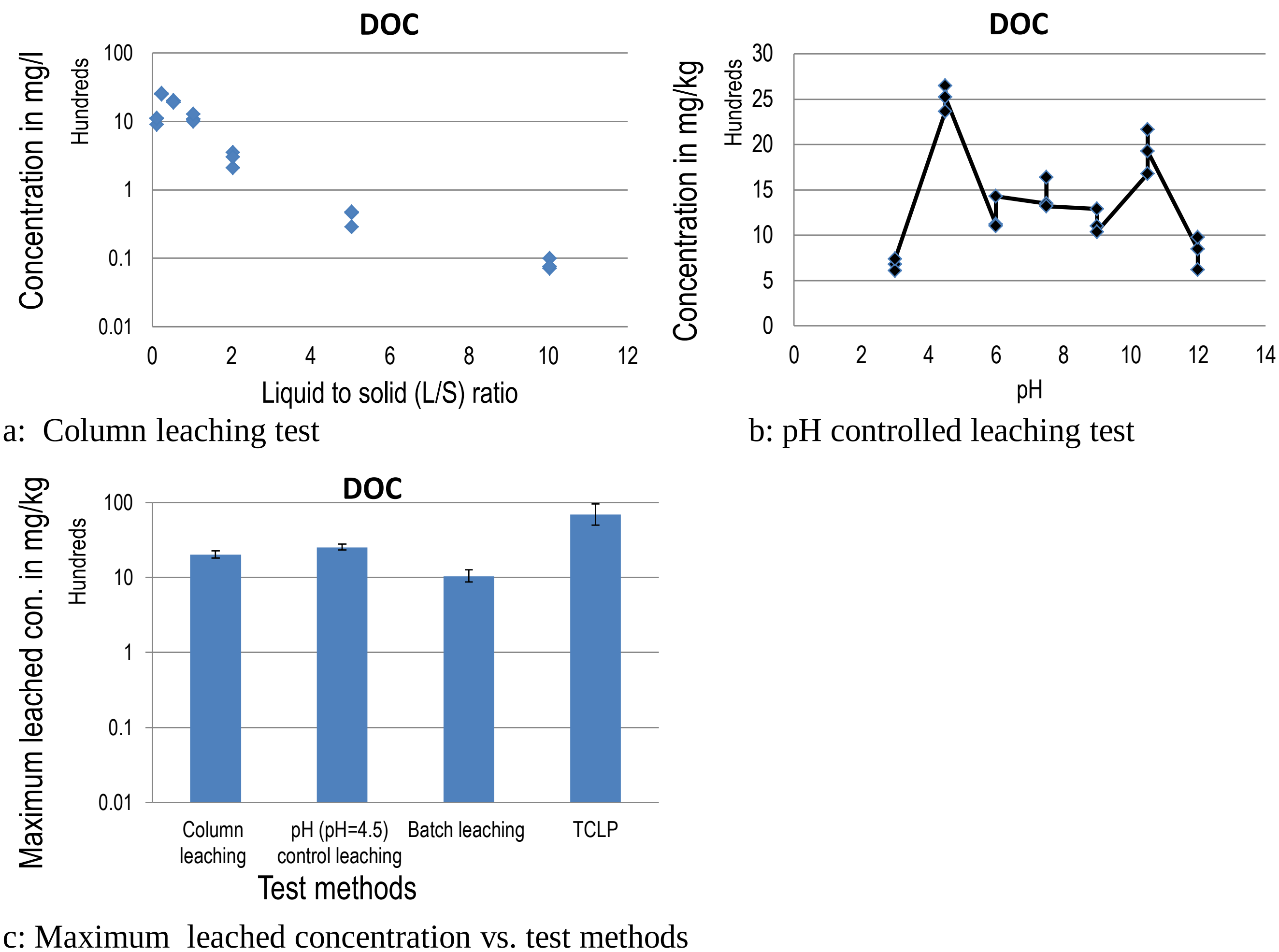
Zinc is the other metal determined by the test methods, and 27.4 ± 0.15 mg/l Zn was obtained as a maximum value by the TCLP test method (Figure 4), which is much higher than results reported on WEEE, including personal computer components, (14.9 mg/l) (Visvanathan and Yin, 2009). This discrepancy is likely because the previous study was conducted on different types of WEEE, which would alter concentrations, leaching availability and leaching solution pH.

The data show that Pb and Cd content of printed circuit boards exceeded the regulatory limit according to TCLP, which are 5 and 1 mg/l, respectively. In addition, Pb, Cd, Zn, Mn and Cu content of the test material compare well with the Dutch standard of heavy metals reference values in soil and groundwater. For instance, the standard's reference value of Pb, Cd, Zn, Mn and Cu in soil is 85, 0.8, 140, 35 and 36 mg/kg, respectively (Dutch Standard, 2000). However, the leached concentrations of the metals highly exceeded the reference values (Figure 4).

This study showed that metals were better mobilized in the lower L/S ratios and high amounts of Pb. Zn, Cr, Mn, Cu and Ni leached before a L/S ratio of 5 (Figures 1 and 2). The exchangeable (water soluble) fraction of metals is generally considered the mobile and available form and, therefore, more susceptible to be released (Xiaoli et al., *2007*). High amount of Cd was leached by the TCLP test when compared with the other test methods (Figure 4). This may be due to the TCLP extraction fluid buffering capacity that enhanced the leaching of Cd.

Studies indicated that the release of DOC primarily controls the leaching of heavy metal like Cu which has a high affinity for DOC (Van der Sloot et al., 2001). In this study, high amount (6934 ± 187 mg/l) of DOC was released from the test material. This may also bind with the other heavy metals and determined their leaching behavior. From the leaching trend, 99.3% of DOC was leached in the lower L/S ratios (up to L/S ratio of 2) (Figure 6a). Therefore, this study suggests that disposal of waste PCBs, in landfill or dumping in the environment with other domestic wastes will be releasing heavy metals and high concentration DOC immediately after the waste materials are in contact with landfill leachate or rain. Long-term DOC increases may have wide-ranging impact on fresh-water biota and drinking water quality.

It is reported that solubility of DOC increases with increasing pH (Cappuyns and Swennen, 2008). However, in this study, the leaching of DOC by the standard pH



a: Column leaching test

b: pH controlled leaching test

c: Maximum leached concentration vs. test methods

Figure 6. DOC leaching patterns under different leaching scenarios and maximum released concentration corresponding to the test methods (all tests are in triplicate).

controlled leaching test seems pH independent (Figure 6b).

Regardless of test methods, the redox potential (Eh) measurements of the metals were between 17.8 and 244 mV, which indicates predominantly oxidative condition and the leachability distribution of the metals were above 50 mV (Figure 5). Under anaerobic condition or in the case of a landfill, the metals bound to carbonate, organic compounds and sulfide are more stable and are retained in the landfill itself, whereas the metals bound to Fe and Mn oxide are unstable. This situation is opposite to an open dumpsite; which is exposed to the atmospheric condition that undergoes different effects due to oxygen diffusion. In a high redox condition, the binding of metals to Mn and Fe oxide increases, whereas binding to carbonate, organic compound and sulfide tend to decrease. With more possibility of oxygen diffusion into the upper layer of a dumpsite and with sufficient moisture content, the degradation rate and the acid buffer capacity of the dumpsite is highly influenced. Under this condition, there is a drop in alkalinity and pH and sulfide oxidation, where heavy metals are easily available and released (Prechthai et al., 2008). Therefore, under open dumping conditions, heavy metals will be easily leaching from waste printed circuit board scraps into the environment.

Regarding the assessment of the test methods used in this study, toxicity characteristics leaching procedure (TCLP) method has been criticized for the over exaggerated test conditions that rarely occur in the natural condition (Li et al., 2009). This test was designed to simulate the worst case landfill scenario of heavy metal leaching by using aggressive extraction fluid at a single pH (4.93). In this study also, maximum leaching rate of all the heavy metals except Pb and Ni was observed in the TCLP extraction condition. On the other hand, for the batch leaching (EN 12457-4) and column leaching (PrEN 14405) tests only de-ionized water was used as extraction fluid to resemble the natural conditions and heavy metals leachability results of these two tests were too small as compared to that of the TCLP extraction test (Figure 4). However, reports indicate that column leaching tests provide a closer approximation to leaching processes that occur under field conditions without compromising reproducibility of experiments (Van der Sloot, *2002*). Likewise, in many respects, leaching behavior as reflected by pH controlled leaching test and related characterization leaching tests provide a better means of assessing environmental impacts than analysis of total composition (Van der Sloot, 1996). All the above facts, therefore, makes the TCLP test over-conservative to assess the leachability of heavy metals from the waste material.

Lastly, the finding of this study suggests that both the disposal and recycling of waste printed circuit boards or other WEEEs are of concern. Hazardous substances can be released from the WEEEs through disposal or informal recycling processes with the potential to expose workers as well as put these chemicals into the environment. Hence, the information of this study, especially the results of the column leaching and the pH controlled leaching tests, can be used in predicting the release of constituents from WEEE in a specific scenario by means of different models for further environmental risk assessment of WEEE recycling and disposal in developing countries.

Conclusion

By combining a set of standard leaching tests methods, different patterns of metals and DOC leaching were observed from waste printed circuit boards. Based on this, Pb, Zn, Cr, Cd, Cu, Ni and DOC are the key pollutants released from printed circuit boards. The results also showed that Pb was the predominant soluble heavy metal and 5617 ± 739 mg/kg Pb was leached by the standard pH controlled leaching condition. The leachability pattern of the other heavy metals was in the order of Cd<Zn<Cr<Ni<Mn<Cu regardless of the leaching test conditions. High amount of heavy metals were leached from the test material in the lower liquid to solid (L/S) ratio or before a L/S ratio of 5. The study also confirmed that, the leaching behavior of Pb, Zn, Cr, Cu, Mn and Ni is strongly pH dependent and Cu showed a relatively v-shaped leaching trend. However, the leaching trend of Cd seems pH independent. The finding of the study indicated that all the heavy metals were leached under oxidative condition.

Recommendations

The results of the study show that there is high content of DOC in printed circuit boards, which may affect the mobility of heavy metals, therefore, it is necessary to investigate the type of DOC (since it is a sum of parameters for all organic species in solution) that comes out from the test materials and observe its long term effect on the mobilization of heavy metals. In addition, the particle size of the sample used for the leaching tests was below 10 mm; under natural conditions waste printed circuit boards might not be crushed to this extent. Therefore, it is recommended to carry out large scale leaching tests on 2 or 3 times shredded scraps separately and by mixing with fresh municipal solid wastes (such as kitchen waste).

Conflict of interest

The authors did not declare any conflict of interest.

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