

HEAVY METALS LEACHABILITY IN FLY ASH REMEDIATED SOIL

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Abstract: This study focused on recycling fly ash, a by-product of power plants, to stabilize heavy metals in contaminated soil. Fly ash collected from the Sejingkat power plant in Kuching, Sarawak, was applied to soil from the roadside. The potential risk associated with the application of fly ash on the soil was assessed in terms of heavy metal concentration in leaching experiments. Deionized water and acidified water (deionized water acidified to pH 4.0 with nitric acid) were used as leaching agents and passed through columns filled with untreated soil and soil-fly ash mixture (9:1). The Cd, Zn, Ni, Mn, Cu and Fe content in leachates were assessed at Day 0, 5, 10 and 15 after stabilization. Leachate from the soil-fly ash mixture with acidified water had low pH range (4.5-6.2) compared to soil-fly ash mixture with deionized water (5.8-6.2), and this affected the solubility of heavy metals. The concentration of the metals in soil-fly ash mixture with acidified water leachate was higher than soil-fly ash mixture with deionized water. In contrast, untreated soil did not show any clear pattern of heavy metal reduction except for Ni. Except for Cr, the concentrations of all the tested heavy metals in treated soil decreased with increasing pH as well as electrical conductivity from Day 0 to 15 of contact time. The results suggested that the solubility of heavy metals in soil leachate was influenced by pH and the type of leaching agent. Therefore, the application of fly ash as a soil remediation agent may be a sustainable option to manage this by-product.

KEYWORDS: Heavy metals, leachate, leaching agents, soil remediation, fly ash.

Introduction

Fly ash, which are small particles of mineral waste formed by the burning of coal to generate electricity in power plants, has become a serious environmental problem (Haynes, 2009; Lima *et al.*, 2012; Kumar *et al.*, 2019). Each year, almost 750 million tons of coal ash are emitted from power plants worldwide, but less than 50% of it only is recycled (Ram & Mastro, 2014). The high content of heavy metals in fly ash hampers its recycling potential (Lima *et al.*, 2012) as it is more convenient to dump it in landfills (Zaco *et al.*, 2014) and fly-ash basins, or wash it out with water into pond ash (Jala & Goyal, 2006; Asl *et al.*, 2019). These methods have limitations that may endanger human health, pollute the environment and degrade the soil (Asl *et al.*, 2018).

Fly ash has been proposed as a remediation or stabilizing agent to improve the physical and chemical properties of soil (Papadimitriou *et al.*, 2008), thereby increasing fertility to boost agricultural yield (Cheng *et al.*, 1998; Ciccu *et*

al., 2003; Dahl *et al.*, 2008; Basu *et al.*, 2009). The application of fly ash generally increases soil concentrations of extractable Ca, Ba, Pb, Se, B, S and Sr, and has the ability to restore nutrient-deficient soil (Haynes, 2009). This is because fly ash contains a high level of macro (P, K, Ca, Mg and S) and micronutrients (Fe, Mn, Zn, Co, B) that are beneficial for plants (Wang *et al.*, 2007; Ram & Mastro, 2014). It has also been suggested as an effective additive to increase organic content while maintaining the optimum pH level, which has been observed to immobilize heavy metals in soil (Sitarz-Palczak & Kalembkiewicz, 2012). Other uses of fly ash include as a cement additive and for land reclamation (Haynes, 2009).

However, one primary concern in using fly ash for soil remediation is the leaching of heavy metals into the soil (Singh *et al.*, 2010). The leaching could cause growth inhibition and uptake of heavy metals in plants (Singh *et al.*, 2016). Leaching of toxic metals, such as As, Se, Pb, Ni, Cr, Co and Mo during soil remediation may also lead to environmental contamination

(Sitarz-Palczak & Kalembkiewicz, 2012). The accumulation of those toxic metals in soil, food and water sources is a risk to human health.

In the light of the potential risk of using fly ash for soil remediation, it is crucial to carefully study the leaching of heavy metals from fly ash. This study aims to determine the effect of fly ash application on soils collected from the roadside by means of column experiments using two leaching solutions; i.e. deionized water and acidified water.

The type of leachate and pH are known to influence the solubility and adsorption of heavy metals onto soil and fly ash surfaces (Wuana *et al.*, 2010; Komonweeraket *et al.*, 2015). This study assessed the concentration of heavy metals, namely Mn, Ni, Cu, Zn, Fe and Cd, in the leachate samples.

Materials and Methods

Sample Collection and Characterization

Coal fly ash samples were obtained from Sarawak Energy Bhd's (SESCO) Sejingkat Power Station in Kuching, Sarawak, Malaysia. Soil samples were collected from roadsides around Kuching, where traffic was heavy. The pH, particle size, organic matter content, specific surface area, electrical conductivity and total heavy metal content of the fly ash and soil samples were characterised. The soil samples were also subjected to cation-exchange capacity analysis.

The pH was measured using a Metler Toledo SG2 pH meter (Metler Toledo, Columbus, Ohio, USA) at 1:2.5 (w/v) ratio of soil (or fly ash) and deionised water. Electrical conductivity was determined using a HI 88733 conductivity meter (Hanna Instruments, Woonsocket, Rhode Island, USA) with the soil to solution suspension ratio

at 1:5 (w/v). The fractions of sand, silt and clay were analysed using the pipette method as described by USDA (1984). Organic matter content was determined by loss of ignition (LOI) method (Sparks, 1996). Specific surface area and cation-exchange capacity (CEC) of soil and fly ash were determined according to Calace *et al.* (2005).

Heavy metal analysis was conducted by subjecting the samples to aqua regia wet digestion method (Kumar *et al.*, 2009) prior to measurement using Perkin Elmer 3310 flame atomic absorption spectrophotometer (Perkin Elmer, Waltham, Massachusetts, USA).

Column Leaching Experiments

Leaching experiment is an efficient method for determining the potential leaching for long term assessment and to predict the immediate risks (Zandi *et al.*, 2007). The soil was air-dried for 72 hours and filtered through a 2 mm sieve. Column leaching experiments were performed in triplicates comprising untreated soil, soil-fly ash mixture with deionized water (pH6) and soil-fly ash mixture with acidified water (pH4) (Table 1).

To set up the experiment, 10 g of each soil sample was placed in 100 mL glass columns with glass wool lining at the bottom to prevent solid fractions from falling out. The leaching agent was passed through the columns (200-250 mL, 0.5 mL/h) and 10 mL aliquots of leachate were collected in a flask placed at the outlet (Calace *et al.*, 2005).

The samples were kept moisturised in their respective leaching agents for 0, 5, 10 and 15 days before the leachates were collected and analysed for pH, electrical conductivity and heavy metal content.

Table 1: Treatments in column leaching experiments.

Treatments	Soil:fly ash ratio (w:w)	Contact time (days)
Untreated soil	-	0, 5, 10, 15
Soil:fly ash mixture with deionized water	9:1	0, 5, 10, 15
Soil:fly ash mixture with acidified water	9:1	0, 5, 10, 15

Statistical Analysis

An analysis of variance (ANOVA) was used to compare the heavy metal concentration of leachates between different columns at different contact times. A probability of $p < 0.05$ was considered significant. Data were analyzed using IBM SPSS Version 15 (IBM, Armonk, New York, USA).

Results and Discussion

Physicochemical Properties of Fly Ash and Soil

The selected properties of fly ash and soil samples are tabulated in Table 2. The soil itself was found to be naturally acidic ($\text{pH} 3.8 \pm 0.10$), which could easily release heavy metals when the leaching process occurred (Kanakaraju *et al.*, 2016; Olaniran *et al.*, 2013).

The fly ash sample had a high pH value (9.1 ± 0.10), which reflected its alkaline characteristic. Fly ash had been reported to have neutralizing capacity in acidic soil (Haynes, 2009). The alkaline property of fly ash was due to the presence of Ca and Mg hydroxides and carbonates (Jambhulkar & Juwarkara, 2009).

The soil samples comprised 55.59% sand, 38.70% clay and 5.71% silt. There was a high percentage of organic matter ($83.21\% \pm 0.37$) and the CEC of the soil was 30 ± 0.08 cmol/kg. The presence of organic matter could affect the

leaching potential of various heavy metals from the soil (Tandy *et al.*, 2009).

The specific surface area for fly ash and soil were 3684.40 ± 15 m²/g and 5001.03 ± 137 m²/g, respectively. A higher metal absorption capacity might be attributed to the higher specific surface area (Kim *et al.*, 2009; Li *et al.*, 2004). Electrical conductivity and moisture content analysis in fly ash produced the values of 0.84 ± 0.02 mS/cm and $0.13\% \pm 0.10$, respectively. Electrical conductivity and moisture content was observed to correlate with each other. The lower value of moisture content produces lower electrical conductivity. Based on the characterisation analysis, fly ash demonstrated low values of electrical conductivity and moisture content.

Fly ash samples contained elevated levels of Mn (177.64 ± 3.46 mg/kg), followed by Fe (176.98 ± 5.02 mg/kg). These two were found to be essential elements in the fly ash. The existence of these elements in high levels and the lesser amount of Cd, Zn, Ni, Cu as well as Cr could be attributed to the type of coal and the burning processes involved in the power plant.

For soil samples, the highest heavy metal concentration was Fe, which constituted about 260.29 ± 4.15 mg/kg, followed by Ni with 125.50 mg/kg and Cd with 118.19 mg/kg. The other elements, such as Zn, Ni, Cu and Cr, existed in smaller quantities in the range of 22.00 to 126.00 mg/kg.

Table 2: Treatments in column leaching experiments.

	Fly Ash	Soil
Particle size		
Sand (%)	-	55.59
Silt (%)	-	5.71
Clay (%)	-	38.70
pH	9.1 ± 0.10	3.8 ± 0.10
Total organic matter	-	83.21 ± 0.37
Moisture content (%)	0.13 ± 0.10	-
Electrical conductivity (mS/cm)	0.84 ± 0.02	0.22 ± 0.01
Specific surface area (m ² /g)	3684.40 ± 15	5001.03 ± 137
Cation exchange capacity, CEC (cmol/kg)	-	30 ± 0.08
Total heavy metals (mg/kg)		
Cd	10.65 ± 1.15	118.19 ± 13.26
Zn	8.65 ± 2.30	37.18 ± 1.15
Ni	17.96 ± 2.00	125.50 ± 12.44
Mn	177.64 ± 3.46	42.50 ± 6.09
Cu	26.61 ± 1.15	21.91 ± 3.45
Cr	21.29 ± 1.18	49.14 ± 1.25
Fe	176.98 ± 5.02	260.29 ± 4.15

Electrical Conductivity and pH of Leachates

Electrical conductivity and pH are important parameters to determine the quality of leachates before entering the groundwater system. In electrical conductivity (Figure 1), the mobility of ions was constantly reduced in all leachates over contact time, except for deionized water. The electrical conductivity increased slightly from 0.11 mS/cm at Day 0 to 0.15 mS/cm after five days of stabilization before declining again.

The initial increase in electrical conductivity could be due to the release of dissolved ions and minerals from the soil. The electrical conductivity decreased to 0.07 mS/cm on Day 15. The loss of electrical conductivity after a

certain period of stabilization was influenced by the greater adsorption of metals onto solid surfaces (Goswami & Mahanta, 2007).

The study by Heikal *et al.* (2004) stated that the decrease in electrical conductivity was due to the formation of hydrated products, thus contributing to the marked consumption of free ions in the leachates. Cations and anions of different electrolytes were simultaneously adsorbed by the soil during leaching experiments (Su-zhen *et al.*, 2009) and, therefore, the quantity of ions released would become lesser than the quantity of ions adsorbed. This would lead to a loss in electrical conductivity after a certain period.

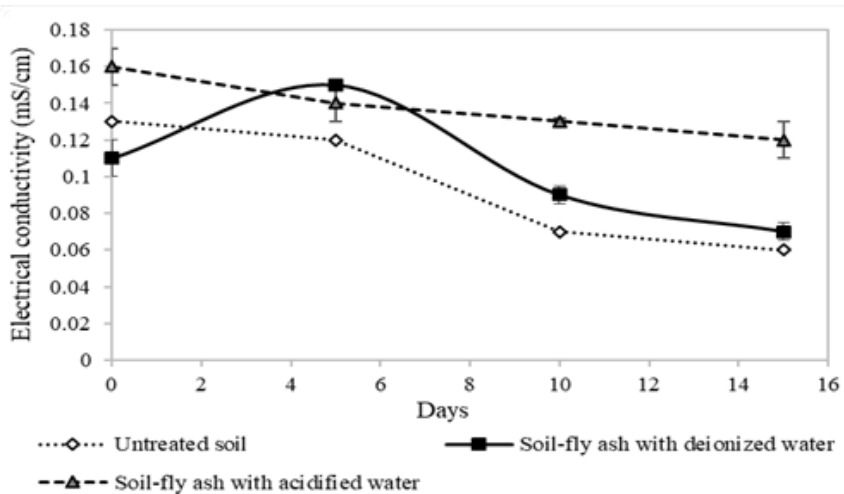


Figure 1: Electrical conductivity of leachates versus contact time (days).

The pH values increased in alleachate solutions from Day 0 to 15 of stabilization (Figure 2). However, the pH increased noticeably from 4.5 to 6.2 in acidified water leachates. The pH of deionized water leachates from soil-fly ash samples also increased from 5.8 to 6.2. In comparison, the leachates from untreated soil showed no changes in pH. The pH equilibrium was achieved after 15 days of stabilization (Figure 2).

The increase in pH value of the soil-fly ash mixtures was due to the presence of lime in fly ash. In fact, the increase of the leachates' pH could be attributed to the alkaline nature of fly ash. The higher neutralization capacity of fly ash was due to its Ca content and the strong adsorption of the elements onto the soil and ash surfaces (Haynes, 2009).

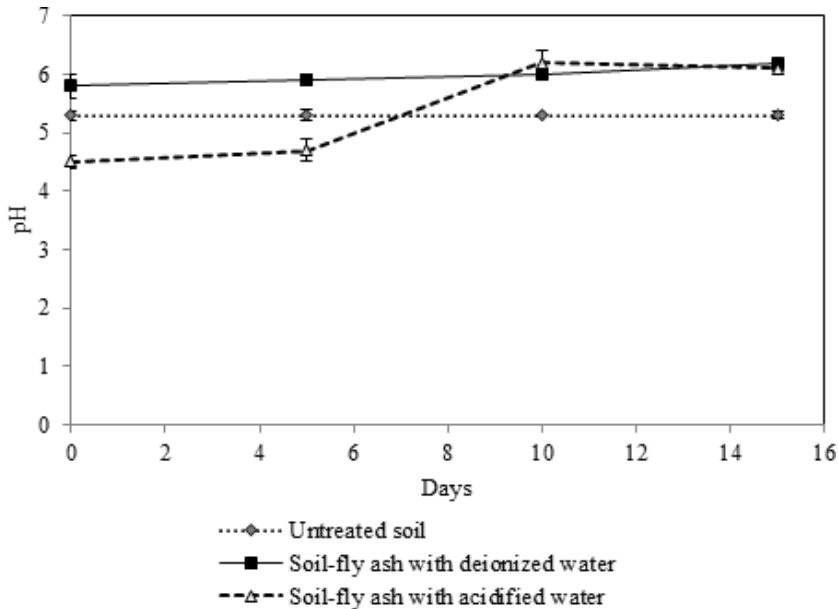


Figure 2: The pH patterns in soil leachates.

Leaching of Heavy Metals from Fly Ash

Leaching of heavy metals from fly ash with deionized water was studied to investigate the effectiveness of fly ash in reducing the concentration of heavy metals from contaminated soil. The concentrations of Cd, Ni, Mn, Cu and Fe decreased after two days of contact time (Figure 3). Only Cr increased from Day 0 to Day 2. Concentrations of Zn fluctuated between contact times. The variations observed among metals might be influenced by the potential leaching and extractability of the individual metals in fly ash samples. For instance, the fly ash samples contained higher levels of Ni and lower level of Cu (Table 2).

Likewise, a higher level of Ni was released in the leachate while Cu was undetected after two days of contact time. The potential leaching of these heavy metals was not only dependent on the total metal content, but also the properties of the fly ash. The mineral and composition of fly ash, size distribution, pH of the leaching solution and test conditions greatly influenced the leaching properties of each heavy metal in fly ash (Singh *et al.*, 2010; Kalembkiewicz & Sitarz-Palczak, 2015; Jiao *et al.*, 2016).

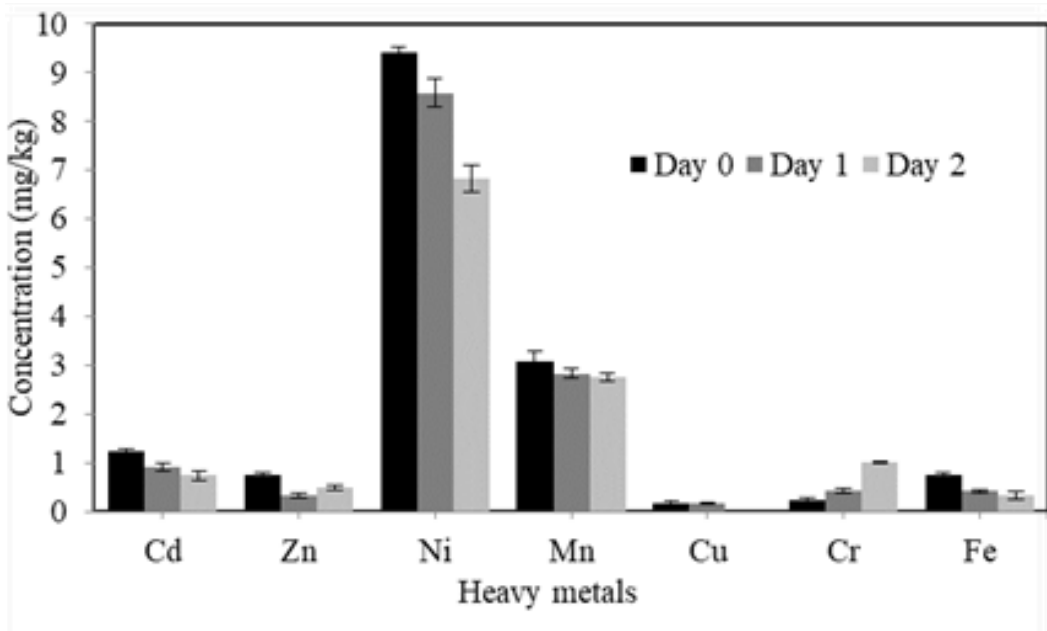


Figure 3: Leaching pattern of heavy metals versus contact time from fly ash using deionized water.

The Release of Heavy Metal Concentrations in Leachates

The addition of fly ash to soil samples greatly influenced the potential leaching of heavy metals. The concentrations of metals in leachates from different leaching experiments are shown in Figure 4. In general, the amount of metals decreases with the addition of fly ash to the soil. The untreated soil did not show any clear pattern of heavy metals in the leachates analysed. The

concentration of heavy metals in leachates is affected by the leaching agents, acidified water and deionized water used and their pH. The pH range obtained for acidic leaching solution (4.5-6.2) was lower compared to deionized water (5.8-6.2). The range of heavy metals concentrations in soil-fly ash with deionized water were 287.50 – 120.83 mg/kg for Cd, 156.25– 55.33 mg/kg for Zn, 425.00 – 103.33 mg/kg for Ni, 104.17 – 52.08 mg/kg for Mn, 235.42 – 35.42 mg/kg for Cu and 483.33 – 152.29 mg/kg for Fe (Figure 4).

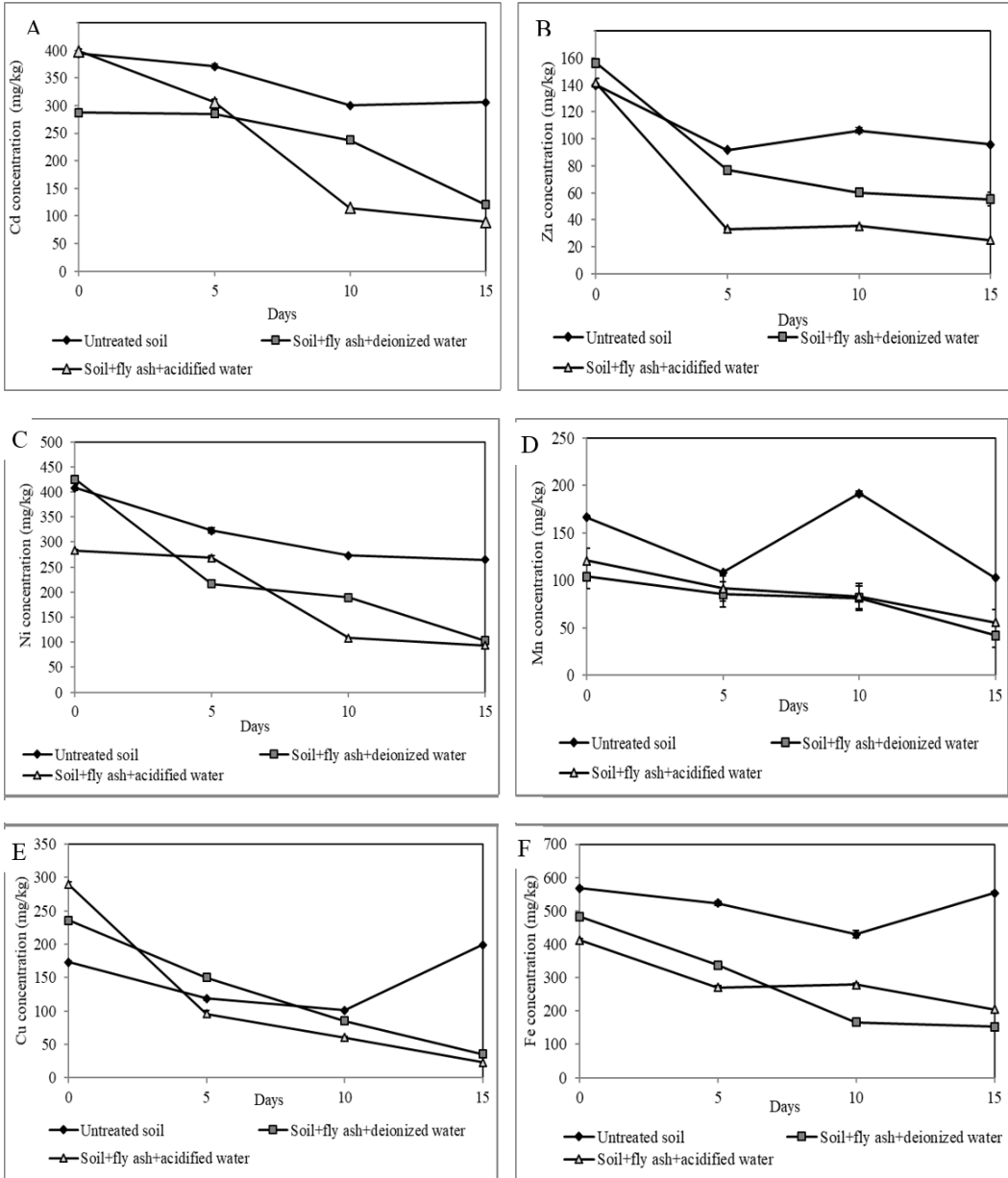


Figure 4: Heavy metals quantified (mg/kg) in leachates from untreated soil, soil-fly ash with deionized water (9:1) and soil:fly ash with acidified water (9:1) at different contact times. They are (a) Cd, (b) Zn, (c) Ni, (d) Mn, (e) Cu and (f) FE

The levels of Cd, Zn, Ni, Mn, Cu and Fe in soil-fly ash with acidified water ranged from 398.33 mg/kg to 89.58 mg/kg, 141.67 mg/kg to 25.00 mg/kg, 283.33 mg/kg to 93.75 mg/kg, 120.83 mg/kg to 55.80 mg/kg, 289.58 mg/kg to 22.92 mg/kg and 412.50 mg/kg to 205.00 mg/kg, respectively (Figure 4).

The solubility of heavy metals is known to decrease at higher pH (Rieuwerts *et al.*, 1998), and nitric acid had the ability to enhance the mobility of some elements. Nitric acid was used in the leaching experiments because it allowed the heavy metals to be detected efficiently (Palumbo *et al.*, 2005). Deionized water was used to simulate conditions where waste and/or soil in contact with leaching agents have a low buffering capacity like rainwater (Al-abad *et al.*, 2006). The presence of fly ash in soil-fly ash mixture had changed the pH of the water and subsequently affected the solubility and adsorption of metals.

Concentrations of Fe, Mn, Ni, Zn and Cd in the untreated soil were higher than soil treated with fly ash and their leaching agents. There were discrepancies in the reduction of metal content in untreated soil leachate. Metals in leachates of untreated soil demonstrated fluctuating contents over time except for Ni (Figure 4c), whereby it was consistently reduced from Day 0 (408.33 mg/kg) to 15 (264.58 mg/kg). The absence of leaching agents to dissolve the metals, low mobility of metals and strong interactions between metal and soil might be the contributing causes of fluctuations in heavy metal content (Wuana *et al.*, 2010; Zheng *et al.*, 2012).

The concentration of Cd decreased gradually from Day 0 to 15 (Figure 4A). The decrease in leachate concentration after 15 days indicated that Cd was strongly bound onto the soil surface and this would reduce its uptake by plants and release into groundwater. Statistical analysis showed a significant difference between untreated soil and soil-fly ash with deionized water at Day 5 of contact time ($P = 0.042$), while at Day 10, there was significant difference ($P = 0.048$) between untreated soil and soil-fly ash with acidified water. At Day 15, there was a significant difference in concentrations of Cd

between all the leachates ($P = 0.000$).

The content of Zn consistently reduced in the soil-fly ash with acidified water leachate where 141.67 mg/kg on Day 0 was reduced to 25.00 mg/kg on Day 15 (Figure 4b).

Concentrations of Zn in leachate solutions also decreased in soil-fly ash with deionized water. Zn is an essential element which acts as a nutrient for plant growth, but only a trace amount of this element could be present in plant tissue. The decrease of Zn concentration in leachates could be attributed to the strong adsorption of this element onto the soil and fly ash surface. Analysis of variance found a significant difference ($p < 0.05$) for soil-fly ash with deionized water and soil-fly ash with acidified water at Day 15 contact time.

Although the concentration of Ni decreased in all metal leaching treatments, soil-fly ash with deionized water demonstrated higher reductions throughout the experiment (Figure 4c). The content of Ni was reduced from 425.00 mg/kg to 103.33 mg/kg. There was a significant difference ($p < 0.05$) between untreated soil and soil-fly ash with acidified water on Day 0 of stabilization. However, there were no significant differences in the concentration of Ni between all column leaching experiments for Day 5, 10 and 15 of stabilization.

A previous study by Chalermyanont *et al.* (2009) reported that Ni was toxic to human health. However, Saffari *et al.* (2015) reported that fly ash can be applied to reduce the Ni content in soil.

The concentrations of Mn also decreased with contact time (Figure 4d). The lower concentration of Mn in column leaching experiments was due to the adsorption of the metal on soil particles. The soil-fly ash with deionized water leachate showed a drastic fall of Mn concentration from Day 0 (104.17 mg/kg) to 15 (42.08 mg/kg).

There was a constant reduction of Cu in soil-fly ash with deionized water and soil-fly ash with acidified water from Day 0 (235.42 mg/kg and 289.58 mg/kg) to Day 15 (35.42 mg/kg and 22.92 mg/kg) (Figure 4e).

In comparison, soil-fly ash with acidified water demonstrated a greater amount of Cu in the leachate on Day 0 (289.58 ± 0.63 mg/kg) to Day 5 (95.83 ± 0.29 mg/kg), and it gradually decreased until Day 15. This result might be due to the interaction of the metal with an acidic leaching solution. Together with Cd, Zn, Ni and Mn, the reduction in concentrations of Cu after 15 days of stabilization indicated that Cu also had strong adsorption onto the soil and ash surfaces, thus reducing the amount of this metal in groundwater. At Day 0 of stabilization, it was already observed that there was a significant difference between leaching experiments ($p < 0.05$).

The most elevated metal concentration in leachates was Fe (Figure 4f). This element displayed a consistent drop in the leachates (Figure 4f). The concentration of Fe found in soil-fly ash with acidified water increased slightly from Day 5 (270.83 mg/kg) to Day 10 (279.17 mg/kg) of contact time, but then continued decreasing up to Day 15. In contrast, soil-fly ash with deionized water showed a steady decrease of Fe in the leachates.

Conclusion

The concentration of metals in leachates was greatly influenced by their adsorption onto the soil and fly ash surfaces, and also on the type of leaching agent. This study highlighted the potential of using fly ash from power stations as a soil amending agent. Its strong alkaline characteristic might require treatment with leaching agents before being applied to soil. It is recommended that future studies should determine the maximum adsorption capacity of heavy metals onto the soil and fly ash surfaces. This would provide more data to ensure that the use of fly ash would not cause heavy metals to leach into the groundwater. The toxicity effects of leachates must also be studied.

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