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Soil Cadmium Contamination and Ecological Implications in a Tropical Urban Ecosystem: A Case Study of Air Hitam Sanitary Landfill in Puchong, Malaysia

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ABSTRACT

Cadmium (Cd) is a mobile heavy metal that is highly toxic to almost all lifeforms. Urban eco-systems are susceptible to Cd contamination due to certain anthropogenic activities. Despite being recognised as an acutely toxic element, its biogeochemical behaviour is still poorly studied and understood, especially in urban ecosystems of tropical countries. Therefore, this study was undertaken to address this knowledge gap. This study was conducted at Air Hitam Sanitary Landfill (AHSL) in Puchong, Malaysia. Samples were collected from various abiotic and biotic factors representing the biogeochemical cycle, including soil, flora, arthropods, atmospheric deposition, leachates, and river water samples. Acid digestion using aqua regia was conducted to determine the total Cd concentration in all samples collected. Cd concentrations at AHSL were relatively high in all biotic and abiotic factors with the concentrations showing a range of 0.019 ppm to 1.568 ppm. The bulk of Cd contamination in the ecosystem was found to eventually end up in rivers. The average Cd concentration in the river samples exceeded several environmental guidelines. There was also evidence of Cd entering food chains via soil arthropods and plants. Thus, Cd poses a credible threat to inhabitants of tropical urban ecosystems.

Key words: **Heavy metals, Malaysia, landfill, municipal solid waste, toxic elements**

INTRODUCTION

Cadmium (Cd) has long been documented as a relatively mobile and heavy metal that is acutely toxic to almost all life forms (Cullen and Maldonado 2012). Natural mobilisation of Cd contained in the crust and mantle occurs primarily due to volcanic eruptions but can also be caused by weathering of parent materials and burning of vegetation. However, the natural biogeochemical cycle of Cd has been significantly altered by anthropogenic sources. This change begun during the industrial revolution, due to an increase in fossil fuel burning and metal extraction. Unfortunately, this phenomenon has only grown worse due to rapid industrialization across the globe.

Soil Cd, like almost all other heavy metals, is mainly from anthropogenically mobilised sources (Rajoo *et al.* 2013). This includes atmospheric deposition due to Cd attaching itself to particles originating from fossil fuel combustion, non-ferrous mining, smelting and manufacturing (Feng *et al.* 2019). Globally, atmospheric deposition of Cd is a major soil contaminant. In China, almost 20% of the total arable lands are believed to be contaminated by atmospheric Cd (Xue *et al.* 2014). In Wales and England, almost 53% of Cd contamination in agricultural soils is from atmospheric deposition (Nicholson *et al.* 2003). To make matters worse, atmospheric Cd often has high bioavailability since up to 84% of the dry deposition of Cd is water soluble with a cation exchangeable fraction (Feng *et al.* 2019). Besides atmospheric deposition, soil Cd contamination can be due to fertilizers and pesticides that are directly applied to the soil (Cullen and Maldonado 2012). In urban ecosystems, a major source of soil Cd contamination is from municipal waste disposal, mainly nickel-cadmium batteries (Kubier *et al*. 2019). Thus, landfills are common locations for soil Cd and groundwater contamination.

Despite being recognised as a highly toxic element, its biogeochemical behaviour is still poorly studied and understood in most ecosystems (Hernandez *et al.* 2022). This is especially true for urban ecosystems, where soil Cd pollution is most prevalent. Even worse, there is relatively no studies in relation to the biogeochemical behaviour of Cd in tropical countries like Malaysia, where the climatic conditions significantly differ from countries where there is such literature (Cullen and Maldonado 2012; Xue *et al.* 2014; Feng *et al.* 2019; Hernandez *et al.* 2022; Rajoo et al., 2023). This knowledge gap has resulted in a poor understanding of how Cd behaves in ecosystems and its potential of affecting human health, either directly or by infiltrating our food chains. Therefore, this study was undertaken to address this knowledge gap. The objectives of this study are: (1) to determine the soil biogeochemical behaviour of Cd in Malaysian urban ecosystems, and (2) to determine the pollution potential of Cd in Malaysian urban ecosystems.

MATERIALS AND METHODS

Study Site: Air Hitam Sanitary Landfill (AHSL)

The AHSL site is located near the Air Hitam Forest Reserve in Mukim Petaling, Daerah Petaling, Puchong (longitude 101° 39’ 55’’ E and latitude 03° 0’ 10’’ N) (Figure 1). The Selangor State Government Council approved Worldwide Sita Environmental Management Sdn. Bhd. to develop this sanitary landfill on 22nd March 1995. ASHL was built in 1995 and was the first engineered sanitary landfill site in Malaysia, covering a total of 42 hectares. During the 11 years ASHL operated, it received approximately 6.2 million tons of domestic waste. ASHL is surrounded by residential housing, highways and manufacturing industries. AHSL was officially closed on 31 December 2006 and the 5-year Landfill Closure and Post-Closure Maintenance Plan (LCPCMP) was put in place (2007-2011).

Figure 1. Location of Air Hitam Sanitary Landfill

AHSL was selected for this study since it is a prime location for soil Cd contamination that will be able to accurately represent its biogeochemical behaviour in an urban ecosystem. As a waste disposal site, there is a high likelihood of Cd accumulation in the soil (Kubier *et al*. 2019). Moreover, since it is surrounded by manufacturing industries and heavy vehicle usage, atmospheric deposition of Cd might be prevalent in this location. AHSL is also equipped with several amenities, including a ground water drainage system, a leachate collection system and treatment plant. Thus, groundwater and leachate samples could be efficiently collected at this site. As the leachate was released into a nearby river, river samples could also be collected at this site. Moreover, AHSL had planted vegetation and soil arthropods, thus the biotic components of the biogeochemical cycle could also be evaluated.

Sample Collection

Samples were collected from various abiotic and biotic factors representing the biogeochemical cycle (Figure 2). Soil sampling was conducted at three locations of AHSL: Phases 1-5, Phase 6 and Phase 7. Six subplots (20 m x 20 m) were randomly established (completely randomised design) at each sampling location. Composite samples were obtained using an auger from each subplot at 0-20 cm, 20-40 cm, 40-60 cm, 60-80 cm and 80-100 cm. All the samples were kept in a standard plastic container and air-dried before being analysed. Samples of the soil was air dried for three days, pounded with a mortar and pestle, and then sieved through a 2-mm mesh. This was done to produce a homogenous mixture for analyses.

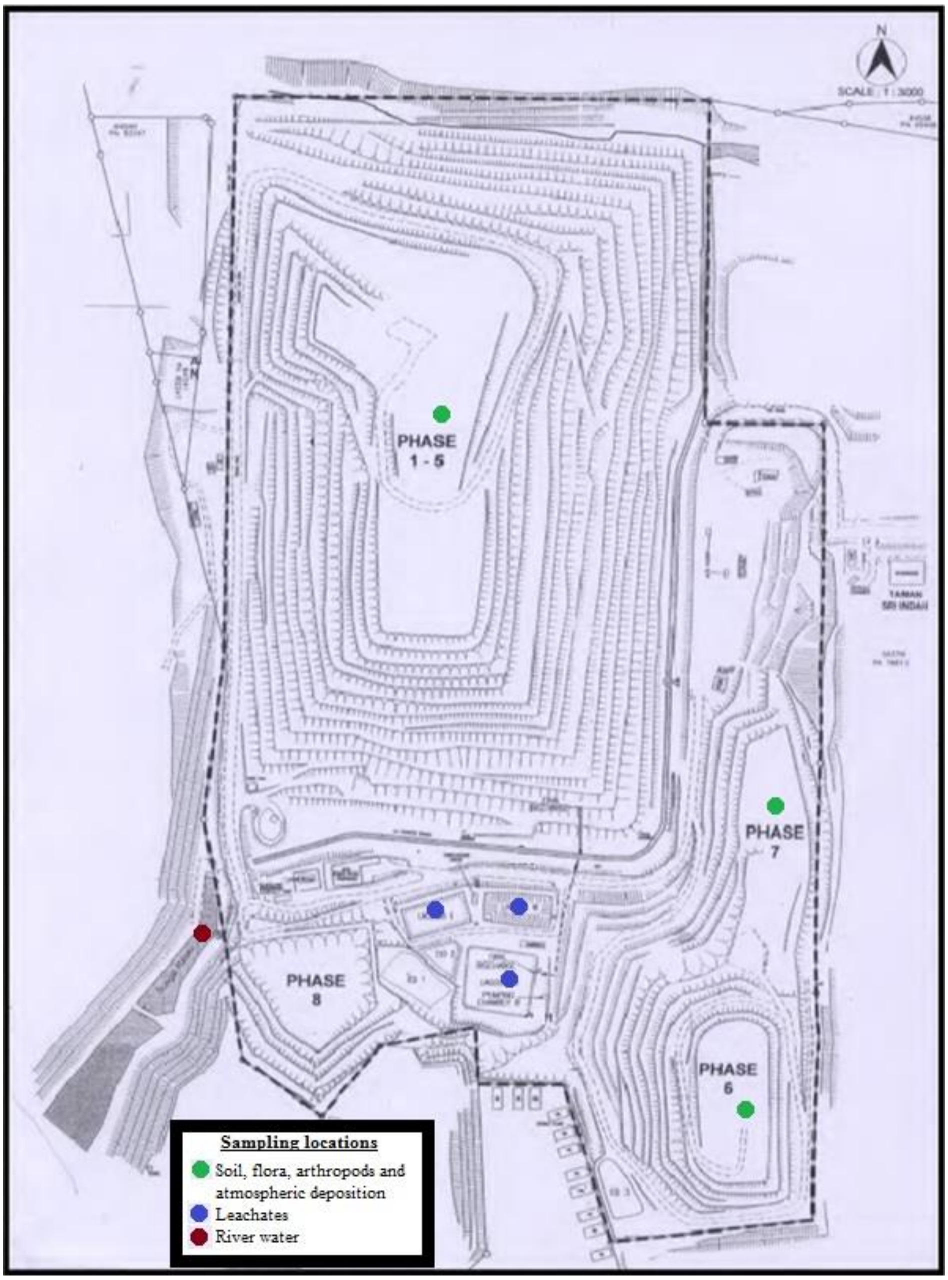

Figure 2. Air Hitam Sanitary Landfill layout and sampling locations

Atmospheric deposition sampling was conducted at the six soil sampling subplots of Phase 1 of AHSL. A wide plastic sheet was placed on the ground and secured with spikes. Sediment (soil) deposited on the plastic sheet was carefully collected using plastic vials twice a day for a month. The process was conducted until a sufficient number of samples were collected for analyses.

Soil arthropod samples were collected using the same soil sampling plots, that is six subplots (20 m x 20 m) were randomly established (completely randomised design). The soil arthropods were collected from these sampling plots using appropriate pit fall methods. Plants were selected within the vicinity of the soil sampling locations. The fresh sample of plants was separated into three parts: roots, stem and leaves. These plant parts were then dried in an oven at 60^{o} C for 24 h and shredded into small pieces before further analysis. Leachate and sludge sampling was conducted at the leachate collection ponds, that is Pond 1, Pond 2 and Pond 3 (Figure 3), using PVC pipes of 10 cm. As shown in Figure 3, the leachate from AHSL is generated to Pond 1, where no treatment is applied. The raw, untreated leachate is then channelled to Pond 2 where aeration is applied and transferred to Pond 3 for further aeration. Leachate that has undergone treatment is discharged into a nearby river. Samples were also collected from the discharged water and from the local river at 25-m intervals, for a total distance of 250 m.

Figure 3. Schematic diagram of leachate flow

Laboratory Analysis
Acid digestion was used to determine the concentration of Cd in all the samples collected (Figure 4). After digestion, the total concentrations of Cd were determined using Atomic Absorption Spectrometer (AAS). Basic physico-chemical analyses were conducted to characterise the soil characteristics (Gupta 2007).

Figure 4. Acid digestion of samples

Statistical Analysis
The data was statistically analysed using the SPSS program (Version 23). Appropriate statistical analyses were conducted to analyse the research data, such as analysis of variance (ANOVA), t-test and regression.

RESULTS AND DISCUSSION

Soil Characteristics

The surface soil of AHSL was added soil to cover the buried municipal waste. The soil was an Oxisol, heavily weathered from being exposed to weathering (Karam *et al*. 2022). The soil pH ranged from 5.47 to 6.21 with the average pH being 5.94. Soil pH influences the availability of heavy metals which are usually more available in a lower pH as most metals are cationic (USDA NRCS 2000). The soil EC at AHSL ranged from 115.1 to 293.00 µS cm^{-1} with the average being 219.4 µS cm^{-1}. In general, soils with EC below 300 µS cm-1 are considered to be sterile soils with little microbial activity, making it 'unhealthy' for plant growth as essential enzymes might not be present for the synthesis of soil nutrients (Karam *et al.* 2013; Masri *et al.* 2022; Shaliha-Jamaluddin *et al.* 2022). This indicates that the plants growing at AHSL are primarily pioneer species, that is plants that grow in disturbed ecosystems. The soil bulk density ranged from 1.40 to1.73 g cm^{-3}; 11.56 to 12.50% moisture content and 34.91 to 47.16% porosity. Generally, soils having a bulk density of between 1.0 g cm^{-3} and 2.0 g cm^{-3} are considered to have a low organic content, which is likely, as AHSL has a highly weathered and exposed soil. The low porosity was due to the soil being compacted to cover the municipal waste buried beneath. This also explains the low moisture content of the soil. The concentration of extractable phosphorus was low in all AHSL phases. Low phosphorus is often associated with high concentration of Fe in the soils, which is characteristic of an Oxisol (Karam et al., 2013). Low organic matter is also often associated with lower NPK levels in soils, which is evident at AHSL (Cavanagh and O'Halloran 2003).

Soil Cd Concentrations

The average soil Cd concentrations ranged from 0.018ppm to 0.029pmm, with the lowest being at Phase 1 and the highest at Phase 7. However, a one-way ANOVA between subjects found no significant different between the phases in Cd concentrations [$F (2, 87) = 1.442$, $p = 0.242$]. Similarly, there was no significant difference in Cd concentrations when it came to soil depth. A linear regression established that soil depth could not predict Cd concentration with statistical significance [$F (1, 99) = 0.287$, $p = >0.05$]. As shown in Figure 5 Cd concentration decreased with soil depth but was not statistically significant. Soil Cd concentration at the study site was not from the buried municipal waste since the waste was buried deep underground and had liners surrounding it. Thus, the Cd was largely from atmospheric deposition, as further explained in the next section.

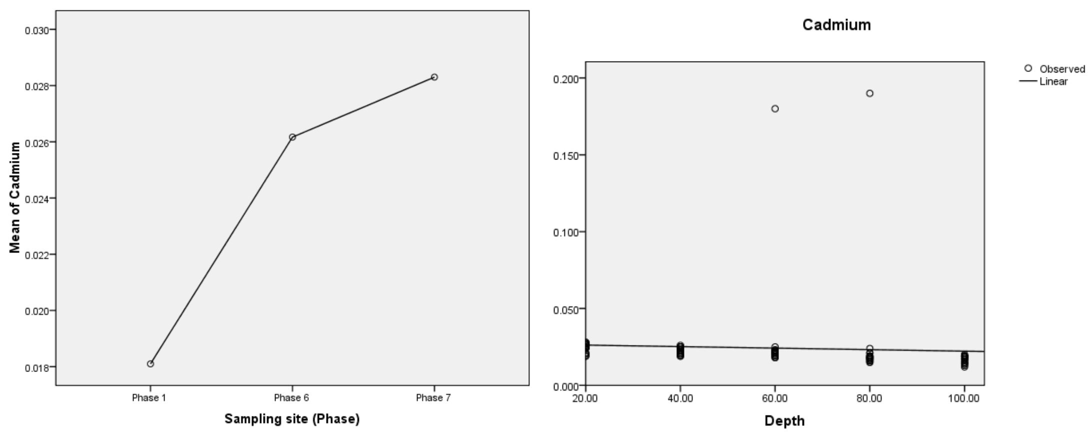

Figure 5. Cd concentrations according to sampling phase (left) and soil depth (right).

Effects of Atmospheric Deposition on Topsoil Cd Concentration

A t-test failed to reveal a statistically reliable difference between the mean concentration of Cd in the top soil (M = 0.021, s = 0.003) and in the dry atmospheric deposition (M = 0.025, s = 0.005), t (16.85) = 1.995, p = 0.062, α = 0.05 (Figure 6). This meant that dry atmospheric deposition was the likely primary contributor to Cd concentration in the topsoil of AHSL (Cullen and Maldonado 2012). At lower pH levels, Cd was found to be highly concentrated in mobile states, indicating that it is very likely to be an environmental hazard through dry atmospheric deposition (Lee *et al*. 2014). As the dissolution of Fe hydroxides can lower pH levels, it is likely that the high Fe concentration found in the dry atmospheric deposition samples was responsible for the mobile state of the Cd, thus contributing to high Cd concentration in the dry atmospheric deposition at AHSL (Lee *et al.* 2014). A high concentration of Cd is common in the dust of urban and industrial areas (Qiu *et al.* 2016). Cd can cause widespread ecological damage that could have severe repercussions on human health, usually affecting kidney, bone and lungs (Qiu *et al.* 2016).

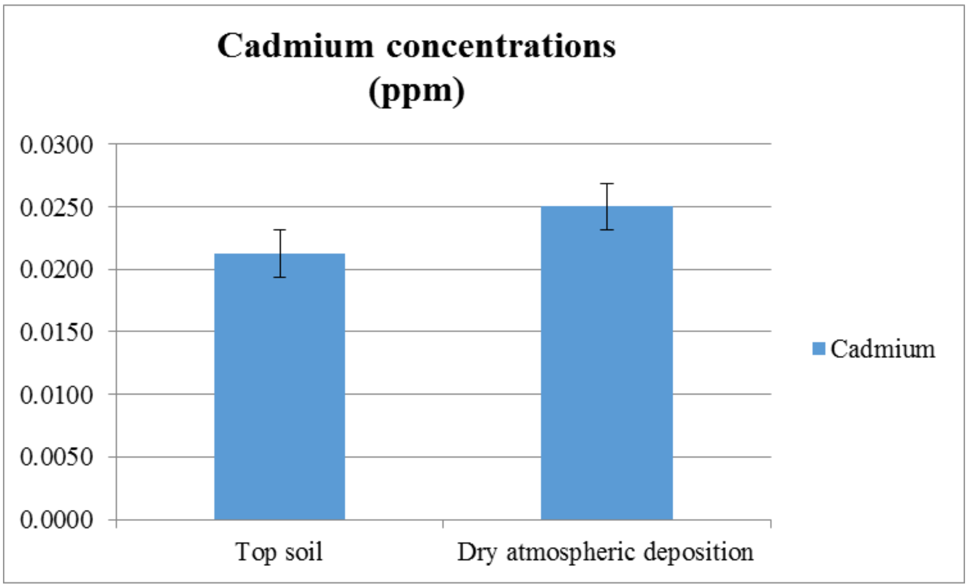

Figure 6: Mean Cd concentrations of topsoil and dry atmospheric deposition samples

Cd Concentration in Leachate Collection Ponds

As seen in Figure 7, there was a significant difference in Cd concentration in the leachate ponds [F (2, 33) = 7200.72, p = 0.0]. A Tukey post-hoc test revealed that the last leachate collection pond (Pond 3) had a statistically significant higher concentration of Cd (0.393 ± 0.015 ppm) compared to Pond 1 (0.005 ± 0.002 ppm, p = 0.0) and Pond 2 (0.013 ± 0.001 ppm, p = 0.0). There was no statistical difference in Cd concentration between Pond 1 and Pond 2 (p = 0.061). This shows that Cd concentration did not decrease after aeration treatment as expected, but actually increased.

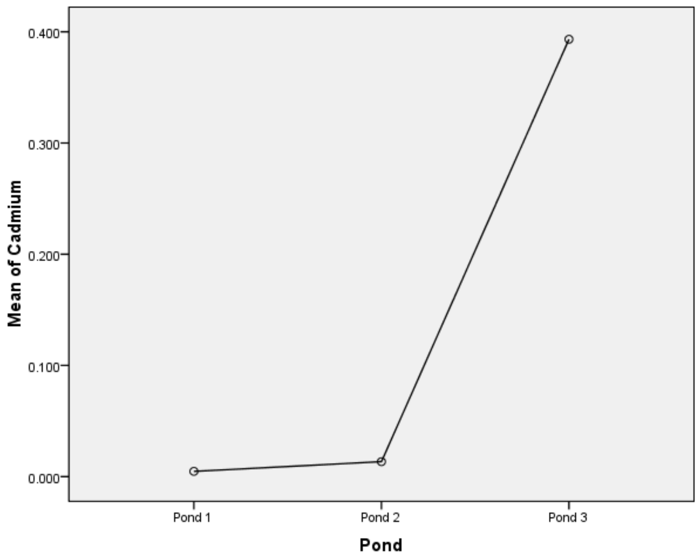

Figure 7. Mean concentration of Cd from leachate collection ponds

As seen in Figure 8, there was a significant difference in Cd concentration in the leachate sediments [$F (2, 33) = 124.938$, $p = 0.0$]. A Tukey post-hoc test revealed that the sediment in the last leachate collection pond (Pond 3) had a statistically significant higher concentration of Cd (1.57 ± 0.12 ppm) than the leachate collection ponds of Pond 1 (0.905 ± 0.08 ppm, $p = 0.0$) and Pond 2 (1.22 ± 0.09 ppm, $p = 0.0$). Pond 1 had statistically higher Cd concentration in its sediments compared to Pond 2 ($p = 0.0$). This shows that Cd concentration increased even after aeration treatment, as in the case of leachate Cd concentration levels (Figure 7).

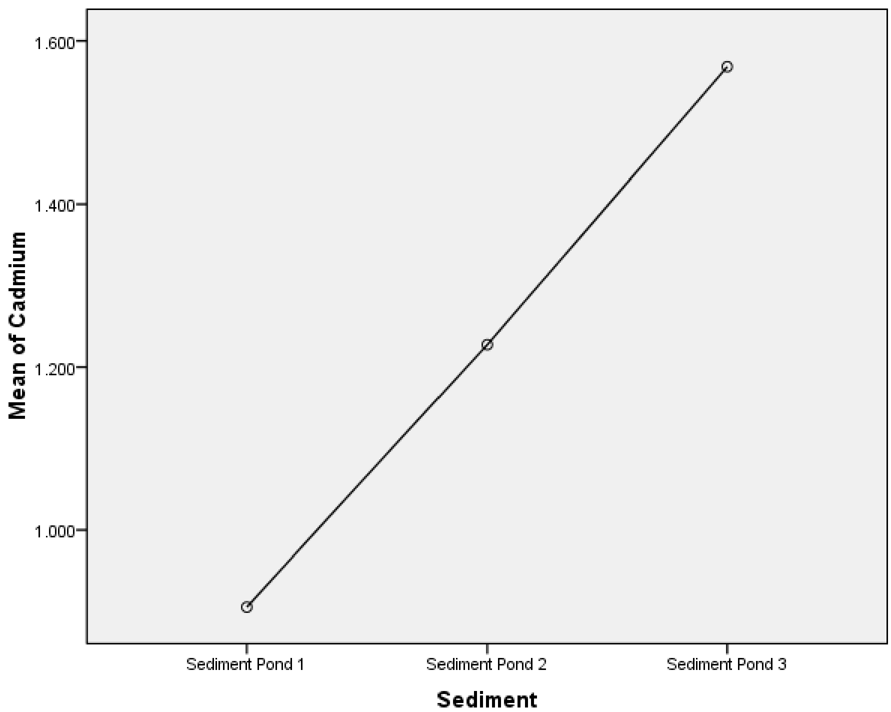

Figure 8. Mean concentration of Cd in sediments from leachate collection ponds

The Cd concentration in the sediments of AHSL leachate ponds was found to be several times higher than in the topsoils of Malaysia, indicating that the aeration treatment was able to solidify these elements. According to Rahman and Zaim (2015), Cd concentration in the sediments was almost 75 times higher than in our topsoils. Aeration treatment can be compared to a common wastewater treatment in the 1990s known as flotation, whereby heavy metals are separated from the liquid phase using bubble attachment (Lundh *et al*. 2000). The heavy metals rise towards the surface of leachate ponds and need to be removed as sludge. Hence, this treatment can only be effective if the sludge is removed, otherwise the sludge sinks to the bottom of the leachate treatment ponds along with the accumulated heavy metals (Lundh *et al.* 2000). The heavy metals then re-enter the leachate, which explains why the treated leachate of AHSL still contains high concentrations of Cd. Hence, for aeration treatment to be efficient in removing Cd, sludge removal also needs to be conducted.

River Cd Concentration

As river samples were collected from 25-m intervals, it can be determined whether river distance affected concentration of Cd. Linear regression analysis was conducted to determine river distance relationship with Cd concentration. Cd concentrations were generally lower with increasing river distance; however, the linear regression established that river distance could not predict cadmium concentration with statistical significance, $F(1, 8) = 14.065$, $p = >0.05$ (Figure 9).

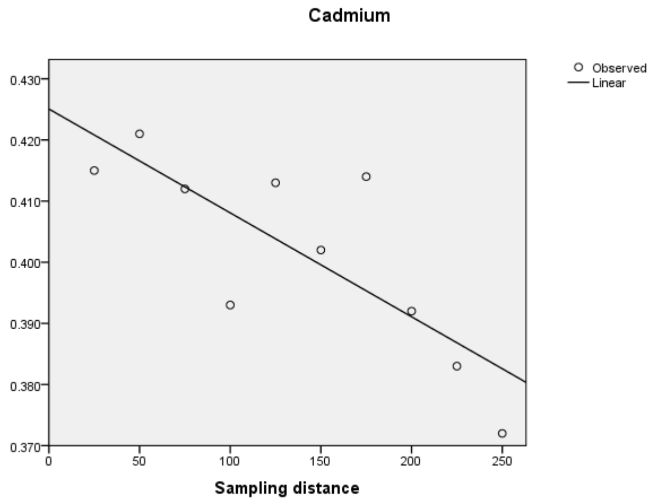

Figure 9. Cd concentration based on river sampling distance (meters)

Cd Content in Soil Arthropods

Two arthropod species were selected for this study, *Orthomorpha coarctata* and *Trigoniulus corallines* (Figure 10). The average density of *Orthomorpha coarctata* was 35.17±7.7 /400m^2, while the average density for *Trigoniulus corallines* was 14.33±5.6 /400m^2. The arthropod samples were divided into washed and unwashed samples. A one-way ANOVA between subjects was conducted to compare the heavy metal concentration in the washed arthropod samples, unwashed arthropod samples and the topsoil. If the results were significantly different, Tukey HSD test was conducted to determine the phases that were significantly different.

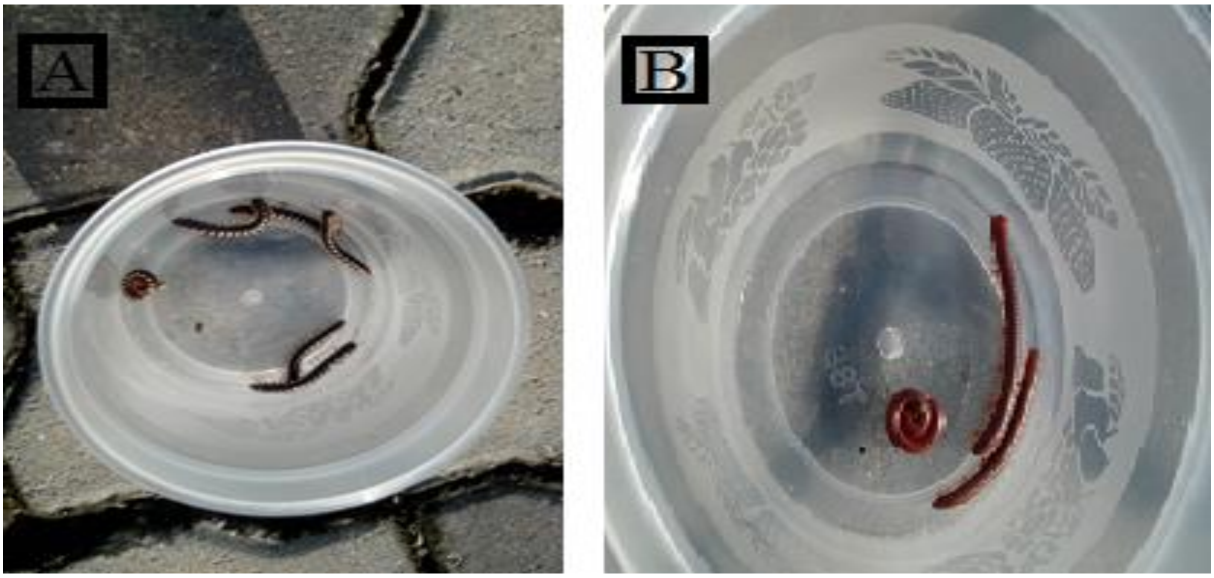

Figure 10. Orthomorpha coarctata (A) *and Trigoniulus corallines (B) samples*

There was a significant difference in Cd concentration [F (2, 33) = 13.081, p = 0.000] between top soil and *Orthomorpha coarctata* (washed and unwashed). A Tukey post-hoc test revealed that the topsoil had a statistically significant lower concentration of Cd (0.021 ± 0.003 ppm) compared to both the *Orthomorpha coarctata* samples, that is the unwashed samples (0.025 ± 0.001 ppm, p = 0.0) and washed samples (0.025 ± 0.001 ppm, p = 0.0). There were no statistical difference in Cd concentration between the unwashed and washed *Orthomorpha coarctata* samples. This indicates that *Orthomorpha coarctata* is an arthropod species that accumulates Cd (Figure 11).

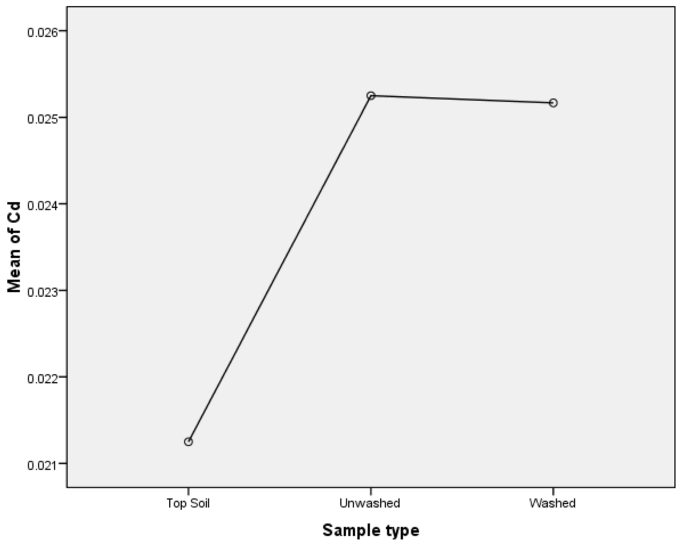

Figure 11. Mean concentration of Cd based on topsoil and Orthomorpha coarctata (washed and unwashed) samples

Similarly, there was a significant difference in Cd concentration [F (2, 33) = 4.149, p = 0.000] between top soil and *Trigoniulus corallinus* (washed and unwashed). A Tukey post-hoc test revealed that the topsoil had statistically significant lower concentration of Cd (0.021 ± 0.003

ppm) than in both the *Trigoniulus corallinus* samples, that is, the unwashed (0.023 ± 0.0008 ppm, p = 0.0) and washed samples (0.023 ± 0.0009 ppm, p = 0.0). No statistical differences were found in Cd concentration between the unwashed and washed *Trigoniulus corallinus* samples. This indicates that *Trigoniulus corallinus* is an arthropod species that accumulates Cd (Figure 12).

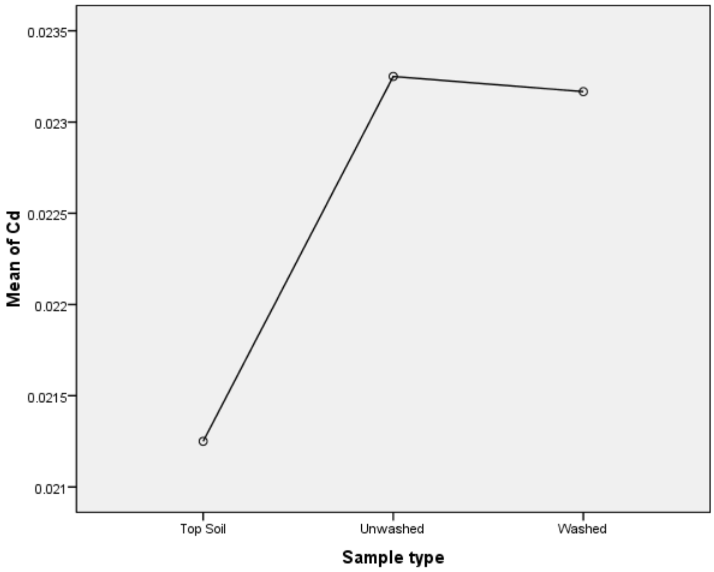

Figure 12. Mean concentration of Cd according to topsoil and Trigoniulus corallinus (washed and unwashed)

The results found that both *Orthomorpha coarctata* and *Trigoniulus corallines* are potential hyperaccumulators of Cd, which is quite a prevalent characteristic in arthropods (Boyd 2009). These two species could be used as bioindicators for Cd, making contamination control easier (Nummelin *et al.* 2007). However, this is also a cause for concern since there is potential for Cd to infiltrate human food chains via these soil arthropods.

Pollution Potential of Cd at ASHL

Land and groundwater contamination in Malaysia is addressed in the Environmental Quality Act 1974 (EQA), a legislation that focuses on the prevention, abatement and control of pollution in the environment of Malaysia (Rajoo *et al*. 2013). Further, there is also a set of guidelines titled 'Contaminated Land Management and Control Guidelines', produced by the Malaysian Department of Environment (DOE). Compliance to the guidelines is purely voluntary; however, DOE is seeking to make it mandatory. Despite these legislations and guidelines, Malaysia still has no specific definition of 'contaminated land and/or groundwater', no soil or groundwater quality standards and almost no government initiative to identify contaminated sites (Ripin *et al*. 2014). However, the Malaysian government did conduct a national study to create a set of standards for water quality in 1985 (DOE 1985). The study was called the 'Development of Water Quality Criteria and Standards for Malaysia' and was headed by a multidisciplinary team of experts from universities throughout the country (DOE 1985). Hence, when it comes to water quality in rivers or lakes, there is a Malaysian standard to refer to, unlike for soils and groundwater. Due to the lack of a standardised environmental quality standard in Malaysia, it is common for soil quality assessment in Malaysia to be conducted by comparing it to the standards of a foreign country; the most common being the Dutch Standard (Ripin *et al*. 2014).

The average concentration of Cd in the topsoil of AHSL was 0.021ppm and 0.025ppm for atmospheric deposition, which was significantly lower than the permissible limits of the Dutch Standard (0.8ppm). However, the average Cd concentration in the river (0.402ppm) exceeded the Dutch Standards target value (0.4ppm). It also significantly exceeded the World Health Organization's Standard for drinking water (0.003ppm) and the Canadian Environmental Quality Guidelines (0.09ppm). Even based on DOE's guidelines for Malaysia, the Cd levels was in the lowest category at Class V (higher than 0.01ppm).

Transference of Cd at Air Hitam Sanitary Landfill

Based on Cd concentrations in various biotic and abiotic factors at AHSL, the nature of Cd could be determined, that is, how it is transferred from one aspect to another and whether Cd accumulates in any of these biotic or abiotic factors. Cd concentration at AHSL is relatively high in all biotic and abiotic factors, with the concentration being in the range of 0.019 ppm to 1.568 ppm (Figure 13). The lowest Cd concentration was in the deep soil while the highest was in the leachate pond sludge. The plant's concentration of Cd was twice higher than in the topsoil and arthropod, meaning the distribution of Cd in the food web was primarily in the plants (Achary *et al.* 2017). This means that Cd contamination of our food chain could be possible and hence attention needs to be given to this aspect. Cd atmospheric deposition was significantly higher compared to topsoil concentration, meaning that Cd contamination via atmospheric deposition was prevalent at AHSL. This is likely due to the various industries and vehicles at Seri Kembangan that release Cd into the atmosphere (Qiu *et al.* 2016). Hence, more environmental measures need to be in place in order to reduce atmospheric release of Cd to prevent further environmental contamination.

The Cd concentration in the leachate pond's sediment was more than 70 times higher than the Cd concentration in most natural Malaysian soils, including the topsoil of AHSL, meaning that leachate aeration treatment was able to solidify Cd (Ripin *et al*. 2014). However, as the leachate sediment was not removed, it will be released into the river along with the treated leachate (Lundh *et al*. 2000). This is evident as reflected by the Cd concentration in the river being beyond the allowable limits of several international environmental guidelines and being in the lowest quality class for Malaysia's environmental guideline (He *et al.* 2015). All this indicates that Cd contamination is a serious concern at AHSL and could possibly be a threat to both the ecosystem and human health, namely to those residing near AHSL.

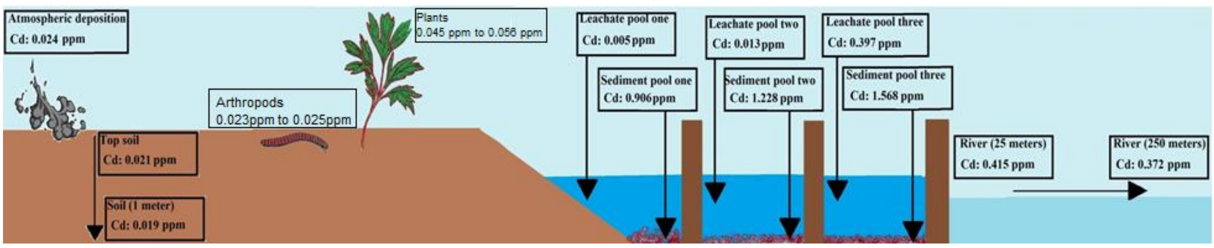

Figure 13. Concentration of Cd in various biotic and abiotic factors of AHSL

CONCLUSION

The major contributor of soil Cd in this urban ecosystem was atmospheric deposition. However, the bulk of Cd pollution came from the municipal waste buried at AHSL, as evident by the significantly high concentration of Cd in the leachate and sediments. Since the leachate and sedimentation are released into a nearby river after aeration treatment, Cd is then transferred into the river ecosystem. The Cd concentration level in this river is of major concern, as demonstrated by its values exceeding several environmental guidelines. Moreover, due to Cd being a highly mobile element, it is very likely that the heavy metal is seeping into our food chain via biotic factors. This is evident from the high Cd concentration in the soil arthropods and plants at AHSL. Further studies need to be conducted on the exact source of the Cd concentration in atmospheric deposition. There is also a need to assess the speciation of Cd to better understand its nature.

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