LEACHING OF HEAVY METALS FROM FLY ASH

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LEACHING OF HEAVY METALS FROM FLY ASH

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A final report submitted in partial fulfilment of the Final Year Project II (STF 3014) course

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2009
DECLARATION

No portion of the work referred to this dissertation has been submitted in support of an application for another degree of qualification of this or any other university or institution of higher learning.

_________________________
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TABLE OF CONTENTS

ACKNOWLEDGEMENT i
TABLE OF CONTENTS ii
LIST OF ABBREVIATIONS iv
LIST OF FIGURES vi
LIST OF TABLES vii
LIST OF APPENDICES viii
ABSTRACT ix

CHAPTER 1: INTRODUCTION 1
  1.1 Introduction 1
  1.2 Statement of Problem 2
  1.3 Objectives 3

CHAPTER 2: LITERATURE REVIEW 4
  2.1 Heavy Metals 4
  2.2 Fly Ash 6
    2.2.1 Physical and Chemical Properties of Fly Ash 7
    2.2.2 Uses of Fly Ash 8
  2.3 Soil 10
  2.4 Leaching Method 14
    2.4.1 Column Leaching Test 14
    2.4.2 Batch Leaching Test 15
  2.5 Previous Studies on Leaching of Heavy Metals from Fly Ash 16

CHAPTER 3: MATERIALS AND METHODS 18
  3.1 Sampling Location 18
  3.2 Characterization of Fly Ash 18
    3.2.1 pH value 18
3.2.2 Moisture Content 18
3.2.3 Electrical Conductivity 19
3.2.4 Specific Surface Area 19
3.2.5 Total Heavy Metals 20

3.3 Characterization of Soil 20
3.3.1 Particle Size 20
3.3.2 pH value 21
3.3.3 Total Organic Matter 21
3.3.4 Specific Surface Area 21
3.3.5 Cation Exchange Capacity 22
3.3.6 Total Metals Content 22

3.4 Experimental Design 22
3.4.1 Leaching Column Experiments 22
3.4.2 Statistical Analysis 23

CHAPTER 4: RESULTS AND DISCUSSION 24
4.1 Fly Ash and Soil Properties 24
4.2 Leaching of Heavy Metals From Fly Ash 27
4.3 Leachates Concentrations of Three Leaching Solutions 28
4.3.1 Cadmium 31
4.3.2 Zinc 32
4.3.3 Nickel 33
4.3.4 Manganese 34
4.3.5 Copper 35
4.3.6 Chromium 36
4.3.7 Iron 37
4.4 Electrical Conductivity and pH of Leachates Solutions 38

CHAPTER 5: CONCLUSION AND RECOMMENDATION 40
REFERENCES 41
APPENDICES 46
**LIST OF ABBREVIATION**

[(Ca, Na)(Al, Si₄)O₈]  Anorthite/Albite  
°C  Degree Celsius  
3CaO.Al₂O₃.3CaSO₄.32H₂O  Ettringite  
Al  Aluminium  
Al₆Si₂O₁₃  Mullite  
ANOVA  Analysis of Variance  
APHA  American Public Health Association  
ASTM  American Society for Testing and Materials  
B  Boron  
Ba  Barium  
BaCl₂  Barium Chloride  
Ca  Calcium  
CaCl₂  Calcium Chloride  
CaO  Calcium Oxide  
CaSO₄  Anhydrite  
Cd  Cadmium  
CEC  Cation Exchange Capacity  
Cr  Chromium  
Cu  Copper  
DOC  Dissolved Organic Carbon  
EPA  Environmental Protection Agency  
FAAS  Flame Atomic Absorption Spectrophotometer  
Fe  Iron  
Fe₂O₃  Iron Oxide  
Fe₃O₄  Magnetite  
g  Gram  
g m⁻²  Gram per Meter Square  
HCl  Hydrochloric Acid  
Hg  Mercury  
HNO₃  Nitric Acid  
K  Potassium  
LOI  Loss of Ignition  
m²g⁻¹  Meter Square per Gram  
Mg  Magnesium  
mg/kg  Milligram per kilogram  
MgO  Magnesium Oxide  
MgSO₄  Magnesium Sulphate  
ml  Milliliter  
mm  Millimeter  
Mn  Manganese
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Abbreviation</th>
<th>Definition</th>
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<tbody>
<tr>
<td>Mo</td>
<td>Molibdenum</td>
<td></td>
</tr>
<tr>
<td>mS/cm</td>
<td>Milisiemens per Centimeter</td>
<td></td>
</tr>
<tr>
<td>MSW</td>
<td>Municipal Solid Waste</td>
<td></td>
</tr>
<tr>
<td>N</td>
<td>Normality</td>
<td></td>
</tr>
<tr>
<td>Na</td>
<td>Sodium</td>
<td></td>
</tr>
<tr>
<td>Na₂EDTA</td>
<td>Ethylenediaminetetraacetic Acid Disodium Salt Dehydrate</td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>Nickel</td>
<td></td>
</tr>
<tr>
<td>OH⁻</td>
<td>Hydroxide Ion</td>
<td></td>
</tr>
<tr>
<td>Pb</td>
<td>Lead</td>
<td></td>
</tr>
<tr>
<td>S</td>
<td>Sulphur</td>
<td></td>
</tr>
<tr>
<td>Se</td>
<td>Selenium</td>
<td></td>
</tr>
<tr>
<td>SiO₂</td>
<td>Silica</td>
<td></td>
</tr>
<tr>
<td>SPLP</td>
<td>Synthetic Precipitation Leaching Procedure</td>
<td></td>
</tr>
<tr>
<td>Sr</td>
<td>Strontium</td>
<td></td>
</tr>
<tr>
<td>TCLP</td>
<td>Toxicity Characteristics Leaching Procedure</td>
<td></td>
</tr>
<tr>
<td>USDA</td>
<td>United States Department of Agriculture</td>
<td></td>
</tr>
<tr>
<td>wt.%</td>
<td>Weight Percentage</td>
<td></td>
</tr>
<tr>
<td>Zn</td>
<td>Zinc</td>
<td></td>
</tr>
</tbody>
</table>
LIST OF FIGURES

Figure 4.1: Leaching Behavior of Selected Elements from Fly Ash 28
Figure 4.2: Leaching Behavior of Cadmium After 15 Days of Stabilization 31
Figure 4.3: Leaching Behavior of Zinc After 15 Days of Stabilization 32
Figure 4.4: Leaching Behavior of Nickel After 15 Days of Stabilization 33
Figure 4.5: Leaching Behavior of Manganese After 15 Days of Stabilization 34
Figure 4.6: Leaching Behavior of Copper After 15 Days of Stabilization 35
Figure 4.7: Leaching Behavior of Chromium After 15 Days of Stabilization 36
Figure 4.8: Leaching Behavior ofIron After 15 Days of Stabilization 37
Figure 4.9: Electrical Conductivity Behavior of All Leaching Experiments 38
Figure 4.10: pH Behavior for All Leaching Experiments 39
## LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Table 3.1</td>
<td>Treatments for Column Leaching Experiments</td>
<td>22</td>
</tr>
<tr>
<td>Table 4.1</td>
<td>Physical and Chemical Characterization of Fly Ash and Soil</td>
<td>24</td>
</tr>
<tr>
<td>Table 4.2</td>
<td>Released Metals Amount (mg/kg) by Untreated Soil Leaching (Treatment 1) at Different Contact Time</td>
<td>29</td>
</tr>
<tr>
<td>Table 4.3</td>
<td>Released Metals Amount (mg/kg) by Leaching of Soil-Fly Ash with Deionized Water as the Leaching Solution (Treatment 2) at Different Contact Time</td>
<td>30</td>
</tr>
<tr>
<td>Table 4.4</td>
<td>Released Metals Amount (mg/kg) by Leaching of Soil-Fly Ash with Acidified Water as the Leaching Solution (Treatment 3) at Different Contact Time</td>
<td>30</td>
</tr>
</tbody>
</table>
LIST OF APPENDICES

APPENDIX A: Raw Data for Physical and Chemical Characteristics of Fly Ash 46

APPENDIX B: Raw Data for Physical and Chemical Characteristics of Soil 47

APPENDIX C: Leachates Concentrations, pH and Electrical Conductivity for all treatments 49

APPENDIX D: Statistical Analysis of Variance (One-Way ANOVA) for Leachates in all treatments 59
Leaching of heavy metals from fly ash

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ABSTRACT

Disposal of coal fly ash is a major issue due to its potential to cause surface and groundwater contamination. Stabilization is one of the effective methods of dealing with heavy metals in contaminated soil. In this study, the stabilization was evaluated using leaching tests. Fly ash samples was applied to the agricultural soil in the ratio of 1:9 in order to investigate the concentration of available metals (Cd, Zn, Ni, Mn, Cu, Cr and Fe) in leachates from 0 – 15 days of stabilization. The different mobilization media, namely deionized water and acidified water (deionized water acidified with nitric acid solution to pH = 4.0) was passed through to the soil-fly ash mixtures in the columns. Soil and fly ash samples were characterized for their physical and chemical properties as their properties greatly influenced the leaching potential. The heavy metals concentrations were analyzed by using Flame Atomic Absorption Spectrophotometer (FAAS). The results obtained from the leaching experiments showed that the addition of fly ash to agricultural soil had reduced the concentrations of available metals except for Cr. pH and electrical conductivity of leachates were analyzed and results found that pH increased while electrical conductivity decreased from 0 day to 15 days of contact time. Statistical analysis showed significant differences (P < 0.05) for particular metals between all treatments for almost at all contact time.

Keywords: Fly ash, heavy metals, soil, mobilization media, leaching, column.

ABSTRAK

Pembuangan abu terbang merupakan satu isu penting kerana keupayaannya menjadi punca pencemaran pada permukaan dan bawah tanah. Penstabilan merupakan salah satu teknik efektif yang berkaitan dengan kehadiran unsur-unsur di dalam tanah yang tercemar. Dalam kajian ini, penstabilan telah diuji dengan menggunakan ujian larut resap. Abu terbang diaplikasikan pada tanah yang digunakan khusus untuk bidang pertanian dalam nisbah 1:9 yang bertujuan untuk mengkaji kepekatan unsur-unsur yang dikehendaki (Cd, Zn, Ni, Mn, Cu, Cr dan Fe) di dalam larutan larut resap untuk 0 – 15 hari penstabilan. Larutan yang berbeza sifat untuk proses larut resap ini lait larutan yang dinyahcas dan larutan yang berasid (larutan yang dinyahcas ditambah dengan asik nitrik kepada pH = 4.0) telah melanai campuran tanah dan abu terbang di dalam kolum. Sifat-sifat kimia dan fizikal tanah serta abu terbang tersebut telah dianalisis kerana sifat-sifat tersebut mempengaruhi keupayaan proses larut resap. Kepekatan unsur-unsur telah dikaji dengan menggunakan alat Spektroskopi Penyera Panuan Nyalakan Atom. Keputusan yang diperolehi daripada eksperimen larut resap menunjukkan bahawa penambahan abu terbang ke atas tanah telah mengurangkan kepekatan unsur-unsur yang dikehendaki kecuali untuk unsur Cr. Nilai pH dan pengaliran elektrik larutan larut resap telah dianalisis dan keputusan mendapat bahawa nilai pH bertambah semantap nilai pengaliran elektrik berkurang dari hari 0 hingga ke hari 15 semasa proses larut resap berlaku. Kajian statistik menunjukkan bahawa terdapat perbezaan penting (P < 0.05) pada unsur-unsur yang dikehendaki di antara semua kaedah rawatan pada hampir keseluruhan hari penstabilan.

Kata kunci: Abu terbang, unsur, tanah, media untuk proses larut resap, proses larut resap, kolum.
CHAPTER 1

INTRODUCTION

1.1 Introduction

Nowadays, the consumption of energy had increased with the increasing demand. According to Kikuchi (1999), the global primary energy consumption has increased from 2500 Mtoe (million tons of oil equivalents) in 1971 to 3500 Mtoe in 1996. Coal is an important energy resource and it accounts for about 25% of total global primary energy. Solid wastes such as fly ash and bottom ash are discharged in large quantities by firing the coal to obtain energy. Almost 500 million tons per year of coal ash has been emitted from the thermal power plants, but only 15% constitutes the global recycling rate of coal ash. Recycling of these solid wastes is the efficient way to save the mineral resources. However, fly ashes are mostly deposited in landfills or coal mines. Recycling of these solid wastes also important in order to transform the coal ash into different useful forms such as zeolite, potassium fertilizer and desulfurizing agent.

Fly ash that produced from power generation can be considered as a waste or as a resource, yet to be fully utilized for subsequent useful application (Haynes, 2009). Environmental problems which occurred from the exploitation of waste materials such as fly ash can be avoided if the characteristics of these materials are well known. Fly ash can be considered as an important and feasible method of re-using waste products and restoring degraded areas and at the same time, organic matter and other essential nutrients that present in the material can be stabilized (Arbestain et al., 2008).
Fly ash has demonstrated beneficial use as soil amendment. This is due to the potential leaching of heavy metals when large amount of fly ash is applied into the soils. Therefore, the addition of fly ash to contaminated soil can help to increase the soil porosity, water-holding capacity, pH, conductivity and dissolved sulphate, carbonate, bicarbonate ion and other basic cations in soils. Other than that, addition of fly ash to soil can promote sequestration of carbon dioxide in soil (Palumbo et al., 2005).

Value-added applications of fly ash would help to reduce the concern of fly ash disposal. In addition, fly ash has been proved to increase the physical and chemical properties of soils in order to stabilize the soil for further usage. Fly ash will be mixed with wet soil and then, the mixture will be compacted and allowed to cure to gain strength (Cheung and Venkitachalam, 2000). Therefore, productive use of fly ash would have considerable environmental benefits such as reducing air and water pollution (Sezer et al., 2006).

1.2 Statement of Problem

Generally, fly ash is landfilled after stabilization process. High heavy metals content in fly ash is one of the main issues conditioning its utilization. So, it is crucial to detoxify this residue for wider ranges of reutilization or applications, thus avoiding deposition in landfills which can lead to other environmental problems.

Hence, this research is vital to acquire better knowledge about our local fly ash properties and its leaching characteristics. Studies have shown to be successful with fly ash as soil amendment for agriculture purposes. Hence, this study was conducted to examine the application of fly ash to soil and also the leaching of heavy metals from fly ash through column experiments.
1.3 Objectives

The objectives of this study are to:

a) characterize soil and fly ash samples for particle size, pH, organic matter, specific surface area, cation exchange capacity (CEC), total metals content, moisture content, electrical conductivity (specific conductivity) and heavy metals content (Cd, Zn, Ni, Mn, Cu, Cr and Fe).

b) evaluate the effects of fly ash on agricultural soil by conducting the column leaching experiments.

c) compare the concentrations of heavy metals in leachates with the application of both de-ionized water and de-ionized water acidified with nitric acid (HNO₃) by one-way ANOVA analysis.
CHAPTER 2

LITERATURE REVIEW

2.1 Heavy metals

Heavy metal fractions are defined as “mobile” fraction, “mobilisable” fraction and pseudo total content. Mobile fraction indicates that the metals are effectively soluble, very active and bioavailable. Mobilisable fraction means that the metals are potentially bioavailable, leachable and also partly active, while pseudo total content indicates that the metals are inactive and inert (Calace et al., 2005).

The existence of heavy metals may be in a variety of forms, such as chloride, fluoride and sulphate. Most of them are toxic, carcinogenic and mutagenic. Heavy metal-bearing wastes pose a serious problem to human and animal health. This is due to the concentration of heavy metals in wastes which may vary in a wide range and sometimes may exceed the acceptance limit of the environment (Chen et al., 2009).

According to Chalermyanont et al. (2009), industrial and mining activities have introduced the heavy metals directly into the surrounding environment, thus contributing to extensive heavy metals contamination. Lead (Pb), cadmium (Cd), zinc (Zn), nickel (Ni) and chromium (Cr) are some of the elements that constitute one of the contaminant groups which have been considered as noxious to human health. They are commonly found in several types of waste and landfill leachates.

There are certain heavy metals that most likely to be the target environmental concerns, namely cadmium (Cd), lead (Pb) and mercury (Hg). They have no known biological
function and somehow, they are toxic to living organisms and have a potential risk for human and environmental health. The accumulation of these heavy metals in forest ecosystems is influenced by filtering effects in which high capacity of canopies to intercept the aerosols which are either one reach the soil through litterfall or washed down by rain. The other fraction of these heavy metals is released and distribute in the soil profile through physical and biological migration processes (Ernst et al., 2008).

The solubility of metal in soils are mostly depends on various parameters such as pH, liquid-to-soil ratio, type of metal and also contact time. From these parameters, pH value of soil and type of metal are the most important limiting factors that determine the metal solubility. The pH determines the equilibrium solubility that can be achievable, while the metal indicates its potential solubility at a given pH (Cao et al., 2009).

In landfills, heavy metals have the potential to leach slowly into the soil, groundwater or the surface of water. It is generally known that Cd, Ni, Zn, Cu, Pb, Cr and Hg are the heavy metals that identified in Municipal Solid Waste (MSW) landfill leachates (Karnchanawong and Limpiteeprakan, 2008). Most heavy metals are known to have low mobility in soil (Asci et al., 2008). Various heavy metals have been generally known to be dangerous to human health and also to the wildlife if there are high concentrations of heavy metals occur in the surrounding (Krishna and Govil, 2004). Toxic metals are hazardous to living organisms and it is determined by the chemical forms of the metals rather than the total concentration of the heavy metals (Madrid et al., 2007).
2.2 Fly Ash

Fly ash is one of the most plentiful and versatile industrial byproducts. It is a solid waste product created by the combustion of coal, carried out of the boiler by the flue gases and extracted by electrostatic precipitators or cyclone separators and filter bags (Sezer et al., 2006). The term ‘ash’ is referring to the inorganic material in a general sense. But, in a more precise term, ‘ash’ refers to the material that does not combust under prescribed set of conditions (Syed, 2007).

There are two major classes of fly ash namely Class C and Class F which were specified on the basis of their chemical composition resulting from the type of coal burned. Class F fly ash is normally produced from burning of bituminous coals or anthracite. This type of fly ash has pozzolanic properties and contains small amounts of lime and materials such as siliceous or siliceous and aluminous, which possesses little or no cementitious value and classified as low-lime. For Class C which was produced from burning of lignite or sub-bituminous coals, it usually contains a significant amount of calcium hydroxide along with pozzolanic materials and cementitious properties, thus classified as high-lime (Sezer et al., 2006).

The concentrations and distribution of some trace elements in fly ash are determined by the type of coal and combustion process. Through the combustion process, the concentrations of trace elements in fly ash can be enriched up to 30 times relative to the feed coal. Moreover, the trace element leachability and toxicity are depending on their distribution and speciation in the fly ash. The enrichment of heavy metals in fly ash is strongly depends on the fuel, where it is found that the hazardousness of MSW is expected to be higher. Fuel gas temperature is also giving a big influence to the dictation of heavy metals integration on the
fly ash fraction. It is found that the concentration of element such as cadmium is depends on the fuel and the fuel gas temperature (Lima et al., 2008).

2.2.1 Physical and Chemical Properties of Fly Ash

According to Miravet et al. (2006), fly ash is produced in a large amount by coal-fired power stations and industrial waste incinerators, and has been distributed widely in the environment. The particles of fly ash are in the form of inhomogenous, highly diverse, dispersed in a broad range of sizes and have different morphologies.

Fly ash has a very fine structure, glass-like particles which have particle size in the range of 0.01 to 100 mm. The nature of coal, type of emission control devices, conditions of combustion process and methods for storage and handling greatly influence the chemical, physical and mineralogical properties of fly ash. Fly ash also has a low bulk density, light texture, high surface area and some of them can have neutralizing capacity of acid soils. Ashes with a high neutralization capacity can increase the pH value of ash/soil and ash/organic matter mixtures when soil or organic wastes are mixed with the surface layers of the fly ash. They will also maintain a high pH within the substrate of the ash disposal site for a long period of time and buffer acidifying reactions, for example nitrification process within the ash deposit (Haynes, 2009). It appears from light to dark-gray powder of predominantly silt-size (Sezer et al., 2006).

A study conducted by Chien and Juu (2001) found that fly ash contains inorganic chemical constituents such as oxides of silicon, alumimium, iron and calcium. Smaller amounts of magnesium, sulfur, sodium, potassium as well as very small quantities of trace elements also presence in fly ash. In addition, fly ash contains an abundant amount of alkaline
materials such as calcium oxide (CaO), magnesium oxide (MgO) and also hydroxide ion (OH\(^-\)). Therefore, due to these compositions, pH value had increased during the adsorption process.

According to Mukherjee et al. (2008), fly ash consists mainly of aluminosilicate glass, mullite (Al\(_6\)Si\(_2\)O\(_{13}\)), silica (SiO\(_2\)), magnetite (Fe\(_3\)O\(_4\)), anorthite/albite [(Ca, Na)(Al, Si\(_4\))O\(_8\)], anhydrite (CaSO\(_4\)), ettringite (3CaO.Al\(_2\)O\(_3\).3CaSO\(_4\).32H\(_2\)O), opal (SiO\(_2\)), iron oxide (Fe\(_2\)O\(_3\)), and in varying amount of lime (CaO). The chemical composition and physical properties of fly ash depends very much on some factors such as composition of the coal and its source, the particle size of the coal, burning condition and temperature of the boiler.

Fly ash also contains a high level of nutrients that are beneficial for the growth of plants because it can supply some elements such as potassium and phosphorus to replace the commercial fertilizer. In addition, fly ash supplies micronutrients such as Ca, Na, K, Mg, B, S and Mo. These elements are suitable for plants growth and to alleviate the condition of nutrient deficiency in the soil. But, if the nutrients cannot be recycled, it can be a waste of resources (Wang et al., 2007). Quartz, mullite and hematite/maghemite are the major constituent of minerals in the fly ash (Lima et al., 2008).

### 2.2.2 Uses of Fly Ash

Fly ash is produced from the burning process of lignite coal. It contains a relatively lower amount of silica, therefore, it has been used for incorporation in cement and concrete. (Vasilopoulos et al., 2009).

Fly ash may be used as a raw material for various industrial products as well as limiting material because fly ash acts similar to lithospheric (soil-like) materials (Mukherjee et
al., 2008). According to Wang et al. (2007), fly ash could be used as a liming agent in agricultural field due to the composition of lime which is also known as calcium oxide that mainly used as a common agent to increase the pH of soil. The alkaline properties of fly ash is good for soil liming material for agriculture and help to eliminate the residual pathogens that present in the sludge compost (Papadimitriou et al., 2008).

Studies have shown that alkaline fly ashes could be used as a stabilizer or binding reagent for fixing of heavy metals and also nutrients that present in hazardous and organic wastes. Other than that, fly ashes also functioning as adsorbents for wastewater treatment or air pollution control. When it is converted to zeolite or zeolite-like material with a large amount of cation exchange capacity (CEC) and also specific surface area, it becomes one of the effective ways for the removal of pollutants (Chien and Juu, 2001).

Fly ash is frequently used in cement and concrete, structural fills, stabilized roadbase, flowable fills, in waste stabilization and also in soil stabilization. The addition of fly ash to soils could permanently alter the soil texture, increase the micro porosity and thus, improving the water retention capacity. It has certain physical and chemical properties that suitable for soil amendment. Therefore, it is used in a number of countries for stabilization of soils and for the construction of layers of hydraulically bound aggregates (Kolias et al., 2005).

In the aspect of agriculture, fly ash in combination with other by-products such as sewage sludge or bio-solids have the capability to decrease the infiltration rate, lower bulk density as well as to overcome some nutrient deficiency in soils, thus improve the physical properties of soils for better crop production. Fly ash also contains mostly all elements for plant growth in varying concentrations except nitrogen (Mukherjee et al., 2008). Before ash
residue can be utilized for subsequent use, its chemical as well as leaching characteristics must be known because fly ash has a high potential to be the agent for soil remediation by improving the soil fertility (Dahl et al., 2008). According to Papadimitriou et al. (2008), fly ash has the capacity to improve the physical and chemical properties of soils.

2.3 Soil

Soil is the collection of natural bodies on the earth’s surface containing living matter and capable of supporting plants. Soil formation takes place whenever hard rocks weathers and accumulates in sufficient quantities to form a medium for plant growth. This accumulation may take place by the rock weathering or it can be caused by the actions of wind, water or ice. Once these weathering products accumulate, soil formation starts (Sopher and Baird, 1982).

There are three types of soil, namely clay, silt and sand soil. Soil pores vary greatly in size ranging from less than 2 mm. The smallest pores can act as a filter for metal binding of microorganisms, thus, transport of the metal through the soil is prevented (Asci et al., 2008). Humic substances are significant constituents of the organic matter in soils (Adani et al., 2007).

According to Odlare et al. (2008), the quality of soil is determined by chemical, physical and biological components of the soil. Soil quality is also define as the capacity of a soil to sustain biological productivity, maintain environmental quality, and promotes plant and animal health. Changes in the soil microbial occur more rapidly compared with the changes in the other soil characteristics. Therefore, soil microbial processes are thought to be sensitive indicators of changes in soil quality. Organic waste also play a role in enhance the soil quality. It can be a valuable conditioner of soil and source of plant nutrients.
The major risks due to metal pollution of soil consist of “mobile” form of metals which can lead to groundwater and play a potential toxic role to plants and/or animals by taking place in the food chain (Calace et al., 2005). The contamination of soil from these heavy metals is due to the use of sludge or urban composts, pesticides, fertilizers and emissions from municipal waste incinerators and metal smelting industry. Heavy metals are highly persistent in soil. Thus, the accumulation of these heavy metals can have a deleterious effects on soil fertility, affect the ecosystem stability and constitute a health risk to human and other living organisms (Sun et al., 2001).

There are various in-situ and ex-situ techniques including excavation, stabilization, solidification, soil washing, electroremediation and phytoremediation that have been employed in order to contradict the impact of metals in the soil environment. The stabilization technique could be employed if waste-derived organic matter is used. This technique relies on the ability of organic matter to bind metals, subsequently reducing their bioavailability and allowing the vegetation to take place. Revegetation helps to diminish the spread of pollution by wind or water erosion. Thus, reduces water uptake and subsequent metal leaching (Tandy et al., 2009).

Total elemental content represents the maximum levels of heavy metals in the soil or sludge and is used as an indication of a worse case scenario in assessing the potential for heavy metal and other trace element contamination. On the other hand research found that the complexes of cadmium with fulvic acids were positively correlated with the soil organic matter content and the cation exchange capacity value of the soil. Trace metal extractability from, and mobility in, contaminated soils depend not only on total concentration in the soil but also on the metal speciation in the soil solution. The mobility of trace metals is controlled by the chemical forms or complexes in which they occur in the pore water of soils (Stephan et al.,
The chemical behavior of heavy metals in soils is mainly controlled by some criteria, such as the release of metal cation from contamination source materials, for example, fertilizer, sludge and etc, the cation exchange capacity, specific adsorption onto surfaces of minerals and soil organic matter and also on the precipitation of secondary minerals (Voegelin et al., 2003).

Soils are also known as the effective agents for metal sorption because of their high surface area and the presence of various surface functional groups. Soil properties, especially the content of Fe, Al, and Mn oxides and hydroxides and the content of clay and organic matter, are believed to be the main controls of heavy metal sorption (Gong and Donahoe, 1997). In soils, the transportation of pollutants is affected by the capacity of soil to impede the passage of contaminants to the groundwater and to the subsurface runoff. The physical and chemical properties of soils determine the extent to which the soil retards downward movement of contaminants, and the rate of which contaminants in the soil are biodegraded or otherwise neutralized (Wong, 2003).

According to Houba et al. (2001), the cation exchange capacity is defined as the total sum of exchangeable cations that a soil, soil constituent or other materials can adsorb at a specific pH. CEC is an index of the soil’s capacity to exchange cations with the soil solution. It affects the ability of the soil to adsorb and retain cations and heavy metals. Soils that have high CEC and organic matter can exchange and retain large amounts of cations released by the manure mineralization process. Cations are held to the soil particles by adsorption and can be returned to the soil solution for plant use by the exchange process.