

**A COMPARATIVE STUDY OF THE TRACE ELEMENTS  
CONTAMINATION AROUND THE E-WASTE RECYCLING SITES AND  
THEIR TREATMENT WITH AGRICULTURE WASTE**



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(2015)**

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**A thesis submitted in partial fulfilment of the requirement for the degree of**

**Master of Science  
in  
Environmental Science**

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## **CERTIFICATE OF APPROVAL**

This dissertation submitted by **Mr. Azhar Iqbal** is accepted in its present form, by the Institute of Environmental Sciences and Engineering (IESE), School of Civil and Environmental Engineering (SCEE), National University of Sciences and Technology (NUST), Islamabad as satisfying the partial requirement for the degree of Master of Science in Environmental Science.

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*I dedicate this thesis to my wife for her encouragement and support during my studies, to my children Irsa, Mahad and Moeed. I am also thankful to my mother who prayed a lot for my success.*

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## **LIST OF ABBREVIATIONS**

E-waste	Electrical and Electronic waste
WEEE	Wastes from Electrical and Electronic Equipment
TCLP	Toxicity Characteristic Leachate Procedure
USEPA	United States Environmental Protection Agency
AAS	Atomic Absorption Spectrometer
PCBs	Printed Circuit Boards
CRT	Cathode Ray Tube
PWB	Printed Wiring Board
RH	Rice Husk
PH	Peanut Hull
Pb	Lead
Cd	Cadmium
Cr	Chromium
Ni	Nickel
Cu	Copper
Zn	Zinc
Co	Cobalt
Mn	Manganese

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## ABSTRACT

The electronic waste (e-waste) is generated by collection, transportation, dismantling, crushing, disposal and informal recycling of electrical and electronic devices. The legal and or illegal export of electronic devices from developed countries to developing countries including Pakistan, India and Sri Lanka has become a subject of growing environmental concern. The informal recycling and unsafe disposal of e-waste is causing health risks due to its effects on the environment. This is first ever study conducted in Pakistan to assess the heavy metals contamination of the soil/ash collected from e-waste recycling sites in Karachi, Rawalpindi and Kamoki area. Eight heavy metals namely Pb, Cd, Cr, Ni, Cu, Zn, Co and Mn were studied using USEPA approved method “*Toxicity Characteristic Leaching Procedure (TCLP)*”. The analysis performed on Atomic Absorption Spectrometer (AAS) showed that the concentration Cd, Cr, Ni and Cu were higher as compared to the permissible limits of EU, USEPA and Indian Standard. It may be noted that there is no standard in Pakistan for permissible limit of heavy metals in soil. The concentration was found significantly different at the selected sites, which is attributed to the complex type of waste and process adopted. The concentration of heavy metals found from different sites revealed that computer, circuit boards, monitors, home appliances and drill machines are being burned at Karachi, computer, circuit boards, monitors, mobile phones are being burned at Rawalpindi and computer, circuit boards and wires are being burned at Kamoki.

The removal of heavy metals was also studied by using locally available, low cost agriculture waste like rice husk and peanut hull. Batch sorption studies were conducted to find the effect of pH, contact time and dosage of adsorbent. The results revealed that rice husk gives optimum sorption results 80 % at pH 9 with 8 g /100 ml adsorbent dosage at 4 hours contact time, whereas the peanut hull gives better sorption results 91 % at pH 5 with 8 g / 100 ml adsorbent amount at 4 hours contact time respectively at constant temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$ . These results showed that peanut hull act as better adsorbent than rice husk for the removal of heavy metals from e-waste leachate solution.

# Chapter 1

## INTRODUCTION

The use of electronic devices including the computers, tablets computers, mp3 and mp4 players and cellular phones, is prevalent in developed countries and increasingly becoming a fashion in developing countries. These electrical and electronic (EE) devices bring convenience in our daily life, and promote efficiency in ways of communication and information transfer. However, besides these benefits, they have also given rise to a series of environmental, health and socioeconomic issues (Berkhout and Hertin, 2004; Schmidt, 2006; Prakash et al., 2010; Orlins and Guan, 2015; Umair et al., 2015).

The e-waste also contains valuable components such as precious metals (gold, silver and copper etc..) which can be recovered by recycling process (Akcil et al., 2015). However, it also contains hazardous components such as heavy metals and organic pollutants, which are directly and or indirectly threat to the environment and can be toxic to human health if consumed through dietary intake (Labunska et al., 2015).

### 1.1 E-waste

Currently, there is no typical definition for e-waste in the world (Kahhat et al., 2008), however, e-waste is a term used to describe finished or discarded electrical and electronics (EE) devices, for example, home appliances, air-conditioning system, digital toys, desktop computers, laptop computers, tablet computers, computer monitors, Light Emitting Diode (LED) and Liquid Crystal Display (LCD) monitors, Cathode Ray Tube (CRT) Tele-visions (TV), mp3 and mp4 players, cell phones, keyboards, computer mice, printers, and copiers. The European Commission generally terms e-waste as **Wastes from Electrical and Electronic Equipment (WEEE)**. E-waste contains more than 1000 different components, most of these contain heavy

metals which are dangerous to environment and human health (Gaulon et al 2005). E-waste require special handling and recycling because it contains both valuable and hazardous materials. It is also chemically and physically distinct from other forms of municipal or industrial waste.

There is no single approach for management of e-waste, there are different phases commonly used for its management, such as the development of eco-friendly devices, proper collection of e-waste, suitable methods and techniques to recover, recycle, dispose of e-waste, ban on export/import of second-hand electronic and electrical devices to developing countries, and raising of awareness among the stakeholders. (Kiddee et al., 2013).

The technique of “Reduce, Reuse, and Recycle” can be applied here.

- i. **Reduce:** the e-waste through effective procurement system and good maintenance.
- ii. **Reuse:** through donation or selling it to others who can still use it.
- iii. **Recycle:** the products which cannot be repaired through formal processes

## 1.2 Global Perspective

The modern life style of people encourages the use of electronic devices for ease in daily life. Hence, the world is generating a huge number of e-waste and its management is considered as a progressive global issue. This is due to the day by day increasing masses of complicated end-of-life electrical and especially the electronic equipment e.g., computers and mobile phones. It was projected in 1994 that approx. 20 million computers (approx. 07 million tons) became obsolete. However, this figure was to increase to about 100 million personal computers (PCs) by 2004. A study investigated that approximately 500 million PCs reached the end-of-life between 1994 and 2003, this amount of PCs contain about 2,872,000 tons of plastics, 718,000 tons of lead, 1363 tons of cadmium and 287 tons of mercury (Puckett et al., 2002). This fast growing waste stream is accelerating because lifespan of a PC is decreasing rapidly in the global market (Culver, 2005). A study enlightened that there are 47.92 million mobile phones discarded in 2002, and it extended to 739.98 million in China in 2012 (Li et al., 2015). Currently the global production of

e-waste is predicted to be 20–25 million tons per year and Europe, America and Australia are biggest e-waste producers. According to a study, the Eastern Europe, America and China will be the major e-waste producers in next ten years (Robinson, 2009).

The international regulations to control the transboundary movements of hazardous wastes and their disposal including e-waste was developed under the Basel Convention, which enforced in 1992. However, these regulations are not implemented effectively. Though, at global level efforts have been made to recognize past, present and future e-waste, but global perspective is still lacking (Widmer et al., 2005).

### **1.3 Regional perspective**

Currently, informal and unsafe recycling of e-waste is a rising problem in many developing countries worldwide (Wittsiepe et al., 2015). A study shows that millions of e-waste produced in developed countries have been exported to the developing countries like Pakistan, India, Bangladesh, Sri Lanka and China every year for recycling (Agoramoorthy, 2006) due to inexpensive labour and weak enforcement of environmental law. These developing countries are facing several issues due to the generation, legal/illegal imports and overall management that includes recycling of e-waste (Herat and Agamuthu, 2012). Due to lack or absent of legislation, national regulation and / or weak enforcement of present e-waste recycling laws and high cost of recycling, e-waste are handled without considering the adverse effect on environment and human health (Widmer et al., 2005; Ogunseitan et al., 2009). There is also lack of awareness among authorities and deficiency of information about e-waste processes and the impact of improper handling of e-waste in Pakistan (Umair et al., 2015).

### **1.4 National perspective**

Pakistan is one of the big destinations of e-waste, along with Bangladesh, Sri Lanka, China, India, and Vietnam (Wu et al., 2015). The used electrical and electronic equipment enter in to Pakistan by several means e.g., legal and or illegal import and as donations. The devices end up

through informal recycling in Pakistan, which involves manual dismantling, burning of wires, extraction of precious metals through acid dipping (He et al., 2006) and recovery of gold and silver from e-waste (Natarajan and Ting, 2015). The e-waste is legally, formally and safely recycled in developed countries, but these processes required money and resources, hence, to save money, these devices are shipped to developing countries, such as Pakistan. Here these are recycled informally through primitive processes to extract gold, silver platinum and other precious metals (Umair et al., 2015).

In Pakistan, the e-waste recycling processes are carried out in populated areas, also focused in this study, causing environmental, health and socio-economic impacts. These unregulated, informal e-waste recycling activities has economic importance in Pakistan, because for many families informal e-waste recycling is a source of income. The people have created home industries and extract precious metals e.g., gold, silver and copper by using primitive methods. The whole family including women and children also contribute equally in recycling of e-waste. These informal activities are growing very rapidly due to weak enforcement and/or absence of legislation and lack of awareness among authorities. The families, workers and people living near the vicinity of recycling sites are more vulnerable to health problems (Frazzoli et al., 2010).



## **1.5 Hypothesis and objectives of the study**

There are 20-25 million tons of e-waste production in the world and approximately 200,000 tons of e-waste enter in Pakistan, where it recycled by using informal and primitive processes, hence causing serious contamination of heavy metals in soil and low cost agriculture waste can remove heavy metals from e-waste leachate solution.

### **Objectives**

- a) To assess the concentration of heavy metals at e-waste recycling sites
- b) To compare removal efficiency of rice husk and peanut hull using e-waste leachate solution.

### LITERATURE REVIEW

#### 2.1 Types of electronic waste

There is diversity of e-waste, some of the most common types of e-waste include home appliances, computers, monitors, TVs, audio and stereo devices, DVD, MP4 and MP3 players, digital cameras, photocopiers and fax machines, mobile phones and video game consoles and the list is going on. The types and examples of common e-waste in our daily life is listed at Table 1 below;

**Table 1:** Types and examples of e-waste

Types of e-waste	Examples
Large appliances	Ovens, washing machines, refrigerators, and cathode ray tube TVs
Small appliances	Toasters, vacuum cleaners and digital cameras
IT equipment	PCs and printers
Entertainment equipment	TVs, HiFis, portable CD players and MP3 players
Telecommunication	Mobile phones and faxes equipment
Audio and video data storage devices,	Hard drives, blue ray disc, CDs, USBs, DVDs, floppy disks and tapes media
Printer components	Cartridges (Ink & toner), laser printer drums and developer units
Miscellaneous items	Wiring, batteries, cables, and fluorescence lamps

**Source:** Microelectronics and Computer Technology Corporation (MCC). 1996.

#### 2.2 Trace elements in computers

The e-waste composition is very complicated and its management has been limited for both technical and economic reasons due to physicochemical complexity and rapidly growing volume (Zhang and Forsberg, 1997; Jang and Townsend, 2003; White et al., 2003; Zhou et al., 2004). Electronic and electrical devices are made of durable and generally non-biodegradable materials,

such as plastics and metals. The composition locations and materials found in a typical desktop computer (weighing ~27 kg) described in Table 2. The silica and plastic are the two most abundant materials in a desktop computer, whereas the metals including copper (Cu), iron (Fe), nickel (Ni), lead (Pb), zinc (Zn), aluminum (Al) and tin (Sn) are among the highest content in percent of the total weight of a desktop computer with collective weight up to 14.17 kg, accounting for over half of the total weight of a desktop computer. The weight of trace metals, Pb, Cu and Zn in a desktop computer can up to 1.91, 1.72 and 0.6 kg, respectively. The computers and computer accessories are exported to Pakistan for reuse, however, only a small amount has been reused and rest is recycled, this is one of the reason that these trace metals concentration were found higher at the recycling sites in Pakistan.

**Table 2:** Heavy Metals composition in typical desktop computer

Name of Material	Content (% of total weight)	Material Weight in computer (kg)	Used for	Location in computer
Gold	0.002	< 0.1	Conductivity & Connectivity	Connectors and PWB
Silver	0.019	< 0.1	Conductivity	Connectors and PWB
Lead	6.299	1.72	Metal joining	Funnel glass in CRTs, PWB
Copper	6.928	1.91	Conductivity	CRTs, PWBs, connectors
Nickel	0.850	0.23	Structural, Magnetivity	PWB, Housing, CRT
Zinc	2.205	0.6	Phosphor emitter, Battery	PWB, CRT
Manganese	0.032	< 0.1	Structural, Magnetivity	PWB, Housing, CRT
Chromium	0.006	< 0.1	Hardener and Decorative	Housing
Cadmium	0.009	< 0.1	Phosphor emitter, Battery	PWB, Housing, CRT
Cobalt	0.016	< 0.1	Structural, Magnetivity	PWB, Housing, CRT

**Source:** Microelectronics and Computer Technology Corporation (MCC). 1996.

### **2.3 E-waste as an environmental issue**

There have been several studies conducted around the world, wherein it is investigated that improper handling, disposal and informal recycling of e-waste cause environmental issues and air, soil & water are seriously contaminated near the locality of e-waste recycling sites. According to a study, it is reported that due to the absence or weak enforcement of e-waste legislation, lack of latest technologies and unawareness of the recyclers, e-waste is an environmental issue (Orlins and Guan, 2015).

E-waste is contaminating the environment in many ways and e-waste components e.g., home appliances, TVs, computer circuits, chips, gold plated components and plastic in electrical and electronic devices have potential environmental hazards (Wath et al., 2011). The e-waste components, their disposal methods and environmental hazards are given in Table 3.

According to a recent research conducted in South China stated that due to dismantling, open burning and acid digestion of e-waste, air and soil is contaminated by toxic substances like Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs) and is a potential risk to the environment (Ren et al., 2015). A research study explained that soil is contaminated by toxic metal like Ni, Cu, Zn, Cd, Pb, Sn and Sb due to primitive e-waste processing and there is a need for upgradation of e-waste recycling technologies and remediation for soil pollution control (Quan et al., 2015). The illegal and informal recycling activity has caused toxic metal pollution in soil and water, which is investigated near unrestricted e-waste recycling plant in Longtang, South China (Wu et al., 2015).

Li and co-workers have reported that the air of an e-waste processing area of Taizhou, Southeast China is contaminated by organic pollutants PCDDs, PCDFs, PCBs and PBDEs (Li et al., 2008). At a typical E-waste processing site, massive amounts of air pollutant PBDEs were estimated in the atmosphere of Guiyu, China especially in the daytime (Chen et al., 2009).

**Water:** A study conducted at water reservoir in the vicinity of several e-waste recycling workshops in South China, revealed that aquatic species including fish, Chinese Mystery snail, prawn, and water snake are seriously affected by organic pollutants (Wu et al., 2008). A study in China showed that the e-waste recycling sites are required ongoing monitoring and potential remediation to reduce the water contamination of hazardous material (Weber et al., 2008).

**Soil:** The soil near the vicinity of e-waste recycling sites is seriously contaminated by hazardous heavy metals and persistent organic compounds (Tang et al., 2010). A research explored the toxicological effects of the biodegradable chelate, ethylene diamine di-succinic acid (EDDS) and DGS6, the plant growth promoting bacterium on corn fields from e-waste-contaminated soils (Luo et al., 2015). A study in China investigated that the local soil and vegetables are seriously poisoned due to uncontrolled e-waste recycling activities (Luo et al., 2011).

**Plants :** A study on air and flora samples the “Eucalyptus spies and Pinus Massoniana Lamb” suggested that the unsafe e-waste recycling is a significant source of plants toxic substance “Dechlorane Plus (DP)” (Chen et al., 2011). The organic pollutants “Polycyclic Aromatic Hydrocarbons (PAHs)” found in soils and vegetation samples in the neighbourhood of an e-waste recycling place in China (Wang et al., 2012).

**Table 3:** The potential environmental hazards due to the improper disposal of different e-waste components

<b>E-Waste Component</b>	<b>Disposal Methods</b>	<b>Environmental Hazard</b>
CRT used in ATM machines, TVs, computer monitors, , cameras, and more	Breaking and removal of yoke, dismantling then land dumping	Heavy metals like lead and barium seeping into the ground water and release toxic materials
Printed Circuit Boards	Open burning, acid baths, removal of chips and desoldering and;	Air emissions as well as discharge of chemicals, electrical dust, tin, lead, brominated dioxin, beryllium cadmium, and mercury into land and water
Gold plated components	Chemical doffing using nitric acid and hydrochloric acid to dissolve gold and open burning of chips	Heavy metals, chemicals, Hydrocarbons, brominated substances discharged directly into water reservoirs, which acidifying the aquatic life. Contamination of surface and groundwater by tin and lead. Air discharges of brominated dioxins, heavy metals and hydrocarbons
Plastics material from printers, mouse, speakers, keyboards, monitors, etc.	Shredding and melting	Emissions of heavy metals, brominated dioxins, and hydrocarbons
Wires	Open burning and doffing to obtain copper	Hazardous gases and hydrocarbon ashes released into air, water and soil.

**Reference:** (Wath et al., 2011)

#### 2.4 Heavy metal impact and toxicity

Heavy metals are metallic elements having high atomic weight, relatively high density and are very toxic in nature such as lead, mercury, cadmium, chromium and nickel. These metals are harmful for all form of life (Munzuroglu and Geckil, 2002) . Human body needs trace elements (Solomons, 2015), however, intake of toxic metals by any mean even at very low concentrations, affect our enzyme systems and can also disturbed the metabolism of the body. Exposure to these elements may cause immediate as well as delayed adverse health effects like the toxic effects on

human skin (Rovira et al., 2015), respiratory problems (Valiulis et al., 2008) and several kinds of cancer (Igharo et al., 2015; Luo et al., 2015).

In this study, assessment of 8 heavy metals were conducted a various cities of Pakistan at e-waste recycling cites. The toxic effects of these heavy metals are discussed below.

#### **2.4.1 Lead (Pb)**

Lead is a persistent metal and present in water, soil, and air. The lead and lead compounds have toxic effects on all form of life including human health, plants, aquatic life and livestock (Demayo et al., 1982). A review study revealed that even low level exposures to lead environment causes renal impairment, hypertension, neurobehavioral disorders, cognitive dysfunction and neurological damage (Patrick, 2006). A study conducted among the Chicago school children reported that contact to lead has a risk to the educational performance, especially among poor children living in urban areas (Evens et al., 2015).

The lead toxicity in plants have been studied by Sharma and reported that as a result of anthropogenic activities the trace elements contamination of soils is rapidly increasing and lead is a toxic pollutant that accumulates in soils and sediments rapidly (Sharma and Dubey, 2005).

The lead has also serious effects on the aquatic life, a study conducted in Karachi Harbour, Pakistan reported the accumulation of lead in the dolphinfish (Ahmed and Benzer, 2015). A research investigated that heavy metals including lead may accumulate in the tissues of aquatic organisms and cause a range of hazardous effects, which can ultimately become harmful to humans (Sadeghi and Imanpoor, 2015).

#### **2.4.2 Chromium (Cr)**

Chromium (III) compounds are stable and occur naturally in the environment (Brewer and Prasad, 2015; Brownley et al., 2015; DeBruyne et al., 2015). A very small amount of Chromium (III) is required to maintain the glucose, lipid and protein metabolism in our body (Sadeghi et

al., 2015). However, Cr (VI) is toxic, non-biodegradable and is a carcinogenic in nature (Chen et al., 2007; Welling et al., 2015).

A research reported that Cr (III) is less toxic than Cr (VI) (Hilario and Jr, 2015), however, long term contact to a high concentration of Cr (III) may cause poisoning symptoms like allergic skin reactions (Abu-Zurayk et al., 2015). Acute exposure causes nausea and vomiting (Mona and Kaushik, 2015), shock coma and lung cancer (Von Burg and Liu, 1993; Tandon and Nandini, 2009), kidney and liver damage (Mishra and Mohanty, 2008), causes respiratory effects like ulcerated perforation of the nasal septum (Lin et al., 1994), irritation of mucous membranes and general bronchospasm (Moller et al., 1986). In aquatic organisms, Cr (VI) has a high potential for bioaccumulation in their bodies and cause chronic poisoning effects (Senthamilselvan et al., 2015).

### **2.4.3 Cadmium (Cd)**

All cadmium compounds are potentially harmful or toxic and acts as a cumulative poison (Sardella et al., 2015). The signs of acute cadmium poisoning include colitis, vomiting, digestive tract irritation, diarrhoea and prostration (Mahvi and Bazrafshan, 2007). A research investigated that cadmium causes cancer and cardiovascular disease (Larsson and Wolk, 2015). The several studies reported that cadmium toxicity caused kidney damage and also affects the activities of certain enzymes in the body (Zikić et al., 1997; Wasana et al., 2015).

A study described that long time exposure to cadmium causes affecting skin and internal organs such as urinary bladder, lungs and kidney (Elshenawy and El-Kadi, 2015), cadmium and copper in water increased the vulnerability of infectious virus such as hematopoietic necrosis (Hetrick et al., 1979).

In studies conducted on animals, cadmium has shown strong evidence of cancer (Nawrot et al., 2015). The cadmium even in low concentration causes behavioural, growth and physiological problems in aquatic life (Ahmed and Benzer, 2015; Frantzen et al., 2015; Meador, 2015).



#### **2.4.4 Copper (Cu)**

Copper is an essential nutrient for humans and animals, with a Recommended Daily Allowance (RDA) of 0.9 milligrams per day (Erdemir and Gucer, 2015) and is a component of many body proteins (Jaiswal et al., 2015). Acute exposure to copper causes gastrointestinal effects such as nausea, vomiting and diarrhoea and irritable bowel disease (Selmi et al., 2015). The copper also causes Wilson's disease (Bandmann et al., 2015) liver damage (Sampaziotis et al., 2015) and can lead to human death (Copper et al., 1994).

Copper has both acute and chronic effect on aquatic organisms that can result in death. They are known to bio-accumulate in some aquatic species. In mammals, overexposure to copper can damage the liver and kidney (Monser and Adhoum, 2002). A research investigated that the younger fishes severely affected by toxicity of copper, however, the adult fishes can tolerate small concentrations of copper for small periods of time (Ajmal et al., 1998).

#### **2.4.5 Nickel (Ni)**

The small amount of nickel is dietary requirement for many organisms, however, human exposure to extremely nickel-polluted environments may cause a variety of pathologic effects (Kasprzak et al., 2003). Their accumulation in the environment represented a serious hazard to human health and causes lung fibrosis, skin allergies, kidney and cardiovascular system damage and stimulation of neoplastic transformation (Denkhaus and Salnikow, 2002). Many researchers examined that acute nickel toxicity effects are gastrointestinal symptoms like nausea, vomiting, abdominal discomfort and diarrhoea and neurological symptoms like headache, giddiness, cough and shortness of breath. A report exposed that nickel compounds have been associated in the Etiology of human lung and nasal cancers (Costa et al., 2005). A research presented the toxic effects of trace metals including nickel in the aquatic life (Dave and Xiu, 1991). The scientists reported the binding of nickel and copper to fish gills predicts toxicity (Paquin et al., 2000).

#### **2.4.6 Zinc (Zn)**

The quantified amount of zinc is essential for human and Oral Zinc Supplementation (OZS) provides a clinically significant benefit in the treatment of neurological disease like dystonia (Xing and O'Suilleabhain, 2015). However, the excessive exposure to zinc can cause serious toxic effects including nausea, vomiting, lethargy, fatigue and epigastric pain (Fosmire, 1990).

Zinc is also toxic for aquatic life and a study investigated toxicity of zinc tolerance in juvenile brown trout and concluded that the smallest fish were shown to be less sensitive than the largest fish (Diedrich et al., 2015). The zinc pollution in the soil is a major problem for crop production worldwide (Paradisone et al., 2015).

#### **2.4.7 Cobalt (Co)**

Cobalt is a vital element for life in very small amounts and cobalt (LD<sub>50</sub>) value has been estimated to be between 150 and 500 mg/kg, hence, 20 grams amount of LD<sub>50</sub> would be required for a 100 kg person (Donaldson and Beyersmann, 2005). However, cobalt ingestion has caused serious health problems. A study in Canada investigated toxin induced cardiomyopathy caused by cobalt, which is known as beer drinker's cardiomyopathy (Morin et al., 1969; Barceloux and Barceloux, 1999). Cobalt is a major source of contact dermatitis (Basketter et al., 2003; Bregnbak et al., 2015), heart disease in beer drinkers (Kesteloot et al., 1968) and lung disease (Sjögren et al., 1980).

#### **2.4.8 Manganese (Mn)**

Manganese is not available as a free element in nature and is frequently found in combination with iron and other minerals. Manganese compounds are found less toxic (Rogalla and Römheld, 2002). However, even for short periods the contact to manganese powders and fumes should not be increased the maximum value of 5 mg/m<sup>3</sup>. The Manganese poisoning even a minute amount shows severe effects on the kidneys and liver (Young et al., 1996; Ong et al., 1997). In 2005, a

study investigated manganese toxicity in central nervous system in rats (Elsner and Spangler, 2005).

## 2.5 Health consequences of heavy metals

E-waste consist of a large number of hazardous heavy metals, chemicals and compounds, and is more hazardous than many other municipal wastes. Long term exposure to e-waste sites seriously affects the human health including nervous system, bones, kidneys, endocrine and reproductive system. Table 4 shows the sources & components of e-waste, and their effects on human health and environment.

**Table 4:** Sources of heavy metals and their effects on human health and environment

Source of e-waste	Components of e-waste	Environmental Hazard	Effects of Human
CRTs (used in computer monitors, TVs, ATM, Camera, etc.) Batteries, PVC cables, and Paints	Heavy metals such as Lead, Barium	These metals seeping into the ground water and release toxic substances	Insomnia, Anemia and Renal Toxicity,
Batteries, housing and medical equipment,	Metals like Mercury	Air emission as well as discharge into water reservoirs	Cerebral palsy, renal toxicity muscle tumors, mental retardation,
Plastics material from printers, mouse, speakers, keyboards, monitors, etc.	Plasticizer bisphenol as well as DEPH and DBP, plastic compounds	Chlorinated plastic release harmful chemicals into the soil, which leach into ground water or other neighboring water reservoirs which cause serious harmful effects to the species that drink this water	Heart problems, obesity, reproductive disease
Polymer & PVC, paints, inks, electrical transformers and capacitors	PCBs	Induced risky pollution from production, exposure to toxic chemicals during use, hazards from fires	Damage to the liver, nervous, reproductive and immune systems,

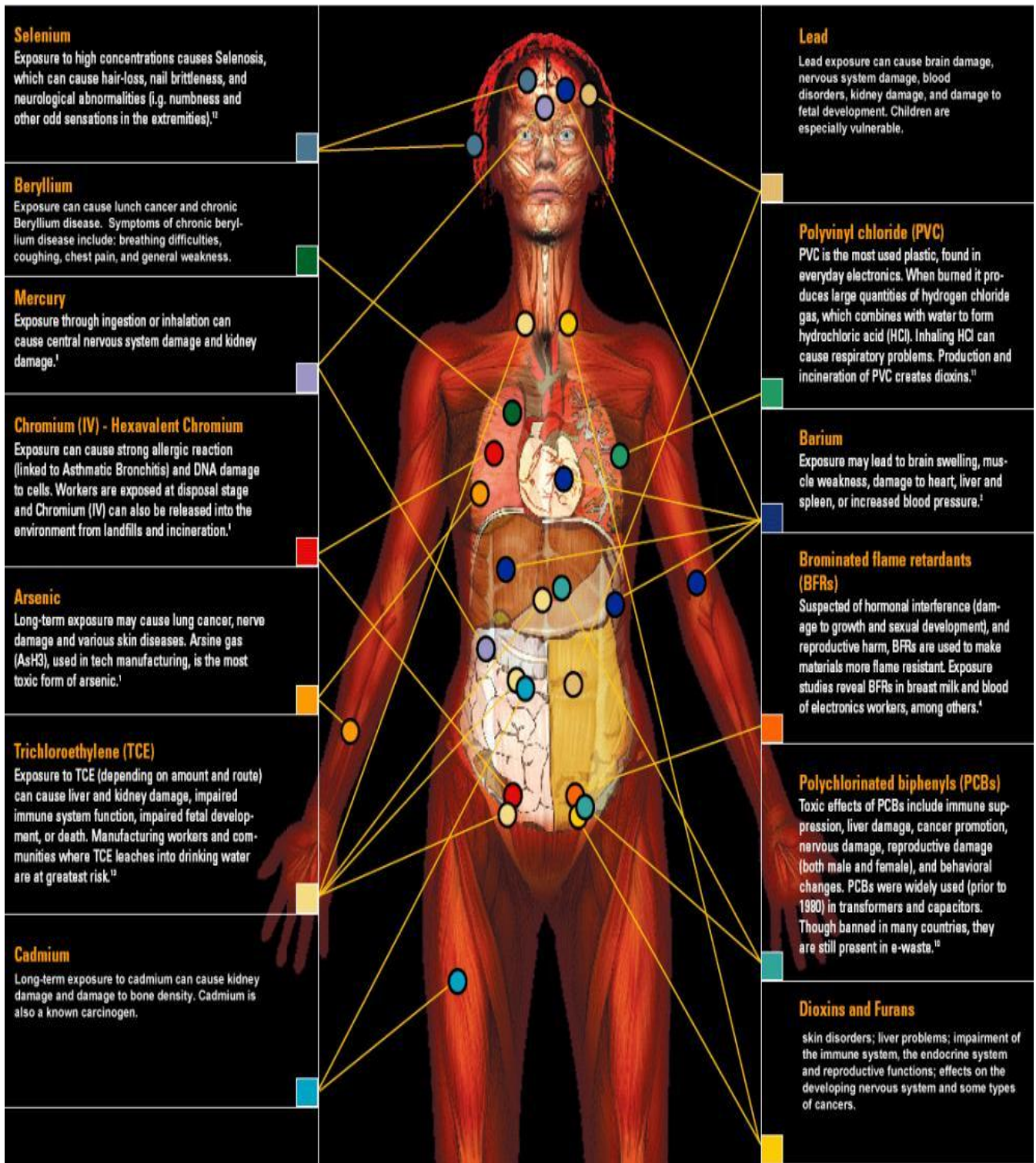
**Reference:** Aditi Rajput (2013)

The health of people residing in the vicinity of e-waste recycling areas are badly affected due to e-waste pollution. A research study in China revealed that the neonatal body length in Guiyu was

increased due to exposure of e-waste pollutant “Polybrominated Diphenyl Ethers” (Xu et al., 2015).

According to a study conducted at rural area in Hung Yen province in Vietnam explained that contamination of e-waste pollutants such as PBDEs, methoxylated PBDEs (MeO-PBDEs), hydroxylated PBDEs (OH-PBDEs), polychlorinated biphenyls (PCBs), hydroxylated PCBs (OH-PCBs) and bromophenols, seriously affect the hormones in the serum of human who are living near these areas (Eguchi et al., 2015). The worker’s health working on the e-waste plants are seriously ill due to unsafely handling of hazardous waste (Chen et al., 2015).

Nigeria is the largest e-waste dump yard in Africa and the management practices are entirely primitive. A recent study documented that the majority (88.8%) of Nigerian e-waste workers worked without personal protective equipment and significantly vulnerable to cancer risk (Igharo et al., 2015). E-waste is extremely toxic especially when not handled effectively. Some of the health consequences of e-waste components are given in Figure 1.



Source: The impact of e-Waste (2015)

Figure 1: Showing human health risks of e-waste components

## 2.6 Standard permissible limits of heavy metals in soil

The pollution of heavy metals in soil is increasing day by day and have serious impact on human health. Keeping in view these hazardous impacts of heavy metals, the Environmental Protection Agencies of various countries have established permissible limits of heavy metals in soil. Some of which is given in Table 5.

**Table 5:** Standard permissible limits of heavy metals in soil

Heavy Metals	Rowell <sup>a</sup>	EU <sup>b</sup>	Indian <sup>c</sup>	USEPA <sup>d</sup>
Pb	50	50-300	250-300	100
Cd	0.5	1-3	3-6	3
Cr	NA	100	NA	100
Ni	25	30-75	75-150	50
Cu	20	50-140	135-270	NA
Zn	80	150-300	300-600	300
Co	NA	20-50	NA	30-100
Mn	NA	NA	NA	2000

**Explanation:** a) (Rowell, 2006) b) (CEC, 1986) c) (Awasthi, 1998; Mushtaq and Khan, 2010) d) (Ewers, 1991; Kabata-Pendias, 2010)

## 2.7 Treatment of trace elements (heavy metals)

There are several methods used for the treatment of heavy metals from aqueous solutions, which may include chemical precipitation (Brbootl et al., 2011), ion exchange (Brower et al., 1997), adsorption and membrane filtration technologies (Blöcher et al., 2003). Among all these technologies, adsorption is cost effective and simple method, thus has been extensively used by researchers. Some of the commonly used methods are discussed below;

### 2.7.1. Chemical precipitation

This is also one of the cost effective techniques for the treatment of heavy metals. In this method a chemical additive is selected in such a way that it will make the metal that is to be removed from the wastewater insoluble. After the metal precipitates in the solution then it can be easily

removed from the water by filtering or settling. However, this method is not effective enough to meet the discharge effluent standards. The major disadvantage of this process is formation of rich sludge which is difficult to handle (Kurniawan et al., 2006).

### **2.7.2. Coagulation/ Flocculation**

Flocculation is one of the famous method used for the removal of suspended particles from wastewater. It is a process in which the metal to be removed is separated out from the solution as flocs or flakes which can then be easily removed. The disadvantage of this process is higher total operation cost and slug formation, which increase the maintenance cost (Hjelmner et al., 1981).

### **2.7.3. Ion exchange method**

Ion exchange is a process in which the heavy metals are removed from the aqueous phase by the exchange of cations or anions between the exchange medium and the wastewater. Even though this is an efficient method for wastewater treatment, it is not much widely used because of its high cost. Another disadvantage of this method is the formation of resins during the ion exchange process and these resins absorb and store all the toxic chemicals which need to be disposed of. Large quantities of salt are also formed which is very difficult to handle (Clifford, 1999).

### **2.7.4. Electrolytic deposition method**

This method is mainly used for the treatment of cyanide containing wastewater. In this process the cyanide wastes are subjected to electrolysis at high temperatures (95°C) for several days. The disadvantage of this process is the treatment of cyanide and cyanate formed as residue during the process (Dean et al., 1972).

### **2.7.5. Evaporation**

Evaporation of plating wastewater has been studied to recover the plating metals. Single stage and multiple stage evaporation have been employed. This method is not commonly used due to high cost of equipment (Ahluwalia et al., 2007).

### **2.7.6. Bio-sorption**

This is a biological sorption method used for heavy metal removal from wastewater and is the cheapest method (Kim et al., 2015). Live or dead microorganisms or their derivatives are used in bio-sorption. In this method the metal ions are complexed by the functional groups present on their outer surfaces through the action of ligands. It is an effective and a cheaper method compared to the other removal techniques (Ramakrishna and Viraraghavan, 1997). However, most of these techniques have some disadvantages such as long time for the removal of heavy metals and the major disadvantage is the production of toxic chemical sludge (Amer et al., 2015).

### **2.7.7. Activated carbon adsorption**

Adsorption by using activated carbon is also widely used method for heavy metal removal from wastewater because of its high efficiency in removing metals (Bansal and Goyal, 2005). This method is not that popular in developing countries because of high initial and operating costs (Monser and Adhoum, 2002).

## **2.8. Adsorption**

Adsorption is a method whereby the solute is removed from solution by using solid granular substance. The solid substance is known as “adsorbent” and the solute removed is known as the “adsorbate”. Adsorption is one of the more effective methods for water treatment (Faust and Aly, 2013). Adsorption is the surface phenomenon of binding molecules or particles in a solution onto a surface. Atoms at the surface of adsorbent are subject to unbalanced forces of attraction which are responsible for the phenomenon of adsorption. Removal of heavy metals by adsorption method with agriculture waste is the cheapest method because it is a method of treatment of waste with waste.



## **2.8.1 Adsorption types**

Adsorption is mainly of two types:

### **2.8.1.1 Physisorption (Physical Adsorption)**

The physical adsorption process does not involve the sharing or transfer of electrons. The adsorbate is linked to the adsorbent micro-pores by weak forces, Van der Waal forces, which increase the interaction between the molecules and the substrate (Maimaiti and Elliott, 2015). Due to the weak forces, the physisorption process is mostly reversible, which means that the adsorption bonds are easily formed and broken.

### **2.8.1.2 Chemisorption (Chemical Adsorption)**

Chemisorption occurs due to interaction between adsorbate molecules and adsorbent surface. In the process of chemisorption ionic and covalent bonds are formed and a high energy of adsorption is released. These bonds are semi-permanent and undergo a chemical change, hence chemisorption is irreversible process (Teng et al., 1993).

## **2.9. Adsorbents**

The adsorbents are normally obtained from natural resources, agricultural wastes or industrial waste products. The main types of adsorbents are activated carbon (Salehin et al., 2015), synthetic polymeric (Mthombeni et al., 2015) and silica based adsorbents (Barakat and Kumar, 2015) and most of these adsorbents are not used widely because of their high cost (Wang et al., 2010). For this reason, the low cost adsorbents for removal of heavy metals are being studied widely with the aim of finding cheap natural adsorbents that are easily available.

The low cost, locally available adsorbents have been used and investigated by many researchers for metal removal. The adsorbent derived from agricultural wastes are very famous among the researcher such as Quetta pine (Rahman et al., 2015), tea waste (Amarasinghe and Williams, 2007) and coffee (Wu et al., 2015), hazelnut straws (Sayan, 2014), peanut hull (Massie et al., 2015; Zhou et al., 2015), saw dusts (Kapur and Mondal, 2015), rice husk (Galletti et al., 2015;

Sudhakar et al., 2015; Xu and Chen, 2015), corn cobs (Mahmoud, 2015), Maize cob (Okafor et al., 2015), tomato waste (Humelnicu et al., 2015), apple juice industrial wastes (Chand et al., 2015; Chand and Pakade, 2015), sugarcane bagasse (Bhagyalakshmi and Sarma, 2015), wool fibres (von Holstein et al., 2015), cassava peel (Simate and Ndlovu, 2015), Maringa pods (Matouq et al., 2015), tea waste (Zhang et al., 2015), banana (Ali and Saeed, 2015), orange peels (Sha et al., 2015), papaya seeds (Chithra et al., 2014; Yadav et al., 2014), tomato leaf powder (Gutha et al., 2015), grape stalk wastes (Liu et al., 2015) and other different cheaply available agricultural waste material (Sarma et al., 2015). In this research removal efficiency of rice husk and peanut hull was studied.

### **2.10 Mechanism of metal adsorption**

Adsorption is a famous method used for the treatment of heavy metals from solutions, especially when these are derived from lignocellulosic materials (Elham et al., 2010). The mechanisms involved in this process includes physisorption, chemisorption, ion exchange and surface adsorption (Brown et al., 2000a; Volesky, 2001).

In adsorption process, the functional groups found in the cell wall of adsorbents are responsible for metal binding. The cell walls of plants (agriculture waste) are built by cellulose molecules, lignin and pectin along with small amounts of protein (Nobel, 1991; Carrott and Carrott, 2007). The polymers include cellulose, hemicelluloses, pectin, lignin, extractives and protein and are adsorbents for a wide range of solutes, particularly divalent metal cations (Laszlo and Dintzis, 1994). The nature of lignocellulosic are hygroscopic, hence, have an affinity for water due to larger surface area. Thus, by the process of absorption and adsorption, the aqueous solution comes into contact with a very large surface area of different cell wall components.

It is reported that the materials contain cellulose are more efficient sorbent for the removal heavy metals from solution (Acemioglu and Alma, 2001) due to swelling capacity by water absorption, the higher the amount of water adsorption, the higher is the swelling, which resultant higher removal efficiency.

A research study investigated that the adsorption mechanisms of heavy metals onto the adsorbent is depend upon the metals under investigation, the degree of functionalization and the type of adsorbent (Hashem et al., 2006a). Most agriculture based adsorbents interact with metallic species through binding of the metal ion and cellulose/lignin units in the active sites (Abdel-Halim et al., 2006). This can be achieved through binding of two hydroxyl groups in the cellulose or lignin units or through binding the cellulose units together. Hydrogen bonding has also been found to be responsible for the adsorption mechanism (Hashem et al., 2006b) in some systems of heavy metal removal, and chelate formation between the adsorbent and adsorbate can immobilize a wide range of metallic species in water (Taty-Costodes et al., 2003). It has also been found that the larger the ionic valence number of the metal, the stronger the attractive forces, suggesting that, for instance,  $\text{Fe}^{3+}$  will be strongly adsorbed than a  $\text{Cd}^{2+}$ , when they are together in a multi-element aqueous medium.

### **2.10.1 Metal adsorption by rice husk**

Pakistan is major producer of rice and large amount of rice husks is produced every year as a waste of the rice milling industry which is creating environmental problems. However, In recent years modified and / or unmodified rice husk is widely used as a sorbent for the removal of heavy metals (Agrafioti et al., 2014; Lata and Samadder, 2014; Dai et al., 2015; Galletti et al., 2015; Sudhakar et al., 2015) . Structurally, rice husks consist of cellulose (35 %), hemicellulose (25 %), lignin (20 %), ash (including silica 17 %) and crude protein (3 %), hence it is very suitable for metallic cations removal (Sobhanardakani et al., 2013).

As rice husks are insoluble in water, have good chemical stability and structural strength due to high silica content (Lee et al., 1983), hence, rice husk is favourite for researcher for the treatment of heavy metal from aqueous solutions.

### **2.10.2 Metal adsorption by peanut hull**

Peanut is commercially produced in Pakistan since 1950 and peanut shell/hull is a waste product of industry as the peanut is the main ingredient of food industry. Agricultural by-products showing high adsorption capacities for metals tend to contain cellulose, hemicellulose, pectin, lignin, and protein (Kumar, 2006). Peanut hull is a good renewable resource (Brown et al., 2000b) and has been also frequently used in the removal of heavy metals. Peanut shells are composed of lignin, cellulose, hemicellulose, polysaccharides, proteins, and lipids, having functional groups such as amino, carbonyl, carboxyl and hydroxyl with characteristic chemical structures. Peanut hulls have similar components, making them a good candidate for use in wastewater treatment. The structures of these compounds have functional groups that invite metal ion exchange and metal chelation. Cellulose is a polymer of glucose units, which are composed of primary and secondary alcohols, joined by ester linkages. Lignin is a complex polymer composed of individual aromatic units. The structure is not known precisely because of lignin's complexity. There are many binding possibilities between the three phenyl propane monomers of which lignin is composed.

# METHODOLOGY

### 3.1 Study Area

This study was carried out in the following e-waste recycling sites in Pakistan;

- a) Shershah, Karachi
- b) Raja Bazar, Rawalpindi
- c) Kamoki, Gujranwala

#### 3.1.1 Shershah, Karachi

Pakistan has become a dumping yard for obsolete electrical and electronic devices, the main scrap market of e-waste is located at Shershah in Karachi. In this market computers and other electronic items are collected, transported, dismantled, crushed and burned by primitive processes without any protective measures to avoid environmental pollution. The first information regarding the biggest recycling site in Pakistan was gathered from “*Kabaria*” (auctioneers) and e-waste guide, an international portal for e-waste information (Methias, 2009).

The e-waste is being used to extract the precious metals, gold and silver, thus there is a restricted entry to e-waste processing sites for sampling and surveying. The recyclers were hesitant to disclose the information regarding collection, processing and disposal of e-waste. A random sampling technique was followed and six samples were collected from two sections of the same site. The soil/ash samples from top, middle and bottom (0 cm, 6 cm and 10 cm) were collected. The unburned plastic waste, printed circuit boards (PCBs) and stone were excluded from soil/ash samples. All of the samples were collected in polythene sampling bags and stored at ambient temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$  for further analysis.



**Figure 2:** Sampling location map of e-waste recycling sites in different cities of Pakistan

### **3.1.2 Rawalpindi**

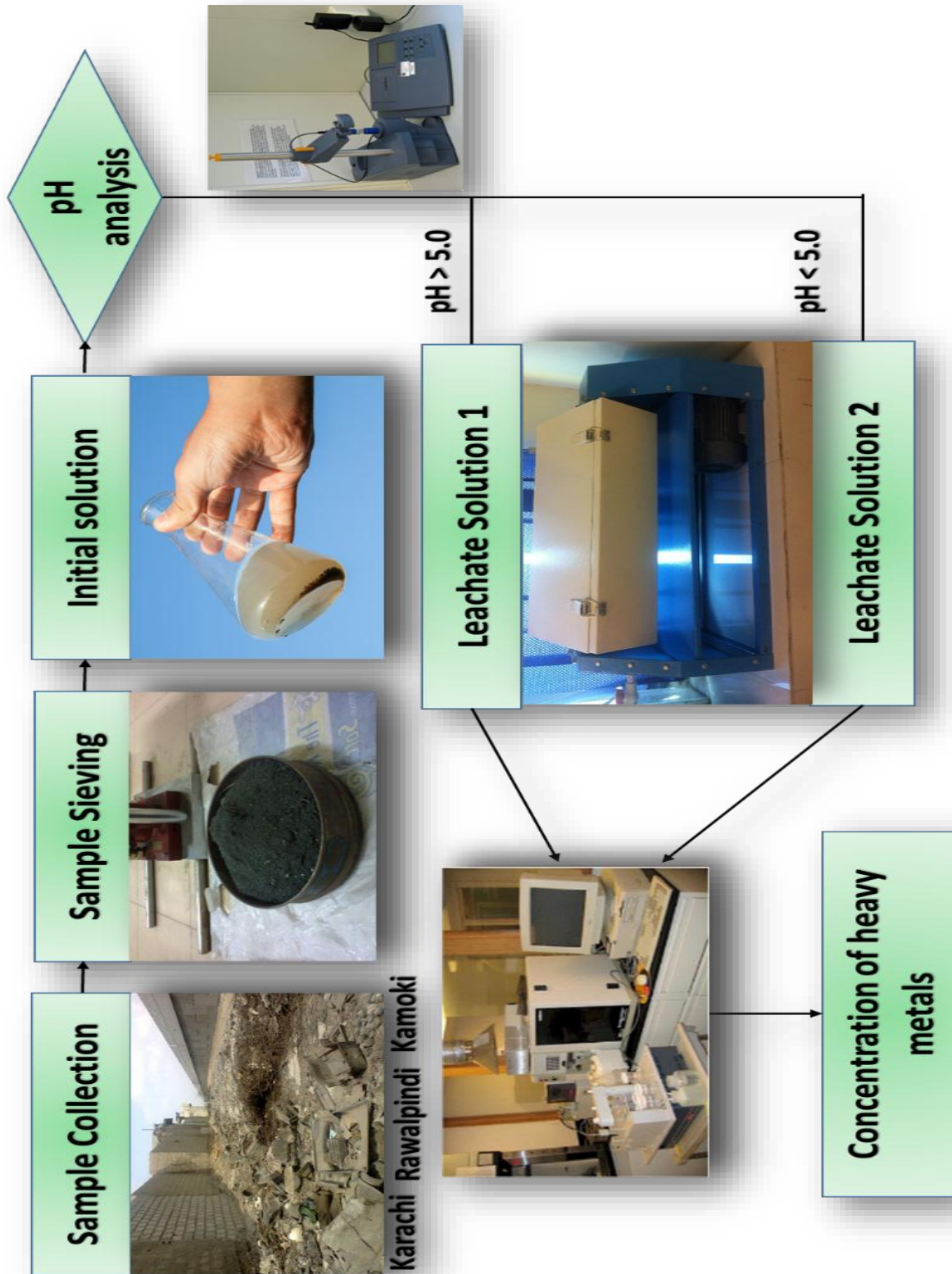
The College Road, Raja Bazar, Rawalpindi is a big market of used electronics including computers, computer accessories, mobile phone and electrical devices e.g., UPS and batteries. A large number of waste computers circuits are unsafely collected, transported, dismantled and even burned here and exported to different recyclers. A random sampling technique was followed, soil and ash samples from dismantling and burning sites were collected. The unburned plastic waste/PCBs and stone were excluded from soil/ash samples. Due to restricted access only four samples from Rawalpindi were collected and packed manually. All of the samples were put in polythene samples bags.

### **3.1.3 Kamoki**

Kamoki is a rural area located in the district of Gujranwala, near Lahore. Kamoki is also a big site, where extraction of precious metals including gold, silver and copper are processed with primitive methods. A random sampling technique was followed and samples were collected from the different sections of the same site. The soil/ash samples of top soil (0–10 cm) were collected. The unburned plastic waste/PCBs and stone were excluded from soil/ash sampling. Due to restricted access, only two samples from Kamoki were collected and packed manually. All of the samples were put in polythene sampling bags.

## **3.2 Leaching solution preparation by TCLP method**

Leachate solution was prepared by using the US EPA method “Toxicity Characteristic Leaching Procedure (TCLP)”. Two types of leachate solutions were obtained having different pH.



**Figure 3:** Process flow diagram showing the steps from samples collection, solution preparation and analysis of heavy metals by Atomic Absorption Spectrometer



### **3.3 Quantitative Analysis**

#### **3.3.1 Sample solution preparation**

The soil/ash samples were crushed and sieved with 9.5 mm standard sieve to get homogenized particle size of the sample. A solution is prepared by using Toxicity Characteristic Leaching Procedure (TCLP), a USEPA approved method for the extraction of leachate from solids, USEPA, TCLP 1311-1, (1992).

The 100 gm of the solid sample was taken in an extraction bottle. Slowly added extraction fluid in extraction vessel, tighter at rotatory agitation device and rotate at 150 r.p.m.  $\pm$  2 r.p.m. for 18 hrs  $\pm$  2 hrs at ambient temperature 25 °C  $\pm$  3 °C. Then the solution was filtered through glass fibre filter. The mixture was vigorously stirred for 5 min using magnetic stirrer and pH of each solution noted. The TCLP leaching solution 2 was applied for those samples, wherein pH was found above 5 and leaching solution 1 was applied, wherein pH found less than 5.

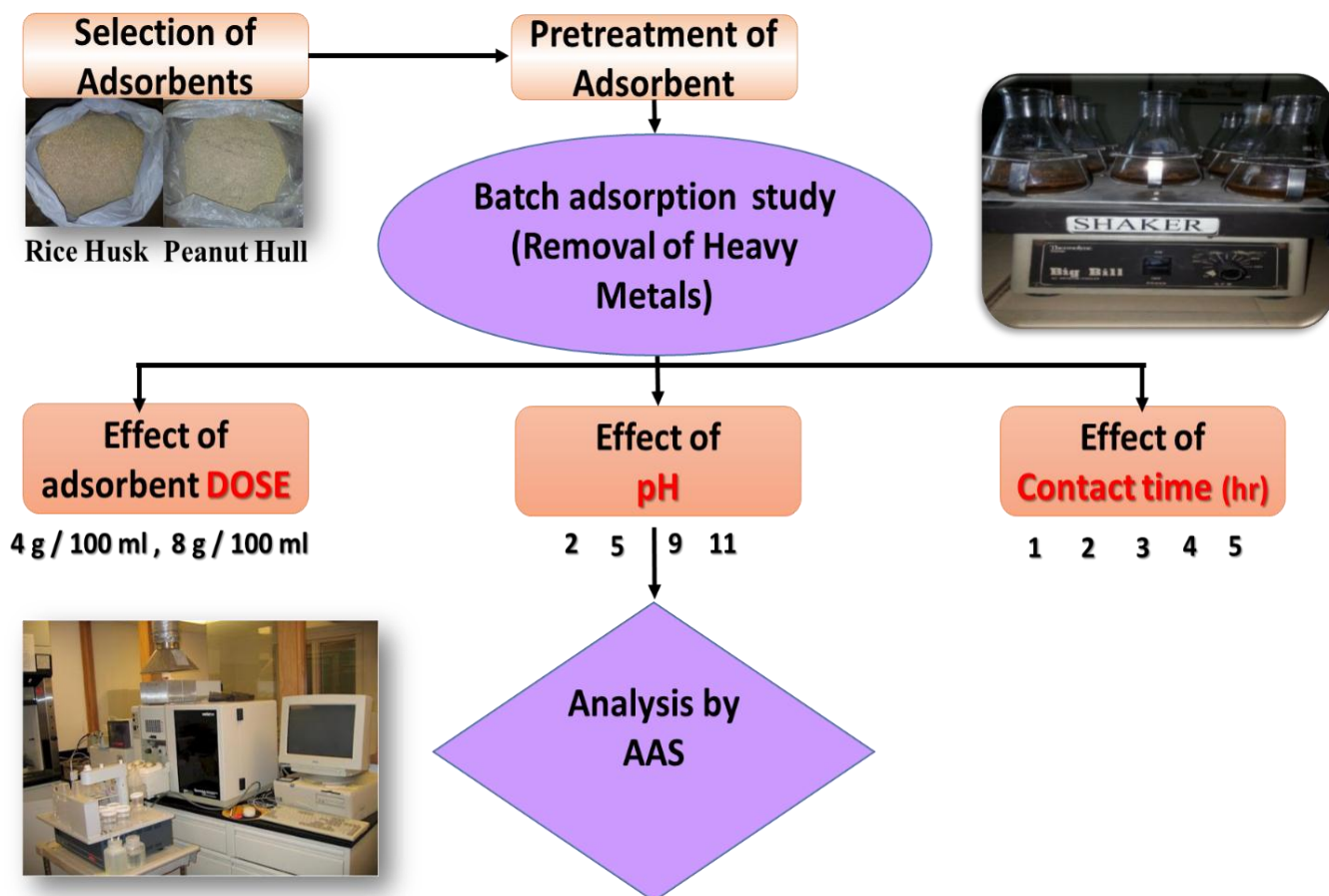
#### **3.3.2 Atomic absorption spectroscopy (AAS)**

The quantitative analysis of metals in soils/ash collected from e-waste sites were determined by flame atomic absorption spectrometer with air/acetylene burner. Each soil/ash sample was digested by using the digestion reagents as specified by USEPA TCLP method. The calibration curves for all the eight heavy metals showed the good linearity on the concentration range of heavy metals (0.1 to 20.0 mg/L) with a correlation co-efficient regression range of 0.996 to 0.999.

### **3.4 Batch adsorption studies**

There are several methods used for the removal of heavy metals from leachate solution. Most of the researchers used fixed bed column method (Khitous et al., 2015), or batch and column method (Hu et al., 2015), for the removal of toxic metals from aqueous solution (Qaiser et al., 2009) as well as wastewater (Konsowa et al., 2003). In this study batch method was used for removal of heavy metals from e-waste leachate (prepared by TCLP method) using two different

low-cost agriculture wastes i.e. Rice Husk and Peanut Hull at ambient temperature ( $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$ ). The experimental setup is given at Figure 4.



**Figure 4:** Experimental setup flow diagram showing batch adsorption process

The following experiments were performed using rice husk and peanut hull by using 100 ml leachate volume at constant temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$ , contact time and rotate at 150 r.p.m:

**Table 6:** Experiments performed at different pH, contact time and dosage of rice husk and peanut hull

Contact Time (Hrs)	Rice Husk Dose (gm)	Peanut Hull Dose (gm)	pH
1	4 8	4 8	2
2			5
3			9
4			11
5			

### **3.5 Process of adsorbent preparation**

In this study two adsorbents were selected based on the cost effectiveness and ready availability from the local markets of Rawalpindi.

#### **3.5.1 Rice husk**

The rice husk purchased from the local market in Rawalpindi. These are dried brown in colour with size of long grain rice. Rice husk were washed with fresh water, cleaned and dried in the oven at 80 °C for overnight. Then the dried rice husk was taken out crushed and sieved using a 200 mm sieve so that the fines from it were removed. The larger rice husk was put in pre-cleaned polypropylene air tight bottles. These prepared rice husk were used for the experiments.

#### **3.5.2 Peanut hull**

The peanut hull purchased from the local market, Rawalpindi and washed with fresh water, cleaned and dried in the oven at 80 °C for overnight and then sieved using a 200 mm sieve so that the fines from it were removed. The cleaned rice husk was put in pre-cleaned polypropylene air tight bottles. These prepared rice husk were used for the experiments.

### **3.6 Reagents**

All reagents were of analytical grade and deionized water was used throughout the experiments. The other chemicals including sodium hydroxide, hydrochloric acid and nitric acid were of analytical grade. 1 M HNO<sub>3</sub> and 0.5 M NaOH solutions were prepared and used for adjusting the pH of the solution. All pH measurements were carried out using a pH meter.

#### **3.6.1 Preparation of standard solutions for calibration curves**

Standard solutions of lead, chromium, cadmium, zinc, copper, nickel, cobalt and manganese were prepared from the 1000 ppm reference solutions suitable for flame atomic absorption spectrometry analysis. These standards were diluted to the concentrations ranging from 0.1 ppm to 50 ppm and stored in polypropylene bottles for use.

### 3.7 Removal efficiency

The removal efficiency of agriculture adsorbents was calculated by using the following formula;

$$\text{Removal efficiency} = \frac{C_o - C_e}{C_o} \times 100$$

Where;

$C_o$  = Initial Concentration of adsorbate (metals ion concentration)

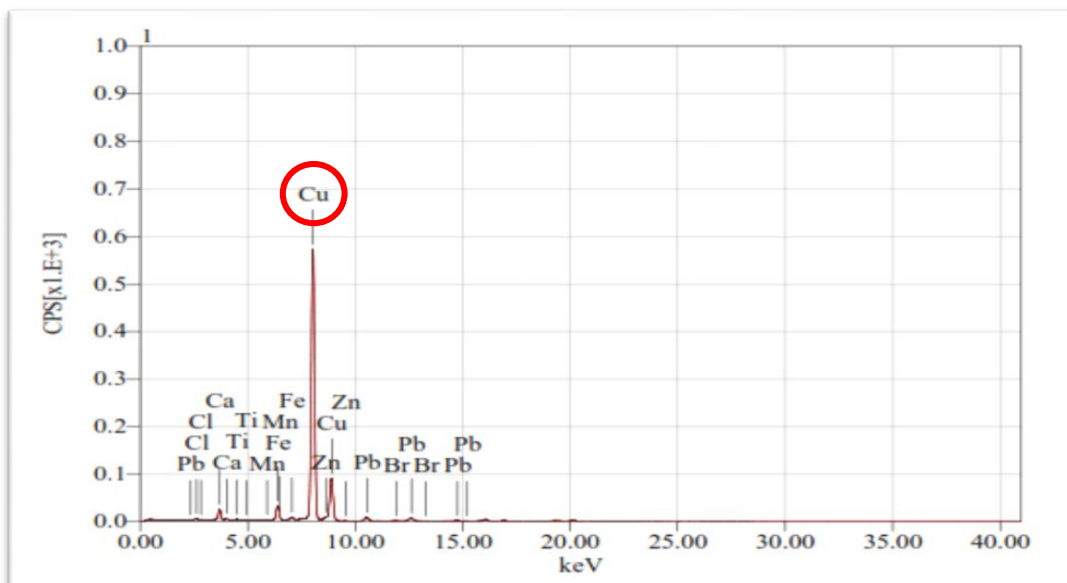
$C_e$  = Equilibrium Concentration of adsorbate (metals ion concentration)

### RESULTS AND DISCUSSIONS

The study was carried out with objectives to assess the contamination of trace elements (heavy metals) at informal e-waste recycling sites in various cities of Pakistan and their treatment with low cost agriculture waste. The assessment of trace elements of soil/ash samples was performed by using different analytical techniques including XRF analysis and Atomic Absorption Spectrometer (AAS). The treatment/removal of heavy metals were conducted by using locally available low cost agriculture waste i.e., Rice Husk and Peanut Hull.

#### 4.1 Elemental XRF Analysis

The qualitative analysis of soil/ash samples were carried out by XRF in order to analyse the presence of selected metals (Pb, Cd, Cr, Ni, Cu, Zn, Co and Mn) collected from three e-waste sites. All results of all samples showed the presence of toxic trace elements. In which the copper has higher percentage (Kamoki) as compared to other metals against wavelength and counts. XRF spectrum of one such is shown in Figure 5.



**Figure 5:** XRF Spectrum of Soil/Ash Sample

## **4.2 Heavy metals content**

A comparative study of the trace metals (heavy metals) contamination at e-waste recycling sites was conducted by analysing the soil/ash samples with AAS. The concentration of each of the 8 heavy metals are discussed below;

### **4.2.1 Concentration of Lead (Pb)**

Figure 6 (a) shows the concentration of all samples of Pb at three e-waste sites. It gives a clear view about the levels of heavy metals, wherein the concentration of Pb in Kamoki is slightly higher than Karachi and Rawalpindi. The mean concentration of Pb was found 10.7 mg/kg, 7.2 mg/kg and 24.4 mg/kg in Karachi, Rawalpindi and Kamoki respectively. The concentration of Pb from all the e-waste sites was lower than the threshold limits defined in EU, USEPA and Indian standards given in Table 5. Lead is located in computer in funnel glass of CRTs and PWBs. The CRTs are separated during the dismantling/disassembling and resale, hence, contents of Lead may be reduced due to this reason. Lead may also be removed during the extraction of precious metals from PWBs, hence, the burning site have less amount of lead and found within the limits.

### **4.2.2 Concentration of Cadmium (Cd)**

Figure 6 (b) describes the concentration of cadmium in e-waste soils/ash samples among the three cities of Pakistan. This demonstrates the level of cadmium in three different e-waste sites, wherein the concentration of cadmium in Karachi was found higher than the Rawalpindi and Kamoki. The mean concentration of cadmium in Karachi samples was 224 mg/kg, Rawalpindi 69.25 mg/kg and Kamoki 143.80 mg/kg. The concentration of Cd from all the e-waste sites was at higher level as compare to the threshold limits defined in permissible limit of EU, USEPA and Indian standards given in Table 5.

### **4.2.3 Concentration of Chromium (Cr)**

The concentration of chromium in e-waste soils/ash samples among the three cities of Pakistan is described in Figure 6 (c). This demonstrates the level of chromium in three different e-waste sites, wherein the concentration of cadmium in Karachi and Kamoki was found higher than the Rawalpindi. The mean concentration of chromium in Karachi samples was 244.2 mg/kg, Rawalpindi 100.26 mg/kg and Kamoki 282.2 mg/kg. The concentration of chromium from Karachi and Kamoki cities was at higher level as compare to the threshold limits defined in EU, USEPA and Indian standards given in Table 5, however, the chromium concentration level is slightly higher at Rawalpindi.

### **4.2.4 Concentration of Nickel (Ni)**

The concentration of nickel in e-waste soils/ash samples among the three cities of Pakistan is shown in Figure 6 (d). This demonstrates the level of nickel in three different e-waste sites, in which concentration of nickel in Karachi and Kamoki was found higher than the Rawalpindi. The mean concentration of nickel in Karachi samples was 239.5 mg/kg, Rawalpindi 72.05 mg/kg and Kamoki 314.15 mg/kg. The concentration of nickel from all the e-waste sites was at higher level as compare to the threshold limits defined in EU, USEPA and Indian standards given in Table 5.

### **4.2.5 Concentration of Copper (Cu)**

The concentration of copper in e-waste soils/ash samples among the three cities of Pakistan is shown in Figure 6 (e). This demonstrates the level of copper in three different e-waste sites, in which concentration of copper in Kamoki was found higher than the Karachi and Rawalpindi. The mean concentration of copper in Karachi samples was 193.37 mg/kg, Rawalpindi 58.13 mg/kg and Kamoki 702.85 mg/kg. The concentration of copper from all the e-waste sites was at higher level as compare to the threshold limits defined in EU, USEPA and Indian standards given

in Table 5, however, copper concentration at Rawalpindi is within the permissible limit defined in standard of EU, USEPA and Indian standards.

#### **4.2.6 Concentration of Zinc (Zn)**

The concentration of zinc in e-waste soils/ash samples among the three cities of Pakistan is shown in Figure 6 (f). This demonstrates the level of copper in three different e-waste sites, in which concentration of zinc in Karachi was found higher than the Rawalpindi and Kamoki. The mean concentration of zinc in Karachi samples was 98.30 mg/kg, Rawalpindi 54.58 mg/kg and Kamoki 59.55 mg/kg. The concentration of zinc from Karachi was at higher level than permissible limit standard (a) given in Table 5, however, from Rawalpindi and Karachi sites the concentration was found at lower level as compare to the threshold limits defined in EU, USEPA and Indian standards given in Table 5.

#### **4.2.7 Concentration of Cobalt (Co)**

The concentration of cobalt in e-waste soils/ash samples among the three cities of Pakistan is shown in Figure 6 (g). This demonstrates the level of cobalt in three different e-waste sites, in which concentration of cobalt in Karachi was found higher than the Rawalpindi and Kamoki. The mean concentration of cobalt in Karachi samples was 91.94 mg/kg, Rawalpindi 33.95 mg/kg and Kamoki 30.10 mg/kg. The concentration of cobalt from all the e-waste sites was within the range as compare to the threshold limits defined in EU, USEPA and Indian standards (d) given in Table 5.

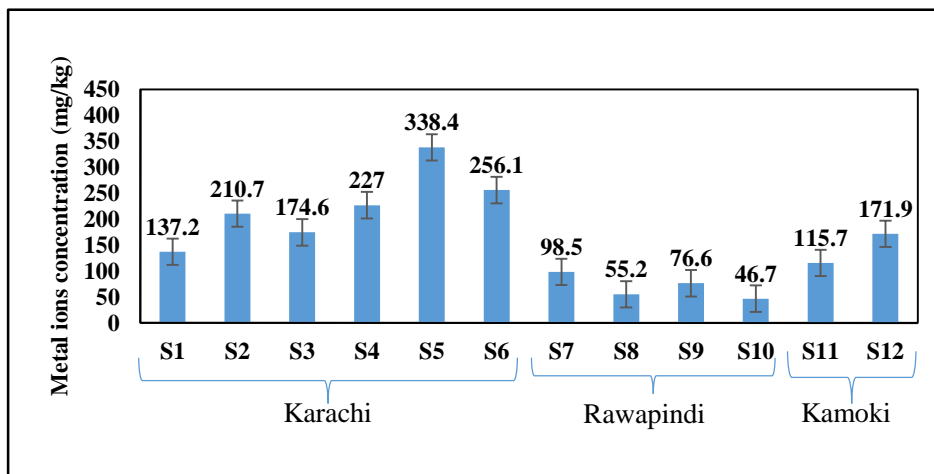
#### **4.2.8 Concentration of Manganese (Mn)**

The concentration of manganese in e-waste soils/ash samples among the three cities of Pakistan is shown in Figure 6 (h). This demonstrates the level of manganese in three different e-waste sites, in which concentration of manganese in Karachi was found higher than the Rawalpindi and Kamoki. The mean concentration of manganese in Karachi samples was 28.87 mg/kg, Rawalpindi 18.06 mg/kg and Kamoki 16.85 mg/kg. The concentration of manganese from all

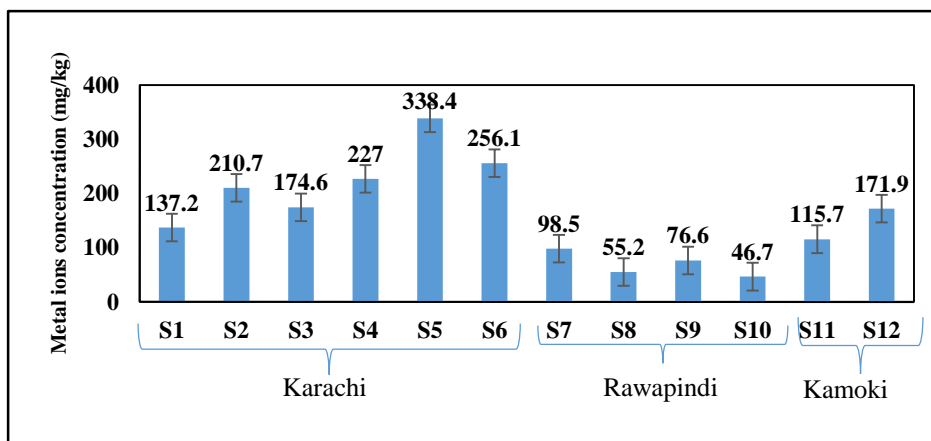


the e-waste sites was at lower level as compare to the threshold limits defined in EU, USEPA and Indian standards (d) given in Table 5.

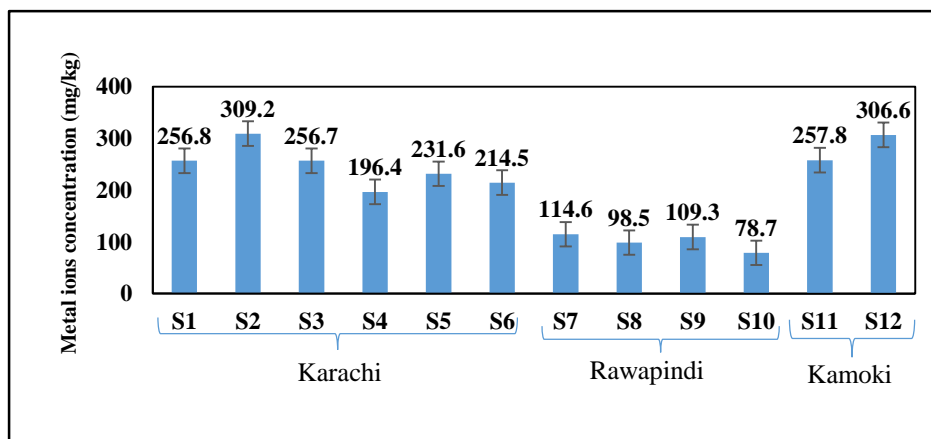
(a)



(b)

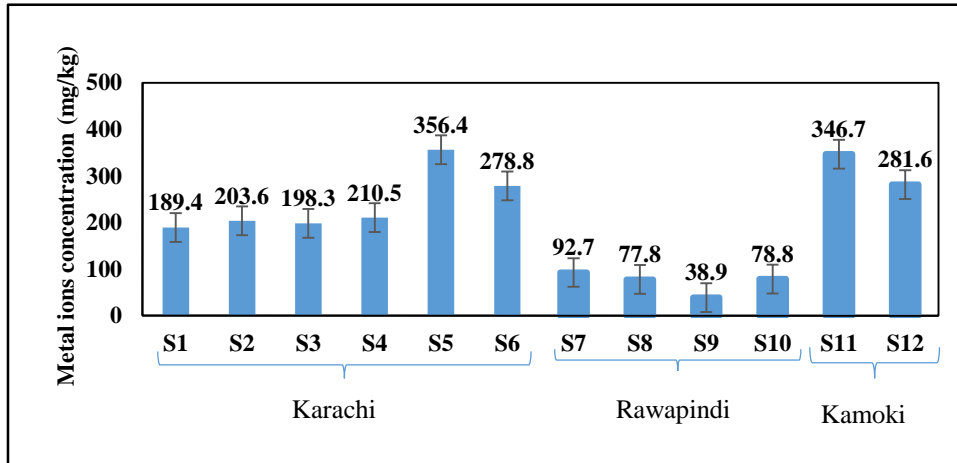


(c)

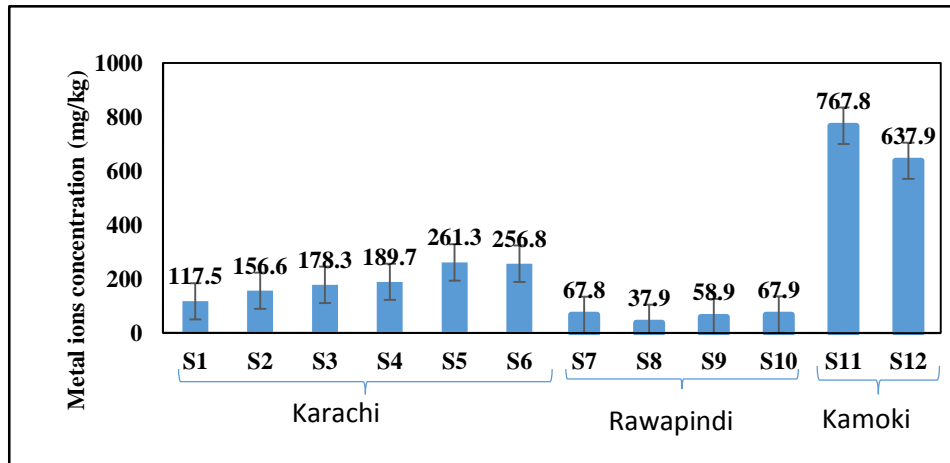


Samples sites and samples numbers

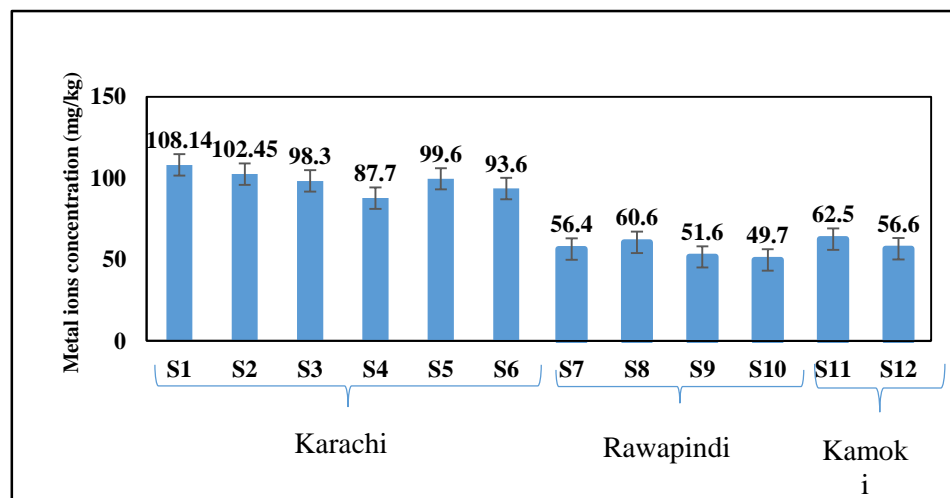
(d)



(e)

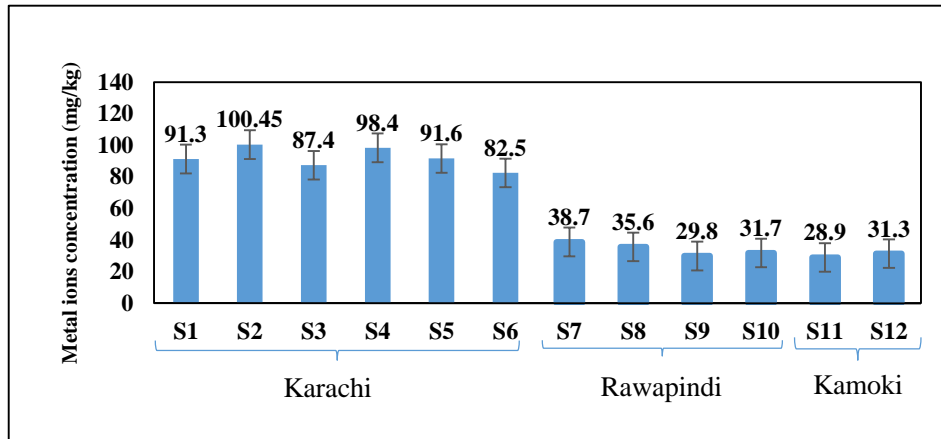


(f)

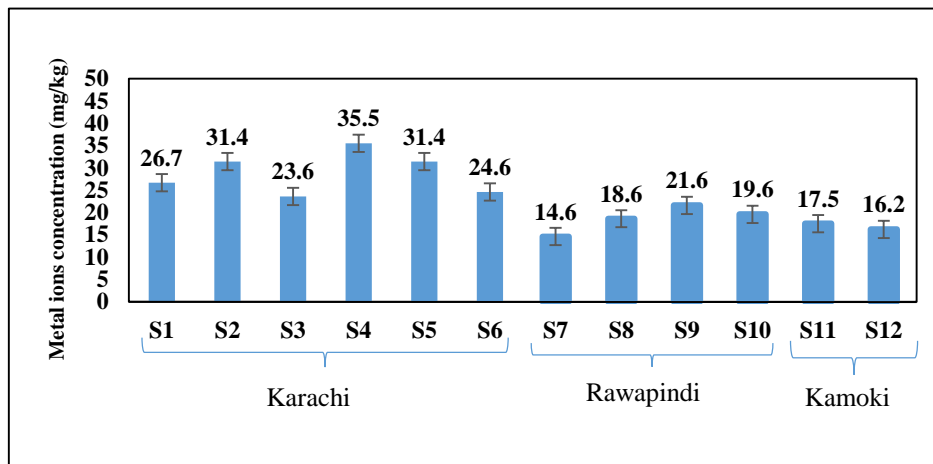


Samples sites and samples numbers

(g)



(h)



Samples sites and samples numbers

**Figure 6:** Concentration of heavy metals in various cities of Pakistan (a) concentration of Pb (b) concentration of Cd (c) concentration of Cr (d) concentration of Ni (e) concentration of Cu (f) concentration of Zn (g) concentration of Co (h) concentration of Mn

### **4.3 Batch sorption studies**

Batch sorption studies were conducted to identify the efficiency of Rice Husk and Peanut Hull at different pH, contact time and adsorbent dosage at constant temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$  and shaking at 150 rpm.

#### **4.3.1 Effect of pH**

The experiments to identify the effect of pH on the removal of heavy metals (Pb, Cd, Cr, Ni, Cu, Zn, Co and Mn) were carried out at different pH covering 2, 5, 9, and 11. The significantly better results were observed at 4 hours contact time, 8 gm /100 ml adsorbent dose, at 150 rpm and constant temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$ , also given in Table 6. The removal rate of metals was found different at changing pH. The literature also evident that adsorption of heavy metals with agricultural adsorbent depends upon the solution pH (Georgieva et al., 2015; Yadav et al., 2015).

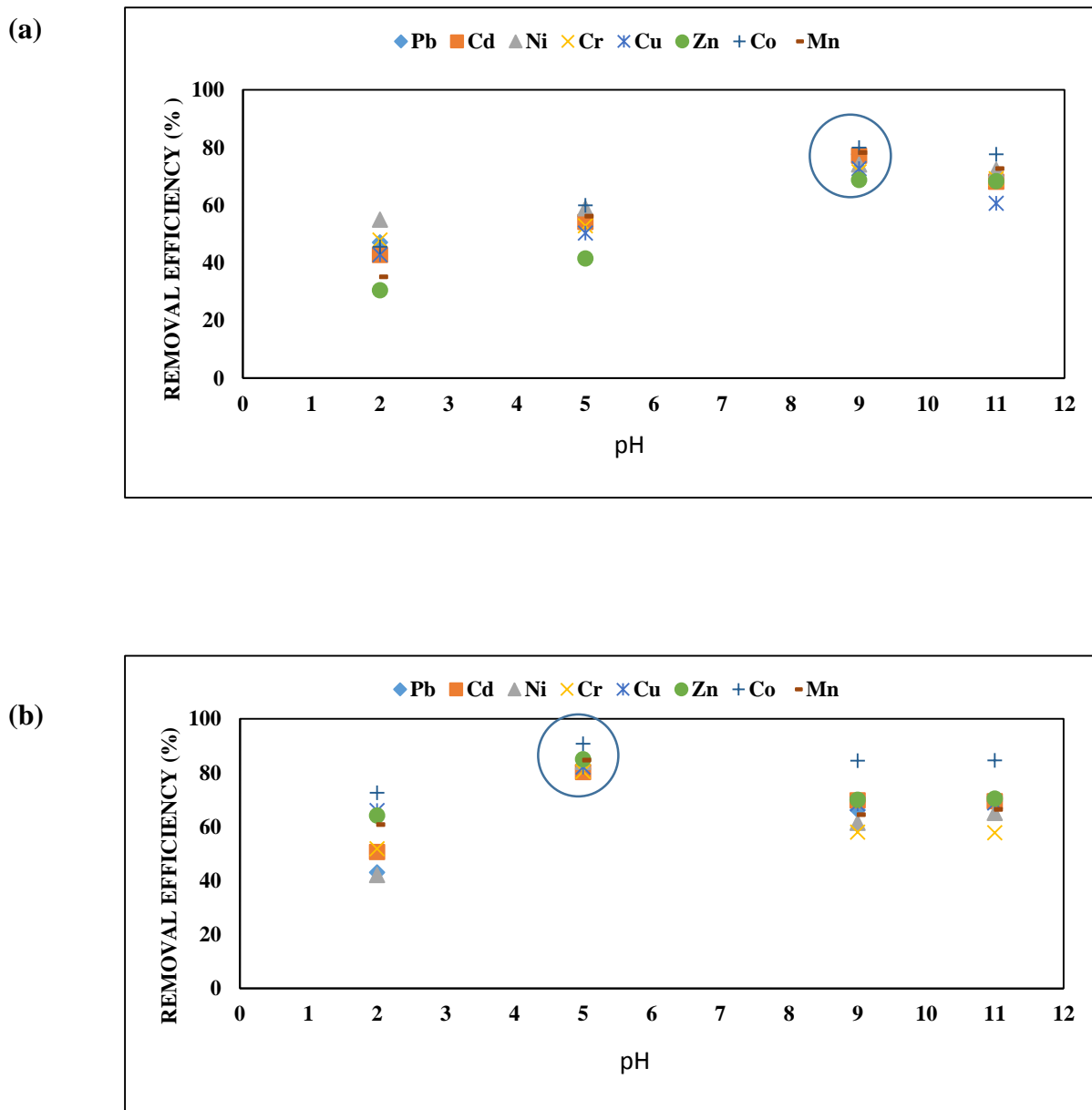
##### **4.3.1.1 Rice Husk**

The results of heavy metals removal using rice husk at different pH are presented in Figure 7 (a). The results showed that the sorption amount of heavy metals increases with the increase of solution pH and the highest increase in the adsorption rate of metals ions onto the rice husk were found at a pH 9.0. Our results are concurrent with the findings of previous studies such as Elham et al., (2010) and Galletti et al., (2015). The removal efficiency of rice husk was found between 72 % to 80 %. The removal efficiency of rice husk for Pb was 78 %, Cd 77 %, Cr 74 %, Ni 72 %, Cu 73 %, Zn 79, Co 80 % and Mn 78 %. The removal efficiency of rice husk was significantly better for cobalt among all heavy metals at pH 9.

##### **4.3.1.2 Peanut Hull**

The results of heavy metals removal using peanut hull at different pH are presented in Fig. 7 (b). Our results showed that the highest increase in the adsorption rate of metals ions on peanut husk were observed at a pH 5. The removal efficiency decrease with increase in pH, hence the optimum

pH for best removal is considered pH 5. The percent sorption of heavy metals by peanut hull reached maximum between 80 % to 91 %. Our results are similar with the studies of other previous research such as Gong et al., (2005) and Oliveira et al., (2010). The Pb showed 83 %, Cd 80 %, Cr 84 %, Ni 81 %, Cu 82 %, Zn 85 %, Co 91 % and Mn 85 %. The removal efficiency of rice husk was significantly better for cobalt among all heavy metals at pH 5.

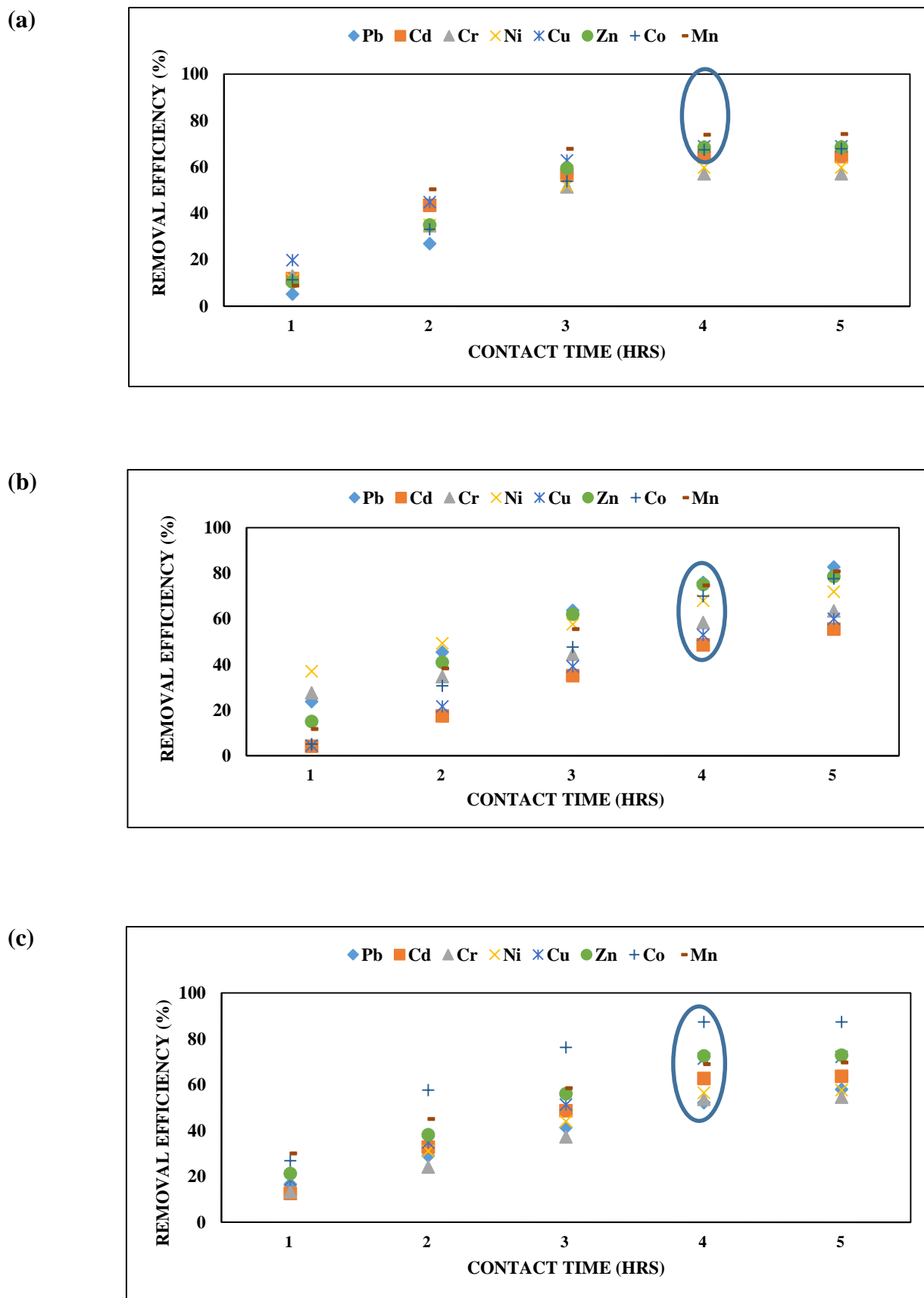


**Figure 7:** Effect of pH using different agriculture wastes at constant temperature 25 °C ± 3 °C and shaking at 150 r.p.m (a) rice husk (b) peanut hull

### 4.3.2 Effect of contact time

The experiments to identify the effect of contact time on the removal efficiency were carried out using 1, 2, 3, 4 and 5 hours as contact time under the conditions given in Table 6, with initial pH value and concentration keeping the temperature at  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$ . The adsorption is dependent on contact time (Ajmal et al., 2003) and increase with increase in contact time (Kannan and Sundaram, 2001) till attain the equilibrium. The amount of metal adsorbed by an adsorbent at a particular time is one of the factors governing the efficiency of adsorption (Imaga and Abia, 2015).

The observed removal rates of heavy metals at different concentration are presented in Figure 8. Our results showed that the sorption efficiency increased with the increase in contact time till equilibrium is attained. This is concomitant with previous studies such as Lokendra and Pradeep, (2013). The removal efficiency of rice husk observed 60 % to 74 % at 4 hours contact time with 4 gm / 100 ml dosage of the adsorbent by samples collected from Karachi given at Figure 8 (a), however, the maximum removal 56 % to 82 % observed at 4 hours contact time with 8 gm / 100 ml rice husk dosage for samples collected from Kamoki and Rawalpindi given at Figure 8 (b). The optimum agitating time for adsorption of heavy metals was observed maximum at 4 hours. The removal efficiency of peanut hull from all sites observed 52 % to 90 % at 4 hours contact time with 8 gm / 100 ml dosage of peanut hull given in Figure 8 (c). The removal efficiency peanut hull (90 %) is observed significantly better than rice husk (82 %).



**Figure 8:** Effect of pH using different agriculture wastes at constant temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$  and shaking at 150 r.p.m (a) rice husk Karachi samples (b) rice husk Rawalpindi & Kamoki (b) peanut hull all cities.

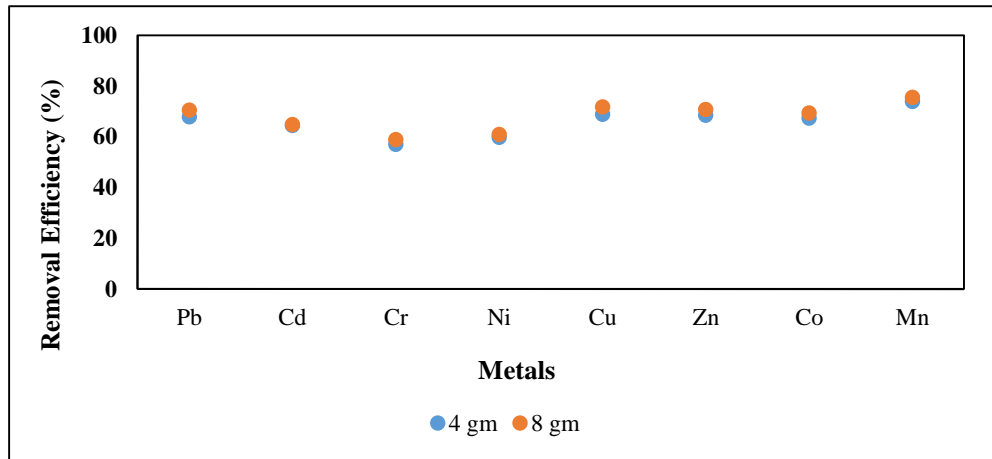
### 4.3.3 Effect of sorbent dose

The effect of variation of sorbent does on the removal of metals ions by rice husks is presented in Figure 9 (a) & (b) and peanut hull in Figure 9 (c). Two different dosages 4gm / 100 ml and 8 gm / 100 ml of sorbent were studied at different contact time 1, 2,3, 4 and 5 hours contact time at an initial metals ion concentration and pH of the leachate solution. The adsorbent dose of 4 gm / 100 ml at 4 hours contact time gives maximum removal efficiency (60 % to 74 %) for samples collected from Karachi given at Figure 9 (a). The adsorbent dose of 8 gm / 100 ml at 4 hours contact time gives maximum removal efficiency (56 % to 82 %) for samples collected from Rawalpindi and Kamoki given at Figure 9 (b). It is observed that the metal ion concentration in solution decreases with increasing sorbent amount for a given initial metal concentration. The removal efficiency of peanut hull from all sites observed 52 % to 90 % at 4 hours contact time with 8 gm / 100 ml dosage of peanut hull given in Figure 9 (c). The adsorption is proportional to specific area, which can be defined as the portion of the total area that is available for bio-sorption (Giles et al., 1960). These results showed that the removal efficiency of heavy metals increase with increasing doses of adsorbent. This is similar with previous studies such as Yalçın and Sevinc, (2000) and Bishnoi et al., (2004).

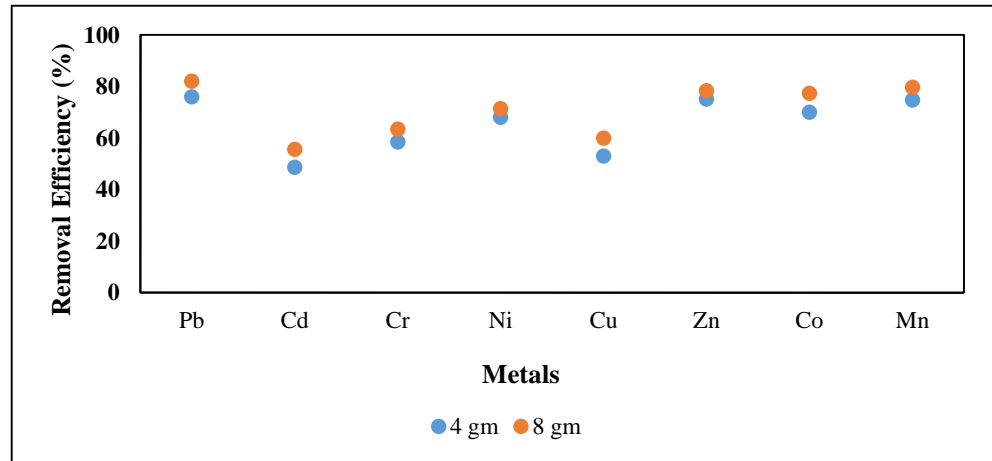
It is also observed that different metals showed different removal behaviours and metals adsorption efficiency was increased with increase in adsorbent dose, which exposed that the adsorption sites remain unsaturated during the adsorption reaction whereas the number of sites available for adsorption site increases by increasing the adsorbent dose (Desta, 2013).



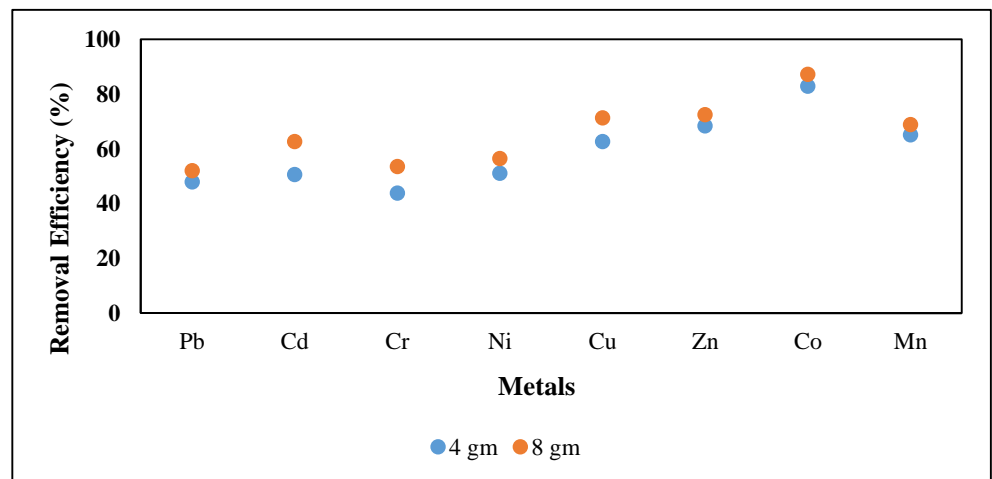
(a)



(b)



(c)



**Figure 9:** Effect of adsorbent dosage using agriculture wastes at constant temperature  $25\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$  and shaking at 150 r.p.m (a) rice husk results from Karachi samples (b) rice husk results from Rawalpindi & Kamoki samples (c) peanut hull results from all cities

### CONCLUSIONS

- a) The concentrations of trace elements, Cd, Cr, Ni and Cu were found higher than permissible limits of EU, USEPA and Indian Standard. These higher limits of metals are risk to human health due to its effect on environment, especially for the workers and people living in the vicinity of e-waste recycling sites.
- b) The adsorption is dependent on pH, contact time and adsorbent dosage.
- c) Comparing the two adsorbents the heavy metals percentage removal efficiency of peanut hull (91 %) is higher than rice husk (82 %).

### RECOMMENDATIONS

- a) Monitoring of emissions released near the vicinity of e-waste recycling and dumping sites may also be studied.
- b) There is a need to develop e-waste handling and disposal rules and guidelines in Pakistan considering its effects on environment and human health.
- c) Commercial scale studies of agriculture waste as heavy metals removal substances may be conducted based on this study.

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