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HEAVY METALS CONTAMINATION IN PADDY SOIL AND ASSOCIATED HEALTH RISK AMONG THE FARMERS IN TANJUNG KARANG, KUALA SELANGOR

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BY

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ABSTRACT

HEAVY METALS CONTAMINATION IN PADDY SOIL AND ASSOCIATED HEALTH RISK AMONG THE FARMERS IN TANJUNG KARANG, KUALA SELANGOR

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Introduction: Heavy metal contamination of soils has been a rising environmental problem affecting agricultural and threatening human health and exposed to direct contact with dermal can give adverse health effect to human. The contamination of heavy metals in agriculture field is commonly related to fertilizers as some fertilizers are known to contain certain heavy metals. **Objectives:** The objective of this study was to determine the concentration of the selected of heavy metals (Cd, Pb, Cu, As, Zn, Ni, Cr) in paddy soil and their associated health risks among the farmers in Tanjung Karang. **Methodology:** A cross-sectional study was done in Kampung Sawah Sempadan. A set of questionnaire was distributed among paddy farmers to know the gender, the average body weight (kg), exposure frequency (events/year), exposure duration (years), and averaging time (days) to calculate the health risk of the farmers for hazard quotient (HQ) and dermal cancer risk (DCR). 72 soil samples were collected from 24 different paddy plots from A to X and extracted using Aqua regia procedure. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) PerkinElmer brand was used for analyzing the samples. **Result and Discussion:** Result shows that the mean concentrations of heavy metals of As, Cr, Zn, Pb, Cu, Ni, and Cd were 17.13 mg/kg, 14.80 mg/kg, 7.21 mg/kg, 6.64 mg/kg, 3.37 mg/kg, 1.82 mg/kg and 0.06 mg/kg respectively in study area. Based on the health risk assessment on paddy farmers, the hazard quotient (HQ) for all selected heavy metals did not exceed 1 and dermal cancer risk (DCR) calculated were ranged from 3.0×10^{-4} to 1.3×10^{-4}. The mean concentration of all element selected were below than the recommended standard except for As which exceed the standard from Dutch Target value for mean concentration and exceed the Kabata-Pendias & Pendias for maximum concentration. **Conclusion:** The concentrations of selected heavy metals were below the value recommended by soil standards except for As which has exceed the Dutch Target Value and Kabata-Pendias & Pendias standard and also the hazard quotient (HQ) and dermal cancer risk (DCR) show that there were no significant carcinogenic and non-carcinogenic health risks of farmers due to dermal exposure. Some of the elements studied do not have cancer slope factor and reference dose, so their respective DCR and HQ cannot be determined.

Keywords: Heavy metals, paddy soil, Aqua regia, risk assessment, dermal

ABSTRAK

PENCEMARAN LOGAM BERAT DALAM TANAH PADI DAN RISIKO KESIHATAN YANG BERKAITAN DALAM KALANGAN PETANI DI TANJUNG KARANG, KUALA SELANGOR

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Pengenalan: Pencemaran logam berat meningkat dalam tanah padi telah menjadi masalah persekitaran yang mempengaruhi kesihatan manusia pertanian dan ancaman juga mendedahkan kepada hubungan langsung dengan dermis yang boleh memberi kesan kesihatan yang buruk kepada manusia. **Objektif:** Objektif kajian ini adalah untuk menentukan kepekatan yang dipilih logam berat (Cd, Pb, Cu, As, Zn, Ni, Cr) dalam tanah padi dan risiko kesihatan yang berkaitan di kalangan petani di Tanjung Karang. **Metodologi:** Satu kajian keratan rentas telah dijalankan di Kampung Sawah Sempadan. Satu set soal selidik telah diedarkan di kalangan petani padi untuk mengetahui jantina, berat badan purata (kg), kekerapan pendedahan (peristiwa / tahun), tempoh pendedahan (tahun), dan masa purata (hari) untuk mengira risiko kesihatan petani untuk bahaya quotient (HQ) dan risiko kanser kulit (DCR). 72 sampel tanah telah diambil daripada 24 petak-petak sawah yang berbeza dari A ke X dan diekstrak menggunakan prosedur regia Aqua. Pasangan Plasma Induktif Mass spektrometri (ICP-MS) berjenama PerkinElmer telah digunakan untuk menganalisis sampel. **Keputusan dan Perbincangan:** Hasil kajian mendapati kandungan purata logam berat As, Cr, Zn, Pb, Cu, Ni dan Cd adalah 17.13 mg / kg, 14.80 mg / kg, 7.21 mg / kg, 6.64 mg / kg, 3.37 mg / kg, 1.82 mg / kg dan 0.06 mg / kg masing-masing di kawasan kajian. Berdasarkan penilaian risiko kesihatan ke atas petani padi, hasil bahagi bahaya (HQ) untuk semua logam berat dipilih tidak melebihi 1 dan risiko kanser kulit (DCR) yang dikira telah berkisar dari 3.0×10^{-4} kepada 1.3 × 10-4. Kepekatan min semua elemen yang dipilih adalah di bawah daripada piawaian yang dicadangkan kecuali Seperti yang melebihi standard daripada nilai sasaran Belanda untuk kepekatan dan melebihi Kabata-Pendias & Pendias untuk kepekatan maksimum. **Kesimpulan:** Kepekatan logam berat yang dipilih adalah di bawah nilai yang disyorkan oleh piawaian tanah kecuali As yang mempunyai melebihi standard Belanda Nilai Sasaran dan Kabata-Pendias & Pendias dan juga hasil bahagi bahaya (HQ) dan risiko kanser kulit (DCR) menunjukkan bahawa tiada risiko kesihatan karsinogenik dan bukan karsinogen yang ketara petani disebabkan oleh pendedahan kulit. Beberapa elemen yang dikaji tidak mempunyai faktor cerun kanser dan dos rujukan, jadi DCR dan HQ masing-masing tidak dapat ditentukan.

Kata kunci: Logam berat, tanah padi, Aqua regia, penilaian risiko, dermal

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LIST OF ABBREVIATIONS

AAS	Atomic Absorption Spectrometry
ABS_d	Dermal absorption fraction
AF	Adherence factor of soil to skin (mg/cm^2-event)
As	Arsenic
AT	Averaging Time (days)
BW	Body weight (kg)
Cd	Cadmium
CF	Conversion factor (10^{-6} kg/mg)
Cr	Chromium
C_{soil}	Chemical concentration in soil (mg/kg)
Cu	Copper
DAD	Dermal Absorbed Dose (mg/kg-day)
DAevent	Absorbed Dose per event (mg/cm^2-event)
DCR	Dermal Cancer Risk
ED	Exposure duration (years)
EF	Exposure frequency (days/ year)
EV	Event frequency (events/ day)
F-AAS	Flame Atomic Absorption Spectrophotometer
GFAAS	Graphite Furnace Atomic Absorption Spectrometry
H_2SO_4	Sulphuric Acid

HCl	Hydrochloric Acid
$HClO_4$	Perchloric Acid
HNO_3	Nitric Acid
HQ	Hazard Quotient
ICMM	International Council on Mining & Metals
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICP-OES	Inductively Coupled Plasma Optical Emission Spectrometry
N/A	Not Available
NH_4CH_3OO	Ammonium Acetate
Ni	Nickel
N-P-K	Nitrogen- Phosphorus- Potassium
N/A	Not Available
Pb	Lead
PPE	Personal Protective Equipment
RfD	Reference Dose
R^2	Linear regression coefficient
SA	Skin surface area available for contact (cm^2)
SF	Absorbed cancer slope factor (mg/kg-day)
USEPA	United States Environmental Protection Agency
WDM	Wet Digestion Method
Zn	Zinc

CHAPTER 1

INTRODUCTION

1.1 Research Background

Agriculture in Malaysia was the key development and become familiar mostly around the Malay communities over several generations in the past. Paddy is the most familiar in the agriculture because of their high demands and important crops in Malaysia after oil palm (Khairiah et al., 2013). Most essential crops are paddy was highly sowed in Peninsular Malaysia such in Kedah, Selangor, Perak, Negeri Sembilan and also in the East of Malaysia, in Sabah and Sarawak. Every year, an estimated amount of 2.5 million tons of the agrochemicals are used in agriculture worldwide and Malaysia produced approximately 2,665,098 tonnes comprising 2,311,978, 145,194 and 207,925 tonnes from peninsular Malaysia, Sabah and Sarawak respectively (Khairiah et al., 2013).

Over the decades, paddy field has becomes more attractive area for the research project about the pollution involved in the soil and water. Surged of improved technologies and high demands which encourage more people to explore the sector and persuade farmers to develop more real and this is because of this sector involved used of the agrochemicals such as pesticides and fertilizers that contribute to the formation of heavy metals (Luo & Han, 2001). Generally, both natural such as weathering, erosion of parent rocks, atmospheric deposition and volcanic activities and anthropogenic for example sewage irrigation, addition of manures, fertilizers and pesticides which was the activities that were responsible sources of soil and crops contamination with heavy metals (Khan et al., 2013).

Based on Liu, Zhang, & Tran, (2011) the assessment of total trace elemental concentrations in agricultural soils is required to evaluate the potential risk of paddy soils due to contamination of toxic heavy metals such as Cd and Pb. Lu, Wang, Teng, & Yu, (2015) was reported that the phosphate fertilizers generally the major source of trace metals among all inorganic fertilizers, and much attention had also been paid to the concentration of Cd in phosphate fertilizers. Agriculture use of pesticides and fertilizers was another source of heavy metals in soils from non-point source contamination. In other hand, some of the farmers were practiced without proper personal protective equipment such as glove and boot when in contact with the soil and water at paddy field.

In Malaysia, the phosphatic fertilizers have been used for long time or period in some regions (Luo & Han, 2001). Zarcinas et al., (2004) indicated that these fertilizers contain many impurities that can contaminate soils. Therefore, it is important to assess the impact of the heavy metals and the concentration of heavy metals in soil and in this study the main focused area in paddy field area. The increase the use of the fertilizers for paddy crop may affect the contaminant in paddy soil. In high amount concentration of heavy metals, it can cause harmful to the human's health and give adverse effects. Not all heavy metals are toxic to human because in a small quantity, heavy metals such as Cu, Zn, Fe and Mn are essential for human and good for health. Heavy metals such as Pb also good for industrial ingredients however, most of the heavy metals become toxic when do not get metabolized by the body and end up accumulating in soft tissues.

Previous study reported from Efremova & Izosimova (2000) that the use of fertilizers significantly increase Cd in soil. As a result from human activity such as use fertilizers, pesticide and other agrochemicals increases of Cd levels that used on agriculture fields (Micó, Peris, Sánchez, & Recatalá, 2006). There are active ingredients for the fertilizers are N-P-K which represents nitrogen, phosphorus and potassium (Luo & Han, 2001). The nitrogen will help the plant for greener foliage and an overall healthy plant. Phosphorus stimulates the development and growth of roots and the development of seeds and blossoming of the flower and for potassium, it will improve the plant's immune system while improving the plant in building protein and completing photosynthesis. The functions of the active ingredient in the fertilizers mostly give the good effect to the plant. But, on the other hand it can give adverse effect to the environment and adverse effect to human in contact when use it repeatedly to soil and plant and heavy metals can accumulate and end up in soil.

1.2 Problem statement

Most of heavy metals exist at contamination such as in sewage, mining, agricultural, naturally occur in environment such as soil, water, and air and others places (Luo & Han, 2001). Generally, in agriculture such as in paddy field, anthropogenic activities such as fertilizers and pesticide was applied on the crops where it potentially the sources of heavy metals parameters relating to soil and plant (Khairiah et al., 2013). According to Aziz, Rahim, Sahid, Mohd, & Idris, (2015), heavy metal contamination of soils has been a rising environmental problem affecting agricultural and threatening human health.

As the anthropogenic activities can increase the heavy metals level in soil, Khairiah et al., (2013) reported that heavy metals can accumulate in paddy soil and plant due to utilization of urea and fertilizers for paddy fields as it used over decades in Malaysia and it's become a rising concern. Furthermore, according to Zarcinas et al., (2004) majority agricultural soil in Malaysia are heavily fertilized by phosphatic fertilizer and contain many impurities such as Cd, Zn, Cu, and As that can contaminate the soil (Zarcinas et al., 2004).

There are many sources of heavy metals contaminant in the soil that difficult to differentiate because of the naturally occur of heavy metals in soil and man-made activities. Zarcinas et al., (2004) reported that there were lack of the consistent investigation and monitored into contaminants in agricultural soils in Malaysia and make it difficult to identify the source of heavy metals. The use of the personal protective equipment such as glove and boot among farmers can give big effect to because it can lead to the health risk to human when exposed directly to the soil without any protection to the dermal and skin. From that problem, this study wants to know the use of the personal protective equipment can cause the health risk effect on dermal to human or not.

1.3 Study Justification

The heavy metals in our environmental can exist both naturally and come from the accumulation in pollution and according to (Lu et al., 2015), heavy metals that have been identified in the polluted environment include As, Cu, Cd, Pb, Cr, Ni, Hg and Zn. A number of previous studies had reported the heavy metals contamination in paddy or agricultural in Malaysia such as in Langkawi, Kedah and Tumpat, Terengganu. To the best of author's knowledge, there is no study on the heavy metals contamination in paddy soil in Kampung Sawah Sempadan, Tanjung Karang.

Previous study on heavy metals in soils and crops in Southeast Asia was conducted by Zarcinas et al., (2004) was to evaluate the normal ranges of heavy metals in agricultural soils of Peninsular Malaysia. Generally, the study found that the maximum value of heavy metals in soil were Zn> Pb> Cr> As> Cu> Ni> Co> Hg> Cd. Another study conducted by Aziz et al., (2015) found that the concentrations of some heavy metals such as Cr, and Ni in Ranau Valley, Sabah were exceed maximum allowable concentration in soil (Kabata-Pendias and Pendias) which is Guidelines values for agricultural soils with a pH > 7.

The effect use of pesticide and fertilizer in the agriculture sector become vigorous among the paddy farmers and unidentified can cause the contaminant to the environment such as in soil or water and will give the adverse effect to human health when exposed directly to the skin. The farmers are likely exposed to the soil that contaminant by the heavy metals when the accumulation of the chemical in pesticide and fertilizers can increase the toxicity of the heavy metals in soil for a long-term. In this issues, when the heavy metals was accumulated in soil, the farmers will exposed with dermal or skin contact through paddy soil and paddy water.

Since there are farmers in Tanjung Karang that do not wear appropriate and full PPE while working with paddy soil, they are exposed to direct contact to skin with heavy metals that contain in paddy soil. In addition, it also important to assess the health risk that is imposed on the farmers by the exposure. This study is conduct to investigate and create a better understanding on the concentration of heavy metals in paddy soil and to determine the dermal health risk associated to the farmers with the respective selected heavy metals.

1.4 Research questions

The research questions are as follows:

i. What is the concentration of heavy metals (Cu, Cr, Zn, Ni, Pb, As and Cd) in soil samples collected from Tanjung Karang paddy field?

ii. Is there any health risk due to occupational exposure of heavy metals in paddy soil among the farmers in Tanjung Karang?

iii. Is the concentration of the heavy metals in paddy soil were exceed the standard of Netherland - Dutch target value, Trace Elements in Soils and Plants (Kabata-Pendias & Pendias), and Australian Ecological Investigation Levels (EILs)?

1.5 Objectives

To determine the concentration of the selected of heavy metals (Cd, Pb, Cu, As, Zn, Ni, Cr) in paddy soil and their associated dermal health risks among the paddy farmers in Tanjung Karang, Kuala Selangor.

The specific objectives are as follows:

i. To determine the concentration of the selected heavy metals (Cd, Pb, Cu, As, Zn, Ni, Cr) in paddy soil collected from Kampung Sawah Sempadan, Tanjung Karang.

ii. To assess the health risk of the paddy farmers in Tanjung Karang due to occupational exposure of heavy metals in paddy soil.

iii. To compare the concentration of the heavy metals in soil with the standard of Netherlands - Dutch target value, Trace Elements in Soils and Plants (Kabata-Pendias & Pendias), and Australian Ecological Investigation Levels (EILs).

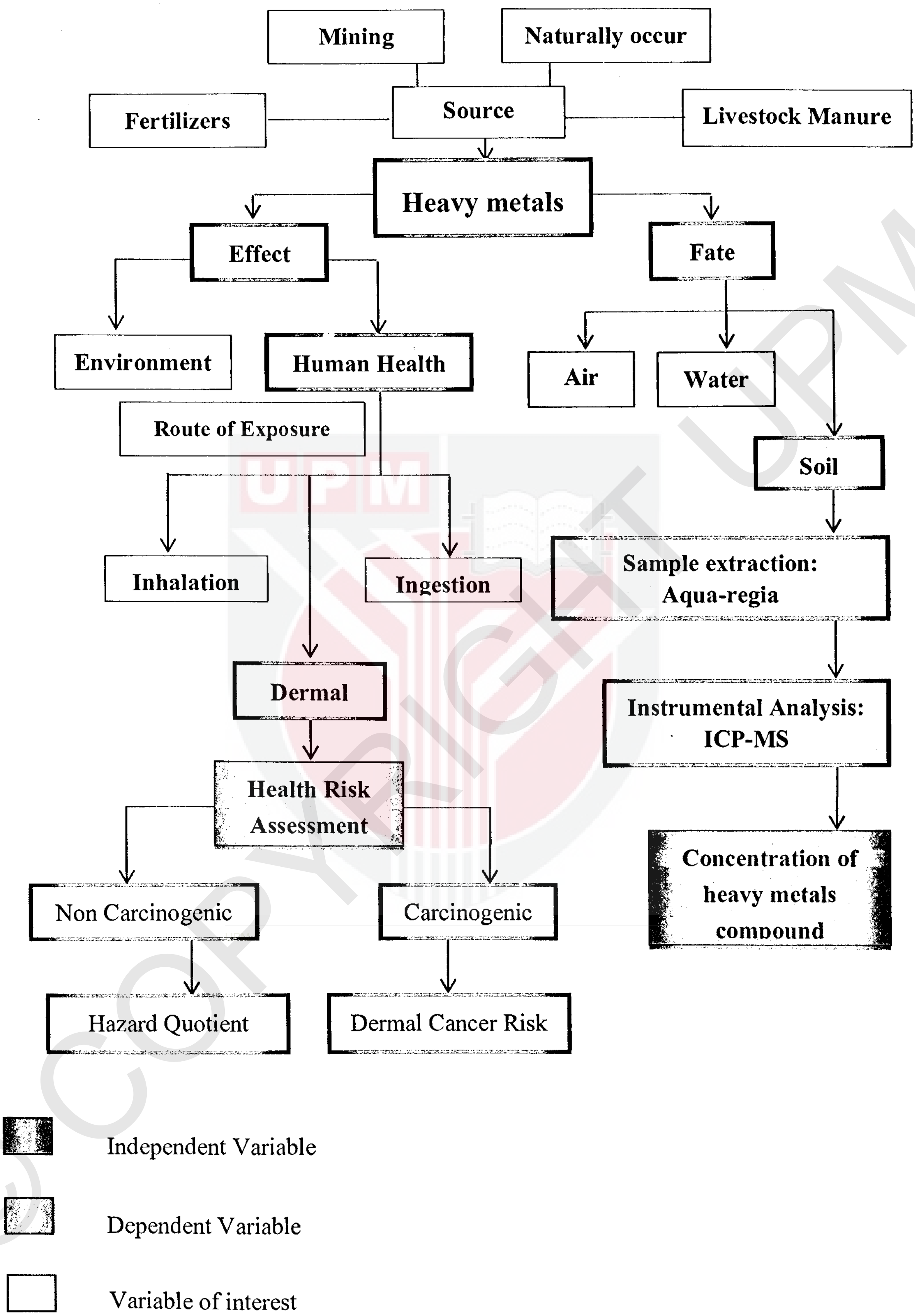

Figure 1.1: The Conceptual framework

CHAPTER 2

LITERATURE REVIEW

2.1 Heavy Metal in Soil and Agricultural Fields

Many population are depends on the paddy as staple food in developing country like Malaysia for survive and growth. To increase the production, many activities have been identified and could potentially cause to the heavy metals contaminant in paddy field soils such as the use of fertilizers and pesticide. One of the major threats to paddy and aquatic organisms are fertilizers (Sow, Ismail, & Zulkifli, 2013). Contaminants can enter the food chain from industrial, urban and agricultural sources (Zou B, P Zhuang, Li N Y & Li, 2009). The potential effects on human health and the possible long-term sustainability of food production in contaminated areas due to the contamination of agricultural soils and crops by heavy metals is causing concerns (Luo & Han, 2001).

In Malaysia, the traditional way of growing the paddy rice that is being used nowadays by farmers involves soil puddling, followed by rice transplantation, and growing the crop in a submerged environment from crop establishment to close to harvest (Gong, 1983; Sahrawat, 2005). By applying the pesticides and fertilizers regularly, paddy field soils are likely to accumulate the pollutant, since heavy metals in the soil are bound to the various phase components of the soil like carbonates, clay minerals, sulfides, and organic or biological substances (Yap, Adezrian, Khairiah, & Ismail, 2009). Mining process and industries also contribute to heavy metals contaminations in soil (Luo & Han, 2001).

Soil contains many heavy metals that are naturally bound to various phase components of the soil, by administering pesticides and fertilizers extensively; these contaminants and pollutants will accumulate (Khairiah et al., 2013). Due to their non-biodegradable and persistent nature, heavy metals are generally accumulated in the soil or sediment, and thus, pose potential risks to the ecosystem. More importantly, they can enter the food chain and subsequently threaten human health (Micó et al., 2006)

There are non-carcinogenic and carcinogenic can be classify in heavy metals. Therefore, the source of the heavy metals in soil can give adverse health effect to human health. The contaminated paddy soil could exert long-term risks to ecosystem and also to human health through the increased uptake and accumulation of heavy metals in paddy soil (Efremova & Izosimova, 2000)

Previous study reported that cadmium is a highly toxic heavy metal causing cancer and has been polluting the paddy fields due to indiscriminate application of inorganic fertilizers, and being persistent, it enters into food chains causing potential threat to human beings (Reddy, Satpathy, & Dhiviya, 2013). According to Reddy et al., (2013), accumulation of excessive Cd in soils cannot only result in poor growth of the crop plants but also reduce the crop productivity and affect human beings.

2.2 Heavy Metals and Health Effects

Contamination of heavy metal can affect the biological systems in harmful ways since it does not undergo biodegradation even in low concentration. According to WHO, (2014) contact dermatitis is the most important skin disease caused by direct contact with external irritants and/or allergens. 10% of all occupational diseases in Europe and the USA reported that occupational skin disease represents for workers in occupations as well as direct effects such as irritation, urticarial, acne, cancers and phototoxicity which also presented.

2.2.1 Cadmium, Cd

Metals like Cr, Cd and Ni can be very toxic and with their high accumulation inside living bodies over time can cause serious harmful diseases (Khan et al., 2013).

Cd known as human carcinogens can cause severe damage to the lungs and may occur through breathing high levels of Cd. Cd is a highly toxic heavy metal causing cancer and has been polluting the paddy fields due to indiscriminate application of inorganic fertilizers, and being persistent, it enters into food chains causing potential threat to human beings (Reddy et al., 2013).

2.2.2 Lead, Pb

According to United States Environmental Protection Agency, USEPA Pb is a naturally occurring element found in small amounts in crust of the earths. In other hand, it also can be toxic to humans and animals causing of health effects. In addition, EPA states that all parts of our environment such as air, soil and water, Pb also can be found but much of the exposure comes from human activities.

Today, the most common sources of Pb exposure in the United States are lead-based paint and possibly water pipes in older homes, contaminated soil, household dust, drinking water, Pb crystal, Pb in certain cosmetics and toys, and lead-glazed pottery (Järup, 2003). EPA has determined Pb in human carcinogen and can affect every organ and system in the body for long-term exposure of adults can result functions of the nervous system; weakness in fingers, wrists, or ankles; small increases in blood pressure and anemia.

2.2.3 Arsenic, As

As is odorless and tasteless where can be found naturally in the environment in larger quantities through the releasing of volcanic activity, erosion of rocks, forest fires, and human activity. Human activity also can include farmers that use fertilizers in paddy fields (Liu et al., 2011). Long-term exposure to high level of inorganic As exhibit several symptoms include pigmentation, skin lesions and hyperkeratosis.

2.2.4 Chromium, Cr

Cr is very persistent in sediments in water but the Cr compounds bind to soil and are not likely to migrate to ground water (Järup, 2003). Cr are mostly found in rocks, animals, soil, plants, and can be a liquid, solid or gas. Usually, Cr was used in metal alloys such as stainless steel, magnetic tapes, cement, paper, rubber, composition floor covering and other materials. Cr (VI) compounds are toxins and known human carcinogens, whereas Cr (III) is an essentials nutrient. The effect of the Cr such as skin contact can cause skin ulcers and allergic reactions consisting of severe redness and swelling of the skin have been noted (Järup, 2003).

2.2.5 Nickel, Ni

Ni is the one from many carcinogenic metals known to be an environmental and the occupational pollutant. According to (Nazir, Khan, Masab, Rehman, & Rauf, 2015), Ni has been considered to be an essential trace element for human and animal health. However, the exposure to the Ni introduces to free radicals which can lead to oxidative damage and may also affect the kidneys and liver. Ni can occur naturally in the environment at low levels. Metals like Cr, Cd and Ni can be very toxic and their high accumulation inside living bodies over time can cause serious harmful disease (Khan et al., 2013).

The data indicate that absorption of Ni following dermal contact with various Ni compounds can take place, but to limited extent, and with a large part of the applied dose remaining on the skin surface or in stratum corneum (ICMM, 2007).

2.2.6 Zinc, Zn

Zin is the relatively harmless compared to several other metal ions with similar chemical properties. Exposure to high doses has toxic effects making acute Zn intoxication a rare event. According to Khan et al., (2015) the higher concentration of Zn can be toxic to the organism. However, according to (Khan et al., 2013), the sufficient amount of Zn is very important for normal body functions and the deficiency of the Zn can cause anorexia, diarrhea, dermatitis and depression, immune dysfunction and poor wound healing.

According to ICMM, (2007) based on the cumulative amount recovered from the receptor fluids at 72 hours, penetration of Zn was below 1 %. However, it may become available at a later stage when the amount retained in the skin should regard as being absorbed. Unfortunately, since wounded skin was investigated, or suction blisters were raised, impairing the intactness of the skin, the human data presented are not considered valid (ICMM, 2007).

2.2.7 Copper, Cu

Exposure to Cu can be from air, water, food, and by skin contact. Cu is a very common substance that occurs naturally in the environment and spreads through the environment via natural phenomena and it also applied in the industries and in agriculture. Cu toxicity is a fundamental cause of Wilson's disease (Samuel et al. 2011). High dose of the Cu can cause the mental diseases such as Alzheimer's (Khan et al., 2013).

According to ICMM, (2007), the dermal absorption factor of 0.3 % for soluble and poorly soluble Cu substances and also stated that the available studies provides no consistent evidence that dermal absorption is greater for soluble than for poorly soluble Cu substances.

The production of Cu has lifted over the last decades and due to this, the quantities of the Cu in environment have increased. It will then up mainly in soils, may also contain large quantities of Cu after from the air has settled. Phosphate fertilizers production is the examples from human activities that contribute to Cu release have already been named that may contribute to adverse human health. According to Nazir et al., (2015) Cu also can accumulates in the liver and brain.

2.3 Methods to determine heavy metals in soil sample

2.3.1 Extraction heavy metals in soil

Previous study, Khan et al. (2013) stated that the chosen method for extract the soil sample was wet digestion method (WDM) and the chemicals used to extraction were HNO_3, H_2SO_4 and $HClO_4$ in the ratio 5:1:1. The digest extracts was filtered into clean volumetric flasks and diluted up to 100 mL volume using highly purify deionized water and the dilution was keep at room temperature for further analysis. The method which was Aqua regia extraction leads to the maximum soluble acid amount of metals with recoveries from 89 to 110 % for some metals in soils and sediment (Sastre, Sahuquillo, Vidal, & Rauret, 2002).

According to Khairiah et al., (2013), the soil was extracted using ammonium acetate, NH_4CH_3OO (pH 7) for extraction method and the samples were then filtered using 0.45 μm milipore filter paper. Other studies, which using microwave digestion by Micó et al., (2006) and Melaku, Dams, & Moens, (2005). Other than that conventional method also applied to extract the soil. For conventional method, using oven heating is time consuming compared to microwave digestion. Microwave digestion method was more expensive and requires some experience (Melaku et al., 2005). Furthermore, the cross contamination of the sample due to open digestion may occur. Conventional method may cost in termed of the apparatus and equipment used, despite the disadvantages.

2.3.2 Instrumental analysis

Previous study stated that the most selected instrument for soils extracts were analyzed using graphite furnace atomic absorption spectrometry-GFAAS (Micó et al., 2006), atomic absorption spectrophotometer Perkin Elmer Model 3300 (Khairiah et al., 2013) and Perkin Elmer AAS-700 (Khan et al., 2013). Cu, Cr, Pb, Ni, and Cd concentrations were determined by AAS (Liu et al., 2011). According to

Feng, Zhao, & Bian, (2012) total concentration Pb , Cd, Cr and Zn samples were determined using F/GAAS.

Based on Sue et al., (2014), the instruments available and used by the researchers in their studies to determine the concentration of heavy metals are Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) and Flame Atomic Absorption Spectrophotometer (F-AAS). Furthermore, compared to ICP-OES, ICP-MS has lower detection limit for heavy metals element in soil and water.

2.4 Legal Requirement & Guidelines

According to Zarcinas et al., (2004) there were currently no soil quality reference values for heavy metals or toxic metals and there is a need to establish levels of heavy metals found in soils commonly used for agricultural production in Malaysia. However, the available standard of Netherlands for soil protection - Dutch target value are based on natural (background) soil levels and on negligible risk concentrations (Zarcinas et al., 2004), Trace Elements in Soils and Plants (Kabata-Pendias & Pendias, 2011) and Australian Ecological Investigation Levels (EILs) (NEPM, 2013) was used to compared the values of the concentration of heavy metals in paddy soil at Kampung Sawah Sempadan, Tanjung Karang.

Table 2.1: Soil reference values (Dutch target values, Kabata-Pendias and Pendias and Australian EIL) for heavy metals in soil. All value are in mg/kg.

	Soil reference value		
Metals	Dutch target value [a]	Kabata-Pendias and Pendias [b]	Australian EIL [c]
As	29	6.83	100
Cd	0.8	0.41	20
Cr	100	59.5	100
Cu	36	38.9	6000
Ni	35	27	400
Pb	85	29	300
Zn	140	70	7400

[a] Recommended Concentration of Heavy Metals in Soil by Netherlands for soil protection - Dutch target value, [b] Recommended Concentration Background Contents of Trace Elements in Continental Crust and Soil Surface by Kabata-Pendias & Pendias (2011), [c] Recommended Concentration of Heavy Metals in Soil by Australian Ecological Investigation Levels (EILs) (NEPM, 2013)

2.5 Health Risk Assessment

Health risk assessment can be defined as methodological approach to identify, characterize, and analyze dose-response relationship of the chemicals toxicities to generate a numerical data. There are two types which health risk assessment can be performed, which are qualitative risk assessment and quantitative risk assessment. Quantitative health risk assessment is used to estimate the potential health risk due to the exposure of carcinogenic or non-carcinogenic substances.

There are two categories for the potential chronic health risk, which are non-carcinogenic and carcinogenic risk. For the non-carcinogenic risk, there is a level of exposure below which no adverse effects will be observed known as threshold dose (U.S. EPA 1989). Non-carcinogenic risk can be calculated using hazard quotient (HQ). HQ is the ratio of exposure level to non-carcinogenic elements to the reference dose (RfD).

According to United States Environmental Protection Agency (USEPA), if the value of HQ is less than one, then there is no significant risk of non-carcinogenic effects. However, if the values calculated exceed one, there may be a significant concern for potential non-carcinogenic health risk. Carcinogenic health risk estimates represent the incremental probability that an individual will develop cancer over a lifetime as a result of a specific exposure to a carcinogenic chemical (U.S. EPA 1989) and can be estimated using dermal cancer risk (DCR) equation.

CHAPTER 3

METHODOLOGY

3.1 Study Design

This study was classified as a cross sectional study in which to determine the concentrations of seven selected element of heavy metals in Kampung Sawah Sempadan, Tanjung Karang and health risk assessment among paddy farmers. The sample population was choose randomly based on the inclusion criteria stated in this research and the sample collection also choose randomly in 24 different block which consist A to X block.

3.2 Study Area

There were few consideration in selecting the location of the study, these included (i) the location must be an agricultural area in which the main crop is paddy, (ii) the location must be at a short distance from the laboratory, and (iii) the location should comprised of residents who are willing to cooperate with this study. Kampung Sawah Sempadan, Tanjung Karang, Kuala Selangor which was known by its vigorous rice cultivation activity has been selected as the study area. The location and maps of study area was depicted in Figure 3.1 and Figure 3.2.

Figure 3.1: Maps of the sampling sites at Kampung Sawah Sempadan, Tanjung Karang, Kuala Selangor

Figure 3.2: Location of the sampling sites at Kampung Sawah Sempadan, Tanjung Karang, Kuala Selangor

3.3 Sample location

A total 72 soil samples were collected at 0 – 20 cm depth from 24 different plots from A to X at Kampung Sawah Sempadan paddy field. This depth was choosing based on the accumulation of the heavy metals in soil which known as plough layer. Soil Samples were collected in zip-locked bag and transported on ice back to the laboratory for analysis immediately.

3.4 Sample population

The sample study was comprised of paddy farmers who work directly and handled the fertilizers. The required sample population comprised of paddy farmers that handle the crop by coming in contact with soil directly. The record from district office of Tanjung Karang which was the total population of paddy farmers in the Tanjung Karang was approximately 7679, and the population of the paddy farmers in the study area (Kampung Sawah Sempadan) was 1147. Below was the sample size estimation calculated according to Social Research Methods (Lemeshaw, 1990). The sample size was determined by using formula as below.

$$N = \frac{\left[Z_{1-\alpha}\sqrt{2P\,(1-P)} + Z_{1-\beta}\sqrt{P_1\,(1-P_1) + P_2\,(1-P_2)}\right]^2}{P_1 - P_2} \qquad \text{(Equation 3.1)}$$

Where,

N = Sample size

$Z_{1-\alpha} = 1.282$

$Z_{1-\beta} = 0.842$

$P_1 = 0.21$

$P_2 = 0.1$

$P = \frac{P_1 + P_2}{2}$

For 95% level of confidence, the margin error is ± 2 times the standard error. Therefore, the error (e) is 0.05.

The value of prevalence P_1 is 0.21 based on the study by (WHO, 2014) on the occupational dermal disease and P_2 is 0.1 from the study by Vuong TA, van der Hoek W, Ersbøll AK, Nguyen VT, Nguyen DT, Phung DC, (2007) on dermatitis among farmers engaged in peri-urban aquatic food production in Hanoi, Vietnam.

$$N = \frac{\left[1.282\sqrt{2(0.155)(1-0.155)} + 0.842\sqrt{0.21(1-0.21) + 0.1\,(1-0.1)}\right]^2}{0.21 - 0.1}$$

The minimum sample size will be rounded up to 20% to recover the loss of respondents throughout the study. Thus,

20% of 97

= 20 00

= 97 + 20

= 117 respondents. Thus, the total sample size population is 117.

The name list of paddy farmers was obtained from Tanjung Karang Farmers Organization Office. However, only those who fulfilled the inclusion criteria were included in the study.

Inclusion Criteria:

- Paddy farmers within the age group of 18 to 70 years old
- Paddy farmers who have been working for > 6 months in the paddy field
- Paddy farmers who work directly to the paddy soil

3. 5 Instrumentations

3.5.1 Questionnaire

In this study, the questionnaires was developed to acquire information on the respondents' gender, the average body weight (kg), exposure frequency (events/year), exposure duration (years), and averaging time (days) to calculate the health risk of the farmers. This information then incorporated into the equation (Equation 3.4) to calculate the dermal risk exerted by the farmers.

The questionnaire developed was based on (NOSQ, 2002). These references were selected due to the similarities of the study conducted in terms of dermal risk assessment. The questionnaire consists of three parts for farmers to answer which is (i) Part A: Personal Information (ii) Part B: Exposure to Heavy Metals and (iii) Part C: The Use of Personal Protective Equipment.

3.5.2 Extraction of Paddy Soil

The soil was collected in paddy field at Tanjung Karang, Kuala Selangor. The soil samples were passed through a 2 mm sieve, homogenized, and extracted with their manually occurring. The aliquot was stored at -20 °C until further analysis. In this experiment, the soil was extracted using Aqua-Regia Digestion Method. According method by Miroslav (1998), 130 mL of HCl (R & M Marketing, Essex, U.K) was added to 120 mL water and mix it. The 150 mL of the solution was added to 50 mL HNO_3 (R & M Marketing, Essex, U.K) and mixed it. The solution were prepared and handled with great care.

3.5.3 Instrumental analysis

The instrument Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (PerkinElmer, U.S.A) brand was used to determine the concentration of the selected heavy metals (Cd, Cr, As, Ni, Zn, Pb, Cu). The parameter settings of the ICP-MS were as followed:

Parameter Settings of the instrument Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (PerkinElmer, U.S.A):

Nebulizer Gas Flow	:	0.87
Lens Voltage	:	6.50
ICP RF Power	:	1050.00
Analog Stage Voltage	:	-1750.00
Pulse Stage Voltage	:	1050.00
Discriminator Threshold	:	40.00
AC Rod Offset	:	-6.00

Table 3.1: Instrument detection limit for ICP-MS PerkinElmer (mg/kg)

Heavy metals	Instrument Detection Limit (ICP-MS) (mg/kg)
Cd	0.09
Cu	0.2
Cr	0.2
Zn	0.3
Ni	0.04
Pb	0.04
As	0.06

According to Miroslav (1998), the advantages of ICP-MS methods for trace metals determination are due to their low detection capabilities and fast method to test for few elements in just a single analytical run. In addition, the used of ICP-MS to analyze the sampled has many advantages such as has good detection limits, linearity of calibration curves, compatibility with different acid mixtures for samples dissolution and low sensitivity to matrix effects (Bettinelli, Beone, Spezia, & Baffi, 2000).

3.6 Quality control

According to Sue et al, (2014) all the apparatus and glass wares were soaked in acid bath and cleaned with detergent, rinsed abundantly with ultrapure water and left air dried to prevent any contamination on the samples. To obtain an accurate measurement of heavy metals, blank extraction for the sample extraction will include for every batch of sample extraction (Sue, 2014).

The blanks were prepared using same method with the sample to be analyzed but the sample was not added to the blank. Calibration curve was obtained by prepare the calibration standards for metals Pb, Zn, Cr, Cd, Ni, Cu and As. Calibration curve was constructing for each metal by plotting the graph of the intensity against concentrations of heavy metals. The ranged of linearity will determine by referring to the linear regression coefficient (R^2) of the calibration curve. To ensure the results accurate, the R^2 obtained must be more than 0.995 (Sue, 2014).

The extraction recovery was conducted in order to validate the analytical procedure and it was done by spiking the samples with varied amounts of standard solutions of the metal (Sue, 2014). The standard used to spike was multi-element calibration standard 3 (PerkinElmer, U.S.A). To compare the concentration of each heavy metal spiked before sample digestion, the percentage of recovery was calculated. Its concentration spiked after sample digestion in the same sample matrix using Equation 3.2.

$$\% \, Recovery\ of\ extraction = \frac{Standard\ spiked\ before\ extraction}{Standard\ spiked\ after\ exraction} \times 100 \qquad \text{(Equation 3.2)}$$

The recovery of extraction for heavy metals was shown in Table 3.2. The extraction recovery was conducted in order to validate the analytical procedure and it was done by spiking the samples with varied amounts of standard solutions of the metal (Sue, 2014). To compare the concentration of each heavy metal spiked before sample digestion, the percentage of recovery was calculated. The recovery of each element of heavy metals in this study shows more than 80 percent. The highest recovery is Ni (89.9%) followed by Cr (89.1%), As (88.7%), Cu (88.3%), Pb (86.5%), Cd (82.7%) and Zn (81.3%).

Table 3.2: The recovery of extraction for heavy metals

Heavy metals	Mean concentration spiked before extraction (mg/kg)	Mean concentration spiked after extraction (mg/kg)	Recovery (%)
Cd	2.34	2.83	82.7
Cu	2.68	3.04	88.2
Cr	0.90	1.01	89.1
Zn	6.07	7.47	81.3
Ni	3.20	3.56	89.9
Pb	4.00	4.62	86.6
As	3.70	4.17	88.7

In this study, the ranged of linearity was determined by referring to the linear regression coefficient (R^2) of the calibration curve. According to Sue et al,. (2014) to ensure the results accurate, the R^2 obtained from the graph must be more than 0.995. Calibration curve was constructed for each metal by plotting the graph of the intensity against concentrations of heavy metals. In this study, R^2 shows that for each element has more than 0.995. The linear regression coefficient (R^2) of the calibration curve was shown in Table 3.3.

Table 3.3: Linear regression coefficient (R^2) of the calibration curve

Heavy metals	Correlation Coefficient (R^2)
Cd	0.9999
Cu	0.9999
Cr	0.9992
Zn	0.9998
Ni	0.9999
Pb	0.9981
As	0.9999

3.7 Health Risk Assessment

3.7.1 Risk Characterization

The risk was quantified under the present of environmental conditions at farmers at Kampung Sawah Sempadan for the selected exposure scenarios. The risk characterization was considered separately as carcinogenic and non-carcinogenic effects.

3.7.1.1 Non- carcinogenic risk

The non-carcinogenic risk was calculated using Hazard Quotient (HQ) which is a ratio of exposure dose to the compounds-specific reference dose (RfD). The dermal RfD for the seven selected heavy metals were adjusted from oral toxicity factor. This adjustment accounts for the absorption efficiency in the critical study, which forms the basis of the RfD. The value for dermal RfD were adapted from the Risk Assessment Information System (Hashmi et al., 2014) based on USEPA studies in Tables 3.4.

The dermal HQ was defined based on (USEPA, 2004) the equation as follows:

$$HQ = \frac{DAD}{RfD} \qquad \text{(Equation 3.3)}$$

Where,

DAD = Dermal Absorbed Dose (mg/kg-day)

RfD = Reference dose (mg /kg-day)

DAD is defined by the following equation USEPA, (2004):

$$DAD = \frac{DAevent \times EV \times ED \times EF \times SA}{BW \times AT}$$ (Equation 3.4)

Where:

DA_{event} = Absorbed dose per event (mg/cm^2-event)

SA = Skin surface area available for contact (cm^2)

EV = Event frequency (events/day)

EF = Exposure frequency (days/year)

ED = Exposure duration (years)

BW= Body weight (kg)

AT = Averaging time (days)

Dermal Absorbed Dose per Event by following equation (USEPA, 2004):

$$DAevent = Csoil \times CF \times AF \times ABSd$$ (Equation 3.5)

Where,

DA_{event} = Absorbed dose per event (mg/cm^3-event)

C_{soil} = Chemical concentration in soil (mg/kg)

CF = Conversion factor (10^{-6} kg/mg)

AF = Adherence factor of soil to skin (mg/cm^2-event)

ABS_d = Dermal absorption fraction

Table 3.4: The dermal reference dose (RfD) for selected heavy metals

Elements	RfD (mg/kg-day)
Cu	1.2×10^{-2}
Cr	7.5×10^{-3}
Zn	6.0×10^{-2}
Ni	5.4×10^{-3}
Pb	N/A
As	1.2×10^{-4}
Cd	5.0×10^{-6}

N/A = Not available

3.7.1.2 Carcinogenic risk

Dermal cancer risks were estimated using Lifetime Cancer Risk (LCR) equation, which represent incremental probability that an individual will develop cancer over a lifetime as a result of a specific exposure to a potential carcinogen. The slope factors for the seven selected heavy metals are shown in Table 3.5.

$$Dermal\ cancer\ risk = DAD \times SF \qquad \text{(Equation 3.6)}$$

Where,

DAD = Dermal Absorbed Dose (mg/kg-day)

SF = Absorbed cancer slope factor (mg/kg-day)

Table 3.5: The dermal slope factor for selected heavy metals

Elements	SF (mg/kg-day)
Cu	N/A
Cr	N/A
Zn	N/A
Ni	N/A
Pb	N/A
As	3.66
Cd	N/A

N/A = Not available

CHAPTER 4

RESULT & DISCUSSION

4.1 Socio-demographic Background of Respondents

The socio demographic data of respondents are summarized in Table 4.1. Respondents' age ranged from 24 to 70 years old as in inclusion criteria for this study with a mean of 49.8 ± 12.17 years old. The mean body mass indexes (BMIs) of the respondents fell in the healthy and normal range of 25.8 ± 4.27 kg/m^2. Exposure duration of the respondents towards the occupation as paddy farmers in Kampung Sawah Sempadan, Tanjung Karang was in the range of 25.5 ± 14.51 years.

From the questionnaire, the information on the use of Personal Protective Equipment (PPE) among the farmers, only 36.8% of the farmers used full PPE such as boots and gloves which covered hand and feet of paddy farmers. Besides that, the frequency of the farmers changing the clothes after farming was 95.7%. In addition, the percentage of the farmers take shower after farming was 81.2%.

Table 4.1: Sociodemographic information (n=117)

Variable	Mean ± SD	Median	Min Value	Max Value
Age	49.8 ± 12.17	51	24	70
Weight (kg)	70.3 ± 12.77	70	45	120
Height (m)	1.65 ± 0.07	1.66	1.52	1.86
BMI (kg/m^2)	25.8 ± 4.27	26.0	17.6	39.2
Exposure Duration (years)	25.5 ± 14.51	23	1	55
Event Frequency (days/ year)	1.21 ± 0.43	1	1	3
Variable	**Category**	**Frequency**	**Percentage,%**	**Cumulative percentage**
Wash up/Shower after farming	Yes	95	81.2	81.2
	No	22	18.8	100
Change clothes after farming	Yes	112	95.7	95.7
	No	5	4.3	100
Use full PPE (boots and gloves)	Yes	43	36.8	36.8
	No	74	63.2	100

4.2 Concentration of Heavy Metals in Soil Samples

In this study, 24 paddy blocks were chosen as sampling point representing each block of Kampung Sawah Sempadan starting from block A to X. The soil samples were collected at each plot and the concentrations of 7 heavy metals (Cu, Cr, Zn, Ni, Pb, As and Cd) were determined. The results for concentration of selected heavy metals in soil samples are summarized in Table 4.2.

Among the selected heavy metals, As has the highest mean concentration 17.13 mg/kg followed by Cr (14.80 mg/kg), Zn (7.21 mg/kg), Pb (6.64 mg/kg), Cu (3.37 mg/kg), Ni (1.82 mg/kg) and Cd (0.06 mg/kg). By comparing the mean concentration of the heavy metals in soil to the available standard of Netherlands for soil protection - Dutch target value (Zarcinas et al., 2004), Trace Elements in Soils and Plants (Kabata-Pendias & Pendias, 2011) and Australian Ecological Investigation Levels (EILs) (NEPM, 2013), the As has exceeded the standard from Netherlands for soil protection - Dutch target value (Zarcinas et al., 2004) and the maximum value has exceeded the Trace Element in Soils and Plants by (Kabata-Pendias & Pendias, 2011). The maximum value of As that exceed the limit was found in block T1.

The maximum concentrations of the heavy metals in this study were arranged in hierarchy from high to low as follows: As> Cr> Zn> Pb> Cu> Ni> Cd.

Table 4.2: The heavy metals concentration in soil samples of the study areas (mg/kg) and the comparison with available standards

Variables	Cr	Ni	Cu	Zn	As	Cd	Pb
Mean ± SD	14.80 ± 6.61	1.82 ± 0.74	3.87 ± 1.92	7.21 ± 3.17	17.13 ± 7.11	0.06 ± 0.04	6.64 ± 2.70
Median	14.54	1.65	3.51	6.72	17.94	0.05	6.04
Min value	3.90	0.88	0.77	1.91	4.42	0.02	2.31
Max value	36.72	4.63	7.98	20.92	31.49	0.29	16.05
Soil Standards	100^a	35^a	36^a	140^a	29^a	0.8^a	85^a
	59.5^b	27^b	38.9^b	70^b	6.83^b	0.41^b	29^b
	100^c	400^c	6000^c	7400^c	100^c	20^c	300^c

[a] Recommended Concentration of Heavy Metals in Soil by Netherlands for soil protection - Dutch target value, [b] Recommended Concentration Background Contents of Trace Elements in Continental Crust and Soil Surface by Kabata-Pendias and Pendias, [c] Recommended Concentration of Heavy Metals in Soil by Australian Ecological Investigation Levels (EILs)

The average concentration of Cr in the paddy was 14.80 ± 6.61 mg/kg and has the maximum reading of 36.72 ± 0.07 mg/kg at B2 and Cr has the lowest reading at M2 with the concentration 3.90 ± 0.02 mg/kg. The mean concentration of Ni was 1.82 ± 0.74 mg/kg with maximum value of concentration recorded at W1 (4.63 ± 0.08 mg/kg). The average concentration of Cu was 3.87 ± 1.92 mg/kg and the maximum concentration was 7.98 ± 0.19 mg/kg at X2. Zn recorded an average concentration of 7.21 ± 3.17 mg/kg with the maximum concentration of 20.92 ± 0.22 mg/kg at D1. The mean concentration of Pb in paddy soil was 6.64 ± 2.7 mg/kg and the maximum concentration was recorded at Q2 (16.05 ± 0.52 mg/kg). As shows the highest mean concentration across all heavy metals 17.13 ± 7.11 mg/kg with the maximum concentration from sample T1 (31.49 ± 0.7 mg/kg). In this study, Cd shows the lowest mean concentration (0.06 ± 0.04 mg/kg) and the maximum level of Cd was from sample A3 (0.29 ± 0.004 mg/kg).

In general, the concentration of heavy metals in this study was below the standards and in comparison to the other studies in paddy cultivation area except for As. Based on previous study, accumulation of heavy metals in the top soil were believed come from the anthropogenic sources (Aminah, A. et al., 2009). Referring to the total mean concentration of heavy metals present in soil samples in Kampung Sawah Sempadan, there were believed that the heavy metals concentration comes from the anthropogenic sources practiced by the paddy farmers. It is because of according to Liu et al., (2011) the initial sources of heavy metals in soil are the parent materials from which the soil were derived mainly from pedogenetic parent materials and since lower mean concentration among selected heavy metals in this study, metals accumulation based on human activities were not significant related to the concentration of heavy metals in soil samples in Tanjung Karang.

The concentrations of heavy metals in all samples were less than the recommended maximum level except for As. Today, the farmers in Kampung Sawah Sempadan at Tanjung Karang are mainly active in paddy cultivation activity and there are no other anthropogenic activities such as industrial or mining around the area. There also has no such polluted by livestock manure from near area. This may explain the result of lower concentration of heavy metals in paddy soil as the area is not polluted from the discharge of anthropogenic activities.

Previous study in Beijing (China) by Lu et al., (2015) stated that contents of fertilizers and pesticides can be the causes of to the accumulations of heavy metals in soil. However, the concentration of heavy metals in this study were not exceed any of the standard indicating there was no overused of pesticides and fertilizers from farmers.

In this study, Cd shows the lowest concentration which ranged from 0.02 to 0.29 mg/kg. Previous study by Aminah, A. et al., (2009) stated that the minimum concentration of Cd in paddy soil was originated from natural soil deposits. As compared to study from Peninsular Malaysia, the mean concentrated of Cd in soil was 0.12 mg/kg which more than the mean concentration of Cd in Tanjung Karang. The ranged reported was 0.01 to 2.02 mg/kg in soil in Peninsular Malaysia. According to Aminah et al., (2009) citation manufactured fertilizers were usually not sufficiently purified, and could contain several impurities including heavy metals. By repeating application of fertilizers could increase the Cd content in soils but since the Cd shows the lowest concentration in Tanjung Karang, it can be related to the natural sources in the environment.

As has the highest mean concentration (4.42 - 31.49 mg/kg) in paddy soil at Tanjung Karang and exceed the Dutch Target Value and also the maximum value of the As concentration was exceed the Kabata-Pendias & Pendias, (2011) standard which can be apply for all soil in the world. According to Liu et al., (2011), the paddy soil was irrigated with As polluted ground water and deposited in soil. In previous study, Zarcinas et al., (2004) reported that mean concentration for As in all Malaysian soils (Peninsular) was 16.8 mg/kg where lower than the research study area in Tanjung Karang, Kuala Selangor. The ranged present was 0.28 – 280 mg/kg which was quite higher.

4.3 Health Risk Assessment on Farmers

Heavy metals contaminations are important as they can pose potential toxicity to the environment and metals have the ability to accumulate in the human body system and cause damage to system organs. Thus, it is important to assess the health risk of the people who are exposed to the heavy metals in soil. To assess the health risk among the paddy farmers, the information on paddy farmer's exposure to paddy soil was collected throughout questionnaire that has been distributed among them. Some of the information was done via interviewed the farmers at Kampung Sawah Sempadan, Tanjung Karang. The important of the questionnaire was to know the event frequency the farmers exposed to soil while working at paddy field, the exposure duration, the exposure frequency, averaging time and also body weight of the paddy farmers. The information were used to calculate the health risk of the farmers exposed directly to the seven selected heavy metals such as As, Cr, Ni, Pb, Cd, Zn and Cu.

Health risk assessment was done on farmers for dermal exposure of selected heavy metals. The information needed to calculate the health risk was obtained by the interview using the questionnaire. The results were averaged for each parameter to represent the farmers in Kampung Sawah Sempadan. The data for the health risk evaluation were listed as in Table 4.3.

Table 4.3: Information on Farmer's Exposure to paddy soil

Parameters	Average
Event frequency (events/day)	1[a]
Exposure duration (years)	25.5[a]
Exposure frequency (days/year)	220[a]
Body weight (kg)	70.3[a]
Averaging time (days)	5659.5[a]
Skin surface area (hands) for adult male (cm^2)	990[b]
Skin surface area (feet) for adult male (cm^2)	1310[b]
Skin surface area (hands and feet) for adult male (cm^2)	2300[b]

[a] Information obtained from questionnaires administered to the respondents
[b] Adopted from U.S EPA Exposure Factor Handbook (2011)

Three situations has been separate regarding to the use of PPE and the skin surface area that are exposed to the heavy metals in the paddy soil. The first situation is where the farmers wear gloves but do not wear the boots, hence exposing the feet to the paddy soil. Second situation is where farmers wear boots but do not wear the gloves and exposing the hands to the paddy soil. Meanwhile, the third situation is where the farmers do not wear both gloves as well as boots thus exposing hands and feet to the paddy soil that contain heavy metals.

The risks were categorized into non-carcinogenic and carcinogenic and the effects were considered for each heavy metal by calculating HQ and DCR respectively.

Table 4.4: The health risk assessment (value for HQ and DCR) on human health in Tanjung Karang

Heavy Metals	Hand exposure		Feet exposure		Hand and feet exposure	
	HQ	DCR	HQ	DCR	HQ	DCR
Cr	5.4×10^{-3}	N/A	7.1×10^{-3}	N/A	12.5×10^{-3}	N/A
Ni	8.1×10^{-3}	N/A	10.7×10^{-3}	N/A	18.7×10^{-3}	N/A
Cu	2.2×10^{-3}	N/A	2.9×10^{-3}	N/A	5.1×10^{-3}	N/A
Zn	1.6×10^{-4}	N/A	2.2×10^{-4}	N/A	3.8×10^{-4}	N/A
As	2.9×10^{-1}	1.3×10^{-4}	3.9×10^{-1}	1.7×10^{-4}	6.8×10^{-1}	3.0×10^{-4}
Cd	1.1×10^{-1}	N/A	1.5×10^{-1}	N/A	2.7×10^{-1}	N/A
Pb	N/A	N/A	N/A	N/A	N/A	N/A

The hazard quotient (HQ) and dermal cancer risk (DCR) on human health of farmers in Kampung Sawah Sempadan, Tanjung Karang was below 1 and less than $1x10^{-4}$. By using some proper Personal Protective Equipment such as glove and boot can eliminate all potential dermal or skin disease to the farmers.

The farmers in Kampung Sawah Sempadan exposed to the paddy soil for average 2 hours per day depending on the weather and the peak season. The average exposure duration of the respondents was 25.5 ± 14.51 years old. The ranged of the age for the respondents were 24 to 70 years old. Majority of the respondents are full time farmers while some of them have another job and only involve in the paddy cultivation activities as part time and started to become farmers at the late age.

The paddy cultivation activities take 110 days per season with 2 seasons per year which make up the exposure frequency of 220 days/year. The average body weight of the respondents were 70.3 kg and almost similar to the body weight that were recommended by USEPA which are 70 kg. For dermal exposure to soil, 50th percentile body surface area values are used to estimate contact rates (USEPA, 1989). The skin surface areas were considered based on the area that the farmers were exposed while handling the paddy soil. The areas are feet and hands as some of the farmers do not wear the appropriate PPE such as boots and gloves.

The HQ calculated for feet exposure, hands exposure and for both hand and feet exposures were less than 1 for all heavy metals. According to USEPA, the value of HQ less than one indicates that there is no significant risk of non-carcinogenic effects Although the average exposure duration exposed to heavy metals were as high as 25.5 years, the risk is not significant as the concentration of heavy metals in the paddy soil were in low concentrations. Among the selected heavy metals, only As has the reported dermal slope factor, thus the DCR was only calculated for As. The DCR calculated were in the range of 3.0×10^{-4} to 1.3×10^{-4} shows the carcinogenic risk are in the acceptable risk level of 10^{-4} to 10^{-6} (MAEH, 2012).

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

Overall, the present study showed the selected heavy metals in the paddy soil at Kampung Sawah Sempadan present in low concentration from the recommended concentration of heavy metals by comparing the mean concentration of the heavy metals in soil to the available standard of Netherlands for soil protection - Dutch target value, Trace Elements in Soils and Plants and Australian Ecological Investigation Levels (EILs), except for As which exceeded the recommended value for mean concentration of Netherlands for soil protection - Dutch target value and the maximum concentration of As which in T1 block was exceed the Trace Elements in Soils and Plants. As shown that the highest average concentration was 17.13 mg/kg compared to other selected heavy metals in this study. The highest mean concentration of the selected heavy metals was As followed by Cr> Zn> Pb> Cu> Ni> Cd. The mean concentration of As was 17.13 mg/kg followed by Cr, Zn, Pb, Cu, Ni, Cd which are 14.80 mg/kg, 7.21 mg/kg, 6.64 mg/kg, 3.37 mg/kg, 1.82 mg/kg and 0.06 mg/kg respectively. The factors that related to the existent of the heavy metals in soil was the used of fertilizers and pesticide applied to paddy plant and paddy soil by the farmers that can end up in soil was described from the result. Here, the most possible source of pollution was the accumulation of the heavy metals in natural environment. Anthropogenic activity such as fertilizers and pesticides used by the farmers was still under the control because since concentrations of heavy metals in soil were low. The others reason for As exceed the level for Dutch Target Value and Kabata-Pendias & Pendias standard when the paddy soil was irrigated with As polluted ground water and deposited in soil. The HQ for all heavy metals were below 1 thus indicate that the risk of non-carcinogenic dermal health risk to heavy metals in paddy soil among farmers were negligible. There was also no significant carcinogenic risk for arsenic exposure as the DCR obtained was ranged from 3.0×10^{-4} to 1.3×10^{-4}.

5.2 Limitation and Recommendation

The recommendation for the farmers in Kampung Sawah Sempadan, Tanjung Karang is they should wear the high-cut boots or gloves when handling the fertilizers or pesticides and when in contact with the soil that contaminated by heavy metals at paddy fields. To encourage the farmers to wear the PPE in paddy fields and in contact with the paddy soil, health promotion in study area should be conduct to educate the farmers about the important of wear PPE. The used of PPE is important because the findings of this study shown that the mean concentration of As was exceeded the standard which the long-term exposure to high level of inorganic As exhibit several symptoms such as pigmentation, skin lesions and hyperkeratosis to human. There was limitation in this study where some of the elements selected or studied do not have cancer slope factor and reference dose, so their respective dermal cancer risk and hazard quotient cannot be determined.

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APPENDICES

JAWATANKUASA ETIKA UNIVERSITI UNTUK PENYELIDIKAN MELIBATKAN MANUSIA (JKEUPM)
UNIVERSITI PUTRA MALAYSIA, 43400 UPM SERDANG, SELANGOR, MALAYSIA

BORANG B1: PENERANGAN DAN PERSETUJUAN RESPONDEN

Sila baca maklumat berikut dengan teliti. Sekiranya anda mempunyai sebarang pertanyaan, sila kemukakan kepada penyelidik.

1.TAJUK KAJIAN

Pencemaran Logam Berat Dalam Tanah Padi Dan Risiko Kesihatan Yang Berkaitan Dalam Kalangan Petani Di Tanjung Karang, Kuala Selangor.

2. PENGENALAN

Penilaian pencemaran logam berat dalam tanah padi dan risiko kesihatan yang berkaitan dalam kalangan petani di Tanjung Karang, Kuala Selangor adalah proses dimana peneyelidik menganggar kebaraangkalian terjadinya penyakit bahaya jika terdedah pada bahan kimia di persekitaran yang tercemar disebabkn oleh penggunaan baja di kawasan sawah. Pesawah merupakan golongan yang banyak terdedah pada penggunaa baja yang mempunya bahan yang aktif seperti nitrogen, phosporus dan potassium. Terdapat kajian yang sebelum ini membuktikan kehadiran logam berat didalam tanah adalah disebabkan oleh penggunaan baja dan membawa risiko yang bahaya kepada penyakit berbahaya. Oleh itu, kajian ini dijalankan untuk mengenalpasti sekiranya wujud logam berat didalam tanah yang membawa penyakit kronik dlam kalangan pesawah. Sekiranya ada, pesawah dinasihatkan supaya berhati-hati ketika musim membaja.

3. APAKAH YANG PERLU ANDA LAKUKAN?

Anda perlu menjawab soalan soal selidik yang akan diedarkan bagi tujuan mendapatkan informasi mengenai berat badan, tempoh dan kekerapan pendedahan kepada baja.

4. SIAPA YANG TIDAK BOLEH MENYERTAI KAJIAN INI?

Penduduk kampung sawah sempadan yang tidak terlibat dengan aktiviti pertanian padi dan pesawah yang tidak menggunakan baja dalam tanaman. Selain itu, kanak-kanak dibawah 18 tahun juga tidak dibenarkan terlibat dalam kajian ini.

5. APAKAH FAEDAH MENYERTAI KAJIAN INI?

a)KEPADA ANDA SEBAGAI PESERTA?

Kajian ini akan menilai jika terdapat risiko kesihatan kulit apabila anda terdedah kepada penggunaan baja. Selepas selesai kajian, anda akan diberitahu sekiranya penggunaan baja adalah berisiko untuk mendapat penyakit kanser atau bukan kanser melalui pendedahan terhadap logam berat.

b) KEPADA PENYELIDIK?

Melalui kajian ini, risiko pesawah untuk mendapat penyakit kulit kronik sama ada berkaitan kanser atau tidak dapat ditentukan. Data yang didapati daripada kajian ini boleh digunakan lebih lanjut untuk kajian yang lebih mendalam dan spesifik mengenai penyakit yang dikenalpasti. Selain itu, hubungkait antara risiko, kekerapan penggunaan baja, dan penggunaan kelengkapan pelindung diri juga dapat dikenalpasti.

6. ADAKAH IA BERISIKO?

Tidak. Proses kajian yang akan dijalankan hanya memerlukan anda untuk menjawab soal selidik sahaja.

7. ADAKAH MAKLUMAT DAN IDENTITI SAYA KEKAL RAHSIA?

Ya. Maklumat dan identiti anda akan dirahsiakan, ini tidak akan mengganggu proses kajian.

8. SIAPA YANG SAYA PERLU HUBUNGI SEKIRANYA SAYA MEMPUNYAI SOALAN TAMBAHAN SEMASA MENGIKUTI PENYELIDIKAN INI?

Sekiranya terdapat apa-apa pertanyaan, anda boleh hubungi saya di nombor dan emel di bawah:

Nama: Intan Idayu binti Abd Kharni

No. Tel: 019-9948615

Emel: intankmm@yahoo.com.my

Sila tandatangan di sini sekiranya anda telah membaca dan memahami kandungan halaman ini _______

9. PERSETUJUAN

Saya... No Kad Pengenalan. .... beralamat.. ....dengan ini bersetuju untuk mengambil bahagian secara sukarela dalam penyelidikan yang tersebut di atas *(kajian klinikal/percubaan ubat-ubatan/rakaman video/kumpulan sasaran/temuduga/ soal selidik).

Saya telah diberi penjelasan secara menyeluruh mengenai penyelidikan ini dari segi metodologi, risiko dan komplikasi (seperti tertulis pada Helaian Penerangan Responden). Saya memahami bahawa saya berhak menarik diri dari penyelidikan ini pada bila-bila masa tanpa memberi sebarang alasan.Saya juga memahami bahawa sebarang maklumat yang berkaitan identiti saya akan dirahsiakan.

Saya* berminat / tidak berminat untuk mengetahui keputusan kajian yang melibatkan saya.

I setuju/tidak bersetuju untuk imej/gambar/rakaman video/ rakaman suara digunakan dalam apa jua bentuk penerbitan atau pembentangan. (sekiranya berkaitan).

*potong yang tidak berkenaan

Tandatangan .... (*Responden*)

Tarikh :..

Tandatangan .... (*Saksi*)

Nama :..

No. K/P: ....

Saya mengesahkan bahawa saya telah menerangkan kepada responden ini sifat dan tujuan penyelidikan yang tersebut di atas.

Tarikh ....

Tandatangan .... (*Penyelidik*)

JABATAN KESIHATAN PEKERJAAN & PERSEKITARAN

FAKULTI PERUBATAN DAN SAINS KESIHATAN

UNIVERSITI PUTRA MALAYSIA

PENCEMARAN LOGAM BERAT DALAM TANAH PADI DAN RISIKO KESIHATAN YANG BERKAITAN DALAM KALANGAN PETANI DI TANJUNG KARANG, KUALA SELANGOR

ARAHAN SOALAN:

1. Borang soal selidik ini mengandungi **TIGA (3)** bahagian iaitu:

   Bahagian A : Maklumat Diri

   Bahagian B : Pendedahan kepada Logam Berat

   Bahagian C : Penggunaan Kelengkapan Pelindung Diri

2. Anda diminta menjawab semua soalan yang ada dalam buku soalan ini
3. Buku soalan ini hendaklah dikembalikan kepada pengkaji setelah selesai menjawab

PENDEDAHAN KEPADA LOGAM BERAT DALAM KALANGAN PESAWAH DI KAMPUNG SAWAH SEMPADAN, TANJUNG KARANG

KEGUNAAN PENYELIDIK

BAHAGIAN A: MAKLUMAT DIRI

1. Umur: ………………… Tahun

2. Jantina:
   - [ ] Lelaki
   - [ ] Perempuan

3. Bangsa:
   - [ ] Melayu
   - [ ] Cina
   - [ ] India
   - [ ] Lain-lain (sila nyatakan): …………………………

4. Berat: ………… kg

5. Jisim berat badan: …………… kg/m^2

6. Adakah anda merokok?
   - [ ] Ya
   - [ ] Tidak

BAHAGIAN B: PENDEDAHAN KEPADA LOGAM BERAT

7. Pekerjaan sekarang: …………………………………..

8. Tempoh bekerja di tempat sekarang: ……………….. bulan/ tahun

9. Adakah anda mandi selepas ke tempat kerja?
   - [ ] Ya
   - [ ] Tidak

SULIT

10. Adakah anda menukar pakaian kerja setiap hari?

☐ Ya
☐ Tidak

11. Nyatakan kekerapan anda mengendalikan atau terdedah kepada tanah padi:

……………… kali/ hari
atau
……………… kali/ minggu
atau
………………. kali/bulan
atau
………………. kali/ tahun

BAHAGIAN C: PENGGUNAAN KELENGKAPAN PELINDUNG DIRI

12. Adakah anda menggunakan kelengkapan pelindung diri semasa bekerja?

☐ Ya
☐ Tidak

13. Apakah jenis kelengkapan pelindung diri yang digunakan semasa bekerja?

a. But	
b. Apron	
c. Baju kalis air	
d. Alat penafasan	
e. Sarung tangan	
f. Cermin mata	
g. Topeng	
h. Penutup kepala	

PENDEDAHAN KEPADA LOGAM BERAT DALAM KALANGAN PESAWAH DI KAMPUNG SAWAH SEMPADAN, TANJUNG KARANG	KEGUNAAN PENYELIDIK

14. Jika "Ya", berapa kerapkah anda memakai kelengkapan pelindung diri?

a. Sentiasa (setiap kali turun ke sawah)	
b. Hampir sentiasa (2-4 kali seminggu)	
c. Jarang (sekali seminggu)	
d. Sangat jarang (kurang daripada 4 kali sebulan)	

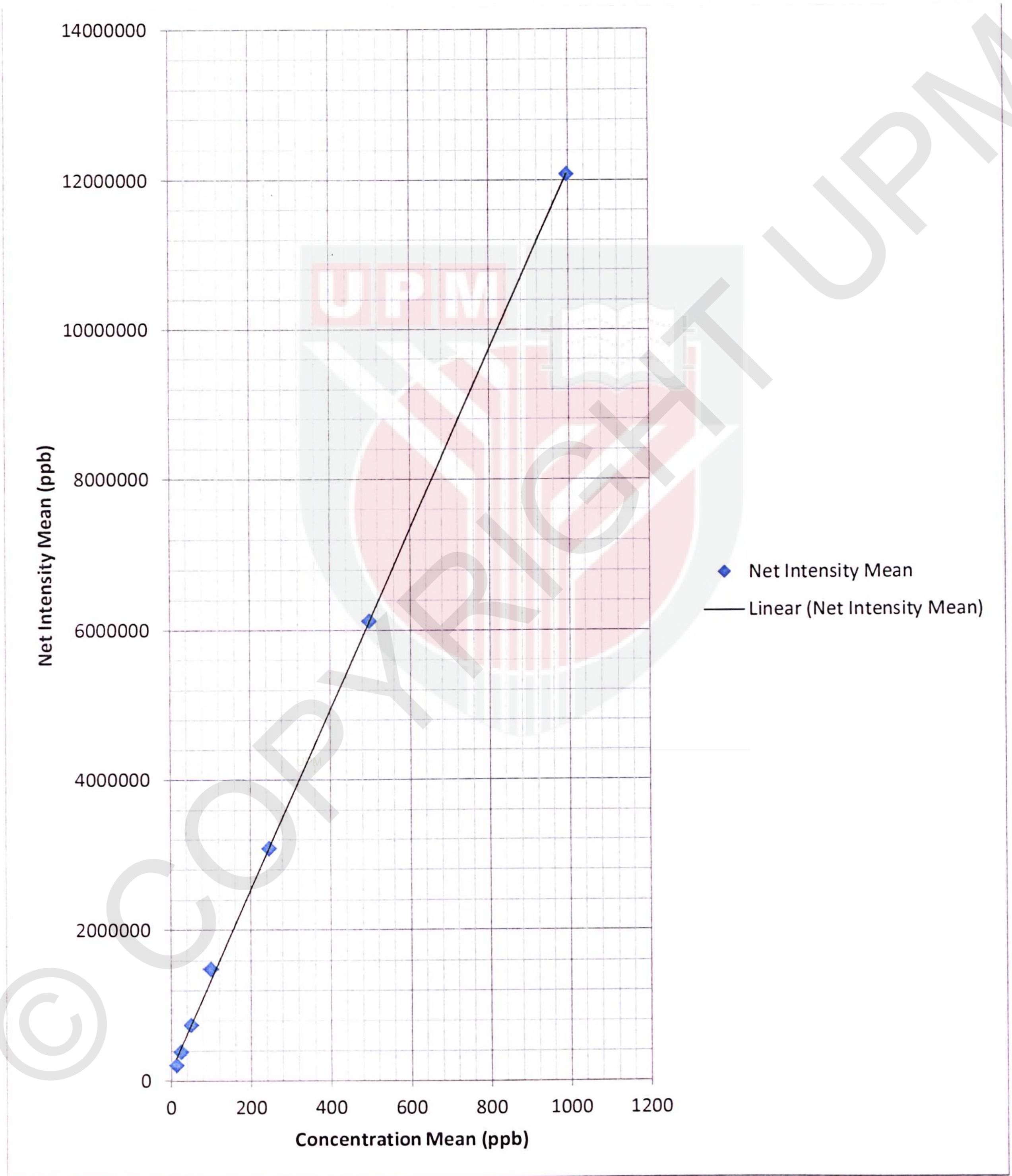
15. Kekerapan anda menukar kelengkapan pelindung diri:

a. Setiap hari	
b. Seminggu sekali	
c. Dua minggu sekali	
d. Sebulan sekali	
e. Lain-lain (sila nyatakan)	

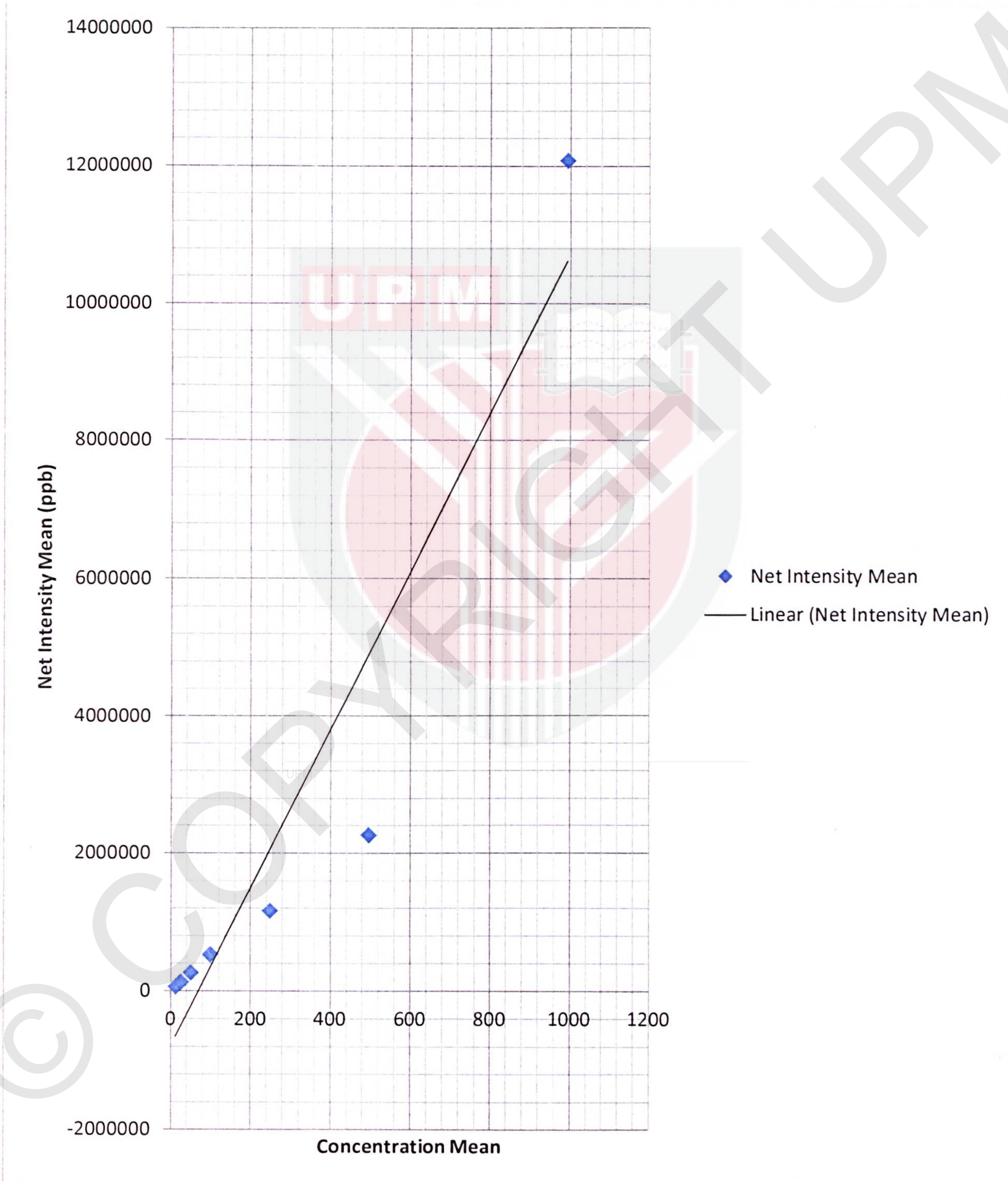
~Tamat-soal-selidik-

SULIT

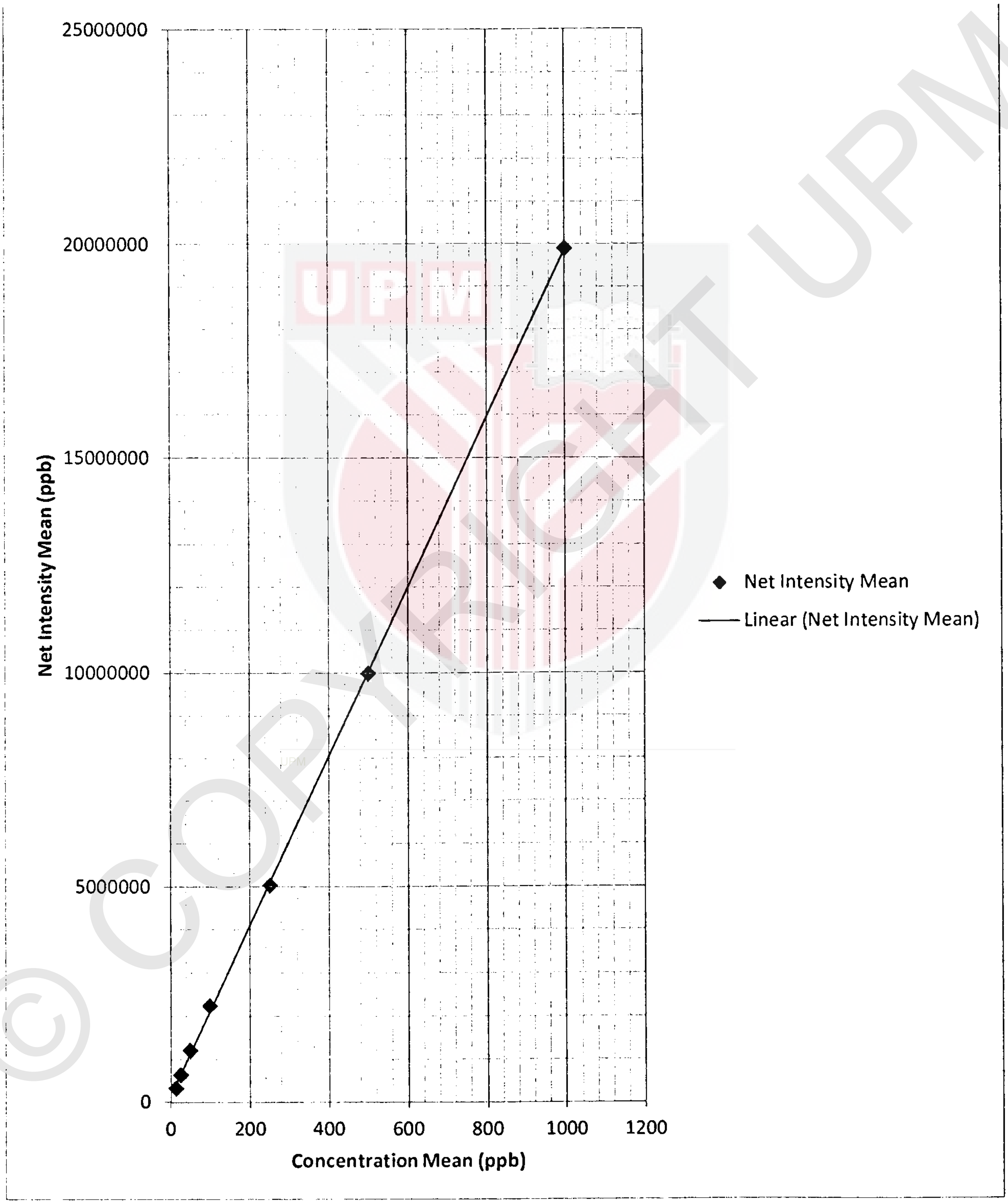
The Calibration Curved for Cu



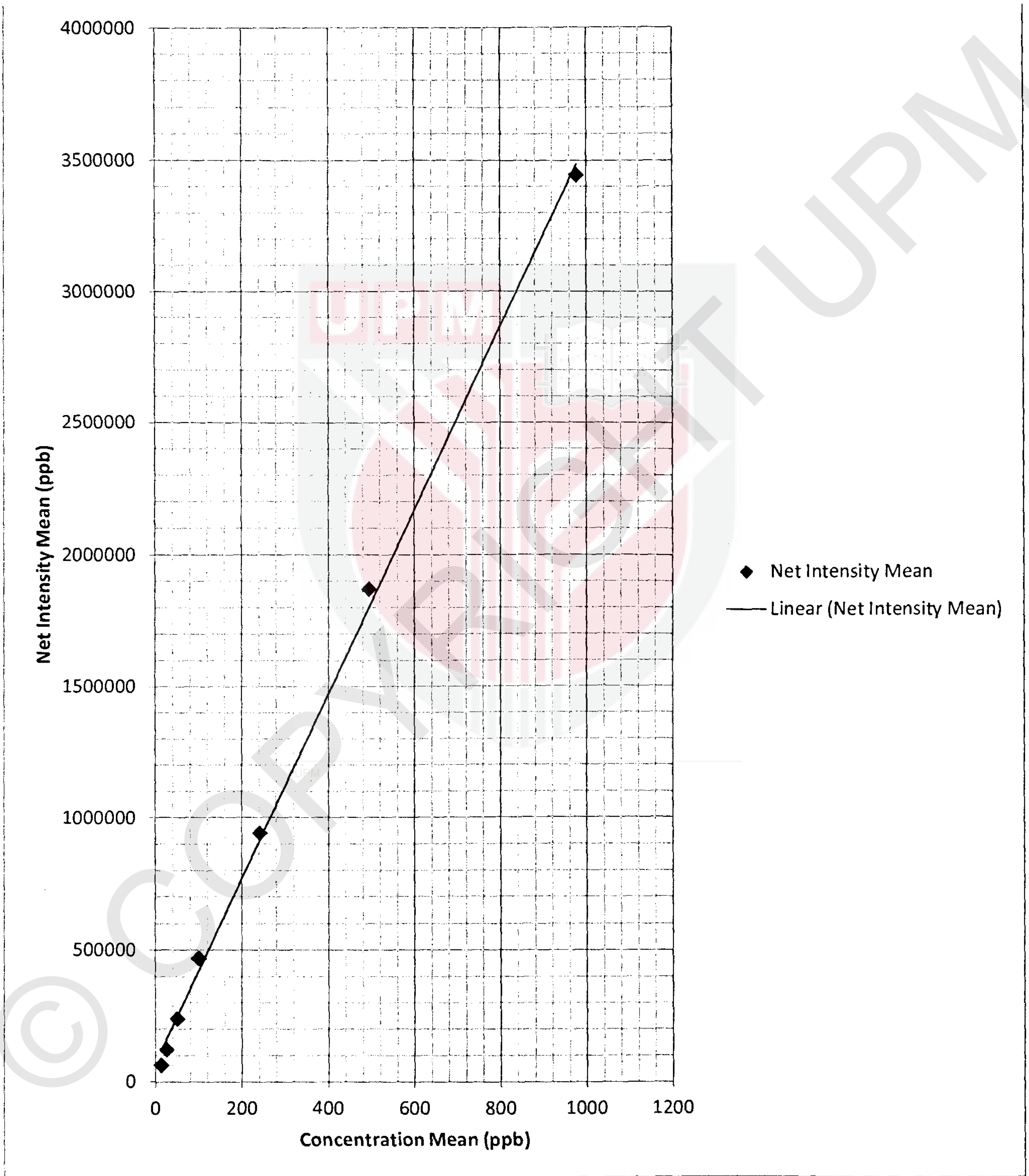
The Calibration Curved for Cd



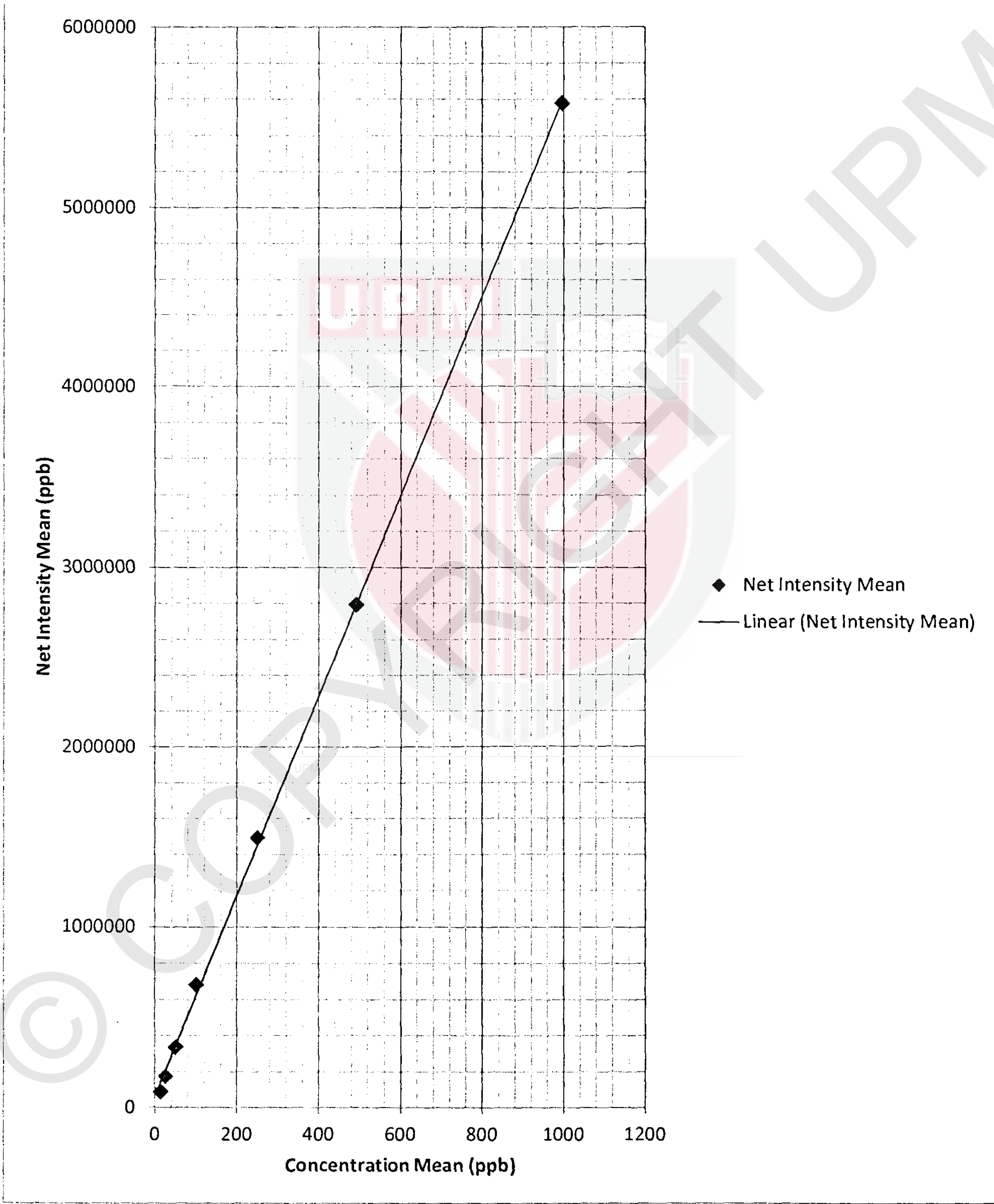
The Calibration Curved for Cr



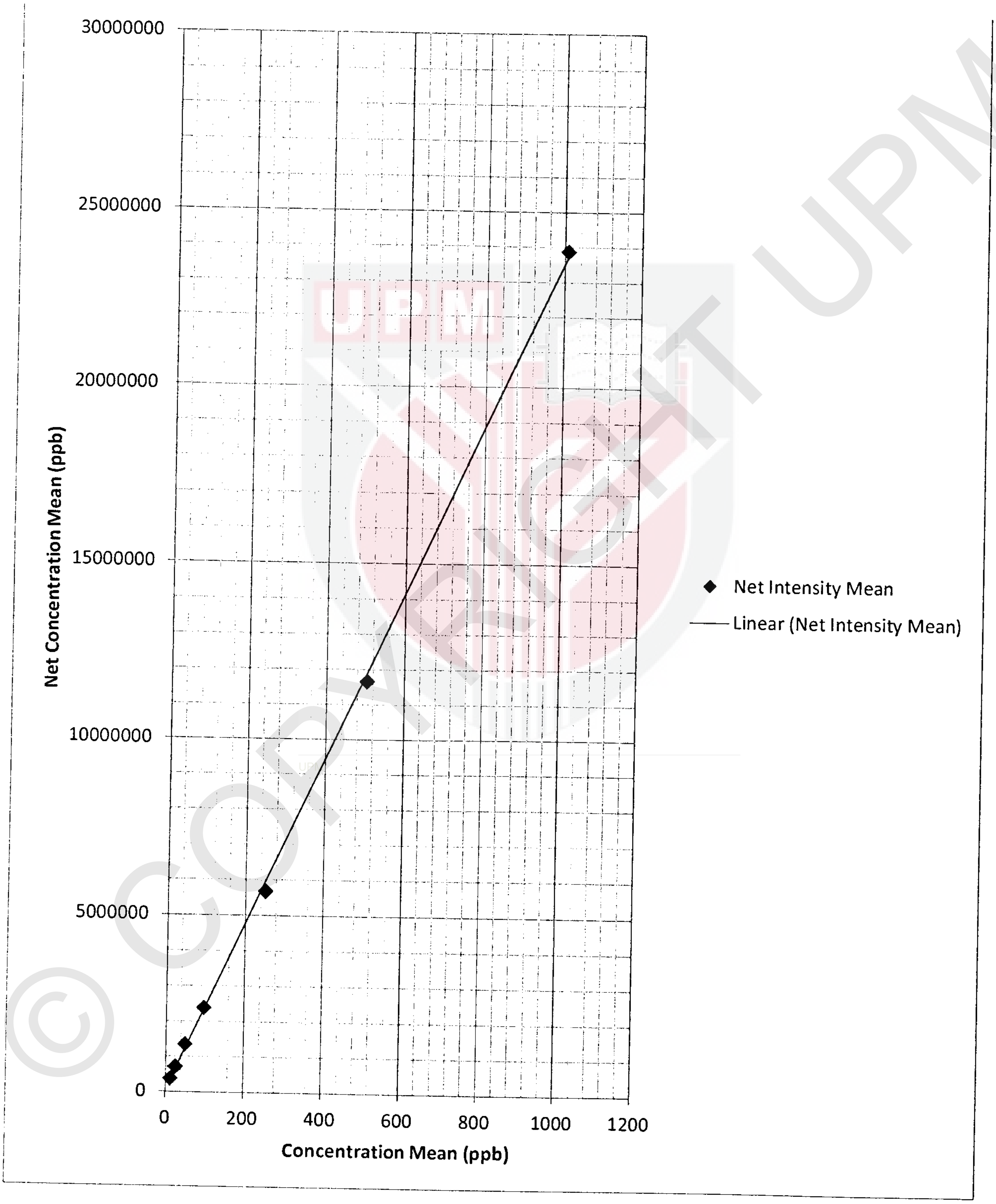
The Calibration Curved for Zn



The Calibration Curved for Ni



The Calibration Curved for Pb



The Calibration Curved for As