**UNIVERSITI PUTRA MALAYSIA**

***BIOAVAILABLE HEAVY METAL CONCENTRATION IN CLASSROOM DUST AND RELATED HEALTH RISK ASSESSMENT OF PRIMARY SCHOOL CHILDREN IN RAWANG, MALAYSIA***

**TAN SOCK YIN**

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# BIOAVAILABLE HEAVY METAL CONCENTRATION IN CLASSROOM DUST AND RELATED HEALTH RISK ASSESSMENT OF PRIMARY SCHOOL CHILDREN IN RAWANG, MALAYSIA

By

**TAN SOCK YIN**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy**

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

**BIOAVAILABLE HEAVY METAL CONCENTRATION IN CLASSROOM DUST AND RELATED HEALTH RISK ASSESSMENT OF PRIMARY SCHOOL CHILDREN IN RAWANG, MALAYSIA**

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**Chairman : Associate Professor Sarva Mangala Praveena, PhD**
**Faculty : Medicine and Health Sciences**

Children are vulnerable to heavy metals in classroom dust. A total of 51 classroom dust samples were collected from children's palms using wet tissue wiping method from April to June 2016. Physiologically Based Extraction Test (PBET) was applied to determine bioavailable heavy metal concentrations and potential health risks among the school children were estimated. The highest mean of bioavailable heavy metal concentration in classroom dust was Zn (12.5103 µg/g), followed by Cu (0.9585 µg/g), Ni (0.5340 µg/g), Cr (0.0472 µg/g), Co (0.0234 µg/g), As (0.0177 µg/g), Cd (0.0096 µg g), and Pb (0.0050 µg/g). Principal Component Analysis (PCA) was used to determine heavy metal sources in classroom dust. Heavy metals in PC1 (As, Cd, Co, Cu, Zn) were mostly linked with natural and anthropogenic sources, while PC2 (Cr, Ni) were more related to anthropogenic activities (industrial activities, traffic congestion). Hierarchical cluster has indicated three clusters, namely Cluster 1 (S3, S4, S6, S15) as residential areas, Cluster 2 (S7, S9, S10, S12) as industrial area and Cluster 3 (S1, S2, S5, S8, S14, S11, S13, S16, S17) as a mixed land use area (residential, industrial, plantation). Emissions from vehicles, plantations and industrial activities were the main heavy metal sources in classroom dust. The relationship between bioavailable heavy metal concentrations in classroom dust with school and classroom characteristics was done using Spearman's Rho. Only Cu ($r$ = 0.767, $p$ = 0.016) was found significant related with distance between school and traffic road, while Cd ($r$ = -0.725, $p$ = 0.027) was found negatively related to classroom floor level. There were no potential health risks (non-carcinogenic and carcinogenic) of ingestion pathway reported.

**Keywords:** heavy metal, classroom dust, children, health risks, carcinogenic, non-carcinogenic

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

# BIOAVAILABILITI LOGAM BERAT DI DALAM HABUK BILIK DARJAH DAN PENILAIAN RISIKO KESIHATAN TERHADAP KANAK-KANAK SEKOLAH RENDAH DI RAWANG, MALAYSIA

Oleh

**TAN SOCK YIN**

**Julai 2020**

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Kanak-kanak mudah terdedah kepada logam berat dalam habuk bilik darjah. Sebanyak 51 sampel habuk bilik darjah telah dikumpul dari tapak tangan kanak-kanak dengan kaedah mengelap pada April hingga Jun 2016. *Physiologically Based Extraction Test (PBET)* telah digunakan untuk menentukan bioavalabiliti logam berat dan potensi risiko kesihatan di kalangan kanak-kanak telah dianggarkan. Purata bioavalabiliti logam berat dalam habuk bilik darjah ditunjukkan dalam susunan berikut: Zn (12.5103 µg / g) > Cu (0.9585 µg / g) > Ni (0.5340 µg / g) > Cr (0.0472 µg / g) > Co (0.0234 µg / g) > As (0.0177 µg / g) > Cd (0.0096 µg g) > Pb (0.0050 µg / g). Analisis Komponen Utama (PCA) digunakan untuk menentukan sumber logam berat dalam habuk bilik darjah. Kebanyakan logam berat dalam PC1 (As, Cd, Co, Cu, Zn) dihubungkan dengan sumber semula jadi dan antropogenik, sementara PC2 (Cr, Ni) lebih berkaitan dengan aktiviti antropogenik (aktiviti industri, kesesakan lalu lintas). Analisis kluster hierarki telah menunjukkan tiga kelompok, iaitu Kluster 1 (S3, S4, S6, S15) diklasifikasi sebagai kawasan perumahan, Kluster 2 (S7, S9, S10, S12) sebagai kawasan perindustrian dan Kluster 3 (S1, S2, S5, S8, S14, S11, S13, S16, S17) sebagai kawasan guna tanah campuran (perumahan, perindustrian, perladangan). Pelepasan dari kenderaan, perladangan dan aktiviti perindustrian didapati sebagai sumber utama kewujudan logam berat dalam habuk bilik darjah. Hubungan antara bioavalabiliti logam berat dalam bilik darjah dengan ciri-ciri sekolah dan bilik darjah telah dijalankan melalui ujian *Spearman's Rho*. Hanya tembaga, Cu ($r$ = 0.767, $p$ = 0.016) didapati mempunyai hubungan signifikan dengan jarak antara sekolah dan jalan raya, sementara kadmium, Cd ($r$ = -0.725, $p$ = 0.027), didapati secara negatif berkaitan dengan tingkat bilik darjah. Tiada potensi risiko kesihatan (bukan karsinogenik dan karsinogenik) dilaporkan dalam kajian ini.

**Kata kunci:** logam berat, habuk bilik darjah, kanak-kanak, risiko kesihatan, karsinogenik, bukan karsinogenik

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This thesis was submitted to the Senate of the Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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| | |
|---|---|
| As | Arsenic |
| BARGE | Bioaccessibility Research Group of Europe |
| Cd | Cadmium |
| Co | Cobalt |
| Cr | Chromium |
| Cu | Copper |
| HI | Hazard Index |
| HQ | Hazard Quotient |
| HRA | Health Risk Assessment |
| ICPMS | Inductively Coupled Plasma Mass Spectrometer |
| IVG | In-vitro Gastrointestinal Method |
| LCR | Lifetime Cancer Risk |
| Ni | Nickel |
| Pb | Lead |
| PBET | Physiologically Based Extraction Test |
| SBET | Simplified Bioavailability Extraction Test |
| TLCR | Total Lifetime Cancer Risk |
| USEPA | United State of Environmental Protection Agency |
| WHO | World Health Organization |
| Zn | Zinc |

# CHAPTER 1

# INTRODUCTION

## 1.1 Background of Study

Rapid urbanisation and areas development have resulted in the increasing of industrial and road traffic activities; which allowed the formation of dust particulate and contributed to air pollution (Wong et al., 2006; Han et al., 2014). Dust can be divided into two types; outdoor dust and indoor dust. Outdoor dust consists of solid particulates (less than 100 µm) which are accumulated on the ground naturally by wind, whereas indoor dust present as a heterogeneous mixture of organic and inorganic particles in different sizes and densities which originated from indoor and outdoor sources (Adekola and Dosumu, 2001). Street dust, traffic dust, road dust and mining dust are the examples of outdoor dust, while classroom dust and household dust are the examples of indoor dust. Studies showed that dust becomes as a carrier of inorganic and organic pollutants such as heavy metals, pesticides, polychlorobiphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) (Al-Rajhi et al., 1996; Al-Khashman, 2004; Maertens et al., 2004; Alahmr et al., 2012; Hassan, 2012; Saeedi et al., 2012; Hussain et al., 2015; Neisi et al., 2016; Wang et al., 2016). Heavy metals are the primary pollutants in dust and the accumulation of heavy metals in dust occurred through sedimentation, impaction and interception from motor vehicles and industrial discharges (Maertens et al., 2004; Lu et al., 2009; Han et al., 2014).

Indoor dust has become a focus because people spend up to 90% time in indoor environments such as homes, schools and offices (Klepeis et al., 2001; Schweizer et al., 2007; Tran et al., 2012; Latif et al., 2014). Besides that, the United States Environmental Protection Agency (USEPA, 2017) has indicated that indoor pollutant level is two to five times higher than outdoor pollutants. The pollutants contained in settled airborne particle in an indoor environment may originate from interior and exterior sources (Rashed, 2008; Turner; 2011). The common anthropogenic sources of heavy metal accumulation in dust are transportation emission, industrial activities, combustion, excessive use of pesticides and fertilizers, smoking, cooking, waste disposal and corrosion of building materials (Maertens et al., 2004; Kurt-Karakus, 2012). Butte et al. (2002) revealed that the adsorption and adherence of suspended pollutants to dust are related to the types and size of particles.

Apart from the pollutants in indoor dust, heavy metals are of more concerns in human health due their high toxicity and non-degradable properties (Chen et al., 2014). A long-term exposure to dust contaminated by heavy metals can pose human health risks, such as renal dysfunction, osteoporosis, bone fractures, dermatitis and lung cancer (Staessen et al., 1999; Jarup, 2003; Verougstraete et al., 2003; Mazinanian et al., 2013). Also, studies showed that heavy metals in dust such as Cr, Cu, Cd, Ni, Pb and Zn cause adverse health effects to human (Al-Rajhi et al., 1996; Vincent, 2005). Hence, health risk assessment is used to determine the human health risk in term of carcinogenic and non-carcinogenic risks.

## 1.2 Problem Statement

Rawang, as an urban area in Malaysia is well known for its high population and industrial activities (Angel et al., 2014; Othman et al., 2014). The annual temperature of Rawang is 32.7 °C, and the average daily temperature during daytime and night are 33.0 °C and 23.3 °C respectively. The driest month in Rawang is June, whereas April is the rainiest month (Praveena, 2018). Up to date, Mohamad Ismail et al. (2018) reported that Rawang has a potential to experience dust pollution due to the release of pollutants from industrial and traffic activities, as high average $PM_{10}$ (119.31 µg/ $m^3$) and $PM_{2.5}$ (35.44 µg/ $m^3$) concentration were reported. Mohamad et al. (2018) also revealed that dust pollution had affected the health of community in Rawang, as 51% of respondents complained having watery and sore eyes and 74% of respondents experienced respiratory symptoms and illnesses (cough, phlegm, sore throat, nasal congestion, breathing difficulties and asthma), but no further heavy metal exposure analysis was done. Besides, Praveena and Aris (2018) and Praveena (2018) had investigated heavy metal exposure on road dust in Rawang, but not in school environment. Therefore, there is a lack of further information on the impact of dust pollution and heavy metal sources in dust in school environment. School environment is where most of the time being spent by children indoors instead of home, thus classroom dust can act as the indicator of heavy metals amount that children are exposed to (Poopola et al. 2012, Latif et al. 2014). Children are more vulnerable to heavy metals because of their higher respiratory rates, hand-to-mouth behaviour or crawling on the floor while playing, which increase the risk of ingesting heavy metals in dust (Tong and Lam, 2000).

Studies on classroom dust have been conducted in the school environment such as in China (Lu et al., 2014), Hong Kong (Tong and Lam, 1998), Mexico (Meza-Figueroa et al., 2007), Nigeria (Poopola et al., 2012; Olujimi et al., 2015), and Malaysia (Tahir et al., 2007; Yap et al., 2011; Darus et al., 2012; Latif et al., 2014; Praveena et al., 2015) which showed that classroom dust is the major pathway of children to be exposed to heavy metal. Heavy metal concentration in classroom dust are varied depends on the land use, dustiness and ventilation system of the classroom (Tong and Lam, 1998; Tahir et al., 2007; Popoola et al., 2012; Latif et al., 2014; Praveena et al., 2015). Contaminated dust can be brought into classroom through wind dispersion and children footprints, which causing dust accumulation in classroom area such as floor, windows, walls, fans, desks, and chairs through settlement (Poopola et al., 2012; Latif et al., 2014). Children can expose to contaminated classroom dust by touching the surface of desks, chairs, walls and windows. Poopola et al. (2012) revealed that heavy metals found in classroom dust were linked to the infiltration of contaminated dust into classroom from outdoor sources. Classroom dust samples collected in area with high road traffic and industrial activities were found higher heavy metal concentration compared to those collected from rural area (Tahir et al., 2007; Darus et al., 2012). Olujimi et al. (2015) also reported that heavy metals concentration in classroom dust were related to the emissions of vehicle and industrial activities. Moreover, classroom with less cleaning frequency and open natural ventilation system (doors, windows) tends to have higher amount of dust accumulated in classroom, thus increasing the risk of children exposed to heavy metals in classroom dust (Tong and Lam, 1998; Darus et al., 2012; Praveena et al., 2015).

In Malaysia, previous studies on classroom dust involved the collection of dust samples from window sills, fans, or corners of classrooms (Tahir et al., 2007; Yap et al., 2011; Darus et al., 2012; Latif et al., 2014; Praveena et al., 2015), which can be misleading regarding the actual amount of heavy metals in classroom dust that come into direct contact with children compared to classroom dust collection from children's palms. Heavy metals in classroom dust can be determined by total heavy metal concentration using strong acid digestion and bioavailable heavy metal concentration using in-vitro digestion model. Although Latif et al. (2014) in Bandar Baru Bangi and Kajang, Malaysia collected dust from children's palms, the determination of the total heavy metal concentration involving the use of acids combination such as *aqua regia* (combination of nitric acid and hydrochloric acid) may over-extract the maximum amount of heavy metal from the dust sample and lead to the overestimation of the actual health risks (Luo et al., 2012; Okoro et al., 2012).

## 1.3 Study Justification

This study intends to be a baseline study of heavy metal concentration focusing on classroom dust in Rawang. The outputs of this study may contribute to the filling of gaps on research concerning heavy metal exposures to children in school environment. By knowing the sources of heavy metal in classroom dust, the risk of children's health may be minimised, by reducing the chance of children approaching to the sources.

Classroom dust samples collected from classroom surroundings such as floors, fans, windowsills, desks, chairs and cupboards will over reflect the actual dust which enter children's bodies. Hence, to obtain a more realistic actual amount of heavy metal exposure to children, classroom dust was collected from children's palms using wet tissue instead of classroom area. This study also focused in determining bioavailable heavy metal concentration in classroom dust as it can show the actual heavy metal exposure in classroom dust toward children. Besides, the school and classroom characteristics (distance between school and traffic road, classroom floor level, number of windows in classroom, number of fans in classroom) also being studied to know whether school and classroom characteristics can cause the differences of bioavailable heavy metal concentration in classroom dust.

Studies conducted in Malaysia (Tahir et al., 2007; Yap et al., 2011; Darus et al., 2012; Latif et al., 2014; Praveena et al., 2015) through the determination of the total heavy metal concentration found high heavy metal concentrations in classroom dust in urban areas. However, studies on obtaining the right heavy metal form in classroom dust to be incorporated with health risks in Malaysia are fairly scarce. Most of the past studies conducted health risk assessment using total heavy metal concentration which potentially over-estimate the health risk. Thus, this study determines heavy metal in classroom dust through in-vitro digestion model approach, known as physiologically based extraction test (PBET) to provide more realistic results in estimating heavy metals daily intake and children's health risks using the health risk assessment calculation by USEPA.

## 1.4 Conceptual Framework

Figure 1.1 shows the conceptual framework of the present study. Dust can be formed in outdoor and indoor environment. Indoor dust such as classroom dust in this study was collected from children's palms using wet tissue wiping. Classroom dust contains heterogeneous compounds such as human skin scalp, heavy metals, organic compounds and microorganism. Heavy metals such as As, Cd, Co, Cu, Cr, Ni, Pb and Zn were determined in this study due to its high toxicity and vulnerability to children. Heavy metals can be identified through bioavailability (using in-vitro digestion model which mimics human gastrointestinal tract) and total heavy metal (using strong acid digestion). The bioavailable heavy metal concentration in this study was done using Physiologically Based Extraction Test (PBET). The confounding factors such as age, weight and height of children were obtained through questionnaires, which the collected data was used in health risk assessment calculation. Health risks (non-carcinogenic risk and carcinogenic risk) for ingestion pathway was calculated using the bioavailable heavy metal concentration data.

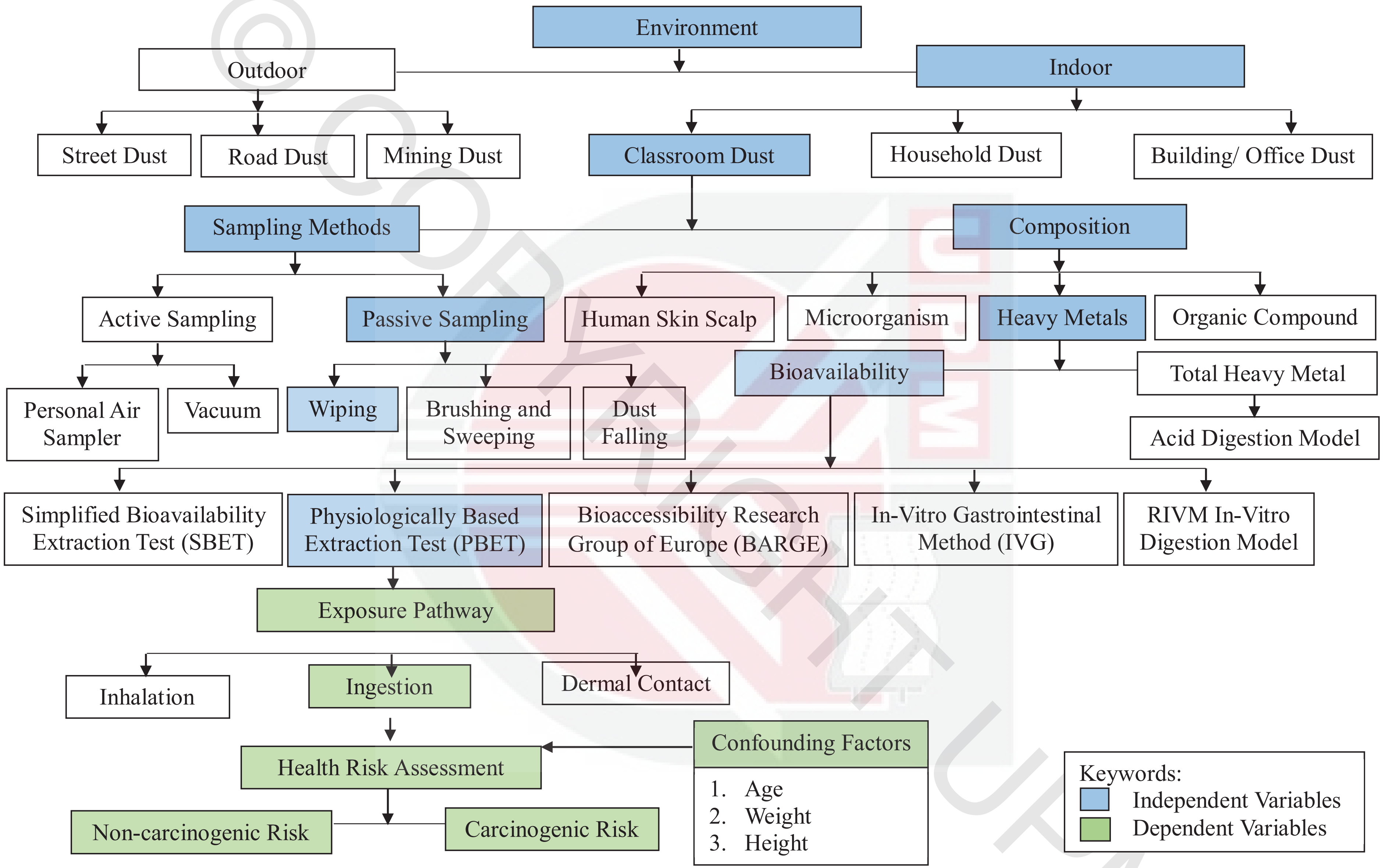


**Figure 1.1 : Conceptual framework**

### 1.5 Research Objectives

#### 1.5.1 General Objective

To assess bioavailable heavy metal concentration, heavy metal sources in classroom dust, relationship with school characteristics and its potential health risks to children (non-carcinogenic and carcinogenic risks).

#### 1.5.2 Specific Objectives

The specific objectives of this research are: -

1. To determine bioavailable heavy metal concentrations (As, Cd, Co, Cu, Cr, Ni, Pb, Zn) in classroom dust collected from children's palms in Rawang.
2. To determine heavy metals sources in classroom dust collected from children's palms.
3. To classify the land use of school location based on bioavailable heavy metal concentrations in classroom dust.
4. To assess the relationship between bioavailable heavy metal concentration in classroom dust with the school and classroom characteristics.
5. To evaluate potential health risks (non-carcinogenic and carcinogenic) of heavy metals in classroom dust that children are exposed to.

### 1.6 Research Questions

1. What is the level of bioavailable heavy metal concentrations in classroom dust collected from children palms?
2. What are the sources of heavy metals in classroom dust collected from children's palms?
3. What is the land use of school location based on bioavailable heavy metal concentrations in classroom dust?
4. What is the relationship between bioavailable heavy metal concentration in classroom dust with school and classroom characteristics?
5. Are there any potential health risks (non-carcinogenic and carcinogenic) of heavy metals in classroom dust toward children?

### 1.7 Research Hypothesis

1. There is a significant relationship between bioavailable heavy metal concentration in classroom dust with school and classroom characteristics (distance between school and traffic road, classroom floor level, number of windows in classroom, number of fans in classroom).

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