

KWAME NKRUMAH UNIVERSITY OF SCIENCE AND TECHNOLOGY

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Exposures and risks of children to some toxic heavy metals in packaged drinking water

By

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# KNUST



## DECLARATION

I hereby declare that this submission is my own work towards the award of Master of Science in Food Quality Management and that, to the best of my knowledge, it contains no material previously published by another person nor material which has been accepted for the award of any other degree of the University, except where due acknowledgement has been made in the text.

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## DEDICATION

This work is dedicated to my late father, Mr Samuel Ofori Abankwah, who always encouraged me to push further in my education and to my daughter, Josephine Kusiwaa Opoku.

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## **ACKNOWLEDGEMENTS**

My source of inspiration and strength, Jesus Christ. All glory and honour for seeing me through to the end of my period of study

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### **ABSTRACT**

The exposure of children from the Kumasi Metropolis, Ghana to Arsenic (As), Cadmium (Cd), Lead (Pb), Mercury (Hg) were estimated in packaged drinking water. The purpose of the study was to estimate the exposure and risk associated with the ingestion of packaged drinking water due to long term exposures. The water samples were obtained from Adum, Bantama, Kejetia and Sofoline markets. Digestion was carried out using the Nitric-Sulphuric acid method. The digestates were then analysed using AAS, to quantify the heavy metals. Monte Carlo Simulation of hazard data was performed using dataset of heavy metals. Regulatory recommended values were used for contact rate of ingestion



of water and body weight of children. Non-carcinogenic risk was evaluated for Cd and Hg. Additionally, carcinogenic risk of As and Pb were also evaluated. The results revealed that oral exposure to the Cd and Hg does not pose a health hazard threat to children who consume packaged drinking water. In dealing with Pb, the results revealed that the oral exposure to Pb in 50% of the packaged water does not pose a health risk; however, 5% of the packaged water samples pose a health risk to children who consume drinking water. Finally, the results for As revealed that oral exposure to As in 95% of the packaged drinking water poses a health risk to children who consume packaged drinking water. Therefore, this suggests that packaged water consumed in the study area may pose a significant health risk to children in relation to the presence of Pb and As.

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## LIST OF ABBREVIATIONS

AWWA	American Public Health Association American Water Works Association
(mg/kg-day) <sup>-1</sup>	milligram per kilogram per day concentration
USEPA	United States Environmental Protection Agency
FAO/WHO	Food and Agriculture Organization/World Health Organization

## CHAPTER ONE

### 1.0 INTRODUCTION

#### 1.1 Background

Water, essential to sustain life, is a natural resource without which man cannot live. A safe supply must be made available to all (WHO, 2017). However, unsafe water exists which contains all sorts of contaminants, such as toxic chemicals which cause unique health effects. Children, who are part of the sensitive group of the population, have the right to access drinking water that is safe (WHO/UNICEF, 2012), since the amount of water they consume in relation to their body weight is high (USEPA, 2002). Studies have reported that they are more susceptible to the effects of toxic metals because full development of most of their organs involved in the removal of toxins has not taken place (Obiri *et al.*, 2010; Cobbina *et al.*, 2013). Therefore, provision of safe drinking water would ensure better health with positive longer term consequences for their lives (WHO, 2017).

Inclusive of the World Health Organization (WHO) list of chemicals of that have drawn much concern in the public domain are toxic heavy metals which include Arsenic (As), Cadmium (Cd) Lead (Pb) and Mercury (WHO, 2017). It is reported that these heavy metals are known to bio-accumulate in the body thus there has been massive public outcry the world over due to their presence in drinking water (Alves *et al.*, 2014; Zhang *et al.*, 2012). Zhang *et al.* (2014) indicated ingestion of these metals have recorded a lot of cases of adverse health effects even though they enter the body through different ways. For example, WHO (2017) reported that in Bangladesh residents were discovered drinking water which was contaminated with As at alarming levels hence this exposure affected the skin by lesions appearing on it. Indeed heavy metals are of concern since they may cause cancer when the exposure takes place after a long time (WHO, 2017). In view of this, safe

drinking water must be free from concentrations of chemical substances that may be highly toxic and can negatively affect health (WHO, 2017).

Heavy metals may enter water systems through two main ways which are geologic or anthropogenic activities (Khan *et al.*, 2013). In Ghana, contamination of natural water body sources with high concentrations of heavy metals have been reported (Asante *et al.*, 2007; Obiri, 2007). Further studies on water sampled from Tinga and Nangodi (two rural communities in the Northern region), Tarkwa and the Obuasi municipality estimated the adverse health effects brought about by continual usage of such contaminated water sources on both adults and children (Cobbina *et al.*, 2013; Bortey-Sam and Nakayama, 2015; Obiri *et al.*, 2010). It was recommended that provision of safer drinking water alternatives would be of immense benefit. Therefore, it is reported that in an attempt by most Ghanaians in urban areas to ensure they have access to an alternative of drinking water which they perceive to be quality and safe, packaged water is highly sought after (Dada, 2011).

## **1.2 Problem Statement and Justification**

In urban areas in Ghana there continues to be an increase of packaged drinking water production. Due to this, most studies on packaged water have focused on assessing its microbiological and physicochemical properties (Ackah *et al.*, 2012; Fisher *et al.*, 2015). Also studies have determined whether the packaged water has certain trace elements such as manganese and calcium in the right quantities (Oyelude and Ahenkorah, 2012). However, not enough studies have been carried out on determining the exposures of toxic metals that may be present in the packaged drinking water and the risks they come along with. According to USEPA (2008) since the organs of children responsible for removing toxins are at the developmental stages and the amount of water they consume in relation

to their body weight is high (USEPA, 2002) their risk value will be higher than adults. Therefore, there is the need to assess whether the toxicity of the metals that may be in packaged drinking water are enough to pose risks to children by appropriately monitoring of the exposure and risks of such heavy metals in drinking water.

### **1.3 Objective**

This study sought to determine the exposure and risks of children to As, Cd, Pb and Hg in packaged drinking water within the Kumasi metropolis.



## **CHAPTER TWO**



## 2.0 LITERATURE REVIEW

### 2.1 Safety and quality of drinking water

The concerns with the provision and accessibility of safe drinking water occurs the world over. Safe drinking water, a basic need and a substance without access to would infringe on a person's human right, should be void of physical, microbial and chemical contaminants (WHO, 2017, ). When ingested over a long period it should also not cause negative effects on a person's health (WHO, 2017). Therefore, Ertuo and Mirza (2005) have reported that judgment of water as safe or not safe should be on the basis of its usage and contact.

Ensuring the quality and safety of water are of the highest standards should be the number one priority (WHO, 2017) due to the immense benefits. Hunter *et al.* (2014) attributed the decrease in the rate of absenteeism by school children in Cambodia Ihsanullah to the provision of drinking water which was safe. In addition, Shuaibu *et al.* (2014) further acknowledged that there was a decrease in children affected by diarrhoea as a result of safe water stored in homes.

Studies have shown that oral intake of contaminated water has a huge negative impact on health (Smith *et al.*, 2010; Ashbolt, 2015; Akhtar *et al.*, 2018). Ntengwe (2003) also reported that the existence of high levels of contaminants in drinking water renders the quality of the water as poor, causing negative health effects to be introduced.

Physical, microbial and chemical parameters of drinking water help to indicate quality and safety of the water (Mustapha, 2008) but a study (Doria, 2010) revealed although guidelines for quality and safe drinking water are present, perceptions of quality water



such as organoleptic properties of the water and information provided by the mass media exist. Doria (2010) also reported that organoleptic perceptions may cause consumers to patronize unwholesome sources of drinking water and desist from aesthetically unappealing but otherwise safe supplies. Kulinkina (2017) reported due to perceptions, some rural communities in Ghana have desisted from using safe but unappealing groundwater boreholes rather opting to use contaminated surface water.

## **2.2 Sources of drinking water**

Water can be obtained from different sources. One source is groundwater. Groundwater is water below the water table and in zones of saturation found below the surface. Springs, well water and borehole water are grouped under sources of groundwater (FAO, 2016). Kortatsi *et al.* (2008) reported that in Ghana, more than 60% of the population depends on groundwater for drinking purposes. In addition, surface water, comprising rivers, canals and low land reservoirs and streams are also sources of water (FAO, 2016). Most rural communities such as in the Northern region of Ghana, source their water from surface water (Cobbina *et al.*, 2015). Finally, rainwater, water collected from the atmosphere whenever it rains (FAO, 2016) is another widely used source of drinking water supply however Kortatsi (1994) indicated it is not relied upon much due its system not being continuous.

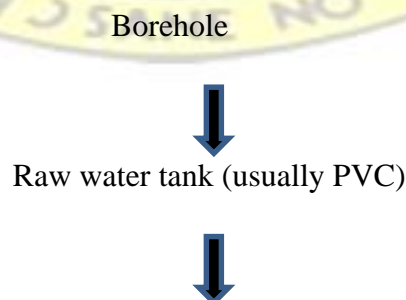
In Ghana, the responsibility of pipe-borne water supply to urban communities rests on Ghana Water Company Limited (GWCL) (Ainuson, 2009), however, a study revealed that while some communities do not receive this supply of water others have pipe-borne systems that are not functioning (Asare *et al.*, 2007). Ngmekpele and Hawkins (2015) also revealed that sometimes the supply from GWCL is interrupted resulting in water shortages. Hence since supply of piped water cannot be trusted, large proportion of people depend

on packaged water for drinking purposes (Dada, 2011; Stoler *et al.*, 2013). Studies (Okioga, 2007 ) in some parts of Ghana have confirmed that most people depend on the plastic bagged form of packaged water as their main source of drinking water.

### 2.2.1 Packaged water

The industries that produce packaged water sold as bottled water or bagged plastic sachets can be seen all over in many countries (Dada, 2011). Its patronage continues to grow in Kumasi and other places in Ghana. Packaged water is water which is for consumption mainly and packaged in different containers such glass, plastic sachets and plastic bottles (WHO, 2017).

The plastic bagged form of packaged water sold as sachets is produced as follows according to Dada (2011) and Stoler *et al.* (2012). A reservoir collects raw water which is then treated with chlorine tablets. Then pumping of the water into an overhead tank through four sets of filters with a pore size of 5µm each takes place. The water flows with pressure through a given area into four sets of filters which have different pore sizes. The water then passes through carbon to stainless steel ultraviolet machine before finally passing through a packaging machine. Large rolls of pre-printed high-density polyethylene (HDPE) are sterilized using UV light as they pass through the sachet machine and normally feature the company name, contact information and regulatory information. The water is finally pumped into the plastic tube and is heat-sealed to create each individual sachet.



Industrial modules (consisting of sand bed filter and activated carbon)



Treated water tank (usually PVC)



Micro-filters ( $5\mu$ - $2\mu$ - $0.5\mu$ )



UV sterilizer (attached to sachet water machine)



Sachets stocked in bigger bags and ready for distribution

**Figure 1: Flow chart of the production of bagged packaged water (Dada, 2011)**

The process involved in the production of bottled packaged water is as follows according to Warburton (1992). Raw water to be processed is also collected in tanks. A known quantity is pumped into the above tank where the water is dozed with alum for coagulation. After coagulation, the water is allowed to settle for an hour. The impurities may be removed by Reverse Osmosis techniques also. The supernatant water is taken to the chlorination tank where primary disinfection is brought about by bubbling chlorine gas. The water is then passed through sand filters for trapping of un-dissolved impurities. The water after sand filtration is passed through Carbon filters for removal of odour, colour and also for de-chlorination. It is then passed through series of micro fillers of various sizes followed by ultraviolet disinfection system for terminal disinfection. Packing is done in PET bottles of 1 litre capacity through an automatic rinsing, filling, and capping machine fitted with an Ozone generator. The bottles are capped and packed in corrugated boxes of one dozen each.

Ahimah and Ofosu (2012) reported that packaged water producers use water mainly obtained from groundwater and pipe-borne water. Therefore, contamination of any sort when found in packaged drinking water may originate from these water sources (Leeuwen, 2000). Contaminants may enter water sources through various ways such as, natural weathering of the earth's surface or through manufacturing processes releases and sewer overflows (Newcomb and Rimstidt, 2002; USEPA, 2017). Obiri (2007) reported that when these effluents are found in aquifers, they affect the groundwater. Also, during distribution, pipes through which the water flows may contaminate the drinking water (WHO, 2017). Therefore, since the processing and nature of the water source also may affect the safety and quality of the drinking water (Ilodigwe *et al.*, 2013) and this may then affect the health of consumers (David *et al.*, 2013), safety assessments should take place to assure consumers that indeed the packaged water is safe.

### **2.3 Risk assessment of drinking water**

According to Gerba (1999) risk assessment is the process of estimating over a period of time the adverse effects occurring due to the human exposure to a hazard. A hazard is an agent, be it biological, chemical or physical, which has the potential to cause adverse effects when an organism, system or (sub) population is exposed to it (FAO, 2009). When both biological and chemical hazards are present in drinking water its safety is questionable however, health concerns of chemical hazards in drinking-water differs from those associated with biological hazards. This is due to the ability of chemical hazards to cause serious health effects after prolonged periods of exposure (WHO, 2017). Obiri *et al.*, (2010) have reported that the presence of chemical hazards such as toxic heavy metals in surface and groundwater is a risk to health based on the risk assessment they carried out on sources of drinking water in the Obuasi municipality. A risk is the probability of an adverse effect in an organism, system or (sub) population caused under specified



circumstances by exposure to an agent (FAO, 2009). Risk assessment consists of hazard identification, hazard characterization (dose-response assessment), exposure assessment and risk characterization (Gerba, 1999).

## **2.4 Hazard identification**

Hazard identification involves the identification of known or potential health effects and its association with a particular biological, chemical or physical agent (Codex Alimentarius Commission, 2013). In this stage the toxicity of chemicals with their effect on the human body are examined (Obiri *et al.*, 2010). Chemicals, such as heavy metals, may be present in packaged water through the filling with contaminated water (Fakhri *et al.*, 2015) or the tap through which raw water is obtained (WHO, 2017).

### **2.4.1 Heavy metals**

Various studies have reported that the heavy metals have no known beneficial health effects rather their toxicity in the body has negative health effects (Guha Mazumder *et al.*, 1998; Smith *et al.*, 1992). Heavy metals which include As, Pb, Cd and Hg are metals that gather together within the (Suresh *et al.*, 2014) body and have toxic effects on humans since the availability of a known homeostasis mechanism is absent (Morais *et al.*, 2012).

Their presence brings about a lot of worry thus have been given a lot of thought of late since damage the liver, kidney, digestive system, and nerve system of humans are some of their trademarks (Zhang *et al.*, 2012). In children the harm they cause is damaging on their health (Vracko *et al.*, 2007) as numerous evidence present has linked toxic metals such as Hg, Pb, As, and Cd to cancers of all sorts and of damage to mental abilities (Obiri *et al.*, 2010).



## **2.4.2 Hazardous effects of heavy metals**

### **2.4.2.1 Arsenic (As)**

As exists in different forms and its contamination of drinking water sources occurs mainly when minerals and ores dissolve naturally. Groundwater is mainly contaminated with inorganic As (WHO, 2017). In the body, studies have reported that its main effects include lowering the defense mechanism of antioxidants and producing oxidative stress which eventually leads to the death of cells (Flora *et al.*, 2008). Its classification as a group 1 agent by the International Agency for Research in to Cancer (IARC) (WHO, 2017) is not surprising since studies have revealed that exposure to As at different levels through drinking-water may lead to cancer development at several organs, especially, lung, skin and bladder. It also causes cardiovascular diseases (Smith *et al.*, 1992). Furthermore, studies have reported that the effects on children who ingest As contaminated water sources include intelligence reduction, development of skin lesions, as well as both renal and neurologic effects (Guha Mazumder *et al.*, 1998 ; Huy *et al.*, 2014).

### **2.4.2.2 Cadmium (Cd)**

Contamination of drinking-water by Cd may be caused by corrosion of galvanized pipes and industrial spillage (WHO, 2011). Studies have shown that when Cd is ingested by humans, as it is transported to the liver through the blood, bonding takes place at the liver. The complexes formed are then transported to the kidneys where they accumulate (Lenntech, 2015). This accumulation eventually destroys the mechanisms responsible for filtering and renders the kidney damaged. Also excretion of essential proteins and sugars from the body occurs. Therefore, excretion of Cd from the human body takes a very long time once it is stored in the kidneys (Lenntech, 2015) since it has a lengthy biological half-life (WHO, 2017). In addition, studies have also shown that Cd interacts with essential nutrients, decreases haemoglobin and deposits in bones (WHO (2017). Although IARC

has placed cadmium and its compounds in Group 2A (probably carcinogenic to humans) it is reported that evidence of carcinogenicity by the oral route does not exist (WHO, 2017). Shuaibu *et al.* (2014) also confirmed this by reporting a correlation between renal failure and the presence of Cd in drinking water.

#### **2.4.2.3 Lead (Pb)**

Pb contaminants drinking water mainly due to its presence in household plumbing systems containing Pb or the service connections to homes (WHO, 2017). Flora *et al.* (2012) reported that Pb is a potent occupational toxin and a non-biodegradable substance. Pb is capable of taking the position of calcium and zinc in body mechanisms and it contributes mainly to neurological dysfunction (Flora *et al.*, 2012). Some of its effects are causing the kidney not to function properly, damage to the nervous system and inducing cardiovascular effects (WHO, 2017). When in children, Pb primarily accumulates in the skeletal system (WHO, 2017).

#### **2.4.2.4 Mercury (Hg)**

Belonging to Group 2A of the IARC classification (WHO, 2017), is present in its inorganic form in surface water and groundwater. When ingested via water its effects include acute oral poisoning which immediately affects the gastrointestinal tract and ultimately damages the kidney due to its accumulation at that organ (WHO, 2017).

### **2.5 Hazard characterization (HC)**

Hazard characterization involves quantifying the adverse health effects associated with biological, chemical and physical agents which may be present (CAC, 2013). It is the second stage after hazard identification has been carried out. Evidence obtained from the

health effects are then used to determine the critical effect, which is the first significant adverse effect observed as the hazards dose is increased (Gerba, 1999).

When dealing with effects due to toxicity, a dose below which adverse effects will not occur known as the threshold is identified. This dose is described as the no-observed adverse-effect level (NOAEL) or no-observed-effect level (NOEL). NOAEL/NOEL can be said to be an estimation of the threshold which exists for that particular chemical when it is causing a specific effect. Usually, the NOAEL /NOEL is used as a point to which references are made when dealing with the risk characterization. However, chemicals and other contaminants are not equivalent in the mode within which they cause adverse effects (Gerba, 1999).

For some effects of toxic chemicals, the body is able to return to its normal state of health from the exposure due to its threshold. These thresholds are represented by the reference dose (RfD) of a substance. There are also other toxic chemicals for which there may be no threshold, therefore repetition of their exposure even at low doses adds up represented by a potency factor (Gerba, 1999).

#### **2.5.1 Reference dose (RfD) and potency factor (PF)**

RfD is the maximum intake of the substance per unit body weight per day (mg/ kg/ day) that is likely to pose no risk to human populations, including children who belong to sensitive groups, due to the natural ability of the body to not only repair itself naturally but also remove toxins (Gerba, 1999).

The potency factor also referred to as the slope factor is the risk produced by the lifetime average dose of 1mg/kg-day. It is usually derived from institutional compendium (Gerba,

1999). The provision of an estimate for increased cancer risk is made possible by the potency factor (Ferguson *et al.*, 2018).

## 2.6 Exposure assessment

Exposure assessment evaluates the amount a particular agent that gets to a population in a specific frequency for a defined duration (Gerba, 1999). Cobbina *et al.* (2013) also reported that it is used for the estimation of the contaminants rate of intake by an organism. Studies have shown that heavy metals may enter the human body when they are inhaled, ingested through food or water and through contact (Ferguson *et al.*, 2018). Ryan *et al.* (2000) confirmed that drinking water is a pathway for exposure to these heavy metals. In addition, hazardous chemicals that occur in drinking-water are of concern because some chemicals produce negative health effects just after a short while of exposure (WHO, 2017). When the concentration of the hazard in drinking water is high the exposure will also be high thus WHO has provided maximum levels of contaminant (MCL) for the hazards in drinking water (WHO, 2017). Exposure to contaminants depends on several factors which allow exposure to be estimated (Čupr, 2015). In exposure assessment, the chronic daily intake (CDI) is determined as it helps to estimate exposure. CDI is amount of the hazard ingested per body weight per day (Gerba, 1999). When determining the CDI by calculation the following parameters are involved,  $Cr$  which is the volume of water ingested per day (L/day),  $C$ , the concentration of hazard in the drinking water (mg/L),  $EF$  is the exposure frequency (days/year),  $ED$  is the exposure duration (years),  $BW$  is the body weight (kg) and  $AT$  is the averaging time (days) (Gerba, 1999).

When dealing with children, Kleiner (1999) reported that the amount of fluid required by children in terms of proportion is higher than adults. Therefore the exposure of the youngest child to toxic substances in drinking water is higher because they consume more water per unit of their body weight (USEPA, 2002) Also, Black *et al.* (2005) reported that



when comparing children to adults, children higher intakes coupled with lower bodyweights of children result in increased dosing rates. The quantity of water children ingest per their body weight is at a maximum in the first month of life and as they become older the quantity they ingest also decreases (USEPA, 2002).

## **2.7 Risk characterization**

Finally, the risk characterization process involves using the information obtained from the hazard identification, hazard characterization and exposure assessment to establish the dangers of specific intakes of individual contaminants (CAC, 2013). Thus, the negative health effects that might result due to exposure to carcinogenic and non-carcinogenic chemicals (Obiri *et al.*, 2007) Health risks classified as non-carcinogenic refers to harm done to the central nervous and other adverse health effects with the exception of cancer, brought about by exposure to neurotoxic chemicals (Cobbina *et al.*, 2013).

### **2.7.1 Hazard Quotient (HQ)**

When dealing with non-carcinogenic health risk, the hazard quotient (HQ) is used to quantify the risks. The HQ is the ratio of the chronic daily intake to the reference dose.

When the value for the HQ is greater than 1.0 it signifies exposure is above non carcinogenic health effects, however a value of 1.0 or less indicates that there is no noncarcinogenic risk as the risk is below levels associated with such non-carcinogenic health effects (USEPA, 2008). According to Obiri *et al.* (2010), ingesting water in the Obuasi Municipality, the results of the non-cancer human health risk for both resident adults and children were also found in most cases to be greater than the USEPA's acceptable noncancer human health hazard index of 1.0. When dealing with multiple hazards in the drinking water, Gerba *et al.* (1999) indicates that hazard index (HI) should



be used. HI is the addition of the different HQ for each substance under assessment in a study (Gerba *et al.*, 1999)

### **2.7.2 Lifetime risk (R)**

The risk for carcinogens is determined as lifetime risk (R). It makes use of the integrated product of the potency factor (PF) and the human exposures as the PF and the corresponding chronic daily intake are multiplied (Gerba, 1999). USEPA (2011) has provided an acceptable cancer risk range of  $1 \times 10^{-4}$  to  $1 \times 10^{-6}$ , which implies the risk ranges from “extremely low” (near 1 in a million chance) to “low” (near 1 in ten thousand) (USEPA, 2011).

## **2.8 Future outlook**

In Ghana, a lot of studies have been carried out on assessing the quality of drinking water with focus being on physicochemical and microbial properties of drinking water and packaged water. Studies have reported the quality of packaged drinking water in different regions and cities of the country. Moreover, studies in Ghana have carried out risk assessment of drinking water sources in mining areas from exposure to heavy metals. In spite of the fact that studies have shown that chemicals are known to have adverse effects from long term exposure as opposed to short term, the toxicity of chemicals in packaged drinking water have not been assessed. This limitation in studies of the determination of the levels of heavy metals in packaged drinking water have resulted in scanty information for stakeholders to make decisions about the toxicity in relation to packaged drinking water. In other countries although studies have been carried out in determining the presence of heavy metals in packaged water, there still remains little information on exposures and risks of the heavy metals in the packaged drinking water especially in relation to children.

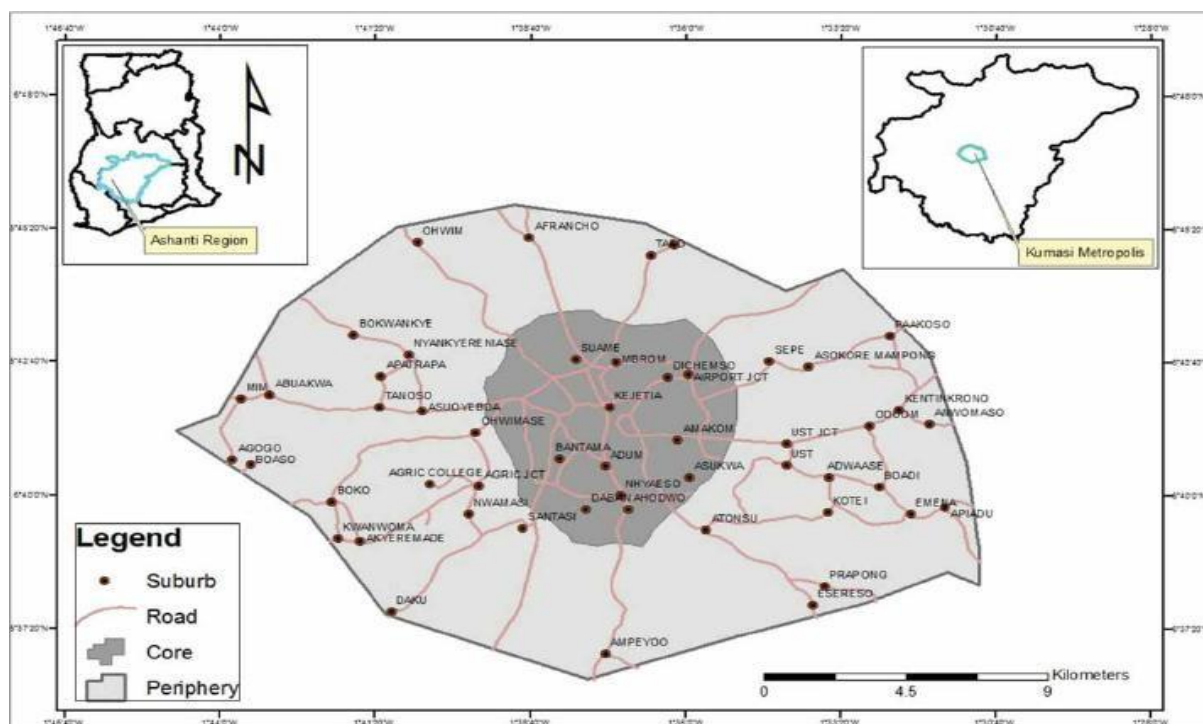
## **CHAPTER THREE**

### **3.0 MATERIALS AND METHODS**

#### **3.1 Materials**

##### **3.1.1 Description of study area**

The study was conducted in the Kumasi Metropolis. It is located within the Ashanti region with a population of over two million thus, making it the most heavily populated district in the Ashanti Region according to the 2010 population census (GSS, 2013). It is an important, commercial, and educational city (GSS, 2014). It is dominated by the middle Precambrian rock with a few small scale mining in the area (GSS, 2014). Water for drinking in the metropolis is mostly obtained from pipe-borne from GWCL (74.1%), boreholes (12.6%) and protected wells (6.4%). Finally, children constitute the largest proportion of the household members (GSS, 2014).



**Figure 2: Map showing the Kumasi metropolis**

### 3.1.2 Sampling of water

Drinking water samples were collected from different markets (Sofoline, Bantama, Adum and Kejetia) in the Kumasi metropolis using simple random sampling. In all thirty different brands of packaged water (sachet and bottled water) were purchased.

## 3.2 Methods

### 3.2.1 Digestion of samples

Packaged water samples were digested using the Nitric-Sulphuric acid method according to Cobbina *et al.* (2013), Obiri *et al.* (2010) and American Water Works Association (AWWA), (1998). For the digestion of Pb, As and Cd, 5 mL each of concentrated nitric acid ( $\text{HNO}_3$ ) and sulphuric acid ( $\text{H}_2\text{SO}_4$ ) were mixed with 100 mL of the water samples. Heating of the mixture took place until there was a reduction in the volume to about 15 to 20 mL, allowing the acids to become concentrated. The digested samples were cooled at room temperature and filtered through filter paper of size  $0.45\mu\text{m}$ . Then the final volume was adjusted to 100 mL with double distilled water and stored for analysis.

For the digestion of Hg, 5 mL of concentrated  $\text{H}_2\text{SO}_4$  was added to 100 mL water sample. Then to the mixture, 2.5 mL of concentrated  $\text{HNO}_3$  was added and after each addition, the contents were thoroughly mixed. 15 mL of 5% w/w potassium permanganate ( $\text{KMnO}_4$ ) was also added to the mixture. The mixture was shaken consistently for at least fifteen minutes until a purple colour persisted. About 8 mL of 5% w/w potassium persulphate ( $\text{K}_2\text{S}_2\text{O}_8$ ) was then added and this solution was heated for two hours on a water bath at  $95^\circ\text{C}$ . It was then cooled and the  $\text{KMnO}_4$  content reduced with the addition of 6 mL of 12% w/v hydroxylamine hydrochloride ( $\text{HONH}_2\cdot\text{HCL}$ ). The digested solution was then stored for analysis.

### 3.2.2 Water Sample Analysis

The analyses of water samples for the levels of As, Cd, Pb and Hg were determined according to Bakirdere *et al.* (2013), Fakhri *et al.* (2015) and Obiri *et al.* (2010) and American Water Works Association (AWWA), (1998). The concentrations of Pb and Cd in the blank and digested water samples were determined using flame AAS Shimadzu model 6401F. In the determination of As in the digested packaged water samples, 5 mL of 0.5% Sodium borohydride ( $\text{NaBH}_4$ ) and 5 mL of 0.5M  $\text{HCl}$  were added to each of the digested water samples to reduce all the As in the samples to arsine gas, in the arsine gas generator, which was coupled to the flame AAS Shimadzu model 6401F. Mercury concentrations in the blank and digested packaged water samples were determined as follows: A carrier solution containing 3% v/v  $\text{HCl}$  and a reducing agent 1.1% m/v  $\text{SnCl}_2$  in 3% v/v  $\text{HCl}$  were automatically sucked into a mixing chamber to reduce the mercury in the +2 state to its elemental state. The mercury vapour generated was directed to the cold vapour cell mounted on the AAS and the mercury concentrations were measured automatically.



Analytical grade reagents were used for all analysis and replicate measurements were done to ensure reproducibility and good quality control. The average concentration of the metal present was obtained in mg/L by the instrument after extrapolation from the standard curve.

### 3.2.4 Data analysis

To estimate the CDI of the hazards in the packaged water the Monte Carlo Palisade @risk software was utilized to obtain the result as a Microsoft add-in (Huy *et al.*, 2014). The concentrations of the chemical hazards (Pb, As, Hg and Cd) in the water as obtained from the AAS analysis were fitted to their respective statistical distributions. The values of the other parameters were taken from USEPA (2014) default values estimating CDI. The values were then used to estimate the CDI based on Equation 1.

$$CDI = \frac{C \times Cr \times ED \times EF}{BW \times AT} \quad (1)$$

Below are the indices used for Equation 1 with their corresponding values used obtained from USEPA (2014):

- CDI is the chronic daily intake (mg/kg/day)
- C is the geometric mean concentration (mg/L) of heavy metal
- Cr is the water intake rate (0.78 L/day for children)
- EF is the exposure frequency (365 days/year)
- ED is the exposure duration (6 years non-carcinogenic, 70 years carcinogenic)
- BW is the average body weight (15 kg)
- AT is the average time (25,550 days, i.e., 70 years  $\times$  365 days/year (carcinogenic) and 2190 days, i.e., 6 years  $\times$  365 days/year (non-carcinogenic))



When dealing with non-cancerous risk, the values for EF, ED and AT were equivalent hence Equation 2 which is a modification of Equation 1 was utilized (Kavcar *et al.*, 2009).

$$CDI = \frac{C \times Cr}{BW} \quad (2)$$

Where CDI is the chronic daily intake (mg/kg/d), C is the drinking water contaminant concentration (mg/L), Cr is the average daily intake rate of drinking water (L/day), and

Bw is the body weight in (kg).

The non-cancer risk, the HQ was estimated using Equation 3 (USEPA, 2008). The oral reference doses used for determining non-cancer health risk for this study due to Cd and Hg exposure in the packaged drinking water were taken from the regional screening level (RSL) generic table released in November, 2017 (USEPA, 2017).

**Table 1: Reference doses of Cd and Hg**

Heavy metal	Reference dose
Cd	$5 \times 10^{-4}$ mg/kg-day
Hg	$3 \times 10^{-4}$ mg/kg-day

Source: USEPA, 2017

For the risk assessment of a mixture of chemicals, the individual HQs are summed to form the hazard index (HI). It is the addition of the different hazard quotients for each substance (chemical) under assessment in a study based on Equation (4) (Gerba *et al.*, 1999).

$$HQ = \frac{CDI}{RfD} \quad (3)$$

$$HI = \sum(HQ_1 + HQ_2) \quad (4) \quad \text{The lifetime}$$

cancer risk resulting from lead and arsenic exposure was estimated using Equation 5 (USEPA, 2008).

$$R = PF \times CDI \quad (5)$$

The values for the potency factors which were used in determining cancer health risk from exposure to Pb and As in this study were obtained from USEPA (2017).

**Table 2: Potency factors for Pb and As**

Heavy metal	Potency factor (mg/kg-day) <sup>-1</sup>
Pb	$8.05 \times 10^{-3}$
As	1.5

Source: USEPA, 2017

The R values were iterated using the Palisade @risk software (Huy *et al.*, 2014) to give lifetime cancer risk curves. For the regression study, the regression option was selected and the regression co-efficient was displayed.

## CHAPTER FOUR

### 4.0 RESULTS AND DISCUSSION

#### 4.1 Hazard exposure in packaged drinking water

The hazards all presented different statistical distributions as shown in Table 3. The results revealed that the hazard with the highest minimum concentration in the water was As ( $1.91 \times 10^{-4}$  mg/L) compared to Pb, Hg and Cd which all followed a similar trend of negligible minimum concentrations. As, also, had the highest maximum concentration of  $1166.00 \times 10^{-4}$  mg/L. In general, As maximum concentration was the highest in the packaged water followed by Cd > Pb > Hg.

**Table 3: Statistical distribution of toxic heavy metals in packaged drinking water**

Hazard ( $\times 10^{-4}$ mg/L)	Statistical distribution	Min	Max	Mode	Mean	5 <sup>th</sup>	50 <sup>th</sup>	95 <sup>th</sup>
Cd	Extvalue(0.0007627,0.00052738)	0	71.54	7.73	10.67	1.84	9.56	23.29
Pb	Extvalue(0.00036437,0.00027718)	0	37.23	3.62	5.24	0.63	4.66	11.88
Hg	Normal(0.00039261,0.0001516)	0	10.52	3.95	3.93	1.43	3.93	6.42
As	Loglogistic(0.00010697,0.0004751,2.2004)	1.09	1166.00	4.13	7.92	2.32	5.82	19.20

It was observed that the 5<sup>th</sup> percentile of the packaged water had As having the highest concentration of  $2.32 \times 10^{-4}$  mg/L while 50% of the packaged water and 95<sup>th</sup> percentile of

the packaged water had Cd having the highest concentrations of  $9.56 \times 10^{-4}$  mg/L and  $23.29 \times 10^{-4}$  mg/L respectively. The presence of heavy metals in packaged drinking water may pertain to the mixed natural and anthropogenic sources infiltrating the water table from which the raw water used in packaged water production is obtained (Khan *et al.* 2013). WHO (2017) has set the safe maximum contaminant levels (MCL) of As, Cd, Pb and Hg in drinking water as follows: As-0.01 mg/L, Cd-0.005 mg/L, Pb- 0.01 mg/L, and Hg-0.006 mg/L. Based on the results all the maximum concentrations of the heavy metals in the packaged water were below the MCL. Studies carried out by Bakirdere *et al.* (2012) and Khaniki *et al.* (2011) also had the levels of heavy metals in their packaged water below the WHO safe limits. In addition, a study on bottled form of packaged water in Accra, Ghana revealed that while some heavy metals were not detected the others were below the MCL as provided by WHO. Levels of heavy metals in packaged drinking water may pertain to the treatment process during the production of the packaged water (Mohamed *et al.*, 1998). The hazards in this study were all within the permissible limits set by WHO, in spite of this due to their detection in the packaged water they are important in the point view of health due to their ability to accumulate and produce negative health effects as a result of their toxicity (Khaniki *et al.* (2011).

#### **4.2 Chronic exposures**

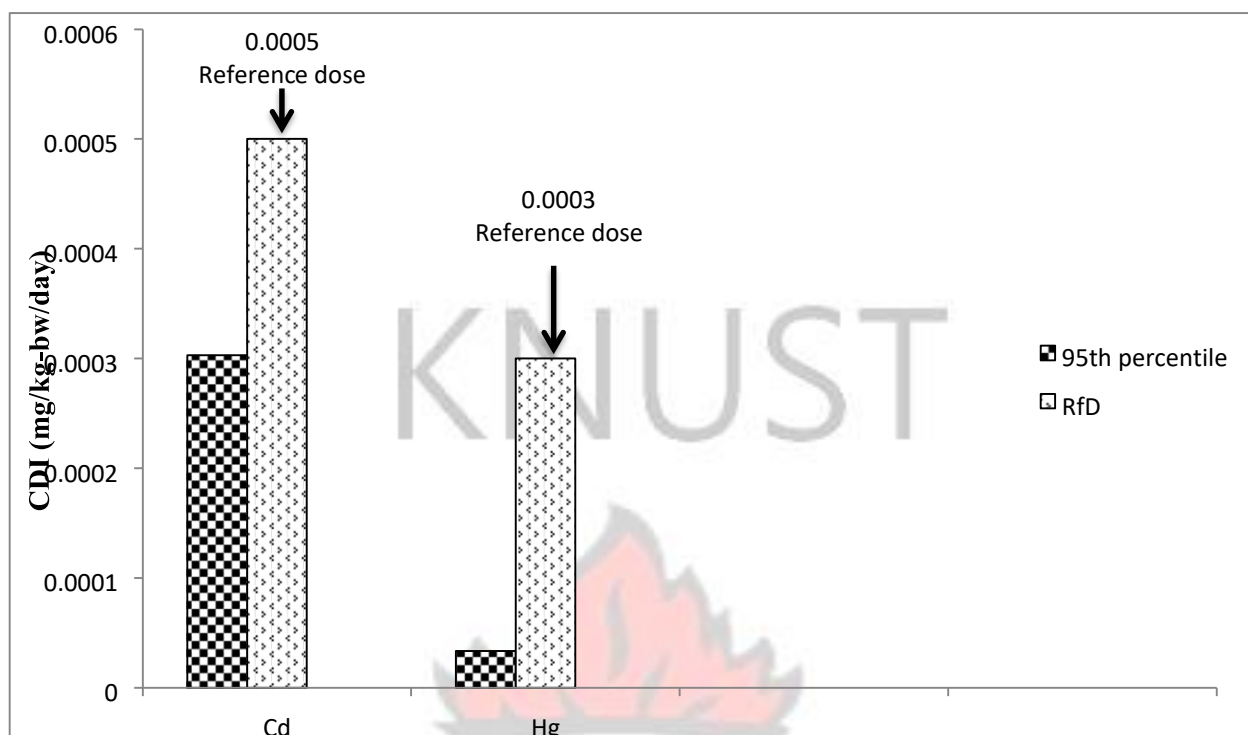
The degree of toxicity of heavy metals to human beings depends upon their daily intakes. Table 4 summarises the estimated CDI values for children (15kg) based on noncarcinogenic and carcinogenic exposures from consumption of drinking water. The quantities of CDI for the hazards under focus are found in the order of  $As > Pb > Cd > Hg$ .



**Table 4: Chronic daily intake of children for non-carcinogenic and carcinogenic risk in packaged water**

Percentile	Non-carcinogenic exposures		Carcinogenic exposures	
	Cadmium mg/kg-bw/day	Mercury mg/kg-bw/day	Lead mg/kg-bw/day	Arsenic mg/kg-bw/day
5 <sup>th</sup>	$2.39 \times 10^{-5}$	$7.45 \times 10^{-6}$	$1.32 \times 10^{-3}$	$5.07 \times 10^{-3}$
50 <sup>th</sup>	$1.24 \times 10^{-4}$	$2.04 \times 10^{-5}$	$1.02 \times 10^{-2}$	$1.27 \times 10^{-2}$
95 <sup>th</sup>	$3.03 \times 10^{-4}$	$3.34 \times 10^{-5}$	$2.59 \times 10^{-2}$	$4.19 \times 10^{-2}$

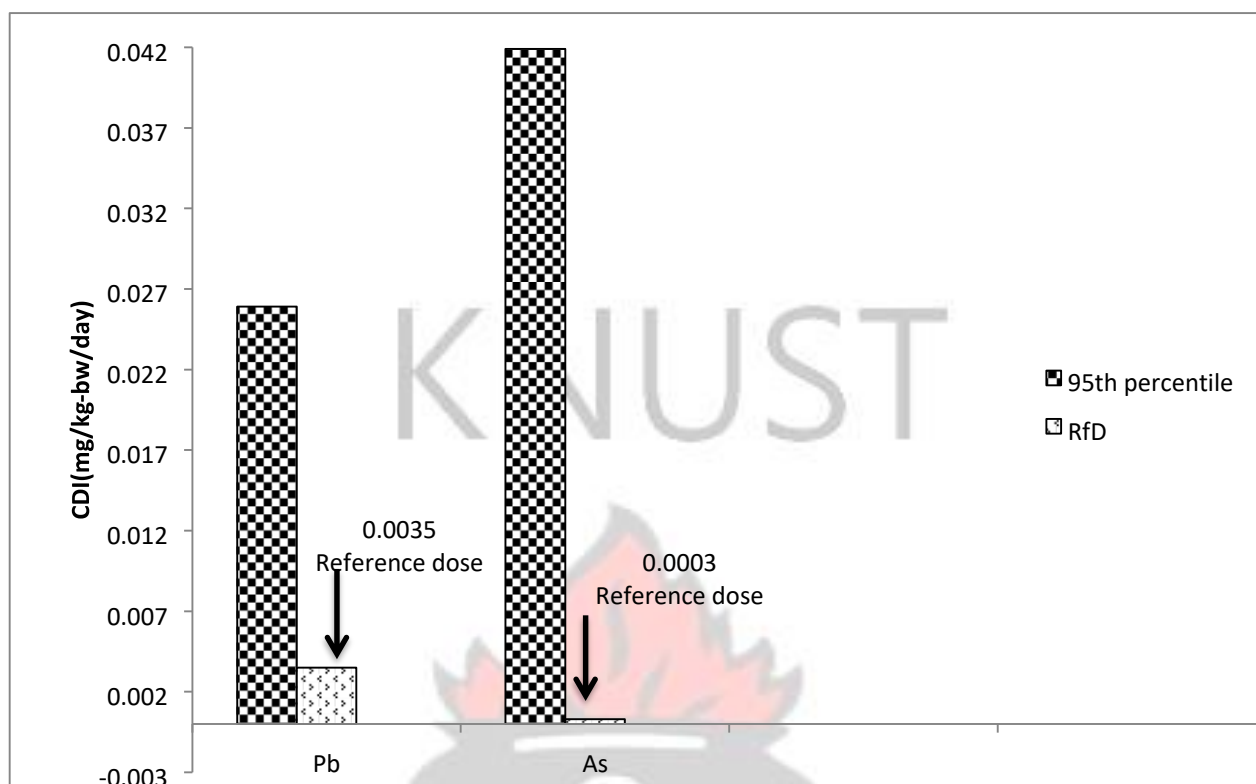
The CDI for Cd and Hg (non-carcinogenic) ranged from  $7.45 \times 10^{-6}$  to  $3.03 \times 10^{-4}$  mg/kg-bw across the day. The 50<sup>th</sup> and 95<sup>th</sup> percentiles' chronic daily non-carcinogenic exposures were in the range of  $2.04 \times 10^{-5}$  to  $3.03 \times 10^{-4}$  mg/kg-bw/day. The CDI for Cd from this study was lower than in a previous CDI study in Swat, Pakistan (Khan *et al.*, 2013) which reported the lowest CDI of children to be  $3.5 \times 10^{-4}$  mg/kg-bw/day. Differences may pertain to the Bw of the child (32.7 kg) used in Swat in contrast to that of the Bw of the child (15 kg) in this study. Fakhri *et al.* (2015) reported a lower CDI for Cd in Minab Iran, than that of the 95<sup>th</sup> percentile of this study. This may also be attributed to the fact that the CDI was in relation to an adult (72 kg) in contrast to this study which was of a child (15 kg). Finally, the 95<sup>th</sup> percentile chronic intakes of Hg ( $3.34 \times 10^{-5}$  mg/kg-bw/day) and Cd ( $3.03 \times 10^{-4}$  mg/kg-bw/day) from this study were lower relative to the RfD of Hg and Cd (Figure 3). Therefore, this implies that chronic intakes of both Hg and Cd in packaged drinking water do not pose a risk to the children.



**Figure 3: CDI (non-carcinogenic exposure) and USEPA oral R<sub>f</sub>D for Cd and Hg**

The cancer exposures ranged from  $1.32 \times 10^{-3}$  to  $4.19 \times 10^{-2}$  mg/kg bw across the day, while the 50<sup>th</sup> and 95<sup>th</sup> percentile chronic daily intakes were in the range of  $1.02 \times 10^{-2}$  to  $4.19 \times 10^2$  mg/kg bw/day. Maigari *et al.* (2016) reported a CDI for Pb (4.9 mg/kg/day) which was lower than the 5<sup>th</sup> percentile CDI of this study. Differences may pertain to levels of Pb in the drinking water of this study

Finally the 95<sup>th</sup> percentile chronic intakes of As and Pb from this study were higher relative to the R<sub>f</sub>D of As and Pb (Figure 4). Therefore, chronic intakes of both As and Pb in packaged drinking water poses a risk to the children. When CDI exceeds R<sub>f</sub>D there is the tendency of intakes to cause harm but this harm cannot be assured and the assurance of no potential harm following intakes that are lower than the UL or R<sub>f</sub>D cannot also be guaranteed (USEPA, 1993).



**Figure 4: CDI (carcinogenic exposure) and USEPA oral RfD for Pb and As**

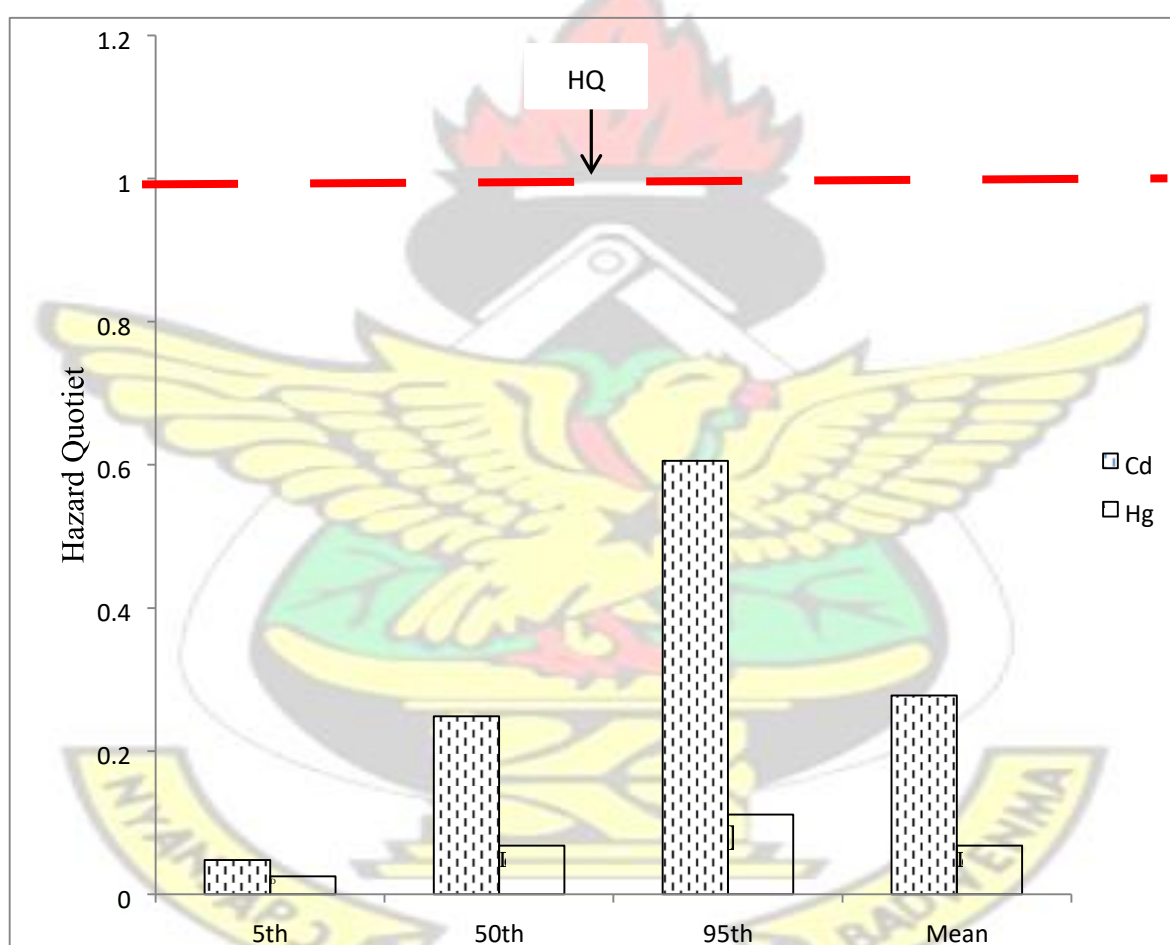
The CDI of the various heavy metals were below their corresponding RfD with the exception As and Pb. This could implicate risk to consumers. Generally, the exposures of children are high because of their low body weight. Hence, the exposure of children to toxic substances in drinking water is high because in relation to their body weight the amount of water they consume is high (USEPA, 2002). Finally, parameters used in estimating CDI are the concentration of chemical hazard and other factors such as body weight and contact rate. Thus it is these factors that are responsible for the variations in exposures to the chemical hazards from this study and other studies.

#### 4.3 Risk characterization

##### 4.3.1 Hazard quotient

The results for non-cancer health risks for children from exposure to Cd and Hg in packaged drinking water have been presented in Figure 5. With respect to Cd exposure,

the 5<sup>th</sup> percentile did not show any risks because the recorded HQ values which were less than 1 across the day. The median (50<sup>th</sup> percentile) presented a trend across the day from the Cd exposure because they also showed HQ values less than 1. Finally, the 95<sup>th</sup> percentile also showed HQ levels less than 1. A similar trend was seen in a study where HQ for children exposed to Cd in drinking water was also less than 1.0 (Bortey-Sam and Nakayama, 2015). Hence children are not likely to experience any health risks as a result of exposure to Cd in packaged drinking water.



**Figure 5: Estimated hazard quotient of Cd and Hg in packaged drinking water**

With respect to Hg exposure, the 50<sup>th</sup> percentile consumers and the 95<sup>th</sup> percentile consumers also recorded HQ values less than 1.0. In a study by reported by Cobbina *et al.*



(2013), children in the Obuasi were ingesting water with the maximum HQ greater than 1. Generally, the HQs of mercury by resident children in Obuasi were greater than 1.0. The differences in HQ for mercury may be attributed to different CDI of the children in both areas and differences in concentration of Hg in the drinking water.

In addition, the additive effect of Cd and Hg in the packaged water indicated an HI less than 1.0. Bortey-Sam and Nakayama (2015) reported that heavy metals present in drinking water in the town of Huniso in Tarkwa had oral HI from their studies that the additive effect of heavy metals and metalloid when taken into consideration, raise concerns about the non-carcinogenic adverse health effects of drinking water in that town. Emenike *et al.* (2017) reported a HI of less than 1 indicating that the ingestion of heavy metals in the packaged water within the study has no potential health risk to the children in the area.

The HQ values of the Hg and Cd in these water samples may be due to their low concentration. Therefore, children in this study showed safe levels of Cd and Hg exposure since the hazard index is less than 1. Therefore, children are not prone to adverse health effects associated with exposure to both Cd and Hg in the packaged drinking water.

#### 4.3.2 Lifetime risk

The results for estimated cancer health risks for children from exposure to Pb and As are shown in Table 3.

**Table 5: Estimated lifetime risk for Pb and As**

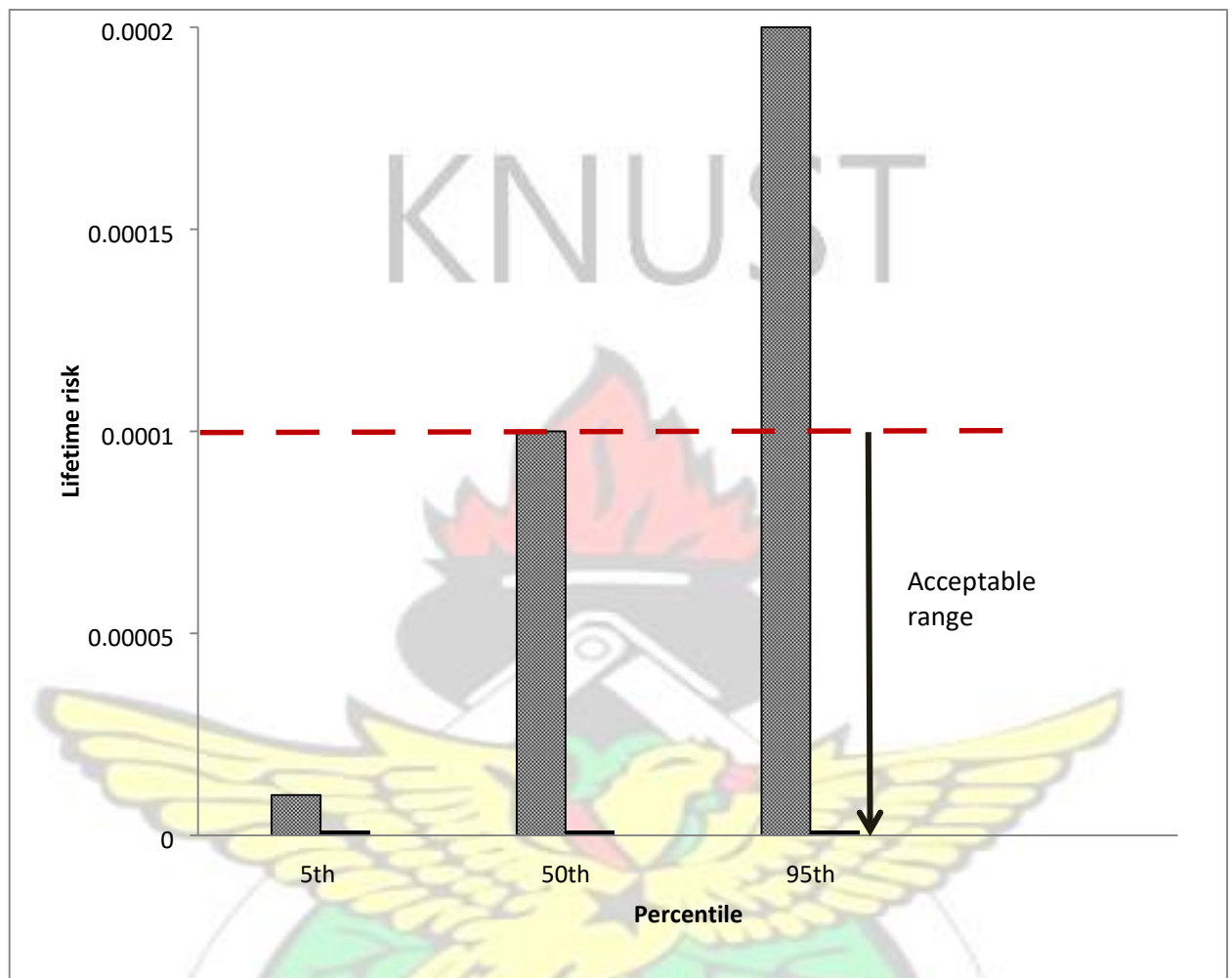
Percentile	Lead	Arsenic
5 <sup>th</sup>	$1.12 \times 10^{-5}$	$7.59 \times 10^{-3}$
50 <sup>th</sup>	$8.65 \times 10^{-5}$	$1.91 \times 10^{-2}$
95 <sup>th</sup>	$2.20 \times 10^{-4}$	$6.28 \times 10^{-2}$

The 5<sup>th</sup>, 50<sup>th</sup> and 95<sup>th</sup> percentile consumers' lifetime risks were in the range of  $1.12 \times 10^{-5}$  to  $2.20 \times 10^{-4}$  with respect to the Pb exposure in the packaged drinking water. The 5<sup>th</sup>, 50<sup>th</sup> and 95<sup>th</sup> percentile consumers recorded cancer risk levels which were higher than the de-minimus ( $10^{-6}$ ). However, the lifetime cancer risk levels for the 5<sup>th</sup> and 50<sup>th</sup> percentile consumers were within the USEPA acceptable range of  $1 \times 10^{-4}$  to  $1 \times 10^{-6}$ , therefore, the risks ranged from “extremely low” to “low” (USEPA, 2011) as shown in Figure 5 with the exception of the 95<sup>th</sup> percentile. The 95<sup>th</sup> percentile packaged water samples are at risk due to exposure of Pb being higher than the acceptable risk.

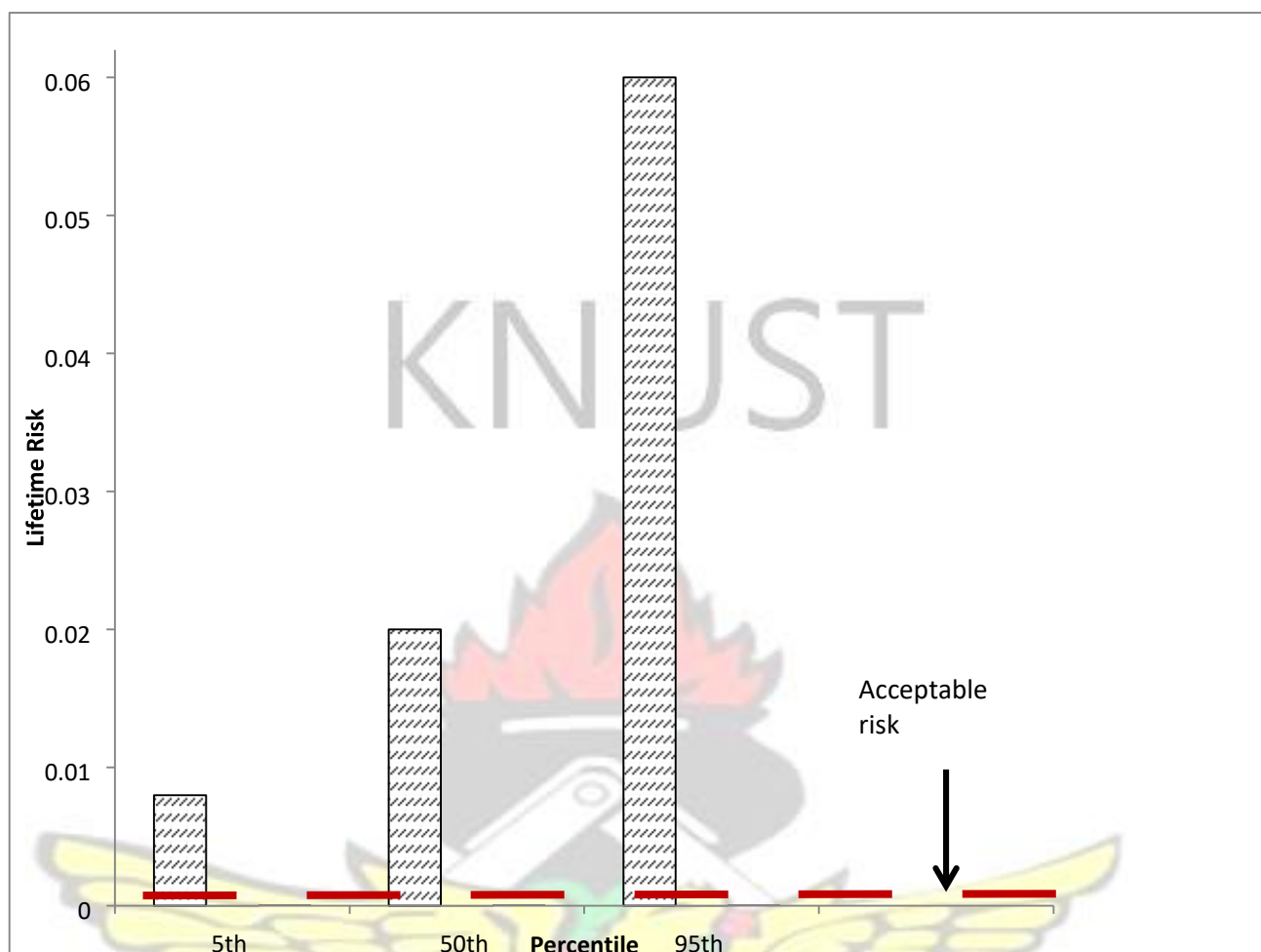
The 5<sup>th</sup>, 50<sup>th</sup> and 95<sup>th</sup> percentile for As in drinking water were in the range of  $7.59 \times 10^{-3}$  to  $6.28 \times 10^{-2}$ . The 5<sup>th</sup> percentile consumers presented cancer risk levels which were higher than the de-minimus ( $10^{-6}$ ) but were not within the USEPA acceptable range of  $1 \times 10^{-4}$  to  $1 \times 10^{-6}$ . As shown in Figure 6. A similar trend was seen in a study conducted by Obiri *et al* (2010) in Obuasi where the children had high cancer risks. In a study carried out in Tarkwa on children ingesting water with heavy metals, the estimated As lifetime cancer risks (Bortey-Sam and Nakayama 2015) were within the USEPA acceptable range. The cancer risk levels of As for children, through drinking water, ranged from  $5.53 \times 10^{-6}$  to  $1.81 \times 10^{-4}$  at certain towns, with the reported exposure to As estimated at  $6.1 \times 10^{-7}$ .

Therefore, since children have low body weight with most of their organs responsible for removing toxins still at the developmental stages, their risk value will be higher than adults and the implication of the risk will be greater (USEPA, 2008). Observations from the regression studies indicate that the variable that contributed mostly to the cancer risk was

the concentration of the toxic metals due. Due to this the concentration of the hazard must be dealt.



**Figure 6: Estimated lifetime Risk for Pb and USEPA acceptable range**



**Figure 5: Estimated lifetime risk for As and USEPA acceptable range CHAPTER FIVE**

## **5.0 CONCLUSION AND RECOMMENDATIONS**

The estimated lifetime risk resulting from exposure to As and the estimated lifetime risk due to Pb (95<sup>th</sup> percentile) exposure in this study, was higher than the acceptable risk range provided by USEPA. This indicates adverse health effects for children who consume packaged water. Also, since the concentration of the hazards in the packaged drinking water contributed the most to the health risk of the children, potential water sources should be properly assessed before boreholes are dug. In addition, there should be continual monitoring of the water sources being used and not just the final product by monitoring agencies. Moreover, modern treatment methods such as the use of technology should be undertaken during the production of packaged water to ensure the absence of heavy metals



in the water. Finally, packaged water producers within the Kumasi metropolis should be made aware, by continuous education, of the toxicity of As and Pb and their dangers.

# KNUST

The logo of KNUST (Kwame Nkrumah University of Science and Technology) is centered in the background. It features a red flame above a black and white open book, which is flanked by two yellow wings. Below the wings is a yellow shield with a black and white design. At the bottom, a yellow banner contains the text 'KNUST' in black letters.

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