

An-Najah National University

Faculty of graduate studies

**Environmental Exposure Assessment of
Cadmium, Lead, Copper and Zinc in
Different Palestinian Canned Food**

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**This Thesis is Submitted in Partial Fulfilment of the Requirements for
the Degree of Master of Public Health, Faculty of Graduate Studies,
An-Najah National University, Nablus, Palestine**

2016

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III

Dedication

To my parents who made the best for me and gave me the opportunity to have this master degree.

To my beloved husband; Ossayd, thank you for encouraging me and for making me strong when I became upset. Thank you for being near to me all the time and by your deep belief in me and my endeavours.

To all my family who supported me in my way through during this Master and thesis.

Acknowledgement

Firstly, I must thank God (Allah) for his graces and blessing on me to complete this research.

I would like to express my sincere thanks and gratitude to my supervisors, Dr. Hamzeh Al Zabadi and Prof. Shehdeh Jodeh for their encouragement and great guidance throughout this study. Their inputs were of great values.

I would like also to offer all respects and appreciation to my instructors in the Public health Master program who afforded their best efforts to enlighten my way in the public health world. Special thanks to my instructors in the chemistry laboratories at An-Najah National University.

الإقرار

أنا الموقع أدناه مقدم الرسالة التي تحمل العنوان:

Environmental Exposure Assessment of Cadmium, Lead, Copper and Zinc in Different Palestinian Canned Food

أقر بأن ما اشتملت عليه هذه الرسالة إنما هي نتاج جهدي الخاص، باستثناء ما تمت الإشارة إليه حيثما ورد، وأن هذه الرسالة ككل، أو أي جزء منها لم يقدم من قبل لنيل أية درجة علمية أو بحث علمي أو بحثي لدى أية مؤسسة تعليمية أو بحثية أخرى.

Declaration

The work provided in this thesis, unless otherwise referenced, is the researcher's own work, and has not been submitted elsewhere for any other degree or qualification.

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التاريخ: 2016 / 4 / 24

Table of Contents

Dedication	III
Acknowledgement	IV
Declaration.....	V
Table of Contents	VI
List of Figures	VIII
List of Tables.....	IX
List of Abbreviations.....	X
Hazardous Substances Data Bank.....	X
Abstract	XI
Chapter One.....	1
Introduction	1
1.1 Background	1
1.2 Significance of the study	2
1.3 Study objectives	3
1.3.1 General objective.....	3
1.3.2 Specific objectives.....	3
Chapter Two	4
Literature Review.....	4
2.1 What is Canning?	4
2.2 Cadmium.....	4
2.3 Lead	5
2.4 Zinc	6
2.5 Copper.....	7
2.6 The Food Industry Sector in the West Bank	10
2.7 Previous Studies	11
Chapter Three	15
Materials and Methods	15

3.1 Sampling	15
3.2 Samples preparation and digestion:	15
3.3 Apparatus	16
3.4 Calibration.....	16
Chapter Four	17
Results	17
4.1 Introduction.....	17
4.2 Samples origin.....	19
4.3 Corn results	20
4.4 Chickpeas results.....	23
4.5 Beans results	26
4.6 Mushroom results.....	29
4.7 Comparison of all canned food types.....	32
5.3 References values for the studied elements.....	33
Chapter Five	35
Discussion.....	35
5.1 Background	35
5.2 Main study findings.....	35
Conclusion:	39
Recommendations:	40
الملخص	ب

List of Figures

No.	Title	Page
Figure 4.1	Calibration Curve of Lead	17
Figure 4.2	Calibration Curve of Cadmium	18
Figure 4.3	Calibration Curve of Zinc	18
Figure 4.4	Calibration Curve of Copper	19
Figure 4.5	Lead concentrations in canned corn types	21
Figure 4.6	Cadmium concentrations in canned corn types	22
Figure 4.7	Zinc concentrations in canned corn types	22
Figure 4.8	Copper concentrations in canned corn types	23
Figure 4.9	Lead concentrations in canned chickpeas types	24
Figure 4.10	Cadmium concentrations in canned chickpeas types	25
Figure 4.11	Zinc concentrations in canned chickpeas types	25
Figure 4.12	Copper concentrations in canned chickpeas types	26
Figure 4.13	Lead concentrations in canned beans types	27
Figure 4.14	Cadmium concentrations in canned beans types	27
Figure 4.15	Zinc concentrations in canned beans types	28
Figure 4.16	Copper concentrations in canned beans types	28
Figure 4.17	Lead concentrations in canned mushroom types	30
Figure 4.18	Cadmium concentrations in canned mushroom types	30
Figure 4.19	Zinc concentrations in canned mushroom types	31
Figure 4.20	Copper concentrations in canned mushroom types	31
Figure 4.21	Average concentrations in the studied canned food	33

List of Tables

No.	Title	Page
Table 2.1	Concentrations of heavy metals in Lebanese food samples	12
Table 2.2	Lead concentrations in Study conducted in Riyadh	13
Table 4.1	Origin of food types used in the study	20
Table 4.2	Concentrations of heavy metals contents in canned corn	21
Table 4.3	Concentrations of heavy metals contents in canned chickpeas	24
Table 4.4	Concentrations of heavy metals contents in canned beans	26
Table 4.5	Concentrations of heavy metals contents in canned Mushroom	29
Table 4.6	Average concentrations of heavy metals in studied canned food types	32

List of Abbreviations

ATSDR	The Agency for Toxic Substances and Disease Registry
CDC	Centers for Disease Control and prevention.
EPA	Environmental Protection Agency.
EU	European Union.
FAO	Food and Agriculture Organization.
HSDB	Hazardous Substances Data Bank
HACCP	Hazard Analysis & Critical Control Points.
IQ	Intelligence Quotient.
IARC	International Agency for Research on Cancer.
ICP-AES	Inductively coupled plasma atomic emission spectroscopy.
KSA	Kingdom of Saudi Arabia.
NAS	National Academy of Sciences.
ND	Not Detected
PPM	Parts Per Million (mg/l), (mg/kg).
SD	Standard Deviation.
UAE	United Arab Emirates.

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Abstract

Many chemical elements that present in the human diet are essential for human life at low concentrations but can be toxic at high concentrations and at chronic exposure. The aim of this study was to determine the levels of heavy metals in some canned food that are sold in the Palestinian market and compare them with the recommended international levels.

The contents of investigated metals in canned food in this study were found to be in the range of (0.089-1.17 mg/L) for Pb, (0.019-0.32 mg/L) for Cd, (2.05-10.6 mg/L) for Zn and (0.79-3.97 mg/L) for Cu. Cadmium and Copper results were higher than international permissible levels. Whereas Lead and Zinc levels were within the permissible levels. The results necessitate continuous monitoring of Cd and Cu levels and controlling of canning process in canned food to obtain food safety.

This study demonstrates that Cd and Cu levels were higher than international permissible levels and this is an important alarm in public health. We recommend further future studies that asses the levels of the studied elements in human body and selecting samples from hair ,nails and blood.

Chapter One

Introduction

1.1 Background

One of the most important environmental health issue facing the world is contamination of the environment by inorganic, organic, and organo-metallic materials. There has been growing interest in the monitoring of heavy metals in the bio-organs ^{[1] [2]}. The main source of exposure of humans to heavy metals is through ingestion of food. Many chemical elements that present in the human diet are essential for human life at low concentrations but can be toxic at high concentrations and at chronic exposure ^{[3] [4]}.

Lead and Cadmium are from the prevalent toxic elements in food and environment that have a long half-life after absorption. In humans and animals; they can make unpleasant effects such as damage to internal organs. Cadmium accumulates in the human body and especially in the kidneys which could lead to dysfunction of the kidney with impaired re-absorption of some molecules such as: proteins, glucose, and amino acids^[5]. Exposure to lead affects multiple health outcomes and physiological systems, including: hypertension, the gastrointestinal system, anemia, nephropathy, vitamin D metabolism, decreased growth, the immune system, the nervous system, behavioral/cognitive/IQ, nerve conductive effects, hearing loss, effects on reproduction and development and death from encephalopathy ^[6].

Trace heavy metals are significant in nutrition, either for their essential nature or their toxicity. Copper and Zinc are known to be essential and may enter the food materials from soil through mineralization by crops or environmental contamination with metal-based pesticides. The adult human body contains about 1.5 to 2 ppm (parts per million) of Copper ^[7] and 33ppm of Zinc ^[8]. Excessive intake of either Copper or Zinc has been reported to be toxic ^[9].

Heavy metal ions are essential micronutrients for plant metabolism but when present in excess, can become extremely toxic. Heavy metals are dangerous because they tend to bio-accumulate. Bioaccumulation means an increase in the concentration of a chemical in a biological organism over time, compared to the chemical's concentration in the environment ^[10]. Copper and Zinc are essential micronutrients if consumed in adequate amounts, but they might become toxic when consumed excessively. In contrast, Cadmium (Cd) and Lead (Pb) are toxic metals and are harmful at low concentration and are not easily biodegradable^[11]. Therefore, it is important to assess the concentrations of these metals in the human food chain as environmental exposure assessment technique in order to evaluate their actual concentrations in the environmental medium (food) and whether these concentrations perform dangerous effects on the consumers when compared to the reference permissible values.

1.2 Significance of the study

Canned food is popular food sources in Palestine because they are inexpensive and affordable. There are no studies conducted in the

Palestinian market regarding the determination of heavy metals Pb, Cd and trace metals Cu, Zn in canned food. This study will shed the light to any exposure levels to these elements in our diet and compare it to the permissible exposure limits. This would further suggest a preventive measures and alternatives to these food sources with the policy and decision makers at the national level.

1.3 Study objectives

1.3.1 General objective

To determine the concentrations of heavy metals (Cd, Pb) and trace elements (Cu, Zn) in the food chains in the Palestinian market.

1.3.2 Specific objectives

1. To determine the concentrations of the certain well-known toxic heavy metals (Cadmium and Lead) and trace elements (Cu ,Zn) in four food species (canned beans, canned chickpeas, canned corn and canned mushroom) in the Palestinian market.
2. To compare the concentrations of the different four elements with the different companies of the same product.
3. To compare the measured values of these four elements with the permissible exposure limits stipulated by international health agencies.

Chapter Two

Literature Review

2.1 What is Canning?

Canning is a method of preserving food by first sealing it in air-tight jars, cans or pouches, and then heating it to a temperature that destroys contaminating microorganisms that can either be of health or spoilage concern because of the danger posed by several spore-forming thermo-resistant microorganisms. The process of canning is sometimes called sterilization because the heat treatment of the food eliminates all microorganisms that can spoil the food and those that are harmful to humans, including directly pathogenic bacteria and those that produce lethal toxins^[12].

2.2 Cadmium

Cadmium has been widely dispersed into the environment through the air by its mining and smelting and by other man-made routes such as: usage of phosphate fertilizers, presence in sewage sludge, and various industrial uses such as the Nickel–Cadmium (Ni-Cd) batteries, plating, pigments and plastics. Then Cadmium compounds can be associated with respirable-sized airborne particles and can be carried long distances. Once on the ground, Cadmium moves easily through soil layers and is taken up into the food chain by plants' uptake such as leafy vegetables, root crops, cereals and grains^[13].

The total Cadmium body burden at birth is non-detectable .It gradually increases with age to about 9.5 mg to 50 mg ^[13].The kidneys and liver together contain about 50% of the body's accumulation of Cadmium ^[35].

The biological half-life of Cadmium in the kidney is estimated to be between 6 to 38 years; the half-life of Cadmium in the liver is between 4 and 19 years .These long half-lives reflect the hypothesis that humans do not have effective pathways for Cadmium elimination. Cadmium has no known biologic function in humans. Bioaccumulation appears to be a by-product of increasing industrialization. Any excessive accumulation in the body should be regarded as potentially toxic. Due to slow excretion, Cadmium accumulates in the body over a lifetime and its biologic half -life may be up to 38 years ^[13].

Cadmium is known to increase the oxidative stress by being a catalyst in the formation of reactive oxygen species, increasing lipid peroxidation and depleting glutathione and protein-bound sulfhydryl groups. It also can stimulate the production of inflammatory cytokines and down regulates the protective function of nitric oxide formation ^[13].

2.3 Lead

Lead occurs naturally in the environment. However, most of the high levels found throughout the environment come from human activities. Environmental levels of Lead have increased more than 1,000-fold over the past three centuries as a result of human activity. The greatest increase occurred between the years 1950 and 2000, and reflected the increasing worldwide use of Leaded gasoline. Lead can enter the environment through

releases from mining Lead and other metals, and from factories that make or use lead, lead alloys, or Lead compounds.

The amount of lead found in canned foods decreased 87% from 1980 to 1988 in the United States, which indicates that the chance of exposure to Lead in canned food from Lead-Soldered containers has been greatly reduced. However, Lead-Soldered cans are still used in some other nations. The effects of Lead are the same whether it enters the body through breathing or swallowing. The main target for Lead toxicity is the nervous system, both in adults and children. Long-term exposure of adults to Lead at work has resulted in decreased performance in some tests that measure functions of the nervous system^[31]. Lead exposure may also cause weakness in fingers, wrists, or ankles. Lead exposure also causes small increases in blood pressure, particularly in middle-aged and older people. Lead exposure may also cause anemia. At high levels of exposure, Lead can severely damage the brain and kidneys in adults or children and ultimately cause death. In pregnant women, high levels of exposure to Lead may cause miscarriage. High level exposure in men can damage the organs responsible for sperm production ^[14].

2.4 Zinc

Zinc is an essential element needed by our body in small amounts. We are exposed to Zinc compounds in food. The average daily Zinc intake through the diet ranges from 5.2 to 16.2 milligrams. Food may contain levels of Zinc ranging from approximately 2 parts of Zinc per million (2 ppm) parts of foods (e.g., leafy vegetables) to 29 ppm (meats, fish, poultry)^[15].

The National Academy of Sciences (NAS) estimates the Recommended Dietary Allowances (RDA) for Zinc of 11 mg/day (men). Eleven mg/day is the same as 0.16 mg per kilogram (kg) of body weight per day for an average adult male (70 kg). An RDA of 8 mg/day, or 0.13 mg per kg of body weight for an average adult female (60 kg), was established for women because they usually weigh less than men. Lower Zinc intake was recommended for infants (2-3 mg/day) and children (5-9 mg/day) because of their lower average body weights. The RDA provides a level of adequate nutritional status for most of the population. Extra dietary levels of Zinc are recommended for women during pregnancy and lactation. An RDA of 11-12 mg/day was set for pregnant women. Women who nurse their babies need 12-13 mg/day^[15].

Prolonged oral exposure to Zinc has been shown to decrease the absorption of Copper from the diet, resulting in the development of Copper deficiency. At low doses (~0.7–0.9 mg Zinc/kg/day) and intermediate exposure durations (6–13 weeks), the effect is minor and manifests as subclinical changes in copper-sensitive enzymes, such as superoxide dismutase. At higher exposure levels (~2 mg Zinc/kg/day) for chronic duration, more severe symptoms of Copper deficiency, including anemia, have been reported^[16].

2.5 Copper

Copper can be found in plants and animals, and at high concentrations in filter feeders such as mussels and oysters. Copper is also found in a range

of concentrations in many foods and beverages that we eat and drink, including drinking water ^[32].

Copper is readily absorbed from the stomach and small intestine. After nutritional requirements are met, there are several mechanisms that prevent Copper overload. Excess Copper absorbed into gastrointestinal mucosal cells induces the synthesis of and binds to the metal binding protein metallothionein. This bound Copper is excreted when the cell is sloughed off. Copper that eludes binding to intestinal metallothionein is transported to the liver. It is stored in the liver bound to liver metallothionein, from which it is ultimately released into bile and excreted in the feces ^[33]. Although Copper homeostasis plays an important role in the prevention of Copper toxicity, exposure to excessive levels of Copper can result in a number of adverse health effects including liver and kidney damage, anemia, immunotoxicity, and developmental toxicity. Many of these effects are consistent with oxidative damage to membranes or macromolecules. Copper can bind to the sulfhydryl groups of several enzymes, such as glucose-6-phosphatase and glutathione reductase, thus interfering with their protection of cells from free radical damage. One of the most commonly reported adverse health effect of Copper is gastrointestinal distress. Nausea, vomiting, and/or abdominal pain have been reported, usually occurring shortly after drinking a Copper Sulfate solution, beverages that were stored in a Copper or untinned brass container, or first draw water (water that sat in the pipe overnight). The observed effects are not usually persistent and gastrointestinal effects have not been linked with other health effects.

Animal studies had also reported gastrointestinal effects (hyperplasia of forestomach mucosa) following ingestion of Copper Sulfate in the diet. Copper is also irritating to the respiratory tract. Coughing, sneezing, runny nose, pulmonary fibrosis, and increased vascularity of the nasal mucosa have been reported in workers exposed to Copper dust ^[17].

The liver is also a sensitive target of toxicity. Liver damage (necrosis, fibrosis, abnormal biomarkers of liver damage) have been reported in individuals ingesting lethal doses of Copper sulfate. Liver effects have also been observed in individuals diagnosed with Wilson's disease, Indian childhood cirrhosis, or idiopathic copper toxicosis (which includes Tyrollean infantile cirrhosis). These syndromes are genetic disorders that result in an accumulation of Copper in the liver; the latter two syndromes are associated with excessive copper exposure. Inflammation, necrosis, and altered serum markers of liver damage have been observed in rats fed diets with Copper Sulfate levels that are at least 100 times higher than the nutritional requirement. Damage to the proximal convoluted tubules of the kidney has also been observed in rats. The liver and kidney effects usually occur at similar dose levels; however, the latency period for the kidney effects is longer than for the liver effects. There is some evidence from animal studies to suggest that exposure to airborne Copper or high levels of Copper in drinking water can damage the immune system. Impaired cell-mediated and humoral-mediated immune function have been observed in mice. Studies in rats, mice, and mink suggest that exposure to high levels of Copper in the diet can result in decreased embryo and fetal growth. The

carcinogenicity of copper has not been adequately studied. An increase in cancer risk has been found among Copper smelters; however, the increased risk has been attributed to concomitant exposure to arsenic. Increased lung and stomach cancer risks have also been found in Copper miners. However, a high occurrence of smoking and exposure to radioactivity, silica, iron, and arsenic obscure the association of Copper exposure with carcinogenesis^[34]. Animal studies have not found increased cancer risks in orally exposed rats or mice. The International Agency for Research on Cancer(IARC) has classified the pesticide, Copper 8-hydroxyquinoline, in Group 3, unclassifiable as to carcinogenicity in humans and Environmental Protection Agency (EPA) has classified Copper in Group D, not classifiable as to human carcinogenicity ^[17].

2.6 The Food Industry Sector in the West Bank

The food processing industry is considered one of the oldest industries in Palestine. In its early days, this industry was limited to the production of few processed foods and sweets. Currently, this industry is a major contributor to the Palestinian economy and its gross domestic product. In addition, this sector has created job opportunities in the local market ^[18].

According to the Palestinian Food Industries Union, the following are the most important food processing industries in the West Bank, related to agro products targeted in the project ^[19]:

- **Canned vegetables and fruits:** In Palestine, there are 18 factories specialized in the production of canned vegetables and fruits.

- **Oils and vegetable fats:** There are 13 factories specialized in the production of oils. Within this industry, 3 factories have received the ISO 22000 certification, while 10 olive mills have received the Hazard Analysis & Critical Control Points (HACCP) certification.
- **Wheat flour and grains:** There are 9 factories working within this industry.
- **Pasta and vermicelli:** There are 5 factories working in the production of pasta and vermicelli.

2.7 Previous Studies

A study conducted in Saudi Arabia to measure the concentration of 27 elements of mineral and toxic heavy metals in fresh and canned food, it included Cu, Zn and Pb. The results showed that the mean ranges of the elements analyzed in (mg kg^{-1}) between the fresh and canned food (Fresh food - canned food) are as follows: Iron (34.35 – 164.1 mg/kg), Aluminium (6.63 – 41.14 mg/kg), manganese (11.73 – 17.95 mg/kg), Lead (2.31 – 7.11 mg/kg), Zinc (24.14 – 26.76 mg/kg), Copper (6.22 – 8.03 mg/kg), Ca (1611 – 8557 mg/kg), Magnesium (1669 – 1206 mg/kg), sodium (9918 – 23787 mg/kg), respectively. Some of the measured values were found, not only relatively high in canned compared to fresh food samples, but also exceeds the international tolerance levels^[20].

Another study conducted in Lebanese market to assess heavy metals in canned food. Lead had the highest levels in corn and fava beans. Thirty percent of vegetables and legumes and 45% of fish samples had Cd levels

above European Union (EU) permissible level (0.1 $\mu\text{g/g}$), The concentrations of elements in food samples are shown in Table 1 [21].

Table 2.1: Concentrations ($\mu\text{g/g}$)of heavy metals in Lebanese food samples⁽²¹⁾

Food type	Zinc	Copper Mean	Lead	Cadmium
Canned fish	8.43	0.65	0.065	0.021
Tuna	6.57	0.49	0.062	0.26
Sardine	11.4	0.92	0.071	0.021
Meat	15	0.97	0.007	0.025
Luncheon	15.2	0.66	0.01	0.025
meat	12.75	0.70	0.005	0.024
Hot dog	19.5	2.3	0.002	0.025
Liver paste				
Vegetable and legumes	7.42	1.63	0.057	0.003
Vegetables	6.72	1.28	0.058	0.003
Legumes	8.5	2.14	0.056	0.075
Fava Beans	ND*	ND*	-	0.483
Chickpeas	ND*	ND*	-	0.125
Mushroom	ND*	ND*	-	0.185
Permissible levels	11	0.9	1	0.1

*ND: Not Detected

A study was carried out to determine Lead contamination in 104 of the representative food items in the Saudi diet and to estimate the dietary Lead intake of Saudi Arabians. Three samples of each selected food items were purchased from the local markets of Riyadh city, the capital of Saudi Arabia. Each pooled sample was analyzed in triplicate by Inductively coupled plasma atomic emission spectroscopy (ICP-AES) after thorough

homogenization. Sweets (0.011–0.199 $\mu\text{g/g}$), vegetables (0.002–0.195 $\mu\text{g/g}$), legumes (0.014–0.094 $\mu\text{g/g}$), eggs (0.079 $\mu\text{g/g}$), meat and meat products (0.013–0.068 $\mu\text{g/g}$) were the richest sources of Lead. Considering the amounts of each food consumed, the major food sources of Lead intake for Saudi can be arranged as follows: vegetables (25.4%), cereal and cereal products (24.2%), beverages (9.7%) sweets (8.2%), legumes (7.4%), fruits (5.4%) milk and milk products (5.1%). The daily intake of Lead was calculated taking into account the concentration of this element in the edible part of the daily consumption data which were derived from two sources, (a) the Kingdom of Saudi Arabia (KSA) food sheet provided by the Food and Agriculture Organization (FAO) and (b) from questionnaires distributed among 300 families in Riyadh city. The results showed that the daily intakes of Lead according to the two sources are 22.7 and 24.5 $\mu\text{g/person/day}$ respectively ^[22]. Table 2 shows the concentrations of Lead in canned food samples in this study.

Table 2.2 Lead concentrations ($\mu\text{g/g}$ dry weight food) in Study conducted in Riyadh \pm SD

Food	Mean Lead concentration \pm SD
Canned beans	0.019 \pm 0.006
Beans	0.014 \pm 0.002
Canned peas	0.048 \pm 0.029
Canned kidney beans	0.018 \pm 0.009
Canned green peas	0.011 \pm 0.007
Canned Spinach	0.007 \pm 0.003

Another study in Turkey performed on 10 canned foods from Turkish markets were determined by flame and graphite furnace atomic absorption spectrometry. The contents of the investigated trace elements in canned foods were found to be in the range of 2.85–7.77 $\mu\text{g/g}$ for Copper, 8.46–21.9 $\mu\text{g/g}$ for Zinc, 6.46–18.6 $\mu\text{g/g}$ for manganese, 27.5–79.6 $\mu\text{g/g}$ for iron, 0.05–0.35 $\mu\text{g/g}$ for selenium, 0.93–3.17 $\mu\text{g/g}$ for aluminum, 0.19–0.52 $\mu\text{g/g}$ for chromium, 0.18–0.75 $\mu\text{g/g}$ for nickel, and 0.20–1.10 $\mu\text{g/g}$ for cobalt, the levels of (trace metals) in canned foods were higher than the reported vegetable samples^[23].

Another important study was performed in Egypt to determine heavy metals Cd and Pb and trace elements, Cu and Zn contents in legumes, cereals, cereal products and fried potatoes purchased from the Egyptian market were carried out using atomic absorption spectrometry, the levels of Pb in broad beans, common bean, lupine, fenugreek, rice, wheat, spaghetti and pasta were found to be above the permissible levels. The heavy metal amounts detected in legumes ranged from (0.010 to 0.178 mg.kg^{-1}) of Cd and (0.013 to 0.281 mg.kg^{-1}) of Pb. However, the trace elements ranged from (2.839 to 8.012 mg.kg^{-1}) of Cu, and (6.111 to 15.861 mg.kg^{-1}) of Zn. In the case of cereals, they ranged from (0.091 to 0.142 mg.kg^{-1}) of Cd, (0.116 to 0.398 mg.kg^{-1}) of Pb, (0.241 to 1.962 mg.kg^{-1}) of Cu, and (4.893 to 15.450 mg.kg^{-1}) of Zn. The highest values of Cd, Pb, Cu and Zn in cereal products were observed in popcorn (0.194 mg.kg^{-1}), pasta (0.299 mg.kg^{-1}), salted biscuits (1.386 mg.kg^{-1}) and pizza crust (13.70 mg.kg^{-1}), respectively^[24].

Chapter Three

Materials and Methods

3.1 Sampling

A total of sixteen canned food samples including beans, chickpeas, corn and mushroom were collected from a popular supermarket in Nablus city during 2015. We have chosen randomly 4 companies of each product. The different samples were transported to the chemistry laboratory at An-Najah National University and stored in a clean dry place prior to the digestion and analysis.

3.2 Samples preparation and digestion:

Each can was opened and the contents were mixed in blender and homogenized. We weighted 5 g of each can and transferred it to 100 ml flask. In fume hood we added approximately 5 ml of concentrated HNO_3 (Riedek-deHaen No. 30713) and heated with Bunsen burner until first vigorous reaction (black residue appeared). Then, 2 ml of H_2SO_4 (Batch No: P110001671) were added with continue heating, maintaining oxidizing conditions by adding concentrated HNO_3 in small increments until the solution is clear (yellow-orange) and no solid residue remains. A total of 25-30 ml of HNO_3 was added and this took 1.5- 2 hours for each sample. Then, the solution was transferred to 50 ml volumetric flask and rinsed appropriately to ensure quantitative transfer of the sample. Then it was diluted with distilled water to 50 ml. All the plastic and glassware were cleaned and rinsed with distilled water prior to use.

The stock solutions were prepared by taking the required amounts of Pb (NO₃)₂ salt (1.598 g), Cadmium salt (0.27g), ZnSO₄.7H₂O salt (4.397g) and CuNO₃ salt (0.465g), and dissolved in de-ionized water to give 1000 mg/L standard solution.

3.3 Apparatus

Flame Atomic absorption spectrometer IcE-3000 SERIES, Serial number c113500021 designed in UK AA Spectrometer with a hollow cathode lamp for Cadmium ,Lead, Copper and Zinc. The wavelength, slit width and lamp current of each metal was adjusted according to the description given in the manufacturer manual for determination of Cd, Pb, Cu and Zn ^[25].

3.4 Calibration

The Calibration was done by introducing water as blank to adjust reading of the instrument. Stock solution was used to prepare different concentrations 0, 2,5,10 and 20 mg/L of heavy metals Cd, Pb, Zn and Cu. After calibration, we introduced each sample into the instrument recording the steady reading obtained, and then apparatus was washed after each introduction with water as a blank solution to check that the reading returns to its initial setting.

Chapter Four

Results

4.1 Introduction

The results of this study were devoted to determine heavy metals (Cadmium, Lead, Zinc, Copper) in some canned food sold in Palestinian market, based on three repetitions of the experiment for each canned product.

The Calibration curves were constructed by plotting values of absorbance versus concentration, as shown in the following figures (Figures 4.1- 4.4).

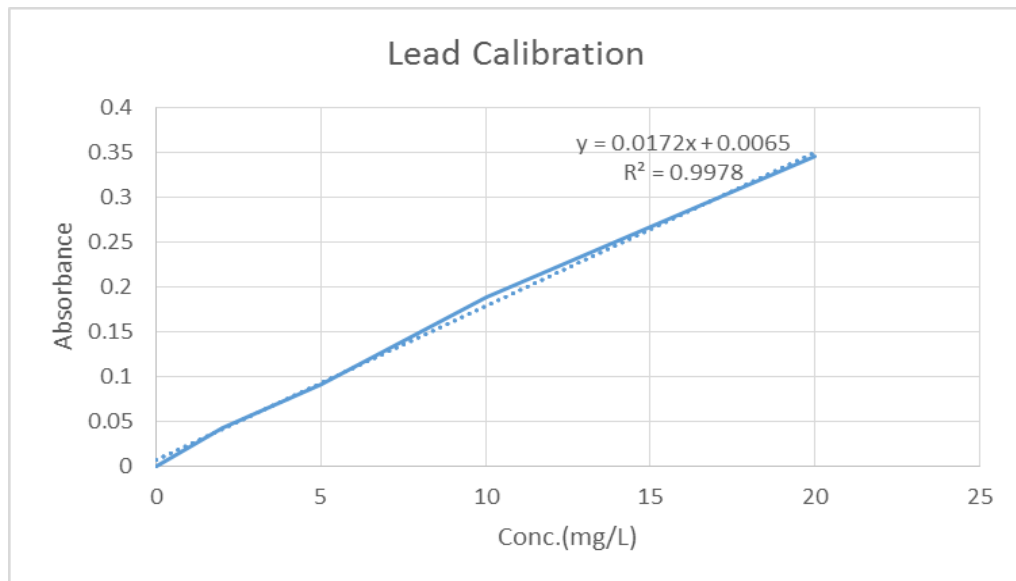


Figure 4.1. Calibration Curve of Lead.Conc: concentration.

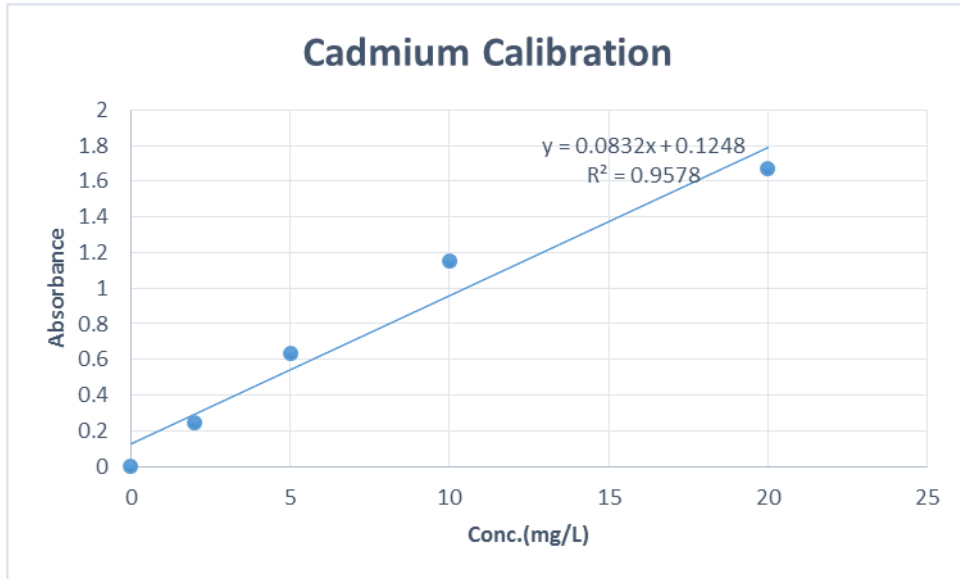


Figure 4.2: Calibration Curve of Cadmium. Conc: concentration.

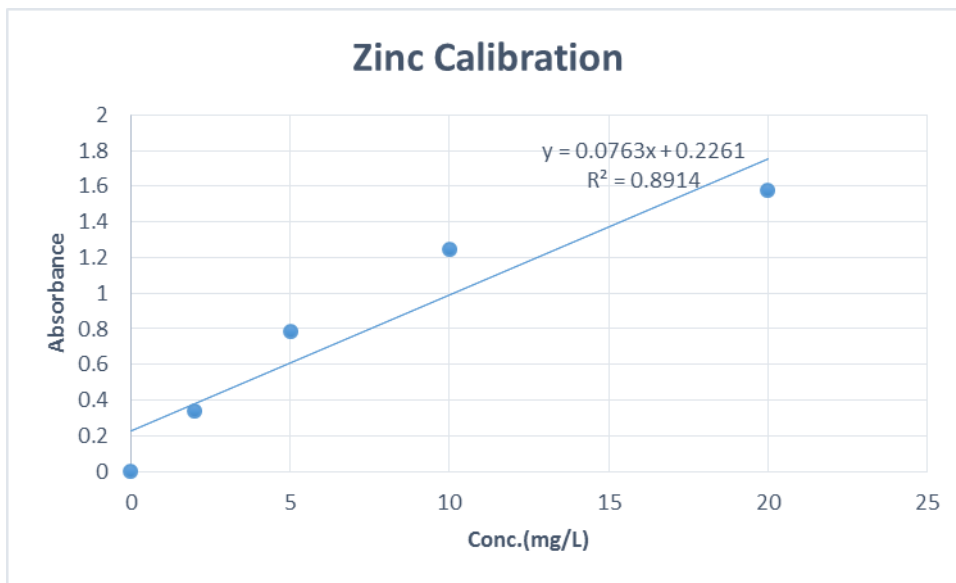


Figure 4.3: Calibration Curve of Zinc. Conc: concentration.

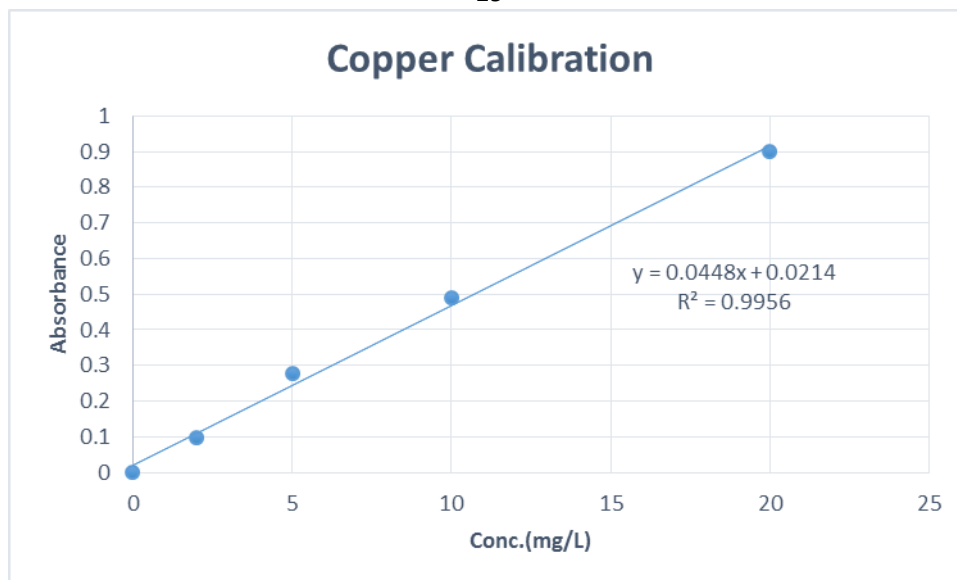


Figure 4.4: Calibration Curve of Copper. Conc: concentration.

4.2 Samples origin

As shown in table 4.1 below, the canned food sources in this study were from five different countries: Thailand, China, United Arab Emirates (UAE), Jordan and Palestine.

Table 4.1: Origin of food types used in the study.

Food Type	Company	Origin
Corn	Marina	Thailand
	Sahten o Afia	China
	Americana	UAE
	Freshly	Thailand
Chickpeas	Americana	UAE
	Kaseeh	Jordan
	Kfr Qare	Kfr Qare
	Affoleh	Affoleh
Beans	Kaseeh	Jordan
	Americana	UAE
	Shelleh	China
	Delmonaty	UAE
Mushroom	Big Sea	China
	Marina	China
	Sahten o Afia	China
	Americana	China

UAE: United Arab Emirates.

4.3 Corn results

As shown in table 4.2 below, the Marina corn had the highest Lead concentration (1.17 mg/L), but freshly corn had the lowest one. Cadmium concentration in all corn types was very low. However, Sahten o Afia had the highest concentration of Cadmium. All corn cans had convergent concentrations of Zinc. Americana corn Copper concentration was the highest one compared to other canned corn origins.

Table 4.2: Concentrations (mg/L) of heavy metals contents in canned corn.

Food Type	Pb	Cd	Zn	Cu
		*Conc.± SD		
Marina	1.17 ± 0.10	0.07 ± 0.01	5.49± 0.31	0.85 ± 0.08
Sahten o Afia	0.50 ± 0.04	0.09 ± 0.01	5.71± 0.45	0.92 ± 0.06
Americana	0.47 ± 0.05	0.05± 0.01	6.80 ± 0.5	1.05 ± 0.10
Freshly	0.30 ± 0.02	0.01 ± 0.01	6.69 ± 0.47	0.79 ± 0.05
Average	0.61 ± 0.05	0.06 ± 0.01	6.17 ± 0.36	0.91 ± 0.07

***Conc.±SD : Concentration± Standard Deviation**

Figure 4.5 below, represent the Lead concentration± SD (standard deviation) among corn food types. Marina canned corn had the highest concentration of Lead and Freshly had the lowest one.

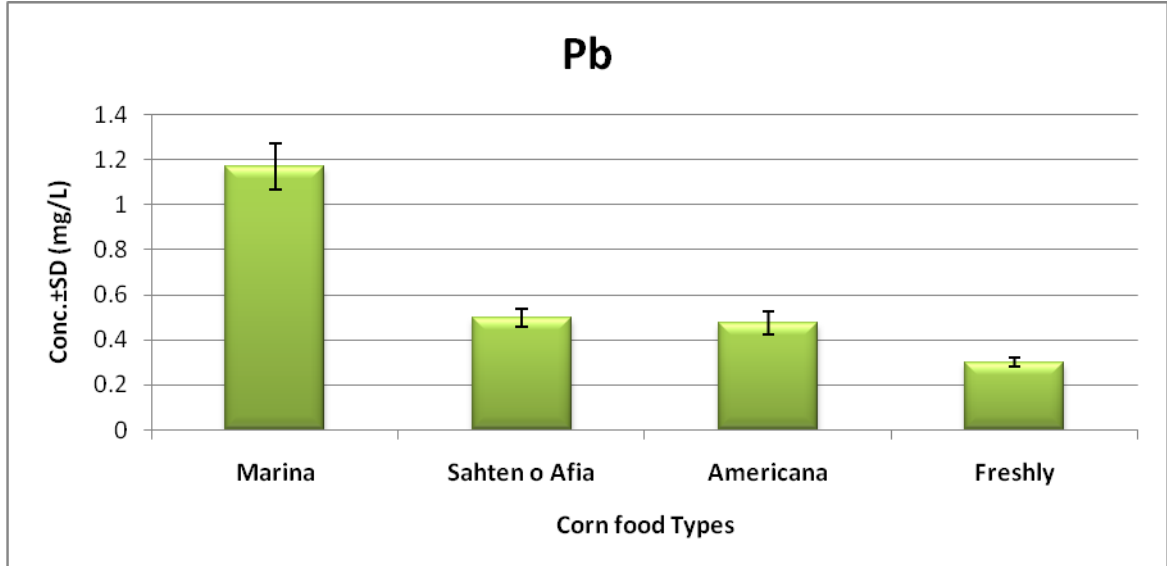


Figure 4.5: Lead concentrations in canned corn types. The error bars represent ± SD.

Conc.: concentration; SD: Standard Deviation

Regarding Cd in canned corn, the highest concentration was observed in Sahten o Afia corn (figure 4.6).

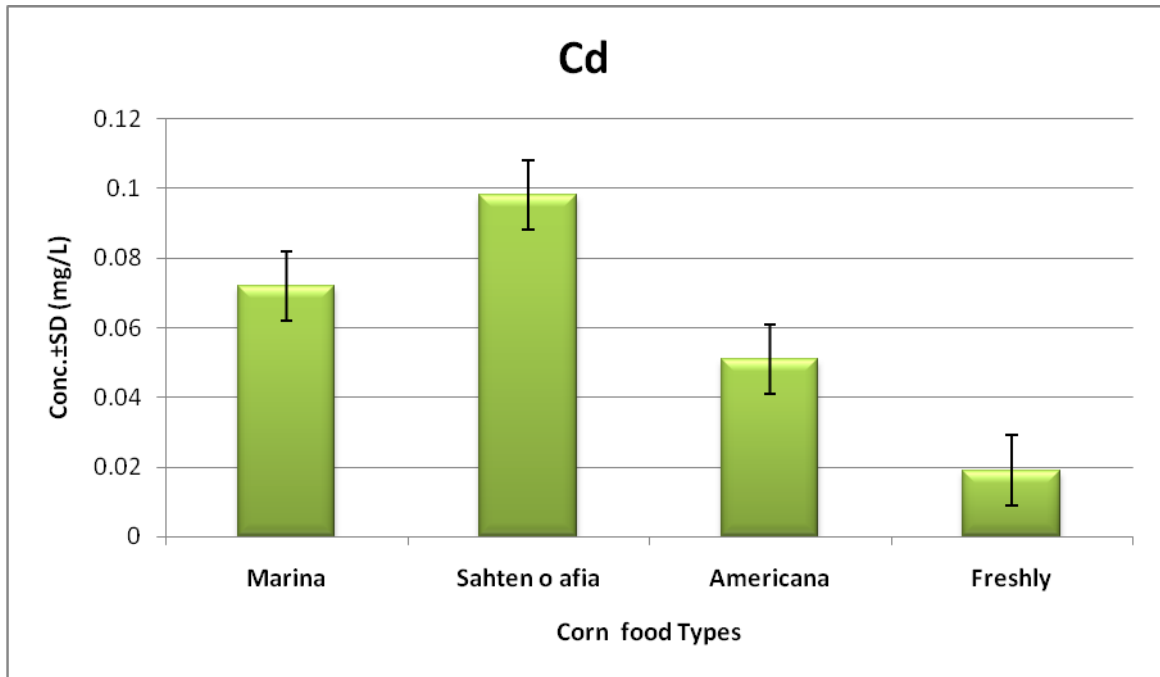


Figure 4.6: Cadmium concentrations in canned corn types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation

In regard to Zn concentrations in canned corn, the levels were nearly approximate each other as show in figure 4.7 below.

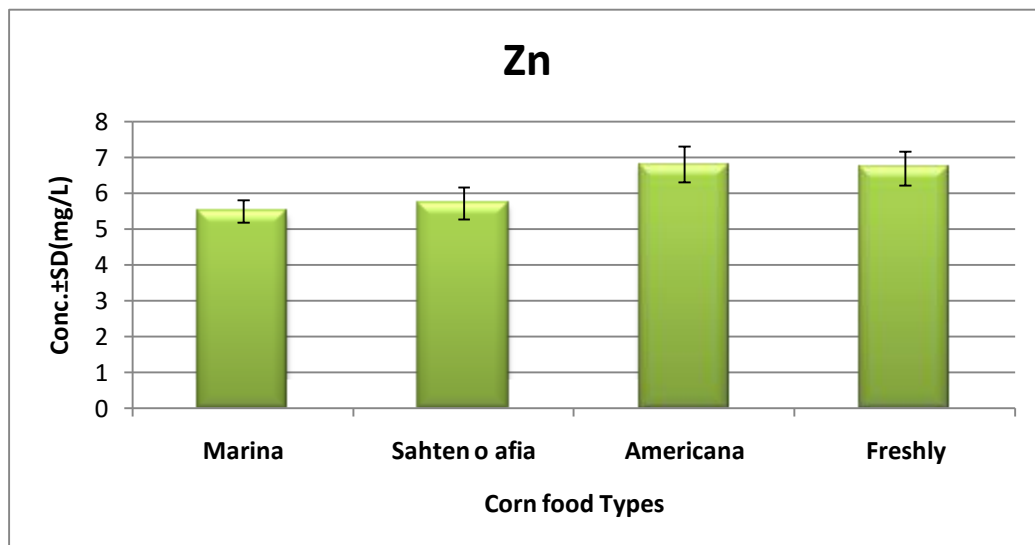


Figure 4.7: Zinc concentrations in canned corn types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation

Americana canned corn had the highest concentration of Cu as shown in figure 4.8 below.

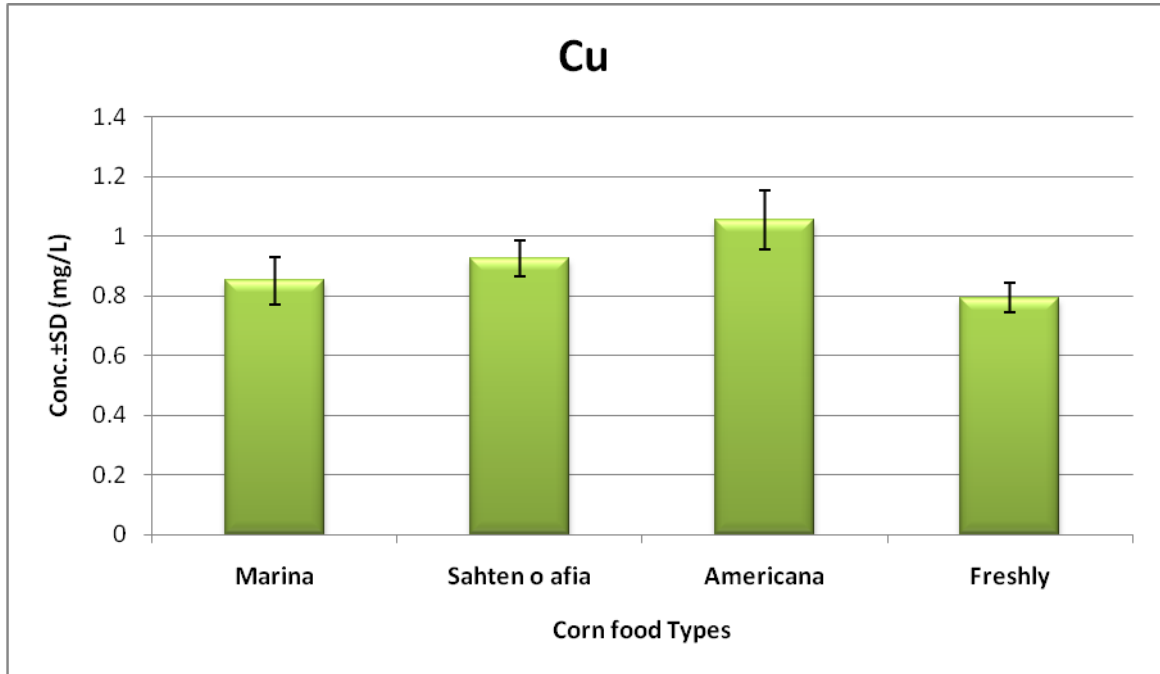


Figure 4.8: Copper concentrations in canned corn types. The error bars represent \pm SD.

Conc.: concentration; SD: Standard Deviation

4.4 Chickpeas results

As shown in table 4.3 below, Kaseeh and Kfr Qare Lead concentrations were the highest in comparison to other chickpeas origins, but Affoleh chickpeas had the lowest concentration of Lead. Chickpeas had low concentrations of Cd but Zn concentrations were very high. All chickpeas cans had convergent concentrations of Cu and the highest one was Affoleh.

Table 4.3: Concentrations (mg/L) of heavy metals contents in canned chickpeas.

Food Type	Pb	Cd	Zn	Cu
	*Conc. ± SD			
Americana	0.25 ± 0.03	0.20 ± 0.02	10.39 ± 0.6	1.99 ± 0.15
Kaseeh	0.75 ± 0.06	0.16 ± 0.03	8.90 ± 0.7	1.97 ± 0.12
KfrQare	0.70 ± 0.05	0.20 ± 0.02	10.0 ± 0.9	1.93 ± 0.10
Affoleh	0.12 ± 0.01	0.10 ± 0.02	10.66 ± 0.6	2.05 ± 0.25
Average	0.46 ± 0.03	0.17 ± 0.02	9.99 ± 0.7	1.98 ± 0.15

***Conc±SD.: Concentration ±Standard deviation**

Figure 4.9 below represents Lead concentration in canned chickpeas ±SD, the highest concentration of Lead was in Kaseeh chickpeas and the lowest was in Affoleh.

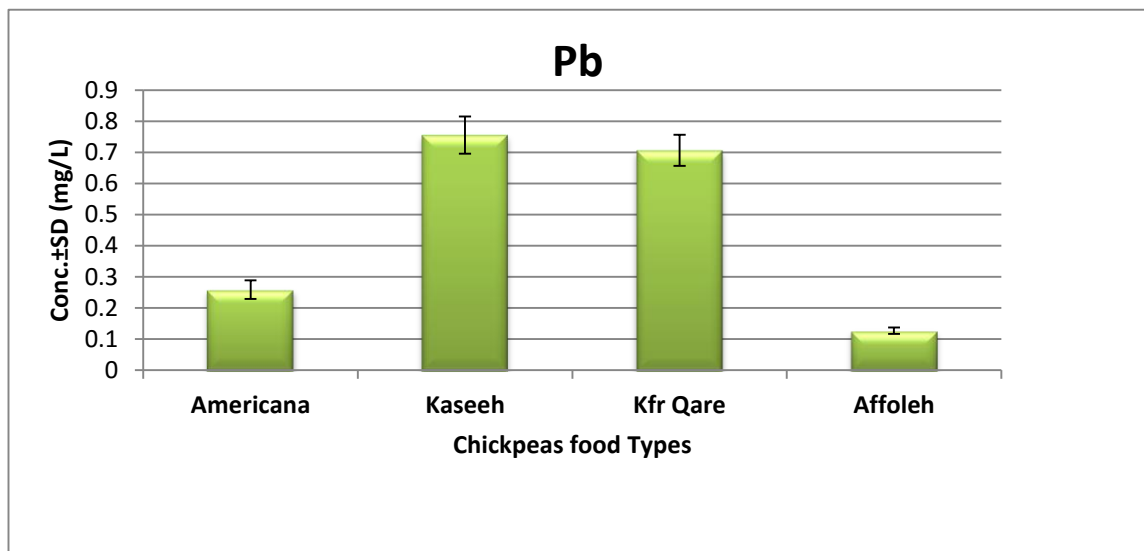


Figure 4.9: Lead concentrations in canned chickpeas types. The error bars represent ± SD. Conc.: concentration; SD: Standard Deviation

Regarding to Cd, Americana and Kfr Qare chickpeas had nearly approximate concentrations of Cadmium as shown in figure 4.10 below.

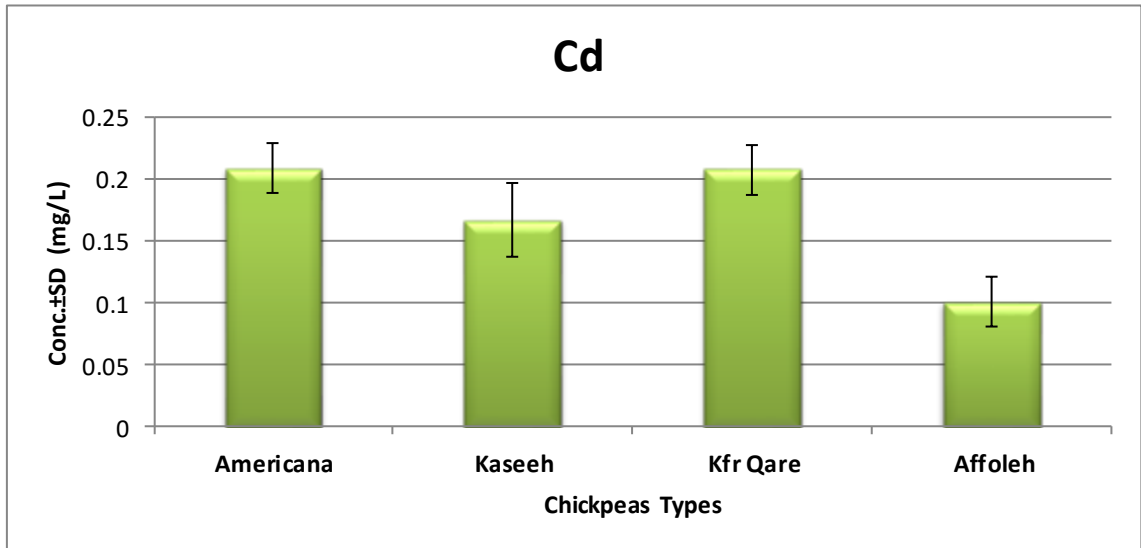


Figure 4.10: Cadmium concentrations in canned chickpeas types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation.

As we can see in figure 4.11 below, Affoleh chickpeas had the highest concentration of Zinc.

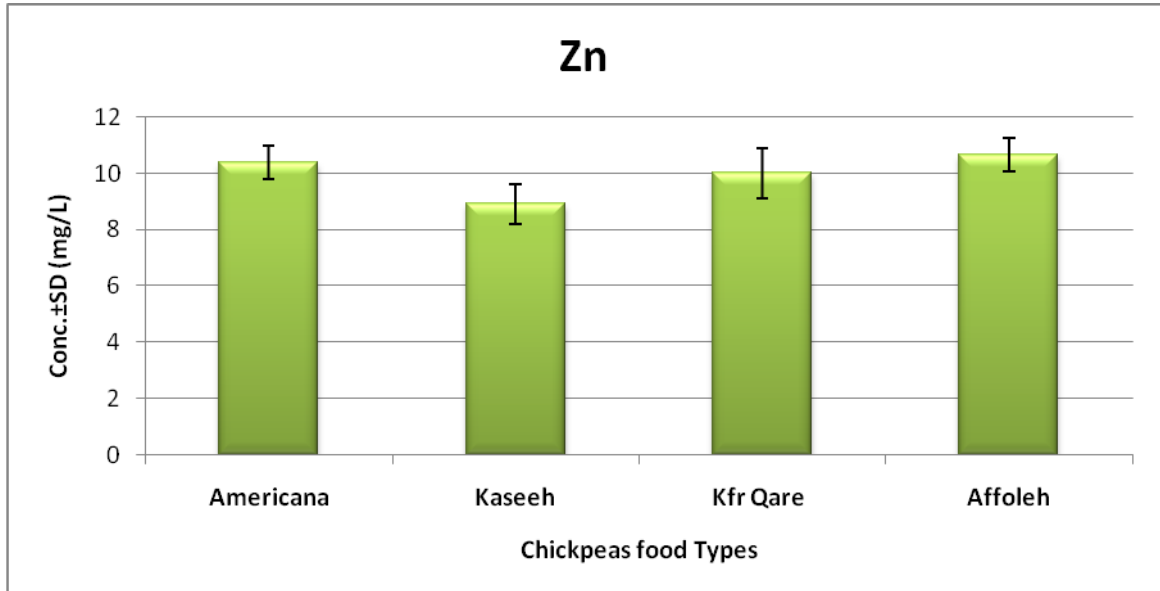


Figure 4.11: Zinc concentrations in canned chickpeas types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation

Also figure 4.12 below show that Affoleh chickpeas also had the highest concentration of Copper.

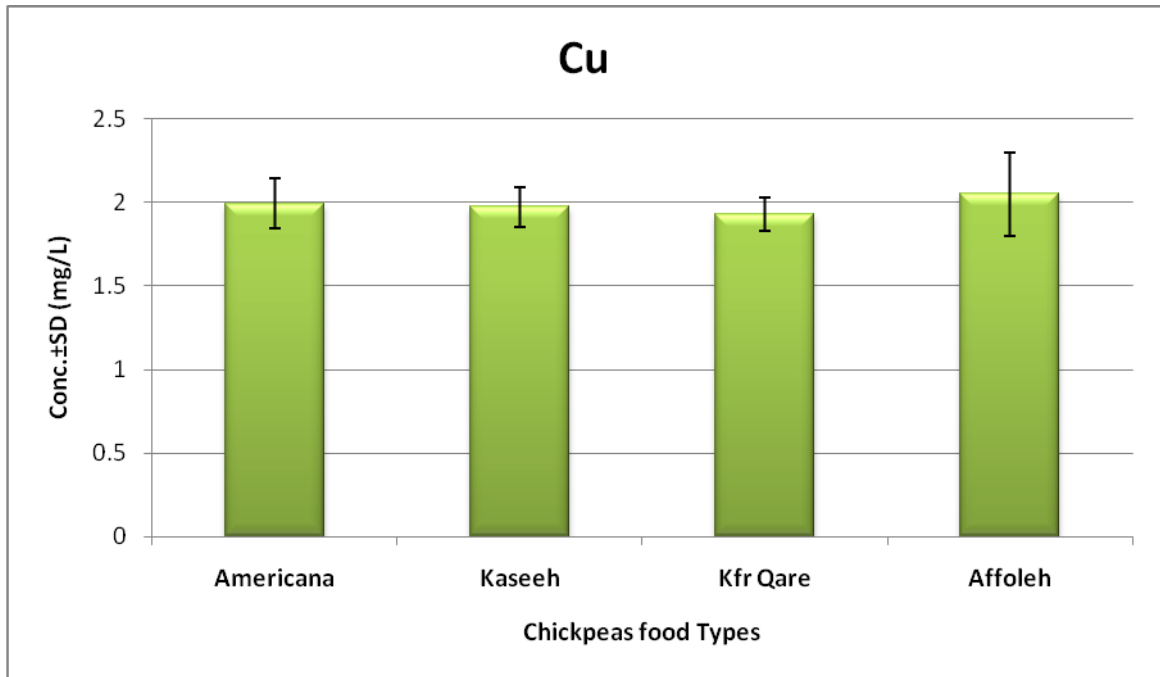


Figure 4.12: Copper concentrations in canned chickpeas types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation

4.5 Beans results

As shown in table 4.4 below, canned beans samples had very low concentrations of Lead and in some of them it was negative. Cadmium concentrations were almost high, especially in Kaseeh sample. In contrast Kaseeh had the lowest concentration of Zn and Cu. Americana Beans had the highest levels of Cu, and shelleh had the highest one of Zn.

Table 4.4: Concentrations (mg/L) of heavy metals contents in canned beans.

Food type	Pb	Cd	Zn	Cu
	*Conc. \pm SD			
Kaseeh	0.350 \pm 0.03	0.32 \pm 0.05	6.55 \pm 0.51	1.68 \pm 0.12
Americana	ND*	0.30 \pm 0.03	8.45 \pm 0.7	3.25 \pm 0.19
Shelleh	ND*	0.24 \pm 0.02	9.26 \pm 0.9	2.59 \pm 0.5
Delmonaty	0.089 \pm 0.01	0.23 \pm 0.01	8.67 \pm 0.8	2.97 \pm 0.3
Average	0.22 \pm 0.02	0.27 \pm 0.095	8.24 \pm 0.72	2.63 \pm 0.27

*Conc. \pm SD: Concentration \pm Standard Deviation

*ND: Not detected

Figure 4.13 below represents concentrations of Lead \pm SD. Kaseeh canned beans had the highest concentration of Lead, but Americana and Shelleh types had no detected Lead in them.

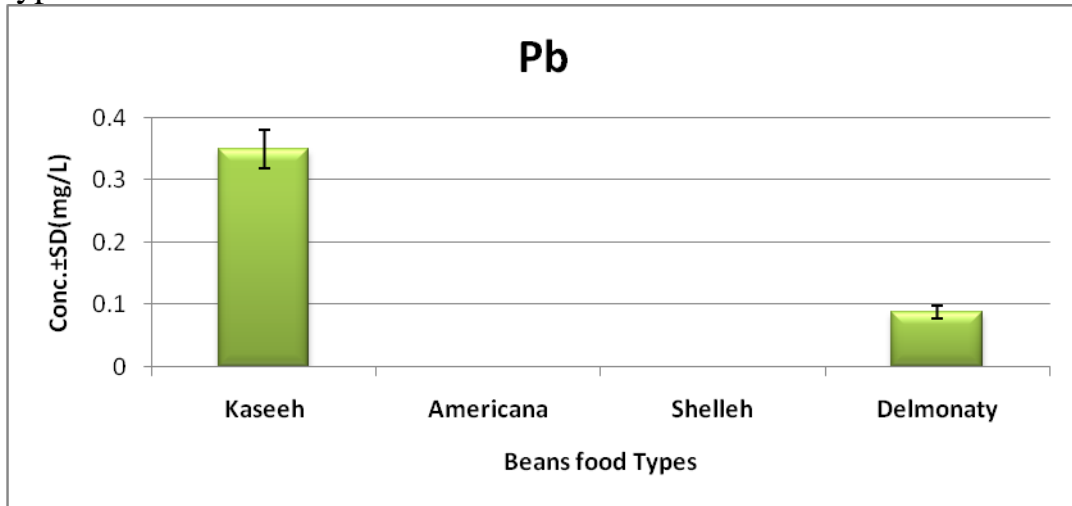


Figure 4.13: Lead concentrations in canned beans types. The error bars represent \pm SD.

Conc.: concentration; SD: Standard Deviation

Also Kaseeh canned beans had the highest concentration of Cadmium as shown in figure 4.14 below.

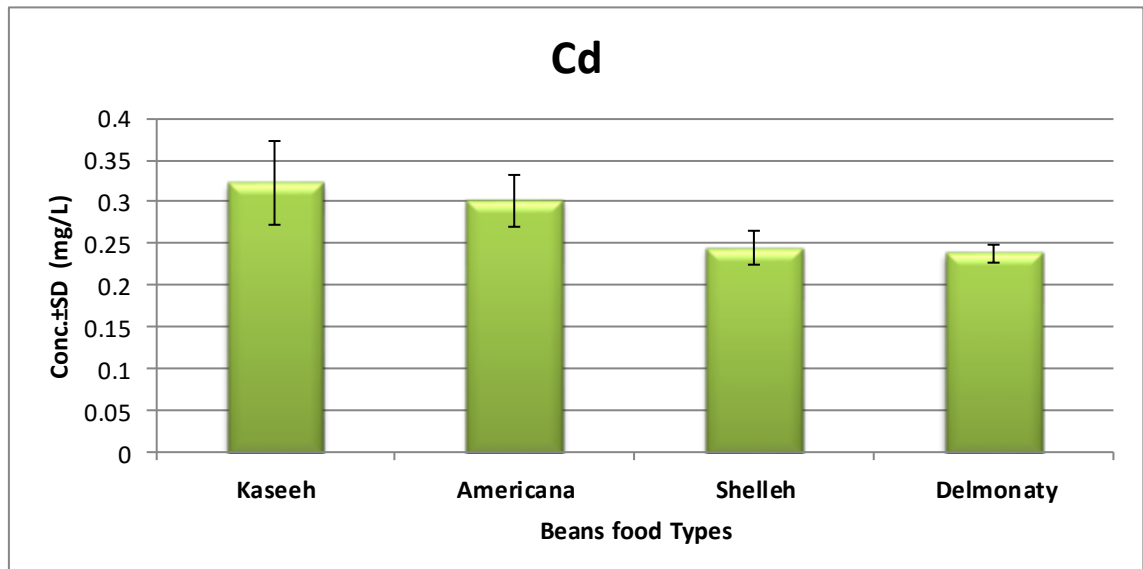


Figure 4.14 : Cadmium concentrations in canned beans types. The error bars represent \pm

SD. Conc.: concentration; SD: Standard Deviation.

In regarding to Cu, Americana and Delmonaty canned beans had the highest concentration of Cu as shown in figure 4.15 below.

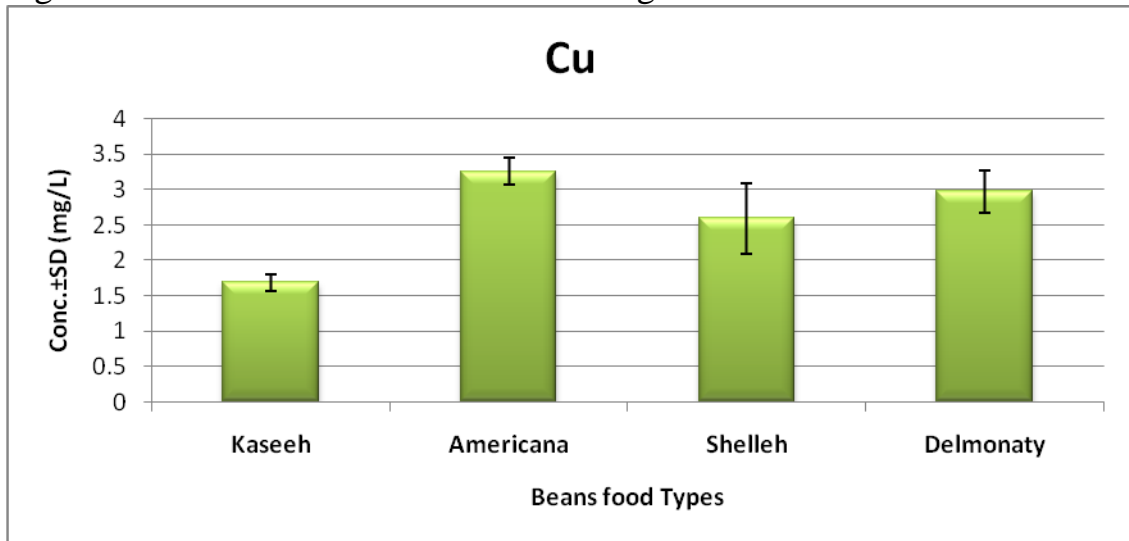


Figure 4.15: Copper concentrations in canned beans types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation.

Concerning to Zn, canned beans had approximate concentrations of Zinc as shown in figure 4.16 below.

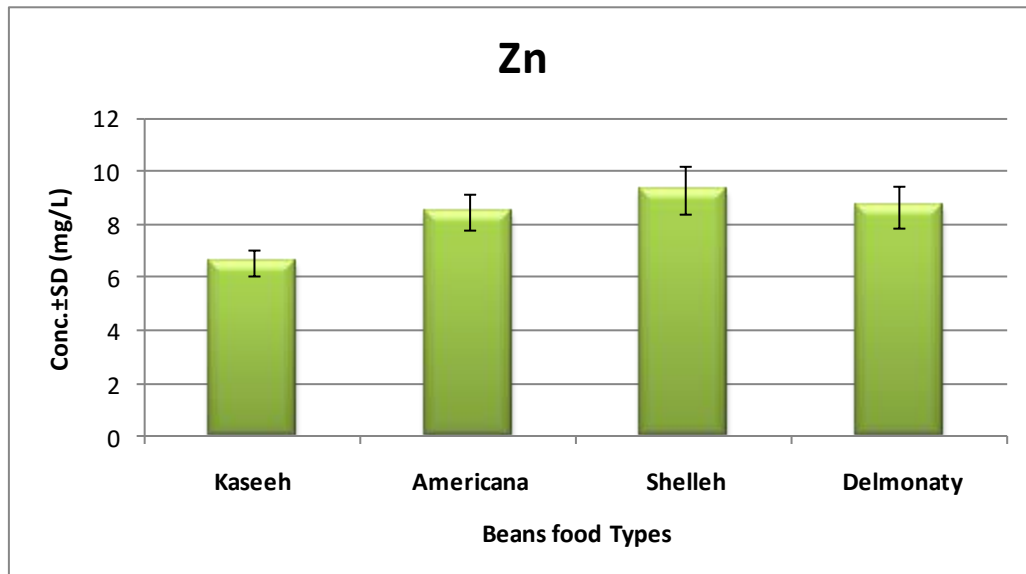


Figure 4.16: Zinc concentrations in canned beans types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation

4.6 Mushroom results

As shown in table 4.5 below, the canned mushroom had low concentrations of Lead, and Marina Mushroom was the lowest. Mushroom had very low concentrations of Cd, Marina and Americana had negative values. The Zn and Cu concentrations were generally low.

Table 4.5: Concentrations (mg/L) of heavy metals contents in canned Mushroom.

Food Type	Pb	Cd	Zn	Cu
		Conc. \pm SD*		
Big sea	0.549 \pm 0.05	0.065 \pm 0.02	2.058 \pm 0.27	1.005 \pm 0.20
Marina	0.293 \pm 0.03	ND*	3.678 \pm 0.51	1.438 \pm 0.34
Sahteen o afia	0.439 \pm 0.04	0.023 \pm 0.01	4.157 \pm 0.062	0.815 \pm 0.05
Americana	0.454 \pm 0.06	ND*	5.670 \pm 0.1	1.280 \pm 0.12
Average	0.433 \pm 0.045	0.044 \pm 0.05	3.89 \pm 0.23	1.134 \pm 0.17

***Conc. \pm SD: Concentration \pm standard deviation. *ND:Not detected**

Figure 4.17 below represents Lead concentrations \pm SD in mushroom canned food .Big Sea had highest concentrations in contrast,Marina had the lowest one.

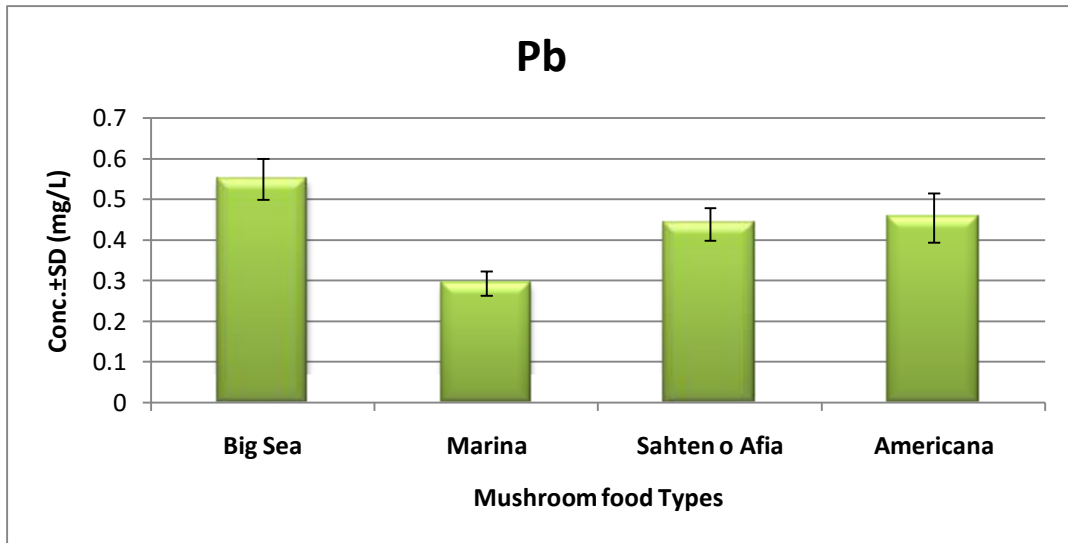


Figure 4.17: Lead concentrations in canned mushroom types. The error bars represent \pm SD.

Conc.: concentration; SD: Standard Deviation.

Regarding to Cd, Big Sea canned mushroom had highest concentration of Cadmium, however, Marina and Americana types had no detected Cadmium in them as shown in figure 4.18 below.

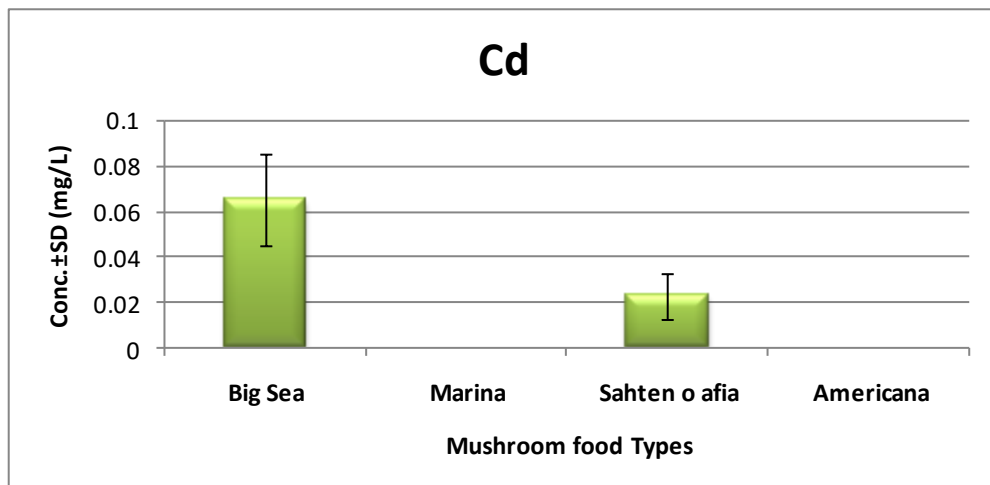


Figure 4.18: Cadmium concentrations in canned mushroom types. The error bars represent \pm SD.

Conc.: concentration; SD: Standard Deviation

In regard to Cu, Marina canned mushroom had the highest concentration of Cu and Sahten o Afia had the lowest one as shown in figure 4.19 below.

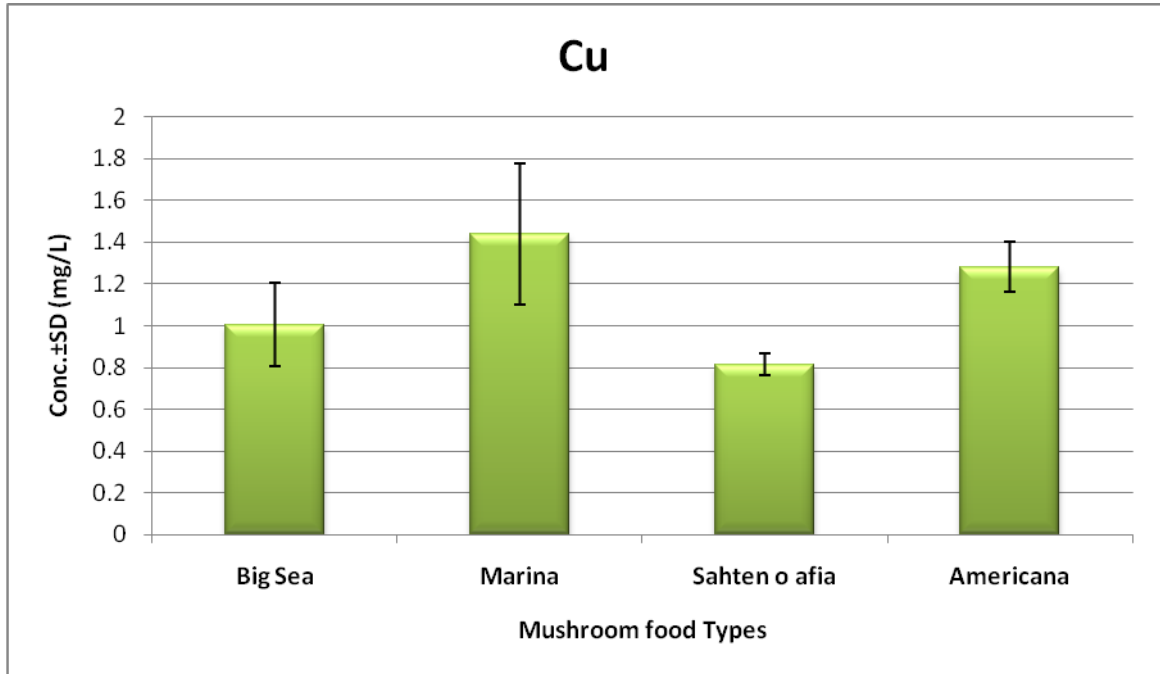


Figure 4.19: Copper concentrations in canned mushroom types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation.

Referring to Zn, Americana canned mushroom had the highest concentration of Zn as shown in figure 4.20 below.

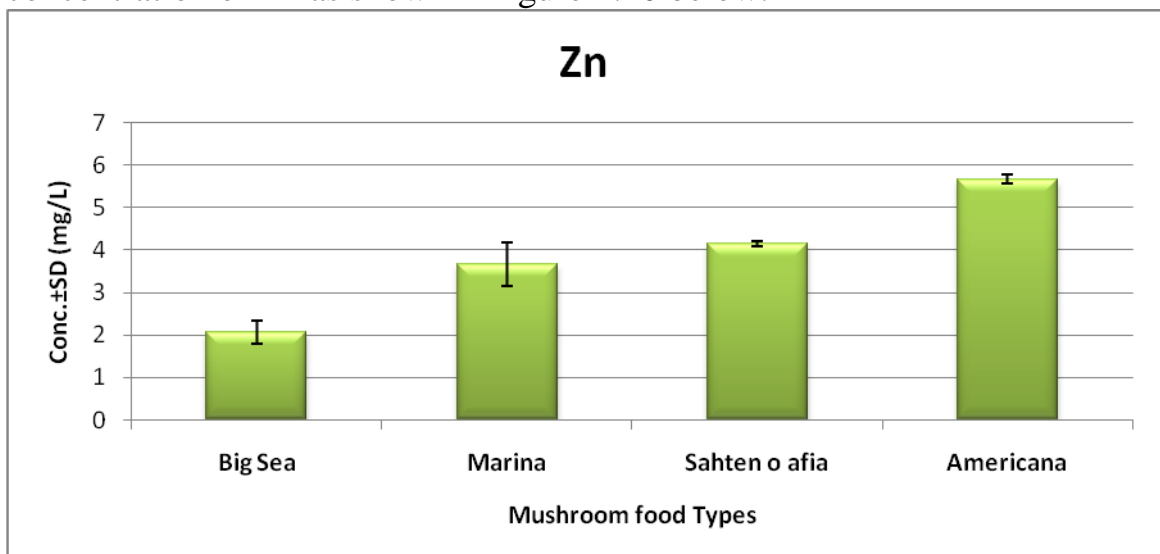


Figure 4.20: Zinc concentrations in canned mushroom types. The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation.

4.7 Comparison of all canned food types

Table 4.6 and figure 4.21 below show the concentrations of all measured elements in the study among all canned food origins. As shown, chickpeas has the highest average concentration of Zn followed by canned beans. While Mushroom has the lowest average concentration on Zn.

Regarding Pb, it was found that canned corn has the highest average concentration while beans the lowest. In addition, mushroom and chickpeas has nearly the same approximate Pb levels of concentrations. However, canned Beans has the highest concentration of Cd followed by canned chickpeas, but corn and mushroom had very low concentrations of Cd.

Canned Beans also has the highest concentration of Cu. In contrast canned corn has the lowest. Chickpeas has higher concentration of Cu than mushroom.

**Table 4.6: Average concentrations (mg/L) of heavy metals in the study
canned food**

Food Item	Pb	Cd	Zn	Cu
	*Conc. ± SD			
Corn	0.61 ± 0.05	0.06 ± 0.01	6.176 ± 0.36	0.91 ± 0.07
Chickpeas	0.46 ± 0.03	0.17 ± 0.02	9.9905 ± 0.7	1.98 ± 0.15
Beans	0.22 ± 0.02	0.27 ± 0.09	8.24 ± 0.72	2.62 ± 0.27
Mushroom	0.43 ± 0.04	0.044 ± 0.05	3.89 ± 0.23	1.13 ± 0.17

***Conc.± SD: Concentration ±Standard deviation**

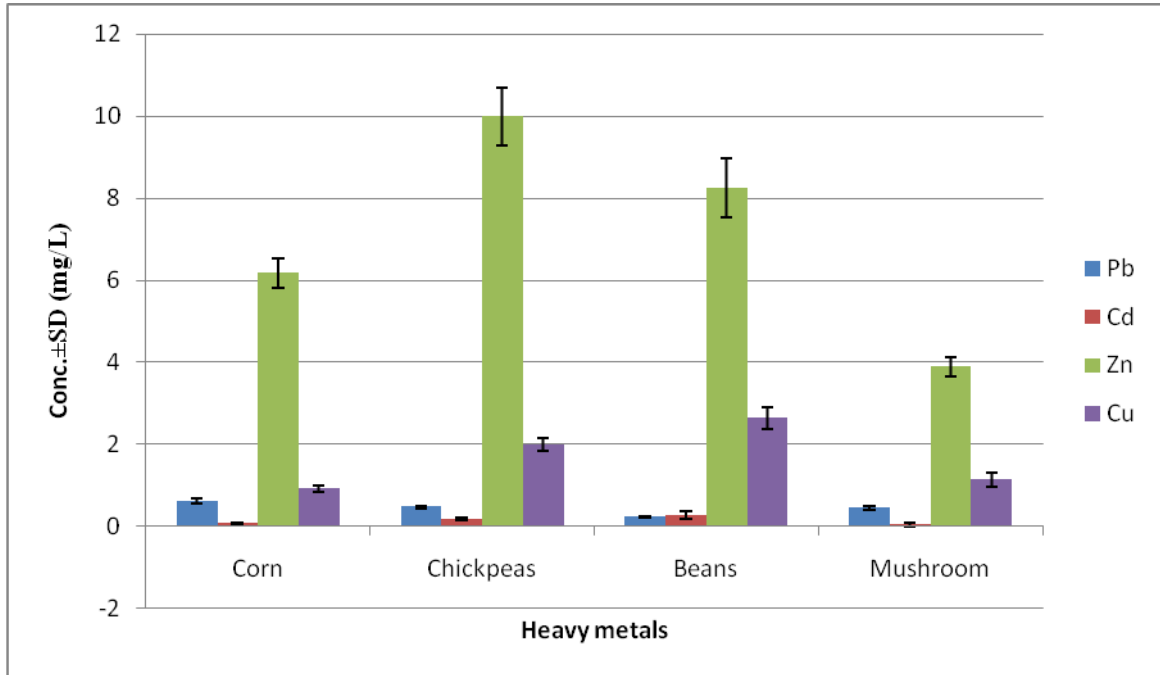


Figure 4.21: Heavy metals average concentrations in the study canned food items . The error bars represent \pm SD. Conc.: concentration; SD: Standard Deviation

5.3 References values for the studied elements

The ATSDR stated that the chronic durational oral minimal risk level (MRL) for Cd is $0.1\mu\text{g}/\text{kg}/\text{day}$ based on its renal effect (ATSDR). If we take 60 kg person , he needs $6\mu\text{g}/\text{day}$ to reach the permissible level which is lower than our results for Cd, whereas our ranges of Cd in our study was (0-0.322)mg/L which were higher than the permissible level (0.006 mg/L)^[13] . Therefore, canned food may harm human health in chronic use and this maybe one of the risk factors for kidney diseases in the society, because Cd had a very high effect on kidneys, and this is an alarm in public health for kidney diseases.

Expert Committee on Food Additives has suggested a provisional tolerable intake of 400–500 μg Pb per week for man^[26]. However, the Food and Drug Administration (FDA) has set an action of 0.5 $\mu\text{g}/\text{ml}$ for Pb in products

intended for use by infants and children, and has banned the use of Lead-Soldered food cans. (FDA 1994, as cited in ATSDR 1999)^[14]. The Food and Agriculture Organization (FAO) has set (1mg/kg) as maximum level of Lead in canned food^[27]. In our study, the maximum level of Lead was 1.17mg/L in Marina corn which is higher than the permissible level of Pb in canned food. But the other samples were below the permissible level.

The recommended dietary allowance (RDA) of Cu is 0.9mg/day for adult has recently published by the ATSDR ^[17]. Our Cu ranges was (0.7-3.25) mg/L which is higher than the permissible levels, and this is an important result in the time that excessive levels of Copper can result in a number of adverse health effects including liver and kidney damage, anemia, immunotoxicity and developmental toxicity^[17]. Also ,the Food and Nutrition Board of the Institute of Medicine in Washington DC has developed recommended dietary allowances (RDAs) of 340 µg of Copper per day for children aged 1-3 years, 440 µg/day for children aged 4-8 years, 700 µg/day for children aged 9-13 years, 890 µg/day for children aged 14-18 years ^[17] .

Permissible levels of Zinc in food as reported in ATSDR are 8mg/day for female and 11mg/day for male. In our study, concentration of Zinc range was (2.058-10.65)mg/L which is higher than permissible level ^[16] .

Chapter Five

Discussion

5.1 Background

Environmental pollution with heavy metals and its effects on human health have been important issue all over the world. Minerals are important components required by humans in our daily food need. These includes more than 22 mineral elements, some of them are required in large amounts such as: Calcium(Ca), Magnesium (Mg), and Potassium (K). But others such as Copper (Cu) and Zinc (Zn), are required in trace amounts because higher concentrations of these elements can be harmful ^[20].

In our study we assessed Pb, Cd, Cu and Zn in canned foods which include corn, chickpeas, beans and mushroom that are sold in Palestinian market at Nablus city in 2015.

5.2 Main study findings

The contents of investigated metals in canned food in this study were found to be in the range of (0.089-1.17mg/L) for Pb, (0.019-0.322mg/L) for Cd, (2.05-10.6mg/L) for Zn and (0.79-2.97mg/L) for Cu. According to these data, Zn had the highest concentrations, followed by Cu then Pb .As expected, the lowest metal in our study canned food was Cd.

Cadmium had no known biological function in humans; it almost accumulates in kidneys and liver, also it has long half-life (4-19 years)^[13]. In our study the highest concentrations of Cd were in “kaseeh”canned

beans (0.322mg/L) and this result agree with Korfali, Hamadan et al.2013^[21]who reported that Cd in Fava beans level as (0.483µg/g) . On the other hand, the lowest concentration of Cd in canned beans was in “Delmonaty” 0.23 mg/L and this is higher than levels of Cd reported in Radwan et al.2005^[24] in legumes at the range of (0.01-0.178) mg/kg. Mushroom had generally low concentrations of Cd (0 - 0.065mg/L in comparison to all canned food in our study and these results almost agree with S.Abd_Al-Wahab et al. 2014^[29] results that reported (Not Detected) levels of Cd in canned mushroom. High concentrations of Cd was detected in the present study for canned chickpeas (0.208mg/L for Americana , 0.166 mg/L for Kaseeh , 0.207 mg/L for KfrQare, 0.100 mg/L for Affoleh) and this disagree with S.Abd Al_waahab et al. 2014^[29]that reported (Not Detected) Cd levels in canned chickpeas. In contrast, our results agree with Korfali, Hamadan et al 2013 ^[21] levels of Cd which was reported as (0.125µg/g) in canned chickpeas .

Lead is toxic metal and even in low concentration. It causes health hazards since it is not biodegradable and it may cause kidney damage as mentioned in Korfali, Hamadan et al. 2013 ^[21]. The presence of (Pb) in canned food may due to use of Pb in product package materials or due to absorbed Lead by plants in limit level^[29].

In general, beans had the lowest concentrations of Pb in comparison to canned food types in this study and this agree with a study conducted in Riyadh to measure Pb levels in canned beans which was reported as 0.019 µg/g in Othman ZAA et al .2010^[22]. Our reported levels of Pb in Marina

corn (1.17 mg/L) which produced in Thailand was higher than that indicated in S.Abd Al_Wahab, et al .2014^[29] for sweet corn (0.75ppm) even they are from same origin. In contrast, our reported results for Pb in mushroom (0.549 mg/L for BigSea , 0.293mg/L for Marina , 0.439mg/L for Sahten o Afia , 0.454 mg/L for Americana) were lower than levels reported in S.Abd Al-Wahab et al.2014^[29] which was (1ppm) , even they are from the same origin (China).

In a study conducted in Saudi Arabia for Z.thagafi, H.Arida et al. 2014^[20] to measure heavy metals in fresh and canned food, the highest level of Pd in canned food was 7.11mg/kg which is very high in comparison to our results, whereas the highest level of Pb in our study was 1.17mg/L in Marina corn.

Lead was determined in legumes in a study performed in Egypt for Radwan et al .2005 ^[24], ranged from (0.013-0.281)mg/kg, which is lower than our results in chickpeas (0.127-0.756mg/L) but approximate to our beans results (0.089-0.35)mg/L.

Copper is known as vital and toxic for many biological systems and may enter food from soil through mineralization by crops, food processing or environmental contamination, such as Copper-based pesticides ^[30].

The minimum Copper value in our study was found as 0.793mg/L for “Freshly” corn and maximum value as 3.25mg/L for ”Americana” beans. Our results for Copper were low in comparison to Tuzen, Soylak et al.2007^[23] study which reported a concentration of 4.8 µg/g in Mushroom, 3.52µg/g in corn and 7.77 µg/g in canned Beans.

Copper value in canned food in a study performed in Saudi Arabia for Z.thagafi, H.Arida et al. 2014^[20] reported a range level of (6.22-8.03 mg/kg) which is very high in comparison to our results (0.79-3.97mg/L). Whereas in a study conducted in Lebanon for Kofrali, Hamdan et al .2013^[21], Cu concentration was reported as not detected in chickpeas ,mushroom and fava beans.

Zinc is one of the most important trace metals for normal growth and development of humans. Deficiency of Zinc can result from inadequate dietary intake, impaired absorption, excessive excretion or inherited defects in Zinc metabolism^[15].

Zinc had the highest levels in the present study. The maximum level of Zn was in chickpeas with average 9.9mg/L, and lowest was in mushroom with average 3.89mg/L. These results disagree with Tuzen and Soylak et al .2007^[2]who reported that mushroom had the highest levels of Zn (21.9µg/g) followed by beans (12.8µg/g) and corn (8.5µg/g), while Zn levels in our study was 6.17mg/L for corn and 8.2mg/L for beans.

The present study reported a range of (2.05-10.6mg/L) of Zn in all food samples whereas it is almost lower than levels of Zn reported in Radwan et al. 2005^[24] with a range of (6.11-15.86mg/kg) in legumes. In contrast to the study performed in Lebanon for Kofrali, Hamdan et al .2013^[21] which reported Zn concentration as not detected in the canned food .

Conclusion:

This study has assessed the metal content (essential and toxic) in different canned food categories and brands sold in Palestinian market. For toxic metals (Pb,Cd), Lead concentrations in our study were within international permissible levels, but Cadmium results were higher than international permissible levels and this is an important issue in public health for food safety and heavy metals accumulation in the human body. For the essential metals (Zn,Cu), Zinc levels were within the permissible levels, in contrast ,Copper levels were higher than the international permissible levels.

Canned beans had the highest concentrations of Cd and Cu. In contrast, it has the lowest concentrations of Pb .Furthermore, canned chickpeas had the highest concentrations of Zn. Canned Marina corn had the highest levels of Pb .On the other hand, Mushroom had the lowest levels of Cd.

In conclusion, we didn't assess or correlate the environmental exposure levels of canned food elements with human biological biomarkers or adverse effects. Therefore, conclusions whether these levels were toxic at the biological levels should be drawn with caution as we only assess some exposure concentrations of which they might be different when reaching the human body.

Recommendations:

1. Efforts should be made by the government to control processing of canning raw materials.
2. The study findings necessitate continuous monitoring of Cd and Cu levels in canned food to obtain food safety.
3. A better selection of the fresh material, including an analysis for toxic elements prior to processing, could surely improve the situation.
4. The government should control the use of fertilizers by farmers which may increase Cu levels.
5. This study suggests more investigation studies to determine the contamination of canned food with heavy metals in different canned food.
6. We recommend further future studies that assess the levels of the studied elements in human body and selecting samples from hair, nails and blood and correlate this concentrations with the individual intake of the studied food items to find out whether there will be a significant correlations or not.

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جامعة النجاح الوطنية
كلية الدراسات العليا

تقييم التعرض البيئي للكاديوم، الرصاص، النحاس والزنك في
بعض الأغذية الفلسطينية المعلبة

اعداد

غدير غسان سايح

بإشراف

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أ. د. شحده جوده

قدمت هذه الأطروحة استكمالاً لمتطلبات الحصول على درجة الماجستير في الصحة العامة بكلية
الدراسات العليا في جامعة النجاح الوطنية في نابلس-فلسطين

2016

ب

تقييم التعرض البيئي للكاديوم، الرصاص، النحاس والزنك في بعض الأغذية الفلسطينية المعلبة

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الملخص

مقدمة:

واحدة من اهم قضايا الصحة البيئية التي تواجه العالم ، هو تلوث البيئه من خلال المواد العضويه وغير العضويه، المصدر الرئيسي لتعرض الانسان للعناصر الثقيله هو عن طريق تناول الطعام. هناك العديد من العناصر الكيميائيه الموجوده في الغذاء ضروريه لحياة الانسان بتراكيز منخفضه، ولكن يمكن ان تكون سامه اذا كانت بتراكيز عاليه او من خلال الاستمرار في تناولها لفترات طويله. الاغذيه المعلبه متواجده بشكل كبير في الاسواق وتعتبر رخيصه الى حد ما، لذلك الكثير من الناس يفضلون تناولها. الهدف من هذه الدراسه هو تحديد مستويات العناصر الثقيله في بعض المواد الغذائيه المعلبه التي تباع في السوق الفلسطيني ومقارنتها مع المستويات الدوليه.

طريقة البحث:

تم قياس مستوى تركيز العناصر الثقيله (الرصاص، الكاديوم،الزنك والنحاس) في 16 عينه من مختلف المواد الغذائيه المعلبه (الذره،الحمص، الفول والبطر) التي تباع في السوق الفلسطيني وتشمل شركات ومصادر مختلفه، عن طريق جهاز الامتصاص الذري اللهبه

نتائج الدراسة:

تراكيز العناصر الثقيله في المعلبات تراوحت في الرصاص بين (0.089- 17.1)ملغم/لتر، الكاديوم بين (0.019-0.322) ملغم/لتر، الزنك بين (2.05-10.6)ملغم/لتر، والنحاس بين (2.97-0.79)ملغم /لتر.

ت

كانت تراكيز الرصاص ضمن حدود المستويات المسموح بها دولياً، بينما نتائج الكاديوم كانت اعلى من المستويات المسموح بها، في حين الزنك كان ضمن المستوى المسموح به دولياً، وبالمقابل مستوى النحاس كان اعلى من المستوى المسموح به دولياً. هذه النتائج تتطلب الرصد المستمر لمستويات الكاديوم والسيطره على عملية التعليب في الاغذية المعلبه للمحافظة على سلامة الغذاء.

الخلاصة

تشير هذه الدراسه الى مزيد من الدراسات لتحديد تلوث الاغذية المعلبه بالعناصر الثقيله، واجراء المزيد من الدراسات المستقبليه التي تقيس مستويات العناصر في جسم الانسان واخذ عينات من الشعر والاذافر والدم، وربط هذه التراكيز مع تناول الفرد من المواد الغذائيه ودراسة اذا كان هناك ارتباط ذو دلالة مع العينات البيولوجية ام لا.