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INVESTIGATION OF TOXIC CHEMICALS IN PLANT-BASED MILK ALTERNATIVES

DISSERTATION

Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in the Graduate School
of Texas Southern University

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2021

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INVESTIGATION OF TOXIC CHEMICALS IN PLANT-BASED MILK ALTERNATIVES

By

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Milk is a dietary component consumed all over the world and is known to be essential for development in children. Despite all the nutritional advantages associated with milk consumption, several issues like lactose intolerance, and high lipid content, etc., negatively affects the perception of cow milk consumption, and has grown into general health concerns. Issues with cow milk consumption drove for a healthier alternative which led to plant-based milk alternatives (PBMA). Several studies have linked consumption of these PBMAs with other negative effects such as allergy, poor nutrient profile, and the presence of toxic chemicals. This study's main objective is to investigate the chemical profile of milk samples to detect the presence of toxic chemicals such as heavy metals and pesticides. This study includes the quantitation of essential elements present in milk samples.

This study uses instrumental analysis such as an inductively coupled plasma mass spectrometry (ICP-MS) to analyze the presence of heavy metals as well as essential elements in milk samples. For organic contaminants which include pesticides, the gas chromatography (GC) with electron capture detector (ECD) is used. Results obtained for heavy metals using ICP-MS showed trace concentration of Pb, Cd and Cr in the majority of the PBMA samples, specifically hemp milk (1.0 μg/l for Pb; 1.2 μg/l for Cr) and soy milk (0.4 μg/l for Cd), while heavy metal concentration for cow milk was below detection limit. Organochlorine pesticide (OCPs) residue limit was present in all milk samples. The concentrations for the OCPs banned by the Stockholm Convention were higher than the residue limits set by EPA and FAO-WHO. HCB, Mirex, toxaphene, and Chlordane had residue concentrations above 200 μg/kg. This is alarming because most banned OCPs are classified as carcinogens, even at trace concentrations. Overall, cow milk had no significant difference in pesticide residue limits compared to PBMAs like almond, coconut, soy, and oat. Almond, coconut, and soy milk were more fortified in essential elements than cow milk. However, heavy metal concentration in cow milk was below detection limits when compared to most of the PBMAs.

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LIST OF ABBREVIATIONS

µg/l- microgram per liter

ANOVA- analysis of variance

BDL- below detection limit

BHC- beta-hexachlorocyclohexane

Ca- calcium

CB- carbamate pesticide

CCB- continuing calibration blank

CCV- continuing calibration verification

Cd- cadmium

Cr- chromium

Cu- copper

DDT- dichlorodiphenyltrichloroethane

DRI- daily recommended/reference intake

ECD- electron capture detector

EPA- environmental protection agency

FAO- food and agriculture organization

Fe- iron

GC-MS- gas chromatography mass spectrometry

GMO- genetically modified organism

GPC- gel permeation chromatography

HCB- Hexachlorobenzene

ICB- initial calibrating blank

ICP-MS- inductively coupled plasma mass spectrometry

ICV- initial calibrating verification

K- Phosphorus

LCS- laboratory control samples

LLE- liquid-liquid extraction

MCL- maximum contamination limit

Mg- magnesium

mg/l- milligram per liter

Mn- manganese

Na- sodium

OCP- organochlorine pesticide

OPP- organophosphate pesticide

PAH- polyaromatic hydrocarbon

Pb- lead

PBMA- plant-based milk alternative

POPs- persistent organic pollutants

PPB- parts per billion

PPM- parts per million

PTFE- polytetrafluoroethylene

QUECHERS- quick, easy, cheap, rugged, and safe

SC- Stockholm Convention

SPE- solid phase extraction

WHO- World Health Organization

Zn- zinc

VITA

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CHAPTER 1

INTRODUCTION

Cow Milk

Milk is a dietary component consumed all over the world where it is known to be essential for development in children and it is important in the diets of children and adults (frieslandcampina, 2013; Heinrichs, Jones and Bailey, 2016). Some of the essential nutrients in milk such as magnesium, calcium, manganese, and macromolecules (e.g., proteins, vitamins, and fat) are required by the human body to accomplish daily tasks and sustain life (frieslandcampina, 2013). Milk is believed to boost energy, quench thirst, reduce fatigue and increase bone development to name a few. Milk is processed to be consumed commercially as a beverage which mostly comes from cows and goats. However, in some cases, it is used to produce other dairy products such as chocolates, cheese, and yoghurt etc. (frieslandcampina, 2013). With shortage of adequate nourishment and balanced diet around the world, the consumption of milk has been supported worldwide as a means of improving diet and attaining essential nutrients required to nourish the human body. The ministry or department of health of several countries have suggested an average of 1-2 daily servings of 240 ml of milk is required for children in the age range of 1-3 years old and 530 ml for children aged 4-5 years old (frieslandcampina, 2013). Quality of milk being consumed is usually measured via fat-protein ratio, which is also affected by the species of the herd. Here in the United States (US), a 1.34 herd species of Guernsey and a 1.28 herd species of Jersey yield the highest fat-protein ratio according to the United States Department of Agriculture (Heinrichs, Jones and Bailey, 2016). There are four other factors other than herd type that affect the quality of milk fat-protein ratio which are listed as the following:

1. Age of cattle (fat-protein % decreases with age).
2. Stage of lactation which is low at 20-50 days (about one and a half months) after calving but usually peaks at 250 days (about 8 months).
3. Seasons (milk quality is usually low in summer possibly because of agitating heat but peaks during fall/winter).
4. Health of the cattle (presence of infection such as mastitis can affect milk quality) according to Heinrichs, Jones and Bailey, (2016).

Despite all the nutritional advantages associated with milk consumption over the years, several issues that negatively affect the perception of cow milk consumption have grown into public health concerns. Some of these issues include but are not limited to allergic response, metabolism of lactose and hypercholesterolemia. The allergic response to milk is significant because milk is regarded as one of the highest allergens inducing dietary product when compared to shellfish, gluten, and nuts (Ho *et al.,* 2018; Lee *et al.,* 2018). The metabolism of individuals who are lactose intolerant were not able to metabolize lactose, a major constituent of cow milk (Ho *et al.,* 2018; Lee *et al.,* 2018; Sethi *et al*., 2016). Hypercholesterolemia is where the consumption of cow milk leads to high cholesterol (Ho *et al*., 2016; Sethi *et al*., 2016). Other issues include the increasing need to consume low calorie containing products for health reasons. The ecological footprint is an issue associated with commercial animal husbandry especially with high production of greenhouse methane. Cruelty activism such as the growing animal rights against cruel treatment of farm animals, excessive grazing of land covering grass and presence of other xenobiotic in milk matrix (Ho *et al.,* 2018; Lee *et al.,* 2018).

Plant-based Milk Alternatives

Issues with cow milk consumption after years of investigation drove for a healthier alternative according to Sethi *et al.,* (2016). The alternative led to plant-based milk which is a more accepted replacement for cow's milk in many homes. Plant-based milk alternatives (PBMA) can be classified into categories based on its plant source of origin. According to Sethi *et al.,* (2016), these alternatives include:

1. Cereal based alternatives: Corn milk and oat milk.
2. Nut based alternatives: Almond, coconut, and hazelnut.
3. Seed based alternatives: Sunflower, sesame, and hemp.
4. Legume based alternatives: Soy, Cowpea, and lupine.

For this research, the plant-based milk alternatives (PBMA) of focus are almond, coconut, rice, hemp, oat, cashew, and soy milk. These are the most widely consumed plant-based alternative milk, internationally.

Almond Milk

Almonds are rich in taste and has recently gained more popularity over soy as the alternative for cow milk especially in North America, Europe, and Australia (Vanga and Raghavan, 2017). Almonds are perceived to be a rich nutritional source for proteins, fiber, vitamin E, manganese, and monounsaturated fatty acids (MUFA) (Vanga and Raghavan, 2017; Sethi *et al.,* 2016). Almond milk is made by soaking and grinding almond nuts in excess water until a milky white liquid is obtained following filtration of the nut shafts. Commercially, the process is followed by high pressured homogenization and pasteurization to increase shelf life as well as stability (Vanga and Raghavan, 2017). The main bioactive components found in almond milk include α-tocopherol and arabinose associated with the health benefits of antioxidant protection against free radicals and prebiotic properties (Sethi *et al.,* 2016).

Coconut Milk

Coconut milk consumption is extremely popular amongst south-eastern Asia cuisines, both as a beverage and as an ingredient for making a variety of sweet and savory delicacies. Coconut milk is a rich source of fiber, vitamin C and E, iron, calcium, potassium, magnesium, and zinc (Sethi *et al.,* 2016). It exhibits anti-microbial and anti-carcinogenic properties, promotes brain development and is rich in lauric acid. The lauric acid is abundant in human breast milk. It helps to promote high density lipoprotein formation which in turn reduces dangerous low-density lipoproteins (Vanga and Raghavan, 2017; Sethi *et al.,* 2016). Coconut milk rarely presents any chance of allergic reactions, despite its many health benefits. The consumption of coconut milk is limited due to its elevated levels of saturated fats. Coconut milk is prepared by grating the white meat of the coconut and mixing homogenously until milky liquid is formed (Vanga and Raghavan, 2017).

Soy Milk

Soy milk has traditionally been associated as the pioneer plant-based alternative to cow milk as it considered to be rich in protein (~45%) and fat (~20%) and heavily incorporated in the diets of vegetarians (Vanga and Raghavan, 2017). Popularly consumed in South-Asia for thousands of years either as soy milk or as its derivative products such as miso, tempeh, tofu, or soybean paste (Vanga and Raghavan, 2017). Soy milk is considered also to be rich in isoflavones such as daidzein and genistein which have been significantly linked with anti-cancer properties (Vanga and Raghavan, 2017; Sethi *et al.,* 2016). However, the poor beany taste has seen its decline in demand over the years.

Rice, oat, hemp, and cashew are reliable sources of carbohydrates and fiber. Oat milk is popular in the United States and some parts of Europe (Vanga and Raghavan, 2017). However, these milk alternatives are fortified with sugar/sweeteners to make the taste palatable during consumption and this can lead to dental problems (Aydar *et al.,* 2020).

Disadvantages of PBMAs

Despite the advantages of some of these plant-based milk alternatives (PBMA), several studies have linked consumption of these plant-based derivatives with other negative effects such as allergic reactions associated with soy and almond consumption, improper nutritional balance for coconut milk with high saturated fats, and low protein and calcium content in comparison to cow milk (Vanga and Raghavan, 2017). Soy is considered as a source of allergen to some people and its seeds have high estrogen content (Vanga and Raghavan, 2017). In fact, some of these commercially available plant-based milk alternatives may need nutrient fortification to be comparable with cow milk (Vanga and Raghavan, 2017; Sethi *et al.,* 2016). In addition, PBMAs requires a high amount of water to homogenize the grated shafts/seeds/beans of these plant-based products into milk, which means that high ecological footprint is associated with commercial production of plant-based milk alternatives (Vanga and Raghavan, 2017). Other factors include the genetic make-up of these commercially farmed plants which are being used for milk production that has led to the debate surrounding ethical application of genetically modified organisms (GMO) versus organic farming. In addition, there are several studies done that identified contaminants in PBMA, for example *Streptococcus mutans* biofilm contamination in soy and vanilla almond milk by Lee *et al.,* (2018). The investigation by Ferrer *et al.,* (2009) shows the presence of phytoestrogens and its trimethylsilyl derivatives present in soy milk. This is also supported by another study conducted by Dwyer *et al.,* (1994) that suggest soy contains phytoestrogens.

Because PBMAs are on the rise, it is necessary to investigate the environmental aspects of each plant-based milk alternative to determine if it is beneficial for human dietary intake and if there should be a concern for the presence of toxic metals and organics. For this research, the PBMAs and cow milk were analyzed quantitatively for essential elements and heavy metals using inductively coupled plasma and mass spectrometer (ICP-MS) and organochlorines using gas chromatography electron capture detector (GC-ECD).

Instrumental Analysis

Several studies have identified the presence of inorganic contaminants in cow milk such as lead, arsenic and cadmium which are toxic metals (Lutfullah *et al.,* 2014; Meshref *et al.,* 2014). With organics, the contamination with pesticide residues, hormones and PAHs are mostly detected in cow milk (Avancini *et al.,* 2012; Rusu *et al.,* 2016; Matraszek-Zuchowska *et al.,* 2016; Grova *et al.,* 2002). However, for plant-based milk alternatives (PBMA), there is a gap in the knowledge of research to determine if there is a presence of toxic organics for safe consumption. Therefore, a comparative study of cow milk and PBMAs to address contaminants whether it is safe for consumption. To analyze the chemical profile of PBMAs as well as cow milk, the use of instrumental analysis will be vital to this study.

Inductively Coupled Plasma and Mass Spectrometer (ICP-MS) for Elemental Analysis of Milk

An instrument employed for analysis of inorganic contaminants present in milk is the inductively coupled plasma mass spectrometer (ICP-MS) shown in figure 1. The application for the ICP-MS was first introduced in 1980 by Houk *et al.* The inductively coupled plasma is the most common used plasma ion source where it is also the most adaptable trace, ultra-trace elemental and isotope instrumental analysis available now (Becker, 2007). The advantages of the ICP-MS include high sample throughput, high sensitivity, accuracy, and multi-element determination (Becker, 2007). Mass spectrometric systems work by utilizing ion sources that produces a sufficient intensity ion beam that interact with sample, separating ions based on mass to charge ratio, ion separation (vacuum chamber) and ion detection system. According to Becker (2007), the principle of ion source in mass spectrometry, is to evaporate solid samples, dissolve or vaporize liquid samples, and to atomize gaseous molecules to generate ions for analysis.

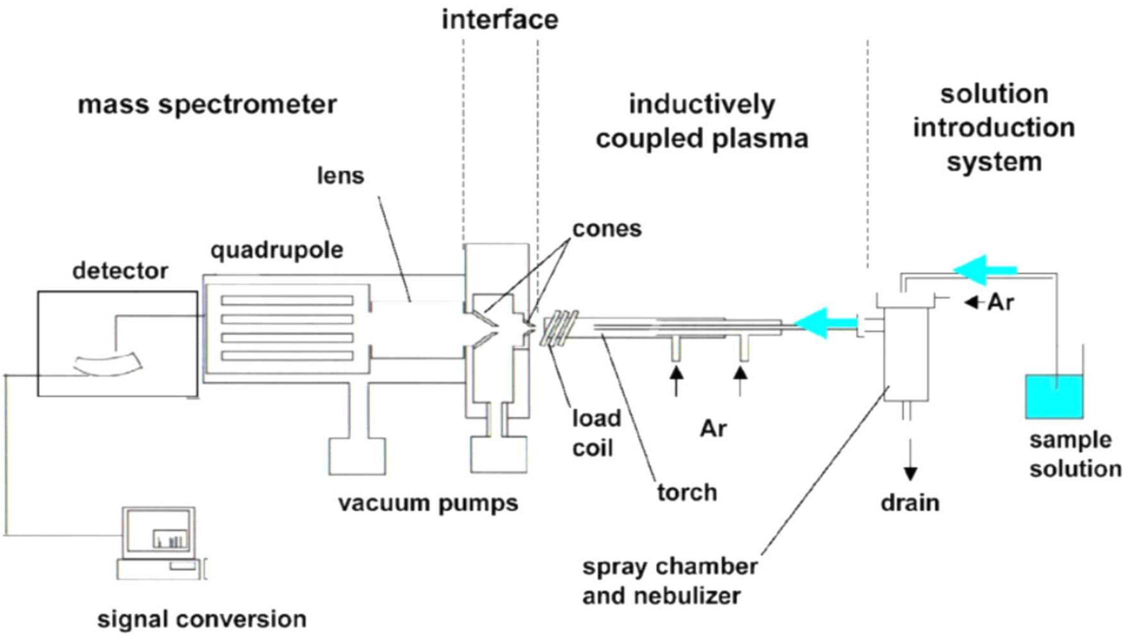


Figure 1. A Schematic of Inductively Coupled Plasma (Gilstrap and Allen, 2009).

The ICP-MS works by introducing the sample through the spray-nebulizer system where it is carried by inert gas, argon, to the inductively coupled plasma where analytes are atomized by heat generated from the load coil. Excitement from the plasma leads to sample dissolution and evaporation into atoms. The atoms will collide with Ar^+, excited Ar, or electrons in the electrostatic collimator to form ions that will enter the MS. The ions are introduced into the quadrupole mass vacuum via the double cone interface to separate ions according to its mass-to-charge ratio then the ions reach the detector. The detector converts the signal into an electrical response (Becker, 2007). An example of the ICP-MS is in figure 1.

Gas Chromatography and Electron Capture Detector (GC-ECD) for Organic Contaminant Analysis

The instrumentation used for the analysis of organic contaminants such as pesticide residues, hormones, etc. in milk samples is gas chromatography and electron capture detector (GC-ECD). The GC-ECD series 6890 (Agilent Technologies, US) is used primarily for the analysis of complex organic-biochemical mixtures. The GC component separates volatile and semi-volatile compounds while the coupled ECD is responsible for detailing ionic charge on these compounds such that it can be identified and quantitized (Hussain and Maqbool, 2014). The GC operates via a carrier gas which is usually inert and non-reactive with the sample, known as the mobile phase. The stationary phase is microscopically lined with a layer of lipid/polymer/inert solid support of the glass or metal tube called a column. In the mobile phase, inert gas such as helium is used to sweep the sample through the column where components of the sample begin separating from one another where some would elute faster than others (Hussain and Maqbool, 2014).

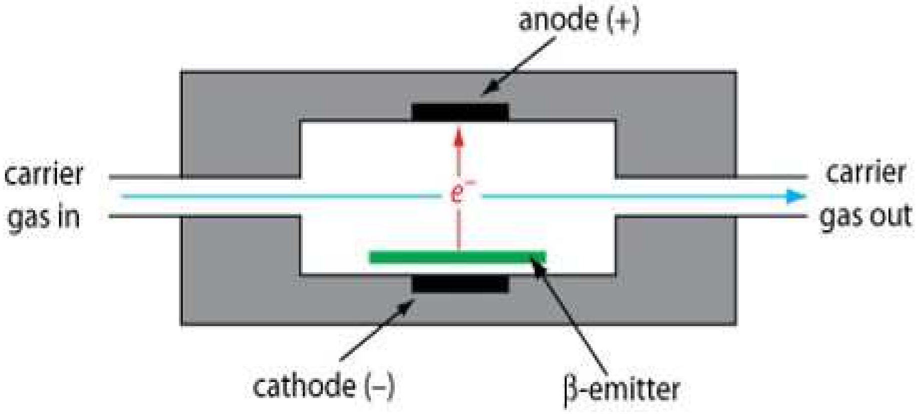


Figure 2. A Schematic Diagram of a GC-ECD (Harvey, 2013).

The ECD is a detector that receives the sample matrix from a carrier gas (nitrogen for this study) after it elutes from the stationary phase lined column and into the detector chamber. The detector is lined with a β-emitter which ionizes the sample matrix. There is also a cathode and anode side that separates the ions that are formed from the sample matrix. The ionized sample matrix is then separated into electrons which are captured by the anode (+), that generates a current for measurement, and sent to a signal converter which produces peaks for matrix electrons being measured.

Metals of Interest

Essential and Toxic Elements

The essential elements are nutrients taken in by the body to sustain life and carry out cellular function. It is important to note that the dose at which intake occurs is also vital to the organism in question as lower dosage may lead to nutrient deficiencies and higher doses may elicit toxic effects (Fairweather-Tait and Cashman, 2015). Due to this phenomenon, it is important to classify essential elements based on the size of the dose required for intake (Fairweather-Tait and Cashman, 2015). This leads to categorization of macronutrients and micro/trace elements (Prashanth *et al.,* 2015). According to various nutritional charts including those from Fairweather-Tait and Cashman, 2015; Olivares and Uauy, 2009, macronutrients are listed as Ca, Na, Mg, P, K and S. These macro-elements are required in larger amounts (greater than 100 mg/day) because of its involvement in several intracellular and extracellular functions which includes structural rigidity (bones, teeth, and cellular membrane), information transmission (neural, intracellular phosphorylation and hormonal), electrolyte balance and muscle contraction (Fairweather-Tait and Cashman, 2015). Macro-elements are required in higher concentrations (greater than 100 mg/day) but at a balanced rate because the effects on human health. It can be catastrophic when there is a deficiency of these nutrients to support basic biochemical and molecular function as well as when these nutrients are present in excessive amount beyond the recommended DRI (Prashanth *et al.,* 2021; Fairweather-Tait and Cashman, 2015 and Olivares and Uauy, 2009). Micro/trace nutrients are those elements that are required for vital cellular functions, but at a trace intake level (less than 100 mg/day). According to nutritional charts, the list of known trace nutrients includes Cu, Zn, Fe, Mn, Al, and F (Fairweather-Tait and Cashman, 2015 and Olivares and Uauy, 2009). The biochemical and molecular functions associated with trace nutrients include enzyme cofactors, oxygen transport, gene expression and redox reaction (Prashanth et al., 2021; Fairweather-Tait and Cashman, 2015 and Olivares and Uauy, 2009). Because micronutrients are required in trace concentration intake compared to macronutrients, the biomarker for detecting deficiency is limited. Therefore, micronutrient deficiency is only discovered when disease or cellular function failure manifests (Olivares & Uauy, 2009).

Enriched milk is a reliable source of balance diet especially within the demographics that consume milk/dairy products the most which are Children and elderly. The presence of these essential elements in fortified milk gives an opportunity for children to sustain rapid development and high cellular activities while the elderly gets to maintain cellular function required to support life. The ICP-MS analysis of the various milk samples was done to yield the respective concentrations of essential elements.

The doses to which these essential elements are added as nutrient fortifiers in milk matter because excess concentrations render these elements toxic and may leave residues in the system after consumption (Lutfullah et al., 2014; Pilarclyzk et al., 2013). With regards to PBMAs, essential nutrients in high concentrations can be due to farming activities, production processing, and preparation. There are other elements that are toxic and pose detrimental effects in the human system (Lutfullah et al., 2014; Pilarclyzk et al., 2013). Elements like iron, zinc, copper, lead, arsenic, and mercury are known as heavy metals which have a density of 6 g/cm^2 and these heavy metals once in the system can cause variety of effects such as forming covalent bonding with organic macromolecules to alter certain cellular activities. In addition, elevated levels of these metals in milk will lead to excess adsorption of these metals and these heavy metals can bioaccumulate over a prolonged period leading to more chronic ailments and system dysfunction (Pilarclyzk et al., 2013). Irregular inorganic fertilizer applications can pollute the soil where the plants used for PBMAs are grown, processed, packaged, and transported. These PBMA products may also contribute to pollution with toxic metals. Since children are the highest consumers of milk, it is necessary to determine and regulate the concentration of toxic elements that may be present in PBMAs. In conclusion, this study hypothesizes that certain residues of organic and inorganic constituents may be present in PBMA, regardless of quality control protocols established in the production line and this study aims to determine the concentration of these contaminants by investigating the chemical profile analysis of several commercially available PBMAs brands available at the local grocery store.

Pesticide Residue

The milk samples are analyzed for pesticide residues which are the primary organic contaminants of interest along with phytoestrogens and poly-aromatic hydrocarbons PAHs. Organic pesticides are a mixture of active and inert ingredients that are used to rid and control disease carrying vectors (e.g., insects and rodents) and prevent the loss or damage of crops (WHO, 2019). The chemical components in pesticides are designed in such a way that the active ingredient is made to target pest of interest (animals, microbes, or weeds) and the inert ingredient helps to improve the pesticides longevity, ease of use, ease of penetration into pest, and protection against degradation (EPA, 2019). Pesticides are usually classified in two major forms which includes the chemical makeup of the pesticide and the target of the pesticide (WHO, 2019; EPA, 2019). Classification based on its target is the most popular insecticides targeting insects, herbicides targeting unwanted plants and fungicides targeting fungal growth. Other popular classifications based on chemical makeup are the class of organo-chlorine pesticide (OCPs), organo-phosphorus pesticide (OPPs), pyrethroids and carbamate (CB) (Akhtar & Ahad, 2017). Some of the pesticides in the OCP and OPP (organo-phosphorus pesticide) class have been identified as persistent organic pollutants also known as the "dirty dozen" and have been banned from application by the regulatory bodies in many countries. These pesticides are considered a public health threat because of the following characteristics:

1. It is not easily degradable.
2. It is associated with long distance travel away from the source point.
3. Some pesticides display non-targeted detrimental effects such as endocrine disruption and developmental disorders especially in children.
4. Pesticides have the immense potential to bioaccumulate across the food chain by transferring from organism to the other (Akhtar &0 Ahad, 2017).

Pesticides get introduced into our systems usually through ingestion (food consumption and drinking water) as well as partially through inhalation upon application. The same can be considered for farm animals as the grass or silage it feeds on may be due to pesticide application and the barn has a presence or residue of pesticide to ward off pest (Akhtar and Ahad, 2017).

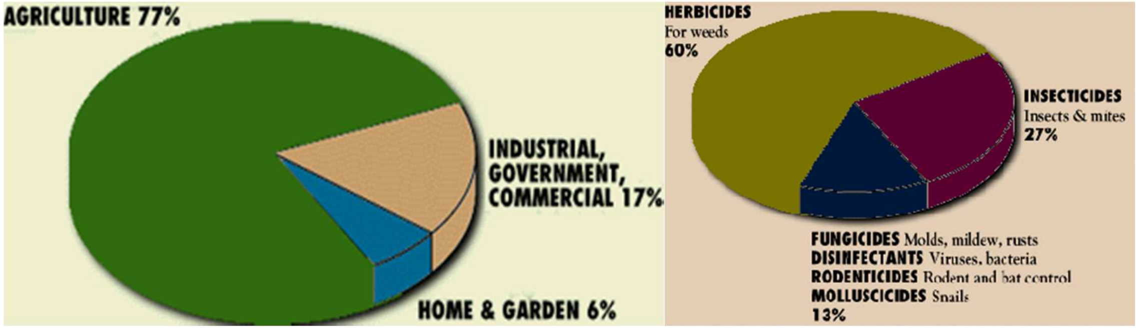


Figure 3. The rate of pesticide used per sector in the US and commonly used pesticide (EPA, 2017).

As seen in figure 3, agriculture as an individual sector contains the most pesticide application over the other sectors and herbicides are the most common pesticides applied by commercial farmers. With regards to cow milk, pesticide residues can make its way to cow milk as grass and silage are sprayed and insecticides are applied in barns to ward off disease carrying vectors. Therefore, we hypothesize that commercial farming of crops to produce PBMAs will also be treated with pesticides and pesticide residues will be present in trace quantities in these milk alternatives.

CHAPTER 2

LITERATURE REVIEW

Recently, the consumption of cow milk has been declining based on several concerns ranging from high calorie and fat content, allergies, and consumer's own inability to metabolize lactose. While these issues continue to persist, some research studies have shown other validation for concerns of consuming cow milk. Pilarczyk *et al.,* (2013) used an inductively coupled plasma emission atomic spectrometry where the study showed that the concentrations of heavy metals in raw cow milk differed across two different cattle species which are Simmental specie and Holstein-Friesian specie in the northwestern Lubuskie province of Poland. According to the study, the level of cadmium (Cd) and lead (Pb) were significantly higher ($P<0.001$) in Holstein-Friesian species than in Simmental species (0. 0040 ppb in milk samples from Holstein-Friesian compared to 0.0035 ppb for Cd in milk samples from Simmental). In addition, 0.00412 ppb of Pb was detected in Holstein-Friesian while 0.0366 ppb of Pb was detected in the latter. The study also mentioned no significant difference in the level of nutritional elements such as Ca, Mn, and Se. A study conducted in Poland by Sujka *et al.,* 2019 consisted of a total of 40 dairy products (milk, butter milk, cream, kefir, yoghurt, cheese spread, and cottage cheese samples) were analyzed for the presence of toxic/heavy metals purchased from various locations in Poland. The study used ICP-MS to analyze the concentration of heavy metals and the result showed that 50% of the samples contained Cd concentration ranging from 0.0067 to 0.0058 mg/kg. The concentration of Pb was highest in samples from the Silesia region due to high industrialization in that locality.

Cheese spread and cottage cheese had higher concentrations of heavy metals analyzed compared to other samples (Sujka *et al.,* 2019). According to Mesherf, Moselhy and Hassan (2014), lead and cadmium are quite common air pollutants from industrial emissions which is it major source. When these emissions enter soil, water, and plant (which are considered as the primary source for life sustenance), the toxic metals become incorporated with the food chain through a process known as biomagnification. The consumption of these toxic metals in food can pose detrimental health effects to the human body, especially in children and elderly adults with vulnerable body defense and who are ironically among the largest consumers of milk and dairy products.

In agriculture, the use of pesticide on crops occurs in three stages which are preharvest, postharvest and storage. The pre-harvest is when the seeds are germinating, sprouting, and yielding produce such as fruits, grains, or leaves. Post-harvest is after seeds have yielded a mature crop/plant with fully developed ripe fruits, nuts, grains, etc. The storage phase is where the matured crop produce has been plucked from farmland and saved in preparation for next planting season or sold for consumption. The fate and transport of these pesticides from lower plants to higher animals on the food chain comes through the process of feeding also known as bioaccumulation. In addition, pesticides can be introduced to farm animals through direct spraying to minimize pest infestation. Whether pesticide contaminates feed or the direct spraying of cow with pesticides, both lead to the persistence of pesticide residues in animal products such as milk, meat, and eggs (Ahktar and Ahad, 2017). A study by Pastor Ciscato, Gerbara and de Souza Spinosa (2002) using a combination of gas chromatography with electron capture detector (ECD), nitrogen/phosphorus detector (NPD), and flame photometric detector (FPD) analyzed 132 samples of milk (38 raw milk and 94 pasteurized milk) for pesticide residue contamination using DFG S19 multi-residue analytical method for sample preparation. The results showed the presence of HCH (alpha isomers) and endosulfan (alpha and beta isomers) residues in 0.76% and 10.6% of the samples analyzed. However, organophosphorus pesticides, carbamates, herbicides, and fungicides were not detected. Even though, the use of HCH is banned and the use of endosulfan is restricted, its residues were still present in cow milk. A study by Avancini *et al.,* (2012) consisted of 100 samples of bovine milk gathered from various locations in Brazil. These samples were analyzed for the presence of organochloride pesticide (OCPs) residues. Samples were analyzed using solid phase extraction (SPE) and residue concentrations were identified using a GC-ECD. From the 100 samples analyzed, the following OCPs were identified as 44% Aldrin, 36% DDT, 34% Mirex, 32% endosulfan, 17% chlordane, 14% dicofol, 11% heptachlor and 11% dieldrin. Also, according to Avancini *et al.,* (2012), some of the concentrations of these residues present in the samples were above the established limits set by regulation for residue limits in food.

Aside from the presence of pesticide residues in milk samples, another commonly known organic contaminant is hormone/growth factor. Hormones and growth factors are part of biological molecules found in blood plasma for transport to areas where it is needed. Hormones can also exist in the mammary gland because it can be synthesized locally or penetrate through the plasma because of its high lipophilic properties (Matraszek-Zuchowska *et al*., 2016). Another explanation for the fate of hormones in milk could be from veterinary application to animals to either hasten development, to meet profit target, or for therapeutic purposes to prevent animal diseases (Matraszek-

Zuchowska *et al*., 2016). The continuous presence of natural and synthetic hormone residues in milk can cause detrimental health effects among different population groups where these effects can range from target receptor blocking to hormone related cancer (Ganmaa and Sato, 2005). The study by Ganmaa and Sato (2005) using stepwise multiple regression analysis correlated the incidence rates of certain types of cancer (breast, ovarian and corpus uteri) to food intake. It was found that milk was the second most linked food ($r = 0.817$) to breast cancer after meat ($r = 0.827$). Milk is denoted as first in relation to ovarian cancer ($r = 0.779$) and then followed by animal fat and cheese ($r = 0.717$ and $r = 0.697$, respectively). Lastly, milk and cheese were revealed to have the highest significant contribution to corpus uteri cancer incidences ($r = 0.861$) (Ganmaa and Sato, 2005). Maruyama, Oshima and Ohyama (2010) stated in their study that consumption of cow's milk for men and children suppressed the secretion of gonadotropin, decreased testosterone secretion which is important for sexual maturation in pre-pubertal children; however, there was an increase in the urine of serum estrone (E1), estradiol, estriol and pregnanediol. The exogenous estrogens in cow milk are easily adsorbed in young men and children and as a result, it leads to suppression of significant role-playing endogenous hormones such as testosterone, serum luteinizing hormone and follicle stimulating hormone (Maruyama, Oshima and Ohyama 2010).

As stated earlier, this review of literature shows validation of the need for an alternative for cow milk, which has led to the increase in the demand for PBMAs and the presumption that PBMAs are a healthier replacement for cow milk. On the contrary, not much research is available that has investigated the presence of hazardous chemicals in milk that are plant-based. Agriculture has the biggest usage of pesticides; therefore, application in the commercial farming of plants for milk production is inevitable. The use of machinery, fertilizer, manure, and other sources of toxic metals, including abundance in farm soil can also contribute to the possible presence of these metals in plant-based milk.

CHAPTER 3

METHODOLOGY

Rationale

In chapter one and two, milk consumption has been associated with but not limited to endocrine disruption, obesogenic conditions (hypercholesterolemia) and high calorie composition which may subsequently pose detrimental effects to the health of consumers who are mostly children (Matraszek-Zuchowska *et al.,* 2016; Meshref *et al.,* 2014**)**. During the process of milk production, efforts are made to rid the final product off microbial contamination through pasteurization and quality control protocols. However, trace concentrations of hormones, heavy metals and pesticide that may have been introduced through the process of feeding of cattle, veterinary care for cattle and farming process for plant-based alternatives. These contaminants may be present in the final product and can be unknowingly consumed. Some research studies have linked the presence of steroid hormones in cow milk from hormones fed to cows to increase development (Courant *et al.,* 2008; Fischer *et al.,* 2011). While other studies have linked the presence of phytoestrogens in plant-based milk, especially soy milk (Ferrer *et al.,* 2009; Dwyer *et al.,* 1994). It is imperative to investigate quality of milk production, as the processing of cow milk is at its highest around winter and early spring for lactating cows during which quality may be affected because of seasonal changes, making fresh grazing often impossible. This indicates that cows are fed with graze that may have been harvested, processed with pesticides, and stored for the winter season. Another reason to investigate the quality of milk is that barns where cattle are kept may have routine pesticide applications to hinder the build-up of disease carrying vectors. In chapter one, bioaccumulation was mentioned as a pesticide characteristic. Bioaccumulation is a process where an organism is directly exposed to a toxicant from their immediate environment, either through ingestion, inhalation, or dermal contact. Plant-based milk may also be subjected to pesticide application due to farming practices leading to bioaccumulation of residues before and after harvest; hence, the need to investigate the quality of PBMAs.

The quality of milk needs constant investigation because of long term exposure to trace amounts of pesticides and toxic metals in a regular dietary component such as milk can lead to several detrimental health effects such as cognitive and developmental disorders, oncogenesis, and endocrine disruption (Matraszek-Zuchowska *et al.,* 2016). Therefore, continuous analysis, detection, and regulation of processed products are necessary before consumption (Fischer *et al.,* 2011). Being that children are the highest consumers of milk, studies have shown most of these inorganic and organic contaminants are present in cow milk; however, literature for the analysis of PBMAs are little to none. Therefore, it poses a gap in the knowledge that needs to be addressed. Following this rationale, the research question is: “Should there be a need for analyzing the toxic chemicals in plant-based milk alternatives in order to determine its safety for consumption”? It is hypothesized that a toxic chemical analysis and comparison with cow milk would help determine a safe source of milk for daily consumption from either cow milk or plant-based milk. To test our hypothesis, the following specific aims are investigated:

- Aim 1: To analyze the concentration of essential elements (macro/micro) in milk samples (cow versus plant-based) using an inductively coupled plasma mass spectrometer.
- Aim 2: To analyze the concentration of heavy metals present in milk samples (cow milk versus plant-based milk) using an inductively coupled plasma mass spectrometer.
- Aim 3: To analyze the presence of organochlorine pesticide residues in milk samples (cow milk versus plant-based) using a gas chromatography and electron capture detector (GC-ECD) for analysis.

Specific Aim 1 and 2: Analysis of Essential Elements and Toxic Metal Presence in both Cow Milk and Plant-based Alternatives using ICP-MS

The use of instrumentation is vital for the quantitative analysis of inorganic elements in milk and the specific instrumentation employed for this study is the inductively coupled plasma mass spectrometers (Agilent, Series 7900). The ICP-MS was used to investigate specific aim 1 and 2 using sample preparation and materials.

Sample Preparation and Materials

Since little to no literature has described the possible inorganic contaminants that are present in plant-based milk alternatives when compared to cow milk. This investigation would bridge this gap in knowledge. One of the study’s objectives is to analyze quantitatively the presence of essential nutrients and toxic inorganic contaminants present in nine different milk samples. A total of 24 commercially available milk brands were purchased as samples from local stores in Houston, Texas which are H.E.B store on Shepard drive and Wholefood’s store on Westheimer Road. After purchasing, the 24 samples were prepared for microwave digestion according to the milk protocol for CEM Mars 6 (CEM, US). Each sample was prepared by transferring 4 ml aliquot of each milk sample into a 15 ml tube and 10 ml of 70 % HNO_3 reagent grade was dispensed into the same tube. Each milk brand sample listed in table 3 was prepared in triplicates. The samples were then transferred into a CEM Mars 6 digestion vessel and placed in a turn table stand. Temperature was set for 200 ^oC with a ramp of 15 mins, a hold set at 15 mins, with a pressure at 800 psi and power that is set between 900-1000 (W). After digestion, sample triplicates were transferred to 15 ml tubes individually. The digested samples were cooled. The multi-element reagent standards (Sigma Aldrich) for the inductively coupled plasma mass spectrometry was prepared where the elements where at a 1000 mg/l stock concentration. The standard consists of Ag, Al, As, Au, Ba, Be, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, Hg, Ir, K, Li, Mg, Mn, Na, Ni, Os, Pb, Pd, Pt, Rb, Re, Rh, Ru, Se, Sr, Te, Tl, U, V, Zn, and Sc. Through serial dilution from the stock, the calibration standard concentrations ranged from 0.05 ppb, 0.1ppb , 1 ppb, 10 ppb 50 ppb and 100 ppb, respectively while the internal calibration standard (ICV) and the internal calibration blank (ICB) were also prepared at 50 ppb. Before placing sample in the ICP-MS (Series 7900 by Agilent technologies, US) sample chamber, the sample were further de-acidified from 70% to 2% using Equation 1. The de-acidification process involved dilution with Millipore 18 Ω distilled water. Table 1 and table 2 shows the ICP-MS parameter settings and sample uptake rate for analyzing milk samples. See table 1 for the settings of the ICP-MS for calibration prior to analysis. See table 2 for ICP-MS sample uptake and probe rinse and time settings. The ICP-MS analysis of essential nutrients and heavy metals was repeated twice to acquire data accuracy and data was analyzed using

©Microsoft Excel (Microsoft, US) to plot graphs while statistical significance was determined using a pooled T-Test ($P<0.05$) between milk brands and averaged to represent each milk sample.

Milk Samples

Milk samples were purchased from H.E.B. and Wholefoods, both located in Houston, Texas. Samples included cow milk, plant-based milk alternatives (rice, oat, hemp, almond, soy, coconut, and cashew) and coffee creamer. See table 3 for list of milks and coffee creamer analyzed. Codes were assigned to protect manufacturer's brand. The CEM MARS 6 sample preparation for digestion starts with adding 4 ml of sample to10 ml of 70 % HNO_3 where this is a total of 14 ml.

$$\text{Eq 1. } C_1V_1=C_2V_2$$

Equation 1 was used to deacidify the sample from 70 % acidification (V_1) to 2% at 10 ml. It was determined that 0.3 ml of the sample was needed and then micro pipetted into a 15 ml vial containing distilled water. The sample is diluted with 9.7 ml of distilled 18 Ω Millipore water. The prepared sample was vortexed for 1 min then arranged in the ICP-MS sample holder and labelled during batch creation. Different concentrations of multi-element standards are also arranged in the allotted holder. Thereafter, the ICP-MS is calibrated to begin analysis. Helium gas was used as a carrier gas for the samples and initial calibration verification (ICV) and initial calibration blank (ICB) both at a concentration of 50 ppb were set for after every 10 samples, a quality control protocol to ensure results generated fall within ±10 % of expected concentration range. The ICV and ICB checks are followed by the continuing calibration verification (CCV) and continuing calibration blank (CCB) for the same reason. Results generated from the ICP-MS will be exported to ©Microsoft Excel for further statistical analysis and representation. The sample volume of 4 ml and the volume for ICP-MS of 10 ml leads to a 2.5 factor that is applied to the ICP-MS readings. The experiment was repeated twice, and a single factor analysis of variance (ANOVA) was done to compare the mean for milk samples with more than two brands, while an S-pooled, T-Test in Equation 2 was applied for milk samples with only two brands (See Appendix D). ICP-MS was used to analyze individual milk brands in triplicates to obtain concentrations of elements (essential and heavy metals) of interest.

$$\text{Eq } 2\text{: } t = \frac{\overline{X_1}-\overline{X_2}}{\sqrt{(S^2\frac{1}{n_1}+\frac{1}{n_2}}}$$

The resulting concentrations of the milk brands were averaged, standard deviation recorded, and results compiled to represent each milk sample for comparison. ANOVA analysis of some milk brands showed that the concentrations of essential elements and heavy metals varied; however, the goal of this study is to compare concentrations in milk samples and not the brands. The average calculated concentrations were obtained and used to represent each milk sample (See Appendix D). P value less than 0.05 indicates there are no significant differences between milk samples.

Table 1. The ICP-MS Parameter Settings

Parameter	Settings
Plasma	HMI-4
RF Power (W)	1600
Sampling depth	8 mm
Carrier gas flow (L/min)	0.8
Lens tune	Autotune
Helium flow rate (ml/min)	4.3
H_2 Flow rate (ml/min)	6
Number of elements	24 elements, 6 internal standards

Table 2. The Sample Uptake and Rinse Time

	Time taken (s)	Pump speed (%)
Sample load	8	50
Stabilize	15	5
Probe rinse sample	20	5
Probe rinse standards	20	5
Loop rinse	30	50

Specific Aim 3: Analysis of Pesticide Residues Presence in Milk Samples

This study analyzed nine different milk samples as shown in table 3 below. A total of 24 milk brands were commercially purchased for investigation.

Table 3. List of Milk Samples and Brands Purchased for Analysis.

Milk sample	Brand + Code
Cow Milk (CM)	(CM1) (CM2) (CM3) (CM4)
Almond Milk (AM)	(AM1) (AM2) (AM3) (AM4)
Coconut Milk	(CCM1)

(CCM)	(CCM2) (CCM3)
Soy Milk (SM)	(SM1) (SM2) (SM3)
Oat Milk (OM)	(OM1) (OM2)
Rice Milk (RM)	(RM1) (RM2)
Hemp Milk (HM)	(HM1) (HM2)
Cashew Milk (CAM)	(CAM 1) (CAM2)
Coffee Creamer (CRM)	(CRM 1) (CRM 2)

After acquiring samples from the local grocery stores, samples were stored in 6 °C refrigerators and later transferred into individual 1 L amber glass bottles. SW-846-8081 B was the preparation and extraction method utilized for the analysis of OCP residues in milk samples (USEPA, 2007). The detector used was an electron capture detector (ECD) for the GC (GC ECD series 6890 by Agilent Technologies, US). According to USEPA (2007), 8081 B method determines the concentration of OCPs in extracts from liquid matrices and solids. Based on this method, samples are prepped using hexane-acetone (1:1) solvent that is pesticide grade quality. Extraction was done following the 3546 methods for microwave digestion, this method is used to extract water insoluble/slightly soluble organic compounds from various matrices. Microwave energy is used to generate elevated temperature and pressures in a closed vessel, as described in the method for specific aim 1 and 2. Standards used for calibrating the GC-ECD instrument are the EPA 500 series- 17 analyte count multi-pesticide component, and reagents used were of analytical grade. The stock, composite, calibration, internal and surrogate solutions prepared were stored at 6 °C or less in polytetrafluoroethylene (PTFE) sealed containers. After extraction, sample cleanup was done using method 3630, using silica gel as a regenerative adsorbent in column chromatography to separate OCP components from other interferants with different chemical polarity. Samples were introduced into the GC using a dual one injection port split into two open tubular silica fused capillary columns. Dual columns were used to ensure that the sample analytes are adequately separated.

Results generated are confirmed by identifying if the sample extract peaks fall within the daily retention time window. Another form of confirmation is by using a column with a different stationary phase component, then determine the agreement between the data generated by the two different columns. Quality control is applied by including a method blank, matrix spike and a laboratory control sample (LCS) within each analytical sample batch. The LCS consists of an aliquot of a control matrix like that of sample matrix with similar weight or volume. The LCS is spiked at the same concentration as the sample. The LCS verifies that the instrument can perform a clean matrix analysis. The EPA 500 series standard used for this study contained the following compounds and its respective concentration: 500 µg/ml of trans-chlordane, 500 µg/ml of cis-chlordane, 1000 µg/ml of endosulfan II, 1000 µg/ml of delta-BHC, 1000 µg/ml of 4,4'-DDE, 1000 µg/ml of endrin aldehyde, 1000 µg/ml of alachlor, 1000 µg/ml of 4,4'-DDT, 1000 µg/ml of heptachlor epoxide - isomer b, 1000 µg/ml of endosulfan I, 1000 µg/ml of heptachlor, 1000 µg/ml of endrin, 1000 µg/ml of atrazine, 1000 µg/ml of 4,4'-DDD, 1000 µg/ml of beta-BHC (beta-HCH), 1000 µg/ml of gamma-BHC (lindane), 1000 µg/ml of endosulfan sulfate, 1000 µg/ml of dieldrin, 1000 µg/ml of methoxychlor, 1000 µg/ml of alpha-BHC (alpha-HCH) and 1000 µg/ml of aldrin The parameters for the GC-ECD were set where a dual split column was used. Column 1 from Restek Corporations, US, was the Rtx-CL pesticides column with internal diameter of 0.32 mm, film thickness of 0.32 µm. Column 2 from Restek Corporation, US, was the Rtx-CL pesticides 2 with internal diameter of 0.32 mm, and film thickness of 0.25 µm. The carrier gas was nitrogen which was set at a flow rate of 4.3 ml per minute. The run time was 9.44 minutes, and the pressure was set to 12.2 psi. The temperature was 250ºC for the injector, 340 ºC for the detector, 120 ºC for the initial temperature, with a hold time of 3 minutes, and 310 ºC for the final temperature, with a hold time of 1 minute.

Final concentration and data analysis were performed following Method 8081 B protocol to quantify sample analytes. Data generated was referenced through an ECD ionic charge library and dilution factor was applied using the 8081 B protocol (USEPA, 2007). Data generated for the individual milk brands were averaged, the standard deviation recorded, and compiled to represent each milk sample. A one-way factor ANOVA was done for milk samples with more than two brands. The S-pooled T-Test was done for milk samples with just two brands. Graphs were compiled using MS Excel spread sheet.

CHAPTER 4

RESULTS AND DISCUSSION

As previously discussed, cow milk and PBMAs are consumed by humans because these milks consist of nutrients known as essential elements which are vital for cellular function and sustaining life. These essential elements can be further categorized into macro- and micronutrients where the higher concentration of the nutrients is believed to be enriched. The essential elements contained in milk contributes as a source for a balance diet where concentrated nutrients lead to more enrichment. In addition to these essential elements, there is also a need to be certain that toxic elements are not present or below the maximum contamination limit (MCL) which are established by regulatory standards. The toxic elements can be dangerous for consumption even at low concentrations and can accumulate which can lead to detrimental effects to human health. Another concern in the production of milk is the presence of residue concentrations of organic contaminants such as pesticides which is also addressed and investigated to ensure that the milk is safe for consumptions. The regulatory standards that are used to compare results obtained for essential elements are based on EPA (EPA, 2002) and National Academy of Science (NAP, 2019). The organochlorine pesticides (OCP) in plant-based milk are compared the concentration to the residue limit standard set by EPA's maximum contamination limit (MCL) and United Nations' Food and Agriculture Organization's (FAO) Codex Alimentarius.

Essential Elements

Specific Aim 1

Milk samples were analysed using an ICP-MS to investigate the concentrations of essential elements and heavy metals. The experiment was run twice and the results from both runs were averaged. The averages were tabulated using ©Microsoft Excel sheet. Results for the essential elements were grouped into macro- and micronutrients with tables highlighting milk samples with the highest concentrations of elements in ppb and graphs showing visual representation of concentration difference between each sample for individual elements.

Macronutrients

Table 4 represents the average concentration of macronutrients analyzed in milk samples in ppb. The milk samples with the highest concentrations of macronutrients compared to other milk samples are highlighted in bold fonts in table 4. Macronutrients are nutritionally vital elements required for consumption above 100 mg/day, and elements in this category include sodium, calcium, magnesium, phosphorus, sulfur, and potassium (Prashanth *et al.,* 2015). If the dose for these elements is not maintained, it can lead to the development of severe health conditions. The data from table 4 will be used to generate graphs that will visually compare the concentrations of each macronutrients in cow milk and PBMA samples. These concentrations will be compared to the dietary reference intake (DRI) value recommended by the National Academy of Science (NAP, 2019). The error bar indicates percentage error, see Appendix C and D for concentrations and relative standard deviation (%)

Table 4. List of Macronutrients and Concentrations (ppb) in Milk Samples Using an ICP-MS.

Samples	**Na (ppb)**	**Mg (ppb)**	**Ca (ppb)**	**K (ppb)**
Cow Milk	11206.6	2333.5	12389.0	54905.1
Almond	**22749.6**	1371.2	**27563.3**	45346.5
Soy	10398.4	**4697.6**	11235.7	81900.2
Coconut	6575.0	1530.0	16149.5	21488.5
Creamer	8738.6	1129.7	9027.6	**90128.3**
Rice	15684.1	435.5	8221.7	5060.4
Oat	14794.7	973.6	18155.4	89815.9
Hemp	17433.1	3401.9	9575.0	15544.0
Cashew	9394.4	1782.3	1659.1	11058.3

*numbers in bold fonts represent highest concentration of each macronutrient in milk samples.

The Concentration of Sodium (Na) in Milk

Data from figure 4 suggests that sodium is significantly higher in almond milk at a value of 22.7 ppm. Other milk samples such as hemp milk, rice milk and cow milk had concentrations of 17.4 ppm, 15.7 ppm, and 11.2 ppm, respectively. Coconut milk had the lowest concentration of Na at 6.5 ppm. None of the milk samples had concentrations above 100 mg based on the dietary dose recommendations for a macronutrient. However, Na is supplemented through other foods to achieve the dietary dose recommendations.

Na is macronutrient and serves as a principal cation in extracellular fluid transport (Olivares and Uauy, 2009). The presence of sodium in the body is necessary in keeping acid-base balance, maintaining intra/extra cellular fluid volume and osmolality as well as cellular membrane electrochemical gradient and muscle contraction (Meyers, Hellwig and Otten, 2006). The average dietary reference intake (DRI) for Na in children ranging from age 1 to 8 years old is 900 mg/day, while the average DRI for male and female adult is 1450 mg/day according to the National Academy of Science (NAP, 2019), a private, non-profit institution that aids government agencies such as the United States Centre for Disease control, United States Department of Agriculture, and Canadian National Institute of Health, to solve problems and advocate policies related to science, medicine and engineering. The major source of Na intake in the body is in the form of salt (NaCl) which is used in food seasoning and not necessarily via milk consumption. Na consumption can be of concern because there are certain health conditions associated with the deficiency of sodium. Some of these health conditions include central nervous system dysfunction, dehydration, hyponatremia, neuromuscular spasms, metabolic acidosis, and headaches. Also, excess concentration of Na can also lead to development of hypertension (Olivares and Uauy, 2009; Fairweather-Tait and Cashman, 2015). The results obtained indicate that these milk samples have Na concentrations that are below the DRI. For an element to be considered a macronutrient, it must be consumed at a concentration of a 100 mg/kg/day where 100 mg/kg is 100 ppm in terms of the ICP-MS analysis. This concentration cannot be obtained through plant-based milk samples alone. Therefore, individuals who are dependent on milk or plant-based milk products need another supplement for sodium intake. With regards to milk consumption, the milk samples do not provide the recommendation of 100 mg/kg/day dietary reference by National Academy of Science (NAP, 2019).

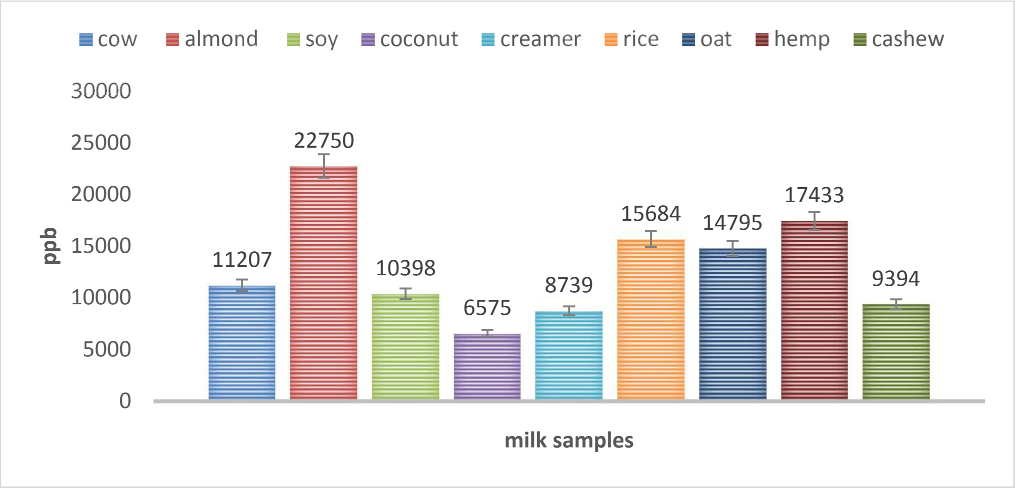


Figure 4. Na concentration (ppb) in Milk Samples from ICP-MS Analysis.

The Concentration of Magnesium (Mg) in Milk

Figure 5 shows that Mg contains the highest concentration in soy milk with a value of 4.7 ppm than the other samples. Hemp, cow, and cashew milk follow the soy milk sample at concentrations of 3.4 ppm, 2.3 ppm and 1.8 ppm, respectively. Rice milk had the lowest concentration of Mg with 0.435 ppm.

Mg is a macronutrient that plays a vital role as a cofactor for over 300 enzymes, helps in the enzymatic metabolism of carbohydrate, lipid, and nucleic acid, assists in the skeletal development and mineralization and cellular permeability (Olivares and Uauy, 2009). Mg is also vital for the regulation of K and Ca levels in the body (Meyers, Hellwig and Otten, 2006). The average DRI for Mg in children age range 1-8 years old is 145 mg/day, while the average DRI for both male and female adults is 385 mg/day and 312 mg/day, respectively according to National Academy of Science (NAP, 2019). The concentration of Mg across the milk samples is below the DRI and is not a concern. The primary dietary source of Mg is high fibrous fruits and vegetables (Meyers, Hellwig and Otten, 2006). Although, Mg in milk and PBMAs are lower than the recommendations, it can contribute to the dietary needs of children and adults. The health conditions that may rise because of Mg deficiency include hypertension, type 2 diabetes onset, cardiovascular related problems, muscle cramps and seizures (Meyers, Hellwig and Otten, 2006; Olivares and Uauy, 2009). Excess Mg has been associated with diarrhea, abdominal cramps and metabolic alkalosis (Meyers, Hellwig and Otten, 2006).

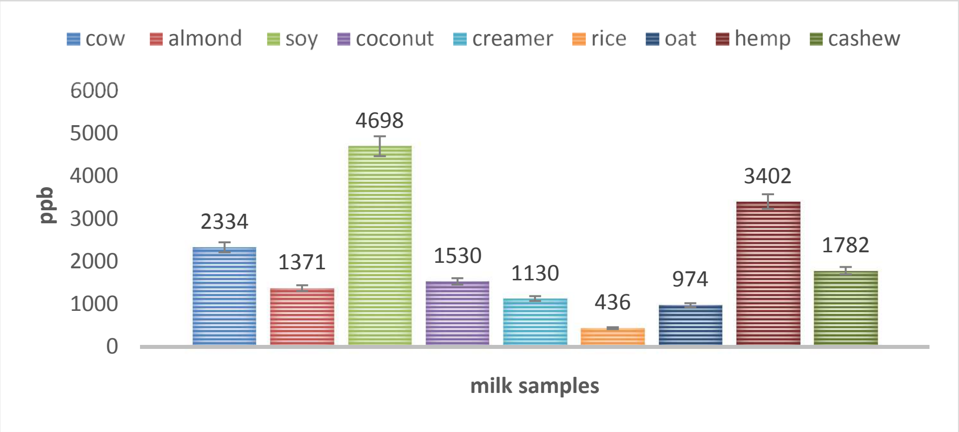


Figure 5. Mg concentration (ppb) in Milk Samples from ICP-MS Analysis.

The Concentration of Calcium (Ca) in Milk

In Figure 6, almond milk contains the highest concentration of Ca with a value of 27.6 ppm than other milk samples. Consequently, other milk samples that proceed the almond milk such as oat milk, coconut milk, and cow milk have concentrations of 18 ppm, 16 ppm and 12.4 ppm, respectively proceed the almond milk. Cashew milk had the lowest Ca concentration at 1.6 ppm.

Calcium is a macronutrient that plays a vital role in the skeletal and dental development. Calcium is also essential in muscular, neuromuscular function (Meyers, Hellwig and Otten, 2006). The average dietary recommended intake for Ca according to the National Academy of Science in children between 1-8 years old is 850 mg/day, while the average DRI for adult male and female is 1100 mg/day (National Academic Press, 2019). The concentration of Ca across all milk samples falls below the DRI and is not a concern. The primary dietary source of calcium are dairy and dairy products such as yogurt and cheese, while other sources include calcium rich vegetables such as Chinese-cabbage, kale, and broccoli (Meyers, Hellwig and Otten, 2006). Health conditions associated with calcium deficiency include osteoporosis, elevated risk of bone fracture and poor intestinal absorption. And excess amount of Ca can lead to conditions such as hypercalcemia, kidney stones and renal insufficiency (Meyers, Hellwig and Otten, 2006; Olivares and Uauy, 2009).

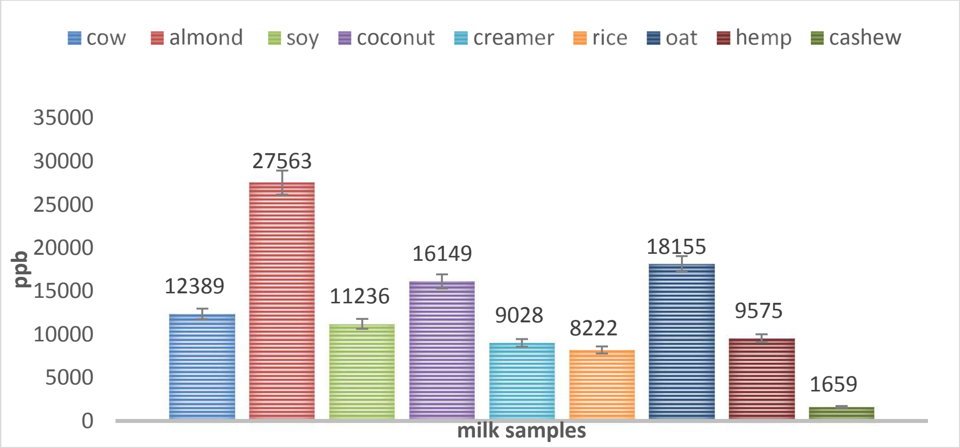


Figure 6. Ca Concentration (ppb) in Milk Samples from ICP-MS Analysis.

Micro-nutrients

According to Prashanth *et al.,* 2015, micronutrients are elements required in trace doses, usually below 100 mg/day. Therefore, these essential elements are sometimes referred to as trace elements. Elements that fall in the micronutrient category include iron, copper, zinc, manganese, cobalt and selenium. Data was obtained from the average of two experimental runs. Data in table 5 shows concentration of micronutrients in cow milk and PBMAs. The cow milk and PBMA samples with the highest concentrations of micronutrients are highlighted in bold fonts. Table 5 was used derive figures that show a visual comparison of concentrations between cow milk and PBMA samples for each trace element. The concentrations of the micronutrients are compared to the dietary reference intake recommended by the National Academy of Science (NAP, 2019) for daily consumption.

Table 5**.** A list of Micronutrients and the Concentration (ppb) Analysed from the Milk Samples using an ICP-MS.

Samples	**Fe (ppb)**	**Cu (ppb)**	**Zn (ppb)**	**Mn (ppb)**
Cow Milk	13.2	36.6	**145.7**	1.2
Almond	77.9	52.4	27.9	16.5
Soy	**213.9**	**105.4**	129.0	**83.9**
Coconut	100.3	43.4	56.0	10.1
Creamer	24.8	61.9	114.0	1.3
Rice	41.6	36.1	24.6	16.0
Oat	68.7	50.5	46.6	41.5
Hemp	118.0	40.8	114.8	62.1
Cashew	86.9	53.3	68.2	24.5

*numbers in bold fonts represent highest concentration of each micronutrient in milk samples.

The Concentration of Iron (Fe) in Milk

In Figure 7, soy milk contains the highest concentration of Fe with approximately 0.214 ppm. Other milk samples such as hemp, coconut and cashew had concentrations of 0.12 ppm, 0.1 ppm and 0.087 ppm, respectively. Fe is micronutrient because its recommended dietary intake should be below 100 mg/day (Prashanth *et al.,* 2015). Cow milk had the lowest concentration of iron with 0.013 ppm.

Fe plays a vital role as a component of various molecular structures in the body such as in enzymes like cytochromes, in protein such as myoglobin in the muscle tissues and with hemoglobin found in erythrocytes to support oxygen transport (Meyers, Hellwig and Otten, 2006). Metabolized iron can exist in multiple states of oxidation with ferric (Fe^{3+}), ferrous (Fe^{2+}) and ferryl (FeO_4^{2-}) states being the most common (Meyers, Hellwig and Otten, 2006). Iron is also essential for executing many cellular mechanisms of action such as oxidative phosphorylation, metabolism of neurotransmitters and synthesis of DNA (Olivares and Uauy, 2009). The average dietary recommended intake of iron according to National Academy of Science for children within the age range of 1-8 years is 8.5 mg/day while in adults, the average ranges from 12.5 mg/day for females and 8.9 mg/day for males (National Academic Press, 2019). The concentration of Fe for all milk samples investigated in this study is below the DRI and is not a concern. The primary source of iron comes from the consumption of meat, poultry and fish which makes heme-iron readily bioavailable for absorption while nonheme-iron can also be obtained from vegetables, fruits, and whole grain foods. The nonheme-iron is not readily absorbed through digestion according to Meyers *et al.* (Meyers, Hellwig and Otten, 2006). A health condition associated with iron deficiency is anaemia. Iron can lead to conditions such as hemochromatosis and iron-poisoning when excess amounts are above the recommendation is consumed regularly (Wada., 2004; Meyers, Hellwig and Otten, 2006; Olivares and Uauy, 2009).

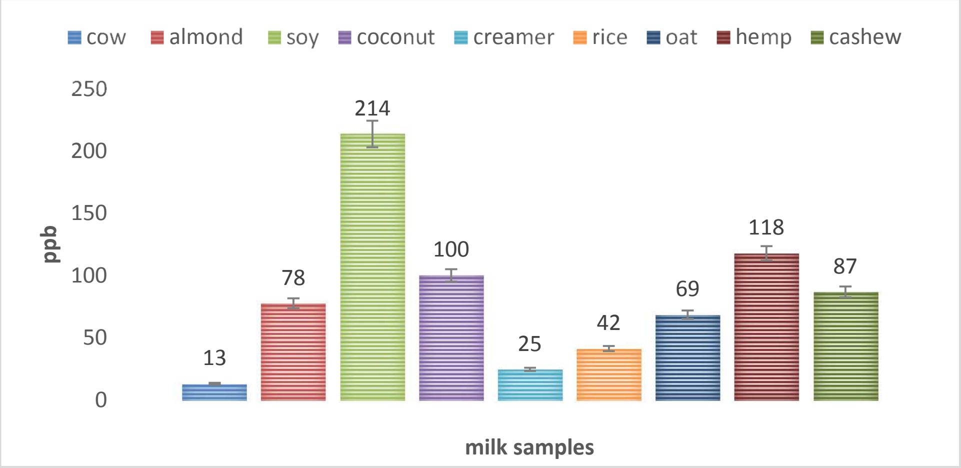


Figure 7. Fe Concentration (ppb) in Milk Samples using ICP-MS Analysis.

The Concentration of Copper (Cu) in Milk

In Figure 8, soy milk contains the highest concentration of Cu with a value of 0.11 ppm. Consequently, other samples such as coffee creamer, cashew and almond milk had concentrations of 0.062 ppm, 0.053 ppm and 0.052 ppm, respectively. Rice milk had the lowest concentration at 0.036 ppm. Copper's daily recommendation is less than 100 mg/day which makes it a micronutrient.

Copper plays a vital role in the formation of biochemical molecules where it is a component of metalloenzymes or as plasma Cu which participates in several oxidizing and reduction reactions (Wada, 2004; Meyers, Hellwig and Otten, 2006). Examples include diamine oxidase used in allergic reaction response to inactivate histamines and ferroxidases and a copper enzyme found in plasma that helps bind iron to transferrin (ferrous iron oxidation reaction) (Wada, 2004; Meyers, Hellwig and Otten, 2006;

Olivares and Uauy, 2009). Copper is metabolized in the small intestine where it is adsorbed and then trafficked through the MNK; ATP7A pathway, commonly known as the Menkes P-type ATPase (Meyers, Hellwig and Otten, 2006). Most of the copper content found in the human body is deposited in the skeleton and muscle, while the liver maintains a high plasma copper concentration. The average dietary recommended intake by National Academy of Science for copper in children with age range of 1 to 8 years is 390 µg/day (390 ppb), while 865 µg/day (865 ppb) is the average for both males and females (National Academic Press, 2019). Thus, the concentrations of Cu in milk samples are below the DRI and pose no concern to public health. Primary dietary sources of copper are from plant such as nuts, seeds, and grains. Copper is readily available in the soil, while seafood, organ meat (e.g., liver and kidney), and dietary supplements are also viable sources to support dietary needs (Meyers, Hellwig and Otten, 2006). The deficiency of copper in human is quite rare except in malnourished infants. It has been observed that other metals in excess doses can compete with copper for participation in cellular function especially metals like zinc and iron (Wada, 2004; Meyers, Hellwig and Otten, 2006). Health conditions associated with copper deficiency includes osteoporosis, neutropenia, and leukopenia. An excess intake of copper can lead to acute or chronic copper toxicity (depending on the duration of exposure), which may result in health defects such as liver damage, gastrointestinal illness, abdominal cramps, pain, and nausea. Diseases that could result due to excess copper are such as Wilson's disease, idiopathic copper toxicosis (ICT) and Indian childhood cirrhosis (ICC) (Wada, 2004; Meyers, Hellwig and Otten, 2006; Olivares and Uauy, 2009).

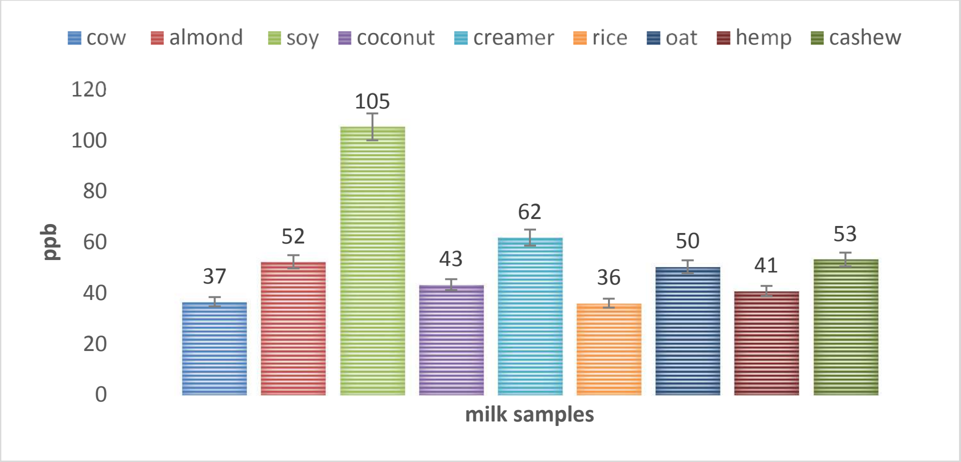


Figure 8. Cu Concentration (ppb) in Milk Samples using ICP-MS Analysis.

The Concentration of Manganese (Mn) in Milk

In Figure 9, soy milk contains the highest concentration of Manganese with a value of 0.084 ppm. Other milk samples such as hemp, oat, and cashew milk with concentrations of 0.062, 0.042 and 0.025 ppm, respectively. Cow milk had the lowest concentration at 0.0012 ppm. Manganese is required in doses below 100 mg/day (Prashanth et al., 2015), and is vital in trace amounts. It is essential in bone formation, and the metabolism of amino acids, cholesterol, and carbohydrate. It is also a necessary component of metalloenzymes such as arginase and manganese superoxide dismutase (Wada, 2004; Meyers, Hellwig and Otten, 2006; Fairweather-Tait and Cashman, 2015).

There's no official reference intake reported for Mn; however, the average DRI for children ranging from ages 1-8 is 1.4 mg/day, while the average recommendation for both male and females are 2.3 mg/day and 1.8 mg/day (National Academic Press, 2019). Concentrations of Mn across milk samples are below the DRI and it's of no concern to one's health. The primary dietary sources of manganese intake include plants, beverage, and dietary supplement pills (Meyers, Hellwig and Otten, 2006). Several elements compete with manganese include calcium, iron and phytate. The presence of these competitive elements at a high concentration can lead to no absorption of Mn (Meyers, Hellwig and Otten, 2006). People who suffer from manganese depletion appear to develop the following health conditions; scaly dermatitis, impaired reproductive function, hypocholesterolaemia, and impaired growth due to poor skeletal development (Wada, 2004; Meyers, Hellwig and Otten, 2006). Manganese toxicity resulting from excess intake can lead to conditions related to neurotoxicity. As seen in patients who inhale manganese dust, it tends to develop central nervous system defects with symptoms like that of Parkinson's disease (Wada, 2004; Meyers, Hellwig and Otten, 2006).

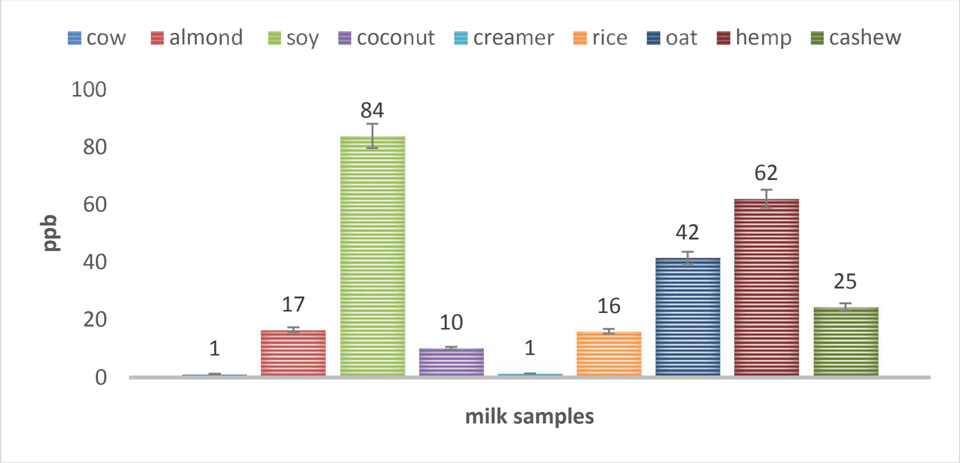


Figure 9. Mn Concentration (ppb) in Milk Samples using ICP-MS Analysis

Toxic Elements

Specific Aim 2

According to Prashanth *et al.,* 2015, elements that are not essential and whose presence can induce toxicity are referred to as toxic elements. Heavy metals fall under this category of toxic elements. As earlier described, heavy metals are a group of naturally occurring metals with large atomic weight (63.5 – 200 g/mol) and density five times higher than water (Srivastava and Majumder, 2008; Tchounwou *et al*., 2014). Aside from being naturally occurring, heavy metals also have multiple agricultural, domestic, and industrial applications which increases the potential exposure to these heavy metals and the hazardous effects these metals have on human health (Srivastava and Majumder, 2008; Tchounwou *et al*., 2014). Heavy metals do not easily biodegrade and can persist either in its active state or inactive state for a long time in the environment, until naturally occurring activities such as atmospheric deposition, metal corrosion, leaching into surface or ground water, or sediment re-deposition takes place (Bradl, 2005; Tchounwou *et al*., 2014). Anthropogenic activities such as mining and industrial manufacturing as well as waste discharge play an active role in exposure to these heavy metals (Bradl, 2005). Heavy metals such as arsenic, lead, cadmium, chromium, and mercury are known to exhibit significant levels of toxicity. These are of public health concern because of the carcinogenic mechanism of action when exposed (Beyersmann and Hartwig, 2008; Tchounwou *et al*., 2014). For this study, the toxic elements of interest are Lead (Pb), Cadmium (Cd) and Chromium (Cr), due to the relative standard deviation (less than 20%) and concentration obtained from the ICP-MS that were more statistically acceptable (see Appendix C). Other heavy metals were not reported due to low limit of detection. Heavy metals that are naturally occurring were analyzed for its presence in cow milk and PBMA samples which may arise from plant nutrient uptake or animal grazing on contaminated soil. Agricultural application, poor wastewater treatment and proximity to industrial point sources are considered anthropogenic factors for heavy metal presence in milk samples.

Table 6 was obtained from the compiled average of two experimental runs (see appendix D). Table 6 shows the concentration of heavy metals in cow milk and PBMA samples. The highest concentrations of individual heavy metals in milk samples are highlighted in bold fonts and milk samples with concentrations too low to be detected are identified as BDL (below detection limit) in table 6. The data in table 6 was used to derive figures for visual comparison in concentrations between cow milk and PBMA samples. These concentrations are also compared to the maximum contamination limit value set by United States Environmental Protection Agency (EPA) for drinking water, a component for comparing PBMAs, as there are no set regulations for heavy metals specifically for milk (EPA, 2002).

Table 6. List of Toxic Elements and Concentrations in ppb across Milk Samples Analyzed Using an ICP-MS.

Samples	**Pb (ppb)**	**Cd (ppb)**	**Cr (ppb)**
Cow Milk	0.4	**BDL**	**BDL**
Almond Milk	0.4	**BDL**	**BDL**
Soy Milk	0.7	**0.4**	0.6
Coconut Milk	0.3	**BDL**	0.2
Coffee Creamer	0.7	**BDL**	0.5
Rice Milk	0.4	0.1	0.7
Oat Milk	0.4	0.2	0.7
Hemp Milk	**1.0**	0.2	**1.2**

*BDL represents below detection limit

*numbers in bold fonts represent highest concentration of heavy metal in milk samples

The Concentration of Lead (Pb) in Milk

A bluish-grey metal that found within the earth's crust is one of the most significant heavy metal of public health concern (Srivastava and Majumder, 2008; Tchounwou *et al*., 2014). Although, the application and usage of lead has dwindled over the years, the exposure to lead contamination persists. In the past, the application of lead was included in manufacturing of metal products (solders and pipes), ammunition, fossil fuel burning, paints, lead-acid batteries, and x-ray protection component to name a few (Bradl, 2005; Tchounwou *et al*., 2014). As of 2004, an estimated 1.5 million metric ton of lead was used across various sectors in the United States of America (Tchounwou *et al*., 2014). Human exposure to lead primarily comes from inhalation of lead contaminated dust and aerosol and ingestion of lead contaminated food and water (Beyersmann and Hartwig, 2008). Once absorbed into the body, the metabolism of lead is dependent on the age and physiology of the person with children being the most vulnerable. Lead is metabolized first in the kidney, followed by the liver, and ends with other soft organs like the heart and brain. The skeleton and the nervous system are at the most risk towards lead contamination (Tchounwou *et al*., 2014). Exposure to lead contamination among pregnant women's unborn baby and young children is extremely dangerous. This is because lead toxicity can easily overwhelm unborn babies and children developing systems leading to several cognitive and carcinogenic health defects (Beyersmann and Hartwig, 2008). Upon absorption, lead displays a unique characteristic which aids in its exertion of toxicity, where this characteristic enables it to inhibit or mimic the action calcium in biochemical pathways. This includes binding with proteins and enzymes, which alters the configuration and function (Beyersmann and Hartwig, 2008). The effect of acute or chronic exposure to lead among children and some adults has been documented and are said to include diminished intelligence (low-IQ), poor attention span, growth retardation, speech, and hearing impairment, decreased sperm count, spontaneous miscarriage, and damage to vital organs such as kidney, brain, liver, and the central nervous system (Srivastava and Majumder, 2008; Tchounwou *et al*., 2014).

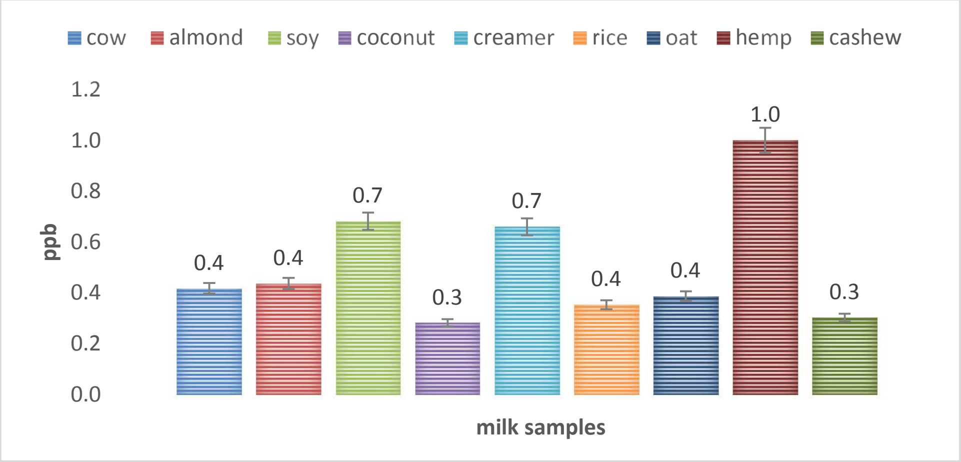


Figure 10. Pb Concentration (ppb) in Milk Samples using ICP-MS Analysis.

In Figure 10, hemp contains the highest concentration of lead with a value of 1 ppb. Soy milk and coffee creamer have the second highest concentration where both are 0.7 ppb. Cow milk had the same concentration of Pb as almond, rice, and oat milk, while coconut and cashew milk had the lowest concentration of lead with 0.3 ppb. There are no set regulations for heavy metals in food; however, the contamination limit set by EPA for lead in water which is a component used in manufacturing milk is 1.5 ppb (EPA, 2021). The concentration of Pb in all the milk samples should not be of concern because it is below the regulatory standard set; however, it may lead to the development of acute or chronic conditions if the lead-daily intake is higher than limits set by EPA.

The Concentration of Cadmium (Cd) in Milk

Cadmium is widely deposited among sedimentary rocks and marine phosphates (Kabata-Pendias, 2000). Application of cadmium is significantly tied to the industrial sector and its use ranges from alloy production, pigment formation to battery construction (Tchounwou *et al*., 2014). Because of public health concerns, the use of cadmium has been drastically reduced in developing countries as in lead (Beyersmann and Hartwig, 2008). The main sources of potential exposure to cadmium are via inhalation and ingestion of Cd contaminated dust, aerosol, food, and water. One may be exposed by skin absorption, but it is rare (Tchounwou *et al*., 2014). With regards to Cd exposure, bioavailability is a primary reason for high Cd concentration in the human body with foods such as dried seaweed, shellfish, mushroom, liver, and cocoa where cocoa holds the highest concentration (Kabata-Pendias, 2000). Cd metabolism occurs primarily in the kidneys, while absorption occurs through the gastrointestinal tract (GIT) which causes significant localized damage to renal function as well as to the gastrointestinal tract as exposure and toxicity persist (Tchounwou *et al*., 2014). There is no regulatory limit set on cadmium in food; however, Cd toxicity is not to be taken lightly based on the international agency for research in cancer and the U.S (United States) national toxicology program. Cd is a human carcinogen that targets the pulmonary system (Beyersmann and Hartwig, 2008).

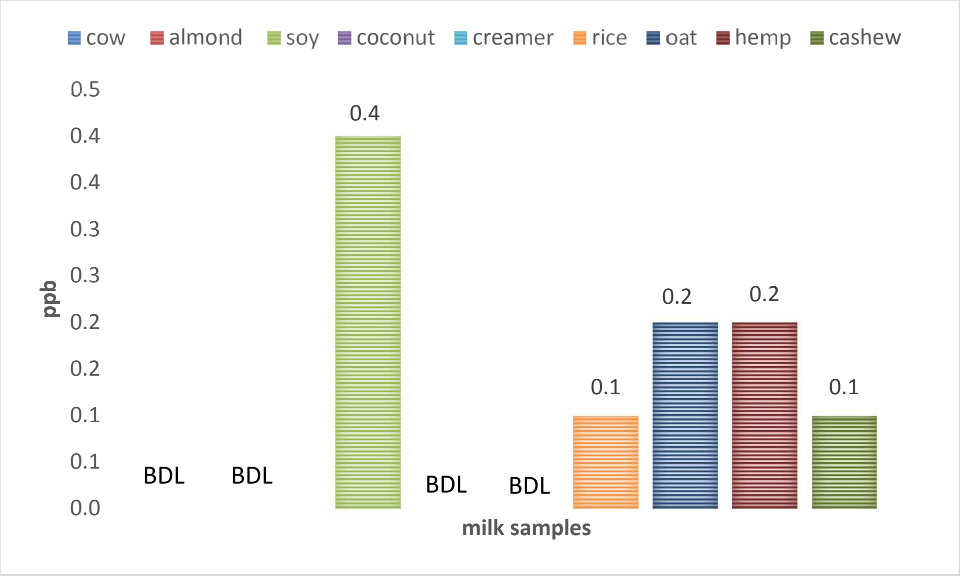


Figure 11. Cd Concentration (ppb) in Milk Samples using ICP-MS Analysis.

In Figure 11, soy had the highest concentration of Cd at 0.4 ppb, while cow milk, almond coconut and creamer had concentrations that were below detection limits (BDL) of the ICP-MS. The maximum contamination limit set by EPA for Cd concentration in water is 5 ppb (EPA, 2021). The concentration of Cd present in milk samples is below the regulatory standard limit which is not of concern. However, continuous exposure to these concentrations may accumulate into severe health complications if exposed above the limits set by EPA.

The Concentrations of Chromium (Cr) in Milk

It exists naturally in the earth crust under different oxidation states such as Chromium II and Chromium VI, however, it can also exist as Cr (III) in its ore state, otherwise known as Ferro-chromite (Kabata-Pendias, 2000; Tchounwou *et al*., 2014). Of all the heavy metals, chromium has the most diverse sources of exposure, either from natural or anthropogenic activities because of its existence in multiple oxidation states where it can easily move between environmental matrices like air, water, and soil

(Kabata-Pendias, 2000; Tchounwou *et al*., 2014). Industrial application of Cr includes the production of stainless-steel welding, metal processing, and chrome pigmentation (Tchounwou *et al*., 2014). Cr (VI) or hexavalent Cr is produced as a by-product/waste from industrial activities where Cr (VI) is toxic to human health and has been classified as a human carcinogen (Meyers, Hellwig and Otten, 2006; Beyersmann and Hartwig, 2008). Aside from anthropogenic activities, Cr (VI) has been found naturally in ground and surface water (Tchounwou *et al*., 2014). The primary sources of exposure to Cr are from industrial activities and ingestion of Cr contaminated food and water. Chromium once ingested via food or water or inhaled via occupational dust could initiate the action of insulin and lipid regulation (Meyers, Hellwig and Otten, 2006; Tchounwou *et al*., 2014). Chromium toxicity may also lead to DNA strand break, lipid peroxidation, chromosomal abnormalities and other conditions that may develop into carcinogenicity (Beyersmann and Hartwig, 2008).

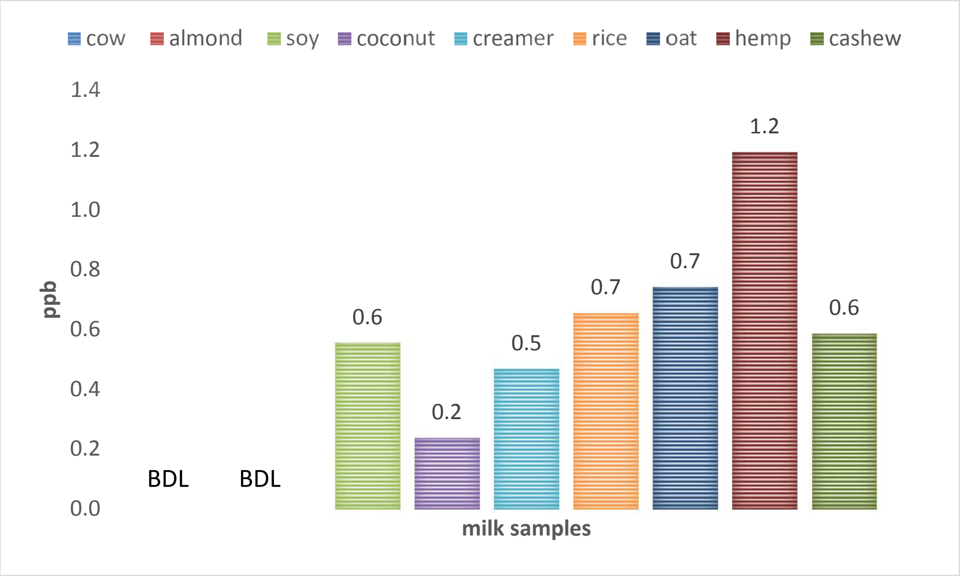


Figure 12. Cr Concentration (ppb) in Milk Samples using an ICP-MS Analysis.

In Figure 12, hemp contains the highest concentration of Cr at 1.2 ppb. Cow milk and almond milk are the only milk samples that had concentrations that were below detectable limits (BDL) in the ICP-MS. The maximum concentration of Cr in water set by EPA is 100 ppb (EPA, 2021). The concentration of Cr present in milk samples is not of public health concern because it is below the 100 ppb (0.1 mg/l) as set by regulatory standards. However, continuous exposure to these Cr above the regulatory standards when consuming milk may lead to health complications.

Table 7 shows a list of the elements detected using ICP-MS, milk sample with the highest concentration and the intracellular function these elements play upon absorption. It also shows the dietary recommended intake (DRI) for essential elements and maximum contamination limit (MCL) for heavy metals.

Table 7. List of Heavy Metals and Concentrations (ppb) in Milk Samples

Element	**Milk sample with highest concentration**	**Intracellular function**
Na	Almond (22.7 ppm) DRI: 1450 mg/day	Vital for Osmolality, acid-base balance, and intracellular fluid volume.
Mg	Soy (4.7 ppm) DRI: ≥350 mg/day	Vital as enzyme cofactor, intracellular permeability and skeletal development and mineralization.
Ca	Almond (27.6 ppm) DRI: 1100 mg/day	Vital for skeletal and dental development. Plays a role in muscular function.
K	Coffee creamer (90.1 ppm) DRI: 1150 mg/day	pH balance regulation, intracellular catalytic activator, energy generation

		and storage
Fe	Soy milk (0.214 ppm) DRI: ≥10.7 mg/day	Vital as enzyme cofactor like in cytochrome. Protein cofactors like in myoglobin. Oxygen transport
Zn	Cow milk (0.146 ppm) DRI: ≥9.5 mg/day	Vital as a catalyst for enzymatic reaction, structural components of enzymes and proteins. Can induce apoptosis via PKC activity
Cu	Soy (0.105 ppm) DRI: 865 µg/day	Component of metalloenzymes. Influences certain Redox reactions
Mn	Soy (0.083 ppm) DRI: 1.9 mg/day	Component of metalloenzymes, vital for amino acid cholesterol and carbohydrate metabolism
Pb	Hemp (1.0 ppb) MCL:0.0015 ppb	Calcium inhibition/mimicry, cognitive impairment, growth and development inhibition, carcinogenicity.
Cr	Hemp (1.2 ppb) MCL: 0.1 ppm	Potentiate insulin regulation, DNA strand breaks, lipid peroxidation and chromosomal abnormalities
Cd	Soy (0.4 ppb) MCL: 0.005 ppm	Pulmonary adenocarcinoma, renal function deregulation, GIT injury

Pesticides

Specific Aim 3

It is customary practice in agriculture to apply pesticides in the commercial planting of crops like fruits, nuts, grains, and vegetables, to prevent loss, infection, death, or depreciation of quality. Livestock are also introduced to pesticide residue from application within their vicinity to prevent proliferation of disease-causing vectors or parasites. It is common knowledge that pesticides are synthetic chemicals used to treat crops or livestock vicinity and are classified based on the type of pest it targets, e.g., insecticide for insects, herbicides for weed, rodenticide for rodents and fungicides etc. (Akhtar and Ahad, 2017). However, pesticides can also be grouped based on the key/active ingredient of composition, and in this category, the organophosphate and organochloride groups of pesticides are the most common (Genius, Lane and Birkholz, 2016). Some pesticides are of public health interest because of the ability to move up the food chain (bioaccumulation); from crops to animals due to high lipophilic affinity (Akhtar and Ahad, 2017). This makes it easy for pesticides to be stored in fat cells and penetrate cellular membrane. Some pesticides also take a longer time to degrade, and therefore, can persist in different environmental layers such as soil, air, and water for an extensive period, causing high toxicity to the species exposed (Akhtar and Ahad, 2017; Genius, Lane and Birkholz, 2016). Individuals that are at risk to pesticide exposure are the pesticide application workers, where they are in direct contact due to inhalation and dermal absorption that is due to not having adequate personal protection equipment being utilized (Genius, Lane and Birkholz, 2016). However, young children, pregnant women and elderly are highly susceptible to pesticide exposure from ingesting food and beverages contaminated with pesticide residues because of their vulnerable immunity; therefore, leading to onset of pesticide toxicity induced health complications (Genius, Lane and Birkholz, 2016).

Organochlorine pesticides (OCPs) are a group of pesticides with chlorinated hydrocarbons as the active ingredient. Some of the OCP groups belong to the persistent organic pollutants (POPs) list established by the United Nation's Stockholm Convention in 2001, because it can lead to bioaccumulation, long half-life, long transport, and slow degradation (Genius, Lane and Birkholz, 2016; Jayaraj, Megha and Sreedev 2016). Due to these possible outcomes and its ability to induce acute or chronic toxicity in humans, some of these pesticides have been banned from use in the United States of America (Genius, Lane and Birkholz, 2016; Jayaraj, Megha and Sreedev 2016). Some examples of OCPs under the initial persistent organic pollutants (POP) category include, aldrin, chlordane, dichlorodiphenyltrichloroethane (DDT), and its analogues, endrin, dieldrin, heptachlor, mirex, hexachlorobenzene and toxaphene (Genius, Lane and Birkholz, 2016). The primary effect of these OCPs in human health are carcinogenicity, endocrine disruption, cognitive and developmental impairment, and organ damage (Jayaraj, Megha and Sreedev 2016). Although, these OCPs have been banned from use in the US, the residues can still be found in food and beverages today. This study involves the analysis milk samples (cow milk, PBMAs, and creamers), using a GC-ECD to detect the presence of OCP residues. The results were obtained from each milk sample to detect OCPs and its average concentration in parts per billion.

This study aims to investigate the average concentration of several OCPs, compare the concentration across milk samples and compare the concentration to the residue limit standard set by regulatory bodies which are EPA’s maximum contamination limit (MCL) and United Nations’ Food and Agriculture Organization’s (FAO) Codex Alimentarius.

Table 8. List of OCPs of interest, milk samples, and the average concentration in ppb

OCPs	**Cow milk (ppb)**	**Almond**	**Coconut**	**Soy**	**Oat**	**Rice**	**Hemp**	**Cashew**	**Creamer**	**residue limit (ppm)**
Alpha-chlordane	35	36	34	36	36	20	4	4	4	0
Gamma-chlordane	26	26	25	26	26	129	3	3	3	0
4,4-DDD	37	37	36	38	37	20	4	4	4	0.002
4,4-DDE	51	52	49	52	51	28	5	5	5	0.002
4,4-DDT	68	69	66	69	68	38	7	7	7	0.002
a-BHC	14	14	14	14	14	8	1	1	1	0
Aldrin	28	29	27	29	29	16	3	3	3	0.006
b-BHC	47	47	45	48	47	26	5	5	5	0
Chlordane	237	240	229	241	238	131	25	25	24	0
d-BHC	48	49	47	49	48	27	5	5	5	0
Dieldrin	35	36	34	36	36	20	4	4	4	0.006
Endosulfan I	48	49	47	49	48	27	5	5	5	0.01
Endosulfan II	40	40	38	40	40	22	4	4	4	0.01
Endosulfan sulfate	35	36	34	36	36	20	4	4	4	0.01
Endrin	55	56	53	56	55	31	6	6	6	0.002
Endrin aldehyde	58	59	56	59	58	32	6	6	6	0.002
Endrin ketone	47	47	45	48	47	26	5	5	5	0.002

g-BHC	21	22	21	22	21	12	2	2	2	0
Heptachlor	47	47	45	48	47	26	5	5	5	0.006
Heptachlor epoxide	37	37	36	38	37	20	4	4	4	0.006
Methoxychlor	81	82	78	82	81	45	8	8	8	0.04
Mirex	345	349	333	351	346	190	36	36	35	0
Toxaphene	237	240	229	241	238	131	25	25	24	0
Dicofol	946	957	914	962	948	523	98	98	96	0.05
Hexachlorobenzene	372	376	359	378	373	206	39	39	38	0
Kepone	335	339	323	341	336	185	35	35	34	0.3

Table 8 shows the list of OCPs analyzed from cow milk and PBMA samples where the average concentrations are reported in ppb with a comparison to residue limits set by regulatory bodies (EPA/WHO-FAO). The list of OCPs is written in bold fonts on the left of the table, and the milk samples are in bold fonts at the top of the table, while the regulatory MCL are on the right of the table. Data for table 8 was analyzed by averaging the concentrations of the milk brand to represent each milk sample. Table 8 shows that dicofol had the highest residue concentration at ~900 ppb for cow, almond, soy, coconut, and oat milk samples. The concentration of dicofol is lower for rice, hemp, cashew, and creamer but remains the higher than the maximum contamination limit set by the Codex Alimentarius (0.05 ppb). Hexachlorobenzene (HCB), kepone, mirex, toxaphene and chlordane followed the same trend as dicofol, where concentration ranges from 370 ppb to 240 ppb in cow, almond, soy, coconut, and oat. Although, dicofol application has been banned by the European Union, its production and use in the US is still ongoing as a potent insecticide. This is the likely reason for its high concentration especially in milk samples (Krieger, 2010). Methoxychlor is another insecticide that has two registered production labels in the US, even though it is banned in Europe. It is believed to have a shorter environmental half-life; however, there are no established literature for sensitization and irritation effects on animals (Krieger 2010). From the original 12 chemicals banned from use by the Stockholm Convention (SC), Hexachlorobenzene (HCB) had the highest concentration that is ~350 ppb for the following: soy, almond, coconut, oat, and cow milk. The concentration was lower for rice, hemp, cashew, and coffee creamer which was less than 38.7 ppb. Soy milk had the highest concentration of HCB with 379 ppb and coffee creamer with lowest concentration at 37.5 ppb. The second SC banned OCP with the highest concentration is Mirex, which concentration is greater than 330 ppb, observed in soy, cow, almond, coconut, and oat. The concentration is lower for rice, hemp, cashew, and creamer where its concentration is less than 35.9 ppb. Oat milk had the highest concentration of mirex at 359 ppb while coffee creamer had the lowest concentration of mirex at 34.8 ppb. Toxaphene and chlordane had high concentrations greater than 200 ppb in cow milk, almond, coconut, soy, and oat milk as the above- mentioned OCPs (HCB and mirex) and less than 25 ppb for rice, hemp, cashew, and creamer. Soy milk had the highest concentration of toxaphane with 241 ppb while coffee creamer was the least at 23.9 ppb. Chlordane residue was highest in soy milk at 240.8 ppb and coffee creamer was the least at 23 ppb. All the pesticides analysed have concentrations higher than the maximum residue contamination limit set by EPA and the *codex alimentarius international food standards* set by the Food and Agriculture Organization of the United Nations in collaboration with the World Health Organization (FAO, WHO). Table 8 was used to derive figures to draw visual comparison between cow milk, coffee creamer and PBMA samples. Error bars on figures indicate percentage error, that is reported in appendix D for standard deviation statistics of each milk sample.

Figure 13 shows the OCPs from the initial Stockholm Convention list of POPs. In the group, HCB had the highest residue concentration across all milk samples (see table 8). Figure 13 also shows the concentration trend across the milk samples with high residue concentrations in cow milk, almond, coconut, soy and oat milk and low concentration for rice, hemp, cashew, and creamer. Other OCPs from the SC initial POPs list with high residue concentrations in milk samples are shown in the graph and these include mirex, toxaphane, chlordane, and kepone. The MCL set for HCB, mirex, toxaphane, and chlordane is 0 ppb, while the MCL set for kepone is 0.03 ppb (see table 8).

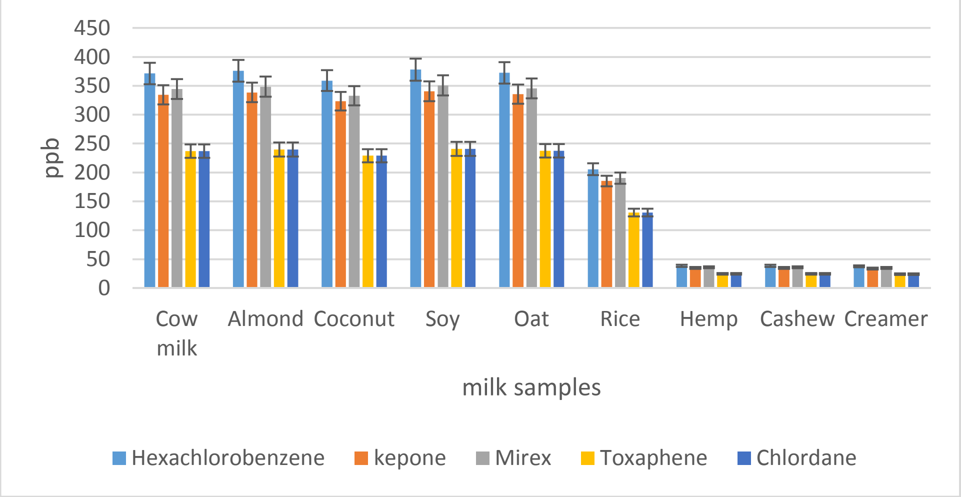


Figure 13. Stockholm Convention banned OCPs with the highest residue concentrations.

In Figure 14, data shows other OCPs that were part of the SC initial list of POPs with the varying residue concentration. From figure 14, methoxychlor had the highest residue concentration with concentrations greater than 80 ppb in cow, almond, coconut, soy, and oat. DDT follows with residue concentrations above 68 ppb where there is a lower concentration for rice, hemp, cashew, and coffee creamer. Other pesticides from the figure 14 include endrin, hepatochlor, dieldrin, and aldrin. The MCL set for methoxychlor is 0.04 ppm, DDT and endrin is 0.002 ppm, while MCL set for aldrin and dieldrin is 0.006 ppm (see table 8).

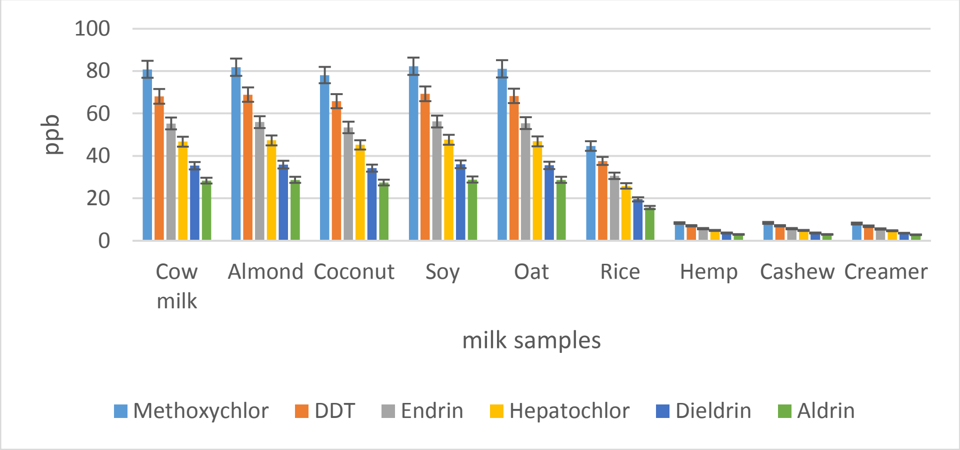


Figure 14. Residue concentrations of other Stockholm convention banned OCPs

In Figure 15, chlordane residue was present in all milk samples with concentrations above 200 ppb in cow almond, coconut, soy, and oat. Other residues of chlordane analogues such as alpha-chlordane and gamma-chlordane were present. Chlordane pesticide has been banned from application since the inception of the Stockholm Convention by the United Nations, yet its residue and that of its analogues persist in milk samples. MCL for chlordane and its analogues is 0 ppm.

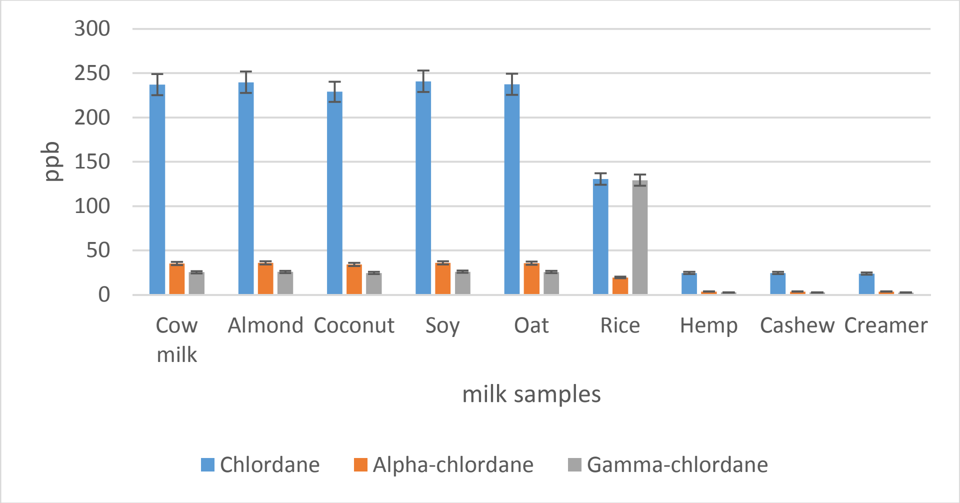


Figure 15. Residue concentration of chlordane and its analogues.

In Figure 16, the presence of DDT residue was present in all milk samples above 68 ppb for cow, almond, coconut, soy, and oat. The concentration of DDT analogues is greater than 50 ppb for 4,4-DDE in cow, almond, coconut, soy, and oat milk. The concentration is greater than 30 ppb for 4,4-DDD in the same milk samples mentioned prior. The use of DDT has been banned since the inception of the Stockholm Convention and its residues and that of its analogues persist in milk samples. MCL set for DDT is 0.002 ppm.

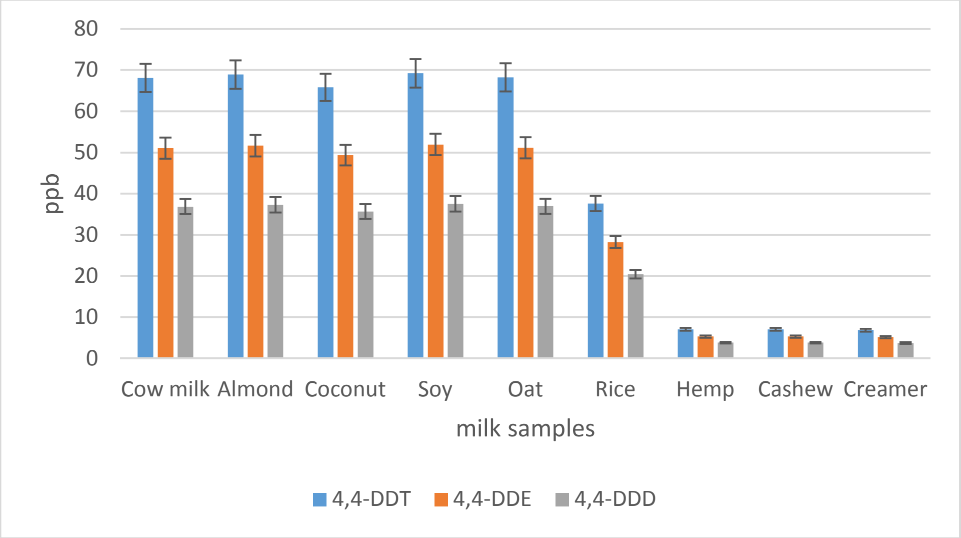


Figure 16. Residue concentration of DDT and its analogues

Hexachlorocyclohexane also known as BHC residue was observed in all milk samples with a concentration greater than 40 ppb in cow, almond, coconut, soy, and oat milk. Figure 17 shows the presence of other analogues of BHC such as delta-BHC, gamma-BHC, and alpha-BHC were also observed in all milk samples. Delta-BHC had the highest concentrations of hexachlorocylohexane derivatives at 48 ppb in cow, almond, coconut, soy, and oat. BHC application has been banned; however, its residue and that of its analogues are still present in milk samples. MCL set for BHC is 0 ppm.

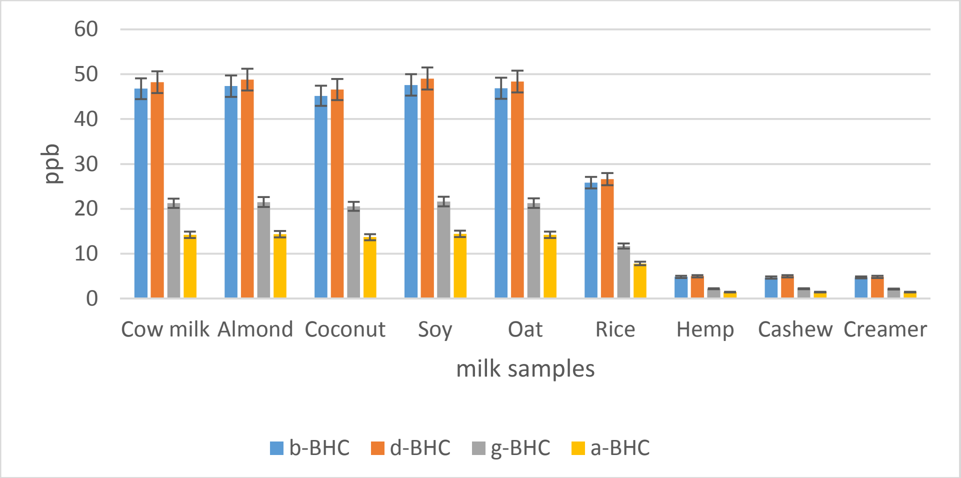


Figure 17. Residue concentration of hexachlorocyclohexane (BHC) and its analogues

In Figure 18, endosulfan residue concentration was present in all milk samples along with some of its analogues. Endosulfan contains concentrations greater than 48 ppb in cow, almond, coconut, soy, and oat milk. Endosulfan II contains concentrations greater than 39 ppb for the same milk samples mentioned prior. Endosulfan application has been banned by the Stockholm Convention and yet its residue persists in milk samples. MCL set for endosulfan is 0.01 ppm.

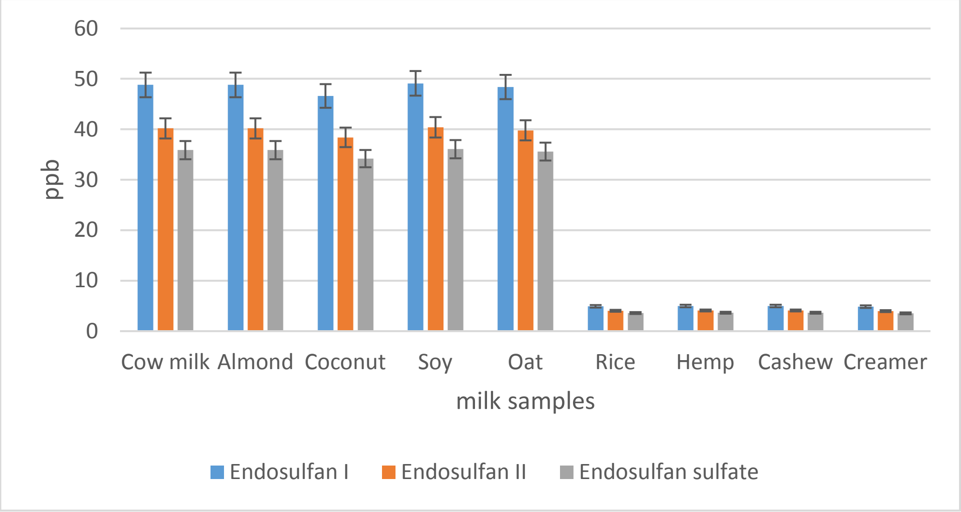


Figure 18. Residue concentration of endosulfan and its analogues.

Figure 19 shows the residue concentration of endrin along with some its analogues across all milk samples. Endrin concentration was greater than 50 ppb in cow, almond, coconut, soy, and oat. Endrin aldehyde had the highest concentration at ~60 ppb in cow, almond, coconut, soy, and oat. Endrin application has been banned since the inception of the Stockholm Convention; however, its residue persists in milk samples. MCL set for endrin is 0.002 ppm.

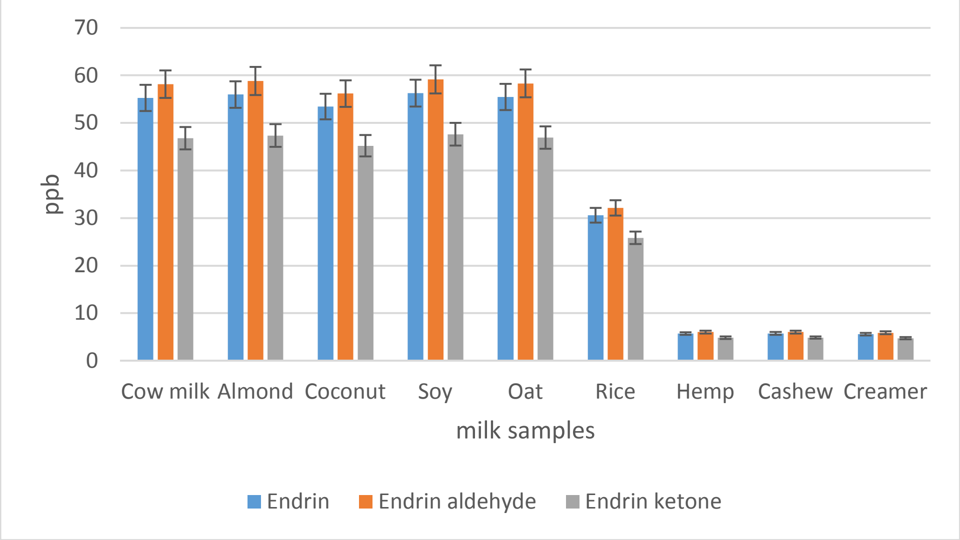


Figure 19. Residue concentration of endrin and its analogues

Figure 20 shows the residue concentration for hepatochlor that is greater than 45 ppb across cow, almond, coconut, soy, and oat milk samples. The analogue of hepatochlor is hepatochlor epoxide with concentrations greater than 38 ppb in cow, almond, coconut, soy, and oat milk. Hepatochlor has been banned from application since the inception of the Stockholm Convention but its residue still exists in milk samples. MCL set for hepatochlor is 0.006 ppm.

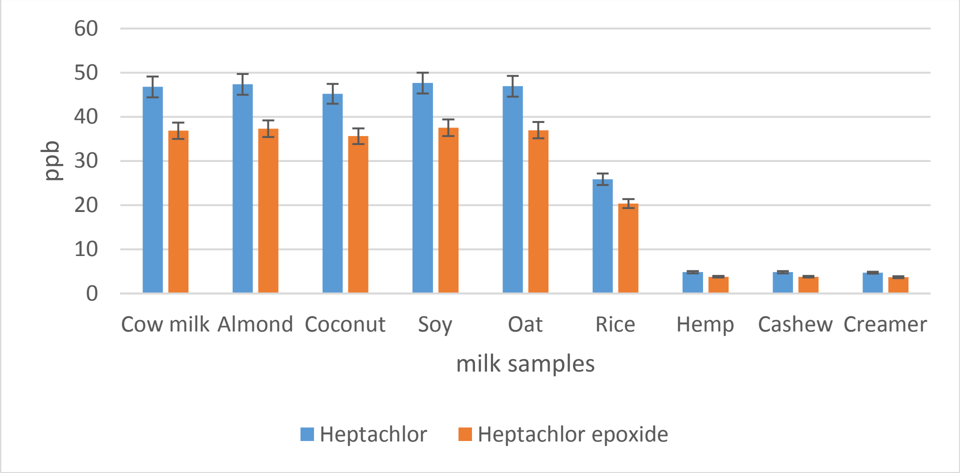


Figure 20. Residue concentration of hepatochlor and its analogues

Figure 21 shows the trend that concentration of OCPs is lower in rice milk, hemp milk, cashew milk, and creamer; however, OCPs are higher than the MCL regulations. The milk sample with highest concentration of OCPs with exception for Mirex and Dicofol is soy milk. Oat milk had the highest concentration of mirex at 359 ppb, while cow milk had the highest concentration of dicofol at 957.47 ppb. Dicofol is the only OCP that is not banned from production and application by the Stockholm Convention. As earlier observed in table 8, all milk samples have concentrations higher than that of the residue contamination limits set by regulatory standards which ranged from 0 – 0.04 ppm. While these OCPs mentioned above have been banned from production and application in the United States for several years, the residues concentration still surpasses the limit set for food according to the FAO-WHO Codex Alimentarius and EPA for residues in food (FAO/WHO, 2021; EPA, 2021). A probable reason for this may be the importation of pesticide contaminated produce (nuts and grains) from jurisdictions without a ban or strict regulation compliance (Genius, Lane and Birkholz, 2016).

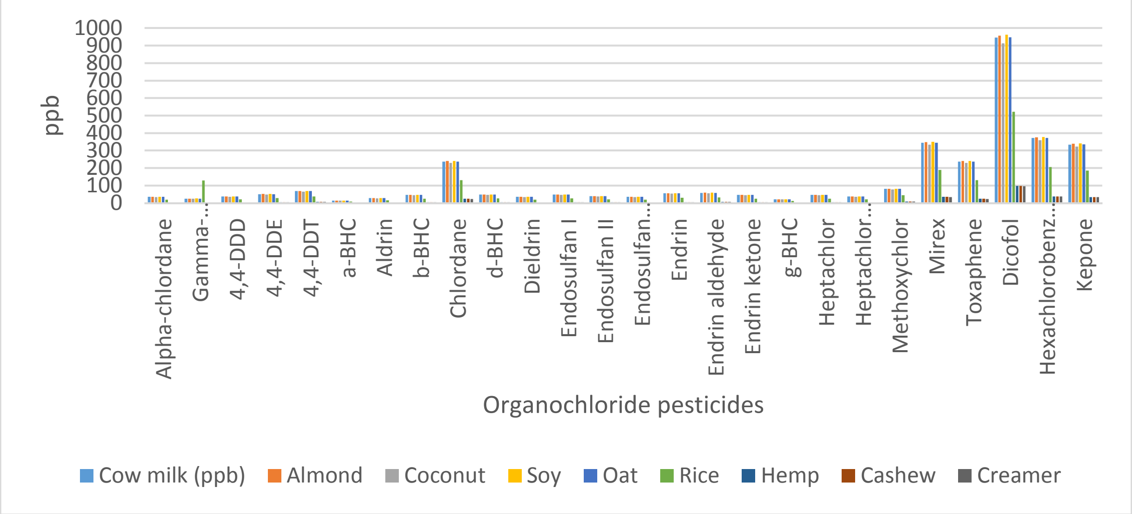


Figure 21. Average Concentration of OCPs (ppb) in all the Milk Samples using GC-ECD Analysis.

The potential health detriment associated with pesticide toxicity are acute neurotoxins, metabolism dysregulation, intracellular oxidative stress, endocrine disruption, cell death, synergistic effects in combination with other xenobiotic toxicants and carcinogenicity (Genius, Lane and Birkholz, 2016; Jayaraj, Megha and Sreedev 2016). The population with the most risk of suffering from OCPs residue in food including milk are children, elderly, and pregnant mothers. Pregnant women risk the transfer of OCPs to fetus (Genius, Lane and Birkholz, 2016; Jayaraj, Megha and Sreedev 2016). In the United States, regulations such as Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA), Federal Food, Drugs, and Cosmetic Act (FFDCA), Food Quality Protection Act of 1996 (FQPA) (EPA, 2021) and the FAO-WHO Codex Alimentarius for international food standards exists to address environmental concerns such as OCPs (FAO/WHO, 2021). The residue limits detected in these milk samples are alarming considering that some of these OCPs have been banned from production and application since the 1970's. More monitoring and quality control are recommended to reduce the potential health and economic burden on people who consume milk daily or frequently.

CHAPTER 5

CONCLUSION

In conclusion, milk consumption has been established as a vital source of balance diet because of the presence of essential elements (macronutrients and micronutrients); therefore, it is believed to help in children's growth and development. These essential elements play a vital role in intracellular molecule formation, intracellular function, intracellular signalling plus other vital functions such as metabolism of biomolecules, mineralization, and development of bone and teeth. Milk fortified with essential elements is encouraged for consumption; however, due to rising concerns with calorie intake, lactose intolerance and other issues mentioned, the shift from cow milk to other alternative plant-based milk sources has increased over time. A comparative analysis of milk samples for the presence of essential elements and heavy metals showed that several plant-based milk samples were more fortified than cow milk with regards to macronutrients concentrations and micronutrients concentrations, except in zinc, cow milk had the highest concentration of zinc at 0.145 ppm. PBMAs like hemp milk had the highest concentration of Pb and Cr at 1.0 ppb and 1.2 ppb, respectively and soy milk with the highest concentration for Cd at 0.4 ppb, compared to cow milk. Cow milk had concentrations below detection limits for Cr and Cd, and a concentration of 0.4 ppb for Pb. From the result, it is observed that almond and soy milk were rich in essential elements compared to cow milk and other plant-based milk alternatives. However, cow milk can be deduced as a safer option for consumption once the heavy metal analysis is factored in. Cow milk has the lowest concentration or below detection limit concentrations for almost all the heavy metals of interest. Hemp milk, rice and soy repeatedly have the highest concentrations of heavy metals, even though these concentrations were in trace amount and well below the maximum contamination limit set by EPA. Presence of heavy metals in plant-based milk alternatives may be attributed to nutrient intake as many of these heavy metals are found naturally in soil. However, anthropogenic activities in the agricultural process or manufacturing process may also be a contributing factor to the residue presence in final milk product. Lastly, while it can be observed that cow milk may be safer for consumption judging from heavy metal concentrations; however, it does not reflect the other primary concerns associated with cow milk consumption such as lactose intolerance, high calorie intake per serving, etc. Certain PBMAs such as almond and coconut milk also displayed high fortification when compared to other PBMAs. In addition, heavy metal presence ranged from trace concentrations to below detection limit concentration. This study also focused on the presence of OCPs residues in milk sample because of the characteristics of some of these OCPs and the fact that most have been banned from utilization years ago.

Results from pesticide analysis showed all the initial OCPs identified as POPs by the Stockholm Convention consist residue concentrations in all the milk samples. The residue concentrations detected were above the regulatory standard MCL (0 ppm - 0.04 ppm). This is of concern as these OCPs have been banned from use for over two decades. The reason for the ban is that these OCPs were discovered to have a negative effect on the ecosystem by affecting non-target organisms, causing eggshell thinning among wildlife birds, diminishing local wildlife population in surrounding ecosystem, causing cognitive dysfunction, etc. (SCPOP, 2008). At trace concentrations, these OCPs are potent enough to bioaccumulate in fat cells and could lead to inducing acute or chronic toxicity in the body. Other OCPs that are not banned by the Stockholm Convention had high residue concentrations. Dicofol had the highest concentration among all milk sample. Even though, the active ingredient for dicofol is DDT, it is not classified as hazardous to humans (FAO/WHO, 2021) and it continues to be in high production and use within the agricultural sector. More strictness on regulation can be used to mediate the concentrations of banned OCPs in milk samples and frequent testing of imported produce can also be an additional quality control protocol. This study highly recommends rigorous quality control on pesticide application especially in the agricultural sector to monitor the pesticide transport up the food chain. Strict adhesion to regulatory guidelines such as MCL is also recommended for residue concentrations of heavy metals and pesticides in food.

Future Studies

Further chemical profile analysis to study in the future will include the presence of hormones. Since PBMAs are derived from plants, it may contain various forms of Phyto-hormones that can compete with or disrupt natural endocrine function. Hormones are messenger biomolecules that play a vital role of homeostasis in living organisms. Hormone feedback signalling is a mechanism used by the body to elicit response, trigger the commencement of a function, monitor the function, and relay messages to end the function once the goal is attained (Hiller-Sturmhöfel and Bartke, 1998). According to Walker *et al.,*2005, environmental estrogens and androgens exist and can disrupt the human endocrine system by imitation of natural estrogens by binding to its receptor and stimulating the induction of estrogenic processes and by inhibiting natural estrogen function which binds to its receptor and preventing it from initiating any function. Pseudo hormones like phytoestrogens in plants and growth factors in cow milk can threaten the mechanism of the body's natural hormone called endocrine disruption. This can lead to onset of detrimental health conditions like obesity, cardiovascular disease, diabetes, etc.

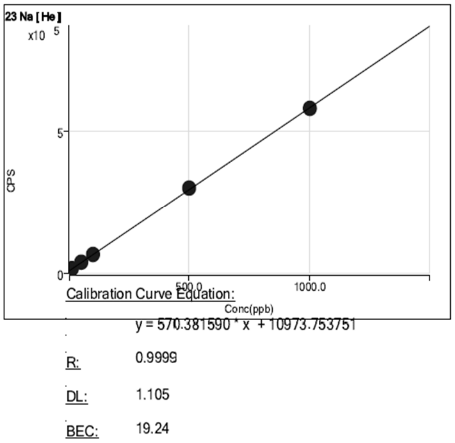
To analyse the presence of hormone residue in milk samples, the Agilent QUECHERS protocol would be used during the sample preparation period. The QUECHERS protocol stands for quick, easy, cheap, rugged, and safe. A three-step technique developed by the Agilent technical and research team. There are different traditional ways of sample preparation for GC-MS analysis such as liquid-liquid extraction (LLE) which requires low cost to perform, has a short development time and can easily remove inorganic salts. The LLE is labor intensive, forms emulsion and requires the use of large volumes of organics. Another popular sample preparation method is the solid phase extraction (SPE), according to Stevens and Jones (2010). This is advantageous because it yields a high recovery, can be reproducible, effective with samples with different matrix concentration and it is selective. However, it is complex to execute, requires a lengthy development and can be costly. Other methods for analyses are gel permeation chromatography (GPC) and soxhlet extraction. The QUECHERS protocol which was introduced in 2003 by Anastassiades *et al.*, 2003, validated in 2005 and modified in 2007 has become a popular streamlined approach to sample preparation for many investigators because it is easier and less expensive to analyse most organic residues in food especially pesticides and hormones (Stevens and Jones, 2010). There are three basic methods to executing the QUECHERS protocol to suit study aims. These methods include AOAC method (2007.01), European method (EN 15662) and the original method (Stevens and Jones, 2013). Every method involves three steps where the first step consists of extraction of sample by homogenizing sample in tube, adding acetonitrile and then extracting salt packet. The second step involves centrifuging samples and transferring into a dispersive tube and then centrifuge again. The last step is extracting the supernatant and transferring into a GC/LC-MS vial for analysis. According to Stevens and Jones (2010), the purpose of the extracting salt in the first step is to prevent exothermic reactions by inhibiting the degradation of sample to ensure maximum recovery yield. The extraction salt for the EN method is made up of 4 g $MgSO_4$, 1 g NaCl, 1 g NaCitrate and 0.5 g disodium citrate sesquihydrate (Stevens and Jones, 2010).

The mass spectrometry (MS) serves as the detector for the GC. As the sample moves through the column to the detection, it then enters the MS. The substances undergo electron ionization to convert to ions and produce fragments for identification. The ions are sorted based on the mass to charge to form a pattern of fragmentation where each fragmentation pattern is unique and characteristically used to identify each component present in the sample (Hussain and Maqbool, 2014).

APPENDIX

APPENDIX A

CALIBRATION CURVE FOR ICP-MS STANDARDS



Calibration Curve Equation:
y = 570.381590 * x + 10973.753751

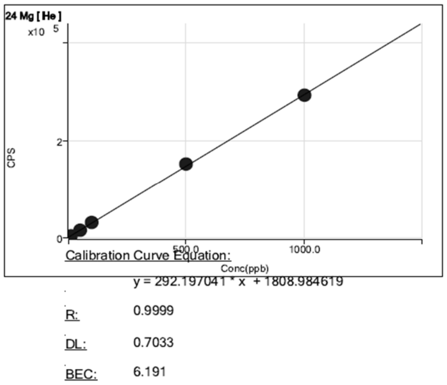
R: 0.9999

DL: 1.105

BEC: 19.24

Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-4.305	8518.10		Pulse	2.47
2	False	1.000	-0.935	10440.45		Pulse	0.75
3	False	10.000	8.582	15868.67		Pulse	0.77
4	False	50.000	50.815	39957.58		Pulse	1.61
5	False	100.000	102.247	69293.46		Pulse	0.34
6	False	500.000	509.692	301692.83		Pulse	0.33
7	False	1000.000	994.905	578449.00		Pulse	0.24

Figure 1. Calibration curve for Na standard



Calibration Curve Equation:
y = 292.197041 * x + 1808.984619

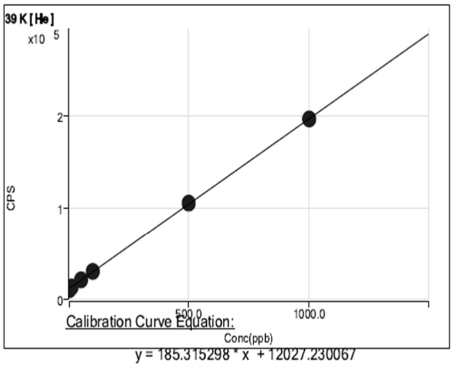
R: 0.9999

DL: 0.7033

BEC: 6.191

Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-4.122	604.46		Pulse	11.33
2	False	1.000	-1.483	1375.63		Pulse	5.39
3	False	10.000	8.467	4282.89		Pulse	1.58
4	False	50.000	51.277	16791.94		Pulse	1.06
5	False	100.000	101.256	31395.79		Pulse	1.76
6	False	500.000	511.554	151283.68		Pulse	0.79
7	False	1000.000	994.051	292267.78		Pulse	0.46

Figure 2. Calibration curve for Mg standard



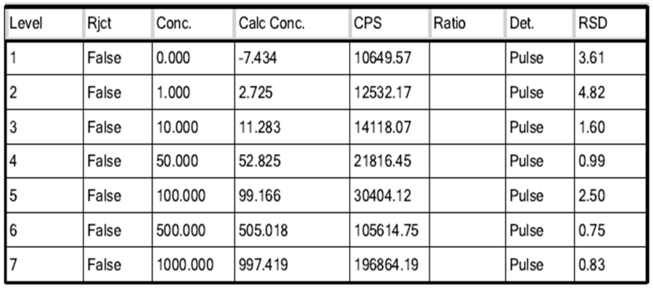
Calibration Curve Equation:

y = 185.315298 * x + 12027.230067

R: 0.9999

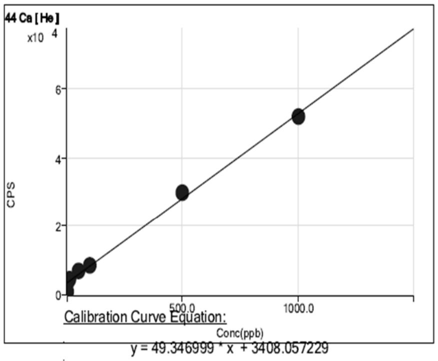
DL: 6.219

BEC: 64.9



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-7.434	10649.57		Pulse	3.61
2	False	1.000	2.725	12532.17		Pulse	4.82
3	False	10.000	11.283	14118.07		Pulse	1.60
4	False	50.000	52.825	21816.45		Pulse	0.99
5	False	100.000	99.166	30404.12		Pulse	2.50
6	False	500.000	505.018	105614.75		Pulse	0.75
7	False	1000.000	997.419	196864.19		Pulse	0.83

Figure 3. Calibration curve for K standard



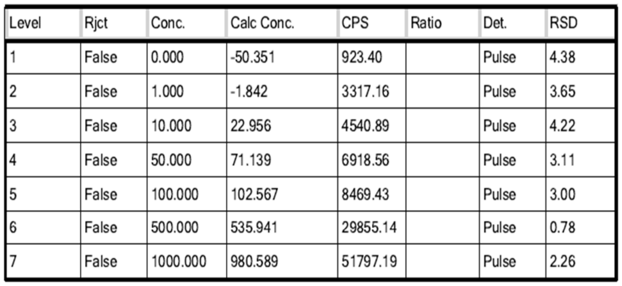
Calibration Curve Equation:

y = 49.346999 * x + 3408.057229

R: 0.9972

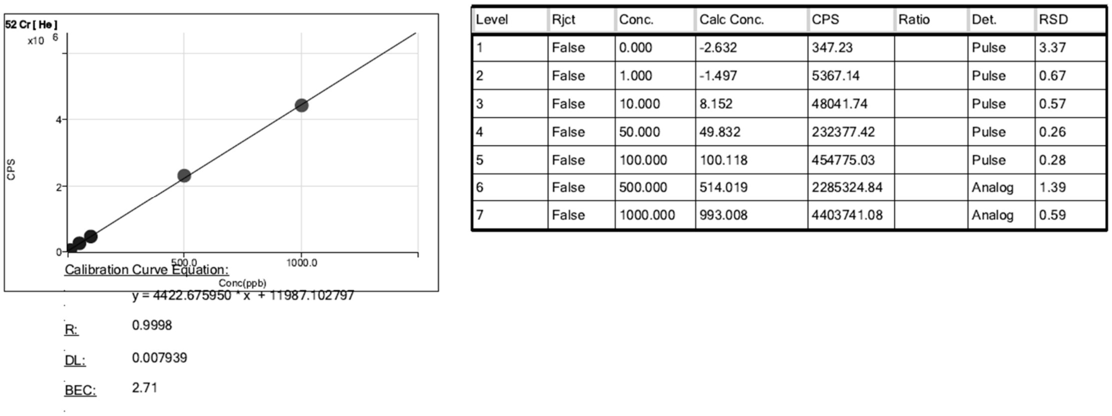
DL: 2.457

BEC: 69.06



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-50.351	923.40		Pulse	4.38
2	False	1.000	-1.842	3317.16		Pulse	3.65
3	False	10.000	22.956	4540.89		Pulse	4.22
4	False	50.000	71.139	6918.56		Pulse	3.11
5	False	100.000	102.567	8469.43		Pulse	3.00
6	False	500.000	535.941	29855.14		Pulse	0.78
7	False	1000.000	980.589	51797.19		Pulse	2.26

Figure 4. Calibration curve for Ca standard



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-2.632	347.23		Pulse	3.37
2	False	1.000	-1.497	5367.14		Pulse	0.67
3	False	10.000	8.152	48041.74		Pulse	0.57
4	False	50.000	49.832	232377.42		Pulse	0.26
5	False	100.000	100.118	454775.03		Pulse	0.28
6	False	500.000	514.019	2285324.84		Analog	1.39
7	False	1000.000	993.008	4403741.08		Analog	0.59

Calibration Curve Equation:

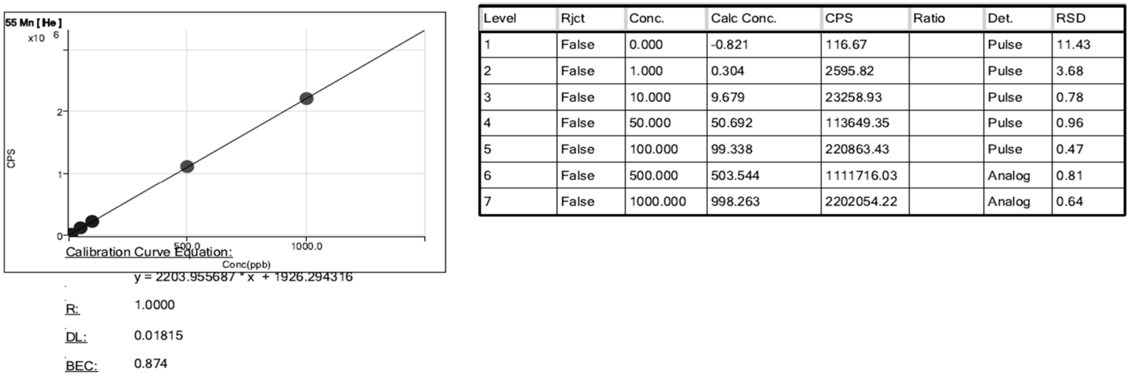
y = 4422.675950 * x + 11987.102797

R: 0.9998

DL: 0.007939

BEC: 2.71

Figure 5. Calibration curve for Cr standard



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-0.821	116.67		Pulse	11.43
2	False	1.000	0.304	2595.82		Pulse	3.68
3	False	10.000	9.679	23258.93		Pulse	0.78
4	False	50.000	50.692	113649.35		Pulse	0.96
5	False	100.000	99.338	220863.43		Pulse	0.47
6	False	500.000	503.544	1111716.03		Analog	0.81
7	False	1000.000	998.263	2202054.22		Analog	0.64

Calibration Curve Equation:

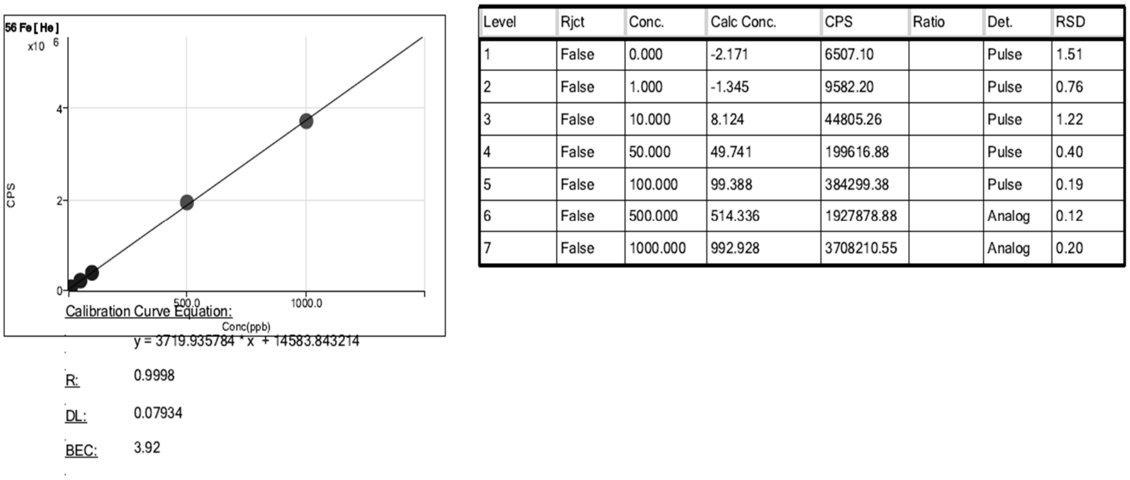
y = 2203.955687 * x + 1926.294316

R: 1.0000

DL: 0.01815

BEC: 0.874

Figure 6. Calibration curve for Mn standard



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-2.171	6507.10		Pulse	1.51
2	False	1.000	-1.345	9582.20		Pulse	0.76
3	False	10.000	8.124	44805.26		Pulse	1.22
4	False	50.000	49.741	199616.88		Pulse	0.40
5	False	100.000	99.388	384299.38		Pulse	0.19
6	False	500.000	514.336	1927878.88		Analog	0.12
7	False	1000.000	992.928	3708210.55		Analog	0.20

Calibration Curve Equation:

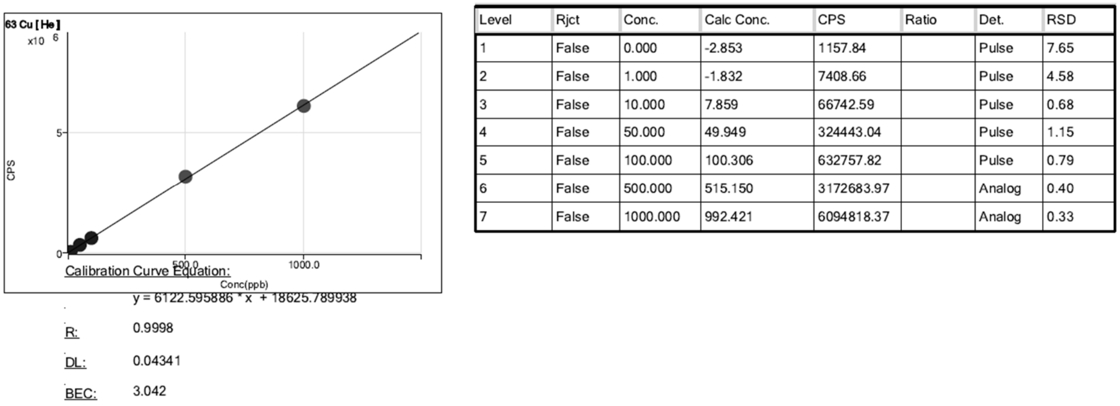
$y = 3719.935784 * x + 14583.843214$

R: 0.9998

DL: 0.07934

BEC: 3.92

Figure 7. Calibration curve for Fe standard



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-2.853	1157.84		Pulse	7.65
2	False	1.000	-1.832	7408.66		Pulse	4.58
3	False	10.000	7.859	66742.59		Pulse	0.68
4	False	50.000	49.949	324443.04		Pulse	1.15
5	False	100.000	100.306	632757.82		Pulse	0.79
6	False	500.000	515.150	3172683.97		Analog	0.40
7	False	1000.000	992.421	6094818.37		Analog	0.33

Calibration Curve Equation:

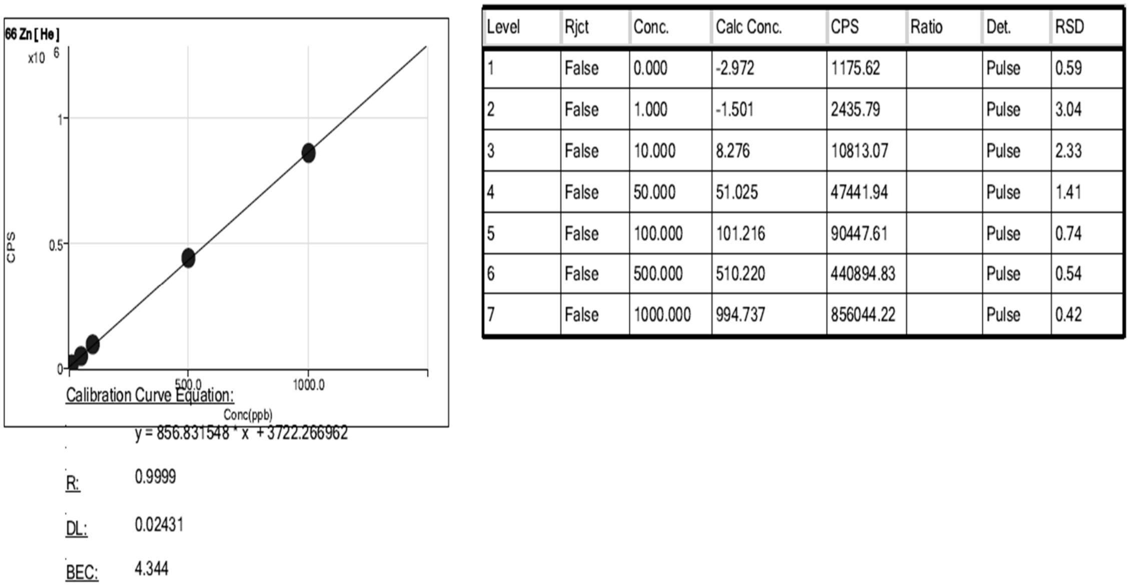
$y = 6122.595886 * x + 18625.789938$

R: 0.9998

DL: 0.04341

BEC: 3.042

Figure 8. Calibration curve for Cu standard



Calibration Curve Equation:

y = 856.831548 * x + 3722.266962

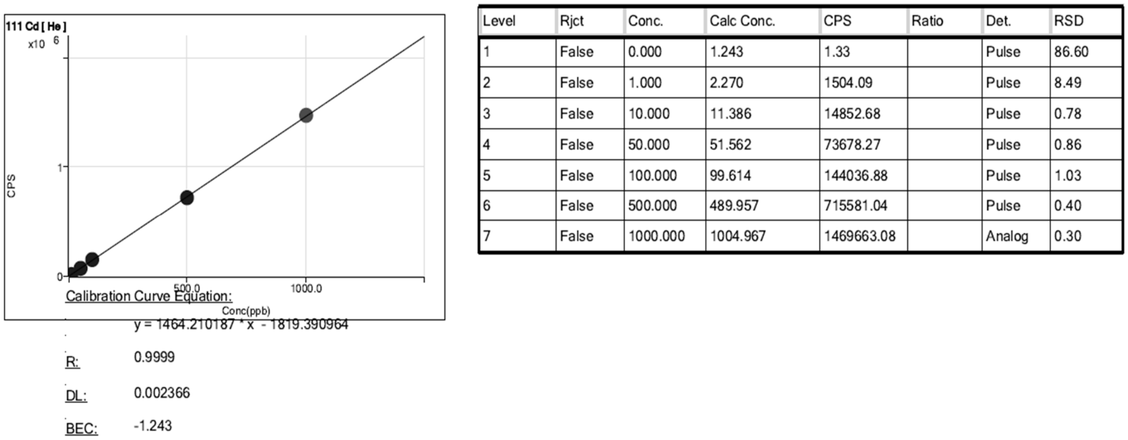
R: 0.9999

DL: 0.02431

BEC: 4.344

Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-2.972	1175.62		Pulse	0.59
2	False	1.000	-1.501	2435.79		Pulse	3.04
3	False	10.000	8.276	10813.07		Pulse	2.33
4	False	50.000	51.025	47441.94		Pulse	1.41
5	False	100.000	101.216	90447.61		Pulse	0.74
6	False	500.000	510.220	440894.83		Pulse	0.54
7	False	1000.000	994.737	856044.22		Pulse	0.42

Figure 9. Calibration curve for Zn standard



Calibration Curve Equation:

y = 1464.210187 * x - 1819.390964

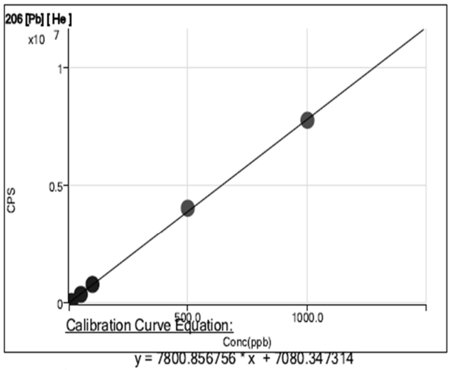
R: 0.9999

DL: 0.002366

BEC: -1.243

Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	1.243	1.33		Pulse	86.60
2	False	1.000	2.270	1504.09		Pulse	8.49
3	False	10.000	11.386	14852.68		Pulse	0.78
4	False	50.000	51.562	73678.27		Pulse	0.86
5	False	100.000	99.614	144036.88		Pulse	1.03
6	False	500.000	489.957	715581.04		Pulse	0.40
7	False	1000.000	1004.967	1469663.08		Analog	0.30

Figure 10. Calibration curve for Cd standard



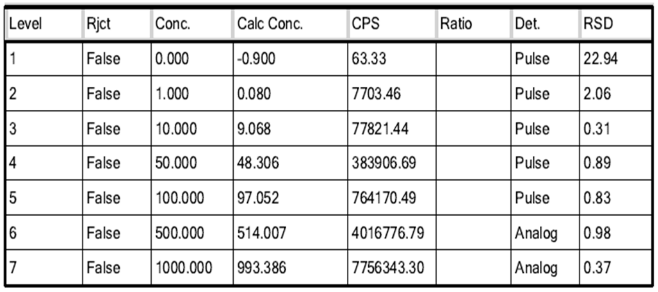
Calibration Curve Equation:

y = 7800.856756 * x + 7080.347314

R: 0.9999

DL: 0.005587

BEC: 0.9076



Level	Rjct	Conc.	Calc Conc.	CPS	Ratio	Det.	RSD
1	False	0.000	-0.900	63.33		Pulse	22.94
2	False	1.000	0.080	7703.46		Pulse	2.06
3	False	10.000	9.068	77821.44		Pulse	0.31
4	False	50.000	48.306	383906.69		Pulse	0.89
5	False	100.000	97.052	764170.49		Pulse	0.83
6	False	500.000	514.007	4016776.79		Analog	0.98
7	False	1000.000	993.386	7756343.30		Analog	0.37

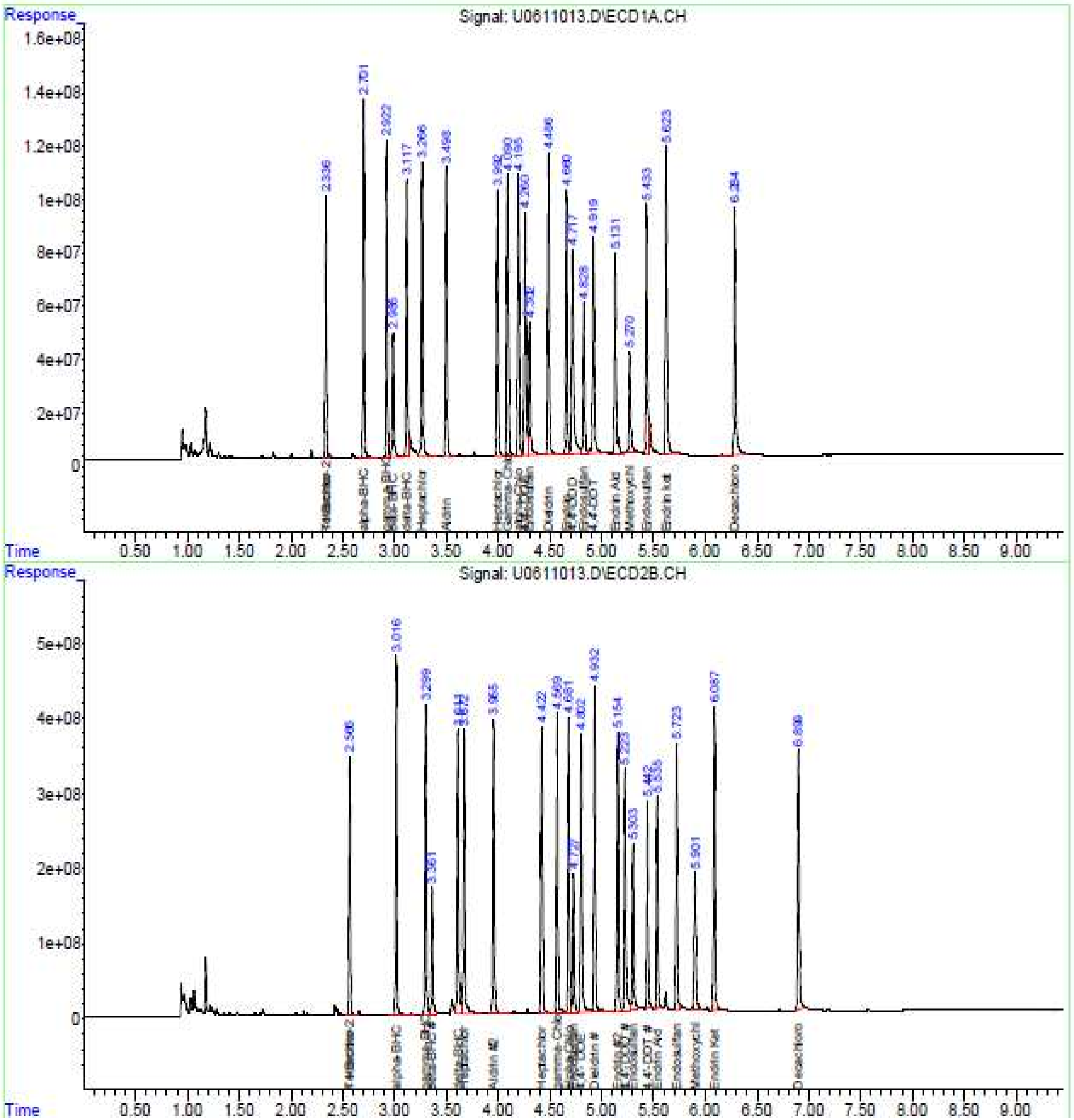
Figure 11. Calibration curve for Pb standard

APPENDIX B

CHROMATOGRAPH FOR THE GC-ECD STANDARDS

```
Integration File signal 1: AUTOINT1.E
Integration File signal 2: autoint2.e
Quant Time: Jun 11 16:15:42 2021
Quant Method : C:\msdchem\1\methods\PESTU0611.M
Quant Title  : SW846 8081A Pesticides - MultiMix
QLast Update : Fri Jun 11 16:05:45 2021
Response via : Initial Calibration
Integrator: ChemStation   6890 Scale Mode: Large solvent peaks clipped

Volume Inj.     :
Signal #1 Phase :                  Signal #2 Phase:
Signal #1 Info  :                  Signal #2 Info :
```



APPENDIX C

ICP-MS CONCENTRATIONS FOR MILK SAMPLES

Table 1. Cow Milk Brands, ICP-MS Concentrations for Macronutrients (1^{st} run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Cm 1.1	2944.145	0.305567	1010.046	0.687376	10232.47	0.675222	2650.936	1.433115
Cm 1.2	2771.929	1.337092	952.2898	0.419836	9498.592	1.595024	2484.855	0.630383
Cm 1.3	3580.369	3.949326	984.0027	3.17246	12065.27	5.202788	3128.624	4.874268
Cm 2.1	4310.332	4.465697	1094.447	0.080962	17040.41	6.455866	5117.583	7.382504
Cm 2.2	5737.947	2.589328	1124.773	0.396945	23025.74	4.387099	6881.468	3.351765
Cm 2.3	7731.984	1.629419	1086.648	0.12195	27714.39	3.430936	8151.031	2.13812
Cm 3.1	6927.547	1.681113	971.789	0.237517	31646.2	1.411661	10305.83	1.160965
Cm 3.2	7820.177	2.050885	1074.804	0.236687	34635.95	0.5866	11556.89	1.980322
Cm 3.3	8152.789	1.849272	1240.624	0.67426	39289.92	0.635792	13470.17	0.569584
Cm 4.1	6831.3	1.794961	998.5365	0.669135	33606.32	0.316384	8551.269	0.74421
Cm 4.2	7271.272	0.696873	936.5872	0.335501	34629.44	0.383129	8493.154	1.672851
Cm 4.3	7529.984	0.795709	969.9379	0.938421	36901.98	1.005502	9134.333	1.277182

Table 2. Cow Milk Brands, ICP-MS Concentrations for Micronutrients (1^{st} run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CM 1.1	0.186774	23.26177	3.887121	2.345148	1.19664	2.829882	28.96954	1.562786
CM 1.2	0.164676	9.208924	2.491672	4.101622	10.98649	1.782525	27.54933	3.054769
CM 1.3	0.253053	14.86833	5.950884	8.189717	14.02237	3.800951	32.4809	4.625522
CM 2.1	0.3635	7.653475	4.679914	6.815426	18.48833	4.693404	61.60594	6.035198
CM 2.2	0.457896	8.219099	8.418079	2.613628	20.74449	4.465179	68.85229	3.483164
CM 2.3	0.530199	6.912088	14.08868	2.532839	32.92336	2.590195	84.0031	1.865486
CM 3.1	0.672788	12.67532	10.75483	3.669118	28.75132	2.261026	137.0656	2.248772
CM 3.2	0.753117	6.248716	11.13748	3.761476	32.19612	2.24942	150.069	0.860553
CM 3.3	0.791278	8.557665	12.32297	2.442663	36.00339	0.644643	155.0461	1.659941
CM 4.1	0.514125	7.804013	9.761981	2.265184	28.5078	1.048219	75.95804	2.051209
CM 4.2	0.486014	3.118527	7.269151	1.824174	29.66896	0.878815	73.52482	1.815831
CM 4.3	0.558311	19.01144	9.878191	0.913762	49.59721	0.233268	80.06442	2.085035

Table 3. Cow Milk Brands, ICP-MS Concentrations for Heavy Metals (1st run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CM 1.1	0.064537	8.247468	0.010265	0	0.009907	58.07565	0.047245	13.83219
CM 1.2	0.044559	22.61449	0.003422	173.2051	0.019052	48.49742	0.20926	3.00201
CM 1.3	0.083995	16.99594	0.010265	0	0.01829	62.5	0.104105	7.720699
CM 2.1	0.073753	22.04714	0.003422	173.2051	0.029721	20.35193	0.089681	9.111185
CM 2.2	0.145973	4.209385	0.023952	65.46537	0.022862	45.82576	0.149888	2.301383
CM 2.3	0.20897	9.386029	0.010265	100	0.051822	24.29811	0.307936	6.370069
CM 3.1	0.085532	19.87119	0.02053	0	0.019814	24.01922	0.125952	5.110813
CM 3.2	0.125482	5.09996	0.02053	50	0.028197	26.06392	0.216577	7.498908
CM 3.3	0.104483	5.880883	0.030795	0	0.025911	22.2054	0.139226	1.84333
CM 4.1	0.217679	9.211715	0.037639	103.2529	0.027435	8.333333	0.212709	2.350897
CM 4.2	0.123948	16.83426	0.037639	62.98367	0.025149	18.18182	0.140376	7.602841
CM 4.3	0.10346	16.88635	0.037639	41.65978	0.047249	29.565	0.26372	2.221536

Table 4. Almond Milk Brands, ICP-MS Concentrations for Macronutrients (1st run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Am 1.1	9480.846	1.379628	571.0881	0.34468	18522.87	1.137124	15083.93	1.074589
Am 1.2	9319.44	0.64837	559.3062	1.123517	18302.38	0.937025	14801.48	1.328393
Am 1.3	9500.512	0.695525	550.1416	0.480589	17745.06	1.240533	14446.52	1.192083
Am 2.1	16467.62	1.484097	579.4026	0.789231	17113.81	0.174555	16513.23	1.090382
Am 2.2	268.4251	0.966819	18.48395	1.734471	285.8079	3.795919	241.294	5.898894
Am 2.3	507.3004	0.671234	39.3891	2.543337	332.2512	1.207259	408.8766	5.584001
Am 3.1	11881.55	0.937899	537.3176	2.059154	26042.69	0.446044	19027.85	1.086394
Am 3.2	11438.26	0.59171	526.4935	3.22221	24813.28	0.461166	18259.06	0.488996
Am 3.3	11885.06	0.518787	538.6959	0.463321	25348.71	0.286437	18750.5	0.888816
Am 4.1	12895.38	0.42548	610.8929	0.748692	26956.24	3.118267	17919.34	1.03418
Am 4.2	26974.13	0.961867	1057.315	1.642512	42290.73	1.144621	33151.75	1.055217
Am 4.3	27595.4	1.387215	1079.526	0.641831	43270.27	0.345038	34082.11	0.710709

Table 5. Almond Milk Brands, ICP-MS Concentrations for Micronutrients (1st run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Am 1.1	9.213624	3.730819	87.07045	0.858407	25.74254	1.344973	15.95293	0.68989
Am 1.2	9.111166	4.311945	82.58019	0.575435	35.9243	1.1407	15.35565	3.685203
Am 1.3	9.058925	3.824891	106.2711	1.889844	47.19171	0.753014	14.87359	6.446115
Am 2.1	7.692859	2.595259	22.44883	0.774459	41.94292	2.086217	22.95873	2.472955
Am 2.2	0.196812	12.74746	2.818563	0.951839	32.14531	1.494295	4.416161	9.204791
Am 2.3	0.28719	14.73586	2.823439	6.889857	34.51284	0.599277	3.949908	11.69766
Am 3.1	9.400459	1.032048	30.7067	0.540529	31.25585	0.993058	17.89688	2.347528
Am 3.2	9.187494	2.784236	28.75734	0.744604	29.45799	1.172545	13.48511	2.177554
Am 3.3	8.799781	2.074397	26.50519	1.402698	31.26372	1.489573	16.31449	1.184022
Am 4.1	10.49943	2.412815	28.31606	0.604558	38.96246	0.21822	15.95293	4.096986
Am 4.2	16.9534	3.367359	45.15655	2.369938	39.14887	0.649594	29.12677	1.86753
Am 4.3	17.38146	2.553922	76.4654	1.245357	13.85526	1.472814	30.64665	2.710939

Table 6. Almond Milk Brands, ICP-MS Concentrations for Heavy Metals (1st run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Am 1.1	0.119335	0.744863	0.167664	9.352195	0.027435	38.18813	0.167867	3.220873
Am 1.2	0.141876	17.27272	0.061591	28.86751	0.021338	12.37179	0.151665	0.860937
Am 1.3	2.547807	2.573932	0.11976	47.20775	0.035818	35.15471	0.223475	0.567103
Am 2.1	0.583893	5.475842	0.044482	81.04349	0.126506	20.31291	0.196925	1.519102
Am 2.2	0.054802	15.44394	0.003422	173.2051	0.012955	36.73528	0.132118	6.061842
Am 2.3	0.057878	24.08842	0.037639	56.77271	0.005335	24.74358	0.104733	2.949097
Am 3.1	0.310386	1.714851	0.082121	33.07189	0.027435	8.333333	0.138285	2.26795
Am 3.2	0.250456	6.040986	0.034217	45.82576	0.028959	12.05941	0.155428	6.661815
Am 3.3	0.297578	6.224841	0.051326	40	0.021338	22.30356	0.16933	8.372732
Am 4.1	0.297578	8.676096	0.092386	19.24501	0.022862	36.05551	0.142048	4.561269
Am 4.2	0.727822	4.029525	0.065013	9.116057	0.127268	10.97623	0.321735	5.676053
Am 4.3	0.727822	0.79896	0.061591	28.86751	0.105929	25.74896	0.195253	2.596293

Table 7. Soy Milk Brands, ICP-MS Concentrations for Macronutrients (1st run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Sm 1.1	7528.092	1.177327	1695.741	0.373821	29937.95	0.607313	10738.7	1.019111
Sm 1.2	7920.67	0.703125	1863.167	0.744745	32874.19	0.539849	11764.97	0.961605
Sm 1.3	6741.481	1.377028	1805.22	0.401024	39133.05	0.95358	8179.387	1.660067
Sm 2.1	6863.264	0.635996	1662.947	0.723401	30024.44	1.083239	10601.57	2.123724
Sm 2.2	6906.535	0.625724	1865.242	0.64673	39574.86	1.221673	8364.25	0.619941
Sm 2.3	7822.993	0.261522	1800.835	0.337588	38841.69	1.99194	8035.326	0.689466
Sm 3.1	2646.986	0.820803	1889.637	2.404071	44282.79	0.989456	1607.306	2.564093
Sm 3.2	2954.395	0.094843	1982.8	1.037421	47017.57	1.908172	1650.806	2.153388
Sm 3.3	4209.537	0.067945	1977.92	0.255702	47982.75	0.964601	1585.338	3.024912

Table 8. Soy Milk brands, ICP-MS Concentrations for Micronutrients (1st run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Sm 1.1	36.71503	0.797489	124.6456	0.728288	49.23829	0.266883	64.66796	0.444116
Sm 1.2	39.80828	0.553315	137.6373	0.645222	72.82141	0.285398	70.04801	2.628504
Sm 1.3	47.41835	1.36847	111.8295	0.718769	85.6723	0.703561	72.1036	0.841165
Sm 2.1	36.64467	0.956127	135.8358	0.649163	63.49885	0.109883	65.74818	1.762107
Sm 2.2	47.23722	0.905571	108.8124	1.265666	66.88788	0.46114	73.60858	0.830003
Sm 2.3	46.40028	1.544743	94.99562	1.331746	69.02696	1.323217	74.42138	2.132802
Sm 3.1	48.95546	1.877073	107.6974	0.3458	58.79918	0.767802	73.66102	1.277292
Sm 3.2	50.91527	0.814039	116.3961	1.236872	59.96266	0.318531	79.5924	0.647028
Sm 3.3	51.77251	0.617286	117.9966	0.659542	71.02522	0.781742	79.80734	0.961005

Table 9. Soy Milk Brands, ICP-MS Concentrations for Heavy Metals (1st run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
Sm 1.1	0.254042	4.02612	0.092386	55.55556	0.167658	6.148977	0.332919	3.365221
Sm 1.2	0.296045	5.198275	0.068434	56.78908	0.163848	22.07711	0.270201	2.63203
Sm 1.3	0.247383	7.930633	0.071856	37.79645	0.188234	9.196613	0.223894	2.430235
Sm 2.1	0.297578	3.155129	0.092386	0	0.123457	11.11111	0.260166	0.594679
Sm 2.2	0.192581	2.437141	0.061591	33.33333	0.172993	13.03846	0.315567	2.839307
Sm 2.3	0.220752	6.867292	0.061591	33.33333	0.230911	10.04841	0.23142	7.518921
Sm 3.1	0.144946	2.449335	0.088965	17.62529	0.139461	14.57081	0.180723	3.210598
Sm 3.2	0.162874	4.110576	0.054747	39.03124	0.162324	9.758033	0.324138	4.078552
Sm 3.3	0.186432	11.21911	0.075278	20.82989	0.144796	11.42239	0.315671	4.470717

Table 10. Coconut Milk Brands, ICP-MS Concentrations for Macronutrients (1st run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CCM 1.1	1604.501	0.431935	348.0205	0.495525	6979.535	0.214052	6514.22	1.59139
CCM 1.2	1455.736	0.693113	311.8312	3.123703	6231.706	0.588521	5829.468	1.00598
CCM 1.3	1221.847	0.612471	364.2793	0.194903	7287.331	0.798704	6846.729	0.921683
CCM 2.1	5604.925	0.341675	284.849	0.821397	20557.87	1.219471	18748.36	0.657437
CCM 2.2	5236.199	0.357959	271.9944	0.190598	19980.65	2.385379	17807.75	0.243838
CCM 2.3	6922.554	1.606344	285.4567	0.285542	20745.71	1.054422	18838.07	1.805202
CCM 3.1	2261.896	0.716839	1111.784	0.22923	4229.112	0.300308	4919.04	0.379309
CCM 3.2	3632.264	0.539172	1092.845	0.942019	4252.828	0.256595	4973.2	1.139832
CCM 3.3	2565.525	0.80476	1235.85	0.570143	4436.759	0.895489	5101.425	0.766811

Table 11. Coconut Milk Brands, ICP-MS Concentrations for Micronutrients (1st run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CCM 1.1	6.421285	1.224841	37.38843	0.472551	32.3324	0.215443	8.528706	2.119769
CCM 1.2	5.728254	1.510949	37.56557	0.777241	36.09186	0.473233	12.69393	3.673891
CCM 1.3	6.907413	3.424214	38.24234	0.983195	37.41715	1.293019	9.618457	2.287803
CCM 2.1	11.30106	2.823065	108.4605	0.354036	17.50519	0.57123	10.50916	10.67784
CCM 2.2	10.48935	1.533792	103.0322	0.657082	21.94019	1.520933	9.702297	3.922842
CCM 2.3	11.34527	1.476459	108.8148	0.869811	12.46016	0.781469	10.56155	0.648859
CCM 3.1	8.150893	4.696221	23.23043	1.354256	19.50338	2.163961	67.58858	0.75842
CCM 3.2	8.436155	1.868343	28.82764	1.24462	20.56129	0.331987	75.23433	3.474681
CCM 3.3	8.850004	3.204327	26.04008	1.001856	22.5717	0.294015	73.55087	0.845319

Table 12. Coconut Milk Brands, ICP-MS Concentrations for Heavy Metals (1st run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CCM 1.1	0.207436	7.143233	0.157399	24.69091	0.064015	21.42857	0.165776	10.13456
CCM 1.2	0.236117	5.832966	0.136868	88.84678	0.052584	50.14158	0.197239	5.254109
CCM 1.3	0.206407	10.89763	0.112917	39.62635	0.032007	12.37179	0.151037	2.803626
CCM 2.1	0.157755	9.049276	0.136868	31.22499	0.037342	18.70439	0.37285	2.039642
CCM 2.2	0.157755	9.803758	0.11976	43.98516	0.026673	17.84285	0.184487	3.022836
CCM 2.3	0.191045	1.672939	0.164242	16.53595	0.05106	37.55148	0.194417	5.243636
CCM 3.1	0.27709	6.084398	0.092386	19.24501	0.020576	11.11111	0.119575	1.091542
CCM 3.2	0.469161	7.766555	0.054747	75.77722	0.044201	41.48693	0.324034	4.90845
CCM 3.3	0.248406	5.944235	0.078699	27.15217	0.050297	12.02614	0.831476	1.107568

Table 13. Coffee Creamer Brands, ICP-MS Concentrations for Macronutrients (1st run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CRM 1.1	7322.361	0.629418	909.6498	0.79596	37435.08	1.526357	8890.513	0.952936
CRM 1.2	7177.186	0.696833	922.8142	0.354272	38921.92	0.213236	9337.92	1.074024
CRM 1.3	9618.576	1.171505	1043.004	0.333896	44204.45	0.602822	10654.65	1.151347
CRM 2.1	1871.665	0.504764	145.2074	1.059923	73784.02	0.389872	2651.804	2.179443
CRM 2.2	1919.308	0.613826	142.5617	1.351601	70012.44	1.613346	2662.358	0.738691
CRM 3.3	1994.185	0.478822	144.9742	0.951774	74893.79	0.915691	2736.754	2.780101

Table 14. Coffee Creamer Brands, ICP-MS Concentrations for Micronutrients (1st run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CRM 1.1	0.662745	14.96514	10.08889	1.320721	46.16715	1.208568	85.09931	1.707651
CRM 1.2	0.574373	6.057227	10.60713	2.270439	36.80778	1.318483	90.33412	1.168272
CRM 1.3	0.636633	8.794077	12.40169	3.062341	64.75023	1.477922	110.4167	0.820965
CRM 2.1	0.433791	6.3646	7.864777	2.239654	51.92196	0.46751	39.81334	3.183365
CRM 2.2	0.361487	5	9.97992	3.354042	49.1846	0.642476	43.46156	1.500538
CRM 3.3	0.447847	13.06755	8.651728	1.811959	40.54858	0.532328	57.99333	0.784291

Table 15. Coffee Creamer Brands, ICP-MS Concentrations for Heavy Metals

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CRM 1.1	0.159291	8.699309	0.061591	44.09586	0.01829	57.2822	0.412365	3.728654
CRM 1.2	0.128558	6.584964	0.061591	16.66667	0.020576	72.86043	0.16055	3.032117
CRM 1.3	0.185922	9.532104	0.102651	45.82576	0.038104	6.928203	0.295914	3.865346
CRM 2.1	0.190532	14.67182	0.054747	10.82532	0.029721	42.82896	0.209782	2.446539
CRM 2.2	0.223312	8.710933	0.085543	27.71281	0.029721	30.76923	0.190236	5.362507
CRM 3.3	0.237141	7.728633	0.068434	56.78908	0.045725	21.79449	0.315253	3.860832

Table 16. Rice Milk Brands, ICP-MS Concentrations for Macronutrients (1^{st} run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
RM 1.1	8813.551	0.987627	239.0889	1.016024	4289.1	0.624263	2889.338	2.953666
RM 1.2	9754.639	1.154639	250.4472	1.125622	4647.101	0.337884	3118.903	2.322919
RM 1.3	9196.195	1.578758	275.1162	1.521156	4204.534	0.58488	6669.675	0.691961
RM 2.1	11626.54	1.204418	196.9742	1.026795	2267.505	1.192839	7092.837	1.396231
RM 2.2	10367.58	0.922471	186.8157	0.396214	2308.742	1.182806	6795.481	1.259674
RM 3.3	10417.5	0.318193	177.1436	0.389621	2219.929	0.141509	6927.198	3.638335

Table 17. Rice Milk Brands, ICP-MS Concentrations for Micronutrients (1^{st} run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
RM 1.1	4.589334	5.226314	8.644475	3.157455	26.37866	0.590941	9.964253	1.938333
RM 1.2	6.533768	3.180888	77.97665	9.034741	27.47873	0.466541	11.28453	4.236119
RM 1.3	12.21325	2.122781	21.61517	0.760925	28.82091	0.298655	20.17102	0.500921
RM 2.1	12.80597	2.804246	22.09135	0.856407	21.86051	1.448841	22.00511	4.536268
RM 2.2	12.75372	2.195864	24.0775	0.329052	29.46669	1.18553	19.53173	1.657719
RM 3.3	12.47643	1.715296	21.3789	1.195534	32.2969	1.234083	17.48819	2.70305

Table 18. Rice Milk Brands, ICP-MS Concentrations for Heavy Metals (1^{st} run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
RM 1.1	0.202314	6.891186	0.87938	6.637635	0.04807	30.19934	0.347449	2.904967
RM 1.2	0.353919	4.880076	0.923862	11.60034	0.032688	26.95633	0.327275	2.185479
RM 1.3	0.30782	13.63609	1.002562	2.955718	0.040379	37.79645	0.258493	3.821298
RM 2.1	0.217676	0.816705	0.35928	15.11858	0.030765	57.2822	0.200061	3.584503
RM 2.2	0.245846	4.998688	0.372967	15.15845	0.021151	31.49183	0.230374	5.717603
RM 3.3	0.244823	9.081216	0.33875	8.017428	0.036533	9.116057	0.166821	7.74808

Table 19. Oat Milk Brands, ICP-MS Concentrations for Macronutrients (1st run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
OM 1.1	9567.935	0.313973	482.0932	0.992117	49102.25	0.676653	15488.9	0.642719
OM 1.2	10282.18	1.006916	494.5942	0.39442	51998.53	0.648689	16196.83	0.795703
OM 1.3	9712.784	0.825419	471.528	1.433057	50541.74	0.405864	15692.4	1.219596
OM 2.1	7766.203	0.588009	376.5982	0.509102	58868.07	0.574189	7723.358	0.638574
OM 2.2	8803.527	0.164073	428.6204	0.701435	65942.22	0.835711	8853.582	1.022501
OM 2.3	9874.483	0.822811	439.3229	0.448252	67996.27	0.645581	9008.824	1.492177

Table 20. Oat Milk Brands, ICP-MS Concentrations for Micronutrients (1st run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
OM 1.1	33.60214	2.60366	38.25319	0.67628	35.61093	0.586607	34.75034	1.873205
OM 1.2	34.86693	1.108387	37.39199	1.910676	34.92049	0.819003	45.10749	0.86831
OM 1.3	34.2335	1.048165	36.44352	0.751052	30.66785	1.742273	39.98115	2.679803
OM 2.1	16.90722	1.986558	45.74747	0.564227	40.88903	0.47558	18.02788	3.776727
OM 2.2	19.34894	1.659531	52.83998	1.120184	40.97601	0.347393	25.28019	2.145382
OM 2.3	19.58004	3.062311	53.61443	0.700969	43.5694	0.299538	26.33356	2.076732

Table 21. Oat Milk Brands, ICP-MS Concentrations for Heavy Metals (1st run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
OM 1.1	0.536771	9.41367	0.11976	32.4509	0.143272	5.604071	0.354349	4.215485
OM 1.2	0.592602	0.539078	0.078699	19.92424	0.124982	12.97792	0.214068	1.22847
OM 1.3	0.524479	1.860724	0.11976	17.84285	0.134127	4.289679	0.22797	1.906001
OM 2.1	0.358529	10.99702	0.147134	29.0465	0.043439	22.94157	0.179887	3.23965
OM 2.2	0.44765	1.726975	0.171086	21.07131	0.070874	31.10855	0.246367	0.847273
OM 2.3	0.508601	1.98142	0.150555	43.30127	0.06935	30.63156	0.354036	3.046586

Table 22. Hemp Milk Brands, ICP-MS Concentrations for Macronutrients (1^{st} run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
HM 1.1	11353.99	1.860748	2848.632	3.045312	17354.52	0.750538	11328.51	1.940539
HM 1.2	12115.81	0.268295	3035.736	0.630768	18126.67	0.776987	11994.38	0.739943
HM 1.3	12005.54	0.863833	3131.71	0.638801	19363.41	0.450503	12354.19	2.119045
HM 2.1	9677.417	0.50704	94.44092	0.256054	1058.613	0.80925	1038.818	6.080799
HM 2.2	10339.71	2.715825	100.5544	0.46344	1164.399	0.375749	1108.915	2.228008
HM 2.3	10826.45	0.819132	100.9647	2.930317	1235.102	0.853307	1147.345	3.839627

Table 23. Hemp Milk Brands, ICP-MS Concentrations for Micronutrients (1^{st} run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
HM 1.1	75.16716	0.897369	133.2013	0.316413	50.0011	0.525634	116.1674	0.900673
HM 1.2	79.79796	0.491967	136.4022	0.337401	24.03638	1.534866	122.26	2.338523
HM 1.3	82.22139	0.151394	143.8004	0.981849	56.70819	0.486782	127.8387	1.255284
HM 2.1	0.389611	12.96774	14.60098	0.451668	7.527042	1.062565	3.190301	9.998999
HM 2.2	0.47597	14.59836	15.43189	0.396298	14.82493	1.519304	4.594277	3.426551
HM 2.3	0.538224	11.65	14.89653	2.335176	25.19697	0.854054	3.87655	9.630891

Table 24. Hemp Milk Brands, ICP-MS Concentrations for Heavy Metals (1^{st} run)

	Cr [He]		As [He]		Cd [He]		208 Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
HM 1.1	1.124789	3.142468	0.547474	10.6617	0.070112	10.48223	0.25473	4.889235
HM 1.2	1.201108	5.452502	0.434557	5.455278	0.064777	17.41017	0.197448	1.840848
HM 1.3	1.31175	2.513529	0.526944	16.56799	0.059442	11.53846	0.218668	3.00937
HM 2.1	0.115239	6.928203	0.037639	15.74592	0.025149	24.05228	0.161595	0.784627
HM 2.2	0.142896	8.534682	0.065013	48.23764	0.039628	26.0149	0.231419	3.125766
HM 2.3	0.153655	17.32363	0.075278	67.26658	0.015242	37.74917	0.204451	3.169485

Table 25. Cashew Milk Brands, ICP-MS Concentrations for Macronutrients (1st run)

	Na [He]		Mg [No Gas]		K [He]		Ca [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CAM 1.1	10726.12	0.45763	893.887	0.845441	7010.474	0.368211	1523.663	1.798977
CAM 1.2	10435.18	0.28832	842.1092	0.428396	6829.678	0.343367	1428.209	0.409428
CAM 1.3	10597.78	1.094284	929.4046	0.631675	6858.633	0.699064	1554.921	1.325506
CAM 2.1	1618.016	0.442917	834.7678	0.765824	8002.118	0.478956	819.2421	5.181908
CAM 2.2	1716.127	0.758202	846.2082	1.137596	7713.063	0.487086	757.5951	2.022358
CAM 2.3	1405.938	1.01746	813.9181	0.898618	7202.782	0.58052	855.1333	1.863445

Table 26. Cashew Milk Brands, ICP-MS Concentrations for Micronutrients (1st run)

	Mn [He]		Fe [He]		Cu [He]		Zn [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CAM 1.1	14.31094	1.070107	61.79868	0.46064	26.61709	0.687588	44.0486	1.212114
CAM 1.2	13.75436	2.80517	59.55902	0.276315	46.58249	1.472843	46.78499	1.484758
CAM 1.3	14.21446	1.891944	62.41576	0.771717	58.31648	0.224568	44.31079	1.120121
CAM 2.1	18.12301	1.25498	52.97955	0.832831	22.78181	0.739953	43.67127	0.576211
CAM 2.2	18.11499	6.225264	53.38135	1.122351	48.95006	0.467428	52.54102	3.91839
CAM 2.3	18.09285	0.470036	53.54769	0.736766	22.64484	0.666658	43.92285	0.530021

Table 27. Cashew Milk Brands, ICP-MS Concentrations for Heavy Metals (1st run)

	Cr [He]		As [He]		Cd [He]		Pb [He]	
	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD	Conc. [ppb]	Conc. RSD
CAM 1.1	0.465578	8.420471	0.037639	62.98367	0.021338	16.36634	0.166821	2.03336
CAM 1.2	0.441501	1.607044	0.02053	50	0.020576	22.22222	0.191908	2.287644
CAM 1.3	0.421016	8.225712	0.034217	17.32051	0.019052	13.85641	0.193789	3.158809
CAM 2.1	0.423576	4.236178	0.054747	39.03124	0.024387	44.30452	0.173302	3.835592
CAM 2.2	0.398478	6.858306	0.044482	26.64694	0.028959	22.79014	0.197552	2.47905
CAM 2.3	0.343164	9.394669	0.075278	39.36479	0.01448	36.46423	0.153546	1.505739

APPENDIX D

ICP-MS CALCULATIONS FOR COW MILK

Table 28. Calculated Concentration of Macronutrients in Cow Milk (Run 1)

	Na		**Mg**		**K**		**Ca**	
Cow Milk	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %
CM 1.1	2944.14504	0.305567	1010.046	0.687376	10232.47	0.675222	2650.936	1.433115
CM 1.2	2771.92871	1.337092	952.2898	0.419836	9498.592	1.595024	2484.855	0.630383
CM 1.3	3580.36873	3.949326	984.0027	3.17246	12065.27	5.202788	3128.624	4.874268
Average	3098.8	1.9	982.1	1.4	10598.8	2.5	2754.8	2.3
Dilution factor (2.5)	7747.0		2455.3		26496.9		6887.0	
CM 2.1	4310.33224	4.465697	1094.447	0.080962	17040.41	6.455866	5117.583	7.382504
CM 2.2	5737.94667	2.589328	1124.773	0.396945	23025.74	4.387099	6881.468	3.351765
CM 2.3	7731.98426	1.629419	1086.648	0.12195	27714.39	3.430936	8151.031	2.13812
Avg	5926.8	2.9	1102.0	0.2	22593.5	4.8	6716.7	4.3
D.F (2.5)	14816.9		2754.9		56483.8		16791.7	
CM 3.1	6927.54692	1.681113	971.789	0.237517	31646.2	1.411661	10305.83	1.160965
CM 3.2	7820.17749	2.050885	1074.804	0.236687	34635.95	0.5866	11556.89	1.980322
CM 3.3	8152.7888	1.849272	1240.624	0.67426	39289.92	0.635792	13470.17	0.569584
Avg	7633.5	1.9	1095.7	0.4	35190.7	0.9	11777.6	1.2
D.F (2.5)	19083.8		2739.3		87976.7		29444.1	
CM 4.1	6831.29957	1.794961	998.5365	0.669135	33606.32	0.316384	8551.269	0.74421
CM 4.2	7271.27165	0.696873	936.5872	0.335501	34629.44	0.383129	8493.154	1.672851
CM 4.3	7529.98444	0.795709	969.9379	0.938421	36901.98	1.005502	9134.333	1.277182
Avg	7210.9	1.1	968.4	0.6	35045.9	0.6	8726.3	1.2
D.F (2.5)	18027.1		2420.9		87614.8		21815.6	
D.F Avg	14918.7		2592.6		64643.1		18734.6	

Table 29. Calculated Concentration of Micronutrients in Cow Milk (Run 1)

	Fe		**Cu**		**Zn**		**Mn**	
	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %
CM 1.1	3.88712134	2.345148	1.19664	2.829882	28.96954	1.562786	0.186774	23.26177
CM 1.2	2.49167203	4.101622	10.98649	1.782525	27.54933	3.054769	0.164676	9.208924
CM 1.3	5.95088448	8.189717	14.02237	3.800951	32.4809	4.625522	0.253053	14.86833
Average	4.1	4.9	8.7	2.8	29.7	3.1	0.2	15.8
Dilution factor (2.5)	10.3		21.8		74.2		0.5	
CM 2.1	4.67991387	6.815426	18.48833	4.693404	61.60594	6.035198	0.3635	7.653475
CM 2.2	8.41807896	2.613628	20.74449	4.465179	68.85229	3.483164	0.457896	8.219099
CM 2.3	14.0886786	2.532839	32.92336	2.590195	84.0031	1.865486	0.530199	6.912088
Avg	9.1	4.0	24.1	3.9	71.5	3.8	0.5	7.6
D.F	22.7		60.1		178.7		1.1	
CM 3.1	10.7548305	3.669118	28.75132	2.261026	137.0656	2.248772	0.672788	12.67532
CM 3.2	11.1374834	3.761476	32.19612	2.24942	150.069	0.860553	0.753117	6.248716
CM 3.3	12.3229695	2.442663	36.00339	0.644643	155.0461	1.659941	0.791278	8.557665
Avg	11.4	3.3	32.3	1.7	147.4	1.6	0.7	9.2
D.F	28.5		80.8		368.5		1.8	
CM 4.1	9.76198129	2.265184	28.5078	1.048219	75.95804	2.051209	0.514125	7.804013
CM 4.2	7.26915126	1.824174	29.66896	0.878815	73.52482	1.815831	0.486014	3.118527
CM 4.3	9.87819068	0.913762	49.59721	0.233268	80.06442	2.085035	0.558311	19.01144
Avg	9.0	1.7	35.9	0.7	76.5	2.0	0.5	10.0
D.F	22.4		89.8		191.3		1.3	
D.F Avg	21.0		63.1		203.2		1.2	

Table 30. Calculated Concentration of Pb in Cow Milk (run 1)

	Pb	
	Conc ppb	RSD %
CM 1.1	0.0472446	13.83219
CM 1.2	0.20925955	3.00201
CM 1.3	0.10410519	7.720699
Average	0.1	8.2
Dilution factor (2.5)	0.3	
CM 2.1	0.08968074	9.111185
CM 2.2	0.14988765	2.301383
CM 2.3	0.30793637	6.370069
Avg	0.2	5.9
D.F	0.5	
CM 3.1	0.12595164	5.110813
CM 3.2	0.21657682	7.498908
CM 3.3	0.13922625	1.84333
Avg	0.2	4.8
D.F	0.4	
CM 4.1	0.21270936	2.350897
CM 4.2	0.14037577	7.602841
CM 4.3	0.26371952	2.221536
Avg	0.2	4.1
D.F	0.5	
D.F Avg	0.4	

Table 31. Calculated Concentration of Macronutrients in Cow Milk (Run 2)

	Na		**Mg**		**K**		**Ca**	
	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %
CM 1.1	3673.0	3.3	1050.694	1.696446	21045.16	0.748417	2749.557	3.784518
CM 1.2	3601.3	6.1	1043.05	0.82566	21002.32	0.827705	2726.181	2.138775
CM 1.3	3399.1	7.0	952.8568	7.87353	19908.94	0.696015	2477.733	2.11818
Average	3557.8	5.5	1015.5	3.5	20652.1	0.8	2651.2	2.7
Dilution factor (2.5)	8894.5		2538.8		51630.3		6627.9	
CM 2.1	3309.7	7.6	911.4841	0.355975	18540.23	0.414345	2652.049	4.255147
CM 2.2	3646.1	3.8	1087.267	2.694751	21152.44	0.527758	3001.317	10.23563
CM 2.3	4124.5	9.2	1256.67	9.027806	24219.14	1.103218	3357.691	0.830271
Avg	3693.4	6.9	1085.1	4.0	21303.9	0.7	3003.7	5.1
D.F (2.5)	9233.6		2712.9		53259.9		7509.2	
CM 3.1	2400.7	4.9	621.5457	0.994497	14178.37	0.40661	2197.755	3.015536
CM 3.2	2454.5	7.7	675.584	3.515099	14877.31	0.413341	2181.168	0.971279
CM 3.3	2375.1	4.5	574.3839	13.6091	15891.32	0.905318	2391.595	0.955227
Avg	2410.1	5.7	623.8	6.0	14982.3	0.6	2256.8	1.6
D.F (2.5)	6025.3		1559.6		37455.8		5642.1	
CM 4.1	2388.2	15.8	588.637	0.696227	15433.76	0.393105	1804.71	1.806898
CM 4.2	2301.9	6.6	578.7443	1.111507	14878.12	0.228413	1687.259	1.490465
CM 4.3	2299.8	1.4	616.6412	1.082821	15675.43	1.013751	1780.893	2.4084
Avg	2330.0	7.9	594.7	1.0	15329.1	0.5	1757.6	1.9
D.F (2.5)	5824.9		1486.7		38322.8		4394.1	
D.F Avg	7494.6		2074.5		45167.2		6043.3	

Table 32. Calculated Concentrations of Micronutrients in Cow Milk (Run 2)

	Fe		**Cu**		**Zn**	
	Conc ppb	RSD %	Conc ppb	RSD %	Conc ppb	RSD %
CM 1.1	2.826658	2.450541	7.115554	1.909634	37.15827	5.892181
CM 1.2	1.733559	4.100325	7.064046	1.1557	39.35575	5.96662
CM 1.3	1.179833	8.050411	6.595754	2.788941	35.16582	8.794139
Average	1.9	4.9	6.9	2.0	37.2	6.9
Dilution factor (2.5)	4.8		17.3		93.1	
CM 2.1	6.206588	5.624737	6.868379	5.558079	36.88523	8.746687
CM 2.2	1.611037	16.67403	7.042018	9.301768	42.39636	6.950546
CM 2.3	2.502317	3.437297	7.200353	0.995963	49.40106	6.572097
Avg	3.4	8.6	7.0	5.3	42.9	7.4
D.F	8.6		17.6		107.2	
CM 3.1	1.89894	6.879766	0.807093	5.956234	37.86807	4.812044
CM 3.2	1.429474	1.353758	0.89544	4.116963	37.75111	5.645047
CM 3.3	3.15282	1.939877	0.90074	1.928168	41.69234	8.294038
Avg	2.2	3.4	0.9	4.0	39.1	6.3
D.F	5.4		2.2		97.8	
CM 4.1	1.056144	11.73284	1.439945	2.804188	22.17877	6.40632
CM 4.2	1.088218	2.707822	1.362836	3.119147	21.39651	3.749826
CM 4.3	1.046435	2.518408	1.15243	3.976321	22.08099	4.477145
Avg	1.1	5.7	1.3	3.3	21.9	4.9
D.F	2.7		3.3		54.7	
D.F Avg	5.4		10.1		88.2	

Table 33. Overall Concentrations of Essential Elements and Heavy Metals in Cow Milk Analyzed with ICP-MS

		Na	**Mg**	**K**	**Ca**
		Conc ppb	Conc ppb	Conc ppb	Conc ppb
Cow milk	Run 1	14918.7	2592.6	64643.1	18734.6
	Run 2	7494.6	2074.5	45167.2	6043.3
	Avg Conc	11206.6	2333.5	54905.1	12389.0
		Fe	**Cu**	**Zn**	**Mn**
		Conc ppb	Conc ppb	Conc ppb	Conc ppb
Cow milk	Run 1	21.0	63.1	203.2	1.2
	Run 2	5.4	10.1	88.2	BDL
	Avg Conc	13.2	36.6	145.7	1.2
		Pb	**Cd**	**Cr**	
		Conc ppb	Conc ppb	Conc ppb	
Cow milk	Run 1	0.4	BDL	BDL	
	Run 2	BDL	BDL	BDL	
	Avg Conc	0.4			

Table 34. Single Factor ANOVA for Sodium Concentration in Milk Brands

	Sodium (Na)										
CM 1	CM 2	CM 3	CM 4								
2944.15	4310.33	6927.547	6831.3		Anova: Single Factor						
2771.93	5737.95	7820.177	7271.272								
3580.37	7731.98	8152.789	7529.984		SUMMARY						
					Groups	*Count*	*Sum*	*Average*	*Variance*		
					CM 1	3	9296.442	3098.814	181335.7		
					CM 2	3	17780.26	5926.754	2953662		
					CM 3	3	22900.51	7633.504	401439.5		
					CM 4	3	21632.56	7210.852	124778		
					ANOVA						
					Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
					Between Groups	37657540	3	12552513	13.71404	0.001612	4.066181
					Within Groups	7322430	8	915303.8			
					Total	44979970	11				

APPENDIX E

OCP AVERAGE CONCENTRATION FOR MILK SAMPLES

Table 35. The Average Concentration of OCPs in Cow Milk

OCPs	CM1	CM2	CM3	CM4	Avg Conc	STD DEV ±
Alpha-chlordane	35.9	35.7	36.2	33.9	35.4	1.0
Gamma-chlordane	25.8	25.7	26.1	24.4	25.5	0.8
4,4-DDD	37.3	37.1	37.7	35.3	36.9	1.1
4,4-DDE	51.7	51.4	52.2	48.9	51.1	1.5
4,4-DDT	68.9	68.6	69.6	65.2	68.1	2.0
a-BHC	14.4	14.3	14.5	13.6	14.2	0.4
Aldrin	28.7	28.6	29.0	27.2	28.4	0.8
b-BHC	47.4	47.1	47.8	44.8	46.8	1.3
Chlordane	240.0	239.0	242.0	227.0	237.0	6.8
d-BHC	48.8	48.6	49.3	46.2	48.2	1.4
Dieldrin	35.9	35.7	36.2	33.9	35.4	1.0
Endosulfan I	48.8	48.6	49.3	46.2	48.2	1.4
Endosulfan II	40.2	40.0	40.6	38.0	39.7	1.2
Endosulfan sulfate	35.9	35.7	36.2	33.9	35.4	1.0
Endrin	56.0	55.7	56.5	52.9	55.3	1.6
Endrin aldehyde	58.9	58.6	59.4	55.7	58.2	1.7
Endrin ketone	47.4	47.1	47.8	44.8	46.8	1.3
g-BHC	21.5	21.4	21.7	20.4	21.3	0.6
Heptachlor	47.4	47.1	47.8	44.8	46.8	1.3
Heptachlor epoxide	37.3	37.1	37.7	35.3	36.9	1.1
Methoxychlor	81.8	81.4	82.6	77.4	80.8	2.3
Mirex	349.0	347.0	352.0	330.0	344.5	9.9
Toxaphene	240.0	239.0	242.0	227.0	237.0	6.8
Dicofol	957.4	952.9	966.7	905.5	945.6	27.4
Hexachlorobenzene	376.0	374.0	380.0	356.0	371.5	10.6
Kepone	339.0	337.0	342.0	320.0	334.5	9.9

Table 36. The Average Concentration of OCPs in Almond Milk

OCP	AM 1	AM 2	AM 3	AM 4	Avg Conc	STD DEV ±
Alpha-chlordane	34.4	36.8	37.1	35.2	35.9	1.3
Gamma-chlordane	24.8	26.5	26.7	25.4	25.9	0.9
4,4-DDD	35.8	38.2	38.6	36.6	37.3	1.3
4,4-DDE	49.5	52.9	53.5	50.7	51.7	1.9
4,4-DDT	66.1	70.6	71.3	67.6	68.9	2.5
a-BHC	13.8	14.7	14.9	14.1	14.4	0.5
Aldrin	27.5	29.4	29.7	28.2	28.7	1.0
b-BHC	45.4	48.5	49.0	46.5	47.4	1.7
Chlordane	230.0	246.0	248.0	235.0	239.8	8.7
d-BHC	46.8	50.0	50.5	47.9	48.8	1.7
Dieldrin	34.4	36.8	37.1	35.2	35.9	1.3
Endosulfan I	46.8	50.0	50.5	47.9	48.8	1.7
Endosulfan II	38.5	41.2	41.6	39.4	40.2	1.5
Endosulfan sulfate	34.4	36.8	37.1	35.2	35.9	1.3
Endrin	53.7	57.4	57.9	54.9	56.0	2.0
Endrin aldehyde	56.4	60.3	60.9	57.7	58.8	2.1
Endrin ketone	45.4	48.5	49.0	46.5	47.4	1.7
g-BHC	20.6	22.1	22.3	21.1	21.5	0.8
Heptachlor	45.4	48.5	49.0	46.5	47.4	1.7
Heptachlor epoxide	35.8	38.2	38.6	36.6	37.3	1.3
Methoxychlor	78.4	83.8	84.7	80.3	81.8	3.0
Mirex	334.0	357.0	361.0	342.0	348.5	12.7
Toxaphene	230.0	246.0	248.0	235.0	239.8	8.7
Dicofol	917.9	980.9	990.6	939.5	957.2	34.3
Hexachlorobenzene	361.0	385.0	389.0	369.0	376.0	13.2
Kepone	325.0	347.0	350.0	332.0	338.5	12.0

Table 37. The Average Concentration of OCPs in Coconut Milk

OCP	CCM1	CCM2	CCM 3	Avg Conc	STD DEV±
Alpha-chlordane	33.9	34.7	34.1	34.2	0.4
Gamma-chlordane	24.4	25.0	24.5	24.6	0.3
4,4-DDD	35.3	36.1	35.5	35.6	0.4
4,4-DDE	48.9	50.0	49.1	49.3	0.6
4,4-DDT	65.2	66.7	65.5	65.8	0.8
a-BHC	13.6	13.9	13.6	13.7	0.2
Aldrin	27.2	27.8	27.3	27.4	0.3
b-BHC	44.8	45.8	45.0	45.2	0.5
Chlordane	227.0	232.0	228.0	229.0	2.6
d-BHC	46.2	47.2	46.4	46.6	0.5
Dieldrin	33.9	34.7	34.1	34.2	0.4
Endosulfan I	46.2	47.2	46.4	46.6	0.5
Endosulfan II	38.0	38.9	38.2	38.4	0.5
Endosulfan sulfate	33.9	34.7	34.1	34.2	0.4
Endrin	52.9	54.2	53.2	53.4	0.7
Endrin aldehyde	55.7	56.9	55.9	56.2	0.6
Endrin ketone	44.8	45.8	45.0	45.2	0.5
g-BHC	20.4	20.8	20.5	20.6	0.2
Heptachlor	44.8	45.8	45.0	45.2	0.5
Heptachlor epoxide	35.3	36.1	35.5	35.6	0.4
Methoxychlor	77.4	79.2	77.7	78.1	1.0
Mirex	330.0	338.0	331.0	333.0	4.4
Toxaphene	227.0	232.0	228.0	229.0	2.6
Dicofol	905.5	926.4	909.5	913.8	11.1
Hexachlorobenzene	356.0	364.0	357.0	359.0	4.4
Kepone	320.0	328.0	322.0	323.3	4.2

Table 38. The Average Concentration of OCPs in Soy Milk

OCP	SM 1	SM 2	SM 3	SM 4	Avg Conc	STD DEV ±
Alpha-chlordane	36.6	35.4	35.4	36.9	36.1	0.8
Gamma-chlordane	26.3	25.5	25.5	26.6	26.0	0.6
4,4-DDD	38.0	36.8	36.8	38.4	37.5	0.8
4,4-DDE	52.7	50.9	50.9	53.2	51.9	1.2
4,4-DDT	70.2	67.9	67.9	70.9	69.2	1.6
a-BHC	14.6	14.2	14.2	14.8	14.5	0.3
Aldrin	29.3	28.3	28.3	29.6	28.9	0.7
b-BHC	48.3	46.7	46.7	48.8	47.6	1.1
Chlordane	244.0	236.0	236.0	247.0	240.8	5.6
d-BHC	49.8	48.1	48.1	50.2	49.1	1.1
Dieldrin	36.6	35.4	35.4	36.9	36.1	0.8
Endosulfan I	49.8	48.1	48.1	50.2	49.1	1.1
Endosulfan II	41.0	39.6	39.6	41.4	40.4	0.9
Endosulfan sulfate	36.6	35.4	35.4	36.9	36.1	0.8
Endrin	57.1	55.2	55.2	57.6	56.3	1.3
Endrin aldehyde	60.0	58.0	58.0	60.6	59.2	1.4
Endrin ketone	48.3	46.7	46.7	48.8	47.6	1.1
g-BHC	22.0	21.2	21.2	22.2	21.7	0.5
Heptachlor	48.3	46.7	46.7	48.8	47.6	1.1
Heptachlor epoxide	38.0	36.8	36.8	38.4	37.5	0.8
Methoxychlor	83.4	80.7	80.7	84.2	82.3	1.8
Mirex	356.0	344.0	344.0	359.0	350.8	7.9
Toxaphene	244.0	236.0	236.0	247.0	240.8	5.6
Dicofol	976.1	943.9	943.9	985.7	962.4	21.7
Hexachlorobenzene	383.0	371.0	371.0	387.0	378.0	8.2
Kepone	345.0	334.0	334.0	349.0	340.5	7.7

Table 39. The Average Concentration of OCPs in Oat Milk

OCP	OM 1	OM 2	Avg Conc	STD DEV ±
Alpha-chlordane	35.7	35.4	35.6	0.21
Gamma-chlordane	25.7	25.5	25.6	0.14
4,4-DDD	37.1	36.8	37.0	0.21
4,4-DDE	51.4	50.9	51.2	0.35
4,4-DDT	68.6	67.9	68.3	0.49
a-BHC	14.3	14.2	14.3	0.07
Aldrin	28.6	28.8	28.7	0.14
b-BHC	47.1	46.7	46.9	0.28
Chlordane	239.0	236.0	237.5	2.12
d-BHC	48.6	48.1	48.4	0.35
Dieldrin	35.7	35.4	35.6	0.21
Endosulfan I	48.6	48.1	48.4	0.35
Endosulfan II	40.0	39.6	39.8	0.28
Endosulfan sulfate	35.7	35.4	35.6	0.21
Endrin	55.7	55.2	55.5	0.35
Endrin aldehyde	58.6	58.0	58.3	0.42
Endrin ketone	47.1	46.7	46.9	0.28
g-BHC	21.4	21.2	21.3	0.14
Heptachlor	47.1	46.7	46.9	0.28
Heptachlor epoxide	37.1	36.8	37.0	0.21
Methoxychlor	81.4	80.7	81.1	0.49
Mirex	347.0	344.0	345.5	2.12
Toxaphene	239.0	236.0	237.5	2.12
Dicofol	952.9	943.9	948.4	6.37
Hexachlorobenzene	374.0	371.0	372.5	2.12
Kepone	337.0	334.0	335.5	2.12

Table 40. The Average Concentration of OCPs in Rice Milk

OCP	RM 1	RM 2	Avg Conc	STD DEV ±
Alpha-chlordane	35.5	3.6	19.6	22.5
Gamma-chlordane	256.0	2.6	129.3	179.2
4,4-DDD	37.0	3.8	20.4	23.5
4,4-DDE	51.2	5.2	28.2	32.5
4,4-DDT	68.2	7.0	37.6	43.3
a-BHC	14.2	1.5	7.8	9.0
Aldrin	28.4	2.9	15.7	18.0
b-BHC	46.9	4.8	25.9	29.8
Chlordane	237.0	24.3	130.7	150.4
d-BHC	48.3	5.0	26.6	30.7
Dieldrin	35.5	3.6	19.6	22.5
Endosulfan I	48.3	5.0	26.6	30.7
Endosulfan II	39.8	4.1	21.9	25.3
Endosulfan sulfate	35.5	3.6	19.6	22.5
Endrin	55.5	5.7	30.6	35.2
Endrin aldehyde	58.3	6.0	32.1	37.0
Endrin ketone	46.9	4.8	25.9	29.8
g-BHC	21.3	2.2	11.7	13.5
Heptachlor	46.9	4.8	25.9	29.8
Heptachlor epoxide	37.0	3.8	20.4	23.5
Methoxychlor	81.0	8.3	44.7	51.4
Mirex	345.0	35.4	190.2	218.9
Toxaphene	237.0	24.3	130.7	150.4
Dicofol	948.3	97.1	522.7	601.9
Hexachlorobenzene	373.0	38.1	205.6	236.8
Kepone	336.0	34.4	185.2	213.3

Table 41. The Average Concentration of OCPs in Hemp Milk

OCP	HM 1	HM 2	Avg Conc	STD DEV ±
Alpha-chlordane	3.64	3.71	3.68	0.05
Gamma-chlordane	2.62	2.67	2.65	0.04
4,4-DDD	3.79	3.86	3.83	0.05
4,4-DDE	5.24	5.35	5.30	0.08
4,4-DDT	6.99	7.13	7.06	0.10
a-BHC	1.46	1.48	1.47	0.01
Aldrin	2.91	2.97	2.94	0.04
b-BHC	4.80	4.90	4.85	0.07
Chlordane	24.30	24.80	24.55	0.35
d-BHC	4.95	5.05	5.00	0.07
Dieldrin	3.64	3.71	3.68	0.05
Endosulfan I	4.95	5.05	5.00	0.07
Endosulfan II	4.00	4.16	4.08	0.11
Endosulfan sulfate	3.64	3.71	3.68	0.05
Endrin	5.68	5.79	5.74	0.08
Endrin aldehyde	5.97	6.09	6.03	0.08
Endrin ketone	4.80	4.90	4.85	0.07
g-BHC	2.18	2.23	2.21	0.04
Heptachlor	4.80	4.90	4.85	0.07
Heptachlor epoxide	3.79	3.86	3.83	0.05
Methoxychlor	8.30	8.46	8.38	0.11
Mirex	35.40	36.10	35.75	0.49
Toxaphene	24.30	24.80	24.55	0.35
Dicofol	97.12	99.05	98.09	1.36
Hexachlorobenzene	38.10	38.90	38.50	0.57
Kepone	34.40	35.00	34.70	0.42

Table 42. The Average Concentration of OCPs in Cashew Milk

OCP	CAM 1	CAM 2	Avg	STD DEV ±
Alpha-chlordane	3.70	3.68	3.69	0.01
Gamma-chlordane	2.66	2.65	2.66	0.01
4,4-DDD	3.84	3.82	3.83	0.01
4,4-DDE	5.32	5.30	5.31	0.01
4,4-DDT	7.09	7.06	7.08	0.02
a-BHC	1.48	1.47	1.48	0.01
Aldrin	2.96	2.94	2.95	0.01
b-BHC	4.88	4.50	4.69	0.27
Chlordane	24.70	24.60	24.65	0.07
d-BHC	5.03	5.00	5.02	0.02
Dieldrin	3.70	3.68	3.69	0.01
Endosulfan I	5.03	5.00	5.02	0.02
Endosulfan II	4.00	4.12	4.06	0.08
Endosulfan sulfate	3.70	3.68	3.69	0.01
Endrin	5.76	5.74	5.75	0.01
Endrin aldehyde	6.06	6.03	6.05	0.02
Endrin ketone	4.88	4.85	4.87	0.02
g-BHC	2.22	2.21	2.22	0.01
Heptachlor	4.88	4.85	4.87	0.02
Heptachlor epoxide	3.84	3.82	3.83	0.01
Methoxychlor	8.42	8.38	8.40	0.03
Mirex	35.90	35.70	35.80	0.14
Toxaphene	24.70	24.60	24.65	0.07
Dicofol	98.58	98.12	98.35	0.33
Hexachlorobenzene	38.70	38.50	38.60	0.14
Kepone	34.90	34.70	34.80	0.14

Table 43. The Average Concentration of OCPs in Coffee Creamer

OCP	CRM 1	CRM 2	Avg	STD DEV ±
Alpha-chlordane	3.52	3.64	3.58	0.08
Gamma-chlordane	2.53	2.62	2.575	0.06
4,4-DDD	3.66	3.79	3.725	0.09
4,4-DDE	5.07	5.24	5.155	0.12
4,4-DDT	6.76	6.99	6.875	0.16
a-BHC	1.41	1.46	1.435	0.04
Aldrin	2.82	2.91	2.865	0.06
b-BHC	4.65	4.8	4.725	0.11
Chlordane	23.5	24.3	23.9	0.57
d-BHC	4.79	4.95	4.87	0.11
Dieldrin	3.52	3.64	3.58	0.08
Endosulfan I	4.79	4.95	4.87	0.11
Endosulfan II	3.94	4.08	4.01	0.10
Endosulfan sulfate	3.52	3.64	3.58	0.08
Endrin	5.49	5.68	5.585	0.13
Endrin aldehyde	5.77	5.97	5.87	0.14
Endrin ketone	4.65	4.8	4.725	0.11
g-BHC	2.11	2.18	2.145	0.05
Heptachlor	4.65	4.8	4.725	0.11
Heptachlor epoxide	3.66	3.79	3.725	0.09
Methoxychlor	8.03	8.3	8.165	0.19
Mirex	34.2	35.4	34.8	0.85
Toxaphene	23.5	24.3	23.9	0.57
Dicofol	93.91	97.12	95.515	2.27
Hexachlorobenzene	36.9	38.1	37.5	0.85
Kepone	33.2	34.4	33.8	0.85

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