5-6-2024

Toxic Chocolate: An Investigation of Heavy Metal Concentrations in Chocolate Products

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Honors Thesis

2024

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By: Meredith Hoff

May 6th, 2024

Abstract

Many news organizations and scientific journals have recently reported high concentrations of lead and cadmium in chocolate products. The consumption of lead, cadmium, and other toxic metals can lead to numerous health impacts and impair development in children.[1,2] Previous studies have specifically focused on the quantification of lead and cadmium in mainstream chocolate products, neglecting small businesses and the quantification of other heavy metals. This study addresses this disparity by analyzing the concentrations of lead, cadmium, barium, strontium, thallium, and mercury in lesser-known chocolate products from Castronovo Chocolate, Meridian Cacao Co., and Lidl. Two ICP-OES instruments and a direct mercury analyzer were used to quantify the heavy metals in the samples. Little to no lead was detected in the samples. Many samples contained mercury at ppb concentrations, which were not high enough to be immediate health risks. Cadmium was reliably detected in Castronovo's The Lost City Honduras dark chocolate and Meridian's Organic Cocoa Powder at average concentrations of 780 ppb and 364 ppb, respectively. Barium, strontium, and thallium were detected in many of the chocolate products, ranging from 1.97-7.71 ppm, 3.01-6.70 ppm, and 3.82-6.07 ppm, respectively. Consuming chocolate with these metal concentrations could result in psychological issues, paralysis, numbness, and other health impacts based on EPA recommendations.[3-6] The high concentrations of the heavy metals in the chocolate products indicate that the metals are entering the cacao beans through polluted soil or that manufacturing processes are introducing the metals into the finished chocolate products.

Acknowledgements

First, I would like to thank my honors thesis advisor, Dr. Grace Schwartz. She has graciously spent her time inside and outside of the lab with me, teaching me how to operate unfamiliar instruments and guiding me through the writing process. She has been a wonderful mentor, and I am grateful for her continuous support, patience, and encouragement over the past year.

I would also like to thank my thesis committee members, Dr. Zachary Davis and Dr. Kelli Carroll. Dr. Davis has fostered my love for analytical chemistry and instrumental analysis, and he has been a “rockin!” advisor and teacher since my freshman year. I am grateful for Dr. Carroll’s expertise and time spent reviewing my work, and I am honored to have her on my committee.

I am thankful to Dr. Jun Ohata at North Carolina State University for allowing me to work in his lab for a summer and his constant support and advice during the past two years. I would also like to thank my research mentor at NCSU, Mohammad Nuruzzaman, for working closely with me and helping develop my research skills.

Additionally, I would like to thank Dr. Christy Haynes at the University of Minnesota-Twin Cities for being a caring and trusted advisor and giving me the opportunity to join her group for a summer. I am grateful to my mentor at UMN, Clare Froehlich, for her guidance and for allowing me to flourish as an independent researcher.

I am thankful to my closest friends- Sydney, Taylor, and Maddie- for always encouraging me and making me laugh, especially when times were stressful. They have kept me grounded over the past four years, and I am forever grateful to them.

Lastly, I thank my family- Mom, Dad, and Meleah- for their unwavering support of my academic and personal endeavors. Without their support and guidance, I would not be where I am today. Everything I do is to make them proud.

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Dedicated to my family: Mom, Dad, and Meleah

Chapter 1

Introduction

Recently, many news sources and scientific journals have reported increased concentrations of heavy metals, specifically cadmium and lead, in popular dark chocolate brands. Consumer Reports' article entitled "Lead and Cadmium Could Be in Your Dark Chocolate" brought great notice to this issue, as they found alarming levels of these toxic metals in Hershey's, Trader Joe's, Lindt, Godiva, and other notable chocolate brands.[7] High levels of lead in adults have been known to cause high blood pressure and kidney damage. Children are even more susceptible to health issues caused by lead poisoning, especially with the development of their brains and nervous systems.[2] Cadmium is known to cause reproductive harm alongside damage to the liver, kidneys, and bones.[1] As many people consume dark chocolate daily, further research must be done to decipher which chocolate brands contain levels of lead and cadmium that exceed allowable doses.

Heavy metals typically infiltrate chocolate products through contaminated soil or post-harvest practices. When cacao beans are planted in contaminated soil, the metals can be absorbed through the plant's roots and eventually reside in the cacao pod itself. Soil pH, phosphorus content, and fertilizer application all impact the uptake of cadmium into the plant. Furthermore, the post-harvest transformation of cacao from seeds to beans and eventually nibs can introduce metals, especially in the roasting and dehulling stages as the shells can contain 2-4 times the concentration of a given heavy metal than the nibs. Manufacturing processes can also introduce heavy metals.[8] Figure 1 illustrates how lead and cadmium commonly infiltrate chocolate products.[9] It does not seem that the U.S. Food and Drug Administration (FDA) or any other government entity regularly tests chocolate products for heavy metal concentrations, which puts American consumers at risk of heavy metal exposure from chocolate consumption.

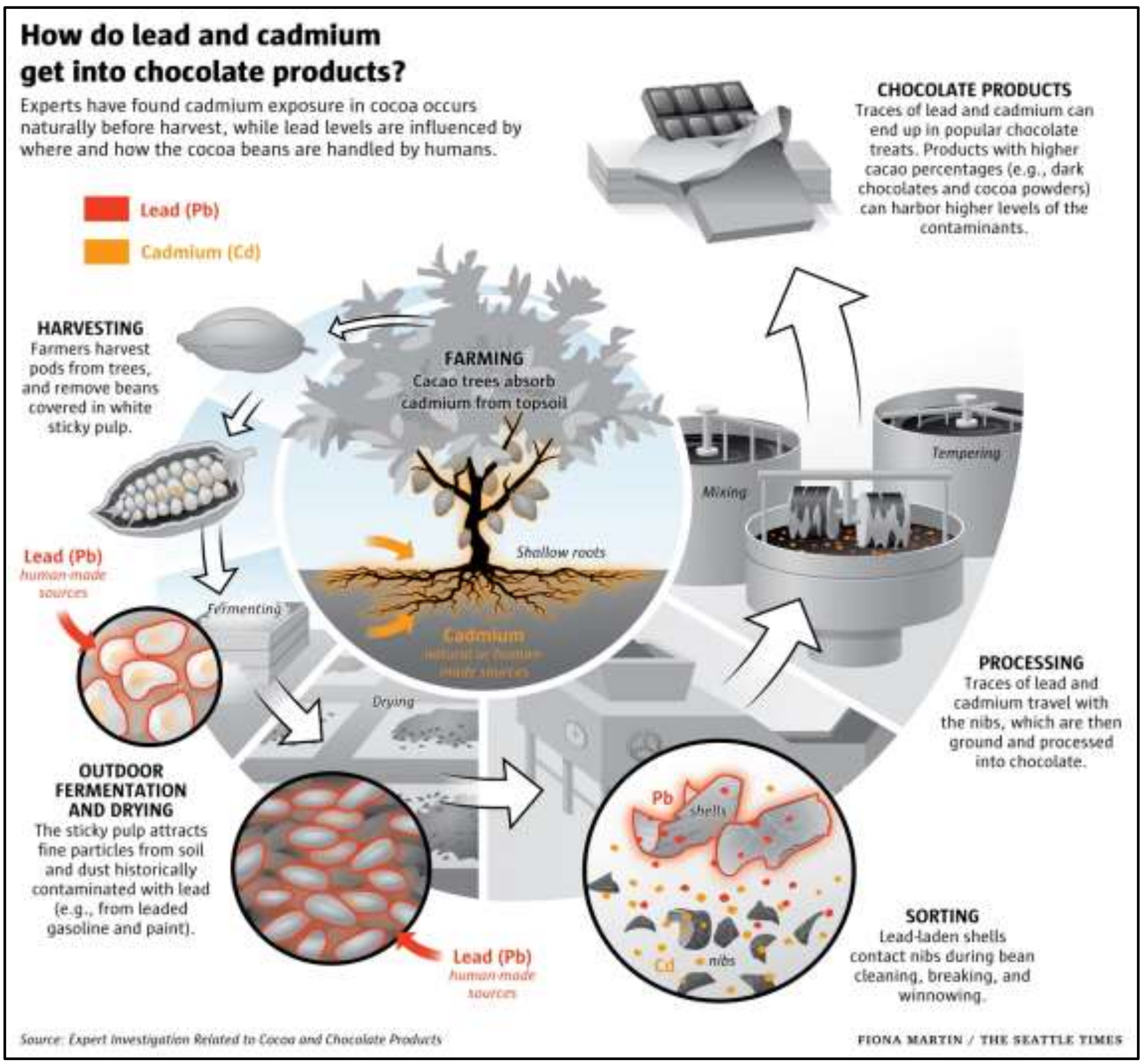

Figure 1: Heavy Metal Infiltration of Chocolate Products[9]

Previous reports have disproportionately studied popular chocolate companies, while consumers frequently ingest chocolate products from local or regional businesses. Therefore, this study analyzes the toxic metal contents of chocolate from two small businesses and brings awareness to this ongoing and dangerous issue. Additionally, many brands source their cacao from countries in West Africa and Central America, so this study investigates if the soil of specific regions results in increased concentrations of lead, cadmium, and other toxic metals in finished chocolate products. The metal contents of dark chocolate bars were tested using two inductively coupled plasma optical emission spectroscopy (ICP-OES) instruments and a direct mercury analyzer, with a special emphasis on screening heavy metals other than lead and cadmium.

Chapter 2

Methodology

2.1 Supplies and Instrumentation

Chocolate products were purchased from Castronovo Chocolate, Meridian Cacao Co., and Lidl (Table 2.1.1). Standard reference material (SRM) Montana II Soil and DORM-5 Fish Protein were purchased from the National Institute of Standards and Technology and the National Research Council Canada, respectively, and a 1000 μg/mL mercury SRM was purchased from Thermo Scientific. For simplicity, the chocolate products and other samples were given abbreviations (Table 2.1.2). The instrumentation used in this study includes an Environmental Express HotBlock SC154 from Wofford College, a Perkin Elmer Optima 8000 ICP-OES from Greenville Water, a Teledyne Prodigy 7 ICP-OES from Furman University, and a LUMEX RA915-LAB Mercury Analyzer from Wofford College. As two ICP-OES instruments were used in this study, ICP-OES Method 1 will refer to the Perkin Elmer Optima 8000 ICP-OES, and ICP-OES Method 2 will refer to the Teledyne Prodigy 7 ICP-OES.

Table 2.1.1: Chocolate Product Information

Chocolate Brand	Product Label	Country of Origin/ Type of Cacao*	% Cocoa
Castronovo Chocolate	Sierra Nevada Colombia	Colombia/ Criollo & Trinitario Cacao	72%
	The Lost City Honduras	Honduras/ Rainforest Cacao	72%
	Zorzal Dominican Republic	Dominican Republic/ Amelonado Cacao	72%
	Chuao Venezuela	Venezuela	72%
Meridian Cacao Co.	Organic Cacao Nibs	Kokoa Kamili, Tanzania	N/A
	Organic Cocoa Powder	Kokoa Kamili, Tanzania	N/A
Lidl Preferred Selection	Dark Chocolate	N/A	85%
	Madagascar Dark Chocolate	Madagascar	70%
	Peru Dark Chocolate	Peru	60%
	Grenada Dark Chocolate	Grenada	46%

*Not all products have the cacao's country of origin or type of cacao listed.

Table 2.1.2: Sample Abbreviations

SNC = Sierra Nevada Colombia	DC = Dark Chocolate
LCH = The Lost City Honduras	MA = Madagascar Dark Chocolate
ZDR = Zorzal Dominican Republic	PE = Peru Dark Chocolate
CV = Chuao Venezuela	GR = Grenada Dark Chocolate
NIB = Organic Cacao Nibs	WB = Water Blank
PO = Organic Cocoa Powder	-S = Spiked Sample

2.2 Acid Digestion for ICP-OES

To prepare the chocolate bars and nibs for acid digestion, small fragments of the products were crushed into a powder using a mortar and pestle. Three samples of each product were prepared by weighing out approximately 1 gram in 50 mL digestion vessels. Two samples each of water blanks (approx. 1 g H_2O) and standard reference material soil (approx. 0.5 g) were also prepared in 50 mL digestion vessels. Two additional samples of a random chocolate product were prepared and spiked with 100 µg of Pb for a matrix study.

The acid digestion procedure was adapted from EPA Method 3050B.[10] First, 5 mL (1:1) HNO_3 + deionized (DI) water was added to each sample and swirled. The samples were covered with ribbed watch glasses and heated in the HotBlock at 95±5 $^{\circ}$C for 15 minutes without boiling. Then, the samples were cooled to room temperature. 2.5 mL concentrated HNO_3 was added to each sample, and samples were refluxed at 95 $^{\circ}$C for 30 minutes. This was repeated until no brown fumes were given off by the samples. Once no brown fumes appeared, the samples were heated with ribbed watch glasses to a volume of about 5 mL or for 2 hours at 95±5 °C, ensuring that the samples did not boil or go dry. The samples were removed from the HotBlock and cooled completely.

DI water (1 mL) and 1.5 mL 30% H_2O_2 were slowly added to each sample, and it took around 5-10 minutes for the consequent exothermic reactions to finish. Once the reactions were complete, the samples were placed back in the HotBlock with the ribbed watch glasses. When effervescence occurred, the samples were lifted out of the HotBlock, allowing the reactions to continue without additional heat. Care was taken to ensure that the samples did not foam out of the digestion vessels. Additional aliquots of 0.5 mL 30% H_2O_2 were added to the samples with heating until the samples remained unchanged in color (no longer than 30 minutes). No more than 5 mL of 30% H_2O_2 total was added to each sample. The samples were heated for another 2 hours or until the volume was reduced to approximately 5 mL.

Concentrated HCl (5 mL) was added to each sample, and the samples were covered with ribbed watch glasses. Samples were refluxed at 95 °C for 15 minutes. After cooling, the samples were diluted to the 50 mL mark with DI water and filtered to remove insoluble material. The Castronovo Chocolate and Meridian Cacao Co. products were analyzed using ICP-OES Method 1, while the Lidl products were analyzed with ICP-OES Method 2 due to instrument access issues.

2.3 Standard and Sample Preparation for Mercury Analyzer

Mercury standards were prepared to calibrate the instrument using a procedure from Lumex Instruments.[11] The (1:1) nitric acid solution was prepared by adding 50 mL of concentrated nitric acid to 50 mL of deionized water with stirring. The dilution solution was prepared by placing 500-600 mL of deionized water in a 1 L volumetric flask. Then, 200 mg of potassium dichromate ($K_2Cr_2O_7$) was added and mixed until complete dissolution. Lastly, 50 mL of the (1:1) nitric acid solution was added with stirring. After cooling, the mixture was diluted up to 1L with deionized water.

A standard curve consisting of 10 ppm (mg/L), 1000 ppb (μg/L), 100 ppb, and 10 ppb mercury stock solutions was prepared using the SRM of mercury ion solution (C(Hg)=1.0 g/mL) and the potassium dichromate dilution solution. The formula $C_{Hg} = C_{SRM} \times V_a/V_k$ was used to prepare the stock solutions, where C_{Hg} is the concentration of mercury in the stock solution, C_{SRM} is the concentration of the mercury SRM, V_a is the volume of mercury SRM added, and V_k is the capacity of the volumetric flask.

Chocolate samples were prepared by weighing out 0.05-0.2 g of crushed bars/nibs into the analyzer sample boats. Samples of the fish tissue SRM were also prepared to ensure the instrument's accuracy. Once samples were properly prepared, they were inserted into the mercury analyzer. The direct analyzer measures mercury by decomposing the sample according to a programmable temperature mode, releasing gaseous products that are carried into a heated analytical cell. The analytical cell has an atomic absorption spectrometer measuring absorption of the 254 nm resonance radiation by the mercury atoms.[12]

Chapter 3

Lead

3.1 Overview and Toxicity

While lead is naturally occurring in Earth's crust, anthropogenic uses (mining, smelting, manufacturing, etc.) have resulted in increased human exposure and environmental contamination. Inhalation of lead particles and ingestion of lead-contaminated food and water are common routes of exposure. Once lead enters the body, it can disrupt the brain, kidneys, bones, and liver. Young children are most vulnerable to the impacts of the toxic metal as they absorb 4-5 times more ingested lead than adults, and they can suffer significant and permanent health impacts, particularly with the development of the brain and nervous system. In adults, lead poisoning can increase the risk of high blood pressure, kidney damage, and cardiovascular issues. High level of lead exposure in pregnant women can cause miscarriage, stillbirth, and premature birth.[2]

According to the Centers for Disease Control and Prevention (CDC), a blood lead level of 3.5 µg/dL or above is a cause for concern and indicative of significant lead exposure.[13] The U.S. Food and Drug Administration (FDA) set an interim reference level (IRL) for lead of 2.2 µg/day for children and 8.8 µg/day for females of childbearing age, depicting the upper limit of the amount of lead that can be consumed without potential health concerns. The IRL for lead was set to be 10x lower than the amount of lead a child or adult would have to consume to reach the CDC's blood lead level.[14] Nevertheless, lead can impair development and have harmful effects at even lower levels, especially in children. There is no known safe exposure level, so the ingestion and inhalation of lead should be avoided by all ages and genders.[13]

3.2 Previous Studies

Many studies have identified significant amounts of lead in dark chocolate. An article entitled "Perspective on Cadmium and Lead in Cocoa and Chocolate" reported lead levels ranging from 0 to 2,480±190 ppb in products from varying countries.[15] Considering the FDA's suggested serving size of 30 grams for chocolate products, the upper end of the samples tested in this article could result in 74.4 µg of lead being ingested in one serving. Furthermore, a study in India detected an average of 1,920 ppb of lead in chocolates, resulting in 57.6 µg of lead being ingested in one serving.[16] These values greatly surpass the FDA's suggested IRLs for lead, meaning regular ingestion of this amount of lead could result in serious health impacts.

3.3 ICP-OES Method 1 Results

The concentrations of lead in Castronovo Chocolate products (SNC, LCH, CV, ZDR) and Meridian Cacao Co. products (PO, NIB) were quantified using ICP-OES Method 1 (Table 3.3.1). The SRM and spiked duplicate percent recovery values were also determined (Table 3.3.2).

Table 3.3.1: ICP-OES Method 1 Results for Lead in Castronovo Chocolate and Meridian Cacao Co. Products

Sample	Pb (ppb)	Sample	Pb (ppb)
WB 1	0	CV 3	0
WB 2	0	ZDR 1	0
SNC 1	0	ZDR 2	0
SNC 2	0	ZDR 3	0
SNC 3	0	PO 1	0
LCH 1	0	PO 2	0
LCH 2	0	PO 3	0
LCH 3	0	NIB 1	0
CV 1	0	NIB 2	0
CV 2	0	NIB 3	0

Table 3.3.2: ICP-OES Method 1 Results for Lead in SRM and Spiked Samples

Sample	Pb (ppm)	Percent Recovery
SRM 1	13.1	95.1%
SRM 2	12.3	97.2%
SNC-4S	1.94	97.0%
SNC-5S	1.92	96.0%

3.4 ICP-OES Method 2 Results

The Lidl chocolate products (MA, PE, GR, DC) were tested using ICP-OES Method 2 (Table 3.4.1). The SRM and spiked samples percent recovery values were determined (Table 3.4.2). Figure 3.4 shows the average lead levels found in the products using ICP-OES Method 2, with the error bars representing one standard deviation from the triplicated data.

Table 3.4.1: ICP-OES Method 2 Results for Lead in Lidl Products

Sample	Pb (ppb)	Sample	Pb (ppb)
WB 1	0	PE 3	0
WB 2	0	GR 1	0
MA 1	0	GR 2	0
MA 2	166	GR 3	59.5
MA 3	0	DC 1	139
PE 1	0	DC 2	0
PE 2	0	DC 3	0

Table 3.4.2: ICP-OES Method 2 Results for Lead in SRM and Spiked Samples

Sample	Pb (ppm)	Percent Recovery
SRM 1	12.6	88.4%
SRM 2	13.0	86.6%
DC-4S	1.71	85.3%
DC-5S	1.73	86.4%

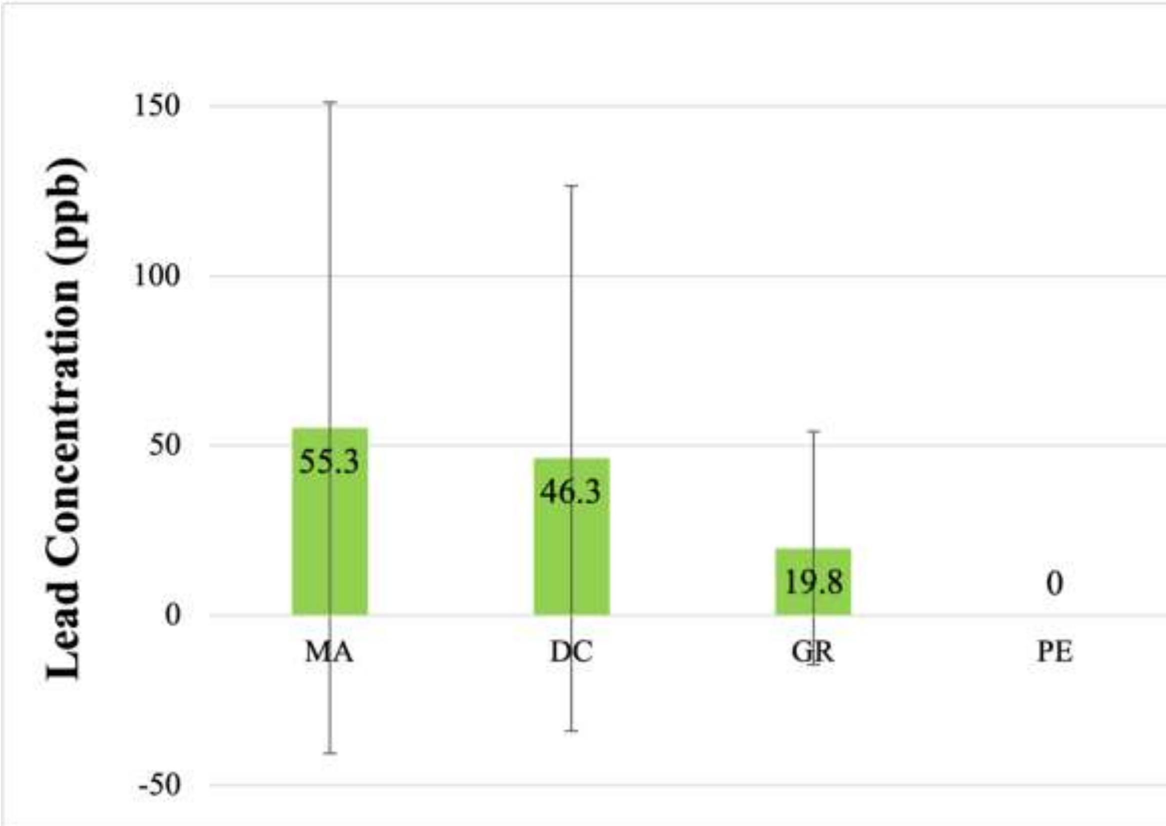

Figure 3.4: Average Lead Concentrations Found in Lidl Products

3.5 Conclusion

Overall, very little to no lead was found in the chocolates tested using ICP-OES Methods 1 and 2. ICP-OES Method 1 provided adequate recovery rates, demonstrating the technique's accuracy. However, ICP-OES Method 2 had substantially lower percent recovery values, ranging from 85.3-88.4%. Consequently, the results from ICP-OES Method 2 shown in Table 3.4.1 and Figure 3.4 are not reliable, especially as samples of the same chocolate product had no signal, resulting in large standard deviations. More so, these samples were under the instrument's estimated limit of detection (LOD) of 5 ppb before the concentrations were adjusted for dilution. Even if that data was reliable, the greatest average concentration found was 55.3 ppb, which only equates to 1.66 µg of lead in one serving. This value does not surpass the FDA's IRL for lead. Therefore, it can be concluded that the chocolate products tested in this study have very little to no lead present.

Chapter 4

Cadmium

4.1 Overview and Toxicity

Cadmium levels in soil have been increasing in recent years due to its use in various industrial processes, including the production of batteries, solar cells, plastic stabilizers, and electroplating. Cadmium typically enters the human body through inhalation, ingestion, or the skin. The toxic metal is known to cause cancer and disrupts the cardiovascular, gastrointestinal, renal, reproductive, neurological, and respiratory systems.[17] Like lead, cadmium poisoning is especially dangerous for children and can cause developmental issues.[18]

The Agency for Toxic Substances and Disease Registry (ATSDR) states that cadmium's chronic durational minimal risk level is 0.1 μg per kg of body weight per day based on renal effects.[19] In comparison, the FDA's IRL range for cadmium is 0.21-0.36 μg per kg of body weight per day based on health effects to the bones and kidneys.[18] While both levels are different, it can be concluded that the consumption of cadmium should be limited to a maximum of 0.36 μg per kg of body weight per day. For body weights of 20 kg and 70 kg, this corresponds to 7.2 µg and 25.2 µg of cadmium per day, with the lower value being estimated for a child and the larger for an adult.

4.2 Previous Studies

Previous studies have detected dangerous amounts of cadmium in chocolate products from around the world. A review article identified cadmium concentrations ranging from 0 to 1,833±20 ppb, leading to 55.0 µg of cadmium potentially being ingested in one serving.[15] Another study found an upper end of 3,150 ppb cadmium in chocolate products or 94.5 µg of cadmium in one serving.[20] Both 55.0 µg and 94.5 µg surpass the ATSDR and FDA guidelines for cadmium consumption, exemplifying the dangers chocolate products pose to human health.

4.3 ICP-OES Method 1 Results

The concentrations of cadmium in Castronovo Chocolate products (SNC, LCH, CV, ZDR) and Meridian Cacao Co. products (PO, NIB) were quantified using ICP-OES Method 1 (Table 4.3.1). The SRM percent recovery values were also determined (Table 4.3.2). Figure 4.3 illustrates the average cadmium levels found in the products, with the error bars representing one standard deviation from the triplicated data.

Table 4.3.1: ICP-OES Method 1 Results for Cadmium in Castronovo Chocolate and Meridian Cacao Co. Products

Sample	Cd (ppb)	Sample	Cd (ppb)
WB 1	0	CV 3	245
WB 2	0	ZDR 1	0
SNC 1	0	ZDR 2	0
SNC 2	0	ZDR 3	0
SNC 3	0	PO 1	351
LCH 1	768	PO 2	371
LCH 2	800	PO 3	369
LCH 3	772	NIB 1	0
CV 1	0	NIB 2	0
CV 2	229	NIB 3	0

Table 4.3.2: ICP-OES Method 1 Results for Cadmium in SRM Samples

Sample	Cd (ppb)	Percent Recovery
SRM 1	560	105%
SRM 2	520	106%

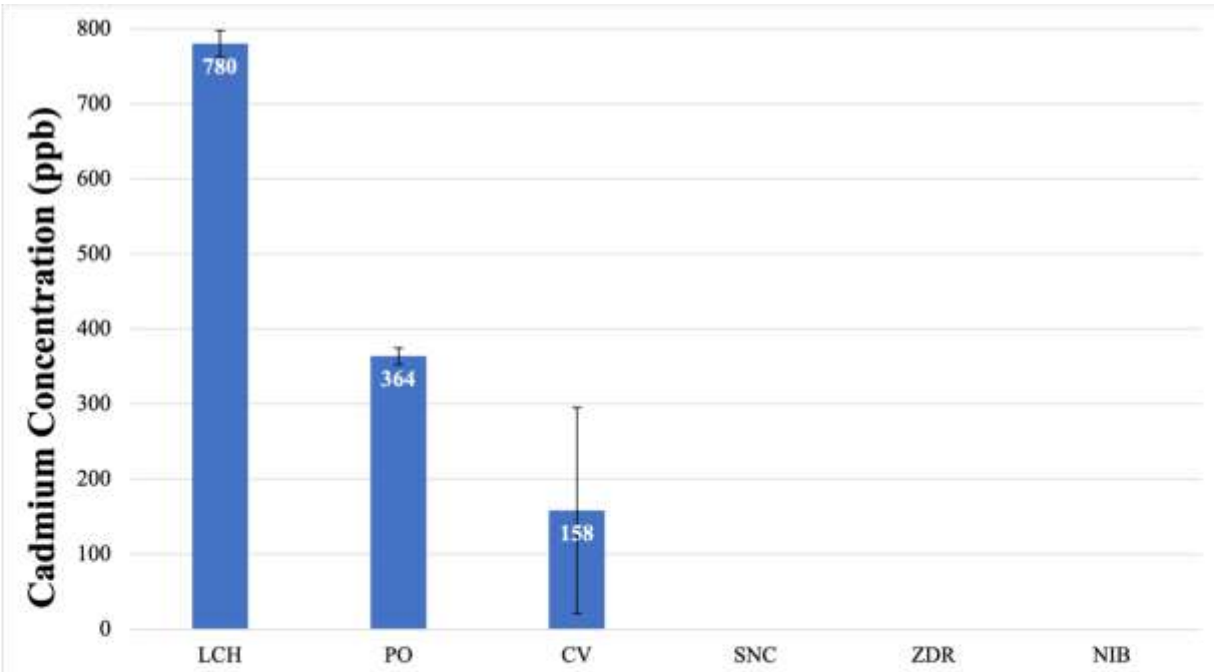

Figure 4.3: Average Cadmium Concentrations Found in Castronovo Chocolate and Meridian Cacao Co. Products

4.4 ICP-OES Method 2 Results

The Lidl chocolate products (MA, PE, GR, DC) were tested using ICP-OES Method 2 (Table 4.4.1). The SRM samples percent recovery values were determined (Table 4.4.2).

Table 4.4.1: ICP-OES Method 2 Results for Cadmium in Lidl Products

Sample	Cd (ppb)	Sample	Cd (ppb)
WB 1	0	PE 3	0
WB 2	0	GR 1	0
MA 1	0	GR 2	0
MA 2	0	GR 3	0
MA 3	0	DC 1	0
PE 1	0	DC 2	0
PE 2	0	DC 3	0

Table 4.4.2: ICP-OES Method 2 Results for Cadmium in SRM Samples

Sample	Cd (ppb)	Percent Recovery
SRM 1	39.7	73.4%
SRM 2	41.4	76.5%

4.5 Conclusion

While ICP-OES Method 2 did not detect any cadmium in the samples, some products tested using ICP-OES Method 1 had elevated levels of cadmium. Two of the CV samples contained cadmium. However, these samples were near the instrument's LOD of 5 ppb before the concentrations were adjusted for dilution, and one CV sample did not contain detectable cadmium, resulting in a high standard deviation. Nonetheless, both LCH and PO had reliable and appreciable amounts of cadmium, with LCH having an average of 780 ppb and PO with an average of 364 ppb. These concentrations correspond to 23.5 μg and 10.9 μg of cadmium in one serving of chocolate, respectively. The cadmium found in LCH is well above the recommended daily intake for the average child and is near the maximum intake for an average adult. The cadmium found in PO is also above the guideline for children but may not be significantly hazardous for adults. As the recovery values for cadmium with ICP-OES Method 1 are acceptable, it can be concluded that both the LCH and PO chocolate products contain levels of cadmium dangerous for consumption.

Chapter 5

Other Heavy Metals

5.1 Overview and Toxicity

Previous studies pertaining to the concentrations of heavy metals in chocolate have primarily focused on lead and cadmium contents, neglecting other equally toxic and harmful species. To address this issue, the Castronovo Chocolate and Meridian Cacao Co. products underwent a full-panel screening using ICP-OES Method 1, where the concentrations of 19 other toxic and non-toxic metals were determined. This study will focus on the barium, strontium, and thallium levels determined by the full-panel screening. Additionally, a direct mercury analyzer was used to screen mercury contents in the chocolate products.

Barium is naturally found in air, water, and soil, but industrial plants often contaminate the surrounding air, nearby waterways, and soil with additional barium. Ingesting large amounts of barium can impact heart rhythm and paralysis in humans. The EPA has determined that the concentration of barium in drinking water and food should not exceed 2 ppm.[3]

Strontium is another naturally occurring element, with stable strontium found in soil, coal, surface and underground water, air, plants, and animals. Stable strontium is not harmful in humans at the levels typically found in the environment. The EPA recommends that food and drinking water levels of stable strontium should not exceed 4 ppm. However, nuclear reactors and explosions of nuclear weapons can release radioactive strontium, which can damage bones and surrounding soft tissues. The inhalation or ingestion of radioactive strontium should be avoided at all costs.[4] Radioactive strontium is relatively rare in the environment and will not be considered in this study.

Thallium is a trace element in the earth's crust and is released into the environment as a by-product of smelting other metals and coal burning. Thallium does not decompose and remains in the air, soil, and water where it is often absorbed by plants where it enters the food chain. The metal can impact the nervous system, causing numbness in fingers and toes, and can affect the lungs, liver, heart, and kidneys.[5] Thallium concentrations in food should not exceed 2-4 ppm.[6]

Mercury levels in the environment vary, and the element is distributed by geological weathering, fossil fuel combustion, and manufacturing. Humans are exposed to mercury through seafood, with fish often having mercury in the ppm range. Ingesting mercury can cause psychological issues, numbness, and muscle weakness. Fetuses, infants, and children are particularly vulnerable as mercury can impact brain development.[21] The EPA and FDA state that mercury intake should be limited to 0.7 µg per kilogram of body weight per week.[22] For body weights of 20 kg and 70 kg, this corresponds to 14 µg and 49 µg of mercury per week, with the lower value being estimated for a child and the larger for an adult.

5.2 ICP-OES Method 1 Full-Panel Results

The concentrations of barium, strontium, and thallium in Castronovo Chocolate products (SNC, LCH, CV) and Meridian Cacao Co. products (PO, NIB) were quantified using ICP-OES Method 1 (Table 5.2). Figure 5.2 illustrates the average metal levels found in the products, with the error bars representing one standard deviation from duplicated (CV, PO, NIB) or triplicated (SNC, LCH) data. However, there was no SRM to estimate the completeness of extraction for these metals.

Table 5.2: ICP-OES Method 1 Results for Barium, Strontium, and Thallium in Select Castronovo Chocolate and Meridian Cacao Co. Products

Sample	Ba (ppm)	Sr (ppm)	Tl (ppm)
WB 1	0	0	0
WB 2	0	0	0
SNC 1	6.41	6.65	3.38
SNC 2	6.41	6.66	3.76
SNC 3	6.23	6.77	4.33
LCH 1	7.78	4.35	4.89
LCH 2	7.53	4.42	4.74
LCH 3	7.82	4.39	4.10
CV 1	3.05	3.14	4.46
CV 2	3.21	3.21	4.17
PO 1	2.13	3.15	4.32
PO 3	2.07	3.13	4.90
NIB 2	1.96	2.98	6.02
NIB 3	1.99	3.03	6.12

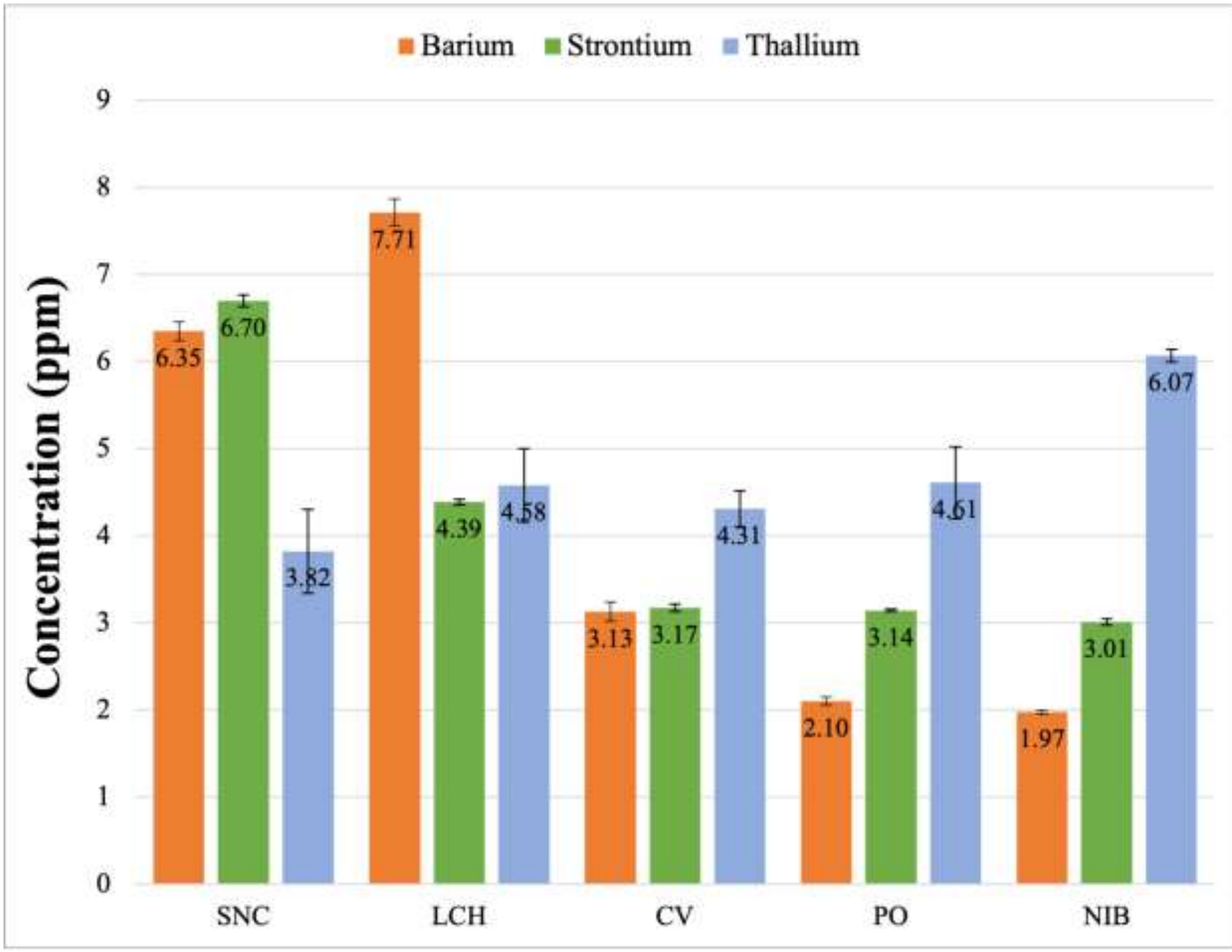

Figure 5.2: Average Concentrations of Barium, Strontium, and Thallium in Select Castronovo Chocolate and Meridian Cacao Co. Products

5.3 Mercury Analyzer Results

The concentrations of mercury in all chocolate products were determined on single replicates using a direct mercury analyzer (Table 5.3.1). The fish tissue SRM was analyzed with each sample run and percent recovery values were determined (Table 5.3.2). Figure 5.3.1 shows the mercury metal levels found in the products. Figure 5.3.2 is a scatterplot for the Lidl chocolate products displaying the correlation between percent cocoa and concentration of mercury.

Table 5.3.1: Mercury Analyzer Results for All Chocolate Products

Sample	Hg (ppb)	Sample	Hg (ppb)
SNC	0	NIB	2.06
LCH	1.53	DC	6.16
CV	0	MA	5.56
ZDR	0	PE	1.32
PO	9.66	GR	0

Table 5.3.2: Mercury Analyzer Results for Fish Tissue SRM Samples

Sample	Hg (ppb)	Percent Recovery
SRM 1	342	108%
SRM 2	301	95.1%
SRM 3	335	106%
SRM 4	314	99.3%

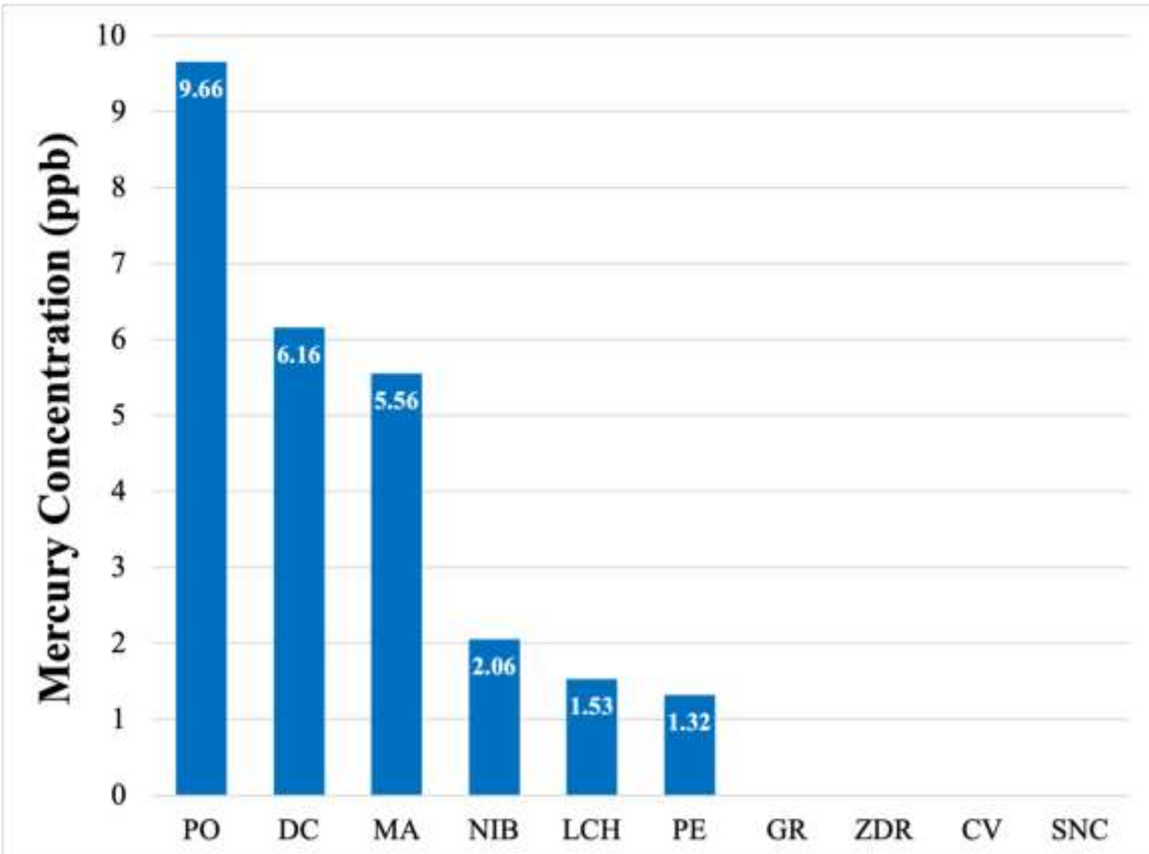

Figure 5.3.1: Concentrations of Mercury in All Chocolate Products

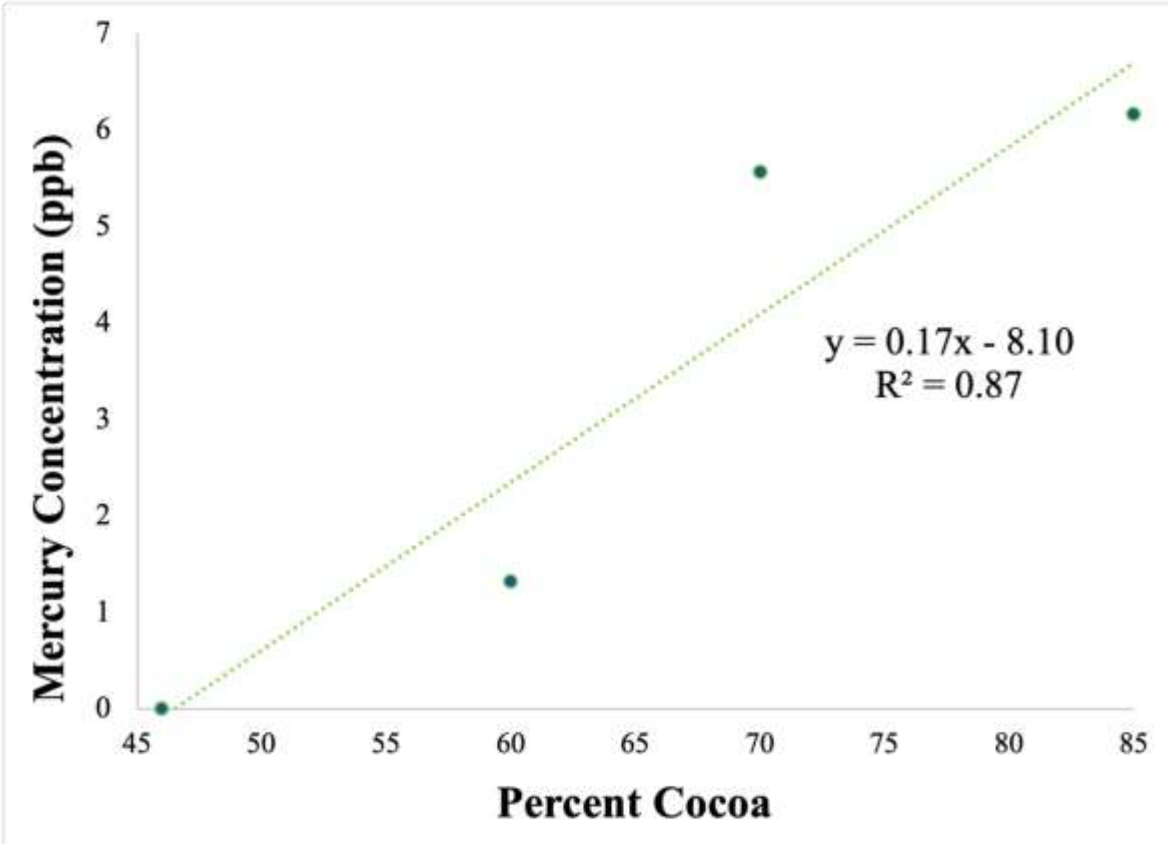

Figure 5.3.2: Scatterplot Relating Mercury Concentration and Percent Cocoa in Lidl Products

5.4 Conclusion

All the chocolate products tested in the ICP-OES Method 1 full-panel screening contained elevated levels of barium, strontium, and/or thallium. Nearly all the samples exceeded the recommended maximum concentration of barium, with NIB being the only sample with an average below 2 ppm. Furthermore, all the samples were within or above the 2-4 ppm recommended maximum for thallium. Both SNC and LCH demonstrated high levels of strontium, with SNC and LCH having averages of 6.70 ppm and 4.39 ppm, respectively. Therefore, the regular consumption of the SNC, LCH, CV, PO, and NIB chocolate products could result in negative health impacts.

Many of the chocolate products also contained mercury, with concentrations ranging from about 1-10 ppb. These concentrations are not immediate health risks, as even the greatest mercury concentration detected only results in around 0.290 µg mercury in one serving, which is well below the EPA's and FDA's recommended weekly intake. To surpass the EPA's and FDA's guidelines, a child with a body weight of 20 kg would have to eat 48 servings of this chocolate in one week, and an adult with a body weight of 70 kg would have to eat 169 servings in one week.

For the Lidl chocolate products, which varied in percent cocoa, there was a correlation between mercury concentration and percent cocoa values. This implies that the elevated levels of mercury are probably caused by the cacao beans' uptake of mercury from the soil rather than additives or manufacturing processes. However, the linear trend likewise implies that all the cacao bean varieties contain similar levels of mercury, meaning that the locations where the beans were grown had little to no impact on mercury concentration. The Castronovo Chocolate bars all contain the same percent cocoa of 72%, implying that their increased concentrations of mercury and other heavy metals are likely due to the soil where their beans are grown.

Chapter 6

Conclusion

In this study, the concentrations of various toxic metals in chocolate products were quantified using two ICP-OES instruments and a direct mercury analyzer. While previous studies have focused on lead and cadmium levels, this study expanded upon the scope of past work by screening the samples for 19 other toxic and non-toxic metals using ICP-OES Method 1. Moreover, the chocolate products were primarily purchased from small businesses which have been repeatedly overlooked in previous works.

The full-panel screening revealed high and potentially dangerous levels of barium, strontium, and thallium in many of the Castronovo Chocolate and Meridian Cacao Co. products. Elevated levels of cadmium and mercury were also found in a few products, while very little to no lead was detected in any sample. In general, the Lost City Honduras dark chocolate from Castronovo Chocolate had the highest concentrations of toxic metals, with its cadmium, barium, strontium, and thallium levels all above recommended intakes. The Sierra Nevada Colombia and Chuao Venezuela Castronovo Chocolate products likewise contained high concentrations of metals. Meridian Cacao Co.'s Organic Cocoa Powder contained elevated levels of cadmium, barium, and thallium, and the company's Organic Cacao Nibs had a high concentration of thallium. These results indicate that the soil in Honduras, Colombia, Venezuela, and Tanzania where the cacao beans of the contaminated chocolates were grown might be polluted with the toxic metals quantified in this study alongside other heavy metals. It is also possible that the manufacturing processes of the businesses could be introducing metals into the finished chocolate products. Overall, consumers should be cognizant of how much chocolate they consume to avoid significant exposure to heavy metals, especially as government agencies do not seem to be monitoring chocolate products for toxic metal concentrations.

In the future, research should be conducted to confirm the presence of barium, mercury, strontium, thallium, and other metals in chocolate products and elucidate the dangers these metals pose rather than focusing on just cadmium and lead. Additionally, it would be beneficial to run a full-panel ICP-OES screening of all the chocolate products included in this study; only five products were tested due to limited time and resources. Similarly, the Lidl products were analyzed with ICP-OES Method 2 instead of ICP-OES Method 1. As ICP-OES Method 2 does not seem to be as sensitive as ICP-OES Method 1, it is possible that cadmium and lead could have remained undetected in these samples. Future work should address this disparity by analyzing the Lidl products with the same ICP-OES instrument.

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