Trace Elements in Gluten-free Pastas and Flours from Markets Located in the Las Vegas, Nevada Area

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Abstract

The popularity of gluten-free foods has been increasing across the United States and abroad. A significant reason for this trend involves marketing efforts targeted towards individuals seeking to avoid the consequences of celiac disease or a perceived gluten intolerance. Many gluten-free food products originate in regions of the world where irrigation with metal-contaminated waters is common. Calcium, Fe, Mg, Ti and Zn were detected at various levels across all food products. Cadmium was detected in 96.8% of U.S. and 54.5% of Asian gluten-free foods with gluten containing foods above reported averages (216 μg kg⁻¹ Cd); as was Co (140μg kg⁻¹) in 48.4 % of U.S., 72.7% of Asian gluten-free foods, and 40% of the gluten containing foods; Cr was in 54.8% of the U.S., 72.5% of Asian gluten-free foods, and 100% of gluten containing food products; while Ca, Fe, Mg, Ti and Zn were greater than 10,000 μg kg⁻¹ with Ba, Cd, Co, Mo, and Ni above reported averages. Finally, trace metals were more commonly detected in the gluten containing foods overall. It was found that trace elements were more commonly found in the gluten containing products; however, none of the higher than expected levels pose a significant health risk to consumers.

Keywords

gluten-free foods; nutrients; metals; celiac; pasta; flour; Las Vegas

1. Introduction

Gluten is a protein found in breads, pastas, and products of wheat, rye, barley, and triticale. According to literature, the proteins glutenin and gliadin are the main products of gluten
with gliadin responsible for most of the negative health effects (Basha et al., 2014). Gluten helps food maintain its shape while also binding it together (Basha et al., 2014). For person with a gluten allergy, gluten is known to cause small intestine irritation, among other symptoms, in sufferers of Celiac disease, a genetic based immune disorder (Basha et al., 2014). According to other researchers, celiac disease is characterized by an immune response of the T-lymphocytes in the small intestine to gluten peptide bonds (i.e. Basha et al., 2014, Howdle, 2003, Miedico et al., 2017, Orecchio et al., 2014, Thompson, 1999). While medically debated, many people claim to be gluten sensitive and report allergic response to its protein.

Research suggests that celiac disease occurs in approximately 1% of the general population (Niewinski, 2008, Thompson, 1999, 2000, Thompson et al., 2005). In addition to sufferers of celiac disease, many people avoid gluten because of a wheat allergy or they believe they have a gluten intolerance, despite there being limited medical evidence that such a medical disorder exists (Elli et al., 2015, Catassi et al., 2013). Current studies indicate that celiac disease is increasing across all age groups, especially in the senior population (Curiel et al., 2014). The only currently available treatment to this disease is a strict gluten-free diet, which can lead to the restoration of the atrophied intestinal villi (Curiel et al., 2014, Niewski, 2008).

The consumption and/or baking with gluten-free products have become much more prevalent due to product availability (Orecchio et al., 2014). Gluten-free consumers can purchase premade gluten-free products such as breads, pastas, flour, and desserts. Today, many foods contain gluten-free starch and flour made of corn, potato, tapioca, or rice. Gluten-free baking mixes and flours are becoming accessible to consumers so that they can bake their own breads, pastas, cake, and other foods, which in turn alleviates the need for them to consume a diet consisting of mostly raw and green foods (Orecchio et al., 2014).

Trace metals have a vital role in biology and are essential micro-nutrients that play an active role in biochemical functions of living organisms (Miedico et al., 2017, Orecchio et al., 2014). While some metals like Al, Cr, and Fe are essential to biological functions at low levels, they can be toxic at higher levels. Other metals (i.e. Cd, Pb, Se, V) do not occur naturally in the body, and therefore, their presence is usually the result of ingestion from foods. Exposure to such metals can be harmful to human health, especially in children and the elderly who tend to be more sensitive to these toxins (Miedico et al., 2017).

Gluten-free food products may contain higher and lower levels of trace metals depending on the product or where they are grown in the world (Orecchio et al., 2014, Thompson et al., 2005). Studies have identified that Cd-contaminated wastewater from industrial sites have been used in Asia’s agricultural fields (Takagi et al., 2004, Vahter et al., 2007, Yamagami et al., 2006). Consuming foods containing Cd over a lifetime can result in a mild to severe mitochondrial dysfunction called Itai–itai disease (Vahter et al., 2007, Yamagami et al., 2006, Inaba et al., 2006). Extended intake of certain pollutants (i.e. I, Pb, As, Cd, Hg) can lead to other disorders in the gastrointestinal tract. The World Health Organization has determined acceptable levels of trace metals in food in cooperation with other international

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There are several gluten free cereal products that are primary ingredients for these foods such as rice (Oryza sativa), maize (Zea mays), sorghum (Sorghum bicolor), and buckwheat (Fagopyrum esculentum Moench) according to Orecchio et al. (2014). While there is nutritional value in these foods, they are only part of an overall balanced diet. Buckwheat, for instance, is known as a source of protein, amino acid, vitamins, dietary fiber, minerals, and essential trace elements (Orecchio et al., 2014, Howdle, 2003). Buckwheat; however, has lower concentrations of essential metals (Zn, Fe) when compared with wheat (Orecchio et al., 2014, Vahter et al., 2007). Moreover, the grinding processes to turn such products into flour can also alter the nutrient composition of the food product (Thompson, 1999).

Trace metal concentration in mainstream food products has been well studied however, little information exists for gluten-free foods (e.g. Aziz et al., 2015, Orecchio et al., 2014, Ertl and Coessler, 2018, Parra-Orobio et al., 2017). This research measured trace metals, both toxic and non-toxic, in gluten-free foods (n =42), and the daily intake concentrations. A limited number (n=5) of gluten containing foods were analyzed for comparison. The overall study objectives were to evaluate the levels of trace metals (Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Ti, Zn) found in selected gluten-free food products (i.e. pastas, flours) purchased in the Las Vegas, Nevada (U.S.) market. This study will: 1) identify crustal (Al, Ca, Fe, Mg, Zn) and toxic trace metals (Ba, Cd, Co, Cr, Cu, Mn, Mo, Ni, Ti) in gluten-free food products; 2) determine the levels of trace metals in specific gluten-free product types; and 3) if possible, evaluate the level of exposure (i.e. chronic exposure) to consumers of these foods in the literature and discuss possible effects due to low-level chronic-long term exposure.

1.1 Metals in the Food Chain

It is known that toxic elements in foods can be from both natural and anthropogenic source (Orecchio et al., 2014, Baker et al., 2018). For instance, Orecchio et al. (2014) determined that metal contamination of food products is related to the soils it grows in and rarely the manufacturing (milling) process. Other researchers have examined the uptake of metals (e.g. Cu, Ni, Fe, Co, Zn, U, Pb) in vegetation, animal tissues, and the transfer of said metals via the food chain (Miedico et al., 2017, Kipp et al., 2009, Clemente et al., 2005). High levels of Pb tend to be present in broad leaf plants (Elmleaf, Blackberry and Rubus ulmifolius) grown in contaminated soils as documented by Clemente et al. (2005). The bioaccumulation of As, and resulting phytotoxicity, was observed in areas that had several metal-tolerant plant species (e.g., Angophora floribunda, Cassinia laevis, and Chrysocephalum apiculatum) that colonized the periphery of contaminated sites or were irrigated with contaminated waters (Clemente et al., 2005, Han et al., 2006).

It is known that high levels of metals are possible in grasses, melons, coconuts, fruits, and even in rice, corn, and wheat where effluent from factories or mining is utilized for irrigation (Nicholson et al., 2003, Vazquez et al., 2008, Wang et al., 2007). Crops grown in the mining region of the Sierra Madrona Mountains of southern Spain showed higher concentrations of trace metals (Pb, Zn, Cd, Cu, As and Se) (Orecchio et al., 2014). It has been reported that...
higher concentrations of trace metals in vegetation were more prevalent in crops like rice and wheat where irrigation originated from polluted waters (Reglero et al., 2008).

It is known that contaminated crops affect the food chain and ultimately human health (Inaba et al., 2006, Yamagami et al., 2006). Cadmium contaminated wastewater from industrial sites was used for growing rice in Japan and ultimately impacted local health (Takagi et al., 2004, Vahter et al., 2007). Studies found that residents in the area who consumed 3.1 g to 3.8 g of Cd in rice over a lifetime developed mild to severe mitochondrial dysfunction called Itai–Itai disease (Inaba et al., 2006). The source of this disease was found to be the direct result of Cd-enriched wastewater discharged to local rivers, and subsequently used in the rice fields. The bioaccumulation of Cd in the tissue of local residents affected their tubular epithelial cells after 80-weeks of exposure by ingestion of the contaminated rice (Nation Academy of Science [NAS], 1973).

2. Materials and Methods

Forty-two (42) samples of gluten-free foods (pastas, flours) and five (5) gluten containing foods (pastas) were purchased in Las Vegas markets in June 2018. Products (“samples”) were representative of what is available in the Las Vegas Valley with 31 samples manufactured in the U.S. and eleven (11) imported from Asia. The five (5) gluten containing samples were manufactured in the United States (U.S.). While manufacturing location was known, the agricultural source of the plant material was not. Each sample was homogenized with an acid washed mortar and pestle, immediately sub-sampled and digested with hot aqua-regia acid to reduce degradation. All gluten-free samples were prepared in triplicate, digested, and analyzed per USEPA methods as described below. Data was evaluated for standard deviation (σ), mean (x), minimum (min), and maximum (max) values seeking patterns within each group of products (i.e. gluten free vs. gluten containing) and across groups.

2.1 Sample Preparation and Analysis

Forty-two gluten-free and 5 gluten containing samples (n=47) were purchased from local markets and analyzed for trace metals (Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Ti, Zn). Approximately 1.0g (±0.001g) of homogenized food material (gluten-free [e.g. rice, corn] and gluten containing [i.e. wheat]) was processed according to USEPA Solid Waste 846 (SW-846) protocols, including quality control (USEPA, 1997). Fourteen trace elements (Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Ti, Zn) were extracted using USEPA method 3050B (hot aqua-regia acid digestion) followed by USEPA method 6010B for inductively coupled plasma-optical emission spectrometer (USEPA, 1997). Instrument calibration consisted of 6 points across a concentration range (including a blank) and fitted by linear regression with >0.995 R² (USEPA, 1997). Instrument integrity was verified with a certified reference sample purchased from a USEPA certified supplier with the sample falling within certified windows in order to meet requirements. Samples were analyzed in triplicate with a required relative standard deviation (RSD) of less than 20% to qualify as acceptable (USEPA, 1997). Method detection limits (MDL) are defined as 3 time the standard deviation (σ), where the level of the analyte approaches zero (0). Limit of
quantitation (LOQ) is defined as 10 times \( \sigma \) with an uncertainty of \( \sim 30\% \) at the 95% confidence level.

### 3. Results and Discussion

Trace metal concentrations (Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Ti, Zn) for 42 gluten-free and 5 gluten containing food samples (n=47) are presented in Table 1 and reported in \( \mu g \, kg^{-1} \), dry weight. Table 2 shows statistics for all of the data in three separate formats; overall data, U.S. gluten free, Asian gluten free, and gluten containing foods of pasta and flour. Samples were processed in triplicate utilizing USEPA quality control measures for accuracy and averaged for reporting purposes. Aluminum was detected at 400 to 6750 \( \mu g \, kg^{-1} \) (mean (\( \bar{x} \)) of 1754) across all products with a U.S. market gluten-free foods \( \bar{x} \) of 1708 \( \mu g \, kg^{-1} \). Asian gluten-free foods were 550 to 3000 \( \mu g \, kg^{-1} \) (\( \bar{x} = 1736 \mu g \, kg^{-1} \)) while gluten containing foods ranged between 1450 and 3350 \( \mu g \, kg^{-1} \) (\( \bar{x} = 2080 \mu g \, kg^{-1} \)).

U.S. market foods were highest in samples 7 (6750 \( \mu g \, kg^{-1} \) [pasta]) and 23 (5900 \( \mu g \, kg^{-1} \) [flour]). Asian foods were highest in samples 45 and 47 measuring 2150 and 3350 \( \mu g \, kg^{-1} \) Al, respectively, with 850 to \( 3.8 \times 10^4 \) \( \mu g \, kg^{-1} \) reported by other authors (Loska and Wiechuya, 2003, Yokel et al., 2005).

With Al at a level of 6750 \( \mu g \, kg^{-1} \), it is important to understand that other studies have established that oral exposure to Al is not considered harmful as it is poorly absorbed and is excreted through feces and urine (Finberg et al., 1986). It has been reported; however, that ingestion of high amounts of Al can lead to nausea, mouth and skin ulcers, skin rashes, vomiting, diarrhea, and arthritic pain (Jaishankar et al., 2014). These symptoms are generally mild and short lived although people with renal disease, especially children, are at risk of developing Al toxicity (Finberg et al., 1986, Jaishankar et al., 2014). Finally, the Food and Drug Administration (FDA) has determined that aluminum used as food additives, are generally safe; however, they have set an exposure limit for Al at 0.2 mg L\(^{-1} \) in bottle water according to ATSDR (2008).

Barium was detected at 350 to \( 1.2 \times 10^4 \) \( \mu g \, kg^{-1} \) across all samples (\( \bar{x} = 2606 \mu g \, kg^{-1} \)). The U.S. gluten-free samples ranged between 500 and \( 1.2 \times 10^4 \) \( \mu g \, kg^{-1} \) (\( \bar{x} = 2494 \mu g \, kg^{-1} \)) while the Asian samples ranged from 350 to 4050 \( \mu g \, kg^{-1} \) (\( \bar{x} = 1477 \mu g \, kg^{-1} \)), and gluten containing foods from 3700 to 9700 \( \mu g \, kg^{-1} \) (\( \bar{x} = 5790 \mu g \, kg^{-1} \)). Literature has shown cereal products (both gluten and gluten-free) tend to encompass higher Ba levels such as bran flakes at 3900 \( \mu g \, kg^{-1} \) (Mertz, 1986). Current Ba data falls within stated ranges although 11.9% of samples exceeded Mertz (1986) average in samples 14 (7100 \( \mu g \, kg^{-1} \)), 19 (6800 \( \mu g \, kg^{-1} \)), 23 (12100 \( \mu g \, kg^{-1} \)), 28 (9000 \( \mu g \, kg^{-1} \)), and 37 (4050 \( \mu g \, kg^{-1} \)). Data also falls within stated ranges reported by Millour et al. (2012).

Although our findings indicate that the highest level of Ba was detected at \( 1.2 \times 10^4 \) \( \mu g \, kg^{-1} \), according to the ATSDR (2008), Ba toxicity depends on its solubility in water and bodily fluids. Insoluble barium salts are nontoxic, whereas the barium ion (Ba\(^{2+} \)) and compounds that can dissolve in water or can be diluted in hydrochloric acid, such as chloride, nitrate, carbonate, and hydroxide, are toxic. The predominant effect of barium toxicity is...
hypokalemia that can result in ventricular tachycardia, hypertension and/or hypotension, muscle weakness, and paralysis (ATSDR, 2008).

Calcium ranged from $1.4 \times 10^4$ to $2.09 \times 10^6 \mu g \ kg^{-1}$ with an overall $\bar{x}$ of $3.55 \times 10^5 \mu g \ kg^{-1}$. Gluten-free products from the U.S. markets ranged between $1.4 \times 10^4$ and $2.09 \times 10^6 \mu g \ kg^{-1}$ ($\bar{x} = 4.24 \times 10^5 \mu g \ kg^{-1}$) while the Asian samples were between $6.5 \times 10^4$ and $3.36 \times 10^5 \mu g \ kg^{-1}$ ($\bar{x} = 3.9 \times 10^5 \mu g \ kg^{-1}$). It has been noted that Ca in gluten-free foods ranged between 5000 and $1.68 \times 10^6 \mu g \ kg^{-1}$ and was higher for gluten containing foods (Orecchio et al., 2015). Prolonged exposure to Ca is a concern as calcification in the kidneys (nephrocalcinosis) can lead to the formation of nephrolithiasis; i.e. kidney stones (Patel and Goldfarb, 2010, Ross et al., 2011). The current study falls above Orecchio et al. (2015) established ranges for the U.S. market gluten-free foods and therefore, may pose an issue with chronic exposure.

Calcium is vital for normal body functions and studies have established that it is difficult for a normal, healthy adult to consume excess Ca by diet alone (Patel and Goldfarb, 2010, Ross et al., 2011). If Ca toxicity (e.g. hypercalcemia) is an issue however, signs may include anorexia, weight loss, polyuria, heart arrhythmias, fatigue, and soft tissue calcification (Patel and Goldfarb, 2010). Other authors have found that tissues most at risk of calcification include vascular tissues, lungs, and kidneys. Additionally, calcification of the kidneys (nephrocalcinosis) may lead to the formation of nephrolithiasis (i.e. kidney stones) according to Patel and Goldfarb (2010) and Ross et al. (2011). Studies have established that children under the age of 4 have an Upper Intake Level (UIL) of Ca that varies by life stages. For instance, infants 0–6 months in age have an UIL of 1000mg/day while the UIL in adults >51 years is 2000mg/day (Patel and Goldfarb, 2010, Ross et al., 2011). This study shows that the gluten free and gluten containing have levels far below the mg/day intake that would produce damage.

Cadmium ranged from 150 to $600 \mu g \ kg^{-1}$ with a $\bar{x}$ of $278 \mu g \ kg^{-1}$ across all products. The U.S. gluten free foods ranged between 150 and $600 \mu g \ kg^{-1}$ ($\bar{x} = 270 \mu g \ kg^{-1}$), Asian products between 250 to $500 \mu g \ kg^{-1}$ ($\bar{x} = 333 \mu g \ kg^{-1}$), and gluten containing products between 200 and $350 \mu g \ kg^{-1}$ ($\bar{x} = 260 \mu g \ kg^{-1}$). It is worth noting that 45% of the products from the Asian markets contained no detectable Cd (samples 33, 34, 36, 39, 40). Other studies have established a range for Cd in plant materials including those used for food stocks to be 2 to $216 \mu g \ kg^{-1}$ (EC, 2006, Skrbic et al., 2013). Many of the samples in this study were above this range. With almost 55% of U.S. market foods, 36% of Asian, and 80% for gluten containing foods above the established range. Moreover, six samples (samples 5, 21, 26, 27, 32, 41) were $1\sigma$ above the $\bar{x}$ ($379 \mu g \ kg^{-1}$) with gluten containing foods falling below.

Data show that gluten free and gluten containing foods have levels far below the mg/day for Cd intake that would produce harm. However, it is important to state that other studies indicate that soluble Cd compounds of $SO_4^{2-}$ and $Cl^-$ salt forms have been shown to cause both acute and chronic intoxication (Jaishankar et al., 2014, Tchounwou et al., 2012). Moreover, acute cadmium ingestion can cause gastrointestinal irritation leading to GI tract
Cobalt (Co) levels ranged from 150 to 400 μg kg\(^{-1}\) (\(\bar{x} = 252 \mu g \text{ kg}^{-1}\)) for all products including the U.S. market, Asian, and gluten containing foods. The U.S. market ranged from 150 to 400 μg kg\(^{-1}\) (\(\bar{x} = 247\)), Asian ranged from 200 to 450 μg kg\(^{-1}\) (\(\bar{x} = 275 \mu g \text{ kg}^{-1}\)), and gluten containing products at 200 μg kg\(^{-1}\) for the minimum, maximum and \(\bar{x}\). Within the gluten-free foods (Asian / U.S. market), 4 samples were 1\(\sigma\) (i.e. 333 μg kg\(^{-1}\) Co) above the \(\bar{x}\); (samples 14, 36, 41 [pastas], 23 [flour]). The highest Co was found in Asian gluten-free foods containing 200 to 450 μg kg\(^{-1}\) while the U.S. gluten-free foods contain between 200 and 450 μg kg\(^{-1}\). Most samples (53.2%) were within 4 to 140 μg kg\(^{-1}\) established by other researchers (Podio et al., 2013).

Cobalt is an essential micronutrient consumed in the form of vitamin B\(_{12}\). While this study indicates that Cr is at levels low enough as to not pose an acute effect unless a significant amount is consumed (Leyssens et al., 2017). Cobalt toxicity can occur acutely in large doses or cumulatively with long-term low level exposure according to Leyssens et al. (2017) and Simonsen et al. (2012). Water-soluble cobalt salts are rapidly absorbed from the small intestine. Co causes erythropoiesis by increasing erythropoietin production. It is also known for increasing the oxygen-carrying capacity of the blood and therefore enhancing physical endurance (Leyssens et al., 2017, Simonsen et al., 2012). Finally, it is low-level chronic exposure that can pose a toxicity issue to the consumer (Simonsen et al., 2012).

Chromium levels ranged from 100 to 1950 μg kg\(^{-1}\) (\(\bar{x} = 575 \mu g \text{ kg}^{-1}\)) while the U.S. gluten free ranging between 100 and 550 μg kg\(^{-1}\); Asian foods ranged from 150 to 1950 μg kg\(^{-1}\) while gluten containing products varied from 1650 to 1800 μg kg\(^{-1}\). For Cr, no U.S. gluten-free were above the overall \(\bar{x}\) of 575 μg kg\(^{-1}\); however, both the Asian and the gluten containing foods were above this level. Of the Asian and U.S. gluten-free foods, only 1 sample of a flour product (sample 39) was 1\(\sigma\) above the overall \(\bar{x}\) (i.e. 575 μg kg\(^{-1}\)). Current results exceed the literature established value by Tchounwou et al. (2012). It is important to point out that the highest Cr (1650 – 1800 μg kg\(^{-1}\)) detected was in gluten containing products; twice that established in the literature.

The health hazards associated with Cr depend greatly on its oxidation state. This study considered Cr\(^{3+}\) and not the more harmful Cr\(^{6+}\) due to its reduction to Cr\(^{3+}\) in a relatively short time and therefore, these foods would not contain the more harmful form of Cr\(^{6+}\) (Jaishankar et al., 2014). The most common forms are trivalent (Cr\(^{3+}\)), a nutrient, and hexavalent (Cr\(^{6+}\)), a known toxin (Jaishankar et al., 2014). Trivalent (III) Cr is readily oxidized to hexavalent (VI) due to the excess oxygen in the environment. Hexavalent (VI) Cr is highly water soluble, making it extremely toxic compared to the trivalent (III) form (Jaishankar et al., 2014, Tchounwou et al., 2012). Trivalent Cr is the typical form found in foods and usually not an issue. However, the uptake of Cr\(^{6+}\) by plants is known to happen;

Jaishankar et al., 2014. It can also cause hepatic, renal, pulmonary damage, and coma. Chronic low-level exposure can cause cadmium to be deposited in the kidneys, leading to renal disease (Tchounwou et al., 2012). Data shows that the consumption of Cd through gluten-free foods is unlikely according to Inaba et al. (2006) as it would require 3,100 to 3,800 μg of Cd over a lifetime to cause mitochondrial dysfunction disease.
however, it is usually reduced by 96.3% to Cr³⁺ within 96 hours according to Shugaba et al. (2012).

Copper in gluten-free foods in U.S. products ranged between 600 and 2.25 × 10⁴ μg kg⁻¹, Asian products ranged from 250 to 3800 μg kg⁻¹, and gluten containing foods ranged from 3100 to 6100 μg kg⁻¹ with an overall x̄ of 4333 μg kg⁻¹. Four samples (9, 11, 24, 28) were 1σ above the overall x̄ (i.e. 4333 μg kg⁻¹ Cu). However, the data is within established ranges (1800 to 1 × 10⁴ μg kg⁻¹) according to Ertl and Goessler (2018) and the WHO (2004) maximum per day intake of 1 × 10⁴ μg kg⁻¹ for ages 19 to 70 years. Copper is an essential nutrient as it serves as a co-factor for several oxidative stress-related enzymes (Kahn and Line, 2010a, 2010b). It is also involved in the formation of hemoglobin, carbohydrate metabolism, catecholamine biosynthesis, and the formation of cross-links in collagen, elastin, and keratin in the hair (Tchounwou et al., 2012). The maximum exposure to Cu per day is recommended at 0.5 mg kg⁻¹ according to the ATSDR (2008). Excess exposure to Cu leads to bioaccumulation in the liver, resulting in chronic hepatitis and cirrhosis according to Kahn and Line (2010a) and Tchounwou et al. (2012).

Iron concentrations in gluten-free products from the U.S. markets ranged from 100 to 6.32 × 10⁴ μg kg⁻¹; Asian foods contained the lowest Fe (550 to 2.04 × 10⁴ μg kg⁻¹); gluten containing foods ranged from 3.08 × 10⁴ to 5.02 × 10⁴ μg kg⁻¹. Data is within the established limits (1.3 × 10⁴ to 1.03 × 10⁵ μg kg⁻¹) for wheat and gluten-free food products as described by Ertl and Goessler (2018). Iron is an important nutrient as it is a vital cofactor for many proteins and enzymes; deficiency can lead to anemia and decreased oxygen delivery to tissues enzymes (Jaishankar et al., 2014, Kahn and Line, 2010b). Studies show that 20 mg kg⁻¹ per day can lead to nausea, vomiting, and stomach pain, especially on low protein diets (Jaishankar et al., 2014).

Magnesium is another important part of a daily diet and vital for healthy functions. Mg however, was detected at 1.32 × 10⁴ to 1.96 × 10⁶ μg kg⁻¹ with an overall x̄ of 6.12 × 10⁵ μg kg⁻¹. U.S. gluten-free foods were within the overall range, Asian products ranged between 2.54 × 10⁴ and 1.39 × 10⁶ μg kg⁻¹ while gluten containing foods were between 2.44 × 10⁵ and 1.16 × 10⁶ μg kg⁻¹ (x̄=5.49 × 10⁵ μg kg⁻¹). Gluten-free products were 1σ above the overall x̄ (i.e. 1.147 × 10⁶ μg kg⁻¹ Fe) in 14% of samples (9, 11, 18, 24, 28, 34). Findings are comparable with other studies (350 to 1.4 × 10⁴ μg kg⁻¹) as provided by Podio et al. (2013). The risk of magnesium toxicity is increased in individuals with decreased renal function or with renal failure as the kidney is responsible for removing excess magnesium from the body (Higdon et al., 2014). The Food and Nutrition Board (FNB) Institute of Medicine (1997), and Ertl and Goessler (2004) state that consuming 3 × 10⁵ and 4.2 × 10⁵ per day is healthy and therefore, findings are within established norms.

Manganese detected in U.S. foods ranged from 400 to 8.95 × 10⁴ μg kg⁻¹ with an overall x̄ of 2.78 × 10⁴ μg kg⁻¹. Asian foods ranged from 400 to 2.09 × 10⁴ μg kg⁻¹ while gluten samples ranged from 1.5 × 10⁴ to 4.83 × 10⁴ μg kg⁻¹. Samples contained Mn at 1σ above the overall x̄ in 14% of the gluten-free samples (9, 11, 18, 24, 28, 34, and 45). Manganese is an essential nutrient, but can have toxic consequences if too much is ingested. The primary oral route for intoxication is through contaminated water as its bioavailability is higher in
water (Higdon et al., 2014). The risk of manganese neurotoxicity is set at 2mg/day for children 1–3 years; 3mg/day for children 4–8 years; 6mg/day for children 9–13 years; 9mg/day for adolescents 14–18 years, and 11mg/day for adults >19 years (O’Neal and Zang, 2015). These findings are analogous with other published works (350 to $1.4 \times 10^4 \mu g kg^{-1}$) for Mn in gluten-free and gluten containing foods (Podio et al., 2013). If a person has a Fe deficient diet, there is an increased risk of Mn toxicity as Fe deficiency enhances the gastrointestinal absorption of Mn, leading to possible cognitive deficits (Higdon et al., 2014, FNB, 1997).

Molybdenum was detected in 78.7% of foods with a range of 250 and 6100 μg kg$^{-1}$ with an overall \( \bar{x} \) of 1294 μg kg$^{-1}$. Gluten-free products (U.S. markets) ranged between 250 and 6100 μg kg$^{-1}$, Asian products ranged from 350 to 2100 μg kg$^{-1}$, and gluten containing foods ranged between 700 and 1050 μg kg$^{-1}$. Data show that 9.5% of samples were 1σ above the overall \( \bar{x} \) (2580 μg kg$^{-1}$) for pasta gluten-free foods (samples 2, 5, 6, 22). Molybdenum toxicity is relatively rare in humans as it functions as a co-factor in four Mo-dependent enzymes necessary for health (Higdon et al., 2014, O’Neal and Zang, 2015). Other studies have set a daily intake of 300 μg /day for children 1–3 years; 600 μg /day for children 4–8 years; 1.1mg/day for children 9–13 years; 1.7mg/day for adolescents 14–18 years, and 2.0mg/day for adults >19 years (O’Neal and Zang, 2015, Suchowilska et al., 2012).

Although the Institute of Medicine found little evidence of adverse health effects related to excess Mo consumption in healthy people, they have set a daily UIL at 300 μg /day for children 1–3 years, 600 μg /day for children 4–8 years, 1.1 mg/day for children 9–13 years, 1.7 mg/day for adolescents 14–18 years, and 2.0 mg/day for adults >19 years (Higdon et al., 2014). Levels are within limits set in other studies (420 to 580 μg kg$^{-1}$ Mo) for gluten-free and gluten containing foods.

Nickel was detected at 150 to 5000 μg kg$^{-1}$ with an overall \( \bar{x} \) of 1757 μg kg$^{-1}$. Gluten-free products from the U.S. ranged between 150 and 4350 μg kg$^{-1}$, Asian foods ranged from 300 4350 μg kg$^{-1}$, and gluten containing foods ranged between 4950 and 5350 μg kg$^{-1}$. In 14.3% of samples, Ni was 1σ above the overall \( \bar{x} \) (1757 μg kg$^{-1}$) across a mix of pasta and flour food products (samples 5, 9, 14, 23, 41, 42). Additionally, four gluten containing pastas (samples 43–45, 47) were 1σ above the overall \( \bar{x} \). In the literature, Ni detected in foods is normally lower than 1000 μg kg$^{-1}$ as described by Ekholm et al. (2007); placing these finding above acceptable ranges in 33% of gluten-free foods and 60% of gluten containing foods. The risk of Ni toxicity is low due to its limited intestinal absorption according to ATSDR (2008).

Titanium ranged from 7000 to $3.1 \times 10^5 \mu g kg^{-1}$ with an overall \( \bar{x} \) of $5.53 \times 10^4 \mu g kg^{-1}$. Gluten-free products (U.S. market) ranged from 7000 and $2.17 \times 10^5 \mu g kg^{-1}$, Asian products ranged between 8350 and $3.1 \times 10^5 \mu g kg^{-1}$, and gluten containing foods ranged between $1.03 \times 10^4$ and $3.62 \times 10^4 \mu g kg^{-1}$. Compared with other elements, Ti was found to be high and more in line with crustal metals (i.e. Ca, Mg) with 14.3% of the samples (7, 14, 23, 26, 36, 41) 1σ above the overall \( \bar{x} \) for gluten-free pastas and flours. Other authors have determined that the typical exposure to food-grade TiO$_2$ in an average adult to be on the order of 1000 μg kg$^{-1}$ per day (Weir et al., 2012); placing this data above the established
norms in 100% of samples. It is important to note that Weir et al. (2012) stated there is an important difference between total Ti (\(\gamma\)Ti), TiCl\(_4\) and food-grade TiO\(_2\). Other researchers have shown that TiCl\(_4\) is the harmful form of Ti; however, the forms of Ti found in foods are TiO\(_2\) MPs (micro-particles) and TiO\(_2\) NPs (nanoparticles) thus, no limit is specified in the literature at this time (ATSDR, 2008).

Zinc ranged from 5350 to 8.0 \(\times\) 10\(^4\) μg kg\(^{-1}\) with an overall \(\bar{x}\) of 3.9 \(\times\) 10\(^4\) μg kg\(^{-1}\) in analyzed foods. Gluten-free (U.S. market) ranged between 5750 and 8.0 \(\times\) 10\(^4\) μg kg\(^{-1}\); Asian products ranged between 5350 and 3.7 \(\times\) 10\(^4\) μg kg\(^{-1}\); while gluten containing foods ranged from 2.8 \(\times\) 10\(^4\) and 6.8 \(\times\) 10\(^4\) μg kg\(^{-1}\). Zinc was also in line with crustal metals (i.e. Ca, Mg) with 23.8% (samples 5, 6, 9, 11, 14, 15, 22–24, 28) \(1\sigma\) above the overall \(\bar{x}\) (6.26 \(\times\) 10\(^4\) μg kg\(^{-1}\)) with sample 45 (gluten containing pasta) significantly greater than \(1\sigma\) of the overall \(\bar{x}\). Other studies have determined that the average Zn in foods range between 9400 and 5 \(\times\) 10\(^4\) μg kg\(^{-1}\) (Ertl and Coessler, 2018). It is known that Zn is an important trace element to the human diet and is therefore required as part of our food chain (Bermudez et al., 2011). The levels of Zn that can produce an adverse health effect are much higher than the Recommended Dietary Allowances (RDAs) of 11 mg/day for men and 8 mg/day for women (ATSDR, 2008). Likewise, data places Zn in the gluten-free foods above the established range in 35.7% in the gluten-free samples and 20% of gluten containing foods. Therefore, the levels of Zn that can produce an adverse health effect are much higher than the Recommended Dietary Allowances (RDAs) of 11 mg/day for men and 8 mg/day for women which this data is below (ATSDR, 2008).

4. Conclusions

This study produced similar results to those of Orecchio et al. (2014, 2015) where they evaluated trace elements in gluten-free foods across Italy. Most elements were detected in all samples with Ca, Fe, Mg, Ti and Zn being those with the greatest \(\bar{x}\) concentrations across food products but, below harmful levels with a normal, healthy diet. Cadmium was detected (0–600 μg kg\(^{-1}\)) in 96.8% of the U.S., 54.5% of Asian gluten-free foods, and in 100% of gluten containing foods, although all below action limits in the literature (2004). Cobalt was present in 48.4% of the U.S., 72.7% of Asian gluten-free foods, and 40% of the gluten containing foods; Cr was detected in 54.8% of the U.S., 72.7% of Asian gluten-free foods, and 100% of the gluten containing products. Chromium in gluten-free foods was below reported averages of 799 μg kg\(^{-1}\) except in sample 39 and all of the gluten containing foods; however, even these levels are below the action limits reported by the WHO (2004). There was little difference in trace metal content observed between the pasta or flour products however, gluten containing food had higher levels of metals than those deemed gluten free.

It has been suggested that gluten-free foods may at times contain higher levels of metals than gluten containing foods (Orecchio et al., 2014, Thompson, 2000). This study found elevated trace metals were more commonly found in the gluten containing products, but at limits below WHO (2004) action levels. This is likely the product used for gluten free (i.e. rice) do not have an affinity to up-take metals as does other products such as wheat and barely. It is possible that low-level exposure to metal laden gluten-free and gluten containing foods, even at low levels, can lead to chronic toxicity with long-term consumption.
Additional research is required to better understand the sources and pathways of trace elements in these foods.

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Table 1.
Data covering overall pasta and flour product data set: United States Market, Asian Market, and Gluten foods

| Source    | Type  | ID | Al  | Ba  | Ca  | Cd  | Co  | Cr  | Cu  | Fe  | Mg  | Mn  | Mo  | Ni  | Ti  | Zn  |
|-----------|-------|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| US market | Pasta | 1  | 1400| 600 | 375400 | 150 | -  | -  | 600 | 100 | 13250 | 400 | -  | 200 | 30200 | 5950 |
| US market | Pasta | 2  | 1950| 1750| 332300 | 200 | 150 | 100 | 7300| 39600| 739000| 20850| 4600| 1000| 36950| 57100 |
| US market | Pasta | 3  | 2800| 2650| 198200 | 250 | -  | 200 | 3500| 27450| 842500| 43300| 1050| 500 | 96350| 54600 |
| US market | Pasta | 4  | 550 | 1200| 109400 | 150 | 150 | 150 | 2500| 9250 | 804000| 58100| 900 | 300 | 13650| 43250 |
| US market | Pasta | 5  | 1050| 3600| 274750 | 400 | 150 | 150 | 7500| 54300| 756500| 25750| 6100| 3600| 30800| 70000 |
| US market | Pasta | 6  | 2350| 1950| 453050 | 350 | 200 | 200 | 6650| 42750| 864500| 35050| 2650| 750 | 54200| 75350 |
| US market | Pasta | 7  | 6750| 2800| 618350 | 200 | 250 | 200 | 3150| 26700| 704500| 32950| 400 | 500 | 124000| 40300 |
| US market | Pasta | 8  | 700 | 1150| 82650  | 300 | -  | -  | 2050| 6850 | 631000| 46700| 1000| 350 | 20250| 36100 |
| US market | Flour | 9  | 550 | 750 | 303250 | 200 | -  | 550 | 1850| 57100| 1817500| 83100| 450 | 4050 | 14800| 64250 |
| US market | Flour | 10 | 400 | 1450| 253700 | -  | -  | -  | -  | 28950| 1050 | -  | 150 | 7000| 5750  |
| US market | Flour | 11 | 600 | 1100| 273200 | 250 | 300 | 400 | 22550| 63200| 1871000| 74600| 750 | 1850| 82600| 79550 |
| US market | Flour | 12 | 850 | 1550| 455900 | 150 | -  | -  | 1250| 14750| 2580500| 1900 | 1200| 600 | 35200| 13300 |
| US market | Flour | 13 | 650 | 600 | 402200 | 150 | -  | 100 | 2900| 1650 | 974610| 2850 | 250 | 150 | 32500| 15750 |
| US market | Pasta | 14 | 2950| 7100| 856000 | 200 | 400 | 200 | 7650| 46800| 984000| 63300| 1650| 3850| 162400| 77150 |
| US market | Pasta | 15 | 2150| 1300| 246300 | 200 | 300 | -  | 7100| 39800| 669000| 25000| 1350| 1800| 57550| 69550 |
| US market | Pasta | 16 | 400 | 650 | 424300 | 350 | -  | -  | 750 | 7400 | 202000| 3000 | 500 | 550 | 51700| 14500 |
| US market | Pasta | 17 | 950 | 1900| 104300 | 300 | 300 | -  | 1950| 7250 | 365550| 22800| 900 | 400 | 24450| 38950 |
| US market | Pasta | 18 | 2050| 1650| 176700 | 350 | 150 | 200 | 3150| 26650| 1223000| 79900| 850 | 1000| 42200| 52400 |
| US market | Pasta | 19 | 1050| 6800 | 277050 | 150 | -  | -  | 1950| 7200 | 312600| 13150| 600 | 950 | 22700| 24000 |
| US market | Pasta | 20 | 1150| 1600| 128650 | 250 | -  | -  | 2550| 8700 | 850000| 54100| 750 | 250 | 17200| 47600 |
| US market | Pasta | 21 | 1100| 1750| 107750 | 400 | -  | 200 | 2450| 9650 | 687000| 39850| 1250| 500 | 50050| 51850 |
| US market | Pasta | 22 | 2550| 2500| 237100 | 250 | 250 | -  | 7550| 52100| 590500| 19800| 4500| 2100| 59750| 66400 |
| US market | Flour | 23 | 5900| 12100| 840000 | 200 | 350 | 150 | 8600| 49600| 1138000| 89500| 1750| 4350| 217800| 77300 |
| US market | Flour | 24 | 1250| 3450 | 2094500| 250 | 250 | 300 | 9700| 32100| 1967500| 36300| 550 | 800 | 39800| 66750 |
| US market | Flour | 25 | 900 | 500 | 651000 | 300 | -  | -  | 1800| -  | 158100| 15100| 400 | 450 | 24500| 25800 |

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| Source           | Type   | ID | Al  | Ba  | Ca   | Cd  | Co  | Cr  | Cu  | Fe   | Mg  | Mn  | Mo  | Ni  | Ti  | Zn  |
|------------------|--------|----|-----|-----|------|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|-----|
| US market Flour- | 26     | 1100 | 1550 | 14300 | 450 | -   | 150 | 2350 | 12350 | 614500 | 22050 | 900 | 850 | 120100 | 34800 |
| US market Flour- | 27     | 3250 | 800  | 96700 | 600 | 250 | -   | 2300 | 34900 | 865000 | 29200 | 1100 | 2300 | 86400 | 50050 |
| US market Flour- | 28     | 1950 | 9000 | 1436500 | 300 | 250 | 250 | 13850 | 46000 | 1804500 | 69900 | 300 | 2100 | 55250 | 80400 |
| US market Flour- | 29     | 1050 | 650  | 1936000 | 250 | -   | -   | 1100 | 49200 | 276300 | 26300 | 750 | 300 | 19900 | 25500 |
| US market Flour- | 30     | 900  | 950  | 188600 | 200 | -   | -   | 1200 | 5300 | 370050 | 23500 | 250 | 400  | 24350 | 23350 |
| US market Flour- | 31     | 1700 | 1900 | 191600 | 350 | 200 | 2250 | 13350 | 593500 | 19400 | 1200 | 900  | 85700 | 37850 |
| Asian Market Pasta- | 32   | 950  | 1300 | 65000 | 500 | -   | -   | 3500 | 3550 | 121550 | 20900 | 900 | 300  | 20200 | 30300 |
| Asian Market Flour- | 33    | 650  | 3450 | 336050 | -   | 200 | -   | 300  | 550  | 56900 | 8550  | 2100 | -   | 11100 | 6400  |
| Asian Market Flour- | 34    | 2400 | 450  | 137550 | -   | 200 | 200 | 3800 | 20450 | 1392500 | 18650 | -  | 500  | 8350  | 37900 |
| Asian Market Flour- | 35    | 1600 | 1200 | 100800 | 250 | 200 | 650 | 1000 | 2000 | 115300 | 14400 | 1750 | 350 | 30650 | 30550 |
| Asian Market Pasta- | 36    | 2150 | 2200 | 118400 | -   | 350 | 300 | 1600 | 4050 | 42350 | 4800  | 850 | 2150 | 148650 | 26450 |
| Asian Market Pasta- | 37    | 3000 | 4050 | 1416500 | 250 | -   | 400 | 850  | 7200 | 86900 | 3850  | 350 | 1850 | 94850 | 16550 |
| Asian Market Pasta- | 38    | 950  | 1100 | 145100 | 300 | 200 | 150 | 1300 | 1400 | 129700 | 13000 | -  | -   | 26450 | 27700 |
| Asian Market Pasta- | 39    | 1950 | 350  | 109100 | -   | 250 | 1950 | -   | 6700 | 31700 | 400  | -   | 500  | 24350 | 5350  |
| Asian Market Pasta- | 40    | 550  | 650  | 194800 | -   | 350 | -   | -    | -    | 33300 | -    | -   | -    | 11050 | 6750  |
| Asian Market Pasta- | 41    | 2000 | 850  | 84300 | 450 | 450 | 300 | 250  | 2650 | 39550 | 1550 | -   | 5600 | 311500 | 10200 |
| Asian Market Pasta- | 42    | 2900 | 650  | 115800 | 250 | -   | 700 | 300  | 4700 | 25400 | 1000 | -   | 5900 | 59350 | 7900  |
| Gluten Foods Pasta- | 43    | 1450 | 4750 | 209300 | 250 | -   | 1700 | 3700 | 34950 | 433500 | 19850 | 700 | 5350 | 10350 | 28000 |
| Gluten Foods Pasta- | 44    | 1600 | 5250 | 207750 | 250 | 200 | 1700 | 3150 | 33650 | 457800 | 15400 | 1050 | 4950 | 22050 | 47850 |
| Gluten Foods Pasta- | 45    | 2150 | 9700 | 323100 | 250 | 200 | 1800 | 5000 | 43750 | 1168000 | 48300 | -   | 5100 | 22350 | 68050 |
| Gluten Foods Pasta- | 46    | 1850 | 5550 | 571500 | 200 | -   | 1650 | 6100 | 50200 | 244850 | 15000 | -   | -   | 22550 | 29300 |
| Gluten Foods Pasta- | 47    | 3350 | 3700 | 659000 | 350 | -   | 1650 | 3100 | 30800 | 441950 | 18550 | -   | 5150 | 36250 | 47450 |

NOTE: Data is reported in ug kg⁻¹. Sample Identification (ID). When data is less than instrument detection level (IDL), it is represented as a "-". Sample data to be valid must be above limit of quantitation (LOQ). Statistical data shown is broken into four (4) areas; overall data, United States market gluten free foods (USM), Asian gluten free foods (Asian), and gluten containing foods (control), source of food type: r = rice products, w = wheat products.
Table 2.
Statistical data covering overall data set, United States products (USM), Asian products (Asian), and Gluten containing products (Control)

| Source | Statistical parameter | Al  | Ba  | Ca  | Cd  | Co  | Cr  | Cu  | Fe  | Mg  | Mn  | Mo  | Ni  | Ti  | Zn  |
|--------|-----------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
|        | σ                     | 1275| 2628| 4388.31| 101 | 81 | 614 | 4645 | 1950.7 | 53520.4 | 24212 | 1286 | 1824 | 58792 | 23223|
| Overall | x                     | 1754| 2606| 355133 | 278 | 252 | 575 | 4333 | 23651 | 612717 | 27890 | 1294 | 1757 | 55328 | 39407|
| data   | MIN                   | 400 | 350 | 14300 | 150 | 150 | 100 | 250  | 100   | 13250  | 400   | 250  | 150  | 7000  | 5350 |
|        | MAX                   | 6750| 12100| 2094500 | 600 | 400 | 1950 | 22550 | 63200  | 1967500 | 89500 | 6100 | 5900 | 311550 | 80400 |
|        | IDL                   | 5   | 2   | 5    | 2   | 2   | 2   | 2    | 5     | 3      | 3    | 3    | 3    | 5     |
|        | LOQ                   | 15  | 6   | 15   | 6   | 6   | 6   | 6    | 10    | 15     | 10   | 10   | 10   | 15    |
|        | σ                     | 1465| 2685| 515270 | 104 | 77  | 136  | 5276 | 19746 | 524697 | 25700 | 1406 | 1234 | 47479 | 23186|
| USM    | x                     | 1708| 2494| 424905 | 270 | 247 | 241  | 5223 | 27381 | 773434 | 34798 | 1341 | 1221 | 56139 | 45982|
|        | MIN                   | 400 | 500 | 14300 | 150 | 150 | 100  | 600  | 100   | 13250  | 400   | 250  | 150  | 7000  | 5750 |
|        | MAX                   | 6750| 12100| 2094500 | 600 | 400 | 550  | 22550 | 63200  | 1967500 | 89500 | 6100 | 4350 | 217800 | 80400 |
|        | σ                     | 867 | 1240| 73306  | 113 | 96  | 587  | 1342 | 5733  | 401170 | 7578  | 715  | 2335 | 91634 | 12044|
| Asian  | x                     | 1736| 1477| 140777 | 333 | 275 | 581  | 1433 | 5325  | 188650 | 8710  | 1190 | 2144 | 67888 | 18732|
|        | MIN                   | 550 | 350 | 65000  | 250 | 200 | 150  | 250  | 550   | 25400  | 400   | 350 | 8350 | 5350  |
|        | MAX                   | 3000| 4050| 336050 | 500 | 450 | 1950 | 3800 | 20450 | 1392500 | 20900 | 2100 | 5900 | 311550 | 37900 |
|        | σ                     | 758 | 2296| 209500 | 55  -61 | 1305 | 8058 | 356646 | 14060 | 247   | 165  | 9175 | 16410 |
| Control| x                     | 2080| 5790| 394130 | 260 | 200 | 1700 | 4210 | 38670 | 549220 | 23420 | 875  | 5138 | 22710 | 44130|
|        | MIN                   | 1450| 3700| 207750 | 200 | 200 | 1650 | 3100 | 30800 | 244850 | 15000 | 700  | 4950 | 10350 | 28000 |
|        | MAX                   | 3350| 9700| 659000 | 350 | 200 | 1800 | 6100 | 50200 | 1168000 | 48300 | 1050 | 5350 | 36250 | 68050 |

NOTE: Data is reported in ug kg⁻¹. When data is less than instrument detection level (IDL), it is represented as a “-”. Sample data to be valid must be above limit of quantitation (LOQ). Statistical data shown is broken into four (4) areas; overall data, United States market gluten free foods (USM), Asian gluten free foods (Asian), and gluten containing foods (control); mean: X; standard deviation: σ