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Comparison of heavy metal residue in selected processed canned tomato paste and bottled tomato sauce using atomic absorption spectrometry

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Abstract

Heavy metals are used to manufacture various types of packaging materials such as cans and glass bottle containers. Determination of heavy metal concentration presence in processed food products is crucial to identify specific possibly hazard metal contaminants and create public awareness upon consumption. A total of ten samples consisting of five different brands of canned tomato paste and bottled tomato sauce sold in Malaysia were analysed for the concentration of lead (Pb), cadmium (Cd), iron (Fe), zinc (Zn), chromium (Cr) and nickel (Ni) metals residues by Atomic Absorption Spectrometry (AAS). The average heavy metals concentration of all samples was arranged in increasing order, Cd > Ni > Cr > Pb > Zn > Fe. The highest level of metal was Fe (69.93 mg/kg) while the lowest was Cd (0.01 mg/kg). The concentration of Pb, Cr and Ni in all samples exceed the permissible limit set by the World Health Organisation 2011, the European Commission 2006 and the Malaysia Food Act and Regulation 1985 except Zn. There is a significant difference between heavy metal concentrations in canned tomato paste and bottled tomato sauce ($p<0.05$). Therefore, this study suggests efficient monitoring and inspection of both agricultural environment quality and industrial processing procedures.

1. Introduction

Tomato is a nutritious fruit that is commonly processed into canned and bottled tomatoes, fruit, juice, sauce, paste, pickled tomato and dried tomato slices, which can be stored longer. Processed canned tomato paste and bottled tomato sauce are important food products used in dishes to enhance the flavour of food. They are widely consumed by humans in daily life as food sources as they are nutritious to the human body. Despite being a nutritious food source for humans, processed canned tomato paste and bottled tomato sauce products may accumulate and be contaminated with heavy metal elements as a result of industrialisation, agricultural and human actions may induce harmful effects on human wellness.

Heavy metals are categorised into non-essential and essential metals. Non-essential heavy metals are those metals acknowledged as toxic compounds even in small quantities. Examples of non-essential heavy metals are lead, cadmium, tin, aluminum and arsenic. In addition, non-essential heavy metals are required in trace amounts as there is no biological function to the body (Neha *et al*., 2022) Toxicity of non-essential metals increases with increasing absorption from the polluted environment leading to adverse health effects (Serafim *et al*., 2012) In contrast, essential metals are important components for biochemical, physiological and metabolisms of living organisms (Abbasi *et al*., 2020). Examples of essential metals include iron, zinc, copper, nickel, chromium and magnesium. Insufficient amounts of essential metals will cause micronutrient deficiencies while significant concentrations produce toxicity to living organisms (Yilmaz *et al*., 2010).

Rapid globalisation, industrialisation and population growth led to critical air, water, environment and cropland pollution worldwide. Hazard heavy metals such as copper, lead, zinc, cadmium, iron, tin and arsenic are abundantly present inside the wastewater discharged from various sectors and industries. Through irrigation practice, those heavy metals bio-accumulate into soil particles and water resources will be uptake by tomato plants, which indirectly deteriorates the quality and safety of the tomato fruits (Osma *et al*., 2012).

Besides that, canned food is often endangered by heavy metal contamination no matter the raw materials, addition of preservatives, stabilizers and synthetic colouring agents, and leaking during the canning process and packaging materials used for instance, lead

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contamination causes adverse effects on human health, which due to use of solder in manufacturing of the cans. There are also reports regarding contaminated canned food products causing heavy metal poisoning (Al-Thagafi *et al*, 2014). In addition, prohibited or undeclared food packaging materials used by manufacturers will ease the migration of certain poisonous metals into the food products. Long storage periods and acidic food particles will eventually accelerate the rate of corrosion of food containers used. Fault processing processes and the use of unpermitted cans and glass bottle containers caused the leaching of heavy metal into the tomato sauce and paste (Keller and Heckman, 2015).

There are several previous studies conducted to inspect heavy metal residue in processed canned and bottled tomato products. According to a study conducted by Massadeh and Al-Massaedh (2017) in Northern Jorden, results obtained indicated that Pb, Cd, Cr, Ni and As presented in canned tomato paste exceed the permissible level set by health organizations of Codex standard 2010. Meanwhile, a study conducted by Baysal *et al*. (2011) revealed that the level of Pb, Cr, and Ni in canned tomato sauce exceeded the maximum permissible level set by WHO (Baysal *et al*., 2011). Apart from that, results obtained from a study conducted by Iwegbue *et al*. (2012) in Nigeria showed there was a significant difference between the levels of heavy metals in different brands of canned tomato paste. Pb and Cr levels found were above the permissible limit of human food set by WHO. Nevertheless, according to Muniz *et al*. (2018), the result showed the level of Cr in bottled tomato sauce was above the maximum permissible level.

This study was conducted to determine the heavy metal residue in processed tomato products using Atomic Absorption Spectrometry and made the comparison between selected processed canned tomato paste and bottled tomato sauce, to create awareness among the public and policymakers. Effective monitoring of the environment, food quality and safety of the tomatoes should be implemented to identify specific types of hazardous metals present in the food supply chain. Besides that, prevention actions should be introduced to reduce the heavy metal level in the environment and food products, which eventually decrease adverse impacts on livelihood. Researchers should work together in studying heavy metal contamination of air, water and food sources; and evaluate safety permissible limits (Tchounwou *et al*., 2014).

2. Materials and methods

2.1 Sample collection

A total of ten samples consists of processed canned tomato paste and bottled tomato sauce purchased from local convenience stores in Malaysia. Different popular brands of canned tomato paste and bottled tomato sauce products that are widely purchased by Malaysians were randomly purchased and used in this research. Canned tomato paste samples were coded as S1, S2, S3, S4 and S5 while bottled tomato sauce samples were coded as B1, B2, B3, B4 and B5 respectively for each of every different brand's product.

2.2 Method description

All glassware was soaked overnight in 10% (v/v) nitric acid prior used in sample preparation. Acid-washed glassware was rinsed with deionised water and dried before use (Boadi *et al*., 2012).

2.3 Preparation of solutions

A blank solution was prepared with 5 mL of nitric acid (HNO_3) and 2 mL of sulphuric acid (H_2SO_4). Then, the solution was diluted with deionised water, which made up to 50 mL in the volumetric flask. From the 50 mL of stock solution, 15 mL of sample solution was poured into 15 mL centrifuge tube. Then, serial dilution of 10^{-1}, 10^{-2}, 10^{-3} and 10^{-4} were conducted.

Stock solutions for each type of heavy metals Pb, Cd, Zn, Fe, Cr and Ni were prepared at a concentration of 1000 mg/L. Stock solutions used were cadmium nitrate ($Cd(NO_3)_2$), nickel (II) nitrate ($Ni(NO_3)_2$), iron (III) nitrate ($Fe(NO_3)_3$), zinc nitrate ($Zn(NO_3)_2$), lead (II) nitrate ($Pb(NO_3)_2$) and chromium (III) nitrate ($Cr(NO_3)_3$). Standard calibration solutions for each metal at concentrations ranging from 5 μg/L to 100 μg/L were prepared by diluting the stock solution of heavy metals with deionised water. Then, the calibration curve for each metal was constructed by plotting the absorbance versus concentration (Zabadi *et al*., 2018).

2.4 Sample preparation and analysis

The canned and bottled tomato sauce samples were poured out and mixed thoroughly with a blender. Then, samples were dried in oven at 70°C for 24 hrs. After drying, dried samples were ground into fine powder by using a blender (Zabadi *et al*., 2018).

Then, 5 g of dried sample was weighed into 100 mL beaker. In the fume hood, 5 mL of concentrated HNO_3 (69%) was added to the sample and heated with digital hot plate until the first vigorous reaction where dark blackish-red residue appeared. Then, 2 mL of sulphuric

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acid (H_2SO_4) was added and the mixture was continuously heated until a dark reddish residue formed (Zabadi *et al*., 2018). After that, about 25 mL to 30 mL of HNO_3 was added in small increments to maintain the oxidising conditions until the solution became clear with reddish-orange and no solid residue remained. Throughout the heating process, there was yellow-orange flame emitted. The procedure was conducted for about 1.5 to 2 hrs to completely digest the sample (Jaishankar *et al*., 2014).

After that, the solution was allowed to cool in the fume hood. The digested solution was filtered by using 0.90 mm filter paper together with a glass funnel. Then, the filtrate was syringe-filtered with 0.45 mm PTFE filter. The beaker was rinsed appropriately with deionised water to ensure quantitative transfer of the sample. After that, the mixture was diluted with sufficient amount of deionised water and was marked up to 50 mL. Next, 50 mL sample was transferred into 50 mL centrifuge tube as a stock solution. Then, 15 mL of stock sample was transferred into a 15 mL centrifuge tube as a sample solution. Then, the sample solution was serially diluted into 10^{-1}, 10^{-2}, 10^{-3} and 10^{-4} accordingly. All the procedures were repeated for all the canned tomato paste and bottled tomato sauce samples respectively (Zabadi *et al*., 2018).

All the solution was stored in the chiller at 2°C prior for the analytical process. All the six types of heavy metals (Pb, Cd, Zn, Fe, Cr and Ni) were determined by using Atomic Absorption Spectrometry. In addition, the data was presented in mg/L and was converted into mg/kg by using the conversion formula (Al-Thagafi *et al*., 2014).

3. Results and discussion

Table 1 clearly shows that the heavy metal with the highest concentration was iron (69.93 mg/kg) while the lowest was cadmium (0.01 mg/kg). Meanwhile, metal concentrations of Pb, Zn, Fe, Cr and Ni for all canned tomato paste samples were higher than bottled tomato sauce samples. Furthermore, ranking of the average concentration of all the metals presence in the samples were Fe > Zn > Pb > Cr > Ni > Cd, in that order of decreasing magnitude. In addition, the heavy metal concentrations studied in each sample were compared with the permissible limit set by Malaysia Food Act 1983 and Regulation 1985, European Commission 2016 (EC, 2016; FAO/WHO, 2011).

In addition, one-way ANOVA analysis revealed that there is significant difference between all heavy metal concentrations in canned tomato paste and bottled tomato sauce ($p<0.05$). Mean concentration of Cd, Pb, Zn, Fe and Ni metals in canned sample C1 was 0.14 mg/kg, 5.15 mg/kg, 18.80 mg/kg, 69.93 mg/kg and 1.48 mg/kg respectively, and C5 was 1.85 mg/kg, which were significantly highest as compared to other samples. The difference in metal concentration of samples were probably due to differences sources of tomato fruit brought by differences samples' brands, and probably due to difference in processing of the sauce and paste products.

Among the five canned tomato sauce samples, highest mean concentration of Cd was in C1 (0.14 mg/kg) and lowest in C2 (0.01 mg/kg). Meanwhile, among five bottled tomato paste samples, highest mean concentration of Cd was in B3 (0.11 mg/kg), and lowest in B2 and B5 (0.06 mg/kg). The concentration of Cd was consistently below the maximum permissible limit set by Malaysia Food Regulation (1985), which is 1.00 mg/kg

Table 1. Level of heavy metal concentration in canned tomato paste and bottled tomato sauce samples.

Type of sample	Concentration of heavy metal (mg/kg)					
	Cd	Pb	Zn	Fe	Cr	Ni
C1	0.14±0.0058	5.15±0.1473	18.80±0.3157	69.93±0.4431	1.54±0.0987	1.48±0.1744
C2	0.01±0.0058	3.45±0.1950	14.75±0.2082	53.49±0.3885	1.73±0.2194	1.37±0.1405
C3	0.10±0.0153	4.43±0.0252	15.21±0.6564	62.70±0.3612	1.82±0.1852	1.35±0.2272
C4	0.10±0.0000	3.53±0.2261	11.96±0.2868	51.64±0.4078	1.53±0.1415	1.16±0.2359
C5	0.02±0.0100	3.11±0.2316	15.53±0.1650	54.41±0.3156	1.85±0.2021	1.38±0.2043
B1	0.10±0.0153	2.17±0.1601	3.78±0.0436	29.63±0.3089	1.43±0.1514	0.58±0.1229
B2	0.06±0.0153	3.32±0.1168	3.87±0.0208	22.16±0.3300	1.17±0.1550	0.81±0.1808
B3	0.11±0.0153	2.68±0.0404	2.38±0.0379	19.79±0.0700	1.18±0.1818	0.63±0.1002
B4	0.07±0.0100	0.41±0.1222	3.98±0.0153	31.87±0.3175	1.14±0.0777	0.43±0.2801
B5	0.06±0.0058	3.06±0.1026	3.58±0.0462	13.44±0.1172	0.66±0.1852	0.42±0.0436
WHO/FAO	0.05	1.5	30	40	1	0.3
EC 2006	0.05	0.05	-	20	-	-
MFA	1	1	100	-	-	-

Values are presented as mean±SD.

for all sauce and paste samples. All the ten samples differ in average cadmium concentration ranging from 0.01 mg/kg to 0.14 mg/kg. The percentage range value of mean concentration below the permissible limit for these samples was 86% to 99%. This result indicated concentration of Cd was barely determined by AAS among samples as the concentration of Cd existed in very low level.

However, all samples exceed the permissible limit set by WHO/FAO 2011 and the European Commission 2006 where both the regulations,0.05 mg/kg except for canned paste samples C2 and C5. This indicated that the consumption of canned tomato paste and bottled tomato sauce is hazardous to human health, whereby permitted daily intake should be taken into consideration. In comparison with earlier study conducted by Massadeh and Al-Massaedh (2017) in Northern Jorden and David *et al.* (2008) indicated concentration of Cd in metallic can tomato paste samples was high and exceeded the permissible limit set by WHO regulation and Codex standard 2010. It suggests that Cd accumulates in certain types of fruit and vegetables due to the uptake of metals by the roots from the cultivation soil area. In general, metal residues present in fruits and vegetables are difficult to reduce via washing and peeling. Effective monitoring of heavy metal residues in canned tomato paste will aid in evaluating and improving technology development and food safety (Baysal *et al*., 2011).

The mean Pb concentration was highest in C1 (5.15 mg/kg) and lowest in C5 (3.11 mg/kg) while among five bottled tomato paste samples, the highest mean Pb concentration was B2 (3.32 mg/kg), and lowest in B4 (0.41 mg/kg). The canned tomato paste samples have higher Pb concentration compared to bottled tomato sauce samples. The high mean concentration of Pb in canned sauce C1 suggests that the soil used to cultivate tomato plants is contaminated with lead. Contaminated land fields were possibly due to industrial waste discharge from batteries and paint factories. Apart from that, Pb is mainly found in pewter and tin materials in the impurity form (Cederberg *et al*., 2015).

In comparison with the previous study conducted by Al-Thagafi *et al*. (2014), the result showed that the average Pb concentration range was 5.40 mg/kg to 3.20 mg/kg, which is similar to the result obtained from this study. The observation suggests that preservative additives added, types of containers used and corrosion of containers after a long storage period cause high Pb residue present in tomato sauce products. On top of that, acidic tomato sauce, and the long contact period between can and bottle containers causes Pb migration into tomato sauce (Muniz *et al*., 2018). Excessive consumption of Pb is toxic to the human body and cause numerous diseases such as cardiovascular, kidney and nervous system disorders. Besides that, the sources of Pb also come from coal combustion, mining of ore, solder in food containers, and lead-arsenate pesticides used by various sectors. As a result, runoff of Pb strongly binds to soil particles and sediment due to atmospheric deposition and weathering. Ungradable Pb contributes to the high concentration of Pb in the environment, whereby Pb is re-suspended, re-deposited and bio-concentrated into the soil profile (Onwuka *et al*., 2019).

Among the five canned tomato sauce samples, the highest and lowest Zn concentrations was in C1 (18.80 mg/kg) and C4 (11.96 mg/kg) respectively; while among five bottled tomato paste samples, the highest and lowest Zn concentration was in B4 (3.98 mg/kg) and B3 (2.38 mg/kg) respectively. Besides that, Zn was found second highest level in all the tomato sauce and paste samples, among the heavy metals analysed. This result agreed with a previous study conducted by Zabadi *et al*. (2018), results showed zinc was the highest Zn had the highest concentrations which were 2.05 mg/kg to 10.6 mg/kg (Zabadi *et al*., 2018). This observation suggests that Zn will enter the food products from soil because of crops' mineralization and environment contaminated with Zn-based chemical substances. This is because Zn is widely used in various industries such as electroplating, smelting and ore processing industries. Moreover, zinc sulfate is used as a zinc supplement for feeding animals, a zinc-based fertiliser in the agricultural sector to boost soil nutrients and spray agents to control disease. Zn is an essential micronutrient for the growth and metabolism of plants and tend to be bio-accumulated in soil profile (Alves and Reis, 2016).

Nevertheless, according to Cederberg *et al*. (2015), high Zn concentration in processed canned and bottled food products might be due to galvanized iron containers used to fill in the acidic tomato paste and sauce products (Dhuey *et al*., 2021). Zn is usually used as a coating material to prevent corrosion of the can containers. Indeed, acidic tomato sauce will react with zinc coating forming salt compounds that are readily absorbed by the human body and might cause mild sickness and other disorders. There are zinc poisoning cases have been reported due to acidic drink packed in galvanized iron containers being consumed. However, results showed that all bottled tomato sauce products are safe to be consumed as the average zinc concentration was 3.52 mg/kg, which is below the dietary intake limit. This is because Zn is an important trace metal needed by the human body as well as plants to have normal growth, excretion, immunity and metabolism development in adequate amounts. However, extremely high levels and long-term exposure to Zn would pose a risk to human

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health (Zabadi *et al*., 2018).

The highest concentration was in C1 (69.93 mg/kg) and the lowest in C4 (51.64 mg/kg); while among five bottled tomato paste samples was in B4 (31.87 mg/kg) and lowest in B5 (13.44 mg/kg). The mean concentration of iron presence in all canned tomato paste samples detected ranging of 51.64 mg/kg to 69.93 mg/kg was above the maximum permissible limit set by WHO/FAO. This result is equivalent with a previous study conducted by David *et al.* (2008) on canned tomato paste purchased from the Romania market. The concentration of Fe in those samples was very high, in a range value of 17.53 mg/kg to 219.58 mg/kg. This observation suggests that environment pollution causes Fe contamination in plant and crop yield. Primarily, Fe is highly generated from iron ore mining and processing plant industries. Deposition, weathering and runoff of Fe cause high levels of Fe in cropland. The plant tends to uptake Fe for metabolism function and continuous growth. This led to high Fe detected in all the samples.

Furthermore, according to research conducted by Greger and Baier (2010), results showed that the mean Fe concentration detected in canned tomato sauce samples was 69.00±1.4 mg/kg, which exceeded the permissible limit of WHO/FAO (40.00 mg/kg). This value was also higher than bottled tomato sauce samples, which was 28.00±0.060 mg/kg. These observations suggest that the can containers might not coated with lacquer on their interior surfaces. Indeed, some can containers are lacquered with resin coating and also show slightly high levels of Fe due to the reason of high acidity of tomato sauces stored inside the can containers. Meanwhile, dents and scratches of the interior can container together with acidic tomato sauce accelerate corrosion and migration of Fe into food particles. Oxidizing agents, cupric salt, pigment substances and sulfur compounds cause disintegration of Fe and Sn metals. As a result, food comes in contact with those hazardous metals and leads to high contamination. Besides that, a higher level of Fe was detected in canned samples than in bottled samples, which is similar to the study conducted by Cederberg *et al.*, 2015. This is due to the higher contact surface for canned products compared to glass bottled products as iron metal is only present on the lids and closures of glass bottles and jars.

The highest mean concentration of Cr was in C5 (1.85 mg/kg) and lowest in C4 (1.53 mg/kg); while among five bottled tomato paste samples was in B1 (1.43 mg/kg), and lowest in B5 (0.66 mg/kg). High chromium concentrations detected in analysed canned food samples were similar to a previous study conducted by Massadeh and Al-Massaedh (2017) with range mean concentration of Cr was 0.66 mg/kg to 0.94 mg/kg, which exceeded the permissible limit set (0.10 mg/kg). These results indicate that high Cr concentrations are possibly due to the corrosion of can containers used and the leaching of Cd from unlacquered cans used. As a result, it is suggested that authoritarian control and analysis procedures should be taken in order to reduce the risk of heavy metal contamination. Advances in packaging technology and on-going research and development are crucial (Massadeh and Al-Massaedh, 2017).

Furthermore, a study conducted Diviš *et al.* (2017) shown the average concentration of Cr detected in the canned fruit samples was 1.37 mg/kg and exceeded the permissible limit. This observation suggests that tin coating applied on steel plates to avoid corrosion and damage to the tinplate container has caused the dissolution of other metallic elements such as Cr, Zn, Cd and Pb into the inner food particles. Indeed, thin chromium oxide layer might apply when manufacturing a can container to avoid corrosion. As a result, high amounts of hazardous metals are used when canning the food product increases the possibility of food inside being contaminated with those metals. Consequently, according to Anastácioa *et al*. (2018), canned fruit juices that have chromium concentrations that exceed the maximal permissible values are still can be consumed and do not cause risk to human health. The reason for this statement is the amount of fruit juice drunk per day is expected lesser than the amount of water. This same goes for the amount of canned tomato paste consumed per day is expected lesser than the amount of rice.

Ni has the highest concentration in C1 (1.48 mg/kg) and lowest in C4 (1.16 mg/kg); while among five bottled tomato paste samples was in B2 (0.81 mg/kg) and B5 (0.42 mg/kg) respectively. A higher level of nickel detected in all samples indicated that possible contaminated raw tomato fruit was used and improper processing procedures in the factory. A study conducted by Osma *et al.* (2012) indicated that the Ni level detected in fresh tomato fruit planted in suburban areas was 1.06 mg/kg, which is less than in industrial locations, 1.69 mg/kg. In addition, the study also proved that proper washing of raw tomato fruit prior to processing can reduce metal residue on fruits. Ni concentration detected in the unwashed tomato sample was 1.69 mg/kg, which is much higher than the washed tomato sample of 1.60 mg/kg. Thus, these observations suggest that standard manufacturing guidelines should be obeyed by food manufacturers in order to produce quality and safety-assured products for consumption.

The high concentration of Ni detected in canned tomato paste samples might be due to the wide usage of

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Ni alloy in the plating industry prior to cans production Cederberg *et al.* (2015). Ni is used to produce high quality alloys with good corrosion resistant properties together with other metals such as Fe, Al, Cr and Zn. Ni containing stainless steel equipment such as tomato sauce tanks and packing containers such as metallic cans used in manufacturing processes. Apart from that, a study conducted by Onwuka *et al*. (2019) showed that exceeded mean concentration Ni in canned food products. This is because Ni releases as wastewater discharge from the mining, oil-burning and coal-burning industries into the environment, and ends up in soil and sediment as it strongly binds to substances containing iron and manganese. Subsequently, Ni is readily uptake and bio-accumulated in the tomato plant and its fruits.

4. Conclusion

In this study, the levels of copper, cadmium, lead, nickel, chromium, manganese, iron, cobalt and zinc in canned tomato paste were investigated. The result showed that some heavy metals were present in selected canned tomato paste. Considerable differences were found among the various concentrations of canned tomatoes. The levels of cadmium and cobalt complied with the permissible limit. Chromium, manganese cadmium and zinc showed some degree of elevation. It is strongly suggested that heavy metal levels in canned tomato paste should be monitored periodically to prevent heavy metals burden.

Conflict of interest

The authors declare no conflict of interest.

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