

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.jfda-online.com

Original Article

Investigation of aluminum content of imported candies and snack foods in Taiwan



Tai Sheng Yeh^{*}, Yeng-Ting Liu, Pei-Jyun Liou, Hong-Ping Li, Ching-Chuan Chen

Department of Food Science and Nutrition, Meiho University, Pingtung, Taiwan, ROC

ARTICLE INFO

Article history:

Received 8 September 2015

Received in revised form

6 April 2016

Accepted 12 April 2016

Available online 21 June 2016

Keywords:

aluminum

candy

dietary exposure

snack food

ABSTRACT

Candies, chewing gums, dried fruits, jellies, chocolate, and shredded squid pieces imported from 17 countries were surveyed for their aluminum content. The samples were bought from candy shops, supermarkets, and convenience stores, and through online shopping. Sample selection focused on imported candies and snacks. A total of 67 samples, including five chewing gums, seven dried fruits, 13 chocolates, two jellies, two dried squid pieces, and 38 candies, were analyzed. The content of aluminum was analyzed by inductively coupled plasma optical emission spectrometry (ICP OES). The limit of quantitation for aluminum was 1.53 mg/kg. The content of aluminum ranged from not detected (ND) to 828.9 mg/kg. The mean concentrations of aluminum in chewing gums, dried fruits, chocolate, jellies, dried squid pieces, and candies were 36.62 mg/kg, 300.06 mg/kg, 9.1 mg/kg, 2.3 mg/kg, 7.8 mg/kg, and 24.26 mg/kg, respectively. Some samples had relatively high aluminum content. The highest aluminum content of 828.9 mg/kg was found in dried papaya threads imported from Thailand. Candies imported from Thailand and Vietnam had aluminum contents of 265.7 mg/kg and 333.1 mg/kg, respectively. Exposure risk assessment based on data from the Taiwan National Food Consumption Database was employed to calculate the percent provisional tolerable weekly intake (%PTWI). The percent provisional tolerable weekly intake of aluminum for adults (19–50 years) and children (3–6 years) based on the consumption rate of the total population showed that candies and snacks did not contribute greatly to aluminum exposure. By contrast, in the exposure assessment based on the consumers-only consumption rate, the estimated values of weekly exposure to aluminum from dried papaya threads in adults (19–50 years) and children (3–6 years) were 4.18 mg/kg body weight (bw)/wk and 7.93 mg/kg bw/wk, respectively, for 50th percentile consumers, and 6.26 mg/kg bw/wk and 12.88 mg/kg bw/wk, respectively, for 95th percentile consumers.

Copyright © 2016, Food and Drug Administration, Taiwan. Published by Elsevier Taiwan LLC. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

^{*} Corresponding author. Department of Food Science and Nutrition, Meiho University, 23, Pingguang Road, Neipu, Pingtung 91202, Taiwan, ROC.

E-mail address: x00010091@meiho.edu.tw (T.S. Yeh).

<http://dx.doi.org/10.1016/j.jfda.2016.04.004>

1021-9498/Copyright © 2016, Food and Drug Administration, Taiwan. Published by Elsevier Taiwan LLC. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

Aluminum is the third most abundant element on earth. Aluminum additives are widely used in many food products such as flour, baking powder, firming agents, coloring agents, and anticaking agents. Food is the major source of aluminum exposure to humans [1]. Sedman [2] reported that aluminum intoxication was an iatrogenic disease and could cause encephalopathy, metabolic bone disease, and microcytic anemia. Excessive aluminum exposure to humans had been associated with adverse neurologic, hematopoietic, skeletal, respiratory, immunologic, and other health effects [3–5]. The European Food Safety Authority (EFSA) has established a tolerable weekly intake for aluminum, which is 1 mg aluminum/kg body weight (bw)/wk, and for highly exposed consumers the intake was estimated to be 2.3 mg/kg bw/wk [6]. In the 2011 Joint FAO/WHO Expert Committee on Food Additives (JECFA) report, the provisional tolerable weekly intake (PTWI) for aluminum was established to be 2 mg aluminum/kg bw/wk [7]. The dietary exposure estimates of children to aluminum-containing food additives could exceed the PTWI by up to twofold, according to the 2011 JECFA report. Codex [8] and EFSA regulated the maximum content of aluminum-containing additives in different specified food items to reduce the dietary exposure to aluminum. However, food additives containing aluminum were generally recognized as safe (GRAS) by the US Food and Drug Administration (FDA) when used as a salt substitute in accordance with good manufacturing practice. For foods more likely to be highly consumed by children, the Codex General Standard for Food Additive (GSFA) established the maximum permissible level of aluminum in various food category with added aluminum-containing additives as follows: 100 mg/kg in chewing gums, 100 mg/kg in crackers, 100 mg/kg in ordinary bakery products, 60 mg/kg in dairy-based drinks, 40 mg/kg in steamed breads and buns, and 40 mg/kg in mixes for bread and ordinary bakery wares [8]. The Commission Regulation (EU) No. 380/2012 permitted the maximum level of aluminum coming from all aluminum lakes to be up to 30 mg/kg in potato-, cereal-, flour-, or starch-based snacks; up to 70 mg/kg in confectioneries, and candied fruits and vegetables; up to 200 mg/kg, as aluminum sulfate, in candied cherries; and up to 300 mg/kg in chewing gums. In 2016, the Taiwan FDA specified the application scope and the maximum permissible level of food additives containing aluminum, including ammonium aluminum sulfate (INS523), aluminum potassium sulfate (INS 522), sodium aluminum sulfate (INS521), aluminum sulfate (INS520), and acidic sodium aluminum phosphate [INS541(i)]. These food additives have been restricted for use only in specified categories of food, with the maximum limits being 500 ppm in processed mollusks, crustaceans, and echinoderms products; 500 ppm in seaweed; 300 ppm in fried puffed foods; 300 ppm in pastries; 200 ppm in pickled vegetables; and 40 ppm in mixes for bread and ordinary bakery wares.

According to Sato et al [9], daily intakes of aluminum from sugar and confections/savories for younger children (1–6 years old) and children of 7–14 years of age in Japan were 0.83 mg/person/d and 0.7 mg/person/d, respectively. Only in

small children the aluminum exposure for the percentages of 95th percentile (P95) to PTWI (2.0 mg/kg bw/wk) exceeded 100%, according to Sato et al [9]. Children are more susceptible to aluminum overexposure per kilogram of body weight than adults [6,7,10]. The study by Guo et al [11] found that children in China had the highest risk of aluminum exposure, with 22.8% having an aluminum intake higher than the JECFA PTWI. By contrast, only 3.2% of adults exceeded the PTWI in the same study.

The Taiwan FDA conducted a survey on aluminum content in domestic food products in 2012. The survey focused on aluminum-rich foods, including ordinary bakery products, fried puffed foods, pastry products, sugar-coated desserts, processed jelly fish products, mixes for bread and ordinary bakery wares, mung bean vermicelli, and cheese- and cocoa-based products. Previously, no surveillance and monitoring of aluminum content for imported candies and snack foods in Taiwan were carried out [12]; hence, a study to assess the dietary exposure of aluminum from candies and snack foods is very important.

2. Methods

2.1. Reagents and chemicals

Aluminum (1000 µg/mL, ISO Guide 34 Certified Reference Material) was obtained from High-Purity Standards (Charleston, SC, USA). Nitric acid (Selectipur-UPS, 70% purity) was obtained from BASF SE (Ludwigshafen, Germany). Hydrogen peroxide (Perdrogen 30% H₂O₂ (w/w), reagent grade, ISO, reagent grade) was acquired from Sigma-Aldrich (St Louis, MO, USA).

2.2. Equipment

The high-speed pulverizing machine RT-02A was acquired from Rong Tsong Precision Technology Co. (Taichung City, Taiwan). The ASX-500 Series Auto Sampler was obtained from Agilent Technologies (Santa Clara, CA, USA) and the heating block BHW-09C from Kohan Instruments Co., Ltd. (Taipei, Taiwan). Microwave digestion of the sample was performed by CEM MARSXpress (CEM Corp., Matthews, NC, USA). Horiba Jobin Yvon-Ultima 2 inductively coupled plasma optical emission spectrometry (ICP OES) (HORIBA Jobin Yvon S.A.S., Longjumeau, France) was employed for the determination of aluminum.

2.3. Sample collection and pretreatment

The samples were bought from candy shops, supermarkets, and convenience stores, and through online shopping. Sample selection focused mainly on imported candies and snacks. A total of 67 samples, including five chewing gums, seven dried fruits, 13 chocolates, two jellies, two dried squid pieces, and 38 candies, were analyzed.

The samples were first cut into small pieces and homogenized using the high-speed pulverizing machine RT-02A. About 0.25 g homogenized sample was placed in a microwave digestion vessel, and 5 mL concentrated nitric acid was added. The temperature of the heating block was set at 105°C

for 30 minutes for predigestion prior to microwave digestion. After the digestion vessel was cooled down, 2 mL H₂O₂ was added to it. The digestion vessel was then placed in the microwave digester for digestion and heating by the temperature program in two parts. In the first part of the digestion program, the temperature was ramped to 170°C within 20 minutes, followed by a hold time of 5 minutes under microwave irradiation at 1600 W. In the second part of the digestion program, the temperature was ramped to 200°C within 6 minutes, followed by a hold time of 30 minutes under microwave irradiation at 1600 W.

When the digestion vessel was cooled down to room temperature, the content was placed into a 25-mL volumetric flask and deionized water added up to the 25-mL mark. The sample solution was filtered through a filter paper before ICP OES analysis.

Operation conditions for ICP OES were as follows: The radio frequency (RF) power was 1000 W. Argon gas flow rates for the plasma, auxiliary, and nebulizer flow were 12 L/min, 0 L/min, and 0.67 L/min, respectively. The elemental wavelength for aluminum detection was 396.152 nm.

3. Results and discussion

3.1. Method validation

3.1.1. Linearity of calibration curve and limit of quantitation
The calibration curve was linear for the aluminum concentrations of 100 ng/mL, 250 ng/mL, 500 ng/mL, 750 ng/mL, and 1000 ng/mL. The slope and intercept of the calibration curve were 12.032 and 877.75, respectively. The correlation coefficient of the calibration curve was 0.9949. The limit of quantitation (LOQ) was 1.53 µg/g, as determined by the standard deviation of signals (*s*) and slope (*m*) in the calibration curve, i.e., $LOQ = 10 \times s/m$ [13].

3.1.2. Precision and accuracy

From our previous work and the work by Stahl et al [14], cocoa-based products such as chocolate could have a high aluminum content. From the work of Sato et al [9], the recovery of aluminum did not vary greatly for fish/shellfish, sugar and confections/savories, fruits, vegetables, and seaweeds. Therefore, a recovery test was conducted for chocolate. The recovery rate was evaluated by spiking 300 ng/mL aluminum standard solution into chocolate in triplicate for 3 days. The recovery rate was between 85.6% and 108.9%, and the mean recovery rate was 94.9% for the spiked samples. The coefficient of variation was checked by spiking 300 ng/mL and 500 ng/mL aluminum standard solutions into chocolate in triplicate for 3 days. The coefficient of variation was 4.7% and 7.0%, respectively, for 300 ng/mL and 500 ng/mL.

3.2. Aluminum content in candies and snacks

Aluminum content in different types of candy and snack samples are shown in Table 1. For the present study, in 81% of the samples, the concentration of aluminum was greater than the LOQ. For calculating the mean concentrations, it had been assumed that in samples with aluminum concentrations less

Table 1 – Aluminum content in different types of candy and snack samples.

Type	No.	Content (mg/kg)	Mean ^a (mg/kg)	Median (mg/kg)
Chewing gums	5	19.7–54.3	36.62	40.70
Dried fruits	7	2.3–828.9	300.06	18.3
Chocolate	13	1.4–23.0	9.1	8.2
Jellies	2	2.4–2.4	2.3	2.3
Dried squid pieces	2	7.2–8.4	7.8	7.8
Candies	38	0.8–333.10	24.54	3.8

LOQ = limit of quantitation.

^a A value of 1/2 LOQ is assigned for samples with aluminum concentration below LOQ when calculating the mean concentration.

than the LOQ, the concentrations were equal to 1/2 LOQ, according to the World Health Organization (WHO) risk assessment principle [15]. Dried fruits were found to have the highest mean aluminum content, followed by chewing gums and candies. The highest median aluminum content was found in chewing gums, followed by dried fruits and chocolate. Aluminum content, mean and median, in samples from different world regions are shown in Table 2. The products imported from the EU had the lowest mean and median aluminum content. This could be due to stricter regulations in EU countries.

The dried fruit samples included one dried mango, three raisins, and three dried papaya threads. The highest aluminum contents were found in three dried papaya threads from Thailand, which were 828.9 mg/kg, 646.7 mg/kg, and 576.2 mg/kg. A dried papaya thread is actually a dried fruit product that imitates a dried fig (*Ficus carica*) thread, and mislabeling has been previously reported in the news media. Taiwan FDA had demanded the product vendors to provide correct labeling. The mean aluminum concentration of the three raisins was 15.4 mg/kg, and the concentration of dried mango from Thailand was 2.3 mg/kg. The aluminum content in raisins was higher than 5.92–8.76 mg/kg reported by Bratakos et al [16]. Altundag and Tuzen [17] reported that the average aluminum content was 0.83–12.02 mg/kg in dried fruits, 7.69–10.53 mg/kg in raisins (*Vitis vinifera* L.), and 0.83–1.06 mg/kg in figs (*F. carica* L.). González-Weller et al [18] reported that the mean aluminum content in hazelnuts and

Table 2 – Aluminum content, mean and median, in samples from different world regions.

Region	Number	Content (mg/kg)	Mean (mg/kg)	Median (mg/kg)
America ^a	3	12.7–28.6	19.7	18.3
East Asia ^b	18	0.8–54.3	14.0	5.9
Europe ^c	12	0.8–8.7	3.0	1.1
South East Asia ^d	31	0.8–829.9	96.0	6.9
West Asia ^e	3	5.4–15.3	10.1	9.6

^a America: US, Chile.

^b East Asia: China, Korea, Japan.

^c Europe: Czech Republic, France, Germany, Hungary, Italy, the Netherlands.

^d South East Asia: Indonesia, Malaysia, Thailand, Vietnam.

^e West Asia: Iran, Turkey.

dried figs was 4.89 mg/kg. Ekholm et al [19] found that the aluminum content of fruits and berries was 5–32 mg/kg. For crystallized fruit samples and figs, the aluminum content was 1.0–8.9 mg/kg and 3.6–7.2 mg/kg, respectively, in the study by Tóth et al [20]. The maximum limit for aluminum sulfate was 300 mg/kg in crystallized fruits in Portugal; however, the aluminum content in the study by Tóth et al [20] was much lower than the Portugal regulatory limit. Usually the reported aluminum content in fruits and dried fruits is quite low, but there were reports about products containing a relatively high level of aluminum in Asian countries. According to Chen et al [21], the average aluminum content in glacé fruits was 62 mg/kg, and the highest aluminum content in their study was 236 mg/kg. Jiang et al [22] detected 3.3–209.0 mg/kg aluminum in preserved fruits. Li et al [23] found that in nearly 50% of preserved fruits in Shijiazhuang city, the aluminum content was found to range from 60 mg/kg to 851 mg/kg. The Commission Regulation (EU) No. 380/2012 [24] regulated the limit for aluminum coming from all aluminum lakes to 70 mg/kg for candied fruits and vegetables. Similarly, the maximum level of aluminum sulfate in candied cherries was regulated at 200 mg/kg to reduce the aluminum exposure level from these products.

The aluminum content of 19.7–54.3 mg/kg in chewing gums was similar to the level 36–64 mg/kg determined by Viñas et al [25]. The aluminum content of 610–900 mg/kg reported by Kupchella and Syty [26] and 132–515 mg/kg by Lione and Smith [27] was higher than that found in the present study. Even though the aluminum content in a single stick of chewing gum could amount to 20% of normal daily intake, Lione and Smith [27] found that the aluminum extracted by chewing would contribute only 2.2% at most to the estimated daily intake of aluminum from all sources. As the presence of aluminum in chewing gums was mostly due to the use of aluminum-containing food additives, the Commission Regulation (EU) No. 380/2012 regulated the maximum limit for aluminum coming from all aluminum lakes to be 300 mg/kg. The maximum limit for aluminum silicate (INS 559), calcium aluminum silicate (INS 556), and sodium aluminosilicate (INS 554) in chewing gums was 100 mg/kg, according to Codex General Standard for Food Additives [8]. In the recent survey by Marín-Martínez et al [28], the mean aluminum content in chewing gums without sugar and chewing gums with sugar was 40.63 mg/kg and 54.55 mg/kg, respectively. The lower aluminum content surveyed in chewing gums in recent years could be a consequence of international regulations and the use of new aluminum-containing food additives.

Relatively high aluminum content in candies could be detected due to the use of aluminum-containing additives. Yang et al [29] reported that in orange color-coated candies the highest Ponceau 4R lake dye concentration was 353.8 mg/kg, and in red color-coated candies the highest Allura Red lake dye concentration was 322.7 mg/kg. There were two candies with an aluminum content of >200 mg/kg in this study. These two samples were fruit-flavored candies with a hard coating of different colors. The aluminum concentration in the different colors of highest mean concentration fruit-flavored candy sample was yellow color 799.0 mg/kg, blue color 704.3 mg/kg, orange color 686.2 mg/

kg, and pink color 325.9 mg/kg respectively. Aluminum content in the sample with the second highest aluminum mean concentration was as follows: 310.9 mg/kg in blue color, 259.6 mg/kg in purple color, and 259.1 mg/kg in red color fruit-flavored candies. In Belgian food samples, Fekete et al [30] detected 0.05–560.35 mg/kg aluminum in sugar and confectionaries. For food samples in Hong Kong, Wong et al [31] found that confectioneries with a coating could have aluminum content in the range of 1–201 mg/kg. Stahl et al [14] reported confectioneries with an aluminum content of 1–184 mg/kg in German food products. Most of the other previous studies had shown that the aluminum level in candies was <30 mg/kg. Müller et al [32] reported an aluminum content of 3.4–12 mg/kg in candies, Schäfer and Seifert [33] reported 3.6 mg/kg in candies, Jalbani et al [34] reported 10.87–21.7 mg/kg in sugar-based candies, Millour et al [35] reported 0.472–3.05 mg/kg in sugar and sugar-based products, Bratakos et al [16] reported 0.43–11.10 mg/kg in sweets and sugars, and Sato et al [9] reported 17.03–21.73 mg/kg in sugar and confections.

The aluminum content of 19.7–54.3 mg/kg in chocolate was similar to the level of 1.77–54.1 mg/kg in chocolate reported by Millour et al [35]. The aluminum level was higher in cocoa and chocolate products from previous studies; in particular, chocolate products with a higher cocoa content could have higher aluminum content. The aluminum content was 9.4–103 mg/kg in cocoa and cocoa products, as reported by Müller et al [32]; 9.9–111 mg/kg in cocoa and cocoa products, as reported by Schäfer and Seifert [33]; 48.84–184.3 mg/kg in cocoa-based chocolate and 20–38.4 mg/kg in milk-based chocolate, as reported by Jalbani et al [34]; 6–150 mg/kg in chocolate, as reported by Stahl et al [14]; and 30–312 mg/kg in cocoa powder, as reported by Stahl et al [14].

3.3. Estimation of risk of exposure to aluminum from candies and snacks

In order to estimate the aluminum exposure through consumption of candies and snacks intake, the observed individual mean approach currently used by the EFSA to estimate the long-term exposure of food additives was adopted [15,36]. In the observed individual mean approach, all the relevant foods consumed on a person-day, which are present in the food consumption database, are multiplied by the mean concentration of a chemical in that food. Two estimates based on different model populations are calculated: the total population (including consumers and non-consumers) and the consumers only model [11,37]. The total population model was suitable for total diet study [38]. For commodity not eaten very frequently, the consumers-only model was a preferable approach for estimating consumer risk. We compared the percentage of estimated weekly intake (EWI) with the PTWI set by JECFA to characterize the extent of exposure. The food consumption rate (g/d) and the body weight data (kg) were taken from the Taiwan National Food Consumption Database [15,39] to evaluate the exposure of aluminum from candies and snacks. The mean and standard deviation of the consumption rate (g/d) for different candy and snack categories in the total population and in consumers only are listed in Table 3. The mean

Table 3 – Mean and SD of consumption rate (g/d) of different candy and snack categories in the total population and in consumers only.

Age group	Type	Total population ^a		Consumers only ^b	
		Mean (g/d)	SD (g/d)	Mean (g/d)	SD (g/d)
3–6 y	Candy ^c	2.92	17.88	24.19	32.54
	Chewing gum ^c	2.92	17.88	24.19	32.54
	Chocolate ^d	1.52	8.19	23.41	14.14
	Snack ^e	2.51	15.94	24.06	41.58
	Jelly ^f	17.33	94.78	138.05	117.72
	Dried mango ^g	0.05	0.43	4.64	2.88
	Raisin ^h	0.54	3.66	13.74	13.19
	Dried papaya thread ⁱ	0.35	4.74	35.56	10.72
	19–65 y	Candy ^c	1.6	18.24	26.97
Chewing gum ^c		1.6	18.24	26.97	42.54
Chocolate ^d		2.05	36.93	64.21	71.54
Snack ^e		1.23	11.12	25.16	34.72
Jelly ^f		5.63	63.19	140.37	185.91
Dried mango ^g		0.15	1.78	18.05	19.05
Raisin ^h		0.68	7.12	18.06	33.81
Dried papaya thread ⁱ		0.08	2.22	56.74	14.17

NFCD = National Food Consumption Database; SD = standard deviation.

^a Consumption rate from the total population.

^b Consumption rate from consumers only.

^c Taiwan NFCD food item category K201.

^d Taiwan NFCD food item category K202.

^e Taiwan NFCD food item category K303.

^f Taiwan NFCD food item category K304.

^g Taiwan NFCD food item category I402.

^h Taiwan NFCD food item category I202.

ⁱ Taiwan NFCD food item category I102.

chocolate daily intake of 1.52 g for children 3–6 years of age in the total population was lower than the average chocolate daily intake of 19 g in Germany [14], but the mean chocolate daily intake of 23.41 g in consumers only was comparable with the German consumption. Similarly, the mean candy intake of 2.92 g/d for children 3–6 years of age in the total population was lower than the average confectionery intake of 21 g/d in Germany [14], but the mean candy intake of 24.19 g/d in consumers only was comparable with the German consumption. The serving size for dried papaya threads with the highest aluminum content was 35 g, which was comparable with the mean consumption rate of 35.56 g/

respectively. The exposure scenario for the average consumer was estimated by the 50th percentile (P50) consumption rate and that for high consumers was estimated by the P95 consumption rate from the respective total population and consumers only consumption data. The P50 and P95 food consumption rates could be determined from the National Food Consumption Database by lognormal distributions. The mean concentration of aluminum in different sample types was adopted to calculate the EWI, as suggested by the WHO and EFSA dietary exposure assessment principle [36,40]. The P50 and P95 EWI and %PTWI were calculated by the following equations:

$$\text{EWI (mg/kg bw/wk)} = \frac{\left[\text{mean Al concentration} \left(\frac{\text{mg}}{\text{kg}} \right) \right] \times \text{CR} \left(\frac{\text{g}}{\text{day}} \times \frac{1 \text{ kg}}{1000 \text{ g}} \right) \times 7 (\text{d/wk})}{[\text{body weight (kg)}]} \quad (1)$$

d for children aged 3–6 years in consumers only. The serving size for fruit-flavored candies with the highest aluminum content was 20 g, which was comparable with the mean consumption rate of 24.19 g/d for children aged 3–6 years in consumers only.

According to the Taiwan National Food Consumption Database, the mean body weights for children aged 3–6 years and adults aged 19–65 years are 20.56 kg and 63.05 kg,

$$\% \text{PTWI} = \frac{\text{EWI}}{\text{PTWI}} \times 100 \quad (2)$$

The values for estimated weekly exposure of aluminum (mg/kg bw/wk) for adults (19–65 years) and children (3–6 years) based on the consumption rate in the total population are given in Table 4. The calculated P50%PTWI for both children (3–6 years) and adults (19–65 years) was

Table 4 – Estimated weekly exposure to aluminum (mg/kg bw/wk) for children (3–6 years) and adults (19–50 years) based on the consumption rate in the total population.

Age group	Type	Mean Al (mg/kg)	P50 CR ^a (g/d)	P95 CR ^b (g/d)	P50 EWI ^c (mg/kg bw/wk)	P95 EWI ^d (mg/kg bw/wk)	P50%PTWI ^e (%)	P95%PTWI ^f (%)
3–6 y	Candy ^g	24.5	0.47	10.90	0.0039	0.0909	0.20	4.55
	Chewing gum ^g	36.6	0.47	10.90	0.0059	0.1359	0.29	6.79
	Chocolate ^h	9.1	0.28	5.76	0.0009	0.0179	0.04	0.89
	Snack ⁱ	7.8	0.39	9.32	0.0010	0.0248	0.05	1.24
	Jelly ^j	2.3	3.12	65.61	0.0024	0.0514	0.12	2.57
	Dried mango ^k	2.3	0.01	0.18	0.0000	0.0001	0.00	0.01
	Raisin ^l	15.4	0.08	1.99	0.0004	0.0104	0.02	0.52
19–65 y	Dried papaya thread ^m	683.9	0.03	1.10	0.0060	0.2570	0.30	12.85
	Candy ^g	24.5	0.14	5.28	0.0004	0.0144	0.02	0.72
	Chewing gum ^g	36.6	0.14	5.28	0.0006	0.0215	0.03	1.07
	Chocolate ^h	9.1	0.11	5.94	0.0001	0.0060	0.01	0.30
	Snack ⁱ	7.8	0.14	4.29	0.0001	0.0037	0.01	0.19
	Jelly ^j	2.3	0.50	18.66	0.0001	0.0048	0.01	0.24
	Dried mango ^k	2.3	0.01	0.49	0.0000	0.0001	0.00	0.01
	Raisin ^l	15.4	0.06	2.29	0.0001	0.0039	0.01	0.20
Dried papaya thread ^m	683.9	0.00	0.20	0.0002	0.0152	0.01	0.76	

Al = aluminum; bw = body weight; CR = consumption rate; EWI = estimated weekly intake; NFCD = National Food Consumption Database; P50 = 50th percentile; P95 = 95th percentile; PTWI = provisional tolerable weekly intake; %PTWI = percent provisional tolerable weekly intake.

^a Fifty percentile of CR in the total population.

^b Ninety-five percentile of CR in the total population.

^c P50 EWI = mean Al × P50 CR × 7/bw/1000.

^d P95 EWI = Mean Al × P95 CR × 7/bw/1000.

^e P50%PTWI = (P50 EWI/PTWI) × 100.

^f P95%PTWI = (P95 EWI/PTWI) × 100.

^g Taiwan NFCD food item category K201.

^h Taiwan NFCD food item category K202.

ⁱ Taiwan NFCD food item category K303.

^j Taiwan NFCD food item category K304.

^k Taiwan NFCD food item category I402.

^l Taiwan NFCD food item category I202.

^m Taiwan NFCD food item category I102.

<1%. For children (3–6 years), the highest P95%PTWI was 12.85% from consuming dried papaya threads, followed by 6.79% from chewing gums and 4.55% from candies. However, for adults (19–65 years), the highest P95%PTWI was 1.07% from consuming chewing gums, followed by 0.76% from dried papaya threads, and 0.72% from candies. The results indicated that for the total population aluminum exposure from candies and snacks would not pose any health risk.

Table 5 illustrates the aluminum exposure risk from candies and snacks for consumers only. Dietary exposure in consumers only was higher than that in the total population, as reported in the dietary exposure study by Guo et al [11]. The P95 aluminum exposure of children among consumers only was 7.61 mg/kg bw/wk and that for the total population was 6.50 mg/kg bw/wk. The calculated P50% PTWI values from consuming dried papaya threads were 396.38% and 208.99% for children (3–6 years) and adults (19–65 years), respectively; the P95%PTWI values from consuming dried papaya threads were 643.87% and 313.21% for children and adults, respectively. For both the children and adults, EWI from consuming dried papaya threads exceeded the aluminum PTWI suggested by JECFA. Owing to the high aluminum content in dried papaya threads, these results could be expected. For children (3–6 years), the

second highest P95%PTWI was 47.85% from chewing gums, followed by 32.03% from candies. A similar trend was observed in adults (19–65 years), but the exposure for adults was lower than that for children. For adults (19–65 years), the second highest P95%PTWI was 18.45% from chewing gums, followed by 12.35% from candies.

The %PTWI values of aluminum exposure from chocolate for average and high consumers among children were 17.29% and 59.15%, based on the PTWI value of 1 mg/kg bw/wk according to Stahl et al [14]. The %PTWI of the previous work converted to the PTWI value by JECFA in 2011 was 8.15 and 29.57%. Although chocolate consumption in consumers only was similar to that reported by Stahl et al [14], the corresponding %PTWI values in this work were 3.10% and 7.77%, based on the PTWI value by the 2011 JECFA report. The %PTWI was lower in the present study, because the aluminum content was lower in our chocolate samples. The %PTWI values for aluminum exposure from candies for average consumers among children, according to Stahl et al [14] and the present work, were 1.91% and 6.02% based on the 2011 JECFA reported PTWI value. The higher %PTWI was due to the higher aluminum content in our candy samples.

The %PTWI values of aluminum exposure for children in Australia, France, and UK, according to the PTWI established by the JECFA in 2011, were 15.4%, 21.8%, and 43.1%,

Table 5 – Estimated weekly exposure to aluminum (mg/kg bw/wk) for adults (19–50 years) and children (3–6 years) based on the consumption rate in consumers only.

Age group	Type	Mean Al (mg/kg)	P50 CR ^a (g/d)	P95 CR ^b (g/d)	P50 EWI ^c (mg/kg bw/wk)	P95 EWI ^d (mg/kg bw/wk)	P50%PTWI ^e (%)	P95%PTWI ^f (%)
3–6 y	Candy ^g	24.5	14.43	76.80	0.12	0.64	6.02	32.03
	Chewing gum ^g	36.6	14.43	76.80	0.18	0.96	8.99	47.85
	Chocolate ^h	9.1	20.04	50.15	0.06	0.16	3.10	7.77
	Snack ⁱ	7.8	12.05	83.38	0.03	0.22	1.60	11.07
	Jelly ^j	2.3	105.04	354.36	0.08	0.28	4.11	13.87
	Dried mango ^k	2.3	3.94	10.08	0.00	0.01	0.15	0.39
	Raisin ^l	15.4	9.91	37.45	0.05	0.20	2.60	9.82
19–65 y	Dried papaya thread ^m	683.9	34.05	55.30	7.93	12.88	396.38	643.87
	Candy ^g	24.5	14.44	90.79	0.04	0.25	1.96	12.35
	Chewing gum ^g	36.6	14.44	90.79	0.06	0.37	2.93	18.45
	Chocolate ^h	9.1	42.89	187.98	0.04	0.19	2.17	9.50
	Snack ⁱ	7.8	14.76	80.69	0.01	0.07	0.64	3.49
	Jelly ^j	2.3	84.58	442.89	0.02	0.11	1.08	5.65
	Dried mango ^k	2.3	12.41	51.52	0.00	0.01	0.16	0.66
	Raisin ^l	15.4	8.51	64.01	0.01	0.11	0.73	5.47
	Dried papaya thread ^m	683.9	55.05	82.50	4.18	6.26	208.99	313.21

Al = aluminum; bw = body weight; CR = consumption rate; EWI = estimated weekly intake; NFCD = National Food Consumption Database; P50 = 50th percentile; P95 = 95th percentile; PTWI = provisional tolerable weekly intake; %PTWI = percent provisional tolerable weekly intake.

^a Fifty percentile of CR in consumers only.

^b Ninety-five percentile of CR in consumers only.

^c P50 EWI = Mean Al × P50 CR × 7/bw/1000.

^d P95 EWI = Mean Al × P95 CR × 7/bw/1000.

^e P50%PTWI = (P50 EWI/PTWI) × 100.

^f P95%PTWI = (P95 EWI/PTWI) × 100.

^g Taiwan NFCD food item category K201.

^h Taiwan NFCD food item category K202.

ⁱ Taiwan NFCD food item category K303.

^j Taiwan NFCD food item category K304.

^k Taiwan NFCD food item category I402.

^l Taiwan NFCD food item category I202.

^m Taiwan NFCD food item category I102.

respectively [41]. The respective %PTWI values for children in Japan [9] and Shenzhen China [42] were 43.1% and 163.6%, respectively. The %PTWI of aluminum exposure for children in Asian countries was, on average, higher than that in western countries.

4. Conclusion

From the result of the present work, the aluminum content of some candies and snacks could expose children to a high level of aluminum. In the present study, for consumers only, the P50 and P95 %PTWI values for consumption of dried papaya threads were 396.38 and 643.87 in children and 208.99 and 313.21 in adults, according to the 2011 JECFA reported PTWI value.

Children were at a higher risk of exposure to aluminum from candies and snacks compared with adults. To protect children from high aluminum exposure and as a health precautionary measure, the level of aluminum in some products should be reduced and regulated in Taiwan. Aluminum sulfate was commonly employed as a hardening or firming agent in the production of candied fruits. A hardening or firming agent without aluminum could be employed as an alternative for the manufacturing process.

5. Conflicts of interest

The authors declare that they have no conflicts of interest.

Acknowledgments

This project was sponsored financially by the Department of Health in Taiwan (MOHW104-FDA-F-113-000324).

REFERENCES

- [1] Yokel RA, Florence RL. Aluminum bioavailability from the approved food additive leavening agent acidic sodium aluminum phosphate, incorporated into a baked good, is lower than from water. *Toxicology* 2006;227:86–93.
- [2] Sedman A. Aluminum toxicity in childhood. *Pediatr Nephrol* 1992;6:383–93.
- [3] Bondy SC. Prolonged exposure to low levels of aluminum leads to changes associated with brain aging and neurodegeneration. *Toxicology* 2014;315:1–7.
- [4] Krewski D, Yokel RA, Nieboer E, Borchelt D, Cohen J, Harry J, Kacew S, Lindsay J, Mahfouz AM, Rondeau V. Human health

- risk assessment for aluminium, aluminium oxide, and aluminium hydroxide. *J Toxicol Environ Health Part B* 2007;10:1–269.
- [5] Willhite CC, Karyakina NA, Yokel RA, Yenugadhathi N, Wisniewski TM, Arnold IMF, Momoli F, Krewski D. Systematic review of potential health risks posed by pharmaceutical, occupational and consumer exposures to metallic and nanoscale aluminum, aluminum oxides, aluminum hydroxide and its soluble salts. *Crit Rev Toxicol* 2014;44(Suppl. 4):1–80.
- [6] European Food Safety Authority (EFSA). Safety of aluminium from dietary intake [1]-scientific opinion of the panel on food additives, flavourings, processing aids and food contact materials (AFC). *EFSA J* 2008;754:1–34.
- [7] Food and Agriculture Organization/World Health Organization (FAO/WHO). Evaluation of certain food additives and contaminants. Seventy-fourth report of the Joint FAO/WHO Expert Committee on Food Additives. Introduction. WHO technical report series 966. Geneva: World Health Organization; 2011. p. 1.
- [8] Food and Agriculture Organization/World Health Organization (FAO/WHO). CODEX STAN 192–1995 (Rev. 2015) general standard for food additives. Available from: http://www.fao.org/input/download/standards/4/CXS_192_2015e.pdf.
- [9] Sato K, Suzuki I, Kubota H, Furusho N, Inoue T, Yasukouchi Y, Akiyama H. Estimation of daily aluminum intake in Japan based on food consumption inspection results: impact of food additives. *Food Sci Nutr* 2014;2:389–97.
- [10] Crisponi G, Fanni D, Gerosa C, Nemolato S, Nurchi VM, Crespo-Alonso M, Lachowicz JI, Faa G. The meaning of aluminium exposure on human health and aluminium-related diseases. *Biomol Concepts* 2013;4:77–87.
- [11] Guo J, Peng S, Tian M, Wang L, Chen B, Wu M, He G. Dietary exposure to aluminium from wheat flour and puffed products of residents in Shanghai, China. *Food Addit Contam Part A* 2015;32:2018–26.
- [12] Chen H-C, Huang H-L, Chen S-C, Lu W-L, Tung C-H, Shiau H-W, Wang J-Y, Huang H-W, Shyu J-F, Feng R-L, Chiang Y-M. Border inspections of imported food and related products in Taiwan from 2011 to 2013. *J Food Drug Anal* 2015;23:161–3.
- [13] Konieczka P, Namiesnik J. Quality assurance and quality control in the analytical chemical laboratory. New York: CRC Press; 2009.
- [14] Stahl T, Taschan H, Brunh H. Aluminium content of selected foods and food products. *Environ Sci Eur* 2011;23:37.
- [15] Hsieh DPH, Huang HY, Ling MP, Chen YS, Huang LL, Wu CH, Ni SP, Hung HC, Chiang CF. Total dietary studies and food safety assessment in Taiwan—food preservatives as an illustration. *J Food Drug Anal* 2012;20:744–63.
- [16] Bratakos SM, Lazou AE, Bratakos MS, Lazos ES. Aluminium in food and daily dietary intake estimate in Greece. *Food Addit Contam Part B Surveill Commun* 2012;5:33–44.
- [17] Altundag H, Tuzen M. Comparison of dry, wet and microwave digestion methods for the multi element determination in some dried fruit samples by ICP-OES. *Food Chem Toxicol* 2011;49:2800–7.
- [18] González-Weller D, Gutiérrez AJ, Rubio C, Revert C, Hardisson A. Dietary intake of aluminum in a Spanish population (Canary Islands). *J Agric Food Chem* 2010;58:10452–7.
- [19] Ekholm P, Reinivuo H, Mattila P, Pakkala H, Koponen J, Happonen A, Hellström J, Ovaskainen M-L. Changes in the mineral and trace element contents of cereals, fruits and vegetables in Finland. *J Food Compos Anal* 2007;20(6):487–95.
- [20] Tóth IV, Rangel AOSS, Santos JLM, Lima JLFC. Determination of aluminum(III) in crystallized fruit samples using a multicommutated flow system. *J Agric Food Chem* 2004;52:2450–4.
- [21] Chen M-C, Sun L, Lin S-Y. Risk evaluation of pollution level of aluminum in glace fruit. *Food Nutr China* 2010;10:13–5 [In Chinese, English abstract].
- [22] Jiang Q, Wang J, Li M, Liang X, Dai G, Hu Z, Wen J, Huang Q, Zhang Y. Dietary exposure to aluminium of urban residents from cities in South China. *Food Addit Contam Part A Chem Anal Control Expo Risk Assess* 2013;30:698–704.
- [23] Li Q, Liu S-J, Fang C-G. Research progress on the content and harm of aluminum in food. *J Food Saf Qual* 2016;7:14–9 [In Chinese, English abstract].
- [24] European Commission. Regulation (EU) no 380/2012 of 3 May 2012 amending Annex II to Regulation (EC) no 1333/2008 of the European Parliament and of the council as regards the conditions of use and the use levels for aluminium-containing food additives. *Official J Eur Union* 2012;L 119:14–38.
- [25] Viñas P, Campillo N, López García I, Hernández Córdoba M. Determination of aluminium in chewing gum samples using electrothermal atomic-absorption spectrometry and slurry sample introduction. *Fresenius J Anal Chem* 1995;351:695–6.
- [26] Kupchella L, Syty A. Determination of nickel, manganese, copper, and aluminum in chewing gum by nonflame atomic absorption spectrometry. *J Agric Food Chem* 1980;28:1035–6.
- [27] Lione A, Smith JC. The mobilization of aluminium from three brands of chewing gum. *Food Chem Toxicol* 1982;20:945–6.
- [28] Marín-Martínez R, Barber X, Cabrera-Vique C, Carbonell-Barrachina ÁA, Vilanova E, García-Hernández VM, Roche E, García-García E. Aluminium, nickel, cadmium and lead in candy products and assessment of daily intake by children in Spain. *Food Addit Contam Part B* 2016;9(1):66–71.
- [29] Yang Y, Zhang J, Zhao S, Shao B. Simultaneous determination of five aluminum lake dyes in coated candy by ultra performance liquid chromatography. *Chin J Food Hyg* 2013;25:148–51 [In Chinese, English abstract].
- [30] Fekete V, Vandevijvere S, Bolle F, Van Looc J. Estimation of dietary aluminum exposure of the Belgian adult population: evaluation of contribution of food and kitchenware. *Food Chem Toxicol* 2013;55:602–8.
- [31] Wong WW, Chung SW, Kwong KP, Yin Ho Y, Xiao Y. Dietary exposure to aluminium of the Hong Kong population. *Food Addit Contam Part A Chem Anal Control Expo Risk Assess* 2010;27:457–63.
- [32] Müller M, Anke M, Illing-Günther H. Aluminium in foodstuffs. *Food Chem* 1998;61:419–28.
- [33] Schäfer U, Seifert M. Oral intake of aluminum from foodstuffs, food additives, food packaging, cookware and pharmaceutical preparations with respect to dietary regulations. *Trace Elem Electrolytes* 2006;23:150–61.
- [34] Jalbani N, Kazi TG, Khan Jamali M, Arain MB, Afridi HI, Sheerazi ST, Ansari R. Application of fractional factorial design and Doehlert matrix in the optimization of experimental variables associated with the ultrasonic-assisted acid digestion of chocolate samples for aluminum determination by atomic absorption spectrometry. *J AOAC Int* 2007;90:1682–8.
- [35] Millour S, Noël L, Kadar A, Chekri R, Vastel C, Sirot V, Leblanc JC, Guérin T. Pb, Hg, Cd, As, Sb and Al levels in foodstuffs from the 2nd French total diet study. *Food Chem* 2011;126:1787–99.
- [36] European Food Safety Authority (EFSA). Refined exposure assessment for Quinoline Yellow (E 104). *EFSA J* 2015;13(4070):33.
- [37] Jensen BH, Andersen JH, Petersen A, Christensen T. Dietary exposure assessment of Danish consumers to

- dithiocarbamate residues in food: a comparison of the deterministic and probabilistic approach. *Food Addit Contam Part A Chem Anal Control Expo Risk Assess* 2008;25:714–21.
- [38] Boorman JL, Baines J, Hambridge TL, Abbey JL. Dietary exposure assessment in a total diet study. In: Moy GG, Vannoort WR, editors. *Total diet studies*. New York, NY: Springer; 2013. p. 179–90.
- [39] National Health Research Institutes (NHRI). National Food Consumption Database. Available from: <http://intakes.nhri.org.tw/food-intake-data/2014>. [Accessed 07 Sep 2015].
- [40] World Health Organization (WHO). Principles and methods for the risk assessment of chemicals in food. Chapter 6 dietary exposure assessment of chemicals in food. *Environ Health Criteria* 2009;240:19–21.
- [41] Arnich N, Sirot V, Rivière G, Jean J, Noël L, Guérin T, Leblanc JC. Dietary exposure to trace elements and health risk assessment in the 2nd French Total Diet Study. *Food Chem Toxicol* 2012;50:2432–49.
- [42] Yang M, Jiang L, Huang H, Zeng S, Qiu F, Yu M, Li X, Wei S. Dietary exposure to aluminium and health risk assessment in the residents of Shenzhen, China. *PLoS One* 2014;9:1–8.