

Risk assessment of daily intakes of artificial colour additives in food commonly consumed in Korea

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Summary

The aim of this study was to determine the artificial colours in foods and evaluate the dietary intake of artificial colour additives in foods commonly consumed by different age groups in Korea. The content level of artificial food colour was experimentally determined by high performance liquid chromatography with a photodiode array detector. Of the 643 food items analysed, 503 (approximately 78%) contained artificial colorants. Consumers aged 13–19 years displayed the greatest consumption of colour additive-containing foods, but the amount was much lower than the acceptable daily intakes (ADI) established by FAO/WHO Joint Expert Committee on Food Additives (JEFCA). Both mean and high (95th percentile) intakes of permitted artificial colour additives for all population groups were markedly lower than the ADI, because the Korean food industry has widely substituted natural colour additives for artificial colorants. However, the issue of consumption of colour additives by targeted groups such as children is of concern, so further studies to provide data on dietary intake of artificial colour additives on such vulnerable groups are needed.

Keywords

dietary intake; artificial colour; assessment; risk; Korea

Colour additives are frequently used in food-stuffs to improve appearance [1]. Natural colours have several demerits such as a high price, extraction difficulty and discolouration during processing. On the other hand, artificial colours are inexpensive and are superior to natural extracts specifically in tinctorial strength, hue and stability [2]. Therefore, although consumer awareness of health-related risks of artificial colour additives has increased, artificial colours are used more frequently than natural colours in many processed foods [1, 3]. Food additives such as artificial colours have increasingly come under investigation for evaluation of their safety [4]. In general, the amount of artificial colour used in processed foods is less than 500 mg·kg⁻¹ (with some exceptions) [5, 6]. Artificial colours that are allowed by many countries for use as additives have proven safety through toxicity tests. On the contrary, some artificial colours have been prohibited due to their toxicity or carcinogenicity [2].

The number of artificial colours permitted differs by country. For instance, FAO/WHO Codex Alimentarius permits 14 artificial colours, European Union (EU) 15 colours, Japan 12 colours, USA 9 colours, and Korea 9 colours (Tab. 1). Some regulations, such as EU directives and Codex Alimentarius, regulate not only which artificial colours are permitted but also the maximum permitted levels [5, 6]. Korea, on the other hand, has only established the artificial colours permitted for use. The Korea Food and Drug Administration (KFDA), a Korean government regulatory agency, conducts colour tests in food products to understand the appropriateness of colour usage. But these tests provide only qualitative data on artificial colours in foods and do not provide information on their levels in foods. Because colour is one of the important factors determining consumer preference, the usage of artificial colour additives may be increased to attract consumers. Thus, not only qualitative information on colour additives in

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Tab. 1. Regulations for permitted artificial colours in Korea, European Union, USA, Codex Alimentarius and Japan.

Artificial colours	Synonyms	E number	Korea [18]	EU [5]	USA [19]	CA [6]	Japan [20]
Tartrazine	FD & C Yellow No. 5	E102	+	+	+	+	+
Sunset Yellow FCF	FD & C Yellow No. 6	E110	+	+	+	+	+
Amaranth	FD & C Red No. 2	E123	+	+	–	+	+
Erythrosine	FD & C Red No. 3	E127	+	+	+	+	+
Allura Red AC	FD & C Red No. 40	E129	+	+	+	+	+
Ponceau 4R	Cochineal Red A	E124	+	+	–	+	+
Fast Green FCF	FD & C Green No. 3	–	+	–	+	+	+
Brilliant Blue FCF	FD & C Blue No. 1	E133	+	+	+	+	+
Indigotine	Indigocarmine	E132	+	+	+	+	+
Quinoline Yellow	Quinoline Yellow	E104	–	+	–	+	–
Patent Blue V	Patent Blue	E131	–	+	–	–	–
Green S	Green S	E142	–	+	–	–	–
Phloxine	Phloxine	–	–	–	–	–	+
Rose Bengal	Rose Bengal	–	–	–	–	–	+
Acid Red	Acid Red	–	–	–	–	–	+
Red 2G	Red 2G	E128	–	–	–	+	–
Orange BI	Orange B	–	–	–	+	–	–
Brilliant Black	Black PN	E151	–	+	–	+	–
Azo Rubine	Carmoisine	E122	–	+	–	+	–
Brown HT	Chocolate brown HT	E155	–	+	–	+	–
Brown FK	Brown FK	E154	–	+	–	–	–
Citrus Red No. 2	Citrus Red No. 2	–	–	–	+	–	–

CA – Codex Alimentarius. + – permitted, – – not permitted.

food products but also data on the exact amounts of colour additives in foods should be collected. Therefore, it is imperative that artificial colour contents in foods should be analysed and their safety in diet evaluated.

In this regard, this study experimentally determined content of nine artificial colours permitted in Korea in foods, namely, Tartrazine, Sunset Yellow, Amaranth, Erythrosine, Allura Red, Ponceau 4R, Brilliant Blue, Indigotine and Fast Green, and assessed the estimated daily intake of artificial colour additives in foods commonly consumed by Koreans.

MATERIALS AND METHODS

Consumption data of coloured foods

Food consumption data for estimating of artificial colorant intakes were obtained from the Korea Health and Nutrition Survey [7]. A total of 25 628 subjects older than 1 year of age were

selected randomly nationwide for the survey and were surveyed individually. The dietary intake survey involved a 1 day, 24 h dietary recall. Foodstuffs containing colours were grouped to 10 categories among all food items in the survey: bakery products, snacks, candies, chocolates, chewing gums, jam, ice cream, drinks, salted vegetables (salted cucumber, salted eggplant, etc.) and alcoholic beverages. Average intake of each of 10 food categories consumed by different age group was collected to calculate the dietary intake of artificial colour in foods (Tab. 2).

Analysis

A total of 10 food categories comprising 643 foodstuffs were selected as commonly consumed source of artificial colorants in the Korean diet. All foodstuffs were purchased nationwide, including imported food. Purchased samples were stored at various temperatures depending on the foodstuff until tested; for instance, ice creams were stored at –20 °C, perishable foods at 4 °C and dried foodstuffs at room temperature. These

Tab. 2. Consumption data for food in which artificial colorants are used, for average and high consumers in Korea.

Food category	Total	Average consumption [g·d ⁻¹]								High consumption [g·d ⁻¹]	
		Consumers age [yr]								90 th percentile	95 th percentile
		1-2	3-6	7-12	13-19	20-29	30-49	50-64	> 65		
Bakery products	2.4906	2.9369	3.4159	2.7392	11.6197	1.7994	1.9115	1.9482	0.6817	174.0	217.9
Snacks	2.6088	2.4298	3.7374	4.5007	11.7872	1.3655	0.5626	0.2820	0.0000	90.0	98.5
Candies	0.5519	1.1224	0.7524	0.8040	0.9447	0.1504	0.0799	0.0948	0.2024	35.0	54.6
Chocolates	0.2585	0.2950	0.5642	0.3435	0.2461	0.0675	0.0105	0.0095	0.0000	65.0	80.0
Chewing gum	0.0341	0.0812	0.0528	0.0366	0.0555	0.0042	0.0049	0.0008	0.0000	9.3	15.0
Jams	0.1892	0.2637	0.2037	0.2538	0.7400	0.1014	0.0839	0.0329	0.0206	38.4	42.7
Ice cream	5.4274	4.6946	7.6474	10.0741	22.7754	3.6812	1.0913	0.5965	0.1257	165.0	180.0
Drinks	20.8137	16.4645	22.3738	24.6426	90.9357	26.6110	10.8159	6.6379	4.5436	340.8	433.1
Salted vegetables	2.3521	0.7709	1.7808	1.8743	10.3583	4.1901	2.0743	0.8888	0.5986	60.2	80.2
Alcoholic beverages	7.0284	0.0000	0.0000	0.0000	8.0667	8.8284	11.7536	10.9084	6.7399	426.6	682.6

samples were analysed to determine the content of 9 artificial food colours permitted as colour additives in Korea. Colour standards for these colour additives were purchased from Kasei (Tokyo, Japan). Hydrochloric acid and ammonium acetate were obtained from Sigma-Aldrich (St. Louis, Missouri, USA). Methanol, acetonitrile and water were of high performance liquid chromatography (HPLC) grade (Merck, Darmstadt, Germany). Each colour standard was prepared as a 100 $\mu\text{g}\cdot\text{ml}^{-1}$ stock solution in water. Mixture of each solution was adjusted to 0, 1, 2, 10, 50, and 100 $\mu\text{g}\cdot\text{ml}^{-1}$ with 0.01 $\text{mol}\cdot\text{l}^{-1}$ ammonium acetate to form the working standards. Purity of each colour standard was determined by scanning and measuring on a UV-VIS spectrophotometer (Shimadzu, Japan) as described by WALFORD [8]. The working standard solutions were individually filtered through a 0.45 μm membrane (Millipore, Bedford, Massachusetts, USA) pre-wetted with 10ml of methanol and 10ml of 0.1% tetra-butyl ammonium phosphate (TBAP), stored in a dark and cool space (at 4 °C in a refrigerator) and used within 1 week. Ten microlitres of each working standard was injected into the HPLC system. The mobile phase containing 0.01 $\text{mol}\cdot\text{l}^{-1}$ ammonium acetate was used.

Extraction and clean-up

Artificial colours from foods were extracted with 0.01 $\text{mol}\cdot\text{l}^{-1}$ ammonium acetate solution that was slightly modified solution from that described previously [9]. Samples of 5–10g were weighed in a 50ml glass tube. The samples were thoroughly extracted for 10 min in an ultrasonic bath (Whasin, Seoul, Korea) with the ammonium acetate solution. In cases of solid food, the sample was ground or cut into approximately 5mm pieces before extraction. Fat-containing samples were treated three times with 50ml petroleum ether to remove the fat prior to extraction. Each sample was subjected to three extractions with 20ml of 0.01 $\text{mol}\cdot\text{l}^{-1}$ ammonium acetate solution until the sample was colour-free. Candies were extracted with 20ml distilled water. All extracts were evaporated and cleaned up by filtration through a 0.45 μm membrane filter (Millipore) [10]. The membrane filter was pre-wetted with 10ml of methanol and 10ml of 0.1% TBAP. All treated samples were analysed by HPLC system with photodiode array detection. The HPLC system consisted of a Nanospace SI-2 separation module (Shiseido, Tokyo, Japan) equipped with Nanospace SI-2 photodiode array detector (PDA; Shiseido) controlled by Millennium32 chromatography manager data acquisition system (Shisei-

Tab. 3. HPLC conditions for analysis of nine artificial colour additives.

HPLC system	Micro HPLC		
Column	Capcell-Pak C ₁₈ (250 mm x 4.6 mm, 5 μ m)		
Gradient program	Time [min]	Solvent A	Solvent B
	0	95	5
	15	50	50
	20	95	5
	25	95	5
	30	Stop	–
Injection volume	10 μ l		
Detector	Photodiode array		
Wavelengths for testing of each food colour	420 nm	Tartrazine, Sunset Yellow	
	520 nm	Amaranth, Erythrosine, Allura Red, Ponceau 4R	
	620 nm	Brilliant Blue, Indigotine, Fast Green	

Solvent A – 0.01 mol·l⁻¹ ammonium acetate; solvent B – acetonitrile.

do). The column used for the separation conducted at room temperature (20 °C) was a Capcell-Pak C₁₈ Column (250 mm × 4.6 mm, 5 μ m; Shiseido). The flow rate was 1000 μ l·min⁻¹. The detector wavelengths were set based on the maximum absorption wavelength of each food colour: 420 nm for Tartrazine and Sunset Yellow; 520 nm for Amaranth, Erythrosine, Allura Red and Ponceau 4R; and 620 nm for Brilliant Blue, Indigotine and Fast Green. HPLC conditions are summarized in Tab. 3. All samples and standards were run in duplicate and the retention times were 4.3 min for Tartrazine, 9.9 min for Sunset Yellow, 5.6 min for Amaranth, 21.5 min for Erythrosine, 12.0 min for Allura Red, 9.4 min for Ponceau 4R, 18.5 min for Brilliant Blue, 6.3 min for Indogotine and 17.6 min for Fast Green.

Quality control

In order to verify the precision and linearity of HPLC separation, calibration was carried out using external standards by 10 measurements of a standard solution containing 9 artificial food colours. Recovery was determined for each food category by spiking at a level of 10 mg·kg⁻¹ with 9 artificial colorants. All samples were analysed in triplicate. Mean recoveries of Tartrazine, Sunset Yellow, Amaranth, Erythrosine, Allura Red, Ponceau 4R, Brilliant Blue, Indigotine and Fast Green were 84.9–100.9%, 94.7–98.3%, 87.4–100.4%, 85.1–97.7%, 91.7–99.1%, 87.5–98.1%, 92.3–102.1%, 79.0–95.4%, and 90.0–101.3%, respectively. The recoveries from each food category of the 9 artificial colorants are shown in Tab. 4. The detection limits of each colorant were determined by direct injection of standard solution. Detection limits were calculated

from calibration curve ($y = ax + b$) by multiplying 2 by standard deviation (SD) of the b/a , and quantification limits were calculated by multiplying 10 by SD of b/a . The detection limits for Tartrazine, Sunset Yellow, Amaranth, Erythrosine, Allura Red, Ponceau 4R, Brilliant Blue, Indigotine and Fast Green were 0.003, 0.008, 0.004, 0.003, 0.003, 0.007, 0.001, 0.002 and 0.002 mg·kg⁻¹, respectively.

Assessment of daily intakes of colour additives

The daily intakes of the colour additives were calculated by coupling content of 9 artificial food colours from 10 food categories and individual food consumption data for average consumers by age group and 95th percentile high consumers. The contents of colorants from each food category were determined by analysis of 10 food categories and 643 food items, comprising 30 samples of bakery products, 54 samples of snacks, 309 samples of candies, 40 samples of chocolates, 56 samples of chewing gums, 3 samples of jam, 46 samples of ice creams, 46 samples of salted vegetables, 46 samples of drinks and 13 samples of alcoholic beverages. Overall mean content of the 9 artificial colours in each food category were calculated from all sample values for estimating the colorant intakes. Non-detected concentration of colorants was considered as “0”. Average food consumption data per food category for total population and age group were obtained from the 2001 Korea Health and Nutrition Survey [7]. The 95th percentile consumption data of the population were obtained from the same survey to estimate intake of colorants by high consumers as well. The calculation methods of food colour intakes were very similar to those reported previously [11]. The calculated colorant intakes were adjusted

Tab. 4. Recoveries of artificial colour additives from food items.

Artificial colour	Recovery [%]									
	Bakery products	Snacks	Candies	Chocolates	Chewing gum	Jams	Ice cream	Drinks	Salted vegetables	Alcoholic beverages
Tartrazine	94.5 ± 0.3	84.9 ± 1.5	95.2 ± 0.6	100.2 ± 1.5	96.2 ± 0.1	94.2 ± 0.3	100.2 ± 2.0	100.9 ± 0.1	96.3 ± 1.1	96.9 ± 1.0
Sunset Yellow	97.2 ± 0.7	95.3 ± 2.7	94.7 ± 1.1	98.0 ± 0.9	97.0 ± 0.4	95.0 ± 0.7	98.3 ± 0.5	97.2 ± 2.6	94.9 ± 3.5	97.6 ± 0.3
Amaranth	90.5 ± 0.1	87.4 ± 2.2	90.5 ± 1.0	97.9 ± 1.6	99.1 ± 0.5	97.6 ± 1.5	97.2 ± 1.0	98.6 ± 1.7	92.7 ± 0.9	100.4 ± 2.4
Erythrosine	88.7 ± 2.4	85.1 ± 3.0	92.2 ± 0.9	94.5 ± 2.5	97.7 ± 0.7	92.5 ± 0.3	91.1 ± 1.4	97.2 ± 2.6	96.2 ± 2.7	95.2 ± 0.9
Allura Red	92.8 ± 0.1	92.3 ± 1.8	91.7 ± 0.3	96.6 ± 0.2	95.1 ± 0.6	96.8 ± 0.8	97.4 ± 0.5	99.1 ± 1.7	94.3 ± 1.2	96.5 ± 2.0
Ponceau 4R	87.5 ± 0.1	88.4 ± 1.5	93.7 ± 0.2	94.2 ± 0.3	92.3 ± 0.5	94.5 ± 1.2	94.4 ± 0.3	98.1 ± 1.5	97.1 ± 2.3	97.2 ± 2.1
Brilliant Blue	93.5 ± 1.7	92.3 ± 1.6	95.9 ± 2.5	93.4 ± 1.6	98.4 ± 1.5	95.6 ± 2.3	96.7 ± 1.3	97.4 ± 3.0	95.0 ± 1.9	102.1 ± 1.3
Indigotine	85.2 ± 4.6	89.4 ± 0.5	83.5 ± 0.5	79.0 ± 3.2	92.0 ± 0.3	88.6 ± 1.7	88.7 ± 2.1	95.1 ± 3.3	85.9 ± 2.4	95.4 ± 1.1
Fast Green	92.7 ± 0.8	90.0 ± 0.8	93.8 ± 1.0	94.9 ± 0.9	95.2 ± 1.2	93.1 ± 1.6	94.8 ± 1.1	101.1 ± 2.5	98.4 ± 3.0	101.3 ± 2.1

Values represent means ± standard deviation. Spike levels were 10 mg·kg⁻¹.

to the standard body weight of the Korea Health and Nutrition Survey – Health examination part [12] for total population and age group. To evaluate the daily intakes of artificial food colours, the colorant intakes adjusted to the body weight were compared with acceptable daily intake (ADI) of each colorant established by the FAO/WHO Joint Expert Committee on Food Additives (JECFA).

RESULTS AND DISCUSSION

Content of artificial colour additives

Among the 643 food items analysed, 503 samples (approximately 78%) contained the artificial colorants Tartrazine, Sunset Yellow, Amaranth, Erythrosine, Allura Red, Ponceau 4R, Brilliant Blue, Indigotine or Fast Green, which are permitted in Korea. One hundred forty samples (approximately 22%) did not contain any artificial colorant. Fig. 1 shows the distribution of the artificial colorants most frequently detected above limit of quantification (*LOQ*) in the food categories analysed. Tartrazine (23.3%) was the most commonly used artificial colorant, followed by Brilliant Blue (19.1%), Allura Red (18.2%) and Sunset Yellow (12.1%). The results are similar to those of a study conducted in Hong Kong [13], which reported that Tartrazine (27.5%) and Sunset Yellow (24.0%) were the most commonly used synthetic colorants, reported in two other studies [9, 14].

Tab. 5 shows the contents of 9 artificial colorants determined in food samples that are most commonly consumed in Korea. Of the coloured food items analysed, the most artificial colours were detected in candies and chocolates. Artificial colours in candies were detected in the range of 3.0–190.3 mg·kg⁻¹ and the content of Sunset Yellow

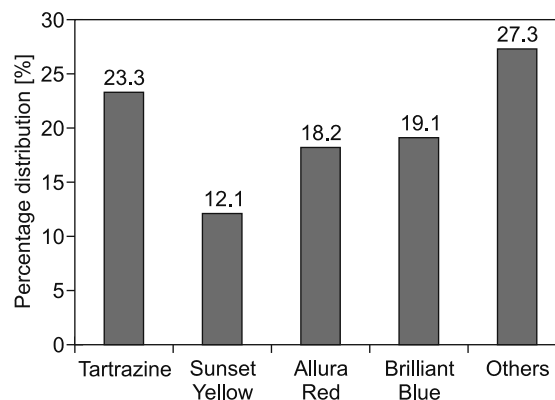


Fig. 1. Percentage distribution of artificial colorants in the food categories studied.

Tab. 5. Contents of artificial colorants in various food items vs permitted levels in Codex alimentarius and EU.

Artificial Colour		Food category [mg·kg ⁻¹]									
		Bakery products	Snacks	Candies	Chocolates	Chewing gum	Jams	Ice cream	Drinks	Salted vegetables	Alcoholic beverages
Tartrazine	DR	< LOQ – 2.7	< LOQ – 38.2	< LOQ – 69.3	< LOQ – 67.6	< LOQ – 22.6	< LOQ	< LOQ – 18.1	< LOQ – 67.4	< LOQ – 60.6	< LOQ – 5.0
	CA	300	300	300	300	300	500	300	300	500	300
	EU	200	–	300	–	–	–	150	100	–	200
Sunset Yellow	DR	< LOQ	< LOQ – 21.0	< LOQ – 190.3	< LOQ – 102.5	< LOQ – 21.3	< LOQ	< LOQ – 4.4	< LOQ – 19.0	< LOQ	< LOQ
	CA	300	550	400	100	400	500	300	300	500	300
	EU	50	–	50	–	–	100	50	50	–	200
Amaranth	DR	< LOQ	< LOQ – 3.7	< LOQ – 15.6	< LOQ	< LOQ	< LOQ	< LOQ – 20.0	< LOQ	< LOQ	< LOQ – 5.1
	CA	300	300	100	100	300	300	300	100	–	300
	EU	200	–	300	–	–	–	150	100	–	200
Erythrosine	DR	< LOQ	< LOQ	< LOQ – 3.0	< LOQ – 31.7	< LOQ – 0.2	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
	CA	300	300	300	300	200	400	300	300	300	300
	EU	200	–	300	–	–	–	150	100	–	200
Allura Red	DR	< LOQ – 2.4	< LOQ – 18.2	< LOQ – 80.2	< LOQ – 77.3	< LOQ – 39.2	< LOQ – 57.4	< LOQ – 20.4	< LOQ – 63.5	< LOQ – 1.0	< LOQ – 35.4
	CA	300	200	348	300	467	500	300	300	500	300
	EU	200	–	300	–	–	–	150	100	–	200
Ponceau 4R	DR	< LOQ	< LOQ – 1.8	< LOQ – 21.4	< LOQ	< LOQ – 0.9	< LOQ	< LOQ	< LOQ – 2.3	< LOQ	< LOQ
	CA	200	200	300	150	300	500	150	100	50	200
	EU	50	–	50	–	–	50	50	50	–	50
Brilliant Blue	DR	< LOQ – 0.0	< LOQ – 5.9	< LOQ – 31.8	< LOQ – 18.0	< LOQ – 6.7	< LOQ	ND – 13.3	ND – 21.2	ND – 4.1	ND – 3.3
	CA	100	200	300	100	300	500	150	100	100	200
	EU	200	–	300	–	–	–	150	100	–	200
Indigotine	DR		< LOQ	< LOQ – 6.7	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
	CA	300	300	450	450	300	500	300	300	–	300
	EU	200	–	300	–	–	–	150	100	–	200
Fast Green	DR	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	LOQ	< LOQ – 0.8	< LOQ	< LOQ	< LOQ
	CA	100	–	100	–	300	400	100	100	–	100
	EU	–	–	–	–	–	–	–	–	–	–

DR – detected range, CA – Codex Alimentarius permitted level [6]), EU – EU permitted level [5, 21], LOQ – limit of quantification, ND – not detected.

in candies (maximum content of $190.3 \text{ mg}\cdot\text{kg}^{-1}$) was approximately four times the maximum permitted level in the EU Directive ($50 \text{ mg}\cdot\text{kg}^{-1}$). In addition, 8 artificial colorants except Fast Green were found in candies. We recognised the fact that most of the artificial colours permitted in Korea were used in candies. Artificial colours in the range of $18.0\text{--}102.5 \text{ mg}\cdot\text{kg}^{-1}$ were detected in chocolates as well. Sunset Yellow was the most detected colour in chocolates, at a maximum of $102.5 \text{ mg}\cdot\text{kg}^{-1}$, which exceeded the amount of the Codex Alimentarius maximum permitted level ($100 \text{ mg}\cdot\text{kg}^{-1}$). Furthermore, the content of Allura Red, Tartrazine, Erythrosine and Brilliant blue in chocolates did not exceed the amounts recommended by Codex Alimentarius or EU regulations, but the results showed that, compared to other food products, they were contained quite extensively. Subsequently, drinks, chewing gum and snacks were the food products in which some colorants were used. Allura Red, Tartrazine, Sunset Yellow, Brilliant Blue and Ponceau 4R in those three foods were detected in ranges of $2.3\text{--}67.4 \text{ mg}\cdot\text{kg}^{-1}$, $0.2\text{--}39.2 \text{ mg}\cdot\text{kg}^{-1}$, and $1.8\text{--}38.2 \text{ mg}\cdot\text{kg}^{-1}$, respectively. Alcoholic beverages were found to contain mainly Allura Red as a colorant at a maximum content of $35.4 \text{ mg}\cdot\text{kg}^{-1}$. Only small quantities of Tartrazine and Allura Red were detected in bakery products and no other colorants were detected. Among the 10 food categories in this study, bakery products contained the least amount of artificial colorants.

On the one hand, we recognised that Fast Green was not detected in any food products except ice cream ($0.8 \text{ mg}\cdot\text{kg}^{-1}$); thus it was the least used artificial colorant.

Analysing the above results, we found that the maximum detected amounts of Sunset Yellow in candies and chocolates were higher than the maximum permitted levels set by EU and Codex Alimentarius. This is derived from the fact that Korea has only defined what artificial colours are permitted and not the maximum permitted levels.

As a consequence, we recommend that Korean Regulatory Authorities should define not only which artificial colours are permitted in food products, but also the permitted levels of usage just as Codex Alimentarius and EU have done. This would prevent the inappropriate usage of artificial food colours through such regulation, for the benefit of health and safety of the Korean public.

Assessment of daily intakes of artificial food colours

Daily intakes of the 9 artificial colour additives for different age groups are summarized in Fig. 2. The intakes of Allura Red, Amaranth and Tartrazine presented the highest amount for consumers aged 13–19 years, representing 0.1%, 0.1% and 0.1% of FAO/WHO ADI. The highest Sunset Yellow and Erythrosine intakes were found in consumers aged 1–2 years (0.1% and 0.0% of the ADI, respectively). Red and yellow artificial colours were mainly consumed by Korean teenagers, while green and blue colours were seldom consumed by this age group, similar to previous findings [15, 16]. Also, the daily intakes of artificial colour additives were highest among those aged 13–19 and 1–2 years, but the intakes were quite safe, at $< 0.1\%$ of the particular ADI. It has been

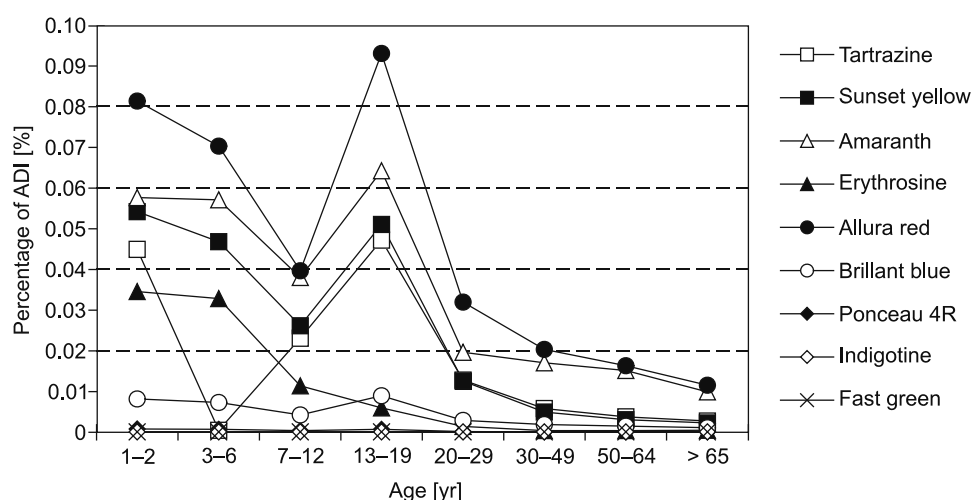


Fig. 2. The consumption (percentage of ADI) of artificial colorants in different age groups.

Tab. 6. Estimated daily intakes of artificial food colour for average and high (95th percentile) consumers in Korea.

Artificial colour	ADI (set by JECFA)		Average consumer		High consumer (95 th percentile)	
	[mg·kg ⁻¹]	Reference	Estimated intake [μg·kg ⁻¹]	Percentage of ADI	Estimated intake [μg·kg ⁻¹]	Percentage of ADI
Tartrazine	0 – 7.5	[22]	0.9102	< 0.01	61.3929	0.8
Sunset Yellow	0 – 2.5	[23]	0.3227	< 0.01	29.5746	1.2
Amaranth	0 – 0.5	[24]	0.1157	< 0.01	9.4096	1.9
Erythrosine	0 – 0.1	[25]	0.0055	< 0.01	4.2225	4.2
Allura Red	0 – 7.0	[26]	1.9790	< 0.01	129.8242	1.9
Ponceau 4R	0 – 4.0	[27]	0.0056	< 0.01	0.4873	< 0.01
Brilliant Blue	0 – 12.5	[28]	0.3336	< 0.01	23.5093	0.2
Indigotine	0 – 5.0	[29]	0.0002	< 0.01	0.0455	< 0.01
Fast Green	0 – 25	[30]	0.0019	< 0.01	0.1448	< 0.01

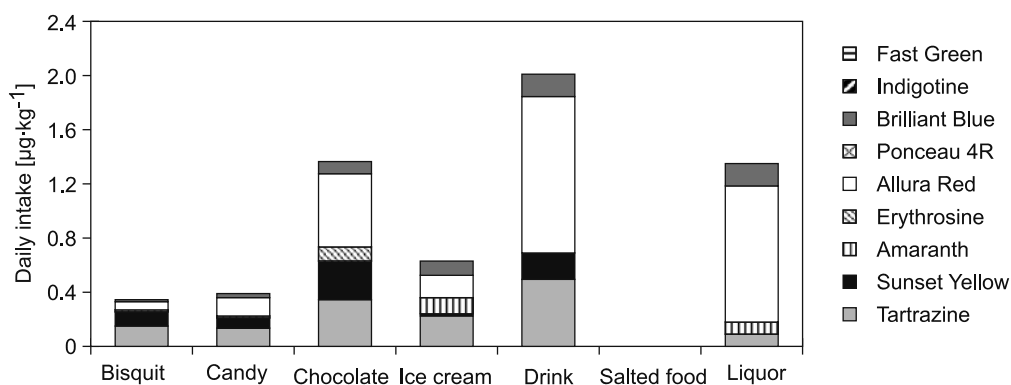
ADI and consumption is expressed per kilogram of body weight per day.

reported that, in Brazilian children, the intakes of Amaranth, Sunset Yellow and Tartrazine were relatively higher than those of the other colour additives (24%, 3% and 0.4% of the JECFA-established ADI, respectively) [15]. This consumption of artificial colorants differed appreciably from the present findings. In particular, the intake of Amaranth differed approximately 400 times between Brazil and Korea. Moreover, a study conducted with children in Kuwait aged 5–14 years reported that the consumption of Tartrazine, Sunset Yellow, Carmoisine and Allura Red exceeded ADI levels [16]. These results also differed considerably from the present findings. Although the other cited studies had a slight discrepancy in the age groups, the present findings from the Republic of Korea demonstrated the lowest consumption of the colour additives, compared with the previous studies conducted in Brazil and Kuwait. This may

reflect the fact that the food industry in Korea has widely substituted natural colour additives for artificial colours. The amount of imported artificial colour additives has gradually decreased from 138 834 kg in 2002, to 104 611 kg in 2003 and 91 798 kg in 2004 [17].

The daily intakes of each artificial food colour for average and high (95th percentile) consumers are presented in Tab. 6. Intakes for average consumers ranged from 0.0002 μg·kg⁻¹ to 2 μg·kg⁻¹ of body weight per day, corresponding to a range of < 0.01% of their ADI, as determined by JECFA. Allura Red was the highest consumed colour among the nine tested artificial food colours, followed by Amaranth, Sunset Yellow and Tartrazine (< 0.01% of the ADI).

The major contributing foods for average colour consumption were drinks, ice creams, liquors, biscuits and candies (Fig. 3). Although

**Fig. 3.** Contribution to consumption of artificial colour additives by an average consumer in different food categories.

Daily intake is expressed per kilogram of body weight per day.

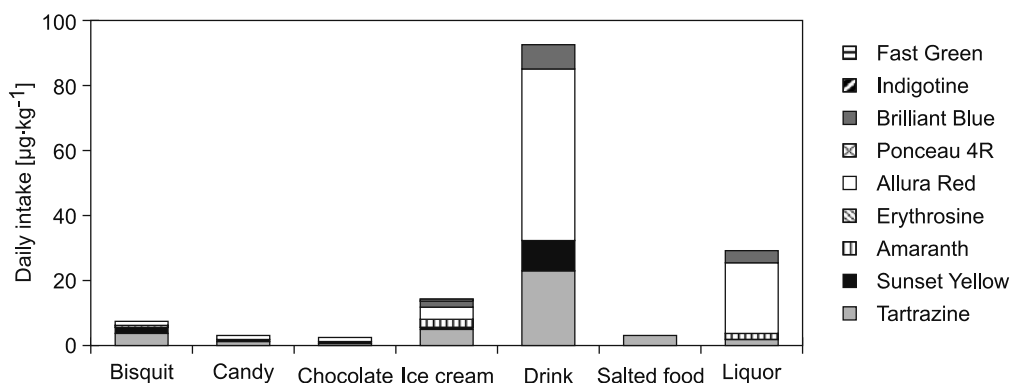


Fig. 4. Contribution to consumption of artificial colour additives by a high consumer in different food categories.

Daily intake is expressed per kilogram of body weight per day.

chocolates contained the highest levels of certain artificial food colours such as Tartrazine, Sunset Yellow, Erythrosine, Allura Red and Brilliant Blue (Tab. 5), the daily intake of each artificial colour from chocolates was not higher than with other samples, due to the low intake of chocolates among the surveyed consumers. Similar to the present findings, a Hong Kong-based study [13] reported that many children were frequently exposed to drink, ice cream and sweets with artificial colours, while the mean intakes of artificial food colours were below ADI.

The intake of colour additives for high, 95th percentile consumers ranged from 0.05 µg·kg⁻¹ to 129.8 µg·kg⁻¹ of body weight per day, and ranged from < 0.1% to 4.2% of the ADI set by JECFA (Tab. 6). Erythrosine was the most consumed colour additive (4.2% of ADI), followed by Amaranth, Allura Red and Sunset Yellow (1.9%, 1.9% and 1.2% of ADI, respectively). Nevertheless, the amount of colour additives for the high consumer group in Korea still constituted no more than 5% of ADI. The major contributing foods for high colour consumption were drinks, chocolates, liquors, ice cream, candies and biscuits (Fig. 4).

This study was to assess the dietary intake of artificial colour additives by different age groups for an average consumer and high consumer in Korea. The intake amounts of colour additives for both the average consumer and the high consumer in Korea are lower than ADI. As a conclusion, the intake levels of artificial colorants in Korea are safe.

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REFERENCES

1. Rowe, K. S. – Rowe, K. J.: Artificial food colouring and behaviour – a dose response effect in a double-blind placebo controlled repeated measures study. *Journal of Pediatrics*, 125, 1994, pp. 691–698.
2. Newsome, R. L.: Natural and artificial colouring agents. In: Food additives. Branen, L. A. – Davidson, P. M. – Salminen, S. (Ed.): New York: Marcel Dekker, 1990, pp. 328–345. ISBN 0-8247-8046-9.
3. Yoshioka, N. – Ichihashi, K.: Determination of 40 synthetic food colors in drinks and candies by high-performance liquid chromatography using a short column with photodiode array detection. *Talanta*, 74, 2008, pp. 1408–1413.
4. Evaluation of certain food additives. 21st report of the Joint FAO/WHO Expert Committee on Food Additives. WHO technical report series No. 617. Geneva: World Health Organization, 1978. 41 pp. ISBN 92-4-120617-9.
5. European Parliament and Council Directive No 94/36/EC of 30 June 1994 on colours for use in foodstuffs. *Official Journal of the European Communities*, L237, 1994, pp. 13–28.
6. Codex STAN 192-1995. Codex general standard for food additives (GSFA). In: Codex general standard for food additives (GSFA) [online database]. Rome: FAO/WHO Codex Alimentarius Commission, 1995, updated 2011 [cit. 17 February 2011]. <<http://www.codexalimentarius.net/gsfaonline/additives/results.html?techFunction=9&searchBy=tf>>.
7. The second Korea national health and nutrition examination survey – Nutrition survey. In: The second Korea national health and nutrition examination survey. Seoul: The Korean Ministry of Health and Welfare, 2002, pp. 85–154. (in Korean)
8. Walford, J.: Developments in food colours. II. London: Applied Science Publishers, 1980. 259 pp. ISBN 0853348812.
9. Sawaya, W. – Hysain, A. – Al-Otaibi, J. – Al-Foudari, M. – Hajji, A.: Colour additive levels in foodstuffs commonly consumed by children in Kuwait.

- Food Control, 19, 2008, pp. 98–105.
10. Spears, K. – Marshall, J.: Qualitative analysis of artificial colourings in food. *Journal of Association of Public Analysts*, 25, 1987, pp. 47–54.
 11. Suh, H. J. – Chung, M. S. – Cho, Y. H. – Kim, J. W. – Kim, D. H. – Han, K. W. – Kim, C. J.: Estimated daily intakes for BHA, BHT, and TBHQ in Korea. *Food Additives and Contaminants*, 22, 2005, pp. 1176–1188.
 12. The second Korea national health and nutrition examination survey – Health examination survey. In: *The second Korea national health and nutrition examination survey*. Seoul: The Korean Ministry of Health and Welfare, 2002, pp. 54–197. (in Korean)
 13. Lok, K. Y. W. – Chung, W. Y. – Benzie, I. F. – Woo, J.: Colour additives in snack foods consumed by primary school children in Hong Kong. *Food Additives and Contaminants*, part B, 3, 2010, pp. 148–155.
 14. Rao, P. – Sudershan, R. V. – Krishna, T. P. – Naidu, N.: Exposure assessment to synthetic food colours of a selected population in Hyderabad, India. *Food Additives and Contaminants*, 21, 2004, pp. 415–421.
 15. Toledo, M. C. F. – Guerchon, M. S. – Ragazzi, S.: Potential weekly intake of artificial food colours by 3–14-year-old children in Brazil. *Food Additives and Contaminants*, 9, 1992, pp. 291–301.
 16. Husain, A. – Sawaya, W. – Al-Omair, A. – Al-Zenki, S. – Al-Amiri, H. – Ahmed, N. – Al-Sinan, M.: Estimates of dietary exposure of children to artificial food colours in Kuwait. *Food Additives and Contaminants*, 23, 2006, pp. 245–251.
 17. Dietary intake of food additive by Korean population – Coal tar dye combined diet intake. In: *Dietary intake of food additive by Korean population*. Seoul: Korea Food and Drug Administration, 2006, pp. 55–57. (in Korean)
 18. Korea Food Additives Code. In: *KFDA* [online]. Ohsong: Korea Food and Drug Administration, 2007, updated 2010 [cit. 15 April 2011]. <http://www.kfda.go.kr/fa/index.do?nMenuCode=12&page_gubun=1&gongjeoncategory=1> (in Korean)
 19. US FDA/CFR-Code of Federal Regulation Title 21, Part 74 Listing of color additives subject to certification. In: *U.S. Food and Drug Administration* [online]. Maryland : US Food and Drug Administration, updated 1 April 2011 [cit. 10 April 2011]. <<http://www.accessdata.fda.gov/scripts/cdrh/cfdocs/cfcfr/CFRSearch.cfm?CFRPart=74>>
 20. Standards for use of food additives, 7th ed, 2000. In: *The Japan Food Chemical Research Foundation* [online]. Tokyo : The Japanese Ministry of Health, Labour and Welfare, updated 30 November 2006 [cit. 20 February 2011]. <<http://www.ffcr.or.jp/zaidan/FFCRHOME.nsf/pages/stanrd.use>>
 21. Scientific opinion on the re-evaluation of Sunset Yellow FCF (E110) as a food additive. *EFSA Journal*, 7, 2009, issue 11:1330. doi:10.2903/j.efsa.2009.1330.
 22. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Tartrazine. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_2228.htm>
 23. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Sunset Yellow FCF. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_2217.htm>
 24. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Amaranth. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_94.htm>
 25. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Erythrosine. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_700.htm>
 26. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Allura Red AC. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_55.htm>
 27. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Ponceau 4R. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_1959.htm>
 28. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Brilliant Blue FCF. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_236.htm>
 29. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Indigotine. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_1117.htm>
 30. Summary of Evaluations Performed by the Joint FAO/WHO Expert Committee (JECFA) on Food Additive – Fast Green FCF. In: *IPCS INCHEM website* [online]. Geneva: International Programme on Chemical Safety, updated 9 May 2003 [cit. 2 February 2011]. <http://www.inchem.org/documents/jecfa/jecval/jec_849.htm>

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