Report

Surveillance of Cadmium Concentration in Chocolate and Cocoa Powder Products Distributed in Japan

Yohei KATAOKA*, Takahiro WATANABE, Kyoko HAYASHI and Hiroshi AKIYAMA

National Institute of Health Sciences:
3–25–26 Tonomachi, Kawasaki-ku, Kawasaki 210–9501, Japan;
*Corresponding author

Chocolate and cocoa are manufactured from cacao beans produced by the cacao tree (*Theobroma cacao*). These products may contain cadmium (Cd), which originates from contaminated soil. Here, we surveyed the Cd concentrations in dark chocolate, milk chocolate, white chocolate and cocoa powder products purchased at retail stores in Japan, using inductively coupled plasma mass spectrometry. The Cd concentrations in these chocolate and cocoa powder products ranged from 0.00021 to 2.3 mg/kg and from 0.015 to 1.8 mg/kg, respectively. A weak positive correlation was found between the Cd concentration and the content of cocoa solids stated on the product labels. A comparison between these results and the maximum levels (MLs) set by the European Union revealed that the Cd concentrations in chocolate and cocoa powder products on the Japanese market exceeded the MLs for eight of the 180 chocolate products and 26 of the 140 cocoa powder products.

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Key words: cadmium; chocolate; cocoa powder; surveillance

Introduction

Cadmium (Cd) is a heavy metal element that exists widely in nature, in minerals and soil. It is commercially used for metal plating to prevent rusting of iron materials, in batteries, and in alloys with lead and tin. Since Cd coexists at high concentrations in mineral deposits containing mainly zinc and copper, it can be obtained as a by-product when these metals are mined and refined[1)–3)].

Cd contamination of foods can occur through numerous pathways, including industrial activities such as metal mining and refining, and geological events. For example, ash released into the atmosphere during volcanic eruptions is a major source of Cd contamination in some countries with strong volcanic activity[4)–6)]. The Cd found in water and soil is accumulated in various vegetables, cereals, livestock, fish and shellfish, etc. Consequently, humans can be exposed to Cd by consuming foods derived from these agricultural commodities. The concentration of Cd in foods has received much attention because of its association with toxicity[7),8)]. Continued consumption of foods contaminated with high levels of Cd carries an increased risk of adverse health effects, such as kidney disorder.

In contrast to concerns about the adverse health effects of Cd, foods containing ingredients derived from cacao beans are considered to have beneficial health effects because of their high levels of antioxidants. These foods, such as cocoa liquor, cocoa mass, cocoa butter, cocoa powder, and chocolate, are manufactured from cacao beans produced by the cacao tree (*Theobroma cacao*). However, high concentrations of Cd have been found in beans from cacao trees grown in soil with high Cd concentrations[9),10)]. It is highly likely that high Cd concentrations are retained in the finished products made from these beans, because reducing the Cd concentration during the manufacturing process is difficult[11)]. For this reason, as a measure to manage the risk associated with the consumption of chocolate and cocoa powder products, the European Union (EU) announced plans to implement regulations governing chocolate and cocoa powder products containing excessive Cd levels (No. 488/2014), which go into effect on January 1, 2019.

The detection of Cd in chocolate and other cocoa powder products has been reported[12)–21)]. However, information on Cd analysis and concentration in chocolate and cocoa powder products remains insufficient. In the present study, we surveyed Cd concentrations in various types and brands of chocolate and cocoa powder products available on the Japanese market, using validated analytical methods. For chocolate products, the relationship between Cd concentration and cocoa solid content was also evaluated.

Materials and Methods

Samples

From April 2015 to August 2016, 320 samples, including 180 chocolate products (dark, milk, and white choco-

*ykataoka@nihs.go.jp

late products) and 140 cocoa powder products, were purchased online or at supermarkets in Tokyo and surrounding areas. A portion of each product was homogenized and used as an analytical sample. The concentrations of Cd in these products were then measured using inductively coupled plasma mass spectrometry (ICP-MS).

Reagents

Double deionized water (Milli-Q 18.2 MΩ/cm resistivity; Merck Millipore, Darmstadt, Germany) was used for all dilutions. HNO_3 (specific gravity 1.40) for ultra-trace analysis was purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan), and H_2O_2 (30%) of ultrapure quality was purchased from Kanto Chemical Co., Inc. (Tokyo, Japan). All vessels, which were made of polypropylene, were cleaned by soaking in diluted HNO_3 and rinsed with distilled water before use. All solutions were appropriately diluted with 1% nitric acid solution unless otherwise stated. The element standard solutions used for calibration were prepared by diluting 1000 mg/L stock solution (Sigma-Aldrich Co., St. Louis, MO, USA). An internal standard solution was prepared by diluting a 1,000 mg/L stock solution of indium (In).

Instruments

The ICP-MS instrument (iCAP Q; Thermo Fisher Scientific, Waltham, MA, USA) was used under the following conditions: Ar auxiliary gas flow rate 0.8 L/min; Ar cooling gas flow rate 14.0 L/min; quartz cyclonic chamber and perfluoroalkoxy alkane nebulizer (1.0 mL/min); water cooling; radiofrequency power 1,550 W. The sensitivity and resolution of the apparatus were optimized with the aid of a customized standard from Thermo Fisher Scientific. A deionized water sample was used to calibrate the instrument.

The Ethos-One (Milestone, Bergamo, Italy) closed-vessel microwave digestion system was used to prepare the samples. Teflon and quartz reaction vessels were used in all digestion procedures. The reaction vessels were immersed in HNO_3 (10% v/v) overnight and then rinsed well with double deionized water before each sample digestion.

Digestion procedure and measurement

A 0.5 g mass of each homogenized sample was weighed into the quartz reaction vessel and 5 mL of concentrated HNO_3 and 2 mL of H_2O_2 were added. Next, the quartz reaction vessel was placed in the Teflon reaction vessel with 5 mL of double deionized water and 2 mL of H_2O_2 prior to digestion. The operation program of the microwave digestion system was as follows: the temperature was raised from room temperature to 70℃ for 2 min, 50℃ for 1 min, and 200℃ for 15 min, and then maintained at 200℃ for 10 min and subsequently cooled to room temperature. The digested product was rinsed in a volumetric flask with HNO_3 (2% v/v), and 0.5 mL of 10 mg/L internal standard solution was added, after which the mixture was diluted to 50 mL with HNO_3 (2% v/v) to prepare the sample solutions.

Blank digests were performed for each quartz reaction vessel used in the experiments in the same way. The Cd concentrations were measured using ICP-MS. The limit of quantitation was 0.00005 mg/kg.

Performance evaluation of the analytical method

To evaluate the performance of the analytical method, a blank sample of a chocolate product containing a low Cd concentration was spiked with 0.005, 0.2, and 1.0 mg/kg of Cd standard. The two blank samples and two spiked samples of each concentration were analyzed in parallel per day, and this analysis was repeated for 5 days. All blank and spiked samples were subjected to the same preparation and analysis procedures. The trueness, repeatability, and intralaboratory reproducibility of the method were estimated from the analytical results, as were the measurement uncertainties.

Results and Discussion

Validation of the analytical method

The trueness, repeatability, and intralaboratory reproducibility estimated based on the analytical results for the spiked samples at three different levels are shown in Table 1. The estimate for the trueness ranged from 101% to 103% through the three concentrations. The repeatability and intralaboratory reproducibility through the three concentrations, expressed as RSD%, were estimated to be in the range of 1.3–4.1 and 1.3–5.3%, respectively. From these results, we considered that this method is applicable for the analysis of Cd in chocolate and cocoa powder products.

Table 1. Trueness, repeatability and intralaboratory reproducibility estimated by analysis of spiked samples

Spiked concentration (mg/kg)		Concentration (mg/kg)					Average (mg/kg)	Trueness (%)	Repeatability (RSD %)	Intralaboratory reproducibility (RSD %)
		1st day	2nd day	3rd day	4th day	5th day				
0.005	Portion 1	0.0048	0.0051	0.0047	0.0054	0.0053	0.0051	101	4.1	5.3
	Portion 2	0.0051	0.0052	0.0048	0.0048	0.0054				
0.2	Portion 1	0.20	0.21	0.21	0.20	0.21	0.21	103	1.5	1.6
	Portion 2	0.20	0.20	0.20	0.20	0.21				
1.0	Portion 1	1.0	1.0	1.0	1.0	1.1	1.0	103	1.3	1.3
	Portion 2	1.0	1.0	1.0	1.0	1.0				

Table 2. The concentrations of Cd in chocolate products

Sample No.	Concentration of Cd (mg/kg)	Product type	Cocoa solids content (%)*	Conformity to MLs in EU***	Sample No.	Concentration of Cd (mg/kg)	Product type	Cocoa solids content (%)*	Conformity to MLs in EU***
1	2.3	Dark chocolate	70	No	91	0.12	Dark chocolate	73	
2	2.0	Dark chocolate	70	No	92	0.12	Dark chocolate	70	
3	1.9	Dark chocolate	—**	No	93	0.12	Dark chocolate	64	
4	1.0	Dark chocolate	71	No	94	0.11	Dark chocolate	70	
5	1.0	Dark chocolate	70	No	95	0.11	Dark chocolate	60	
6	0.75	Dark chocolate	75		96	0.10	Dark chocolate	72	
7	0.74	Dark chocolate	61		97	0.10	Dark chocolate	54	
8	0.71	Dark chocolate	92		98	0.10	Dark chocolate	53	
9	0.62	Dark chocolate	65		99	0.10	Dark chocolate	—**	
10	0.58	Dark chocolate	63		100	0.10	Dark chocolate	73	
11	0.56	Dark chocolate	71		101	0.093	Dark chocolate	70	
12	0.55	Dark chocolate	62		102	0.092	Dark chocolate	72	
13	0.54	Dark chocolate	85		103	0.091	Dark chocolate	—**	
14	0.50	Dark chocolate	70		104	0.091	Dark chocolate	—**	
15	0.48	Dark chocolate	85		105	0.088	Dark chocolate	52	
16	0.46	Dark chocolate	71		106	0.087	Dark chocolate	48	
17	0.45	Dark chocolate	—**	Possible nonconformity	107	0.085	Dark chocolate	77	
18	0.42	Dark chocolate	72		108	0.084	Milk chocolate	—**	
19	0.42	Dark chocolate	80		109	0.083	Dark chocolate	70	
20	0.41	Dark chocolate	75		110	0.080	Dark chocolate	70	
21	0.38	Dark chocolate	70		111	0.077	Dark chocolate	72	
22	0.38	Dark chocolate	71		112	0.071	Dark chocolate	70	
23	0.36	Dark chocolate	70		113	0.070	Dark chocolate	72	
24	0.36	Dark chocolate	71		114	0.069	Dark chocolate	64	
25	0.36	Dark chocolate	99		115	0.069	Milk chocolate	—**	
26	0.36	Dark chocolate	95		116	0.068	Dark chocolate	—**	
27	0.34	Dark chocolate	70		117	0.064	Dark chocolate	56	
28	0.34	Dark chocolate	75		118	0.061	Dark chocolate	—**	
29	0.33	Dark chocolate	80		119	0.060	Dark chocolate	60	
30	0.33	Dark chocolate	100		120	0.060	Dark chocolate	72	
31	0.32	Dark chocolate	62		121	0.060	Milk chocolate	38	
32	0.31	Dark chocolate	—**	Possible nonconformity	122	0.059	Dark chocolate	—**	
33	0.31	Dark chocolate	70		123	0.059	Milk chocolate	—**	
34	0.31	Milk chocolate	50	No	124	0.058	Milk chocolate	—**	
35	0.31	Dark chocolate	56		125	0.058	Dark chocolate	50	
36	0.31	Dark chocolate	70		126	0.056	Dark chocolate	61	
37	0.30	Dark chocolate	80		127	0.054	Dark chocolate	85	
38	0.30	Dark chocolate	—**		128	0.053	Dark chocolate	—**	
39	0.29	Dark chocolate	81		129	0.053	Dark chocolate	85	
40	0.28	Dark chocolate	67		130	0.053	Milk chocolate	—**	
41	0.27	Dark chocolate	71		131	0.052	Milk chocolate	—**	
42	0.26	Dark chocolate	—**		132	0.052	Milk chocolate	39	
43	0.25	Dark chocolate	80		133	0.051	Dark chocolate	55	
44	0.24	Dark chocolate	73		134	0.050	Dark chocolate	70	
45	0.24	Dark chocolate	95		135	0.048	Dark chocolate	—**	
46	0.24	Dark chocolate	—**		136	0.047	Dark chocolate	57	
47	0.23	Dark chocolate	75		137	0.046	Milk chocolate	35	
48	0.23	Dark chocolate	65		138	0.046	Dark chocolate	59	
49	0.23	Dark chocolate	70		139	0.046	Dark chocolate	58	
50	0.22	Dark chocolate	85		140	0.045	Dark chocolate	50	
51	0.22	Dark chocolate	—**		141	0.044	Dark chocolate	50	
52	0.22	Dark chocolate	66		142	0.044	Dark chocolate	—**	
53	0.22	Dark chocolate	75		143	0.044	Dark chocolate	54	
54	0.22	Dark chocolate	86		144	0.041	Dark chocolate	48	
55	0.21	Dark chocolate	70		145	0.041	Dark chocolate	54	
56	0.21	Dark chocolate	73		146	0.036	Dark chocolate	—**	
57	0.20	Dark chocolate	74		147	0.035	Milk chocolate	—**	
58	0.20	Dark chocolate	80		148	0.035	Dark chocolate	—**	
59	0.19	Dark chocolate	70		149	0.033	Milk chocolate	33	
60	0.19	Dark chocolate	70		150	0.033	Dark chocolate	60	
61	0.19	Dark chocolate	68		151	0.033	Milk chocolate	37	
62	0.18	Dark chocolate	70		152	0.033	Milk chocolate	—**	
63	0.18	Dark chocolate	66		153	0.033	Dark chocolate	52	
64	0.18	Dark chocolate	64		154	0.030	Milk chocolate	—**	
65	0.18	Dark chocolate	62		155	0.029	Dark chocolate	—**	
66	0.18	Milk chocolate	49		156	0.028	Milk chocolate	—**	
67	0.18	Dark chocolate	70		157	0.025	Dark chocolate	48	
68	0.17	Dark chocolate	60		158	0.023	Milk chocolate	30	
69	0.17	Dark chocolate	65		159	0.023	Milk chocolate	30	
70	0.17	Dark chocolate	60		160	0.019	Milk chocolate	35	
71	0.17	Dark chocolate	70		161	0.019	Milk chocolate	—**	
72	0.15	Dark chocolate	49		162	0.019	Milk chocolate	31	
73	0.15	Dark chocolate	72		163	0.018	Milk chocolate	58	
74	0.15	Dark chocolate	72		164	0.017	Milk chocolate	31	
75	0.15	Dark chocolate	99		165	0.017	Milk chocolate	32	
76	0.15	Dark chocolate	80		166	0.015	Milk chocolate	32	
77	0.14	Dark chocolate	—**		167	0.015	Milk chocolate	31	
78	0.13	Dark chocolate	70		168	0.015	Milk chocolate	38	
79	0.13	Dark chocolate	—**		169	0.015	Milk chocolate	35	
80	0.13	Dark chocolate	99		170	0.015	Milk chocolate	30	
81	0.13	Dark chocolate	63		171	0.014	Milk chocolate	36	
82	0.13	Dark chocolate	85		172	0.014	Milk chocolate	—**	
83	0.13	Dark chocolate	72		173	0.013	Milk chocolate	35	
84	0.13	Dark chocolate	70		174	0.013	Milk chocolate	—**	
85	0.13	Dark chocolate	62		175	0.012	Milk chocolate	—**	
86	0.12	Dark chocolate	72		176	0.00158	White chocolate	29	
87	0.12	Dark chocolate	70		177	0.00065	White chocolate	29	
88	0.12	Dark chocolate	72		178	0.00028	White chocolate	28	
89	0.12	Dark chocolate	55		179	0.00021	White chocolate	31	
90	0.12	Dark chocolate	70		180	0.00021	White chocolate	35	

*Information described in the product.
**No information in the product.
***The blank is the conforming product.

Table 3. Summary of the concentrations of Cd in chocolate products and cocoa powder products

	Concentration (mg/kg)				
	Chocolate				Cocoa powder ($n=140$)
	Total ($n=180$)	Dark chocolate ($n=140$)	Milk chocolate ($n=35$)	White chocolate ($n=5$)	
Maximum value	2.3	2.3	0.31	0.0016	1.8
Minimum value	0.00021	0.025	0.012	0.00021	0.015
Mean	0.20	0.25	0.043	0.00059	0.32
Median	0.12	0.15	0.023	0.00028	0.16
Standard deviation	0.30	0.32	0.056	0.00059	0.38

Table 4. The concentrations of Cd in cocoa powder products

Sample No.	Concentration of Cd (mg/kg)	Conformity to MLs in EU***	Sample No.	Concentration of Cd (mg/kg)	Conformity to MLs in EU***
1	1.8	No	71	0.16	
2	1.6	No	72	0.16	
3	1.6	No	73	0.16	
4	1.5	No	74	0.16	
5	1.4	No	75	0.15	
6	1.31	No	76	0.15	
7	1.29	No	77	0.15	
8	1.28	No	78	0.15	
9	1.21	No	79	0.15	
10	1.20	No	80	0.15	
11	0.95	No	81	0.15	
12	0.93	No	82	0.15	
13	0.92	No	83	0.15	
14	0.84	No	84	0.15	
15	0.79	No	85	0.15	
16	0.77	No	86	0.15	
17	0.77	No	87	0.15	
18	0.76	No	88	0.15	
19	0.73	No	89	0.15	
20	0.72	No	90	0.15	
21	0.68	No	91	0.15	
22	0.66	No	92	0.14	
23	0.63	No	93	0.14	
24	0.61	No	94	0.14	
25	0.61	No	95	0.14	
26	0.60	No	96	0.14	
27	0.57		97	0.14	
28	0.57		98	0.14	
29	0.48		99	0.14	
30	0.47		100	0.13	
31	0.46		101	0.132	
32	0.44		102	0.125	
33	0.43		103	0.123	
34	0.42		104	0.121	
35	0.37		105	0.121	
36	0.37		106	0.118	
37	0.36		107	0.116	
38	0.36		108	0.115	
39	0.36		109	0.105	
40	0.35		110	0.100	
41	0.35		111	0.095	
42	0.35		112	0.089	
43	0.35		113	0.087	
44	0.33		114	0.074	
45	0.32		115	0.066	
46	0.29		116	0.056	
47	0.26		117	0.054	
48	0.25		118	0.053	
49	0.20		119	0.050	
50	0.20		120	0.050	
51	0.19		121	0.050	
52	0.19		122	0.049	
53	0.18		123	0.043	
54	0.18		124	0.041	
55	0.18		125	0.039	
56	0.18		126	0.038	
57	0.17		127	0.037	
58	0.17		128	0.036	
59	0.17		129	0.036	
60	0.17		130	0.035	
61	0.17		131	0.034	
62	0.17		132	0.034	
63	0.17		133	0.033	
64	0.17		134	0.031	
65	0.17		135	0.028	
66	0.17		136	0.026	
67	0.16		137	0.020	
68	0.16		138	0.019	
69	0.16		139	0.018	
70	0.16		140	0.015	

*Blank indicates a conforming product.

Table 5. Summary of Cd concentrations in chocolate products classified with cocoa solid contents

	Concentration (mg/kg)				
		Products with information on cocoa solids contents			
	Total ($n=140$)*	Cocoa solids content			
		<50% ($n=23$)	50%≧, <70% ($n=41$)	70%≧, <80% ($n=55$)	80%≧ ($n=21$)
Maximum value	2.3	0.18	0.74	2.3	0.71
Minimum value	0.013	0.013	0.018	0.050	0.053
Mean	0.22	0.040	0.17	0.32	0.28
Median	0.13	0.023	0.12	0.19	0.25
Standard deviation	0.30	0.044	0.17	0.41	0.16

*The number of the products with cocoa solid contents in the indicated range.

Analytical results for the chocolate and cocoa powder products

Cd concentrations were determined in 180 kinds of chocolate products using the validated ICP-MS method. The results are shown in Table 2. The statistics regarding the Cd concentrations measured in the chocolate products are summarized in Table 3. The Cd concentrations ranged from 0.00021 to 2.3 mg/kg, and the maximum concentration (2.3 mg/kg) was found in a dark chocolate product. The Cd concentrations in the chocolate products found in the present study are in a similar range to those of previous reports[12–21]. As in the previous reports, Cd concentrations measured in dark chocolate products were higher than those in milk chocolate products.

The Cd concentrations determined in 140 different cocoa powder products are shown in Table 4. The statistics regarding the concentrations are summarized in Table 3. The Cd concentrations ranged from 0.015 to 1.8 mg/kg.

The statistics regarding the Cd concentrations in chocolate and cocoa powder products are shown in Table 3. The highest mean Cd concentration was found in cocoa powder (0.32 mg/kg), followed by dark chocolate (0.25 mg/kg) and milk chocolate (0.043 mg/kg) products. The Cd concentrations detected in the white chocolate products were all below the limit of quantitation.

Correlation between percentage of cocoa solids and Cd concentration in chocolate products

Table 5. shows the relationship between detected Cd concentrations and the contents of cocoa solids stated on the labels. Information regarding the contents of cocoa solids (percentage) was found for 140 of the 180 chocolate products. These 140 products were classified into the following four groups according to the percentage of cocoa solids they contained: <50%, from 50 to <70%, from 70 to <80%, and ≥80%. The medians were then calculated as representative values of the Cd concentration for each classification. As shown in Fig. 1, the Cd concentration appeared to increase with increase in the percentage of cocoa solids. Correlation analysis between the Cd concentration and the content of cocoa solids showed only a weak positive correlation (Fig. 2). These findings suggest that the Cd concentration in chocolate products depends on not only the content of cocoa solids, but also on the Cd concentration in the cacao beans used

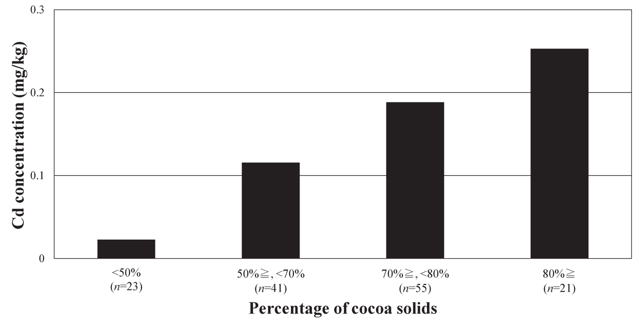

Fig. 1. Measured Cd concentrations(median) *vs.* percentage of cocoa solids stated on the labeling for chocolate products ($n=140$)

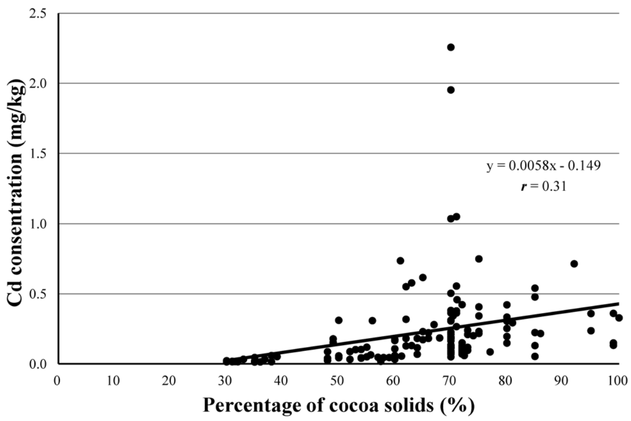

Fig. 2. Correlation between percentage of cocoa solids stated on the labeling of chocolate products and the measured concentrations of Cd ($n=140$)

Compliance with EU standards

The determined values of Cd concentrations were compared with the maximum levels (MLs) of Cd concentration for specific cocoa and chocolate products, enforced as EU standards from January 2019. The EU regulations are summarized in Table 6. The Cd concentration exceeded the MLs for eight of the 180

Table 6. The EU's cadmium MLs in chocolate and cocoa products, enforced from January 2019

Chocolate products	Total dry cocoa solids (%)	MLs (mg/kg)
Milk chocolate	＜30	0.10
	≧30	0.30
Chocolate	＜50	0.30
	≧50	0.80
Cocoa powder sold to the final consumer or as an ingredient in sweetened cocoa powder sold to the final consumer (drinking chocolate)		0.60

chocolate and 26 of the 140 cocoa powder products. The proportions of nonconforming chocolate and cocoa powder products were 4.4 and 18.6%, respectively. These results include the Cd concentrations in two chocolate products for which information regarding cocoa solid content was unknown (we assumed that the total cocoa solid content was less than 50% in these two products).

Conclusion

In the present study, we found that the Cd concentrations in chocolate and cocoa powder products purchased on the Japanese market from 2015 to 2016 ranged from 0.00021 to 2.3 mg/kg and 0.015 to 1.8 mg/kg, respectively. The Cd concentrations detected in the examined products varied greatly, showing a range of approximately 10,000-fold for chocolate and about 100-fold for cocoa powder products. A weak positive correlation was found between the Cd concentration and the content of cocoa solids, indicating that the Cd concentration in chocolate tends to increase with increasing cocoa solids content.

We also examined whether the Cd concentrations in 180 chocolate and 140 cocoa powder products complied with the MLs set by the EU. Among them, 4.4% of chocolate products and 18.6% of cocoa powder products had Cd concentrations greater than the MLs.

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