A multi-year heavy metal analysis of 72 dark chocolate and cocoa products in the USA

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Introduction: Contamination of cocoa containing products, such as dark chocolate, with heavy metals including lead, cadmium and arsenic has been reported in the US. However, a formal exploration into the significance of this contamination, nor multi-year trends in the degree or scope remain unresolved.

Methods: From 2014 to 2022, 72 consumer cocoa-containing products were purchased and analyzed for heavy metal contamination with lead (Pb), cadmium (Cd), and arsenic (As) in 4 distinct cohorts (2014, 2016, 2019, 2022). The thresholds used to assess heavy metal contamination were set to Prop 65 maximum allowable dose levels (MADLs) of 0.5 mcg/day, 4.1 mcg/day, 10 mcg/day for Pb, Cd, and As, respectively.

Results and discussion: Our analysis reports that 43, 35, and 0% of the products tested exceed Prop 65 MADLs for heavy metal concentrations, respectively, of Pb, Cd, and As, while 97.2% (70 of 72) fall below US FDA IRL limits established for Pb. Median concentrations of each metal tested were lower than even the conservative Prop 65 MADLs, indicating a potentially large effect of product outliers. This indicates that heavy metal contamination—in more than half of products tested—may not pose any appreciable risk for the average person when consumed as a single serving; however, consuming some of the products tested, or more than one serving per day in combination with non-cocoa derived sources heavy metals, may add up to exposure that would exceed the Prop 65 MADLs. Notably, "organic" products were significantly more likely to demonstrate higher levels of both Cd and Pb.

KEYWORDS

lead, cadmium, metal, cocoa, cocoa and chocolates

Introduction

The contamination of consumer cocoa-containing products such as dark chocolate with heavy metals, including lead (Pb), cadmium (Cd), and arsenic (As), has been reported by prominent consumer media outlets and independent testing agencies (1–4). Such findings may significantly alter consumer dietary behaviors, as they may opt against the consumption of cocoa-containing products owing to purported health risks associated with heavy metal contamination. The ramifications nor the clinical veracity of these claims have been explored in the literature. Recent research has further underscored the global concern regarding heavy

metal contamination in food products, highlighting the need for stringent monitoring and regulation (5).

Heavy metal contamination in consumer foods remains a significant issue globally, though there is evidence of progress in the domestic United States (US), with serum levels of heavy metals, particularly Pb, falling by almost 95% since 1976 (6–10). Heavy metal contamination has been demonstrated to be pervasive across nations and has been detected in nearly every food category from grain to meat-products, with vegetable and cereal products most affected by industrial and environmental sources of contamination (11–16). Pb, Cd, As and Hg are typically evaluated for contamination as they represent the most common health threats to consumers (9–23). National initiatives, including the US Food and Drug Administration (FDA) Closer to Zero Program (C2Z), have been implemented and aim to reduce the net level of contamination in foods, especially those consumed by sensitive groups such as toddlers and expectant mothers, to the lowest possible level while maintaining access to nutritious foods (24). The C2Z works by establishing an interim reference level (IRL) or maximum allowable intake corresponding to a serum target roughly 10-fold below the accepted reference level or value for a particular toxicant. In the case of Pb, for example, the IRL is derived from the blood Pb reference value (BLRV) or the 97.5% of blood lead levels (BLLs) in children 1–5 years of age identified during US Center for Disease Control (CDC) surveillance of the National Health and Nutrition Examination Survey (NHANES). As of 2022, the IRL for has been amended to 2.2 mcg/day for children under 7 years of age and 8.8 mcg/day of Pb for women of child-bearing age (24, 25). This is a 64% decrease from the previous cutoff for Pb of 6 mcg/day for young children and 25 μg/day for pregnant or lactating women in the early 1990's, but this is still well above the California Proposition 65 (Prop 65) maximum allowable dose levels (MADLs).

While federal regulations pertaining to consumer Pb contamination have received, perhaps, the most visibility, proposed reference ranges have been offered by several agencies for both As and Cd. MADLs set on daily consumption by Prop 65 mandate that total Cd and As consumption be 4.1 and 10 mcg, respectively (26–28).

Cocoa-containing products are notoriously rich in metals owing to cultivation and manufacturing practices required to produce such products (29). Dark chocolate in particular is a rich source of earth metals and contamination in the course of its processing has been well documented. Specifically, it is thought that the proximity of cocoa-farms and processing plants apparent proximity to other processing and industrial facilities, which have been demonstrated to be significant sources of pollution, is a driver of heavy metal contamination in dark chocolate. While several authors have noted that heavy metal contamination of dark chocolate and other cocoa products is chiefly the result of post harvesting contamination, the precise level of contamination and the clinical implications of this contamination for domestic dark chocolate and cocoa-containing products is largely unexplored in the literature (6, 30).

Our aim is to assess the heavy metal contamination of the most popular cocoa-containing consumer products each year for several years to assess trends. In this study, we report the findings from a multi-year heavy metal contamination survey (2014–2022) of 72 cocoa-containing products purchased in the domestic US, which were identified through annual consumer surveys of product popularity. This data serves to provide a window into the extent of the contamination problem in US cocoa-containing products such as dark chocolate.

Methods

From 2014 to 2022, 72 consumer cocoa-containing products were purchased from retail sources and analyzed for heavy metal contamination with Pb, Cd, and As in 4 distinct cohorts (2014, 2016, 2019, 2022). Pb, Cd and As were selected as they tend to be the most common sources of heavy metals in cocoa containing products owing to their presence in soil and their role as industrial waste products in and around cocoa processing sites.

The thresholds used to assess heavy metal contamination were set to Prop 65 MADLs of 0.5 mcg/day, 4.1 mcg/day, 10 mcg for Pb, Cd, and As, respectively. All heavy metal concentrations are listed in mcg/g and were scaled to mcg/day based on listed servings per product and assuming one serving per day. Seals and certifications (e.g., Organic, Fairtrade, Non-GMO) were recorded.

All products were purchased by ConsumerLab.com from third party online retailers (e.g., Amazon, iHerb), physical retailers (e.g., GNC, Whole Foods) and manufacturer or distributor websites. All products were domestic or European produced and obtained domestically. Products were selected largely according to a yearly ranked product survey taken by 8,000–10,000 respondents through the online site, ConsumerLab.com, an independent testing company committed to providing transparency and clinically-guided knowledge of nutritional supplementation to consumers and healthcare providers. Product selection in each year was affected by previous findings: Products with higher levels of heavy metal contamination were often excluded from subsequent testing, consistent with ConsumerLab's stated mission "To help consumers and healthcare professionals find the best quality health and nutrition products through independent testing and evaluation."

Two independent commercial laboratories in the domestic US were enlisted by ConsumerLab.com for analysis employing acid or microwave digestion and Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS). For the lead analysis, nominal 0.5 gram weights of the samples were digested with trace metals grade concentrated nitric acid, hydrochloric acid and hydrogen peroxide using a PerkinElmer Multiwave 3,000 Anton Paar Microwave Reaction System. After digestion, the samples were brought to a final volume of 50 mL with DI water. For arsenic and cadmium analysis, nominal 1 g weights of the samples were digested on a hotplate with trace metals grade concentrated nitric acid. After digestion, the samples were brought to a final volume of 200 mL with DI water. Samples were analyzed for metals on a PerkinElmer NexIon 350X ICPMS. Reporting limits were 0.01ug/g for lead and 0.02 ug/g for arsenic and cadmium. Analytic techniques used for testing of Pb, Cd and As in analyzed products are consistent with AOAC 2015.01 methods for the determination of heavy metal in food products. QC limits were 75%–125% for spiked recoveries. Calibration standards for arsenic were 0.001, 0.05, 0.1, and 0.5 ppm. Calibration standards for lead and cadmium were 0.0005, 0.002, 0.010, and 0.02 ppm. QC limits of variation 75%–125% and ±10% for second source continuing check standard of 0.1 ppm for arsenic and 0.01 ppm for lead and cadmium. Samples exceeding the pre-specified Prop 65 thresholds were retested at one of two independent laboratories for confirmation of heavy metal contamination.

A series of multivariate linear regressions with fixed and random effects were generated to evaluate the impact of 'number of attained third party certifications' and specific certifications (e.g., USDA Organic), controlling for cohort testing year, on total heavy metal concentrations for Pb, As, and Cd. Marginal linear predictions

TABLE 1 Summary statistics.

	N	Mean	Median	CA Prop65 limit	Min	Max
Lead (mcg/g)	72	0.062	0.05	–	0.009	0.269
Lead (mcg/serving)	72	0.615	0.375	0.5	0	3.136
Lead (mcg/day)	72	0.615	0.375	0.5	0	3.136
Cadmium (mcg/g)	72	0.396	0.239	–	0.016	2.2
Cadmium (mcg/serving)	55	6.986	3.03	4.1	0.1	92.4
Cadmium (mcg day)	37	4.358	2.725	4.1	0.29	14.12
Arsenic (mcg/g)	55	0.094	0.05	–	0.017	0.2
Arsenic (mcg/serving)	37	0.931	0.75	10	0.056	2.695
Arsenic (mcg/day)	37	0.931	0.75	10	0.056	2.695
Lead (mcg/g) (2014)	17	0.098	0.1	–	0.025	0.269
Arsenic (mcg/g) (2014)	NA	NA	NA	–	NA	NA
Cadmium (mcg/g) (2014)	17	0.512	0.224	–	0.06	1.52
Lead (mcg/g) (2016)	17	0.06	0.05	–	0.05	0.141
Arsenic (mcg/g) (2016)	17	0.196	0.2	–	0.132	0.2
Cadmium (mcg/g) (2016)	17	0.593	0.288	–	0.1	2.2
Lead (mcg/g) (2019)	21	0.044	0.03	–	0.025	0.106
Arsenic (mcg/g) (2019)	21	0.058	0.05	–	0.037	0.2
Cadmium (mcg/g) (2019)	21	0.295	0.225	–	0.054	0.982
Lead (mcg/g) (2022)	17	0.05	0.055	–	0.009	0.113
Arsenic (mcg/g) (2022)	17	0.038	0.037	–	0.017	0.078
Cadmium (mcg/g) (2022)	17	0.208	0.24	–	0.016	0.521

Summary of mean, median concentrations of heavy metals from 2014 to 2022 for all available samples. Arsenic was not tested as mcg/g in 2014.

were derived and used to assess the impact of cohort year on heavy metal contamination. Bonferroni correction was applied in head-to-head marginal comparison. Hausman testing of endogeneity was used to determine the suitability of fixed vs. standard random effects. Robust standard errors were assumed. A multivariate Ordinary Least Squares model with robust standard errors was also generated and used as a robustness check to evaluate the effect of year on metal contamination. KW testing was employed to ascertain significant differences in the content of each metal by year of testing. Finally, Welch's T-test with unequal standard errors was performed to better isolate the contribution of "organic" in the context of all metals. All statistics were calculated using StataBE 18 (Stata Corp, College Station, Tx).

Results

Overall, mean concentrations per listed serving of both Pb (0.615 mcg/serving) and Cd (4.358 mcg/serving) exceeded Prop 65 MADLs (0.5mcg/day and 4.1 mcg respectively) among all products tested (Table 1). Mean As concentrations (0.931) were universally compliant and well-below 10 mcg Prop 65 MADL. Among all products tested per serving, 31 of 72 (43%) exceeded Prop 65 limits for Pb, while 13 of 37 (35%) exceeded Prop 65 limits for Cd. Importantly, however, median concentrations of Pb (0.375 mcg/day) and Cd (3.03 mcg/day) tested below Prop 65 standards,

indicating a potentially large effect associated with the inclusion of outliers (Figures 1–3). Of note, while FDA IRLs have not been established for Cd or As, mean concentrations of Pb still resulted in levels more than 12-fold lower than the relatively less conservative US federal limits for expectant mothers and nearly 4-fold lower than those set for children younger than 7 (7).

Neither trade certifications, nor "organic" designation, resulted in lower concentrations of Pb, Cd, or As (Tables 2, 3). Those products certified as organic were, unexpectedly, associated with a statistically significant 3.163 greater mcg/serving greater concentration of Cd per serving of cocoa or dark chocolate; though, this association was tenuous and varied by specification (Table 2). When controlling for the mcg/g concentration, however, products noted as "organic" were significantly more likely to display higher levels of Cd (0.4 mcg/g) and Pb (0.02 mcg/g; Table 3, $p < 0.001$). Welch's T-testing demonstrated that organic products were not significantly more likely to present with elevated levels of Pb nor As. In contrast, Cd concentrations were again significantly greater in organic products [t = −2.1, Pr (T < t) = 0.02, Table 4].

As a whole, products in later cohorts (2016, 2019, 2022) demonstrated lower concentrations of all heavy metals tested as compared with those in 2014, with significantly lower concentrations of lead documented between the years 2022 vs. 2014 and the years 2019 vs. 2014 (Table 5). KW testing confirmed that year of testing was jointly associated with tested levels of Pb and As ($p < 0.001$).
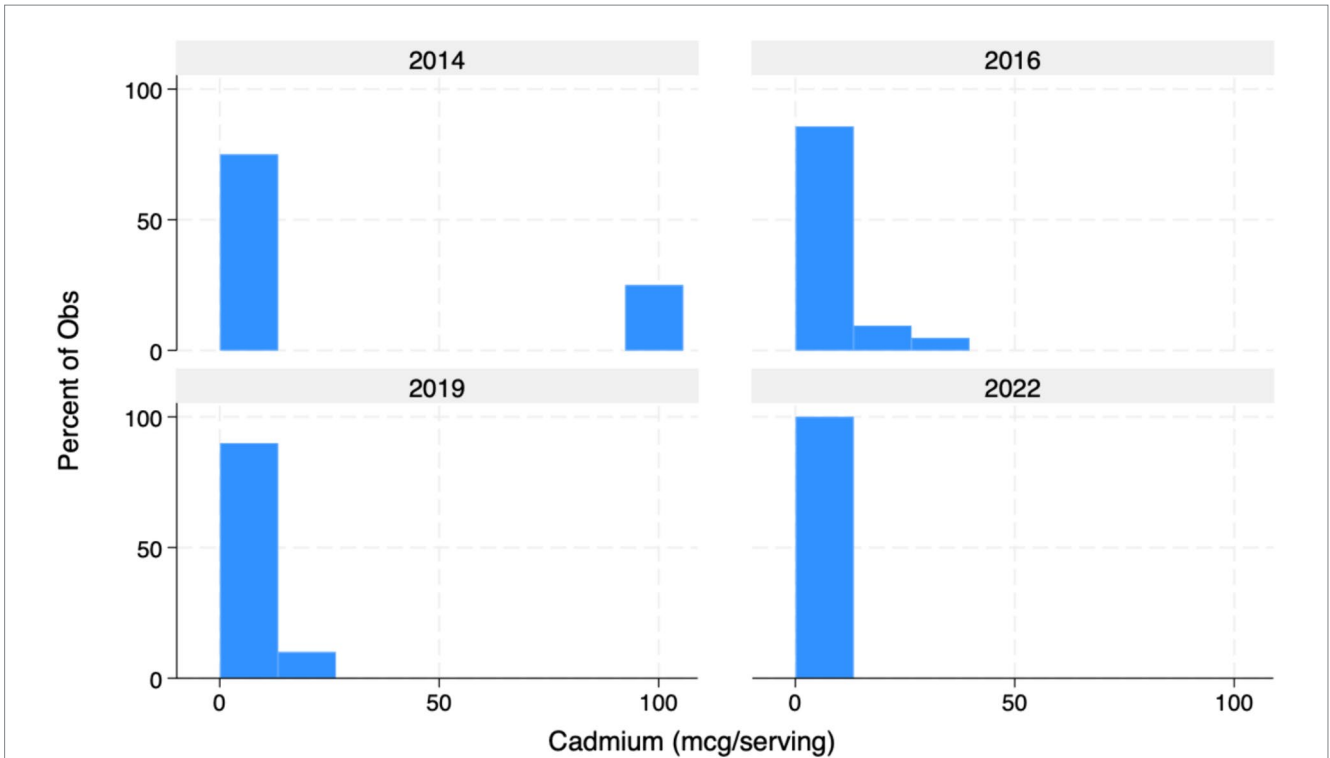
FIGURE 1
Cadmium (Cd) in mcg/serving of listed products (%). Graph of Cd concentrations detected in 72 samples. The presence of outliers in 2014 skews the apparent improvement of Cd contamination in cocoa products from 2014 to 2022.
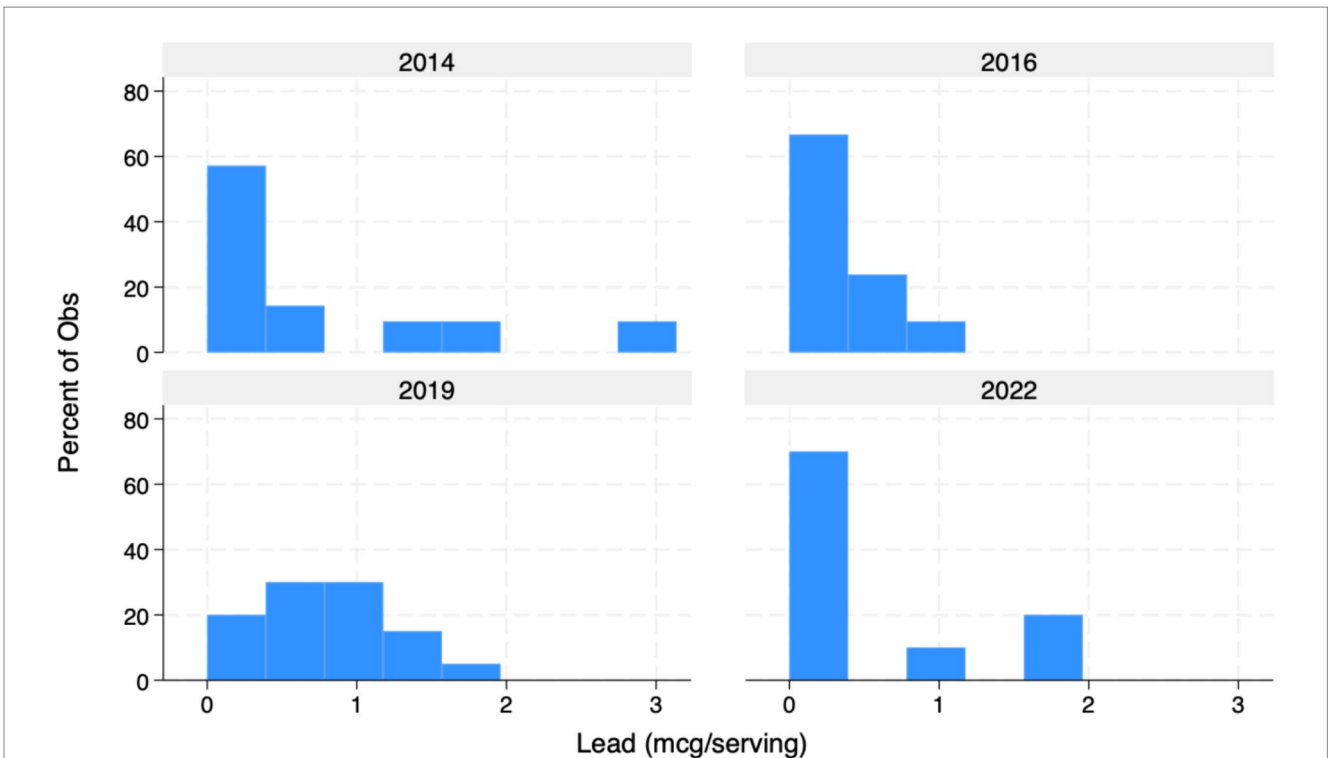

FIGURE 2
Lead [Pb in mcg/serving of listed products (%)]. Graph of Pb concentrations detected in 72 samples.
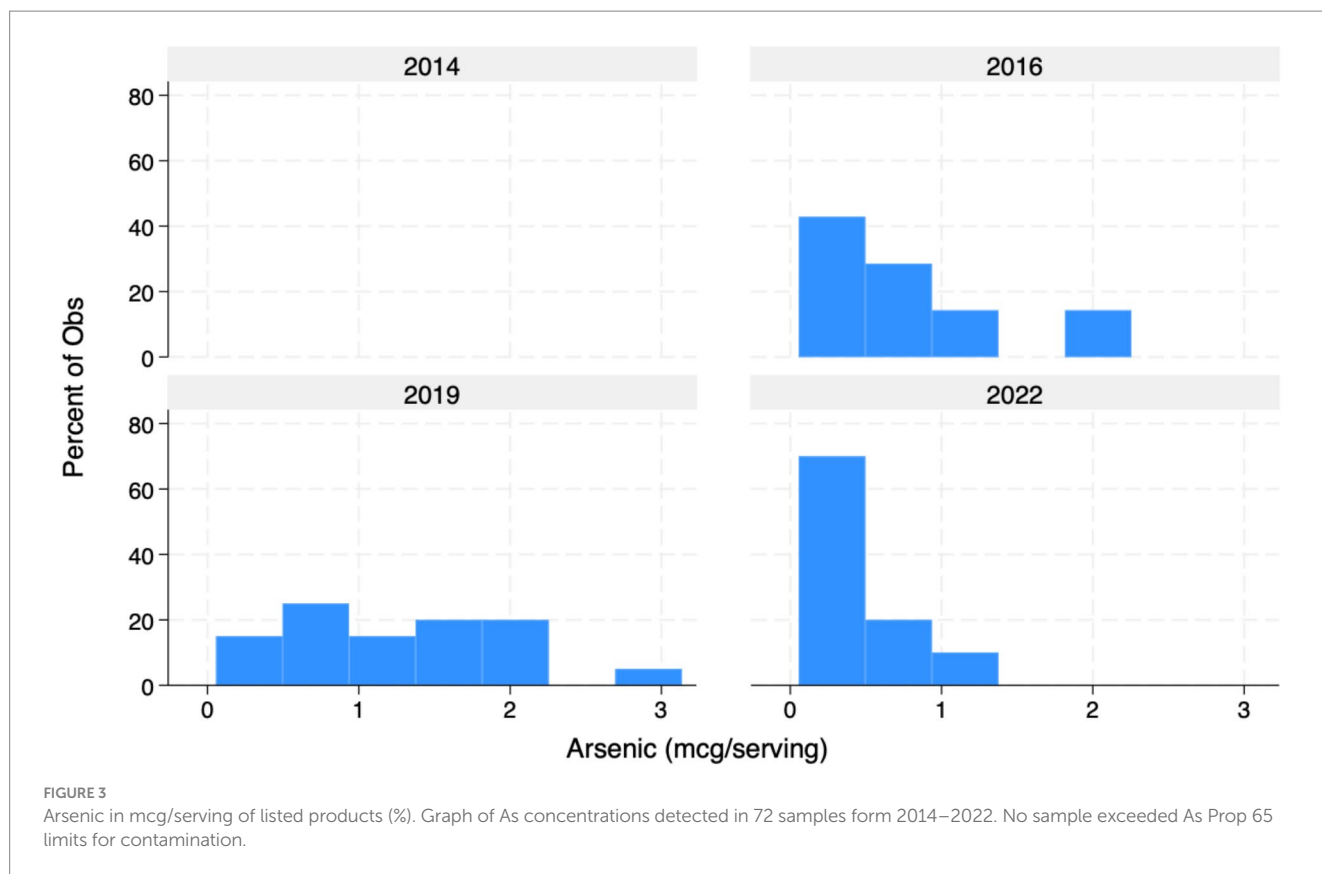
FIGURE 3
Arsenic in mcg/serving of listed products (%). Graph of As concentrations detected in 72 samples form 2014–2022. No sample exceeded As Prop 65 limits for contamination.

TABLE 2 Panel regression with FEs robust SE.

Variables	(1) Lead (mcg/serving)	(2) Cd (mcg/serving)	(3) Arsenic (mcg/serving)
Organic	0.0263	3.163**	0.108*
	(0.194)	(0.335)	(0.0121)
Certifications (quantity)	0.0195	−0.674	−0.0659
	(0.109)	(0.928)	(0.0478)
Serving (g or mL)	0.0108	0.330	0.0364
	(0.00879)	(0.150)	(0.00718)
Observations	71	54	37
R-squared	0.070	0.142	0.686
Number of years tested	4	3	2

Multivariate linear regression of heavy metals from 2014 to 2022 for all available samples in mcg/serving. Several samples were dropped owing to one or more incomplete entries required for the model. Robust standard errors in parentheses. ***$p < 0.01$, **$p < 0.05$, *$p < 0.1$.

Discussion

We report the concentrations of Pb, Cd, and As for 72 popular cocoa-containing consumer products (e.g., dark chocolate) sampled over a period of 8 years.

Our analysis suggests that less than 50% of the products tested exceed Prop 65 MADLs for heavy metal concentrations of Pb and Cd, while 97.2% (70 of 72) fall below US FDA IRL limits established for Pb. Moreover, median concentrations of each metal tested were lower than even the conservative Prop 65 MADLs. This indicates that the heavy metal contamination, in the majority of products surveyed, may

not pose a risk for the average person when consumed as a single serving; however, consuming more than one serving per day and/or in combination w other sources of heavy metals (e.g., seafood) may cumulatively add up to exposure that exceed the Prop 65 MADLs.

The question of "hazard" associated with consumer consumption of heavy metals contained in cocoa-containing products is complex. Taken at their mean or median concentrations, the risk associated with Pb consumption, for example, is "low" or almost 4-fold lower than the most stringent US FDA IRL of 2.2 mcg/day of Pb for children under 7 (30). Yet, significant outliers (maximum tested Pb sample = 3.13 mcg/serving) do, in fact, begin

TABLE 3 OLS Regression with Robust SEs.

Variables	(1) Lead (mcg/gram)	(2) Cd (mcg/gram)	(3) Arsenic (mcg/gram)
Organic?	0.0202***	0.401***	0.0204
	(0.00715)	(0.148)	(0.0141)
Certifications (quantity)	−0.00896***	−0.0745*	−0.00432
Year (compared with 2014)	(0.00300)	(0.0415)	(0.00368)
2016	−0.0391**	0.0678	
	(0.0167)	(0.178)	
2018	−0.0492***	−0.198*	−0.136***
	(0.0161)	(0.102)	(0.00966)
2022	−0.0466***	−0.335***	−0.159***
	(0.0161)	(0.101)	(0.00624)
Observations	72	72	55
R-squared	0.260	0.240	0.900

Multivariate linear regression of heavy metals from 2014–2022 for all available samples. Arsenic was not tested as mcg/g in 2014. Robust standard errors in parentheses. ***$p < 0.01$, **$p < 0.05$, *$p < 0.1$.

TABLE 4 Two-sample Welch's T-test for cadmium (mcg/day) for not organic and organic.

Group	Observations	Mean	Std. err.	Std. dev.	[95% conf. interval]
Not organic	22	3.176	0.835	3.914	1.44–4.91
Organic	15	6.09	1.064	4.123	3.80–8.38
Combined	37	4.36	0.690	31.5013	2.96–5.76
Diff	–	−2.91	1.352	–	−5.67−−0.157
T-stat	−2.1560	Pr (T < t)	0.0195	-	

TABLE 5 Pairwise comparison testing of heavy metals by year with BE correction to adjust for multiple comparisons from 2014 to 2022.

Year	Contrast	Bonferroni standard error	t	P > t
Pb (mcg/g) pairwise comparison (batch testing by year)				
2016 vs. 2014	−0.0391111	0.0166747	−2.35	0.132
2019 vs. 2014	**−0.0492426**	**0.0161305**	**−3.05**	**0.020**
2022 vs. 2014	**−0.046642**	**0.0161116**	**−2.89**	**0.031**
2019 vs. 2016	−0.0101315	0.0089885	−1.13	1.000
2022 vs. 2016	−0.007531	0.0091391	−0.82	1.000
2022 vs. 2019	0.0026005	0.0090927	0.29	1.000
Cadmium (mcg/g) pairwise comparison (batch testing by year)				
2016 vs. 2014	0.0677999	0.1781246	0.38	1.000
2019 vs. 2014	−0.1975056	0.1018891	−1.94	0. 341
2022 vs. 2014	**−0.3348801**	**0.1005712**	**−3.33**	**0.009**
2019 vs. 2016	−0.2653055	0.1506569	−1.76	0.497
2022 vs. 2016	−0.40268	0.1496009	−2.69	0.054
2022 vs. 2019	−0.1373745	0.0625568	−2.20	0.190
As (mcg/g) pairwise comparison (batch testing by year)				
2019 vs. 2016	−0.1360507	0.009663	−14.08	<0.0001
2022 vs. 2016	−0.1585439	0.0062441	−25.39	<0.0001
2022 vs. 2019	−0.0224932	0.0097177	−2.31	0.074

Bold values means Bonferroni correction.

to exceed the parameters of conservative IRLs. Further, it should be remarked that there has never been a safe serum level of Pb consumption identified with any value, even those under the 3.5 mcg/day threshold (0.5 IQ points), are associated with adverse effects across a range of systems (30, 31). Indeed, prior authors have demonstrated that the steepness of the slope between IQ and BLL, for instance, may be more prominent at lower serum Pb levels. Thus, any intake of Pb, especially among vulnerable populations, should be heavily scrutinized.

Similarly, while there has yet to be a federal limit identified for Cd consumption, the concentrations found in several samples significantly exceeded multiple regulatory recommendations for safe limits (32, 33). The biological effects associated with Cd consumption, even at levels below regulatory standards, indicate that Cd levels are associated with cardiovascular disease, kidney dysfunction, cognitive deficit, diabetes, cancer, osteoporosis, and a panoply of related conditions (34–46). Therefore, like Pb, the maximum reasonable reduction of dietary exposure, especially for developing children and expectant mothers, is highly advisable.

Though the results of this analysis suggest that the overall consumer market may be, on average, without biologically significant contamination as individual servings, the presence of outliers as well as the potential additive exposure through other foods, such as teas and spices, is potentially problematic (17–23, 47). More interesting was the observation that "organic" products were significantly more likely to demonstrate higher levels of both Cd and Pb. More striking, the number of trade certifications (e.g., Non-GMO, Fairtrade) did not significantly alter the levels of heavy metals found among products surveyed. Thus, even those consumers opting to purchase "higher" quality products retained no exposure protection. Such heterogeneity invites potentially hazardous exposures, especially among biologically susceptible groups, necessitating enhanced surveillance. Cocoa-containing products are a rich source of dietary polyphenols namely flavonoids such as catechins and anthocyanidins, which may be beneficial to health, though the benefits associated with cocoa-containing products have been typically been modest (46, 48, 49, 50).

Since it has been documented that heavy metal contamination of dark chocolate and other cocoa products is chiefly the result of post harvesting contamination, better quality control practices during harvesting and manufacturing may help eliminate the problem.

Conclusion

The results of our analysis suggest that many products contain Pb and Cd in amounts that may exceed certain, stringent regulatory requirements (Prop 65). Therefore, enhanced surveillance may be warranted. Further, additional research into cumulative heavy metal exposure from the diet as a whole would help put this work into context to best inform public health policy and interventions. For instance, if contaminated products as a whole are consumed in small amounts and infrequently by most, these contaminants may not be a public health concern (though, perhaps still an individual concern); in contrast, if many such products are consumed fairly regularly by the average consumer, the additive exposure may be a public health concern. However, high mean contamination and the

presence of outliers with greater contamination should motivate greater testing of all consumer products, particularly cocoa, to better address significant gaps in quality control. Additionally, this must be placed in the context of dietary patterns and public health concern.

Data availability statement

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

Author contributions

JH: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. MA: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. TC: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. JB: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. LF: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

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Conflict of interest

MA, TC and JB were employed by ConsumerLab.com.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

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