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Implications of distribution of cadmium between the nibs and testae of cocoa beans on its marketability and food safety assessment

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RESEARCH ARTICLE

Abstract

There is increasing concern globally, regarding the consumption of foods contaminated with heavy metals such as cadmium (Cd), with consequent implementation of stringent food safety standards for consumer protection. Cd contents of cocoa beans can affect whether shipments of beans can be sold and whether cocoa products manufactured from beans with Cd can meet food safety standards. Cd determinations in bean exports are usually carried out on whole beans, which are comprised of nibs and the covering testae/shells, the latter being impossible to completely remove from the nibs used in chocolate and cocoa powder manufacture. The aim of this study was to evaluate the distribution of Cd levels between the nibs and shells of cocoa beans. This can allow for assessment of possible implications of the analytical protocols used for Cd determination in cocoa beans and the possible consequences for the safety of cocoa products made from such beans. Fermented and dried cocoa bean samples from different cacao-growing areas in Trinidad and Tobago were separated into nibs and shells and analysed for Cd by flame atomic absorption spectrometry, following exhaustive acid digestion. Shells of fermented and dried beans were found to contain significantly higher ($P<0.05$) and on average between 1 to 3 times as much Cd than the cocoa nibs. Analysis of whole cocoa beans, as well as incorporating shells in chocolates can thus result in Cd levels being significantly higher than using nibs only. These finding imply that the analysis of whole beans currently employed to determine their Cd contents, can affect the marketability and prices of cocoa beans. Questions also arise on whether the allowed incorporation of shells in the manufacture of cocoa products can affect their safety for human consumption.

Keywords: cadmium, cocoa beans, cocoa products, food safety, nibs, testae, shells

1. Introduction

Cadmium (Cd), a non-essential and toxic metal, can contaminate foods naturally through uptake from soils. Soils may contain Cd naturally from volcanic activity (Burnfred, 2009), or through human activities, including farming (Scott *et al.*, 2000), manufacture/processing (Burnfred, 2009; Dahiya *et al.*, 2005; Minifie, 1999) or in automobile exhaust emissions (ATSDR, 2004; USEPA, 2003). With agricultural produce being a major source of Cd in foods (Chaudri *et al.*, 2001), its intake by humans is generally unavoidable and may present significant health risks if consumed in highly contaminated foods such as rice (Nogawa *et al.*, 1983; Reilly, 2002; Singh, 2005) and other cereals (Adams *et al.*, 2004; Wieczorek *et al.*, 2005), vegetable (Beccaloni *et al.*, 2013; Nabulo *et al.*, 2011) and shellfish (Olmedo *et al.*, 2013). Consumption of highly Cd-contaminated foods can cause abdominal cramps, headaches, vomiting and diarrhoea (ATSDR, 1993). While only 5-10% of ingested Cd may be absorbed by the body, it may accumulate in some tissues over time, with renal damage being caused by long-term exposure (Harrison, 2001).

Recent research in food safety has raised concerns and actions over the levels of Cd in chocolates and other cocoa-based products. For example, the American Environmental Safety Institute took legal action against chocolate manufacturers for excessive levels of Cd in chocolates (Anderson, 2002).

In addition, the European Commission's recent assessment of Cd in different type of foodstuffs reported chocolate as being highest in Cd (Dickson, 2010). Not surprisingly, the issue of Cd contamination in chocolates and cocoa products is of great public significance (Dahiya *et al.*, 2005; Jalbani *et al.*, 2009; Mounicou *et al.*, 2002, 2003).

In response to such findings, international legislative bodies, as well as chocolate manufacturing countries have introduced new Cd regulations for the protection of the health of their consumers (Ducos *et al.*, 2010; FSA, 2008; ICCO, 2012). While there is as yet no maximum permissible level (MPL) for cocoa nibs, the MPL of Cd in chocolate and cocoa powder set by chocolate producers in Europe are country-dependent, but vary between 0.4 and 0.5 μg/g (Mounicou *et al.*, 2002). However, the EU is currently preparing to implement even more stringent regulations for Cd in cocoa products (EFSA, 2011).The Cd in chocolates and other cocoa-based products have been attributed to cocoa beans used in their production, with Cd in beans likely to originate from soils (Fauziah *et al.*, 2001; Mounicou *et al.*, 2003). Some soils, especially those of volcanic origins, can contain high levels of Cd, which can be taken up by the cacao plant and become concentrated in the beans (Burnfred, 2009).

Cocoa beans are covered by the outer testae or shells, which enclose the cotyledons or nibs, which are used in chocolate manufacture (Fowler, 1999). The Cd content in chocolates is a function of the Cd in cocoa nibs and diminishes in the order: bitter chocolate (used for baking), dark chocolate (bittersweet and semi-sweet), milk chocolate and white chocolate (Mounicou *et al.*, 2002). Although the removal of cocoa shells is a prerequisite for making good quality chocolates (Burnfred, 2009), a percentage of shell material may still be incorporated in chocolates (Fowler, 1999). Since it is difficult to remove all shells during winnowing of roasted beans, Codex allows a maximum of 5% cocoa shell in chocolates and other cocoa products (Codex Alimentarius Commission, 2001). Consequently, if the Cd concentrations of nibs and shells differ, the inclusion of shells in chocolates will influence the Cd contents of the manufactured products.

While the protocol for analysis of cocoa beans does not mention the use of the whole beans, checks with commercial laboratories which provide such analyses for purchasing agents and manufacturers (OLEOTEST, personal communication; Société Générale de Surveillance (SGS) Laboratories, personal communication) have confirmed this practice.

The objective of this investigation was to determine how Cd levels are distributed between the nibs and shells of local cocoa beans obtained all over Trinidad and Tobago, a producer of fine/flavour cocoa beans used for high quality chocolates worldwide (ICCO, 2006). Local cocoa bean producers have already been affected by the determination of Cd in whole cocoa beans, with buyers either rejecting or demanding lower prices for beans deemed to contain excessive levels of Cd (Cocoa and Coffee Industry Board of Trinidad and Tobago, personal communications). The results of this study will also allow local farmers to decide whether buyers offer fair market value of their cocoa beans, as well as to evaluate possible implications of incorporation of shells with nibs in meeting food safety regulations for cocoa products.

2. Materials and methods

All water used for sample preparation and cleaning of glassware in this study was glass-distilled and then deionised. In order to avoid trace metal contamination, laboratory glassware and other utensils used in all analyses were washed with a suitable detergent, soaked in an acid bath of 2 M nitric acid for at least 24 h, rinsed in distilled deionised water and dried in an oven at 50 °C. All reagents used in this study were of analytical grade (Avantor Performance Materials, Center Valley, PA, USA; Sigma Aldrich, St. Louis, MO, USA).

Sampling and sample preparation

Forty-five fermented and dried cocoa bean samples from different cacao-growing areas in Trinidad and Tobago were obtained directly from local farmers and buying agents of beans between 2006 and 2009. Each sack containing cocoa bean samples was thoroughly mixed to provide a uniform sample from which representative 1 kg samples of beans were taken. All samples were placed in plastic bags, tied at the mouths, immediately labelled for identification and then transported to the laboratory for processing. Each batch of cocoa beans was oven-dried in aluminium foil at 105 °C to constant mass (about 5 h) to remove residual moisture. Dried beans were left to cool and later carefully cracked to separate the nibs from the shells, which were ground separately in acid-washed ceramic mortars and pestles to particle sizes <1 mm diameter, mixed well and stored in clean plastic bags for analysis.

Chemical analysis

Portions of each finely divided sample of cocoa nib and shell was weighed out in triplicate (0.5 g) into boiling tubes for analysis. A sample extraction protocol for Cd, previously optimised using ground cocoa nib and shell samples was used for all samples. To each analysis sample, 10 ml concentrated analytical grade nitric acid (J.T. Baker, Center Valley, PA, USA) was added and mixed well in a ventilated fume hood. Sample tubes were covered with clean glass plates to prevent aerial contamination and allowed to pre-digest overnight at room temperature. Reagent sample blanks were prepared simultaneously. The pre-digested samples were then refluxed on a dry-block heater set at 130-135 °C for 6 h, to completely digest samples. Samples were swirled at intervals during the process, to ensure that all the material was digested.

After acid digestion, samples were cooled to room temperature, 5 ml deionised water was added to dilute the contents of each boiling tube and the extract filtered through Whatman no. 542 filters (GE Healthcare Bio-Sciences, Pittsburgh, PA, USA) into 25 ml volumetric flasks (Corning, New York, NY, USA). Each solution was made up to volume with deionised water rinses of the residues and mixed thoroughly. The digests were then analysed for their Cd concentrations by flame atomic absorption spectrometry (FAAS), fitted with high-sensitivity UltrAA hollow cathode Cd lamp (Varian SpectrAA 880; Varian, Mulgrave, Australia). Certified Reference Cd standard solutions (Spectrosol; BDH Ltd., Poole, UK) were used to make up diluted Cd calibration solutions between 0.025 and 1.0 μg/ml shortly before use.

Quality control

The test method used was validated with National Institute for Standards and Technology (NIST) standard reference material, SRM 1570a spinach leaves, since no chocolate or cocoa sample certified for Cd content was commercially available. However, internal quality control materials (IQCM) comprised of finely ground and sieved (<1 mm) local cocoa nibs (IQCM A) and cocoa shells (IQCM B) were analysed together with the NIST SRM, to determine their Cd contents. These two IQCM and SRM 1570a were subsequently analysed in triplicate with every batch of nib and shell samples (n=10), to monitor the consistency of Cd analyses (Table 1 and 2). Sample blanks were also prepared with every batch of samples and nitric acid used, to correct sample readings for any background levels of Cd in reagents, filters or water used. Calibration check standard solutions (0.1 and 0.5 μg/ml) were analysed at regular intervals (at least 1 in every 10 samples) during sample analyses, to ensure consistent responses of the FAAS.

3. Results and discussion

The results in Table 1 demonstrate that the Cd analysed in SRM 1570a compared well with its certified value, with a 102% recovery, over 10 × 3 analyses of this sample over a period of 12 months. The limit of quantitation of the test method was calculated (IUPAC, 1997) as 0.3 μg/g based on the standard deviation of sample blank measurements of a 0.5 g sample made up to 25 ml. Similarly, mean Cd concentrations of IQCM A (Nibs) and IQCM B (Shells) under the same conditions (Table 2) were highly consistent, with relative standard deviations (%RSD) of pooled analyses of each sample being all less than 5%, indicating acceptable precisions of analyses for Cd levels in the SRM and IQCM cocoa nib and shell samples.

Table 2. Mean cadmium concentrations (μg/g) from 10 batches of IQCM A (cocoa nibs) and IQCM B (cocoa shells).

Analyte	Sample	Analysis mean ± %RSD
Cd	IQCM A	1.56±3.85
	IQCM B	5.62±3.74

RSD = relative standard deviation.

Variations of cadmium concentrations between nibs and shells of cocoa bean samples

Detectable Cd concentrations (>0.3 μg/g) in nibs and shells of fermented and dried cocoa beans samples analysed are summarised in Table 3. Paired data analysis (Minitab, 2010) showed that Cd concentrations between nibs and shells differed significantly (P<0.05) over the concentration ranges of 0.5 to 2.34 μg/g for nibs and 1.07 to 4.58 μg/g for shells. Correlation analysis (Figure 1) similarly demonstrated a positive and significant (P<0.05) correlation of 0.63 between the Cd concentrations of the fermented and dried cocoa nibs and shells, with cocoa shells of fermented and dried beans containing between 1 to 3 times as much Cd as the cocoa nibs.

These findings are in agreement with previous reports (Lee and Low, 1985; Mounicou *et al.*, 2003), which indicated that Cd is preferentially bound to the shell of the cocoa bean, but were reported for only a narrow concentration range, whereas our results show a linear relationship between nibs and shells of cocoa beans over a wide concentration range. Thus if whole beans (nibs + shells) are analysed, the Cd values are likely to be over-estimated. While an MPL

Table 1. Mean cadmium concentration (μg/g) and % recovery from 10 batches of certified reference material SRM 1570a.

Analyte		Certified value ± %RSD (μg/g)	Analysis mean (n = 10 × 3) ± %RSD (μg/g)	% recovery ±%RSD
Cd	SRM 1570a (spinach leaves)	2.89±2.42	2.72±1.47	102.03±3.82

RSD = relative standard deviation.

Table 3. Total cadmium concentrations (µg/g; ± standard deviation) in nibs and shells of cocoa bean samples.

Sample	Nibs (n=3)	Shells (n=3)
1	1.15±0.24	2.26±0.15
2	1.26±0.31	2.77±0.16
3	0.64±0.01	1.11±0.23
4	1.36±0.49	2.46±0.12
5	0.50±0.11	1.07±0.03
6	1.19±0.36	3.37±0.17
7	1.10±0.20	1.59±0.10
8	1.84±0.55	2.01±0.53
9	0.90±0.15	1.59±0.04
10	1.42±0.06	2.37±0.11
11	0.77±0.13	1.11±0.03
12	0.92±0.11	1.04±0.21
13	0.76±0.04	1.97±0.18
14	2.00±0.28	5.24±0.06
15	0.50±0.04	1.36±0.24
16	1.23±0.14	3.56±0.33
17	0.75±0.12	1.63±0.04
18	1.18±0.21	2.40±0.29
19	0.51±0.08	1.71±0.07
20	0.92±0.07	0.96±0.06
21	0.71±0.04	1.67±0.09
22	0.56±0.01	1.23±0.09
23	0.55±0.15	0.99±0.15
24	1.06±0.08	1.49±0.08
25	0.50±0.18	0.92±0.06
26	0.51±0.40	0.94±0.02
27	0.61±0.15	1.69±0.76
28	2.34±0.25	4.58±0.07
29	0.50±0.24	1.24±0.07
30	0.99±0.08	2.58±0.07
31	0.66±0.10	1.28±0.10
32	0.89±0.15	0.92±0.05
33	1.10±0.99	3.19±0.25
34	0.50±0.09	1.87±0.09
35	1.36±0.41	3.13±0.36
36	1.86±0.12	4.72±0.03
37	0.86±0.01	3.85±0.28
38	0.61±0.07	1.37±0.13
39	0.64±0.11	1.46±0.12
40	0.52±0.04	1.16±0.06
41	1.25±0.13	2.40±0.08
42	1.75±0.16	3.45±0.33
43	0.65±0.08	1.47±0.11
44	0.93±0.02	2.60±0.15
45	1.14±0.13	2.34±0.34

for cocoa nibs has not yet been set, MPL for edible cocoa products (chocolates, powders) do exist. Consequently, based on the weight percentage nibs incorporated in cocoa products, buyers of cocoa beans and cocoa product manufacturers can estimate the Cd concentrations in the final products, using concentration values of Cd of beans and may use such values to bargain for lower prices of beans from suppliers, as has been experienced locally. Since the shells comprise 11 to 12% of the weight of whole cocoa beans, whole bean analyses can make the determined Cd values significantly higher than the nibs alone, to the detriment of cocoa producers.

Cocoa beans from Malaysia have been reported to contain between 0.29 to 1.09 µg/g Cd (Fauziah *et al.*, 2001), but it was not clear whether whole beans or nibs were analysed. Nevertheless, similar implications may exist for other cocoa-producing regions, especially with those with volcanic soils such as Ecuador, Venezuela (Burnfred, 2009) and Papua New Guinea (Wood, 2008).

Food safety implications for nib and shell cadmium distribution

At present, some European Union (EU) countries have adopted a MPL of 0.4-0.5 µg/g for Cd in chocolates (Mounicou *et al.*, 2002), but have not yet imposed a limit for cocoa beans (nibs assumed). Such countries are primary markets for export of local beans, which are sold as 100% fine/flavour beans (ICCO, 2006) primarily used in the production of fine or single-origin dark chocolates. Dark chocolates are mandated by the EU to contain no less than 35% cocoa nib solids in their production (EC, 2000). However, Codex allows up to 5% cocoa shell in chocolates and other cocoa products, due to the difficulty of totally removing shells during winnowing (Codex Alimentarius Commission, 2001).

Based on a minimum of 35% cocoa nib incorporation in dark chocolates, it is estimated that only 10% of the samples analysed can produce chocolate Cd concentrations greater than the MPL of 0.5 µg/g. On the other hand, if 5% of cocoa shells of the samples analysed in this study were incorporated with their nibs for the production of chocolates, up to 50% of these could have contained Cd levels above this MPL. This has clear implications for cocoa products meeting food safety standards if Cd-containing beans are used and can result in the decline in cocoa farming in areas affected by Cd in soils.

4. Conclusions

This study has demonstrated that the distribution of Cd between nibs and shells can result in analysed values in whole cocoa beans being significantly higher than in the nibs. This may have adverse and unfair effects on the prices of cocoa beans and the economic viability of cocoa farms, as well as for the food safety of cocoa products, based on their current MPL. Such distributions may affect whether

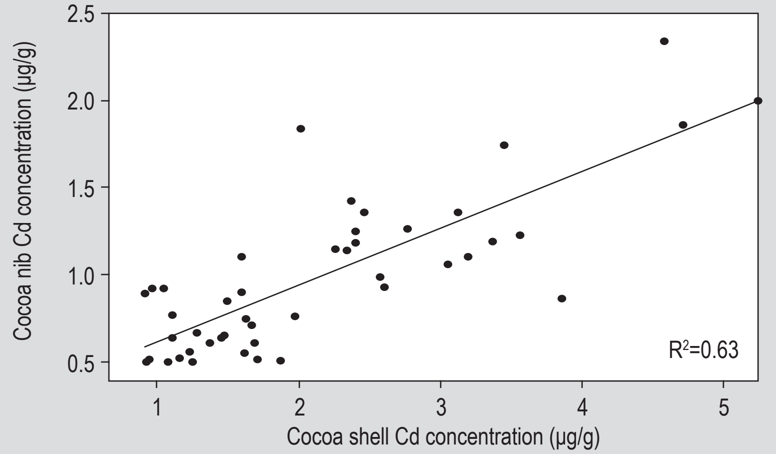

Figure 1. Correlation-plot of cadmium concentrations in fermented and dried cocoa nibs versus shells.

cocoa products can meet food safety standards and may also have a negative impact on the exports of cocoa beans and the welfare of cocoa farmers. It is necessary that regulators of Food Safety Standards as well as chocolate manufacturers be aware of such Cd distributions. If they are not, it is quite likely that cocoa samples from producing areas affected by Cd contamination may not meet the international standards and this can seriously impact on cocoa production, particularly in fine/flavour producing countries and put at stake the livelihoods of many cocoa farmers. Modification of the current protocols used by manufacturers and food safety regulatory bodies to assess Cd levels in cocoa is recommended, to protect the interests of all stakeholders in the cocoa industry. The removal of the testae from cocoa used in the manufacture of products for consumption is also strongly advocated, to safeguard the health of consumers of cocoa products.

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