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Measurement of Heavy Metals in Commercially Available Soybean and Palm Oils and Relevant Health Risk Assessment in Bangladesh

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Background: Soybean and palm oils are widely consumed in Bangladesh.

Objective: The study aimed to investigate the levels of heavy metals and estimate their health risks in nationally representative samples of branded and unbranded soybean and palm oils sold in retail stores in Bangladesh.

Methods: A total of 1,521 soybean and palm oil samples were collected from eight administrative divisions. National composites of branded oil were prepared by combining at least 12 samples for each brand. In the case of unbranded oil, composites were prepared for each administrative division. A total of 44 composite samples, including 23 soybean oil samples (19 branded and 4 unbranded) and 21 palm oil samples (13 branded and 8 unbranded), were tested. Twenty-five individual samples (11 crude and 14 refined) collected from the refineries were also analyzed to trace the origin of the heavy metals. Market samples were analyzed for arsenic (As), cadmium (Cd), chromium (Cr), and lead (Pb) and only mercury (Hg) in both market and refinery samples using various atomic absorption spectrophotometric techniques. The possible adverse health effects of exposure to heavy metals content in edible oil were estimated using the tools of daily exposure (D_E) and noncarcinogenic risk assessment hazard quotient (HQ).

Results: The median values of As, Cd, Cr, Pb, and Hg content in soybean and palm oil respectively ranged between 6.9 and 8.8 μg/kg (As), 4.3 and 6.9 μg/kg (Cd), 12.3 and 42.3 μg/kg (Cr), 19.4 and 27.8 μg/kg (Pb), and 1.73 and 5.11 mg/kg (Hg). The differences in heavy metal contents between branded and unbranded oils were not statistically significant. Except for Hg, all other metal concentrations were within national and international standard limits. The estimated D_E of Hg through edible oil represented a considerable risk for noncarcinogenic health effects (HQ > 1). The ranking orders of HQ for the oil samples were as follows: unbranded soybean oil (3.99) > branded soybean oil (3.50) > branded palm oil (2.61) > unbranded palm oil (1.69).

Discussion: The present study evaluated the level of heavy metal contamination in soybean and palm oils and conducted risk assessments associated with their consumption in Bangladesh. It appears that the source of this contamination is the imported crude oil. Strong and effective monitoring infrastructure is needed to regulate the import of safe crude oils for refineries. https://doi.org/10.1289/JHP1072

Introduction

Edible oil from plant sources is an indispensable part of the normative diet all over the world. Given its versatile functions in providing essential fatty acids, fat-soluble vitamins, and health-promoting phytosterols, vegetable oil is acknowledged as a safe and wholesome source of dietary fat.[1] Vegetable oils are also preferred over animal fat for their absence of cholesterol.[2] According to a recent report, worldwide consumption of vegetable oil was 210.30 million metric tons in 2022–2023.[3] National surveys have reported that in Bangladesh, vegetable oil consumption increased from 20.7 g/d in 2010 to 27.0 g/d in 2016,[4] with soybean and palm oils being the two most widely consumed edible oils in the country. According to the UN Food and Agriculture Organization (FAO), soybean and palm oils respectively account for 20% and 70% of edible oil consumption in Bangladesh.[5] A more recent study also reported soybean and palm oils to be the predominant edible oils in Bangladesh.[6] Edible oil in Bangladesh is distributed either in labeled containers as branded oil or as bulk oil in unlabeled large containers or drums, which is considered to be unbranded oil. A previous study reported that unbranded oil accounts for ~65% of the overall edible oil market share and is consumed by more than two-thirds of the population, primarily because of its affordability.[5]

A specific concern about vegetable oils is that they readily react with substances such as oxygen and heavy metals.[2] Therefore, it is important to analyze the presence of various heavy metals as part of the assessment of edible oil quality with respect to freshness, tolerability, rancidity, and toxicity. Heavy elements, such as arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), mercury (Hg), and nickel (Ni), are known to be toxic. Being nonbiodegradable, and in the absence of an adequate mechanism of elimination, they accumulate in various parts of the body, particularly in adipose tissue.[7] The long biological half-lives of these elements and their ability to persist in human tissues without degradation, leading to gradual accumulation over time, render them especially harmful.[8] Accumulated toxic heavy metals have been implicated in cancer, cardiovascular, kidney, neurological, and bone diseases.[9]

Previous studies have reported that heavy metals can enter the food supply chain via contaminated water, soil, and fertilizer and accumulate in various parts of plants.[10] A study in southern Nigeria reported the bioaccumulation of metals in palm oil.[11] The study emphasized the correlation between soil metal accumulation and their presence in palm oil, specifically

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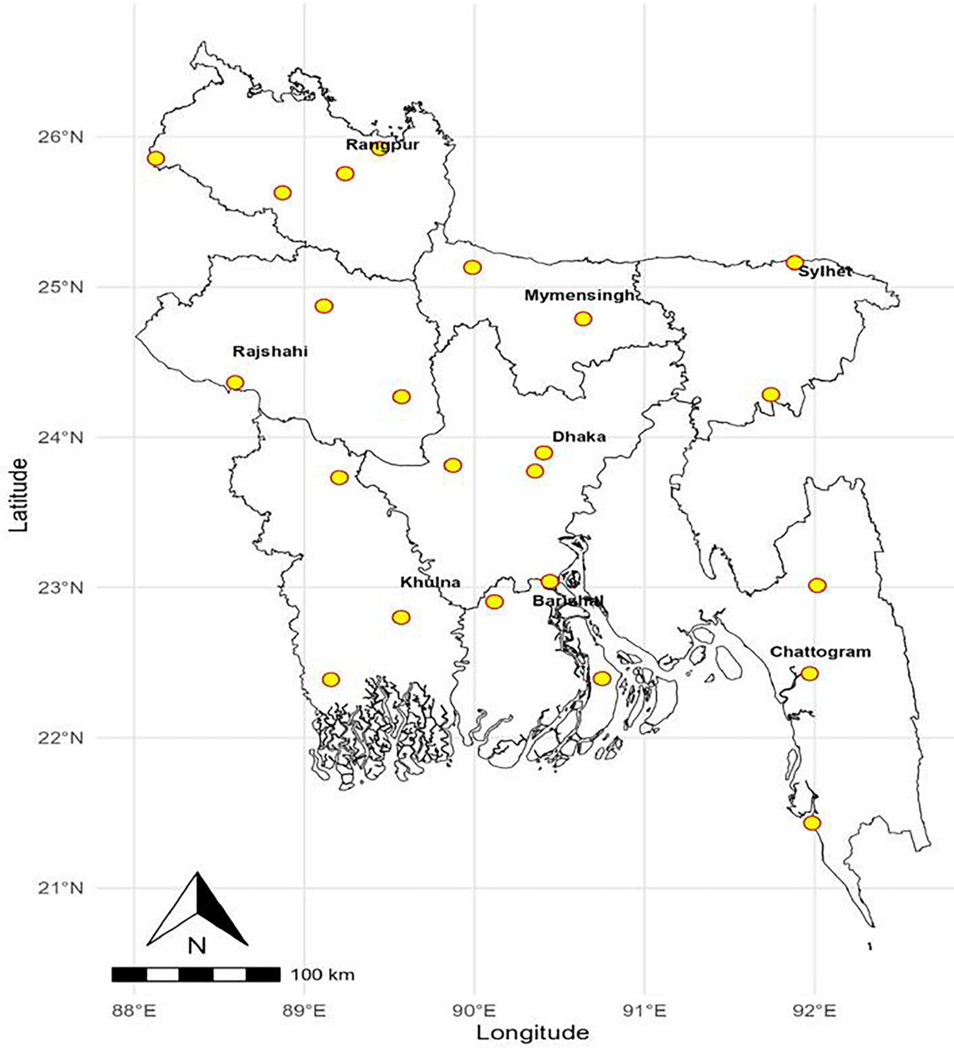

Figure 1. Map showing the districts ($n = 23$) from where edible oil samples were collected. This map was generated by plotting GPS points of the data and sample collections spots in a map of Bangladesh [R software (R version 4.1; R Development Core Team)]. The map was obtained from an open source and no permission is required. Note: GPS, global positioning system.

highlighting potential health risks from elevated levels of Cd and Cr in contaminated palm oil.[11] In earlier Bangladeshi studies, the presence of As, Cd, and Pb was reported in cereals, vegetables, fruits, milk, and fish samples.[12–14] Countries worldwide have confirmed the presence of different heavy metals in edible oil.[15] Recently, heavy metals were detected in nine different vegetable oils from China.[2] Dietary exposure to high levels of heavy metals and minerals may be associated with serious public health issues.[16] Consequently, the increase in consumption of metal-contaminated edible oil increases the risk of health hazards.[16] However, no study in Bangladesh has yet been conducted to measure the presence of heavy metals in edible oils and their health implications.

The objective of the present study was to evaluate the contents of selected toxic heavy metals (As, Cd, Cr, Pb, and Hg) in commercially available branded and unbranded soybean and palm oils collected from across the country. In addition, the study estimated the potential health risks exerted by the heavy metals present in edible oil according to the guidelines of the US Environmental Protection Agency (EPA).[17] The compliance of the heavy metal contents measured in the oil samples with the national guidelines set by the Bangladesh Standard and Testing Institute (BSTI) is also assessed in this study.[18]

Methods

Oil samples were collected from the whole country, as well as from various stages of the refining, supply chain, and marketing processes. Details of the sample collection, preparation, and analysis are described below.

Sample Collection and Preparation of Composite Samples

Soybean and palm oil samples were collected from all over the country. Ninety urban and rural study clusters, each comprising 250–300 households, were previously selected for a food security and nutrition surveillance study and are described in multiple previous articles.[19,20] From the aforementioned clusters, we randomly selected 70 clusters (29 urban and 41 rural) in 23 districts (Figure 1), covering all eight administrative divisions of Bangladesh. Grocery shops located within or nearest to the selected clusters were identified by the study team by walking along the main road of the clusters. Whenever multiple grocery shops were identified, we selected the ones selling more varieties of edible oil. The willingness of the shop owners or salespersons to share data was also taken into consideration in selecting the shops. A total of 1,521 branded and unbranded soybean and palm oil samples were collected. The samples were collected in two rounds: *a*) during the winter (February–March 2021), and *b*) during the summer (June–August 2021).

The sample collection and preparation methods have been described previously in a recently published article.[21] In the case of branded oil, the vendor-defined top (i.e., most sold) three brands were collected from each vendor. The smallest available pack-size bottles were purchased from the shops, placed inside black polyethylene bags, and wrapped tightly to avoid light and sun exposure. Unbranded samples were collected from the same selected shops in two 50-mL Falcon tubes wrapped with black masking tape to prevent light exposure. Samples were transferred to the Institute of Nutrition and Food Science, University of Dhaka, for further processing. Composite samples were prepared to reduce the time and cost required for analyzing large numbers of individual samples. National composites of branded oil were prepared by pooling together an equal volume of at least 12 samples of each brand to make one composite, according to the guidelines of the US Department of Agriculture's National Food and Nutrient Analysis Program.[22] The guidelines were also followed for the development of the Food Composition Table for Bangladesh.[23] In the case of unbranded oil, equal volumes of all available samples collected from both retail and wholesale stores were used to prepare the composite samples. Upon analysis of the market samples, we found that the concentrations of all heavy metals, except Hg, were within the national standard limits. To identify the source of Hg contamination, we collected 25 individual samples, both crude and refined, from five refineries with a major market share, as identified through the market survey conducted in this study. A detailed description of the sampling procedure is provided in Figure S1, which was adapted from a recently published article.[21]

Sample Preparation

The edible oil samples were mineralized in an acid medium prior to analysis. Each $\sim 0.5 \pm 0.1$ g oil sample was placed in a Teflon-coated digestion vessel with concentrated nitric acid (HNO_3; 65%) and hydrogen peroxide (H_2O_2; 30%) in a ratio of 5:2. The digestion was performed in a microwave digester (Ethos One, Milestone) at 180°C for 45 min. The digestate was further diluted using Class I (18 MΩ) deionized water to make the appropriate volume.[24] (All reagents employed in the sample preparation for heavy metal analysis were of Analar grade from Merck.)

Determination of Heavy Metals

Heavy metals were assayed by atomic absorption spectrophotometry (AA-7000, Shimadzu) using three separate atomization techniques. The graphite furnace atomizer (model GFA-7000A, Shimadzu) was used for the analysis of Cd, Cr, and Pb. Digested samples were treated with matrix modifiers solution [0.2% wt/vol ammonium dihydrogen phosphate ($NH_4H_2PO_4$), 0.5% vol/vol Triton X-100, and 0.2% vol/vol HNO_3 in deionized water], before being injected into graphite tubes. As was assayed using the technique of the generation of arsine gas (AsH_3) through the hydride vapor generator (HVG-1, Shimadzu). Briefly, 10 mL of acidified samples were first treated with 1 mL of 5 M hydrochloric acid (HCl) and 1 mL of 20% potassium iodide (KI). The treated samples were then heated in a water bath at 80°C for 30 min to reduce the As(V) to As(III). The treated samples were then introduced into the HVG with a continuous flow (flow rate 0.8 mL/min) of 1 M HCl (32%, Merck) and 0.4% sodium borohydride ($NaBH_4$; >98%, Sigma-Aldrich) in 0.1% sodium hydroxide (NaOH) to convert the As(III) to AsH_3 in the reaction coil. Argon gas was used to carry the AsH_3 into the absorption cell for atomization.[25] Hg in the digested edible oil sample was determined by cold-vapor atomic absorption spectrometry with a Hg vaporizer unit (MVU-1A, Shimadzu). In this method, elemental Hg vapor was generated by applying 10% tin(II) chloride dihydrate ($SnCl_2 \cdot 2H_2O$; Scharlau) to the acidic sample.[26] The Hg vapor was flown through the quartz absorption cell by continuous aeration after passing through a drying column of magnesium perchlorate [$Mg(ClO_4)_2$; Sigma-Aldrich].

All analytical parameters were validated in the laboratory prior to the routine analysis of the samples. The National Institute of Standards and Technology (NIST) traceable certified reference materials (CRMs), Trace CERT ($1{,}000 \pm 4$ mg/L of each As, Cd, Cr, and Pb) from Sigma-Aldrich and AAHG1 ($1{,}000 \pm 10$ mg Hg/L) from Inorganic Ventures were used for the validation and daily calibration of the analytical methods. The recovery of the heavy metals during digestion and the reproducibility of the measurements were validated by analyzing NIST Standard Reference Material (NIST SRM 1568b: rice flour, and NIST SRM 1849a: infant/adult nutritional formula) during the method validation. Because edible oil-based reference materials are not available for heavy metals, rice flour and infant/adult nutritional formula were used as reference materials. Both the rice flour and the infant/adult nutritional formula were digested in the same way as edible oil samples. When reference materials were not available for the same matrix as the sample, different matrix-based reference materials were used, as suggested by AOAC[27] and in an earlier paper.[28] The limits of detection (LODs) were calculated from the slopes of the corresponding calibration plots and found to be 0.36 μg As/L, 0.015 μg Cd/L, 0.19 μg Cr/L, 0.16 μg Pb/L, and 0.1 μg Hg/L. Limits of quantitation were 1.00 μg As/L, 0.50 μg Cd/L, 0.50 μg Cr/L, 1.00 μg Pb/L, and 0.3 μg Hg/L. The validation data for the heavy metals are shown in Tables S1 and S2.

Quality Control of Analysis

The internal quality control was strictly followed to ensure the integrity of the data. A 4-point calibration curve was prepared in every lot from the CRM Trace CERT (1,000 mg/L, Sigma-Aldrich), which is traceable to NIST. A mid-range calibration standard was run in every set of 15 analytical measurements in the entire analytical series to confirm the calibration slope and to ensure the overall consistency of instrument responses. In addition, reagent blanks, digested reagent blanks, in-house standards, and standard spikes of oil samples were also run during the day-to-day analytical activities to evaluate the data consistency. The quality control data for heavy metal analysis are shown in Table S1 for As, Cd, Cr, and Pb and in Table S2 for Hg analysis.

Health Risk Assessment of Heavy Metals in Edible Oil

The possible adverse health effects of exposure to heavy metal content in edible oil were estimated using the tools of exposure assessment and noncarcinogenic risk assessment and expressed as hazard quotients (HQs). The daily exposure (D_E) or estimated daily intake of the population and the HQ were estimated using the formulas shown in Table 1. The estimation of D_E was calculated using the average consumption of edible oil by the Bangladeshi population as reported by the Household Income and Expenditure Survey of Bangladesh. A $HQ \geq 1$ indicates that there might be an adverse health risk associated with the oral exposure of toxic metals. On the other hand, an $HQ < 1$ indicates that there is no adverse health effect.

Number of Samples Exceeding the Standardized Limit

The standardized limits set by both the international institute Codex Alimentarius Commission (CAC)[29] and the national institute BSTI[18] were used to determine which samples exceeded the acceptable limit for As, Cd, Pb, and Hg. Cr levels in oil samples

Table 1. Health risk assessment of heavy metals in edible oil.

Indexes	Calculation	Purpose
D_E or EDI of the population	$D_E = \frac{C_M \times IR_{Oil}}{BW}$ mg/kg BW per day[a]	Assess heavy metal-induced health risk through the consumption of oil
HQ	$HQ = D_E/RfD$	Identify noncarcinogenic health risks for each metal through oil consumption

Note: The average intake of edible oil is 0.027 kg/person-day according to the 2016 Household Income and Expenditure Survey in Bangladesh.[4] The RfD of the individual elements As, Cd, Cr, and Pb are 0.0003, 0.001, 1.3, and 0.007 mg/kg BW per day, respectively.[17] According to the Joint Food and Agriculture Organization/World Health Organization Expert Committee on Food Additives, the RfD of mercury is considered the provisional tolerable weekly intake, which is 4 μg/kg BW.[17] BW, body weight; C_M, concentration (mg/kg) of metals in edible oil; D_E, daily exposure; EDI, estimated daily intake; HQ, hazard quotient; IR_{Oil}, intake rate (mg/day) of edible oil; RfD, reference dose for orally consumed metals, expressed as mg/kg BW per day.
[a]Considering an average body weight of 60 kg per person.

were compared with World Health Organization (WHO)[30] and US EPA guidelines for drinking water[31]; standard limits of Cr for edible oil or relevant commodities were not available in the Codex or BSTI guidelines. BSTI limits for As, Cd, Pb, and Hg in edible oil are 0.1 mg/kg, 1.0 mg/kg, 0.1 mg/kg, and 0.25 mg/kg, respectively.[18] Codex limits for As and Pb for edible fat and oil are 0.1 mg/kg and 0.08 mg/kg, respectively. The Codex limit for both Cd and Hg for the relevant commodity is 0.1 mg/kg.[29] The WHO and the US EPA limits for Cr in drinking water are 0.05 and 0.1 mg/kg, respectively. (A standard limit for Cr for edible oil or relevant commodities was not found in Codex or BSTI guidelines.[30,31])

Statistical Analysis

The data are presented as the median and interquartile range (IQR). Differences between branded and unbranded samples were observed by performing the Mann–Whitney *U*-test. Values were considered significant at the $p < 0.05$ level. Data analysis was performed using SPSS (version 26.0; IBM).

Results

Figure 2 and Table S3 show an overview of the average heavy metal (As, Cd, Cr, and Pb) levels in the analyzed market oil samples ($N = 44$). The Cr levels (in micrograms per kilogram) in branded palm oils were significantly higher than in unbranded samples (palm branded vs. unbranded: 38.0 vs. 33.3 μg/kg; $p < 0.05$). Although the levels of Cr in the branded soybean oil were higher than in the unbranded ones, the differences were not statistically significant (soybean branded vs. unbranded, Cr: 42.3 vs. 32.2 μg/kg; $p > 0.05$). The levels of As, Cd, Pb, and Hg were lower in branded soybean oil than the unbranded soybean oil; however, none of these variations were statistically significant.

Figure 3A and Table S3 show the average Hg levels for market samples ($N = 44$), and Figure 3B and Table S4 show Hg levels for refinery samples ($N = 25$). Table S4 also shows that there was no statistically significant difference based on refining. In the overall analysis, the mean Hg content in crude soybean oil samples was higher than in refined samples (crude vs. refined soybean: 2.4 vs. 1.8 mg/kg; $p > 0.05$). In palm oil, the Hg concentration in refined samples (1.8 mg/kg) exceeded that in crude samples (1.5 mg/kg).

Table 2 provides a comparison between the heavy metal concentrations found in our market oil samples and those reported in previous studies conducted in different countries. In our soybean oil samples, the ranges of Cd, Cr, and Pb contents were 0.0027–0.0083 mg/kg, 0.0235–0.1680 mg/kg, and 0.0102–0.0424 mg/kg, respectively. These values were lower than those reported in earlier studies, which ranged from 0.0019 to 0.0480 mg/kg, 0.018 to 1.7559 mg/kg, and 0.0068 to 0.1631 mg/kg for Cd, Cr, and Pb, respectively. Similarly, we found the average concentrations of Cd, Cr, and Pb in palm oil samples to be 0.0053 mg/kg, 0.0363 mg/kg, and 0.0238 mg/kg, respectively, which again were respectively lower than the values reported in

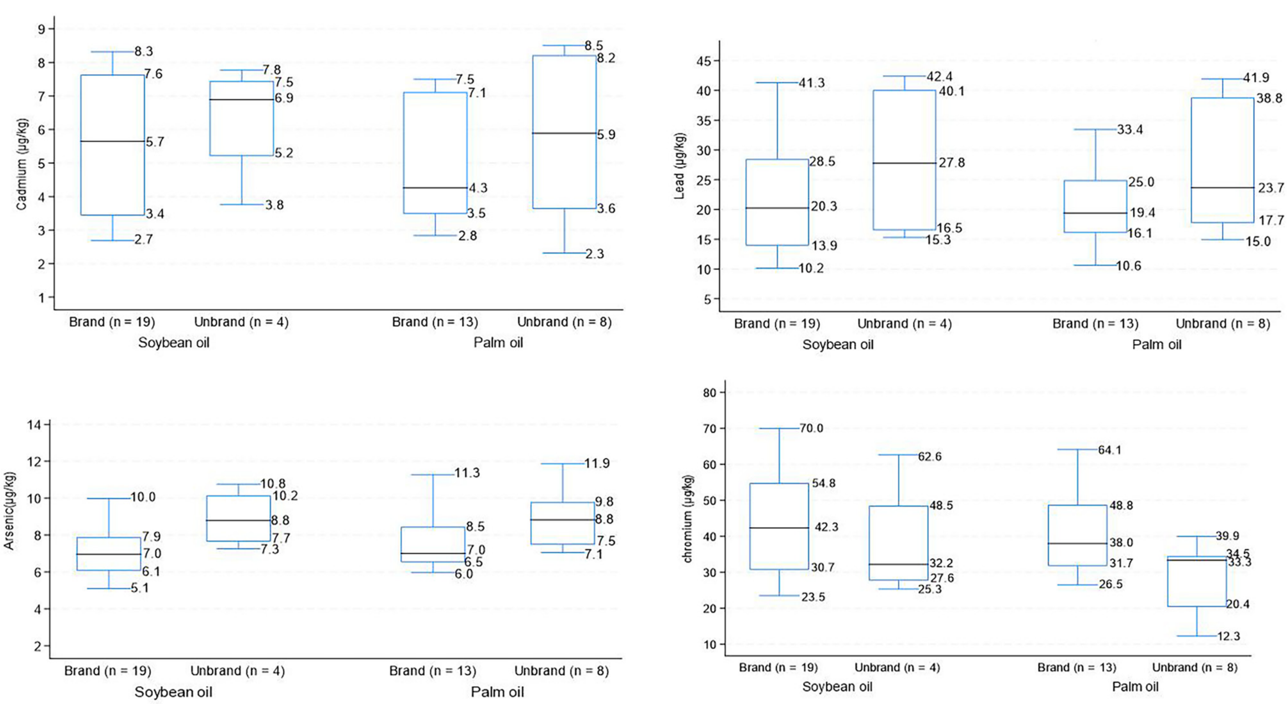

Figure 2. Heavy metal concentration (Cd, Pb, As, Cr) in edible oil (market sample). Whiskers represent range, box limits represent the interquartile range (IQR), and midline of the box is the median. Note: As, arsenic; Cd, cadmium; Cr, chromium; Pb, lead.

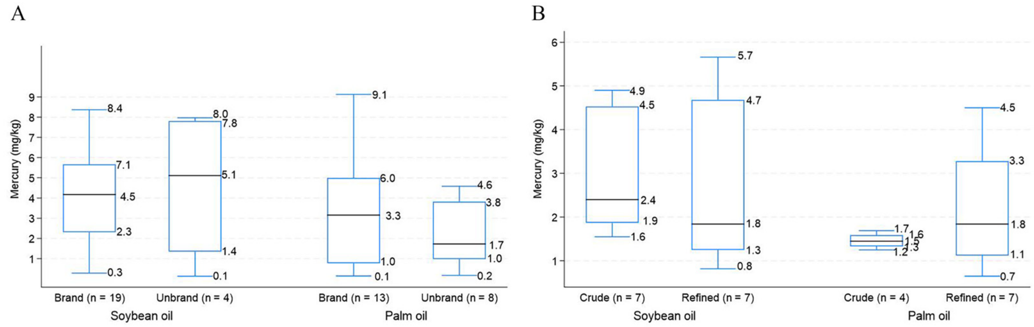

Figure 3. Mercury concentration in edible oil samples: (A) market sample, (B) refinery sample. Whiskers represent range, box limits represent the interquartile range (IQR), and midline of the box is the median.

earlier studies (0.0220 mg/kg, 2.3319 mg/kg, and 0.1780 mg/kg). The average values of As in our soybean and palm oil samples were 0.0078 mg/kg and 0.0084 mg/kg, respectively, which were lower than the levels found in earlier studies (i.e., 0.0180 mg/kg and 0.8200 mg/kg, respectively). The average Hg levels in both soybean and palm oil samples in our study were 5.30 mg/kg and 3.40 mg/kg, respectively, whereas previous studies reported Hg levels below the LOD for both soybean and palm oil samples.

Figure 4 presents the compliance of Hg concentration with the BSTI-established limit of 0.25 ppm.[18] All of the branded soybean oil samples and 92.3% (12 of 13) of the palm oil samples exceeded the limit. Unbranded oils showed a slightly lower percentage exceeding the BSTI-set limit of 0.25 ppm[18] with unbranded soybean vs. palm: 75.0% (3 of 4) vs. 87.5% (7 of 8) compared with branded soybean vs. palm: 100.0% (19 of 19) vs. 92.3% (11 of 12). Compared with the Codex limit for Hg (0.1 mg/kg),[29] all samples exceeded the permissible limit (Table S5). Table S5 shows that the concentrations of As, Cd, and Pb were within Codex-[29] and BSTI-approved[18] safe limits and that Cr concentrations were within the US EPA maximum limit,[31] but not always within the WHO limit.[30]

Estimated Risk Assessment of Heavy Metals

Table 3 presents the estimated levels of D_E to the analyzed heavy metals through consumption of the examined oils. The median D_E of Hg was higher than that of other analyzed elements in both soybean and palm oil. Median D_E to Hg was observed to be higher through the intake of unbranded soybean oil compared with branded oil (soybean oil branded vs. unbranded: 2.00×10^{-3} vs. 2.28×10^{-3} mg/kg BW per day). However, a reverse scenario was observed in palm oil (palm oil branded vs. unbranded: 1.49×10^{-3} vs. 7.69×10^{-4} mg/kg BW per day).

The estimated HQ of As, Cd, Cr, and Pb did not indicate a significant risk of adverse health effects. However, as presented in Table 4, the estimated D_E of Hg through edible oil represented a considerable risk for noncarcinogenic health effects (HQ > 1). The ranking orders of HQ for the oil samples were as follows: unbranded soybean oil (3.99) > branded soybean oil (3.50) > branded palm oil (2.61) > unbranded palm oil (1.35).

Discussion

To our knowledge, this study represents the first examination of the presence of heavy metals in soybean and palm oils available across Bangladesh for consumption. This research aimed to assess concentrations of heavy metals (As, Cd, Cr, Pb, and Hg) in composite samples of branded and unbranded soybean and palm oils from retail markets nationwide, along with Hg assessment in a limited number ($n = 25$) of refined and crude samples collected from refineries to find the source of contamination. The findings revealed that levels of As, Cd, Cr, and Pb were within the acceptable limits set by national and international standards. However, the concentration of Hg in both soybean and palm oil exceeded the limits set by Codex and BSTI, irrespective of the kind of packaging and refinement extent. This elevated level of Hg in the oil suggests potential adverse health effects among the Bangladeshi population.

Because of the potential adverse impacts on human health, exposure to environmental toxic metals of hydrogeochemical origin, such as As, Cd, Cr, Pb, and Hg, has recently gained attention from the public health community worldwide.[38,39] The concentrations of As, Cd, Cr, and Pb in our oil samples were lower than those reported in previous studies conducted in different countries.[32–37] These variations could be due to differences in agricultural practices[10,11] or to the levels of heavy metals present in the soil, influenced by anthropogenic or geological phenomenon[40] in the studied countries or the countries from which the edible oils were originally imported. Other factors, such as differences in refining, packaging, or handling practices between the studied countries and Bangladesh may also have contributed to these variations.

The concentrations of As, Cd, and Pb fell within the limits established by the Codex Alimentarius.[29] However, the study noted that there is no established standard limit for Cr in edible

Table 2. Heavy metal concentration found in soybean and palm oil in various studies.

Heavy metals	Standard value[29] (mg/kg)	Our findings in market samples Average (range) (mg/kg)		Heavy metal content (mg/kg) Average (range) found in other studies (mg/kg)		Country
		Soybean oil ($n = 23$)	Palm oil ($n = 21$)	Soybean oil	Palm oil	
Pb	0.08	0.0228 (0.0102–0.0424)	0.0238 (0.0106–0.0488)	0.0068–0.1631	0.1780	Turkey,[32] Nigeria,[33] Iran[34]
Cd	0.1	0.0057 (0.0027–0.0083)	0.0053 (0.0023–0.0085)	0.0019–0.0480	0.0220	Turkey,[32] Nigeria,[33] Iran[34]
Hg	0.1	5.3000 (0.1300–44.300)	3.4000 (0.1400–23.200)	<LOD	<LOD	Spain,[35] Brazil[36]
Cr	NA	0.0426 (0.0235–0.1680)	0.0363 (0.0123–0.0641)	0.018–1.7559	2.3319	Nigeria,[33] China,[37] Spain[35]
As	0.1	0.0078 (0.0051–0.0168)	0.0084 (0.0060–0.0145)	0.0150	0.8200	China,[37] Brazil[36]

Note: As, arsenic; Cd, cadmium; Cr, chromium; Hg, mercury; LOD, limit of detection; NA, not applicable; Pb, lead.

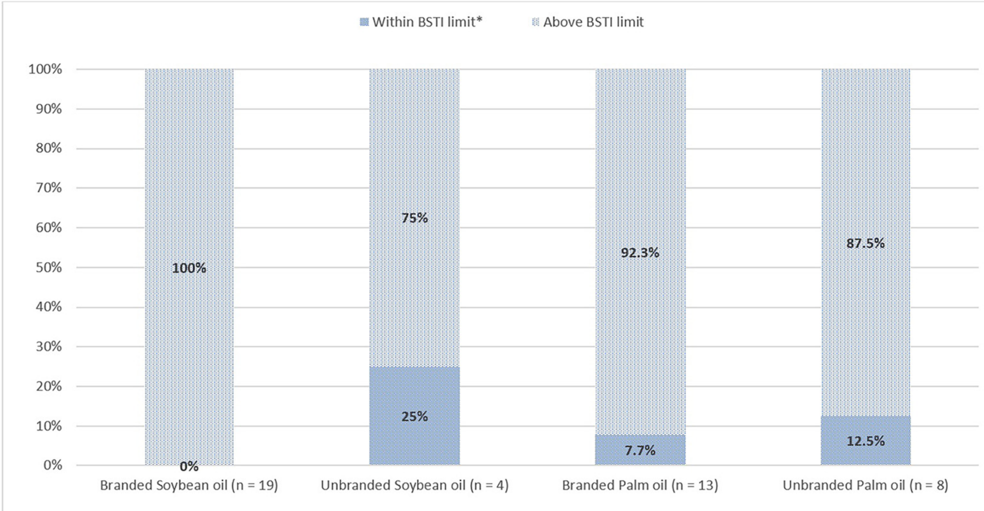

Figure 4. Hg in the analyzed composite market oil samples exceeding the Bangladesh Standard and Testing Institute (BSTI) level. *The BSTI limit for total Hg (0.25 ppm); therefore, "within BSTI limit" represents the number of samples having Hg levels <0.25 ppm. Note: Hg, mercury.

oil or relevant commodities according to Codex guidelines. The Cr levels in a quarter of the samples exceeded the maximum allowable concentration (MAC) for drinking water set by the WHO, which is 0.05 mg/kg.[30] However, the levels in all samples were <0.1 mg/kg, the standard limit defined by the US EPA for drinking water.[31] Moreover, the Cr levels of the samples were all within the limit when they were compared with the MAC (2.3 kg/mg) of fruits and vegetables.[41]

The study particularly focused on the Hg concentration in soybean and palm oil available in Bangladesh. The findings demonstrated that most of the analyzed samples exceeded the BSTI limit for Hg (0.25 mg/kg) in oil.[18] Soybean oil exceeded the BSTI limit for Hg by 17–20 times, whereas palm oil exceeded it by 6–14 times. An earlier Nigerian study found Hg in the range of 0.01–0.055 μg/g in 24 of 25 samples of palm oil.[42] An Indian study also reported the presence of Hg in branded sunflower oil at 71.4–88.0 mg/kg, which was assumed to be due to product adulteration with palm oil.[43] Another Spanish study reported the absence of Hg in edible sunflower and olive oils.[44] Previous studies have detected heavy metals in various food items in Bangladesh; specifically, cereals, milk, fruits, and vegetables were found to have high levels of As and Pb.[45] Fish and shellfish were also found to contain other metals.[46] It is noteworthy that these foods were locally grown. In contrast, soybean and palm oils consumed in Bangladesh are imported from different countries.[3] This contrast might explain why we did not detect As and Pb in oil samples. Instead, we found Hg contamination, likely originating from the source countries or occurring during transportation from the importing countries to Bangladeshi ports or from port to refineries. In the future, samples from these stages of the supply chain should be analyzed to identify the source of the contamination. To the best of our knowledge, heavy metal analysis in edible oils using nationally representative samples has not been conducted previously in Bangladesh.

The unusually high level of Hg found in this study even in the crude and refined oil samples collected from the refineries indicate that the oils might have been contaminated either at the production level, during the industrial extraction process or during transportation, and the contaminant persisted throughout the production, transportation, processing, and marketing chain. It is further evident that the industrial refining technique has little impact on the Hg levels in refined soybean and palm oils as compared with crude ones. Because crude oil was found to be the source of contamination, it was obvious that there would be no difference in Hg levels between branded and unbranded oil samples, given their common origin from the same crude oil. However, to show any potential contamination introduced from packaging materials and handling during distribution, transport, and marketing, we present the results separately for branded and unbranded oil. This is particularly relevant for unbranded oils, given that they are often transported and stored in containers that may not be made of food-grade materials or may have been previously used for transporting or storing various chemicals.[5]

Several studies from various countries have reported the presence of Hg in vegetable oil, although the levels observed were not considered hazardous to health.[42,47] Such contamination can occur during processing, harvesting, or packaging.[40] Both geogenic and anthropogenic activities contribute to global Hg emissions, which can deposit Hg in long-lasting soil pools and deep ocean regions.[48] Bangladesh, being an importer of palm oil mainly from Malaysia and Indonesia and of soybean oil from Latin America, including Argentina and Brazil,[3] may not be the primary source of the

Table 3. Estimation of median daily exposure (D_E of heavy metals, mg/kg BW per day).

	Median D_E (mg/kg)				
Route of exposure	As	Cd	Cr	Pb	Hg
Soybean oil					
Branded ($n = 19$)	3.10×10^{-6}	2.56×10^{-6}	1.89×10^{-5}	9.03×10^{-6}	2.00×10^{-3}
Unbranded ($n = 4$)	3.92×10^{-6}	3.07×10^{-6}	1.43×10^{-5}	1.24×10^{-5}	2.28×10^{-3}
Palm oil					
Branded ($n = 13$)	3.12×10^{-6}	1.90×10^{-6}	1.69×10^{-5}	8.64×10^{-6}	1.49×10^{-3}
Unbranded ($n = 8$)	3.93×10^{-6}	2.63×10^{-6}	1.48×10^{-5}	1.06×10^{-5}	7.69×10^{-4}

Note: As, arsenic; BW, body weight; Cd, cadmium; Cr, chromium; Hg, mercury; Pb, lead.

Table 4. Estimation of hazard quotient (HQ) of heavy metals.

	HQ				
Route of exposure	As	Cd	Cr	Pb	Hg
Soybean oil					
Branded ($n = 19$)	1.03×10^{-2}	2.52×10^{-3}	1.45×10^{-5}	1.29×10^{-3}	3.50
Unbranded ($n = 4$)	1.31×10^{-2}	3.07×10^{-3}	1.10×10^{-5}	1.77×10^{-3}	3.99
Palm oil					
Branded ($n = 13$)	1.04×10^{-2}	1.90×10^{-3}	1.30×10^{-5}	1.23×10^{-3}	2.61
Unbranded ($n = 8$)	1.31×10^{-2}	2.63×10^{-3}	1.14×10^{-5}	1.51×10^{-3}	1.35

Note: As, arsenic; Cd, cadmium; Cr, chromium; Hg, mercury; Pb, lead.

identified Hg contamination in edible oil. Rather, the soil of the producer country, potentially affected by Hg from sources such as mining sites and coal-based power plants, could be a significant origin.[40,49] Oil plants, for instance, palm plants, can uptake trace elements and heavy metals, such as Hg, from soil.[40,50] The repeated use of agrochemicals on the soil may further promote the accumulation of Hg, which can subsequently transfer its way into the oil.[40] In addition, earlier research suggested that heavy metals may be introduced during the refining process through the chemicals used at various stages and also from the equipment of refining plants.[40] Crude oil might also be contaminated with Hg during shipment. As the entire value chain was not examined in our study, the exact source of Hg in oil could not be identified, highlighting the need for further research to address this gap in understanding.

HQs represent the relationship between the detected pollutant dose and a predefined reference dose level. An HQ >1 suggests that the exposed population is at a higher risk of adverse health effects.[14] The HQ values for As, Cd, Cr, and Pb in the present study were within acceptable limits, although higher than the values reported in a previous Iranian study.[51] However, the D_E of Hg was estimated at 0.12–0.14 mg/d and 0.04–0.08 mg/d from the consumption of soybean and palm oil, respectively. The corresponding HQ value ranged from 1.35 to 3.99, indicating a higher risk of adverse health effects for the exposed population.[14] A previous study reported that chronic exposure to Hg can damage the brain, heart, kidneys, lungs, and immune systems of people of all ages.[52] Chronic Hg poisoning is also found to be associated with renal damage, proteinuria, tubular necrosis,[53] and adverse effects on pancreatic beta cells,[54] potentially contributing to conditions such as type 2 diabetes mellitus.[55,56] Another study reported that long-term exposure to neurotoxic organic Hg compounds leads to Minamata disease.[57] This justifies keeping Hg within the acceptable limit to protect population health.

The present research has notable strengths, including the analysis of nationally representative market samples, which allows generalization of the results to the entire Bangladeshi population. We report, to our knowledge for the first time, the presence of high levels of Hg in soybean and palm oils available for consumption in Bangladesh. However, samples could not be collected from the bulk oil carrier at the port of entry into Bangladesh, preventing the identification of the origin of Hg contamination. In addition, the study did not estimate the concentrations of methyl Hg and dimethyl Hg, which have more significant health impacts. In this study, the form of Hg could not be identified (i.e., organic or inorganic); therefore, the carcinogenic risk posed by Hg could not be identified owing to a lack of data.

Conclusions

This study is the first attempt to investigate the toxic heavy metals in edible oils consumed by the Bangladeshi population. The health risk assessment analysis found no evidence of significant health risks associated with the levels of As, Cd, Cr, and Pb. However, an HQ ranging from 1.35 to 3.99 suggests the Hg levels pose higher health risks. The presence of Hg, much of which is above the regulatory limits both in crude and refined oils, strongly suggests that the edible oils being consumed in Bangladesh were contaminated at the source, either at the primary production level, during the subsequent transport, or through the industrial processing chain.

The heavy metals reported in this study are considered control contaminants, and their presence in foodstuffs is unavoidable.[58] We argue that internationally recommended good agricultural practices and good manufacturing practices should be applied at all levels of primary production and processing to reduce the level of contamination through exogenous and endogenous sources to the allowable standard limits of these control parameters for edible oils.

Because Hg exposure through the consumption of food and dietary supplements is a global public health concern, further research is necessary to quantify and speciate the presence of more harmful methyl and dimethyl Hg and other ecotoxic heavy metals in edible oils.

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