



Evaluation of trace metallic element levels in coffee by icp-ms: a comparative study among different origins, forms, and packaging types and consumer risk assessment

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Abstract

Trace elements (TE) contamination of foods and beverages constitutes a public health issue. In this context, the main objective of this study was to determine metals and metalloids content in coffee and to assess the health risks associated with contaminated coffee consumption. To this end, 44 samples of coffee from different origins, forms, and packaging types were analyzed. TE analysis was performed by ICP-MS after digestion. The data analysis was based on principal components analysis (PCA) and analysis of variance (ANOVA). Health risk assessment was determined by the estimated daily intake (EDI), target hazard quotient (THQ), and hazard index (HI). The findings showed that TE levels in coffee varied widely. The highest levels were related to aluminum (Al) (59.88 ± 54.86 mg/kg), manganese (Mn) (16.26 ± 24.59 mg/kg), copper (Cu) (11.60 ± 11.55 mg/kg), and cadmium (Cd) (9.92 ± 10.32 mg/kg). In terms of coffee form and packaging type, a significant difference ($P < 0.0001$) was observed in nickel (Ni), chromium (Cr), zinc (Zn), cobalt (Co), Cu, Mn, and Al content. The highest EDI was found in Al (0.0109 mg/kg BW/day) in ground coffee packaged in capsules. In terms of chronic daily intake (CDI), Cd and Al were above the reference dose (RfD). THQ of these elements were greater than 1.0, and HI was above the value of 1.0 in different forms of coffee. More interdisciplinary research on the relationships between the metal concentrations in coffee samples and those in feed, water, and soil would be quite interesting.

Keywords Heavy metals · Coffee · ICP-MS · Health risk assessment · Food safety

Introduction

The coffee plant is usually a woody perennial tree that grows at higher altitudes; it belongs to the family Rubiaceae and genera *Coffea*, comprising 70 different species [1]. Coffee is the leading worldwide beverage after water; its trade exceeds US \$10 billion, and approximately 500 billion cups of coffee

are consumed annually all over the world [2, 3]. Therefore, coffee consumption exhibits a dichotomy. On the one hand, coffee beverages may be consumed not only for their stimulating effect but also for their good taste, pleasant aroma, and beneficial effects [1]. Indeed, coffee prevents diabetes mellitus, various cancer lines, Parkinsonism, and Alzheimer's disease. Moreover, it ameliorates oxidative stress and helps in proper cognitive functionality [4–8]. On the other hand, excessive coffee consumption could cause myocardial and cerebral infarction [9], insomnia [10], and cardiovascular complications [11].

Coffee, like all daily-consumed products, must be subjected to quality control to ensure its safety for consumers' health, primarily through physico-chemical analysis as well as the determination of agricultural, microbiological, plastic, and heavy metal contaminants [12, 13]. From previous studies, exposure to heavy metals such as cadmium (Cd), lead (Pb), mercury (Hg), nickel (Ni), chromium (Cr), arsenic (As), copper (Cu), and zinc (Zn) may induce cognitive impairments and neurological disorders [14]; nephrotoxicity,

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fetal toxicity, and teratogenicity in humans [15]; disruptions of the blood and circulatory systems; alterations in detoxification pathways (colon, kidney, liver, and skin); and gastrointestinal, reproductive, and mental system disorders [16, 17]. Their presence was related to many consumed products, such as food (vegetables, grains, fruit, fish, shellfish, and meat) and beverages (milk, cheese, water, tea, and coffee) [18]. Moreover, foods of both plant and animal origin can be contaminated during food processing and packaging [19]. Heavy metals can migrate through foods high in fatty acids, such as fast food, sausages, and pizza [20].

The hazard level of exposure to heavy metals could be quantitatively evaluated by human health risk evaluation models. Health risk assessment has been widely applied in the health evaluation of heavy metal contamination; it quantitatively predicts the health effects of contaminants via numerical models [21–23]. The chronic daily intake and non-cancerous risks due to the intake of heavy metals are assessed by the target risk quotient (THQ) and hazard index (HI) [24].

In Algeria, as in several other developing countries, the increase in heavy concentrations from anthropogenic sources (e.g., fuel combustion, the proximity of roads, mining and industrial areas, municipal and agricultural wastewater, and solid waste) has raised concern about their effect on ecosystems [24–27]. Moreover, due to their bioaccumulation and bio-magnification in the food chain, their effects may be exacerbated [28].

In light of the FAO's recommendations and the fact that coffee is widely consumed in Algeria, where consumption reached 0.44 kg per capita for instant coffee and 1.60 kg per capita for roast coffee [29], the determination of heavy metal levels in coffee, which is considered a widely consumed food, is of great interest to protect consumers' health. To our knowledge, no health risk assessment study has ever been undertaken in Algeria to estimate the non-carcinogenic risks following the consumption of coffee. Hence, this study aimed to (i) determine nickel (Ni), chromium (Cr), lead (Pb), zinc (Zn), copper (Cu), cobalt (Co), cadmium (Cd), arsenic (As), manganese (Mn), and aluminum (Al) levels in coffee products purchased from Chlef regions in Algeria and compare the results of this study with data reported in published studies from different countries across the world, and (ii) evaluate the health risks associated with consuming coffee products.

Materials and Method

Study Design and Sampling

The present cross-sectional analytical study was conducted in the region of Chlef, an area located between

35°50'00" and 36°33'00" North and 00°42'00" and 01°45'00" East, in the northwest of Algeria in North Africa, and bordered on the north by the Mediterranean Sea. A total of 44 coffee samples were randomly collected from several local groceries and divided into three groups: (i) group 1: consisted of 20 instant coffee samples imported from different origins, such as France (8 samples), Italy (4 samples), Sweden (3 samples), Switzerland (2 samples), UK (1 sample), Poland (1 sample), and USA (1 sample); (ii) group 2: included 12 instant coffee samples from local packaged brands; and (iii) group 3: included 12 roasted coffee samples (unpackaged coffee marketed in bulk) (Table 1).

All samples were obtained using a randomized design from random Chlef marketplaces (Algeria). Samples were collected from 3th to 13th September 2022.

The collected packets of coffee were initially visually checked to ensure their integrity, impermeability, and that they were still within their shelf life. The coffee was kept at room temperature in its original, firmly sealed packages prior to analysis. The roasted coffee beans were ground by a laboratory mill (Lab Mill 3310, Perkin Elmer, USA) and then stored at room temperature in glass bottles. For each sample, a sampling sheet indicating the coffee form, packaging, manufacturing company, and origin was established.

Trace Elements (TE) Analysis

Reagents and Solutions

All chemicals used in this study were of analytical reagent grade. Nitric acid 65% (HNO₃) (CAS: 7697-37-2) was purchased from (Sigma Aldrich, St. Quentin Fallavier, France). Potassium hydroxide (KOH) was purchased from (Honeywell, Charlotte, North Carolina, USA).

Digestion Procedure

A quantity of 3 g of each coffee sample was digested using a volume of 5 mL of nitric acid (HNO₃) at 65% and 1 mL of potassium hydroxide (KOH) at 5% in ceramic capsules.

These capsules were placed in a microwave digestion system (MPS 320 microwave: MR-50/16, Perkin Elmer, USA) at 200 °C in 3 steps (power (W); ramp (min); hold (min); fan): (i) step 1: 600 W, 10 min, 15 min; (ii) step 2: 850 W, 10 min, 10 min; (iii) step 3: 0 W, 10 min, 10 min. Thereafter, the digested coffee samples were filtered through a 0.45-μm membrane filter (Merck, Île-de-France, France), then transferred into a 25-mL volumetric flask, and diluted to 25 mL with distilled deionized water.

Table 1 Detailed description of the samples examined in this paper. Analyses were carried out in triplicate on all samples

	Caps (<i>N</i> = 8)	Cartons (<i>N</i> = 20)	Glass bottles (<i>N</i> = 4)	In bulk (<i>N</i> = 12)
Sweden (<i>N</i> = 3)	Ground (<i>N</i> = 2)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)
Switzerland (<i>N</i> = 2)	Ground (<i>N</i> = 1)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)
Italy (<i>N</i> = 4)	Ground (<i>N</i> = 1)	Ground (<i>N</i> = 2)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)
Algeria (<i>N</i> = 24)	Ground (<i>N</i> = 2)	Ground (<i>N</i> = 9)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 12)
France (<i>N</i> = 8)	Ground (<i>N</i> = 2)	Ground (<i>N</i> = 4)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 2)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)
USA (<i>N</i> = 1)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)
Poland (<i>N</i> = 1)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)
UK (<i>N</i> = 1)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)	Ground (<i>N</i> = 0)
	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 1)	Instant (<i>N</i> = 0)	Instant (<i>N</i> = 0)
	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)	Beans (<i>N</i> = 0)

Trace Elements (TE) Quantification

Levels of 10 trace elements (TE): nickel (Ni), chromium (Cr), lead (Pb), zinc (Zn), copper (Cu), cobalt (Co), cadmium (Cd), arsenic (As), manganese (Mn), and aluminum (Al) were determined in triplicate using an inductively coupled plasma mass spectrometry (ICP-MS) (ELAN® DRC II, Perkin Elmer, USA).

The following parameter settings were used during the measurements: ICP radio frequency power: RF power: 1.35 kW; RF matching 1.7 V; carrier gas: 0.90 L/min; make-up gas: 0.27 L/min; sampling depth: 8 mm; S/C temp: 2 °C; nebulizer pump: 0.1 rps; torch horizontal: 1.2 mm; torch-vertical: 1.4 mm; extract 1: 4.3 V; extract 2: − 99 V; Omega Bias -ce: − 16 V; Omega Lens-ce: 2.6 V; cell entrance: − 26 V; cell exit: − 20 V; channel spacing: 0.02; QP focus: 5 V. VG Instrument type is a PlasmaQuad PQ2+, detector type has a Channeltron multiplier, and nebulizer type has a V-groove.

Quality Control Procedure

The certified reference materials INC-TEL-1 Black Tea Leaves (Institute of Nuclear Chemistry and Technology, INCT, Poland) were used for quality control procedures.

Relative standard deviation (% RSD) values had to be less than 5% for all mean concentrations; otherwise, the results were disregarded and the digestion was repeated. Recoveries from analyses were in the range of 97.0–102.0%.

In order to determine the limit of detection (LOD) and limit of quantification (LOQ), ten blank samples were measured. For each element, LOD and LOQ were quantified as three times and ten times the standard deviation (SD) of the blank values, respectively [30, 31]. The limits of detection (LODs) were as follows: Ni (0.008), Cr (0.02), Pb (0.02), Zn (0.03), Cu (0.02), Co (0.001), Cd (0.02), As (0.01), Mn (0.002), and Al (0.06) mg/kg. The limits of quantification (LOQs) were as follows: Ni (0.025), Cr (0.06), Pb (0.06), Zn (0.1), Cu (0.06), Co (0.005), Cd (0.06), As (0.03), Mn (0.006), and Al (0.18) mg/kg.

Health Risk Assessment

Estimated Daily Intake (EDI)

In order to evaluate the health risks related to coffee consumption, the following indices were used: The estimated daily intake (EDI) was calculated as follow:

$$EDI = MS_i \times C / BW_i$$

where MS_i is the mass of a selected consumed coffee (5 g) by adults (kg); C is the trace element concentration (mg/kg); and BW_i is the average body weight (kg), which is assumed to be 69.6 kg [32].

Chronic Daily Intake (CDI)

The chronic daily intake (CDI) (mg/day/kg BW) was calculated according to the following formula:

$$CDI = EDI \times EF_r \times ED_{tot} / BW_i \times AT$$

Where EF_r is the frequency of exposure (365 days/year); ED_{tot} is the exposure duration (years); AT is the average period of exposure (365 days/year); and BW_i is the average body weight (kg).

Target Hazard Quotient Determination

The target hazard quotient (THQ) for non-carcinogenic risk was calculated using the equation provided by USEPA [33]:

$$THQ = CDI / RfD$$

where RfD (reference dose) is the daily reference dose that enables the individual to sustain a level of exposure over a long period without experiencing any harmful effects. RfD for Ni, Cr, Pb, Zn, Cu, Cd, As, Mn, and Al are, respectively, 0.002, 0.0003, 0.0035, 0.03, 0.04, 0.0005, 0.0003, 0.14, and 0.0004 mg/kg BW/day [33].

An $THQ > 1.0$ represents adverse non-carcinogenic effects, while an $THQ < 1.0$ represents an acceptable level.

Hazard Index Determination

To determine the total danger of various metals, present in coffee, the hazard index (HI) was used. According to the following equation, it was estimated by adding the THQ of each metal in this study [34].

Hazard index (HI), since more than one toxicant is present, the interactions are considered:

$$HI = \sum HQ_i$$

If $HI < 1.0$, the non-carcinogenic adverse effect of this exposure pathway or chemical is assumed to be insignificant [35].

Data Analysis

Recorded data were subjected to descriptive statistical analysis using a trial version of XLSTAT 2022, and the results are expressed in the form of mean \pm SD (standard deviation). Correlation between TE was demonstrated

using Pearson correlation matrix. In order to identify the different relationships between TE content, coffee form (beans, ground, and instant coffee), origin, and packaging (in caps, in cartons, and in glass bottles), centered, and reduced principal component analyses (PCAs) were applied. Active individuals were introduced according to the sample grouping criteria (form of coffee, packaging, and product origin) for each ACP. Ten variables (Ni, Cr, Pb, Zn, Cu, Co, Cd, As, Mn, and Al) were retained as active variables. Indeed, the principal components are sufficiently explanatory provided that their explained variance exceeds 60%.

When the conditions of normality and homogeneity of the variances were observed, data were analyzed using one-way analysis of variance (ANOVA) followed by Tukey's post hoc test to assess heavy metal variation according to coffee forms, origin, and packaging.

Pearson correlation test was carried out to measure the statistical relationship between heavy metal levels. The statistical significance level was set at $P 0.05$. Data visualization was performed using R 4.2.2 software [36].

Results

Heavy Metal Levels in the Collected Samples

The findings showed that TE levels in coffee varied widely. The most dominant heavy metals were Al (59.88 ± 54.86 mg/kg), Mn (16.26 ± 24.59 mg/kg), Cu (11.60 ± 11.55 mg/kg), and Cd (9.92 ± 10.32 mg/kg) (Fig. 1 and Table 2).

The Pearson correlation matrix showed a correlation between Al, Ni, Zn, Cu, and Co, and also a correlation between As, Pb, Cr, and Cd, which is very consistent with the PCA results (Table 3).

The principal components analysis (PCA) showed that the first two axes explained 78.69% of the total variance. The first axis, with 61.42% of explained variance, was significantly positively correlated with Ni ($R = 0.92$), Cu ($R = 0.91$), Al ($R = 0.87$), Zn ($R = 0.85$), Co ($R = 0.83$), and Cr ($R = 0.74$), while the second axis (17.28% of explained variance) was positively correlated with As ($R = 0.65$) and Pb ($R = 0.52$) and negatively correlated with Mn ($R = -0.44$), Al ($R = -0.39$), and Zn ($R = -0.36$) (Fig. 2).

In terms of coffee samples, the PCA indicated a high correlation between coffee form and TE levels. In this context, the highest concentrations of TE were recorded in ground coffee samples, while the lowest levels were observed in coffee beans (Fig. 2). The PCA also revealed a high correlation between TE levels and coffee packaging. In this respect, the samples of coffee packaged in capsules contain high concentrations of Cu, Co, Zn, Mn, and Al, while

Fig. 1 TE content found in all coffee samples (mg/kg)

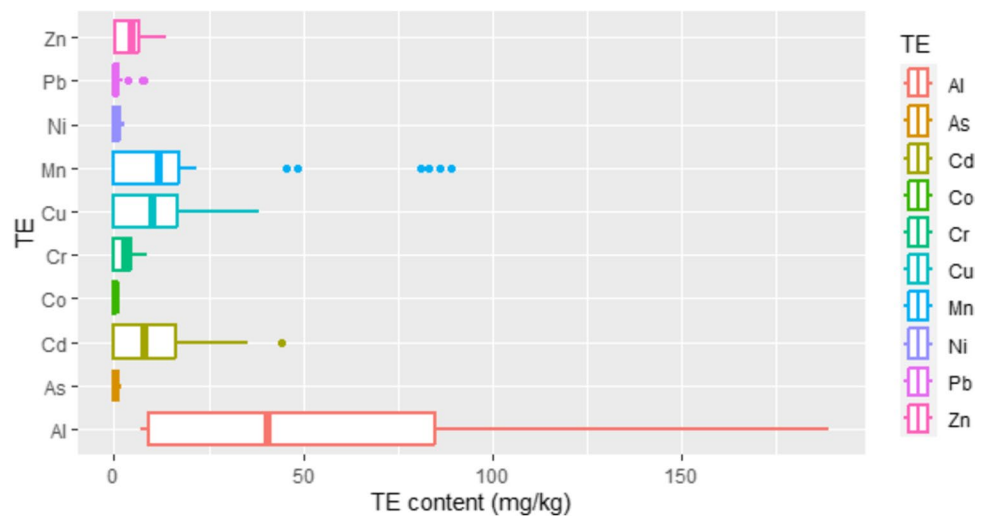


Table 2 Descriptive statistics of the amount of TE found in coffee samples (mg/kg)

Descriptive statistics	Ni	Cr	Pb	Zn	Cu	Co	Cd	As	Mn	Al
MPL (mg/kg) Omeje et al. [37]	0.10	0.10	1.0	-	30	0.043	0.1	1.4	2.0	-
Pigozzi et al. [38]										
1st quartile (mg/kg)	0.01	0.04	0.08	0.40	0.06	0.005	0.058	0.12	0.0075	9.00
3rd quartile (mg/kg)	1.42	4.32	1.33	6.54	16.32	0.67	16.00	0.25	16.65	84.50
Mean (mg/kg)	0.82	2.71	1.14	4.51	11.60	0.35	9.92	0.25	16.27	59.88
SD (mg/kg)	0.81	2.52	1.99	4.03	11.56	0.30	10.32	0.19	24.60	54.87

MPL, maximum permissible limits established by international regulators (FAO, WHO, Food and Nutrition Board); SD, standard deviation; N, sample size (N = 44)

Table 3 Pearson correlation matrix between the detected TE

Variables	Ni	Cr	Pb	Zn	Cu	Co	Cd	As	Mn	Al
Ni	1.00	0.68	0.54	0.75	0.79	0.75	0.62	0.54	0.64	0.83
Cr	0.68	1.00	0.58	0.49	0.51	0.45	0.68	0.70	0.30	0.48
Pb	0.54	0.58	1.00	0.30	0.41	0.44	0.61	0.65	0.38	0.32
Zn	0.75	0.49	0.30	1.00	0.86	0.74	0.39	0.33	0.69	0.89
Cu	0.79	0.51	0.41	0.86	1.00	0.79	0.55	0.34	0.77	0.88
Co	0.75	0.45	0.44	0.74	0.79	1.00	0.51	0.34	0.57	0.73
Cd	0.62	0.68	0.61	0.39	0.55	0.51	1.00	0.56	0.25	0.44
As	0.54	0.70	0.65	0.33	0.34	0.34	0.56	1.00	0.16	0.24
Mn	0.64	0.30	0.38	0.69	0.77	0.57	0.25	0.16	1.00	0.75
Al	0.83	0.48	0.32	0.89	0.88	0.73	0.44	0.24	0.75	1.00

the samples packaged in cartons contain mostly As, Pb, Cr, and Cd, whereas the coffee samples stored in glass bottles and the coffee sold in bulk contain very low concentrations (Fig. 2). It was also noted that the ground coffee packaged in cartons and produced in Algeria contains high levels of As, Pb, Cr, and Cd (Fig. 2).

In order to compare the TE content depending on the form of coffee, packaging, and product origin, an ANOVA was performed, and the results showed highly significant differences ($P < 0.01$) between the different forms of coffee in

terms of Ni, Cr, Zn, Cu, Co, Cd, Al, As, and Mn content, and a significant difference in terms of Pb ($P < 0.05$) (Table 4).

Regardless of the TE, the highest concentrations were always detected in the ground coffee, while the lowest concentrations were recorded in the coffee beans. Also, significant differences ($P < 0.01$) in metal content were observed between the various types of packages, especially in terms of Ni, Cr, Zn, Cu, Co, Mn, Cd, and Al content, and a significant difference in terms of As ($P <$

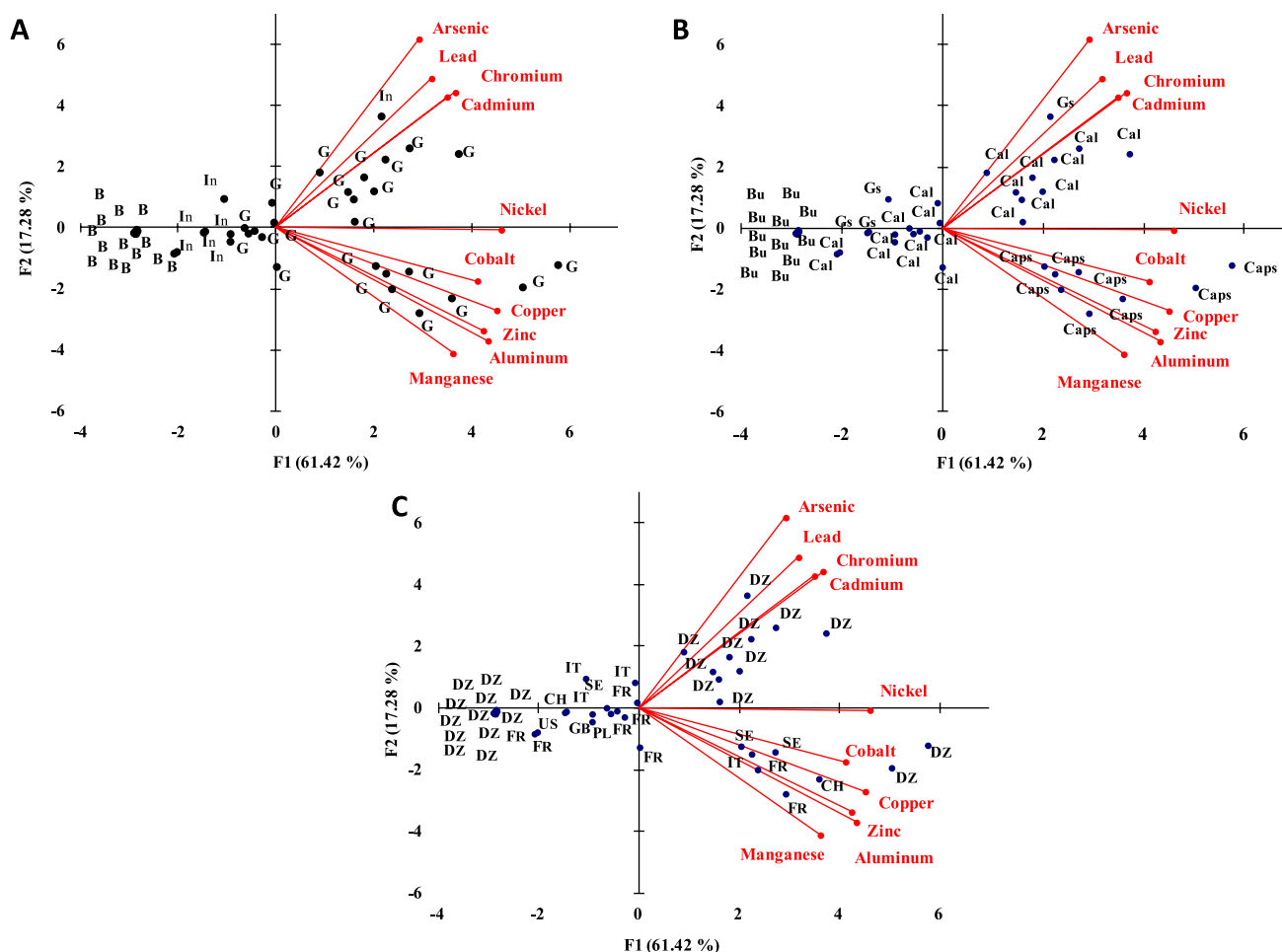


Fig. 2 Principal component analysis (PCA) plot showing the correlation between heavy metals and the variation in TE levels in 44 samples of coffee: **A** in various forms (G, ground coffee; In, instant coffee; B, coffee Beans). **B** According to their packaging types (Gs,

coffee packaged in glass bottles; Caps, coffee in capsules; Cal, coffee conserved in cartons; Bu, coffee in bulk). **C** According to their origin (DZ, Algeria; FR, France; US, United States of America; GB, United Kingdom; SE, Sweden; CH, Switzerland; PL, Poland; IT, Italy)

Table 4 Heavy metal levels (mg/kg) in coffee samples according to the coffee form

Form	Ni	Cr	Pb	Zn	Cu	Co	Cd	As	Mn	Al
Ground	1.43 ^a	4.39 ^a	1.73 ^a	7.19 ^a	19.24 ^a	0.53 ^a	15.52 ^a	0.33 ^a	28.45 ^a	99.78 ^a
Instant	0.39 ^b	2.54 ^b	1.33 ^{ab}	2.77 ^b	9.62 ^b	0.42 ^a	11.28 ^a	0.25 ^{ab}	6.49 ^{ab}	21.71 ^b
Beans	0.009 ^b	0.034 ^c	0.057 ^b	0.97 ^b	0.049 ^c	0.026 ^b	0.035 ^b	0.11 ^b	1.12 ^b	13.42 ^b
<i>P</i> -value	< 0.0001	< 0.0001	0.0406	< 0.0001	< 0.0001	< 0.0001	< 0.0001	0.002	0.001	< 0.0001

Means which are denoted by lowercase different letters (a, b, and c) indicate significant difference between coffee form (ground, instant, and beans) (Tukey's post hoc test, $P < 0.05$). *N*, sample size ($N = 44$)

0.05), while there was no difference between the different packages in lead (Pb) content (Table 5).

The highest TE concentration was observed in coffee packaged in capsules and cartons, especially for Al, Mn,

Cu, Cd, and Zn. Therefore, with the exception of Pb, TE levels could also be linked to coffee packaging. Finally, no significant differences in metal content were observed between the various origins of the product (Table 6).

Table 5 Heavy metal levels (mg/kg) in coffee samples according to the coffee packaging type

Packaging	Ni	Cr	Pb	Zn	Cu	Co	Cd	As	Mn	Al
Caps	1.89 ^a	3.22 ^a	1.53 ^a	11.28 ^a	30.82 ^a	0.78 ^a	13.38 ^a	0.23 ^{a,b}	50.58 ^a	152.75 ^a
Cartons	0.93 ^b	4.16 ^a	1.41 ^a	4.87 ^b	11.23 ^b	0.35 ^b	13.45 ^a	0.33 ^a	15.49 ^b	63.90 ^b
Glass bottles	0.51 ^{b,c}	2.53 ^{a,b}	2.23 ^a	1.44 ^c	9.77 ^b	0.55 ^{a,b}	15.00 ^a	0.28 ^{a,b}	0.29 ^b	8.25 ^c
In bulk	0.009 ^c	0.029 ^b	0.06 ^a	0.41 ^c	0.045 ^c	0.003 ^c	0.038 ^b	0.11 ^b	0.004 ^b	8.50 ^c
<i>P</i> -value	< 0.0001	< 0.0001	0.142	< 0.0001	< 0.0001	< 0.0001	0.0005	0.012	< 0.0001	< 0.0001

Means which are denoted by lowercase different letters (a, b, and c) indicate significant difference between coffee form (ground, instant, and beans) (Tukey's post hoc test, $P < 0.05$). *N*, sample size ($N = 44$)

Table 6 Heavy metal levels (mg/kg) coffee samples according to the coffee origin

Origin	Ni	Cr	Pb	Zn	Cu	Co	Cd	As	Mn	Al
Sweden	1.47	3.04	0.35	9.09	16.31	0.58	12.00	0.22	15.40	109.33
Switzerland	0.82	2.48	0.18	6.84	21.42	0.68	10.00	0.22	40.67	73.50
Italy	0.92	4.53	0.58	4.19	14.87	0.36	9.50	0.21	10.74	59.50
Algeria	0.801	2.82	1.84	3.41	9.63	0.26	11.56	0.29	13.55	50.50
France	0.71	1.76	0.205	6.27	13.02	0.43	6.38	0.18	23.44	78.00
<i>P</i> -value	0.7551	0.5547	0.2359	0.1054	0.5993	0.1708	0.8357	0.7243	0.5865	0.4357

No significant difference between coffee origin (Sweden, Switzerland, Italy, Algeria, and France) (Tukey's post hoc test, $P < 0.05$). *N*, sample size ($N = 44$)

Health Risk Assessments

Regarding the health risks associated with coffee consumption, the highest EDI was shown by Al and particularly in ground coffee caps (0.0109 mg/kg BW/day). The EDI and CDI related to Ni, Pb, Zn, Cu, Co, As, and Mn were below the oral reference dose (RfD) [33] regardless of the form of the product (caps, cartons, and in bulk), while in terms of aluminum and cadmium both EDI and CDI were way above the RfD, and consequently, the hazard quotient (THQ) of both TE were greater than 1 and especially in ground coffee packaged in cartons and ground coffee caps.

Finally, the hazard index (HI) was well above 1 in the three forms of coffee (Table 7).

Discussion

Heavy Metal Levels

Coffee is a very popular and appreciated beverage all around the world, mainly for its flavor and stimulating properties [1]. Thus, it must be subject to quality control and health risk assessment to ensure the safety of consumers and avoid exposure to harmful substances such as acrylamides, drug residues, pesticide residues, and TE. Indeed, some TE, such as As, Cd, and Pb, can disrupt human metabolic functions by accumulating in vital organs such as the liver, heart, kidney,

and brain, interfering with normal biological functioning [39].

Quite recently, particular attention has been paid to TE in coffee collected from different countries across the globe, such as Lebanon [40], Brazil [18, 40], Ethiopia, India, Kenya, Columbia [18], Nicaragua [41], Poland [40], and Bosnia and Herzegovina [40, 42]. Indeed, through local samples of coffee, these studies revealed the presence of various metals and metalloids in coffee (Table 8). Moreover, the results reported here showed that all heavy metals tested were higher than the maximum permissible limits established by international regulators for Ni, Cr, Pb, Co, Cd, and Mn, whereas Cu and As present lower concentrations [37] [38].

According to the results of the present study, TE presence was mainly recorded in ground coffee packaged in cartons and capsules, unlike coffee beans sold in bulk, which contain the lowest TE levels, suggesting that this contamination may principally occur during molding and packaging processes.

The highest levels were shown mainly by Al (59.88 ± 54.86 mg/kg), Mn (16.27 mg/kg), Cu (11.60 mg/kg), and Ca (9.92 mg/kg), these high contents were also reported in more pronounced levels in Brazil (Al = 181.8 ± 190.6 mg/kg, Mn = 71.7 ± 12.4 mg/kg, Cu = 29.67 ± 9.54 mg/kg, Ca = 1354.5 ± 904.7 mg/kg) [18], Ethiopia (Al = 158.2 ± 164.3 mg/kg, Mn = 45.1 ± 10.3 mg/kg, Cu = 24.85 ± 2.62 mg/kg, Ca = 1047.4 ± 257.2 mg/kg) [18], and Colombia (Al = 70.9 ± 48.3 mg/kg, Mn = 78.2 ± 23.1 mg/kg, Cu = 30.81 ± 8.39 mg/kg, Ca = 1073.4 ± 408.8 mg/kg) [18]. The high

Table 7 Health risk assessment from TE exposure through coffee consumption

Groups	TE	TE content (mg/kg)	EDI (mg/kg BW/day)	RfD (mg/kg BW/day) [33]	CDI (mg/kg BW/day)	THQ	HI
Coffee beans in bulk (<i>N</i> = 12)	Ni	0.0090	6.46E-08	0.002	6.49E-08	0.0003	1.59
	Cr	0.028	2.01E-07	0.0003	2.02E-07	0.006	
	Pb	0.062	4.45E-06	0.0035	4.47E-06	0.012	
	Zn	0.409	2.93E-06	0.03	2.94E-06	0.0009	
	Cu	0.045	3.23E-07	0.04	3.24E-07	8.12E-06	
	Co	0.002	1.43E-08	-	-	-	
	Cd	0.037	2.65E-07	0.0005	2.66E-07	0.053	
	As	0.114	8.18E-07	0.0003	8.22E-07	0.002	
	Mn	0.0035	2.51E-08	0.14	2.52E-08	1.8E-6	
	Al	8.5	6.1E-05	0.0004	6.1E-05	1.525	
Ground coffee packaged in cartons (<i>N</i> = 15)	Ni	1.011	7.26E-06	0.002	7.26E-06	0.036	25.79
	Cr	4.457	3.2E-05	0.0003	3.2E-05	10.66	
	Pb	1.647	1.1E-05	0.0035	1.1E-05	0.0031	
	Zn	5.143	3.6E-05	0.03	3.6E-05	0.012	
	Cu	12.74	9.1E-05	0.04	9.15E-05	0.0022	
	Co	0.384	2.75E-06	-	-	-	
	Cd	15.41	0.0011	0.0005	0.0011	2.2	
	As	0.364	2.61E-06	0.0003	2.62E-06	0.873	
	Mn	16.22	0.0011	0.14	0.0011	0.007	
	Al	67.41	0.0048	0.0004	0.0048	12	
Ground coffee caps (<i>N</i> = 8)	Ni	1.89	1.3E-05	0.002	1.3E-05	0.065	30.28
	Cr	3.22	2.3E-05	0.0003	2.3E-05	0.76	
	Pb	1.52	0.0001	0.0035	0.0001	0.028	
	Zn	11.28	0.00081	0.03	0.00081	0.027	
	Cu	30.81	0.0022	0.04	0.0022	0.055	
	Co	0.78	5.60E-06	-	-	-	
	Cd	13.37	0.0009	0.0005	0.0009	1.8	
	As	0.22	1.58E-06	0.0003	1.58E-06	0.052	
	Mn	50.58	0.0036	0.14	0.036	0.257	
	Al	152.75	0.0109	0.0004	0.0109	27.25	

EDI, estimated daily intake; RfD, reference dose; CDI, chronic daily intake; THQ, target hazard quotient; HI, hazard index; Msi = 0.005 kg/day; BWI = 69.6 kg for Algerian population [32]; Efr = 356 per year; EDtot = 70 years; AT = 365 per year; *N*, sample size; -, not defined. Bold data indicate values with significant effects

content of Al may be related to the fact that this metal is largely used in coffee packaging combined with other metals and minerals (such as Cu, Zn, and Mn). According to Alamri [44], food-packaging interactions may compromise the quality and/or safety of foods [44]. Since Al is not essential in enzymes functionality or any metabolic process, high content of Al in human tissues has been associated with many disorders such as dialysis encephalopathy, osteodystrophy, microcytic anemia [44], inflammatory neural degeneration, behavioral impairment, and cognitive decline [45, 46].

Mn was the second major metal (16.27 mg/kg) in coffee available on the Algerian market; unlike Al, Mn is an essential nutrient for intracellular activities; it operates as a cofactor for a variety of enzymes, including arginase, glutamine

synthetase (GS), pyruvate carboxylase, and Mn superoxide dismutase (Mn-SOD) [47]. However, overexposure to Mn (through industrial exposure or contaminated water) may lead to its toxic accumulation in the brain, which has been associated with neurological impairment [48].

The third metal was Cu with a value of 11.60 ± 11.55 mg/kg, very similar to the concentration reported by Adler et al. [42] in Bosnia and Herzegovina (11.4 ± 4.13 mg/kg) and Varady et al. [41] in Nicaragua (14.1 ± 3.16 mg/kg). Cu is an essential TE in both humans and animals; it is a cofactor of many redox enzymes, but excessive amounts of Cu in the body can result in liver disease and severe neurological defects [49].

Cd was also among the major TE of coffee in Algeria (9.92 ± 10.32 mg/kg), though at a much lower level

Table 8 TE content in coffee samples reported in different parts of the world

Origin	Analytical method	N	TE content (mean \pm SD) (mg/kg)	Reference
Lebanon	ICP-MS	1	Mn (6.19 \pm 0.02); Co (0.066 \pm 0.007); Ni (0.8423 \pm 0.023); Cr (0.035 \pm 0.001); Ag (0.127 \pm 0.008); Pb (0.693 \pm 0.019)	[40]
Brazil		3	Mn (4.97 \pm 0.10); Co (0.07 \pm 0.02); Ni (0.88 \pm 0.08); Cr (0.05 \pm 0.02); Ag (0.08 \pm 0.04); Pb (0.71 \pm 0.13)	
		15	Mg (4705.8 \pm 565.9); Ca (1354.5 \pm 904.7); Fe (161.9 \pm 47.5); Al (181.8 \pm 190.6); Mn (71.7 \pm 12.4); Cu (29.67 \pm 9.54); Ba (7.50 \pm 4.24); Sr (5.84 \pm 1.08); Zn (6.99 \pm 8.80); Cr (2.12 \pm 0.64); Pb (1.93 \pm 1.80); Ni (0.75 \pm 0.29); V (1.03 \pm 1.06); Co (0.58 \pm 0.14); Ga (0.35 \pm 0.14); U (0.23 \pm 0.51); Cd (0.26 \pm 0.44); Ag (0.15 \pm 0.18); Li (0.05 \pm 0.05); In (0.12 \pm 0.29); Bi (0.04 \pm 0.04)	[18]
Ethiopia		6	Mg (4721.2 \pm 537.2); Ca (1047.4 \pm 257.2); Fe (132.2 \pm 109.1); Al (158.2 \pm 164.3); Mn (45.1 \pm 10.3); Cu (24.85 \pm 2.62); Ba (14.74 \pm 2.47); Sr (8.13 \pm 1.85); Zn (7.46 \pm 4.89); Cr (2.03 \pm 0.54); Pb (2.55 \pm 1.40); Ni (0.86 \pm 0.31); V (0.72 \pm 1.09); Co (0.27 \pm 0.08); Ga (0.67 \pm 0.10); U (0.54 \pm 0.66); Cd (0.29 \pm 0.23); Ag (0.22 \pm 0.17); Li (0.20 \pm 0.36); In (0.13 \pm 0.14); Bi (0.02 \pm 0.02)	
India		7	Mg (4423.0 \pm 402.9); Ca (1061.1 \pm 272.0); Fe (196.0 \pm 30.1); Al (87.0 \pm 97.1); Mn (67.0 \pm 8.9); Cu (30.32 \pm 6.23); Ba (11.68 \pm 3.34); Sr (8.34 \pm 2.24); Zn (8.57 \pm 9.48); Cr (4.06 \pm 1.94); Pb (1.44 \pm 1.79); Ni (2.67 \pm 1.10); V (1.62 \pm 1.39); Co (0.97 \pm 0.40); Ga (0.53 \pm 0.14); U (0.00 \pm 0.00); Cd (0.10 \pm 0.12); Ag (0.50 \pm 0.92); Li (0.10 \pm 0.12); In (0.03 \pm 0.05); Bi (0.06 \pm 0.04)	
Kenya		11	Mg (4640.8 \pm 437.7); Ca (1640.1 \pm 1390.7); Fe (173.5 \pm 72.0); Al (153.3 \pm 160.1); Mn (86.2 \pm 39.1); Cu (27.42 \pm 3.67); Ba (18.92 \pm 10.39); Sr (11.97 \pm 3.47); Zn (10.57 \pm 10.30); Cr (2.72 \pm 1.42); Pb (1.38 \pm 0.97); Ni (1.18 \pm 0.59); V (0.86 \pm 0.62); Co (0.46 \pm 0.22); Ga (0.92 \pm 0.43); U (0.13 \pm 0.43); Cd (0.14 \pm 0.09); Ag (0.14 \pm 0.18); Li (0.07 \pm 0.05); In (0.04 \pm 0.05); Bi (0.07 \pm 0.07)	
Columbia		17	Mg (4347.7 \pm 753.5); Ca (1073.4 \pm 408.8); Fe (163.7 \pm 66.7); Al (70.9 \pm 48.3); Mn (78.2 \pm 23.1); Cu (30.81 \pm 8.39); Ba (18.24 \pm 8.03); Sr (13.76 \pm 6.48); Zn (5.99 \pm 6.93); Cr (2.51 \pm 1.89); Pb (1.04 \pm 0.55); Ni (1.16 \pm 0.42); V (0.98 \pm 0.76); Co (0.89 \pm 0.31); Ga (0.95 \pm 0.58); U (0.20 \pm 0.82); Cd (0.15 \pm 0.18); Ag (0.16 \pm 0.16); Li (0.06 \pm 0.08); In (0.04 \pm 0.12); Bi (0.07 \pm 0.10)	
		25	Hg (0.0007 \pm 0.00003); Cu (10.9 \pm 2.33); Pb (0.13 \pm 0.02); Cr (0.06 \pm 0.01); Cd (0.15 \pm 0.02); Ni (0.63 \pm 0.07); Al (4.27 \pm 0.53)	[41]
Nicaragua		24	Hg (0.0007 \pm 0.00001); Cu (14.1 \pm 3.16); Pb (0.10 \pm 0.04); Cr (0.06 \pm 0.01); Cd (0.01 \pm 0.01); Ni (0.48 \pm 0.03); Al (3.28 \pm 0.34)	
Poland		23	Cd (3.300 \pm 2.464) in dry coffee and (3.101 \pm 2.471) in infusion; Pb (48.22 \pm 16.27) in dry coffee (46.21 \pm 15.85) in infusion	[43]
		8	Cd (0.058); Pb (0.140); Hg (0.006), As (< LOQ = 0.1)	[13].
		4	Mn (6.28 \pm 1.01); Co (0.06 \pm 0.02); Ni (0.89 \pm 0.07); Cr (0.06 \pm 0.04); Ag (0.11 \pm 0.04); Pb (0.86 \pm 0.17)	[40]
Bosnia and Herzegovina	GFAAS	8	Na (18.6 \pm 11.31); K (19898 \pm 445.48); Ca (789 \pm 132.23); Mg (1758 \pm 35.36); Fe (60 \pm 21.14); Cu (14 \pm 6.43); Zn (3.6 \pm 0.67); Al (4.2 \pm 2.06); Ni (0.415 \pm 0.04); Pb (0.076 \pm 0.0956); Cd (0.015 \pm 0.0005) in green coffee Na (23 \pm 8.80); K (23817 \pm 4779.12); Ca (869 \pm 64.30); Mg (1992 \pm 153.91); Fe (41.1 \pm 5.80); Cu (11.4 \pm 4.13); Zn (5.41 \pm 0.50); Al (4.19 \pm 1.47); Ni (0.88 \pm 0.22); Pb (0.0169 \pm 0.0041); Cd (0.0140 \pm 0.0041) in roasted coffee	[42]
	ICP-MS	3	Mn (4.97 \pm 0.10); Co (0.07 \pm 0.02); Ni (0.88 \pm 0.08); Cr (0.05 \pm 0.02); Ag (0.08 \pm 0.04); Pb (0.71 \pm 0.13)	[40]
Algeria	ICP-MS	44	Ni (1.59 \pm 0.65); Cr (5.62 \pm 1.60); Pb (3.62 \pm 2.49); Zn (6.42 \pm 2.69); Cu (16.23 \pm 8.64); Co (0.52 \pm 0.27); Cd (23.08 \pm 9.03); As (0.46 \pm 0.23); Mn (27.10 \pm 32.63); Al (92.5 \pm 48.79) in local samples packaged in cartons and caps. Ni (0.009 \pm 0.0013); Cr (0.028 \pm 0.009); Pb (0.062 \pm 0.019); Zn (0.409 \pm 0.077); Cu (0.045 \pm 0.021); Co (0.002 \pm 0.001); Cd (0.0375 \pm 0.022); As (0.114 \pm 0.015); Mn (0.0035 \pm 0.002); Al (8.5 \pm 1.507) in local roasted coffee beans marketed in bulk	The present study

GFAAS, graphite furnace atomic absorption spectrophotometer; LOQ, limit of quantification; LOD, limit of detection; N, sample size. Note: Different units included in the literature are all converted into mg/kg

compared to the amounts reported for Brazil, India, Columbia, Ethiopia, and Kenya [18]. Contamination of food and beverages with Cd is of high health risk, which is classified in group I of human carcinogens by the International Agency for Research on Cancer (IARC), its accumulation may cause neurodegenerative disorders, end-stage renal disease (ESRD), breast cancer, prostate cancer, demineralization of bones, and diabetes [39].

The content of Zn was slightly lower (4.50 ± 4.03 mg/kg) compared to the previous TE and to the amounts reported in Kenyan (10.57 ± 10.30 mg/kg), Ethiopian (7.46 ± 4.89 mg/kg), and Indian (8.57 ± 9.48 mg/kg) coffee [18], and almost similar to the amount registered in Columbian (5.99 ± 6.93 mg/kg) coffee [18, 41]. Despite the fact that Zn is essential for numerous biochemical and physiological functions, metabolism including absorption, excretion, and homeostasis [50], Zn excess perturbs Iron (Fe) and Cu homeostasis, leading to increasing levels of Fe and decreasing levels of Cu in the cell [51].

As, Co, and Ni were the least present TE in collected coffee samples with 0.25 ± 0.18 mg/kg, 0.35 ± 0.30 mg/kg, and 0.81 ± 0.81 mg/kg, respectively. As is a toxic metalloid [52, 53] and its chronic ingestion can cause arsenicosis, psychological effects decreased mental performance, hypertension, cardiovascular disease risk, carotid atherosclerosis, diabetes mellitus, and lung cancer. Arsenic compounds (organic and inorganic) are classified by IARC as carcinogenic in group 2B (organic) and group 1 (inorganic) [39, 54].

Compared to the findings reported by Nedzarek et al. [40], the level of Co measured in this study was much higher than that reported in Poland (0.06 ± 0.02 mg/kg) and Bosnia and Herzegovina (0.07 ± 0.02 mg/kg), which may induce various adverse health effects including neurological (hearing and visual impairment), cardiovascular, and endocrine deficits.

Ni content was almost perfectly similar to that recorded in Lebanon (0.8423 ± 0.023 mg/kg) [40], Brazil (0.88 ± 0.08 mg/kg) [18, 40], Poland (0.89 ± 0.07 mg/kg) [40], and Bosnia and Herzegovina (0.88 ± 0.08 mg/kg) [40]. Ni contamination can occur during food production, processing, or packaging. The tolerable upper intake level for Ni is 1 mg/day [40] and its accumulation in the human body may lead to a variety of adverse effects, such as allergic contact dermatitis, oral hypersensitivity, risk of gingival hyperplasia, lung fibrosis, kidney, cardiovascular diseases, and the respiratory tract cancer. As a result, Ni was classified by the International Agency for Research on Cancer (IARC) as a carcinogenic substance [39, 55].

The level of Cr was slightly high (2.71 ± 2.51 mg/kg) at levels similar to those observed in Brazil (2.12 ± 0.64 mg/kg), Ethiopia (2.03 ± 0.54 mg/kg), Kenya (2.72 ± 1.42 mg/kg), Columbia (2.51 ± 1.89 mg/kg) [18], and way above those reported in Poland (0.06 ± 0.04 mg/kg) and Nicaragua (0.06 ± 0.01 mg/kg) [41]. Although considered as

an essential element for human life [56, 57], it can reduce blood pressure and improve malondialdehyde levels in patients with type 2 diabetes [58] and lead to numerous pathophysiological defects such as allergic reactions, anemia, burns, and damage the sperm and the male reproductive system [59].

Finally, a significant level of Pb equal to 1.13 ± 1.99 mg/kg was recorded in the analyzed samples of coffee, consistent with the results mentioned in the Brazilian (1.93 ± 1.80 mg/kg), Indian (1.44 ± 1.79 mg/kg), Kenyan (1.38 ± 0.97 mg/kg), and Columbian (1.04 ± 0.55 mg/kg) coffee, while being way below that recorded in Poland (48.22 ± 16.27 mg/kg) [18], according to Meyer et al. (2008); Wani et al. (2015) lead is among the most important toxic heavy element, with devastating effects on the human body, adverse health effects occurs at blood lead level above $10 \mu\text{g/dL}$ [60, 61] and long-time exposure may damage the heart, kidneys, brain, affect intelligence, cause lung, and bladder cancer; this heavy metal is classified as carcinogenic for humans (group 2A) by IARC [62].

Health Risk Assessment

Maximal estimated daily intake (EDI) and maximal chronic daily intake (CDI) were calculated in adults, using mass consumed (Msi) of 5 g for coffee and an average body weight (BW) of 69.6 kg for the Algerian population in adults [32]. The maximum EDI and mean CDI in adults were below the reference dose (RfD) [63] for coffee in its different forms. This indicates no risk for consumers through the consumption of coffee, which is in agreement with the data published by Winiarska-Mieczan [43].

Results recorded in this study showed that health risk assessment related to Al and Cd largely exceeded the reference dose (RfD), with a hazard quotient (THQ) way beyond the acceptable level and the overall hazard index (HI) being above 1, especially in ground coffee caps (HI = 30.28) and ground coffee packaged in cartons covered inside with Al (HI = 25.79), which indicate a cumulative risk of more than one metal contained in coffee samples. Moreover, it can be concluded that Al and Cd are the major TE contaminants representing a potential health risk in Algeria.

These results are higher compared to those reported by Taghizadeh et al. [64] in coffee samples collected from Iran market. This situation is even worse because the coffee consumers are exposed to many pollutants at the same time, which may result in additive and/or interactive effects [65]. However, the knowledge of combined or interactive effects remains limited; therefore, it is essential that more research efforts be dedicated in this respect.

Conclusion

The results of the present study indicated that coffee could be contaminated by TE. The comparison between the different forms and packaging types in terms of TE content indicated that ground coffee packaged in cartons and capsules contained higher levels of TE than the samples of coffee beans sold in bulk, suggesting that most of the TE contamination occurred during the molding and packaging processes.

Coffee consumption could have adverse non-carcinogenic effects on consumers' health in the long term; THQ values were higher than 1 for Al, Cd, and Cr. Consequently, HI was higher than 1 for ground coffee packaged in cartons and capsules.

Finally, health risk evaluation using THQ and HI can be used in risk management. The identified or potential risk could lead to some useful recommendations for reducing heavy metal emissions and pollution control methods.

To our knowledge, no studies have been conducted to ascertain the long-term consequences of heavy metals in coffee sold in Algeria. Furthermore, more interdisciplinary research on the relationships between the metal concentrations in coffee samples and those in feed, water, and soil would be quite interesting. This will make it easier to comprehend how heavy metals transfer to and move across various matrices, which will help make it clearer how human exposure occurs. As a result, it will be simpler and more sustainable to apply management measures and policies. For storage, glass bottles are advised in order to prevent TE contamination during packaging. Additionally, new eco-friendly instruments and techniques for conventional chemical inputs used in agriculture must be developed as a preventive measure and to lessen the contamination of coffee crops by heavy metals. Finally, it is strongly advised to use all possible treatments, including phytoremediation, chlorination, adsorption, thermal treatment, ion-exchange, chemical extraction, membrane separation, electrokinetics, and bioleaching, to remove heavy metal contamination from soil, water, and sediments.

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Data Availability The data collected and analyzed for this study can be shared upon request.

Declarations

Conflict of Interest The authors declared no competing interests.

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