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REVIEW ARTICLE



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The concentration of potentially toxic elements (PTEs) in the coffee products: a systematic review and meta-analysis

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Abstra 🕧

Coffee is one of the most consumed products globally, and its contamination with potentially toxic elements (PTEs) occurs throughout the production chain and production. Therefore, the current meta-analysis study aimed to estimate the concentration of essential elements (Cu and Co) and the contamination of PTEs (Ni, Cr, Pb, As, and Cd) in coffee. The recommended databases, including PubMed, Scopus, and ScienceDir 19 were investigated to collect data regarding the contamination of PTEs in coffee products from 2010 to 2021. Among 644 retrieved citations in the identification step, 34 articles were included in the meta-analysis. The pooled mean concentration of essential elements in coffee products is much higher than that of toxic elements (Co (447.106 µg/kg, 95% CI: 445.695–448.518 µg/kg) > Ni (324.175 µg/kg, 95% CI: 322.072–326.278 µg/kg) > Cu (136.171 µg/kg, 95% CI: 134.840 33 7.503 µg/kg) > Cr (106.865 µg/kg, 92 CI: 105.309–108.421 µg/kg) > Pb (21.027 µg/kg, 95% CI: 20.824–21.231 µg/kg) > As (3.158 µg/kg, 95% CI: 3.097–3.219 µg/kg) > Cd (0.308 µg/kg; 95% CI: 0.284–0.332 µg/kg)). Results showed high differences between pooled concentrations of all PTEs in coffee products of different countries.

Keywords Potential toxic elements · Coffee products · PTEs · Systematic review · Meta-analysis

Introduction

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Chemical pollution of the environment (Guan et al. 2021; Li et al. 2021; Lin et al. 2021; Liu et al. 2011, 2020; Quan et al. 2621; Shi et al. 2021) followed by food contamination (Liu et al. 2022; Sun et al. 2022; Wang et al. 2022) has become a global concern. Coffee is an attractive beverage worldwide because of its pleasant aroma and taste. Coffee consumption varies according to geographical location (Godos et al. 2014). Flanders in Northern Europe and Bosnia and Herzegovina in Southern Europe are the largest consun 61s of coffee. The World Coffee Organization states that 500 billion 140s of coffee are consumed annually (Dieng et al. 2017). Coffee and tea have been consumed for hundreds of years and have become an important part of cultural traditions and social life. In addition, people use coffee drinks to increase alertness and work productivity (Samet et al. 2018). The highest caffeine consumption in a typical meal is coffee, energy drinks, and caffeinated pills. The intestate level is tea, and the lowest amount of soda (Rattan et al. 2015; van Dam et al. 2020). Caffeine is an alkaloid belonging to methylxanthine. It is also a chemical stimulant found naturally (Wolde 2014). Studies showed that coffee consumption has



chronic and acute effects on human health (Nędzarek et al. 2013). Caffeine ca 28 ct as an antioxidant to prevent diseases. Caffeine also can reduce the risk of several chronic diseases such as liver cancer, diabetes, coronary heart disease, osteoporosis, and gastritis (Nartea et al. 2022). Moreover, the absorption of many vital nutri 32s such as vitamin B₆ is reduced by caffeine intake and the absorption of essential minerals such as magnesium (Mg), iron (Fe), and calcium (Ca) (de Mejia and Ramirez-Mares 2014; Escott-Stump 2008; Wolde 2014). However, consuming large amounts of this drink is not recommended because of some contaminants, including mycotoxins and potentially toxic elements (PTEs), besides 27 adverse effects of caffeine (Barrea et al. 2021; Batrinou et al. 2020; Berman et al. 2022; Khaneghah et al. 2019; Nourbakhsh and Tajbakhsh 2021; Pavithra 2021; Yazdanpanah et al. 2022).

It needs about 20 essential elements for the proper functioning of the human body. The list of meta 34 elements that are essential for the human body include sodium (Na), potassium (K), Mg, Ca, manganese (Mn), Fe, cobalt (Co), copper (Cu), zinc (Zn), molybdenum (Mo), and elements like tin (Sn), nickel (Ni), and vanadium (V), and are assumed not to be necessary for humans, according to chromium (Cr); based on experimental animal results, these metals were considered essen 43 for the human body in the past (Zoroddu et al. 2019). Cu and Co are esse 72 l elements in the human body, a small amount of which plays a vital role in the body. The amount of Co we need is minimal, and only about 2 mg of it is needed for humans. High levels of Cu can cause cardiovascular disease such as cardiac arrhythmias. In addition, high leve 13 f Cu lead to dementia, lung cancer, and lymphoma (Bost et al. 2016). The presence of Cu in the body interferes with the nervous system and causes Wilson's disease, and also, the presence of Cu in the brain leads to brain damage (Kalita et al. 2019).

PT30 are a serious and worrying environmental problem (Bounar et al. 2020; Gao et al. 2022; Mirmahdi et al. 2022). They are widely distributed in the environment a711 thus intake by humans through water, air, soil, and even the food chain (Alkherraz et al. 2019; Heshmati et al. 2020). Accumulating PTEs in the body has negatively affected human health (Fu and Xi 2020; Mirmahdi et al. 2021). These effects include interfering with the immune and nervous systems, endocrine disorders, and malformations (Khaneghah et al. 2020). Organic compounds of lead (Pb) can be absorbed through the skin and enter the brain, causing toxins in the central nervous system. Pb also causes oxidative stress and causes cells to lose antioxidants (Fu and Xi 2020). Adverse effects of Pb include chromosome aberrations, mutations, DNA5 reakdown, and inhibition of DNA synthesis (Ibrahem et al. 2020). Cadmium (Cd) is a toxic and dangerous metal. The Mrld Health Organization limited the tolerable weekly Cd exposure to 50 µg

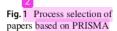
and recommended it as 0.007 111 g/kg body weight (World Health Organization 2000). Depending on the route of exposure and duration, Cd can cause damage to the lung, liver, kidney, bone, testicle, and placenta (Hocaoğlu-Özyiğit and Genç 2020). Arsenic (As) is another P 39 that causes damage and changes in DNA synthesis and cancer, cardiovascular and lung diseases, reproductive outcomes, and cognitive impairment in adults and children (Hu et al. 2022). Studies show that chronic exposure to As causes lung cancer, breast cancer, and bladder, kidney, and larynx diseases. It causes respiratory prob 48 s, infertility in adults, and premature birth in infants (Delgado Quezada et al. 2020; Khan et al. 2020; Pullella and Kotsopoulos 2020). Cr has a different effect on human health depending on the target dose and organ. Effects include respiratory problems, asthma, lung cancer, skin diseases such as dry skin, allergies, dizziness, general weakness, eye irritation, kidney stones, liver problems, gastrointestinal disorders, heart problems, blood disorders, reproduction problems, growth problems, nostril problems, and corneal damage (Ameri et al. 2021). Dental malformations such as discoloration and erosion are accompanied by high concentrations of Cr in the tongue papilla (Achmad and Auerkari 2017; Martineli et al. 2022; Teklay 2016). Ni is an element that may be widely derived from natural resources and human activities in the environment, air, water, and soil. Ni exposure causes digestive diseases, lung problems, lung cancer, and cardiovascular disease (Genchi et al. 2020).

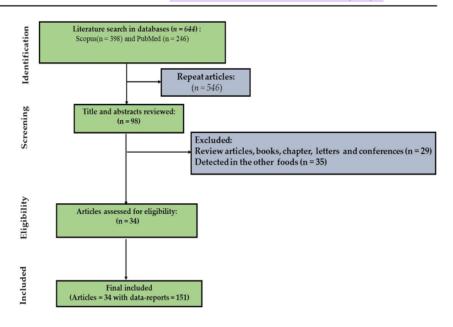
This study systematically reviewed the prevalence of essential elements (Cu and Co) and potentially toxic elements (46 Cr, Cd, Pb, and Ni) in coffee products. Also, another goal of this study was to meta-analyze the results and identified the elements and geographical areas with the highest prevalence.

Material and method

Searching strategy

According to PRISMA guidelines, our study conducted the search strategy (Fig. 1) (Moher et al. 2010). International databases, including Scopus and PubMed (January 2000 and 20 June 2021), were screened to get papers on PTE concentrations in coffee products. Keywords referred to in the title and abstract of citati 81, were ["heavy metal" OR "potentially toxic elements" OR "toxic elements" OR "elements" OR "coffee products" OR "coffee" OR "caffeine" OR "cappuccino" OR "espresso"]. The reference list of papers was assessed for possible papers missed in the title and abstract screening step.





Inclusion and exclusion criteria and extraction of data

Our criteria included the following: detection of PTE concentrations in coffee products, full text available, contamination or prevalence investigation, no language limitation, no restriction for the study location, and present of mean, standard deviation, and/or range concentration of PTEs. One study was excluded because it did not meet the above criteria. One of the authors extracted the data in the retrieved par 51 Data extracted from each article included the country, sample size, the mean and standard deviation of concentration, and measurement method.

8 Meta-analysis of data

The concentration 31 PTEs in coffee products was metaanalyzed based on the standard error (SE) of the equation:

$$SE = \frac{SD}{\sqrt{n}}$$
(1)

where SD is the stan 28 d deviation and n is the number of samples when the f2 index is higher than 50% and heterogeneity is considerable (Higgins et al. 2008; Higg 15 and Thompson 2002; Quan and Zhang 2003); hence, the random-effect model was used for the meta-analysis of the concentration of PTEs in coffee products. PTE concentration's mean and standard deviation were converted to $\mu g/kg$.



A data meta-analysis was performed using Stata, version 14 (Stata Corporation, College Station, TX).

Result and discussion

Our study inclu 19 thirty-four papers with 151 data reports (Fig. 1). The overall rank order of PTEs in the coffee products was as follows: Co (447.106 µg/kg, 95% CI: 445.695–448.518 µg/kg) > Ni (324.175 µg/kg, 95% CI: 322.072–326.278 µg/kg) > Cu (136.171 µg/kg, 95% CI: 134.840–137.503 µg/kg) > Cr (106.865 µg/kg, 95% CI: 105.309–108.421 µg/kg) 33 b (21.027 µg/kg, 95% CI: 20.824–2.231 µg/kg) > As (3.158 µg/kg, 95% CI: 3.097–3.219 µg/kg) > Cd (0.308 µg/kg; 95% CI: 0.284–0.332 µg/kg). As it is obvious from this part of the result, the pooled mean concentration of essential elements in coffee products is much higher than that of toxic elements. Thus, the distribution of PTEs in coffee products is uneven because their content depends on numerous factors (Bost et al. 2016).

It should bear in mind that coffee plants need essential elements (like Co, Ni, and Cu) for their development, so they absorb them from water and soil selectively durin 79 rowth which is influenced by the soil characteristics (chemical composition) in addition to the overall environmental growth conditions (climate, temper 24 e, and agricultural practices) (Barbosa et al. 2014; Habte et al. 2016; Pigozzi et al. 2018). Petrović et al. (2020) also reported that the accumulation of



PTEs in plants relates to the plant's affinity to take up the appropriate element. Furthermore, there is usually a higher concentration of essential elements in the soil than toxic elements since they exist naturally in the rocks and release into water and soil through rock weathering and human activities (like mining) (Atamaleki et al. 2020; Pigozzi et al. 2018). Higher Co, Ni, and Cu pooled concentrations in the coffee products could also be attributed to the greater availability of these PTEs in the soils used for coffee plantation. Therefore, due to the active transmission of essential elements through the soil-root interface and their high content in the soil used for coffee cultivation, they might be readily absorbed by the plant and accumulated in the coffee beans compared with other PTEs 21a Silva et al. 2017). As also evident in a study of da Silva et al. (2017), the elements of Cu, Ni, and Zn are necessary to the growth of the coffee plant, being utilized in the soil or coffee leaf, and are subsequently present in the coffee beans. Furthermore, some PTEs like Cu exist in pesticide ingredients applied in cultivation which assists the uptake of the 58 TEs by plants, validating its presence in coffee beans (da Silva et al. 2017). Albals et al. (2021) also noted that Co and Cu mainly exist at high concentrations in most plants when cultivated in contaminated soil. Cd is found at lower concentrations in such products than in fish.

PTE contamination in coffee products also has diverse forms associated with size, charge, hydrophobicity, activity, bioaccessibility, coffee variety, and type of 40 ocessing (Alkherraz et al. 2019; Pohl et al. 2014; Semen et al. 2017). The concentration of PTEs in coffee is also related to its processing, especially the roasting of coffee beans and coffee beverages' preparation method (Tiep 2014). When it comes

to coffee infusions, several researchers reported that the releasing amount of different PTEs to coffee infusions from the solid sample is different based on coffee type, preparation method, and nature of the metal (leachability and rate of complex formation or interaction with 15 er components) (Ashu and Chandravanshi 2011; da Silva et al. 2017; Jeszka-Skowron et al. 2016; Welna et al. 2013; Winiarska-Mieczan et al. 2021).

Concentration of As

As, a widespread toxic environmental contaminant, enters into the environment (air, water, and soil) through natural and anthropogenic (mainly pesticides used in agricultural practices) sources and is found in t 59 hatural world in organic, inorganic, and gaseous forms (Fu and Xi 2020; Petrović et al. 2020). Inorganic and gaseous forms of As are highly toxic, while organic As is considered relatively nontoxic in very low concentrations (Pullella a 52 Kotsopoulos 2020). Exposure to As is of great concern. It has been related to adverse health effects, including cancer (skin, bladder, and lungs), damaged neurological growth, cardiovascular dis 6 der, and diabetes (Pullella and Kotsopoulos 2020).

The rank order of countries based on the concentration of As in the coffee products was observed in the following: Brazil (48.882 µg/kg) > Netherland (74.981 µg/kg) > Greece (40.000 µg/kg) > Austria (20.050 µg/kg) > France (16.452 µg/kg) > Turkey (10.000 µg/kg) > Ethiopia (6.000 µg/kg) > Ita (17.250 µg/kg) > Germany (0.259 µg/kg) > Indonesia (0.100 µg/kg) ~ Romania (0.100 µg/kg) ~ Rwanda (0.100 µg/kg) ~ Slovenia (0.100 µg/kg)

Table 1 Meta-analysis soncentration of As in the coffee products (μg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	<i>I</i> ² (%)
Austria	20.050	2.350	59.152	4.43	58,897.79	1	< 0.001	100.00
Brazil	178.882	128.720	229.043	0.39	4232.54	2	< 0.001	100.00
Ethiopia	6.000	5.689	6.311	1.64	0.00	0		
France	16.451	15.290	17.612	7.27	92,121.16	2	< 0.001	100.00
Germany	0.259	0.181	0.336	11.55	6968.40	5	< 0.001	99.90
Greece	40.000	39.678	40.322	1.59	0.00	0		
Indonesia	0.100	0.094	0.106	2.84	0.00	0		
Italy	1.250	1.035	1.465	6.07	9419.07	4	< 0.001	100.00
Netherland	74.981	6.382	143.580	1.62	1795.05	1	< 0.001	99.90
Romania	0.100	0.097	0.103	5.68	0.00	1	1	0.00
Rwanda	0.100	0.096	0.104	2.84	0.00	0		
Slovenia	0.100	0.098	0.102	5.68	0.00	1	1	0.00
Turkey	10.000	8.303	11.697	0.12	0.00	0		
Western Balkan	0.100	0.094	0.106	48.28	554.78	18	< 0.001	96.80
Overall	3.158	3.097	3.219	100.00	300,000.00	48	< 0.001	100.00



kg) ~ Western Balkan (0.100 μ g/kg) (Table 1). Since the maximum permitted level (MPL) of As in coffee is 1000 μ g/kg (Petrović et al. 2020), it could be concluded that the As 70 centration of coffee products in all investigated countries does not show a risk to human health concerning this metal.

Concentration of Ni

Ni is naturally present in the soil and widespread distribution 50 the environment (Genchi et al. 2020). This metal has commercial and industrial applications, including metallurgy, electronics, batteries, jet turbines, and medical tools (Şaylan et al. 2020). As an essential element for all living things, Ni has several beneficial impacts on different body organs' functions and metabolism, up to the permitted level. In this regard, Ni influences the liver and pancreas performances, the Fe absorption/metabolism, and the hematopoietic process (Gogoasa et al. 2013). However, in elevated concentrations, Ni can be dangerous for human health by creating respiratory disorders, including lung embolism, respiratory malfunction, asthma, chronic bronchitis, and allergic reactions (Nedzarek et al. 2013). As high long-time exposure to Ni is related to the development of different kinds of cancer such as lung, larynx, and prostate, it is recognized by the International Agency for Research on Cancer (IARC) as a carcinogen (Nędzarek et 272013; Voica et al. 2016).

The rank order of countries based on the concentration of Ni in the coffee products was as follows: Turkey (9695.861 μg/kg) > France (8615.085 μg/kg) > Netherland (5881.104 μg/kg) > India (3665.463 μg/kg) > Italy (2846.081 μg/kg) > Greece (2540.000 μg/kg) > Austria (2274.536 μg/kg) > Romania (1782.844 μg/kg) > Kenya (1630.881 μg/kg) > Brazil (1128.809 μg/kg) > Poland (1046.963 μg/kg) > Lebanon (842.300 μg/kg) > Bosnia (780.000 μg/kg) > Ethiopia (777.455 μg/kg) > Honduras (580.784 μg/kg) > Pakistan (410.000 μg/kg) > Hawaii (356.879 μg/kg) > Colombia (34.796 μg/kg) > Germany (5.047 μg/kg) > Western Balkan (4.471 μg/kg) > Slovenia (0.100 μg/kg) (Table 2).

There is no MPL for Ni in coffee beans, although Brazilian legislation authoriz an MPL of 5000 μ g/kg for Ni in typical foods (da Silva et al. 2017; Pigozzi et al. 2018). Our results showed that the Ni concentrations in the coffee samples of all countries do not surpass this maximum level except for the coffee samples of Turkey, France, and the Netherland.

Table 2 Meta-analysis concentration of Ni in the coffee products (μg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	$I^{2}(\%)$
Austria	2274.536	344.647	4204.426	2.63	23,640.73	1	< 0.001	100.00
Bosnia	780.000	731.668	828.332	0.18	0.00	0		
Brazil	1128.809	1033.256	1224.361	5.40	7,300,000.00	8	< 0.001	100.00
Colombia	34.796	3.540	101.104	3.95	146.58	1	< 0.001	99.30
Ethiopia	777.455	774.516	780.394	13.63	1,400,000.00	7	< 0.001	100.00
France	8615.085	1490.616	16,000.000	0.03	3581.61	1	< 0.001	100.00
Germany	5.047	4.391	5.704	7.05	87,862.69	5	< 0.001	100.00
Greece	2540.000	2514.223	2565.777	0.53	0.00	0		
Hawaii	356.879	124.475	589.284	0.18	48.95	4	< 0.001	91.80
Honduras	580.784	565.420	596.147	1.16	0.12	1	0.732	0.00
India	3665.463	3429.303	3901.623	0.01	0.04	1	0.836	0.00
Italy	2846.081	2002.296	3689.867	3.33	35,250.89	5	< 0.001	100.00
Kenya	1630.881	1584.530	1677.232	0.19	0.05	1	0.83	0.00
Lebanon	842.300	833.284	851.316	1.71	0.00	0		
Netherlands	5881.104	5516.598	6245.610	0.07	4.38	1	0.036	77.20
Pakistan	410.000	300.242	519.758	0.04	0.00	0		
Poland	1046.963	698.597	1395.328	4.60	13,874.64	8	< 0.001	99.90
Romania	1782.844	1304.206	2261.481	2.59	1295.84	5	< 0.001	99.60
Slovenia	0.100	0.097	0.103	5.01	0.00	1	1	0.00
Turkey	9695.861	1993.787	17,000.000	0.17	43,968.77	3	< 0.001	100.00
Western Balkan	4.471	4.339	4.604	47.56	360,000.00	18	< 0.001	100.00
Overall	324.175	322.072	326.278	100.00	310,000,000.00	91	< 0.001	100.00



Concentration of Cr



Both kinds of Cr (Cr3+ and Cr6+) could enter the air, water, and soil through natural processes and industrial activities (like steel, leather, chemical, electronic, painting, and textile manufacturing) (Teklay 2016). Cr3+, an essential element in the body, influences metabolism by adjusting blood sugar levels in r65 tively small amounts (Gogoasa et al. 2013; Nędzarek et al. 2013). It is worth noting that Cr⁶⁺ is more active and toxic than Cr3+. This element occurs at many high levels in industrial activities, which could release Cr6+ into the environment (Achmad and Auerkari 2017). However, Cr3+ is ubiquitous in the environment (water and soil) and 63 logical systems (Voica et al. 2016). It is reported that exposure to a high concentration of Cr could result in several physiological effects, including skin harm; respiratory injuries; gastrointestinal disorders; cardiovascular diseases; hepatic failure; carcinogenic, genotoxic, and neurotoxic mutagenic effects; and reproductive and developmental tox 6 ity (Teklay 2016).

The rank order of countries based on the concentration of Cr in the coffee products was as 20 pws: Netherland $(34,000.000 \mu g/kg) > France (20,000.000 \mu g/kg) > Egypt (2500.000 \mu g/kg) > Kenya (714.922 \mu g/kg) > India (610.321 \mu g/kg) > Indonesia (600.000 \mu g/kg) > Honduras (290.000 \mu g/kg) > Ethiopia (246.854 \mu g/kg) > Hawaii (239.039 \mu g/kg) > Brazil (150.930 \mu g/kg) > Colombia$

(121.610 $\mu g/kg$) > Poland (60.000 $\mu g/kg$) > Bosnia (40.000 $\mu g/kg$) > Lebanon (35.000 $\mu g/kg$) > Germany (0.643 $\mu g/kg$) > Italy (0.134 $\mu g/kg$) > Austria (0.100 $\mu g/kg$) ~ Greece (0.100 $\mu g/kg$) (Table 3).

Similar to Ni, there are no reported maximum levels for the existence of the Cr metal in coffee products. At the same time, Brazilian regulation has established a MPL of 100 µg/kg for this element in the food (da Silva et al. 2017). Our results showed that the pooled concentrations of Cr in coffee products of all investigated countries exceed the maximum permitted level except Poland, Bosnia, Lebanon, Germany, Italy, Austria, and Greece.

Concentration of Cu

Cu is a vital element in numerous human body functions like antioxidant defense, Fe metabolism, enzyme functions, energy production, bone formation, and skin health in trace amounts (Bost et al. 2016; Gogoasa et al. 2017). Furthermore, lacking Cu may cause anemia, hair whitening, and bone distortion (Petrović et al. 2020). However, exposure to high concentrations of Cu is toxic and could result in stomach pain, nausea, diarrhea, peroxidation of macromolecules, and tissue injury (Anosike and Oranusi 2018; Bost et 3 2016).

The rank order of countries based on the concentration of Cu in the coffee products was Germany (38,000.000 µg/

Table 3 Meta-analysis concentration of Cr in the coffee products (µg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	$I^{2}(\%)$
Austria	0.100	0.098	0.102	11.70	0.00	1.00	1	0.00
Bosnia	40.000	28.181	51.819	1.34	0.00	0.00		
Brazil	150.930	139.951	161.908	13.08	54,518.83	8.00	< 0.001	100.00
Colombia	121.610	102.443	140.778	11.82	3178.71	5.00	< 0.001	99.80
Egypt	2500.000	2428.432	2571.568	0.05	0.00	0.00		
Ethiopia	246.854	209.473	284.236	19.86	1759.86	7.00	< 0.001	99.60
France	20,000.000	220.000	46,000.000	0.01	4284.30	1.00	< 0.001	100.00
Germany	0.643	0.258	1.028	12.37	30,137.84	5.00	< 0.001	100.00
Greece	0.100	0.097	0.103	5.85	0.00	0.00		
Hawaii	239.039	54.928	423.150	2.30	142.07	4.00	< 0.001	97.20
Honduras	290.000	279.191	300.809	1.65	0.00	1.00	1	0.00
India	610.321	308.647	911.996	1.62	744.43	3.00	< 0.001	99.60
Indonesia	600.000	511.357	688.643	0.03	0.00	0.00		
Italy	0.134	-0.170	0.438	11.71	18,762.12	4.00	< 0.001	100.00
Kenya	714.922	691.221	738.624	0.41	0.52	1.00	0.472	0.00
Lebanon	35.000	34.409	35.591	5.80	0.00	0.00		
Netherlands	34,000.000	233.240	79,000.000	0.00	4397.72	1.00	< 0.001	100.00
Poland	60.000	36.362	83.638	0.40	0.00	0.00		
Overall	106.865	105.309	108.421	100.00	28,000,000.00	58.00	< 0.001	100.00



kg) > Austria (20,000.000 μ g/kg) > Kenya (19,000.000 μ g/k4) ~ Netherland (19,000.000 μ g/kg) > Honduras (17,000.000 μ g/kg) > India (14,000.000 μ g/kg) > Colombia (13,000.000 μ g/kg) ~ Costa Rica (13,000.000 μ g/kg) ~ Guatemala (14,000.000 μ g/kg) ~ Poland (13,000.000 μ g/kg) > Nicaragua (12,000.000 μ g/kg) > Peru (12,000.000 μ g/kg) > Ethiopia (4207.608 μ g/kg) > Italy (6598.028 μ g/kg) > Greece (6520.000 μ g/kg) > Nigeria (3970.000 μ g/kg) > France (3538.513 μ g/kg) > Brazil (2804.322 μ g/kg) > Egypt (2200.09 μ g/kg) > Hawaii (146.762 μ g/kg) > Indonesia (90.000 μ g/kg) (Table 4).

Our results showed that the concentrations of Cu in coffee products of all studied countries meet the MPL for Cu, determined by Brazilian legislation (30,000 µg/kg) (Alkherraz et al. 2019; Anosike and Oranusi 2018), except in Germany. Cu is usually used in the composition of some pesticides used in coffee fields as an active substance in different forms (Pigozzi et al. 2018). It is necessary for plant growth and is absorbed by plants during cultivation. Senkondo et al. (2014) observed that Cu concentrations in the coffee samples grown in the Cu-based pesticide soils were much higher compared with those in untreated soils, probably du 38) more availability of Cu for plant uptake, which could be the main reason for observed differences in Cu concentrations in different

countries in the present study. However, the soil properties, climate conditions, and differences in 75 free bean varieties are also influential factors (Alkherraz et al. 2019; Senkondo et al. 2014).

Concentration of Co

Co, another essential element, is a constituent of vitamin B₁₂, which is vital 32 creating red blood cells (RBCs). It also participates in the appropriate functions of the nervous system, the self-defense of the body, activation of specific enzymes, and prevention of anemia (Gogoasa et al. 2013; Voica 78 al. 2016). However, a high Co intake could pr 73 ce toxic effects on human and animal health, especially in the

he and liver (Nędzarek et al. 2013; Zoroddu et al. 2019).

The rank order of countries based on the concentration of Co in the coffee products was Ethiopia (2686.563 µg/kg) > France (865.526 µg/kg) > Austria (589.968 µg/kg) > Poland (428.061 µg/kg) > Italy (419.919 µg/kg) > Netherland (314.897 µg/kg) > Germany (308.238 µg/kg) > Hawaii (221.329 µg/kg) > Turkey (140.20) µg/kg) > Slovenia (124.982 µg/kg) > Greece (110.000 µg/kg) > Lebanon (66.000 µg/kg) > Bosnia (60.000 µg/kg) > Brazil (34.913 µg/kg) > Romania (0.102 µg/kg) (Table 5). There are no safety

Table 4 Meta-analysis concentration of Cu in the coffee products (µg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	<i>I</i> ² (%)
Austria	20,000.000	6970.172	34,000.000	0.00	2702.34	1	< 0.001	100.00
Brazil	2804.322	2712.544	2896.099	25.44	33,702.51	11	< 0.001	100.00
Colombia	13,000.000	10,000.000	15,000.000	0.00	345.48	6	< 0.001	98.30
Costa Rica	13,000.000	11,000.000	15,000.000	0.00	0.00	0		
Egypt	2200.000	2164.216	2235.784	0.14	0.00	0		
62 lopia	9097.608	9004.660	9190.556	34.79	350,000.00	18	< 0.001	100.00
France	3538.513	2483.860	4593.166	13.73	17,261.65	3	< 0.001	100.00
Germany	38,000.000	27,000.000	48,000.000	0.35	66,115.44	5	< 0.001	100.00
Greece	6520.000	6420.113	6619.887	0.02	0.00	0		
Guatemala	13,000.000	9995.342	16,000.000	0.00	5.05	1	0.025	80.20
Hawaii	146.762	53.569	239.955	0.02	0.05	4	1	0.00
Honduras	17,000.000	16,000.000	17,000.000	0.00	0.08	1	0.783	0.00
India	14,000.000	11,000.000	16,000.000	0.00	184.80	4	< 0.001	97.80
Indonesia	90.000	76.704	103.296	0.93	0.00	0		
Italy	6598.028	6425.142	6770.915	23.38	64,400.57	6	< 0.001	100.00
Kenya	19,000.000	19,000.000	20,000.000	0.00	0.14	1	0.704	0.00
Netherlands	19,000.000	16,000.000	21,000.000	0.00	115.12	1	< 0.001	99.10
Nicaragua	12,000.000	10,000.000	13,000.000	0.00	1.39	1	0.238	28.20
Nigeria	3970.000	3958.240	3981.760	1.17	0.00	0		
Peru	12,000.000	11,000.000	14,000.000	0.00	0.00	0		
Poland	13,000.000	12,000.000	14,000.000	0.00	0.00	0		
Overall	136.171	134.840	137.503	100.00	1,300,000.00	83	< 0.001	100.00



Table 5 Meta-analysis concentration of Co in the coffee products (µg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	I ² (%)
Austria	589.968	2.354	1217.157	4.34	8914.82	1	< 0.001	100.00
Bosnia	60.000	53.556	66.444	2.23	0.00	0		
Brazil	34.913	2.478	103.251	5.80	256.51	1	< 0.001	99.60
Ethiopia	2686.563	2464.899	2908.227	20.99	4,300,000.00	10	< 0.001	100.00
France	865.526	738.133	992.919	1.20	75.70	1	< 0.001	98.70
Germany	308.238	257.686	358.790	17.02	5516.66	5	< 0.001	99.90
Greece	110.000	106.778	113.222	3.41	0.00	0		
Hawaii	221.329	140.513	302.144	0.48	56.44	4	< 0.001	92.90
Italy	419.919	291.202	548.637	13.43	16,640.50	4	< 0.001	100.00
Lebanon	66.000	63.256	68.744	3.59	0.00	0		
Netherlands	314.897	89.501	540.293	4.24	1003.74	1	< 0.001	99.90
Poland	428.061	1.654	863.858	2.41	333.91	2	< 0.001	99.40
Romania	0.102	0.047	0.156	12.72	815.60	5	< 0.001	99.40
Slovenia	124.982	56.383	193.581	7.50	1693.64	1	< 0.001	99.90
Turkey	140.000	123.831	156.169	0.64	0.00	0		
Overall	447.106	445.695	448.518	100.00	4,800,000.00	49	< 0.001	100.00

ES effect size (mean of concentration)

limits established for Co concentration in coffee products. However, there is an MPL of 10 μ g/kg for this element in tea (Yaqub et al. 2018). Our results revealed that Co concentration in coffee samples of all countries is above this limit (except Romania), representing possible health risks to the population through coffee consumption.

Concentration of Cd

Cd is the most toxic PTE (Gogoasa et al. 2013; Voica et al. 2016). Research has not revealed any positive impacts of Cd on human an 26 nimal health (Alkherraz et al. 2019), while they could be exposed to Cd through food and water consumption or work conditions. This element could accumulate a high amount in the human organs during lifespan. It is detrimental to kidney function and could raise bone demineralization, even at low concentrations. Cd also could cause lung cancer with long-term exposure (Alkherraz et al. 2019). Given its water solubility trait, Cd could be released and spread quickly to the environment through natural rock weathering, agricultural practices, and industrial activities, including batteries, refining ores, metal alloys, and plastic, dye, ceramic, and glass production. This element is also present in phosphate manures and pesticides at small levels and subsequently could be taken up by plants from treated soils wit 6 such compounds (Alkherraz et al. 2019).

The rank order of countries based on the concentration of Cd in the coffee pro 56 ts was Indonesia (300.000 µg/kg) > Brazil (61.541 µg/kg) > Honduras (50.000 µg/kg) > India (40.000 µg/kg) > Colombia (25.010 µg/

kg) > Poland (13.000 μ g/kg) > Kenya (1(200 μ g/kg) > Netherland (5.041 μ g/kg) > Italy (1.160 μ g/kg) > 17 many (0.676 μ g/kg) > Ethiopia (0.183 μ g/kg) > Austria (0.100 μ g/kg) ~ France (0.100 μ g/kg) 2 Greece (0.100 μ g/kg) ~ Jamaica (0.100 μ g/kg) ~ Lebanon (0.100 μ g/kg) ~ Malawi (0.100 μ g/kg) ~ Pakistan (0.100 μ g/kg) (Table 6).

The MPL for Cd set by the Europea 47 Jnion (EU) is 100 µg/kg in coffee products (da Silva et al. 2017). The results showed that the pooled concentration of Cd in coffee products of all studied countries meets the regulation limit except Indonesia.

To this end, geographical location in waste zones and industrial units that release Cd to the environment are critical in Cd concentration in food products. In this regard, synthetic fertilizers in farming sections, contaminated water, and agricultural fields with Cd could increase human exposure to this toxic element through food consumption (Fu and Xi 2020; Hocaoğlu-Özyiğit and Genç 2020).

Concentration of Pb

Pb, another highly toxic element, could accumulate in the body (Gogoasa et al. 2013). It is a non-essential metal for living things (Wadhwa et al. 2014). Pb is present everywhere in the environment (Lee et al. 2020). Plants like coffee beans 74 ld easily absorb Pb from the soil, water, and air (Lee et al. 2020). The significant harmful effects of these PTEs on human and animal health are carcinogenicity, neurotoxicity, hepatotoxicity, endocrine toxicity, reproductive and developmental toxicity, cardiovascular diseases, and gastrointestinal



Table 6 Meta-analysis concentration of Cd in the coffee products (µg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	<i>I</i> ² (%)
Austria	0.100	0.098	0.102	7.28	0.00	1	1	0.00
Brazil	61.541	45.578	77.503	0.26	3208.74	7	< 0.001	99.80
Colombia	25.010	3.658	73.714	3.64	622.50	1	< 0.001	99.80
Ethiopia	0.183	0.158	0.207	42.52	2660.06	13	< 0.001	99.50
France	0.100	0.098	0.102	7.28	0.00	1	1	0.00
Germany	0.676	0.593	0.759	9.01	1743.32	5	< 0.001	99.70
Greece	0.100	0.097	0.103	3.64	0.00	0		
Honduras	50.000	48.544	51.456	0.03	0.00	1	1	0.00
India	40.000	36.494	43.506	0.00	0.00	1	1	0.00
Indonesia	300.000	255.679	344.321	0.00	0.00	0		
Italy	1.160	0.982	1.339	7.61	7633.71	4	< 0.001	99.90
Jamaica	0.100	0.096	0.104	3.64	0.00	0		
Kenya	10.000	9.649	10.351	0.44	0.00	1	1	0.00
Lebanon	0.100	0.094	0.106	3.64	0.00	0		
Malawi	0.100	0.096	0.104	3.64	0.00	0		
Netherlands	5.041	2.647	14.739	3.73	580.20	1	< 0.001	99.80
Pakistan	0.100	0.091	0.109	3.63	0.00	0		
Poland	13.000	10.581	15.419	0.01	0.00	0		
Overall	0.308	0.284	0.332	100.00	25,346.57	53	< 0.001	99.80

ES effect size (mean of concentration)

and hepatic disorders (da Silva et al. 2017; Pigozzi et al. 2018).

The rank order of countries based on the concentration of Pd in the coffee products was Indonesia (3900.000 µg/kg) > Syria (2492.013 µg/kg) > Egypt (1900.000 µg/kg) > Bosnia (1050.000 µg/kg) > Lebanon (693.000 µg/kg) > Austria (350.000 µg/kg) > Netherland (274.986 µg/kg) > Italy (221.898 µg/kg) > Brazil (203.618 µg/kg) > Greece (200.000 µg/kg) > Germany (183.279 µg/kg) > T4 key (120.000 µg/kg) > India (92.630 µg/kg) > Slovenia (7(4)00 µg/kg) > Poland (60.934 µg/kg) > France (23.829 µg/kg) > Pakistan (20.000 µg/kg) > Honduras (15.039 µg/kg) > Colombia (13.48 µg/kg) > Kenya (9.949 µg/kg) > Hawaii (8.895 µg/kg) > Romania (0.459 µg/kg) > South Korea (0.240 µg/kg) > Western Balkan (0.137 µg/kg) > Ethiopia (0.104 µg/kg) (Table 7).

Polish food legislation, Brazilian legislation, and the EU have set the MPLs for Pb in coffee products as 1000 μg/kg, 500 μg/kg, and 200 μg/kg, respectively (Albals et al. 2021; Azam et al. 2021; Jeszka-Skowron et al. 2016). It could be concluded that the investigated coffees of all countries are not dangerous to human health except for Indonesia, Syria, Egypt, and Bosnia. However, using Brazil and EU safe limits, one country (Lebanon) and 5 other countries (Lebanon, Austria, Netherland, Italy, Brazil, and Greece) do not meet these regulation limits, respectively.

As observed previously, there are high differences between pooled concentrations of all PTEs in coffee products of different countries. Different factors are influential in these differences, which are discussed in the following.

First of all, coffee characteristics include variety, species (Arabica or Robusta), ripeness, type (brand/raw or roasted/ground or instant/infusion or powder), size, and processing conditions such as grinding, drying, roasting, mixing, and brewing methods which are effective in the obse 57 d difference in the content of PTEs in coffee products in different 80 ntries (Albals et al. 2021; Gure et al. 2017). Furthermore, the origin of the coffee regarding environmental conditions, including soil properties (type of soil, organic matter concentration, pH, drainage grade), geographical location (climate condition), and agricultural practices (using fertilizers and 53 ticides, organic or conventional farming), has a crucial impact on the 60 tent of PTEs in coffee beans (da Silva et al. 2017; Habte et al. 2016).

It is worth noting that both PTEs' gains and losses may occur during coffee processing which can be another source of observed differences in their concentration in different countries. The type of water used for infusion is also effective (Ashu and Chandravanshi 2011). In the case of the type of coffee products, it is reported that mixtures of coffee substitutes or surrogates (cereal grains) with coffee (20–66%) intermediate exhibit contents. At the same time,



Table 7 Meta-analysis concentration of Pb in the coffee products (µg/kg)

Country	ES	Lower	Upper	Weight (%)	Heterogeneity statistic	Degrees of freedom	P value	<i>I</i> ² (%)
Austria	350.000	310.801	389.199	0.19	74.00	1	< 0.001	98.60
Bosnia	1050.000	919.991	1180.009	0.00	0.00	0		
Brazil	203.618	199.095	208.142	4.41	2,700,000.00	11	< 0.001	100.00
Colombia	13.488	2.091	24.886	3.46	853.99	5	< 0.001	99.40
Egypt	1900.000	1896.422	1903.578	0.28	0.00	0		
Ethiopia	0.104	0.084	0.123	23.89	2152.79	12	< 0.001	99.40
France	23.829	22.977	24.681	4.94	23,091.15	3	< 0.001	100.00
Germany	183.279	117.326	249.233	3.94	41,156.25	5	< 0.001	100.00
Greece	200.000	196.778	203.222	0.33	0.00	0		
Hawaii	8.895	3.437	14.352	0.46	7.36	4	0.118	45.70
Honduras	15.039	3.457	44.322	2.85	1396.88	1	< 0.001	99.90
India	92.630	46.659	138.602	2.19	575.47	3	< 0.001	99.50
Indonesia	3900.000	3323.822	4476.178	0.00	0.00	0		
Italy	221.898	125.044	318.752	2.39	24,069.10	4	< 0.001	100.00
Kenya	9.949	3.546	29.371	2.19	99.00	1	< 0.001	99.00
Lebanon	693.000	681.772	704.228	0.03	0.00	0		
Netherlands	274.986	4.560	617.980	0.80	10,395.51	1	< 0.001	100.00
Pakistan	20.000	9.265	30.735	0.04	0.00	0		
Poland	60.934	38.272	83.597	3.04	1529.02	3	< 0.001	99.80
Romania	0.459	0.338	0.579	10.41	11,222.07	6	< 0.001	99.90
Slovenia	70.000	69.089	70.911	2.19	0.00	1	1	0.00
South Korea	0.240	0.198	0.282	1.95	0.00	0		
Syria	2492.013	2231.473	2752.552	0.00	0.56	1	0.453	0.00
Turkey	120.000	103.026	136.974	0.01	0.00	0		
Western Balkan	0.137	0.116	0.158	30.01	4431.90	18	< 0.001	99.60
Overall	21.027	20.824	21.231	100.00	4,100,000.00	104	< 0.001	100.00

ES effect size (mean of concentration)

lower amounts a 49 letermined in coffee surrogates without coffee addition (Oliveira et al. 2012).

In the case of the brewing method, it is reported that the extraction levels of most PTEs are highest in the Turkish coffee meth 67 in which coffee is boiled together with water (Nędzarek et al. 2013; Şemen et al. 2017). Regarding the type of coffee in terms of raw or roasted, it is noteworthy that the concentrations of PTEs in green coffee are typically lower than those determined in roasted coffees, probably due to 15 moving water and volatile constituents (Amorim Filho et al. 2007; da Silva et al. 2017). Moreover, Şemen et al. (2017) concluded that studied PTEs are leached to the infusion more difficult from roasted coffee than from green coffee. Ashu and Chandravanshi (2011) also observed that the infusions con 24n lower PTEs than the roasted coffee. Moreover, Welna et al. (2013) noted that the transfer degree of PTEs into brew might vary probably re66 ed to coffee type (instant and ground coffees). Amorim Filho et al. (2007) also observed that the concentration of Fe and Cr is highest in steel grinders because of releasing these metals into the coffee samples via the mill overheating. Additionally, Nędzarek et al. (2013) reported that potential contamination with PTEs may originate from packaging materials containing metals during coffee storage. There is also a negative relationship between the size of the coffee powder and the extraction rate of PTEs into the infusion (Pigozzi et al. 2018).

Moreover, Ashu and Chandravanshi (2011) report 35 hat the concentration of PTEs in coffee varies among coffee varieties cultivated in diverse parts of the world with different environ 35 nts, soil, and fertilizers.

Further, differences in sampling, sample preparation, and analytical techniques used in different studies significantly impact the concentration of PTEs (Ashu and Chandravanshi 2011). The contents of PTEs in the ground coffee are different from those in instant (soluble) coffee, probably due to processing differences (Vo 77 et al. 2016). There are different kinds of methods used for the determination of PTEs in co 22 products, including atomic absorption spectrometry (AAS), inductively coupled plasma-optical emission

spectrometry (ICP-OES), inductively coupled plasma-mass spectroscopy (ICP-MS), atomic fluctoring cence spectrometry (AFS), and electrodes. Among these methods, ICP-OES and ICP-MS are the most remarkably applied because of their high sensitivity, specificity, selectivity, low detection limits (LODs), ease of setup, less time, and low cost (Albals et al. 2021; Alkherraz et al. 2019). However, to overcome the disadvantages of AFS methods, 41 me pre-concentration methods could be utilized during sample preparation, including solid-phase extraction (SPE), liquid-liquid extraction (LLE), as well as microextraction procedures like dispersive liquid-liquid microextraction (DLLME) (Şaylan et al. 2020).

Conclusion

This study meta-analyzed the prevalence of potentially toxic elements in coffee products in different countries. The pooled mean concentration of essential elements in coffee products is much higher than that of toxic elements (Co > Ni >Cu>Cr>Pb>As>Cd). The result showed that the pooled mean concentration of essential elements in coffee products is much higher than that of toxic elements. Also, high differences between pooled concentrations of all PTEs were reported in coffee products of different countries. Various concentrations of potentially toxic elements in different studies may be due to several factors, including environmental growth conditions (climate, temperature, and agricultural practices), type of processing, coffee variety, preparation method, agricultural practices (using fertilizers and pesticides, organic or cor42 ntional farming), and analytical techniques. Therefore, more studies are needed to investigate the effect of mentioned factors on the prevalence of PTEs in coffee products in the future.

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Data availability Not applicable.

Declarations

Ethics 10 oval and consent to participate Ethics approval is not applicable. The authors declare their consent to participate in this article.

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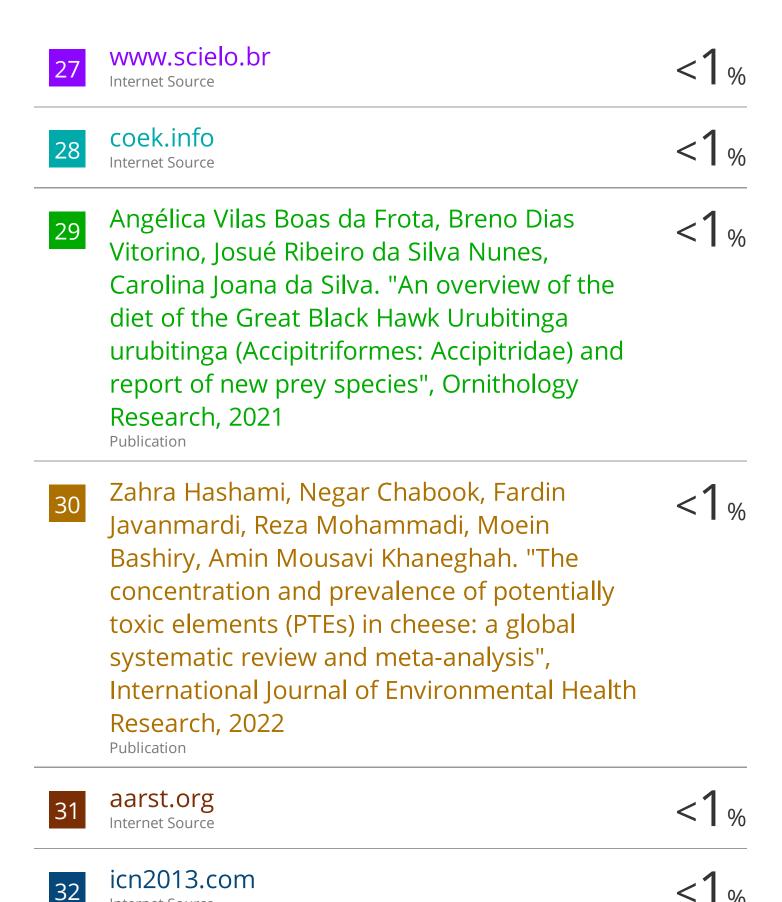
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