Toxic metals in packed rice: Effects of size, type, origin, packing season, and storage duration

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ABSTRACT

The aim of our study was to quantify toxic metals in rice, to determine factors affecting its contamination, and evaluate exposure from its consumption in Lebanon and United Arab Emirates (UAE). For this, a market screening was done and all brands (107 from Lebanon and 129 from UAE) were collected twice and tested using ICP-MS. For As, Cd, Hg and Pb, in UAE, 9%, 1%, 100%, and 69% of samples exceeded the international limits, while in Lebanon, 25%, 73%, 100%, and 69% of samples were above limits, respectively. For As, in Lebanon, brown rice, long grains and brands from developed countries were significantly more contaminated, while in UAE, packing season, country of origin, and collection time had significant effect. For Cd, collection time had significant effect in Lebanon and UAE. Regarding Cr, in Lebanon, country of origin, grain size, rice type, and time between packing and purchasing had significant effect, while in UAE, collection time had significant effect. For Hg, collection time had significant effect in Lebanon and UAE. Alarming exposure levels for Hg and Pb from rice were detected in the UAE. Future studies must assess the effects of handling and cooking to better assess exposure to toxic metals from rice consumption.

1. Introduction

Heavy metals are toxic elements that occur naturally and have high density and atomic weight. Recently, heavy metals' contamination has become a growing hazardous issue worldwide, affecting 235 million hectares of farmlands around the world (Cong et al., 2019). Lead (Pb), chromium (Cr), arsenic (As), cadmium (Cd), and mercury (Hg) are the main metals studied for their high levels of toxicity (Tchounwou et al., 2012). Rice accumulates more metals than other cereals (TatahMentan et al., 2020) and has been extensively studied worldwide, especially because it is a staple food for more than three billion individuals around the world (Mosleh et al., 2015). It is cultivated in more than 100 countries, with the majority (90%) of cultivation and production originating from Asia (Fukagawa & Ziska, 2019). In the UAE, there have been recent trials for planting rice in the desert of the Emirate of Sharjah, using desalinated water along with hybrid rice strains that are grown in saltwater (United States Department of Agriculture, Foreign Agricultural

Service, 2021). On the other hand, Food and Agriculture Organization of the United Nations (FAO) reported that the weather in Lebanon favors cereal growing but expensive inputs stand as obstacles preventing such agricultural practices (FAO, 2020). Therefore, both countries depend mainly on imports for satisfying their local rice demands.

The main sources of toxic metals are geochemical, industrial, and to a limited extent, herbicides and farm animals' growth promoters (Garbinski et al., 2019). The repeated utilization of agrochemicals in agriculture, such as fertilizers and pesticides, may contribute to heavy metals' accumulation in the soil (Zhao et al., 2015). Additionally, sewage sludge and animal manures may contain high amounts of heavy metals contaminating agricultural soils (Chamannejadian et al., 2013). For instance, chromium can distinctively leach from stainless steel utensils and equipment, especially in the presence of high acid food (EFSA CONTAM Panel, 2014). Lead can contaminate food through lead-glazed or lead-soldered containers, and water through leaded pipes (WHO, 2021a, 2021b). Generally, in addition to industrial products and

emissions, smoking, waste incineration, drinking water, agricultural irrigation, and contaminated crops can be important common routes of contamination with toxic metals (Sommella et al., 2013).

Heavy metals are highly toxic to the human health and environment, and their toxicity varies according to their dose, type, way of exposure, age, gender, genetics and nutritional status of the exposed person (Tchounwou et al., 2012). Even at low levels of exposure, they are considered as carcinogens and may cause multiple organ damage (Tchounwou et al., 2012). Consuming contaminated food with toxic metals can also deplete body stores of iron, vitamin C, and other nutrients that are crucial for the human body (Kumar et al., 2020).

Inorganic arsenic compounds (i.e. arsenite and arsenate) are toxic, whereas organic arsenic, usually found in fish and shellfish, is not considered toxic (Olsen et al., 2004; Shraim, 2017). The Agency for Toxic Substances and Disease Registry (ATSDR) has placed As on top of its Hazardous Substances list, not because it is more toxic than other metals, but because of several interrelated factors related to frequency, toxicity, and human exposure potential of the metal. It is linked to several diseases such as cancer, diabetes, cardiovascular diseases, neurological disorders, and chronic kidney disease (Garbinski et al., 2019; Medda, De & Maiti, 2021). Concerning cancer, it is classified as a group I human carcinogen by the International Agency for Research on Cancer (IARC), particularly linked to cancers of the lung, skin, bladder, kidney, and liver (Bielecka et al., 2020). Furthermore, according to Garbinski et al. (2019), the recommendation of the U.S. Food and Drug Administration is to limit infant consumption of food prepared from arsenic-contaminated rice due to the risk of serious developmental problems.

Cadmium chloride ($CdCl_2$) and cadmium oxide (CdO) are toxic (Vallero, 2014) and although only a small percentage of Cd is absorbed via the digestive tract, it has an alarming long biological half-life of 15 years (Bielecka et al., 2020; JECFA, 2010). According to Genchi et al. (2020), Cd has been associated with renal and hepatic dysfunction, osteomalacia, pulmonary edema, adrenal damage, hematopoetic system disruption, altered lipid profile, and coronary heart disease. It is classified as a group I human carcinogen by IARC, mainly related to lung, prostate, endometrium, pancreas, urinary bladder, breast, and naso-pharynx cancers (Bielecka et al., 2020; Genchi et al., 2020). Cd may also hinder the activity of antioxidant enzymes (Genchi et al, 2020).

Cr (III) compounds are commonly used as nutritional supplements due to their association with a decreased risk of diabetes (Tumolo et al., 2020). However, the European Food Safety Authority (EFSA) Panel on Dietetic Products, Nutrition, and Allergies (NDA) (2014) questions its health benefits. The panel points that the evidence for the role of Cr in human metabolism and physiology is not convincing; thus, it considers no proof that it is an essential element (EFSA NDA, 2014). On the other hand, Cr (VI) exposure has been associated with several adverse health effects, particularly related to the skin and respiratory system (Ferreira et al., 2019). Cr (VI) may impose a risk for liver and kidney damage, respiratory disorders, and internal hemorrhage (Tumolo et al., 2020). Furthermore, Cr (VI) compounds have been classified as lung carcinogens by the IARC (group I carcinogen) and the National Toxicology Program (NTP) (Ferreira et al., 2019).

Each form of Hg has its own toxic effect. MeHg is considered the most toxic among the organic Hg forms (EFSA, 2012). The main target organ for the toxicity of Hg is the brain, in addition to having adverse effects on the nervous, renal, and muscular systems and gut lining (Bernhoft, 2012; Bielecka et al., 2020). Prenatal mercury poisoning may range from neurodevelopmental delays and cognitive deficits to cerebral palsy, whereas postnatal exposure may range from paresthesias, ataxia, visual, auditory, and extrapyramidal impairments to clonic seizures in severe exposures (Bernhoft, 2012).

All forms of Pb are toxic, but its organic form Tetra-ethyl Pb is more toxic than the others (Kumar et al., 2020). Upon absorption, Pb accumulates in the blood, bones, liver, kidneys, brain, and skin (Zulkafflee et al., 2019). It may disrupt the development of the nervous system,

especially during the prenatal period through childhood (Mason et al., 2014). During early stages of pregnancy, a high level of serum Pb may hinder fetal growth, whereas in infants and children, it affects cognitive performance, behavior, and postnatal growth, in addition to delaying puberty and altering hearing ability. Furthermore, it may cause cardiovascular and central nervous impairments in adults, along with fertility and kidney problems (Kumar et al., 2020).

In Lebanon and UAE, Hassan et al. (2022a) and Hassan et al. (2022b) assessed the safety of rice in terms of mycotoxins. To date, no study assessed the safety of rice in Lebanon and UAE in terms of toxic metals. The aim of this study was to determine As, Cd, Cr, Pb, and Hg levels in rice marketed in both countries, and to estimate exposure of the Lebanese and Emirati populations to these toxic metals. Also, this is the first study to assess effect of different variables (packing season, country of packing, grain size, rice type, food safety management system certificate, and time between packing and purchasing) on toxic metals in rice.

2. Materials and methods

2.1. Sample collection

Markets in Lebanon and UAE were screened in 2021 for white, parboiled, and brown rice brands. Two packs with different production dates were purchased for each rice brands and the packs were stored in the freezer until analysis. In Lebanon, the first and second collections consisted of 54 and 53 packed rice brands, respectively. Eight brands were not found in the market during the second collection, while seven others were additionally collected. In UAE, the first and second collections consisted of 63 and 66 brands, respectively. Twenty-four brands were not found in the market during the second collection and 27 additional new brands were collected. In total, 236 samples were screened for toxic metals, with 107 samples being from Lebanon and 129 from UAE.

2.2. Sample preparation

Arsenic, cadmium, mercury, and lead standard solutions (Merck, Darmstadt, Germany), Hydrogen peroxide solution 30% (Sigma Aldrich, Germany), Nitric acid 69% (BDH Laboratory supplies, England), Hydrochloric acid 37% (AnalaR Normapur, France) were used in this research. The certified reference material (Rice flour, IRMM-804) was obtained from the Institute for Reference Materials and Measurements (Geel, Belgium) to validate the accuracy of the method. All solutions were prepared with analytical reagent grade chemicals and ultrapure water (18 MV.cm) obtained by purifying distilled water using a Milli-Q water purification system (Millipore S.A., St Quentin-en-Yvelines, France). Rice samples (0.5 g) were digested using a Multiwave ECO microwave digestion system (Anton Paar GmbH, Graz, Austria) equipped with a rotor for 16 pressure-activated-venting vessels (Rotor 24HVT50, Anton Paar GmbH, Graz, Austria) after adding 8 mL of 69% nitric acid and 2 mL of 30% hydrogen peroxide to each of the samples. The sample digestion procedure was performed as follows: (1) 850 W at 180°C for 10 min and (2) 850 W at 22°C for 1 min for cooling. Operating conditions of microwave oven digestion are stated in Table 1. After microwave digestion, 2 mL HCl were added to sample solutions and the digested samples were then transferred into 50 mL polypropylene tubes. The contents were diluted 5 times with 3% nitric acid prepared with ultrapure deionized water and were stored at 4°C until ICP-MS analysis.

Spiking solutions were prepared for two concentrations (0.5 and 5

Table 1
Operating conditions of microwave oven digestion.

	Temperature (°C) ° (°C)	Ramp (min)	Hold (min)	Fan
Step 1	180	20	10	1
Step 2	22	10	0.5	3

ppm) for each metal with 0.5 g of rice sample (TatahMentan et al., 2020; Gedik et al., 2017; Rubio-Armendáriz et al., 2021).

2.3. Sample analysis

Inductively Coupled Plasma-Mass Spectrometry (ICP-MS iCAP Q / iCAP RQ ICP-MS, Thermo Fisher Scientific Inc., Bremen, Germany) operating with argon gas of spectral purity (99.9995%) was used for the determination and quantification of toxic metals in rice. Operating conditions and acquisition parameters are illustrated in Table 2. Before each experiment, the instrument was tuned using iCAP Q/RQ TUNE aqueous multi-element standard solution in 2% HNO_3 + 0.5% HCl solution (Thermo scientific, Bremen, Germany). The torch position, ion lenses, gas output, resolution axis (10% of peak height) and background (<20 shots) were optimized with the tuning solution (1 $mg.L^{-1}$) The correlation coefficients for all the calibration curves were at least 0.9999, reflecting good linear relationship throughout the ranges of concentrations under study. All measurements were conducted using the full quantitative mode analysis while measuring several isotopes of the elements and checking the isotopic ratio in the digested samples to confirm the absence of polyatomic interferences. The limit of detection (LOD) for each element was calculated by blank determination as 3 times standard deviation of 20 blank replicates. Similarly, the limit of quantification (LOQ) was calculated as 2 times LOD for each element. The precision of the heavy metal analysis by ICP-MS was verified by applying the spiking methodology (Jorhem & Engman, 2000) at different concentrations and calculating the recovery percentages for each metal and calculating the recovery percentages (Shim et al., 2018). The Spiking technique involves adding known amounts of a standard to the sample solution. This approach compensates for a sample constituent that enhances or depresses the analyte signal, thus producing a different slope from that of the calibration standards. Spiking solutions were prepared at two metal concentrations (0.5 and 5 ppm) with 0.5 g of rice sample.

Table 2
ICP-MS operating conditions and acquisition parameters.

Operating Conditions	
Spectrometer	Thermo Scientific, ICAP RQ. ASX-280, autosampler ICP-MS
Nebulizer	Borosilicate glass concentric with 0.4 mL/min
Spray chamber	2.70ºC. Quartz cyclonic
Cell geometry	Octopole
Sampling cone	Nickel, 1.1 mm diameter orifice
Skimmer cone	Nickel, 0.75 mm diameter orifice
RF power	400–1600 W
Reflected power	< 10 W
Standard Mode	
Plasma gas flow	15 L/min
Nebulizer gas flow	1.03 L/min
Auxiliary gas flow	0.81 L/min
Expansion stage	2.01mbar
Intermediate stage	10^{-4}mbar
Analyzer stage	10^{-6}mbar
He mode (collision cell mode)	
He gas flow	4.0 mL/min
Octopole bias (CCT bias)	-21 V
Quadrupole bias (pole bias)	-18 V
Acquisition Parameters	
Field	Virtual hyperbolic
Frequency	2 MHz
Mass range	2–290 a.m.u.
Dwell time	0.04 s
Number of sweeps	5
Number of replicates	3
Total Acquisition time	220 s

2.4. Determination of exposure to heavy metals from rice consumption

The average consumption of rice (22.9 g/d of dry rice) and average body weight (70 kg) in Lebanon was acquired from a study published by Hassan et al. (2022a). For UAE, average consumption of rice (315 g/day/person) was calculated based on the total consumption of rice in the country (1.15 MMT (million metric tons), population (9.991 millions), and the average weight of adults (76 kg) in 2021 (United States Department of Agriculture, Foreign Agricultural Service, 2021; United Nations, 2021; World Obesity Observatory, 2022).

To estimate the exposure levels to toxic metals represented as estimated daily intake (EDI) (mg/kg body weight/day), the average concentration of each toxic metal (mg/kg dry weight) was multiplied by the average daily amount of rice consumption (kg/day) divided by the average body weight of the adult population (Chu et al., 2021; FAO/WHO, 2014). To estimate weekly intake (EWI), EDI was multiplied by 7 (Chu et al., 2021).

On the other hand, Hazard Quotient (HQ) was calculated by dividing calculated exposure (estimated as daily, weekly or monthly) with the reference exposure. Reference exposure is PTWI (provisional tolerable weekly intake), BMDL (benchmark dose lower confidence limit) or PTMI (provisional tolerable monthly intake) for each metal according to international guidelines. HQ > 1 indicates an increased risk (Chu et al., 2021).

2.5. Statistical analysis

Concentration values for the five different metals (As, Cd, Cr, Hg, and Pb) were entered into SPSS V27. Rice related data (packing season, country of packing, grain size, rice type, food safety management system certificate, and time between packing and purchasing) were also coded and entered into the software. The metals' concentrations distributions were tested for normal distribution and when departure was detected, analyses accounted for non-normality using bootstrapping estimates of the standard errors. Differences in mean metal concentrations between the different rice variables were tested using ANOVA F test while correcting for non-homogeneity of variance was done using the Welch F test. Differences in collections dates were tested using the paired t-test. All analyses were carried at the 0.05 significance level.

3. Results and discussion

3.1. Concentrations of heavy metals in rice samples

To our knowledge, this is the first study in Lebanon and UAE to assess arsenic, cadmium, chromium, lead, and mercury levels in rice and the estimated exposure of the Lebanese and Emirati populations to these toxic metals. Only one study was conducted in Lebanon by Nasreddine et al. (2006), assessing the contamination of foods and drinks with lead and cadmium and their associated exposure. It is important to note that since Lebanon and the UAE depend exclusively on imports to provide their populations' rice needs, the results are not representative of any local rice cultivation. All metals in this study were analyzed in their total forms. The total number of samples analyzed was 236 samples (107 from Lebanon and 129 from UAE). Limits of detection and quantification of each metal in the ICP-MS are stated in Table 3. Table 4 shows the mean, minimum, and maximum values of heavy metals in the studied rice samples from collections 1 and 2 in Lebanon and UAE. Table 5 shows the numbers and percentages of rice brands contaminated with heavy metals exceeding international limits in both countries. Figs. 1 and 2 report the mean concentrations of toxic metals in the rice samples from Lebanon and UAE, respectively. Remarkably, our study revealed significant differences between the values of toxic metals' contamination within the same brands between both collections, taking place approximately three months apart, in both countries for most of the toxic metals. This signifies that the same brands might have been more

Table 3
Limits of detection and quantification of each metal in ICP-MS.

As		Cd		Cr		Hg		Pb	
LOD	LOQ	LOD	LOQ	LOD	LOQ	LOD	LOQ	LOD	LOQ
0.00005	0.0001	0.00003	0.00005	0.00003	0.0001	0.00004	0.00008	0.00004	0.00008

LOD: limit of detection, LOQ: limit of quantification

Table 4
Mean, standard deviation, minimum and maximum values of heavy metals (mg/kg) in dry rice collected from Lebanon and UAE.

		Lebanon	UAE
As	Mean	0.24	0.18
	Standard Deviation	0.11	0.09
	Minimum	0.11	0.06
	Maximum	0.51	0.62
Cd	Mean	0.29	0.07
	Standard Deviation	0.13	0.04
	Minimum	0.12	0.00
	Maximum	0.50	0.23
Cr	Mean	0.34	0.23
	Standard Deviation	0.13	0.11
	Minimum	0.11	0.08
	Maximum	0.67	0.55
Hg	Mean	0.15	0.17
	Standard Deviation	0.05	0.05
	Minimum	0.03	0.04
	Maximum	0.29	0.25
Pb	Mean	0.27	0.24
	Standard Deviation	0.12	0.09
	Minimum	0.01	0.06
	Maximum	0.57	0.52

Table 5
Numbers and percentages of rice samples from Lebanon and UAE that are contaminated with toxic metals exceeding international limits.

	Lebanon		UAE	
	n (%) of brands > CAC limits	n (%) of brands > EC limits	n (%) of brands > CAC limits	n (%) of brands > EC limits
As	26 (25%)	NA	12 (9%)	NA
Cd	26 (25%)	78 (73%)	0	1 (1%)
Cr	NA	NA	NA	NA
Hg	NA	107 (100%)	NA	129 (100%)
Pb	74 (69%)	74 (69%)	82 (64%)	82 (64%)

n: number, NA: Not applicable.
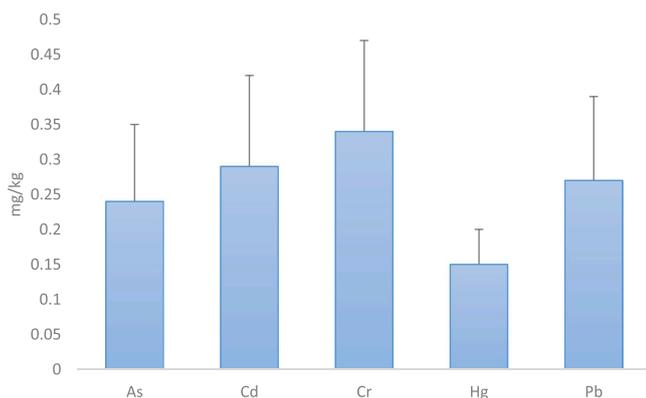
Fig. 1. Mean concentrations of toxic metals in the rice samples from Lebanon.

contaminated among different lot numbers, indicating inconsistency. In general, the packing season and having FSMS did not significantly affect the levels of most of the metals in both countries, indicating that
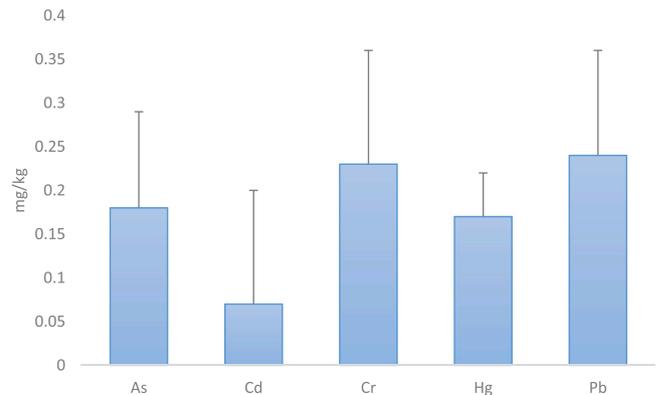
Fig. 2. Mean concentrations of toxic metals in the rice samples from UAE.

pollution and contamination might have affected the samples regardless of season or attainment of a food safety certification.

3.1.1. Arsenic

Among rice samples from Lebanon, the mean of As was 0.24 ± 0.11 mg/kg dry weight, which was less than the MRL of 0.3 mg/kg set by Codex Alimentarius (CAC, 2012). Out of the 107 samples, 26 (25%) were above CAC limits. On the other hand, for the UAE samples, the mean was 0.18 ± 0.09 mg/kg dry weight, which is below CAC limits. Out of the 129 samples, 12 (9%) exceeded CAC limits. As has widely attracted the attention of investigators of toxic metals in rice worldwide due to its widespread contamination in groundwater (Shraim, 2017). Our study has shown that the mean level of As in rice products available in the Lebanese market was 0.24 ± 0.08 mg/kg dry weight. Overall, this level was less than the MRL of 0.3 mg/kg set by Codex, but 25% of the samples exceeded this limit (CAC, 2012). In the samples from the UAE market, the mean was 0.18 ± 0.09 mg/kg dry rice, thus below the CAC limit, with 9% of the samples exceeding it. Both results were higher than most levels reported in the literature. Two other studies, one conducted in KSA and the other in Malaysia, reported mean values of As in rice of 0.03 and 0.087 mg/kg of As in rice, respectively (Mohamed et al., 2017; Praveena & Omar; 2017). A study conducted in Bangladesh found that the mean value of As in irrigated and rain-fed rice was 0.144 ± 0.091 mg/kg, with irrigated rice containing higher levels (0.153 ± 0.112 mg/kg) (Jahiruddin et al., 2017). Similarly, Bielecka et al. (2020) reported that the average As level in rice and rice products collected in Poland was 0.123 ± 0.077 mg/kg. In Vietnam, Chu et al. (2021) found that the mean contamination of Vietnamese rice with As was 0.115 ± 0.049 mg/kg. Another study conducted in Romania found that the mean As contamination in rice was 0.042 mg/kg (Voica et al., 2012). Rabbani et al. (2015) reported that As was not detected in any imported or local rice sample in Iran. However, some other studies presented higher As levels than our results. Thai rice cultivated in the Si Banphot district in Thailand was more contaminated with As (0.192 ± 0.073 mg/kg) than our samples from the UAE (Srinuttrakul et al., 2018). Likewise, in Italy, most rice types assessed by Sommella et al. (2013) were found to be contaminated with As levels (Rice types: Arborio: 0.23 ± 0.01, Carnaroli: 0.23 ± 0.02, Ribe/Roma parboiled: 0.20 ± 0.01, Roma: 0.19 ± 0.10, and Vialone Nano: 0.28 ± 0.03 mg/kg). These levels were higher than those reported in our study for the UAE

samples but less than those reported for the Lebanon samples. A study conducted in Cambodia detected high levels of As contamination in rice in Preak Russey (315 ± 150 μg/kg) and Kendal provinces (158 ± 33 μg/kg), both being higher than our results from both countries (Murphy et al., 2018).

Brown rice samples in our study were significantly more contaminated with As than white and parboiled rice in the Lebanese samples ($p = 0.02$). The measured values were 0.27 ± 0.05, 0.22 ± 0.08, and 0.20 ± 0.13 mg/kg for brown, parboiled and white rice, respectively. These results are comparable to the results from a study conducted in the USA reporting that USA brown rice (0.243 mg/kg) contained more As than white rice from USA, Thailand, India, and Italy ($p < 0.05$) (TatahMentan et al., 2020). In South Korea, Choi et al. (2014) also found that brown rice contained 101 ± 34.6 μg/kg, which was more than that of white (91.7 ± 28.1 μg/kg). Zeng et al. (2015) reported that As content of Chinese brown rice was 0.34 ± 0.25 mg/kg, which was higher than our results for samples collected in Lebanon. Another Chinese study also revealed a high As contamination level of 0.52 ± 0.03 mg/kg in brown rice in Hunan Province (Fan et al., 2017). This is due to germ layer of brown rice that retains higher amounts of toxic metals (TatahMentan et al., 2020).

Long rice grains contained significantly higher amounts of As, when compared to short/medium grains ($p = 0.002$). The reason behind this finding could be that long rice grains have a larger surface area, resulting in the accumulation of more heavy metals from the agricultural stage. Moreover, rice brands originating from developed countries were more contaminated with As when compared to ones imported from developing countries in the Lebanese samples ($p < 0.001$). Similar results were expressed by TatahMentan et al. (2020), as US white rice was more contaminated with As than white rice from Thailand and India. Shraim (2017) found comparable results when studying rice imported into KSA. The author found that the rice imported from USA contained higher amounts of As (0.257 ± 0.142 mg/kg) when compared to rice imported from Thailand (0.200 ± 0.025 mg/kg), Pakistan (0.147 ± 0.055 mg/kg), India (0.103 ± 0.048 mg/kg), and Egypt (0.097 ± 0.044 mg/kg). This could be possibly attributed to higher levels of industrial pollution in developed countries, specifically in USA, as evidenced by the findings of the aforementioned studies. However, in our UAE samples, rice brands originating from developing countries were more contaminated with As than developed countries. The reason could be the lack of adequate regulations controlling pesticides use in developing countries, especially that pesticide use is considered as a contributor to food security to many of these countries (Sarkar et al., 2021).

3.1.2. Cadmium

Mean of Cd was 0.29 ± 0.13 and 0.07 ± 0.04 mg/kg in Lebanon and UAE, respectively. Both were lower than CAC and LIBNOR limit of 0.4 mg/kg each, but EC limit of 0.2 mg/kg was exceeded by the samples from Lebanon (CAC, 2019; EC, 2006; LIBNOR, 2013). Of the samples from Lebanon, 26 (25%) and 78 (73%) were above CAC and EC limits, respectively. Among samples from UAE, all samples were complaint with CAC limits, with only one (1%) exceeding EC limits. Our study has revealed that the mean levels of Cd were 0.29 ± 0.13 and 0.07 ± 0.04 mg/kg in rice collected from Lebanon and the UAE, respectively. Both were lower than CAC and LIBNOR limit of 0.4 mg/kg, but the Lebanese samples exceeded the EC limit of 0.2 mg/kg (CAC, 2019; EC, 2006; LIBNOR, 2013). In Lebanon, 25% of the samples were contaminated with levels exceeding CAC limits, with 73% being above the EC limit. In contrast, all UAE samples were compliant with CAC limits, with only one (1%) exceeding EC limits for Cd in rice. In the UAE, this meager percentage of samples exceeding EC limit might be attributed to the high governmental control over imported food products.

In the study previously conducted in Lebanon by Nasreddine et al. (2006), the reported level of Cd of (0.0006 mg/kg in rice and rice products was less than that presented in our current study for Lebanon

(0.29 ± 0.13 mg/kg). This increase could be attributed to the global rise in the population growth and the demand for rice consumption, thus the utilization of more agrochemicals and mechanical cultivation leading to more toxic metals contamination of crops (Zakaria et al., 2021). Another reason might be that since 2019, Lebanon has been suffering from a major economic crisis, leading to importing several new affordable brands, with question marks surrounding their safety, especially in the absence of adequate governmental control.

Rice samples from Lebanon contained a high amount of Cd but was remarkably less than the level reported by a study conducted in Malaysia for local and imported rice (0.72 mg/kg) (Abd Rashid et al., 2019). However, in general, Cd levels in samples from Lebanon were higher than the ones reported in most studies assessing Cd content in rice. Horiguchi et al. (2020) stated that the mean Cd content of rice collected from Japan were 0.158 and 0.109 mg/kg, respectively. Moreover, Chu et al. (2021) stated that Vietnamese rice samples contained a mean of 0.111 ± 0.105 mg/kg. Mohamed et al. (2017) also presented lower Cd content in rice imported into KSA (0.03 mg/kg). Voica et al. (2012) reported an even lower mean Cd content of 0.003 mg/kg in rice samples collected in Romania. This indicates that Lebanon might be importing rice brands from unreliable suppliers, especially with the lack of adequate governmental control.

Cd levels in our UAE samples were close to levels detected by Rabbani et al. (2015) for Iranian rice (0.06 ± 0.05 mg/kg) but higher than the ones reported by Lien et al. (2021) for Taiwanese rice and Bielecka et al. (2020) for rice and rice products in Poland. Lien et al. (2021) indicated that, on average, Taiwanese rice samples contained 0.04 ± 0.04 mg/kg, while Bielecka et al. (2020) reported a mean Cd content of 25.7 ± 26.5 μg/kg (0.0257 ± 0.0265 mg/kg).

Among the Lebanese samples, long rice grains had higher Cd content of 0.31 ± 0.11 mg/kg than short/medium grains ($p < 0.001$). One of Lebanon's mostly consumed long grain rice types is Basmati rice, which is most commonly imported from India. This could be linked to the findings of a study testing heavy metals in Indian rice in India, reporting one of the highest Cd levels in the literature. The authors found that the mean contamination was 0.99 ± 0.05 mg/kg, even though samples were washed with distilled water before testing (Sharma et al., 2018). The results were also comparable to the findings of Naseri et al. (2015), which assessed domestic and imported rice in Iran and found that several long-grain rice brands imported from Thailand and India were more contaminated with Cd than some other locally cultivated short-grain brands (Naseri et al., 2015).

There was a borderline significance for the effect of grain type on Cd contamination among the samples from Lebanon ($p = 0.054$), with brown rice containing the highest amount (0.32 ± 0.10 mg/kg). Zeng et al. (2015) reported a similar high mean Cd content of 0.312 ± 0.434 mg/kg in brown rice in Hunan province in China. The authors associated this high level of contamination with the fact that Hunan province has a long history of metal mining and smelting. Still, we could also argue that the outer grain layer on brown rice retains higher amounts of toxic metals, thus interpreting this result even more.

Those effects were not significant among UAE samples, indicating that UAE typically imports from relatively more reputable suppliers, regardless of the type of rice.

3.1.3. Chromium

Among the samples from Lebanon, mean of Cr was 0.34 ± 0.13 vs. 0.23 ± 0.11 mg/kg in the samples from UAE. No maximum allowed limits for Cr content in rice have been identified at the international level (Zulkafflee et al., 2022). The mean concentration of Cr was equal to 0.34 ± 0.13 mg/kg in the Lebanese samples, whereas in the UAE samples, it was 0.23 ± 0.11 mg/kg. No maximum limits have been set for Cr content in rice at the international level (Zulkafflee et al., 2022). Nevertheless, Cr values in our results from UAE samples were the same as the reported mean value of 0.23 mg/kg in KSA (Mohamed et al., 2017). Similar results were also observed by Sommella et al. (2013),

assessing Cr levels in several Italian rice types. The Lebanese results for Cr were relatively higher and were close to those reported by Chu et al. (2021) in Vietnam (0.30 mg/kg). However, our results for both countries were lower than the ones published by Sharma et al. (2018) reporting 19.98 ± 2.10 mg/kg, Mahvi et al. (2016) reporting 0.631 ± 0.43 mg/kg, Jahiruddin et al. (2017) reporting 1.06 ± 0.84 mg/kg, and Praveena & Omar (2017) reporting 2.7 mg/kg.

For the samples collected from Lebanon, just like with Cd and As, Cr contamination was significantly higher in samples originating from developed countries (p = 0.006), providing more evidence that developed countries contribute to more toxic metal contamination in rice than developing countries, owing it to the abundance of industrial practices and pollution. Among the Lebanese samples, brown and long rice types were more contaminated than their counterparts (p < 0.001). The findings of Naseri et al. (2015) further demonstrated that some imported long-grain rice brands contained higher Cr levels when compared to local small-grain brands in Iran, with the highest amount being reported in long-grain rice from India (0.55 ± 0.05 mg/kg). Furthermore, as mentioned earlier, long grain Basmati rice is the primary type of Indian rice imported into Lebanon, and Sharma et al. (2018) reported a high Cr contamination level of 19.98 ± 2.10 mg/kg in Indian rice. The highest level of contamination was also identified in samples where the time between packing and purchasing was equivalent to 30 weeks and above (p < 0.001), indicating that more extended periods could have led to more Cr contamination, possibly by cross-contamination, poor barrier properties of packaging, and poor storage conditions.

Cr contamination in rice brands collected from the UAE was not affected by any of the studied variables. This can again be attributed to the fact that the UAE typically imports rice under strict supervision.

3.1.4. Mercury

Mean of Hg in the samples from Lebanon was 0.15 ± 0.05 mg/kg. This level exceeded the limit of 0.01 mg/kg set by the EC (EC, 2018). All the samples were contaminated with Hg levels exceeding the EC limit. CAC has not set a limit for Hg in rice.

In samples from UAE, mean Hg content was 0.17 ± 0.05 mg/kg, exceeding the EC limit. All of the samples were contaminated with Hg levels exceeding this limit. The averages of total Hg were 0.15 ± 0.05 mg/kg and 0.17 ± 0.05 mg/kg in the Lebanese and UAE samples, respectively. All samples from both countries contained Hg amounts exceeding the 0.01 mg/kg limit set by the EC (EC, 2018). CAC has not established an MRL for Hg in rice. Both Hg levels were remarkably higher than levels reported by Rothenberg et al. (2015) in Madagascar (0.0011 mg/kg), Bielecka et al. (2020) in Poland (0.0028 ± 0.0026 mg/kg), and Chu et al. (2021) in Vietnam (0.007 ± 0.003 mg/kg).

Hg levels in UAE samples were not affected by any of the studied variables. In contrast, significantly higher levels were detected in the Lebanese samples for long grain and brown rice (p = 0.019 and p = 0.012, respectively). The case in Lebanon was similar to findings of Batista et al. (2012) assessing heavy metals in Brazilian rice. Batista et al. (2012) reported 3.9 ± 3.3 ng/g in parboiled brown vs. 3.5 ± 4.3 and 2.3 ± 2.3 ng/g in parboiled white and white rice, respectively. This was also evidenced by the difference in the findings of two Chinese studies. Huang et al. (2013) found that polished rice contained 0.0056 ± 0.003 mg/kg of Hg, which was less than 0.069 ± 0.060 mg/kg reported by Zeng et al. (2015) for brown rice. Nevertheless, the levels of Hg in brown rice in the Lebanese and UAE samples of 0.17 ± 0.02 and 0.07 ± 0.39 mg/kg, respectively, were higher than most values presented in the previously mentioned studies. These findings should be eye opening to both governments opting to avoid importing rice contaminated with this highly toxic metal.

3.1.5. Lead

Mean of Pb in the samples from Lebanon was 0.27 ± 0.10 mg/kg.

This level is above the 0.2 mg/kg limit set by CAC, EC, and the national Lebanese standards LIBNOR (CAC, 2019; EC, 2006; LIBNOR, 2013). Out of the samples, 74 (69%) contained levels higher than this limit.

The mean of Pb in the samples from UAE was 0.24 ± 0.09 mg/kg, exceeding the CAC and EC limits. Eighty-two (64%) samples contained Pb amounts higher than the permitted limit.

In general, among the Lebanese samples, 11 (10%) samples exceeded CAC limits for As, Cd, and Pb at the same time, while 53 (50%) samples exceeded EC limits for Cd, Hg, and Pb. Among the UAE samples, none of the samples exceeded CAC limits for As, Cd and Pb at the same time, with one sample (1%) exceeding EC limits for Cd, Hg, and Pb. Our results revealed that the Pb level in Lebanese rice was 0.27 ± 0.10 mg/kg vs. 0.24 ± 0.09 in UAE samples. Both levels exceeded the limit of 0.2 mg/kg set by CAC, EC, and the Lebanese national standards LIBNOR (CAC, 2019; EC, 2006; LIBNOR, 2013). The majority (69% and 64% of the Lebanese and UAE samples, respectively) exceeded this limit. In the study conducted in Lebanon in 2006 by Nasreddine et al. (2006), Pb level was 4.1 µg/kg, which was remarkably lower than the tested level of our current rice samples from Lebanon. Furthermore, unlike the case of As, Cd, Cr and Hg in our study, rice brands imported from developed countries did not contain higher Pb amounts than brands from developing countries. This could be attributed to the great efforts exerted by some developed countries, such as the United States, to reduce lead exposure, whereas, on the contrary, health problems caused by environmental lead pollution continue to impose a serious public health issue in many developing countries, including China (Lin et al. 2011). Therefore, the increase in Pb levels in imported rice in Lebanon since 2006 could be related to the issue of the ongoing rise in lead pollution in developing countries.

Our result for Pb contamination was higher than the level of 0.039 mg/kg reported by Voica et al. (2012) studying heavy metals in rice in Romania, and higher than the level of 0.04 mg/kg reported by Mohamed et al. (2017) assessing rice imported into KSA. However, Wang et al. (2019) reported a similar level of 0.0341 ± 0.0430 mg/kg in Chinese rice and rice products. Similarly, in Poland, Bielecka et al. (2020) reported 37.5 ± 29.3 µg/kg Pb content in imported rice. Chu et al. (2021) stated that Pb content in Sri Lankan rice was 0.075 ± 0.050 mg/kg, which was also lower than the results we contracted. On the other hand, Mahvi et al. (2016) reported a higher Pb level of 0.320 ± 0.230 mg/kg in Indian rice. Another study conducted in Iran reported higher Pb levels in Iranian (0.6416 ± 0.3055 mg/kg) and non-Iranian (0.8088 ± 1.0796 mg/kg) rice (Rabbani et al., 2015). In Malaysia, Abd Rashid et al. (2019) presented an even higher Pb content of 2.19 mg/kg.

No statistical significance was apparent for the effect of any of the variables on Pb content of samples collected from Lebanon and UAE.

3.2. Effect of different variables on heavy metals' levels in rice

The effect of different variables on the heavy metals' contamination of rice in Lebanon and UAE are presented in Tables 6 and 7, respectively.

3.2.1. Arsenic

Among brands collected in Lebanon, there was no statistical significance for the effect of the packing season (p = 0.341), the presence of a food safety management system certificate (FSMS) (p = 0.098), and the time between packing and purchasing (p = 0.362) on As content. However, the results were statistically significant for the country of origin (p < 0.001), with higher levels (0.27 ± 0.12 mg/kg) being from brands originating from developed countries (Italy, France, USA, and Spain) vs. 0.19 ± 0.05 mg/kg for brands originating from developing countries (India, Pakistan, Thailand, and China). There was a significant difference between As levels in white, parboiled, and brown rice (p = 0.02), with the highest content detected in brown rice (0.27 ± 0.05 mg/kg), and between long rice and short/medium rice grains (p = 0.002), with the highest content being in long grains (0.21

Table 6
Effect of different independent variables on heavy metals levels in rice samples collected from Lebanon.

Independent variable	N	As			Cd		Cr		Hg		Pb	
		Mean ± SD[a]	p-value		Mean ± SD	p-value	Mean ± SD	p-value	Mean ± SD	p-value	Mean ± SD	p-value
Packing season												
Fall/winter	62	0.25 ± 0.12			0.27 ± 0.11		0.35 ± 0.13		0.15 ± 0.06		0.28 ± 0.13	
Spring/Summer	23	0.22 ± 0.10			0.33 ± 0.11		0.33 ± 0.13		0.15 ± 0.05		0.28 ± 0.11	
Not known	21	0.22 ± 0.08	0.341		0.29 ± 0.11	0.147	0.34 ± 0.14	0.796	0.16 ± 0.05	0.637	0.24 ± 0.11	0.445
Country of origin												
Developed	75	0.27 ± 0.12			0.30 ± 0.11		0.36 ± 0.14		0.15 ± 0.06		0.27 ± 0.12	
Developing	23	0.19 ± 0.05			0.24 ± 0.09		0.31 ± 0.1		0.14 ± 0.05		0.28 ± 0.12	
Not available	9	0.21 ± 0.07	< 0.001		0.31 ± 0.1	0.055	0.23 ± 0.08	0.006	0.14 ± 0.05	0.437	0.29 ± 0.1	0.906
FSMS[b]												
Yes	35	0.22 ± 0.11			0.30 ± 0.11		0.37 ± 0.14		0.16 ± 0.05		0.28 ± 0.12	
No/Not known	72	0.26 ± 0.11	0.098		0.28 ± 0.11	0.536	0.32 ± 0.13	0.078	0.14 ± 0.06	0.113	0.27 ± 0.12	0.785
Grain size												
Long	71	0.26 ± 0.12			0.31 ± 0.11		0.37 ± 0.14		0.16 ± 0.06		0.28 ± 0.12	
Short/medium	36	0.21 ± 0.06	0.002		0.23 ± 0.08	< 0.001	0.27 ± 0.09	< 0.001	0.13 ± 0.04	0.012	0.26 ± 0.13	0.403
Type												
White	41	0.22 ± 0.08			0.25 ± 0.1		0.29 ± 0.11		0.14 ± 0.04		0.26 ± 0.12	
Parboiled	55	0.20 ± 0.13			0.30 ± 0.11		0.28 ± 0.12		0.16 ± 0.07		0.26 ± 0.12	
Brown	11	0.27 ± 0.05	0.02		0.32 ± 0.1	0.054	0.39 ± 0.15	< 0.001	0.17 ± 0.02	0.019	0.29 ± 0.09	0.436
Time between packing and purchasing												
1–9 weeks	22	0.27 ± 0.12			0.28 ± 0.09		0.25 ± 0.06		0.16 ± 0.06		0.25 ± 0.1	
10–19 weeks	18	0.21 ± 0.1			0.26 ± 0.12		0.34 ± 0.11		0.13 ± 0.05		0.29 ± 0.14	
20–29 weeks	16	0.27 ± 0.12			0.28 ± 0.11		0.33 ± 0.14		0.14 ± 0.05		0.29 ± 0.12	
30 weeks and above	29	0.26 ± 0.12	0.362		0.32 ± 0.11	0.314	0.41 ± 0.13	< 0.001	0.16 ± 0.06	0.166	0.28 ± 0.13	0.647
Lot number for common brands between both collections												
Collection 1	47	0.30 ± 0.12			0.33 ± 0.1		0.35 ± 0.15		0.16 ± 0.06		0.27 ± 0.12	
Collection 2	47	0.19 ± 0.06	< 0.001		0.22 ± 0.09	< 0.001	0.36 ± 0.1	0.67	0.14 ± 0.05	0.001	0.28 ± 0.13	0.775

[a] Standard deviation.
[b] Food safety management system certificate.

± 0.12 mg/kg). Results further revealed a significant statistical difference between As levels in brands collected twice ($p < 0.001$).

Among samples from the UAE, there was a statistical significance for the effect of the packing season ($p = 0.011$). A statistical significance was also found for the effect of country of origin ($p = 0.016$), with brands originating from developing countries (India, Pakistan, Sri Lanka, Philippines, Vietnam, Egypt, and Thailand) having significantly higher levels of As (0.24 ± 0.06 mg/kg) than those from developed countries (USA, Italy, UK, and Belgium) (0.18 ± 0.091 mg/kg). Grain size, type, presence of FSMS, and time between packing and purchasing had no statistically significant effects on As content in rice brands collected from the UAE. Levels of As were significantly different among samples collected twice ($p < 0.001$).

3.2.2. Cadmium

No statistically significant differences were identified for packing season ($p = 0.147$), presence of FSMS ($p = 0.536$), and time between packing and purchasing ($p = 0.314$) on Cd content of rice collected from

Lebanon. There was a borderline significant difference for the country of origin ($p = 0.055$) and type of rice ($p = 0.054$) variables. Long rice grains (0.19 ± 0.09 mg/kg) had a significantly higher Cd content than short/medium grains (0.21 ± 0.06 mg/kg) ($p < 0.001$). Another statistically significant difference was between Cd levels from samples collected twice ($p < 0.001$).

Regarding the samples from UAE, there was no statistical significance for the effects of packing season ($p = 0.627$), country of origin ($p = 0.356$), presence of FSMS ($p = 0.073$), time between packing and purchasing ($p = 0.659$), grain size ($p = 0.485$), and rice type ($p = 0.439$). Levels of Cd were statistically different between the samples collected twice ($p = 0.008$).

3.2.3. Chromium

Among the samples from Lebanon, there was no significant effect for the packing season ($p = 0.796$), presence of FSMS ($p = 0.078$), and among samples collected twice ($p = 0.67$). Contamination was significantly higher in samples from developed countries (0.36 ± 0.14 mg/kg) than those from developing countries (0.31 ± 0.10 mg/kg) ($p = 0.006$).

Table 7

Effect of different independent variables on heavy metals levels in rice samples collected from UAE.

Independent variable	N	As Mean±Sd[a]	As p-value	Cd Mean±Sd	Cd p-value	Cr Mean±Sd	Cr p-value	Hg Mean±Sd	Hg p-value	Pb Mean±Sd	Pb p-value
Packing season											
Fall/winter	75	0.20 ± 0.1		0.07 ± 0.05		0.24 ± 0.1		0.17 ± 0.05		0.25 ± 0.1	
Spring/Summer	51	0.16 ± 0.07	0.011	0.07 ± 0.04	0.627	0.23 ± 0.12	0.717	0.16 ± 0.06	0.114	0.23 ± 0.08	0.458
Country of origin											
Developed	113	0.18 ± 0.092		0.07 ± 0.04		0.23 ± 0.11		0.17 ± 0.06		0.24 ± 0.09	
Developing	14	0.24 ± 0.06	0.016	0.08 ± 0.06	0.356	0.23 ± 0.08	0.993	0.16 ± 0.06	0.786	0.23 ± 0.08	0.534
FSMS[b]											
Yes	60	0.18 ± 0.08		0.06 ± 0.04		0.24 ± 0.11		0.17 ± 0.06		0.24 ± 0.09	
No/Not known	67	0.19 ± 0.1	0.382	0.07 ± 0.05	0.073	0.23 ± 0.11	0.469	0.17 ± 0.06	0.801	0.24 ± 0.1	0.661
Grain size											
Long	76	0.19 ± 0.09		0.07 ± 0.04		0.23 ± 0.1		0.16 ± 0.06		0.24 ± 0.1	
Short/medium	51	0.18 ± 0.1	0.857	0.07 ± 0.05	0.485	0.24 ± 0.11	0.78	0.18 ± 0.06	0.251	0.25 ± 0.08	0.428
Type											
White/ Parboiled	98	0.19 ± 0.08		0.07 ± 0.04		0.24 ± 0.11		0.17 ± 0.05		0.24 ± 0.09	
Brown	29	0.18 ± 0.12	0.899	0.07 ± 0.05	0.439	0.23 ± 0.11	0.817	0.16 ± 0.07	0.392	0.23 ± 0.1	0.68
Time between packing and purchasing											
1–9 weeks	9	0.14 ± 0.1		0.08 ± 0.06		0.28 ± 0.13		0.16 ± 0.05		0.30 ± 0.09	
10–19 weeks	48	0.19 ± 0.11		0.06 ± 0.03		0.23 ± 0.1		0.17 ± 0.06		0.23 ± 0.1	
20–29 weeks	29	0.19 ± 0.07		0.07 ± 0.04		0.20 ± 0.07		0.18 ± 0.06		0.24 ± 0.08	
30 weeks and above	40	0.18 ± 0.08	0.568	0.07 ± 0.05	0.659	0.26 ± 0.12	0.072	0.16 ± 0.05	0.182	0.24 ± 0.09	0.266
Lot number for common brands between both collections											
Collection 1	39	0.21 ± 0.08		0.05 ± 0.05		0.32 ± 0.09		0.15 ± 0.05		0.27 ± 0.1	
Collection 2	39	0.18 ± 0.08	0.008	0.09 ± 0.04	0.001	0.15 ± 0.03	< 0.001	0.18 ± 0.06	< 0.001	0.20 ± 0.08	0.001

[a] Standard deviation.
[b] Food safety management system certificate.

Brown rice and long grain had significantly higher Cr levels than their counterparts (0.39 ± 0.15 and 0.37 ± 0.14 mg/kg of Cr, respectively) ($p < 0.001$). The highest level of contamination was also identified in samples where the time between packing and purchasing was equivalent to 30 weeks and above ($p < 0.001$).

In the samples from UAE, the only statistical significance on Cr was found among brands collected twice ($p < 0.001$).

3.2.4. Mercury

Among samples from Lebanon, long grain and brown rice samples appeared to be more significantly contaminated with Hg ($p = 0.012$ and $p = 0.019$, respectively). A statistically significant difference was identified among samples collected twice ($p < 0.001$). No statistical significance was found for the effects of packing season, country of origin, presence of FSMS, and time between packing and purchasing.

The only statistical significance on Hg contamination of rice samples collected from UAE was among samples collected twice ($p < 0.001$).

3.2.5. Lead

In Lebanon, no significant effect on Pb contamination was found for the different independent variables. As for UAE, there was a statistically significant difference among brands collected twice ($p = 0.001$, respectively).

3.3. Exposure to heavy metals from rice consumption

According to our calculations, UAE rice consumption in 2021/2022 was 315 g/capita/day. On the other hand, according to Hassan et al. (2022a), the average rice consumption in Lebanon in 2021 was estimated to be 22.9 g/capita/day. Table 8 shows the exposure levels of the Lebanese and UAE populations to heavy metals from rice in comparison to international safe levels of exposure. Table 9 shows the recovery of the spiked samples and the certified reference material to assess any potential loss of analyte during the digestion process.

3.3.1. Arsenic

Based on the equations explained in the methodology section, the calculated EDI of As from rice consumption in Lebanon is 0.08 µg/kg bw/day, with HQ = 0.01–0.3 (calculated according to EFSA BMDL). While in UAE, EDI was identified as 0.76 µg/kg bw/day, with HQ = 0.1–2.5. According to JECFA (2011a, 2011b, 2011c), the previously set PTWI of 2.1 µg/kg bw/day was withdrawn since it was observed that it was in the same range of BMDL0.5 (benchmark dose lower confidence limit of 2–7 µg/kg bw/day from inorganic As) for a 0.5% increased risk for lung cancer. According to EFSA CONTAM Panel (2009), the BMDL01 for a 1% increased risk for cancers of the lung, skin and bladder, and skin lesions from As is 0.3–8 µg/kg bw/day. EDI in Lebanon was 0.08 µg/kg bw/day, with a hazard quotient of 0.01–0.27 (according to EFSA

Table 8

Exposure levels of the Lebanese and UAE populations to heavy metals from rice in comparison to international safe levels of exposure.

	Lebanon Exposure Levels (µg/kg bw) (Hazard Quotient)	UAE Exposure Levels (µg/kg bw) (Hazard Quotient)	Safe Levels of Exposure According to FAO/WHO (µg/kg bw)	Safe Levels of Exposure According to European Food Safety Authority (µg/kg bw)
As	0.08 per day; 0.56 per week (0.01–0.27)	0.76 per day; 5.32 per week (0.095–2.5)	2.1 per week was withdrawn	$BMDL_{01}^a$: 0.3–8 per day
Cd	2.7 per month (0.108–0.252)	4.5 per month (0.18–0.42)	25 per month	2.5 per week
Cr	7.79 µg/day (0.03)	72.5 µg/day (0.29)	250 µg/day	250 µg/day
Hg	0.35 per week (0.09)	4.96 per week (1.23)	4 per week	4 per week
Pb	0.088 per day (0.059)	0.998 per day (0.67)	25 per week was withdrawn	$BMDL_{01}^b$: 1.50 per day

[a] Benchmark dose lower confidence limit for a 1% increased risk for cancers of the lung, skin and bladder, and skin lesions.
[b] Benchmark dose lower confidence limit for a 1% increased risk for cardiovascular effects and nephrotoxicity in adults.

Table 9

Recovery range of elements from spiked samples and the Rice flour certified reference material.

Element	Recovery range % from spiked samples	Recovery range % from CRM
Arsenic	90.1–112.4	96.7–99.5
Cadmium	87.4–105.7	87.4–104.5
Mercury	76.5–101.9	84.1–109.2
Lead	72.1–114.6	86.2–112.5

BMDL01), indicating that EDI of the Lebanese population is less than BMDL01 proposed by EFSA. In contrast, UAE results indicate an unsafe level of exposure equivalent to 0.76 µg/kg bw/day, ranging within BMDL01 range for a 1% increased risk for cancers of the lung, skin and bladder, and skin lesions. HQ of UAE population exposure was 0.095–2.5, as calculated according to BMDL01 for a 1% increased risk for cancers of the lung, skin and bladder, and skin lesions. This indicates that the exposure of the UAE population to total As is considered slightly unsafe, as the EDI stands within the range of BMDL01 (close to the lower end) and HQ exceeds 1 for the lower end. However, in a report issued by the EFSA, 600 rice samples were analyzed for their contamination with As, of which they deduced that 70% of the total As content was iAs (EFSA, 2014). Since iAs is the toxic species and we only tested for tAs, the actual iAs levels could be considered less worrying as a result.

Exposure of the UAE population to tAs (0.76 µg/kg bw/day, equivalent to 57.76 µg/day) is comparable to the findings of Jahiruddin et al. (2017) reporting the As exposure of Bengali population as 57.6 ± 36.4 µg/day, but less than levels reported by Chu et al. (2021) (0.0012 and 0.0015 mg/kg bw/day of Vietnamese men and women, respectively). However, in KSA, the country neighboring UAE, Mohamed et al. (2017) reported that exposure to As was remarkably lower (8.18 µg/day) than the one we calculated for the UAE (57.76 µg/day), but was comparable to the Lebanese exposure of 5.6 µg/day.

3.3.2. Cadmium

The calculated exposure level to Cd from rice in Lebanon was 0.09 µg/kg bw/day, equivalent to 2.7 µg/kg bw/month or 0.63 µg/kg bw/week, with HQ = 0.108 according to JECFA PTMI, and HQ = 0.252 according to EFSA PTWI. In the UAE, exposure was 0.15 µg/kg bw/day, equivalent to 4.5 µg/kg bw/month or 1.05 µg/kg bw/week, with HQ = 0.18 (using JECFA PTMI), and HQ = 0.42 (using EFSA PTWI). JECFA withdrew its previously established PTWI of 7 µg/kg bw due to Cd's long half-life of 15 years in human kidneys, so a monthly value (PTMI) of

25 µg/kg bw was considered more appropriate (JECFA, 2013). The EFSA CONTAM Panel (2011) has set a value of 2.5 µg/kg bw for PTWI for Cd. The calculated exposure level to Cd from rice in Lebanon was 2.7 µg/kg bw/month or 0.63 µg/kg bw/week, with HQ = 0.108 according to JECFA PTMI, and HQ 0.252 according to EFSA PTWI, implying that Lebanese exposure was lower than tolerable values and does not impose Cd associated health risks. In the UAE, EDI was 0.15 µg/kg bw/day, equivalent to 4.5 µg/kg bw/month or 1.05 µg/kg bw/week. UAE HQs were equivalent to 0.18 (using JECFA PTMI) and 0.42 (using EFSA PTWI), indicating that exposure to Cd is acceptable in UAE.

Nasreddine et al. (2006) reported that in Lebanon, exposure to Cd was 0.3 µg/day, based on 50.1 g/day consumption of cooked rice. This level was lower than the current studied exposure because of the significantly higher detected levels of contamination of rice with Cd in our study. Results from Italy were also lower, reporting 0.16 µg/week from brown rice and 0.1 µg/week from white rice (Pastorelli et al., 2018). In Bangladesh, the assessed exposure was 19.7 ± 20.0 µg/day for an adult male weighing 60 kg (equivalent to 0.33 µg/kg bw/day), thus being higher than our estimated results for Lebanon and the UAE (Jahiruddin et al., 2017). Rabbani et al. (2015) found that daily exposure to Cd from Iranian rice was 0.00035 vs. 0.00025 mg/kg bw/day from non-Iranian rice. Lien et al. (2021) found that exposure to Cd from Taiwanese rice was 0.06 µg/kg bw/day for male rice consumers between the ages of 19–65 years, thus more than the exposure in Lebanon and less than that in the UAE.

3.3.3. Chromium

In Lebanon, EDI of Cr was 0.11 µg/kg bw/day, equivalent to 7.79 µg/day for an adult with an average body weight of 70 kg. HQ was 0.03, according to WHO and EFSA guidelines. In the UAE, EDI was 72.5 µg/day for 76 kg adult, with HQ = 0.29. A tolerable upper limit for chromium is not available, but WHO has set 250 µg/day as a maximum level for supplemental intake of Cr (WHO, 1996). The EFSA ANS panel endorsed this limit for total Cr intake (EFSA ANS, 2010). In Lebanon, EDI of Cr was 0.11 µg/kg bw/day, equivalent to 7.79 µg/day for a 70 kg adult, with HQ = 0.03, according to WHO and EFSA limit. In the UAE, exposure amounted to 72.5 µg/day for a 76 kg adult, with HQ = 0.29. Both were significantly lower than the limits.

The reported daily Cr intake of 56.93 µg/day in KSA was higher than those reported for both countries in our study (Mohamed et al., 2017). Furthermore, significantly higher Cr intake levels were presented in Bangladesh (423 ± 334 µg/day) and Vietnam (0.0039 and 0.003 mg/kg bw/day for men and women, respectively) (Chu et al., 2021; Jahiruddin et al., 2017).

3.3.4. Mercury

Exposure to Hg in Lebanon was 0.05 µg/kg bw/day equivalent to 0.35 µg/kg bw/week, with HQ = 0.09 (according to JECFA and EFSA PTWI). In the UAE, exposure was 0.71 µg/kg bw/day, equivalent to 4.96 µg/kg bw/week, with HQ = 1.23. PTWI for Hg is 4 µg/kg bw according to JECFA and ESFA CONTAM (JECFA, 2011a, 2011b, 2011c; EFSA CONTAM, 2012). In our study, EDI of Hg in Lebanon was 0.05 µg/kg bw, with EWI being 0.35 µg/kg bw and HQ equivalent to 0.09, indicative of low and safe levels of exposure to Hg in Lebanon. In the UAE, EDI was 0.71 µg/kg bw/day, equivalent to EWI of 4.96 µg/kg bw, with HQ = 1.23, signifying a high and unsafe level of exposure to Hg in the UAE through exceeding weekly tolerable limits. Zeng et al. (2015) observed a comparable but lower level of exposure to Hg from brown rice in China (0.5 µg/kg/day; 3.5 µg/kg/week). Another study in China estimated lower exposure levels to Hg from rice ranging from 0.03 µg/kg bw/day for adults to 0.04 µg/kg bw/day for children (Huang et al., 2013). In Brazil, significantly lower levels of exposure of 0.22 µg/day from Brazilian rice were reported (Batista et al., 2012).

3.3.5. Lead

EDI of Pb in Lebanon was 0.088 µg/kg bw/day, with HQ = 0.059

(according to EFSA BMDL), whereas in the UAE, EDI was 0.998 μg/kg bw/day, with HQ = 0.67. JECFA Committee concluded that the previously established PTWI of 25 μg/kg bw could no longer be considered health protective. Accordingly, it was withdrawn. Moreover, because there is no indication for the threshold for the critical effects of lead, the Committee further deduced that it was not possible to establish a new PTWI (JECFA, 2011a, 2011b, 2011c). EFSA, on the other hand, has set a BMDL01 of 1.50 μg/kg bw/day for the association with a 1% extra risk for cardiovascular effects and nephrotoxicity in adults from Pb exposure (EFSA, 2012). Consequently, according to JECFA guidelines, the exposure to Pb in Lebanon of 0.088 μg/kg bw/day could be a cause of concern. However, according to EFSA, and with HQ = 0.059, this level of exposure is less than BMDL01 associated with a 1% increased cardiovascular effects and nephrotoxicity in adults resulting from Pb exposure. Exposure levels to Pb in the UAE were higher with a value of 0.998 μg/kg bw/day and HQ = 0.67. Similar to Lebanon's case, those results could not be considered safe according to JECFA, but they are still below EFSA's proposed BMDL.

The Pb exposure level from rice and rice products reported 16 years ago for the Lebanese population was 0.2 μg/day, thus lower than our current estimated level (Nasreddine et al., 2006). Internationally, our results for Lebanon and the UAE were higher than the ones reported in Batista et al. (2012) in Brazil (0.44 μg/day) and Bielecka et al. (2020) in Poland (0.00019 mg/day).

UAE results were also higher than the ones reported by Mohamed et al. (2017) in KSA (9.36 μg/day) and Chu et al. (2021) in Vietnam (0.0008 mg/kg bw/day for men and 0.001 mg/kg bw/day for women). Nevertheless, exposure of 0.0864 μg/kg bw/day assessed by Wang et al. (2019) in China was similar to the Lebanese exposure. Other studies reported higher levels of exposure than our revealed levels for Lebanon and the UAE. Two Iranian studies published such high levels; Rabbani et al. (2015) stated that exposure was 0.0035 and 0.004 mg/kg/day from Iranian and non-Iranian rice, respectively, and Mahvi et al. (2016) reported exposure from Indian rice Iran equivalent to 0.88 mg/kg bw/week. Likewise, higher levels were stated by Jahiruddin et al. (2017) in Bangladesh (74.1 ± 43.5 μg/day) and Orisakwe et al. (2012) in Nigeria (0. 3775 g/day).

Overall, our most alarming results were related to Hg in Lebanon and Pb and Hg in the UAE. Both metals are extremely toxic to the human body. The brain is the most affected by Hg intoxication, followed by the nervous, renal, and muscular systems (Bielecka et al., 2020). Exposure to Hg particularly imposes greater threats to the development of children in utero and early in life (WHO, 2021a, 2021b). Pb may disrupt the development of the nervous system, especially during the prenatal period throughout childhood (Mason et al., 2014). According to WHO 2021a, 2021b, lead exposure was responsible for 900,000 deaths and 21.7 million years of healthy life lost from around the world, especially in low and middle-income countries. Pb has detrimental effects on human health, such as causing central nervous system impairment and negatively affecting fertility and the kidneys (Kumar et al., 2020). Young children are the most vulnerable to the toxic effects of Pb and can suffer from permanent adverse health impacts, especially altering the development of the brain and nervous system (WHO, 2021a, 2021b).

Fortunately, toxic metals contamination is preventable. Importing from reputable suppliers and actively assessing and testing imported rice by governments and stakeholders could be of great benefit. Moreover, rinsing and soaking rice can significantly reduce toxic metals content in food. According to a study conducted in the USA by TatahMentan et al. (2020), washing white rice reduced levels of toxic metals such as Pb and Cd by 57% and 46%, respectively. Sharafi et al. (2019) provided more evidence for this effect by proving that washing rice 5 times, then cooking it with excess water, was the most effective cooking method. The authors reported that the reduction in toxic metal levels was 42.3% for As, and 42.9% and 27.6% for Pb. Al-Saleh & Abduljabbar (2017) found that soaking or rinsing rice grains with water reduced Pb and Cd levels in all brands to safe levels. Furthermore, cooking methods

(boiling, steaming, and frying among others) can alter the levels of toxic metals in food by several ways, such as the evaporation of water and volatile substances, solubility of the element, in addition to the binding of the metal to other macronutrients present in food, such as carbohydrates, lipids, and proteins (Kobia et al., 2016). Therefore, exposure levels might be lower than estimates, depending on the handling and cooking methods, but preliminary precautions must necessarily take place.

Our study has several strengths. The first strength was the utilization of ICP-MS, which is considered the gold standard in heavy metals testing, detecting heavy metals at very low concentrations. Second, samples were representative of the rice available in the Lebanese and UAE markets. Third, all sample preparations and testing procedures were validated through replication steps. We also clearly compared our results to the most updated international limits, a requirement that is mostly not met by other related studies. Nevertheless, some limitations could be pointed out. Since washing and cooking methods affect rice's toxic metal concentrations, our results might differ at the consumption level. Another limitation would be that our study only assessed total toxic metals concentrations rather than performing speciation analysis for each metal. In addition, due to the lack of data on consumption of rice in UAE via food frequency questionnaires (FFQ), the evaluated exposure in UAE could be under or overestimated as it was calculated based on total national imports and consumption rates rather than a representative FFQ.

4. Conclusion

In conclusion, our findings demonstrated that all tested rice samples, representing rice marketed in Lebanon and the UAE, were contaminated with total As, Cd, Cr, Hg, and Pb. In UAE, the percentages of samples exceeding international limits were 9%, 1%, 100%, and 69% for As, Cd, Hg and Pb, respectively. Among the samples from Lebanon, 25%, 73%, 100%, and 69% were above limits for As, Cd, Hg and Pb, respectively. Average concentrations of Hg and Pb in UAE and Lebanon were exceeding international limits, with Cd in Lebanon exceeding the European limit. Moreover, alarming findings of above PTWI and BMDL exposure levels for Hg and Pb from rice were detected in the UAE due to high rice consumption. In Lebanon, general exposure levels to Cd, Hg, and Cr were not considered alarming as the Lebanese rice consumption rate is lower than in the UAE, despite the high levels of contamination of rice with Hg and Cd. No provisional tolerable limits are currently recommended for Pb. Therefore, any level of exposure could also be considered unsafe, especially with the high levels of contamination observed in the Lebanese samples. This could also mean that people consuming higher amounts of rice might be at risk of suffering from the deleterious health effects associated with the consumption of these highly toxic metals. Future studies should focus on assessing the major handling and cooking methods used explicitly in the country and, consequently, assessing metal contamination and exposure from cooked rice to provide more representative results. Furthermore, studies performing speciation analysis for each metal should be conducted. In Lebanon, it is recommended to assess unpackaged rice sold at some traditional markets since it could be more contaminated than packaged brands as it is exposed to the environmental pollution. It is also recommended that FFQ based studies to be conducted in the UAE for accurate figures on rice consumption patterns.

CRediT authorship contribution statement

Elias Akoury: Conceptualization, Data curation, Formal analysis, Methodology, Validation, Writing – original draft, Writing – review & editing. **Najwa Mansour:** Data curation, Methodology, Validation, Writing – original draft, Writing – review & editing. **Ghina Abdul Reda:** Data curation, Writing – original draft, Writing – review & editing. **Hani Dimassi:** Formal analysis, Writing – original draft, Writing – review &

editing. **Layal Karam:** Funding acquisition, Writing – original draft, Writing – review & editing. **Nisreen Alwan:** Funding acquisition, Writing – original draft, Writing – review & editing. **Hussein F. Hassan:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Methodology, Project administration, Supervision, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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