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Exposure to Toxic Metals from the Consumption of
Rice in Lebanon and the United Arab Emirates

By

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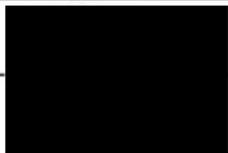
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Exposure to Toxic Metals from the Consumption of Rice in Lebanon and the United Arab Emirates

Ghina Abdul Reda

ABSTRACT

Our study aimed to quantify toxic metals (Arsenic, As, Cadmium, Cd, Chromium, Cr, Mercury, Hg and Lead, Pb) in rice, determine the factors affecting its contamination, and evaluate the dietary exposure from its consumption in Lebanon and UAE. A market screening was done and subsequently, all the brands (total of 236 samples, with 107 from Lebanon and 129 from UAE) were collected and tested. Inductively Coupled Plasma-mass Spectrometry (ICP-MS) was used. In Lebanon, the average \pm standard deviation in samples was 0.24 ± 0.08 , 0.29 ± 0.13 , 0.34 ± 0.13 , 0.15 ± 0.05 , and 0.27 ± 0.10 mg/kg for As, Cd, Cr, Hg and Pb, respectively. In UAE, concentrations were 0.18 ± 0.09 , 0.07 ± 0.04 , 0.23 ± 0.11 , 0.17 ± 0.05 , and 0.24 ± 0.08 mg/kg for As, Cd, Cr, Hg and Pb, respectively. In UAE, 9, 1, 100 and 69% of samples exceeded the international limits for As, Cd, Hg and Pb, respectively. In Lebanon, 25, 73, 100 and 69% of samples were above limits for As, Cd, Hg and Pb, respectively. No limits were set for Cr for comparison. In Lebanon, for As, brown rice was significantly more contaminated than white and parboiled rice ($p=0.02$), long rice grains were significantly more contaminated than short/medium grains ($p=0.002$), and rice brands originating from developed countries were significantly more contaminated compared to those from developing

countries ($p < 0.001$). In UAE, for As, packing season, country of origin, collecting same brands at two different times had significant effect ($p = 0.011, 0.016, \text{ and } < 0.001$, respectively). In Lebanon, for Cd, collecting same brands at two different times and grain size had significant effect ($p < 0.001$). In UAE, for Cd, collecting same brands at two different times had a significant effect ($p = 0.008$). Regarding Cr, in Lebanon, country of origin, grain size, rice type, and time between packing and purchasing had a significant effect ($p = 0.006, p < 0.001, p < 0.001 \text{ and } p < 0.001$, respectively). In UAE, for Cr, the only statistically significant variable was collecting same brands at two different times ($p < 0.001$). For Hg in Lebanon, grain size and type had a significant effect ($p = 0.019 \text{ and } p = 0.012$, respectively), while collecting same brands at two different times had a significant ($p < 0.001$) effect in both Lebanon and UAE. None of the variables had a significant effect on Pb in samples from Lebanon. For Pb in UAE, statistical significance was observed for collecting same brands at two different times and country of origin ($p = 0.004 \text{ and } 0.001$, respectively). Exposure levels to Hg from rice were higher than both FAO/WHO and European Food Safety Authority limits were detected in UAE. In Lebanon, exposure levels were not considered alarming for Cd, Cr and Hg, as the Lebanese rice consumption rate is relatively lower than in UAE, despite the high levels of contamination. No provisional tolerable limits are currently set for As and Pb; thus, the observed exposure to both metals could be a cause of concern in both countries. Future studies must assess the effects of handling and cooking to better assess exposure to toxic from rice exposure in both countries.

Keywords: Rice, Toxic metals, ICP-MS, Exposure, Lebanon, United Arab Emirates

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List of Abbreviations

AAS: Atomic absorption spectrometry

ADAFSA: The Abu Dhabi Agriculture and Food Safety Authority

ADFCA: Abu Dhabi Food Control Authority

As: Arsenic

BDL: Below detection level

BMDL: Benchmark dose lower confidence limit

CAC : Codex Alimentarius Commission

Cd: Cadmium

Cr: Chromium

CV-AAS: Cold vapor atomic absorption spectrometry

DMA: Direct mercury analyze

EC: European Commission

EDI: Estimated daily intake

EFSA ANS: European Food Safety Authority Panel on Food Additives and Nutrient Sources added to Food

EFSA CONTAM: European Food Safety Authority Panel Panel on Contaminants in the Food Chain

EFSA NDA: European Food Safety Authority Panel on Dietetic Products, Nutrition and Allergies

EFSA: European Food Safety Authority

EWI: Estimated weekly intake

F-AAS: Flame atomic absorption spectrometry

FAO: The Food and Agriculture Organization of the United Nations

FFQ: Food frequency questionnaire

GF-AAS: Graphite furnace atomic absorption spectrometry

Hg: Mercury

HG-AAS: Hydride-generating atomic absorption spectrometry

HG-AFS: Hydride-generating atomic fluorescence spectrometry

HQ: Hazard quotient

iAs: Inorganic Arsenic

ICP- DRC-QMS: Inductively coupled plasma-dynamic reaction cell-quadrupole mass spectrometry

ICP-AES: ICP atomic emission spectrometry

ICP-DRC-QMS: Inductively coupled plasma-dynamic reaction cell-quadrupole mass spectrometry

ICP-MS: Inductively coupled plasma mass spectrometry

ICP-OES: Inductively coupled plasma optical emission spectrometry

JECFA: Joint FAO/WHO Expert Committee on Food Additives

LIBNOR: Lebanese Standards Institution

LOD: Limit of detection

LOQ: Limit of quantification

MRL: Maximum residue levels

n: number

NA: Not applicable

NAA: Neutron activation analysis

Nd: Not detectable

Pb: Lead

PTMI: Provisional tolerable monthly intake

PTWI: Provisional tolerable weekly intake

Q-ICP-MS: Quadruple inductively coupled plasma mass spectrometry

Sd: Standard deviation

tAs: Total arsenic

UAE: United Arab Emirates

USDA FAS: United States Department of Agriculture Foreign Agricultural Service

WHO: World Health Organization

CHAPTER 1

LITERATURE REVIEW

1.1 HEAVY METALS

1.1.1 Definition

Heavy metals, also known as toxic metals, are elements that occur naturally and have high density and atomic weight. They are highly toxic to the human health and environment, and their toxicity varies according to their dose, type and way of exposure, age, gender, genetics and nutritional status of the exposed person (Tchounwou et al., 2012). 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, chromium, arsenic, cadmium and mercury are the main metals studied for their high levels of toxicity (Tchounwou et al., 2012).

Arsenic (As) is an omnipresent non-essential toxic metalloid of which its primary sources are geochemical, meaning related to the chemistry of earth, and anthropogenic sources, that are related to agricultural and industrial techniques (Garbinski et al., 2019). Inorganic arsenic compounds (i.e. arsenite and arsenate) are toxic, whereas organic arsenic, usually found in fish and shellfish, is not considered toxic (Olsen & Mørland, 2004; Shraim, 2014).

Cadmium (Cd) is a non-essential toxic transition metal. It naturally occurs in the environment as a byproduct of agricultural and industrial activities (Genchi et al., 2020). Cadmium chloride (CdCl_2) and cadmium oxide (CdO) are toxic in particular (Vallero, 2014).

Chromium (Cr) is a transition metal that exists in nature mostly in the form of Cr (III) (Ferreira et al., 2019), which is a nontoxic-nutritionally-essential trace element (Tumolo et al., 2020), and Cr (VI) to a lesser extent (Ferreira et al., 2019). Cr is widely available in foods, such as meat, oils, cereals, pulses, fish and breads (European Food Safety Authority Panel on Dietetic Products, Nutrition and Allergies [EFSA NDA Panel], 2014). Cr (VI), usually produced from Cr (III), is broadly used in textile dyes, paints, inks, plastics, corrosion inhibitors, leather tanning agents and wood preservatives (Ferreira et al., 2019). Deposition from air, surface runoff, and release of municipal and industrial wastewaters are sources for the contamination of Cr in surface waters. Using stainless steel containers, equipment and utensils in food processing may contribute to the release of Cr (III) in food, especially acidic food (EFSA Panel on Contaminants in the Food Chain [CONTAM], 2014). Several systems in the body reduce some amount of Cr (VI) to Cr (III) (Boon et al., 2010). Cr (VI) is the most toxic form of chromium (Tumolo et al., 2020).

Lead (Pb) is a toxic heavy metal that accumulates in the blood, bones, liver, kidneys, brain and skin (Zulkafflee et al., 2019). All forms of Pb are toxic, but its organic form Tetra-ethyl Pb is more toxic than the others (Kumar et al., 2020). Pb is a component of many chemicals, such as pesticides (Phrukphicharn et al., 2021). It has several industrial applications, mainly in the production of sulfuric acid, cable covers, connecting materials, shields for atomic reactors, containers for radioactive materials, paint and ceramics, chemical and construction industries, printing fonts, and aviation gasoline (Zulkafflee et al., 2019). It is also used in the soldering and petroleum industries, mixing oil and fuel, producing batteries, making ammunition, and controlling the

loudness of the sound of machinery (Phrukphicharn et al., 2021).

Mercury (Hg) is a toxic heavy metal (Bernhoft, 2012). The three chemical forms of Hg are metallic (Hg^0), inorganic (Hg^{2+} , Hg_2), and organic Hg (methylmercury (MeHg), dimethyl mercury, ethyl mercury, phenyl mercury) (Bielecka et al., 2020). Atmospheric elemental mercury (mainly from coal burning and mining) contaminates water where microorganisms convert it to methyl or ethyl mercury that is then ingested by fish (highest concentrations in tuna, swordfish and shark) (Bernhoft, 2012).

1.1.2 Factors leading to toxic metals contamination and consequent human exposure

The main sources of toxic metals are geochemical and 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). For instance, the decomposition of compost fertilizers in flooded rice fields results in the mobilization of arsenic into arsenite, leading to subsequent uptake by the plants (Diyabalanage et al., 2016). Sewage sludge and animal manures may contain high amounts of heavy metals contaminating agricultural soils as well (Chamannejadian et al., 2013). Moreover, by widely existing in industrial, agricultural, technological, medical and domestic applications, they rapidly spread to the environment. Industrial sources include coal burning, metal processing, petroleum combustion, high-tension lines, plastic, textiles, paper processing, and wood preservation industries (Tchounwou et al., 2012). Heavy metals such as Cr, Pb, Hg, and Cd can be released from waste incineration as well (Wu, Lin & Zeng, 2014). Environmental contamination could occur due to the re-suspension

of residues and evaporation of metals from water resources into soil and ground water (Tchounwou et al., 2012). For mercury in particular, human exposure is mainly from fish consumption, along with inhaling elemental methyl mercury vapor from occupational exposure, and dental amalgam (Bernhoft, 2012). Municipal sewage also contains mercury species, including MeHg (Wang & Mao, 2019). For Cd, smoking is also a specific route of exposure (Bielecka et al., 2020). Moreover, 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, 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. Vegetarians and children are particularly at higher risks of exposure (Sommella et al., 2013).

1.1.3 Health implications

Being non-biodegradable and persistent, toxic metals can have detrimental effects on the environment and health (Zulkafflee et al., 2019). Even at low levels of exposure, heavy metals are considered as carcinogens and may cause multiple organ damage (Tchounwou et al., 2012). Consuming food (i.e., rice) contaminated 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). Upon the consumption of contaminated food and water, heavy metals accumulation can be extremely problematic.

Arsenic: The Agency for Toxic Substances and Disease Registry (ATSDR) has put

arsenic 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 (Garbinski et al., 2019). It is linked to several diseases such as various types of cancer, diabetes and 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: 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, adrenals damage, hematopoietic 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 nasopharynx cancers (Bielecka et al., 2020; Genchi et. al., 2020). Cd may also hinder the activity of antioxidant enzymes (Genchi et. al, 2020).

Chromium: 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 that there is 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).

Mercury: 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, extrapyramidal impairments to clonic seizures in severe exposures (Bernhoft, 2012).

Lead: Upon absorption, Pb accumulates in the blood, bones, liver, kidneys, brain and skin (Zulkafflee et al., 2019). Pb 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, 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).

1.2 RICE

Rice Varieties and origin of cultivation

1.2.1 Rice has been detected in archeological sites originating to 8000 BC; however, there is a persisting debate regarding its original date of domestication (Sweeney & McCouch, 2007). The most commonly cultivated rice species nowadays are *Oryza sativais* (Asian rice) and *Oryza glaberrima* (African rice) (Rathna Priya et al., 2019). *Oryza sativai* is the most abundant species grown worldwide, whereas *Oryza glaberrima* is cultivated in smaller amounts in West Africa (Khush, 1997; Rathna Priya et al., 2019). There are more than 110,000 varieties of cultivated rice (Fukagawa & Ziska, 2019). Rice is cultivated between 55°N and 36°S latitudes, in wet environments, and under distinct conditions such as irrigated, rain-fed lowland or upland, and flood-prone ecosystems (Khush, 1997; Saleh et al., 2019). It is cultivated in more than 100 countries around the world, with the majority (90%) of cultivation and production being from Asia (Fukagawa & Ziska, 2019). To a lesser extent, it is produced in the Americas, Europe, Africa, and Australia (Khush, 1997). In general, rice is categorized based on its shape and processing: refined (white or polished rice) or whole-grain (brown, red, and black rice) (Harvard, 2021; Rathna Priya et al., 2019). Its shape varies by having a long (i.e., Basmati rice), medium (i.e., Arborio rice), or short (i.e., sushi rice) length (Harvard, 2021).

Nutritional Value

Each type of rice has a different quality and nutritional content (Fukagawa & Ziska, 2019). It is considered as a main source of energy, carbohydrates, protein and other essential nutrients, especially in developing countries (Saleh et al., 2019). It is a source of phosphorous, manganese, iron, selenium, and B vitamins, specifically, folic acid, thiamin and niacin (Fukagawa & Ziska, 2019). When compared to white rice, and because most nutrients are concentrated in the outer brown, brown rice is richer in dietary fiber, proteins, fats, vitamins and minerals, along with phenolic acids, flavonoids, γ -oryzanol, aminobutyric acid (GABA), α -tocopherol, and γ -tocotrienol (Saleh et al., 2019). Brown rice is particularly considered a good source of magnesium and phosphorus and an excellent source of manganese. Red and black rice contain the antioxidant anthocyanin, and red rice types are rich in iron and zinc, whereas black rice types are especially high in protein, fat and fiber (Rathna Priya et al., 2019). Brown rice, germinated brown rice and derivatives have various potential health benefits that include having antioxidant, neuroprotective, anticancer, and cholesterol lowering properties (Saleh et al., 2019). However, it is imperative to note that whole grain rice may contain higher levels of heavy metals. Brown rice is contaminated with more inorganic arsenic due to its germ layer that retains higher amounts of the metal (TatahMentan et al., 2020).

Consumption and Production Pattern

Rice is a staple food for more than three billion individuals around the world, accounting for around 50% of the world's total population (Mosleh et al., 2015). According to the latest report issued in 2021 by the United States Department of

Agriculture (USDA), Foreign Agricultural Service (FAS), the total global rice production years 2020-2021 was 503,167 thousand metric tons, of which only 2,857 thousand metric tons produced in the middle east (USDA/ FAS, 2021a). In the UAE, there have been recent trials for planting rice in the desert of the Emirate of Sharjah, using desalinated water along with using hybrid rice strains can be grown in saltwater (USDA/FAS, 2021b). 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 in preventing such agricultural practices (FAO, 2020). Therefore, both countries depend mainly on imports for satisfying their local rice demands. Figure 1 demonstrates the monthly imports of rice in the UAE.

Regarding consumption, around two billion people in Asia consume rice as a major staple food (Baldwin et al., 2012). During 2010 through 2011, the annual consumption of rice was around 65 kg per capita, accounting for 19% of the global energy intake (Mohanty, 2013). The total reported annual global consumption of rice in 2020-2021 was 499,562 thousand metric tons and 9,880 thousand metric tons in the Middle East in particular (USDA/FAS, 2021a). USDA/FAS further forecasted that specifically UAE rice imports and consumption would reach 1.15 MMT (million metric ton) in MY 2021/22 (USDA/FAS, 2021b).

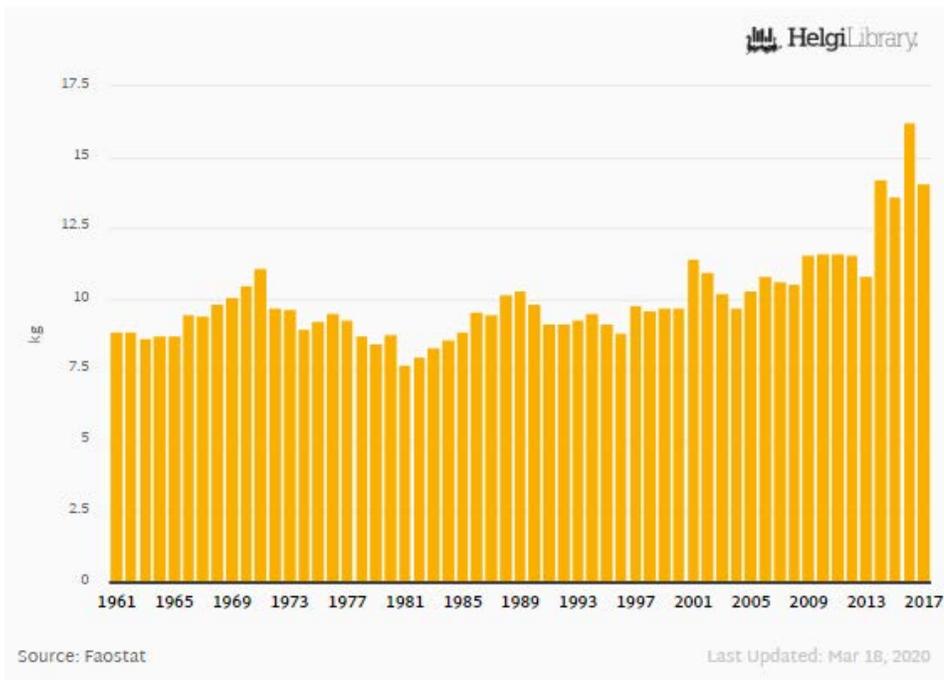


Figure 1. UAE rice monthly imports in 2019 to 2020 (USDA/FAS, 2021b)

In Lebanon, the latest reported rice consumption per capita was 14.1 kg in 2017.

Figure 2 demonstrates the Lebanese rice consumption pattern.

Figure 2. Lebanon rice consumption (kg per capita) (Helgi Library, 2020)



1.3 HEAVY METALS IN RICE

1.3.1 Analytical Methods Used in the Literature

1.3.1.1 Inductively Coupled Plasma Detectors

The inductively coupled plasma (ICP) ion source is a unique type of plasma that utilizes induction from a high-frequency magnetic field in order to sustain its power. (Amr, 2012).

Inductively coupled plasma mass spectrometry (ICP-MS): Mass spectrometry is a powerful analytical method used to determine and quantify analytes from the mass-to-charge ratio of ions generated from a sample (Rockwood et al., 2018). ICP-MS is used for identifying trace and toxic elements in various sample types (Rockwood et al., 2018). It is a convenient technique providing rapid multi-element analysis with low detection limit and wide linear range (Srinuttrakul et al., 2018).

Testing for mercury with ICP-MS is associated with problems related to long washout time and nonlinear calibration curves. Adding 2-mercaptoethanol and L-cysteine as washing reagents could solve this issue.

However, taking into consideration the toxicity and odor of 2-mercaptoethanol, the better option would be adding cysteine to flush the system (Li et al., 2006).

Inductively coupled plasma optical emission spectrometry (ICP-OES), also called ICP atomic emission spectrometry (ICP-AES): In this system, the elemental species are atomized and ionized in the high temperature plasma (Michalke & Nischwitz, 2013). ICP-AES has lower sensitivity than ICPMS but has higher tolerance to samples with high salt content, and can monitor

nonmetallic elements, such as C, S, P, Cl, unlike in ICP-MS (Michalke & Nischwitz, 2013).

Inductively coupled plasma-dynamic reaction cell-quadrupole mass spectrometry (ICP-DRC-QMS): The Dynamic Reaction Cell (DRC) is an enclosed quadrupole, which acts as the interface between the single lens ion optics chamber and the mass analyzer high vacuum chamber in the ICP-MS (Scott & Vladimir, 1999).

1.3.1.2 Atomic absorption spectrometry (AAS)

A method of elemental analysis that has high sensitivity and is comparably inexpensive (Butcher, 2005; Michalke & Nischwitz, 2013). It permits the determination of metals in a variety of samples through the atomic absorption phenomenon that measures the reduction of the intensity of optical radiation after passing through a cell containing gaseous atoms (Michalke & Nischwitz, 2013). There are several types of AAS: flame (F-AAS), cold vapor (CV-AAS), hydride- generating (HG-AAS), and graphite furnace (GF-AAS) systems (Butcher, 2005).

1.3.1.3 Fluorescence Spectroscopy

A form of electromagnetic spectroscopy that allows the analysis of fluorescence from a sample (Chirayil et al., 2017). Absorption spectroscopy is a complementary technique to it (Chirayil et al., 2017). It works by a light-emitting process activated by the absorption of excited radiation of a particular wavelength, allowing the efficient interaction between light and substance (Kohli & Mittal, 2019).

1.3.1.4 Electrochemical Systems

They utilize electrodes made from different material, mainly Mg, Au, Ag, Pt, and carbon (García-Miranda Ferrari et al., 2020). Electrochemical devices are simple to use and reagent-less, and impose low cost (Pujol et al., 2014).

1.3.1.5 Neutron Activation Analysis (NAA)

A nuclear process utilized for determining the concentrations of elements. NAA works based on the excitation by neutrons, allowing the treated sample to emit gamma rays. It is used to precisely identify and quantify elements, including heavy metals (Hamidatou et al., 2013).

1.3.2 Maximum Residues Levels (MRLs) of Heavy Metals Allowed and Maximum Safe Values of Exposure

Table 1. MRLs of heavy metals allowed and maximum safe values of exposure.

	As	Pb	Cd	Hg	Cr
MRLs (mg/kg)	<p>Total As (tAs): Codex Alimentarius Commission (CAC) 0.3 (Codex Alimentarius Commission (CAC), 2012).</p> <p>Inorganic As (iAs)^a: -Polished rice: 0.2 (CAC, 2019). - Husked rice: 0.35 (CAC, 2019).</p> <p>EC regulations are only for iAs^b (EC, 2015).</p>	0.2 in cereal grains (CAC, 2019; The Lebanese Standards Institution [LIBNOR], 2013).	0.4 in polished rice (CAC, 2019; LIBNOR, 2013).	No CAC limits were set in rice (only in fish).	No maximum allowed levels are specified for rice. (Zulkafflee et al., 2022).
Maximum safe values of exposure (µg/kg bw per week, day or month)	<p>The previously set PTWI^c=2.1 was withdrawn.</p> <p>BMDL_{0.5}^d= 2–7 µg/kg bw/ day for iAs (JECFA, 2011).</p> <p>Joint FAO/WHO Expert Committee on Food Additives (JECFA)</p>	Previously established PTWI=25 was withdrawn (JECFA, 2011). No level is considered safe.	PTMI ^e : 25 (JECFA, 2013). PTWI=7 was withdrawn because of its long half-life (JECFA, 2010)	PTWI: 4 (JECFA, 2011). Total Hg: 0.01 in rice (EC, 2018).	250 µg/day (WHO, 1996)

Food Safety Authority (EFSA) ($\mu\text{g}/\text{kg}$ bw per day or week)	BMDL ₀₁ ^f : 0.3-8 $\mu\text{g}/\text{kg}$ b.w./day (EFSA Panel on Contaminants in the Food Chain (CONTAM), 2009)	BMDL ₀₁ ^g = 1.50 (EFSA, 2012).	PTWI: 2.5 $\mu\text{g}/\text{kg}$ b.w (EFSA CONTAM, 2011).	PTWI : 4 $\mu\text{g}/\text{kg}$ b.w (EFSA CONTAM, 2012)	Total Cr: 250 $\mu\text{g}/\text{day}$ (EFSA Panel on Food Additives and Nutrient Sources added to Food (ANS), 2010) Cr (III): 300 $\mu\text{g}/\text{day}$ (EFSA CONTAM, 2014).
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^a tAs is converted to iAs by a factor of 70% (EFSA, 2014).

^b Non-parboiled milled rice (polished or white rice): 0.20 mg/kg; Parboiled rice and husked rice: 0.25 mg/kg

^c PTWI: Provisional tolerable weekly intake.

^d Associated with 0.5% increased incidence of lung cancer

^e PTMI: Provisional tolerable monthly intake.

^f Benchmark dose lower confidence limit associated with 1% extra risk for the incidence of cancer of the lung, skin and bladder, and skin lesions.

^g Benchmark dose lower confidence limit associated with a 1% extra risk for the incidence of cardiovascular effects and nephrotoxicity in adults.

1.3.3 Heavy Metals in Rice Worldwide and their Corresponding Exposure Levels

Table 2. Heavy metals' levels in rice and their corresponding exposure worldwide.

Reference	Country	Number of rice samples	Type and/or country of origin of rice	Analytical method	Range of toxic metals concentrations (Mean \pm SD) ($\mu\text{g}/\text{kg}$ or mg/kg or ng/g)	Mean exposure levels [†]
Nasreddine et al. (2006)	Lebanon	Number of rice samples not specified (105 total food and drink samples)	Rice and rice-based products	ICP-MS	Values in $\mu\text{g}/\text{kg}$: Pb: 4.1–5.5 (4.6) Cd: 3.2–8.8 (6.6)	Pb: 0.2 $\mu\text{g}/\text{day}$ Cd: 0.3 $\mu\text{g}/\text{day}$
Batista et al. (2012)	Brazil	44	Brazilian parboiled brown (PB), white (W) & parboiled white (PW)	ICP-MS	Values in ng/g : -Pb: W: 0.6-8.8 (5.3 \pm 4.1) PW: 0.4-9.5 (3.1 \pm 3.2) PB: 2.8-14.5 (7.8 \pm 5.2) -Hg: W: 0.3-10.4 (2.3 \pm 2.3) PW: 0.6-13.2 (3.5 \pm 4.3) PB: 0.6-13.4 (3.9 \pm 3.3)	Pb: 0.44 $\mu\text{g}/\text{day}$ Hg: 0.22 $\mu\text{g}/\text{day}$
Orisakwe et al. (2012)	Nigeria	Not specified	Nigerian	AAS	Pb: 61.17 mg/kg Cd: Nd	Pb: 0.3775 g/day Cd: -
Voica et al. (2012)	Romania	Not specified	Not specified	ICP-MS	Values in mg/kg : As: 0.042 Cd: 0.003 Pb: 0.039	–

Huang et al. (2013)	China	248	Polished rice Chinese (Zhejiang province)	GF-AAS & HG-AFS	Values in mg/kg: -As: <LOD-0.246 (0.0806±0.051) -Cd: <LOD-0.112 (0.0376±0.015) -Hg: <LOD-0.088 (0.0056±0.003)	Levels in µg/kg bw/day: -As: 0.49 (adults) & 0.34 (children) -Cd: 0.23 (adult) & 0.29 (children) -Hg: 0.03 (adults) & 0.04 (children) -Pb: 0.37 (adult) & 0.47 (children)
Sommella et al. (2013)	Italy	101	Type : Arborio, Carnaroli, Ribe, Ribe/Roma parboiled, Roma, Vialone Nano, Originario, Others (mixture of varieties) Origin: Italy	Q-ICP-MS	Values in mg/kg: -As: Arborio: 0.09-0.47 (0.23±0.01) Carnaroli: 0.10-0.43 (0.23±0.02) Ribe: 0.09-0.39 (0.18±0.01) Ribe/Roma parboiled: 0.14-0.28 (0.20±0.01) Roma: 0.11-0.22 (0.19±0.10) Vialone Nano: 0.20-0.40 (0.28±0.03) Others: 0.07-0.28 (0.18±0.01)	-

					<p>-Inorganic As (As_i) according to region:</p> <p>Lombardia: 0.03-0.17 (0.10±0.03)</p> <p>Piemonte: 0.001-0.20 (0.09±0.04)</p> <p>Emilia: 0.10-0.09 (0.10±0.004)</p> <p>Calabria: 0.02-0.13 (0.06±0.05)</p> <p>-Cr:</p> <p>Arborio: 0.11-0.76 (0.24±0.03)</p> <p>Carnaroli: 0.10-0.43 (0.23±0.02)</p> <p>Ribe/Roma parboiled: 0.14-0.28 (0.20±0.01)</p> <p>Roma: 0.11-0.22 (0.19±0.10)</p> <p>Vialone Nano: 0.20-0.40 (0.28±0.03)</p> <p>Originario: 0.15-0.54 (0.28±0.20)</p> <p>Others: 0.13-1.36 (0.51±0.10)</p> <p>-Cd:</p> <p>Arborio: (0.04±0.01) 0-0.16</p> <p>Carnaroli: (0.03±0.01) 0.1-0.12</p>
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					Ribe: 0-0.15 (0.04±0.01) Ribe/Roma parboiled: 0.01-0.06 (0.0±0.01) Roma: 0-0.08 (0.04 ±0.01) Vialone Nano: 0-0.04 (0.02 ±0) Originario: 0.05-0.1 (0.08 ±0.01) Others: 0-0.06 (0.03 ±0.02)	
Choi et al. (2014)	South Korea	304	Type: White and brown Origin: South Korea, China, USA, Thailand and Australia.	Q-ICP-MS	Values in µg/kg: As: White: (91.7±28.1) Brown: (101±34.6)	—
Shraim (2014)	KSA	84	Type: Yellow, white, long, medium, round. Origin: USA, Thailand, Pakistan, India, Egypt, Surinam, Australia	ICP-MS	Values in mg/kg: - <u>As</u> : USA: 0.127-0.464 (0.257 ± 0.142) Thailand: 0.181-0.250 (0.200 ± 0.025) Pakistan: 0.052-0.250 (0.147 ± 0.055) India: 0.026-0.228 (0.103 ± 0.048) Egypt: 0.039-0.154 (0.097 ± 0.044)	—

				<p>Surinam: (0.290)</p> <p>Australia: (0.188)</p> <p>France: (0.183)</p> <p><u>-Cd:</u></p> <p>USA: 0.005- 0.020 (0.011 ± 0.006)</p> <p>Thailand: 0.007-0.020 (0.012 ± 0.005)</p> <p>Pakistan: 0.007-0.042 (0.025 ± 0.014)</p> <p>India: 0.003- 0.046 (0.018 ± 0.009)</p> <p>Egypt: 0.003- 0.005 (0.004 ± 0.001)</p> <p>Surinam: (0.008)</p> <p>Australia: (<LOQ)</p> <p>France: (0.013)</p> <p><u>-Pb:</u></p> <p>USA: 0.014- 0.098 (0.041 ± 0.030)</p> <p>Thailand: 0.012-0.028 (0.019 ± 0.007)</p> <p>Pakistan: 0.018-0.022 (0.020 ± 0.002)</p>	
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					India: 0.003-0.218 (0.031 ± 0.044) Egypt: 0.015-0.031 (0.021 ± 0.008) Surinam: (0.018) Australia: (<LOQ) France: (<LOQ)	
Naseri et al. (2015)	Iran	210	Type: short & long grain Origin: Iran, India & Thailand	F-AAS & GF-AAS	Values in mg/kg: <u>-Cd:</u> -Domestic: Tarom: 0.34±0.01 Kohmare: 0.27± 0.05 Kamfirooz: 0.41 ±0.02 -Imported: Maryam: 0.34±0.02 Abdossaeid: 0.29±0.01 Tajmahal: 0.36±0.07 Khatereh: 0.43±0.03 Padideh: 0.44±0.02 Thailand: 0.44 + 0.03 Abdossalam: 0.48±0.03 <u>-Pb:</u> -Domestic: Tarom: 0.95 ± 0.21 Kohmare: 1.28 ± 0.30 Kamfirooz: 0.71 ± 0.15	Levels in µg/kg BW: <u>-Cd:</u> - Domestic: Tarom:4.376 Kohmare: 3.465 Kamfirooz : 5.300 - Imported: Maryam: 4.453 Abdossaeid: 3.747 Tajmahal: 4.684 Khatereh: 5.634 Padideh: 5.762 Thailand: 4.453 Abdossalam: 3.747 <u>-Pb:</u> - Domestic: Tarom: 12.256

					<p>-Imported: Maryam: 1.61 ± 0.37 Abdossaeid: 2.00 ± 0.15 Tajmahal: 1.62 ± 0.18 Khatereh: 0.96 ± 0.21 Padideh: 0.92 ± 0.22 Thailand: 0.76 ± 0.19 Abdossalam: 0.88 ± 0.45</p> <p><u>-Cr:</u> -Domestic: Tarom: 0.39 ± 0.03 Kohmare: 0.33 ± 0.03 Kamfirooz: 0.44 ± 0.03</p> <p>-Imported: Maryam: 0.39 ± 0.02 Abdossaeid: 0.36 ± 0.16 Tajmahal: 0.38 ± 0.05 Khatereh: 0.46 ± 0.06 Padideh: 0.53 ± 0.01 Thailand: 0.47 ± 0.07 Abdossalam: 0.55 ± 0.05</p>	<p>Kohmare: 16.491 Kamfirooz : 9.189 - Imported: Maryam: 20.751 Abdossaei d: 25.769 Tajmahal: 20.867 Khatereh: 12.410 Padideh: 11.858 Thailand: 20.751 Abdossala m: 25.769</p> <p><u>-Cr:</u> - Domestic: Tarom: 5.095 Kohmare: 4.325 Kamfirooz : 5.762 - Imported: Maryam: 5.069 Abdossaei d: 4.658 Tajmahal: 4.928 Khatereh: 5.955 Padideh: 6.891 Thailand: 5.069 Abdossala m: 4.658</p>
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Rabbani et al. (2015)	Iran	126	Iranian and non-Iranian	ICP-MS	<p>Values in mg/kg:</p> <p>-<u>As</u>: not found in any of rice samples</p> <p>-<u>Cd</u>: Iranian: (0.0636±0.0539) Non-Iranian: (0.0463±0.06864)</p> <p>-<u>Pb</u>: Iranian: (0.6416±0.3055) Non-Iranian: (0.8088 ± 1.0796)</p>	<p>Levels in mg/day/kg bw:</p> <p>-<u>Cd</u>: Iranian: 0.00035 Non-Iranian: 0.00025</p> <p>-<u>Pb</u>: Iranian : 0.0035 Non-Iranian : 0.004</p>
Rothenberg et al. (2015)	Madagascar	51	Madagascar unpolished rice (with red bran & brown bran)	CV-AFS & gas chromatography (for the separation of Hg species) & ICPMS	<p>Values in ng/g :</p> <p>-Total mercury (THg): 0.36-3.4 (1.1) -MeHg: 0.015-1.1 (0.12)</p> <p>Values in µg/g: -As: 0.074–0.28 (0.16) -Cd: <LOD–0.0061 (0.0024)</p>	–
Zeng et al. (2015)	China	28	Brown rice Chinese (Hunan province)	GF- AAS, F-AAS & HG- AAS	<p>Values in mg/kg:</p> <p>-As: 0.106-1.15 (0.336± 0.248)</p> <p>-Cd: 0.005-2.089 (0.312± 0.434)</p>	<p>Levels in µg/kg/d:</p> <p>-Cd 2.29 -Cr 0.78 -As 2.45 -Pb 0.16 -Hg 0.50</p>

					-Cr: 0.029-0.508 (0.106±0.085) -Pb: 0.003–0.103 (0.022±0.021) -Hg: 0.013-0.226 (0.069±0.060)	
Mahvi et al. (2016)	Iran	100	Indian	F-AAS	Values in mg/kg: -Cd: BDL -Cr: (0.631±0.43) -Pb: (0.320±0.230)	Levels in mg/kg bw/week: -Cd: - -Cr: 1.7 -Pb: 0.88
Pinto et al. (2016)	Portugal	86	Origin: Brands from Portuguese and Spanish market originating from Portugal, Spain, India, Italy, Thailand, Canada and USA. Type: White parboiled, brown and wild rice.	ICP-MS	Values in mg/kg: <u>-As:</u> White: 0.17±0.06 Parboiled: 0.16±0.05 Brown: 0.21±0.05 Wild: 0.18±0.03 <u>-Cr:</u> White: 0.11±0.05 Parboiled: 0.08±0.03 Brown: 0.10±0.05 Wild: 0.13±0.08 <u>-Cd:</u> White: 0.011±0.010 Parboiled: 0.005±0.003 Brown: 0.010±0.007 Wild: 0.009±0.006	Expressed as % of the established PTWI: <u>-As:</u> White: 5.0 Parboiled: 6.4 Brown: 5.0 Wild: 5.4 <u>-Cd:</u> White: 0.6 Parboiled: 0.2 Brown: 0.5 Wild: 0.5 <u>-Pb:</u> White: 0.04 Parboiled: 0.04 Brown: 0.07 Wild: 0.03

					<p>-Pb: White: 0.003±0.002 Parboiled: 0.003±0.002 Brown: 0.005±0.004 Wild: 0.002±0.002</p>	
Al-Saleh & Abduljabb(2017)	KSA	61	<p>Origin: India, Thailand, United States of America, Italy, Indonesia, Iraq, Pakistan, Australia & Spain</p> <p>Rice was soaked for 20 min or rinsed three times with deionized water</p>	AAS, direct Hg Analyzer DMA-80 (for methylmercury (MeHg))	<p>Values in µg/g dry weight: Soaked (rinsed): Pb: 0.034 (0.057) Cd: 0.015 (0.027) MeHg: 0.004 (0.007) tAs: 0.202 (0.183)</p>	<p>Levels in µg/kg bw/week: Soaked (rinsed): Pb: 0.638 (1.068) Cd: 0.279 (0.503) MeHg: 0.271 (0.309) tAs: 3.769 (3.407)</p>
Fan et al. (2017)	China	54	<p>Type: Brown rice</p> <p>Origin: China (Hunan province)</p>	GF-AAS, HG-AAS	<p>Values in mg/kg: -Cd: 0.006-0.241 (0.103 ± 0.007) -Pb: 0.040-1.074 (0.131 ± 0.034) -As: 0.211-0.986 (0.524 ± 0.033)</p>	-
Jahiruddin et al. (2017)	Bangladesh	71	<p>Type: Rain-fed (transplant Aman) and</p>	ICP-MS	<p>Values in mg/kg: -<u>As</u>: Irrigated: 0.047-0.506</p>	<p>Levels in µg/day: -As: 18.6-214</p>

			irrigated (Boro) rice Origin: Bangladesh		(0.153 ±0.112) Rain-fed: 0.047-0.535 (0.140 ±0.080) All: 0.047-0.535 (0.144 ±0.091) -Cd: Irrigated: 0.007-0.297 (0.073 ±0.069) Rain-fed: 0.007-0.189 (0.038 ±0.032) All: 0.007-0.297 (0.049 ±0.050) -Pb: Irrigated: 0.105-0.602 (0.264 ±0.125) Rain-fed: 0.063-0.353 (0.147 ±0.077) All: 0.063-0.602 (0.185 ±0.109) -Cr: Irrigated: 3.983-0.160 (1.208± 0.913) Rain-fed: 0.148-4.616 (0.986 ±0.796) All: 0.148-4.616 (1.058 ±0.836)	(57.6±36.4)) -Cd: 2.6-119 (19.7±20.0)) -Pb: 25.0-241 (74.1±43.5)) -Cr: 59.0-1846 (423±334)
Mohamed et al. (2017)	KSA	8	American, basmati, perfumed brown (Gaba), whole grain.	ICP-MS	Values in mg/kg: As: 0.02–0.07 (0.03) Cd: 0.00–0.20 (0.03) Pb: 0.00–0.13 (0.04)	Levels in µg/d: As: 8.18 Cd: 8.24 Pb: 9.36 Cr: 56.93

					Cr: 0.00–0.65 (0.23)	
Praveena & Omar (2017)	Malaysia	22	<p>Cooked rice (all samples were washed three times with deionised water prior to cooking).</p> <p>Local (Malaysia) : unpolished brown, fragrant, Parboiled, rice-5% broken, Local white, Special glutinous, Milky glutinous, Hill rice.</p> <p>Imported from: Thailand, Japan, India, Pakistan: Siam fragrant, Thai fragrant, Basmathi, white, Thai glutinous, brown, Red,</p>	ICP-OES	<p>Values in mg/kg: As: (0.087) Cr: (2.7) Cd: (0.16) Pb: (0.11)</p>	<p>Bioaccessible concentration (not exposure). Levels in mg/kg: As: (0.016) Cr: (0.11) Cd: (0.027) Pb: (0.022)</p>

			Mix grain, Ponni, Organic black Special Siam, Calrose grain, Steamed rice.			
Murphy et al. (2018)	Cambodia	102	Type: Polished, bran, brown, husk Origin: Cambodia (Preak Russey and Kendal provinces)	ICP-MS	As ($\mu\text{g}/\text{kg}$): Preak Russey: (315 \pm 150) Kendal: (158 \pm 33)	–
Pastorelli et al. (2018)	Italy	78	Italian brown, white & rice-based infant foods	Q-ICP-MS	Cd (mg/kg): Brown: (0.092 \pm 0.051) White: (0.062 \pm 0.035) Rice-based infant foods: (0.026 \pm 0.002)	Cd ($\mu\text{g}/\text{week}$): Brown: 0.16 White: 0.10 Rice-based infant foods: <u>Four months old infant:</u> 0.54 <u>Six months old:</u> 0.96 <u>Eight months old:</u> 0.89
Sharma et al. (2018)	India	13	Indian rice	F-AAS	Values in mg/kg : -Cd: 0.57-1.12 (0.99 \pm 0.05)	Levels in $\text{mg}/\text{kg}/\text{day}$:

					-Cr: 5.82-30.76 (19.98 ±2.10) -Pb: 14.80- 20.30 (17.13 ± 0.40)	-Cd: 1.87E-04- 3.68E-04 (3.24E-04 ± 1.58E- 05) -Cr: 1.91E-03- 1.01E-02 (6.57E-03 ± 6.92E- 04) -Pb: 4.87E-03- 6.67E-03 (5.63E-03 ± 1.33E- 04)
Srinuttrak et al. (2018)	Thailand	30	Sangyod Thai rice	ICP-MS	Values in mg/kg: <u>-As:</u> Bang Kaeo: 0.082±0.036 Khao Chaison: 0.099±0.039 Kong Ra: 0.084±0.020 Mueang : 0.129±0.025 Pak Phayun: 0.108±0.015 Si Banphot: 0.192±0.073 <u>-Cd :</u> Bang Kaeo: 0.031±0.041 Khao Chaison: 0.018±0.022 Kong Ra: 0.055±0.036 Mueang: 0.019±0.029 Pak Phayun: 0.022±0.031	Levels in mg/day: <u>-As:</u> Bang Kaeo: 0.019 Khao Chaison: 0.023 Kong Ra: 0.020 Mueang: 0.030 Pak Phayun: 0.025 Si Banphot: 0.045 <u>-Cd :</u> Bang Kaeo: 0.007 Khao Chaison: 0.004 Kong Ra: 0.013

					Si Banphot: 0.014±0.023	Mueang: 0.005 Pak Phayun: 0.005 Si Banphot: 0.003
Zhang et al. (2018)	China	454	Chinese (Guangzhou city)	GF-AAS	Cd: 2.6-440 (88.0 ± 8.5) µg/kg	Cd: 13.4 µg/day
Abd Rashid et al. (2019)	Malaysia	10	Type: White rice Origin: Malaysia, Thailand & Pakistan	AAS	Cd: 0.72 mg/kg Pb: 2.19 mg/kg	Levels in mg/kg bw/week: Cd: 0.013 Pb: 0.039
Wang et al. (2019)	China	990	Type: Rice and rice products Origin: China	ICP-MS	Pb: 0.0341±0.0430 mg/kg	Pb: 0.0864 µg/kg bw/day
Bielecka et al. (2020)	Poland	99	Type: Rice flakes, white, basmati, parboiled, brown, rice waffles, expanded, rice pasta, rice flour, black, red and wild.. Origin: Thailand, Italy, Pakistan, Burma, India, Cambodia, Vietnam,	ICP-MS	Values in µg/kg: -As: (123.5 ± 77.1); lowest in pasta & highest in red rice) -Cd: (25.7 ± 26.5); highest in flour & lowest in Basmati) -Pb: (37.5 ± 29.3); highest in expanded rice lowest in flakes) -Hg: (2.8 ± 2.6); highest in	Levels in mg/day: -As: 0.00062 (highest in red rice & lowest in pasta) -Cd: 0.00013 (highest is flour & lowest in basmati) -Pb: 0.00019 (highest in, red rice & lowest in

			Belgium, Brazil, USA, Canada, Bulgaria, Spain, France, Holland, and unknown.		parboiled & lowest in flour	expanded rice) -Hg: 0.00001 (highest in parboiled)
Horiguchi et al. (2020)	Japan	1168	Rice and rice products	F-AAS	Cd (mg/kg): Area A: <0.02–0.971 (0.158) Area B: 0.008–0.68 (0.109) Kiritampo: 0.070–0.102 (0.063) Glutinous rice: 0.02–0.32 (0.098) Rice cakes: 0.017–0.182 (0.069) Rice crackers: 0.017–0.263 (0.091)	Cd (µg/day): Farmers in area A: 0.1–289 (28.3) Farmers in area B: 0.1–154 (19.4)
Liu et al (2020)	Sri Lanka	165	Type: Polished rice (including white and red types) All rice types were flushed three times with ultra-pure water Origin: Sri Lanka	ICPMS	Values in mg/kg dry weight: -Cd: 0.003–0.727 (0.080 ± 0.130) -As: 0.019–0.217 (0.077 ± 0.040) -Pb: 0.001–0.345 (0.031 ± 0.050)	Levels in µg/kg bw/day: -Cd: 0.772 -iAs: 0.490 -Pb: 0.306

TatahMenta n et al. (2020)	USA	39	White (USA, Thailand, India & Italy) & brown (USA).	ICP-MS	<p>Values in $\mu\text{g}/\text{kg}$:</p> <p>White rice (USA): -As: 65–202 (129) -Pb: 0.2–32 (5.6) -Cd: 1.7–71 (11)</p> <p>-White rice (Thailand, India & Italy): -As: 58–183 (136) -Pb: 2–96 (14) -Cd: 3.1–27 (12)</p> <p>Brown rice (US): -As: 139–403 (243) -Pb: 1.4–34 (7.4) -Cd: 7.7–65 (24)</p>	-
Chu et al. (2021)	Vietnam	70	Vietnamese	F-AAS, ICP-DRC-QMS, CV-AAS based mercury analyzer	<p>Values in mg/kg:</p> <p>-As: 0.052-0.328 (0.115\pm0.049) -Cd: 0.005-0.480 (0.111\pm0.105) -Cr: <DL-1.223 (0.296\pm0.2965) -Hg: <DL-0.011 (0.007\pm0.003) -Pb: 0.003-0.177 (0.075\pm0.050)</p>	<p>Levels in mg/kg bw/day:</p> <p>-As: Men: 0.0012 Women: 0.0015 -Cd: Men:0.0011 Women: 0.0014 -Cr: Men: 0.0039 Women: 0.003 -Pb:</p>

						Men: 0.0008 Women: 0.001
Kukusamud e et al. (2021)	Thailand	55	Thai: Khaowong Kalasin sticky rice (white glutinous rice), Pka Am Pun rice (unpolished rice), Jek Chuey Sao Hai rice (polished rice), Leb Nok rice (polished rice)	ICP-MS	Values in mg/kg: <u>-As:</u> Khaowong Kalasin sticky rice: (0.16±0.06) Pka Am Pun rice: (0.19±0.09) Jek Chuey Sao Hai rice: (0.11±0.02) Leb Nok rice: (0.24±0.05) <u>-Cr:</u> Khaowong Kalasin sticky: (0.033±0.014) Pka Am Pun: (0.052±0.023) Jek Chuey Sao Hai: (0.073±0.010) Leb Nok: (0.054±0.037) <u>-Cd:</u> Khaowong Kalasin sticky: (0.024±0.022) Pka Am Pun rice: (0.021±0.017) Jek Chuey Sao Hai rice: (0.006±0.004) Leb Nok: (0.009±0.007)	Levels in µg/kg bw/week: <u>-As:</u> Khaowong Kalasin sticky rice: 0.82 (male), 0.88 (female) Pka Am Pun rice: 5.43(M), 5.81(F) Jek Chuey Sao Hai rice: 3.15 (M), 3.36 (F) Leb Nok rice: 6.86 (M), 7.34 (F) <u>-Cr:</u> Khaowong Kalasin sticky rice: 0.24 (M), 0.26 (F) Pka Am Pun rice: 2.12 (M), 2.27 (F) Jek Chuey Sao Hai rice: 2.98 (M), 3.19 (F) Leb Nok rice: 2.21

						(F), 2.36 (M) -Cd: Khaowong Kalasin sticky rice: 0.18 (M), 0.19 (F) Pka Am Pun rice: 0.86 (M), 0.92 (F) Jek Chuey Sao Hai rice: 0.25 (M), 0.26 (F) Leb Nok rice: 0.37 (M), 0.39 (F)
Lien et al. (2021)	Taiwan	1581	Taiwanese	ICP-MS	Cd: (0.04± 0.04) mg/kg	Lifetime average daily dose (LADD) of Cd: 0.06 µg/kg bw/day for male rice consumers between the ages of 19–65 years

†units are specified in each cell.

LOQ: Level of quantification; LOD: Level of detection; BDL: Below detection level; Nd: Not detectable; ICP-MS: Inductively coupled plasma mass spectrometry; GF-AAS: Graphite furnace atomic absorption spectrometry; HG-AAS: Hydride-generating atomic absorption spectrometry; HG-AFS: Hydride-generating atomic fluorescence spectrometry; F-AAS: Flame atomic absorption spectrometry; ICP-OES: Inductively coupled plasma optical emission spectrometry; Q-ICP-MS: Quadruple Inductively coupled plasma mass spectrometry; ICP- DRC-QMS: Inductively coupled plasma-dynamic reaction cell-quadrupole mass spectrometry; CV-AAS based mercury analyzer: Cold vapor atomic absorption spectrometry; DMA: Direct mercury analyzer

CHAPTER 2

AIMS AND HYPOTHESES

2.1 GAPS IN THE LITERATURE

In Lebanon, only one study was conducted in 2006 revealing dietary exposure to Cd and Pb from rice and rice products (Nasreddine et al., 2006). However, it assessed exposure from various food and drink sources, not emphasizing rice in particular, and did not specify rice types and sources. It also did not assess exposure to Cr, Hg and As.

Furthermore, until now, there have been no studies published in the United Arab Emirates assessing exposure to toxic metals in rice. Therefore, because no recent studies have been conducted in this regards in Lebanon and UAE, and no studies have assessed dietary exposure to As, Cd, Cr, Hg and Pb in rice specifically in those countries, it is necessary to conduct such a study.

2.2 RESEARCH OBJECTIVE AND SIGNIFICANCE

Knowing that rice is a popular staple food worldwide, it also has wide popularity in Lebanon and the UAE. Both countries are importers of rice rather than producers/exporters. In the UAE, the most popular rice types are basmati and jasmine, mainly imported from India and Pakistan, and to lesser extents, United States, Australia, Thailand, and the Philippines (USDA/FAS, 2021b). In Lebanon, rice is considered as an ingredient and side dish for plenty of traditional meals. Recently, due to the economic crisis that started in 2019 in Lebanon, numerous new brands of rice from various sources have been introduced to the market. This has brought many question marks regarding the safety of these newly introduced brands along with the ones already available.

Furthermore, being a fulfilling food with lower cost when compared to meat for example, many Lebanese are lately consuming more rice than before. Heavy metals are non-biodegradable and persistent, thus they can have detrimental effects on the environment and health (Zulkafflee et al., 2019). Therefore, it is extremely important to determine the presence of these toxic metals in this staple food.

Nevertheless, the objective of this study was to assess toxic metals in rice, determine the factors affecting its contamination, and evaluate the dietary exposure from rice consumption in Lebanon and UAE.

2.3 HYPOTHESIS

H1: There are high amounts of toxic metals in rice available across the Lebanese market since rice is a major source of contamination and Lebanon lacks proper authority supervision for heavy metals contamination. On the contrary, for instance, Abu Dhabi Food Control Authority (ADFCA) provides full control and inspection over rice imports in order to prevent any type containing impermissible limits of arsenic from being offered in the market (The Abu Dhabi Agriculture and Food Safety Authority [ADAFSA], 2017)

H2: In most countries, As was detected in most rice samples due to its common route of contamination, therefore, rice in Lebanon is contaminated as well, especially in the absence of supervision. This would not be the case in UAE due to the adequate supervision.

H3: Brown rice is more contaminated with heavy metals than white rice since milling whole grain rice removes the hull, germ and bran layers, thus reducing the amount of toxic metals that tends to be in higher concentrations on the outer layer of the rice grain.

H4: Toxic metals content will differ according to country of origin. Rice originating from developing countries would be more contaminated due to the absence of adequate regulations.

H5: Long rice grains will tend to have higher heavy metals content due to the larger surface area, resulting in higher accumulation from the agricultural stage.

H6: Rice packed in facilities certified with a food safety management system, such as HACCP, ISO22000:2018 and FSSC22000, will tend to have lower heavy metals content since these facilities have proper control of suppliers.

CHAPTER 3

MATERIALS AND METHODS

3.1 SAMPLE COLLECTION

The Lebanese and UAE markets were screened for white, parboiled and brown rice brands. Rice brands were purchased in their original packages, and the samples were placed in closed plastic bags and stored in the freezer until the time of experimentation.

In Lebanon, rice brands were identified following a thorough screening of the Lebanese market by nutrition graduate and undergraduate students during September 2020. The first collection of 54 packed rice brands took place during February 2021 and March 2021 from Beirut and Jbeil markets, respectively. The second collection of 53 packed rice brands took place during spring 2021 from Beirut, Jbeil and Jounieh markets. Eight brands were not found in the market during the second collection, while seven others were additionally collected.

In the UAE, the market was screened during March and July 2021. The first collection took place in March 2021, of which 63 rice brands were collected. During the second collection in summer 2021, 66 brands were collected. Twenty-four brands were not found in the market during the second collection so 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.

3.2 SAMPLE PREPARATION

The first and second sample preparation of the first and second Lebanese rice collections took place at the Lebanese American university labs during March and May 2021, respectively. The third and fourth sample preparations of both UAE rice collections occurred during September 2021. Samples were digested using a Multiwave ECO microwave digestion system equipped with a rotor for 16 vessels after adding 8 mL of 69% nitric acid and 2 mL of 30% hydrogen peroxide to each of the samples (0.5 grams). The sample digestion procedure was performed according 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 3. After microwave digestion, 2 mL HCl were added to sample solutions and the digested samples were then transferred into 50 mL flasks. 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.

Table 3. Operating conditions of microwave oven digestion

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

Spiking solutions were prepared: two solutions of 0.5 and 5 ppm concentrations of each metal with 0.5 g of rice sample in each tube (i.e. 0.5 g of rice sample with 5 µL Ars (0.5 ppm) & 0.5 g of rice sample with 50 µL Ars (5 ppm)).

All samples were diluted five times (2 ml of sample in 8 ml 3% HNO₃) prior to conducting the analysis. Two stock solutions were prepared from each metal. Initially, 1000 ppm solution of Arsenic, Lead, Chromium, Cadmium and Mercury were used.

For preparing stock solution 1 of 10,000 ppb (10 ppm) concentration, 50 μL were added from each of the initial metal solutions (1000 ppm) to 4950 μL of 3% HNO_3 each. For stock 2 of 1000 ppb, 5 μL were added from the metal solutions to 4955 μL of 3% HNO_3 . After that, standards were prepared for the calibration curve in ICP-MS. Dilutions for each metal solution were prepared to get 800, 600, 400, 200, 100, 50, 25, 10, 5, 1 and 0.5 ppb.

3.3 SAMPLE ANALYSIS

ICP-MS was used for the determination and quantification of toxic metals in rice.

Operating conditions and acquisition parameters were as illustrated in table 2. A blank tube (containing 3% HNO_3) was put at first in the analysis rack and then the metals standards tubes were placed for the formation of the calibration curve. Then, the diluted samples were placed for analysis, with a blank tube for washing. For As, Cd, Pb and Cr, washing was set to take place between every five tubes. For mercury, 2% cysteine was added to the blank tube and washing was set to take place between every three tubes.

Finally, six spiking tubes (control) were placed for comparison. The results were acquired in counts per second (cps) and were converted into concentrations based on calibration curves (Bielecka et al., 2020). Analysis was repeated twice; a different analysis followed each digestion. 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.

Table 4. 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	2.70 °C. 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	2.01 mbar
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 sec

3.4 DETERMINATION OF EXPOSURE TO HEAVY METALS FROM RICE CONSUMPTION IN LEBANON AND UAE

Three approaches are recommended by the FAO and WHO for assessing dietary exposure to food contaminants: total diet study, duplicate diet study, and selective study of individual foods (FAO and WHO, 1985). In this study, individual food (rice only) was assessed for Lebanon.

The average consumption of rice in Lebanon (g/d) was acquired from a study published by Hassan et al. (2022).

In this study, five hundred participants filled a food frequency questionnaire (FFQ); approximately 53% were females, and 47% were males. Different governorates were proportionally represented according to the number of households in each. The average consumption of cooked rice in Lebanon was 68.7 g/day, while participants' average body weight was 70 kg. In our study, we assessed dry rice but generally, 1 cup of uncooked rice will equal about 3 cups of cooked rice (University of Nebraska-Lincoln, 2022). Accordingly, 68.7g of cooked rice is equivalent to 22.9g dry rice/capita/day (0.0229 kg/capita/day).

For UAE, the assessment of the average consumption of rice per capita represented in g/day was calculated based on the total consumption of rice in the country in 2021, population, and the average weight of adults in that year (equation 1).

- Rice imports and consumption were forecasted to reach 1.15 MMT (million metric tons) in MY 2021/22 (USDA/FAS, 2021b).
- The population in the UAE in 2021 was estimated to be 9.991 million individuals

(United Nations, 2021).

- The average weight of adults in the UAE in 2021 was 76 kg (Global Obesity Observatory, 2022).

Equation 1:

Rice Consumption Per Capita in UAE in MY 2021/2022

$$\begin{aligned} &= \frac{\text{Total consumption of rice in the UAE in 2021 (kg)}}{\text{Population of UAE in 2021}} \\ &= \frac{1,150,000 \times 1000}{9,991,000} \\ &= 115 \text{ kg/year/person; equivalent to } 0.315 \text{ kg/day/person.} \end{aligned}$$

For estimating the exposure levels to toxic metals, represented as estimated daily intake (EDI), the average concentration of each toxic metal detected in the samples was multiplied by the average daily amount of rice consumption divided by the average body weight of the adult population. The calculations (Equation 2) were done according to the equation proposed by FAO (Chu et al., 2021; FAO/WHO, 2014).

Equation 2:

$$\begin{aligned} &\mathbf{EDIRice (mg/kg body weight/day)} \\ &= \frac{\text{(Concentration (mg/kg dry weight))} \times \text{Rice consumption (kg/day)}}{\text{Body weight (Kg)}} \end{aligned}$$

EDI is estimated daily intake. Estimated weekly intake (EWI) was calculated by multiplying the EDI by 7 (Chu et al., 2021).

Equation 3:

Hazard quotient was calculated from the following equation:

$$HQ = \frac{\text{Calculated Exposure}}{\text{Reference exposure}} \text{ (Mohamaed et al. 2017).}$$

Calculated exposure is estimated as daily, weekly or monthly exposure to each metal calculated by this study. Reference exposure is PTWI, BMDL or PTMI for each metal according to international guidelines. $HQ > 1$ indicates an increased risk.

CHAPTER 4

STATISTICAL ANALYSIS

Concentration values for the five different metals (As, Cd, Cr, Hg, and Pb) were obtained from lab analysis and were entered into SPSS V27 for further analysis. 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 SPSS V27. 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.

CHAPTER 5

RESULTS

5.1 CONCENTRATIONS OF HEAVY METALS IN RICE SAMPLES

The total number of samples analyzed was 236 samples (107 from Lebanon and 129 from UAE). Tables 6 and 7 show mean levels of As, Cd, Hg, and Pb in individual Lebanese and UAE rice samples, respectively. Table 8 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 9 shows the numbers and percentages of rice brands contaminated with heavy metals exceeding international limits in both countries.

5.1.1 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 (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.

European Commission regulations are only set for iAs content in rice, and the speciation of iAs was not performed in this research so comparing with EC regulations was not possible (EC, 2015).

5.1.2 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, but EC limit of 0.2 mg/kg was exceeded by the Lebanese samples (CAC, 2019; EC, 2006; LIBNOR, 2013). Of the

samples from Lebanon, 26 (25%) were above CAC limits, with 78 (73%) above EC limits. In samples from UAE, all samples were compliant with CAC limits, with only one (1%) exceeding EC limits.

5.1.3 Chromium

In 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).

5.1.4 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.

5.1.5 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 samples (10%) exceeded CAC limits for As, Cd and Pb at the same time, while 53 samples (50%) 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.

Table 5. Mean \pm standard deviation of As, Cd, Cr, Hg, and Pb in individual Lebanese rice samples in mg/kg dry weight, and limits of detection and quantification of each metal in ICP-MS

	As		Cd		Cr		Hg		Pb	
	LOD	LOQ								
	0.00005	0.0001	0.00003	0.00005	0.00003	0.0001	0.00004	0.00008	0.00004	0.00008
Sample	FC	SC								
1	0.2788 \pm 0.0391	0.3347 \pm 0.0154	0.2717 \pm 0.0063	0.1368 \pm 0.0152	0.2814 \pm 0.0157	0.3932 \pm 0.0161	0.1778 \pm 0.0038	0.1869 \pm 0.0018	0.3553 \pm 0.0101	0.3710 \pm 0.0178
2	0.2534 \pm 0.0296	0.2856 \pm 0.0187	0.3372 \pm 0.0084	0.1613 \pm 0.0049	0.4757 \pm 0.0142	0.3026 \pm 0.0135	0.1811 \pm 0.0042	0.1899 \pm 0.0087	0.2014 \pm 0.0066	0.1902 \pm 0.0168
3	0.3135 \pm 0.0165	0.1493 \pm 0.0115	0.4443 \pm 0.0021	0.3717 \pm 0.0037	0.5593 \pm 0.0199	0.2488 \pm 0.0201	0.2014 \pm 0.0019	0.2007 \pm 0.0026	0.1872 \pm 0.0023	0.0768 \pm 0.0065
4	0.2398 \pm 0.0118	0.2416 \pm 0.0058	0.4634 \pm 0.0131	0.2531 \pm 0.0129	0.5799 \pm 0.0117	0.1699 \pm 0.0043	0.1916 \pm 0.0024	0.1892 \pm 0.0057	0.3679 \pm 0.0147	0.1195 \pm 0.0166
5	0.2275 \pm 0.0084	-----	0.4055 \pm 0.0085	-----	0.5101 \pm 0.0085	-----	0.1871 \pm 0.0035	-----	0.4214 \pm 0.0102	-----
6	0.3443 \pm 0.0054	0.2069 \pm 0.0133	0.4596 \pm 0.0016	0.3148 \pm 0.0021	0.3347 \pm 0.0075	0.2116 \pm 0.0221	0.2031 \pm 0.0380	0.1973 \pm 0.0088	0.2762 \pm 0.0165	0.0658 \pm 0.0058
7	0.3391 \pm 0.0167	0.1358 \pm 0.0135	0.4139 \pm 0.0060	0.2718 \pm 0.0072	0.2285 \pm 0.0119	0.3588 \pm 0.0747	0.1501 \pm 0.0141	0.1147 \pm 0.0097	0.3215 \pm 0.0067	0.1978 \pm 0.0145
8	0.3248 \pm 0.0105	-----	0.4484 \pm 0.0092	-----	0.4922 \pm 0.0191	-----	0.2132 \pm 0.0215	-----	0.2054 \pm 0.0045	-----
9	0.4975 \pm 0.0155	-----	0.4549 \pm 0.0042	-----	0.4368 \pm 0.0117	-----	0.2038 \pm 0.0261	-----	0.3458 \pm 0.0195	-----
10	0.4554 \pm 0.0125	0.1450 \pm 0.0104	0.4375 \pm 0.0075	0.2394 \pm 0.0071	0.3137 \pm 0.0102	0.3369 \pm 0.0325	0.2897 \pm 0.0084	0.2479 \pm 0.0152	0.3154 \pm 0.0103	0.2907 \pm 0.0187

11	0.4855 ± 0.0116	0.1533 ± 0.0172	0.4230 ± 0.0062	0.1919 ± 0.0062	0.5526 ± 0.0177	0.3696 ± 0.0295	0.1399 ± 0.0076	0.1416 ± 0.0147	0.2462 ± 0.0021	0.2494 ± 0.0199
12	0.4718 ± 0.0164	0.2254 ± 0.0155	0.4596 ± 0.0014	0.2177 ± 0.0139	0.6715 ± 0.0101	0.4161 ± 0.0214	0.1965 ± 0.0062	0.1597 ± 0.0128	0.2447 ± 0.0201	0.4320 ± 0.0258
13	0.4814 ± 0.0245	0.1221 ± 0.0201	0.5007 ± 0.0062	0.1832 ± 0.0063	0.4678 ± 0.0124	0.4497 ± 0.0067	0.1915 ± 0.0145	0.1865 ± 0.0098	0.1846 ± 0.0106	0.2284 ± 0.0187
14	0.4727 ± 0.0124	0.1238 ± 0.0194	0.4451 ± 0.0019	0.1985 ± 0.0109	0.5979 ± 0.0106	0.4623 ± 0.0143	0.1517 ± 0.0164	0.1431 ± 0.0087	0.2456 ± 0.0211	0.3819 ± 0.0124
15	0.4503 ± 0.0169	0.1246 ± 0.0197	0.4408 ± 0.0074	0.1849 ± 0.0071	0.5981 ± 0.0138	0.4121 ± 0.0047	0.0954 ± 0.0154	0.1054 ± 0.0068	0.3258 ± 0.0245	0.4119 ± 0.0158
16	0.4466 ± 0.0216	0.1670 ± 0.0019	0.4521 ± 0.0098	0.1349 ± 0.0092	0.5978 ± 0.0182	0.4286 ± 0.0141	0.0454 ± 0.0124	0.0681 ± 0.0077	0.4349 ± 0.0275	0.5693 ± 0.0247
17	0.5042 ± 0.0236	0.1719 ± 0.0185	0.3129 ± 0.0053	0.1623 ± 0.0134	0.5757 ± 0.0118	0.4595 ± 0.0062	0.0418 ± 0.0078	0.0272 ± 0.0087	0.1145 ± 0.0154	0.2224 ± 0.0189
18	0.4470 ± 0.0062	0.1313 ± 0.0068	0.3021 ± 0.0104	0.2684 ± 0.0155	0.5393 ± 0.0052	0.4480 ± 0.0136	0.0732 ± 0.0099	0.0617 ± 0.0047	0.1340 ± 0.0098	0.0440 ± 0.0054
19	0.4417 ± 0.0161	0.2879 ± 0.0157	0.4517 ± 0.0154	0.1838 ± 0.0202	0.3457 ± 0.0382	0.4102 ± 0.0197	0.1053 ± 0.0081	0.0894 ± 0.0068	0.1427 ± 0.0077	0.2185 ± 0.0024
20	0.4876 ± 0.0134	-----	0.3150 ± 0.0221	-----	0.3588 ± 0.0246	-----	0.2105 ± 0.0145	-----	0.1460 ± 0.0065	-----
21	0.4845 ± 0.0112	0.1680 ± 0.0243	0.3442 ± 0.0114	0.2357 ± 0.0223	0.3285 ± 0.0149	0.3502 ± 0.0246	0.1109 ± 0.0022	0.0623 ± 0.0025	0.3319 ± 0.0124	0.3170 ± 0.0217
22	0.4568 ± 0.0182	0.1878 ± 0.0192	0.4166 ± 0.0135	0.2117 ± 0.0132	0.2376 ± 0.0209	0.4503 ± 0.0234	0.1387 ± 0.0024	0.1205 ± 0.0017	0.0971 ± 0.0125	0.3682 ± 0.0258
23	0.5055 ± 0.0113	0.1316 ± 0.0244	0.4245 ± 0.0154	0.1681 ± 0.0124	0.2937 ± 0.0257	0.4253 ± 0.0223	0.1458 ± 0.0025	0.0854 ± 0.0068	0.1418 ± 0.0155	0.4399 ± 0.0288
24	0.2563 ± 0.0106	0.1433 ± 0.0238	0.3116 ± 0.0214	0.2002 ± 0.0221	0.4374 ± 0.0244	0.4403 ± 0.0247	0.1577 ± 0.0021	0.1046 ± 0.0044	0.0119 ± 0.0124	0.2781 ± 0.0124
25	0.2422 ± 0.0124	0.1947 ± 0.0221	0.4316 ± 0.0165	0.2152 ± 0.0223	0.2301 ± 0.0161	0.4206 ± 0.0161	0.1682 ± 0.0047	0.0897 ± 0.0075	0.4022 ± 0.0241	0.4342 ± 0.0198

26	0.3151	0.1932	0.4113	0.2126	0.2233	0.3636	0.1744	0.1254	0.4237	0.4184
	± 0.0139	± 0.0229	± 0.0194	± 0.0135	± 0.0205	± 0.0205	± 0.0041	± 0.0054	± 0.0233	± 0.0222
27	0.1978	0.1652	0.4290	0.1870	0.3285	0.3904	0.2193	0.1269	0.3749	0.4247
	± 0.0124	± 0.0237	± 0.0167	± 0.0152	± 0.0261	± 0.0261	± 0.0078	± 0.0044	± 0.0145	± 0.0175
28	0.2393	0.1674	0.4437	0.1197	0.3708	0.3851	0.2486	0.1699	0.3926	0.3768
	± 0.0167	± 0.0351	± 0.0029	± 0.0207	± 0.0116	± 0.0116	± 0.0054	± 0.0032	± 0.0254	± 0.0188
29	0.2674	0.1569	0.2198	0.2649	0.2773	0.3902	0.2394	0.2506	0.3254	0.3767
	± 0.0103	± 0.0188	± 0.0154	± 0.0163	± 0.0058	± 0.0058	± 0.0088	± 0.0017	± 0.0124	± 0.0057
30	0.2316	0.1055	0.1610	0.2830	0.5251	0.4062	0.2535	0.2667	0.2897	0.3356
	± 0.0068	± 0.0097	± 0.0102	± 0.0198	± 0.0006	± 0.0062	± 0.0214	± 0.0066	± 0.0211	± 0.0065
31	0.2506	0.1770	0.2882	0.1502	0.4104	0.3778	0.2644	0.1208	0.1428	0.3521
	± 0.0046	± 0.0039	± 0.0217	± 0.0167	± 0.0251	± 0.0251	± 0.0184	± 0.0047	± 0.0144	± 0.0158
32	0.2396	0.2464	0.2281	0.3836	0.2789	0.1876	0.2247	0.0871	0.3524	0.3591
	± 0.0538	± 0.0235	± 0.0085	± 0.0196	± 0.0081	± 0.0080	± 0.0199	± 0.0024	± 0.0235	± 0.0211
33	0.3543	0.2290	0.3282	0.3691	0.2328	0.4231	0.2421	0.1254	0.2009	0.3635
	± 0.0034	± 0.0064	± 0.0033	± 0.0145	± 0.0059	± 0.0059	± 0.0204	± 0.0044	± 0.0189	± 0.0189
34	0.2788	0.2091	0.2508	0.3018	0.2158	0.4175	0.0854	0.0468	0.2346	0.3011
	± 0.0122	± 0.0041	± 0.0181	± 0.0109	± 0.0027	± 0.0276	± 0.0125	± 0.0033	± 0.0168	± 0.0174
35	0.1874	0.1648	0.2331	0.1144	0.2016	0.3373	0.0543	0.0687	0.4512	0.4180
	± 0.0031	± 0.0264	± 0.0045	± 0.0186	± 0.0083	± 0.0083	± 0.0021	± 0.0047	± 0.0214	± 0.0154
36	0.2241	0.1881	0.2217	0.2133	0.2478	0.2934	0.0684	0.0715	0.4645	0.3975
	± 0.0033	± 0.0145	± 0.0025	± 0.0045	± 0.0031	± 0.0113	± 0.0087	± 0.0054	± 0.0266	± 0.0187
37	0.1710	0.2598	0.1940	0.1601	0.2108	0.4139	0.0952	0.0959	0.3752	0.3321
	± 0.0041	± 0.0237	± 0.0023	± 0.0233	± 0.0121	± 0.0311	± 0.0054	± 0.0014	± 0.0187	± 0.0147
38	0.2215	0.1452	0.2281	0.2232	0.2188	0.4216	0.0846	0.0932	0.3781	0.3987
	± 0.0020	± 0.0056	± 0.0066	± 0.0064	± 0.0205	± 0.0058	± 0.0068	± 0.0065	± 0.0124	± 0.0183
39	0.2265	0.1297	0.2469	0.1456	0.2313	0.4257	0.1197	0.1131	0.4589	0.3407
	± 0.0059	± 0.0078	± 0.0206	± 0.022	± 0.0044	± 0.0189	± 0.0119	± 0.0037	± 0.0225	± 0.0167
40	0.1098	0.1616	0.1995	0.3125	0.2397	0.4139	0.1299	0.1392	0.1976	0.0962
	± 0.0053	± 0.0061	± 0.0089	± 0.0086	± 0.0047	± 0.0214	± 0.0107	± 0.0089	± 0.0187	± 0.0057

41	0.1560 ± 0.0036	0.1479 ± 0.0021	0.2227 ± 0.0071	0.1254 ± 0.0051	0.2094 ± 0.0025	0.3755 ± 0.0258	0.1317 ± 0.0066	0.1286 ± 0.0106	0.1524 ± 0.0099	0.0835 ± 0.0035
42	0.2997 ± 0.0015	0.3079 ± 0.0105	0.2867 ± 0.0019	0.1224 ± 0.0194	0.3301 ± 0.0045	0.1837 ± 0.0214	0.1290 ± 0.0088	0.1376 ± 0.0142	0.4442 ± 0.0266	0.1184 ± 0.0074
43	0.1996 ± 0.0041	0.3356 ± 0.0047	0.2552 ± 0.0056	0.3205 ± 0.0052	0.2356 ± 0.0037	0.1855 ± 0.0158	0.1354 ± 0.0075	0.1406 ± 0.0174	0.3385 ± 0.0189	0.2229 ± 0.0167
44	0.1946 ± 0.0052	0.3243 ± 0.0038	0.2777 ± 0.0024	0.1588 ± 0.0014	0.1481 ± 0.0025	0.1214 ± 0.0151	0.1429 ± 0.0121	0.1412 ± 0.0144	0.1989 ± 0.0124	0.0854 ± 0.0098
45	0.1984 ± 0.0076	0.2512 ± 0.0177	0.2730 ± 0.0059	0.1067 ± 0.0053	0.2525 ± 0.0042	0.1972 ± 0.0157	0.1592 ± 0.0081	0.1554 ± 0.0098	0.2840 ± 0.0168	0.3245 ± 0.0127
46	0.1293 ± 0.0022	0.1173 ± 0.0048	0.2467 ± 0.0117	0.1677 ± 0.0116	0.3090 ± 0.0113	0.2932 ± 0.0113	0.1669 ± 0.0042	0.1674 ± 0.0197	0.0925 ± 0.0147	0.1611 ± 0.0024
47	0.1510 ± 0.0032	0.2028 ± 0.0097	0.2620 ± 0.0046	0.1637 ± 0.0146	0.2264 ± 0.0134	0.2233 ± 0.0214	0.1544 ± 0.0087	0.1489 ± 0.0098	0.2234 ± 0.0124	0.1518 ± 0.0057
48	0.2747 ± 0.0103	0.1547 ± 0.0065	0.3035 ± 0.0028	0.1291 ± 0.0158	0.1701 ± 0.0187	0.3627 ± 0.0187	0.1623 ± 0.0066	0.1719 ± 0.0047	0.0588 ± 0.0095	0.1423 ± 0.0036
49	0.2100 ± 0.0218	-----	0.3004 ± 0.0052	-----	0.2014 ± 0.0289	-----	0.2145 ± 0.0098	-----	0.2882 ± 0.0047	-----
50	0.2165 ± 0.0068	0.1784 ± 0.0151	0.1904 ± 0.0026	0.3247 ± 0.0210	0.1766 ± 0.0171	0.4148 ± 0.0171	0.1681 ± 0.0204	0.1814 ± 0.0058	0.4360 ± 0.0225	0.2587 ± 0.0147
51	0.1156 ± 0.0033	0.2169 ± 0.0034	0.1873 ± 0.0071	0.4524 ± 0.0188	0.5583 ± 0.0047	0.5439 ± 0.0214	0.1672 ± 0.0147	0.1721 ± 0.0066	0.2706 ± 0.0147	0.0935 ± 0.0057
52	0.2089 ± 0.0047	-----	0.3235 ± 0.0185	-----	0.2118 ± 0.0215	-----	0.1458 ± 0.0068	-----	0.2987 ± 0.0178	-----
53	0.3242 ± 0.0028	0.1996 ± 0.0076	0.3741 ± 0.0063	0.4227 ± 0.0011	0.1892 ± 0.0241	0.2072 ± 0.0241	0.1545 ± 0.0087	0.1661 ± 0.0047	0.1987 ± 0.0147	0.2643 ± 0.0198
54	0.1637 ± 0.0051	-----	0.1648 ± 0.0011	-----	0.1564 ± 0.0215	-----	0.1658 ± 0.0057	-----	0.2687 ± 0.166	-----
55	-----	0.1346 ± 0.0049	-----	0.4057 ± 0.0021	-----	0.1105 ± 0.0058	-----	0.1369 ± 0.0055	-----	0.1215 ± 0.0087

56	-----	0.1236 ± 0.0162	-----	0.3874 ± 0.0042	-----	0.1189 ± 0.0078	-----	0.1787 ± 0.0024	-----	0.3139 ± 0.0068
57	-----	0.2275 ± 0.0036	-----	0.4345 ± 0.0061	-----	0.1656 ± 0.0064	-----	0.1715 ± 0.0036	-----	0.2097 ± 0.0087
58	-----	0.1535 ± 0.0076	-----	0.4493 ± 0.0042	-----	0.2502 ± 0.0098	-----	0.1862 ± 0.0078	-----	0.1097 ± 0.0057
59	-----	0.1503 ± 0.0041	-----	0.3314 ± 0.0088	-----	0.1147 ± 0.0052	-----	0.1756 ± 0.0058	-----	0.1391 ± 0.0072
60	-----	0.1969 ± 0.0137	-----	0.2861 ± 0.0213	-----	0.1372 ± 0.0044	-----	0.1975 ± 0.0047	-----	0.2406 ± 0.0189

FC: first collection SC: second collection LOD: limit of detection
LOQ: limit of quantification

Table 6. Mean ± standard deviation of As, Cd, Cr, Hg, and Pb in individual UAE rice samples in mg/kg fresh weight, and limits of detection and quantification of each metal in ICP-MS

	As		Cd		Cr		Hg		Pb	
	LOD	LOQ								
	0.00005	0.0001	0.00003	0.00005	0.00003	0.0001	0.00004	0.00008	0.00004	0.00008
Sample	FC	SC								
1	0.2268 ± 0.0121	0.3936 ± 0.0174	0.0307 ± 0.0097	0.0632 ± 0.0163	0.2482 ± 0.0127	0.1202 ± 0.0154	0.1294± 0.0028	0.1484 ± 0.014	0.3304 ± 0.0062	0.0841 ± 0.0058
2	0.2589 ± 0.0143	-----	0.0606 ± 0.0123	-----	0.2625 ± 0.0157	-----	0.0725 ± 0.0114	-----	0.2726 ± 0.0235	-----
3	0.1695 ± 0.0221	-----	0.0480 ± 0.0045	-----	0.3905 ± 0.0107	-----	0.0998 ± 0.0052	-----	0.2143 ± 0.0153	-----
4	0.1637 ± 0.0085	-----	0.0233 ± 0.0014	-----	0.3502 ± 0.0164	-----	0.1487 ± 0.0062	-----	0.1652 ± 0.0163	-----
5	0.1937 ± 0.0015	0.1922 ± 0.0347	0.0244 ± 0.0111	0.0343 ± 0.0052	0.4694 ± 0.0179	0.1496 ± 0.0086	0.1956 ± 0.0103	0.1378 ± 0.0087	0.2172 ± 0.0175	0.1177 ± 0.0027
6	0.2395 ± 0.0253	0.2195 ± 0.0315	0.0459 ± 0.0129	0.0648 ± 0.0088	0.3850 ± 0.0127	0.1145 ± 0.0088	0.1258 ± 0.0056	0.0649 ± 0.0063	0.4464 ± 0.0153	0.3322 ± 0.0187

7	0.1503 ± 0.0162	0.1306 ± 0.0168	0.0242 ± 0.0090	0.0265 ± 0.0055	0.5502 ± 0.0241	0.1178 ± 0.0057	0.1475 ± 0.0068	0.1108 ± 0.0104	0.2203 ± 0.0106	0.0967 ± 0.0234
8	0.4941 ± 0.0245	0.4109 ± 0.0181	0.0063 ± 0.0015	0.1427 ± 0.0142	0.3667 ± 0.0133	0.1456 ± 0.0121	0.0789 ± 0.0038	0.1529 ± 0.0112	0.2727 ± 0.0125	0.2332 ± 0.0214
9	0.1521 ± 0.0063	0.1492 ± 0.0214	0.0267 ± 0.0118	0.0801 ± 0.0245	0.3209 ± 0.0068	0.2008 ± 0.0130	0.0737 ± 0.0158	0.1123 ± 0.0158	0.3054 ± 0.0153	0.1192 ± 0.0125
10	0.1819 ± 0.0181	0.2695 ± 0.0163	0.1446 ± 0.0243	0.2318 ± 0.0177	0.3983 ± 0.0185	0.1771 ± 0.0069	0.0429 ± 0.0058	0.1667 ± 0.0132	0.2852 ± 0.0034	0.2991 ± 0.0241
11	0.2269 ± 0.0112	-----	0.0287 ± 0.0009	-----	0.2719 ± 0.0321	-----	0.0457 ± 0.0051	-----	0.1788 ± 0.0156	-----
12	0.3184 ± 0.0294	0.0715 ± 0.0158	0.0058 ± 0.0034	0.0893 ± 0.0146	0.4555 ± 0.0275	0.1364 ± 0.0342	0.1583 ± 0.0242	0.1416 ± 0.0118	0.3207 ± 0.0206	0.2535 ± 0.0157
13	0.1033 ± 0.0311	0.0836 ± 0.0218	0.0142 ± 0.0015	0.0682 ± 0.0142	0.4911 ± 0.0189	0.1541 ± 0.0021	0.1016 ± 0.0198	0.1340 ± 0.0123	0.2903 ± 0.0051	0.2797 ± 0.0182
14	0.1570 ± 0.0063	0.1471 ± 0.0088	0.0147 ± 0.0018	0.0798 ± 0.0172	0.3708 ± 0.0137	0.1217 ± 0.0055	0.1098 ± 0.0257	0.1223 ± 0.0086	0.1948 ± 0.0137	0.2787 ± 0.0237
15	0.1825 ± 0.0291	0.1661 ± 0.0281	0.0775 ± 0.0141	0.0173 ± 0.0154	0.2442 ± 0.0231	0.1324 ± 0.0042	0.1205 ± 0.0125	0.0820 ± 0.0135	0.2567 ± 0.0127	0.2653 ± 0.0134
16	0.1507 ± 0.0344	0.0858 ± 0.0128	0.0529 ± 0.0021	0.0553 ± 0.0114	0.1749 ± 0.0234	0.1716 ± 0.0172	0.1106 ± 0.0063	0.0765 ± 0.0126	0.2173 ± 0.0189	0.2544 ± 0.0214
17	0.1081 ± 0.0282	0.0612 ± 0.0065	0.0119 ± 0.0013	0.0802 ± 0.0046	0.2044 ± 0.0138	0.1631 ± 0.0183	0.1937 ± 0.0125	0.0825 ± 0.0104	0.3313 ± 0.0216	0.2777 ± 0.0249
18	0.0847 ± 0.0422	-----	0.0421 ± 0.0112	-----	0.2453 ± 0.0156	-----	0.1354 ± 0.0087	-----	0.3363 ± 0.0123	-----
19	0.1054 ± 0.0251	0.1452 ± 0.0033	0.0495 ± 0.0201	0.1184 ± 0.0089	0.2554 ± 0.0196	0.1518 ± 0.0204	0.0737 ± 0.0065	0.0952 ± 0.0156	0.3598 ± 0.0109	0.1389 ± 0.0156
20	0.2247 ± 0.0451	-----	0.0824 ± 0.0037	-----	0.2526 ± 0.0082	-----	0.1564 ± 0.0047	-----	0.3268 ± 0.0193	-----

21	0.0881 ± 0.0183	-----	0.1940 ± 0.0114	-----	0.1988 ± 0.0102	-----	0.0847 ± 0.0221	-----	0.3004 ± 0.0324	-----
22	0.2188 ± 0.0144	-----	0.0201 ± 0.0004	-----	0.2003 ± 0.0262	-----	0.0678 ± 0.0097	-----	0.5068 ± 0.0303	-----
23	0.1536 ± 0.0422	0.1169 ± 0.0034	0.0636 ± 0.0012	0.0433 ± 0.0053	0.2279 ± 0.0173	0.1577 ± 0.0141	0.1746 ± 0.0126	0.1928 ± 0.0129	0.4339 ± 0.0131	0.3992 ± 0.0151
24	0.0915 ± 0.0591	-----	0.0454 ± 0.0174	-----	0.2159 ± 0.0278	-----	0.0982 ± 0.0154	-----	0.4727 ± 0.0135	-----
25	0.1231 ± 0.0117	0.1272 ± 0.0069	0.0373 ± 0.0021	0.0651 ± 0.0051	0.2864 ± 0.0126	0.1942 ± 0.0172	0.1857 ± 0.0192	0.2004 ± 0.0105	0.4415 ± 0.0165	0.1871 ± 0.0148
26	0.2295 ± 0.0148	0.2031 ± 0.0128	0.0450 ± 0.0094	0.0657 ± 0.0052	0.2268 ± 0.0103	0.1817 ± 0.0211	0.2219 ± 0.0214	0.2381 ± 0.0114	0.5209 ± 0.0214	0.1855 ± 0.0135
27	0.1809 ± 0.0021	0.1662 ± 0.0162	0.0278 ± 0.0064	0.0746 ± 0.0152	0.2629 ± 0.0123	0.1384 ± 0.0178	0.2239 ± 0.0154	0.2378 ± 0.0098	0.3037 ± 0.0165	0.1085 ± 0.0105
28	0.1480 ± 0.0143	0.1176 ± 0.0211	0.0533 ± 0.0238	0.0737 ± 0.0065	0.3596 ± 0.0124	0.1351 ± 0.0187	0.1476 ± 0.0061	0.2183 ± 0.0109	0.3015 ± 0.0125	0.1013 ± 0.0148
29	0.2506 ± 0.0147	0.3842 ± 0.0141	0.0145 ± 0.0062	0.0689 ± 0.0047	0.2343 ± 0.0111	0.1898 ± 0.0085	0.0981 ± 0.0029	0.2196 ± 0.0096	0.2830 ± 0.0072	0.1489 ± 0.0088
30	0.1905 ± 0.0121	0.1135 ± 0.0091	0.1296 ± 0.0167	0.0646 ± 0.0178	0.3360 ± 0.0107	0.1823 ± 0.0131	0.1235 ± 0.0214	0.2294 ± 0.0091	0.3044 ± 0.0193	0.2634 ± 0.0231
31	0.2641 ± 0.0198	0.2041 ± 0.0048	0.0243 ± 0.0079	0.0730 ± 0.0148	0.2504 ± 0.0124	0.1912 ± 0.0135	0.2151 ± 0.0312	0.2295 ± 0.0101	0.4058 ± 0.0165	0.2843 ± 0.0227
32	0.1523 ± 0.0125	-----	0.1124 ± 0.0179	-----	0.3157 ± 0.0128	-----	0.1897 ± 0.0105	-----	0.4959 ± 0.0132	-----
33	0.1536 ± 0.0143	0.1343 ± 0.0059	0.0563 ± 0.0048	0.0645 ± 0.0164	0.2839 ± 0.0139	0.0822 ± 0.0292	0.1446 ± 0.0109	0.2324 ± 0.0125	0.2632 ± 0.0187	0.2959 ± 0.0158
34	0.1601 ± 0.0112	0.1086 ± 0.0157	0.0473 ± 0.0145	0.0415 ± 0.0147	0.3225 ± 0.0135	0.1082 ± 0.0268	0.1108 ± 0.0065	0.1913 ± 0.0089	0.2047 ± 0.0118	0.3451 ± 0.0214

35	0.1385 ± 0.0122	-----	0.0565 ± 0.0067	-----	0.2750 ± 0.0125	-----	0.1987 ± 0.0062	-----	0.1637 ± 0.0104	-----
36	0.2314 ± 0.0117	0.1786 ± 0.0171	0.0721 ± 0.0062	0.1247 ± 0.0094	0.3501 ± 0.0153	0.1188 ± 0.0108	0.1357 ± 0.0328	0.2154 ± 0.079	0.2028 ± 0.0022	0.1740 ± 0.0187
37	0.2189 ± 0.0311	0.1315 ± 0.0041	0.0666 ± 0.0048	0.0486 ± 0.0056	0.2957 ± 0.0159	0.1755 ± 0.0072	0.2314 ± 0.0214	0.2451 ± 0.098	0.1630 ± 0.0145	0.2509 ± 0.0142
38	0.2335 ± 0.0163	-----	0.1456 ± 0.0134	-----	0.2527 ± 0.0113	-----	0.0789 ± 0.0147	-----	0.2035 ± 0.0168	-----
39	0.2521 ± 0.0011	0.1843 ± 0.0326	0.0234 ± 0.0107	0.1231 ± 0.0073	0.3103 ± 0.0165	0.1702 ± 0.0083	0.1146 ± 0.0264	0.2156 ± 0.0106	0.1554 ± 0.0128	0.1903 ± 0.0161
40	0.2227 ± 0.0283	0.1589 ± 0.0182	0.0460 ± 0.0080	0.1060 ± 0.0176	0.3396 ± 0.0136	0.1227 ± 0.0191	0.1186 ± 0.0136	0.2294 ± 0.0125	0.0946 ± 0.0014	0.0801 ± 0.0099
41	0.2572 ± 0.0259	0.1761 ± 0.0251	0.0285 ± 0.0031	0.1109 ± 0.0164	0.3537 ± 0.0143	0.1243 ± 0.0118	0.1124 ± 0.0114	0.2012 ± 0.0162	0.2070 ± 0.0154	0.1588 ± 0.0145
42	0.6215 ± 0.0139	-----	0.0172 ± 0.0014	-----	0.3136 ± 0.0153	-----	0.1985 ± 0.0089	-----	0.1637 ± 0.0191	-----
43	0.2199 ± 0.0027	0.1582 ± 0.0271	0.0535 ± 0.0132	0.1175 ± 0.0085	0.3290 ± 0.0113	0.1623 ± 0.0097	0.1152 ± 0.0064	0.2325 ± 0.0101	0.0779 ± 0.0149	0.1059 ± 0.0091
44	0.1648 ± 0.0125	-----	0.0347 ± 0.0028	-----	0.3785 ± 0.0161	-----	0.1074 ± 0.0097	-----	0.1399 ± 0.0056	-----
45	0.1618 ± 0.0019	0.1401 ± 0.0164	0.0329 ± 0.0027	0.1403 ± 0.0169	0.3461 ± 0.0137	0.1019 ± 0.0235	0.1056 ± 0.0235	0.2014 ± 0.0123	0.1656 ± 0.0032	0.2989 ± 0.0116
46	0.1748 ± 0.0065	0.1706 ± 0.0214	0.0571 ± 0.0095	0.1198 ± 0.0114	0.3735 ± 0.0114	0.1936 ± 0.0102	0.2242 ± 0.0106	0.2435 ± 0.0084	0.1258 ± 0.0255	0.1827 ± 0.0107
47	0.2487 ± 0.0041	-----	0.0313 ± 0.0189	-----	0.3324 ± 0.0097	-----	0.2145 ± 0.0215	-----	0.1581 ± 0.0118	-----
48	0.1168 ± 0.0112	-----	0.0548 ± 0.0268	-----	0.4406 ± 0.0214	-----	0.2014 ± 0.0194	-----	0.2015 ± 0.0064	-----

49	0.0735 ± 0.0076	0.0729 ± 0.0226	0.0203 ± 0.0031	0.1499 ± 0.0089	0.4353 ± 0.0135	0.0822 ± 0.0118	0.1547 ± 0.0084	0.2292 ± 0.0077	0.1489 ± 0.0045	0.1263 ± 0.0173
50	0.3589 ± 0.0235	-----	0.0252 ± 0.0011	-----	0.4756 ± 0.0161	-----	0.2014 ± 0.0141	-----	0.1437 ± 0.0079	-----
51	0.0844 ± 0.0071	-----	0.0841 ± 0.0115	-----	0.4030 ± 0.0231	-----	0.1897 ± 0.0054	-----	0.1591 ± 0.0203	-----
52	0.1250 ± 0.0311	-----	0.0273 ± 0.0008	-----	0.4932 ± 0.0384	-----	0.1745 ± 0.0268	-----	0.2107 ± 0.0168	-----
53	0.3131 ± 0.0211	0.2748 ± 0.0091	0.0522 ± 0.0035	0.1321 ± 0.0092	0.5059 ± 0.0127	0.0904 ± 0.0154	0.1787 ± 0.0251	0.2348 ± 0.0106	0.2307 ± 0.0192	0.1643 ± 0.0154
54	0.2012 ± 0.0091	0.2214 ± 0.0142	0.0342 ± 0.0053	0.1069 ± 0.0049	0.2315 ± 0.0158	0.1941 ± 0.0209	0.0918 ± 0.0157	0.2314 ± 0.0115	0.2643 ± 0.0102	0.2050 ± 0.0184
55	0.2797 ± 0.0324	0.2904 ± 0.0181	0.0127 ± 0.0037	0.1047 ± 0.0154	0.2371 ± 0.015	0.2057 ± 0.0219	0.1248 ± 0.0109	0.2367 ± 0.0107	0.2962 ± 0.0147	0.1699 ± 0.0192
56	0.2431 ± 0.0214	-----	0.0434 ± 0.0043	-----	0.2908 ± 0.0189	-----	0.1784 ± 0.0187	-----	0.2637 ± 0.0133	-----
57	0.2474 ± 0.0211	0.1380 ± 0.0129	0.1497 ± 0.0079	0.0131 ± 0.0187	0.3407 ± 0.0153	0.1679 ± 0.0122	0.2348 ± 0.0128	0.2145 ± 0.0123	0.2714 ± 0.0106	0.2147 ± 0.0182
58	0.3395 ± 0.0312	0.2510 ± 0.0223	0.1887 ± 0.0054	0.0715 ± 0.0192	0.1558 ± 0.0201	0.1226 ± 0.0042	0.2247 ± 0.0269	0.2378 ± 0.0104	0.2880 ± 0.0118	0.1871 ± 0.0196
59	0.0867 ± 0.0064	-----	0.0226 ± 0.0066	-----	0.3203 ± 0.0139	-----	0.2041 ± 0.0125	-----	0.2395 ± 0.0175	-----
60	0.1429 ± 0.0158	-----	0.0497 ± 0.0147	-----	0.2862 ± 0.0229	-----	0.1874 ± 0.0087	-----	0.2564 ± 0.0198	-----
61	0.1982 ± 0.0041	-----	0.0545 ± 0.0056	-----	0.2440 ± 0.0102	-----	0.1997 ± 0.0051	-----	0.2412 ± 0.0139	-----
62	0.1851 ± 0.0311	-----	0.0566 ± 0.0092	-----	0.2048 ± 0.0092	-----	0.2014 ± 0.0054	-----	0.3097 ± 0.0103	-----

63	0.2439 ± 0.0021	0.1098 ± 0.0191	0.1914 ± 0.0165	0.1203 ± 0.0027	0.1833 ± 0.0219	0.1772 ± 0.0032	0.2315 ± 0.0061	0.2154 ± 0.0088	0.1756 ± 0.0135	0.0588 ± 0.0233
64	-----	0.2048 ± 0.0067	-----	0.0956 ± 0.0182	-----	0.1374 ± 0.0086	-----	0.2301 ± 0.0112	-----	0.0693 ± 0.0254
65	-----	0.1453 ± 0.0154	-----	0.1292 ± 0.0219	-----	0.1605 ± 0.0055	-----	0.1512 ± 0.0103	-----	0.1821 ± 0.0235
66	-----	0.1712 ± 0.0167	-----	0.0695 ± 0.0046	-----	0.1557 ± 0.0168	-----	0.2238 ± 0.0083	-----	0.1594 ± 0.0168
67	-----	0.2141 ± 0.0267	-----	0.1232 ± 0.0182	-----	0.1953 ± 0.0064	-----	0.2423 ± 0.0119	-----	0.1645 ± 0.0145
68	-----	0.0889 ± 0.0334	-----	0.0739 ± 0.0042	-----	0.1133 ± 0.0179	-----	0.2249 ± 0.0121	-----	0.1037 ± 0.0217
69	-----	0.1115 ± 0.0432	-----	0.0996 ± 0.0035	-----	0.3421 ± 0.0202	-----	0.2354 ± 0.0189	-----	0.3528 ± 0.0214
70	-----	0.1379 ± 0.0321	-----	0.0931 ± 0.0132	-----	0.1544 ± 0.0029	-----	0.1126 ± 0.0105	-----	0.3294 ± 0.0125
71	-----	0.2474 ± 0.0114	-----	0.1167 ± 0.0118	-----	0.1565 ± 0.0112	-----	0.1187 ± 0.0116	-----	0.2365 ± 0.0161
72	-----	0.1520 ± 0.0391	-----	0.0941 ± 0.0111	-----	0.1532 ± 0.0104	-----	0.2402 ± 0.0091	-----	0.3105 ± 0.0190
73	-----	0.0612 ± 0.0051	-----	0.0908 ± 0.0151	-----	0.1378 ± 0.0256	-----	0.2126 ± 0.0137	-----	0.2274 ± 0.0051
74	-----	0.0651 ± 0.0035	-----	0.0895 ± 0.0022	-----	0.1875 ± 0.0242	-----	0.2358 ± 0.0124	-----	0.1732 ± 0.0290
75	-----	0.1758 ± 0.0214	-----	0.0148 ± 0.0087	-----	0.1672 ± 0.0144	-----	0.1433 ± 0.0071	-----	0.1672 ± 0.0127
76	-----	0.1258 ± 0.0298	-----	0.0604 ± 0.0127	-----	0.1994 ± 0.0006	-----	0.1784 ± 0.0114	-----	0.1993 ± 0.0021

77	-----	0.4508 ± 0.0218	-----	0.1203 ± 0.0077	-----	0.1569 ± 0.0185	-----	0.1473 ± 0.0102	-----	0.1569 ± 0.0245
78	-----	0.1463 ± 0.0089	-----	0.0011 ± 0.0076	-----	0.1174 ± 0.0112	-----	0.1650 ± 0.0089	-----	0.2970 ± 0.0302
79	-----	0.3001 ± 0.0157	-----	0.1201 ± 0.0024	-----	0.1301 ± 0.0063	-----	0.1583 ± 0.0087	-----	0.2259 ± 0.0165
80	-----	0.3007 ± 0.0463	-----	0.0215 ± 0.0114	-----	0.1771 ± 0.0061	-----	0.1636 ± 0.0124	-----	0.2883 ± 0.0179
81	-----	0.2361 ± 0.0243	-----	0.1323 ± 0.0014	-----	0.1787 ± 0.0034	-----	0.2271 ± 0.0103	-----	0.2942 ± 0.0187
82	-----	0.1113 ± 0.0094	-----	0.0990 ± 0.0109	-----	0.1518 ± 0.0077	-----	0.1713 ± 0.0118	-----	0.3030 ± 0.0109
83	-----	0.1229 ± 0.0220	-----	0.1004 ± 0.0025	-----	0.1542 ± 0.0092	-----	0.1687 ± 0.0126	-----	0.3176 ± 0.0185
84	-----	0.1458 ± 0.0037	-----	0.0275 ± 0.0026	-----	0.1497 ± 0.0231	-----	0.1828 ± 0.0107	-----	0.2826 ± 0.0269
85	-----	0.1227 ± 0.0057	-----	0.0345 ± 0.0091	-----	0.1132 ± 0.0211	-----	0.1967 ± 0.0101	-----	0.2711 ± 0.0141
86	-----	0.0641 ± 0.0078	-----	0.0902 ± 0.0126	-----	0.1419 ± 0.0233	-----	0.1713 ± 0.0089	-----	0.2525 ± 0.0126
87	-----	0.2843 ± 0.0268	-----	0.0836 ± 0.0104	-----	0.1821 ± 0.0214	-----	0.2371 ± 0.0108	-----	0.2514 ± 0.0215
88	-----	0.1114 ± 0.0127	-----	0.0887 ± 0.0184	-----	0.1663 ± 0.0254	-----	0.2415 ± 0.0103	-----	0.2295 ± 0.0276
89	-----	0.0638 ± 0.0097	-----	0.0606 ± 0.0015	-----	0.1385 ± 0.0109	-----	0.2457 ± 0.0114	-----	0.3133 ± 0.0105
90	-----	0.0706 ± 0.0197	-----	0.0771 ± 0.0059	-----	0.1087 ± 0.0214	-----	0.2417 ± 0.0098	-----	0.2587 ± 0.0056

FC: first collection

SC: second collection

LOD: limit of detection

LOQ: limit of quantification

Table 7. 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	0.1055	0.06
	Maximum	0.5055	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	0.0822
	Maximum	0.67	0.5502
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

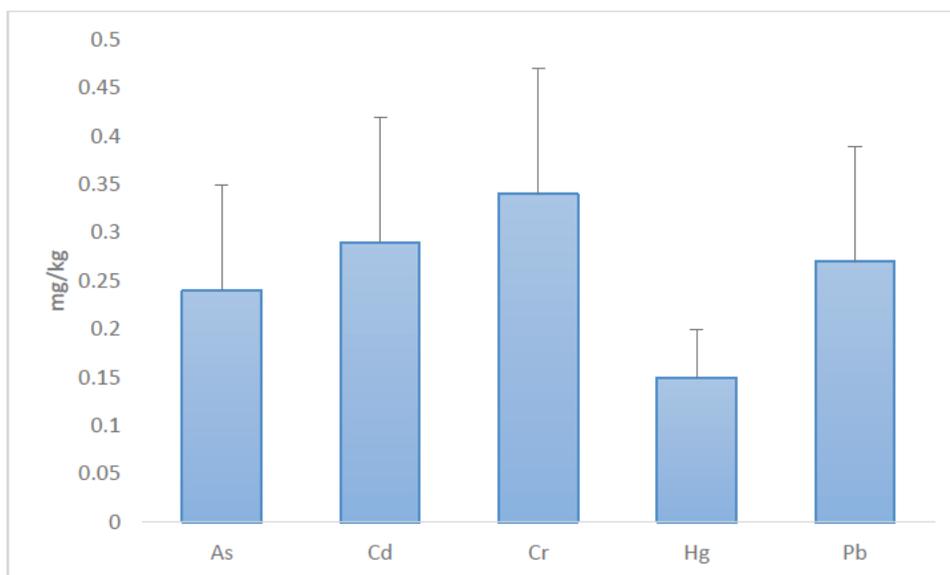


Figure 3. Mean Concentrations of Toxic Metals in the rice samples from Lebanon

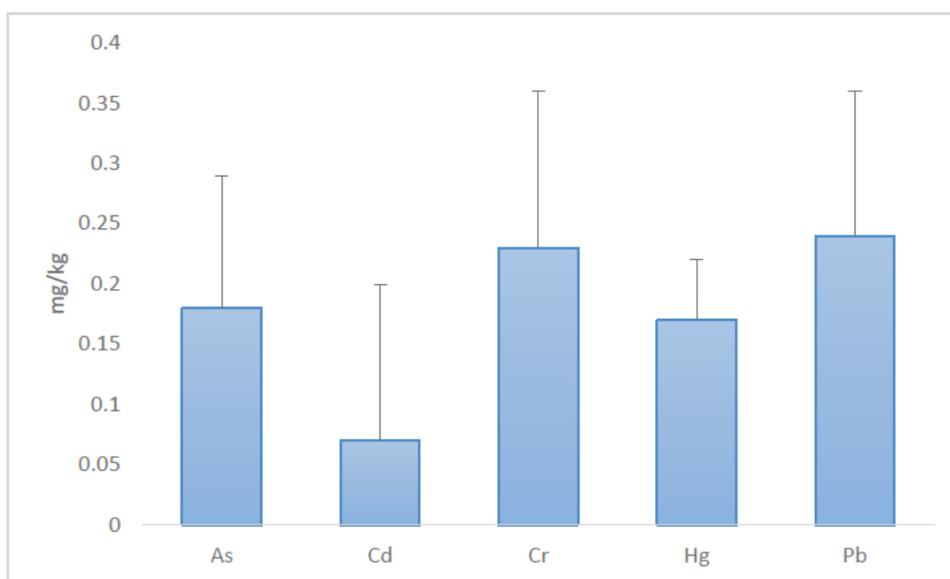


Figure 4. Mean Concentrations of Toxic Metals in the rice samples from UAE

Table 8. Numbers and percentages of rice samples from Lebanon and UAE that are contaminated with toxic metals exceeding international limits

	Lebanon		UAE	
	n (%) of brands exceeding CAC limits	n (%) of brands exceeding EC limits	n (%) of brands exceeding CAC limits	n (%) of brands exceeding 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

5.2 EFFECT OF DIFFERENT VARIABLES ON HEAVY METALS' LEVELS IN RICE

Effect of different variables on the heavy metals contamination of rice in Lebanon and UAE are presented in tables 10 and 11, respectively.

5.2.1 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 ± 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 ($p < 0.001$).

5.2.2 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 statistically 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 ($p = 0.008$).

5.2.3 Chromium

In the samples from Lebanon, there was no 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) ($p = 0.006$). 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 ($p<0.001$).

5.2.4 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 ($p<0.001$).

5.2.5 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 ($p=0.004$ and $p=0.001$, respectively).

Table 9. Effect of different independent variables on heavy metals levels in rice samples collected from Lebanon

Independent variable	As			Cd			Cr			Hg			Pb		
	N	Mean ±Sd ^a	p-value	N	Mean ±Sd	p-value									
Packing season															
Fall/winter	62	0.25±0.12		62	0.27±0.11		62	0.35±0.13		62	0.15±0.06		62	0.28±0.13	
Spring/Summer	23	0.22±0.10		23	0.33±0.11		23	0.33±0.13		23	0.15±0.05		23	0.28±0.11	
Not known	21	0.22±0.08	0.341	21	0.29±0.11	0.147	21	0.34±0.14	0.796	21	0.16±0.05	0.637	21	0.24±0.11	0.445
Country of origin															
Developed	75	0.27±0.12		75	0.30±0.11		75	0.36±0.14		75	0.15±0.06		75	0.27±0.12	
Developing	23	0.19±0.05		23	0.24±0.09		23	0.31±0.10		23	0.14±0.05		23	0.28±0.12	
Not available	9	0.21±0.07	<.001	9	0.31±0.10	0.055	9	0.23±0.08	0.006	9	0.14±0.05	0.437	9	0.29±0.10	0.906
FSMS^b															
Yes	35	0.22±0.11		35	0.30±0.11		35	0.37±0.14		35	0.16±0.05		35	0.28±0.12	
No/Not known	72	0.26±0.11	0.098	72	0.28±0.11	0.536	72	0.32±0.13	0.078	72	0.14±0.06	0.113	72	0.27±0.12	0.785
Grain size															
Long	71	0.26±0.12		71	0.31±0.11		71	0.37±0.14		71	0.16±0.06		71	0.28±0.12	
Short/medium	36	0.21±0.06	0.002	36	0.23±0.08	<.001	36	0.27±0.09	<.001	36	0.13±0.04	0.012	36	0.26±0.13	0.403
Type															
White	41	0.22±0.08		41	0.25±0.10		41	0.29±0.11		41	0.14±0.04		41	0.26±0.12	
Parboiled	55	0.20±0.13		55	0.30±0.11		55	0.28±0.12		55	0.16±0.07		55	0.26±0.12	
Brown	11	0.27±0.05	0.02	11	0.32±0.10	0.054	11	0.39±0.15	<.001	11	0.17±0.02	0.019	11	0.29±0.09	0.436
Time between packing and purchasing															

1 to 9 weeks	22	0.27± 0.12		22	0.28± 0.09		22	0.25± 0.06		22	0.16± 0.06		22	0.25± 0.10	
10 to 19 weeks	18	0.21± 0.10		18	0.26± 0.12		18	0.34± 0.11		18	0.13± 0.05		18	0.29± 0.14	
20 to 29 weeks	16	0.27± 0.12		16	0.28± 0.11		16	0.33± 0.14		16	0.14± 0.05		16	0.29± 0.12	
30 weeks and above	29	0.26± 0.12	0.362	29	0.32± 0.11	0.314	29	0.41± 0.13	<.001	29	0.16± 0.06	0.16 6	29	0.28± 0.13	0.647
Lot number for common brands between both collections															
Collection 1	47	0.30± 0.12		47	0.33± 0.10		47	0.35± 0.15		47	0.16± 0.06		47	0.27± 0.12	
Collection 2	47	0.19± 0.06	<.001	47	0.22± 0.09	<.001	47	0.36± 0.10	0.67	47	0.14± 0.05	0.00 1	47	0.28± 0.13	0.775

^a Standard deviation

^b Food safety management system certificate

Table 10. Effect of different independent variables on heavy metals levels in rice samples collected from UAE

Independent variable	As			Cd			Cr			Hg			Pb		
	N	Mean ±Sd ^a	p-value	N	Mean ±Sd	p-value	N	Mean Sd	p-value	N	Mean Sd	p-value	N	Mean Sd	p-value
Packing season															
Fall/winter	75	0.20± 0.1		75	0.07± 0.05		75	0.24± 0.10		75	0.17± 0.05		75	0.25 ± 0.10	
Spring/Summer	51	0.16± 0.07	0.011	51	0.07± 0.04	0.627	51	0.23± 0.12	0.71 7	51	0.16± 0.06	0.114	51	0.23 ± 0.08	0.458
Country of origin															
Developed	113	0.18± 0.092		11 3	0.07± 0.04		113	0.23± 0.11		113	0.17± 0.06		11 3	0.24 ± 0.09	
Developing	14	0.24± 0.06	0.016	14	0.08± 0.06	0.356	14	0.23± 0.08	0.99 3	14	0.16± 0.06	0.786	14	0.23 ± 0.08	0.534
FSMS^b															

Yes	60	0.18± 0.08		60	0.06± 0.04		60	0.24± 0.11		60	0.17± 0.06		60	0.24 ± 0.09	
No/Not known	67	0.19 0.10	0.382	67	0.07± 0.05	0.073	67	0.23± 0.11	0.46 9	67	0.17± 0.06	0.801	67	0.24 ± 0.10	0.661
Grain size															
Long	76	0.19± 0.09		76	0.07± 0.04		76	0.23± 0.10		76	0.16± 0.06		76	0.24 ± 0.10	
Short /medium	51	0.18 0.10	0.857	51	0.07± 0.05	0.485	51	0.24± 0.11	0.78	51	0.18± 0.06	0.251	51	0.25 ± 0.08	0.428
Type															
White/ Parboiled	98	0.19± 0.08		98	0.07± 0.04		98	0.24± 0.11		98	0.17± 0.05		98	0.24 ± 0.09	
Brown	29	0.18 0.12	0.899	29	0.07± 0.05	0.439	29	0.23± 0.11	0.81 7	29	0.16± 0.07	0.392	29	0.23 ± 0.10	0.68
Time between packing and purchasing															
1 to 9 weeks	9	0.14± 0.10		9	0.08± 0.06		9	0.28± 0.13		9	0.16± 0.05		9	0.30 ± 0.09	
10 to 19 weeks	48	0.19± 0.11		48	0.06± 0.03		48	0.23± 0.10		48	0.17± 0.06		48	0.23 ± 0.10	
20 to 29 weeks	29	0.19± 0.07		29	0.07± 0.04		29	0.20± 0.07		29	0.18± 0.06		29	0.24 ± 0.08	
30 weeks and above	40	0.18± 0.08	0.568	40	0.07± 0.05	0.659	40	0.26± 0.12	0.07 2	40	0.16± 0.05	0.182	40	0.24 ± 0.09	0.266
Lot number for common brands between both collections															
Collection 1	39	0.21± 0.08		39	0.05± 0.05		39	0.32± 0.09		39	0.15± 0.05		39	0.27 ± 0.10	

Collection 2	39	0.18± 0.08	0.008	39	0.09± 0.04	0.001	39	0.15± 0.03	<.00 1	39	0.18± 0.06	<.001	39	0.20 ± 0.08	0.001
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^a Standard deviation

^b Food safety management system certificate

5.3 EXPOSURE TO HEAVY METALS FROM RICE CONSUMPTION

5.3.1 Arsenic

Based on the equations explained in the methodology section, the calculated EDI of As from rice consumption in Lebanon was 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.

5.3.2 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).

5.3.3 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= 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.

5.3.4 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.

5.3.5 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.

CHAPTER 6

DISCUSSION

To our knowledge, this is the first study in Lebanon and the UAE assessing 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.

6.1 CONCENTRATIONS OF HEAVY METALS IN RICE FROM LEBANON AND UAE

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

6.1.1 Arsenic

As has widely attracted the attention of investigators of toxic metals in rice worldwide due to its widespread contamination in groundwater (Shraim, 2014). Our study has shown that the mean level of As in rice products available in the Lebanese market was found to be 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 123.5 ± 77.1 μ g/kg (0.1235 ± 0.0771 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 (Srinuttrak 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 Preah 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 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 & 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 m/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). These results are not surprising as brown rice is more contaminated with arsenic due to its germ layer that retains higher amounts of the metal (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 Taha Mentan et al. (2020), as US white rice was more contaminated with As than white rice from Thailand and India. Shraim (2014) 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).

6.1.2 Cadmium

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. in 2006, the reported level of Cd of 0.6 µg/kg (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.

The Lebanese samples 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 our Lebanese samples 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 area A (Odate city) and area B (Kazuno city and Kosaka town) in 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 $\mu\text{g}/\text{kg}$ (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 reported 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), assessing domestic and imported rice in Iran. They 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 in the Lebanese samples ($p = 0.054$), 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.

6.1.3 Chromium

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, such as 0.23 ± 0.01 mg/kg in Arborio, 0.23 ± 0.02 mg/kg in Carnaroli, 0.20 ± 0.01 mg/kg in Ribe/Roma parboiled, and 0.28 ± 0.03 mg/kg Vialone Nano. 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 ($p<0.001$). These findings were supported, in part, by Pinto et al. (2016) through assessing Cr in local and imported rice collected from

Portuguese and Spanish markets. The authors stated that the obtained values of Cr in brown and wild rice were 0.10 ± 0.05 and 0.13 ± 0.08 mg/kg, respectively, which were higher than 0.08 ± 0.03 mg/kg in parboiled rice, but Cr content of brown rice was slightly less than that of 0.11 ± 0.05 mg/kg in white rice. 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 we have 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.

6.1.4 Mercury

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. 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.

6.1.5 Lead

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 al 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 $\mu\text{g/kg}$ 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 in a small sample of Malaysian, Thai and Pakistani white rice. No statistical significance was apparent for the effect of any of the variables on Pb content of samples collected from Lebanon and UAE.

6.2 EXPOSURE OF THE LEBANESE AND UAE POPULATIONS TO HEAVY METALS FROM RICE

According to our calculations, UAE rice consumption in 2021/2022 MY was 315 g/capita/day. On the other hand, according to Hassan et al, (2022), the average rice consumption in Lebanon in 2021 was estimated to be 22.9 g/capita/day. Table 12 shows the exposure levels of the Lebanese and UAE populations to heavy metals from rice in comparison to international safe levels of exposure.

6.2.1 Arsenic

According to JECFA (2011), the previously set PTWI of 2.1 $\mu\text{g}/\text{kg}$ bw/day was withdrawn since it was observed that it was in the same range of $\text{BMDL}_{0.5}$ (benchmark dose lower confidence limit of 2-7 $\mu\text{g}/\text{kg}$ bw/day from inorganic As) for a 0.5% increased risk for lung cancer. According to EFSA CONTAM panel (2009), the BMDL_{01} for a 1% increased risk for cancers of the lung, skin and bladder, and skin lesions from As is 0.3-8 $\mu\text{g}/\text{kg}$ bw/day. EDI in Lebanon was 0.08 $\mu\text{g}/\text{kg}$ bw/day, with a hazard quotient of 0.01-0.27 (according to EFSA BMDL_{01}), indicating that EDI of the Lebanese population is less than BMDL_{01} proposed by EFSA. In contrast, UAE results indicate an unsafe level of exposure equivalent to 0.76 $\mu\text{g}/\text{kg}$ bw/day, ranging within BMDL_{01} 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 BMDL_{01} 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 BMDL_{01} (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 ($0.76 \mu\text{g}/\text{kg bw}/\text{day}$, equivalent to $57.76 \mu\text{g}/\text{day}$) is comparable to the findings of Jahiruddin et al. (2017) reporting the As exposure of Bengali population as $57.6 \pm 36.4 \mu\text{g}/\text{day}$, but less than levels reported by Chu et al. (2021) (0.0012 and $0.0015 \text{ mg}/\text{kg bw}/\text{day}$ 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 ($8.18 \mu\text{g}/\text{day}$) than the one we calculated for the UAE ($57.76 \mu\text{g}/\text{day}$), but was comparable to the Lebanese exposure of $5.6 \mu\text{g}/\text{day}$.

6.2.2 Cadmium

JECFA withdrew its previously established PTWI of $7 \mu\text{g}/\text{kg bw}$ due to Cd's long half-life of 15 years in human kidneys, so a monthly value (PTMI) of $25 \mu\text{g}/\text{kg bw}$ was considered more appropriate (JECFA, 2013). The EFSA CONTAM panel (2011) has set a value of $2.5 \mu\text{g}/\text{kg b.w}$ for PTWI for Cd. The calculated exposure level to Cd from rice in Lebanon was $2.7 \mu\text{g}/\text{kg bw}/\text{month}$ or $0.63 \mu\text{g}/\text{kg bw}/\text{week}$, with $\text{HQ} = 0.108$ according to JECFA PTMI, and $\text{HQ} = 0.252$ 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 $0.15 \mu\text{g}/\text{kg bw}/\text{day}$, equivalent to $4.5 \mu\text{g}/\text{kg bw}/\text{month}$ or $1.05 \mu\text{g}/\text{kg bw}/\text{week}$. 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.

6.2.3 Chromium

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).

6.2.4 Mercury

PTWI for Hg is 4 µg/kg bw according to JECFA and EFSA CONTAM (JECFA, 2011; 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).

6.2.5 Lead

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, 2011). 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 $\mu\text{g}/\text{kg}$ bw/day could be a cause of concern. However, according to EFSA, and with $\text{HQ} = 0.059$, this level of exposure is less than BMDL_{01} 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 $\mu\text{g}/\text{kg}$ bw/day and $\text{HQ} = 0.67$. 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 $\mu\text{g}/\text{day}$, 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 $\mu\text{g}/\text{day}$) 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 $\mu\text{g}/\text{day}$) 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 $\mu\text{g}/\text{kg}$ 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. in Bangladesh (2017) (74.1 \pm 43.5 $\mu\text{g}/\text{day}$) and Orisakwe et al. (2012) in Nigeria (0.3775 g/day).

Table 11. Exposure levels of the Lebanese and UAE populations to heavy metals from rice in comparison to international safe levels of exposure

	Lebanon Exposure Levels ($\mu\text{g}/\text{kg bw}$) (Hazard Quotient)	UAE Exposure Levels ($\mu\text{g}/\text{kg bw}$) (Hazard Quotient)	Safe Levels of Exposure According to FAO/WHO ($\mu\text{g}/\text{kg bw}$)	Safe Levels of Exposure According to European Food Safety Authority ($\mu\text{g}/\text{kg bw}$)
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 $\mu\text{g}/\text{day}$ (0.03)	72.5 $\mu\text{g}/\text{day}$ (0.29)	250 $\mu\text{g}/\text{day}$	250 $\mu\text{g}/\text{day}$
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.

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). Pb may disrupt the development of the nervous system, especially during the prenatal period throughout childhood (Mason et al., 2014). According to WHO (2021), 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, 2021).

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 & Abduljabb (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 (Joyce 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, 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.

CHAPTER 7

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. Regarding Cr, comparison to international limits was not possible since no limits were set for its content in rice. However, average concentrations of Hg and Pb in the total UAE and Lebanese samples were exceeding international limits, with Cd in Lebanon exceeding the European limit. Since there are no current provisional tolerable limits set for As, any level could be considered unsafe. 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 studies conduct representative FFQs in the UAE.

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