

Research Article

Hanan F. Al-Harbi*, Amal M. Al-Mohaimed, Maha F. El-Tohamy

Assessment of essential elements and heavy metals in Saudi Arabian rice samples underwent various processing methods

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Abstract: Environmental, soil, and groundwater pollution from toxic heavy metals, as well as food safety are all global concerns nowadays. The effect of various processes viz. washing, soaking, and cooking of rice samples (ten rice varieties, 50 samples) on the concentration of essential elements and toxic heavy metals was determined using the inductively coupled plasma-mass spectrometry technique. The concentrations of As, Cd, Cu, and Ni were found to be below the maximum permissible levels. The range of mean concentrations of metals (mg/kg) was recorded as Al (15.495–8.151), Fe (10.358–7.499), Ni (0.399–0.176), Cu (4.518–2.615), Zn (28.635–12.880), As (0.152–0.042), Cd (0.233–0.038), Pb (0.713–0.417), Ti (2.157 > 0.521), Sn (1.406–0.016), and W (1.114–0.017) mg/kg. Pt and Ag metals were not found in all samples. Soaking rice for 2 h was one of the most successful techniques for lowering heavy metal concentrations, followed by overnight soaking, which aided in the elimination of Al, Cd, Pb, and Pb. Heavy metal exposure has a significant impact on human health. This study creates a promising view to use a simple and accurate detection method for minimizing the effect of different processing methods on the essential elements and heavy metal contents.

Keywords: rice grains, ICP-MS technique, essential elements, toxic metals, heavy metals contamination

* **Corresponding author: Hanan F. Al-Harbi**, Department of Botany and Microbiology, College of Science, King Saud University, P.O. Box 93410, Riyadh 11673, Saudi Arabia, e-mail: hhanan@ksu.edu.sa

Amal M. Al-Mohaimed: Department of Chemistry, College of Science, King Saud University, P.O. Box 22452, Riyadh 11495, Saudi Arabia, e-mail: muhemeed@ksu.edu.sa

Maha F. El-Tohamy: Department of Chemistry, College of Science, King Saud University, P.O. Box 22452, Riyadh 11495, Saudi Arabia, e-mail: moraby@ksu.edu.sa

1 Introduction

Toxic metals primarily arising from intensified agriculture, rapid urbanization, and industrialization has become an issue of global concern [1]. Toxic metals such as cadmium (Cd), manganese (Mn), lead (Pb), arsenic (As), chromium (Cr), and antimony (Sb) are naturally present in the earth. They have polluted the environment (water, air, soils, and food), and through the food chains have produced adverse effects on human health [2,3].

Toxic metal contamination of food is one of the most important assessment parameters for food quality assurance. Exceedance of certain threshold levels could lead to several adverse health effects [4–6] considering their tendency for poor biodegradability, accumulation over time, and long biological half-lives. Human exposure to high or low levels of toxic metals (As, Pb, Cd, etc.) through polluted air or diet can cause severe adverse lung, skin, kidney, prostate, and gall bladder cancerous effects [7]. Dietary exposure is the most common route through which toxic metals affect human health [8].

Grains including rice, oats, maize, etc. are the common daily diet that provides nutrients, proteins, essential elements, and carbohydrates to humans. Rice is one of the most prevalent grain crops that has a crucial contribution to fulfilling food requirements across the globe. It represents the dominant staple food for over half of the world's population especially in Asian developing countries [9]. According to the Food and Agriculture Organization reports in recent years, the production of rice has increased to reach 498.3 million metric tons, of which 90% was consumed for food [10].

Because the high soil mobility and availability of the total As, Cd, and Pb mainly derived from the indiscriminate use of pesticides and fertilizers creates a severe warning to produce contaminant free and safe crops worldwide [8,11,12]. Despite rice being widely consumed by humans as a source of certain vitamins, carbohydrates, minerals, and essential elements, it is an important route of toxic metal

exposure due to its ability to accumulate more metals than other cereals [13,14]. Rice plants are more efficient in assimilating As and Cd as toxic heavy metals into their grains than other cereal crops and their content in rice grains depends on cultivation conditions. The bioavailability of these toxic metals is enhanced in flooded (reduced) soil conditions [15,16]. However, with the progress of human activities, mainly industrial processing, these toxic heavy metals have become the most challenging environmental aspects [17].

Different reports have described the toxicity of rice with various heavy metals such as Cd, As, and Pb [18,19]. Others reported the effect of different preparation methods and cooking processes on the reduction of toxic metals of various rice species [20,21]. The influences of different pre-cooking and cooking methods on the concentration of toxic Fe, Co, Zn, As, Pb, and Cd metals in different consumed rice types have been evaluated [22]. All cooking methods can cause a considerable removal of toxic metals from the rice samples. Furthermore, the effect of the par-boiling cooking method on toxic metal content and the nutritional constituents of three rice varieties have been studied. Parboiling has reduced the toxic levels of aluminum, nickel, magnesium, chromium, lead, and arsenic [23]. Multiple studies addressed that the addition of excess water for cooking rice plays an important role in the reduction and removal of As by 15–63% [24–26]. However, the use of excess water for cooking may cause a loss of essential elements such as iron (Fe) by 40–75% according to the type of rice and the technique of cooking [27].

Many analytical techniques such as atomic absorption spectrometry [28], electrochemical atomic absorption spectrometry [29], laser-induced breakdown spectroscopy [30], and inductively coupled plasma optical emission spectroscopy [31] have previously determined the heavy metal contaminations in rice.

Inductively coupled plasma-mass spectrometry (ICP-MS) has been suggested for the quantification of heavy metal concentrations in various rice samples usually used in Saudi Arabia. The identified levels of heavy metals are categorized based on the recommended limit allowed by the FAO/WHO guidelines [32]. Based on the importance of this crop in the food basket of the Saudi Arabia, and in order to find the best and safest methods in the process of preparing rice to avoid contamination with heavy metals as much as possible, this study examined the impact of washing, soaking, and cooking methods on the heavy metal contents of different kinds of rice available in the local markets of Saudi Arabia. Also, this research aims to compare the conditions for utilizing rice as main foodstuff and find out the optimal conditions that preserve the

important components and remove the heavy metals and make suitable for eating based on the dietary preferences of people.

2 Materials and methods

2.1 Sampling

Ten rice varieties of the major brands consumed in Saudi Arabia were purchased from local Saudi Arabia markets. Four samples (three long/white 2016, 2017 and one long/brown 2017, India), two samples (medium/white and long/white 2018, Australia), two samples (medium/yellow and long/white 2018, America), and the last two samples (long/white 2018 Egypt and Pakistan) were selected. Five samples were collected from each rice variety and each sample was analyzed in triplicates. The samples were collected and kept in polypropylene plastic bags at room temperature for further analysis.

2.2 Chemicals and instruments

The multi-elements stock standard solution was used to prepare 10 µg/mL of elemental standard materials. Nitric acid (HNO₃ of purity grade, 65%) was supplied by Sigma-Aldrich (Hamburg, Germany). Ultrapure water was obtained from 18.2 MX cm of a Millipore ultrapure water purification system (Bedfordshire, UK) at ambient temperature.

The analytical measurements of trace metals were performed using an ICP-MS of model NexION 300 D (PerkinElmer, Waltham, USA). The detection was carried out in triplicates in a dwell time of 40 min using lens voltage of 9.55 V, analog stage voltage of –1,745 V, and pulse stage voltage of 950 V. Details of the ICP-MS settings are given in Table 1.

2.3 Preparation of samples

The preparation of the sample was performed by cleaning, drying, and weighing 2 g of rice in a ceramic crucible. The quantity of rice was heated using an electric cooker until the disappearance of fumes and then transferred to a high-temperature oven. The sample was first carbonized at a low temperature and then incinerated at a high temperature of 800°C for 10–12 h. The obtained sample was cooled

Table 1: Instrument operating conditions for the determination of metallic species in rice samples

RF power	1,600 W
Nebulizer gas flow	0.65 L/min
Lens voltage	9.55 V
Analog stage voltage	-1,745 V
Pulse stage voltage	950 V
Number of replicates	3
Reading/replicates	20
Scan mode	Peak hopping
Dwell time	40 ms
Integration	1,200 ms

and treated with 10 mL of 3% (v/v) HNO₃ solution followed by gently heating to near dryness. After cooling, it was transferred into a 100 mL volumetric flask and ultrapure water to the constant mark. Prior to analysis, the resulting solution was shaken well [33].

2.4 Chemical analysis

ICP-MS technique was applied to determine 13 toxic elements of aluminum (Al), iron (Fe), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), silver (Ag), cadmium (Cd), lead (Pb), titanium (Ti), tin (Sn), platinum (Pt), and tungsten (W). The equipment was calibrated using standard reference materials of the elemental mixture. To maintain the precision and accuracy of the sample analysis, blanks and certified reference materials were analyzed with other unknown samples in each batch of rice samples. All rice samples were analyzed in triplicates.

3 Results

Despite the fact that heavy metals are the oldest known poisons detrimental to humans, heavy metal toxicity remains a highly broad issue due to the wide range of symptoms caused by heavy metal poisoning. As, Cd, mercury (Hg), and Pb are some of the most often encountered metals that have been linked to a variety of negative consequences in humans because of their buildup in the human body as a result of any dietary product. Heavy metal exposure frequently causes chronic and subtle symptoms that resemble those of other disorders. As is regarded as one of the most harmful contaminants due to its presence in the environment, poisonous activity, and potential for human exposure. It causes skin damage,

circulatory problems, and enhances the cancer developments [34]. Cd is a very poisonous non-essential element that occurs as a byproduct of zinc manufacture and is commonly found in phosphate fertilizers. Cd may be found literally everywhere due to its extensive technical and industrial applications and it is acknowledged as one of the most harmful trace elements in the environment due to its high soil to plant transference rate. Cd exposure can result in a number of degenerative changes in various organs and tissues. Cd has a broad carcinogenic activity that can impact a variety of organs including the pancreas, kidneys, lungs, urinary bladder, breast, and prostate, as well as cause diabetic problems, hypertension, and osteoporosis [35]. Because lead is a typically cumulative hazardous element, there has been a great deal of concern over lead usage and lead exposure in recent decades. Lead exposure causes toxic consequences in the kidney, neurological, hematological, gastrointestinal systems, male and female reproductive organs, and other soft tissues, with long-term lead deposition mainly accumulating in the bones [36]. Hg is a common and persistent element that may be found almost anywhere. Hg is considered to be one of the most harmful heavy metals for human intake due to high levels of Hg content in a wide range of food and its bioaccumulation in the environment, particularly the aquatic chain. Exposure to Hg causes neurological disorders and kidney damage [37].

The acceptable limits recommended by WHO [38] for some elements that are present in foodstuff are Al (1 mg/kg), Fe (0.8 mg/kg), Ni (0.5 mg/kg), Cu (2–3 mg/day), Zn (40 mg/day), As (0.05 mg/kg), Cd (0.1 mg/kg), Pd (0.2 mg/kg), Ti (1%), Sn (5 mg/kg), and W (50 µg/kg). The obtained results from the conducted study were compared with the acceptable WHO values and are shown in Tables 2–6.

3.1 Heavy metal concentrations in rice samples without washing

The concentrations of heavy metals and other elements were measured without washing in ten rice samples commonly consumed in Saudi Arabia determined in triplicates and are summarized in Table 2. The outcomes revealed that the levels of Al (7.6–36.0 mg/kg), Fe (4.0–22.8 mg/kg), Cu (1.3–4.8 mg/kg), Zn (13.7–75.1 mg/kg), and Pb (0.21–1.38 mg/kg) were higher than those recommended by WHO. However, the other elements such as Ni (0.18–0.67 mg/kg), As (0.02–0.05 mg/kg), Cd (0.03–0.32 mg/kg), Sn (0.006–0.04 mg/kg), and W (0.05–7.76 mg/kg) displayed acceptable concentrations with respect the WHO values.

Table 2: Heavy metals concentration^a in rice samples (without wash)^b (mg/kg ± %RSD) in comparison with WHO standard limit

No.	Al	Fe	Ni	Cu	Zn	As	Cd	Pb	Ti	Sn	W
WHO standard	1.0	0.8	0.5	2-3	40	0.05	0.1	0.2	1%	5.0	50
S1	19.895 ± 1.6	22.299 ± 2.0	0.604 ± 2.7	4.891 ± 3.3	16.2 ± 3.0	0.049 ± 3.5	0.14 ± 2.1	0.96 ± 1.2	0.099 ± 1.3	0.024 ± 1.6	7.767 ± 2.3
S2	13.144 ± 2.2	17.889 ± 1.1	0.471 ± 1.0	3.917 ± 1.0	24.246 ± 1.8	0.048 ± 1.3	0.082 ± 1.5	0.553 ± 0.6	0.399 ± 3.0	0.018 ± 3.1	1.971 ± 0.5
S3	7.665 ± 2.4	5.931 ± 0.4	0.289 ± 1.1	2.678 ± 0.7	14.688 ± 1.0	0.023 ± 2.3	0.093 ± 0.8	1.385 ± 1.5	0.147 ± 2.8	0.012 ± 3	ND
S4	36.045 ± 2.4	13.82 ± 0.4	0.67 ± 1.1	4.61 ± 0.7	22.825 ± 1.0	0.04 ± 3.1	0.32 ± 1.4	0.649 ± 0.8	0.649 ± 1.5	0.026 ± 3.0	ND
S5	8.225 ± 1.0	8.302 ± 0.5	0.288 ± 0.7	3.249 ± 1.2	17.423 ± 0.1	0.029 ± 2.5	0.033 ± 0.7	0.5 ± 2.7	0.329 ± 1.7	0.011 ± 3.0	0.057 ± 1.7
S6	12.106 ± 1.1	9.811 ± 1.2	0.256 ± 2.1	4.426 ± 1.4	75.1 ± 0.6	0.033 ± 0.2	0.12 ± 1.2	0.709 ± 0.8	0.201 ± 3.1	0.013 ± 2.7	1.182 ± 1.5
S7	10.709 ± 3.0	4.079 ± 2.8	0.187 ± 2.4	1.383 ± 2.7	61.801 ± 1.8	0.055 ± 3.0	0.017 ± 1.4	0.214 ± 3.1	0.134 ± 3.2	0.04 ± 2.3	ND
S8	23.298 ± 2	8.24 ± 2.7	0.247 ± 2.8	3.126 ± 0.4	15.018 ± 0.7	0.067 ± 1.2	0.092 ± 0.9	0.7 ± 1.0	0.627 ± 2.6	0.006 ± 3.1	ND
S9	14.175 ± 3.1	4.785 ± 1.8	0.244 ± 1.6	3.246 ± 1.1	25.279 ± 2.3	0.019 ± 3.0	0.103 ± 2.4	0.934 ± 1.9	0.132 ± 2.4	0.005 ± 2.2	ND
S10	9.687 ± 1.9	8.428 ± 1.6	0.288 ± 2.4	2.908 ± 1.6	13.772 ± 2.7	0.055 ± 2.2	0.122 ± 2.1	0.523 ± 1.8	0.511 ± 2.8	0.009 ± 2.9	0.159 ± 0.3

^aThe mean of three measurements.^bPt and Ag are not detected (ND).**Table 3:** Heavy metals concentration^a in rice samples (with washing three time)^b (mg/kg) in comparison with WHO standard limit

No.	Al (µg/g)	Fe	Ni	Cu	Zn	As	Cd	Pb	Ti	Sn	W
WHO standard	1.0	0.8	0.5	2-3	40	0.05	0.1	0.2	1%	5.0	50
S1	14.309 ± 0.9	9.84 ± 1.1	0.414 ± 0.9	2.45 ± 1.3	8.996 ± 1.1	0.046 ± 1.4	0.067 ± 1.8	0.3360 ± 0.6	3.816 ± 1.6	0.006 ± 2.8	0.300 ± 0.8
S2	16.697 ± 1.0	15.818 ± 2.1	0.718 ± 1.5	2.644 ± 1.2	17.741 ± 3.1	0.046 ± 3.1	0.099 ± 1.5	0.375 ± 0.5	1.569 ± 2.4	0.006 ± 3.1	0.259 ± 1.3
S3	6.072 ± 0.6	4.707 ± 2.2	0.239 ± 1.6	1.754 ± 0.7	13.94 ± 0.9	0.028 ± 2.2	0.059 ± 1.2	0.290 ± 0.9	0.499 ± 2.4	0.009 ± 1.2	0.067 ± 3.1
S4	10.332 ± 0.7	9.674 ± 1.9	0.382 ± 0.7	2.563 ± 1.2	7.202 ± 1.6	0.045 ± 2.8	0.033 ± 2.6	0.607 ± 1.2	0.677 ± 2.3	0.090 ± 1.7	0.499 ± 1.4
S5	9.595 ± 2.2	14.123 ± 1.0	0.329 ± 1.1	4.402 ± 1.2	16.095 ± 0.7	0.077 ± 0.7	0.044 ± 0.6	0.984 ± 1.5	0.237 ± 2.4	0.821 ± 0.5	0.058 ± 2.6
S6	3.087 ± 2.3	4.425 ± 1.9	0.244 ± 1.1	2.901 ± 2.6	15.587 ± 1.7	0.032 ± 2.2	0.035 ± 1.3	0.502 ± 1.3	ND	0.007 ± 2.4	0.043 ± 3.0
S7	4.622 ± 2.0	4.143 ± 2.1	0.262 ± 0.3	2.601 ± 1.3	13.18 ± 2.1	0.045 ± 2.3	0.019 ± 3.2	0.438 ± 0.1	0.131 ± 3.1	0.007 ± 2.6	0.038 ± 3.0
S8	2.335 ± 0.4	6.826 ± 3.0	0.209 ± 2.0	2.319 ± 1.7	9.965 ± 1.6	0.085 ± 1.0	0.029 ± 1.5	0.518 ± 0.3	0.586 ± 3.1	0.007 ± 3.0	0.051 ± 2.6
S9	4.742 ± 1.3	5.308 ± 1.7	0.271 ± 1.8	3.027 ± 1.9	15.689 ± 2.0	0.023 ± 2.0	0.089 ± 2.8	1.640 ± 0.4	0.173 ± 2.5	0.006 ± 1.9	0.034 ± 2.7
S10	9.723 ± 2.5	5.062 ± 3.0	0.332 ± 1.5	2.026 ± 2.2	10.303 ± 2.6	0.060 ± 1.0	1.858 ± 1.4	0.449 ± 3.2	0.578 ± 2.5	ND	ND

^aThe mean of three measurements.^bPt and Ag are not detected (ND).

Table 4: Heavy metals concentration^a in rice samples (with soaking for 2 h)^b (mg/kg) in comparison with WHO standard limit

No.	Al	Fe	Ni	Cu	Zn	As	Cd	Pb	Ti	Sn	W
WHO standard	1.0	0.8	0.5	2-3	40	0.05	0.1	0.2	1%	5.0	50
S1	8.170 ± 2.0	4.806 ± 2.5	0.250 ± 1.7	4.580 ± 0.4	8.478 ± 3.2	0.028 ± 2.5	0.046 ± 0.8	0.403 ± 1.5	2.066 ± 2.4	0.006 ± 3.1	0.3 ± 0.8
S2	6.408 ± 1.3	12.773 ± 2.8	0.400 ± 1.3	3.257 ± 1.9	17.413 ± 2.3	0.040 ± 2.5	0.036 ± 1.6	0.295 ± 0.7	0.479 ± 3.1	0.006 ± 1.9	0.035 ± 1.6
S3	15.751 ± 0.6	5.628 ± 3.0	0.188 ± 1.5	4.151 ± 1.3	13.358 ± 2.2	0.025 ± 1.3	0.056 ± 1.5	0.381 ± 0.4	0.185 ± 2.5	0.008 ± 3.0	0.052 ± 1.4
S4	10.233 ± 1.3	6.804 ± 2.0	0.332 ± 1.1	3.521 ± 0.6	6.895 ± 2.0	0.036 ± 1.2	0.028 ± 1.0	0.357 ± 0.2	0.187 ± 2.5	0.011 ± 1.2	0.050 ± 1.2
S5	12.348 ± 0.3	5.481 ± 3.2	0.162 ± 2.5	3.074 ± 0.9	13.889 ± 1.6	0.029 ± 2.4	0.040 ± 2.2	0.256 ± 0.7	0.222 ± 2.5	7.430 ± 3.0	0.055 ± 2.4
S6	4.992 ± 2.6	4.153 ± 2.5	0.156 ± 1.7	3.006 ± 1.2	11.447 ± 2.0	0.025 ± 2.0	0.033 ± 3.1	0.286 ± 0.4	0.167 ± 3.1	0.008 ± 2.1	0.111 ± 0.4
S7	4.699 ± 3.0	10.492 ± 1.7	0.160 ± 2.5	3.306 ± 1.5	13.319 ± 2.3	0.094 ± 1.3	0.026 ± 3.0	0.652 ± 2.2	0.617 ± 3.2	0.005 ± 2.4	0.052 ± 0.5
S8	4.855 ± 1.9	10.605 ± 3.2	0.160 ± 3.0	3.305 ± 2.5	12.315 ± 2.7	0.094 ± 1.9	0.026 ± 3.0	0.651 ± 1.4	0.616 ± 2.5	0.005 ± 0.6	0.052 ± 1.2
S9	10.075 ± 2.5	4.704 ± 3.2	0.224 ± 2.2	2.585 ± 2.4	19.539 ± 1.1	0.023 ± 1.9	0.030 ± 0.9	0.393 ± 2.3	0.059 ± 3.7	0.029 ± 2.2	ND
S10	4.153 ± 1.5	9.549 ± 2.4	0.283 ± 0.6	2.607 ± 1.8	11.303 ± 1.3	0.054 ± 3.0	0.063 ± 0.6	0.500 ± 0.3	0.695 ± 2.5	0.005 ± 2.7	0.246 ± 0.7

^aThe mean of three measurements.^bPt and Ag are not detected (ND).**Table 5:** Heavy metals concentration^a in rice samples (with soaking overnight)^b (mg/kg) in comparison with WHO standard limit

No.	Al	Fe	Ni	Cu	Zn	As	Cd	Pb	Ti	Sn	W
WHO standard	1.0	0.8	0.5	2-3	40	0.05	0.1	0.2	1%	5.0	50
S1	25.086 ± 2.7	5.855 ± 0.8	0.196 ± 0.5	1.935 ± 1.4	8.517 ± 1.4	0.034 ± 2.0	0.041 ± 2.5	0.380 ± 1.8	1.805 ± 1.3	5.488 ± 2.7	ND
S2	12.278 ± 1.8	29.378 ± 1.9	0.343 ± 3.0	3.267 ± 0.3	18.767 ± 1.0	1.173 ± 0.2	0.040 ± 0.6	1.505 ± 0.9	0.739 ± 2.5	2.068 ± 0.2	0.048 ± 1.0
S3	9.096 ± 3.0	3.878 ± 3.2	0.149 ± 1.5	1.395 ± 1.0	12.568 ± 1.3	0.029 ± 2.5	0.037 ± 0.6	0.272 ± 0.6	ND	ND	ND
S4	28.119 ± 1.5	14.537 ± 2.5	0.134 ± 2.6	2.348 ± 1.3	6.363 ± 1.1	0.082 ± 2.1	0.163 ± 1.3	0.672 ± 0.4	5.879 ± 1.9	0.091 ± 1.9	0.065 ± 0.8
S5	15.060 ± 1.0	5.340 ± 2.4	0.262 ± 2.5	2.569 ± 1.2	14.267 ± 2.3	0.026 ± 3.2	0.057 ± 2.6	0.764 ± 0.8	0.205 ± 2.5	0.009 ± 1.2	ND
S6	9.154 ± 2.9	3.769 ± 3.2	0.099 ± 1.7	2.187 ± 2.5	12.661 ± 2.9	0.029 ± 2.5	0.107 ± 0.4	0.309 ± 1.6	0.212 ± 3.2	0.008 ± 2.5	0.111 ± 0.6
S7	5.168 ± 1.3	3.649 ± 2.6	0.108 ± 2.7	2.642 ± 2.1	11.915 ± 1.4	0.039 ± 1.0	0.0179 ± 3.0	0.333 ± 1.1	0.143 ± 2.5	4.962 ± 3.0	0.126 ± 1.2
S8	1.780 ± 0.2	6.701 ± 1.0	0.130 ± 2.1	2.392 ± 1.8	9.628 ± 2.3	0.055 ± 1.4	0.021 ± 3.0	0.407 ± 0.9	0.475 ± 2.5	0.014 ± 0.2	0.109 ± 1.3
S9	11.513 ± 1.4	4.926 ± 2.5	0.161 ± 1.7	5.000 ± 2.3	12.507 ± 1.4	0.021 ± 1.2	0.077 ± 1.6	0.362 ± 0.4	0.144 ± 3.0	0.008 ± 1.2	0.417 ± 1.3
S10	2.320 ± 3.0	7.980 ± 2.5	0.180 ± 3.0	2.419 ± 2.9	11.015 ± 3.1	0.033 ± 1.1	0.028 ± 2.5	0.306 ± 2.1	0.581 ± 3.2	0.006 ± 2.5	0.233 ± 1.5

^aThe mean of three measurements.^bPt and Ag are not detected (ND).

Table 6: Heavy metals concentration^a in rice samples (with cooking)^b (mg/kg) in comparison with WHO standard limit

No.	Al	Fe	Ni	Cu	Zn	As	Cd	Pb	Ti	Sn	W
WHO standard	1.0	0.8	0.5	2–3	40	0.05	0.1	0.2	1%	5.0	50
S1	13.518 ± 2.0	5.007 ± 3.1	0.387 ± 2.3	4.705 ± 2.3	8.619 ± 2.5	0.032 ± 2.8	0.029 ± 0.4	0.444 ± 2.1	0.225 ± 3.1	0.009 ± 0.8	ND
S2	5.719 ± 3.0	11.448 ± 3.1	0.402 ± 2.5	3.363 ± 2.1	20.487 ± 1.0	0.039 ± 2.9	0.041 ± 1.8	0.482 ± 3.0	0.329 ± 2.1	0.007 ± 3.4	ND
S3	7.973 ± 1.8	6.667 ± 0.6	0.289 ± 0.4	3.444 ± 0.9	14.446 ± 1.8	0.035 ± 0.8	0.182 ± 1.2	0.453 ± 0.5	0.245 ± 0.9	0.015 ± 2.5	0.041 ± 1.9
S4	8.948 ± 1.4	4.863 ± 0.9	0.676 ± 1.2	4.512 ± 1.4	8.715 ± 3.0	0.036 ± 2.7	0.036 ± 3.0	0.348 ± 1.4	0.148 ± 3.2	0.013 ± 3.0	ND
S5	11.174 ± 0.6	5.327 ± 1.8	0.239 ± 1.4	3.296 ± 1.3	15.441 ± 1.9	0.026 ± 2.2	0.057 ± 1.5	0.417 ± 0.4	0.254 ± 1.1	0.009 ± 1.8	ND
S6	9.456 ± 0.9	5.339 ± 1.5	0.289 ± 2.3	6.303 ± 0.7	15.300 ± 2.3	0.049 ± 0.2	0.020 ± 3.0	0.723 ± 0.8	2.216 ± 1.7	0.023 ± 0.9	0.042 ± 1.6
S7	10.226 ± 2.4	3.942 ± 1.1	0.307 ± 1.4	4.300 ± 2.1	18.697 ± 0.8	0.098 ± 1.1	0.019 ± 0.9	0.385 ± 0.9	ND	0.009 ± 1.6	ND
S8	6.388 ± 0.6	7.408 ± 2.0	0.200 ± 2.7	4.853 ± 1.4	9.459 ± 2.4	0.079 ± 2.0	0.069 ± 2.2	0.585 ± 0.4	0.667 ± 3.1	0.013 ± 2.0	ND
S9	12.758 ± 2.1	6.427 ± 0.8	0.609 ± 2.3	4.519 ± 1.5	25.340 ± 0.4	0.023 ± 1.8	0.034 ± 1.1	0.379 ± 0.3	0.342 ± 3.1	0.089 ± 0.6	ND
S10	4.544 ± 1.2	10.553 ± 1.0	0.592 ± 2.0	5.885 ± 1.2	18.563 ± 0.6	0.066 ± 1.0	0.035 ± 1.3	0.369 ± 0.3	0.782 ± 2.5	0.016 ± 0.7	0.089 ± 1.4

^aThe mean of three measurements.^bPt and Ag are not detected (ND).

3.2 Heavy metal concentrations in rice samples after three times of washing

The contents of heavy metals and other elements in the same rice samples were evaluated after three times of washing. The results are summarized in Table 3 and are as follows: Al 2.335 ± 0.4 – 16.697 ± 1.0 , Fe 4.143 ± 2.1 – 15.818 ± 2.1 , Ni 0.209 ± 2.0 – 0.718 ± 1.5 , Cu 1.754 ± 0.7 – 4.402 ± 1.2 , Zn 7.202 ± 1.6 – 17.741 ± 3.1 , As 0.023 ± 2.0 – 0.774 ± 0.7 , Cd 0.019 ± 3.2 – 1.858 ± 1.4 , Pb 0.290 ± 0.9 – 1.640 ± 0.4 , Ti 0.131 ± 3.1 – 3.816 ± 1.6 , Sn 0.006 ± 1.9 – 0.821 ± 0.5 , W 0.034 ± 2.7 – 0.499 ± 1.4 . Pt and Ag were not detected in all samples. These results showed decrease in the concentration of all existent elements after washing the rice samples three times with water.

3.3 Heavy metal concentrations in rice samples after soaking for 2 h

The rice samples were subjected to soaking for 2 h and the contents of heavy metals and other elements were estimated. The results are presented in Table 4 and are as follows: Al 4.153 ± 1.5 – 15.751 ± 0.6 , Fe 4.153 ± 2.5 – 12.773 ± 2.8 , Ni 0.156 ± 1.7 – 0.400 ± 1.3 , Cu 2.585 ± 2.4 – 4.580 ± 0.4 , Zn 6.895 ± 2.0 – 19.539 ± 1.1 , As 0.023 ± 1.9 – 0.094 ± 1.9 , Cd 0.026 ± 3.0 – 0.063 ± 0.6 , Pb 0.256 ± 0.7 – 0.652 ± 2.2 , Ti 0.059 ± 3.7 – 2.066 ± 2.4 , Sn 0.005 ± 2.7 – 7.430 ± 3.0 , W 0.035 ± 1.6 – 0.300 ± 0.8 . Pt and Ag were not detected in all samples. The estimated data indicated that soaking process of rice samples for 2 h showed remarkable reduction in some heavy metals such as As, Cd, Ti, Sn, and W. However, slight decrease in the concentrations of Al, Fe, Ni, Cu, and Zn was observed.

3.4 Heavy metal concentrations in rice samples after soaking for 12 h

Another study was performed to evaluate the contents of heavy metals and other essential elements in rice samples after soaking for 12 h. The results are shown in Table 5 and are as follows: Al 1.780 ± 0.2 – 28.119 ± 1.5 , Fe 3.769 ± 3.2 – 29.378 ± 1.9 , Ni 0.099 ± 1.7 – 0.343 ± 3.0 , Cu 1.935 ± 1.4 – 5.000 ± 2.3 , Zn 6.363 ± 1.1 – 19.767 ± 1.0 , As 0.021 ± 1.2 – 1.173 ± 0.2 , Cd 0.017 ± 3.0 – 0.163 ± 1.3 , Pb 0.272 ± 0.6 – 1.505 ± 0.9 , Ti 0.143 ± 2.5 – 5.879 ± 1.9 , Sn 0.006 ± 2.5 – 5.488 ± 2.7 , W 0.048 ± 1.0 – 0.417 ± 1.3 . Pt and Ag were not detected in all the samples. The obtained results after soaking the rice

samples for 12 h displayed a significant decrease in the concentration of heavy metals to half its values.

3.5 Heavy metal concentrations in rice samples after cooking

Cooking rice is one of the most common processing methods used to evaluate the contents of heavy metals and essential elements Table 6 summarizes the resulting values of the tested heavy metals and essential elements: Al 4.544 ± 1.2 – 13.518 ± 2.0 , Fe 4.863 ± 0.9 – 11.448 ± 3.1 , Ni 0.200 ± 2.7 – 0.676 ± 1.2 , Cu 3.296 ± 1.3 – 6.303 ± 0.7 , Zn 8.619 ± 2.5 – 25.340 ± 0.4 , As 0.023 ± 1.8 – 0.098 ± 1.1 , Cd 0.019 ± 0.9 – 0.069 ± 2.2 , Pb 0.348 ± 1.4 – 0.723 ± 0.8 , Ti 0.148 ± 2.2 – 2.216 ± 1.7 , Sn 0.007 ± 3.4 – 0.023 ± 0.9 , W 0.041 ± 1.9 – 0.089 ± 1.4 . Pt and Ag were not detected in all the samples. As shown in Table 6 the levels of the heavy metals were significantly reduced after cooking the rice samples to relatively half its values.

The above outcomes revealed that various concentrations of heavy metals in the selected rice samples were recorded. It was observed that high concentrations of Zn, Fe, Ti, and Cu and fewer concentrations of Sn, Pb, Ni, As, Cd, and W have been recorded after various rice preparation methods. However, Ag and Pt are not detected. According to the recorded data in Table 2, a large variation in the concentrations of Fe and Zn was observed. This could be probably due to the variation in the rice source. Pb concentrations in the rice samples were much higher than the current allowable limits recommended by FAO/WHO (2004) [38]. The obtained results are fully in agreement with those obtained from previously published studies which stated that the levels of Pb are over $0.2 \mu\text{g/g}$ in unwashed samples [39]. The levels of various metals after washing the rice samples three times, soaking the rice for 2 h, overnight soaking, and cooking are summarized in Tables 3–6 as the mean values of three experimental measurements \pm relative standard deviation (% RSD) and are expressed in mg/kg. The recorded concentration levels of the heavy metals including, As, Cd, Cu, and Ni in all studied samples using three different processing methods (washing, soaking, and cooking) were found to be below the maximum permissible levels which were recommended by FAO/WHO. Additionally, the average As was substantially lower than that of the safe limit recommended by FAO/WHO ($0.2 \mu\text{g/g}$) and similar to the previously reported results [40–42]. The recorded results also indicated that W was found in some of the rice batches without a wash (Sample 1) with the highest concentration of $7.767 \mu\text{g/g}$. Throughout all rice processing methods, it

was observed that both Pt and Ag metals have not been detected in any of the rice samples.

3.6 Effect of processing methods of rice samples

3.6.1 Essential element (Zn, Fe, Cu, and Al) levels

The levels of all elements in rice samples were greatly influenced by the processing methods of these samples. The most censorious of these are the levels of essential elements like Zn, Fe, Cu, and Al. The mean percentage recovery levels of Zn extended to 41 mg/kg in rice grains without washing. However, washing three times, soaking the rice samples for 2 h, soaking all night, and cooking decreased its concentration to approximately half 12.7956 , 12.8798 , 11.821 , and 15.515 mg/kg , respectively. The Fe metal concentrations were maintained at 7.9926 , 7.4995 , and 8.6013 mg/kg after washing three times, soaking for 2 h, and soaking the rice all night, respectively. However, the concentration of Fe was greatly affected by cooking, its level decreased from 10.3584 mg/kg prior to washing to 6.6981 mg/kg after cooking. However, Cu concentration in rice grain was found to be 3.443 mg/kg without washing, and the lowest Cu concentrations in rice samples after washing three times, soaking for 2 h, and soaking all night were 2.6687 , 3.3392 , and 2.6154 mg/kg , respectively. The rice samples showed an average high concentration of Cu at 4.518 mg/kg after cooking. Figure 1 shows the level variation of each essential element in rice samples throughout various processing methods.

3.6.2 Reduction of toxic elements in rice samples

Throughout various processing methods, the concentration of toxic elements in the rice samples was greatly influenced and large variations in the concentration of toxic elements were observed. Al, Ni, Pb, Sn, W, and Ti were detected in all samples with the five different processing methods, while Ag and Pt were not detected. The Al element was observed at high levels in all treatment methods with mean percentage recovery of the ten samples and was higher than the levels of Ni, Pb, Sn, W, and Ti. Moreover, the minimal concentration of Al was achieved when washed several times and soaked for 2 h. The minimal concentration of Ni element was obtained after soaking the rice samples overnight, while cooking process caused an increase in its concentration level in only three samples.

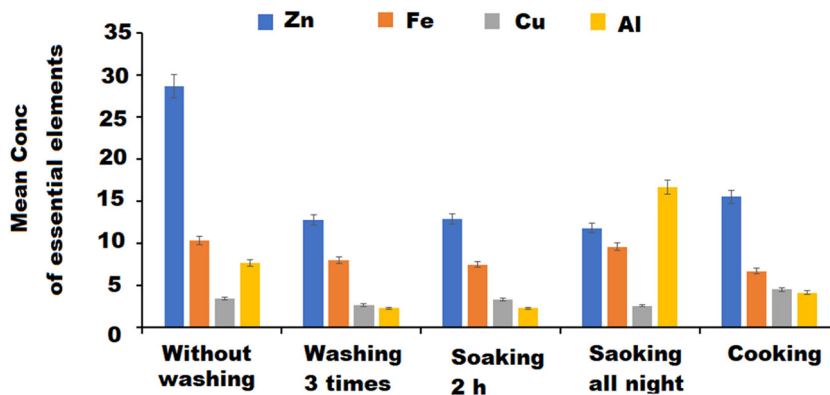


Figure 1: Comparison of mean concentration of essential elements (Zn, Fe, Cu, and Al) contents available in rice samples with the treatment effects.

Soaking overnight and cooking were the most effective processing methods for reducing their levels. Soaking for 2 h gave a decrease in Pb, Ti, and W concentrations and an increase in the concentration of Sn element. Furthermore, soaking overnight increased the As and Sn concentration levels, while cooking was the most effective in reducing their concentrations. It was noticed that the tungsten (W) element was the only toxic heavy metal that extended to the minimum limit as well as the iron (Fe) element after cooking. Figure 2 illustrates the comparative mean concentrations of heavy metals in rice samples after being treated with various processing methods.

4 Discussion

Rice is one of the most important foods in Saudi Arabia. There are many rice varieties that vary in their sources,

essential elements, and contents of toxic metals. There is no doubt that the methods of rice processing before and after cooking play an important role in maintaining or removing many essential and important elements for human health. The current study evaluated the effect of rice processing methods such as washing, soaking, and cooking on the concentrations of essential and heavy elements of ten commonly used rice varieties in the Kingdom of Saudi Arabia using the ICP-MS technique. The obtained results confirmed that the percentage concentration levels of essential elements such as Zn, Cu, and Fe were decreased after using the three processing (washing, soaking, and cooking) methods. The Fe metal concentrations were approximately maintained constant after washing three times, soaking for 2 h, and soaking the rice all night. However, the cooking process has greatly decreased the level of Fe. The Cu concentrations were decreased after washing and soaking, while the Cu

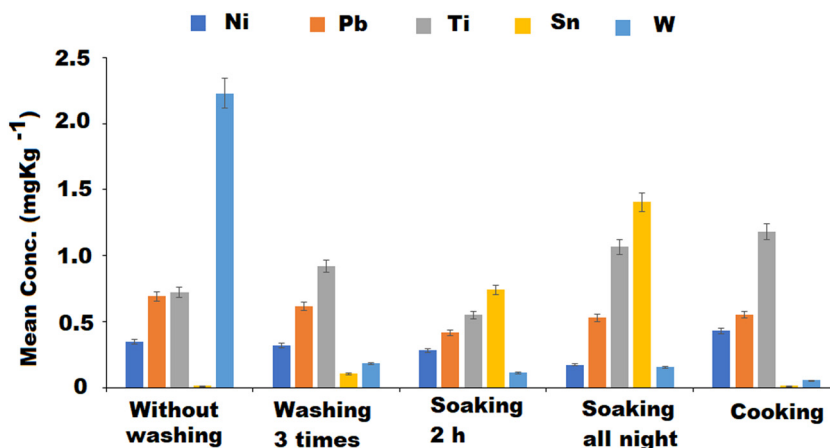


Figure 2: Comparison of mean concentration of heavy metals (Ni, Pb, Ti, Sn, and W) contents available in rice samples with the treatment effects.

level was higher than in other processes. Accordingly, the loss of essential elements is based on their localization on the rice grains' surface, which helps their easy removal through washing, soaking, and cooking processing methods [43]. The washing process once or several times is a common method to prepare rice worldwide. This study showed a different effect on some essential elements such as Fe and Al, and on some toxic elements like Pb, Sn, and Ti. The obtained results also revealed that soaking for 2 h was one of the most potent methods to reduce the concentration of toxic elements except for two toxic elements Ti and Cd, where their concentrations are maintained high. However, soaking the rice overnight removed some toxic elements such as Al, Cd, Pb, Ni, and W. Although, soaking overnight served to remove some concentrations of toxic metals, unfortunately, it increased the concentrations of Ti, As, and Sn. The large variation in the increase or decrease of essential and toxic elements can be attributed to the type of rice, the resistance of the surface layers of rice grains to water infiltration, the elements, and the ability of organic compounds and proteins to form complexes with metals in the rice grains. These outcomes were in agreement with those previously reported by several studies which have focused on the effect of different rice processing methods in decreasing or increasing the number of essential elements [44–46]. Al-Saleh and Abduljabbar [47] stated that soaking or rinsing rice grains with water was served to reduce Pb levels in 36 brands of rice grains to acceptable safe levels. Consequently, the study carried out by Adibi et al. [48] revealed that soaking rice from 1 to 12 h augments the entrance of water in grains and more metals could be dissolved in water.

Some elements may permeate the inner layers of the rice grains, and after a short duration of soaking for 2 h, they are released to the outside and appear on the surface, while soaking for long periods, overnight, are more effective in removing toxic minerals from the rice. So, soaking time affects the reduction of elements in the samples [49].

Washing several times and soaking for 2 h was effective in removing heavy metals from rice. This can be attributed to the increase in soaking time causing a greater quantity of water that penetrates the layers of rice grains and hence, higher removal of toxic metals will be observed. The report [50] revealed that significant quantities of toxic metals (As and Cd) can be removed by discarding the water used for washing and cooking. This removal is enhanced by the fact that a surface region of the grains of about 80 μm thickness is the richest in these elements.

However, important elements like Zn, Mg, K, and Ca (16.75%) were lost in the water after washing several

times. Similar results were obtained in the present study and a decrease in the concentration of essential elements to the Respiratory Distribution Index was observed with an increase in rice cooking water, except for Fe [51]. Various factors including the type of rice, its cultivation conditions (soil quality, irrigation source, and fertilizers), as well as the ratios of water in the cooking process could affect the concentration of toxic and essential metals in rice [52]. Furthermore, the use of heat during cooking enhances protein degradation which may influence the heavy metal concentrations in food. The report [53] addressed that using a large volume of water (6:1 water: rice) for cooking rice and soaking rinsed rice in 2% NaCl solution for 2 h had the greatest effect to decrease As levels in cooked rice. The washing process once or several times is a common method to prepare rice worldwide, our research showed a different effect on some essential elements such as Fe and Al, and some toxic elements like Pb, Sn, and Ti [21]. Additionally, cooking methods reduced the substance of heavy metals, except that the cooking method influenced the concentration of heavy elements negatively, especially Fe. This result is in agreement with the demonstration that cooking rice with extra water (rice to water ratio 1:6) can decrease As and a number of essential elements like K, Ni, Mg, Co, Mn, Ca, Fe, Zn, and others. The obtained outcomes clarified that the concentrations of all essential and toxic metals were found to be less than those reported by Deng et al. [8]. It should be noted that the duration of cooking, the proportions of water used, as well as different sources of rice has a great effect on the decrease or increase of elements after the cooking process [8]. These results are in agreement with several previous studies which stated that the cooking conditions including, boiling time, temperature, and cooking process reduced the toxic elements Cd, Pb, Hg, and As [54,55].

5 Conclusions

The presence of toxic heavy metals and essential elements in rice has been reported in various studies. Few of these studies have been concerned with the effect of different rice processing methods on the concentrations of essential and toxic metals. The present study focuses on the effect of different processing methods of rice including drying, washing, soaking, and cooking on the concentration level of essential elements and toxic heavy metals. The results revealed that washing several times greatly affect the levels of heavy metals and essential elements such as Al, Fe, Ca, and Zn that were decreased from 7.66 to 2.33, 22.29 to 15.0, 4.89 to 4.4, and 75.1 to

17.0 mg/kg, respectively. However, washing several times did not significantly influence the levels of heavy metals such as As, Cd, Pb, and Ti. Meanwhile, the overnight soaking process and cooking decreased the concentration of the previously mentioned heavy metals and essential elements by half. Thus, soaking, washing after overnight soaking and cooking was strongly influencing the concentration level of these metals. The use of low cost ICP-MS technique for the detection of heavy metal residue in rice after different processing methods is recommended since it is relatively cheap, easily used, and provides sensitive detection of heavy metals. This study opens a future promising view for scientific researchers to study the relationship between the processing methods and the removal of toxic elements. Also, to minimize the harmful risk and increase public health awareness of these toxic heavy metals, especially in Saudi Arabia.

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Ethical approval: The conducted research is not related to either human or animal use.

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