



Research article

Relationship between heavy metals and alpha particles as a marker of environmental pollution in rice consumed in Najaf, Iraq

B.A. Almayahi^{a,*}, Naheda Aljarrah^b^a Department of Environment, Faculty of Science, University of Kufa, Najaf, Iraq^b Department of Physics, College of Dentistry, Babylon University, Hilla, Iraq

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ABSTRACT

This study focuses on the emission of alpha particle rates (EAPR) and heavy metal concentrations (HMC) in the rice from Najaf markets. Nuclear Track Detectors and Flame Atomic Absorption Spectrophotometry, were used respectively. This study shows the pollution in the environment through AP and HM and also finds the relationship between AP and HM. The highest EAPR (0.0092 mBq cm⁻²) was found in the Indian basmati rice, whereas, the lowest EAPR (0.0092 mBq cm⁻²) was found in the Indian basmati rice. Highest Fe was found to be about 2.7237 mg/kg in basmati rice, while the lowest Fe (0.3997 mg/kg) was found in the USA basmati rice. Highest Cd was found to be about 0.0468 mg/kg in Iraqi Alnasryah rice, while the lowest Cd (0.0034 mg/kg) was found in Indian basmati rice. The most upper Pb was found to be about 0.2431 mg/kg in Babil Anbar Iraqi rice, while the lowest Pb (0.0695 mg/kg) was found in Indian basmati rice. Pb and Cd were lower than the FAO/WHO recommended limits (Pb, Cd: 0.50 mg/g) and the European Union acceptable dietary limits. In the combination of recent rice consumption data, an estimated weekly intake of toxic element was calculated for the Iraq population. A statistically significant correlation was found between EAPR and HMC in rice at the 0.05 level.

1. Introduction

The examined rice in the present study is the primary food in Iraq. Recently, concerns were raised about the possible pollution of the crop by heavy elements. Many industrialized processes caused the contamination of the soil, food, water, and air through some heavy metals (cadmium (Cd), Iron (Fe) and lead (Pb) (Almayahi et al., 2014a; Alduhaidahawi et al., 2015; Almayahi et al., 2016). Fu et al. (2008) found that the mean Pb in the rice was about 0.69 mg/g in an electronic waste recycling site in China. Huang et al. (2009) showed a high Pb concentration (0.957 mg/g) in rice. Zhao et al. (2010) found Cd concentration in rice with the most significant value of 0.467 mg/g. It was observed in the study of a possible contamination of heavy metals in rice. Cd with a long biological half-life was found to be toxic to the kidney in a human body. The sampled staple food rice was found to contain heavy metals that can cause health risks to consumers (Silici et al., 2008; Saracoglu et al., 2009; Uluozlu et al., 2009; Demirel et al., 2008). Heavy metals are used in many fields such as industrial, agricultural, and technological applications (Bradl, 2002). Environmental contamination is well-known in mining, smelters, foundries, and metal-based industrial operations (Fergusson, 1990; Bradl, 2002; He et al., 2005). Natural uranium consists of 3 radioisotopes

(²³⁸U, ²³⁵U, and ²³⁴U) with a different half-life. These radioisotopes are unstable, which transform into elements either emitting or absorbing the particles. The unstable nucleus loses energy by emitting ionizing radiation to reach a stable state. Alpha particles (α) decay with energy 4.679 MeV. ⁴He atom represents about one alpha particle. This radioactive particle can enter the human body through the ingestion of contaminated food or water, or inhalation of the air. The ²³⁵U (biological T_{1/2} = 15 d) leaves the human body except for the small part, which stay accumulated in the teeth or kidney, and bones. The alpha particles collide with blood cells which may lead to leukemia. The alpha particles collide with an external electron in atom. It produces a free electron and a positive ion. Human tissues that absorb the radiation have the energy to remove the particles in molecule tissue (Valković, 2000; Hada et al., 2011). ²²⁶Ra exists in vegetation and is assimilated by the gut when it is ingested by animals (NCRP, 1999). There are many literature reviews on alpha particles in biological studies that use a technique counting track. It uses nuclear particle detector as it has high-resolution and ability to detect low concentrations. The track detectors are used to measure the alpha and radon in the blood and tissues of humans and animals (Henshaw, 1989; Henshaw et al., 1994; Almayahi et al., 2011, 2012a, b, c; Almayahi et al., 2014b). This study aimed to assess and analyze the alpha particles

* Corresponding author.

E-mail address: basim.almayahi@uokufa.edu.iq (B.A. Almayahi).<https://doi.org/10.1016/j.heliyon.2019.e03134>

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and heavy metals in rice samples. It also aimed to check the health risk concerning daily consumption of rice by adults. Based on the obtained data, Estimated Weekly Intake (EWI) of toxic metals was calculated and then compared with Provisional Tolerable Weekly Intake (PTWI).

2. Materials and methods

Rice samples were collected from the different sites in Najaf city in Iraq. The alpha emission rates in rice were determined using α -sensitive plastic track detector (PADC-TASTRACK CR-39, UK). The commercial name of Nuclear track detector Polyallyl Diglycol Carbonate is CR-39. It has a general component of the monomer on two groups' allele ($CH_2=CH-CH_2-$). CR-39 has a chemical composition of $(C_{12}H_{18}O_7)$ and a density of about 1.31 g cm^{-3} . The CR-39 has a high sensitivity to record the tracks of alpha, proton, and fission fragments because it has the bonds of weak carbon that breaks when exposed to radiation. Nuclear track detector technology is one of the techniques for determining the radioactive substance due to the availability, accuracy, and it does not require sophisticated electrical equipment (Durrani and Bull, 1987). The rice samples were collected from markets and agricultural lands in Najaf city. The rice was transferred to airtight polyethylene bags, labeled, and taken to the environmental lab for further analysis.

Rice was dried and manually grounded by mortar and pestle and passed through a steel sieve of $75 \mu\text{m}$. The samples were weighted and then placed on the sheets of the high-purity plastic α -detector with area of about a $2 \times 2 \text{ cm}^2$. The detectors with samples were put in vacuum-sealed high-density polyethylene bag (to prevent the influx of radon gas and increase the range of alpha particles) and clamped in position. However, the massive and energetic radioactive emissions, of the alpha particles with the shortest in range, had a strong interaction with the matter.

The detectors with samples sealed in a high-density polyethylene bag were placed together in the freezer at $-20 \text{ }^\circ\text{C}$ for 120 days to allow alpha particle tracks in the rice to accumulate on the TASTRACK detectors. In this technique, the alpha particles were measured without the chemical treatments of the sample using the CR-39 detector. After the end of the exposure, the CR-39 was etched under controlled conditions in NaOH as reported elsewhere (Almayahi et al., 2011, 2012a, b, c). Then the CR-39 was washed and dried. The number of α -tracks per unit area for the rice samples was counted using an optical microscope (A. Kruss, Optronic, Germany) with the MDCE-5C camera at $10 \times$ magnification.

The CR-39 efficiency was 85% calculated by the following formula (Almayahi et al., 2014b):

$$\varepsilon = 1 - \frac{V_B}{V_T}, \quad (1)$$

where V_B = bulk etch rate ($\mu\text{m h}^{-1}$), and V_T = track etch rate ($\mu\text{m h}^{-1}$). The alpha emission rate E_α was calculated using the formula:

$$E_\alpha (\text{Bq cm}^{-2}) = \varepsilon \frac{(\rho_s - \rho_b)}{T}, \quad (2)$$

where, T : Exposure time (Seconds), ρ_s : Number of tracks by the samples (track cm^{-2}), ρ_b : Number of background tracks in the detector (track cm^{-2}). The alpha particles quoted in this study include the removal of a mean background track density. For the area of the rice powder surface in contact with the detector, the mean number of recorded alpha particle tracks per cm^2 day was 1.832, with 0.929 as the lowest number and 2.878 as the highest.

The heavy metals in rice were determined using a Flame Atomic Absorption Spectrophotometer (6300 AA, Shimadzu, Japan). About 0.5 g of the sieved samples was digested at $180 \text{ }^\circ\text{C}$ of about 3 h. 5 ml of aqua regia (mixture of nitric acid and hydrochloric acid) and 1 ml of the HF acid (40%) was added. Then the mixture was heated for two hours, and 5 ml of 65% HNO_3 was added and then burned the mix to dry. 50 ml of distilled water was attached to remnants of the beaker and the solution

was filtered for the separation of solid waste to become a sample ready to measure Cd, Pb, and Fe using Atomic Absorption Spectrophotometer.

The human health risk assessment was carried out by considering the following parameters (Onsanit et al., 2010). The EWI and PTWI were jointly established by FAO/WHO, 2004. EWI was calculated using the following Eq:

$$\text{EWI (mg/kg Week}^{-1} \text{ Person}^{-1}) = \frac{C_{\text{rice}} \times (w_{\text{rc}} \text{ rice/body weight})}{\times 10^3} \quad (3)$$

where, C_{rice} = mean of heavy metals concentration in rice (mg kg^{-1}), w_{rc} rice = weekly rice consumption (g week^{-1}) per capita of the Iraqi population ($110 \text{ g per capita per day} \times 7$) and body weight = average of body weight (kg) of the Iraqi people (60 kg).

3. Results

The emission rate of alpha particles ($0.0249 \text{ mBq cm}^{-2}$) was found to be the highest in basmati rice (Southern Iraqi Company), whereas the lowest rate ($0.0092 \text{ mBq cm}^{-2}$) was found in Indian basmati rice (International Company) as shown in Table 1. Figures 1 and 2 show the tracks of studied samples of the rice.

The study revealed that the measurement rates observed in the sampled rice were reasonable and do not cause cancer. No evident increase was observed in the measurement when compared to the set measurement by global research. The rice samples were free of radioactive contamination. The heavy metals concentration (HMC) was found in the sampled rice. HMC for Fe was found to be (2.7237 mg/kg) in basmati rice (Southern Iraqi company), whereas the lowest HMC (0.3997 mg/kg) was found in American basmati rice (General Company for Grain Trade) as shown in Table 2. The HMC for Cd was found to be (0.0468 mg/kg) in Nasiriyah Anbar rice, whereas the lowest HMC (0.0034 mg/kg) was found in Indian basmati rice (International Company). These values were found to be below the wholesome food standards of Cd (0.5 mg/kg) as shown in Table 2. HMC for Pb was found to be (0.2431 mg/kg) in Babylon Anbar rice, whereas the lowest HMC (0.0695 mg/kg) was found in Indian basmati rice (International Company) as shown in Table 2.

The results in Table 3 show statistically significant differences between HMC of Fe, Cd, and Pb ($p > 0.001$). Overall investigation of the cadmium in rice varied from country to country and revealed a range between 0.0008 mg/g to 0.13 mg/g (Watanabe et al., 1996). The lead content in the rice samples from the various countries were observed to be in the range from 0.0016 mg/g to 0.0583 mg/g (mean = 0.0157 mg/g) (Zhang et al., 1996).

The WHO and FAO organizations provided a guideline on the intake of the heavy elements of the human body. PTWI recommended by the

Table 1. Information and rate of emission of alpha particles about the rice from Najaf city.

Company Name	Tracks $\text{cm}^{-2}\text{d}^{-1}$	E_α (mBq cm^{-2})	\pm SE of Mean
Nasiriyah anbr rice	1.768	0.0176	0.0009
Indian basmati rice (public company)	2.121	0.0202	0.0008
Indian rice (Gold Company)	2.095	0.0209	0.0008
Salahuddin anbr rice (Iraqi origin)	1.525	0.0152	0.0015
American basmati rice	2.037	0.0203	0.0020
Basmati rice (Southern Iraqi company)	2.512	0.0249	0.0014
Indian rice (Baghdad Company)	1.499	0.0149	0.0016
Indian basmati rice (International Company)	0.928	0.0092	0.0009
Indian rice (Cart Oriole Astalion)	1.941	0.0194	0.0010
Babylon anbr rice	1.762	0.0176	0.0014
Long grain white rice (Uruguay)	1.159	0.0115	0.0011
American basmati rice (General Company for Grain Trade)	1.313	0.0129	0.0014
Average		0.0169	0.0012

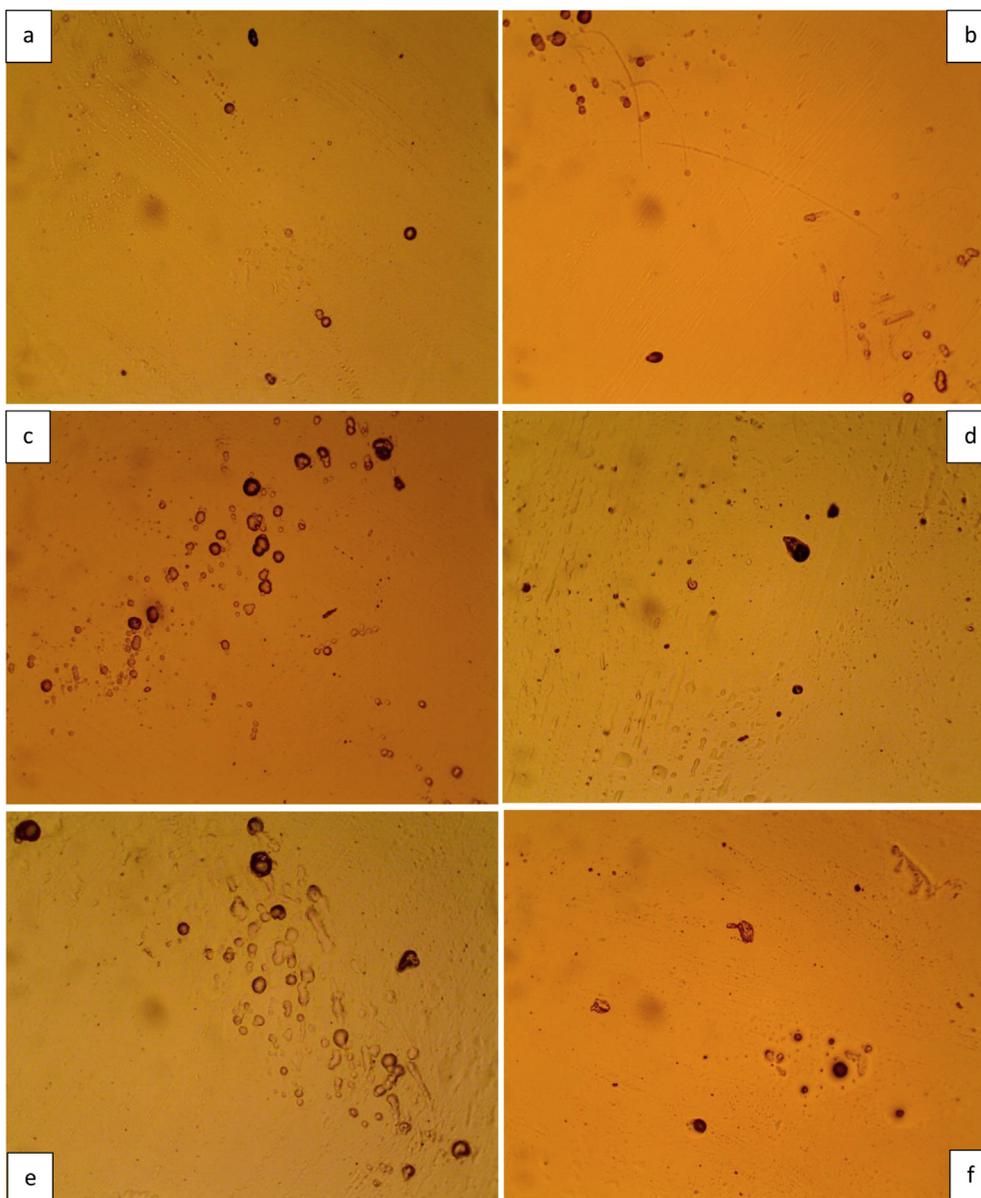


Figure 1. Tracks in the typical rice samples, a: Indian basmati rice (public company), b: Nasiriyah anbr rice, c: Salahuddin anbr rice (Iraqi origin), d: Babylon anbr rice, e: American basmati rice, f: Basmati rice (Southern Iraqi company).

Joint WHO/FAO Expert Committee of Pb and Cd is $25 \mu\text{g kg}^{-1}$ and $7 \mu\text{g kg}^{-1}$, respectively (FAO/WHO, 1972; IARC, 1993). The concentration of heavy metals in rice varied depending on their country of origin. The average level of Cd in Iranian rice was 0.41 mg/g (Afshin et al. 2007). Mean concentrations of Cd and Pb in Australian rice were 0.0075 mg/g and 0.375 mg/g dry weight, respectively (Rahman et al., 2014). Many national and international organizations worldwide (WHO, FAO, USEPA, FSANZ, ANZFA, and IEFES) set tolerable dietary intakes (TDI) limit for the heavy metals depending on body weight and age of the consumers. The EWI of cadmium, lead, and iron was calculated that was based on the average consumption of rice (110 g d^{-1}) as shown in Table 4. Table 4 shows the PTWI for the investigated heavy metals. The estimated intake of Cd, Pb, and Fe in rice consumption was lower than the PTWI (Table 4).

The highest EWI concentration (0.02576 mg/g) for Pb was observed in some imported rice samples in Iran markets (Naseri et al. 2015). Estimated daily intake (EDI) for Cd and Pb of rice in China exceeded the FAO/WHO recommended limit (Zhuang et al., 2009). Health risk will

increase with the consumption of other contaminated food that is not evaluated in this work.

However, rice E_{α} is positively correlated with Fe, Cd, and Pb, as shown in Figure 3. Rice Pb is negatively correlated with Fe and Cd, whereas rice Cd is positively correlated with Fe, as shown in Figure 4. There was a statistically significant correlation between EAPR and HMC in examining rice at $p > 0.05$ (Table 5).

The total heavy metal concentrations of affected rice reported with heavy metals from different countries is presented in Table 6. It was found in this study that heavy metals were within the data of literature reviews.

4. Discussion

These rates in rice were reasonable and alpha particles do not cause cancer, which has no evident increase in these measurements in comparison by global research. So the rice is free of radioactive contamination. The results show statistically significant differences between HMC

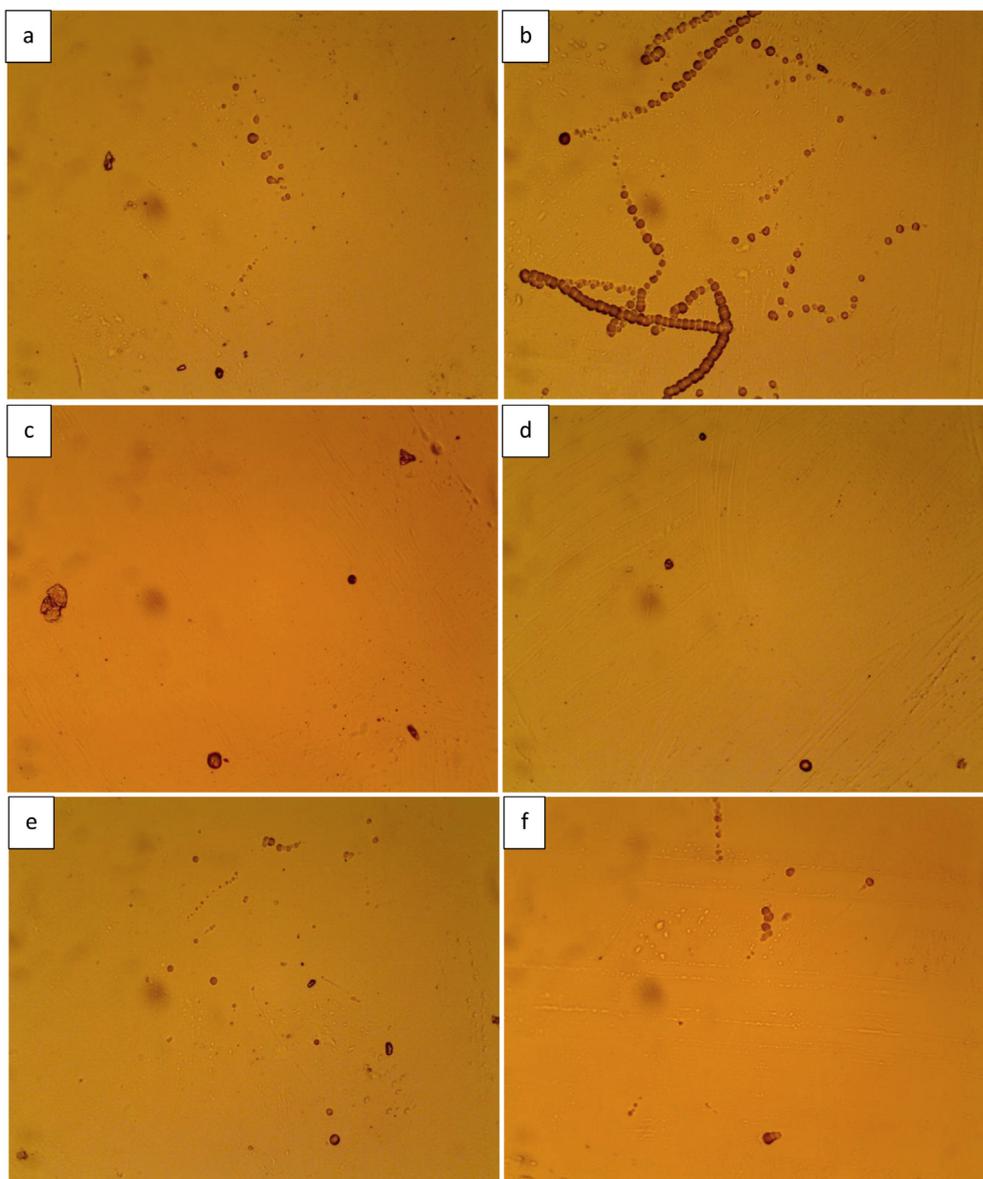


Figure 2. Tracks in the typical rice samples, a: Long grain white rice (Uruguay), b: Indian rice (Cart Oriole Astalion), c: Indian rice (Baghdad Company), d: Indian basmati rice (International Company), e: Indian rice (Gold Company), f: American basmati rice (General Company for Grain Trade).

Table 2. The heavy metals concentration (mg/kg) in the rice samples.

Company Name	Pb	Cd	Fe
Nasiriyah anbr rice	0.0820	0.0468	1.5724
Indian basmati rice (public company)	0.1024	0.0271	1.0321
Indian rice (Gold Company)	0.1434	0.0198	1.0780
Salahuddin anbr rice (Iraqi origin)	0.1332	0.0183	1.0449
American basmati rice	0.0820	0.0227	1.1697
Basmati rice (Southern Iraqi company)	0.2084	0.0196	2.7237
Indian rice (Baghdad Company)	0.1127	0.0212	2.0464
Indian basmati rice (International Company)	0.0695	0.0034	1.5417
Indian rice (Cart Oriole Astalion)	0.1042	0.0088	0.4511
Babylon anbr rice	0.2431	0.0061	0.6395
Long grain white rice (Uruguay)	0.1736	0.0088	0.4340
American basmati rice (General Company for Grain Trade)	0.1389	0.0088	0.3997
Average	0.133	0.018	1.178

Table 3. Analysis of variance results of HMC of Fe, Cd, and Pb.

Source of variation	Sum of squares	Degrees of freedom	Mean square	F ratio	p-value
Between groups	9.805	2	4.902	29.466	0.0000
Within groups	5.490	33	0.166		
Total	15.295	35			

of Fe, Cd, and Pb ($p > 0.001$). Overall investigation of the cadmium in rice from various countries revealed a range between 0.0008 mg/g to 0.13 mg/g. The lead content in the rice from various countries ranged from 0.0016 mg/g to 0.0583 mg/g (mean = 0.0157 mg/g). The WHO and FAO organizations provided a guideline on the intake of the heavy elements of the human body. PTWI was recommended by the Joint WHO/FAO Expert Committee of Pb and Cd. The concentration of heavy metals in rice varied depending on their country of origin. Many worldwide national and international organizations (WHO, FAO, USEPA, FSANZ, ANZFA, and IEFs) set tolerable dietary intakes (TDI) limit for the heavy metals depending on body weight and age of the consumers. The

Table 4. Calculated EWI values for rice types (mg/kg week⁻¹ person⁻¹).

Company Name	Pb	Cd	Fe
Nasiriyah anbr rice	0.0010	0.00060	0.0201
Indian basmati rice (public company)	0.0013	0.00034	0.0132
Indian rice (Gold Company)	0.0018	0.00025	0.0138
Salahuddin anbr rice (Iraqi origin)	0.0017	0.00023	0.0134
American basmati rice	0.0010	0.00029	0.0150
Basmati rice (Southern Iraqi company)	0.0026	0.00025	0.0349
Indian rice (Baghdad Company)	0.0014	0.00027	0.0262
Indian basmati rice (International Company)	0.0008	0.00004	0.0197
Indian rice (Cart Oriole Astalion)	0.0013	0.00011	0.0057
Babylon anbr rice	0.0031	0.00007	0.0082
Long grain white rice (Uruguay)	0.0022	0.00011	0.0055
American basmati rice (General Company for Grain Trade)	0.0017	0.00011	0.0051
PTWI	0.025	0.007	Ns*

* There is no PTWI set for Iron.

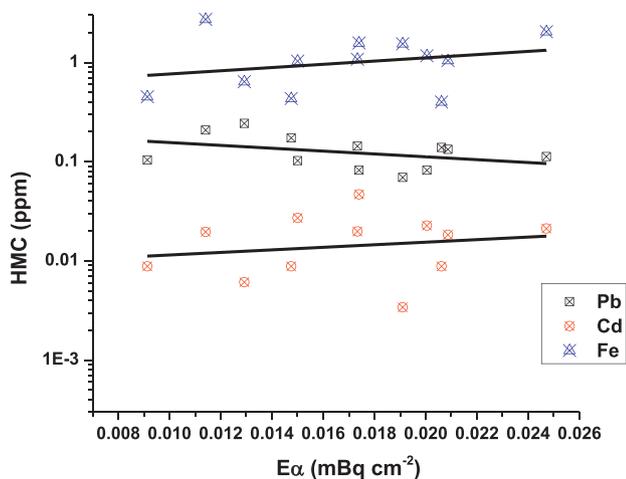


Figure 3. Relationship between EAPR and HMC of Fe, Cd, and Pb in the rice.

EWI of cadmium, lead, and iron was calculated that was based on the average consumption of rice. The estimated intake of Cd, Pb, and Fe in rice consumption was lower than the PTWI. Nevertheless, health risk will increase with the consumption of other contaminated food. However, rice $E\alpha$ is positively correlated with Fe, Cd, and Pb. Rice Pb is negatively correlated with Fe and Cd, whereas rice Cd is positively correlated with Fe. There was a statistically significant correlation between EAPR and HMC in examining rice at $p > 0.05$.

Table 5. Analysis of variance results of HMC and EAPR.

Source of variation	Sum of squares	Degrees of freedom	Mean square	F ratio	p-value
Between groups	11.43558	3	3.8118	30.5468	0.0000
Within groups	5.49065	44	0.1247		
Total	16.92623	47			

Table 6. Comparison of heavy metal concentrations (mg/kg) in rice of the present study and that of some previous studies.

Country	Pb	Cd	Fe	References
Turkey	5.20	Demirel et al. (2008)
Australia	0.4	6.7×10^{-4}	...	Rahman et al., (2014)
Iran	0.916	0.022	...	Ghazanfarirad (2014)
India	...	0.078	...	Meharg et al., (2013)
India	0.003–0.218	Shraim (2014)
Bangladesh	0.713	0.088	...	Ahmed et al. (2015)
China	0.034–0.076	0.15	...	Yu et al. (2016)
USA	0.014–0.098	0.018	...	Shraim (2014)
Japan	...	0.065	...	Kawada and Suzuki (1998)
Korea	...	0.04	...	Meharg et al., (2013)
Spain	...	0.031	...	Kawada and Suzuki (1998)
Iraq	0.133	0.018	1.178	This study

5. Conclusions

The variation in alpha emission rates in the collected rice from Najaf city may depend on the transfer rate of natural radionuclides from water and soil to rice plants. All the heavy metal concentrations were below the recommended limit. Pb, Cd, and Fe varied according to the following order: Fe > Pb > Cd. EWI for cadmium and lead in rice is lower than the PTWI. Thus the results conclude that the best rice as food for humans is Indian basmati rice (International Company) because it has less heavy elements of cadmium and lead that cause poisoning. Also, Indian basmati rice has low alpha emission rate and the relationship between EAPR and HMC is a positive correlation and there is statistically significant at $p > 0.05$. This study showed that alpha particle emission rates are a regular rate of radioactivity present in rice. Overall, the result shows that alpha activities of rice is low and does not cause any dangerous effects on human beings. This means that the rice in this study was free of environmental pollution of the alpha particles. The present work has determined the exposure and intake rates of alpha particles emission and heavy metals. According to the correlations between heavy metals and alpha particles, it can be concluded that this study stands as a real marker of environmental pollution in consumed rice in Najaf city.

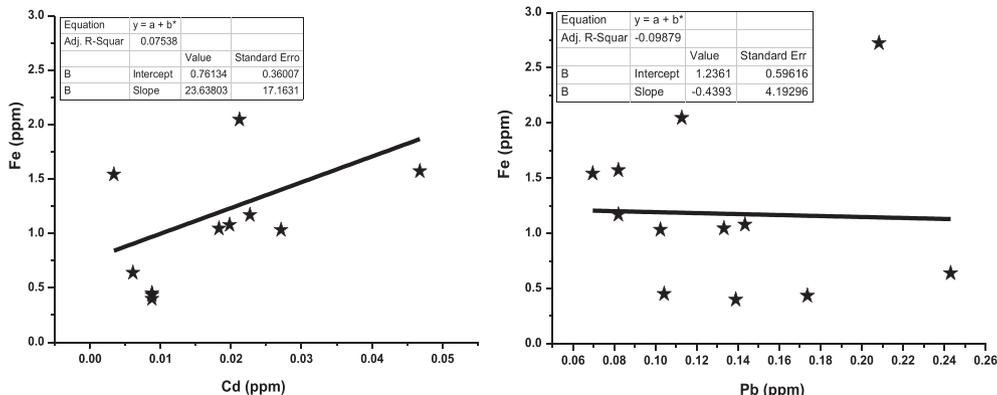


Figure 4. Relationship between HMC of Fe, Cd, and Pb in the rice.

Declarations

Author contribution statement

Basim Almayahi: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Naheda Aljarrah: Contributed reagents, materials, analysis tools or data.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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