Heavy metals in some date palm fruit cultivars in Saudi Arabia and their health risk assessment

Khaled F. Salama, Muhammad Atif Randhawa, Abdulaziz Abdulrahman Al Mulla, and Osama Ahmed Labib

Department of Environmental Health, College of Public Health, Imam Abdul Rahman Bin Faisal University, Dammam, Kingdom of Saudi Arabia

ABSTRACT
Date fruits have ample amount of beneficial minerals but owing to diverse causes may contract with heavy metals and contaminants during field, processing, or transportation posing health challenges to consumers. Hence, current research explored the toxic heavy metals (Al, As, Cr, Pb, Cd, and Sb) levels in seven date varieties (Sakay Mabroum, Sakay Normal, Rashadya Al-qaseem, Barny Al-Madina, Eklas Al-Hassa, Safawy Al-Madina, Kadary) collected from different locations of Saudi Arabia by applying inductively coupled plasma optical emission spectroscopy. Mean values of heavy metals were calculated and expressed. Most of the heavy metals with the exception of As, Pb, and Cd are within safe limit with respect to maximum allowable levels (MAL) in some date cultivars. Estimated daily intake (EDI) of heavy metals through dates consumption (100 gram/person/day) for As and Pb exceeded the Provisional maximum tolerable daily intake from some locations. Although Cd content was exceeding MAL; however, its EDI is well within safe limits of Provisional maximum tolerable daily intake. From EDI data, hazard risk index was also calculated, which predicted that both As and Pb can pose potential health hazards to dates consumers in Saudi Arabia. However, Pb contributed greater health risks due to date fruit consumption as compared to Arsenic.

ARTICLE HISTORY
Received 27 May 2019
Revised 10 September 2019
Accepted 13 September 2019

KEYWORDS
Date palm; Heavy metals; Locations; Daily intake; Health risk

Introduction
Date palm (Phoenix dactylifera) is native to Arab soils and has numerous varieties, each having its own taste and nutritional profile. The preferred habitats considered for date palm is arid and semi-arid territories. It is rich in various minerals and health enhancing entities. Saudi Arabia is the second largest producer of date palm with 1.07 million tons production as compared to other countries. [1] Mineral and some transition metals play important role in health of human beings but surpassing of safety level imposes toxicity, cancer, and even genotoxicity. Among the heavy metals (HMs), chromium (Cr), cadmium (Cd), aluminum (Al), lead (Pb), copper (Cu), zinc (Zn), arsenic (As), boron (B), and metalloids are considered to have bad impact on health and agriculture. Heavy metals (HMs) are substances that have ability to cause toxicity in living tissues and can cause cancer at drastic levels. These HMs are being increased due to numerous environmental factors like mining, fertilizers applications to soils, industrial emissions, metallurgical operations, and non-biodegradation of various metals. These can cause stress in plants leading to augmentation in their tissues conferring negative consequences in consumers. [2] Monitoring of HMs in date palm is very crucial as it has ability to grow in diverse climatic conditions and grows easily along the roads, residential zones, rural areas, and industrial zones where they are vulnerable to pick pollutants. [3–5]
Various elemental analytical instrumental techniques are available now-a-days to assess HMs to safeguard consumers. Among the routine analysis, neutron activation analysis, proton induced X-ray emission, electro thermal atomic absorption, proton induced X-ray emission, and inductively coupled plasma are applied because these methods give good indication of macro and micro minerals from dates. Current study utilized inductively coupled plasma – optical emission spectroscopy (ICP-OES) for determination of HMs in dates.

Date palms fields are irrigated sometimes by using sewage water, which can be hypothesized to HM toxicity. HMs accumulate in various parts of the food crop that leads to a variety of clinical problems in animals and human all over the world. Humans are usually exposed to these metals through ingestion or breathing as involuntary act and when their concentration increases, above the maximum allowable levels (MALs) set by WHO/FAO, on set of various diseases is triggered as HMs are known mutagenic, teratogenic, and carcinogenic materials. The risks by taking HMs in relation to human exposure are poor health, cirrhosis, irreversible brain damage (encephalopathy), renal dysfunction, bone diseases, cardiovascular diseases, hemorrhage, dementia, systemic cancers especially gastrointestinal cancers and also affect sperm motility. Moreover, macromolecular structures are also disturbed by HMs, hence posing threat to DNA, proteins, and fragile tissues of living things. Therefore, for checking safety status of date palm, HMs should be analyzed and compared with permissible limits in order to avoid health issues.

The presence of lead, cadmium, and other metals have been reported in the street dust of Riyadh city and lead (Pb) concentration depends on traffic load and other industrial activities. Aldjain et al. collected dates from 14 locations of Riyadh and found that dates were contaminated with significant levels of lead and cadmium; however, their values were within acceptable limit set by WHO/FAO. The crop variety also affects the lead uptake from soil. Ali and Al-Qahtani analyzed vegetables, cereals, and fruits grown in four major industrial and urban cities of Saudi Arabia and found that the concentration of major elements studied were exceeding the MALs prescribed by WHO/FAO. To the best of our knowledge, no detailed studies are available regarding HMs in dates grown in various regions of Saudi Arabia. The objectives of this study were to

Figure 1. Different locations of collected date cultivars in Saudi Arabia. (a) Alkarj. (b) Al-Qaseem. (c) Al-Madina. (d) Al-Hassa.
determine HMs in different date cultivars and estimation of their daily intake assessment from consumption of dates. The health hazards of metals to human beings are also calculated.

**Material and methods**

**Sample procurement and heavy metals determination**

One kilogram date fruit of seven popular date varieties were procured from cities of Kingdom of Saudi Arabia as detailed under: Sakay Mabroum and Sakay Normal from Alkarj, Rashadya and Kadary from Al-Qaseem, Barny and Safawy from Al-Madina, and Eklas from Al-Hassa. The date fruits from designated varieties were collected during 2017 from different locations of Saudi Arabia (Figure 1) at mature ripened stage, washed in portable water to remove dirt. After washing the dates were air dried and placed in polyethylene zip lock bags and kept under refrigerated conditions and brought to the Environmental Safety Laboratory, College of Public Health at Imam Abdulrahman Bin Faisal University, Dammam. Nearly 0.5 kg deseeded date fruit of each variety was mashed separately to get homogenous and representative sample. After that, 1 g sample was weighed accurately and digested using 12 mL of a mixture (2:1 v/v) of concentrated HCl and HNO$_3$ acids (Sigma-Aldrich, St. Louis, MO, USA). The mixture was heated for 45 min in a microwave accelerated reaction system (MARS 6) at a temperature and pressure not more than 180°C and 200 psi, respectively. After completion of digestion, the solution was passed through Whatman No. 42 ash less filter paper and 25 mL volume made with deionized water. The samples were stored in polyethylene bottles at refrigerated temperature till analysis. HMs (Al, As, Cr, Pb, Cd, Sb) were analyzed on iCAP 6300 Duo Inductively Coupled Plasma – Optical Emission Spectrophotometer (ICP-OES, Thermo Fischer Scientific). The details of instrument standardization and process parameters were as follows: Operating parameters for ICP-OES were so that axial view of metals at their specific wavelength was monitored. Quantification of results was based on the processing of peak area of individual element. The method was found optimum in terms of LOD as well as recovery of metals from spiked samples (Table 1). The detection limit is very important in method validation and trace analysis as it is the lowest concentration of the analyte that can be reliably detected, and is a reflection of the precision of the instrumental response obtained by the method. The recovery was also found more than 90% and $r^2$ value for all the tested metals was 0.999. All the experiments were performed in three replicates and results are expressed as mean.

**Dietary intake assessment**

Daily intake (mg/person/day) of metals was calculated by multiplying concentration of specific metal with the per capita daily consumption of dates and divided by average body weight of human adult.$^{[14]}$ The assessment of daily intake of HMs through consumption of dates were calculated and compared with recommended values of WHO/FAO. According to Al-Mssallem,$^{[15]}$ per capita date consumption in Saudi Arabian adults is about 100 g wet wt./person/day, whereas average body weight of a Saudi Citizen is 71 kg.$^{[16]}$ The Estimated Daily Intake (EDI) values were thus calculated for the seven date varieties collected from different locations and compared with provisional

| Element | Wavelength (nm) | Recovery % (Mean±SD) | LOD (µg/L) |
|---------|----------------|----------------------|------------|
| Al      | 308.22         | 90.25 ± 1.25         | 5.0        |
| As      | 193.70         | 92.65 ± 2.56         | 5.0        |
| Cr      | 267.72         | 97.43 ± 1.86         | 2.5        |
| Pb      | 220.35         | 95.25 ± 1.62         | 5.0        |
| Cd      | 226.50         | 98.27 ± 2.38         | 2.5        |
| Sb      | 206.83         | 101.05 ± 2.49        | 2.5        |
tolerable daily intake (PTDI) or Provisional maximum tolerable daily intake established by world health regulatory agencies (Table 3).

**Health risk index**

Daily intake assessment values of HMs were also used for determining health risk index of HMs in the dates. Health risk index was calculated by dividing daily intake of metals in date varieties by the oral reference dose as described by Khan et al. (2013), and shown below:

\[ \text{HRI} = \frac{\text{DIM}}{\text{RfD}} \]

The HRI > 1 for any metal means that the consumer/population is at health risk.

**Statistical analysis**

Results were analyzed using descriptive statistics where means, median, and ranges were calculated to have estimation of metallic residues from date palm. The calculations were done on Minitab and excel software and results were rounded to three decimal places.

**Results and discussion**

**Screening of toxigenic elements from date palm**

Factorial under CRD was applied to chalk out significance of metal contamination in date as function of location and metal type (Table 2). Analysis revealed highly significant effect of location and metal contamination. Among the individual metals: arsenic, antimony, and cadmium depicted highly significant difference in concentration depending upon locations while aluminum, chromium, and lead illustrated significant effect depending upon locations.

Table 3 describes the mean concentration of all metals in analyzed date cultivars from various locations along with their Maximum Allowed Limits (MAL) prescribed by FAO/WHO (2011). FAO/WHO has not established limits of toxic metals in dates; therefore, general MAL for studied metals

| Source | DF | Aluminum | Arsenic | Chromium | Lead | Cadmium | Antimony |
|--------|----|----------|---------|----------|------|---------|----------|
| Date Types (Location) | 6 | 428.468* | 0.62325** | 0.54572* | 6241.96* | 0.10784** | 0.00534** |
| Error | 14 | 0.625 | 0.00071 | 0.00049 | 4.61 | 0.00016 | 0.00002 |

* = Significant.  
** = Highly significant.

**Table 2. Mean squares for location and heavy metal contents in date palm.**

| Metals | S Mabroum (Al-Karj) | S Normal (Al-Karj) | Rashadya (Al-Qaseem) | Barny (Al-Madina) | Eklas (Al-Hasaa) | Safawy (Al-Madina) | Kadary (Al-Qaseem) | Mean *MAL |
|--------|---------------------|-------------------|----------------------|-------------------|-----------------|-------------------|-------------------|----------|
| Al     | 1.141E              | 5.513C             | 5.099C               | 7.047B            | 15.202A         | 4.151D            | 0.923E            | 5.582B   |
| As     | 0.095DE             | 0.109CD            | 0.079E               | 0.078E            | 0.584A          | 0.132B            | 0.121BC           | 0.171C   |
| Cr     | 0.070B              | 0.042C             | 0.508A               | 0.031C            | 0.071B          | 0.085B            | 0.045C            | 0.122C   |
| Pb     | 2.859C              | 0.493D             | 50.28A               | 0.479D            | 9.417B          | 0.665D            | 1.206D            | 9.343A   |
| Cd     | 0.159B              | 0.203A             | 0.025E               | 0.004F            | 0.084D          | 0.027E            | 0.131C            | 0.090C   |
| Sb     | 0.046B              | 0.017E             | 0.032D               | 0.073A            | 0.038C          | 0.040C            | 0.044B            | 0.041C   |
| Mean   | 0.728E              | 1.063CD            | 9.337A               | 1.285C            | 4.233B          | 0.850DE           | 0.412D            | –        |

Similar lettering in a row shows no significant difference

*MAL means maximum allowable limit (mg/kg) prescribed by WHO/FAO 2011.[54]

– means no limit defined
advised by FAO/WHO in food stuff is used as reference material for comparison and safety evaluation of date cultivars.

Among the metals, aluminum depicted highest concentration 15.202 (mg kg\(^{-1}\)) from Eklas (Al-Hasaa) followed by Barny (Al-Madina) and minimum 0.923 (mg kg\(^{-1}\)) from Kadary (Al-Qaseem). However, Rashadya (Al-Qaseem) and Sakay Normal (Al-Karj) exhibited non-significant differences regarding aluminum concentration. As arsenic is concerned, 0.584 (mg kg\(^{-1}\)) was the maximum content from Eklas (Al-Hasaa) while minimum was 0.078 (mg kg\(^{-1}\)) from Barny (Al-Madina). Moreover, arsenic concentration was in close proximity in Sakay Mabroum (Al-Karj), Sakay Normal (Al-Karj) with levels of 0.095 (mg kg\(^{-1}\)), 0.109 (mg kg\(^{-1}\)), respectively. Meanwhile, arsenic elucidated bit higher values with similar pattern in Safawy (Al-Madina) and Kadary (Al-Qaseem) with values 0.132 (mg kg\(^{-1}\)) and 0.121 (mg kg\(^{-1}\)), correspondingly. Arsenic was below allowable limits 0.1 (mg kg\(^{-1}\)) in Sakay Mabroum (Al-Karj), Rashadya (Al-Qaseem), and Barny (Al-Madina) while it was above in Eklas (Al-Hasaa), Safawy (Al-Madina), and Kadary (Al-Qaseem). Chromium never violated the limit 2.3 (mg kg\(^{-1}\)) and elucidated numerical values ranging from 0.031 (mg kg\(^{-1}\)) in Barny (Al-Madina) to 0.508 (mg kg\(^{-1}\)) Rashadya (Al-Qaseem), respectively. Lead exhibited drastically highest value 50.28 (mg kg\(^{-1}\)) from Rashadya (Al-Qaseem), i.e. very much above the limit 0.3 (mg kg\(^{-1}\)), nevertheless Sakay Normal (Al-Karj) and Barny (Al-Madina) reported minimum and at par values 0.493 (mg kg\(^{-1}\)) and 0.479, respectively. Lead contents were exceeding the maximum allowable limits in all the date types as well as locations. Cadmium depicted escalation in residues near border line 0.2 (mg kg\(^{-1}\)) in one location like Sakay Normal (Al-Karj) 0.203 (mg kg\(^{-1}\)), whereas its concentration was quite below in other spots regarding cadmium. Nonetheless, antimony depicted peculiar behavior and residues were in resembling pattern like 0.046 (mg kg\(^{-1}\)), 0.017 (mg kg\(^{-1}\)), 0.032 (mg kg\(^{-1}\)), 0.073 (mg kg\(^{-1}\)), 0.038 (mg kg\(^{-1}\)), 0.04 (mg kg\(^{-1}\)), and 0.044 (mg kg\(^{-1}\)) in locations viz Sakay Mabroum (Al-Karj), Sakay Normal (Al-Karj), Rashadya (Al-Qaseem), Barny (Al-Madina), Eklas (Al-Hasaa), Safawy (Al-Madina), and Kadary (Al-Qaseem), respectively showing less metal contamination.

Results demonstrated different contamination levels of HMs in different date palm fruits cultivars collected from various locations. Aluminum, chromium, and antimony from all locations and in all cultivars were less than the MAL with mean concentrations of 5.582, 0.122, and 0.041 mg/kg, respectively. This shows that aluminum, chromium and antimony concentrations are in safe limits. Arsenic levels in four date cultivars namely Eklas Al-Ahsaa, Sakay Normal, Safawy Al-Madina, and Kadary are exceeding the MAL with mean values of 0.584, 0.109, 0.132, and 0.121 mg/kg, respectively. The arsenic contents of remaining cultivars is also higher than the half value of MAL, which mean that arsenic contamination can be serious health risk for human beings. In contrast to arsenic, cadmium contents are only found higher than MAL in Sakay Normal (0.203 mg/kg) date cultivar. However, the results for lead are alarming, as higher than MAL is observed in all the cultivars collected from different locations. Sakay Normal is the only cultivar that possessed higher level of arsenic, cadmium, and lead meaning that it can be a potential source of combined risk for mankind. Sakay Normal was procured from Alkarj, which means that this area has more pollution problem because Sakay Mabroum was the other date variety collected from same area and it also contains Ar, Pb, and Cd levels nearly approaching to the MAL but not exceeding the limits. This shows presence of cumulative or combined risk. HMs are natural part of the soil formation and their concentration in soil also increases with increase in anthropogenic activities. In current investigation, soil HM profiling was not carried out and no prior studies are available in literature; therefore, finding out exact cause of HM contamination in this area is not clear.

The boost of lead concentration may be attributed to various causes where automobile contribute to a greater extent followed by the use of lead based pesticides in crop protection. The deposit of lead on edible tissues of crops also depends upon air dust, air pollution, distance from the roads side as well as traffic loads and period of exposure of commodities along roadside for selling. Hussain et al.\textsuperscript{[17]} analyzed dates imported into Kuwait and found that dates collected from different location were differing in Pb contents and some of the samples were exceeding the MAL values. Radwan and Salama\textsuperscript{[18]} during market basket survey for some HMs in Egyptian fruits and vegetables found that Pb contents in dates were 0.22 mg/kg and even higher Pb contents were observed in strawberries, peach, and melon. However, the Cd contents were within
acceptable limit in dates and other fruits. Cadmium can result in diseases of kidney, lungs and also can lead to skeleton problems as it is related with cancerous risk as compared to lead which is non-cancer risk factor. Current study elucidated low level of Cd as compared to Pb revealing uptake of HMs owing to plant ability to accumulate them.\textsuperscript{[19,20]} Cadmium can be deposited in the edible tissues due to agricultural practices as Cd can via usage of polluted sewage water, type of soil, and the application of phosphate fertilizers. Aluminum causes damage to brain cells and damages the bones subsequently causing anemia in individual subjected to hemodialysis. Further, Al also caused toxic outcomes in stems, roots, and green leaves causing wilting and growth retardation also in plants.\textsuperscript{[21,22]} The average concentration levels of Sb ranged between 0.017 in Sakay Normal to 0.073 mg/kg in Barny Al-Madina. Chromium contamination has been attributed to unwashed plants and their ability to concentrate these HMs from soil, environment, and irrigation water.\textsuperscript{[23,24]} Arsenic is toxic to plants especially for palm causing wilting, stunted growth, and necrotic lesions and even cancer to consumers. According to some researchers, its levels above 5 mg/kg impart phytotoxicity.\textsuperscript{[25,26]} Conclusively, metals uptake by plants depends upon species of plants, which introduces these hazards in food chain causing health threats.\textsuperscript{[27–29]}

**Estimated daily intakes of metals from date palm**

Table 4 represents the EDIs of HMs from date fruits. Depending upon the 100 g of date palm consumption on daily basis per person in Saudi Arabia,\textsuperscript{[8]} the EDIs of HMs have been determined for each detected metal in particular location. Whereas, per capita dates consumption data for children is not available in the Kingdom of Saudi Arabia; therefore, health risk estimation for children is a bit difficult. However, children are also consuming dates and health hazard would be similar as mentioned for the adults below.

EDIs have been calculated based on date fruits only, and without considering the pits. Provisional maximum tolerable daily intake values for different HMs studied in current investigation are also shown in Table 4.\textsuperscript{[30–33]} Maximum level of EDI of aluminum was observed in dates collected from Eklas Al-Ahsaa (1520.17 µg/person/day); however, considering Provisional maximum tolerable daily intake for Al, this level is well within safe limits. Likewise, maximum EDIs (mg/person/day/100 g) of Cr and Sb were well within the Provisional maximum tolerable daily intake values. It means that Saudi Consumers do not have any health risk due to presence of Al, Cr and Sb in date cultivars.

EDIs for arsenic showed that only Eklas Al-Ahsaa cultivar is exceeding the Provisional maximum tolerable daily intake value, whereas, higher arsenic levels than MAL were found in four date cultivars including Eklas Al-Ahsaa. EDIs values showed that only Eklas Al-Ahsaa date cultivar is potential threat for human beings as its EDI value (58.37 µg/person/day) is not only higher but more than double of the Provisional maximum tolerable daily intake value (25.2 µg/person/day). On the other extreme, lowest EDIs for arsenic were found in varieties Barny Al-Madina followed by Rashadya Al-Qaseem. For cadmium, EDIs for all the cultivars tested in current study are within safe limits of Provisional maximum tolerable daily intake, although Cd contents in Sakay Normal was higher than MAL. However, EDIs data showed that Saudi citizen do not have any threat due to cadmium intake through this variety, even it is having Cd contents higher than the MAL values. Three date varieties namely

|        | Sakay Normal | Sakay Mabroum | Rashadya Al-Qaseem | Barny Al-Madina | Eklas Al-Ahsaa | Safawy Al-Madina | Kadary     |
|--------|--------------|--------------|--------------------|----------------|---------------|----------------|------------|
| EDIs   |              |              |                    |                |               |                | Provisional maximum tolerable daily intake |
| Al     | 114.05       | 551.26       | 509.89             | 704.70         | 1520.17       | 415.05         | 92.30      |
| As     | 9.51         | 10.90        | 7.93               | 7.75           | 58.37         | 13.18          | 12.10      |
| Cr     | 7.00         | 4.21         | 50.82              | 3.14           | 7.08          | 8.45           | 4.50       |
| Pb     | 285.90       | 49.33        | 5028.00            | 47.85          | 941.69        | 66.45          | 120.60     |
| Cd     | 15.89        | 20.82        | 2.52               | 0.37           | 8.40          | 2.73           | 13.08      |
| Sb     | 4.62         | 1.66         | 3.22               | 7.33           | 3.83          | 4.03           | 4.36       |

*Estimated daily intake µg/person/day/100 g Date Palm Fruit.
**Provisional maximum tolerable daily intake (µg/person/day).\textsuperscript{[30–33]}*
Sakay Mabroum, Rashadya Al-Qaseem, and Eklas Al-Ahsaa are potential threat for Saudi citizens because of higher EDIs of Pb as compared to Provisional maximum tolerable daily intake values. However, it is very interesting to note that all seven varieties were having Pb contents more than the MAL values. Rashadya Al-Qaseem date variety can be toxic for human beings due to exceeding Provisional maximum tolerable daily intake value of Pb by more than 20 folds.

**Health risk index**

In order to really access health risks, health risk index values were calculated based upon the EDIs and oral reference dose of each toxic metals for all of the date cultivars. The results illustrated that no health risk is involved in consumption of date varieties for aluminum, chromium, cadmium, and antimony. Eklas Al-Ahsaa date cultivar can be potential threat with respect to arsenic and lead contents as the health risk index values calculated are exceeding than 1 (Table 5). This date variety is very potential health risk as it exceeded the combined risk of arsenic and lead metals. For Pb, health risk index is maximum in Rashadya Al-Qaseem cultivar (17.704) against the value of 1. Sakay Mabroum date cultivar also exceeded for health risk index for lead contamination.

**Conclusion**

Current study overall revealed that toxic elements were found in date palm fruit samples secured from various points of Saudi Arabia as well as in dates cultivars. Arsenic, lead, and cadmium are exceeding the MAL in some date cultivars, whereas EDIs data suggested that only lead and arsenic are increasing the Provisional maximum tolerable daily intake values. EDI values for all the other metals are well within safe limits of Provisional maximum tolerable daily intake. Health risk index showed that Sakay Mabroum, Rashadya Al-Qaseem, and Eklas Al-Ahsaa date cultivars are unsafe for human consumption by keeping in view the per capita date consumption in Saudi Arabia. This indicted that dates must be monitored for HMs in order to avoid toxicity in humans. No doubt, dates are very good source of the essential elements but exceeding the EDIs and health risk index values can certainly deteriorate the health of consumers, hence demanding more rigorous studies of date palm which are used both in raw and as processed form in various dishes.

**ORCID**

Khaled F. Salama [http://orcid.org/0000-0001-9115-8644](http://orcid.org/0000-0001-9115-8644)

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