



Appraisal of Mineral and Heavy Metal Contents in Peach and Grape Grown at Some Major Agricultural Companies in Saudi Arabia

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ABSTRACT

Extensive use of inorganic fertilizers, insecticides and herbicides may be toxic for plants and cause human health problems. Mineral concentrations determined in peach and grape of three major agricultural Saudi companies. Minerals concentration in peach and grape ranged from 17.43-22.54 mg/kg (Cu), 146-209 mg/kg (Fe), 7.35-11.85 mg/kg (Mn), 11.08-14.62 (Zn), 26.8-25.5 mg/kg (Mg), 122- 177 mg/kg (Ca) and 0.025- 0.18 mg/kg (Cd), 8.22- 12.85 mg/kg (Cu), 109-192 mg/kg (Fe), 7.05-10.15 mg/kg (Mn), 20.31-25.22 mg/kg (Zn), 37.5- 38.3 mg/kg (Mg), 182-302 mg/kg (Ca) and 0.019- 0.12 mg/kg (Cd), respectively. Concentration of (Cu, Fe, and Cd) in fruits was higher than permissible limits.

Key words: Fruits, Heavy metals, Environmental pollution, Provisional tolerable daily intake (PTDI), Tolerable daily upper intake level (UL).

INTRODUCTION

Currently, extensive use of inorganic fertilizers and integrated insect-pest management approach are being adopted for increased fruit production to meet the higher demands for human consumption and to earn more income from the progressive farms. The extensive uses of these resources play an important role in increasing the mineral contents of soils and plant tissues. Large quantities of fertilizers are regularly added

to soils in intensive farming systems to provide adequate N, P, and K for plant growth. The chemical compounds used to supply these elements contain trace amounts of heavy metals (e.g., Cd and Pb) as impurities, which, after continued fertilizer application may significantly increase their content in the soil¹. Some heavy metals are needed in trace amounts as Zn, Cu, Mn and Fe for plants². High concentration of these metals in the agriculture ecosystems may lead to an excessive accumulation of metals, which may be toxic to plants and cause possible health

problems to animals and human³. Contaminated soils represent a real hazard to human health and also limit the area available for agriculture. Turkdogan et al.,⁴ investigated levels of seven different heavy metals in soil, fruit and vegetable samples of Van region in Eastern Turkey, where upper gastrointestinal cancers are endemic. The results showed that the levels of Pb, Cd, Ni, Zn, Cu, Mn, and Co were 409, 25, 53, 44, 70, 160, 269 mg/kg, respectively, which were absolutely higher than the legally permissible limits. Krejpcio et al.,⁵ determined the content of Pb, Cd, Cu and Zn in fresh fruit and juices in the Polish market. It was found that most fruit samples (90.4%) contained low levels of heavy metals. However, the remaining 9.6% contained high concentration of heavy metal (Pb 2.2%, Cd 4.4%, Cu, 1.5%, Zn 1.5%). Most fruit juice sample (88%) met the national standard criteria, but 12% exceeded the permissible limits (as established by the National Ministry of Health of Poland) for Pb and Cd (3% and 9%, respectively). Mahdavian and Somashekar⁶ studied the level of heavy metals in a number of fresh fruits and results showed that the average levels of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were 7.05, 40.93, 75.63, 10.87, 167.39, 61.0, 53.87, 53.87, and 30 mg/kg, respectively. Zahir et al.,⁷ determined levels of major heavy metals in 10 different varieties of fruits in Pakistan, the results were in the range of 7.924-24.674 ug/g Fe, 0.531-7.571 ug/g Pb, 0.013-0.612 ug/g Mn, 0.543-3.234 ug/g Cu, 0.144-5.033 ug/g Ni, 0.173- 0.299 ug/g Cd, 3.268-4.343 ug/g Cr, 0.138-21.409 ug/g Zn, 0.104-1.168 ug/g Co. Furthermore, Pandey et al.,⁸ reported that the metal concentration in soil increased from 8.00 to 12.0 mg/ kg for Cd, and for Zn from 278 to 394 mg/ kg. They also suggested that if the trend of atmospheric deposition is continued, it would lead to a destabilizing effect on sustainable agricultural practice and increase the dietary intake of toxic metals. Human beings are encouraged to consume more fruits, which are a good source of vitamins, minerals, fibers and also beneficial to their health. However, the intake of food contaminated by heavy metals is harmful to human health and several countries have imposed food laws to restrict the presence of heavy metal concentration in food and beverages⁹. Mahdavian and Somashekar⁶ stated that the presence of heavy metals in human body always draws scientific concern as these are considered responsible for affecting health, especially in these days due to the disposal of toxic wastes in the

environment has been increased. The increasing trends in food contamination in urban areas are largely attributed to the polluted environment in urban agriculture, contaminated food transport and supply chains; poor market sanitary conditions, and the use of contaminated or waste water for irrigation purposes.

Healthy plant growth required not only macronutrients (N, P, K, S, Ca, and Mg), but also essential micronutrients such as Co, Cu, Fe, Mn, Mo, Ni, and Zn². Most of the agricultural soils in Saudi Arabia are bare sand with low organic matter, as a result of continuous application of large quantities of fertilizers (inorganic fertilizers and livestock manures) to soils in intensive farming systems to provide adequate nutrients for plants growth. Application of livestock manures (e.g., cattle, sheep, poultry etc.) to land leads to the accumulation of heavy metals such as As, Cd, Cr, Cu, Pb, Hg, Ni, Se, Mo, Zn, Tl, Sb in the soil¹⁰⁻¹¹. Although in poultry industry the Cu and Zn added to diets as growth promoters may also have the potential to cause metal contamination of the soil and if repeatedly applied to restricted areas of land, can cause considerable buildup of these metals in the soil in the long run^{12,13}. Moreover, in integrated insect-pest management, most of the chemicals that are used extensively in agriculture as insecticides and fungicides were based on compounds which contain Cu, Hg, Mn, Pb, or Zn¹. The environmental pollution by heavy metals has not yet reached to critical level in the agriculture ecosystem of Saudi Arabia. A great attention should be paid to highlight the real effects of the possible pollutants induced with the industrial capitals on agricultural areas present near the industrial cities. In some parts of Saudi Arabia, the pollution of the environment with heavy metals has increased dramatically due to the boost in industrial sector¹⁴. A review of literature showed that there is inadequate information available on the mineral composition of agricultural sector in Saudi Arabia to determine toxic levels of some of the heavy metals in fruits for human consumption. Therefore, the main objective of this study was to determine the mineral and heavy metals concentration in peach and grape growing in three major agricultural companies (A, B, and C) in Saudi Arabia and compare its concentrations with the established standards of Joint FAO/WHO¹⁵ the provisional tolerable daily intake (PTDI) and the U.S. National Academy of

Sciences¹⁶ the tolerable daily upper intake level (UL) for human consumption.

MATERIALS AND METHODS

Study Location

Three main agricultural companies (A, B, and C) are the major fruit producers in Saudi Arabia, therefore these three sites were selected for this study.

Collection of Samples

Fruit samples were collected at the peak of the harvest period during May and July, 2012 from farms of the selected companies. The peak of the harvest period was chosen keeping in mind the fact that the metal concentration in plants changes with age of plants and the growing season. Fruits of the selected fruit trees were randomly picked from three locations, wrapped in brown wrapper and transported to analytical laboratory for procurement by drying and analysis.

Preparation of Samples

Fruit Samples were washed first by tap water followed by the de-ionized water, drained properly and shredded before drying. All samples were dried in an air oven at 80°C for about 72 hours, then cooled to ambient temperature, milled by means of a Willy Mill and sieved through 1 mm diameter mesh. The ground samples were packed in airtight plastic containers and stored for analysis. Concentrated HNO₃ (3ml) was added to a 50ml flask containing approximately 0.5 g ground oven-dried plant sample. The flask was swirled gently and the contents digested slowly on an electro-thermal heater to 100°C for 5 hours. The temperature of the digestion mixture was raised gently until it reached 100°C. The digest was then cooled and filtered through 541 Whatman filter paper into a volumetric flask and diluted to 50 ml with distilled water. The residual acid concentration of the digested sample was brought to 1% v/v after digestion.

Analysis of Samples

The digested samples were analyzed for trace metals, using Atomic Absorption Spectrophotometer, Model 451¹⁷. The instrument was calibrated using standard solutions of copper, iron, manganese, zinc, magnesium, Calcium, and

cadmium. The absorbencies obtained were used in calculating the concentrations of the metals in the different samples.

Statistical analysis

Fruit samples were assayed and analyzed individually in triplicate. One way analysis of variance (ANOVA) was used to determine significant difference, considering a level of significance of less than 5% ($p < 0.05$) by using SAS User's Guide¹⁸.

RESULTS AND DISCUSSION

Mineral Concentrations in Fruits of Different Companies

The mineral concentrations (Cu, Fe, Mn, Zn, Mg, Ca, and Cd) of peach and grape fruits grown at different companies are summarized in Table 1 and 2. All the minerals concentration is presented as mg/kg.

Copper (Cu)

Mean concentration of Cu in peach and grape in different companies ranged from 17.4- 22.54 and 8.22-12.85 mg/kg, respectively. Results showed that Cu concentrations in both fruits differ significantly at 0.05 level of significance and the highest concentrations in both fruits were observed in A Company as compared to others. Researchers from different countries in the world have been determined Cu concentrations in different kinds of fruits^{4,5,7,18-22}. While emphasizing the Cu concentrations of peach and grape, it was found that Radwan and Salama²³⁻²⁴ have been reported a mean Cu concentration of 1.46 mg/kg in peach samples collected from Egyptian markets. Tormen *et al.*,²⁵ determined a mean Cu concentration of 150 µg/L in peach juice samples collected from Brazilian markets. Acar²⁴ found that the Cu concentrations of peach juice and peach nectar sampled from different supermarkets in Ankara, Turkey were 5.87 and 5.62 mg/kg, respectively. Sobukola *et al.*,²⁶ also reported a mean Cu concentration of 0.003 mg/kg in grape samples collected from selected markets in Lagos, Nigeria. Bagdatlioglu *et al.*,²⁷ determined a mean Cu concentration of 0.51 mg/kg in grape samples grown in Manisa region, Turkey. Moreover, Elbagermi *et al.*²⁷ reported mean Cu concentrations of 1.87 in peach and 2.13 mg/kg in grape samples of Misurata area in Libya. However, the Cu concentrations of peach

and grape observed in aforementioned studies were much lower than that observed in the present study (Table 3). Moreover; the concentration levels of Cu were higher than the permissible limits according to the Joint FAO/WHO and the U.S. National Academy of Sciences (Table 4). The Cu concentrations of peach in all companies were two times more than the tolerable daily upper intake levels according to the U.S. National Academy of Sciences, while the concentrations were seven times more than the prescribed limits of the Joint FAO/WHO. Average concentration of Cu in the peach and grape from A Company was higher than the tolerable daily intake and the dietary reference intake, and this might be due to the application of micronutrient fertilizers and the widespread use of copper compound as a pesticide or fungicide. Also maybe as a result of application of poultry (chickens and quails) manures since A Company is one of the biggest poultry producers in Saudi Arabia, and in poultry industry the Cu and Zn are added to diets as growth promoters¹²⁻¹³.

Iron (Fe)

Among the investigated minerals, Fe showed the highest concentration in both fruits.

The levels of Fe ranged from 146 to 209 mg/kg in peach and 109 to 192 mg/kg in grape. The highest concentrations of Fe in both fruits were in A Company which was different significantly at 0.05 level of significance. From the results, it can be noted that the levels of Fe obtained in this study were much higher than those reported by Bagdatlioglu *et al.*,²⁸ Tormen *et al.*,²⁵ and Acar²⁶, and were in agreement with Mahdavian and Somashekar⁶ for similar fruits (Table 3). The concentrations of Fe in both fruits at all companies were two to four times higher than the permissible limits according to the Joint FAO/WHO and the U.S. National Academy of Sciences (Table 4), that probably could be referred to the excessive usage of inorganic fertilizers and livestock manures (e.g., cattle, sheep, poultry etc.) which are regularly added to soils in intensive farming systems to meet the higher demands of fruits production for Saudi markets.

Manganese (Mn)

Analyses of peach sample showed the lowest concentration of Mn in B Company as 7.35 mg/kg, while the highest was 11.85 mg/kg in A Company which was significantly different as compared to

Table 1: Average of mineral concentrations (mg/kg) in peach from some major agricultural companies in Saudi Arabia

| Mineral Company | Cu | Fe | Mn | Zn | Mg | Ca | Cd |
|-----------------|---------|--------|---------|---------|--------|--------|---------|
| A | 22.54 a | 209 a | 11.85 a | 14.62 a | 25.5 a | 177 a | 0.18 a |
| B | 19.81 b | 174 b | 8.53 b | 12.51 b | 24.1 a | 152 b | 0.092 b |
| C | 17.43 c | 146 c | 7.35 c | 11.08 c | 26.8 a | 122 c | 0.025 c |
| Mean | 19.93 | 176.33 | 9.24 | 12.74 | 25.47 | 150.33 | 0.099 |

Lowercase letters indicate significant differences ($P=0.05$).

Table 2: Average of mineral concentration (mg/kg) in grape from some major agricultural companies in Saudi Arabia

| Mineral Company | Cu | Fe | Mn | Zn | Mg | Ca | Cd |
|-----------------|---------|--------|---------|---------|--------|--------|---------|
| A | 12.85 a | 192 a | 10.15 a | 25.22 a | 37.5 a | 302 a | 0.12 a |
| B | 10.40 b | 154 b | 8.09 b | 22.42 b | 38.3 a | 198 b | 0.083 b |
| C | 8.22 c | 109 c | 7.05 c | 20.31 c | 38.3 a | 182 b | 0.019 c |
| Mean | 10.49 | 151.67 | 8.43 | 22.65 | 38.03 | 227.33 | 0.074 |

Lowercase letters indicate significant differences ($P=0.05$).

other companies. Results of Mn concentrations of the present study were higher than the study of Tormen *et al.*²³ who found a 120µg/L of Mn in peach juice samples collected from Brazilin markets. On the other hand, mean concentrations of Mn in grape sample ranged from 7.05 – 10.15 mg/kg and the highest concentration of Mn was found in A Company. Mahdavian and Somashekar⁶ studied the level of heavy metals in fresh fruit samples collected from the two main Bangalore City Markets in India. They found the concentrations of Mn in grape sample ranged from 29.03 – 39.31 mg/kg which were higher than the results of the present study (Table 3). The concentrations of Mn in both fruits of B and C companies were within the permissible limits, whereas in A Company concentrations of Mn in both fruits were slightly higher than the WHO recommended 2–9 mg per day for an adult³⁰ and the tolerable daily upper intake levels according to the U.S. National Academy of Sciences (Table 4).

Zinc (Zn)

The mean Zn concentration of peach and grape in different companies ranged from 11.08-14.62 and 20.31- 25.22 mg/kg, respectively. The data showed that peach and grape in different company

were different significantly in Zn concentration at 0.05 level of significance. Also, the highest Zn concentrations in both fruits were in A company than others. Studies from different countries around the world have determined Zn concentrations of peach and grape, Radwan and Salama²⁴ have reported a mean Zn concentration of 6.22mg/kg in peach samples collected from Egyptian markets. Tormen *et al.*²⁵ determined a mean Zn concentration of 180µg/L in peach juice samples collected from Brazilin markets. Acar²⁶ found that the Zn concentrations of peach juice and peach nectar sampled from different supermarkets in Ankara, Turkey were 0.61 and 0.49 mg/kg, respectively. Sobukola *et al.*²⁷ also reported a mean Zn concentration of 0.073mg/kg in grape samples collected from selected markets in Lagos, Nigeria. Bagdatlioglu *et al.*²⁸ determined a mean Zn concentration of 0.41mg/kg in grape samples grown in Manisa region, Turkey. Moreover, Elbagermi *et al.*²⁹ reported mean Zn concentrations of 5.87 in peach and 1.33mg/kg in grape samples of Misurata area in Libya. However, the Zn concentrations of peach and grape observed in above mentioned studies were lower than that observed in the present study (Table 3), Moreover; Mahdavian and Somashekar⁶ studied the level of heavy metals in fresh fruit samples

Table 3: Levels of metals (mg/kg) in fruits in previously published results from other parts of the world

| Minerals Fruits | Cu | Fe | Mn | Zn | Mg | Ca | Cd | Reference |
|--------------------|-------|--------|-------|-------|-------|--------|-------|---|
| Peach | 19.93 | 176.33 | 9.24 | 12.74 | 25.47 | 150.33 | 0.099 | Present study |
| | 1.87 | - | - | 5.87 | - | - | 0.02 | Liyba, Elbagermi et al., (2012) |
| | 1.46 | - | - | 6.22 | - | - | 0.01 | Egypt, Radwan and Salama, (2006) |
| Grape | 10.49 | 151.67 | 8.43 | 22.65 | 38.03 | 227.33 | 0.074 | Present study |
| | 0.51 | 3.58 | - | 0.41 | - | - | nd* | Turkey, Bagdatlioglu et al., (2010) |
| | 0.003 | - | - | 0.073 | - | - | 0.005 | Nigeria, Sobukola et al., (2010) |
| | 2.13 | - | - | 1.33 | - | - | 0.05 | Liyba, Elbagermi et al., (2012) |
| | nd | 206.99 | 39.31 | 17.40 | - | - | 3.63 | India, Bangalore City K.R Market, Mahdavian and Somashekar (2008) |
| | nd | 114.04 | 29.03 | 41.34 | - | - | 12.24 | India, Bangalore City Yeshwantpur Market, Mahdavian and Somashekar (2008) |

*Not detectable

collected from the two main Bangalore City Markets in India. They found the average concentration of Zn in grape sample as 30 mg/kg which was higher than the results of the present study (Table 3). The concentration levels of Zn in both fruits were within the permissible limits according to the Joint FAO/WHO and the U.S. National Academy of Sciences (Table 4).

Magnesium (Mg)

Magnesium concentration ranged from 24.1-26.8 mg/kg in peach and 37.5- 38.8 mg/kg in grape in different companies. Data showed that the Mg concentration in both fruits did not differ significantly at 0.05 level of significance, and was within the permissible limits according to the U.S. National Academy of Sciences (Table 4).

Calcium (Ca)

Mean Ca concentration ranged from 122- 177 mg/kg in peach and 182- 302 mg/kg in grape in different companies. Data showed that the Ca concentration in both fruits were significantly different at 0.05 level of significance. The highest Ca concentration in both fruits was in A Company as compared to others, and was within the permissible limits according to the U.S. National Academy of Sciences (Table 4).

Cadmium (Cd)

Mean Cd concentration of peach and grape in different companies ranged from 0.025- 0.18 and 0.019- 0.12 mg/kg, respectively. The results showed that peach and grape in different companies differed significantly in Cd concentration at 0.05 level

of significance. Also, the highest Cd concentration in both fruits was in A Company than others. There are many reports on Cd concentrations of peach and grape from different countries around the world. It was found that Radwan and Salama²² have reported a mean Cd concentration of 0.015mg/kg in peach samples collected from Egyptian markets. Tormen *et al.*,²⁵ determined a mean Cd concentration of 0.45µg/L in peach juice samples collected from Brazilian markets. Sobukola *et al.*,²⁸ also reported a mean Cd concentration of 0.003mg/kg in grape samples collected from selected markets in Lagos, Nigeria. However, the Cd concentrations of peach and grape observed in aforementioned studies were lower than that observed in the present study (Table 3). Moreover, Elbagermi *et al.*,²⁹ reported mean Cd concentrations of 0.02 in peach and 0.05mg/kg in grape samples of Misurata area in Libya and their results were in agreement with the results of the present study for B Company. Furthermore; Mahdavian and Somashekar⁶ studied the level of heavy metals in fresh fruit samples collected from the two main Bangalore City Markets in India and they found the average concentration of Cd in grape sample was 7.05 mg/kg and was higher than the present study results (Table 3). The concentrations of Cd in both fruits of C Company were within the permissible limits, whereas in B Company were slightly higher than the permissible limits, but in A company Cd concentrations were two to three times higher than the permissible limits according to the Joint FAO/WHO (Table 4). The high concentrations of Cd found in peach and grape samples of A and B Companies probably may be attributed to the excessive usage of fertilizers especially phosphoric fertilizers where Cd is found predominantly in phosphoric fertilizers³¹.

Table 4: The Joint FAO/WHO (Provisional Tolerable Daily Intake PTDI) and the Dietary Reference Intake (Tolerable Daily Upper Intake Levels UL) (mg/day) of minerals for adults

| Minerals | PTDI (mg) | UL (mg/d) |
|----------|-----------|-----------|
| Cu | 3 | 10 |
| Fe | 48 | 45 |
| Mn | 9 | 11 |
| Zn | 60 | 40 |
| Mg | - | 350 |
| Ca | - | 2500 |
| Cd | 0.06 | - |

Table 4 represents the provisional tolerable daily intake (PTDI) of toxic metal and the tolerable daily upper intake level (UL) of essential minerals. Average concentration of (Cu, Fe, and Cd) in the peach and grape from A and B Companies were higher than tolerable daily intake and the dietary reference intake which may pose a health risk to daily consumer and the long term consumption of these fruits should be of concern for the accumulation of the toxic metals especially cadmium in target organs such as kidney and liver³². The average concentrations of the essential metals (Mn, Zn, Mg,

and Ca) were lower than the dietary reference intake. However, at the same time, these fruits can serve as a good source of essential metals (Mn and Zn) for humans.

CONCLUSIONS

The study showed that the consumers are at greater risk of purchasing fresh peach and grape from A and B Companies with high levels

of heavy metals above the legally permissible limits as defined by the Joint FAO/WHO and the U.S. National Academy of Sciences. Moreover, from the results it was clearly seen that there is a lot of potential for further research in evaluating the mineral composition of fruits to mitigate health and environmental hazards likely to be occurred due to the consumption of contaminated fruit especially with heavy metal ions.

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