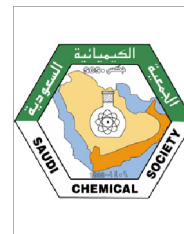




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## ORIGINAL ARTICLE

# Determination of macro, essential trace elements, toxic heavy metal concentrations, crude oil extracts and ash composition from Saudi Arabian fruits and vegetables having medicinal values

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**Abstract** The concentrations of essential elements (Mg, Ca, Na, K, Fe, Zn, Se, Al, Ni, and Cu) and toxic heavy metals (Pb, As, Cr, Cd, and Cr) from Saudi Arabian fruits and vegetables were determined by inductively coupled plasma optical emission spectrophotometry (ICP/OES). Two types of butters, *Caralluma munbayana* and *Caralluma hesperidum*, Vigna (*Vigna unguiculata*), common fig (*Ficus carica*), Annona seeds (*Annonaceae seeds*), Annona fruits (*Annonaceae fruits*), Fennel (*Foeniculum vulgare*), and Fennel flowers (*Nigella sativa*) were investigated, because they are used by indigenous groups as traditional medicines with Soxhlet-extraction and dry-ashing protocol. The estimated daily dietary element intake in food samples was further calculated in order to evaluate the element dietary intake and fruit and vegetable consumption pattern of the indigenes of Saudi Arabia. The crude oil and ash compositions varied widely, but suggested that most of the foods were good sources of oils and minerals. The figures-of-merit of the ICP-OES calibration curves were excellent with good linearity ( $R^2 > 0.9921$ ). The use of ICP-OES in this study allowed the accurate analysis and the detection of the elements at low levels. Essential elements (K, Ca, Na, and Mg) had the highest concentrations while toxic heavy metals (As, Pb, and Cd) had the lowest in the foods. Essential element pairs (Mg-Na, Mg-Ca, Fe-Al) were highly correlated, suggesting that these foods are sources of multiple nutrients. Toxic element pairs (Pb-Cd, Pb-As, and Cd-As), however, were poorly correlated in the foods, suggesting that these elements do not have a common source in these foods. Average consumption of these foods should provide the recommended daily allowances of

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essential elements, but will not expose consumers to toxic heavy metals. The ICP-OES method was validated by determining method detection limits and percent recoveries of laboratory-fortified blanks, which were generally 90–100%.

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## 1. Introduction

Fruits and vegetables continue to be the major sources of nutrients, including proteins, vitamins, macro and essential trace elements, and minerals in human diet for proper growth, body development, and maintenance of overall health and well-being (CMNRIM, 1999; NCI, 1986; NRC, 1989; WCRF, 1997). For instance, Ca and Mg are macro elements that are necessary for proper development of bone and structural tissue formation and play important roles in glucose and protein absorption and metabolism (Agarwal et al., 2011). Ca and Mg are also involved in the regulation and dilation of blood vessels and a regular heartbeat (Agarwal et al., 2011). Deficiency of Ca and Mg has been widely associated with weak bones and structural connective tissue formation, hypertension, and poor glucose absorption and utilization (Kosch et al., 2001). Iron (Fe) is a vital component of heme proteins, hemoglobin, and myoglobin (Fraga, 2005) required for oxygen transportation, proper cellular metabolism, glucose metabolism, and vascular functions (Fernandez-Real et al., 2002). Fe deficiency in humans has been shown to lead to a host of health issues such as a weakened immune system, inhibition of hemoglobin synthesis, which leads to anemia, insomnia, and other health related complications (Tapiero et al., 2001).

Other essential trace elements such as Zn, Cu, Mn, and Se also play important roles in maintaining proper human health. For instance, Zn is an important element in the human body, serving as a cofactor in a number of enzymatic reactions and responses such as metallo-enzymes for carboxyl peptidase, liver alcohol dehydrogenase, and carbonic anhydrase (Prasad, 2012). Copper is a coenzyme and crucial cofactor in Fe utilization, collagen amalgamation, and concealment of free radicals, and required for redox chemical cytochrome oxidase (Arredondo and Nunez, 2005; Naismith et al., 2009). Manganese is needed for the immune system and effective food metabolism, serves as a cofactor in numerous enzymatic responses, and aids in blood clotting and hemostasis (Smith et al., 2013). Selenium is essential for chemical responses for glutathione and thyroxine and has also been shown to have anticancer effects (Bangladesm et al., 2016). Nickel (Ni) is moderately required for proper absorption of Fe in the body (Gupta and Gupta, 2014). In addition to macro and essential trace elements, fruit and vegetables also contain high concentrations of essential oils, phenolics, antioxidants, and pharmacologically active agents with therapeutic effects for the treatment of several diseases such as cancer, diabetes, ulcers, asthma, common cold, and gastrointestinal diseases (Saini et al., 2015). Studies have shown several benefits of a balanced diet such as proper body weight, improved immunity against various diseases including diabetes, stroke, cardiovascular and heart diseases, cancers, and high blood pressure (CMNRIM, 1999; NCI, 1986; NRC, 1989; WCRF, 1997) that incorporate regular and adequate consumption of fruits and vegetables into the human diet.

Despite the important roles that macro and trace elements play in human health, little is known about the elemental composition and nutritional values of numerous fruits and vegetables in many parts of the world. Most importantly, fruits and vegetables may be inadvertently contaminated with chemicals of environmental concern and potentially toxic heavy metals such as Hg, Cd, As, Pb, and Cr. Fruits and vegetables can potentially be contaminated through environmental pollution, industrial activity or the absorption of heavy metals from contaminated soils, industrial effluent, or contaminated irrigation water (Davydova, 2005; Hu et al., 2013; IRAC, 2006; Zaidi et al., 2005). For instance, varying concentrations of heavy metals have been

detected in several food items including beverages, juices, wines, and several food products in both developed and developing countries (Al-Ahmary, 2009; Bua et al., 2016; Goldhabe, 2003; Hu et al., 2013; IRAC, 2006; Licata et al., 2012; Sharma et al., 2009; WHO, 1992; Vadalá et al., 2016; Zaidi et al., 2005). In contrast to macro and essential trace elements, heavy metals have no nutritional value. Heavy metals can also be uptaken, bioaccumulated, and biomagnified in human organs and animal tissues via the food chain and trophic level (Bella et al., 2015; Hu et al., 2013; Naccari et al., 2015; Rodriguez-Iruretagoiena et al., 2015; Salvo et al., 2016; Salvo et al., 2014).

Two types of butters (*Caralluma munbayana* and *Caralluma hesperidum*), Vigna (*Vigna unguiculata*), common fig (*Ficus carica*), Annona seeds (*Annonaceae* seeds), Annona fruits (*Annonaceae* fruits), fennel (*Foeniculum vulgare*), and fennel flowers (*Nigella sativa*) were investigated in this work. Although widely cultivated throughout the world, they are mostly consumed in Saudi Arabia, The Middle East, Africa, Spain, West Asia, Europe, China, Turkey, India, and tropical America. In addition, they are used as traditional medicines for the treatment of various diseases in many countries. For instance, Annona (*Annonaceae*) is used by Arabians as an indigenous therapy for the treatment of cancer disease (Ernhart et al., 1988). Previous studies have also shown the potential application of Annona leave extracts for the treatment of food-borne bacterial diseases and antitumor activity (Chen et al., 2012). Other pharmacological, phenolics, flavonoids, and antioxidant activities of Annona and common fig extracts have been reported (Gajalakshmi et al., 2011; Veberic et al., 2008). Butter has a bitter taste and is mostly used by Saudis and Arabians to regulate blood sugar levels and for the treatment of diabetes. Common fig (*Ficus carica*) is used by the indigenes for the treatment of constipation. Fennel flower (*N. sativa*) is commonly used as a food additive and also used to treat asthma, the common cold, scorpion bites, and other skin wounds. Fennel flower also has other cultural and religious values in The Middle East and Arabian countries including Saudi Arabia. Fennel (*Foeniculum vulgare*) is mostly used by Saudis for the treatment of food indigestion problem, bloating, gas accumulation, and colic problems. Vigna (*Vigna unguiculata*) is used by Saudis for the treatment of cardiovascular and heart diseases.

Distribution of foods and agricultural produce is global and is not limited by borders. The need for effective monitoring and mapping of heavy metal concentrations in food products is therefore not only an environmental, food and agricultural concern, but also a global public health and safety concern. Many health-related issues including cancers, cardiovascular problems, depression, hematic, gastrointestinal and renal failure, osteoporosis, tubular and glomerular dysfunctions have been directly linked to high levels of heavy metals in humans (ASTDR, 2005; EFSA, 2012; Fewtrell et al., 2003; Steenland and Boffeta, 2000; WHO, 2010; Vogtmann et al., 2013). Infants, children, and adolescents are particularly more susceptible to heavy metal poisoning, resulting in improper developmental challenges and low intelligent quotients (Ernhart et al., 1987; Schwartz, 1994; Ernhart et al., 1988; Dapul and Laraque, 2014).

It is therefore imperative to focus on proper food quality assurance and quality control protocols that ensure the intake of adequate amounts of essential trace elements and prevent the consumption of toxic heavy metals from food products. Flame atomic absorption spectroscopy (FAAS) is the most commonly used conventional method of chemical elements analysis (Latimer Jr., 2012; Watson, 1994). However, FAAS suffers from poor detection limit, hindering its use for detection of elements at ultra-low concentrations in food

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