

Residues of heavy metals, PCDDs, PCDFs, and DL-PCBs some medicinal plants collected randomly from the Jeddah, central market.

Yahia Youssef Mosleh^{1,2}, Jelan Mofeed², Omar Abdelhakeem Almaghrabi¹, Nief Mohamed Kadasa¹, Hassan Saeed El-Alzahrani¹ and Michael Paul Fuller³

¹Biology Department, Faculty of Science, King Abdulaziz University, P.O. Box 15758, Jeddah 21454, Saudi Arabia.

²Department of Aquatic Environmental, Faculty of Fish Resources, Suez University, Suez, Egypt.

³Department of Biomedical and Biological Sciences, Faculty of Science and Technology, Plymouth University, Plymouth, PL4 8AA, UK.

Yahia.mosleh@voila.fr

Abstract: The concentrations of PCDDs, PCDFs, and DL-PCBs were determined in samples of eight commonly used medicinal plants, namely "caraway, cumin, anise, sage, rosemary, black tea, ginger and cinnamon" collected randomly from the Jeddah central market between the periods of July and August 2013. Fortunately the result revealed that, the collected samples are free of PCDDs, PCDFs, and DL-PCBs. While, Among the determined heavy metals, the maximum concentration of Fe and Pb were recorded in rosemary (846.2 ± 18.5 and 10.8 ± 0.12 mg.kg⁻¹ respectively), it is noticeable that, the maximum Pb concentration (10.8 ± 0.12 mg.kg⁻¹) in the medicinal plant recorded in Rosemary, followed by 1.3 ± 0.02 mg.kg⁻¹ in black tea with a significant gab. While the maximum concentrations of Zn and Cr were recorded in anise (52.7 ± 1.6 and 3.1 ± 0.1 mg.kg⁻¹ respectively). In contrast to the above, cinnamon characterized by the minimum concentration (0.89 ± 0.03 , 19.5 ± 1.5 , 13.1 ± 1.1 , 0.012 ± 0.001 , and <LOQ mg.kg⁻¹) for six of the recorded eight heavy metals (Cu, Fe, Zn, Ni, Hg and Pb respectively). The study give a good indication about the absence of PCDDs, PCDFs, and DL-PCBs from the collected plants, but on the other hand it sheds light on the importance of follow-up the heavy metals concentration in these commonly used plants.

[Yahia Y. Mosleh, Jelan Mofeed, Omar Almaghrabi, Naif M.S. Kadasa, Hassan Alzahrani and Michael Paul Fuller.

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Keyword: Heavy metals, medicinal plant, PCDDs, PCDFs, and DL-PCBs.

1- Introduction

The term of medicinal plants include a various types of plants used in herbalism and have a medicinal activities. These medicinal plants consider as a rich resources of ingredients which can be used in drug development and synthesis. Medicinal plants are valuable invaluable natural resources; they are exhaustible if overused and sustainable if the juxtaposition of present and future needs takes place within the behavioral pattern of various kind of users. For many decades, traditional remedies were empirically practiced in Saudi Arabia, and indeed in Asia to treat various diseases. Therefore, medicinal plants play an important and vital role in traditional medicine and are widely consumed as a routine treatment and home remedy (Jean *et al.*, 2013 and Olukayode *et al.*, 2004). A total 50,000 plants are used for primary (Schippmann *et al.*, 2002) out of the total plant species, 422,000 species have been reported all over the world (Govaerts *et al.*, 2001). Indigenous plants were used in the treatment of high blood pressure, diarrhea, fever, and so on (Brichard, and Leaderer, 1991 and Roland *et al.*, 2013). WHO has listed over 2000 plants that are known to perform one function or the other (WHO 2002). Plant can

accumulate a number of mineral elements, essential to human nutrition, and on the other hand, it equally accumulates other mineral elements such as Cd, Co, Ag, which are in no direct use to them but injurious to health (Baker and Brooks 1989 and Mark *et al.*, 2000). Trace elements have been reported to have curative and preventive role in medicine (Perman *et al.*, 1993; Ahmed *et al.*, 1994 and Roland *et al.*, 2013). Medicinal plants which known to perform more important role in agronomy production and pharmacy (Rafl 1989; Ali 1993; Kim *et al.*, 1994 and Bibhash *et al.*, 2014) have conducted several studies on the heavy trace metals and macronutrients status in herbal plants. However, limited and scanty information are available with respect to some medicinal plants endemic in Saudi Arabia despite the heavy reliance on such plants as home remedies.

The biological half-lives of these heavy metals are long and have potential to accumulate in different body organs and thus produce unwanted side effects (Ata *et al.*, 2009). Heavy metal analysis is an important aspect of quality control and is a requirement of the clinical trials exemption to ensure that the plant material is not contaminated with toxic metals, such as cadmium, lead and mercury, since

contamination of medicinal plants is always a potential risk, as was reported by (Roland *et al.*, 2013). It was reported that, cadmium is toxic where it accumulates in the body throughout a lifetime (Singh *et al.*, 2006). Lead has been shown to exert toxic effects on the center nervous system, reproductive, renal, haematopoietic and immune systems (Weeden 1984), and it has been identified with impaired quality of life. (Nicaise *et al.*, 2014). Mercury is highly toxic to human health, where it can produce harmful effects on the nervous, digestive and immune systems, lungs and kidneys, and may be fatal. Heavy metal contamination of medicinal plants can occur either during cultivation or the subsequent harvesting and processing procedures. Therefore, medicinal herbs, which cultivated on soil that is relatively free from heavy metal ions, cannot necessarily be guaranteed to be free of contaminants at the final stage of production. While using herbs in medication for various illnesses, one should be aware that, apart from the pharmacological effect they could turn out to be toxic due to the presence of heavy metals and other impurities. Among many methods described in the literature for trace determination of afore mentioned metals in plant material, atomic absorption spectrometry is recommended (Valcho *et al.*, 2008; Khillare *et al.*, 2012; Ingrid 2014 and Nicaise *et al.*, 2014). There are reports available on presence of heavy metals in medicinal plants formulations above regulatory standards, considering this lead, cadmium, arsenic and mercury were analyzed in all drug powders using atomic absorption spectroscopy and found within the limits (WHO 2002).

While, Polychlorinated dibenzo-dioxin/furan (PCDD/Fs) and dioxin-like polychlorinated biphenyls (DL-PCBs) are among the most sinister pollutants that end up in wastewater. These persistent environmental contaminants are potential carcinogens, with a strong tendency to bioaccumulate in various environmental segments (ICON, 2001) along with industrial emissions. Generally, the most common sources for dioxins emission in the world are solid waste and sludge burning. Open burning of domestic and some agricultural solid waste is a quite common practice. On the other hand some of the newly introduced industries such as pulp and paper bleaching and some metallurgic works would constitute another potential source of emission.

The aim of this study was to carry out investigation of heavy metals, Polychlorinated dibenzo-dioxin/furan (PCDD/Fs) and dioxin-like polychlorinated biphenyls (DL-PCBs) in some commonly used medicinal plants collected from different sources in Jeddah city, Saudi Arabia.

Key words: medicinal plants, heavy metals,

Polychlorinated dibenzo-dioxin/furan and polychlorinated biphenyls.

2- Material and methods

2.1. Sample collection

Six samples from each of caraway, cumin, anise, sage, rosemary, black tea, ginger and cinnamon (Table, 1) were collected randomly from the Jeddah central market between the periods of July and August 2013. The collected samples were immediately wrapped in aluminum foil, placed in an ice-chest kept at 4 °C and sent to the laboratory for analysis. In the laboratory, similar samples were wholly bulked together and ground in a warring blender to obtain a homogenous composite.

Table (1): The Common, Scientific names and the used part of the eight examined medicinal plants.

2.3. Heavy metals

Sample preparation

Respective dried plant parts were coarsely ground using a pestle and mortar and passed through a 1000 µm 2 metal sieve. 1 g of powdered sample was dissolved by acid digestion in a 8 mL-mixture of concentrated HCl and concentrated HNO₃ in a 3:1 ratio. The sample was digested for 3 h and the remaining contents were dissolved in ultra high purity (UHP) water, filtered by gravity and diluted to a final volume of 100 mL.

2.3.1. Reagents and chemicals

In this study, UHP water (18 mΩ) from a Millipore Water System (Bedford, MA, USA) was used. CP grade hydrochloric acid (HCl) and AR grade nitric acid (HNO₃) were obtained from Rochelle (Johannesburg, South Africa). Metal working standard solutions were prepared by diluting 1000 ppm stock solutions purchased from Rochelle (Johannesburg, South Africa).

2.3.2. Instrumentation and standards

The atomic absorption spectroscopy (AAS) measurements were performed using an air-acetylene flame atomic absorption spectrometer from Varian (Varian SpectrAA 220FS, Australia). Metal hollow-cathode lamps (Photrons, USA) specific for each of the determined metals were employed as radiation/sources. Working standards of different concentrations as per the instructions of the hollow-cathode lamps were prepared to detect each metal concentration. A five-point calibration curve was automatically generated by the instrument.

2.4. Analyses of PCDD/Fs and PCBs

Sub-samples of 50 g were analyzed for each of the 17 PCDD/Fs and 12 DL-PCBs, for which the WHO developed toxicity equivalency factors (TEFs) (Van *et al.*, 1998 and Loutfy *et al.*, 2010). Seventeen ND-PCBs congeners were analyzed together with PCDD/Fs and DL-PCBs in one analytical procedure.

High-resolution gas chromatography (HRGC)/high-resolution mass spectroscopy (HRMS) HP 6890 plus a gas chromatograph coupled to PCBs in operating in EI mode at 35 eV and with a Micromass Autospec Ultima mass spectrometer resolution of 10,000 (5% valley) was used.

2.4.1. Reagents and Procedure

Reagents used in this study were the same as described earlier (Loutfy *et al.*, 2007). Fifty gram homogenate were spiked with a series of 15 $^{13}\text{C}_{12}$ -labeled 2,3,7,8 PCDD/F mix (EDF8999), a series of 12 $^{13}\text{C}_{12}$ -labeled PCB (EC4937), as internal standards, and then mixed by Speed Matrix. The samples were extracted by sonication with n-hexane. The extracts were spiked with $^{13}\text{C}_{12}$ -labeled 2,3,7,8 PCDD (EDF6999) and with three $^{13}\text{C}_{12}$ -labeled PCB (EC4978) then cleaned up using the automatic three-column system, with pre-packed disposable columns containing multilayer silica, alumina and carbon. From this system two fractions were eluted, one with PCDD/Fs and the other with PCBs.

3. Results

Diversity of medicinal plants rests crucial for human population in providing the numerous indigenous and modern healthcare remedies. Presenting study focused on documentation of commonly used medicinal plants and to investigate the quality and toxicity of frequently used medicinal plants of study area. Study was conducted in one big city of Saudi Arabia (Jeddah).

Inspection of table (2) revealed that, except mercury (which did not detected in all examined plants), all other heavy metals were represented in 96% of the tested plants. It is noticeable that, the

highest concentration of the estimated elements did not recorded in one plant. Among the heavy metals, the maximum concentration of Fe and Pb were recorded in rosemary (846.2 ± 18.5 and 10.8 ± 0.12 mg.kg^{-1} respectively), it is noticeable that, the maximum Pb concentration (10.8 ± 0.12 mg.kg^{-1}) in the medicinal plant recorded in Rosemary, followed by 1.3 ± 0.02 mg.kg^{-1} in black tea with a significant gap. While the maximum concentration of Zn and Cr were recorded in anise (52.7 ± 1.6 and 3.1 ± 0.1 mg.kg^{-1} respectively), while that of Ni (2.1 ± 0.3 mg.kg^{-1}) was recorded in cumin. Cd gave their maximum concentration in cinnamon (0.281 ± 0.02 mg.kg^{-1}). While, the 8 medicinal plant samples and jiggery were subjected to estimation of Cu. Results are shown in table (2). The maximum amount of Cu was found to be in black tea 12.9 ± 1.1 mg. followed by caraway, anise, cumin, sage, ginger, rosemary and cinnamon (10.4 ± 0.9 , 9.1 ± 0.9 , 8.6 ± 0.8 , 5.3 ± 0.1 , 4.5 ± 0.4 , 3.7 ± 0.09 and 0.89 ± 0.03 respectively). In contrast to the above, cinnamon characterized by the minimum concentration (0.89 ± 0.03 , 19.5 ± 1.5 , 13.1 ± 1.1 , 0.012 ± 0.001 , nd and $<\text{LOQ mg.kg}^{-1}$) for six of the recorded eight heavy metals (Cu, Fe, Zn, Ni, Hg and Pb respectively). Among the elements, the maximum concentration of both Na (721 ± 21.2 mg.kg^{-1}) and K (2201 ± 9.92 mg.kg^{-1}) were recorded in ginger.

It is worth mentioning that, the residues of PCDDs, PCDFs, and DL-PCBs in all the samples of the eight medicinal plants collected from Jeddah city - Saudi Arabia (Table, 3), are free of PCDDs, PCDFs, and DL-PCBs.

Table (1): Herbal plants under investigation; name, parts studied and medicinal properties

Plant species	Part used	Medicinal properties
Caraway	Seeds	Caraway is used for digestive problems including <u>heartburn</u> , <u>bloating</u> , gas, loss of appetite, and mild spasms of the <u>stomach</u> and <u>intestines</u> . Caraway oil is also used to help people cough up phlegm, improve control of urination, kill bacteria in the body, and relieve <u>constipation</u> .
Cumin	Seeds	Cumin seeds have traditionally been noted to be of benefit to the digestive system, and scientific research is beginning to bear out cumin's age-old reputation. Research has shown that cumin may stimulate the secretion of pancreatic enzymes, compounds necessary for proper digestion and nutrient assimilation.
Anise	Seeds	Anise it is greatly used in the form of lozenges and the seeds have also been used for smoking, to promote expectoration. The volatile oil, mixed with spirits of wine forms the liqueur Anisette, which has a beneficial action on the bronchial tubes, and for bronchitis and spasmodic asthma, Anisette, if administered in hot water, is an immediate palliative
Sage	Leaves, small stems and flowers	Sage is used for digestive problems, including loss of appetite, gas (<u>flatulence</u>), <u>stomach pain</u> (<u>gastritis</u>), <u>diarrhea</u> , <u>bloating</u> , and <u>heartburn</u> . It is also used for reducing overproduction of perspiration and <u>saliva</u> ; and for <u>depression</u> , <u>memory loss</u> , and <u>Alzheimer's disease</u> .
Rosemary	Leaves and flowers	The herb parts, especially flower tops contain phenolic anti-oxidant rosmarinic acid as well as numerous health benefiting volatile essential oils such as <i>cineol</i> , <i>camphene</i> , <i>borneol</i> , <i>bornyl acetate</i> , <i>α-pinene</i> , etc. These compounds are known to have rubefacient (counterirritant), anti-inflammatory, anti-allergic, anti-fungal and anti-septic properties.
Chinese teas	Leaves	Improve the immune system, slow down aging and can help prevent cancer. Lower blood, cholesterol and blood pressure and prevent arteriosclerosis Prevent tooth decay, freshen the breathe and assist in digestion and also Enhance the eliminating functions of the kidneys
Ginger	Fresh and	Use as medicine for vomiting, diarrhea, indigestive heat, accumulation of mucus, coagulation of blood or

	dried rhizomes	blood circulation problems(due to drop in blood temperature and thus blood become cold and clumpy). Helps to increase appetite and clears the pathways of nutrient absorbing channels. The rhizomes are commonly used in the preparation of herbal tea.
Cinnamon	Inner bark, Leaves and oil	Anti-Clotting It has an anti-clotting effect on the blood. Also used as arthritis relief and anti-bacterial when added to food, it inhibits bacterial growth and food spoilage, making it a natural food preservative. Also smelling cinnamon boosts cognitive function and memory. It is a great source of manganese, fiber, iron, and calcium.

Table (2): residues of heavy metals (mg.kg^{-1}) in the eight tested medicinal plants.

Heavy metals mg.kg^{-1}	Medicinal plant used							
	Caraway	Cumin	Anise	Sage	Rosemary	Chinese teas	Ginger	Cinnamon
Cu	10.4 ± 0.9	8.6 ± 0.8	9.1 ± 0.9	5.3 ± 0.1	3.7 ± 0.09	12.9 ± 1.1	4.5 ± 0.4	0.89 ± 0.03
Fe	416 ± 12.2	236.7 ± 4.5	288.6 ± 2.2	368.1 ± 2.9	846.2 ± 18.5	448.8 ± 41.1	226.7 ± 3.3	19.5 ± 1.5
Zn	44 ± 2.1	46.6 ± 1.5	52.7 ± 1.6	51.2 ± 2.2	25 ± 1.9	32.3 ± 2.1	26.7 ± 2.1	13.1 ± 1.1
Cd	0.024 ± 0.01	0.044 ± 0.001	0.087 ± 0.03	<LOQ	0.008 ± 0.001	0.129 ± 0.01	<LOQ	0.281 ± 0.02
Hg	<LOQ	nd	nd	nd	nd	nd	nd	nd
Pb	<LOQ	0.23 ± 0.005	0.11 ± 0.01	1.3 ± 0.1	10.8 ± 0.12	1.3 ± 0.02	0.46 ± 0.07	<LOQ
Ni	1.8 ± 0.01	2.1 ± 0.3	1.1 ± 0.02	1.3 ± 0.09	0.9 ± 0.12	1.4 ± 0.01	0.72 ± 0.09	0.012 ± 0.001
Cr	2.1 ± 0.02	1.9 ± 0.2	3.1 ± 0.1	0.6 ± 0.05	2.2 ± 0.09	0.9 ± 0.02	1.1 ± 0.01	1.7 ± 0.01
Na	70 ± 3.2	190 ± 2.2	564 ± 5.4	432 ± 11.2	100 ± 2.1	456 ± 21.1	721 ± 21.2	205 ± 1.2
K	1880 ± 22.23	1770 ± 4.3	1988 ± 7.8	1800 ± 6.7	1996 ± 24.9	2060 ± 22.31	2201 ± 9.92	1987 ± 12.4
P	100 ± 4.32	208 ± 5.4	190 ± 2.9	543 ± 5.9	654 ± 11.2	324 ± 5.4	321 ± 10.2	209 ± 2.9

Table (3): Concentrations of PCDDs, PCDFs, and DL-PCBs in medicinal plants collected from local marked in Jeddah, Saudi Arabia

Concentrations of PCDDs, PCDFs, and DL-PCBs	Medicinal plant used							
	Caraway	Cumin	Anise	Sage	Rosemary	Chinese teas	Ginger	Cinnamon
PCDDs 2,3,7,8-TeCDD 1,2,3,7,8-PeCDD 1,2,3,6,7,8-HxCDD 1,2,3,7,8,9-HxCDD OCDD	nd	nd	nd	nd	nd	nd	nd	nd
PCDFs 2,3,7,8-TeCDF 1,2,3,7,8-2,3,4,7,8-PeCDF 1,2,3,4,7,8-HxCDF 1,2,3,6,7,8-HxCDF 2,3,4,6,7,8-HxCDF 1,2,3,7,8,9-HxCDF 1,2,3,4,6,7,8-HpCDF OCDF	nd	nd	nd	nd	nd	nd	nd	nd
Sum PCDDs Sum PCDFs Sum PCDDs/PCDFs Dioxin-like PCBs Non-ortho PCBs 3,4,5,3-TCB 3,4,3,4-TCB 3,4,5,3,4-PeCB 3,4,5,3,4,5-HxCB Mono-ortho PCBs 2,4,5,2,4-CB 2,4,5,3,4-PeCB 2,3,4,5,4-PeCB 2,3,4,3,4-PeCB 2,3,4,3,4,5-HxCB	nd	nd	nd	nd	nd	nd	nd	nd

4- Discussion

The uptake of metals by plants is influenced by various factors, including type of plant, nature of soil, climate and agriculture practices. The concentration of heavy metals is not uniformly distributed throughout the plant. In general, the roots contain the highest levels of heavy metals, followed by vegetative tissue. Sewage sludge, industrial activities, fuel, and automobile tires can also be significant metal sources (Badeae *et al.*, 1999).

There are reports regarding presence of high levels of toxic metals such as lead, mercury, arsenic etc, in the ayurvedic herbal medicinal products manufactured in South Asia (Agnese *et al.*, 2011 and Iris *et al.*, 2013). There are possibilities of plants chelating heavy metals from environment. In the earlier days there were no techniques to determine the actual metal content in these plants. However they were just been utilised for curing certain ailments directly. Now with a better knowledge on the heavy metal toxicity, importance to know the actual content of these elements was also felt. The toxicity for individual elements even at low levels were determined by using the technique of AAS.

There are reports of very high levels of the toxic elements present in the ayurvedic drugs supplied from India to western and European countries. Though, the ayurvedic system doesn't permit the use of these toxic elements at such high levels even in the rasa shastra system, the question "How these high levels could have entered the medicines" needs to be thought of. A possible source of this high level can come from the plants itself. So, in order to ensure that the contribution from the plants is not high, there was need for the analysis of the plants before it is being used for preparation of medicine. Hence a set of commonly used medicinal plants were taken for a preliminary study of their toxic elemental content using the technique of AAS. In the present study, the heavy metals (Cu, Cd, Pb, Hg and Zn) were determined in medicinal plant samples collected from different sources.

Human body requires varieties of nutrients for its functioning. These nutrients include some elements like Cu, Ni, Zn, Fe, Cd, Pb etc, which act as nutrients when present at particular threshold levels, below or above which they affect the functioning of the system, where the higher level than the needed are toxic. These elements also have a curative effect on many ailments. In this connection, many ayurvedic medicines have been developed (Amrita *et al.*, 2011). The source for these elements for preparation of ayurvedic medicines come from medicinal plants, which considered as a good source for bioaccumulation of heavy metals (Liang-Feng *et al.*, 2012 and Fazlin *et al.*, 2014). When the levels of

heavy metals exceed in plants and animals, it can induce a variety of acute and chronic effects in wide range of organisms in various eco-systems. In USA for example, heavy metals have caused natural forest to decline. The control of the heavy metal contents in medicinal plants represents one of the most important factors for the evaluation of the quality. Since these plants originate from different growing areas, great differences in the uptake and concentrations of heavy metals in the plant tissue can be expected. The high heavy metal contents in some medicinal plants arises from their ability to accumulate particular metals especially cadmium. However, high heavy metal uptake can also be found in growing areas located in mountain regions, due to certain properties of these soils, such as acidity and/ or the presence of metal bearing minerals, which favor the mobility of heavy metals in the soil and their availability to plants. In the present study, accumulation of heavy metals (Cd, Cr, Cu Fe, Hg, Ni, Pb and Zn) were estimated within the eight common used medicinal plant from market of Jeddah, Saudi Arabia, caraway, cumin, anise, sage, rosemary, black tea, ginger and cinnamon.

Zn concentration ranged from 13.1 $\mu\text{g}\cdot\text{g}^{-1}$ in cinnamon to 52.7 $\mu\text{g}\cdot\text{g}^{-1}$ in anise. While, Fe concentration found in all the medicinal plants examined was generally relatively high (19.5-846.2 $\text{mg}\cdot\text{kg}^{-1}$), with the three highest concentrations found in cinnamon and rosemary, respectively. The maximum permissible level (MPL) of iron is 1000 $\mu\text{g}\cdot\text{day}^{-1}$ (NRC 1980). In this study, majority of all medicinal plants examined had iron content above the maximum permissible level per 10 g daily⁻¹ dose of medicinal plants or its equivalent formulation. Iron is an integral part of many proteins and enzymes that maintain good health. In humans, iron is an essential component of proteins involved in oxygen transport (Dallman 1986 and Institute of Medicine 2001). It is also essential for the regulation of cell growth and differentiation (Bothwell *et al.*, 1979 and Andrews 1999). A deficiency of iron limits oxygen delivery to cells, resulting in fatigue, poor work performance, and decreased immunity. Iron deficiency anaemia is associated with constipation, nausea, vomiting, and diarrhoea (Bhaskaram, 2001; Haas and Brownlie, 2001 and Institute of Medicine 2001). On the other hand, excess amounts of iron can result in increased risk of free radical damage and cancer and even death (Corbett 1995; Sarita and Rohit 2006 and Street 2012).

Zinc is important during puberty, pregnancy, and menopause. If large doses of zinc (10-15 times higher than the recommended daily intake) which is 8-15 mg/day are consumed, stomach cramps, nausea, and vomiting may occur. In the present study, the maximum Zn concentration were recorded in Anise

($52.7 \pm 1.6 \text{ mg.kg}^{-1}$) followed by Sage ($51.2 \pm 2.2 \text{ mg.kg}^{-1}$) and Cumin ($46.6 \pm 1.5 \text{ mg.kg}^{-1}$). Ingesting high levels of zinc for several months may cause anemia, damage the pancreas, and decrease levels of high-density lipoprotein (HDL) cholesterol. Consuming low levels of zinc is at least as important a health problem as consuming too much zinc. Without enough zinc in the diet, people may experience loss of appetite, decreased sense of taste and smell, decreased immune function, slow wound healing, and skin sores. Too little zinc in the diet may also cause poorly developed sex organs and retarded growth in young men. If a pregnant woman does not get enough zinc, her babies may have birth defects (ATSDR, 2005; Kolachi *et al.*, 2011; Subramanian *et al.*, 2012; Sarah and Susan, 2014). While, copper helps the body to use iron it is also important for nerve function, bone growth, enhanced body use of sugar and protection of cell membranes from destruction by free radicals. A wide range of cardiovascular and blood disorders may be attributed to copper deficiency. Meanwhile, water that contains higher than normal levels of copper may cause nausea, vomiting, stomach cramps, or diarrhea (ATSDR 2005 and Sarah and Susan 2014). The maximum permissible level (MPL) of copper is $12,000 \mu\text{g/ day}$. (Jørgensen 2000) Therefore, the suggested average intake of about 10 g of plant material or its equivalent formulation gives a maximum of $1145 \mu\text{g}$ of copper per day. (Jørgensen 2002). This implies that, the eight medicinal plants evaluated contained safe levels of copper. Although, nickel was present in all medicinal plants, had nickel content higher than the permissible level. This however constitutes only 26%; the majority (74%) had nickel content within acceptable limits.

Regarding cadmium, it was also found in all the plant species, and its concentration ranged from 0.008 in rosemary to 0.281 mg.g^{-1} in cinnamon. Eating food or drinking water with very high cadmium content severely irritates the stomach, leading to vomiting and diarrhea, hypertension, and sometimes death. Accumulation of lower levels of cadmium over a long period of time can lead to a build-up of cadmium in the kidneys and cause kidney damage (ATSDR 2005). While lead can cause neurological damage in fetus and young children. Lead exposure may also cause weakness in fingers, wrists, or ankles, small increases in blood pressure and anemia. At high levels of exposure, lead can severely damage the brain and kidneys in adults or children and ultimately cause death; it also causes miscarriage (ATSDR, 2005). Taking into consideration the present data, the maximum concentration ($10.8 \pm 0.12 \text{ mg.kg}^{-1}$) recorded in Rosemary, followed by $1.3 \pm 0.02 \text{ mg.kg}^{-1}$ in black tea with a significant gap. Mercury causes severe birth defects, abortion and mental retardation.

In addition to permanent damage to the brain and kidneys, mercury can damage the stomach and intestines, producing symptoms of nausea, diarrhea, or severe ulcers. Symptoms included rapid heart rate and increased blood pressure (Hayter 1980). Thus, the benefits of micronutrients may be completely reversed if present at high levels as they may be detrimental to human health. Therefore, the World Health Organisation (WHO) has established levels of metals in foods above which, they should not be consumed. For this reason the levels of trace metals in our food should be of much importance and concern to us. The heavy metal or mineral content of spices is not a well researched area worldwide. So in order to ensure that the contribution from the plants is not high, there was need for the analysis of the plants before it is being used for preparation of medicine.

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Corresponding author

Dr. Yahia Youssef Ismail MOSLEH

¹Biology Department, Faculty of Science, King Abdulaziz University, P.O. Box 15758, Jeddah 21454, Saudi Arabia.

²Department of Aquatic Environmental, Faculty of Fish Resources, Suez University, Suez, Egypt.

Yahia.mosleh@voila.fr

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