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REVIEW ARTICLE

Impact of Toxic Heavy Metals and Their Concentration in *Zygophyllum* Species, *Mentha longifolia*, and *Thymus vulgaris* Traditional Medicinal Plants Consumed in Setif-Algeria

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ABSTRACT
Heavy metals (HM) are essential for living cells to maintain their equilibrium. This survey focuses on the problem of medicinal plant contamination due to environmental pollution produced by many different industrial activities and the atmospheric deposition of some toxic compounds. This analysis is important since plants can easily absorb organic and inorganic compounds from all environmental compartments (water, soil, air), which can enter and be transferred in the trophic chain, up to humans. Medicinal plants are relevant for a study about their interactions with different contaminants, in particular those inorganic persistent as HM, because they are used in the entire world for their beneficial properties and represent a significant part of traditional medicine. This review was undertaken to give readers a comprehensive understanding of chemical contaminants, such as HM, which are significant and frequent pollutants of herbal medicines and pose considerable health concerns to the human body. The information was obtained from several sources to figure out the levels of HM in three traditional medicinal plants used in Algeria's Setif region. The gathered data demonstrated that *Zygophyllum* species, *Mentha longifolia*, and *Thymus vulgaris* accumulate higher quantities of HM when cultivated in polluted soil as opposed to unpolluted soil. The data's conclusions imply that these plants contained different hazardous concentrations of HM over the World Health Organization's allowable limits. Rational herb consumption is necessary for a healthy diet. However, the exact mechanisms through which this HM affect human health are not well understood.

Keywords: Heavy metal, health risk, polluted soil, *Zygophyllum* species, *Mentha longifolia, Thymus vulgaris.*

INTRODUCTION

Heavy metal contamination is a general phenomenon known to pose major dangers to human health and ecosystem stability. The three main challenges are increasing urbanization, real estate transformation, and industrial development, particularly in highly populous and developing nations.[1] Environmental concern over the poisoning of the water and air with toxic metals has affected hundreds of millions of people worldwide. Another concern for human and animal health is the poisoning of food with heavy metals (HM). In this regard, the amount of HM in the resources (water, air, and food) is assessed.[2] Some of these metals can be found in a wide range of amounts. The essential elements (micro- and macro-elements) are crucial for the normal development and growth of living organisms. Additionally, medicinal plants may contain high amounts of HM, which can be poisonous and cause serious metabolic disturbances. Essential metals in the form of micro- and macro-elements (Zn, Cu, Fe, Mn, Na, K, Ca, Cr, and Mg) are required in trace amounts for the healthy functions of enzymatic systems, vitamin synthesis, the production of hemoglobin, and the growth, development, and photosynthesis of plants.[3]

In actuality, air pollution or contact with contaminated soil are the two main causes of natural food contamination. The accumulation of HM in a human body and adipose tissue results in the loss of essential nutrients and deficits in the central nervous system, in addition to heart, digestive, hematological, neurological, hepatocellular, renal, reproductive, immune, and intrauterine growth retardation problems.[1] For many years, using plants as medicine has been a significant component of the global primary health care system. However, several areas still lack information regarding therapeutic plants and their preservation.[4] The safety and toxicity of natural herbs and product formulations on the market have come under scrutiny in tandem with the growing interest in the medicinal advantages of herbal remedies. Although it is generally believed that nat-

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ural herbs and plants are intrinsically harmless, several reports have been reported on toxicity and unfavorable effects associated with the use of plants and their preparations in various parts of the world.[5]

Our goal was to review the literature on the origin, accessibility, toxicity associated with HM, and health effects of HM in some medicinal plants grown in Setif, Algeria, including *Zygophyllum* species, *Mentha longifolia*, and *Thymus vulgaris*. This review will increase our knowledge of their harmful effects on the body's organs and result in better management of metallic intoxications.

HEAVY METAL CONTAMINATION

HM, such as Cr, Hg, Pb, Cu, and Cd are significant environmental pollutants. Heavy metal contamination of the air, water, and soil may have serious negative effects on all living organisms. HM added to soil have adverse effects on food production, population growth rates, and environmental health. Human health can be seriously endangered by the bioaccumulation of HM in food. These substances enter the body through food and breathing. The mobility and bioavailability of the soil influence this pollution.[6] Certain metals (Mn, Cr, Cu and Zn) are present in green plants as necessary metals often in low amounts, and the organisms that consume these plants are unaffected. However, the high levels of HM in plants can rise as a result of environmental contamination.[7] Human and anthropogenic factors are the primary causes of the rising environmental toxicity of HM. The naturally occurring sources of HM include forest fires, volcanic eruptions, wind-blown soil debris, biogeochemical processes, and marine salt. The use of herbicides, pesticides, agricultural practices, sewage evaporation, and industrial processes are among the human-caused factors contributing to the contamination of the HM[8] (Figure 1).

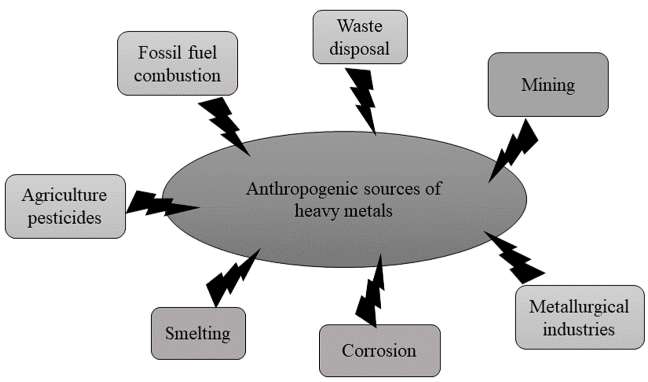

Figure 1. Anthropogenic activity leads to soil contamination by heavy metals.

Numerous factors, including the pH of the soil, the amounts of metals there, the oxidoreduction capacity of the soil, as well as other chemical and physical parameters, all have an impact on the bioavailability of metals. Additionally, contamination could happen at the time of sale or during storage. Herbs can generally become contaminated during cultivation, harvest, and processing. The sources of contamination by HM in herbs may include irrigation water, industrial, emissions, contaminated soil, pesticide and fertilizers, transportation traffic, and harvesting and storage procedures. The average daily dietary consumption affects the health risk posed by metal exposure.[5] HM could be found on the water's surface as a result of either natural or anthropogenic causes. HM ions are found in the form of silicates, phosphates, sulfates, oxides, sulfurs and hydroxydes.[9,10] Other natural source mechanisms that generate heavy metal pollution of water include water interaction with rocks, water interaction with soil, and the deposition of dry and moist air sand. Examples of these compounds are cuprite, calcite, kaolinite, chromite, siderite, and arsenic trioxide. Industry and urbanization are two anthropogenic sources of water contamination, and their fast rise is another.[11–14] These HM in the river can have a severe impact on the aquatic system's biological balance, and as the contamination increases, the variety of aquatic organisms becomes limited.[15] The presence of certain HM causes diseases such as Minamata, an organic Hg poisoning. When HM bioaccumulate, they pose a threat to the human and animal population who drink this water.[16] This necessitates the determination of pollution levels, which allows for the development of strategies to solve the problem [17,18] (Table 1).

Table 1. Permissible limits of heavy metals in water. [19]

Heavy Metals	World Health Organization's (WHO) Permissible Limit (ppm)
Mn	5.00
Zn	5.00
Cr	0.1
Cd	0.001–0.005
Fe	5.00
As	10.00
Pb	0.1
Cu	3.00

Pb, Ni and Cd have a variety of toxic effects on plants at the physiological, morphological, and biochemical levels when they enter cells in high concentrations. The effects of Pb, Ni, and Cd on plants may be caused by direct (metal toxicity from tissue accumulation) and/or indirect factors including the alteration of the photosynthetic process, the disruption of nitrogen/mineral nutrition, and the development of oxidative stress via excessive reactive oxygen species (ROS) generation. [20] ROS, such as superoxide anion (O_2•), hydroxyl radical (•OH), hydrogen peroxide (H_2O_2), and singlet oxygen (O_2) are produced in excess by several HM, including Pb. According to recent research, Pb may interact with oxy-Hb to produce ROS, which can induce peroxidation of erythrocyte membranes. The pro-oxidant/antioxidant equilibrium is upset by the toxic metals, which induce oxidative damage. ROS are produced as a result of lead exposure, which causes oxidative stress, reduces the cellular defense mechanism by depleting glutathione (GSH), inhibits sulfhydryl-dependent enzymes, or increases cellular sensitivity to oxidative damage by altering membrane integrity.[4]

HEAVY METAL-MEDIATED CHANGES IN PLANTS

Plants defends themselves against free radical damage by avoiding the oxidation of some lipid, protein, and nucleic acid components. The strategies used by plants to defend themselves from the effects of HM include the buildup of secondary metabolic products such as phenolic compounds, flavonoids, glutathione, proline, and antioxidant enzymes. These strategies depend on the plant's species used as well as the potential concentration of metal contamination [21] (Figure 2). It's also important to note that plants have a common ROS-scavenging mechanism, called superoxide dismutase (SOD), which converts oxygen to hydrogen peroxide. The H_2O_2 is then detoxified to water molecules H_2O by the enzymes catalase (CAT), peroxidase (POX), or glutathione peroxidase (GPX), which prevent the production of OH• radicals.[22] The main intracellular antioxidant molecule inside the cellule is glutathione. It has been found practically in every cell compartment, including the cytosol, chloroplasts, endoplasmic reticulum, vacuole, and mitochondria. GSH is one of the main non-protein thiol sources in most plant cells, where it plays a crucial role in numerous cellular detoxification processes and protects cells from oxidative stress induced by HM. [22]

POTENTIAL IMPLICATIONS OF HEAVY METALS ON HUMAN HEALTH

In recent years, the impact of heavy metal toxicity on human health has received a lot of attention. The primary pathway for HM from contaminated soil to humans is through plants. HM possess low rates of rein excretion, which means that even at very low amounts, they may have harmful effects on human health. Metals like Mn, Cu, Zn, Cr, and Fe are necessary nutrients because they are crucial for physiological processes. However, if it's consumed more than permissible limits it may become toxic.[24,25] The increased dietary HM intake was generally linked to several health problems, including decreased immune system defenses, fetal deformity, gastrointestinal cancer, heart disease, altered neurological and psychosocial behavior, and many others.[26,27]

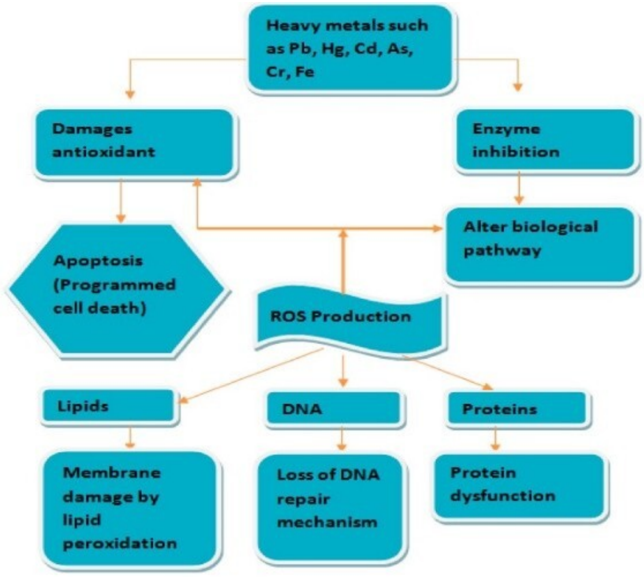

Figure 2. Attack of heavy metals on a cell resulting in the production of reactive oxygen species (ROS) (from Anyanwu et al. [23] according to the provisions MDPI).

Various Heavy Metals and Their Toxicity

The literature was examined for information on the toxic effects of HM. Table 2 outlines the toxic effects of five HM (As, Cr, Hg, Cd, and Pb) on body organs as well as the underlying mechanisms that cause these effects.[28–46]

Excessive exposure to Pb can cause hypertension, appetite loss, stomach pain, headaches, renal dysfunction, arthritic symptoms, lethargy, vertigo, insomnia and hallucinations. Also, Pb can cause urological, respiratory, gastrointestinal, neurological symptoms, muscular, cerebral, as well as reproductive impairments and cardiovascular problems due to immunological modulation, oxidative and inflammation processes.[2,5,8] Pb is classified as a possible carcinogen. It is a protoplasmic toxin with a preference for grey matter in the brain. It infiltrates neurons, destroys cells, nerve synapses, dendrites, and decreases the quantity of oxygen-carrying red blood cells. It binds to phosphorous and enters the bloodstream, where it is transported to the liver, spleen, and kidneys. [47]

Zn is an essential oligo-element required for blood clotting, growth, thyroid function, and DNA and protein synthesis. There is little evidence on the toxicity of Zn; nevertheless, an excess of Zn level has deleterious effects on the immune system, Cu levels, and blood lipoprotein levels. The legal limit for iron in medicinal herbs has not yet been determined. Iron is important in the human body for various reasons, including energy production, oxygen supply, and the immune system.[5] The high level of iron is stored in the pancreas, heart, liver, hypophysis, skeletal muscles, and suprarenal glands. When the organ body receives too much Fe, it escapes from its storage sites and enters the bloodstream, where it is carried to the brain. High levels

Table 2. Comparison of the organ effects, permissible limits, and mechanisms of some heavy metal toxicity.

Toxic Metals	Organ Toxicity	Permissible limits (mg/l)	Disrupted Macromolecule/Mechanism of Action	Reference
As	-Cardiovascular dysfunction - Central nervous system (CNS) injury -Skin and hair changes -Liver damage -Gastrointestinal (GI) discomfort	0.02	-Alterations in neurotransmitter homeostasis -Uncoupled of oxidative phosphorylation (Inhibition of ATP formation) -Damaged capillary endothelium -Thiol binding (GSH conjugation)	28-31
Cr	-Kidney dysfunction -GI disorders -Dermal diseases -Increased occurrence of cancers, including bladder, kidneys, lungs, larynx, testicular, bone, and thyroid	0.05	-DNA damage -Genomic instability -Oxidative stress and ROS generation	32,33
Hg	-CNS injuries -Renal dysfunction -Hepatotoxicity	0.01	-Aquaporin mRNA reduction -Glutathione peroxidase inhibition -Increased c-fos expression -ROS production -Enzyme inhibition -Thiol binding (GSH conjugation)	31,34-37
Cd	-Degenerative bone disease -Kidney dysfunction -Liver damage -Lung injuries -GI disorders -Metabolic syndromes associated with Zn and Cu -Cancer	0.06	-miRNA expression dysregulation -Apoptosis -Endoplasmic reticulum stress -Cd-MT absorption by the kidneys -Dysregulation of Ca, Zn, and Fe homeostasis -Low serum PTH levels -ROS generation -Altered phosphorylation cascades	31,38-42
Pb	-CNS injury -Hematological changes (anemia) -Pulmonary dysfunction -GI colic -Liver damage -Reduced pulmonary function -Cardiovascular dysfunction	0.1	-Enhanced levels of inflammatory cytokines: IL-1β, TNF- ά, and IL-6 in the CNS -Increased serum ET-1, NO, and EPO levels -Inactivation of δ-ALAD and ferrochelatase (inhibition of heme biosynthesis) -Reduced GSH, SOD, CAT, and GPx levels	31,43-46

of iron in the brain destroys neurons, resulting in neurodegenerative diseases and neurological dysfunction with symptoms similar to Alzheimer's disease.[47]

Cd at high doses has a negative toxicological effect on the human body. The kidney is the most vulnerable organ in the exposed population. Cd excretion is slow and it accumulates in the kidney for a long time, resulting in irreversible renal damage. Cd has substantial effects on the liver, vascular and immunological systems, respiratory system, as well as renal and cardiovascular issues. [5,48]

As is a toxic heavy metal that is one of the most serious threats to human health. It is well-known as the king of poisons and the poison of kings.[49] As poisoning, both acute and chronic, is linked to the malfunctioning of various essential enzymes.[2]

The Hg toxicity causes acrodynia or pink disease. Cumulative Hg exposure may alter the structure of the brain, resulting in shyness, cognitive loss, tremors, irritability, and vision or hearing impairment.[2] Exposure to higher concentrations of metallic Hg vapours over a shorter period may cause pulmonary edema, diarrhea, nausea, vomiting, skin eruptions, and an increase in arterial pressure. The symptoms of organic Hg toxicity include fatigue, memory problems, depression, hair loss, headaches, and tremors. Because these signs are frequently combined with other disorders, the circumstances can be difficult to identify.[8]

Heavy Metal-Mediated ROS Generation

The production of free radicals, primarily ROS and reactive nitrogen species (RNS), by toxic metals has the potential to cause oxidative stress. For example, Cd may indirectly produce radicals such as hydroxyls (OH.), O_2.-, and NO., which could weaken the antioxidant defense of cells.[50] Pb significantly decreased antioxidative parameters such as SOD, CAT, GST, GPx and GSH while increasing oxidative parameters such as H_2O_2 and MDA.[51] It has been shown that As produces oxygen (O_2), H_2O_2, O_2.-, nitric oxide (NO.) and peroxyl radicals (ROO.). The production of ROS and RNS induced by Cr decreases the antioxidant cellular capacity causing oxidative stress which increases the toxicity of proteins, lipids and DNA.[52,53] High Hg affinity for -SH groups can inhibit several intracellular receptor signaling as well as reduce glutathione peroxidase capacity. Moreover, Me-Hg increases the activation of phospholipase D (PLD), which is involved in many human diseases such as cancer.[54,55] The toxicity of Hg and Pb induced directly the ROS production or indirectly via reducing the antioxidant cellular system. However, it is believed that the Cd indirectly generates ROS. This may be caused by the substitution of Cd by Fe and Cu in cellular proteins. The result of this excessive accumulation of Fe and Cu is oxidative stress [56,57] (Figure 3).

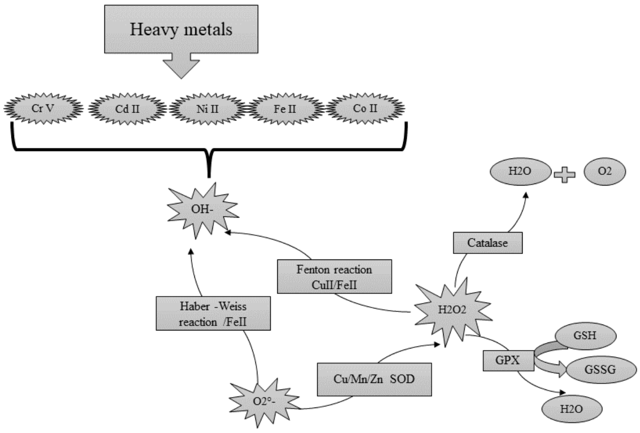

Figure 3. Generation of ROS by heavy metals.

IMPORTANCE OF MEDICINAL PLANTS AND THEIR HEAVY METAL CONTENT

Plant and herbal treatments have begun to receive more attention even in developed nations. For thousands of years, phytotherapy has been used and trusted over the world due to its ease of availability and limited side effects. The therapeutic properties of these plants are usually related to the presence of phytochemical content, with the most important of these phytochemicals being alkaloids, tannins, flavonoids, and phenolic compounds. However, the transportation and storage of medicinal plants may cause the loss of active components, and the production of inactive metabolites and toxic metabolites plays a key role in plant contamination.[31] In the present study, we focused on three different species of plants found in Setif, Algeria, which are *Zygophyllum* species, *Thymus vulgaris*, and *Mentha longifolia* about their chemical composition, medicinal properties as well as HM content.

Zygophyllaceae Family

The Zygophyllaceae is a family of about 25 genera and 240 species adapted to Mediterranean and semi-desert climates.[58] The *Zygophyllum* species are a class of succulent plants that can be resistant to salt and/or dryness tolerant and live in dry and severe climates. Moreover, several authors have listed it as one of the crucial species of desert vegetation.[59] It is believed that *Zygophyllum* species' growth and distribution are influenced by the chemical composition of the soil in their habitats.[59] There are 100 species in the genus *Zygophyllum*, which are found in steppe and desert habitats from the Mediterranean, South Africa, Central Asia, and Australia.[60] It is a perennial herb with fleshy flowers and leaves. The majority of *Zygophyllum* species include *Z. album, Z. simplex, Z. fabago, Z. cocceniem*, and *Z. dumosum*.[61–98] (Table 3). The species of *Zygophyllum* has indicated several biological properties including antidiabetic, antioxidant, antimicrobial, antitumor and anti-inflammatory activities.[62–67] These activities are attributed to their phytochemical compounds including phenolic, flavonoids, essential oils, triterpenes, sterols esters and saponins.[68–73]

The therapeutic uses of *Zygophyllum* species are reported together with that of *Menta longofolia* and *Thymus vulgaris* in Table 3.[62,64,66,67,74–100]

Thymus vulgaris L. (Thym)

T. vulgaris L. (Thym), belonging to the Lamiaceae family, is a living herbaceous plant. The plant is indigenous to southern Italy and the western Mediterranean region. There are 350 different species of thyme grown around the world.[101] Thym has different volatile oils as the primary chemical components. The most significant components of volatile oils in Thym species are thymol and carvacrol, however, this genus also contains several chemotypes. Some other chemical components of the Thymus species are caffeic acid, flavonoids (e.g. thymonin, cirsilineol, and 8-methoxycirsilineol), "*Labiatae* tannin" (rosmarinic acid), triterpenoids, aliphatic aldehydes, and long-chain saturated hydrocarbons.[102] The chemical nature of medicinal plants has a significant impact on their biological activity. Thymol has numerous biological proper-

Table 3. Reported the therapeutic uses of *Zygophyllum* species, *Menta longofolia* and *Thymus vulgaris*.

Medicinal Plants	Therapeutic Uses
***Zygophyllum* species**	
Z. dumosum	Antimicrobial and antifungal[74]
Z. cornutum	Antioxidant[75] ; Antidiabetic[76]
Z. fabago	Antioxidant and cytotoxic activity[77]; Antimicrobial and antifungal[78]; Urease inhibitor [79]
Z. album	Anticancer[80] Anti-acetylcholinesterase[81] Antihyperlipidemic [82] Weight lowering [83] Antihyperglycemic, and the protective hematological proprieties [84] Antidiabetic [64,82, 85] Antioxidant [80,81,85]. Anti-inflammatory [80,84]
Z. coccienium	Antioxidant [86] Anti-hypertensive [87] Antimicrobial and antifungal [88] Cytotoxic activity [89]
Z. gaetulum	Antidiabetic [90,91] Antiinflammatory [92] Hepatoprotective and antioxidant [90]
Z. geslini	Antidiabetic [62,93]
Z. hamiense	Hepatoprotective and antioxidant [94]
Z. macropodum	Analgesic and anti-inflammatory [95]
Z. qatarense	Antimicrobial and antifungal [96]
Z. simplex	Antioxidant [66,97] Anti-inflammatory [97,98] Analgesic [98] Antimicrobial [67]
Thyme (*Thymus vulgaris*)	Antispasmodic and antioxidant, Anthelmintic, as a substitute for cancer prevention [99]
Mint (*Mentha longifolia*)	Antimicrobial, Antispasmodic, Anticancer properties, used as antioxidants in food preservatives [100]

ties, including antioxidant[103], antifungal[104], antibacterial[105], immunomodulating[106], and anti-inflammatory.[107] Carvacrol also has antioxidant antifungal, antimicrobial[108–110], anticarcinogenic and antimutagenic activities.[111]

Mentha longifolia L. (Mint)

Wild Mint (*M. longifolia* L.), also known as *M. sylvestris* L., is a herbaceous plant that belongs to the Lamiaceae family. There are about 20 species of the genus *Mentha*, with numerous variants and subspecies, including a wide range of culinary and medicinal herbs. The species *M. longifolia* is widespread in southern Africa, Eurasia, Egypt, the Atlas Mountains and the Arabian Peninsula.[112] The essential oil and extract of wild mint have a variety of pharmacological properties, including antispasmodic, antimicrobial, and anticancer activities. It can be used as an antioxidant in food preservation products.[100] Moreover, the volatile oil components of mentha have been linked to its biological activity, while phenolic compounds can also have a significant effect. The flavonoid containing is eriocitrin, hesperidin, luteolin-7-O-rutinoside and narirutin. The phenolic acid contained is caffeic acid, rosmarinic acid, and protocatechuic acid.[113]

Content of Heavy Metals in *Zygophyllum* Species

The number of metals found in plant species varies from type to type and from one region to another. Moreover, it is increasingly concentrated in contaminated areas than in non-contaminated ones. Where their concentration varies in seeds, leaves, roots, and flower parts. The concentrations of heavy metal Fe, Zn, Cu, and Al in the *Z. album* from the polluted area were 3.82 $\mu g\ g^{-1}$, 0.615 $\mu g\ g^{-1}$, 0.035 $\mu g.g^{-1}$ and 99.3 $\mu g\ g^{-1}$ compared to 2.35 $\mu g\ g^{-1}$, 0.405 $\mu g\ g^{-1}$, 0 and 56 $\mu g\ g^{-1}$ from the unpolluted area of Southern Sinai, Egypt. While the results of *Z. coccineum* were close to the concentrations of *Z. album*.[114] The highest concentration of Pb, 10.98 μg/g, was found in the root of *Z. album* in the Adrar region, while Cd has the highest concentration, 2.145 μg/g, in the root of *Zygophyllum* sp. in Oued Souf.[115] The higher level of Cd in the root of *Zygophyllum* sp. might be explained by pollution of the soil in the area where the harvest was performed.

Z. album can accumulate and extract 13 HM, including Al, Cu, Fe, Mn, Mo, Zn, Cr, Pb, Co, Ni, Ag, Cd, and Ba from soil that has been contaminated by wastewater used in Jeddah City, Saudi Arabia.[116] Al was the most abundant metal found in the *Z. album* at a location near the sewage water discharge zone, where it had a concentration of 3166 mg/L, compared to 85.2 mg/L in an unpolluted area. Therefore, the species Z. *album* is a hyper-accumulator of the HM.[116] Z. *album* accumulated HM in its leaves at concentrations higher than those found in the stem and root, including Cu, Zn, Al, Ba, Cd, B, Ag, Ni, Mn, Fe, and Cr.

According to Khairia[117], seven plant species *Cassia italika, Cyprus laevigatus, Calotropus procera, Citrullus colocynthis, Argemone maxicana, Phragmite australis,* and *Rhazya stricta* accumulate HM such as Cr, Cd, Ni, Cu, Pb, Co, Fe, Zn in the Reiyad area in Saudi Arabia and the strongest accumulation was found in the roots, followed by the stem, then the leaves, with the exception of Cd, which is about equally accumulated in the stem, root, and leaves.

Al-Sodany et al.[118] found that the Phragmite australis species in Egypt had the strongest accumulations of HM in the root when compared to the shoot. Mazhoudi et al.[119] determined that aerial parts of plant accumulated metals at a lower rate than roots and proposed that the root might play a significant role in the retention of metals by preventing a toxic build-up in the shoot.

Metal accumulators should adopt three criteria related to their ability to store metals. These criteria are improved metal absorption by the root, effectiveness in moving metal from the root to the shoot, and plant tolerance to a high level of this toxic HM.[97] Sathiyamoorthy et al.[120] measured HM from 42 medicinal plants found in the Néguev Desert. The Fe concentration is the highest, with 3020 μg/g in *Gundelia tournefortii* and 2485 μg/g in *Anchusa strigosa.* The levels of iron obtained in *Z. geslini* are comparatively very low, with a maximum of 2.4 g/g at the leaf and 2.16 g/g at the fruit.[115] Zinc concentrations are significantly higher in *Z. geslini* leaves, reaching 119.10 μg/g. [115] This value is rather close to that reported by Lefevre et al.[121] for a nearby species, *Z. fabago*, which has a range of 150 μg/g. The maximum concentration of Mn, 24.89 μg/g, and Nichel, was 19.78 μg/g. is found in *Z. geslini* leaves and *Z. album* root part respectively[115], however Mg is concentrated in the stems in *Z. album*.[118] The Cr concentration obtained is 4.06 μg/g in the stem and 3.94 μg/g in the root of *Z. geslini*. However, the concentration of As, determined in *Z. geslini*, is higher in the root and leaf, 0.07 μg/g, than in the root and fruit.[115] *Z. album* accumulates Al, Cu, Mn and Zn. Zn is higher in the roots while Al, Cu and Mn are more concentrated in the stems.[122] Table 4 shows the content of heavy metals in *Zygophylum* species.[115,116,123,124]

Content of Heavy Metals in *T. vulgaris*

The heavy metal concentrations Cr and Cd in *T. vulgaris* and *T. serpyllum* in the Ash-shoubak region are not detected.[125] The undetected levels of Cr and Cd in thymus herbs might result from low Cd soil content in Jordan's suburban regions or the cultivation of these plants away from industrial operations like the glass and steel industries, which have been demonstrated to be a source of chromium pollution.[126,127] However, a high concentration of Cd has been found along roads in urban areas of Jordan and its level has increased as traffic density has increased.[126,128]

Pb levels in *T. serpyllum* and *T. vulgaris* were 1.26 and 32.03, respectively.[125] The amount of Pb in T. vulgaris was higher than that found in the wild *T. serpyllum* growing in the Ash-shoubak region. However, it was less than that found in the northern

Table 4. Content of heavy metals (mg/g) in *Zygophylum* species.

Locality	Plant	Area	Part uses	The concentration of metals (mg/g) D.W								Reference
				Cd	Cu	Ni	Pb	Zn	Co	Mn	Fe	
South Sinai, Egypt	*Z. album*	unpolluted	Shoot	Nd	Nd	Nd	Nd	0.410 ± 0.021	Nd	Nd	2.35 ± 0.21	114
South Sinai, Egypt	*Z. album*	polluted	Shoot	Nd	0.035 ± 0.007	Nd	Nd	0.623 ± 0.012	Nd	Nd	3.82 ± 0.25	114
South Sinai, Egypt	*Z. coccineum*	unpolluted	Shoot	Nd	0.015 ± 0.007	Nd	Nd	0.303 ± 0.011	Nd	Nd	1.37 ± 0.19	114
South Sinai, Egypt	*Z. coccineum*	polluted	Shoot	Nd	0.232 ± 0.023	Nd	Nd	0.761 ± 0.042	Nd	Nd	2.40 ± 0.007	114
Oued Souf; Algeria	*Zygophylum.spS*	unpolluted	Leaves	0.058	10.60	12844	3.35	Nd	Nd	Nd	Nd	115
Oued Souf; Algeria	*Zygophylum.sp*	unpolluted	Fruit	0.143	9.19	5.52	3.81	Nd	Nd	Nd	Nd	115
Oued Souf; Algeria	*Zygophylum.sp*	unpolluted	Stem	0.093	9.71	8.13	2.99	Nd	Nd	Nd	Nd	115
Oued Souf; Algeria	*Zygophylum.sp*	unpolluted	Root	2.145	9.1	6.89	3.48	Nd	Nd	Nd	Nd	115
Adrar; Algeria	*Z. album*	unpolluted	Leaves	0.209	7.92	8.00	8.35	Nd	Nd	Nd	Nd	115
Adrar; Algeria	*Z. album*	unpolluted	Fruit	0.058	11.42	13.89	10.54	Nd	Nd	Nd	Nd	115
Adrar; Algeria	*Z. album*	unpolluted	Stem	0.131	9.78	7.82	9.39	Nd	Nd	Nd	Nd	115
Adrar; Algeria	*Z. album*	unpolluted	Root	0.107	11.5	19.78	10.98	Nd	Nd	Nd	Nd	115
Ouargla, Algeria	*Z. gelsini*	unpolluted	Leaves	0.34	6.53	5.31	1.12	102.6	Nd	19.86	Nd	115
Ouargla, Algeria	*Z. gelsini*	unpolluted	Fruit	0.23	5.76	6.10	1.30	74.39	Nd	15.58	Nd	115
Ouargla, Algeria	*Z. gelsini*	unpolluted	Stem	0.18	7.10	1.43	1.10	57.33	Nd	13.64	Nd	115
Ouargla, Algeria	*Z. gelsini*	unpolluted	Root	0.09	6.89	2.08	1.17	22.28	Nd	4.27	Nd	115
Yanbu city, Saudi Arabia	*Z. coccinum*	Yanbu Petrolium refinery	Whole plant	19 ± 0.3	Nd	Nd	18 ± 1.0	Nd	16 ± 0.9	33 ± 1.8	Nd	123
Yanbu city, Saudi Arabia	*Z. coccinum*	Sanitary landfill	Whole plant	14 ± 0.2	Nd	Nd	19 ± 0.5	Nd	13 ± 0.8	30 ± 1.5	Nd	123
Yanbu city, Saudi Arabia	*Z. coccinum*	Light industrial park	Whole plant	13 ± 0.5	Nd	Nd	18 ± 0.3	Nd	13 ± 0.3	19 ± 0.4	Nd	123
Yanbu city, Saudi Arabia	*Z. coccinum*	Alsawary area	Whole plant	3.5 ± 0.1	Nd	Nd	10 ± 0.6	Nd	3.4 ± 0.4	9 ± 0.1	Nd	123
Jeddah city, Saudi Arabia	*Z. album*	unpolluted	Leaves	Nd	2.2 ± 0.0003	1.28 ± 0.004	Nd	24.3 ± 0.04	0.08	38.7 ± 0.002	328 ± 32	116
Jeddah city, Saudi Arabia	*Z. album*	polluted	Leaves	0.23 ± 0.001	22.3 ± 0.09	9.4 ± 0.001	8.9 ± 0.002	231 ± 0.04	0.97	88.4 ± 0.008	1493 ± 13	116
Jeddah city, Saudi Arabia	*Z. album*	npolluted	Stems	Nd	1.3 ± 0.0003	1.15 ± 0.003	Nd	9.6 ± 0.01	0.02	13.1 ± 0.001	115 ± 10	116
Jeddah city, Saudi Arabia	*Z. album*	polluted	Stems	0.08 ± 0.001	14.3 ± 0.002	4.2 ± 0.004	19.4 ± 0.001	104 ± 0.01	0.12	43.7 ± 0.003	736 ± 8	116
Jeddah city, Saudi Arabia	*Z. album*	unpolluted	Roots	Nd	4.2 ± 0.0003	1.82 ± 0.001	Nd	5.6 ± 0.05	0.03	12.2 ± 0.022	112 ± 23	116
Jeddah city, Saudi Arabia	*Z. album*	polluted	Roots	0.02 ± 0.001	17.6 ± 0.002	3.53 ± 0.01	14.9 ± 0.02	83.2 ± 0.01	0.45	24.1 ± 0.002	602 ± 12	116
South of Riyadh Capital in Saudi Arabia	*Z. simplex*	Industrial area	Whole plant	0.3	4.76	7.46	2.71	12.1	Nd	Nd	412.46	124

regions.[129] The concentration of Zn in *Thymus spp.* ranged from 16.01 mg kg-1 (*T. fallax*) to 33.71 mg kg-1 (*Thymus praecox*), which was higher than the concentration of Zn in *T. vulgaris* in Turkey (14.30 mg kg-1).[130] In previous studies on this species, the Zn concentration in *T. fallax* decreased in the range (13.7 to 34.7 mg kg-1)[131]. The location of the *T. vulgaris* cultivation region, may help to clarify the contamination of these species by Pb, in particular, as it is well known that motor vehicles are the main source of Pb contamination. However, Pb was below the toxic level in wild *T. serpyllum* growing in its natural environment.[132] Additionally, the availability of the plant to accumulate the metal and store it, as well as the kind of soil and plant species, may all contribute to variations in the amount of Pb in the plants study.[133,134] The Cu concentrations

Table 5. Content of metals (mg/g) in *T. vulgaris*.

Locality	Plant	Area	Part uses	The concentration of metals (mg/g) D.W									Reference
				Co	Cu	Cr	Mn	Ni	Fe	Pb	Zn	Cd	
Jordan	*T. vulgaris*	Ash-shouback	Aerial parts	Nd	13.23 ± 0.13	Nd	15.52 ± 0.16	23.85 ± 0.03	141.3 ± 0.67	32.03 ± 0.04	16.18 ± 0.24	Nd	139
Egypt	*T. vulgaris*	Cairo and Giza governorates	Whole plant	0.151	Nd	0.606	ND	2.55	Nd	11.16	Nd	0.454	140
Morocco	*T. vulgaris*	Ounarha town	Leaves, woods, and flowers	Nd	4.88	8.76	22.4	Nd	405	Nd	14.3	Nd	141
Iraq	*T. vulgaris*	Erbil	Stem, seeds, root, inner bark, flowers and leaves	Nd	0.61 ± 0.05	Nd	6.33 ± 0.55	0.34 ± 0.28	57.57 ± 3.83	0.41 ± 0.03	5.44 ± 0.52	Nd	142
Libya	*T. vulgaris*	Misurata city	Whole plant	Nd	0.99 ± 0.18	0.66 ± 0.18	Nd	Nd	5.26 ± 2.97	0.43± 0.07	6.53 ± 1.08	0.34± 0.52	143
United Arab Emirates	*T. vulgaris*	Market in Dubai	Whole plant	Nd	3.52 -13.16	Nd	Nd	Nd	120.75 -764.51	9.07-23.52	Nd	-0.63	5
Iran	*T. vulgaris*	Tehran drugstores	Herbal drops	Nd	0.782 ± 0.057	Nd	Nd	Nd	Nd	0.264 ± 0.071	Nd	0.012 ± 3.388	144

Table 6. Content of heavy metals (mg/g) in *M. longifolia.*

Locality	Plant	Area	Part uses	The concentration of metals (mg/g) D.W							Reference
				Cu	Cr	Mn	Ni	Fe	Pb	Zn	
Bosnia and Herzegovina	*M. longifolia* L.	Bugojno	Leaves	7.00 ± 0.01	Nd	40.30 ± 0.01	7.00 ± 0.01	645 ± 0.01	0.10 ± 0.01	13.90 ± 0.01	3
Bosnia and Herzegovina	*M. longifolia* L.	Rudo	Leaves	7.30 ± 0.01	Nd	21.30 ± 0.01	3.70 ± 0.01	755 ± 0.01	0.60 ± 0.01	14.50 ± 0.01	3
Bosnia and Herzegovina	*M. longifolia* L.	Sarajevo	Leaves	10.00 ± 0.01	Nd	37.80 ± 0.01	6.60 ± 0.01	659 ± 0.01	0.90 ± 0.01	29.90 ± 0.01	3
Pakistan	*M. longifolia* L.	Chumra derai	Root, shoot, and leaves	Nd	2.00 ± 0.04	Nd	0.38 ± 0.20	Nd	0.40 ± 0.14	20.54 ± 1.14	153
Egypt	*M. longifolia* L.	Polluted canals in summer	Shoot	354.6 ± 168.1	8.2 ± 1.6	98.5 ± 31.2	168.1 ± 9.9	919.3 ± 108.16	2.6 ± 1.0	282.6 ± 70.5	100
Egypt	*M. longifolia* L.	Polluted canals in summer	Root	646.7 ± 193.7	16.0 ± 4.6	156.4 ± 47.2	420.1 ± 47.6	2054.3 ± 436.9	0.6 ± 0.1	544.9 ± 191.7	100
Egypt	*M. longifolia* L.	Unpolluted canals	Shoot	194.7 ± 86.0	3.2 ± 1.1	39.9 ± 27.9	156.4 ± 58.1	573.4 ± 351.9	6.7 ± 6.2	233.7 ± 67.2	100
Egypt	*M. longifolia* L.	Unpolluted canals	Root	372.7 ± 97.3	8.8 ± 2.0	79.4 ± 50.6	288.4 ± 101.4	1270.1 ± 714.2	1.7 ± 2.5	455.9 ± 123.6	100
Montenegro	*M. longifolia* L.	Slopes of Mount Bjelasica	Root	32.8±2.61	Nd	78.8±4.21	2.91±0.35	1 457±92	Nd	58.2±4.47	154
Montenegro	*M. longifolia* L.	Slopes of Mount Bjelasica	Stem	28.2±2.33	Nd	49.1±2.21	0.37±0.03	94.1±12	Nd	10.6±1.32	154
Montenegro	*M. longifolia* L.	Slopes of Mount Bjelasica	Leaf	20.7±2.39	Nd	52.3±4.67	3.06±0.32	183±13	Nd	25.5±4.12	154

in the *Tymus* species were within legal limits (2-20 ppm). The Cu concentration ranged from 13.23 ppm in *T. vulgaris* to 10.4 ppm in *T. serpyllum*[125] This difference in the Cu concentration may be caused by genetic variation across plant species[135] or by varying plant heavy metal selectivity[126], or it influenced by the effect of anthropogenic activities and high heavy traffic levels, which could cause accumulation of Cu metal in the soil.[126,136] The concentration of Zn in *T. vulgaris* was higher than that found in *T. serpyllum*, while the concentrations of Mn were found to be close in *T. vulgaris* and *T. serpyllum.*[125] (Table 5). The high amount of Mn may be attributed to the development of industrial and residential locations rich in Mn and Ni and their use as fuel additives similar to Pb.[137] The optimal range for Co concentration in *T. serpyllum* was between

0.02 and 1.0 mg/kg.[9] The presence of cobalt in *T. serpyllum* and its absence in the other species may be explained by the fact that the occurrence of Co is essentially dependent upon the species. The absorption is controlled by various mechanisms in various species. Physical factors including pH, temperature, the salinity of the environment, and the presence of some metals influence the effect of Co absorption and accumulation in different plant species.[138] Table 5 shows the content of metals in *T. vulgaris*.[139,140–144]

Content of Heavy Metals in *M. longifolia*

The mentha herb may contain harmful elements, such as HM derived from the environment during cultivation, storage or harvesting. Furthermore, contamination of the herb by various HM may also occur during plant cultivation as a result of soil content, the presence of nutritive elements, fertilizers and during the treatment and packaging of herbaceous materials.[5,143,145] Metal concentration ranged from 0.10 mg/g for Pb to 755.00 mg/g for Fe in *M. longifolia L.* (wild species), while metal ranged from 0.40 mg/g for Pb to 1108.40 mg/g for Fe in *M. piperita L.* (cultivated species). Cr and Cd content in wild species were lower than WHO standards.[143,146] Furthermore, the highest concentration was found in Fe, and the lowest concentration was found in Pb. A relatively low Cu concentration in wild species compared to Zn concentration occurs because higher Cu content in soil can reduce Zn availability in plants, due to competition for the same absorption sites in the root of plant.[145,147,148] The highest quantities of trace oligo-elements found in Mentha species are Fe (619.30–1108.40 mg/g), Mn (21.30–94.00 mg/g), Zn (13.90–39.30 mg/g), Cu (4.46–12.50 mg/g) and Cr (0.70–0.90 mg/g). The high amount of Fe is a result of the fact that Fe is mostly found in soil and rock. Cr concentrations in wild *Mentha* were below detection limits.[3] These findings indicate that both *Mentha* species (*M. longifolia* L. and *M. piperita* L.) are good sources of low HM concentrations and are safe for usage as a beverage and in various herbal preparations.[3] HM levels in target species growth in contaminated areas may be doubled or tripled when compared to the noncontaminated area. For example, the Fe content in the root of *M. longifolia* was 14.56 (mg/kg) DW in contaminated soil, and this value was decreased to around half (6.35 mg/kg DW) in non-contaminated soil (Table 6). Sometimes the metal amount of growing plants in a contaminated area yields values that are 10 times higher than those in a non-contaminated area. Furthermore, the metal uptake and accumulation capacities vary significantly among wetland species [123] . This finding may support the concept of employing *M. longifolia* as a phytoremediator, which is consistent with research investigating the phytoremediation potentiality of several species in wetlands.[149–152] Table 6 shows the content of heavy metals in *M. longifolia*.[3,100,153,154]

Our research found that the same medicinal plant species (*Zygophylum, M. longifolia*, and *T. vulgaris*) that grow in different geographical areas accumulate different levels of HM. The concentration of HM varies for different plant species derived from the same geographical location. The amounts of HM detected in the plants collected in some areas were within the legal limits, but levels in other geographical areas exceeded the recommended ranges. As a result, medicinal plants for herbal remedies must be collected from non-polluting natural areas. Our findings also suggest that medicinal plants, whether used for local or medicinal applications, should be obtained from areas free of HM contamination.

CONCLUSION

HM accumulation in plant species is a stress factor that causes a variety of growth problems for plants. As a result, it is critical to avoid harvesting medicinal plants that show signs of stress, because they may be the result of heavy metal accumulation. Toxic metal bioaccumulation has a variety of toxic effects on many organisms, tissues, and organs. Metal poisoning can show as either acute or chronic symptoms. HM interfere with biological functions such as proliferation, growth, differentiation, repair of cellular damage, as well as apoptosis. The main goal of utilizing medicinal plants in disease treatment is to acquire a cure with few or no side effects, yet the presence of HM in plants may pose substantial health risks to consumers if absorbed. Even when they are low toxic levels, those with a long biological half-life tend to accumulate in the body over time, and as a result, long-term absorption of extremely high amounts can result in death.

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