



Heavy Metals Detection in Some Types of Herbs Used in Medical Treatments

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Abstract:

Given the backdrop of the Corona pandemic, the objective of this study was to identify some of the heavy metal presence in specific herbs collected from local markets in Iraq. Eight samples were selected, with four (Chamomile, Laurus nobilis, Artemisia, and Borage) used for COVID-19 prevention and the remaining four (Quince, Clove, Thyme, and Propolis) for treating COVID-19 patients. For the detection of metals in the selected specimens, EDX (energy-dispersive X-ray spectroscopy) was employed. The results revealed the existence of (Ca), (K), (S), (Si), (Al), (Mg), and (Zn) in Chamomile; (K), (Si), (Al), and (Zn) in Laurus nobilis; (Ca), (K), (S), (Si), (Al), (Mg), and (Zn) in Artemisia; (Ca), (K), (P), (Si), (Al), (Mg), and (Zn) in Borage; (Ca), (K), (S), (Si), (Mg), and zinc (Zn) in Quince; (Ca), (K), (Mg), and zinc (Zn) in Clove; (Ca), (K), (Si), (Al), and (Zn) in Thyme; and (Si) in Propolis. All the detected elements are considered essential metals, which are crucial for living biological systems and needed in relatively low concentrations. It is worth mentioning that the selected samples did not contain toxic heavy metals such as (Cd), (Pb), and (Hg), which are regarded as biochemically nonessential. Further analysis for heavy metal content, starting from acid digestion. This process aimed to liberate heavy metals from organic components in the herb samples. The results obtained through atomic absorption confirmed the absence of toxic

heavy elements (Cd, Pb, and Hg). This is considered a positive thing as far as these toxic elements are concerned.

Keywords: Heavy Metals, Herbs, Toxicity, Corona Pandemic.

الكشف عن المعادن الثقيلة في بعض أنواع الأعشاب المستخدمة في العلاجات الطبية

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الخلاصة:

على خلفية جائحة كورونا، كان الهدف من هذه الدراسة هو التعرف على وجود المعادن الثقيلة في أعشاب معينة تم جمعها من الأسواق المحلية في العراق. تم اختيار ثمان عينات، أربع منها (البابونج، لوروس نوبيليس، الشيش، ولسان الثور) تستخدم للوقاية من كوفيد-١٩ والأربعة المتبقية (السفرجل، القرنفل، الزعتر، والعنج) لعلاج مرضى كوفيد-١٩. للكشف عن المعادن في العينات المختارة، تم استخدام (EDX) التحليل الطيفي للأشعة السينية المشتتة. أظهرت النتائج وجود (K)، (Ca)، (S)، (Si)، (Al)، (Mg)، و (Zn) في البابونج. (K)، (Si)، (Al)، و (Zn) في لوروس نوبيليس؛ (Ca)، (K)، (S)، (Si)، (Al)، و (Mg) في الشيش؛ (Ca)، (K)، (P)، (Si)، (Al)، و (Mg) في لسان الثور؛ (Ca)، (K)، (S)، (Si)، و (Mg) في السفرجل؛ (Ca)، (K)، و (Mg)، و (Zn) في القرنفل؛ و (K)، و (Si)، و (Al)، و (Zn) في الزعتر؛ و (Si) في العنج. وتعتبر جميع العناصر المكتشفة معادن أساسية، وهي ضرورية للأنظمة البيولوجية الحية وتحتاج إليها بتركيزات منخفضة نسبياً. ومن الجدير بالذكر أن العينات المختارة لم تحتو على معادن ثقيلة سامة مثل (Cd) و (Pb) و (Hg) والتي تعتبر غير ضرورية من الناحية البيوكيميائية. مزيد من التحليل لمحتوى المعادن الثقيلة تم تنفيذه بدءاً من الهضم الحمضي. تهدف هذه العملية إلى تحرير المعادن الثقيلة من المكونات العضوية في عينات الأعشاب. أكدت النتائج التي تم الحصول عليها من خلال الامتصاص الذري عدم وجود العناصر الثقيلة السامة (الكاديوم والرصاص والزنك). ويعتبر هذا أمراً إيجابياً فيما يتعلق بهذه العناصر السامة.

الكلمات المفتاحية: المعادن الثقيلة، الأعشاب، السمية، جائحة كورونا.

1. Introduction:

Heavy metals are distinguished by their relatively high density (leastwise $5\text{g}/\text{cm}^3$) and their relatively high atomic mass (greater than 23) [1]. In biochemical systems, metals might be classified as essential like (Mn, Fe, Cu, and Zn) because they are significant for living organisms and are needed in the body in very low quantities and nonessential such as (Cd, Pb, and Hg) which up to now, have not been proven to have known essential biological role in living organisms and considered toxic elements [2]. Excess or deficiency of metals in the human body may lead to abnormal conditions or diseases [3]. Accumulation of metals especially heavy ones in plants is highly dependent on the availability or concentration in the soil, also from air pollution as in excused of cars or factories [4] in other words, there is an increased chance of the metal accumulating in plant tissues as the concentration of heavy metals in the soil is bigger [5]. Also, heavy metals percentages are wastewater treatment will increase because of the contaminated soil, they interfere with the plant roots and eventually end up in the food chain, when consumed by living things [6]. Heavy metal is usually absorbed from soil by plants piloe up in the roots and migrates in small portions to all other parts [7]. Several factors affect element migration to plants such as soil chemistry, plant type, kinds of fertilizer used, and climate changes on the earth's surface, but eventually, the most dominant element is metal concentrations and types in soil. The availability of the metals can also be directly affected by the plant independently [8]. Plants which considered medicinal and their sub-products (distillates, extracts, mixtures, etc.) are defined as herbal medicines; at present herbs in various species are extensively used as raw materials in pharmaceutical preparations and widely used in home remedies [9].

Currently, herbal medicines usage has been always increasing mainly because of the popular belief that herbal remedies are natural and consequently, inherently safer if compared to synthetic medicines [10]. Aromatic herbs and volatile oils are known for their antifungal and antibacterial properties. Also, herbal-based medicine originated purely from nature, so it's known to provoke fewer side effects [11]. Herbal medicines and other known natural products have a long history in the treatment of respiratory infections and are deemed as drugs. Mostly, those plant-based medicines have acceptable safety profiles due to their low toxicity in addition; some toxicity of herbal-based plants may be back to contaminants that came from microbes, pesticides, adulterants, and chemical toxins [12].

The main known symptoms of COVID-19 might include cough, fever, and sometimes shortness of breath. Thyme can be one of the remedies for simple lung issues like coughs because, backed by the fact that it contains volatile oils that stimulate tissues inside the lungs,

ease up congestion, and struggle against viruses and bacteria, in addition to stimulating expectoration [13]. Essential oils might be used in different ways like using a diffuser or directly inhaled or used to thin mucus by steam, resulting in better fighting against microbes, expectoration boosting, and increasing the response of the immune system. Plants like *Laurus Nobilis*, have been pleasing smelling oil and might be used specifically for boosting immune cells and exhilarating lymph drainage. Also, Chamomile as an herb is known for its calming abilities that can be consumed to ease pain and reduce inflammations. It is usually marvelous for treating children especially those who are sensitive, irritable, and anxious to pain. Moving towards Borage, which has seed oil known as a plant source of GLA, might actively reduce inflammation. Artemisia oils, besides their antioxidant abilities, inhibit the growth of bacteria, dermatophytes, yeast, and microspores [14].

Cooling and anti-inflammatory effects are also reported in Clove oil, which is also used to clear the breathing passages. This expectorant is useful in many disorders in the respiratory system like asthma, coughs, colds, sinusitis, bronchitis, and tuberculosis. As illustrated in countless studies, Propolis has an exceptional capability to destroy bacteria instantly, not to mention other abilities in fighting against viruses, fungi, and even penicillin-resistant *Staphylococcus*. Researchers have been even looking recently at the abilities of bee propolis to stimulate the immune system. They used a different dose of propolis to measure the response in the immune systems., 3 g/kg of propolis in the daily diet showed a significant boost in antibody levels which otherwise stated increased immune activity [15]. Further looking toward zinc which has been to have the ability to stimulate the immunity system during a virus infection. Moreover, Zn supplements are favored in terms of COVID-19 treatment protocols. The abilities of Zn might be improved by using chloroquine as an ionophore, while Zn if reached inside the infected cell would be able to stop replications of SARS-CoV-2 [16]. Cadmium on the other hand, accumulates in the human body which develops many problems includes for instance; nervous, kidney, cardiovascular, and bone problems extra portions might severely irritate the stomach, which might lead to severe side effects like diarrhea and vomiting, while long-term exposure to fewer amounts might lead to metal accumulations in liver and kidneys. Eventually possible liver, kidney, and obstructive lung disease, and has been also likened to more serious diseases like lung cancer, and osteoporosis in living subjects [17].

2. Aim of the study:

This study aims to determine heavy metals in some types of herbs used in medical treatments. Experimental & Sampling. Eight samples of common herbs that were used through the COVID-19 period from local Iraqi markets in Baghdad include Chamomile, *Laurus nobilis*,

Artemisia, borage, Quince, clove, Thyme, and propolis. The first four samples (Chamomile, Laurus nobilis, Artemisia, and borage) have been chosen on the basis that they were used for preventive purposes from the Coronavirus, while the other four (Quince, clove, Thyme, and propolis) were chosen on the basis that they were used as medicinal herbs for Coronavirus patients.

Table 1: The selected samples with its origin

Sample No.	Herb Type	Country
1	Chamomile	India
2	Laurus nobilis	India
3	Artemisia	Thailand
4	Borage	Thailand
5	Quince	Iran
6	Clove	Iraq
7	Thyme	Iraq
8	Propolis	Iraq

Figures 1 to 14 illustrate the herbs used in this project and their original plants while **Figures 15 and 16** show the beehive and propolis which extracted from it.



Figure 1: Chamomile herb



Figure 3: Laurus Nobilis herb



Figure 5: Artemisia herb



Figure 7: Borage herb



Figure 2: Chamomile flowers



Figure 4: Laurus Nobilis plant

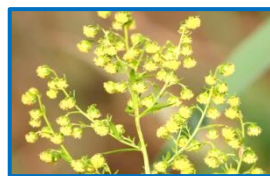


Figure 6: Artemisia flowers



Figure 8: Borage flowers



Figure 9: Quince herb



Figure 11: Clove herb



Figure 13: Thyme herb



Figure 15: Propolis

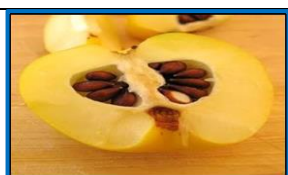


Figure 10: Quince Seeds

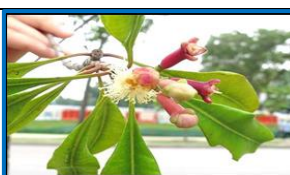


Figure 12: Clove flowers



Figure 14: Thyme plant



Figure 16: Propolis in the beehive

3. Reagents and Chemicals:

Nitric acid (Analytical grade, 65%, Sigma Aldrich) and HCl (Analytical grade, 37%, Sigma Aldrich) were deployed to prepare samples. All the solutions were prepared using deionized water. Reference solutions (Mg, Zn, K, Cu, Cr, Ni, Co, Cd, Pb, and Fe) were prepared from 1000 mg/l in deionized water from Standard Stock Solution of GFS Fishers' (AAS Reference Standard for calibration). Dilution correction as required was implemented for samples during analysis.

3.1 Samples Drying

One gram of each sample was dried from water by putting them separately in watch glasses, then further dried by putting them in an electrical oven at 100 °C and later weighed, then the samples marked with numbers 1, 2, 3, 5, 6, 7, 8 in the same time 0.5g of sample 4 were prepared separately in the same procedure.

3.2 Grinding and Sieving

To increase the surface area for more efficient acid digestion, an electric grinder was used to grind the dried plants. Afterward sieving was implemented for the grinded samples to separate the preferred size of particles for digestion .

3.3 Acid Digestion

A sample of 1g of 1,2,3,5,6,7,8 and 0.5g of 4 of the selected dry samples were placed separately into 200ml beakers. A 15ml of 65% HNO₃ with 10ml of 37% HCL was added. The contents were placed in a beaker and stirred thoroughly on a hot plate surface. The process was ongoing until the contents were mostly dissolved. The digested solution was filtered using Whatman filter paper No.41. After cooling filtrate solutions were later diluted with deionized water to 100ml. The prepared solution was used as samples for spectrophotometric determination of various metal analyses. According to laboratory safety regulations, this procedure was entirely accomplished in the fume hood.

3.4 Energy-Dispersive X-Ray Spectroscopy (EDX) Analysis

Energy-dispersive X-ray spectroscopy (EDS, EDX, or XEDS) Bruker model XFlash6I10, is the analytical technique employed for the elemental characterization of the samples. The tests were conducted at Al-Nahrain University.

4. Result and Discussion:

4.1 EDX Analysis

Chamomile, Laurus nobilis, Artemisia, Borage, Quince, Clove, Thyme, and Propolis herb samples were tested in an EDX instrument to determine elements of interest. Results obtained for this test are shown in **Figures 17, 18, 19, 20, 21, 22, 23, and 24** respectively.

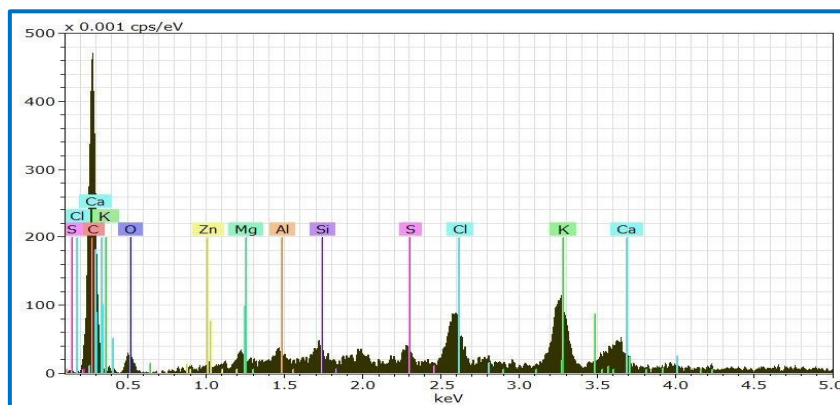


Figure 17. EDX, spectrum for Chamomile herb sample

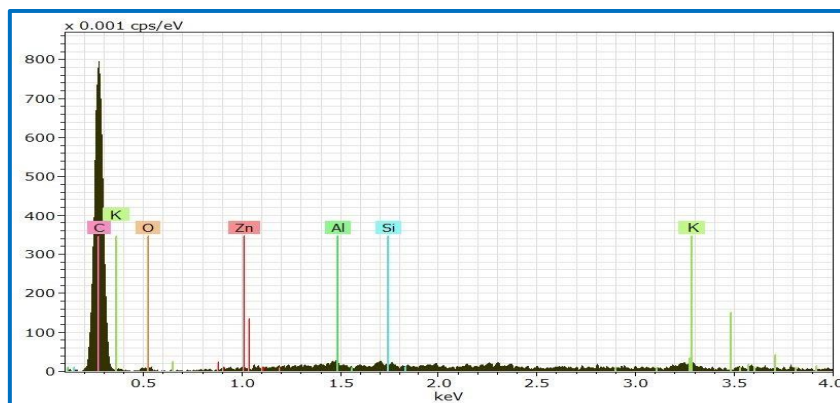


Figure 18. EDX, spectrum for Laurus nobilis herb sample

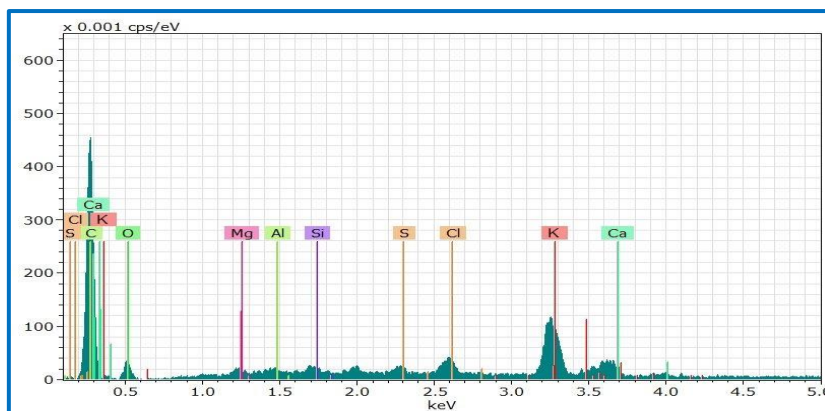


Figure 19. EDX, spectrum for Artemisia herb sample

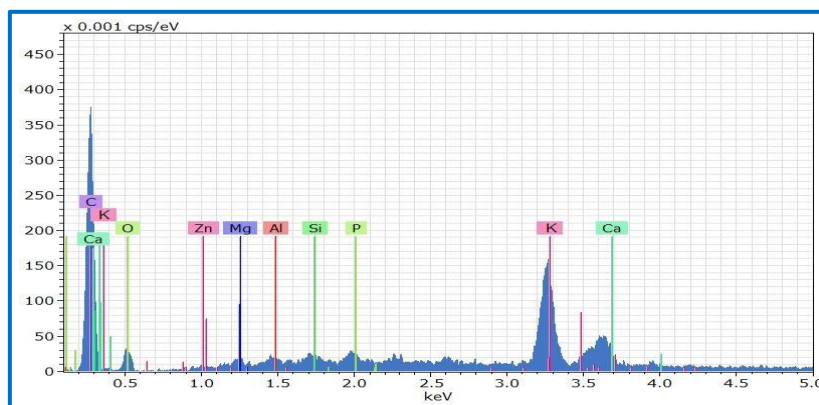


Figure 20. EDX, spectrum for Borage herb sample

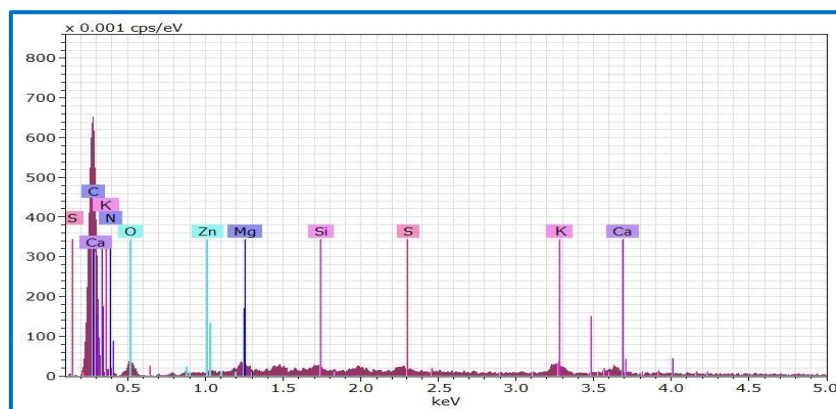


Figure 21.EDX, spectrum for Quince herb sample

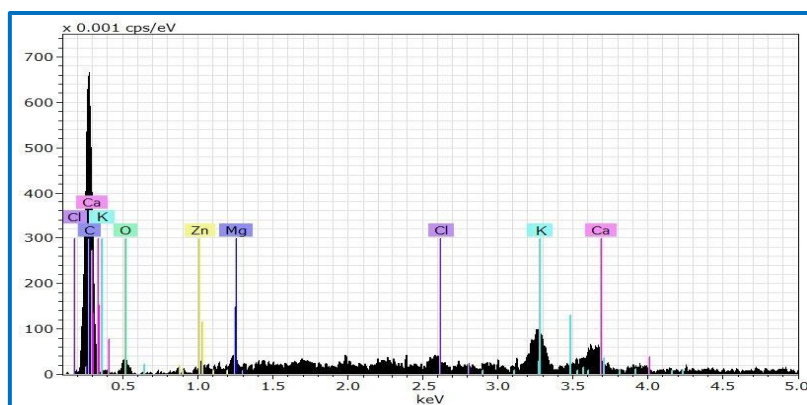


Figure 22.EDX, spectrum for Clove herb sample

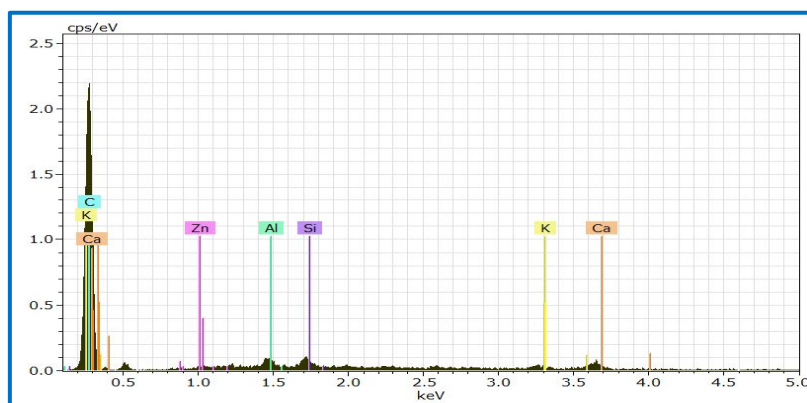


Figure 23. EDX, spectrum for Thyme herb sample

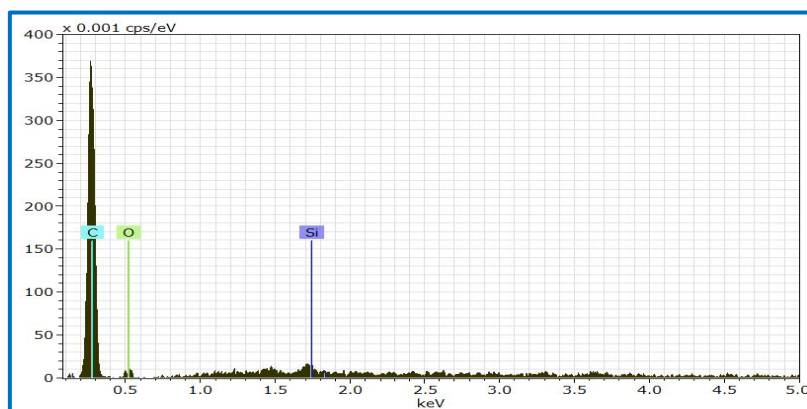


Figure 24. EDX, spectrum for Propolis sample

EDX analysis shows the presence of (Ca, K, S, Si, Al, Mg, and Zn) in Chamomile, (K, Si, Al, and Zn) in *Laurus nobilis*, (Ca, K, S, Si, Al, and Mg) in *Artemisia*, (Ca, K, P, Si, Al, Mg and Zn) in *Borage*, (Ca, K, S, Si, Mg and Zn) in *Quince*, (Ca, K, Mg and Zn) in *Clove*, (Ca, K, Si, Al and Zn) in *Thyme* and (Si) in *Propolis*. All the elements that were detected were considered essential heavy metals which are crucial for living bodies and required in bodies in very low concentrations.

It is very important to notice that the selected samples did not contain heavy metals like (Cd, Pb, and Hg) which are poisonous and are regarded as biochemically nonessential.

The clarification here is that the concentrations of zinc in some herb samples are very simple, while the other samples lack zinc elements. In both cases, all herbs used as prevention from the Coronavirus or curative for Corona patients require the use of zinc separately in a dose estimated by the doctor.

To find out the exact concentrations of the elements that were diagnosed in the EDX device with others that research and studies indicated their presence and may be in low concentrations that were not detected by the EDX due to their concentrations being outside the sensitivity limits of the device, a digestion process was carried out for the selected samples. Acid digestion with a mixture of (65% HNO₃ with 37% HCL) was carried out to free metals from the organic constituents in herb samples to be tested in an atomic absorption instrument for metal detection and concentration determination. Atomic absorption results confirmed the absence of toxic heavy elements (Cd, Pb, and Hg) in the concentration level of mg/Kg as in EDX results.

5. Conclusion:

The eight herb samples that were selected from Iraqi markets were researched using Energy-dispersive X-ray spectroscopy (EDX). EDX data shows the existence of (Ca, K, S, Si, Al, Mg, and Zn) in Chamomile, (K, Si, Al, and Zn) in *Laurus nobilis*, (Ca, K, S, Si, Al, and Mg) in *Artemisia*, (Ca, K, P, Si, Al, Mg and Zn) in *Borage*, (Ca, K, S, Si, Mg and Zn) in *Quince*, (Ca, K, Mg, and Zn) in *Clove*, (Ca, K, Si, Al, and Zn) in *Thyme*, and (Si) in *Propolis*. All the elements detected are considered essential metals which are crucial for living organisms' activities nevertheless in specifically low concentrations. It is very important to notice that the selected samples did not contain heavy metals like (Cd, Pb, and Hg) which even at these low levels are considered very toxic and are categorized as biologically nonessential. Atomic absorption results confirmed the unavailability of the toxic heavy elements (Cd, Pb, and Hg) at the concentration level of mg/Kg as in EDX results.

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