

Original Research Article

X-ray fluorescence spectroscopic determination of heavy metals and trace elements in aerial parts of *Origanum sipyleum* L from Turkey

Cenk Durmuşkahya¹, Hakan Alp², Zehra Sinem Hortoğlu³, Ümit Toktas⁴ and Hüsniye Kayalar^{4*}

¹Department of Forest Engineering, Faculty of Forestry, Izmir Katip Celebi University, Balatçık, İzmir, ²Department of Chemistry, Karadeniz Technical University, Trabzon, ³Center for Materials Research, Izmir Institute of Technology, Urla, ⁴Department of Pharmacognosy, Faculty of Pharmacy, Ege University, Bornova, Izmir, Turkey

*For correspondence: **Email:** husniyekayalar@gmail.com; **Tel:** +905543986021

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Abstract

Purpose: To determine the heavy metal and trace element composition of the powdered aerial parts of *Origanum sipyleum* L. and its water extract.

Methods: The heavy metal and trace elements content of the powdered plant material and 2 % aqueous extract were evaluated by x-ray fluorescence spectroscopy with silicon drift detector SDD at a resolution of 145 eV and 10,000 pulses. The process conditions were 0.1 g sample weight, process time of 300 s at a voltage of 25 kV and 50 kV, and at a current of 0.5 and 1.0 mA under helium atmosphere.

Results: The major elements, K, Ca and Na, known as macronutrients, constituted 11990, 10490 and 970 ppm of the powdered drug and 8910, 2991 and 810 ppm of the water extract, respectively. Among other constituents, arsenic, lead and uranium levels were < 1, 2.1 and < 3 ppm, respectively, in the powdered material while in the aqueous extract, the levels were < 1, < 2 and 200 ppm, respectively.

Conclusion: *O. sipyleum* is a potential source of macro- and micronutrients from which useful food additives and health supplements can be derived.

Keywords: Lamiaceae, *Origanum sipyleum*, Trace elements, Heavy metals, X-ray fluorescence

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INTRODUCTION

Origanum species have been used as spices as well as for protection and treatment of various diseases in traditional medicine for thousands of years [1]. Aerial parts of *Origanum* species are used as diuretic, antitussive, antineuralgic and for improving stomach function and bronchial secretion. *Origanum* species, native to Cyprus, are also traditionally used against common cold and fever, and for spasmolytic actions and antirheumatic purposes via external application [2].

Origanum sipyleum, native to western Anatolia, where it is widely used as a spice and against gastrointestinal disorders and cough. This perennial plant, commonly known as "bayırçayırı", have been used as a medicinal tea, food additive and for the production of essential oil [3,4].

Plants assimilate elements dissolved in water through their roots. In addition to purity, safety and efficacy assessments, a part of quality control studies on medicinal plant is the detection

and quantification of heavy metals. Medicinal plants are taken in different forms such as teas, capsules, syrups and tablets containing ground or powdered form of pharmaceutical materials. Extracts prepared with different solvents are also used as active ingredients.

In the present work, the aim is to obtain a better understanding of the medicinal and nutritive composition of *O. sipyleum*, including heavy metals and trace elements, of the powdered plant material and water extract, using x-ray fluorescence spectroscopy.

EXPERIMENTAL

The aerial parts of *Origanum sipyleum* were collected from Spil Mountain, Manisa, Turkey. Dr Cenk Durmuşkahya from İzmir Katip Çelebi University authenticated the plant material and a voucher specimen (no. 1445) was deposited in the herbarium of Pharmacognosy Department, Faculty of Pharmacy, Ege University.

The aerial parts were collected, air-dried and ground into fine powder. Both the powdered plant material and the water extract prepared as a 2 % infusion, were analysed for heavy metals and trace element using SPectro IQ II instrument equipped with dispersive x-ray fluorescence (XRF) technology at a resolution of 145 eV at 10,000 pulses for the SDD detector. Bragg crystal polarized the primary beam and highly ordered pyrolytic graphite (HOPG) was the target; other conditions include a duration of 300 s at a voltage of 25 kV and 50 kV, 1 mA helium atmosphere and 0.5 and 1.0 mA current.

RESULTS

It was observed that *O. sipyleum* aerial parts and its water extract contain significant values of elements. The results are shown in Table 1. Major elements such as K, Ca and Na, known as macronutrients, are detected at levels of 11990, 10490 and 970 ppm in the powdered drug and 8910, 2991 and 810 (ppm) in the water extract, respectively. Except for trace elements such as Zn, Cu, Br, Mo, Mn and Ag, heavy metal contents were higher in the water extract than in the powdered material. The concentrations of toxic elements such as Pb, Cd and As were low in both the powdered material and its water extract. Aluminum also regarded as a toxic element was present at a high concentration in the powdered material but was much lower in the extract. On the other hand, the water extract

contained higher levels of Cu and Zn than the powdered drug.

Table 1: Metal content of powdered (OS) and water extract (OSw) of *Origanum sipyleum*

Z	Symbol	Element	OS (ppm)	OSw (ppm)
11	Na	Sodium	970	810
12	Mg	Magnesium	1050	643
13	Al	Aluminum	4381	222.7
14	Si	Silicon	782.3	266.7
15	P	Phosphorus	1387	704.4
16	S	Sulfur	1972	1428
17	Cl	Chlorine	578.5	552.4
19	K	Potassium	11990	8910
20	Ca	Calcium	10490	2991
22	Ti	Titanium	21.6	7.5
23	V	Vanadium	5.1	6.5
24	Cr	Chromium	< 5.1	< 5.1
25	Mn	Manganese	120.1	162.8
26	Fe	Iron	74.3	44.2
27	Co	Cobalt	< 3	< 3
28	Ni	Nickel	< 2	< 2.4
29	Cu	Copper	33.7	879
30	Zn	Zinc	49.2	2832
31	Ga	Gallium	< 1	< 1
32	Ge	Germanium	< 1	< 1
33	As	Arsenic	< 1	< 1
34	Se	Selenium	< 1	< 1
35	Br	Bromine	4.9	468
37	Rb	Rubidium	45	145
38	Sr	Strontium	11.7	803
39	Y	Yttrium	2.3	316
40	Zr	Zirconium	< 510	< 510
42	Mo	Molybdenum	13.2	1368
47	Ag	Silver	26.5	3350
48	Cd	Cadmium	< 5.2	< 460
49	In	Indium	< 5.1	< 5.1
50	Sn	Tin	< 6.1	< 6.1
51	Sb	Antimony	< 6.1	< 6.1
52	Te	Tellurium	< 7.1	< 7.1
53	I	Iodine	< 7.1	< 7.1
55	Cs	Cesium	< 8.1	< 8.1
56	Ba	Barium	< 8.1	< 600
57	La	Lanthanum	< 10	< 10
58	Ce	Cerium	< 12	< 12
80	Hg	Mercury	< 2	< 2
81	Tl	Thallium	< 2.9	< 2
82	Pb	Lead	2.1	< 2
83	Bi	Bismuth	< 2	< 2
90	Th	Thorium	< 2	259
92	U	Uranium	<3	200

DISCUSSION

K and Na is crucial for life and also play important roles during excitation and transmission in nerve cells. Ca, which promotes the quality of bones and teeth, also functions in the coagulation process [5,6]. The powdered plant material contained high levels of K (11990 ppm), Na (970 ppm) and Ca (10490 ppm), but were found in lower concentrations in the water extract. Trace elements such as Fe, Cu, Ni, Co,

Zn, Mg, Mn and I, even in minute quantities in medicinal plants can have both curative and toxic effects. Mn, Cu, Cr, and Zn are essential for the effective action of insulin.

Low levels of Zn have been detected in people suffering from diabetes and osteoporosis. The deficiency of this micronutrient is associated with impairment in growth leading to dwarfism [7]. Cu, which acts as a cofactor for a number of oxydase enzymes, plays a major role in Fe metabolism. Cu deficiency results in fragile bones and rupture of major vessels. Fe is an essential element in oxygen and electron transport [8]. Zn and Cu contents were high in the aqueous extract (2832 and 879 ppm), respectively, but Fe level was 74.3 and 44.2 ppm for powdered drug and water extract, respectively.

CONCLUSION

To the best of our knowledge, this is the first report of the levels of some mineral and trace elements of *O. sipyleum* and its water extract. It should be useful to also study the levels of these elements of other traditionally used species from the same genus, as it may provide some insight into the therapeutic actions of the plant in various diseases. The findings show that *O. sipyleum* is a potential source of macro- and micro-nutrients and may be therefore find application as an additive in food and health supplements.

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CONFLICT OF INTEREST

No conflict of interest associated with this work.

CONTRIBUTION OF AUTHORS

We declare that this work was done by the authors named in this article and all liabilities pertaining to claims relating to the content of this article will be borne by the authors.

REFERENCES

1. Fleisher A, Fleisher Z. Identification of Biblical Hyssop and origin of the traditional use of Oregano-group herbs in the Mediterranean region. *Econ Bot* 1988; 42(2): 232–241.
2. Arnold N, Bellomaria B, Valentini G, Arnold HJ. Comparative study of the essential oils from three species of *Origanum* growing wild in the eastern Mediterranean region. *J. Essent. Oil Res* 1993; 5(1): 71–77.
3. Başer KHC, Tümen G, Özek T, Kürkçüoğlu M, Composition of the essential oil of *Origanum sipyleum* of Turkish origin, *J Essent Oil Res* 1992; 4, 139–142;
4. Köksal O, Güneş E, Özer OO, Özden M, Analysis of effective factors on information sources at Turkish Oregano farms, *Afr J Agr Res* 2010; 5(2): 142–149.
5. Seiler HG, Sigel A, Sigel H, Eds. *Handbook on Metals in Clinical and Analytical Chemistry*. Marcel Dekker, New York: 1984; p 531.
6. Khan KY, Khan MA, Niamat R, Munir M, Fazal H, Mazari P, Seema N, Bashir T, Kanwal A, Ahmed SN. Element content analysis of plants of genus *Ficus* using atomic absorption spectrometer. *Afr J Pharm Pharmacol* 2011; 5(3): 317–321.
7. Salgueiro M, Zubillaga M, Lysionek A, Sarabia M, Caro R, De Paoli T, Hager A, Weill R, Boccio J. Zinc as an essential micronutrient: A. review. *Nutr Res* 2000; 20(5): 737–755.
8. Obiajunwa Eli Adeleke CA, Olanrewaju RO. Essential and trace element contents of some Nigerian medicinal plants. *Trad Nucl Chem* 2002; 252(3): 473–476.