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Secondary metabolites in *Hypericum* species and their distribution in different plant parts

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Abstract

The genus *Hypericum* (Hypericaceae) has attracted remarkable scientific interest as its members accumulate significant amounts of various bioactive compounds. In the current study, we investigated accumulation of several bioactive compounds in various parts of *Hypericum hircinum* L. subsp. *majus* (Ainton) N. Robson, *H. pallens* Banks et Sol., *H. russeggeri* (Fenzl) R. Keller and *H. lanuginosum* Lam. The plant materials were harvested at flowering, dissected into different tissues and subsequently subjected to high performance liquid chromatography (HPLC) analyses. Accumulation level of the investigated compounds varied greatly depending on species and plant parts. Among different plant tissues, flowers were found to be superior to leaves with respect to accumulation of the chemicals tested except for neochlorogenic, caffeic and 2,4-dihydroxybenzoic acids which were accumulated mainly in leaves. *H. hircinum* did not produce quercitrin or avicularin, *H. lanuginosum* did not accumulate hyperforins and caffeic acid, rutin was detected only in *H. pallens*.

For the first time, chemical profiles of these Turkish species of *Hypericum* were reported and the results are discussed from a phytochemical point of view. The present data could be helpful in selecting the future targets for phytochemical and biological studies on *Hypericum* genus as well as enriching our current knowledge about *Hypericum* chemistry.

Key words: chemical characterization, flavonols, *Hypericum hircitum*, *H. lanuginosum*, *H. pallens*, *H. russeggeri*, naphthodianthrones, phenolic acids, phloroglucinols.

Introduction

According to the most recent count by Crockett and Robson (2011), the genus *Hypericum* L. (Hypericaceae) includes 484 species that occur naturally on every continent in the world, except Antarctica. In particular, extracts of *Hypericum perforatum* L., the most abundant and well known species, are now widely used in Europe as a drug for the treatment of mild to moderate depression and all species of *Hypericum* have been used as sedatives, antiseptics and antispasmodics (Fiebich et al., 2011; Camas et al., 2014).

Hypericum hircinum L. subsp. *majus* (Ainton) N. Robson is a widespread shrub which grows naturally in damp, shady places besides streams at moderate altitudes of Southern Turkey. Results from recent studies reporting the monoamine oxidases inhibitory (Chimenti et al., 2006), antimicrobial (Nogueira et al., 2013), antioxidant and antiproliferative (Quassinti et al., 2013; Bertoli et al., 2015) activities of *H. hircinum* point out the great potential of this species as a promising medicinal plant.

Hypericum pallens Banks et Sol. and *Hypericum russeggeri* (Fenzl) R. Keller are rare herbaceous perennials, which grow in calcareous and limestone rocks of high altitudes in Southern Turkey. *Hypericum lanuginosum* Lam. is an endemic species from Turkish flora, which grows wild in some calcareous and scrub zones of Southern Turkey. In contrast to *H. hircinum*, neither ethnomedicinal nor pharmacological reports on the last three species are available in the current literature.

Phloroglucinol derivatives, naphthodianthrones, phenolic acids, flavonoids and essential oils are thought to be the main ingredients in *Hypericum* extracts, which possess a wide array of biological activities (Kasper et al., 2010). Among the chemicals, hypericins and hyperforins are considered to be synergistically responsible for the antidepressant activity of *Hypericum* extracts (Çirak, 2006; Du et al., 2006). Hyperforin also exhibits anti-inflammatory (Feisst, Werz, 2004), antitumoral (Schwarz et al., 2003) and antiangiogenic (Dona et al., 2004) effects.

Hypericins, the naturally occurring red pigments have been reported to exhibit significant bioactivities namely, antiviral, antiretroviral, photodynamic, antibacterial, antidepressant and antitumoral activities (Guedes, Eriksson, 2005). Although hyperforin and hypericins have been reported to mainly contribute to the pharmacological effects of *Hypericum* extracts, flavonoids, which are well known as antioxidants have also made an important contribution to the antidepressant activity (Gastpar, Zeller, 2005; Cirak, 2007; Cirak et al., 2010; 2013 a; b; Bertoli et al., 2011; Khan et al., 2011).

The aerial parts of *H. hircinum* subsp. *majus* were reported to contain chlorogenic acid, rutin, hyperoside, isoquercetin, quercitrin, quercetin, hyperforin and hypericin in a previous investigation (Sagratini et al., 2008). In the present paper we aimed to complete chemical characterization of this species with the first detection of several new compounds namely, pseudohypericin, adhyperforin, neochlorogenic acid, caffeic acid, 2,4-dihydroxybenzoic acid amentoflavone, avicularin, (+)-catechin and (−)-epicatechin.

Besides, to our knowledge, no study has been done on the chemistry of *H. pallens*, *H. russeggeri* and *H. lanuginosum*. So in this study, we also aimed to describe these species chemically by investigating the presence of naphthodianthrones hypericin and pseudohypericin, phloroglucinol derivatives hyperforin and adhyperforin, the phenolic acids chlorogenic acid, neochlorogenic acid, caffeic acid, 2,4-dihydroxybenzoic acid, and the flavonoids amentoflavone, hyperoside, isoquercetrin, quercitrin, quercetin, avicularin, rutin, (+)-catechin and (−)-epicatechin in the three species of *Hypericum* for the first time.

Materials and methods

Chemicals. Solvents used were of high performance liquid chromatography (HPLC) grade and purchased from Roth GmbH (Germany). Water was filtered through the grade water preparation cartridge Millipore HPLC (Millipore, USA). Reference substances were purchased from ChromaDex (USA), Sigma-Aldrich (USA) and HWI ANALYTIK GmbH (Germany).

Plant materials. The aerial parts of *Hypericum* plants, which represent a total of 30 individuals for each species were collected at full flowering from Southern Turkey in June, 2015. The species names, their voucher numbers and geographical data of collection sites are shown in Table 1.

Voucher specimens were deposited in the herbarium of Faculty of Agriculture, Ondokuz Mayis University, Turkey. The top of 2/3 plants was harvested. The plant materials were dried at room temperature (20 ± 2°C), and after separated into different tissues subsequently assayed for chemical contents by HPLC.

Preparation of plant extracts. Air-dried plant material was mechanically ground with a laboratory mill to obtain a homogenous drug powder. Samples of approximately 0.1 g (weighed with 0.0001 g precision) were extracted in 10 ml of 100% methanol by ultrasonication at 40ºC for 30 min in a Sonorex Super model RK 225H ultrasonic bath. The prepared extracts were filtered through a membrane filter with pore size of 0.22 µm (Carl Roth GmbH, Germany) and kept in a refrigerator until analysis. The extracts for naphthodianthrones analysis were exposed to light under xenon lamp (765 W m^{-2}) for 8 min due to the photoconversion of protohypericins into hypericins.

High performance liquid chromatography (HPLC) analysis and identification. A separation module system Waters Alliance 2695 (Waters Corporation, USA) equipped with Waters 2487 UV/Vis and Waters 996 PDA diode-array detectors were used for HPLC analysis. Data were analyzed using software chromatographic manager system Empower (Waters Corporation, USA).

Separation of flavonoids, epicatechin and hyperforin was carried out on SunFire C18 column (3.5 µm, 150 × 3.0 mm internal diameter) with 10 mm guard-precolumn. The binary gradient elution method was used for detection of corresponding compounds. The mobile phase consisted of water Milli-Q acidified with 0.3% phosphoric acid as eluent A and acetonitrile containing 0.3% phosphoric acid as eluent B. The elution profile was used as follows: 0–12 min 16% B, 12–18 min (B 16→53%), 18–18.1 min (B 53→97%), 18.1–29 min (B 97→97%) and 29–30 min (B 97→16%). Flow rate was 0.6 mL min^{-1} at a constant 25°C column temperature. The volume of extract injected was 10 µL. Peaks were detected at a wavelength range of 270–360 nm.

The ACE C18 column (5.0 µm, 250 × 4.6 mm internal diameter) (MAC-MOD Analytical Inc., USA) with guard-precolumn was used for separation of phenolic acids, catechin and hypericins. The mobile phase of gradient elution of phenolic acids and catechin was composed of eluent A – water acidified with 0.5% glacial acetic acid, and eluent B – acetonitrile. The separation was performed using the following program: 0–30 min (B 5→35%), 30–36 min (B 35→90%) and

Table 1. Collection sites and habitat of the *Hypericum* species examined

Species	Voucher numbers	Collection sites	Latitude N	Longitude E	Elevation m	Habitat
H. hircinum L.	OMUZF122	Samandağ-Batıayaz	36° 11′	36° 59′	450	shady areas near streams
H. lanuginosum Lam.	OMUZF123	Samandağ-Mağaracık	36° 06′	35° 56′	60	calcareous rocks, Macchie
H. pallens Banks et Sol.	OMUZF124	Samandağ-Mağaracık	36° 06′	35° 56′	60	calcareous rocks
H. russeggeri (Fenzl) R. Keller	OMUZF125	Samandağ-Çevlik	36° 07′	35° 55′	30	calcareous rocks

Note. Species are listed alphabetically.

36–37 min (B 90→5%). The flow rate was 1.0 mL min^{-1} at 25°C column temperature. Peaks were detected at a wavelength range 277–324 nm.

Hypericin and pseudohypericin were analyzed according to modified pharmacopoeial HPLC method (Anonymous, 2010). The mobile phase of isocratic elution of hypericin and pseudohypericin consisted of ethyl acetate, aqueous 0.1 M sodium dihydrogen phosphate solution, adjusted to pH 2.0 using phosphoric acid and methanol (16:17:67% v/v). The flow rate was 1.0 mL min^{-1}; 20 µL of extract was injected. Detection was performed at 560 nm wavelength at 40°C column temperature. Comparing retention times of samples with those of the reference standards identified chromatographic peaks. Furthermore, in order to confirm the identity of the eluted constituents, spectral characteristics of the eluting peaks were recorded with diode-array detector and compared with UV spectra of authentic standards.

Quantification of compounds was carried out by the external standard method. Standards stock solutions at a concentration of 1.0 mg mL^{-1} were prepared freshly in methanol and diluted in appropriate quantities to obtain a set of corresponding concentration ranges for the study of linearity. A calibration curve for each of the compounds was constructed by plotting peak areas versus the respective compound concentration and calculated by linear regression analysis. The regression coefficients ($r^2 \geq 0.999$) of all calibration curves indicated that, in the ranges of standard concentrations analyzed, the peak areas were directly proportional to the concentrations and, thus, methods presented adequate linearity. The precision of the method was demonstrated for all analyses, since all the obtained relative standard deviation (SD) values were lower than 5%. The concentration of compounds was expressed as mg g^{-1} dry mass (DM).

Results and discussion

Results from HPLC analyses of methanolic plant extracts revealed different chemical composition profiles and significant quantitative differences for *H. hircinum* subsp. *majus*, *H. pallens*, *H. russeggeri* and *H. lanuginosum*. Occurrence and accumulation level of the tested compounds varied greatly depending on species and plant parts. Among different plant tissues, generally lower levels of chemical accumulation were observed in stems and some compound, namely, hyperforin, adhyperforin, caffeic acid, 2,4-dihydroxybenzoic acid and avicularin were not detected in this tissue. Flowers were found to be superior to leaves with respect to accumulation of the chemicals tested except for neochlorogenic, caffeic and 2,4-dihydroxybenzoic acids which were accumulated mainly in leaves (Tables 2 and 3).

Regarding the quantitative amount of tested compounds, hypericin and pseudohypericin concentrations varied from trace amounts in flowers of *H. hircinum* subsp. *majus* to 1.05 and 1.74 mg g^{-1} DM in flowers of *H. pallens*, respectively, and those compounds were generally absent in leaves as well as stems. Hyperforin and adhyperforin were not detected in *H. lanuginosum* and similar to hypericins, these compounds reached their highest accumulation levels in flowers of *H. pallens* (3.02 and 1.25 mg g^{-1} DM, respectively). Chlorogenic and neochlorogenic acids contents ranged from 0.01 and 0.07 mg g^{-1} DM in stems of *H. russeggeri* to 76.36 and 5.48 mg g^{-1} DM in leaves of *H. pallens*, respectively.

2,4-dihydroxybenzoic acid accumulation was the highest in leaves of *H. hircinum* subsp. *majus* (0.54 mg g^{-1} DM) and was not observed in flowers of *H. pallens* whose leaves yielded this compound in small amount (0.05 mg g^{-1} DM). Caffeic acid accumulation was observed only in leaves of *H. hircinum* subsp. *majus* and *H. pallens* (0.01 and 0.05 mg g^{-1} DM, respectively). Leaves of *H. russeggeri* did not accumulate amentoflavone and the highest concentration level of this compound was observed in flowers of *H. hircinum* subsp. *majus* (2.79 mg g^{-1} DM) (Table 2).

Hyperoside, isoquercetin, quercetin, (+)-catechin and (−)-epicatechin were detected in all parts of *Hypericum* plants studied. Leaves of *H. lanuginosum* produced the highest level of hyperoside, isoquercetin, quercetin, (+)-catechin and (−)-epicatechin – 5.36, 2.01, 1.96, 1.27 and 3.90 mg g^{-1} DM, respectively. The lowest accumulation levels of hyperoside, isoquercetin, quercetin and (+)-catechin were observed in stems of *H. russeggeri* (0.13, 0.04, 0.01 and 0.01 mg g^{-1} DM, respectively) and in leaves of the same species for (−)-epicatechin (0.36 mg g^{-1} DM).

Table 2. Hypericin (1), pseudohypericin (2), hyperforin (3), adhyperforin (4), chlorogenic acid (5), neochlorogenic acid (6), caffeic acid (7), 2,4-dihydroxybenzoic acid (8) and amentoflavone (9) contents (mg g^{-1} DM) in different plant parts of some *Hypericum* species from Southern Turkey

Species	Plant parts	Compounds								
		1	2	3	4	5	6	7	8	9
H. hircinum L. subsp. *majus* (Ainton)	stem	–	–	–	–	0.65	0.14	–	–	–
	leaf	–	–	–	–	3.21	1.12	0.01	**0.54***	1.27
	flower	trace amount	trace amount	0.02	0.01	4.19	0.12	–	0.10	**2.79***
H. lanuginosum Lam.	stem	–	–	–	–	0.02	0.21	–	–	–
	leaf	–	–	–	–	0.82	**2.03***	–	0.22	0.05
	flower	0.01	0.04	–	–	1.26	1.82	–	0.13	0.42
H. pallens Banks et Sol.	stem	0.01	0.02	–	–	5.59	0.31	–	–	0.01
	leaf	**0.62***	**0.69***	**1.25***	0.12	**76.36***	**5.48***	**0.05***	0.05	0.04
	flower	**1.05***	**1.74***	**3.02***	1.25	**11.56***	1.31	–	–	0.86
H. russeggeri (Fenzl)	stem	–	–	–	–	0.01	0.07	–	–	–
	leaf	–	–	–	–	0.05	**2.91***	–	0.24	–
	flower	0.01	0.01	0.02	0.02	0.29	1.35	–	0.16	0.25

Note. Species are listed alphabetically; * – $P < 0.05$ was considered statistically significant.

Table 3. Hyperoside (10), isoquercetin (11), quercitrin (12), quercetin (13), avicularin (14), rutin (15), (+)-catechin (16) and (−)-epicatechin (17) contents (mg g^{-1} DM) in different plant parts of some *Hypericum* species from Southern Turkey

Species	Plant parts	Compounds							
		10	11	12	13	14	15	16	17
H. hircinum L. subsp. *majus* (Ainton)	stem	0.47	0.24	–	0.14	–	–	0.83	0.43
	leaf	0.78	0.27	–	0.27	–	–	0.97	1.27
	flower	1.14	1.42	–	**1.36***	–	–	**1.34***	**1.99***
H. lanuginosum Lam.	stem	0.50	0.17	0.77	0.15	–	–	0.01	1.25
	leaf	1.61	0.48	**2.27***	0.12	–	–	0.19	1.13
	flower	**5.36***	**2.01***	**2.96***	**1.96***	0.09	–	**1.27***	**3.90***
H. pallens Banks et Sol.	stem	0.36	0.62	0.85	0.04	–	0.93	0.66	0.88
	leaf	1.02	1.42	1.15	0.15	–	**4.45***	0.02	1.17
	flower	1.97	**1.78***	**3.32***	0.85	**0.38***	1.17	**1.23***	**2.18***
H. russeggeri (Fenzl)	stem	0.13	0.04	0.37	0.01	–	–	0.01	0.48
	leaf	0.44	0.12	1.49	0.06	–	–	0.02	0.36
	flower	1.99	0.84	**2.31***	0.24	0.06	–	0.04	**1.74***

Note. Species are listed alphabetically; * – $P < 0.05$ was considered statistically significant.

Rutin was detected only in *H. pallens* whose leaves accumulated the highest amount of this compound (4.45 mg g^{-1} DM). *H. hircinum* subsp. *majus* did not produce avicularin and this compound was detectable only in flowers of the other investigated species in low amounts. *H. hircinum* subsp. *majus* also did not produce quercitrin whose yield ranged from 0.37 mg g^{-1} DM in stems of *H. russeggeri* to 3.32 mg g^{-1} DM in flowers of *H. pallens* (Table 3).

Morphologically, three kinds of secretory structures, including light glands, dark glands and secretory canals are facilitated to characterize *Hypericum* plants (Ciccarelli et al., 2001). These structures are accumulation and/or synthesis sites for different kinds of phytochemicals. For example, hypericins are thought to be present only in the species of *Hypericum* whose aerial parts bear dark glands (Lu et al., 2001) and a positive correlation was reported in dark gland density and hypericin content of leaf in *H. perforatum*, *H. pruinatum* Boiss. & Balansa and *H. avicullariifolium* subsp. *depilatum* var. *depilatum* (Freyn and Bornm.) Robson var. *depilatum* (Cirak et al., 2006). The localization of the secretory structures varies greatly among plant tissues, and for that reason, the levels of phytochemicals in a particular *Hypericum* tissue depend on the relative abundance of these secretory structures on the harvested material (Zobayed et al., 2006). As a result, organ-dependence of a given chemical is common among *Hypericum* species and this phenomenon could explain the great variation observed in the chemical contents among the species investigated as well as their different tissues.

All values are presented as the mean ± standard error (SE) of the mean of n observations, being n the number of studied plants. The results in each group were compared by one-way analysis of variance (*ANOVA*) for multiple comparisons. $P < 0.05$ was considered statistically significant. Software *MATLAB* (MatLab® 8.1.0.604 R2013a) was used for statistical analysis (Table 4).

Accumulation patterns of the chemicals in leaves and flowers of the species in the present paper corresponded in the same way to each other and matched largely those described for other *Hypericum* species. In *H. perforatum*, the well known and most studied species of Hypericaceae, flowers accumulated larger amounts of hypericin, hyperforin, rutin, quercetin and quercitrin and leaves had the highest level of hyperoside (Bagdonaite et al., 2010). Similarly, *H. origanifolium* Willd. and *H. perfoliatum* L. accumulated quercitrin, rutin as well as hypericin, pseudohypericin and hyperforin mainly in their floral buds and flowers, while their leaves produced higher amounts of quercetin, chlorogenic acid and hyperoside (Cirak et al., 2007). Flowers of *H. montbretii* Spach accumulated the highest level of hyperforin, hypericin and apigenin-7-O-glucoside and

Table 4. Descriptive statistics for compound in different plant parts of some *Hypericum* species

	Minimum	Maximum	Mean		SD
	statistic	statistic	statistic	SE	statistic
Hypericin	0.00	1.05	0.14	0.097	0.34
Pseudohypericin	0.00	1.74	0.21	0.15	0.52
Hyperforin	0.00	3.02	0.36	0.26	0.91
Adhyperforin	0.00	1.25	0.12	0.10	0.36
Chlorogenic acid	0.01	76.36	8.67	6.23	21.58
Neochlorogenic acid	0.07	5.48	1.41	0.45	1.57
Caffeic acid	0.00	0.05	0.01	0.001	0.01
2,4-dihydroxybenzoic acid	0.00	0.54	0.12	0.05	0.16
Amentoflavone	0.00	2.79	0.47	0.24	0.84
Hyperoside	0.13	5.36	1.31	0.41	1.42
Isoquercetin	0.04	2.01	0.78	0.20	0.70
Quercitrin	0.00	3.32	1.29	0.34	1.18
Quercetin	0.01	1.96	0.45	0.18	0.62
Avicularin	0.00	0.38	0.04	0.03	0.11
Rutin	0.00	4.45	0.55	0.37	1.30
(+)-catechin	0.01	1.34	0.55	0.16	0.56
(−)-epicatechin	0.36	3.90	1.40	0.28	0.98

SD – standard deviation, SE – standard error

the highest accumulation level of chlorogenic acid was found in leaves (Cirak, Radusiene, 2007). Hypericin and pseudohypericin were detected only in flowers and leaves had the highest accumulation level of chlorogenic acid in *H. scabrum* L. and *H. bupleuroides* Gris. (Cirak et al., 2016). In wild and greenhouse-grown *H. triquetrifolium* Turra plants, flowers accumulated the highest level of hyperoside, quercetin and quercitrin; however, leaves produced higher amount of chlorogenic acid and isoquercetin (Cirak et al., 2013 a). Floral parts had the highest level of hypericin, pseudohypericin, quercitrine and quercetine and leaves accumulated the highest level of chlorogenic acid as well as rutin and hyperoside in *H. aviculariifolium* subsp. *depilatum* var. *depilatum* and *H. orientale* L. (Cirak et al., 2013 b).

Studies on identifying chemical profile may be useful for taxonomic analysis of the genus *Hypericum* (Crockett, Robson, 2011). Because only few morphological characters are available to distinguish between some sections, identifying the individual plants solely based on morphological characters is very difficult. In this regard, chemotaxonomic significance is attributed to some of the *Hypericum* chemicals such as naphthodianthrones hypericins (Kitanov, 2001), dimeric phloroglucinol uliginosin B (Ferraz et al., 2002), flavonoids hyperoside, quercetin, quercitrin (Cirak et al., 2010), rutin and mangiferin (Nunes et al., 2010). Thus, results of the present chemical investigation could be used as an additional tool for taxonomic identification of species in the genus although *H. hircinum*, *H. lanuginosum*, *H. pallens* and *H. russeggeri* fall into different sections within *Hypericum* genus, namely *Androsaemum* (Duhamel) Gordon, *Adenosepalum* Spach, *Triadenioides* Jaub. & Spach. and *Triadenia* Spach. (Davis, 1988).

Conclusion

Increasing interest in recent years for using medicinal and aromatic plants in pharmaceutical, food, biotechnology and cosmetic industries all over the world has resulted in extensive efforts to discover new sources of potential bioactive phytochemicals. In this sense, the present screening data could be helpful in selecting the future targets of new sources of bioactive compounds of *Hypericum* species for phytochemical and biological studies as well as enriching our current knowledge about *Hypericum* genus chemistry. Besides, this is the first report describing the chemical profile of *H. pallens*, *H. russeggeri* and *H. lanuginosum* as well as the occurrence of several new compounds in *H. hircinum* subsp. *majus*. The present data are of great interest to reveal new sources of raw material as potential pharmaceuticals. The chemical evaluation of *Hypericum* species could be used as an additional tool in completing the taxonomy of genus and for understanding the evolution of its diversity.

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Jonažolių rūšių antriniai metabolitai ir jų pasiskirstymas augalų dalyse

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Santrauka

Hypericum genties (Hypericaceae) augalai sulaukia vis didesnio susidomėjimo dėl jų kaupiamų įvairių biologiškai aktyvių junginių. Tyrimo metu buvo nustatyta kai kurių antrinių metabolitų sudėtis *Hypericum hircinum* L. subsp. *majus* (Ainton) N. Robson, *H. pallens* Banks et Sol., *H. russegger* (Fenzl) R. Keller ir *H. lanuginosum* Lam. augaluose jų žydėjimo metu. Surinkta žaliava buvo suskirstyta pagal augalo dalis ir analizuota efektyviosios skysčių chromatografijos metodu. Nustatyti junginių kiekiai įvairavo priklausomai nuo augalų rūšių ir jų dalių. Žiedai sukaupė didesnius kiekius junginių, išskyrus neochlorogeninę, kavos ir 2,4-dihidroksibenzojinę rūgštis, kurios dominavo lapuose. *H. hircinum* nekaupė kvercitrino ir avikularino, *H. lanuginosum* – hyperforino ir kavos rūgšties, o rutinas buvo aptiktas tik *H. pallens* augaluose.
Pirmą kartą buvo nustatyta ir įvertinta Turkijoje augančių jonažolių bioaktyvių junginių sudėtis. Gauti tyrimų duomenys galės būti panaudoti biologiniams ir cheminiams jonažolių genties augalų tyrimams ir papildys žinias apie šios genties augalų cheminę sudėtį.

Reikšminiai žodžiai: cheminė sudėtis, fenolinės rūgštys, flavonolai, floroglucinolai, *Hypericum hircitum*, *H. lanuginosum*, *H. pallens*, *H. russeggeri*, naftodiantronai.

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