



Accepted Article

Title: Review

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for pharmaceutical and food industry

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To be cited as: *Chem. Biodiversity* 10.1002/cbdv.202000097

Link to VoR: <https://doi.org/10.1002/cbdv.202000097>

Balkans' Asteraceae species as a source of biologically active compounds for pharmaceutical and food industry

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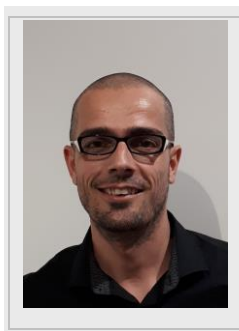
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Herbal drugs are a useful source of different bioactive compounds. Asteraceae species, as the most widespread vascular plants can be used both as food and as medicine due to the great diversity of recorded chemical components – different phenolic compounds, terpenes, carotenoids, vitamins, alkaloids, etc. The Balkan Peninsula is characterized by great diversity of plants from Asteraceae family, including presence of rare and endemic species. In this review, results of the survey of chemical composition and biological activity, mainly focusing on antioxidant, antimicrobial and anticancer effects of selected Balkans' Asteraceae species were provided. In addition, information on edible plants from Asteraceae family is presented, due to growing interest for the so-called "healthy diet" and possible application of Balkans' Asteraceae species as food of high nutritional value or as a source of functional food ingredients.

Keywords: Asteraceae • Bioactive compounds • Balkans • Food industry • Pharmaceutics

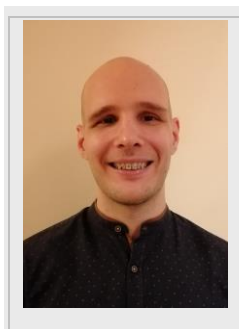
Dr Aleksandar Ž. Kostić is Assistant professor at the Chair of Chemistry and Biochemistry, Faculty of Agriculture, University of Belgrade, teaching in different Chemistry courses. His research area is focused on phytochemistry (with special emphasis on pollen chemistry and nutrition and bioactive compounds in plant materials), food chemistry, water chemistry, health risk assessment for humans expressed through food and water consumption and polyacrylic hydrogels. Until now, he has been the author and co-author of more than a hundred scientific papers in total (thirty-four peer-reviewed articles), two university textbooks and two student handbooks. His articles have been cited more than 200 times, h-index is 10 (Google Scholar). He has reviewed more than 100 articles for top International Journals.



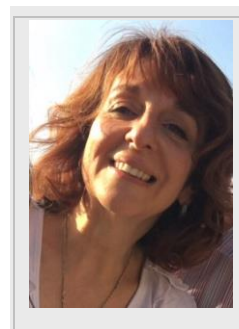
Dr Peđa Janačković is Full professor at Chair of Plant Morphology and Systematics, Faculty of Biology, University of Belgrade, teaching in different Botany courses at bachelor, master and doctoral studies (e.g. Systematics of higher plants, Phytochemistry practicum, History of Botany, Ethnobotany, Systematics and phylogeny of the Asteraceae). He was the supervisor of 3 completed PhD theses, and currently he is the supervisor of 2 PhD students. His research area is focused on plant systematics especially of Asteraceae as well as phytochemistry and ethnobotany (with special emphasis on medicinal plants and bioactive compounds in plant). Until now, he has been the author and co-author of more than one hundred and thirty scientific papers in total (fifty four peer-reviewed articles), one chapter in international monograph and five university textbooks. His articles have been cited more than 444 times (excluding self citations), h-index is 13 (Scopus). He has reviewed more than 30 articles for top International Scientific Journals. Prof. Janačković is a board member of two international journals.



M.S. Stefan M. Kolašinac, is a forth year Ph.D. student at the Faculty of Agriculture, University of Belgrade Chair of Agrobotany. During the bachelor studies, he was awarded as one of the best students. Until now, he has published 7 scientific papers. Currently, he is included in one national project and one bilateral cooperation. Stefan is a demonstrator on courses Agriculture Botany and Systematics of flowering plants. His main interests are based on plant chemistry and chemometrics.



Dr Zora P. Dajić Stevanović is the Head of Department of Agricultural Botany at Faculty of Agriculture, University of Belgrade. She is Full professor teaching several modules in the field of Plant sciences at bachelor, master and doctoral studies; she was a supervisor of 8 completed PhD theses, currently supervisor of 3 PhD students; team leader of research group interested in biodiversity, ethnobotany, morpho-anatomy, phytochemistry, bioactivity of plant metabolites and Raman spectroscopy. She published more than 300 papers, nearly 70 in international peer reviewed journals and 11 chapters in international monographs. Her papers were cited 875 times, h-index 15 (ResearchGate). Prof. Dajić Stevanovic is a reviewer of papers in international journals and a board member in 4 international journals. She is the president of Association for Medicinal and Aromatic Plants of Southeast European Countries (AMAPSEEC) and a member of working groups for medicinal and aromatic plants in international organizations (FAO, ECPGR, IUCN).

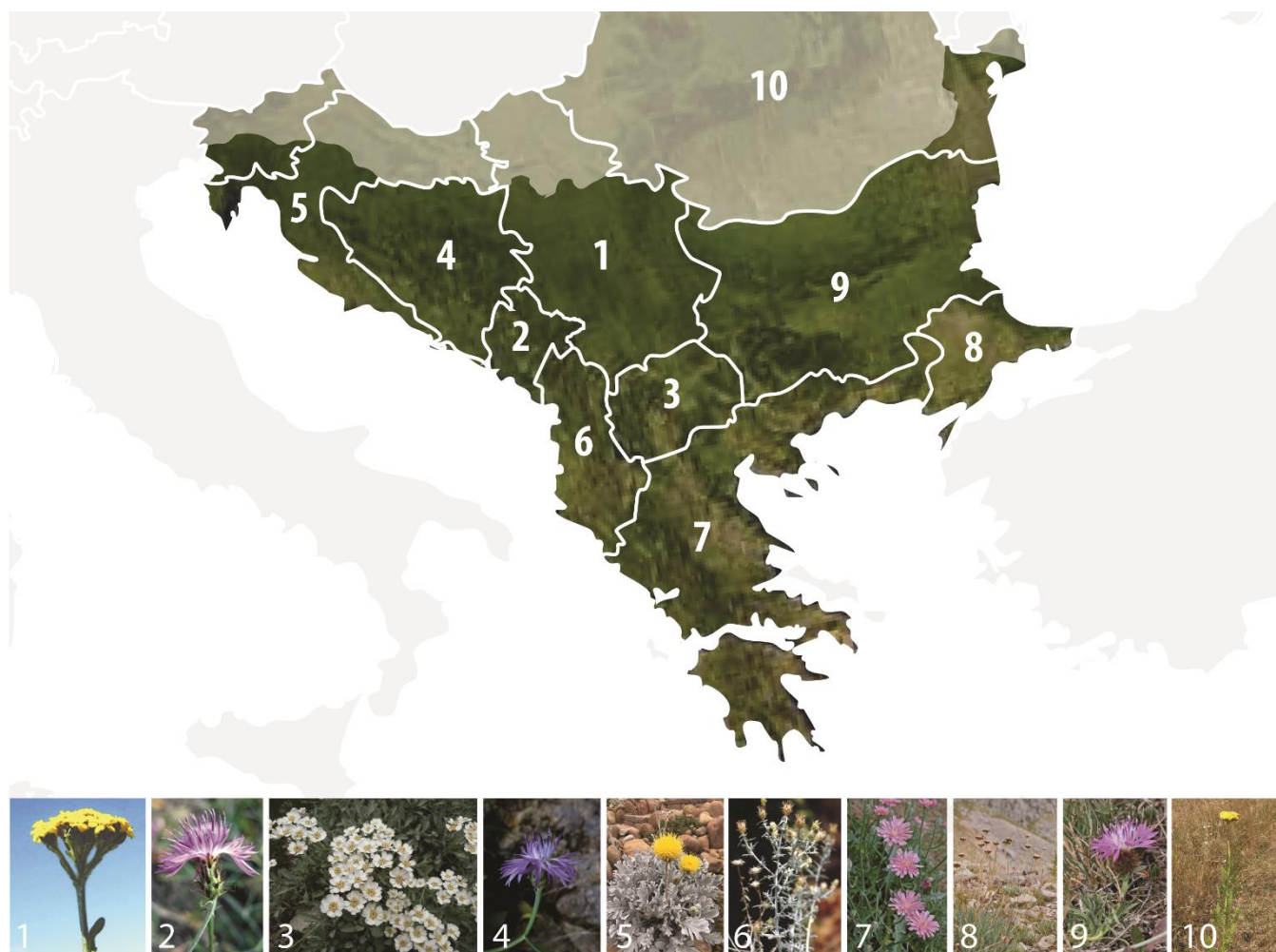


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17 **1. Introduction**

18 Plants have been essential resources for human health from ancient times to the present day. According to the World Health Organization (WHO), the
19 majority of the world's population still depends on traditional medicine for primary health care needs. More than 35 000 plant species are used in herbal
20 medicine and as spices, most of which are of local importance due to traditional use. The flora of the Balkan Peninsula is among the most diverse in Europe,
21 comprising more than 8 000 species of vascular plants, among which 2 600-2 700 are known as endemic species.^[1] The Balkans and Rhodope Mountains are
22 recognized as global Centers of Plant Diversity. The region is also well known for richness in indigenous medicinal and aromatic plant (MAPs) resources
23 and long tradition in use of MAPs and their products. The total number of MAP species in Southeast Europe and the Balkans is very difficult to assess, but it
24 certainly exceeds 1 000 species or even 2 000 upon approximation that one in six species of higher plants is used medicinally.^[2] Asteraceae family is the
25 most abundant in the number of genera and species in the Balkans' flora (**Figure 1**), as it is the largest family of vascular plants. Asteraceae (Compositae),
26 daisy or sunflower family, represents the mega-diverse family containing more than 10% (25 000–35 000) of all today's living angiosperms.^[3] Members of
27 the Asteraceae are distributed on all continents including Antarctica -two introduced species *Nassauvia magellanica* J. F. Gmel. and *Gamochoeta nivalis*
28 Cabrera, according to Smith and Richardson^[4], and occur on almost every type of habitat on planet Earth, having the greatest concentration of species in
29 steppes, prairies, deserts, montanes, and in regions with Mediterranean-like climates.^[3] Newly discovered fossils and the latest molecular clock dating,
30 showed that Asteraceae possible originates in the Late Cretaceous: ~83 MYA.^[3] The family consists of 13 subfamilies and 47 tribes according to Mandel et
31 al.^[3] Asteraceae as one of the largest, natural families (with a combination of specialized morphological structures e.g., capitula, inferior ovaries and unique
32 fruit – cypsela, and also, with a wide diversity of specialized metabolites) has been researched from many different aspects for centuries. Because of its
33 diversity Asteraceae represents immense plant resources. Asteraceae species have wide economic importance, e.g. as medicinal plants, sources of oil,
34 vegetables, insecticides, and as horticultural and garden ornamentals.^[5] Nevertheless, some species of Asteraceae, as noxious weeds, represent a major
35 problem for agriculture.^[6] The Balkan Peninsula extends from Central Europe in the north to the Eastern Mediterranean in the south, and is bounded by the
36 Adriatic, Ionian, Aegean and Black Seas.^[7] The Balkans or the Balkan Peninsula is in the north limited by the rivers Danube and Sava, in the northwest by
37 the Soča (in Slovenian, or Isonzo in Italian) river, in the east by the Black Sea, in the south by the Aegean and Ionian seas, and in the west by the Adriatic
38 Sea.^[8] Although, till today, a complete list of the flora of the Balkan Peninsula has not yet been completed, it can be estimated that the Asteraceae family on
39 the Balkans counts 913-1 081 species (13.52 % of the total) classified in 547 genera^[9–13] and represents one of the most important plant resource of the
40 Balkan Peninsula. The exact number of the Balkans' endemic Asteraceae species is unknown, but could be approximately at least between 350-400 species.

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Figure 1. The Balkan Peninsula with usual appearance of some of the Asteraceae species: 1. *Achillea alexandri-regis* Borrm. & Rudsky, 2. *Centaurea scabiosa* L., 3. *Achillea clavennae* L., 4. *Centaurea derventana* Vis. & Pančić, 5. *Centaurea ragusina* L., 6. *Centaurea soskai* Hayek ex Košanin, 7. *Crepis incana* Sm., 8. *Achillea fraasii* Sch.Bip., 9. *Centaurea parilica* Stoj. & Stef., 10. *Achillea thracica* Velen.

Due to great diversity, these species can be used as an excellent source of different bioactive compounds which can improve functional food properties or as a source in drug development for pharmaceutical industry. In that sense, in this review authors tried to make the first detailed cross-section of the current status of phytochemical analyses of different materials that originated from Asteraceae plants grown specifically on the Balkan Peninsula, and their possible biological activities. Therefore, the Balkans' endemic Asteraceae species were especially emphasized in the review. Additionally, importance and potential application of wild growing Asteraceae plants as an edible food source are reviewed upon available ethnobotanical sources for the Balkan region, in order to indicate to the readers the possibility of using them as healthy food for the future. Consequently, since the number of endemics used as wild edible foods is limited, assessment of edible Balkans' Asteraceae plants included all reported species within ethnobotanical data.

2. Chemical composition and biological activity of Balkans' Asteraceae species

2.1. Chemical composition

Plants are a well-known source of different phytochemicals which are recognized as potential antioxidants for animals and humans.^[14,15] These compounds are mostly specialized (secondary) plant metabolites which are produced from compounds included in central (primary) metabolic processes such as photosynthesis, plant respiration, etc.^[16] There is a great diversity among them but the most important groups of compounds are different phenolics and terpenes, carotenoids, fatty acids, saponins and alkaloids^[16] as well as some bioactive vitamins such as vitamins A, C and E.^[15] It is a challenging approach to match different phytochemical profiles of different taxa with their phylogenetic, morphological and ecological properties. Such was the case, for instance with *Iris* species of the central Balkans where detailed polyphenols screening was done, and where 9 novel compounds were assessed in such a way.^[17] Similarly, phytochemical investigation of endemic species of the western part of the Balkans of genus *Amphoricarpus*, showed presence of a large number of sesquiterpene lactones so called amphoricarpolids (sesquiterpenoids that contain a γ - lactone ring), which are characteristic of this genus, and which are showing: lipophilicity, cytotoxicity^[18] and antimicrobial^[19] activity as well as protective effect on human lymphocyte DNA,^[20] and which are also attributed to a role in enhancing antioxidant activity^[19]. Among all known bioactive compounds, the terpenes are commonly basic components of plant essential oils (EOs) which have regularly been analyzed in plants due to their great potential for application in pharmacy, food industry, aromatherapy and well known in

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65 traditional medicine. Within phenolic compounds, the class of flavonoids is the most important in terms of diversity, stability and potential as chemophenetic
66 markers.^[21] According to Bohm and Stuessy^[21] almost all flavonoid subclasses that are known in the plant kingdom can be found in many Asteraceae genera
67 or species. The most represented flavonoid compounds in the family Asteraceae are anthocyanin pigments, chalcones, aurones, flavones, flavanones and
68 flavonols.^[21] Among “famous” Asteraceae species, sunflower (*Helianthus annuus* L.) pollen from Serbia has already been defined as a better source of
69 flavonoids^[22] compared to some usual vegetables. It makes it suitable for application as a potential food ingredient. Summarized available data regarding the
70 chemical composition of indigenous Balkans’ Asteraceae plants are shown in **Table 1**. Here are provided results only for the Balkan endemic species of
71 Asteraceae, illustrating high variability in phytochemical profiles. The main compounds were indicated according to information provided in cited reference,
72 if any.

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Table 1. Chemical composition (with the main compound(s)) of mostly endemic Balkans' Asteraceae plants.

Scientific species name	Distribution	Part of plants	Analyzed fraction and / or general chemical composition and reference source	Type of extract (s)	Applied technique(s)	The main compound(s)	Remarks
<i>Achillea abrotanoides</i> (Vis.) Vis. ^[6]	Alb, BiH, Cro, Grc, Mkd, Mne, Srb ^[6]	Aerial parts	EOs ^[24] ; sesquiterpene lactones ^[23]	Chloroform	Silica gel column chromatography; ¹ H NMR spectroscopy	desacetoxymatricarin (202 mg)	-3 new lactones (desacetoxymatricarin, desacetylmatricarin and 1 β , 10 β -epoxydesacetoxymatricarin) and one flavonoid (centaureidin) were isolated.
			- EOs; terpenes ^[24]	Hydrodistillation	GC-MS	α -tujone (34.5%) and 1,8-cineol (8.5 - 10.9%)	- 97 terpene and non-terpene compounds were isolated. 50 compounds were reported for the first time in <i>Achillea</i> genus.
			- EOs; terpenes ^[25]	Hydrodistillation	GC; GC-MS	β -thujone (16.8%), α -thujone (15.6%) and 1,8-cineole (11.3%)	-In total, 36 components (monoterpenes and sesquiterpenes) were identified and quantified.
<i>A. ageratifolia</i> subsp. <i>serbica</i> (Nyman) Heimerl ^[6]	Alb, BiH, Srb	Aerial parts	- Sesquiterpene lactones ^[26]	Petrol-ether/Ether/ MeOH (1:1:1)	Column chromatography	No data	- 6 guaianolides were obtained from the extract with 4 new identified compounds.
			- EOs ^[27]	Hydrodistillation	GC-FID; GC-MS; ¹ H NMR spectroscopy	Sabinyl-acetate (30.8 - 37.5%) and <i>trans</i> -sabinol (5.0 - 6.3%)	- 300 components were identified. Sabinyl derivatives were predominant in EOs.
<i>A. alexandri-regis</i> Bornm. & Rudsky ^[6]	Srb	Aerial parts	- Flavonoids, phenolic acids, triterpenoids ^[28,29]	75% MeOH ^[6]	HPLC	Aigenin-7- <i>O</i> -glycoside (EtOAc ^[6] fraction) and caffeic acid (BuOH ^[6] fraction)	- Polyphenols were the main components.
			- Triterpenic derivatives and phenolics ^[30]	Combined chloroform/EtOAc i.e. MeOH/50% MeOH	Silica gel column chromatography; ¹ H NMR spectroscopy and	No data	- In total 15 triterpenic (chloroform/EtOAc) and 4 phenolic compounds (MeOH)

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			- EOs ^[31]	Hydrodistillation	¹³ C NMR spectroscopy; GC-MS GC; GC-MS	α -pinene (14.4 - 15.7%), isopinocampone (5.4 - 23.5%), α -phellandrene epoxide (5.0 - 19.0%) , borneol (3.6 - 7.7%) and spathulenol (1.4 - 9.7%)	were identified. - In total 24 volatile compounds were identified.
<i>A. holosericea</i> Sm. ^[6]	Alb, Grc, Mkd, Srb	Aerial parts	- EOs ^[32-34]	Hydrodistillation	GC-MS	Camphor (14.8% i.e. 20.9%), borneol (16.3 i.e. 30.2%), <i>cis</i> / <i>trans</i> farnesyl acetate (9.6%) ^[32,33] and 1,8- cineole (47.4%) ^[34]	- Ninety five, ^[32] twelve ^[33] i.e. sixteen ^[34] compounds were identified belonging to monoterpenes or sesquiterpenoids classes.
<i>A. baldaccii</i> Degen ^[6]	Alb, Srb	Leaves	- Flavonoids ^[35]	Acetone	TLC	No data	- 5 derivatives (3 in small amounts and 2 as traces) of scutellarein, kaempferol and quercetagenin were isolated and identified.
<i>A. chrysocoma</i> Friv. ^[6]	Alb, Bul, Grc, Mkd, Srb	Aerial parts	- EOs ^[36,37]	Hydrodistillation	HPLC/GC/GC-MS	1,8-cineole (17%) and β - caryophyllene (6.3%) ^[36] i.e. borneol (10.1%), terpinen-4- ol (9.2%) and <i>cis</i> / <i>trans</i> menth-2-en-1-ol (7.9 - 8.2%) ^[37]	- 48 ^[36] i.e. 28 compounds ^[37] were identified.
<i>A. chrysocoma</i> Friv. ^[6]	Alb, Bul, Grc, Mkd, Srb	Flowers and leaves	- Sesquiterpene lactones ^[38]	Chloroform	Silica gel column chromatography	Kapicidin A (10 mg)	- In total 22 lactones were identified in the chloroform extract. Lack of germacranolides as well as guaianolide derivatives of matricarin and achillin was observed.
<i>A. clavennae</i> L.	Mkd, Srb	Aerial parts	- Alkanes, fatty acids, monoterpenes, guaiane sesquiterpenes and flavonoids ^[39] - EOs ^[34]	Ether/Hexane/MeOH (1:1:1) Hydrodistillation	Silica gel column chromatography, GC-MS, NMR spectroscopy GC-MS	No data Camphor (46.9%) and 1,8- cineole (43.9%)	- 1 flavonoid (centaureidin) and 3 terpenes (1,8-cineole, camphor and caryophyllene oxide) were predominant. 12 compounds were identified.

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<i>A. fraasii</i> Sch.Bip. ^[g]	Alb, Grc, Mne, Mkd, Srb, Tur	Aerial parts	- EOs ^[32]	Hydrodistillation	GC-MS	Camphor (16.3%) and 1,8-cineole (11.9%)	- 14 compounds were identified.
<i>A. taygetea</i> Boiss. & Heldr. ^[g]	Grc	Aerial parts	- EOs ^[32]	Hydrodistillation	GC-MS	1,8-cineole (26.6%) and camphor (25.7%)	- 13 compounds were identified.
<i>A. thracica</i> Velen. ^[g]	Bul, Rou	Fresh, <i>in vitro</i> and <i>ex-vitro</i> grown ground parts	- EOs ^[40]	Hydrodistillation	GC-MS	1,8-cineole (35.7%) and germacrene D (33.8%) - <i>in vitro</i> Santolina alcohol (41.2%) and β -eudesmol (12.4%) - <i>in situ</i> Santonia alcohol (26.4%) and β -eudesmol (11.5%) - <i>ex vitro</i>	- Fresh and <i>ex-vitro</i> grown plants EOs differ from EO of <i>in vitro</i> grown plant.
			- Monoterpenes, irregular terpenes, esters and flavonoids ^[41]	Acetone	GC-FID; GC-MS; HPLC	Volatile compounds: Santolina alcohol (31.1%) - natural plant Yonaghi alcohol (21.66%) - <i>in vitro</i> plant Santolina alcohol (28.1%) - <i>in vitro</i> plant Flavonoids: <i>O,O</i> -dimethyl quercetin (6.7%) - natural plant <i>O,O</i> -dimethyl quercetin (2.1%) and <i>O</i> -methyl quercetin (28.4%) - <i>in vitro</i> plant <i>O,O</i> -dimethyl quercetin (38.7%) and <i>O</i> -methyl quercetin (38.1%) - <i>ex vitro</i> plant	- Among volatile compounds 16 irregular monoterpenes were identified and quantified. In acetonic extracts 4 different flavonoids were identified and quantified.
<i>A. depressa</i> Janka	Bul, Grc, Mkd, Rou, Srb	Roots	- Sesquiterpene-coumarin ethers ^[42]	Petrol ether/ether (2:1)	TLC; ¹ H NMR spectroscopy	Acetyldeparnol (7 mg) and farnochrol (6.5 mg)	- 11 compounds were identified with 3 new sesquiterpene-isofraxidin ethers: albartol (bicyclic derivate), deparnol and acetyldeparnol (monocyclic

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		Leaves	- Monoterpenes, sesquiterpene lactones and flavonoids ^[43]	Chloroform	Silica gel column chromatography; ¹ H NMR spectroscopy	Camphor (150 mg)	derivatives). - 5 monoterpenes were isolated (camphor, borneol, bornyl acetate, sabinol and sabinyl acetate), as well as 3 flavonoids (salvigenin, tambulin and 6-methoxy-genkwainin) and 2 well known guaianolides: achillin and β -1,10-epoxyachillin.
		Aerial Parts	- Sesquiterpene and flavonoids ^[44,45]	Chloroform ^[44] ; Petrol ether/ether/MeOH (1:1:1) ^[44]	Silica gel column chromatography; TLC; ¹ H NMR spectroscopy ^[44]	No data ^[44,45]	- 2 monoterpenes (tanaparthin- α -peroxide and 8 α -hydroxy-tanaparthin- α -peroxide), sesquiterpenes (reynosin, ridentin, acrifolide and spirodepressolide ^[44]) 3 flavonoids (cirsimaritin, salvigenin penduletin) and 2 unidentified lactones were isolated from aerial parts of this plant ^[45] .
			- Terpenoids ^[46]	Chloroform	Column chromatography; ¹ H NMR spectroscopy	No data	Five acyclic terpenoids were isolated (as minor components) and identified. Three of them were new natural compounds.
<i>A. umbellata</i> Sibth. & Sm. ^[g]	Grc	Aerial parts	- EOs ^[47]	Hydrodistillation	GC; GC-MS	β -hujone (62.8%) and camphor (8.7%)	- In total, 66 components were isolated and identified from EO. Predominant class of compounds was oxygen-containing monoterpenes.
			- EOs ^[48]	Hydrodistillation	GC-MS	Fragranol and fragranyl acetate (in total make 74.6% of EO)	- 51 compounds were quantified from plant EO. In addition, 14 fragranol esters were identified with 6 compounds identified for the first time from a plant.
<i>Amphoricarpus autariatus</i> subsp.	Alb, Grc, Mne, Srb	Leaves	- Polyphenols and	MeOH	UV-Vis spectroscopy	No data	- Dry MeOH ^[e] extract

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bertisceus Blečić & E.Mayer^[g]

			flavonoids ^[19]					contained the highest quantity of total phenolics and flavonoids.
<i>A. autariatus</i> Blečić & Mayer subsp. <i>autariatus</i> ^[g]	BiH, Mne, Srb	Leaves	- n-alkanes, sesquiterpene lactones ^[49]	Dichloromethane	GC; GC-MS; LC-ESI MS ^[h] ; ¹ H NMR spectroscopy	C ₂₉ alkanes (29.7%) in the non-polar fraction, amphoricarpolide 13 (48.7%) in the more polar fraction		- The non-polar fraction contained C ₂₇ , C ₂₉ and C ₃₁ alkanes as predominant, while 13 sesquiterpene lactones were identified in the more polar fraction.
			- Polyphenols and flavonoids ^[19]	MeOH	UV-Vis spectroscopy	No data		- Among the examined <i>Amphoricarpos</i> species, extracts of this plant had the highest total phenolic and flavonoid content.
<i>A. neumayerianus</i> (Vis.) Greuter ^[g]	Cro, BiH, Mne	Leaves	- Polyphenols and flavonoids ^[19]	MeOH	UV-Vis spectroscopy	No data		- Dry MeOH extract contained the highest quantity of total phenolics and flavonoids.
<i>Anthemis auriculata</i> Boiss. ^[g]	Bul, Grc, Mkd, Tur	No data	- EO ^[50]	Hydrodistillation	GC-MS	α -cubebol (14.5%), <i>trans</i> -hanphyllene (11.1%), and germacrene-D (9.5%)		- Terpenoids consisted of the main class in EO.
<i>A. macedonica</i> Boiss. & Orph. ^[g]	Bul, Grc, Mkd, Srb	Aerial parts	- Sesquiterpene lactones ^[51]	Chloroform	Column chromatography; ¹ H NMR spectroscopy	Parthenolide and stizolin		- 9 compounds (6 germacranolides and 3 eudesmanolides) were identified.
<i>A. sribnyi</i> subsp. <i>tracica</i> Velen. ^[g]	Bul, Grc	Aerial parts	- Sesquiterpene lactones ^[51,52]	Chloroform	Column chromatography; ¹ H NMR spectroscopy	Isospiciformin		- In total, 4 germacranolides were isolated and identified. One of them (lactone form) was recorded for the first time.
<i>A. rumelica</i> (Velen.) Stoj. & Acht. ^[g]	Bul	Aerial parts	- Sesquiterpene lactones, ^[53]	Chloroform	Silica gel column chromatography; ¹ H NMR spectroscopy	No data		- 6 compounds were identified: tanacin, hanphyllin, two guaianolides, centaureidin and santin.
<i>Artemisia arborescens</i> (Vaill.) L.	Grc	Aerial parts	- EO ^[54]	Hydrodistillation	GC-MS	Camphor (30.5%), <i>trans</i> -thujone (18.4%)		- 41 compounds were identified in EO.
<i>A. inculta</i> Delile	Grc	Aerial parts	- EO ^[54]	Hydrodistillation	GC-MS	1,8-cineole (28.1%), <i>cis</i> -thujone (17.5%)		- 37 compounds were identified in EO.
<i>Aster albanicus</i> (Degen) Degen ^[g]	Alb, Srb	Aerial parts	- EO ^[55]	Hydrodistillation	GC/GC-MS	Germacrene D (34.7%), β -		- Chemical analysis of plant

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						pinene (11.9%), Caryophyllene E (10.9%)	EO revealed presence of 111 components (sesquiterpenes and monoterpene hydrocarbons were the main ones).
<i>Carduus candicans</i> subsp. <i>globifer</i> (Velen.) Kazmi ^[g]	Bul, Rou, Srb, Tur	No data	- Water soluble polyphenols and flavonoids ^[56]	EtOH	UV-Vis spectroscopy	No data	- Among the examined species, it was the second one based on total phenolic and flavonoid content.
		Flower heads	- EOs ^[57]	Hydrodistillation	GC-MS	Benzaldehyde (22.1%), pantoic acid (8.1%)	- 50 compounds were identified during EO analysis.
<i>C. kernerii</i> subsp. <i>austro-orientalis</i> Franco ^[g]	Alb, Bul, Grc, Mkd, Mne, Srb	No data	- Water soluble polyphenols and flavonoids ^[56]	EtOH	UV-Vis spectroscopy	No data	- EtOH extract of this plant possessed the lowest content of water-soluble polyphenols and flavonoids.
<i>Centaurea achaia</i> Boiss. & Heldr. ^[g]	Grc	Aerial parts	- Sesquiterpene lactones ^[58]	Cyclohexane/ether/MeOH (1:1:1)	Column chromatography; TLC; ¹ H NMR spectroscopy	No data	- 15 compounds (9 germacranolides, 2 elemanolides, 2 eudesmanolides, and 2 hydroxyesters derivatives) were isolated and identified.
<i>C. attica</i> Nym subsp. <i>attica</i> ^[g]	Grc	Aerial parts	- Sesquiterpene lactones ^[59]	Cyclohexane/ether/MeOH (1:1:1)	Column chromatography; HPLC	Cnicin (101.1 mg)	- 2 germacranolides and 2 elemanolides were isolated and identified from the extract.
<i>C. melanocephala</i> Pančić ^[g]	Alb, Srb	Capitula	- Monoterpenes and sesquiterpenes ^[60]	Dichloromethane	GC; GC-MS	β -caryophyllene (9.0%), caryophyllene oxide (8.1%)	- In total, 61 compounds (monoterpenes and sesquiterpenes) were identified. About 30% of the compounds belonged to monoterpenes. High content of oxygenated sesquiterpenes was also observed.
<i>C. chrysolepis</i> Vis. ^[g]	BiH, Bul, Mne, Srb	Capitula	- Monoterpenes and sesquiterpenes ^[60]	Dichloromethane	GC; GC-MS	Humulonsane (14.1%), β -caryophyllene (12.8%)	- 59 components were recorded in EO with monoterpenes as a minor fraction.
<i>C. grisebachii</i> (Nyman) Heldr. subsp. <i>grisebachii</i> ^[g]	Alb, Bul, Grc, Mkd, Srb	Aerial parts	- Sesquiterpene lactones, flavonoid aglycons,	Cyclohexane/ether/MeOH (1:1:1) and MeOH/aqueous	Silica gel column chromatography/HPLC;	4- <i>epi</i> -malacitenolide	- 10 sesquiterpene lactones, 5 flavonoids, 2 lignans and 1

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			lignans and sterol ^[61]	(5:1)		¹ H NMR spectroscopy	sterol were identified. None of the guaianolides were detected which is a chemotaxonomic characteristic of this plant.
<i>C. galicicae</i> Micevski ^[5]	Mkd	Aerial parts	- Sesquiterpene lactones and flavonoids ^[62]	Petrol ether/ether/MeOH (1:1:1)		¹ H and ¹³ C NMR spectroscopy; LC-ESI-MS	- Lactone (cnicin) was quantified and 6 flavonoids (apigenin, isokaempferide, hispidulin, eupatorin, santoflavone and salvigenin) were identified.
		Fruits	- Fatty acids ^[63]	2-propanol		GC-FID; GC-MS	- 11 fatty acids were identified.
<i>C. gracilentia</i> Velen. ^[5]	Bul	Aerial parts	- EOs ^[64]	Hydrodistillation		GC; GC-MS	- Plant EO (45 identified compounds) constituted mostly sesquiterpenoids, different hydrocarbons and carboxylic compounds (in total, 4 compounds).
<i>C. gloriosa</i> ssp. <i>multiflora</i> Radić	Cro	Aerial parts	- EOs and total polyphenols ^[65]	n-hexane, EtOH, aqueous		GC-MS	- Total phenolic content of aqueous plant extract was the highest compared to n-hexane and EtOH ^[1] extracts.
<i>C. derventana</i> Vis. & Pančić ^[5]	BiH, Mne, Srb	Aerial parts	- Sesquiterpene lactones, triterpenes sterols and flavonoids ^[66]	No data		Silica column gel chromatography	- 4 germacranolides were quantified. In addition, 2 triterpenes (α and β -amyrin), 2 flavonoids (apigenin and eupatillin) and 2 sterols (stigmasterol and β -sitosterol) were also isolated
<i>C. davidovii</i> Urum. ^[5]	Bul	Aerial parts	- EOs ^[67]	Hydrodistillation		GC; GC-MS	- In total, 62 compounds were identified and quantified.
		Fresh, <i>in vitro</i> and <i>ex-vivo</i> grown leaves	- Total phenolics and flavonoids, sesquiterpene lactones ^[68]	80% MeOH (for phenolics); Chloroform (for sesquiterpene lactones)		UV-Vis; HPLC	- The content of total phenolics and flavonoids was higher in fresh leaves compared to those grown under controlled conditions (<i>in vitro</i> and <i>ex-vivo</i>).
<i>C. euxina</i> Velen. ^[5]	Bul	Aerial parts	- EOs ^[69]	Hydrodistillation		GC-MS	- 58 volatile components were

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<i>C. formanekii</i> Halácsy ^[g]	Bul, Grc, Mkd	Aerial parts	- EOs ^[70]	Hydrodistillation	GC-MS	and spathulenol (10.8%) Hexadecanoic acid (13.6%), and δ -elemene (9.1%)	identified and quantified. - 64 compounds were determined in EO.
<i>C. kosaninii</i> Hayek ^[g]	Alb, Mkd, Srb	Aerial parts	- Sesquiterpene lactones, triterpenes sterols and flavonoids ^[66]	No data	Silica column gel chromatography	Mixture of sterols (stigmasterol and β - sitosterol) and triterpenes (α and β -amyrin)	- None of the lactones or flavonoids were isolated from extract of this plant.
<i>C. moesiaca</i> Urum. & J.Wagner ^[e]	Bul, Srb	Aerial parts	- Sesquiterpene lactones, flavonoids and lignan ^[71]	Chloroform	Silica gel column chromatography	Cnicin, 4'-acetylcnicin	- In total, 21 components were isolated from chloroform extract: eleven sesquiterpene lactones, nine flavonoids (derivatives of 6- methoxyflavones and 6- methoxyflanonols) and one lignan (trachelogenin).
<i>C. orphanidea</i> Heldr. & Sart. ex Boiss. subsp. <i>thessala</i> (Hausskn.) Dostál ^[g]	Grc	Aerial parts	- EOs ^[70]	Hydrodistillation	GC-MS	γ -elemene (26.1%), and carvophyllene oxide (13.2%)	- 41 components were identified and quantified.
		Capitula		Hydrodistillation	GC-MS	hexadecanoic acid (33.5%), and heptacosane (6.3%)	- 41 components were identified and quantified.
		Roots		Hydrodistillation	GC-MS	octyl-hexadecanoate (22%), and α -chamigrene (14%)	- 41 components were identified and quantified.
<i>C. thessala</i> Hausskn. subsp. <i>drakiensis</i> (Freyn & Sint.) T.Georgiadis ^[g]	Grc	Aerial parts	- EOs ^[72]	Hydrodistillation	GC-MS	Hexacosane (22.6%), and carvophyllene oxide (7.8%)	- In total 43 EO components were identified.
<i>C. parilica</i> Stoj. & Stef. ^[g]	Grc, Bul	Aerial parts	- EOs ^[60]	Hydrodistillation	GC; GC-MS	Hexadecanoic acid (39.2%), and (Z,Z)-9,12- octadecadienoic acid (14.5%)	- 28 identified compounds.
<i>C. peltia</i> DC. ^[g]	Grc	Aerial parts	- EOs ^[72]	Hydrodistillation	GC-MS	Heptacosane (5.7%), and carvophyllene oxide (4.3%)	- In total 80 EO components were identified.
<i>C. ragusina</i> L. ^[g]	Cro	Flowers	- EOs ^[73]	Hydrodistillation	GC; GC-MS	Gamma-ene D (17.1%), and γ -myrcenolene (9.1%)	- 62 different compounds (sesquiterpenes, hydrocarbons, aldehydes, etc.) were identified and quantified.
			- Sesquiterpene lactones, organic acids and phenolics ^[74]	Aqueous (hot water)	HPLC-ESI-MS	Quinic acid, and citric acid	- Aqueous extracts was characterized with the presence of different lactones, organic acids as well as some

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		Leaves	- EOs ^[73]	Hydrodistillation	GC; GC-MS	Cycloocta-1,3-diene (13.6%), octadec-9-enoic acid (10.6%), and acetophenone (10.3%)	phenolic compounds. - In total, 61 different compounds (sesquiterpenes, hydrocarbons, acids, ketones etc.) were identified and quantified.
		Herbal parts	- Sesquiterpene lactones, organic acids and phenolics ^[74]	Aqueous (hot water)	HPLC-ESI-MS	Solstitialin A-3,13-diacetate, epoxyrepiolide and quinic acid	- Aqueous extract did not contain polyphenol compounds.
<i>C. raphanina</i> subsp. <i>mixta</i> (DC.) Runemark ^[8]	Grc	Aerial parts	- EOs ^[75]	Hydrodistillation	GC-MS	Caryophyllene oxide (10.5%), and <i>epi</i> -bicyclosesquiphellandrene (6.9%)	- EO of aerial plant parts contained 74 different compounds.
<i>C. spinosa</i> L. ^[8]	Grc	Aerial parts	- Sesquiterpene lactones and flavonoids ^[76]	Cyclohexane/ether/MeOH (1:1:1)	Column chromatography; HPLC; ¹ H NMR and UV spectroscopy	4'-acetylnicin (354 mg)	- 10 sesquiterpene lactones (two were new natural products) and 9 flavonoids were isolated and identified in plant extract.
<i>C. spruneri</i> Boiss. & Heldr.	Alb, Grc	Aerial parts	- EOs ^[75]	Hydrodistillation	GC-MS	β -elemene (27.7%)	- 50 compounds were isolated from the plant.
<i>C. sibthorpii</i> Halácsy ^[8]	Grc	Aerial parts	- EOs ^[77]	Hydrodistillation	GC; GC-MS	Hexadecanoic acid (18.6%), and 9,12-octadecadinoic acid (1.2%)	- The obtained plant EO consisted of 63 different compounds. Fatty acids and sesquiterpenes were predominant classes of compounds.
<i>C. soskai</i> Hayek ex Košanin ^[8]	Alb, Mkd	Aerial parts	- Sesquiterpene lactones and flavonoids ^[62]	Pethrol ether/ether/MeOH (1:1:1)	¹ H and ¹³ C NMR spectroscopy; LC-ESI-MS	Santoflavone	- 1 lactone (nicin) was quantified and 6 flavonoids (apigenin, isokaempferide, hispidulin, eupatorin, santoflavone and salvigenin) were identified.
<i>C. tuberosa</i> Vis. ^[8]	Alb, BiH, Bul, Cro, Grc, Mkd, Mne, Srb	Aerial parts	- EOs ^[78]	Hydrodistillation	GC	Caryophyllene-oxide (27.8%), and β -caryophyllene (11.1%)	- 32 compounds were registered in the obtained oil.
<i>C. tomorosii</i> Micevski ^[8]	Mkd	Aerial parts	- Sesquiterpene lactones and flavonoids ^[62]	Pethrol ether/ether/MeOH (1:1:1)	¹ H and ¹³ C NMR spectroscopy; LC-ESI-MS	Santoflavone	- 1 lactone (nicin) was quantified and 7 flavonoids (apigenin, isokaempferide,

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		Fruits	- Fatty acids ^[63]	2-propanol	GC-MS	Linolelaic acid (48.8% i.e. 2.6 g/100g)	hispidulin, eupatorin, santoflavone, cirsimaritin and salvigenin) were identified.
<i>C. vlachorum</i> Hartvig	Alb, Grc	Aerial parts	- EOs ^[79]	Hydrodistillation	GC-MS	Caryophyllene-oxide (11.9 - 26.7%), and spathulenol (9.5 - 15.6%)	- Only 5 fatty acids were identified. - The leaves, flowers and stems of examined plants were characterized with prevalence of oxygenated sesquiterpenes. The following components were accompanying: E-nerolidol and humulene epoxide II in leaves, <i>n</i> -heneicosane and methyl-linoleate in flowers i.e. 13-epi-malooloxide and <i>n</i> -heneicosane in examined stems.
		Seeds	- Lignans and alkaloids ^[80]	No data	¹ H NMR spectroscopy	No data	- The first report about 2 lignans (matairesinol and arctiin) and 2 alkaloids (<i>N</i> - <i>p</i> -coumaroyl-serotonin and moschamine) in seeds of this plant has been made.
<i>C. zuccariniana</i> DC. ^[g]	Alb, Grc	Aerial parts	- EOs ^[72]	Hydrodistillation)	GC-MS	Hexadecanoic acid (6.5%), and caryophyllene oxide (6.2%)	- In total 74 EO components were identified.
<i>Crepis incana</i> Sm. ^[g]	Grc	Aerial parts	- Sesquiterpene lactones and flavonoids ^[81]	Cyclohexane/ether/water (1:1:1) followed by MeOH/water (5:1)	TLC; ¹ H NMR spectroscopy	Ghreosimin and 1- <i>O</i> - β -D-glucopyranosyl ester of taraxinic acid	- 5 different lactones (ghreosimin, crepaside E, crepaside D, taraxinic acid and its ester with glucose) and 3 flavonoids (luteolin and its glucoside and 3- <i>O</i> -glucoside of quercetin) as well as 1 nor-preonid (loliolide) were isolated for the first time in case of this species.
		Roots	- Triterpenoids ^[81]	Cyclohexane/ether/water	TLC; ¹ H NMR	No data	- 2 triterpenoids (lupeol and

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Species	Origin	Part	Compounds	Extraction	Analysis	Findings	Notes
<i>Hieracium orienti</i> A.Kern.	Alb, Bul, Cro, Grc, Mne, Mkd, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	(1:1:1) Dichloromethane followed by MeOH	spectroscopy LC-MS	Chlorogenic acid (18.8 mg/g), luteolin-7- <i>O</i> -glucuronide (17.7 mg/g), luteolin-7- <i>O</i> -glucoside (17.5 mg/g), and luteolin 7- <i>O</i> -glucoxyloside (16.8 mg/g)	oleanolic acid) were identified. - 16 compounds (12 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	None	- 2 guaianolide lactones were quantified but as traces.
<i>H. durmitoricum</i> (Rohlena & Zahn) Niketic ^[81]	Mne, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin-7- <i>O</i> -glucoside (34.0 mg/g), luteolin 7- <i>O</i> -glucoxyloside (16.4 mg/g), and chlorogenic acid (13.5 mg/g)	- 22 compounds (18 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Crepiside E (5.5 mg/g), and guaianolide glucoside (4.7 mg/g)	- 4 lactones were quantified.
<i>H. guentheri-beckii</i> Zahn ^[81]	Alb, BiH, Mne, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin-7- <i>O</i> -glucuronide (10.5 mg/g), chlorogenic acid (19.8 mg/g), Luteolin-7- <i>O</i> -glucoside (15.2 mg/g), and 3,5-dicaffeoylquinic acid (14.5 mg/g)	- 21 compounds (17 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Guaianolide glucoside (11.1 mg/g), and crepiside E (10.4 mg/g)	- 3 lactones were quantified.
<i>H. mirificissimum</i> Rohlena & Zahn ^[81]	BiH, Mne, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin-7- <i>O</i> -glucuronide (74.5 mg/g), chlorogenic acid (30.3 mg/g), luteolin-7- <i>O</i> -glucoside (16.95 mg/g), and luteolin 7- <i>O</i> -glucoxyloside (14.5 mg/g)	- 14 compounds (10 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Crepiside E (10.1 mg/g), and guaianolide glucoside (2.4 mg/g)	- 3 lactones were quantified.

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<i>H. mokragorae</i> (Nägeli & Peter) Nägeli & Peter ^[6]	Alb, BiH, Bul, Mne, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin-7- <i>O</i> -glucuronide (18.5 mg/g), 3,5-dicaffeoylquinic acid (12.8 mg/g), and chlorogenic acid (12.7 mg/g)	- 11 compounds (7 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Crepiside E (0.3 mg/g)	- Guaianolide glucoside and calophyllamine A were quantified in traces.
<i>H. albopellitum</i> (Zahn) Niketić ^[6]	Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin-7- <i>O</i> -glucuronide (26.4 mg/g), luteolin-7- <i>O</i> -glucoside (16.3 mg/g), chlorogenic acid (14.6 mg/g), and 3,5-dicaffeoylquinic acid (9.8 mg/g)	- 16 compounds (12 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Crepiside E (0.5 mg/g)	- Guaianolide glucoside, calophyllamine A and B were quantified in traces.
<i>H. naegelianum</i> Pančić	Mkd, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin 7- <i>O</i> -glucoside (49.2 mg/g), luteolin hexosylpentoside (40.8 mg/g), and luteolin 7- <i>O</i> -glucuronide (34.8 mg/g)	- 15 compounds (11 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Crepiside E (7.9 mg/g), and guaianolide glucoside (3.7 mg/g)	- 3 lactones were quantified.
<i>H. macrodontoides</i> (Zahn) Zahn	Alb, Mne, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	3,5-dicaffeoylquinic acid (29.1 mg/g), luteolin-7- <i>O</i> -glucuronide (26.9 mg/g), chlorogenic acid (24.8 mg/g), and luteolin 7- <i>O</i> -glucoside (21.0 mg/g)	- 12 compounds (8 flavonoids and 4 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	None	- Only 1 lactone (eudesmanolide type i.e. calophyllamine B) was above the limit of quantification.
<i>H. tommasinianum</i> K.Malý	Alb, Bul, Cro, Grc, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	3,5-dicaffeoylquinic acid (28.6 mg/g), luteolin 7- <i>O</i> -	- 12 compounds (8 flavonoids and 4 phenolic acids) were

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			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	None	glucoxyloside (16.1 mg/g), chlorogenic acid (15.7 mg/g), and luteolin-7-O-glucuronide (15.3 mg/g), quantified. It was one of the species where phenolic acids were predominant components compared to flavonoids. - Only 1 lactone (crepiside E) was present, but in low quantity (below 1 mg/g).
<i>H. bupleuroides</i> subsp. <i>pseudoschenkii</i> Rohlena & Zahn	BiH, Mne	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Chlorogenic acid (22.3 mg/g), 3,5-dicaffeoylquinic acid (22.3 mg/g), and luteolin 7-O-glucoxyloside (13.7 mg/g)	- 17 different components (13 flavonoids and 4 phenolic acids) were recorded in the extracts.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Guaiacolide glucoside (10.4 mg/g) and calophyllamine A (5.1 mg/g)	- 3 lactones were quantified.
<i>H. waldsteinii</i> subsp. <i>plumosum</i> (A.Kern.) Freyn	BiH, Mne	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin 7-O-glucuronide (33.9 mg/g), chlorogenic acid (17.8 mg/g), luteolin 7-O-glucoside (14.7 mg/g), and 3,5-dicaffeoylquinic acid (9.5 mg/g) Crepiside E (7.5 mg/g)	- 13 compounds (8 flavonoids and 5 phenolic acids) were quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS		- 3 lactones were quantified.
<i>H. coloriscapum</i> Rohlena & Zahn	Alb, Grc, Mne	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin 7-O-glucuronide (23.7 mg/g), luteolin-7-O-glucoside (23.0 mg/g), chlorogenic acid (21.0 mg/g), and luteolin 7-O-glucoxyloside (15.3 mg/g)	- 18 different components (13 flavonoids and 5 phenolic acids) were isolated and quantified.
			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	Crepiside E (10.1 mg/g)	- 4 lactones were quantified.
<i>H. gymnocephalum</i> Griseb. ex Pant.	Alb, BiH, Grc, Mne, Srb	Aerial parts	- Flavonoids and phenolic acids ^[82]	Dichloromethane followed by MeOH	LC-MS	Luteolin 7-O-glucoxyloside (25.1 mg/g), luteolin 7-O-	- 18 compounds (14 flavonoids and 4 phenolic

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			- Sesquiterpene lactones ^[83]	MeOH	LC-MS	glucoside (20.2 mg/g), chlorogenic acid (15.3 mg/g), and luteolin 7-O-glucuronide (11.9 mg/g) Crepiside E (14.2 mg/g) and guaianolide glucoside (8.2 mg/g)	acids) were recorded. - 3 lactones were quantified.
<i>Jurinea tzar-ferdinandii</i> Davidov ^[g]	Bul, Rou	Aerial parts	- Terpenoids, sesquiterpene lactones, and flavonoids ^[84]	<i>n</i> -hexane Chloroform	Column chromatography; GC-MS; ¹ H NMR spectroscopy	8-(4'-hydroxymethacryloyloxy)-15-oxo- <i>melianol</i> -1(10),4,11(15)- <i>tri</i> -6,12- <i>olide</i> (6.3 mg)	- In total 22 compounds were isolated and identified.
<i>Onopordum laconicum</i> Heldr. & Sart. ex Rouy ^[g]	Grc	Aerial parts	- Flavonoids ^[85]	<i>n</i> -hexane MeOH	Column chromatography; TLC; ¹ H NMR and UV spectroscopy	Chrysoeriol-7- <i>O</i> - β -glucoside (15 mg), and acetin (15 mg)	- 12 flavonoids and derivatives were isolated and identified.
<i>Senecio macedonicus</i> Griseb. ^[g]	Bul, Grc, Mkd	Leaves	- Sesquiterpene lactones ^[86]	Chloroform	Column chromatography; ¹ H NMR spectroscopy	No data	- 4 sesquiterpene lactones were identified. Two of them were new C-8 epimeric secomacrotolides.
		Roots	- Alkaloids ^[87]	EtOH	Column chromatography; GC-MS; ¹ H and ¹³ C NMR spectroscopy	No data	- 6 alkaloids were identified in plant root ethanolic extract.
<i>Tanacetum corymbosum</i> (L.) Sch.Bip.	BiH, Bul, Grc, Cro, Mne, Mkd, Srb, Tur	Whole plant	- Sesquiterpene lactones ^[26]	Chloroform	No data	None	- None of the lactones were identified. This plant is well known as a poor source of these lactones ^[26] .
<i>T. larvatum</i> (Pant.) Hayek ^[g]	Alb, Mne, Srb	Aerial parts	- Parthenolide - sesquiterpene lactone ^[88]	Pethrol ether/ether/MeOH (1:1:1)	Silica gel column chromatography/dry-column flash chromatography	Parthenolide (35 mg) and douglanine (21 mg)	- 2 lactones (germacranolide parthenolide and eudesmanolide douglanine) and 1 flavone (tricin) were isolated and identified.
			- EOs ^[89]	Hydrodistillation	GC	<i>trans</i> -sabinyl acetate (51.2 - 69.7%), β -pinene (4.3 - 7.7%), and camphor (4.3 - 6.3%)	- 47 compounds were isolated and identified. Among them, monoterpenes were the predominant class.

^[a] Alb- Albania; BiH- Bosnia and Herzegovina; Bul- Bulgaria; Cro- Croatia; Grc- Greece; Mkd- North Macedonia; Mne- Montenegro; Srb- Serbia; Tur - Turkey; Rou- Romania^[b] EOs/EO - essential oils/ essential oil; ^[c] EtOAc - ethyl-acetate; ^[d] BuOH - butanol; ^[e] MeOH - methanol; ^[f] EtOH - ethanol ^[g] Balkans' endemic taxa; ^[h] LC ESI MS - Liquid Chromatography-Electrospray Ionization-Mass Spectrometry

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74 It is obvious that there is great diversity regarding the chemical composition of studied plant species from the Balkan Peninsula. As expected, the main
75 classes of chemical compounds of EOs, mainly different mono- and sesquiterpenes were similar to those reported for other taxa belonging to Asteraceae.
76 ^[21,90] In addition, in surveyed Balkans' Asteraceae taxa, the flavonoids were the main phenolic subclass as already reported for large families of vascular
77 plants.^[21,90] The chemical structures of some of the representative bioactive compounds determined in different extracts of Balkans' Asteraceae species are
78 given in **Figure 2** (see supplementary file).

79
80 As shown in **Table 1**, the literature search included 63 species of Asteraceae typical for the Balkan Peninsula for which some phytochemical records already
81 exist. The highest number of species belongs to the genus *Centaurea* (25), followed by genera *Achillea* and *Hieracium* (13 each), *Artemisia*, *Carduus*,
82 *anacetum* (2 each), *Aster* and *Crepis* (1 each). The bioactive and potentially bioactive specialized metabolites from these taxa belong to different classes:
83 EOs (10 oxygenated monoterpenes, 3 monoterpene hydrocarbons and 1 oxygenated sesquiterpene were presented in significant quantities (above 20%) in all
84 Balkans' *Achillea* species),^[91] esters, flavonoids, phenolic acids, sesquiterpene lactones, triterpenoids (from *Achillea* taxa); flavonoids and other polyphenols,
85 n-alkanes, sesquiterpene lactones-amphoricarpolides (from *Amphoricarpos* taxa); EOs (from *Aster albanicus*) and EOs, water soluble phenolic compounds
86 (from *Carduus* taxa); EOs, fatty acids, flavonoids, lignans, monoterpenes, organic acids, phenolics, sesquiterpenes, sesquiterpene lactones, sterols,
87 triterpenes, triterpene sterols (from *Centaurea* taxa); flavonoids, sesquiterpene lactones, triterpenoids (from *Crepis incana*); flavonoids, phenolic acids,
88 sesquiterpene lactones (from *Hieracium* taxa) and EOs and parthenolide - sesquiterpene lactone (from *Tanacetum* taxa). In addition, some organic acids,
89 lignans and alkaloids were recorded in several Balkans' Asteraceae species. A range of specialized metabolites, including some specific components, such as
90 some flavonoids, sesquiterpene lactones and rare fatty acids (γ -linoleic acid, *trans*, epoxy and hydroxy forms) might be of chemophenetic relevance. For
91 instance, several germacranolides are principal components of *Anthemis macedonica*^[52] In addition, linear irregular lactones could be considered as
92 chemophenetic markers for two Asteraceae genera: *Anthemis* and *Maruta*.^[52] Todorova et al.^[92] determined the presence of seven new irregular sesquiterpene
93 dilactones in the flower of *Anthemis auriculata* Boiss. from Bulgaria. Same authors^[44] reported the presence of one unusual bis-norsesquiterpene lactone
94 (proposed name is spirodepressolide) in aerial parts of *Achillea depressa* Janka. Regarding some characteristic flavonoids, literature data reported the
95 presence of pectolinarigenin, eupatilin and jaceosidin for the first time in *J. tzar-ferdinandii* Davidov.^[84] These compounds possibly make a biogenetic
96 relationship between *Jurinea* genus and two other Asteraceae genera: *Onopordon* and *Centaurea*.^[84] Many of the examined Balkans' Asteraceae species are
97 endemic (**Table 1**), and are not used in traditional or official medicine. However, some of Balkan endemics might be of further research interest upon
98 preliminary information on the phytochemical composition and related biological activity (**Table 2**). It should be emphasized that endemic Asteraceae
99 species are mostly composed of small populations with limited distribution. Therefore, it is necessary to implement a range of *in situ* and *ex situ* conservation
100 measures for their possible commercial use.

101

102 2.2. Biological activity

103 Nowadays, the importance of biologically active substances acting as antioxidants is based upon their ability to prevent cell and macromolecule damages
104 caused by free radicals, reactive oxygen (ROS) and nitrogen (RNS) species.^[93] Ability of different compounds to perform as antioxidants is established on
105 the positive redox potential during reaction with different prooxidants allowing them to inhibit "oxidative stress" (OS). The number of articles dealing with
106 antioxidant properties / abilities of different plant foods or food ingredients has become immense during the last few decades.^[94-96] Due to the importance of
107 antioxidants in human diet, there are several antioxidant tests developed for assessment of biological behavior of whole plant extracts and individual
108 components.^[97] The main differences among these assays are related either to the way of performance - *in vitro* and *in vivo*^[93,98] or the principal chemical
109 reaction mechanism. In that sense we can distinguish so-called "hydrogen atom transfer-based" (HAT), "single electron transfer-based" (SET) or mixed
110 assays.^[93,99] Currently, there are 19 *in vitro* and 10 *in vivo* tests used to establish antioxidant potential of plant material.^[100] Among all plant metabolites,
111 polyphenols, carotenoids and vitamins are considered as the most prominent antioxidants^[101,102] as well as valuable antimicrobial agents.^[103] Application of
112 plant extracts and individual components as effective agents against different microorganisms is still in progress and will be even more applied due to known
113 resistance to standard antibiotics.^[103] In **Table 2** available literature sources for antimicrobial activity and *in vitro* and *in vivo* antioxidant effects of selected
114 Balkans' Asteraceae plants are summarized.

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Table 2. Biological activity assays of selected Balkans' Asteraceae plants.

Scientific species name	Part of plant	Biological activity assays ^[b]	Remarks ^[c]
<i>Achillea alexandri-regis</i> Bornm & Rudsky	Aerial parts	- Anti-inflammatory and anti-ulcer activity ^[28] - Antioxidant activity ^[29] - Antioxidant and cytotoxic activity ^[30]	- All applied MeOH ^[a] extracts were successful as anti-inflammatory agents in doses below the dose necessary for the anti-ulcer effect - Both plant extracts (50% EtOH ^[b] and EtOAc ^[c]) were successful in hydroxyl and superoxide radicals neutralization - MeOH ^[c] extract exhibited the highest DPPH activity while combined chloroform and EtOAc extracts showed significant ability to act against adenocarcinoma cells (HeLa)
<i>A. holosericea</i> Sm.	Aerial parts	- Antimicrobial activity ^[32] - Antimicrobial activity ^[39]	- EtO ^[d] obtained from this plant were completely inactive against examined bacteria and fungi species - Plant extract possessed low but measurable activity against 5 bacteria and 2 fungi
<i>A. clavennae</i> L.	Aerial parts	- Antimicrobial activity ^[39]	- Plant extract exhibited significant antimicrobial activity against 5 bacteria and 2 fungi species
<i>A. chrysocoma</i> Friv.	Aerial parts	- Antibacterial activity ^[37]	- EtO showed significant activity against 2 gram-negative bacteria
<i>A. fraasii</i> Sch.Bip.	Aerial parts	- Antimicrobial activity ^[32]	- Extract presented a significant activity against gram negative bacteria
<i>A. taygetea</i> Boiss & Heldr.	Aerial parts	- Antimicrobial activity ^[32]	- Extract presented a moderate activity against gram negative bacteria and pathogenic fungi
<i>A. thracica</i> Velen.	Fresh, in vivo and ex-vitro grown plant parts	- Antibacterial activity ^[41]	- Acetonic extract of naturally obtained plants showed very weak antibacterial activity while the other two extracts can be characterized as moderate antibacterial agents against five bacterial strains
<i>A. umbellata</i> Sm.	Aerial parts	- Antimicrobial activity ^[48]	- The highest inhibitor activity was recorded against <i>Staphylococcus aureus</i> while <i>Pseudomonas aeruginosa</i>

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			- <i>in vivo</i> toxicity and locomotor activity ^[48]	was resistant to EO treatment
<i>Amphoricarpus autariatus</i> subsp. <i>bertisceus</i> Blečić & E.Mayer	Leaves		- Antimicrobial activity	- The first case of mortality between treated mice was recorded at EO dose of 600 mg/kg. Compared to standard drug (diazepam) plant EO exhibited significant activity
			- Antioxidant activity ^[19]	- Examined extracts possessed a significant antimicrobial potential
<i>A. autariatus</i> Blečić & Mayer subsp. <i>autariatus</i>	Leaves		- Antifungal activity ^[49]	- Dry MeOH plant extract showed a higher DPPH activity compared to liquid MeOH and aqueous extracts
			- Antimicrobial and antioxidant activity ^[19]	- Lactone fraction possessed a significant activity against eight examined fungi
<i>A. neumayerianus</i> (Vis.) Greuter	Leaves		- Antimicrobial and antioxidant activity ^[19]	- Examined extracts possessed a significant antimicrobial potential. Dry MeOH plant extract showed a higher DPPH activity compared to liquid MeOH and aqueous extracts
<i>Artemisia. arborescens</i> (Vaill.) L.	Aerial parts		- Antioxidant activity ^[54]	- Examined extracts possessed a significant antimicrobial potential. Dry MeOH plant extract showed a higher DPPH activity compared to liquid MeOH and aqueous extracts
<i>A. inculta</i> Delile	Aerial parts		- Antioxidant activity ^[54]	- DPPH test proved that EO of <i>A. arborescens</i> possessed 40 times higher antioxidant activity compared to EO of <i>A. inculta</i> mostly due to absence of chamazulene in EO of <i>A. inculta</i>
<i>Carduus candicans</i> subsp. <i>globifer</i> (Velen.) Kazmi	No data		- Antioxidant activity ^[56]	- MeOH plant extracts exhibited significant ability to quench DPPH and ABTS free radicals as well as to reduce Fe ³⁺ ions during FRAP assay. In addition, compared to BHT, plant extracts had higher ability to prevent lipid peroxidation
<i>C. kernerii</i> subsp. <i>austro-orientalis</i> Franco	No data		- Antioxidant activity ^[56]	- Several compounds, extracted and isolated from plant extract, showed strong antifungal activity against nine tested fungi: <i>A. niger</i> , <i>A. ochraceus</i> , <i>A. versicolor</i> , <i>A. javus</i> , <i>A. alternata</i> , <i>C. cladosporioides</i> , <i>P. ochrochloron</i> , <i>P. fruticulosum</i> , and <i>T. viride</i> . MIC values were in range 0.1 - 3.0 nmol/L while MFC values were between 0.75 and 6.0 nmol/L except in case of 4- <i>epi</i> -carmanin.
<i>Centaurea achaila</i> Boiss. & Heldr.	Aerial parts		- Antifungal activity ^[58]	- Three different tests were applied: DPPH, ABTS and ORAC. Aqueous plant extract showed significant ability to quench DPPH and ABTS radicals while ORAC test proved that the extracts of this plant can act both as oxidant and prooxidant which is controlled by free radical source
<i>C. gloriosa</i> var. <i>multiflora</i> Radić	Aerial parts		- Antioxidant activity ^[65]	- Disc diffusion method proved a low antimicrobial activity

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		- Antimicrobial activity ^[65]	of plant extract with <i>Candida</i> spp. and <i>Pseudomonas</i> spp. as the most sensitive microbes
<i>C. ragusina</i> L.	Flowers and leaves	- Antimicrobial activity ^[73]	- Based on the results of two <i>in vitro</i> applied methods (disc diffusion and microdilution) the plant extract showed significant antibacterial and antifungal activity at mass concentration of 250 µg/mL. The investigation included 4 gram-positive and 4 gram-negative bacteria as well as 2 fungal strains
	Herba and flowers	- <i>in vitro</i> cytotoxic activity ^[74]	- Examination of aqueous plant extract on 1 bladder and 1 glioblastoma cell lines proved good cytotoxic activity
<i>C. spinosa</i> L.	Aerial parts	- Antibacterial activity; <i>in vitro</i> cytotoxic activity ^[76]	- All lactones, isolated from plant extract, were inactive against selected Gram-negative bacteria while they exhibited some inhibitory effect against three Gram-positive bacteria (<i>S. aureus</i> , <i>B. Cereus</i> , <i>M. flavus</i>). Elemanolide was the most active compound against tested human cell lines (DLD1, SF268, H460, OVCAR3) especially in the case of OVCAR3 line with significant cell growth inhibition at concentration below 5 µM.
<i>C. sibthorpii</i> Halácsy	Aerial parts	- Antibacterial activity ^[77]	There was no any activity of plant EO against 10 examined bacteria
<i>C. vlachorum</i> Hartvig	Seeds	- Antioxidant activity ^[80]	DPPH assay revealed good antioxidant properties for all isolated components (<i>N-p</i> -coumaroyl-serotonin, matairesinol and moschamine) except in case of arctiin.
		- Inhibitor activity ^[80]	During lipid peroxidation inhibition test <i>N-p</i> -coumaroyl-serotonin and arctiin showed high antilipid peroxidation activity
<i>Hieracium</i> spp. (14) ^[6]	Aerial parts	- Antioxidant activity ^[82]	- Matairesinol and moschamine possessed good inhibition properties against soybean lipoxygenase enzyme
		- Inhibitor activity ^[80]	All extracts exhibited promising antioxidant properties in all applied assays (FRAP, DPPH and 2-deoxyribose assay).
<i>Jurinea tzar-ferdinandii</i> Davidov	Aerial parts	Anti-lipase activity ^[84]	The obtained results correlated with the results for phenolic compounds content in plant extracts
			IC ₅₀ values of chloroform extract against two lipase enzymes were 29 -i.e. 39 µg/mL for <i>Candida rugosa</i> lipase (CRL) and porcine pancreas lipase (PPL) respectively.
			Among tested isolated compounds onopordopicrin was the most prominent with IC ₅₀ values at 32 (CRL) and 36 µg/mL (PPL)

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<i>Tanacetum corymbosum</i> subsp. <i>cinereum</i> (Griseb.) Bormm.	Flowers	- <i>in vivo</i> antifeedant activity ^[104]	- MeOH flower extract exhibited significant antifeedant effect against larvae of <i>S. littoralis</i>
<i>T. larvatum</i> (Pant.) Hayek	Aerial parts	- Antioxidant activity ^[105]	- <i>in vitro</i> DPPH assay revealed remarkable antioxidant capacity of plant extracts
		- Anti-inflammatory activity, gastroprotective effects ^[106]	- combined <i>in vivo</i> application of chloroform plant extracts with indomethacin on rats showed improved anti-inflammatory effect with reduced gastric lesions
<i>Senecio macedonicus</i> Griseb.	Roots	- <i>in vitro</i> cytotoxic activity ^[87]	- 9-angeloylplatynecine and mixture of sarracine and neosarracine (1:3 w/w), alkaloids isolated from plant roots extract, possessed promising immunomodulatory activity applied to normal murine spleen lymphocytes and P3U1 mouse myeloma cells.

116 ^[a] MeOH – methanol; ^[b] EtOH – ethanol; ^[c] EtOAc – ethyl-acetate; ^[d] EOs – essential oils; ^[e] the given results are related for fourteen Balkans' endemic species

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Essential oils are one of the most powerful natural antimicrobial agents.^[107] Similarly, some plant metabolites can act as biopesticides as it can be seen in the case of *Tanacetum corymbosum* subsp. *cinereum* (Griseb.) Bornm.^[90] Since plants produced some specific molecules in order to protect themselves from herbivores in this case effectiveness of methanolic plant extract against larvae of *Spodoptera littoralis* can be linked to the presence of several active compounds belonging to germacranolide, eudesmanolide, coumarin, and flavonol subclasses.^[90] Although synthetic antioxidants (such as butylated hydroxytoluene (BHT) and hydroxyanisole (BHA)) are still predominantly used in the food industry due to rising concerns about consumers' safety and healthy diet, there are great opportunities for "back to the nature" by applying natural antioxidants. Among 35 surveyed Asteraceae taxa, some of them showed promising antioxidant properties proved by several assays (DPPH, ABTS, ORAC, FRAP, mainly referring to *Tanacetum larvatum*, *Centaurea gloriosa* var. *multiflora* and several *Hieracium* species) (Table 2). Some authors pointed out the correlation between antioxidant ability and the content of polyphenols in Asteraceae taxa.^[82] In addition to antimicrobial effects, some phenolic compounds of Asteraceae species exhibited antiviral and anti-helminths activity, as reported for caffeic acids and tannins isolated from tarragon (*Artemisia dracunculus*).^[103] *Hieracium* species from the Balkan Peninsula were characterized as good sources of different luteolin derivatives and phenolic acids^[82] which might be of further interest for the testing of their possible antimicrobial activity. It had already been shown that: 3,5-di-*O*-caffeoylquinic acid, luteolin, 4-coumaric acid and quercetin pentosides isolated from the aqueous extract of dandelion (*Taraxacum mongolicum* Hand.-Mazz.) exhibit good antifungal activity against *Candida albicans*.^[108] Additionally, different phenolic compounds (e.g. caffeic acids, tannins, quercetin, chrysin, myricetin and some other flavonoids) were tested for anti-HIV activity.^[103] Due to the presence of several bioactive compounds (curcumin, and different polyphenolic compounds such as flavonoids and tannins) which can neutralize free radicals, medicinal plants can act as natural anti-inflammatory agents.^[109] In this way, these compounds prevent gastric ulcer since free radicals can react with some inflammatory molecules responsible for disease development^[109] such as nitric oxide, nitric oxide synthase and interleukins-1. The observed anti-inflammatory and anti-ulcer activities of *Achillea alexandri-regis* Bornm & Rudsky extract^[28] is mostly related to the presence of sesquiterpene lactones and, probably, some flavonoids^[28]. Similar effect was observed in the case of *Tanacetum larvatum* chloroform extract^[92] since it significantly reduced ulcerogenic effect of the standard drug, indomethacin, used for gastric ulcer treatment. Authors suggested that this important ability is mostly connected to the inhibition of activity NF- κ B protein complex responsible for the transcription process. It is probably caused by some of the active ingredients originated from plant extract^[92]. This is important since modern pharmacy is seeking for alternative sources of anti-ulcer agents to replace synthetic drugs which have some unpleasant side effects such as: nausea, abdominal pain, constipation, etc.^[110] Anti-cancer activity of plant-derived molecules has been recognized and used in order to replace some standard synthetic anti-cancer drugs.^[111] Among examined species cytotoxic activity of *Centaurea spinosa* L. extract is connected with one of the elemanolides' active ingredients. This compound belongs to sesquiterpene lactones and its activity against OVCAR3 cell line probably originated from the presence of aldehyde group.^[76] On the other side, *in vitro* cytotoxic activity of the extract obtained from *Senecio macedonicus* Griseb. roots^[87] is probably caused by the presence of several pyrrolizidine alkaloids which is in accordance with literature data about alkaloids' anti-cancer activity.^[111] Since all Asteraceae plants are a possible good source of phenolic acids and flavonoids, different biological activities of their extracts and of related compounds should be further researched.

148 3. Balkans' Asteraceae species as food source – review from ethnobotanical data

149 3.1. Historical background

Due to historical, political and biological factors, the Southeast Europe, i.e. the Balkan region is a hotspot of biocultural diversity in Europe.^[112] Traditional use of plants in the Balkans is very complex and interesting, considering the rich cultural and historical past and influences coming from the East and West, due to geographical position of the region. The curative and prophylactic use of medicinal herbs in the Balkan peninsula has a long tradition, starting from the first reports of Theophrastus and Dioscorides and several medieval manuscripts, such as Slavic manuscript found in Sinai monastery St Ekaterina (anonymous, XI century, Bulgaria), "Canon Prayer to St. Ivan Rilski and Medical Text" (anonymous, 1845, Bulgaria), "Herbarium" (by Peter Melius Juhasz, 1578, Romania), "Hodosh Codex" (anonymous, XIV century, Serbia), "Chilandar Medical Codex, No 517 (anonymous, XV century, Serbia) as well as some others. There is an important historical background on traditional knowledge of edible plants from the Balkan region. Some recent reports have addressed the use of wild edible plants in the Southeast Europe which is still ongoing in the rural areas.^[113–118] It is thought that nearly 30% of the world flora have edible parts and therefore are used in traditional nutrition.^[119] Many wild herbaceous plants are still collected by local people in the rural areas for their own use or for sale at the local markets.^[114,120] It is important to preserve the knowledge about traditional use of wild edible plants as a source of important nutrients, mainly vitamins, minerals and antioxidants.^[113] The interest in wild leafy vegetables is increasing due to documented health benefits of these herbs in human diet.^[121–124] Many of the wild edible plants belong to Asteraceae family, where preference for their use could be attributed to their satisfying sensory properties, resulting in domestication and cultivation of a range of species of this family for human consumption.^[125] Species of the Asteraceae family (exceeding 80 species) are widely used in the Mediterranean region and the Balkans as edible plants,^[116,117,126,127] (Table 3).

164 3.2. Edible green vegetables

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165 Among the most used wild Asteraceae are: dandelion (*Taraxacum officinale* (L.) Weber ex F.H.Wigg.) and chicory (*Cichorium intybus* L.), readily eaten in
 166 fresh, mixed and garnish salads.^[121] There are several reports on nutritive and phytochemical composition of these species, addressing the high content of
 167 potassium, calcium, manganese, iron, beta-carotene, dietary fibers and vitamin B6.^[123,125,128] The high content of B-sitosterol, quercetin glycosides,
 168 monocaffeoyltartaric acid and sesquiterpene lactones (taraxinic acid β D glucopyranoside) was determined in dandelion leaves,^[123] whereas flavonoids and
 169 hydroxycinnamic acids, with isomers of chicoric acid, chlorogenic acid, caftaric acid, and luteolin hexoside were dominant compounds in the leaves of
 170 chicory.^[122] Besides dandelion and chicory, the other frequently used edible Asteraceae in the Balkans are: yarrow (*Achillea millefolium* agg.), daisy (*Bellis*
 171 *perennis* L.), hawkbits (*Leontodon tuberosum* Batt.), Jerusalem artichoke (*Helianthus tuberosus* L.), stemless carline thistle (*Carlina acaulis*), common
 172 golden thistle (*Scolymus hispanicus* L.), common sow thistle (*Sonchus oleraceus* (L.) L.), and some other thistle plants (usually belonging to the tribe
 173 Cardueae), especially of genera *Carduus*, *Cirsium* and *Onopordum*. Edible thistles are known to have edible basal leaves and the midribs of the basal leaves,
 174 like in *Silybum*, *Cynara*, *Scolymus*, *Onopordum* and *Arctium*.^[129] The highest number of identified edible Asteraceae in the Balkans region is used as green
 175 vegetables, either raw as a salad, or as cooked vegetables (**Table 3**). This is in agreement with the data of Hadjichambis et al.^[126] showing that leaves are the
 176 most used plant parts, followed by shoots and other plant parts. It has already been reported that edible leafy vegetable Asteraceae species contain valuable
 177 dietary phytochemicals, mainly carotenoids, tocopherols and high content of ascorbic acid,^[121,130] in addition to various phenolic compounds and
 178 sesquiterpene lactones.^[131] Therefore, wild fresh leafy vegetables might constitute significant functional food components.^[132] It is known that in southern
 179 Europe and the Balkans, many bitter-tasting Asteraceae species are still used as a food in rural communities (e.g. *Cichorium*, *Hypochaeris*, *Sonchus*,
 180 *Leontodon*, *Arctium*), while being neglected in the northern Europe, due to replacement with species of less bitter taste, such as *Urtica*, *Chenopodium*,
 181 *Aegopodium* and some others.^[119] Bitter taste of leafy Asteraceae species is related to the presence of sesquiterpene lactones, exhibiting putative health-
 182 promoting effects.^[133]

183 3.3. Edible roots

184 Nearly 30 species of Asteraceae family with edible underground parts were recorded (**Table 3**), of which the following have the roots as the only plant part
 185 used: *Carlina acaulis* L., *Chondrilla juncea* L., *Inula helenium* L., *Picnemon acarna* (L.) Cass. and *Scorzonera hispanica* L. Roots of many edible
 186 Asteraceae plants are characterized by high content of fibers and inulin, a complex carbohydrate of sweet taste upon cooking, because of partial breakdown
 187 into fructose.^[119] The most used edible roots of Asteraceae species in the Balkans are dandelion, chicory and stemless carline thistle. Dandelion roots are
 188 known for the presence of carbohydrates, mainly inulin, mucilage, pectin and simple sugars (glucose, fructose and sucrose), carotenoids (lutein), as well as
 189 different phenolic compounds, mainly phenolic acids (chicoric acid, caffeic, chlorogenic acid), taraxasterol, chrysoeriol, luteoline glucoside and some other
 190 compounds.^[123] It has been reported that inulin accounts for up to 70% of the total root carbohydrates in chicory and that together with the Jerusalem
 191 artichoke (*Heliantus tuberosus* L.), it is the most important source of inulin for the industry uses.^[128] The root is also rich in hydroxycinnamic acids, such as
 192 ferulic, caffeic and chicoric acid, which are together with the inulin, responsible for the antidiabetic activity.^[134] Roots of *Carlina acaulis* L. are also very
 193 rich in inulin, in addition to high content of flavonoids and 1–2% of essential oil, with carlina oxide as the main component.^[135]

194 3.4. Edible flowers

195 Edible flowers contribute to the taste, appearance, aesthetic value and aroma of food, simultaneously expressing valuable nutrition and low calorie
 196 characteristics.^[136] Edible flowers are usually used to bring fragrance, flavor and color to main courses, salads, soups, entrees and drinks.^[137] Frequently used
 197 edible flowers of the Asteraceae species are: *Calendula officinalis* L., *Carthamus tinctorius* L., *Centaurea cyanus* L., *Chrysanthemum morifolium* Ramat.,
 198 *Cichorium intybus* L.,^[138] *Bellis perennis* L., *Taraxacum officinale* (L.) Weber ex F.H.Wigg.,^[124] *Scolymus hispanicus* L. and the cultivated *Carthamus*
 199 *tinctorius* L.; both used as saffron substitutes, as well as flowers of *Cynara humilis* L. and *Cynara cardunculus* L. used for making the local cheese
 200 varieties.^[129] It was demonstrated that edible flowers of Asteraceae contain different phenolic compounds, mainly phenolic acids, flavonols and anthocyanins,
 201 all exhibiting strong antioxidant effects.^[139,140] Edible flowers are not much used in the Balkans cuisine. Nevertheless, there are few notes on edible flowers
 202 used either raw, in the salads, as reported for *Bellis perennis* L., *Taraxacum officinale* (L.) Weber ex F.H.Wigg.^[118] and *Cichorium intybus* L.,^[141] or for
 203 seasoning, as in the case of *Achillea millefolium* L., *Artemisia absinthium* L., *Chamomilla recutita* (L.) Rauschert, *Eupatorium cannabinum* L.,^[118] *Calendula*
 204 *officinalis* L. and *Helianthus tuberosus* L.^[142] Finally, it should be added that the flowers of many Asteraceae species are readily used in the Balkans for
 205 preparing herbal teas, which are used not only as a remedy in prophylaxis, but often as a hot beverage. The most used species in the herbal teas are:
 206 *Chamomilla recutita* (L.) Rauschert, *Achillea millefolium* L., *Calendula officinalis* L., *Taraxacum officinale* (L.) Weber ex F.H.Wigg., *Cichorium intybus* L.,
 207 *Matricaria suaveolens* Koch, *Xanthium spinosum* L., *Cirsium vulgare* (Savi) Ten., *Tussilago farfara* L., *Artemisia absinthium* L., *Arnica montana* L.,
 208 *Centaurea jacea* L., *Helichrysum italicum* (Roth) G. Don, *Helichrysum plicatum* DC., and some others.^[115–117,143–145] Instead of a conclusion it is worth to
 209 mention some less common uses of wild Asteraceae in the Balkans, such as for preparation of sweet syrup, called “dandelion’s honey” as in the case of
 210 *Taraxacum*,^[146] and *Arctium* species^[147]; use of chicory and dandelion roots as coffee surrogate^[143] or use of aerial parts of *Arctium lappa* L., *Artemisia*
 211 *absinthium* L., *Cyanus segetum* Hill, *Tanacetum vulgare* L. and *Taraxacum* spp. for fermented beverages, such as wine and beer.^[145] Finally, apart from the
 212 vegetative plant parts and flowers, rarely, some other plant parts of Asteraceae species might be used, such as cypselas or pollen, as in the case of sunflower,

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213 where its pollen was assigned to have much higher flavonoid content and better nutritive features than many other commonly used crops which makes it a
 214 possible functional food ingredient.^[22,148]

215 **Table 3.** Balkans' Asteraceae plants as food source.

Species	Plant part used	Preparation	Country/Sub-region	Reference
<i>Achillea millefolium</i> aggr.	Young shoots	Green vegetable, condiment	BiH ^[a]	[149,150]
	Flowers	Seasoning	Mkd	[118]
<i>Arctium</i> L. spp.	Young shoots	Salad, soup	WB	[147]
	Root	Sweet syrup, flavor, cooked	WB	
		Vegetable, coffee supplement	WB	
<i>Arctium lappa</i> L.	Leaves	Salad	Bul	[144]
	Root	Flour, soup, baked vegetable		
<i>Arctium nemorosum</i> Lej.	Root	Mush	BiH	[149]
	Young leaves	Green vegetable		
<i>Artemisia absinthium</i> L.	Flowers	Seasoning	Mkd	[118]
<i>Artemisia vulgaris</i> L.,	Young shoot	Cooked vegetable, condiment	BiH	[149,150]
<i>Bellis perennis</i> L.	Leaf	Green vegetable	Tur	[120]
		Salad	Bul	[144]
	Young shoot	Salad, green vegetable	WB	[147]
	Flowers	Raw	Mkd	[118]
<i>Bellis sylvestris</i> Cirillo	Leaves	Salad	BiH	[151]
<i>Calendula arvensis</i> (Vaill.) L.	Leaves	Cooked vegetable	Grc	[126]
<i>Calendula officinalis</i> L.	Flowers	Seasoning, decoration, cooked	Bul	[142]
<i>Carduus</i> L. spp.	Young shoots	Salad	Cro	[152]
<i>Carduus acanthoides</i> L.	Young shoots	Salad	WB	[153]
	Root	Cooked vegetable		
<i>Carduus argentatus</i> L.	Young stem	Cooked vegetable	Grc	[154]
<i>Carthamus lanatus</i> L.	Young leaves	Cooked vegetable	WB-Med	[153]
<i>Carlina acaulis</i> L.	Root	Fresh, salad	BiH	[149,150]
		Beverage	BiH	
		Cooked vegetable	Srb	[143]
<i>Carlina involucreta</i> Poir.	Young stems	Cooked vegetable	Grc	[154]
<i>Centaurea calcitrapa</i> L.	Young stems	Cooked vegetable	Grc	[154]
<i>Centaurea jacea</i> L.	Leaves	Cooked vegetable	BiH	[151]
<i>Centaurea hyalolepis</i> Boiss.	Young stems	Cooked vegetable	Grc	[154]
<i>Centaurea scabiosa</i> L.	Young leaves	Green vegetable	WB	[153]
<i>Chondrilla juncea</i> L.	Root	Cooked vegetable	WB	[153]
<i>Cichorium intybus</i> L.	Leaves	Salad, green vegetable	Cro	[127]
			Grc	[126]
		Sweets, soups, green vegetable	Mne	[128]
		Soup, salad, green vegetable	WB	[147]
	Cooked vegetable, coffee surrogate	BiH	[149]	
	Root	Coffee surrogate	Srb	[143]
	Flowers	Raw	Srb	[141]
<i>Cichorium pumilium</i> Jacq.	Leaves	Green vegetable	Grc	[126]
<i>Cicerbita alpina</i> (L.) Wallr.	Young shoots	Cooked vegetable	BiH	[149]
(Syn. <i>Lactuca alpina</i> (L.) A. Gray)	Leaves	Salad	WB	[153]
<i>Cirsium arvense</i> (L.) Scop.	Leaves	Salad, green vegetable	Cro	[127]
<i>Cirsium oleraceum</i> (L.) Scop.	Young shoots	Cooked vegetable Salad	BiH	[151]
			WB	[153]
<i>Cirsium lanceolatum</i> (L.) Scop.	Young shoots	Cooked vegetable	WB	[153]
(Syn. <i>Cirsium vulgare</i> (Savi) Ten.)				
<i>Chamomilla recutita</i> (L.) Rausch.	Flowers	Seasoning	Mkd	[118]
(Syn. <i>Matricaria chamomilla</i> L.)				
<i>Crepis biennis</i> L.	Leaves	Salad, green vegetable	Cro	[127]

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		Salad, cooked and baked vegetable	BiH	[150]
		Coffee surrogate		
	Root		BiH	[150]
<i>Crepis capillaris</i> (L.) Wallr.	Leaves	Salad, green vegetable	BiH	[150]
	Root	Coffee surrogate	BiH	[150]
<i>Crepis sancta</i> (L.) Bomm.	Leaves	Salad, green vegetable	Cro	[127]
<i>Crepis zacintha</i> (L.) Loisel.	Leaves	Salad, green vegetable	Cro	[114]
<i>Cynara cardunculus</i> L.	Young stems	Cooked vegetable	Grc	[154]
<i>Cynara cornigera</i> Lindl.	Young shoots	Cooked and baked vegetable	Grc	[126]
<i>Cynara scolymus</i> L.	Young stems	Cooked vegetable	Grc	[154]
			WB-Med	[153]
<i>Echinops spinosissimus</i> Turra	Young stems	Cooked vegetable	Grc	[154]
<i>Echinops ritro</i> L.	Young leaves	Cooked vegetable	WB-Med	[153]
<i>Eupatorium cannabinum</i> L.	Root	Salad	Mkd	[118]
	Flowers	Seasoning	Mkd	[118]
<i>Gundelia tournefortii</i> L.	Young stems	Cooked vegetable	Grc	[154]
<i>Inula helenium</i> L.	Root	Mush, bread	BiH	[151]
<i>Helianthus tuberosus</i> L.	Tubers	Cooked vegetable	Srb	[143]
		Consumed raw as snack	Srb	[117]
	Flowers	Seasoning, decoration, cooked	Bul	[142]
<i>Hypochoeris radicata</i> L.	Leaves	Green vegetable	WB	[153]
<i>Hypochoeris maculata</i> L.	Leaves	Green vegetable	WB	[153]
<i>Hyoseris scabra</i> L.	Leaves	Salad	WB	[153]
<i>Lactuca seriola</i> L.	Leaves	Salad, green vegetable	Cro	[152]
<i>Lactuca perennis</i> L.	Leaves	Salad, green vegetable	Cro	[127]
<i>Lapsana communis</i> L.	Leaves	Salad, green vegetable	Srb	[143]
			Bul	[144]
<i>Leucanthemum vulgare</i> Lam.	Young shoots, leaves	Salad, soups, green vegetable	Balkan	[147]
<i>Leontodon autumnalis</i> L.	Leaves	Salad, cooked vegetable	BiH	[150]
(Syn. <i>Scorzoneroides autumnalis</i> (L.) Moench)	Root	Coffee surrogate	BiH	[150]
<i>Leontodon hispidus</i> L.	Young leaves	Salad	Bul	[144]
<i>Leontodon taraxacoides</i> (Vill.) Mérat (Syn. <i>Leontodon saxatilis</i> Lam.)		Salad, green vegetable	Cro	[114]
<i>Leontodon tuberosum</i> L.	Leaves	Salad, green vegetable	Cro	[127]
	Shoots	Cooked vegetable	Grc	[126]
	Tubers	Cooked vegetable	WB	[147]
<i>Mycelis muralis</i> (L.) Dum. (Syn. <i>Lactuca muralis</i> (L.) Gaertn.)	Leaves	Salad	WB	[153]
<i>Notobasis syriaca</i> (L.) Cass	Young shoots	Cooked vegetable	Grc	[126]
<i>Onopordum acanthium</i> L.	Young shoot	Green vegetable	WB	[153]
	Root	Salad, cooked vegetable, beverage	BiH	[150]
<i>Onopordum bracteatum</i> Boiss & Heldr.	Young stems	Cooked vegetable	Grc	[154]
<i>Onopordum cyprium</i> Eig.	Young stems	Cooked vegetable	Grc	[154]
<i>Petasites albus</i> (L.) Gaertn.	Young leaves	Cooked vegetables	BiH	[149]
<i>Petasites hybridus</i> (L.) Gaertn. & al.	Young leaves	Cooked vegetable, Beverage, Substitute for tobacco	BiH	[150,151]
<i>Picnomon acarna</i> (L.) Cass.	Root	Cooked vegetable	WB-Med	[153]
<i>Picris echioides</i> L. (Syn. <i>Helminthotheca echioides</i> (L.) Holub)	Leaves	Salad, green vegetable	Cro	[152]
			Grc	[126]
<i>Picris hieracioides</i> L.	Young leaves	Cooked vegetable	WB	[153]
<i>Reichardia picroides</i> (L.) Roth	Leaves	Salad, green vegetable	Cro	[127]
			Grc	[126]
<i>Scolymus hispanicus</i> L.	Root	Cooked vegetable	WB-Med	[153]
	Young stems	Cooked vegetable	Grc	[154]

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	Rhizome	Cooked vegetable	Grc	[126]
<i>Scolymus maculatus</i> L.	Young stems	Cooked vegetable	Grc	[154]
<i>Scorzonera laciniata</i> L. (Syn. <i>Podospermum laciniatum</i> (L.) DC.)	Leaves	Salad, green vegetable	Cro	[114]
<i>Scorzonera hispanica</i> L.	Root	Cooked vegetable, salad	WB	[153]
<i>Scorzonera villosa</i> Scop.	Leaves	Salad, green vegetable	Cro	[114]
<i>Scorzonera rosea</i> L.	Leaves	Salad	WB	[147]
(Syn. <i>Podospermum roseum</i> (Waldst. & Kit.) Gemeinholzer & Greuter)	Root	Cooked vegetable	WB	[151]
<i>Serratula tinctoria</i> L.	Young shoot	Green vegetable	WB	[153]
<i>Sonchus asper</i> (L.) Hill	Leaves	Salad, green vegetable	Cro	[152]
			Grc	[126]
<i>Sonchus arvensis</i> L.	Leaves	Salad	Bul	[144]
<i>Sonchus oleraceus</i> L.	Leaves	Salad, green vegetable	WB	[147]
			Grc	[126]
	Shoot	Cooked vegetable	BiH	[151]
<i>Silybum marianum</i> (L.) Gaertn.	Young leaves, involucrum	Cooked vegetable	WB-Med	[153]
	Young stems	Cooked vegetable	Grc	[154]
<i>Tagetes erecta</i> L.	Flowers	Seasoning, decoration	Bul	[142]
<i>Taraxacum</i> F.H. Wigg spp.	Leaves	Salad, stew	Bul	[144]
	Root	Salad, cooked vegetable	WB	[147]
	Flowers	Raw	Alb	[116]
			Mkd	[118]
<i>Taraxacum campylodes</i> G.E. Haglund;	Flowers	Syrup, "honey"	Rou	[146]
<i>Taraxacum cyprium</i> H. Lindb.	Leaves	Salad	Grc	[154]
<i>Taraxacum megalorrhizon</i> Hand.- Mazz.	Leaves	Salad, green vegetable	Cro	[127]
			Srb	[143]
	Root	Coffee surrogate	Srb	[143]
<i>Taraxacum officinale</i> Weber	Leaves	Salad, green vegetable	WB	[147]
(Syn. <i>Taraxacum</i> sect. <i>Taraxacum</i> F. H. Wigg)	Flowers	Syrup, "honey"	Srb	[141]
	Root	Sweets, coffee	BiH	[149]
<i>Telekia speciosa</i> (Schreb.) Baumg.	Young shoots	Cooked vegetable	BiH	[149]
<i>Tragopogon orientalis</i> L. (Syn. <i>Tragopogon pratensis</i> subsp. <i>orientalis</i> (L.) Čelak.)	Leaves	Salad	BiH	[151]
<i>Tragopogon pratensis</i> L.	Leaves	Salad, green vegetable	WB	[147]
	Root	Cooked vegetable	WB	
	Shoot	Green vegetable	Tur	[120]
<i>Tragopogon sinuatus</i> Avi.-Lall. (Syn. <i>Tragopogon porrifolius</i> L.)	Shoots	Cooked vegetable	Grc	[126]
<i>Tussilago farfara</i> L.	Shoot	Cooked vegetable	Tur	[120]
	Leaves	Cooked vegetable instead cabbage	BiH	
				[150]
<i>Urospermum picroides</i> (L.) Scop. ex F.W.Schmidt	Leaves	Salad, green vegetable	Cro	[127]

216 ^[a] Alb- Albania; BiH- Bosnia and Herzegovina; Bul- Bulgaria; Cro- Croatia; Grc- Greece; Mkd- North Macedonia; Med- Mediterranean; Mne- Montenegro; Romania- Rou; Srb-
217 Serbia; Tur – Turkey; WB- Western Balkans.

218 Concluding remarks

219 Indigenous species of Asteraceae family are valuable sources of bioactive plant compounds, referring to components of essential oils, sesquiterpene lactones,
220 different flavonoids, anthocyanins and other phenolic compounds, as well as complex carbohydrates such as inulin. Southeast Europe, i.e. the Balkan
221 Peninsula is known for traditional uses of plants, both in ethnomedicine and as foods. Very high species diversity also reflects on high variability of
222 specialized metabolites and complex phytochemical profiles which should be further studied using metabolomic and nutrigenomic approaches. High rate of
223 endemism and related high number of endemic species of the Asteraceae family in the Balkans, represent a starting point for future research on highly
224 efficient bioactive compounds and novel phytochemicals to be used as pharmaceuticals or nutraceuticals and components of functional food. Nevertheless,

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225 the final message should address the necessity for conservation of such valuable bioresources and further research of their *ex situ* conservation, *in vitro*
226 propagation and sustainable exploitation.

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Acknowledgements

This study was supported by the Ministry of Education, Science and Technological Development of the Republic Serbia through Project Nos. TR31069 and 451-03-68/2020-14/200178. Authors would like to thank to Miloš Dobrijević for technical support during graphical abstract preparation and Nebojša Banjac for technical support during preparation of chemical formulas.

Author Contribution Statement

A.Ž.K., P.J. and Z.P.D.S. conceptualized Manuscript idea. All authors have conducted literature search and participated in writing of the first Manuscript draft. S.M.K. and A. Ž.K. prepared graphical abstract. A.Ž.K., P.J. and Z.P.D.S. have reviewed and finalized Manuscript. All authors approved final version of Manuscript.

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Entry for the Graphical Illustration

