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Essential oil composition, antioxidant and antimicrobial activity of the galbuli of six juniper species

Valtcho D. Zheljazkov[a,*], Miroslava Kacaniova[b,c], Ivayla Dincheva[d], Tzenka Radoukova[e], Ivanka B. Semerdjieva[f], Tess Astatkie[g], Vicki Schlegel[h]

[a] Crop and Soil Science Department, Oregon State University, 3050 SW Campus Way, 109 Crop Science Building, Corvallis, OR 97331, USA
[b] Department of Microbiology, Faculty of Biotechnology and Food Sciences, Tr. A. Hlinku 2, Slovak University of Agriculture in Nitra, 949 76 Nitra, Slovak Republic
[c] Department of Bioenergetics and Food Analysis, Faculty of Biology and Agriculture, University of Rzeszow, Zelwerowicza St. 4, 35-601 Rzeszow, Poland
[d] Plant Genetic Research Group, AgroBioInstitute, Agricultural Academy, 8 Dragan Tsankov Blvd., 1164, Sofia, Bulgaria
[e] Department of Botany and Methods of Biology Teaching, Faculty of Biology, University of Plovdiv Paisii Hilendarski, 24 Tzar Assen, 4000 Plovdiv, Bulgaria
[f] Department of Botany, Agricultural University, Mendleev 12, 4000 Plovdiv, Bulgaria
[g] Department of Engineering, Faculty of Agriculture, Dalhousie University, PO Box 550, Truro, NS B2N 5E3, Canada
[h] Food Science and Technology Department, University of Nebraska-Lincoln, Food Innovation Center, Lincoln, NE 68588, USA

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ABSTRACT

To respond to the market demand for consistency of supply and quality, there are current efforts to develop junipers as agricultural crop for the production of juniper galbuli (berries). However, comparative data on the galbuli essential oil (EO) composition and bioactivity of different juniper species is limited. The objective of this study was to evaluate the EO composition, antimicrobial and antioxidant activity of the galbuli of the six juniper species naturally distributed in Bulgaria: *Juniperus communis* L. (common juniper), *J. oxycedrus* L. (red juniper), *J. sibirica* Burgsd. (Siberian juniper), *J. sabina* L. (Cossack juniper), *J. pygmaea* C. Koch. (Alpine juniper), *and J. excelsa* M. Bieb. (Gracian juniper). The EO content of the galbuli of the six juniper species varied from 0.47% (in *J. sibirica*) up to 1.6% (in *J. sabina*). The oil profile of the galbuli was also different among species. Differences and similarities in the groups of terpenes were established between the six juniper species. The three oil constituents with the highest concentration (in descending order) in the galbuli of each of the species were as follows: for *J. oxycedrus:* β-Myrcene, α-Pinene, and Germacrene D; for *J. communis:* α-Pinene, Germacrene D, β-Myrcene; for *J. excelsa:* α-Pinene, α-Cedrol, Germacrene D; for *J. sibirica:* α-Pinene, β-Myrcene, Germacrene D; for *J. pygmaea:* α-Pinene, Sabinene, β-Myrcene; and for *J. sabina:* Sabinene, α-Pinene, Terpinene-4-ol. Overall, the antioxidant capacity of the six oils was as follows: *J. sibirica* > *J. communis* = *J. excelsa*. Out of the six EO, *J. oxycedrus* galbuli EO was the most effective against *Clostridium perfringens, J. communis* against *Candida clabrata* and *J. oxycedrus* against *Staphylococcus aureus*. The antioxidant capacity of the juniper galbuli oils were positively affected by the concentrations of β-Elemene, γ-Elemene, and τ-Muurolol. Positive relationships were found between the concentrations of some constituents with the antimicrobial activity of juniper EO against *Staphylococcus aureus*, *Bacillus cereus, Yersinia enterocolitica, Clostridium perfringens,* and *Candida glabrata*. The results contribute to further understanding of juniper galbuli EO, can be used by industry utilizing juniper EOs, and may help with policy making processes with respect to conservation and agriculture.

1. Introduction

The geographic location of Bulgaria, which is betweenmoderate and subtropical geographic belts, and its rich geological history are prerequisites for biodiversity and the abundance of plant species. The variable climatic feature of the country fosters the spreading of plant species with great ecological plasticity and various life forms. Species of the genus *Juniperus* L. are characterized with high ecological adaptability, life span, and have a significant role in the composition of the flora in the censos (Bekele and Hudnall, 2005; Hantemirov et al., 2000; Evangelista et al., 2004; Wesche and Ronnenberg, 2004). The genus *Juniperus* comprises around 68 species and 36 different varieties (Adams, 2014), that are widely distributed in the northern hemisphere (Adams, 2011). Junipers inhabit areas from sea level to high-

* Corresponding author.
E-mail addresses: Valtcho.jeliazkov@oregonstate.edu, Valtcho.pubs@gmail.com (V.D. Zheljazkov).

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mountainside treeless areas (Adams, 2008; Mao et al., 2010). The juniper species are slow growing evergreens, and have the ability to grow on sandy or infertile rocky soils. This is one of the reasons for their use as pioneers performing mechanical protection of the soil, as well as for providing conditions for the development of economically important tree species (Čolić, 1957).

Of the 68 species found worldwide, 10 species are found in Europe (Franco, 1964), and six are distributed in the Bulgarian flora; *J. excelsa* M. Bieb. (forest or Grecian juniper) *J. communis* L. (common juniper), *J. oxycedrus* L. (cade or red juniper), *J. sibirica* Burgsd. (Siberian juniper), *J. sabina* L. (savin or Cossack juniper), *J. pygmaea* C. Koch. (Alpine juniper) (Yordanov et al., 1963). The six species of the genus Jiniperus that are naturally distributed in Bulgaria belong to two taxonomic sections; Section *Juniperus* (*J. oxycedrus, J. communis, J. pygmaea, J. sibirica*) and Section *Sabina* (*J. sabina* L. and *J. excelsa*) (Adams, 1998). Representatives are typical nanofanerophytes (NPh), some of which (*J. oxycedrus*, *J. communis*) under favorable conditions pass into phanerophytes (Ph) (Raunkiaer, 1934). With the greatest ecological plasticity is characterized *J. communis*, which grows from 200 to 1700 m asl, and is also the most commonly found juniper species in the world and in Bulgaria (Adams, 2014; Assyov et al., 2012). *J. oxycedrus* L. generally prevails in the southern parts of the country, from 0 to 1100 m asl. The other three junipers *J. pygmaea*, *J. sibirica* and *J. sabina* are found mainly in the high mountain areas of Bulgaria (Stara Planina, Vitosha, Rila, Pirin) at elevations between 1300 and 1600 m asl. The only homonymous representative of the genus in Bulgaria, phanerophyte (Ph), is *J. excelsa*, distributed in the southern part of Bulgaria, in the conditions of the trans-Mediterranean climate. Fully developed galbuli of *J. communis* L., *J. pygmaea, J. sibirica, J. sabina* L. and *J. excelsa* are spherical, bluish with more or less pronounced greyish scale, whereas the mature galbuli of *J. oxycedrus* are larger, reddish brown, and usually without blemishes (Fig. 1).

Although *J. sabina* is widely used as ornamental and therefore could be seen frequently in towns, its natural habitats are rare and found in rocky outcrops in the mountains, and frequently at inaccessible parts of the high mountains (Fig. 2). Therefore, *J. sabina* and the habitats of *J. excelsa* are listed in the Bulgarian Red Book categories as endangered and critically endangered, respectively (Stoeva, 2015); and they are listed in Appendix № 2 of the biodiversity of Bulgaria (State Gazette, 2002). The specific composition and structure of the habitats in which these two species are included, as well as some of the other species of the genus Juniperus, determines their inclusion in Annex I to Directive 92/43 / EEC Natura 2000 (Official Journal of the European Union (OJEU, 1992), as well as in the Law on the Biodiversity of Bulgaria (State Gazette, 2002).

Junipers are also used as medicinal plants by various cultures around the world, including Mediterranean (Pieroni et al., 2006), and Native Americans (Lim, 2012). Juniper wood is very durable and valued for its color, aroma, and antimicrobial properties (Adams, 1991; Clark et al., 1990). Generally, junipers are relatively slow growing trees and generally, have relatively low economic importance as lumber. Junipers thrive on infertile soils and move to places not favored by other trees. Therefore, some countries such as Bulgaria and United States championed programs for juniper eradication from pasture lands, (e.g. in Oregon, United States of America, http://www.oregon.gov/OWEB/MONITOR/pages/monitor_juniper.aspx).

Junipers galbuli (berries, female cones), leaves (needles), and wood contain significant amount of essential oil (EO), consisting mostly of oxygenated monoterpenes and sesquiterpene hydrocarbons (Pavićević et al., 2016). Therefore, there are 3 types of juniper oil; juniper wood, juniper leaf, and juniper galbuli oil, and surely, mixes in different ratios of the above three. These three types of oil may have differential composition and bioactivity. Juniper oils are utilized in aromatherapy, perfumery, and alcoholic beverages such as gin and Slovakian

Fig. 1. Galbuli of the six *Juniperus* species.