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Source / Izvornik: **Journal of Food Composition and Analysis**, 2015, 44, 128 - 138

Journal article, Accepted version

Rad u časopisu, Završna verzija rukopisa prihvaćena za objavljivanje (postprint)

<https://doi.org/10.1016/j.jfca.2015.08.008>

Permanent link / Trajna poveznica: <https://urn.nsk.hr/urn:nbn:hr:184:529968>

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Download date / Datum preuzimanja: **2024-08-10**



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Chemical markers for the authentication of unifloral *Salvia officinalis* L. honey

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Highlights

- ▶ Chemical characterization of unifloral *Salvia officinalis* L. honey
- ▶ Determination of polyphenolics, carbohydrates and minerals
- ▶ Chemical markers for the authentication of *Salvia officinalis* honey

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1 **ABSTRACT**

2 The objective of the present study was to trace phytochemicals that characterize unifloral
3 Common sage (*Salvia officinalis* L.) honey originating from the Croatian North Adriatic
4 coast. The polyphenolic profiles and total phenolic contents (TPC), the compositions of
5 minerals, sugars and sugar alcohols, and the radical scavenging activities (RSA) of 18
6 unifloral *S. officinalis* honey samples were investigated. The quantitative data on the targeted
7 compounds (25 phenolic compounds, 14 carbohydrates and 25 minerals) together with the
8 TPC and RSA data served as a pool of variables for multivariate analysis, which provided
9 useful information for the accurate authentication of unifloral sage honey and its
10 discrimination from other unifloral types of honey. The proposed markers, together with
11 chemometrics, could further contribute, as a powerful tool, to the quality control of Croatian
12 unifloral *S. officinalis* honey and thus, possibly certify its commercial value.

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16 **Keywords:** Unifloral honey, *Salvia officinalis* L., Chemical markers, Polyphenolics, Sugars,
17 Sugar alcohols, Minerals, Food analysis, Food composition

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19 1. Introduction

20

21 Common sage (sometimes called Great sage or Dalmatian sage, Latin *Salvia*
22 *officinalis* L.) is a circum-Mediterranean nectariferous botanical species common to the
23 Eastern Adriatic and Ionian seas (Ricciardelli D'Albore and Galarini, 2000) with a habitat
24 reaching south into northwest Greece (Karousou et al., 2000). This ~~spontaneous~~ perennial
25 Mediterranean shrub (belonging to the family *Lamiaceae*), widespread in the Mediterranean
26 part of Croatia, spontaneously grows on the hillsides of the North Croatian Littoral and
27 Dalmatian islands as well ~~in~~on the adjacent coastal belt (800–5000 m wide) and has
28 significant beekeeping importance (Flora Croatica Database, 2012). The North–East part of
29 the Adriatic Littoral (North Croatian Littoral) is especially characterized by an abundance of
30 sage-dominated botanical communities. Actually, they sometimes cover areas of several
31 square kilometers, representing practically ~~the~~ by far the most predominant plant of this poor,
32 rocky terrain of the karst region (Šugar et al., 1983). Through the ~~centennial~~ tradition of
33 beekeeping, this area has become well known for its famed unifloral *S. officinalis* honey that
34 has seen widespread use in traditional medicine for the treatment of respiratory problems, as
35 an antiseptic, *etc.* The potential health effects of this unifloral honey are usually ascribed to
36 its phytochemical constituents, which mostly originate from *S. officinalis* nectar (Kenjerić et
37 al., 2008).

38 The objective of the present study was to determine useful chemical markers for the
39 authentication of unifloral *S. officinalis* honey, based on the analysis of the polyphenolic
40 profiles, minerals, sugars and sugar alcohols in 18 honey samples originating from the
41 ~~North~~north–East Adriatic region of Croatia. The phytochemical profiles of the studied
42 honey samples were analyzed by high resolution LC/MS techniques. Quantification of major
43 phenolic compounds was achieved using ~~ultra~~ultra-high-performance liquid chromatography

44 coupled with a diode array detector and a triple quadrupole mass spectrometer (UHPLC DAD–
45 MS/MS). In order to trace the phytochemicals that characterize sage honeys produced in the
46 North Croatian Littoral, this work was focused on the identification of target compounds
47 using ~~ultra-ultra~~-high-performance liquid chromatography coupled with hybrid mass
48 spectrometry, which combined a Linear Trap Quadrupole and OrbiTrap mass analyzer
49 (UHPLC–LTQ OrbiTrap MS). This technique has already proven itself to be reliable for the
50 unambiguous detection of phenolic acids and their derivatives, as well as of the flavonoids
51 aglycones and glycosides. The sugar content was determined using high-performance anion-
52 exchange chromatography with pulsed amperometric detection (HPAEC/PAD). The
53 characterization of ~~Common~~-common sage unifloral honey was further supported by the
54 evaluation of the mineral composition using inductively coupled plasma-atomic emission
55 spectroscopy (ICP-OES) and melissopalynological analysis.

56

57 2. Experimental

58

59 2.1. Sage honey sampling and the authenticity of the samples

60 Representative honey sampling was performed directly at the filling facilities of the
61 primary producer. After samplingcollection, samples were placed into a-glass jars sealed with
62 the-metal lids and kept at temperature of $+4\text{ }^{\circ}\text{C}$ to $+8\text{ }^{\circ}\text{C}$ until analyzed. In order to attain
63 confirmation of the botanical origin of the *S. officinalis* honeys, the samples were subjected to
64 thorough melissopalynological and sensory assessment. Melissopalynological analysis,
65 considered as an analytical tool essential for the verification of the botanical and geographical
66 origin of a honey, was realized according to the method described by Loveaux et al. (1978)
67 and further elaborated by Von der Ohe et al. (2004).

68 The extent to which a honey sample corresponds to a given plant source is determined
69 from the frequencies of the pollen and honeydew elements in it. Since sage pollen is under-
70 represented, and the percentage of sage pollen in the sediment is lower than the percentage of
71 the corresponding nectar in the honey (Ricciardelli D'Albore and Galarini, 2000), the
72 melissopalynological assessment was based on the expression of the pollen representativity
73 within pollen frequency classes: "predominant pollen" (more than 45-% of the pollen grains);
74 "secondary pollen" (16–45 %); "important minor pollen" (3–15 %); "minor pollen" (less than
75 3 %), as well as on the presence of honeydew elements (Loveaux et al., 1978; Von der Ohe et
76 al., 2004). Sensory assessment, as an equally important analytical mechanism for the
77 determination of the unifloral character of a honey (Piana et al., 2004), comprehensive
78 distinctive organoleptic features (visual, taste, odour, tactile) of the samples were determined
79 taking into consideration the extent of their compliance with the organoleptic profile of
80 unifloral sage honey (Lušić et al., 2007).

81

82 2.2. Reagents and standards

83 Acetonitrile and formic acid (both MS grade), methanol (HPLC grade), Folin–
84 Ciocalteu reagent, sodium carbonate, hydrogen peroxide, and hydrochloric and nitric acid
85 were purchased from Merck (Darmstadt, Germany). Trolox (6-hydroxy-2,5,7,8-
86 tetramethylchroman-2-carboxylic acid) was purchased from Sigma Aldrich (Steinheim,
87 Germany). 2,2-Diphenyl-1-picrylhydrazyl (DPPH·) was purchased from Fluka AG (Buchs,
88 Switzerland). The Strata C18–E (500 mg/3mL) SPE cartridges used for the extraction and
89 concentration of samples were obtained from Phenomenex (ThermoFisher
90 Scientific, Torrance, CA). Ultra-pure water (ThermoFisher TKA MicroPure water purification
91 system, 0.055 $\mu\text{S}/\text{cm}$) was used to prepare the standard solutions and blanks. Syringe filters
92 (13 mm, PTFE membrane 0.45 μm) were purchased from Supelco (Bellefonte, PA, USA).

93 *cis, trans*-Abscisic acid and polyphenolic standards were purchased from Fluka AG
94 (Buch~~s~~, Switzerland). Sugar standards were purchased from Tokyo Chemical Industry
95 (~~Zwijndrecht~~TCI, Europe, Belgium) and sugar alcohol standards were obtained from Sigma
96 ~~Sigma~~-Aldrich (Steinheim, Germany).

97

98 2.3. Preparation of standard solutions

99 A 1000 mg/L stock solution of a mixture of all phenolic standards and *cis, trans*-
100 abscisic acid was prepared in methanol. Dilution of the stock solution with methanol yielded
101 the working solutions of concentrations 0.025, 0.050, 0.100, 0.250, 0.500, 0.750, and 1.000
102 mg/L. Calibration curves were obtained by plotting the peak areas of the standards against
103 ~~their concentration~~ Calibration curves were obtained by plotting the peak areas of the
104 ~~compounds identified relative to the peak area against the concentration of the standard~~
105 ~~solution~~. Calibration curves revealed good linearity, with R^2 values exceeding 0.99 (peak
106 areas vs. concentration).

107 The evaluation of the carbohydrate content of the honey samples was obtained from
108 calibration curves of pure compounds. The calibration was performed with standard solutions
109 of sugars and sugar alcohols dissolved in ultrapure water. Each individual standard was
110 dissolved in ultrapure water. Stock solutions with concentrations of 1000 mg/L were prepared
111 and working solutions in the concentration ranges were as follows: for glucose and fructose
112 from 10.0 to 100.0 mg/L; for sucrose from 1.0 to 10.0 mg/L; for isomaltose from 0.5 to 5.0
113 mg/L, while for all the other standards, the concentration range was from 0.1 to 1.0 mg/L.
114 ~~Under these chromatographic conditions, the last compound was detected after approximately~~
115 ~~25 min, and the analysis was ended at 30 min.~~

116 | To analyze the mineral composition of honey, a multi-element plasma standard
117 | solution 4, Specpure, containing 1 g dm^{-3} of each element was utilized for reference
118 | purposes.

119

120 | 2.4. LC–MS/MS analysis

121 | 2.4.1. Preparation of sample extracts

122 | The method previously described by Gasic et al. (2014) was used for extraction and
123 | isolation of phenolics from the honey samples. Prior to UHPLC–DAD MS/MS and UHPLC–
124 | MS/MS Orbitrap analysis, the extracts were filtered through a ~~0.45~~ $45\text{-}\mu\text{m}$ PTFE membrane
125 | filter.

126

127 | 2.4.2. UHPLC–MS/MS Orbitrap analysis of polyphenolic compounds

128 | Separation of the compounds of interest were was performed using a liquid
129 | chromatography system that consisted of a quaternary Accela 600 pump and an Accela
130 | Autosampler, connected to a linear ion trap–orbitrap hybrid mass spectrometer (LTQ
131 | OrbiTrap XL) with a heated–electrospray ionization probe, HESI-II (ThermoFisher
132 | Scientific, Bremen, Germany).

133 | A Synchronis C18 column ($100 \times 2.1 \text{ mm}$, $1.7 \mu\text{m}$ particle size) from Thermo Fisher
134 | Scientific was used as the analytical column for separation. The mobile phase consisted of
135 | ~~(A) water + 0.1% formic acid (A) and acetonitrile + 0.1% formic acid (B)water + 0.1 %~~
136 | ~~formic acid and (B) acetonitrile + 0.1 % formic acid.~~ A linear gradient program at a flow rate
137 | of 0.300 mL/min was used: $0.0\text{--}1.0 \text{ min}$ 5% B, $1.0\text{--}9.9 \text{ min}$ from 5% to 95% (B), $9.9\text{--}10$
138 | min from 95% to 5% (B), then 5% (B) for 3 min . The injection volume was $5 \mu\text{L}$ (Gasic et
139 | al., 2014).

140 The mass spectrometer was operated in the negative ion mode. The HESI-source
141 parameters were given previously (Gasic et al., 2014). ~~Xcalibur software (version 2.1) was~~
142 Xcalibur software 2.1 (Thermo Fisher, Bremen, Germany) was used for instrument control,
143 data acquisition and data analysis. The phenolics were identified according to the
144 corresponding spectral characteristics: mass spectra, accurate mass, characteristic
145 fragmentation, and characteristic retention time. Full scan analysis was employed to detect
146 the monoisotopic mass of unknown compounds, while the fragmentation pathway was
147 obtained by MS/MS. This exact mass search method was based on high resolution MS
148 analysis (Orbitrap), online database search (Patiny and Borel, 2013) and prediction of
149 MS/MS fragmentation using Mass Frontier 6.0 software (Thermo Fisher Scientific).

150

151 2.4.3 UHPLC–DAD MS/MS analysis of polyphenolic compounds

152 The separation, determination, and quantification of the components in the sage honey
153 samples were performed using a Dionex Ultimate 3000 UHPLC system equipped with a
154 diode array detector (DAD) that was connected to TSQ Quantum Access Max triple-
155 quadrupole mass spectrometer (ThermoFisher Scientific, Basel, Switzerland). The elution
156 was performed at 40 °C on a Synchronis C18 column. The mobile phase consisted of ~~(A)~~
157 water + 0.1 % formic acid (A) and acetonitrile (B) + 0.1 % formic acid, and (B)
158 acetonitrile, which were applied in the following gradient elution: 5 % B in the first 2.0 min,
159 2.0–12.0 min 5–95 % B, 12.0–12.2 min from 95 % to 5% B, and 5 % B until the 15th min.
160 The flow rate was set to ~~0.4 mL min⁻¹~~ 0.4 ml min⁻¹ and the detection wavelengths to ~~254–~~
161 and 280 nm ~~254 and 280 nm~~. The injection volume was 5 µL.

162 A TSQ Quantum Access Max triple-quadrupole mass spectrometer equipped with an
163 heated electrospray ionization (HESI) source was used with the vaporizer temperature kept at
164 200 °C, and the ion source settings as follows: spray voltage 5000 V, sheet gas (N₂) pressure

165 40 AU, ion sweep gas pressure 1 AU and auxiliary gas (N₂) pressure 8 AU, capillary
166 temperature 300 °C, and skimmer offset 0 V (Natic et al., 2015). The mass spectrometry data
167 were acquired in the negative ion mode, in the *m/z* range from 100 to 1000. Multiple mass
168 spectrometric scanning modes, including full scanning (FS), and product ion scanning (PIS),
169 were conducted for the qualitative analysis of the targeted compounds. The collision-induced
170 fragmentation experiments were performed using argon as the collision gas, and the collision
171 energy was varied depending on the compound (**Table S1**). The time-selected reaction
172 monitoring (tSRM) experiments for quantitative analysis were performed using two MS²
173 fragments for each compound that were previously defined as dominant in the PIS
174 experiments (**Table S1**).

175 Xcalibur software 2.2 (Thermo Fisher, Bremen, Germany) ~~Xcalibur software (version~~
176 ~~2.2)~~ was used for instrument control. The phenolics were identified by direct comparison
177 with commercial standards. The total amounts of each compound were evaluated by
178 calculation of the peak areas and are expressed as mg kg⁻¹.

180 2.5. Determination of TPC and RSA

181 The samples were prepared according to a previously described method (Gasic et al., 2014).
182 Each honey sample (5 g) was mixed with ultrapure water in a 50-mL volumetric flask. The
183 solution was then filtered through 0.45-μm PTFE membrane and analyzed for determination
184 of TPC and RSA. -The amount of total phenolics was determined according to the Folin-
185 Ciocalteu method, while the radical-scavenging activity of honey extracts was measured
186 using the DPPH· method (Gasic et al., 2014). The TPC and RSA values are expressed as
187 milligram gallic acid equivalents (mg GAE) perequivalents (GAE) per kilogram and
188 micromoles of Trolox equivalents (μmol TE) perequivalents (TE) per kg of honey sample,
189 respectively.

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2.6. HPAEC/PAD analysis of sugars and sugar alcohols

The honey samples were homogenized, weighed (between 0.2 and 0.3 g) and diluted 1000-fold with ultrapure water. The solutions were filtered and transferred to vials.

The sugar and sugar alcohol contents were determined by HPAEC/PAD. ~~high performance anion-exchange chromatography with pulse amperometric detection (HPAEC/PAD)~~. The honeys were analyzed on an ICS 3000 DP liquid chromatograph equipped with a quaternary gradient pump (Dionex, Sunnyvale, CA, USA). The carbohydrates were separated on a CarboPac[®] PA10 ~~pellicular~~ anion-exchange column (4 × 250 mm) at 30 °C. Each honey sample (25 µL) was injected with an ICS AS-DV 50 autosampler (Dionex, Sunnyvale, CA, USA). The carbohydrates were eluted with the flow rate set to 0.7 mL/min, using a gradient program constituted from 600 mM sodium hydroxide (eluent A), 500 mM sodium acetate (eluent B) and ultrapure water (eluent C). The gradient program was as follows: 0.0–20.0 min, 15 % A; 20.1–30.0 min, 20 % A; 0.0–5.0 min, 0 % B; 5.1–12.0 min, 2 % B; 12.1–20.0 min, 4 % B; 20.1–30.0 min, 20 % B, 0.0–5.0 min, 85 % C; 5.1–12.0 min, 83 % C; 12.1–20.0 min, 81 % C; 20.1–30.0 min, 60 % C. Under these chromatographic conditions, the last compound was detected after approximately 25 min, and the analysis was ended at 30 min. The total amounts of each sugar or sugar alcohol ~~was~~ were evaluated according to the method ~~previously~~ described in section 2.3.

2.7. ICP–EOS analysis of minerals in honey samples

To analyze the mineral composition of honey, about 0.6–0.7 g of fresh honey sample ~~was~~ were treated with 7 mL of 65 % HNO₃ and 1 mL of 35 % H₂O₂ in polytetrafluoroethylene (PTFE) vessels. A microwave closed digestion system (ETHOS 1₊;

215 Milestone, Bergamo, Italy) was used for the mineralization process. The final clear solution
216 was made up to 50 mL with ultrapure water. A blank was prepared in the same way.

217 All mineral elements in the digested solutions were determined using an ICP-OES
218 (iCAP 6500 Duo ICP, Thermo Scientific, UK) instrument. The results are expressed as mg of
219 mineralmetal per kg of honey.

220

221 2.8. Statistical analysis

222 Data of all measurements performed in triplicate are expressed as the mean \pm standard
223 deviation (*SD*). Statistical analyses were performed using the Analysis ToolPak from the
224 Microsoft Office Excel 2007 Professional. ~~Statistical analyses were performed with the~~
225 ~~program MS Excel (Microsoft Office 2007 Professional).~~ PCA was realized using the PLS-
226 Tool Box software package for MATLAB 7.12.0 (Eigenvector Research, Inc., Wenatchee,
227 WA, USA)~~MATLAB (Version 7.12.0)~~. All data were group-scaled prior to PCA. The
228 singular value decomposition algorithm (SVD) and a 0.95 confidence level for Q and T^2
229 Hotelling limits for outliers were chosen.

230

231 3. Results and discussion

232

233 3.1. Verification of the sage honey samples

234 A great deal of attention was given to the authenticity of the Croatian Common sage
235 honey samples, especially to their geographical and botanical origin (Persano Oddo and
236 Bogdanov, 2004). Representative honey sampling was realized directly at primary producers'
237 filling facilities, above all taking into ~~the~~ consideration two important criteria: A) that the
238 honey sample extraction occurred ~~el~~osely soon after the sage flowering period (May) when
239 sage flowers were the main bee source of nectar, and B) appropriate apiary locations for

240 sample production. That is to say, particular beehive sites were selected for collection of *S.*
241 *officinalis* honey samples in line with the field observations on the abundance of sage nectar.
242 Furthermore, cartographic data concerning the areas of predominate *Salvia officinalis* L.
243 growth were taken from the comprehensive ~~Vegetation-vegetation~~ maps of Croatia (Šugar et
244 al., 1983), confirming that the production beehives involved were situated deeply inside
245 within the sage-dominated vegetation zones.

246 As a general rule, honey is considered unifloral if it ~~was~~is produced mainly from one
247 plant species, and if the pollen of that particular species predominates. However, the pollen
248 grains of some flowers are under-represented (or over-represented) in unifloral honeys, *i.e.*,
249 the percentage of pollen in the sediment is lower (or higher) than the percentage of the
250 corresponding nectar in the honey_ (Persano Oddo and Bogdanov, 2004). Therefore, the
251 pollen spectrum of other nectariferous and non-nectariferous botanical species should
252 likewise be taken into the consideration, as well as the presence of honeydew elements
253 (Persano Oddo and Bogdanov, 2004; Piazza and Persano Oddo, 2004). The unifloral
254 character of all the sage honey samples in this study was confirmed by thorough
255 melissopalynological and sensory evaluation (**Table 1**). When compared to the representation
256 of other pollen sources in samples, under-representation of *S. officinalis* pollen grains was
257 noted in almost all the studied honey samples_ thereby confirming the natural hypopollenic
258 features of sage (Ricciardelli D'Albore and Galarini, 2000; Flora Croatica Database, 2012).
259 The ~~greatest~~highest portion of the identified pollen in the sage unifloral honey originated
260 from nectariferous species belonging to the families *Rhamnaceae*, *Sapindaceae* (genus *Acer*)
261 and *Fagaceae* (genus *Castanea*). Pollen sources of non-nectariferous producing plants were
262 mostly attributed to *Quercus* spp. (fam-ily *Fagaceae*) and species belonging to the families
263 *Graminaceae* and *Plantaginaceae* (*Plantago* spp.), all sharing the flowering period of sage as
264 well as their areal of distribution. This characteristic pollen profile and specific combination

265 could be considered a valuable indicator of the geographical origin of the sage unifloral
266 honey samples.

267 Sensory assessment, as an equally important analytical mechanism for the
268 determination of the unifloral character of honey (Piana et al., 2004) revealed distinctive
269 organoleptic features (visual, taste, odour, tactility) of the samples, taking into consideration
270 the extent of their compliance with the particular organoleptic profile of unifloral sage honey
271 (Lušić et al., 2007). Based on the results of the melissopalynological and sensory evaluations,
272 all the honey samples in the present study were confirmed to be sage honeys.

273

274 3.2. Phenolic profile of Croatian sage honey samples

275 Although the composition of honey highly depends on the floral source used to collect
276 the nectar, some other factors, including geographic origin, seasonal and environmental
277 factors, bee variety, as well as processing technologies, may also affect the composition of
278 the phenolic compounds in honey (Kaskonienė and Venskutonis, 2010). On the other hand,
279 unifloral honeys have almost never been made from 100 % monofloral nectar, since the
280 nectar from flowers of many various plants contributes to the production of every honey
281 (Persano Oddo and Bogdanov, 2004). Therefore, it was important to analyze a large number
282 of sage honey samples, in order to derive more general rules, and define which compounds
283 and/or groups of compounds mostly characterize the phenolic and sugar profiles, and thus the
284 uniqueness of this autochthonous honey. Phenolic compounds such as flavonoids (Kenjerić et
285 al., 2008), carbohydrates (Primorac et al., 2011), and volatile compounds (Jerković et al.,
286 2006), were previously suggested as possible markers for the determination of Common sage
287 unifloral honey.

288 As it was previously reported, sage leaf extracts contain a wide range of phenolic compounds
289 with the majority of the phenolic acids represented by caffeic acid derivatives and rosmarinic

290 | ~~acid being the dominant one~~ Sage leaf extracts, as hitherto reported, contain a wide range of
291 | ~~phenolic compounds. The majority of the phenolic acids were found to be caffeic acid~~
292 | ~~derivatives, with rosmarinic acid being the dominant one. The study of~~ According to
293 | ~~Generalić et al. (2011);~~ identified rosmarinic, syringic, gallic, *p*-coumaric, caffeic, and *trans*-
294 | ferulic acid as the principal phenolic acids of Common sage extracts. ~~the principal phenolic~~
295 | ~~acids of Common sage extracts are rosmarinic, syringic, gallic, *p*-coumaric, caffeic, and~~
296 | ~~*trans*-ferulic acid.~~ The relative content of rosmarinic acid in the extracts ranged from 94.54-%
297 | to 98.38-%, depending on the phenophase, while the contents of other acids were
298 | significantly lower (Generalić et al., 2011; ~~Generalić et al., 2012~~). Other studies also report
299 | the presence of vanillic acid, salvianolic acids K and I, and methyl rosmarinate in Common
300 | sage (Dragovic-Uzelac et al., 2012; Dent et al., 2013). Flavonoids of *S. officinalis* are mostly
301 | present as flavones (apigenin, luteolin and their corresponding 6-hydroxylated derivatives),
302 | flavone glucosides (6-hydroxyluteolin-7-glucoside, luteolin-7-glucuronide, luteolin-
303 | glucoside, luteolin-3'-glucuronide, apigenin-7-glucuronide and apigenin-7-glucoside),
304 | flavonols (mostly kaempferol and quercetin methyl ethers), and flavonol glucosides
305 | (quercetin-4'-glucoside, rutin), as reported by several authors (Generalić et al., 2011;
306 | Dragovic-Uzelac et al., 2012; Generalić et al., 2012). Stilbenes (*trans*-resveratrol, astringin,
307 | piceid) and catechins ((+)-catechin, (-)-epicatechin) are also present (Generalić et al., 2011;
308 | ~~Generalić et al., 2012~~). *Salvia officinalis* L. is reported to contain also phenolic diterpenes,
309 | including carnosol and carnosic acid (Lamien-Meda et al., 2010). Some of the phenolic
310 | compounds previously determined as constituents of sage leaf extracts were also found in
311 | unifloral *S. officinalis* honeys from this region (Kenjerić et al., 2008), including phenolic
312 | acids (caffeic, rosmarinic, gallic, *p*-coumaric, and ferulic acid), and flavonoids (flavones
313 | apigenin and luteolin, and their corresponding glycosides; flavonols quercetin and
314 | kaempferol and their derivatives (quercetin hexoside and rutin); stilbenes (resveratrol); and

315 catechins (catechin and epicatechin)). It should be borne in mind that previous studies
316 concerning the composition of the phenolics in indigenous Croatian Common sage honey
317 (Kenjerić et al., 2008) usually concentrated on targeted metabolomic analysis that included a
318 limited number of compounds, and that there ~~is~~ are a lack of literature data concerning the
319 complete polyphenolic profiles.

320 On the other hand, the present study gives insight into the profile of the phenolics of sage
321 unifloral honey using the non-targeted metabolomic approach, which resulted in the
322 identification of a significant number of phenolic compounds (**Table 2**). In the absence of
323 standards, the identification of flavonoid glycosides and other phenolics were based on the
324 search for the $[M-H]^-$ deprotonated molecule and its fragmentation using UHPLC–LTQ
325 OrbiTrap MS/MS. The exact mass search and the study of the fragmentation pathways
326 described in the literature enabled as much structural information as possible to be obtained.
327 In this way, it was possible to ~~individuate~~ identify 61 compounds (**Table 2**). The
328 chromatograms of the investigated Common sage honey samples showed similar profiles. A
329 selected base peak chromatogram of a representative sage honey extract (sample No. **SH2**) is
330 shown in **Fig. S1**.

331 Hydroxycinnamic acids, such as caffeic, rosmarinic, ferulic, chlorogenic, and *p*-
332 coumaric acid were detected in the sage honey samples analyzed in the present study. These
333 phenolic acids constituted a significant share ~~to~~ of the total phenolics content of the sage
334 honey samples. Generally, the presence of phenolic compounds in nectar is usually connected
335 with their protective role against microbial infestations (Heil, 2011). However, high
336 concentrations of these compounds could lead to ~~the~~ the nectar's toxic effect, and have a negative
337 influence on pollinators (Adler, 2000). Of the hydroxybenzoic acids, *p*-hydroxybenzoic acid,
338 vanillic, gentisic, and protocatechuic acid were previously reported in sage (Zgórka and
339 Głowniak, 2001), and confirmed in unifloral sage honey samples. All these phenolic acids are

340 considered as potential markers for the authentication of sage unifloral honeys and were
341 therefore included in the subsequent targeted quantitative analyses of the honey samples.

342 The majority of flavonoids in *S. officinalis* are flavones of apigenin and luteolin, and
343 their corresponding 6-hydroxylated derivatives (hispidulin and cirsimaritin), as well as the
344 dihydroflavone hesperetin (Brieskorn and Biechele, 1971; Cuvelier et al., 1996; Lu and Yeap
345 Foo, 2002; Kontogianni et al., 2013), and all of these compounds were evidenced in the
346 analyzed sage honey samples. Of the flavone- glucosides, luteolin and apigenin glycosides
347 are very common in analyzed sage honeys, and some of them were previously found in *S.*
348 *officinalis* (Masterova et al., 1989; Wang et al., 1998; Lu and Yeap Foo, 2000). Interestingly,
349 it is well known that the presence of 6-hydroxy- and 6-methoxy-flavone glycosides clearly
350 differentiates section *Salvia*, which includes *S. officinalis*, from other sections belonging to
351 the genus *Salvia* (Tomás-Barberán et al., 1988). Therefore, the presence of these compounds
352 in honey might be one of the indicators that the honey in question is really of sage floral
353 origin. Flavonols of sage are mostly those of kaempferol and quercetin methyl ethers (Lu and
354 Yeap Foo, 2002), and nectar–pollen derived flavonoids, such ~~are~~ as quercetin, kaempferol,
355 and hesperetin, have been identified in samples of Common sage honey. Of the flavonoids
356 previously identified in sage, stilbene resveratrol and catechins (catechin and epicatechin)
357 were also confirmed in the sage honey samples (Generalic et al., 2011). The following
358 derivatives of catechin and epicatechin were also recorded in the honey samples:
359 gallocatechin, epigallocatechin, gallocatechin gallate, and epigallocatechin gallate.

360 The phenolic diterpenes carnosol and carnosic acid, although present in sage
361 (Kontogianni et al., 2013), were not previously detected in unifloral sage honeys. In the
362 present study, these compounds were identified in ~~the~~ all honey samples, but in trace amounts
363 (**Table 2**).

364

365 3.3. Quantification of targeted phenolics in the honey samples

366 Solid-phase extraction (SPE) combined with ultra-high-performance liquid
367 chromatography with a diode array detector (DAD) and a triple-quadrupole mass
368 spectrometer was used to analyze the content of 25 targeted compounds in the *S. officinalis*
369 honey samples. Three basic criteria for the selection of chemical markers from the group of
370 phenolic compounds were applied: 1) putative sage nectar–pollen derived compounds
371 (phenolic acids and flavonoids); 2) propolis characteristic flavonoids and 3) abscisic acid.

372 Among the quantified compounds in Common sage honeys, some of phenolic acids,
373 *i.e.*, *p*-coumaric, *p*-hydroxybenzoic, and ferulic acid, were present in the highest amounts.
374 Interestingly, rosmarinic acid was present in relatively low amounts in the unifloral sage
375 honeys analyzed in the present study (**Table 3**). It is well known that phenolic acids of sage
376 are mostly based on caffeic acid building blocks (Lu and Yeap Foo, 2002), and that
377 rosmarinic acid is the major phenolic compound in sage leaves. Possible reasons for this
378 could be relatively low concentrations of this compound in the nectar. Gentisic acid was
379 detected only in three samples (SH8, SH15, and SH18). Of the nectar–pollen derived
380 flavonoids quantified herein, quercetin, kaempferol, and hesperetin were abundant and
381 present in significant amounts. Stilbene resveratrol was detected only in four of the sage
382 honey samples (SH2, SH5, SH16, and SH17). Catechins were abundant in the analyzed
383 honey samples, with gallic acid gallate and epigallocatechin gallate being quantified as the
384 dominant compounds from this group. The contents of catechin and epicatechin were low.

385 Pinocembrin, pinobanksin, pinostrobin, galangin, and chrysin are characteristic
386 flavonoids of propolis, and were determined in most of the previously analyzed European
387 honey samples (Tomás-Barberán et al., 2001; Kenjeric et al., 2008). The portion of propolis-
388 derived compounds in the unifloral sage honeys analyzed in the present study was significant,
389 but much less than in a previous study (Kenjeric et al., 2008), which reported a relatively

390 | high portion of galangin and chrysin (51.3-%) in the total identified flavonoids. The sage
391 | honey samples analyzed in the present study were characterized by the significant amounts of
392 | pinobaksin (0.21–2.35 mg/kg) and chrysin (0.06–1.98 mg/kg).

393 | The plant stress hormone abscisic acid (AbBA) is known to be present in floral
394 | nectars of some plants, and is transferred from the nectar to honey. This phytohormone is
395 | present in relatively high amounts in some European honeys (Tomás-Barberán et al., 2001;
396 | Truchado et al., 2008; Bertoncelj et al., 2011), including unifloral sage honey (Kenjeric et al.,
397 | 2008), and was also confirmed in the present study. The presence of abscisic acid in high
398 | amounts (0.26–3.99 mg/kg) is not surprising, since natural rocky habitat of sage is
399 | characterized by periods of drought seasons during the summer, which results in stress-
400 | induced responses in the plants (Bertoncelj et al., 2011).

401

402 | 3.4. Antioxidant activity of Common sage honeys

403 | Antioxidant capacity of *S. officinalis* honey samples was determined by the total
404 | phenolics content (TPC) and the radical scavenging activity (RSA). The results of these
405 | investigations are given in **Table 3**.

406 | The Common sage honey samples were characterized with TPC values ranging
407 | between ~~208.519 to and 747.549~~ mg of gallic acid equivalents (GAE) per kg of honey. The
408 | average content of total phenolics was in a good agreement with the values given in the
409 | literature for sage honeys from the same region (Piljac-Žegarac et al., 2009).

410 | The results of the determination of the RSA of sage honey samples ranged from
411 | ~~351.20 to 894.8275~~ micromoles of Trolox equivalents TE per kg of sample. To determine the
412 | relationship between the content of polyphenols and antioxidant activities of *S. officinalis*
413 | honey samples, the correlation between the TPC and the RSA values was calculated. The
414 | RSA showed a statistically significant ($r = 0.872$; $P < 0.0001$) and positive linear

415 correlation with the TPC ($RSA = 68.08 + 1.10 \times TPC$). A significant and positive linear
416 relationship between the antioxidant activity and total phenolic content of sage honey
417 samples indicated that phenolic compounds could be identified as the chemicals that
418 predominately contributed to the antioxidant activity, which is in accordance with previous
419 investigations ~~reported previously~~ (Piljac-Žegarac et al., 2009; Gasic et al., 2014).

420

421 3.5. Determination of the sugars and sugar alcohols

422 Fourteen different sugars and sugar alcohols were identified and quantified in the
423 analyzed unifloral sage honey samples using the HPAEC/PAD method. Quantification was
424 performed with available standards. The reducing sugars, fructose and glucose, were found to
425 be the major constituents of all the investigated samples (**Table 4**), which confirmed that all
426 honey samples were genuine honeys. In all the analyzed honeys, the value of the glucose plus
427 fructose amounts was around or higher than 60 g per 100 g, which is the value for all honey
428 types required by the European and FAO (Codex Alimentarius) standards (FAO/WHO, 2001;
429 The Council of the European Union, 2002). Another monosaccharide identified in the honeys
430 in relatively low amounts was arabinose.

431 All the sage honey samples had a sucrose content lower than 5 g per 100 g, which is
432 generally taken as the limit value for honeys allowed by European Union Honey Directive
433 (The Council of the European Union, 2002). Apart from sucrose, the other identified
434 disaccharides were trehalose, turanose, maltose and isomaltose. The trisaccharides
435 maltotriose and isomaltotriose were also evidenced. From the group of polyols (sugar
436 alcohols), erythritol, sorbitol, ~~galactitol~~galactitol, and glycerol were identified.

437 The ratio between some carbohydrates is another indicator that may be used to
438 ascertain honey authenticity. Thus, the ratios of fructose/glucose, maltose/isomaltose,
439 sucrose/turanose, and maltose/turanose, maltotriose/raffinose+erlose+melezitose were used

440 for the authentication of some unifloral honeys, and all these studies were reviewed by
441 Kaskonienė and Venskutonis (2010). The fructose/glucose (*FRU/GLU*) ratio in sage honeys,
442 which was recommended for the evaluation of honey granulation because glucose is less
443 ~~water-water-~~soluble than fructose, varied from 1.31 in sample **SH1** to 4.42 in sample **SH87**.
444 One more characteristic of the unifloral sage honeys analyzed in the present study was the
445 relatively low maltose/isomaltose (*MAL/iMAL*) ratio, which ranged from 0.9 (sample **SH11**)
446 to 2.41 (**SH5**).

447

448 3.6. Determination of minerals in *S. officinalis* honeys

449 The concentrations of minerals quantified in the studied sage honey samples are
450 presented in **Table 5**. The most abundant element in all samples was found to be potassium
451 (content ranging from 592 ± 1.68 to 215 ± 1.350 mg/kg), which agrees with other studies and
452 indicates that K is the most common element in honeys (Cantarelli et al., 2008), including
453 unifloral sage honeys (Bilandžić et al., 2014). Phosphorus, sulfur, and calcium were the next
454 most common elements, followed by magnesium and sodium. Among the micro-elements in
455 decreasing amounts, B, Zn, Fe, Mn, Cu, Se, and Ni were found, while Co, Cr, Li, and V were
456 found as trace elements. Therefore, the influence of botanical origin on the elemental
457 composition of the unifloral sage honey was evident for both elements essential for plant
458 growth (macronutrients), such as K, P, S, Ca, Mg and Na, and for micronutrients (trace
459 elements), such as B, Mn, Zn, Fe, *etc.* The essential elements are present in plants in
460 significantly higher amounts than the trace elements, and this observation was also true for
461 the honey samples. On the other hand, the possibility that the mineral composition of honey
462 samples also reflects the environmental and pedological conditions of the geographical
463 locality cannot be excluded (Terrab et al., 2004). Toxic elements (Al, As, Cd, Pb, and Sb) in

464 the tested samples were found in small amounts (allowable concentrations), which excludes
465 the existence of environmental contamination of the honeys.

466

467 3.7. Pearson's correlation analysis

468 Pearson's correlation analysis was performed to evaluate the associations between
469 variables in 18 sage unifloral honey samples (**Table S2**), in order to define some general rules
470 characteristic for unifloral sage honey. Both positive and negative Pearson's correlations
471 were observed between the contents of the different analyzed compounds in the unifloral sage
472 honeys. However, statistically significant correlations were observed in some cases as can be
473 seen from the Tables given in supplementary material (**Tables S2–S4**). High positive
474 correlations were found between propolis-derived compounds. Namely, correlations between
475 CaA and PNB, PNS, CHR, PNC, GLN were in the range from 0.691 to 0.886. Likewise,
476 correlations among PNB, PNS, CHR, PNC, and GLN were also characterized with high
477 positive coefficients (**Table S2**). Statistically significant correlations between CaA and HES
478 ($r = 0.827$, $P_p \leq 0.0005$), FeA and GeA ($r = 0.786$, $P_p \leq 0.0005$), FeA and PrA ($r = 0.652$, P
479 $p \leq 0.005$), C and EC ($r = 0.663$, $P_p \leq 0.005$), and C and EGC ($r = 0.656$, $P_p \leq 0.005$) could
480 be considered as important characteristics of the analyzed sage honeys. It was also observed
481 that AbA was well correlated with FeA and GeA, with $r = 0.890$ ($P_p \leq 0.000001$) and $r =$
482 0.887 ($P_p \leq 0.000001$), respectively. The observed correlations between the phenolic
483 compounds in the analyzed honey samples probably reflected the situation in the sage nectar
484 and/or pollen, which are the main sources of phenolics in honey.

485 Pearson's correlation analysis was also performed between 14 targeted carbohydrates
486 in the unifloral sage honey samples (**Table S3**), whereby the highest positive correlation was
487 observed between maltose and isomaltose ($r = 0.870$, $P_p \leq 0.000005$), which could be
488 considered as a unique characteristic of unifloral sage honey. Moreover, statistically

489 significant correlations were found between MALt and SUC, and MALt and TUR~~maltotriose~~
490 and sucrose, and maltotriose and turanose (Table S3).

491 Regarding the mineral composition of the sage honeys, among all statistically
492 significant correlations, the highest positive ones were between Ca and Mn with $r = 0.858$ (P
493 $p \leq 0.000005$), and between Mg and P with $r = 0.849$ ($Pp \leq 0.000005$). **Table S4** shows the
494 Pearson's correlation analysis of the minerals.

495

496 3.8. Authentication of unifloral sage honey

497 In order to demonstrate the applicability of the present research for the authentication
498 of unifloral sage honey, three types of available unifloral honeys of *Lamiaceae* species were
499 introduced into the analysis as out-groups: mint (*Mentha* spp.) honey, winter savory (*Satureja*
500 *montana* L.) honey, and thyme (*Thymus* spp.) honey. The quantitative data on TPC, RSA,
501 targeted phenolics, sugars and minerals in thyme, mint and winter savory honeys are
502 presented as Supplementary data (**Table S5**). Principal component analysis (PCA) was
503 employed to analyze the quantitative data for TPC, RSA, 25 targeted phenolic compounds, 14
504 carbohydrates and 25 minerals in order to examine their relative variations within different
505 honeys (sage, mint, thyme and winter savory honeys).

506 The combination of all the variables was informative enough to clearly discriminate
507 sage honeys from the honeys of different floral origins. The results showed that the principal
508 factorial 2-dimensional plane captured 32.18-% of the total variability (**Fig. 1**). The first
509 principal component accounted for 17.58-% and the second for 15.60-% of the total variance.
510 Clear differentiation of unifloral sage honey from unifloral thyme, mint and winter savory
511 honeys along PC 1 was observed. The variables responsible for the differentiation of unifloral
512 sage honey from the other studied honeys were identified using the loading plots (**Fig. 1B**).
513 Sage honey samples were distinguished from the other studied honeys based on the

514 significantly higher contents of mineral-boron-B. Most of the samples of sage honeys were
515 characterized with high K contents. Higher contents of TPC, TUR-turanose and KAE
516 kaempferol in the sage honeys compared to the thyme, mint and winter savory honeys further
517 contributed to the separation (Fig. 1B). On the other hand, mint honey was characterized by
518 larger contents of Mn, Ba, and ChA-chlorogenic acid, when compared to the other samples.
519 Only two unifloral sage samples (**SH2** and **SH8**) considerably deviated from the rest of the
520 sage honey samples along PC2, due to higher contents of chrysin, pinocembrin,
521 galanginCHR, PNC, GLN, and CaAcaffeic acid, which were also characteristic for the **MH1**
522 and **WSH2** samples.

523

524 4. Conclusions

525

526 The study of sage (*Salvia officinalis* L.) honey samples showed ~~some~~ interesting
527 results related to their ~~peculiar~~ characteristic phenolic, sugar and mineral contents. Several
528 identified compounds showed significant potential for the characterization of this particular
529 honey ~~intrinsic~~ typical of the Adriatic Littoral of Croatia, especially its northern area. The
530 data suggest clear differentiation of unifloral sage honey from the other unifloral honeys by
531 using groups of chemical markers (phenolic compounds, carbohydrates and minerals).

532 Among all studied unifloral honeys of Lamiaceae species, higher contents of boron and
533 potassium, as well as turanose and kaempferol could be identified as authentication markers
534 of unifloral sage honey. In addition, the application of multivariate statistical analysis ~~to~~ for
535 the authentication and classification ~~was~~ proved to be an important complementary tool for a
536 more reliable identification and quality control method of honey.

537

538

539

540 **Acknowledgement**

541 This work was performed within the framework of the Research Projects Nos. 172017

542 and 173024, supported by the Ministry of Education, Science and Technological

543 Development of the Republic of Serbia and specialized funds of the Department of

544 Environmental Health, Faculty of Medicine, University of Rijeka.

545

Accepted Manuscript

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661

662 **Figure Captions**

663

664 **Fig. 1.** (A) PC scores plot of the honey samples; (B) Loadings plot of the honey samples.665 **Fig. S1.** Base peak chromatogram of Common sage honey (sample No. SH2) extract. Peak666 numbers corresponds to those in **Table 3:** (2) gallocatechin, (3) salvianic acid, (4)667 protocatechuic acid, (7) epigallocatechin, (9) catechin, (11) chlorogenic acid, (12) *p*-

668 hydroxybenzoic acid, (13) feruloyl-hexoside, (14) epicatechin, (17) coumaroyl-

669 hexoside, (22) gentistic acid, (23) luteolin-rutinoside, (24) isorhamnetin-rutinoside,

670 (25) quercetin-hexoside, (27) *p*-coumaric acid, (28) taxifolin, (30) rosmarinic acid,671 (35) *trans, trans*-abscisic acid, (37) monohydroxybenzoic acid, (42) sakuranetin, (44)

672 kaempferol, and (45) rhamnetin.

673

674

675 **Table 1** Apiary locations of the sage honey sample's production. Melissopalynological and
 676 sensory assessment of unifloral sage (*Salvia officinalis* L.) honey samples deriving from the
 677 North Croatian Littoral.

Sample	Location	Year	Melissopalynological assessment of honey samples	Sensory assessment of honey samples	Compliance to sage honey uniflorality
SH1	Croatia, Cres	2013	D	Fair	Complies
SH2	Croatia, Eastern Istria	2013	B	Good	Complies
SH3	Croatia, Cres	2013	C	Fair	Complies
SH4	Croatia, Rab	2012	C	Fair	Complies
SH5	Croatia, Cres	2012	C	Good	Complies
SH6	Croatia, Krk	2011	B	Good	Complies
SH7	Croatia, Klenovica	2011	C	Good	Complies
SH8	Croatia, Krk	2011	B	Fair	Complies
SH9	Croatia, Cres	2010	C	Good	Complies
SH10	Croatia, Krk	2010	B	Good	Complies
SH11	Croatia, Cres	2010	D	Good	Complies
SH12	Croatia, Krk	2010	C	Fair	Complies
SH13	Croatia, Krk	2009	C	Good	Complies
SH14	Croatia, Cres	2009	C	Fair	Complies
SH15	Croatia, Kraljevica	2012	B	Good	Complies
SH16	Croatia, Cres	2010	C	Fair	Complies
SH17	Croatia, Cres	2012	C	Fair	Complies
SH18	Croatia, Krk	2012	B	Good	Complies

678 Pollen frequency classes:

679 A - "Predominant pollen" (more than 45 % of the pollen grains);

680 B - "Secondary pollen" (16–45 %);

681 C - "Important minor pollen" (3–15 %);

682 D - "Minor pollen" (less than 3 %).

683

684 **Table 2** Presence of polyphenolics in the sage (*Salvia officinalis* L.) honeys; number of
 685 identified compound, target compounds, mean expected retention times, exact mass,
 686 calculated mass, mean mass accuracy (ppm), and MS/MS fragments.

Peak No	Compounds	t_R , min	Exact mass, [M-H] ⁻	Calculated mass [M-H] ⁻	Δ pm	MS/MS fragments
1	Gallic acid ^a	2.55	169.01392	169.01425	1.95	125
2	Gallocatechin ^a	3.96	305.06583	305.06668	2.79	219, 261
3	Salvianic acid A	3.97	197.04520	197.04555	1.78	179, 153, 123
4	Protocatechuic acid ^a	4.34	153.01903	153.01933	1.96	109
5	Chlorogenic acid isomer 1	4.48	353.08716	353.08781	1.84	191, 179, 146
6	Caffeoyl-hexoside	4.52	341.08716	341.08781	1.91	179
7	Epigallocatechin ^a	4.65	305.06589	305.06668	2.59	219, 261
8	Dimethoxybenzoic acid	4.75	181.05025	181.05063	2.10	151, 137
9	Catechin ^a	4.92	289.07095	289.07176	2.80	159, 123
10	Eriodictyol-rutinoside	5.01	595.16644	595.16684	0.67	449, 287
11	Chlorogenic acid ^a	5.04	353.08682	353.08781	2.80	191, 179, 146
12	<i>p</i> -Hydroxybenzoic acid ^a	5.09	137.02425	137.02442	1.24	93
13	Feruloyl-hexoside isomer 1	5.30	355.10229	355.10346	3.29	193
14	Epicatechin ^a	5.32	289.07114	289.07176	2.14	159, 123
15	Gallocatechin gallate ^a	5.34	457.07703	457.07763	1.31	305
16	Chlorogenic acid isomer 2	5.37	353.08710	353.08781	2.01	191, 179, 146
17	Coumaroyl-hexoside	5.39	325.09213	325.09289	2.34	163
18	Epigallocatechin gallate ^a	5.46	457.0769	457.07763	1.60	305
19	Caffeic acid ^a	5.48	179.03476	179.03498	1.23	135, 161
20	Feruloyl-hexoside isomer 2	5.61	355.10260	355.10346	2.42	193
21	Rutin ^a	5.94	609.14490	609.14611	1.99	463, 301
22	Gentistic acid ^a	5.96	153.01900	153.01933	2.16	109
23	Luteolin-rutinoside	5.97	593.15045	593.15119	1.25	447, 285
24	Isorhamnetin-rutinoside	6.03	623.16040	623.16176	2.18	461, 315
25	Quercetin-hexoside	6.07	463.08691	463.08820	2.79	301
26	Ellagic acid ^a	6.16	300.99847	300.99899	1.73	283, 200, 175
27	<i>p</i> -Coumaric acid ^a	6.25	163.03984	163.04007	1.41	119

28	Taxifolin	6.47	303.05023	303.05103	2.64	285, 269, 255,217
29	Ferulic acid ^a	6.70	193.05014	193.05063	2.54	175, 139
30	Rosmarinic acid ^a	6.79	359.07635	359.07724	2.48	197, 179, 161
31	Apigenin-rutinoside isomer	6.83	577.15521	577.15628	1.85	431, 269
32	Apigenin-hexoside isomer 1	6.92	431.09775	431.09837	1.44	269
33	Luteolin-hexoside	6.93	447.09274	447.09329	1.23	285
34	Eriodictyol	7.14	287.05551	287.05611	2.09	125
35	<i>trans, trans</i> -Abscisic acid	7.44	263.12814	263.12888	2.81	191, 179
36	Apigenin-hexoside isomer 2	7.52	431.09778	431.09837	1.37	269
37	Monohydroxybenzoic acid	7.70	137.02423	137.02442	1.39	93
38	<i>cis, trans</i> -Abscisic acid ^a	7.73	263.12833	263.12888	2.09	191, 179
39	Luteolin ^a	7.75	285.03989	285.04046	2.00	213, 151
40	Quercetin ^a	7.80	301.03445	301.03538	3.09	151, 179, 121
41	Resveratrol ^a	7.85	227.07056	227.07137	3.57	209
42	Sakuranetin	8.06	285.07623	285.07685	2.17	133
43	Apigenin ^a	8.44	269.04477	269.04555	2.90	149, 151, 173, 183
44	Kaempferol ^a	8.57	285.03970	285.04046	2.67	199, 161, 151, 135
45	Rhamnetin	8.57	315.04996	315.05103	3.40	300, 165, 121
46	Hispidulin	8.66	299.05527	299.05611	2.81	284
47	Pinobanksin ^a	8.67	271.06067	271.06120	1.96	253, 243, 165, 151, 107
48	Isorhamnetin	8.73	315.05057	315.05103	1.46	300, 151, 107
49	Hesperetin ^a	8.77	301.07101	301.07176	2.49	271, 161
50	Quercetin dimethyl ether 1	8.98	329.06656	329.06668	0.36	315, 165
51	Quercetin dimethyl ether 2	9.64	329.06586	329.06668	2.49	315, 166
52	Pinostrobin ^a	9.83	269.08121	269.08193	2.68	151, 179
53	Prenyl caffeate	9.85	247.09703	247.09758	2.23	135, 179
54	Chrysin ^a	10.07	253.05009	253.05063	2.13	101, 151, 181, 209, 143
55	Pinocembrin ^a	10.09	255.06563	255.06628	2.55	213, 211, 151
56	Acacetin	10.14	283.06094	283.06120	0.92	268, 133, 151, 107
57	Caffeic acid phenethyl ester (CAPE)	10.15	283.09714	283.09758	1.55	135, 179
58	Galangin ^a	10.25	269.04489	269.04555	2.45	151, 183
59	Genkwanin	10.62	283.06042	283.06120	2.76	268, 239, 211

60	Carnosol^a	11.72	329.17487	329.17583	2.92	311, 296
61	Carnosic acid^a	12.81	331.19061	331.19148	2.63	287, 269

687 ^a Confirmed using available standards.

688 **Table 3** Quantification of individual polyphenolics (mg/kg), radical scavenging activity
 689 (RSA) and total phenolic content (TPC) in the sage (*Salvia officinalis* L.) honeys.

	SH1	SH2	SH3	SH4	SH5	SH6	SH7	SH8	SH9	SH10	SH11	SH12	SH13	SH14	SH15	SH16	SH17	SH18
Ga	-	0.1	0.13	0.1	0.1	0.18	0.1	0.1	-	0.1	-	0.1	-	0.1	0.1	0.1	0.1	-
A	-	5		2	2		6	9	-	3	-	2	-	7	3	5	2	-
GC	-	0.4	0.15	0.1	0.1	0.16	-	-	0.1	0.2	0.1	0.1	-	-	-	0.2	0.2	0.3
A	-	9		9	6		-	-	6	2	6	5	-	-	-	0	6	7
Pr	0.34	0.7	0.74	0.5	0.2	0.34	0.3	0.6	0.6	0.4	0.4	0.1	0.3	0.4	0.3	0.3	0.4	0.6
A	0.34	0		4	5		1	8	2	8	9	4	1	2	3	9	4	8
EG	0.12	-	0.46	0.1	0.0	0.12	0.0	0.1	0.1	0.1	0.0	0.0	0.1	0.0	0.0	-	0.2	0.1
C	0.12	-		0	8		8	0	4	4	9	7	6	8	8	-	4	6
Ge	-	-	-	-	-	-	-	0.1	-	-	-	-	-	-	-	0.0	-	0.0
A	-	-	-	-	-	-	-		3	-	-	-	-	-	1	-	-	2
HB	1.89	1.9	1.61	1.2	1.8	1.45	3.2	2.1	1.3	1.8	1.2	0.8	1.3	1.5	2.0	1.7	1.6	2.3
A	1.89	3		5	2		8	8	6	2	0	1	4	5	6	3	6	6
Ch	-	0.0	0.01	0.1	0.2	-	0.0	0.0	0.2	0.0	0.0	0.0	0.0	-	-	0.1	0.0	-
A	-	4		0	2		2	5	6	9	4	3	7	-	-	0	3	-
C	0.12	0.0	0.15	0.0	-	-	-	-	0.0	0.0	0.0	-	0.0	-	-	0.0	0.0	-
A	0.12	3		9	-	-	-	-	5	1	4	-	5	-	-	1	6	-
Ca	0.48	1.8	0.56	0.3	0.4	0.59	0.5	0.9	0.5	0.6	0.6	0.6	0.3	0.3	0.4	0.7	0.5	0.2
A	0.48	9		7	8		7	2	4	4	1	2	7	8	7	0	5	6
GC	0.82	0.7	0.75	-	-	-	1.1	1.0	0.7	0.7	0.7	0.6	0.8	0.6	0.7	0.6	0.7	1.0
G	0.82	4		-	-	-	5	5	2	0	2	9	1	9	1	9	3	3
EC	0.10	-	0.03	0.0	-	-	-	-	-	-	-	-	-	-	-	0.0	0.0	-
A	0.10	-		6	-	-	-	-	-	-	-	-	-	-	-	5	7	-
Co	2.73	3.1	3.45	1.8	2.0	1.36	3.6	2.7	2.8	1.3	1.8	2.5	1.0	0.7	0.9	1.6	2.6	1.1
A	2.73	1		9	1		2	8	1	9	2	1	3	7	2	2	8	0
Fe	0.64	1.5	3.09	0.9	0.4	0.50	1.0	4.3	1.3	0.3	0.5	0.7	0.6	0.1	0.8	0.5	0.6	1.4
A	0.64	4		7	4		1	9	1	4	1	7	0	6	3	3	7	0
Ro	-	0.2	0.01	0.2	0.3	0.30	0.3	0.0	0.1	0.0	0.1	0.2	0.1	0.2	0.2	0.1	0.2	0.2
A	-	1		1	7		3	5	8	9	5	3	4	7	5	4	6	9
EG	0.94	1.2	0.97	0.8	0.7	1.11	1.0	0.9	0.8	-	0.9	1.2	0.9	0.5	1.0	1.2	1.1	1.6

CG	5	5	9		0	6	0		6	6	5	8	7	8	4	1		
Ab	1.0		0.5	0.7		0.7	3.9	0.6	0.2	0.4	0.8	0.3	0.6	1.6	0.6	0.4	1.5	
A	0.35	1.89	4	4	1.09	3	9	1	6	8	6	9	4	4	9	6	9	
RE	0.1			0.2											0.0	0.4		
S	-	-	-	2	-	-	-	-	-	-	-	-	-	-	8	6	-	
KA	0.1		0.7	0.4		0.2	0.2	0.5	0.3	0.6	0.0	0.3	0.1	0.2	0.5	0.5	0.2	
E	0.14	0.24	8	6	0.16	0	1	1	8	5	3	8	8	6	4	5	8	
PN	2.3		0.3	0.6		1.0	1.8	0.8	1.3	1.0	2.2	0.4	0.5	1.1	1.1	0.2	0.2	
B	-	0.35	3	3	1.57	0	2	2	0	0	6	4	8	0	4	4	1	
QU	0.3		0.5	0.2		0.1	0.3	0.3	1.0	0.3	0.1	0.3	0.3	0.1	0.5	0.4	0.3	
E	0.07	0.14	8	3	0.12	7	3	6	5	6	1	2	6	4	9	5	0	
CH	1.9		0.2	0.2		0.4	0.9	0.4	0.8	0.7	1.5	0.4	0.4	0.9	0.9	0.2	0.0	
R	0.06	0.41	7	3	0.87	8	5	7	1	3	0	2	7	0	2	5	9	
PN	0.3					0.0	0.1	0.0	0.0	0.0	0.1			0.0	0.0			
S	-	-	-	-	0.07	4	9	1	7	3	9			5	4			
PN	0.8		0.0	0.1		0.2	0.4	0.2	0.4	0.3	0.7	0.1	0.1	0.4	0.4			
C	-	0.09	5	5	0.51	2	6	8	5	5	2	5	7	6	3			
HE	0.8		0.0	0.2		0.1	0.3	0.2	0.3	0.2	0.5	0.0	0.0	0.3	0.3			
S	-	0.19	6	0	0.37	1	8	7	3	0	2	4	9	6	6			
GL	0.3			0.0		0.0	0.2	0.0	0.1	0.0	0.2	0.0		0.0	0.0			
N	-	0.01	-	1	0.11	7	3	4	2	7	2	1	-	9	9			
TP	553.	485	424.	591	522	484.	471	509	538	56	747	417	525	444	379	545	585	208
C	98	.86	92	.08	.33	91	.03	.11	.51	0.7	.49	.73	.39	.97	.54	.40	.28	.51
RS	819.	627	526.	770	548	571.	541	627	610	58	894	474	641	675	416	626	770	351
A	75	.44	81	.65	.55	41	.69	.79	.38	6.0	.87	.17	.38	.23	.81	.20	.11	.20
										4								

690 **GaA** – Gallic acid; **GC** – Gallocatechin; **PrA** – Protocatechuic acid; **EGC** – Epigallocatechin; **GeA** – Gentisic
691 acid; **HBA** – *p*-Hydroxybenzoic acid; **ChA** – Chlorogenic acid; **C** – Catechin; **CaA** – Caffeic acid; **GCG** -
692 Gallocatechin gallate; **EC** – Epicatechin; **CoA** – *p*-Coumaric acid; **FeA** – Ferulic acid; **RoA** – Rosmarinic acid;
693 **EGCG** - Epigallocatechin gallate; **AbA** – *cis, trans*-Abscisic acid; **RES** – Resveratrol; **KAE** – Kaempferol;
694 **PNB** – Pinobanksin; **QUE** – Quercetin; **CHR** – Chrysin; **PNS** – Pinostrobin; **PNC** – Pinocembrin; **HES** –
695 Hesperetin; **GLN** – Galangin; **TPC** – Total phenolic content (mg GEA/kg); **RSA** – Radical scavenging activity
696 ($\mu\text{mol TE/kg}$).

697

698 **Table 4** Quantification of the sugars and sugar alcohols in the sage (*Salvia officinalis* L.) honeys (g/kg).

	SH1	SH2	SH3	SH4	SH5	SH6	SH7	SH8	SH9	SH10	SH11	SH12	SH13	SH14	SH15	SH16	SH17	SH18
ERY	0.04	0.07	0.05	0.05	0.02	0.07	0.06	0.07	0.05	0.07	0.09	0.69	0.06	0.07	0.18	0.12	0.11	0.09
SOR	0.04	0.13	0.03	0.06	0.09	0.08	0.30	0.33	0.20	0.06	0.06	0.03	0.04	0.02	0.18	0.05	0.05	0.02
TRE	1.23	5.70	2.71	2.07	5.34	0.21	5.07	2.76	1.45	5.08	1.82	0.75	1.31	2.39	1.41	0.14	0.63	1.02
ARA	0.04	0.15	0.05	0.10	0.10	0.37	0.12	0.07	0.06	0.07	0.10	0.05	0.09	0.05	0.05	0.06	0.08	0.03
GLU	305.03	272.07	280.3	253.24	212.95	206.81	108.89	245.39	200.35	263.82	227.38	240.15	244.46	252.72	271.21	262.22	277.59	263.08
FRU	399.41	464.01	440.3	420.83	393.63	475.99	480.76	437.51	442.23	462.32	464.06	489.06	461.97	455.68	461.76	464.87	447.35	471.03
SUC	30.41	28.78	11.02	15.69	14.17	14.27	20.71	14.03	11.87	16.54	21.4	27.08	25.64	19.01	11.21	18.59	16.34	16.16
TUR	1.34	0.12	0.32	0.68	0.6	0.83	1.13	0.76	0.88	0.65	0.87	0.82	0.82	0.90	0.11	0.77	1.16	1.06
GLY	1.51	0.11	0.16	0.09	0.12	0.17	0.12	0.16	0.03	0.19	0.10	0.05	0.08	0.07	0.17	0.12	0.12	0.13
GAL	0.06	0.06	0.04	0.09	0.46	0.33	0.20	0.36	0.02	0.07	0.08	0.03	0.02	0.03	0.2	0.06	0.07	0.03
iMAL	4.45	3.11	5.31	14.6	3.63	7.13	7.68	5.40	3.95	12.61	12.34	3.52	7.44	8.51	8.28	10.22	11.62	9.98
iMALt	1.44	2.88	0.73	4.66	2.55	2.76	1.24	1.48	1.11	3.79	5.66	1.11	0.33	0.36	0.31	2.41	3.91	2.44
MAL	6.05	3.74	5.92	16.85	8.76	9.32	11.91	7.32	7.38	11.77	11.09	7.40	8.98	10.06	8.33	11.49	12.31	9.91
MALt	0.11	0.06	0.01	0.02	0.02	0.05	0.06	0.08	0.06	0.04	0.07	0.08	0.09	0.07	0.02	0.09	0.08	0.08
SUM	751.16	780.99	746.95	729.03	642.44	718.39	638.25	715.72	669.64	777.08	745.12	770.82	751.33	749.94	763.42	771.21	771.42	775.06
FRU/GLU	1.31	1.71	1.57	1.66	1.85	2.30	4.42	1.78	2.21	1.75	2.04	2.04	1.89	1.80	1.70	1.77	1.61	1.79

MAL/iMAL 1.36 1.20 1.11 1.15 2.41 1.31 1.55 1.35 1.87 0.93 0.90 2.10 1.21 1.18 1.01 1.13 1.06 0.99

- 699 **ERY** – Erythritol; **SOR** – Sorbitol; **TRE** – Trehalose; **ARA** – Arabinose; **GLU** – Glucose; **FRU** – Fructose; **SUC** – Sucrose; **TUR** – Turanose; **GLY** – Glycerol; **GAL** –
- 700 Galactitol; **iMAL** – Isomaltose; **iMALT** – Isomaltotriose; **MAL** – Maltose; **MALT** – Maltotriose; **SUM** – Summary of quantified sugars and sugar alcohols; **FRU/GLU** –
- 701 Fructose/Glucose ratio; **MAL/iMAL** – Maltose/Isomaltose ratio.

Table 5 Quantification of the minerals in the sage (*Salvia officinalis* L.) honeys (mg/kg).

	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH	SH
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
A	0.0	0.4	0.4	0.4	0.1	0.0	<L	0.2	0.5	0.2	0.3	<L	0.3	0.8	0.0	0.2	0.1	0.1
I	95	33	31	19	45	23	OQ	61	05	66	87	OQ	06	83	59	63	49	11
A	0.0	<L	<L	<L	<L	<L	<L	0.0	<L	<L	<L	<L	<L	<L	<L	0.0	<L	<L
s	03	OQ	O	OQ	OQ	OQ	OQ	06	OQ	OQ	OQ	OQ	OQ	OQ	OQ	13	OQ	OQ
B	1.2	2.3	1.1	1.2	1.8	2.7	2.1	2.5	1.9	1.3	2.0	2.2	2.1	1.7	3.0	2.1	1.4	1.6
B	70	76	34	90	76	60	13	99	07	88	86	01	74	74	29	40	55	81
B	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
a	51	94	84	68	76	66	59	76	67	64	72	76	64	71	71	60	69	62
C	23.	49.	50.	50.	36.	23.	31.	47.	34.	32.	39.	20.	35.	22.	36.	30.	30.	20.
a	823	894	42	471	787	591	557	122	375	747	980	409	098	978	471	205	493	885
C	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
d	03	04	03	04	03	03	03	04	03	03	03	03	03	04	04	03	03	03
C	<L	<L	<L	0.0	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L
o	OQ	OQ	O	12	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ
C	0.0	0.0	0.0	<L	<L	<L	<L	0.0	0.0	0.0	0.0	<L	0.0	0.0	<L	<L	<L	0.0
r	04	05	04	OQ	OQ	OQ	OQ	15	06	10	07	OQ	13	09	OQ	OQ	OQ	03
C	0.1	0.2	0.1	0.1	0.2	0.1	0.2	0.1	0.2	0.1	0.2	0.2	0.1	0.1	0.3	0.1	0.1	0.1
u	01	99	87	26	20	98	83	73	33	55	19	21	66	87	13	52	24	09
F	0.2	0.9	0.5	0.5	0.4	0.4	0.2	0.8	0.3	0.4	2.2	0.4	2.4	0.8	0.1	0.3	0.2	0.4
e	47	28	27	33	57	42	74	53	71	79	78	00	94	06	95	82	48	96
K	2.1	1.2	1.8	2.0	1.3	0.8	0.9	0.9	1.0	1.1	1.8	0.5	1.3	0.7	1.2	1.1	2.1	0.6
"	51	81	38	37	13	44	09	82	26	16	73	92	00	30	13	45	07	75
L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L	<L
i	OQ	OQ	O	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ	OQ
M	5.0	26.	11.	10.	7.9	7.9	8.9	25.	8.0	11.	8.7	5.9	7.1	9.9	10.	7.1	5.7	10.