

HYDROLATES – BY-PRODUCTS OF ESSENTIAL OIL DISTILLATION: CHEMICAL COMPOSITION, BIOLOGICAL ACTIVITY AND POTENTIAL USES

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Hydrolates, also referred to as hydrosols, floral or distillate waters, as well as aromatic waters, are produced in the same isolation process with essential oils by steam distillation. A small amount of essential oil constituents is dissolved in hydrolates providing specific organoleptic properties and flavor, as well as biological activity which makes them useful as raw material in many industries. Their popularity is still on the rise, especially in aromatherapy. The objective in this review is to analyze the chemical compositions of hydrolates and their corresponding essential oils, as well as biological activity of hydrolates (antimicrobial, antioxidant and antiinflammatory) and potential uses, not only in food industry for flavoring, and preservation of fresh-cut fruits and vegetables, but also as functional (soft) drinks. However, hydrolates can be used in aromatherapy and cosmetics, as well as in organic agriculture and aquaculture.

Keywords: Hydrolates, Hydrosols, Aromatic waters, Floral waters, Distillate waters

Introduction

Hydrolates, also referred to as hydrosols, floral or distillate waters, as well as aromatic waters, are produced in the same isolation process with essential oils by steam distillation. During industrial distillation, water is evaporated simultaneously with the essential oil. After condensation of the vapors in contact with cold vessels or tubes, the liquefied components are separated into two phases inside a collecting Florentine vessel: essential oil and the hydrolate [1,2,3]. However, a small amount of essential oil constituents dissolves in hydrolates, precious oxygenated compounds, providing specific organoleptic properties and flavor, as well as biological activity which makes them useful for food and cosmetic industries [4,5].

The essential oil dissolved in hydrolates is usually discarded. This leads to the loss of dissolved essential oil. This phenomenon was observed in many other aromatic crops and attempts were made to recover the dissolved oil from hydrolates. Recovered essential oils are often referred to as secondary oils. The methods used for recovering aromatic oils from hydrolates were: cohobation, extraction with diethyl ether, adsorbing oil constituents on to an adsorbent followed by ethanol extraction and poroplast technique [6]. Furthermore, the loss of oxygenated constituents in distillation water makes the aroma of primary oils incomplete in terms of organoleptic richness and fullness. When the recovered oil was blended with the primary oil, the olfactory evaluation of the blended oil indicated that it had a much more natural and richer aroma than the primary oil. Alternatively, hydrolates could also be used for the isolation

of pure compounds [5].

Many different hydrolates are used commercially, mainly as cosmetic ingredients, as well as in food industry. Their popularity is still on the rise, especially in aromatherapy. These natural products constitute promising, new and valuable sources, which may be approved as raw material in many different products [7].

Coincidentally, short time before work was started on this contribution, a review covering a similar field was published [8]. Not meaning to duplicate this work, the intention was still to complement it. However, the above-mentioned excellent review is mainly devoted to the discussion of the biological activity of hydrolates and their potential as antimicrobials for food applications.

The objective in this review is to analyze the chemical compositions of hydrolates and their corresponding essential oils, as well as biological activity of hydrolates (antimicrobial, antioxidant and antiinflammatory) and potential uses, not only in food industry for flavoring, and preservation of fresh-cut fruits and vegetables, but also as functional (soft) drinks. Furthermore, hydrolates can be used in aromatherapy and cosmetics, as well as in organic agriculture.

Chemical composition

Hydrolates usually contain less than 1 g L⁻¹ (i.e. 0.10%) of water soluble aromatic compounds (i.e. volatile organic compounds) from essential oil, that remain dissolved in the water phase, whereas hydrolates which included 0.17% aromatic compounds that remained after the water fraction,

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are highly aromatic and their composition was rather different than of the essential oil's [9]. However, literature review of absolute quantification of the water-soluble aromatic compounds from different hydrolates in mg L⁻¹ is shown in Table 1.

Table 1. Content of Volatile Organic Compounds (VOCs) in hydrolates (mg L⁻¹)

Plant material	Plant part	Content of VOC (mg L ⁻¹)	Reference
<i>Abies alba</i>	seed	11	[7]
<i>Abies balsamea</i>	branches	190	[10]
<i>Abies koreana</i>	seed	37	[7]
<i>Asarum canadense</i>	root	590	[11]
<i>Citrus aurantium</i>	peel	~212	[1]
<i>Comptonia peregrina</i>	aerial parts	296	[12]
<i>Cymbopogon citratus</i>	leaves	~249	[1]
<i>Eucalyptus citrodora</i>	leaves	~165	[1]
<i>Eugenia caryophyllata</i>	buds	~1012	[1]
<i>Ledum groenlandicum</i>	aerial parts	630	[12]
<i>Melissa officinalis</i>	aerial parts	240	[11]
<i>Mentha piperita</i>	aerial parts	235	[13]
<i>Mentha spicata</i>	aerial parts	~397	[1]
<i>Myrica gale</i>	aerial parts	110	[12]
<i>Ocimum basilicum</i>	aerial parts	~360	[1]
<i>Picea glauca</i>	branches	933	[10, 13]
<i>Picea mariana</i>	branches	260	[10]
<i>Solidago puberula</i>	aerial parts	241	[13]

(-) calculated according to presented data

Table 2. Analysis of essential oils and Volatile Organic Compounds (VOCs) from corresponding hydrolates of different plant samples

Plant material	Plant part	Country of origin	Essential oil	Hydrolate	Reference
<i>Abies alba</i>	seed	Poland	limonene (82.9%) camphene (6.3%)	selin-6-en-4-ol (51.7%) β -himachalol (14.5%) τ -cadinol (10.9%) intermedeol (9.8%) α -terpineol (41.6%)	[7]
<i>Abies balsamea</i>	branches	Canada	β -pinene (33.5%) δ -3-carene (14.0%) α -pinene (11.3%) bornyl acetate (10.0%) β -phellandrene (7.2%) limonene (7.1%)	maltol (8.6%) borneol (6.4%) bornyl acetate (5.1%)	[10]
<i>Abies koreana</i>	seed	Poland	limonene (53.7%) α -pinene (12.1%) bornyl acetate (12.1%) camphene (11.2%)	intermedeol (76.7%) selin-6-en-4-ol (2.4%) bornyl acetate (2.0%)	[7]
<i>Asarum canadense</i>	root	Canada	methyl eugenol (38.5%) linalool (19.4%) α -terpineol (5.9%)	linalool (106 mg L ⁻¹) α -terpineol (115 mg L ⁻¹) methyl eugenol (89 mg L ⁻¹)	[11]
<i>Bunium persicum</i>	seed	Iran	cuminic aldehyde (26.8%) γ -terpinene (24.03%) γ -terpinene-7-al (14.2%) o-cymene (9.2%)	cuminol (32.5%) cuminic aldehyde (29.4%) α -terpinene-7-al (19.4%)	[18]
<i>Cerasus serrulata</i>	flowers	China	benzaldehyde (42.1%) tricosane (27.7%) pentacosane (19.0%)	benzaldehyde (64.3%) mandelonitrile (12.4%)	[19]
<i>Cerasus subhirtella</i>	flowers	China	benzaldehyde (31.2%) tricosane (23.1%) pentacosane (23.2%)	benzaldehyde (67.5%) mandelonitrile (12.5%)	[19]
<i>Citrus aurantium</i>	peel	Egypt	linalool (52.8%) linalyl acetate (9.5%)	linalyl acetate (46.7%) linalool (32.7%)	[1]
<i>Citrus ladanifer</i>	aerial parts	Portugal	α -pinene (26.3%) camphene (19.0%) bornyl acetate (5.0%) limonene (3.8%)	2,6,6-trimethyl cyclohexane (10.8%) E-pinocarveol (5.2%) borneol (6.2%)	[20]
<i>Comptonia peregrina</i>	aerial parts	Canada	β -caryophyllene (27.0%) 1,8-cineole and myrcene (9.7%) myrcene (7.3%)	1,8-cineole (15.8%) E-2-hexen-1-ol (14.2%) Z-3-hexen-1-ol (12.3%) α -terpineol (9.8%) linalool (6.3%) terpinene-4-ol (6.2%)	[12]

<i>Cupressus lusitanica</i>	aerial parts	Portugal	α -pinene (24.8%) sabinene (15.6%) limonene (13.6%) δ -3-carene (12.2%)	umbellulone (47.9%) terpinen-4-ol (23.8%)	[20]
<i>Cymbopogon citratus</i>	leaves	Egypt	geranial (36.9%) neral (34.5%) linalool (9.3%)	geranial (43.8%) neral (31.9%)	[1]
<i>Daucus carota</i> subsp. <i>sativus</i>	aerial parts	Algeria	alismol (15.2%) α -humulene (9.5%) β -ionone (8.2%)	caryophyllene oxide (9.8%) <i>p</i> -cymen-8-ol (8.9%)	[21]
<i>Daucus carota</i> subsp. <i>sativus</i>	root	Algeria	geranyl linalool (50.3%)	myristicine (17.8%) E-methyl-iso-eugenol (16.6%) methyl-eugenol (11.9%)	[21]
<i>Eucalyptus alba</i>	leaves	Senegal	1,8-cineole (37.2%) α -pinene (22.7%) limonene (7.0%)	1,8-cineole (39.1%) E-pinocarveol (19.3%) pinocarvone (6.8%)	[22]
<i>Eucalyptus camaldulensis</i>	leaves	Senegal	1,8-cineole (50.0%) limonene (17.8%) α -pinene (9.3%) <i>p</i> -cymene (7.4%)	1,8-cineole (52.6%) α -terpineol (6.6%) Z- <i>p</i> -mentha-1-(7)-8-dien-2ol (5.1%)	[22]
<i>Eucalyptus cinerea</i>	leaves	Italy	1,8-cineole (83.8%) limonene (4.7%) α -terpineol (2.8%)	1,8-cineole (88.4%) α -terpineol (6.2%)	[23]
<i>Eucalyptus citrodora</i>	leaves	Egypt	citronellal (34.3%) citronellol (8.9%)	citronellal (76.1%) citronelool (9.9%)	[1]
<i>Eucalyptus parvula</i>	leaves	Italy	1,8-cineole (87.8%) limonene (3.3%) α -terpineol (3.1%)	1,8-cineole (89.7%) α -terpineol (5.9%)	[23]
<i>Eucalyptus pulverulenta</i>	leaves	Italy	1,8-cineole (86.4%) limonene (4.0%) α -terpineol (2.1%)	1,8-cineole (90.0%) α -terpineol (4.3%)	[23]
<i>Eucalyptus tereticornis</i>	leaves	Senegal	<i>p</i> -cymene (45.5%) 1,8-cineole (22.9%) limonene (5.4%)	1,8-cineole (30.7%) α -terpineol (8.8%) E-pinocarveol (7.6%) carvacrol (7.0%) eugenol (84.4%)	[22]
<i>Eugenia caryophyllata</i>	buds	Egypt	eugenol (96.6%)	eugenol (84.4%)	[1]
<i>Laurus nobilis</i>	leaves	Argentina	1,8-cineole (45.0%) linalool (12.8%)	1,8-cineole (54.4%) α -terpineol (11.8%) methyl eugenol (10.9%) linalool (8.1%) terpinen-4-ol (7.6%) eugenol (6.0%)	[24]
<i>Lavandula angustifolia</i>	flowers	Poland	linalool (30.6 %) linalyl acetate (14.2 %) geraniol (5.3 %)	linalool (34.1%) α -terpineol (9.9%) borneol (7.1%) terpinen-4-ol (5.9%)	[25, 26]
<i>Lavandula intermedia</i>	flowers	Turkey	linalyl acetate (47.7%) linalool (34.0%)	linalool (55.6%) borneol (13.5%) camphor (13.4%) 1,8-cineole (9.8%) linalool oxide (6.0%)	[27]
<i>Lavandula intermedia</i>	flowers	Italy	linalool (35.8%) 1,8-cineole (19.8%) α -pinene (8.7%) linalyl acetate (7.5%)	1,8-cineole (52.9%) camphor (19.6%) linalool (12.6%)	[28]
<i>Ligusticum porteri</i>	root	Oregon	sabinene (9.0-15.5%) <i>p</i> -cymene (6.5-9.7%) viridene (19.4%) E-sabinyl acetate (22.6%)	terpinen-4-ol (16.5 mg L ⁻¹) hexanal (2.5 mg L ⁻¹) furfural (2.0 mg L ⁻¹) <i>p</i> -cymen-8-ol (2.5 mg L ⁻¹)	[17]
<i>Lippia alba</i>	leaves	Columbia	carvone (41.6%) limonene (40.6%) germacrene D (8.1%)	carvone (92.7%)	[29]
<i>Melissa officinalis</i>	aerial parts	Canada	geranial (24.3%) neral (17.5%) geraniol (8.5%)	geraniol (42 mg L ⁻¹) Z-3-hexen-1-ol (28 mg L ⁻¹) linalool (18 mg L ⁻¹) geranial (13 mg L ⁻¹) α -terpineol (11 mg L ⁻¹) E- <i>p</i> -menth-2-ene-1,8-diol (11 mg L ⁻¹) Z- <i>p</i> -menth-2-ene-1,8-diol (8 mg L ⁻¹)	[11]

<i>Mentha arvensis</i>	aerial parts	Japan	menthol (66.4%) menthone (16.1%) isomenthone (3.5 %)	menthol (371.7 mg L ⁻¹) menthone (52.8 mg L ⁻¹) isomenthone (19.2 mg L ⁻¹)	[30]
<i>Mentha longifolia</i>	aerial parts	Senegal	pulegone (47.2%) menthone (17.8%) 1,8-cineole (12.3%) Isomenthone (11.1%)	pulegone (53.6%) 1,8-cineole (13.8%) isomenthone (9.0%) menthone (7.8%)	[31]
<i>Mentha piperita</i>	aerial parts	Canada	menthol (52.3%) menthone (15.9%)	menthol (67.3%) menthone (3.9%)	[13]
<i>Mentha piperita</i>	aerial parts	Egypt	menthol (48.6.1%) menthone (20.2%) 1,8 cineole (5.3%)	menthol (22.1%) menthone (13.6%) 1,8 cineole (6.7%)	[1]
<i>Mentha pulegium</i>	aerial parts	Morocco	pulegone (68.9%) piperitone (24.8%)	carvacrol (39.4%) piperitone (10.1%)	[16]
<i>Mentha spicata</i>	aerial parts	Morocco	carvone (71.6%)	1,8-cineole (22.9%) camphor (13.5%) α -pinene (5.9%)	[16]
<i>Mentha spicata</i>	aerial parts	Egypt	carvone (75.2%)	carvone (56.2%) limonene (6.7%)	[1]
<i>Mentha spicata</i>	aerial parts	Iran	carvone (28.9%) pulegone (16.3%) piperitone (9.2%)	piperitone (38.3%) carvone (22.1%) pulegone (14.8%) 1,8-cineole (9.7%)	[18]
<i>Mentha suaveolens</i>	aerial parts	Morocco	piperitone oxide (74.7%) γ -muurolene (5.5%)	piperitone oxide (69.3%)	[16]
<i>Monarda citrodora</i>	aerial parts	Italy	thymol (19.6%) <i>p</i> -cymene (15.6%) γ -terpinene (13.5%) α -terpinene (9.2%)	thymol (66.4%) carvacrol (28.6%)	[32]
<i>Myrica gale</i>	aerial parts	Canada	E-E-germacrone (13.5%) myrcene (7.9%) germacrene B (7.5%) β -caryophyllene (5.6%)	Z-3-hexen-1-ol (28.9%) 1,8-cineole (7.1%) terpinen-4-ol (6.3%) α -terpineol (6.3%)	[12]
<i>Nepeta nepetella</i>	aerial parts	Algeria	4 $\alpha\alpha$,7 α ,7 $\alpha\beta$ -nepetalactone (72.4%) 4 $\alpha\alpha$,7 α ,7 $\alpha\alpha$ -nepetalactone (16.3%)	4 $\alpha\alpha$,7 α ,7 $\alpha\beta$ -nepetalactone (49.2%) 4 $\alpha\alpha$,7 α ,7 $\alpha\alpha$ -nepetalactone (7.7%) terpinene-4-ol (6.1%)	[33]
<i>Ocimum basilicum</i>	aerial parts	Egypt	linalool (59.2%) eugenol (26.1%) 1,8-cineol (6.9%)	linalool (65.9%) eugenol (7.0%) 1,8-cineol (5.1%)	[1]
<i>Origanum vulgare</i> (thymol chemotype)	aerial parts	India	thymol (30.8%) γ -terpinene (21.4%) <i>p</i> -cymene (19.0%)	thymol (83.4%) carvacrol (5.9%)	[5]
<i>Origanum vulgare</i> (carvacrol chemotype)	aerial parts	India	carvacrol (42.3%) <i>p</i> -cymene (20.9%) γ -terpinene (23.4%)	carvacrol (94.7%)	[5]
<i>Picea glauca</i>	branches	Canada	β -pinene (20.0%) camphor (20.0%) α -pinene (12.0%) bornyl acetate (12.0%)	camphor (65.0%) borneol (10.0%)	[10]
<i>Picea mariana</i>	bark	Canada	α -pinene (40.6%) β -pinene (29.9%)	α -terpineol (31.4%) terpinen-4-ol (5.4%) verbenone (5.2%)	[34]
<i>Picea mariana</i>	branches	Canada	bornyl acetate (34.2%) camphene (16.4%) α -pinene (12.9%) δ -3-carene (5.9%)	α -terpineol (14.8%) borneol (13.5%) bornyl acetate (7.8%) terpinen-4-ol (6.5%) Z-3-hexen-1-ol (6.5%) camphene hydrate (5.2%)	[10]
<i>Piper chimonanthifolium</i>	leaf	Brazil	piperitone (35.2%) Z- β -ocimene (15.7%) spathulenol (6.6%)	piperitone (86.6%) linalool oxide (10.3%)	[35]
<i>Piper chimonanthifolium</i>	inflorescent	Brazil	linalool (31.1%) piperitone (23.5%) Z- β -ocimene (7.7%) E- β -ocimene (7.7%)	piperitone (60.1%) linalool (25.4%) linalool oxide (11.4%)	[35]
<i>Piper chimonanthifolium</i>	stem	Brazil	piperitone (31.7%) limonene (7.2%) Z- β -ocimene (7.2%) α -pinene (6.8%) humulene epoxide II (5.0%)	piperitone (100.0%)	[35]

<i>Psoralea bituminosa</i>	aerial parts	Algeria	E- β -caryophyllene (19.8%) caryophyllene oxide (15.2%) Z-phytol (14.2%) E-phytol (11.5%) β -copaene (9.2%) citronellol (35.2%) geraniol (22.2%) nonadecane (13.9%) nerol (10.3%)	caryophyllene oxide (35.5%) E-phytol (25.6%) <i>r</i> -cadinol (9.9%) α -cadinol (8.5%)	[36]
<i>Rosa damascena</i>	flowers	Turkey	citronellol (35.2%) geraniol (22.2%) nonadecane (13.9%) nerol (10.3%)	geraniol (30.7%) citronellol (29.4%) phenylethyl alcohol (23.7%) nerol (16.1%)	[37]
<i>Rosa damascena</i>	flowers	Bulgaria	citronellol (31.5%) geraniol (19.1%) nerol (9.7%)	phenylethyl alcohol (43.7%) citronellol (18.2%) geraniol (14.8%)	[38]
<i>Rosmarinus officinalis</i>	leaves	Japan	eucalyptol (18.9%) α -pinene (13.3%) camphor (11.3%) β -pinene (7.3%) borneol (7.1%) bornyl acetate (7.3%) β -myrcene (5.6%)	camphor (29.7%) borneol (23.9%) eucalyptol (20.5%)	[39]
<i>Rosmarinus officinalis</i>	flowers	Columbia	camphor (29.2%) 1,8-cineole (21.1%) α -pinene (10.1%) camphene (8.7%) β -pinene (7.1%)	camphor (51.9%) 1,8-cineole (38.2%)	[29]
<i>Salvia officinalis</i>	aerial parts	Turkey	1,8-cineole (17.9%) camphor (19.9%) Z-thujone (20.1%) E-thujone (9.1%)	1,8-cineole (24.0%) camphor (43.4%) Z-thujone (15.5%) borneole (7.7%)	[9]
<i>Sideritis raeseri</i>	aerial parts	Greece	nuciferol (45.2%) spathulenol (11.6%) caryophyllene oxide (5.6%)	carvacrol (77.1%) nuciferol (8.2%) α -bisabolol (6.6%)	[40]
<i>Solidago puberula</i>	aerial parts	Canada	α -pinene (37.2%) germacrene D (12.4%) limonene (11.9%) myrcene (11.1%)	benzyl alcohol (73.3%) Z-3-hexen-1-ol (7.7%)	[13]
<i>Thymbra capitata</i>	aerial parts	Greece	γ -terpinene (35.3%) <i>p</i> -cymene (23%) carvacrol (20.4%)	carvacrol (946.3 mg L ⁻¹)	[41]
<i>Thymus capitatus</i>	aerial parts	Algeria	carvacrol (69.6%) <i>p</i> -cymene (12.4%)	carvacrol (95.1%)	[42]
<i>Thymus capitatus</i>	aerial parts	Italy	carvacrol (67.6%) β -caryophyllene (7.7%) γ -terpinene (6.8%) <i>p</i> -cymene (6.4%)	carvacrol (93.1%) thymol (6.3%)	[43]
<i>Thymus vulgaris</i>	aerial parts	Columbia	thymol (36.1%) <i>p</i> -cymene (28.4%) γ -terpinene (11.6%)	thymol (98.1%)	[29]
<i>Trachyspermum ammi</i>	seed	Iran	thymol (53.4%) <i>o</i> -cymene (21.1%) γ -terpinene (20.2%)	thymol (90.9%) cuminaldehyde (6.4%)	[18]
<i>Zataria multiflora</i>	aerial parts	Iran	thymol (36.4%) carvacrol (30.7%) <i>o</i> -cymene (11.9%)	carvacrol (55.9%) thymol (40.4%)	[18]

Hydrolates are acidic liquids (pH ranging from 4.5 to 5.5) with pleasant to unpleasant and from similar to dissimilar odor to the essential oil [14, 15]. The quality of hydrolates is determined based on the amount of their soluble volatile compounds. For this reason, the absolute quantification is recommended in the quality control to ensure the optimum quality and to detect adulteration which is easily done by diluting hydrolates with water. Absolute quantification is also useful for providing the processor with full information about the potential application and safety of hydrolates [1]. The chemical composition of essential oils and corresponding hydrosols, according to literature are shown in Table 2.

The volatile components from hydrolates are mainly

monoterpene alcohols, aldehydes and ketones, as well as sesquiterpene alcohols. They are highly polar (hydrophilic) compounds. However, hydrocarbon monoterpenes have very lower solubility in water at pH=7: less than 5 mg L⁻¹ (they are lipophilic compounds). Because of this, they are not observed in the acidic hydrolates [7, 13]. In cases where the monoterpene hydrocarbons are dominant compounds in essential oils, the compositions of hydrolates are very different from the corresponding oils [12, 16]. However, if monoterpene hydrocarbons are present in hydrolates, their concentration is below the detection limit as only oxygenated compounds are observed [17].

Generally, the chemical structure of the component

(polarity factor) determines the extent of hydrogen bonding with water molecules and hence its degree of solubility in water (at constant temperature) [1]. It appears that in some cases the composition of hydrolates at room temperature may change over a two-year period of storage. Possible hydration processes may occur in the hydrolates during shelf-life [12]. In some cases, the main compounds of hydrolates are relatively stable in acidic water over the two-year observation period [13], while in another changes in the aroma and composition appear and on the intrinsic antimicrobial activity of the compounds contain [8].

As it is visible in Table 2, the similarity between the essential oil and the hydrolate composition depends mainly on the relation of hydrocarbons and oxygenated compounds in the essential oil. In materials in which oxygenated compounds are dominant in the essential oil, the degree of similarity between essential oil and hydrolate is very high. Inversely, when hydrocarbons are main essential oil constituents, hydrolate composition differs significantly from the essential oil composition.

A chemical composition of the essential oil depends on several factors e.g. place of geographical origin, habitat, the moment of harvesting, harvesting season, extraction methods, etc. Typically, the hydrolates include some of the water-soluble components of the essential oil along with other water-soluble plant secondary metabolites [44].

According to the data in Table 2, *Mentha* sp. essential oils are rich in oxygenated monoterpenes and are not rich in sesquiterpene. They contain small amounts of sesquiterpene hydrocarbons, which are absent in hydrolates as hydrocarbons not as sesquiterpenes. *M. piperita* and *M. longifolia* essential oils and hydrolates have very similar profiles. Economic importance is the feature of essential oils not mono- and sesquiterpenes [1, 13,16,18, 30, 31].

There are species with similar dominant compounds in essential oils and hydrolates such as *E. caryophyllata* with eugenol, *O. basilicum* with linalool, and *C. citratus* with geraniol and neral [1]. Furthermore, the dominant compound in *L. alba* carvone [29], in *S. officinalis* 1,8-cineole [9] and in *N. nepetella* nepetalactones [33]. Leaves from different *Eucalyptus* sp. mainly contain 1,8-cineole as the dominant compound, both in essential oil and hydrolate [22,23], while *E. citrodora* contain citronellal and citronellol in the essential oil, as well as in the hydrolate [1].

Several *Rosmarinus officinalis* chemotypes have been reported: α -pinene, cineole, p-cymene and camphor-limonene according to dominant compounds in the essential oil. Differences in the chemical composition may be caused by water solubility of aroma compounds. Camphor and borneol were found both in *R. officinalis* essential oils and hydrolates from Japan [39]. However, in the Columbian sample of *R. officinalis*, camphor and 1,8-cineole were dominant in both, essential oil and hydrolates [29]. These facts indicate the possibility of utilizing hydrolates for medicinal usage in the same purpose

as essential oils [39].

In essential oil of *O. vulgare*, were dominated by hydrocarbons, followed by oxygenated compounds. However, hydrolates were dominated only by oxygenated compounds. These variations are observed due to relatively higher solubility of oxygenated compounds in water over the solubility of hydrocarbons. It can be said that the hydrolates of *O. vulgare* should not be discarded, as usually done by introducing cohobation system in the field distillation unit, in order to improve fresh herb and to minimize the loss of valuable components of the pure compounds (thymol and carvacrol). It can also be used as such for disinfection and cosmetic applications [5]. Furthermore, *T. capitatus* belongs to carvacrol chemotype [42, 43], while *T. vulgaris* belongs to thymol chemotype [29], it is visible in the essential oil, as well as in the hydrolate composition. This confirmed the possibility of using thyme and oregano hydrolates as antimicrobial agents similarly to their essential oils [45].

Lavender (*L. angustifolia*) and lavandin (*L. intermedia*) have the same composition but in different proportions. However, hydrolates of both species have linalool as the dominant compound, but they did not contain any linalyl acetate. This can be explained by the fact that linalool is water soluble while linalyl acetate is insoluble in water [27]. Similarly, both essential oil and hydrolate of *C. aurantium* contain linalool and linalyl acetate as the dominant compounds. However, in different proportions [1].

Citronellol and geraniol were the major compounds of *R. damascena* hydrolate and the essential oil [37]. Piperitone was found in all samples of *P. chimonanthifolium*. This monoterpene is the major compound in all hydrolates and in the essential oils of the stem and leaf, and the second most abundant compound in the essential oil of inflorescence [35]. In case of *C. subhirtella* and *C. serrulata* benzaldehyde was the key component of the essential oils, while benzaldehyde, as well as mandelonitrile was the principal compound of hydrolates [19].

Apart from this, there are some species with completely different composition of essential oils and hydrolates (Table 2), such as **Abies sp.**, *A. canadense*, *C. peregrina*, *D. carota* subsp. *sativus*, *L. porteri*, *M. officinalis*, *M. gale*, *Picea* sp., *Psoralea bituminosa*, *Sideritis raeseri* and *S. puberula* [7,10,11,12, 13,17,21, 34, 36, 40].

Biological activity

The biological activity of hydrolates depends on the presence of components that characterize them with their functional groups [8]. The knowledge of the chemotype of the plant material used for the essential oil extraction, as well as hydrosol is of fundamental importance to understand the mechanisms of its biological activity. However, the investigation of hydrolates biological activity is focused mainly on antimicrobial, antioxidant and antiinflammatory activity.

Table 3. Antimicrobial activity of hydrolates (inhibition zone diameter in mm)

Plant species	Reference	<i>Bacillus amyloquelificans</i>	<i>Bacillus brevis</i>	<i>Bacillus cereus</i>	<i>Bacillus subtilis</i>	<i>Candida albicans</i>	<i>Enterobacter aerogenes</i>	<i>Enterococcus faecalis</i>	<i>Escherichia coli</i>	<i>Klebsiella pneumoniae</i>	<i>Listeria monocytogenes</i>	<i>Micrococcus luteus</i>	<i>Pseudomonas aeruginosa</i>	<i>Proteus vulgaris</i>	<i>Salmonella enteritidis</i>	<i>Salmonella galinarum</i>	<i>Salmonella typhimurium</i>	<i>Staphylococcus aureus</i>	<i>Yersinia enterocolitica</i>
<i>Allium tuberosum</i>	[46]				30										25				
<i>Areca catechu</i> (floral axis)	[47]					11.6			12.6/9.9*									12.9	
<i>Nepeta nepetella</i>	[33]			18.0	16.0	32.0/37.0*		12.0	9.0	7.0	10.0		7.0					18.0	
<i>Areca catechu</i> (flowers)	[47]					12.1			11.2/10.2*									12.7	
<i>Areca catechu</i> (root)	[47]					10.3			9.8/8.5*									11.7	
<i>Citrus aurantifolia</i> (fresh peels)	[48]				4	2		1	4									5	
<i>Citrus citratus</i> (dried leaves)	[46]				6	2		-	3									3	
<i>Citrus odorata</i> (fresh leaves)	[47]				3	-		-	-									1	
<i>Cocos nucifera</i> (floral axis)	[47]					14.0			11.7/6.9*									11.3	
<i>Cocos nucifera</i> (flowers)	[45]					14.2			13.8/6.8*									12.3	
<i>Cocos nucifera</i> (root)	[45]					12.5			12.5/7.4*									7.2	
<i>Cuminum cyminum</i>	[48]	-	14	-	-		26		-/19*	-				-	-	-	-	-	-
<i>Lantana camara</i>	[45]			-								9/10/11*						5/6/7*	
<i>Mentha piperita</i>	[48]			/6/10*											12				
<i>Ocimum gratissimum</i> (dried leaves)	[46]				-	1		1	2									1	
<i>Ocimum gratissimum</i> (fresh leaves)	[46]				2	1.5		-	2									2	
<i>Origanum majorana</i>	[48]								19/18*									22	18
<i>Origanum onites</i>	[48]								17/19*									30	16
<i>Origanum vulgare</i>	[46]	27	15	14	20		14		16/19*	20				19	15	14	20	33/15*	16
<i>Origanum vulgare</i>	[45]								13/15*									21	12
<i>Pimpinella anisum</i>	[45]	-	-	-	-				9/-*	-				-	-	-	-	-/12*	-
<i>Piper nigrum</i>	[46]				10										11				
<i>Salvia fruticosa</i>	[47]				15										13				
<i>Satureja hortensis</i>	[47]	14	15	15	14		16		18/10*	16				16	17	16	15	18/16*	17
<i>Thymbra spicata</i>	[47]	12	16	15	-		-		13/-*	-				14	14	14	-	-/10*	13
<i>Thymbra spicata</i>	[48]				20										15				
<i>Thymus serpyllum</i>	[46]								14/12*									18	14
<i>Thymus vulgaris</i>	[47]								14/12*									17	13

*activity depends on bacterial strain
 **activity depends on hydrolate concentration

Antimicrobial activity

The effectiveness of hydrolates as natural antimicrobials depends on the absolute quantity of their major soluble aromatics [1]. However, antimicrobial potential *in vitro* was tested using a paper disk diffusion method. Data about antibacterial and antifungal activities of hydrolates (values are expressed as inhibition zone in mm) is shown in Table 3. Furthermore, some results showed that hydrolates possess a strong antifungal activity based on the inhibition zone, minimal inhibitory concentration or minimal fungicidal concentration against *Candida albicans*, and phytopathogenic and toxic fungi (Table 4).

Antimicrobial properties of hydrolates depend on the microbial strain and concentration, i.e. dilution [30,33, 38, 45]. Despite a higher concentration needed for the same inhibitory effect, the amount of terpenes supplied with hydrolate was lower than in the essential oil. This means a higher hydro-

late effectiveness that is likely to occur due to the hydrophilic environment promoting a higher terpene availability. Lastly, hydrolate exhibited promising results in the control of fungal growth on paper artwork, suppressing the four tested strains at concentrations of 25-50% [32]. The plant part [51] or date of harvest [20], processing plant material (fresh or dry) [47], as well as the extraction method [20], or formulations such as nanoemulsion [28], also influence the chemical composition and further antimicrobial properties. Apart from this, it is established that hydrolate exhibited considerable antibacterial activities against the Gram-positive bacteria, while Gram negative bacteria were found to be resistant. This is most probably due to its outer membrane [43, 52]. Furthermore, only *L. intermedia* hydrolate formulated in nanoemulsion exhibited activity against *E. coli* (MIC value was 0.75%) and *B. cereus* (MIC value 0.60%), whereas pure hydrolate was inactive on both bacteria strains [28].

Table 4. Antifungal activity based on the minimal inhibitory concentration – MIC ($\mu\text{g mL}^{-1}$)

Plant species	Reference	<i>Aspergillus fumigatus</i>	<i>Aspergillus niger</i>	<i>Aspergillus oryzae</i>	<i>Aspergillus parasiticus</i>	<i>Botrytis cinerea</i>	<i>Candida albicans</i>	<i>Cladosporium herbarum</i>	<i>Fusarium oxysporum</i>	<i>Fusarium solani</i>	<i>Penicillium cyclopium</i>	<i>Penicillium italicum</i>
<i>Anthemis nobile</i>	[49]						>800					
<i>Citrus aurantium</i>	[49]						1600					
<i>Cymbopogon citratus</i>	[49]						400					
<i>Eugenia caryophyllata</i>	[49]						400					
<i>Eupatorium japonicum</i>	[49]						>800					
<i>Eupatorium laciniatum</i>	[49]						>800					
<i>Lavandula angustifolia</i>	[49]						3200					
<i>Lindera umbellata</i>	[49]						800					
<i>Matricaria recutita</i>	[49]						>800					
<i>Mentha crispa</i>	[50]	2.5			5.0	<0.32		0.32	5.0		2.5	
<i>Monarda fistulosa</i>	[49]						50					
<i>Pelargonium asperum</i>	[49]						>800					
<i>Tanacetum vulgare</i>	[49]						>800					
<i>Thymus capitatus</i>	[42]		0.5	0.2						0.2		0.1

Investigations of *Thymus capitatus* hydrolate against *Salmonella enterica* show that this plant hydrolate possesses a significant antimicrobial action against both planktonic and biofilms cells of a common foodborne pathogen. The advantages of using hydrolates to disinfect food-contact surfaces are numerous. It is an aqueous solution which can easily be rinsed out from surfaces, it does not have the strong smell of the essential oil and it is a by-product of the essential oil distillation procedure. Consequently, hydrolates could obviously be of great value to combat biofilms and thus improve product safety not only for the food industries but also for many other industries which experience biofilms-related problems [41].

Thymus thymbra hydrolate exhibits sufficient bactericidal effects on bacterial biofilms of *Staphylococcus simulans*, *Lactobacillus fermentum*, *Pseudomonas putida*, *Salmonella enterica* and *Listeria monocytogenes* that formed on stainless steel. Use of natural antimicrobial agents could provide alternative or supplemented ways for disinfecting microbial-contaminated industrial surfaces [53].

These results suggest that the use of some spice hydrolates as antimicrobial agents may be exploitable to prevent the deterioration of stored foods by bacteria, as long as the taste impact is acceptable in the targeted foods [48]. According to this, hydrolates are considered natural food and feed additives to improve gut health in humans and animals [46].

Apart from antifungal and antibacterial activities, the antiviral activity assay indicated that *Thymus vulgaris*

and *Nepeta cataria* hydrolates reduced the porcine reproductive and respiratory syndrome load *in vitro* significantly. Moreover, the mechanisms of action for both *T. vulgaris* and *N. cataria* hydrolates were in both pre-entry and post-entry steps. These results suggest that both hydrolates have therapeutic potential in the prophylaxis and the treatment of porcine reproductive and respiratory syndrome infection [54].

Antioxidant activity

Hydrolates produced from *Hyssopus officinalis*, *Marubium vulgare* and *Artemisia herba-alba* originating from Morocco expressed a good antioxidant activity [14]. Gaharu hydrolate which is produced during the hydrodistillation of resinous wood part of *Aquilaria sp.* exhibited very low antioxidant activity in comparison to quercetin [55].

Thymus vulgaris hydrolate possesses a high antioxidant activity in comparison to synthetic or natural antioxidant used in the formulation of a cosmetic or phytopharmaceutical product, if it is used as therapeutic active principle, it could also allow the preservation of the product with its antioxidant efficacy [29]. Furthermore, the *Syzygium aromaticum* and *Thymus vulgaris* hydrolates, predominantly consisting of eugenol and carvacrol, respectively, were the most effective as antioxidants for the prevention of lipid peroxidation through the measurement of malonaldehyde produced after degradation of hydroperoxides. In case of scavenging superoxide anion radical *Lavandula officinalis* hydrolate in which linalool prevailed was a stronger antioxidant [56].

The iron reduction assay and free radical scavenging

capacity were used for antioxidant evaluation of *Poliointha longiflora* essential oil, hydrolate and solid waste residues. The essential oil presented the highest biological activities for antioxidant and antimicrobial tests while the hydrolate and the solid waste residues had the lowest antioxidant activity and the hydrolate had the lowest antimicrobial activity. However, the data suggests that *P. longiflora* hydrolate and extracts from solid waste residues can still have compounds with antimicrobial and antioxidant capacities [57].

Antiinflammatory properties

Depending on the method of evaluation of the antioxidant activity, the essential oils and hydrolates showed different results. Independently of the method used, the hydrolates exhibited a lower antioxidant activity than the respective essential oils [20].

Study with *Tetragonia tetragonoides* hydrolate showed that the treatment with this hydrolate inhibited the lipopolysaccharide (LPS)-induced nitric oxide (NO) and prostaglandin E2 (PGE2) production in RAW 264.7 cells by suppressing inducible NO synthase (iNOS) and cyclooxygenase-2 (COX-2) expression, respectively. *T. tetragonoides* hydrolates also inhibited the LPS-induced production of interleukin (IL)-1 β and IL-6 in RAW 264.7 cells. These effects could have been exerted by the inhibition of nuclear factor-kappa B (NF- κ B) activation and phosphorylation of the mitogen-activated protein kinases (MAPK) pathways. These findings provide evidence that *T. tetragonoides* hydrolates exhibits potential anti-inflammatory activities [58].

Rosa damascene hydrolate suppressed neutrophil activation induced by lipopolysaccharide (LPS), tumor necrosis factor alpha (TNF- α), and N-formyl-Met-Leu-Phe (fMLP) at 5–15%. It also reduced the LPS- and TNF- α -induced cell surface expression of the adhesion-related molecule. However, it did not affect the migratory capacity of neutrophils with or without chemoattractant. These results suggest that rose water may reduce the pathogenicity of microbes, and attenuate neutrophil stimulation, which is involved in inflammatory responses. These findings suggest that rose water has a potential effect to inhibit skin inflammation caused by microbes [38].

A pilot study aimed to investigate the preventive effect of *Salvia officinalis*, *Thymus vulgaris* and *Mentha piperita* hydrolates oral rinse used in conjunction with basic oral care on chemotherapy-induced oral mucositis show that these hydrolates show promising results in alleviating oral mucositis, and due to this, hydrolates can be recommended for clinical use as they are well tolerated and cost effective [59].

Application

Due to the increased interest for natural products, it is important to understand biological activities for developing new applications in food, human health, and agriculture against insects and pests. Biological and organoleptic properties of hydrolates make them useful. Typically,

in commercial production, essential oils are skimmed off and the hydrolate is discarded as waste. But they can be cohobated back to the source solution, and in that way reduce the waste and environmental pollution. They could also be further extracted with another lipophilic solvent to obtain the secondary oil in opposition to the primary one that naturally evaporates during distillation. Hydrolates have a much softer scent and are less biologically active than corresponding essential oils. Blending the primary and the recovered oil from hydrolates gave more richness and fullness to the oil [5,25,34].

However, according to the research results, hydrolates had great potential for usage in food industry in drinks, for flavoring fruit beverages, confectioneries and soft drinks, and for food preservation, and as a convenient sanitizing agent while washing fresh-cut fruits and vegetables. Apart from this, they can be used for sanitizing other food products such as freshwater fish, fermented meat products (sausage and suck), as well as for disinfection of food and areas in contact with it. Furthermore, hydrolates are often used in cosmetics and perfume industry, as well as in aromatherapy.

Functional (soft) drinks

Traditionally hydrolates are used as drinks and in food products [4]. For example, sage hydrolate is released from a by-product during the distillation in Turkey and is drunk as a natural antibiotic against various bacterial diseases and digestion disorders [9]. Furthermore, in traditional and folk Persian medicine, hydrolates are used as drinks for medicinal purposes to treat different conditions [60, 61]. For example, fennel hydrolate was suggested to start menstruation while yarrow aromatic water to regulate menstruation. These hydrolate are used for women's reproductive and hormonal health. However, scientific investigation of these hydrolates may lead to the development of some functional beverages and soft drinks as a safe use of essential oils or even new lead components or therapeutic agents [3]. Furthermore, *Trachyspermum ammi* are traditional Persian medicines applied for chronic pain and neural ailments such as tremor, paralysis, and palsy. As a by-product *T. ammi* hydrolate can be considered as an almost pure, but inexpensive and easy producible resource of natural thymol (84.33-95.62%). In accordance with the pharmacological properties of thymol, this preparation can be applied in numerous relevant clinical and phytopharmaceutical approaches [15]. The hydrolate of *Citrus aurantium* flowers, commonly known as neroli, has been used in traditional medicine as a remedy for the treatment of mild depression, sedation and as a heart tonic [62], while *Lavandula angustifolia* hydrolates exhibit revitalizing and relaxing properties when consumed in the form of an additive to water or food [63].

Biopreservation of fresh-cut fruits and vegetables (natural food sanitizers)

Investigations have shown that an abundance of fresh fruit and vegetables that are rich in minerals, vitamins and

phytochemicals in a diet promotes a healthy life and decreases the risk of diseases. However, water washing is not effective itself for extending the shelf life of the product by decontamination. Therefore, several sanitizers are used in fresh-cut industry. Chlorine based chemicals are the most common sanitizers with their low cost and proven effectiveness against pathogens. However, nowadays, consumers demand more natural processing of fresh-cut fruit and vegetables. Up to date, a number of physical and chemical fresh-cut sanitization methods have been tested in order to eliminate the use of chlorine from fresh-cut industry, decrease high water requirement, as well as provide high decontamination efficiency [64]. However, investigations show that hydrolates inhibit tyrosinase – the main enzyme responsible for the browning reaction of fruits and vegetables, at varying levels depending on substrate type and concentration [65,66].

Since fresh or minimally processed fruits and vegetables are established as a source of several outbreaks caused by *Escherichia coli* and *Staphylococcus*, strategies based on natural preservatives are being developed to control and decontaminate. The simple and inexpensive production method of hydrolates and their recognition for safe human consumption also focused on these compounds as antimicrobial components [67]. For these reasons, there are many studies with hydrolate treatment of fresh-cut fruits (apple) and vegetables (iceberg lettuce, carrots, tomato, parsley and cucumbers).

Rosmarinus officinalis and *Thymus vulgaris* hydrolates were found to be the most efficient sanitizers in reducing the *Staphylococcus aureus* numbers. Results

demonstrate that predictive models could be utilized for describing the inactivation or survival of *S. aureus* on fresh-cut apple with the effects of the treatments with plant hydrolates [68].

Inhibitory effects of the hydrolates of *Thymus vulgaris*, *Nigella sativa*, *Rosmarinus officinalis*, *Salvia officinalis* and *Laurus nobilis* were investigated against *Salmonella Typhimurium* and *Escherichia coli* inoculated into apple and carrots. *T. vulgaris* showed the highest antibacterial effects on both *S. Typhimurium* and *E. coli*. *L. nobilis* hydrolate significantly reduced *E. coli* population on apple and carrot samples. In conclusion, it was shown that plant hydrolates, especially *T. vulgaris* hydrolate, could be used as a convenient sanitizing agent during the washing of fresh-cut fruits and vegetables [69].

Hydrolates from *Thymus vulgaris*, *Satureja hortensis* and *Origanum onites* were evaluated for their antibacterial activity against *E. coli* inoculated into fresh-cut tomato and cucumbers. The results suggested that hydrolates from these plants have the potential to be used as natural food sanitizers for fresh cut tomatoes and cucumbers to provide their microbial safety without causing any sensorial defect on treated products or health problems in humans [70].

Table 5: Antibacterial activity of the plant hydrolates on the bacterial strains depend on treatment time (20, 40 and 60 min) log 10 cfu g⁻¹

Plant material	Treated	Reference	<i>Escherichia coli</i>			<i>Listeria monocitogenes</i>			<i>Salmonela Typhimurium</i>		
			20	40	60	20	40	60	20	40	60
<i>Laurus nobilis</i>	Iceberg lettuce	[64]	4.99	4.95	4.84	4.89	4.69	4.57	6.46	6.10	6.05
<i>Laurus nobilis</i>	Apple	[69]	5.20	4.97	4.80				5.15	5.01	5.00
<i>Laurus nobilis</i>	Carrot	[69]	4.65	4.67	4.51				5.68	5.51	5.39
<i>Nigella sativa</i>	Apple	[69]	5.15	5.01	4.97				5.04	5.04	5.06
<i>Nigella sativa</i>	Carrot	[69]	4.60	4.53	4.47				5.61	5.57	5.30
<i>Origanum onoites</i>	Iceberg lettuce	[64]	3.86	3.74	3.70	3.73	3.30	2.89	4.29	4.07	3.60
<i>Rosmarinus officinalis</i>	Iceberg lettuce	[64]	4.95	4.13	4.01	4.89	4.69	4.53	6.02	5.85	5.23
<i>Rosmarinus officinalis</i>	Apple	[69]	4.57	4.49	4.35				5.09	4.97	4.77
<i>Rosmarinus officinalis</i>	Carrot	[69]	4.59	4.58	4.51				5.64	5.67	5.37
<i>Salvia officinalis</i>	Iceberg lettuce	[64]	5.18	4.99	4.69	4.54	4.60	4.25	6.30	6.26	6.15
<i>Salvia officinalis</i>	Apple	[69]	4.82	4.75	4.73				5.14	5.10	4.93
<i>Salvia officinalis</i>	Carrot	[69]	4.62	4.63	4.53				5.64	5.44	5.16
<i>Satureja hortensis</i>	Iceberg lettuce	[64]	3.75	2.75	2.45	3.84	3.11	2.84	3.99	4.04	3.94
<i>Sideritis canariensis</i>	Iceberg lettuce	[64]	5.30	4.69	4.45	4.69	4.60	3.52	6.15	6.11	6.01
<i>Thymus vulgaris</i>	Iceberg lettuce	[64]	3.96	3.47	2.99	3.75	3.30	2.54	4.57	4.04	3.74
<i>Thymus vulgaris</i>	Apple	[69]	4.48	4.44	4.44				4.81	4.81	4.73
<i>Thymus vulgaris</i>	Carrot	[69]	4.73	4.58	4.37				4.82	4.40	4.32

The inhibitory effects of *Salvia officinalis*, *Rosmarinus officinalis*, *Origanum vulgare* and *Thymus vulgaris* hydrolates were evaluated against *Escherichia coli* and *Staphylococcus aureus* inoculated into parsley at high and low concentrations. Following the pathogen inoculations, parsley samples were washed with hydrolates, and the reduction of pathogen counts were determined. Decontamination of foodborne pathogens from fresh-cut vegetables can be achieved by hydrolate treatments [67].

Six hydrolates (*T. vulgaris*, *S. hortensis*, *R. officinalis*, *S. officinalis*, *Sideritis canariensis*, *Origanum onites* and *Laurus nobilis*) were used for decontamination of fresh-cut iceberg lettuce inoculated with *Salmonella enterica*, *Listeria monocytogenes* and *Escherichia coli*. *Thymus vulgaris* and *S. hortensis* hydrolates archived the reduction of all bacterial strains. Hydrolate treated samples, especially with *L. nobilis* and *S. canariensis*, were generally accepted by the panellist. This study confirmed that plant hydrolates could be successfully used for sanitizing fresh-cut lettuce to provide their microbiological safety without causing deep sensorial defects of the products [64].

The application of different hydrolates depends on the treatment time. However, hydrolates used in biopreservation of fresh-cut fruits (apple) and vegetables (iceberg lettuce and carrot) depend on time (20, 40 and 60 min) are summarized in Table 5. Values are expressed as log 10 cfu g⁻¹.

Biopreservation of other food products

The *in vitro* antibacterial activity of the *Thymus serpyllum* and *Syzygium aromaticum* hydrolates were tested against *Aeromonas hydrophila*, *Escherichia coli*, *Pseudomonas aeruginosa* and *Pseudomonas fluorescens* which are responsible for the spoilage of freshwater fish. It is found that these hydrolates were effective against all bacteria. Consequently, it is likely that these plant hydrolates may be used as antimicrobial agents to prevent the deterioration of food products [71].

Antifungal agent *Thymus vulgaris* hydrolate was applied to a fermented sausage and suck surface mycobiota was tested. However, *T. vulgaris* hydrolate did not affect the number of lactic acid bacteria and Mycrococaceae which are important microorganism groups for properties such as taste, aroma and color of fermented sausage. In future studies, hydrolates should be tested at different concentrations against specific mold or yeast species which are undesirable on the surface of fermented sausage and suck [72].

Hydrolates meet the requirements to be used as natural antimicrobials in food industry, also as sanitizing solutions for tools, machines and working surfaces. In fact, being aqueous solutions, the hydrolates could be easily rinsed out from surfaces, and do not have a strong persistent smell, contrarily to essential oils. Moreover, hydrolates may provide valuable alternative pathways for the prevention of biofilms formation on working tools and surfaces [8]. *Lippa palmeri* hydrolate has an antibacterial activity against *Listeria monocytogenes*, exhibiting a

concentration-response type of relationship. It could be used as an active ingredient in the disinfection of food and areas in contact with it [73].

Nowadays, people wish to consume value added products as a part of their everyday meal. The addition of *Cymbopogon citratus* hydrolate in the concentration of 3.5% leads to the production of new naturally flavored herbal ice-cream [74]. Furthermore, increased environmental concerns about synthetic packaging have promoted the development of novel, environmentally friendly edible biofilms. Coating eggs with pectin and *Cinnamomum verum* hydrosol led to lower weight loss during storage, better structural homogeneity and microbial quality during storage time during a six-week period [75].

Aromatherapy and cosmetics

Hydrolate therapy has been evolved as a complementary alternative medicinal modality, with the current resurgence of interest in aromatherapy [44,76]. Hydrolates, as water solutions, are extensively used because they can be easily applied topically without dilution [63,65]. They can be used as facial and body sprays mainly to feel cool and refreshed. Apart from this, the investigations conducted with *Cinnamon zeylanicum* hydrolate show that it has the effect on coagulation, useful in the situation when the wound is not clotting [77].

Depending on aromatic intensity of the hydrosol used, they can prevent body odor, especially if they possess antimicrobial properties, appropriately helpful in minimizing foot odor. Furthermore, they can help to ease itching caused by dry skin and dandruff, as well as wound healing. Apart from this, they are helpful with insect bites, but they also act as insect repellents. In addition, when hydrolates are used in skin care or as massage products, it is important to ensure that the amount of the soluble aromatics is not big enough to cause skin sensitivity [1].

Hydrolates are increasingly replacing water in cosmetics, providing additional active ingredients. Hydrolates are used as the aqueous phase in the production of lotions, creams and soaps, or independently as tonics and air fresheners. The application of hydrolates as raw material in the cosmetic industry results in both reducing expenses for sewage disposal and environmental burden as they are no longer discarded to the environment. Lavender hydrolate used as a replacement for water phase in cosmetics may contribute to maintaining microbiological stability of cosmetic formulations. Furthermore, lavender hydrolate can be directly applied on every type of skin, particularly the impure and oily. It experts cooling, hydrating toning action similar to oil, and it alleviates mental fatigue and stress states [63,65].

Apart from all this, hydrolates are often used as air freshening sprays, due to their therapeutic properties, to reduce stress and anxiety, to help induce a sense of calm. They are also used to help purify and cleanse the living area. They find their application in fragrance sheets, towels and linen, as well as clothes, to eliminate

unpleasant musty, smoky or other odors. They are also used for car fresheners.

Organic agriculture

Using the chemical substances in weed control, diseases and pests, without discernment has been responsible for the environmental damage and human health. For these reasons, in the last years the research has intensified its efforts to find alternative agriculture strategies [78]. Furthermore, post-harvest losses, particularly due to fungal invasions, are much more significant for highly perishable fresh fruits and vegetables than field crops. The use of synthetic chemicals to control post-harvest deterioration of food commodities is restricted, due to their possible carcinogenicity, teratogenicity, high and acute toxicity, long degradation periods, environmental pollution and their effects on human beings. Therefore, alternatives to synthetic pesticides are needed from microbial and plant sources [79].

The investigation of antifungal activity of *Ocimum basilicum*, *Cuminum cyminum*, *Echinophora tenuifolia*, *Rosmarinus officinalis* and *Satureja hortensis* against some plant pathogen fungi (*Rhizoctonia solani*, *Fusarium oxysporum* f. sp. *tulipae*, *Botrytis cinerea* and *Alternaria citri*) *in vitro* shows that *C. cyminum*, *S. hortensis* and *E. tenuifolia*. Furthermore, the hydrolate of *Satureja hortensis* showed a fungicidal effect against *Alternaria mali*, *Sclerotinia sclerotiorum* and *Colletotrichum citricolans*. These results may contribute to the development of environmentally safe alternatives to synthetic preservatives to protect spoilage of food products from pathogenic and saprophytic fungi [79, 80, 81].

Citrus sinensis fruits infected by *P. italicum* were treated *in vivo* with hydrolates. It was established that 0.2 µg mL⁻¹ of *T. capitatus* hydrolate was enough to result in the absence of orange infection and causing 100 % mycelial growth inhibition. This activity can be correlated with the chemical composition of extracts which are rich in carvacrol (more than 69%). Therefore, the preventive and curative effects of *T. capitatus* essential oil and its hydrolate could be exploited as an ideal alternative to synthetic fungicides for treating many fungal phytopathogens causing severe destruction to oranges [42].

The hydrolate of *Daucus carota* subsp. *sativus* roots showed the best inhibition effect against *Penicillium expansum* and *Botrytis cinerea*. The results showed that the treatments of strawberry fruit with roots essential oil and hydrolate extract of *D. carota* subsp. *sativus* presented a very interesting protective and preventive activity on the strawberry against *B. cinerea* mold. They prove to be valuable raw material not only for cosmetic industries but also for post-harvest treatment. They can be used for the development of new and effective methods, which are regarded as safe and eco-friendly to control post-harvest diseases [21].

Apart from potential application as an antifungal agent, the investigations are focused also on insecticidal, as well as nematocidal activity of hydrolates and the

possibilities for their use in organic agriculture both in the field production and during storage. The results of investigations on the influence of *Mentha pulegium* and *M. suaveolens* hydrolates on insect pest of citrus *Taxopera aurantii* have shown a high insecticidal effect, and that *M. suaveolens* hydrolate is more effective against citrus pests than *M. pulegium*. Consequently, these natural compounds can be used in the management of aphids on citrus [76]. Furthermore, hydrosols of *Mentha pulegium* and *Melissa officinalis* had the strongest inhibitory effect against *Myzus persicae*, while *Origanum majorana* caused 10-15% aphid mortality. These results clearly showed that potential of hydrosols in pest control ought not to be ignored and should attract the interest of future studies [82].

The nematocidal activity of hydrolates of *Lavandula intermedia*, *L. luisieri*, *Thymus vulgaris* and *T. zygis* was investigated against root-knot nematode *Meloidogyne javanica*. *In vivo* tests on tomato seedlings at sublethal doses of the hydrolates induced a significant reduction of nematode infectivity. In pot experiments, all hydrolates tested on tomato plants significantly affected the infection frequency and reproduction rate of the nematode population. This study demonstrated that hydrolates could be an exploitable source of the potential waste protection products on root-knot nematodes [83].

Chemical herbicides generally used to control weeds have harmful effects on the environment. Therefore, natural compounds which can be used as a bio herbicide are preferred to replace chemical. The phytotoxic activities of the *Halophyllum tuberculatum* hydrolate were evaluated against *Triticum aestivum* and *Raphanus sativus* seed. It was established that these cultivated plants are resistant to the application of *H. tuberculatum* hydrolates and showed high speed germination. It could be concluded that hydrolates of this plant can be used as biological herbicide with *T. aestivum* and *R. sativus*. Furthermore, hydrosoluble compounds can improve the possibilities of applying them as bioherbicides in the future [84]. The effects of different concentrations of *Ocimum basilicum* hydrolate on germination rate and shoot and root length of *O. basilicum* and *Chenopodium quinoa* seeds have been investigated, too. The results show that all the germination of *O. basilicum* and *C. quinoa* seeds were significantly inhibited under laboratory conditions [85].

Another research was conducted with hydrolates of *Ipomea carnea* leaves and flowers and *Lantana camera* leaves focusing on their effect on the growth of wheat, maize and cotton for 15 days. The hydrolate of *I. carnea* flowers can be suited as a fertilizer for maize plants in limited doses [86]. Since hydrolates have low toxicity, they are biodegradable and inexpensive to produce; the possibility of developing hydrolates for use in crop protection may be attractive [87].

Toxicity

Determination of safety study on *Cyprinus caprio* showed that 0.05 ml L⁻¹ doses of *Eucalyptus camaldulensis*

hydrolate do not create any kind of mental disorder. In fish exposed to 0.10 and 0.15 ml L⁻¹ doses, some mental disorders were observed (rolling, numbness) at the end of 5 h, mortality in the trial fish has been determined [88]. In another study, lethal concentration values of *Cinnamomum zeylanicum* on *C. caprio* were investigated. The results indicated that the hydrolate caused swimming changes, lethargy, lack of breath and leaning towards the depth of the aquariums at all concentrations, while the 1 h lethal concentration value of the safe dose of *C. zeylanicum* hydrolate was estimated at 4.39% [89]. Better survival of tambaqui (*Colossomam acropomum*) juveniles was achieved by adding 5% of *Lippia alba* hydrolate during simulated transport. Based on these findings, it is suggested that 5% of *L. alba* hydrolate is an effective sedative in the aquaculture management [90].

Storage

Because of the small concentration of the essential oil dissolved in hydrolates, they are subjected to microbiological proliferation. The reasons for spoilage have to be found in the nature of substrates supporting growth and of microbiological contaminants. Non-volatile water-soluble organic compounds (thermally stable) were likely carried over during distillation by a priming and foaming effect and could be used as nutrients by microorganisms. Contamination by mesophilic bacteria and yeast was observed in the hydrolates [91]. Microbial proliferation at room temperature is commonly observed in hydrolates when stored in a non-sterile container [92]. However, the essential oil concentration in hydrolates or cold storage are not sufficient to ensure microbiological stability. Additional hurdles such as chemical preservatives or aseptic packaging will be necessary to ensure microbial stability [93].

Conclusion

Hydrolates have widely been used in cosmetic industry as replacements for water in lotions, creams, soaps, tonics. Hydrolates can be used in drinks, for flavoring fruit beverages, confectioneries and soft drinks, for food preservation and as a convenient sanitizing agent during the washing of fresh-cut fruits and vegetables. Furthermore, they can be used in organic agriculture for decreased post-harvest losses during storage, as well as in the field production as nematocidal agents, or for weed control. However, their popularity is still on the rise, especially in aromatherapy.

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Izvod

HIDROLATI – NUSPROIZVODI PRILIKOM DESTILACIJE ETARSKIH ULJA: HEMIJSKI SASTAV, BIOLOŠKA AKTIVNOST I MOGUĆNOSTI UPOTREBE

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Hidrolati, koji se takođe nazivaju i hidrosoli, cvetne ili aromatične vode, kao i destilati, proizvode se u procesu destilacije etarskih ulja vodenom parom. Mala količina etarskog ulja nalazi se rastvorena u hidrolatima dajući im specifične organoleptičke osobine i ukus, kao i biološku aktivnost, što ih čini korisnim kao sirovinu u mnogim industrijama. Njihova popularnost poslednjih godina raste, posebno u aromaterapiji. Cilj ovog preglednog rada je da se analizira hemijski sastav hidrolata i njihovih etarskih ulja, kao i biološku aktivnost hidrolata (antimikrobna, antioksidativna i antiinflamatorna), ali i mogućnosti upotrebe, ne samo u prehrambenoj industriji kao prirodnih aroma i konzervanasa sveže isečenog voća i povrća, ali i funkcionalnih (bezalkoholnih) napitaka. Takođe, hidrolat i mogu da se koriste u aromaterapiji i kozmetici kao i u organskoj poljoprivredi i ribarstvu.

Ključne reči: Hidrolati, Hidrosoli, Aromatične vode, Cvetne vode, Destilati