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QUALITATIVE COMPOSITION AND ORGANIC ACIDS CONTENT IN THE ABOVEGROUND PART OF PLANTS FROM FAMILIES *LAMIACEAE*, *ASTERACEAE*, *APIACEAE* AND *CHENOPODIACEAE*

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Background. *Organic acids are the compounds of aliphatic or aromatic orders, which are widespread in flora and have a wide range of biological activity. We studied the qualitative composition and quantitative contents of organic acids in the aboveground part of some unofficial medicinal plants from families Lamiaceae, Asteraceae, Apiaceae and Chenopodiaceae is relevant.*

Objective. *The objects of the research are the aboveground part of unofficial medicinal plants from families Lamiaceae, Asteraceae, Apiaceae and Chenopodiaceae.*

Methods. *Identification of organic acids was performed by means of thin-layer and paper chromatography, their content was determined by means of gas chromatography, the quantitative amount of organic acids was defined by titrimetric analysis.*

Results. *In the studied raw plants the quality of organic acids and their total contents were determined (in terms of malic acid). It is established that the maximum content of organic acids is accumulated in the grass Hyssopus officinalis L. (Lamiaceae), and the minimal is in the leaves of Chrysánthemum xhortorum L. variety Apro (Asteraceae). In all studied raw plants the dominance of aliphatic acids (citric, malic, oxalic and malonic) was determined by means of gas chromatography. Benzoic is predominant among the aromatic acids.*

Conclusions. *In the studied raw plants the quality of organic acids and their total content were determined. The following results can be used in developing the methods of quality control of the studied raw plants and during the study of new bioactive substances.*

KEY WORDS: **organic acids, *Lamiaceae, Asteraceae, Apiaceae, Chenopodiaceae,* grass, leaves, thin-layer chromatography, gas chromatography, paper chromatography.**

Introduction

Organic acids are the biologically active substances which are in plants in the free state, in the form of salts, esters, dimers and compounds with other substances. They are intermediate products of plants' metabolism: involved in the oxidation of carbohydrates, fats, amino acids and proteins; used in the synthesis of amino acids, alkaloids, steroids, etc. [1, 2].

Organic acids have a wide range of biological effects. They enhance the secretory and motor activity of the digestive tract, improving digestion; help to reduce nitration processes in the organism and to reduce chemical carcinogenesis; raise the protective strength and vitality of the organism. The antioxidant, antiallergic, anti-inflammatory, antiseptic properties of these compounds are established [2].

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According to the information above, we consider that the study of the qualitative composition and quantitative contents of organic acids in the aboveground part of some unofficial medicinal plants from families *Lamiaceae*, *Asteraceae*, *Apiaceae* and *Chenopodiaceae* is relevant and is of significant scientific and practical interest [3–5]. This will make it possible to justify the use of these plants in the future pharmaceutical researches.

The aim of our research is to define the qualitative composition and quantitative contents of free organic acids in the aboveground part of medicinal plants from families *Lamiaceae (Hyssopus officinalis* L., *Lophanthus anisatus* L.*)*, *Asteraceae (Bellis perennis* L. — cultivated, *Tagetes tenuifolia* Cav., *Chrysánthemum xhortorum* L. variety *Apro), Apiaceae (Angelica sylvestris* L.*)* and *Chenopodiaceae (Chenopodium album* L.*)*. The leaves of *Chrysánthemum xhortorum* and *Angelica sylvestris* and the grass of the rest species were used for phytochemical analysis. Raw plants for research were harvested during their mass flowering.

Materials and Methods

Identification of free organic acids in raw materials was performed out by means of thin-layer chromatography (TLC), paper chromatography (PC) and gas chromatography (GC) according to [6, 7]. For TLC and PC the aqueous extracts of raw plants were prepared. As standard samples for PC and GC benzoic, oxalic, malic, tartaric, succinic, salicylic, citric acids and the following solvent systems: n-butanol-formic acid-water (10:1:4), 95% ethanol-chloroform-concentrated solution of ammonia-purified water (70:40:20:2); 95% ethanol-concentrated solution of ammonia (16:4.5) and ethyl acetate-formic acid-water (3:1:1) were used. The chromatograms were developed after drying in 0.05% alcohol solution bromothymol blue and 0.1% solution of 2,6-dichlorophenolindophenol sodium salt hydrate. The action of ammonia vapours on chromatograms after a few seconds improved the contrast of spots.

The quantitative contents of organic acids in aqueous extracts of raw plants were determined according to [7] by titrimetric method. The contents of free organic acids (X) in terms of malic acid in absolutely dry raw materials in percentage were calculated by the formula:

$$X = \frac{V \times 0{,}0067 \times 250 \times 100 \times 100}{m \times 10 \times (100 - W)},$$

where: V — volume of 0.1 M sodium hydroxide solution consumed on titration, ml;

0.0067 — the amount of malic acid corresponding to 1 ml of 0.1 M sodium hydroxide solution;

m — mass of raw material, g;

W — loss in weight because of drying, %.

The quantitative contents of individual organic acids were defined by modified methods for determining the fatty acids in raw plants with further detection of organic acids. It is based on getting acid methyl esters (fatty, organic, phenolic) by methylating agent and their removal for further chromatographing by means of the gas chromatograph Agilent Technologies 6890 N with mass spectrometric detector 5973 N. Methyl esters of organic acids were obtained by a modified method of A. Carrapiso [8].

Results

3–5 organic acids were identified in the raw plants of every studied species by TLC and PC methods (Table 1).

According to Table 1, all studied species contain citric, oxalic and malic organic acids; tartaric, salicylic, benzoic and succinic acids were found only in some representatives.

The results of quantitative contents of organic acids determination (in terms of malic acid) are shown in Table 2. According to Table 2, the maximum contents of organic acids accumulate in the grass *Hyssopus officinalis*, the lowest is in the leaves of *Chrysánthemum xhortorum*.

The component composition and quantitative contents of individual organic acids in the aboveground organs of some studied species was defined by means of GS method (Table 3). 13 organic acids in raw plants of *Hyssopus officinalis*, 19 — in *Lophanthus anisatus, 5* — in *Bellis perennis, 5* — in *Chrysánthemum xhortorum, 18* — in *Chenopodium album*, 15 — in *Angelica sylvestris were determined*.

Discussion

According to the results, aliphatic organic acids (citric, oxalic, malonic and malic) were defined in all studied raw plants by means of the methods of thin-layer, gas and paper chroma-

Table 1. Results of Organic Acids Identification in Raw Plants from Families *Lamiaceae, Asteraceae, Apiaceae* and *Chenopodiaceae*

Acid	Types of plants						
	Hyssopus officinalis (grass)	Lophanthus anisatus (grass)	Bellis perennis (grass)	Tagetes tenuifolia (grass)	Chrysánthemum xhortorum (leaves)	Chenopodium album (grass)	Angelica sylvestris (leaves)
succinic	+	+	–	+	–	+	traces
tartaric	–	–	traces	–	–	–	–
citric	+	+	+	+	+	+	+
salicylic	–	traces	+	–	–	–	traces
oxalic	+	+	+	+	+	+	+
malic	+	+	traces	+	+	+	+
benzoic	+	+	–	–	–	+	–

Note: "+" — labels identified compounds, "–" — labels unidentified compounds.

Table 2. Quantitative Contents of Organic Acids in Raw Plants from Families *Lamiaceae, Asteraceae, Apiaceae* and *Chenopodiaceae*

Types of plants	Hyssopus officinalis (grass)	Lophanthus anisatus (grass)	Bellis perennis (grass)	Tagetes tenuifolia (grass)	Chrysánthemum xhortorum (leaves)	Chenopodium album (grass)	Angelica sylvestris (leaves)
Contents of acids, %	3,26±0,03	2,49±0,02	0,69±0,01	2,78±0,16	0,34±0,05	2,37±0,03	0,69±0,29

Table 3. Contents of Main Organic Acids in Raw Plants from Families *Lamiaceae, Asteraceae, Apiaceae* and *Chenopodiaceae*

Acid	Component Content, mg/kg					
	Hyssopus officinalis (grass)	Lophanthus anisatus (grass)	Bellis perennis (grass)	Chrysánthemum xhortorum (leaves)	Chenopodium album (grass)	Angelica sylvestris (leaves)
oxalic	84	370	70	310	20257	0,69±0,29
malonic	3731	3578	1069	557	963	1066
succinic	82	214	–	–	727	37
benzoic	466	101	–	–	81	–
malic	946	590	–	2797	297	1101
citric	3063	1500	438	1562	792	1122

tography. The detection of organic acids is a topical issue of phytochemical researches. The study on composition and contents of these compounds in raw plants was not the urgent matter before [2, 9, 10].

Citric acid is very widespread in nature and is used in medicine as a part of drugs to improve energy metabolism; malic and oxalic acids are used in food industry [2, 9]. We consider that among all investigated objects the grass *Chenopodium album* is the most promising source of oxalic acid, and the grass *Hyssopus officinalis* — of citric and malic.

Malonic acid is the predominant organic acid in all investigated raw plants except the grass *Chenopodium album*. This dicarboxylic acid is an important component of biochemical reactions in a plant organism and a precursor in the synthesis of cinnamic acids and flavonoids; its significant contents indicates the level of metabolism in plants during the preparation of raw plants (in the period of their flowering). Its accumulation in plants depends on the intensity of photosynthetic activity, enzymic reactions, temperature, etc. Studying the role of this acid in plants and its effect on the biological activity of phytosubstances of plants is a promising area for scientific investigations [11].

Benzoic acid is quantitatively dominant among the aromatic acids in the grass family *Lamiaceae* (Table 3), which is rather useful in pharmacy because it has anti-inflammatory, antibacterial and immunotropic properties [2]. the grass *Hyssopus officinalis* is the most promising for benzoic acid among the studied species.

Conclusions

The quantitative content of organic acids was studies and determined in the aboveground parts (grass or leaves) of the plants from families *Lamiaceae*, *Asteraceae*, *Apiaceae* and *Chenopodiaceae*.

The component composition of free organic acids in raw plants from families *Lamiaceae* (*Hyssopus officinalis*, *Lophanthus anisatus*), *Asteraceae (Bellis perennis* — cultivated, *Tagetes tenuifolia*, *Chrysánthemum xhortorum* variety *Apro)*, *Apiaceae (Angelica sylvestris)* and *Chenopodiaceae (Chenopodium album*) were analysed by means of gas chromatography for the first time. The dominance of aliphatic acids (citric, malic, oxalic and malonic) was determined. The following results can be used to develop the methods of quality control of the studied raw plants and during the study of new bioactive substances.

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