PHARMACIA

Pharmacia 69(3): 865–872
DOI 10.3897/pharmacia.69.e87549

Research Article

# Assessment of morphological pharmacognostic characteristics of the content and label information of dried herbs marketed as food supplements in Bulgaria

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**Received** 8 June 2022 ♦ **Accepted** 24 July 2022 ♦ **Published** 16 September 2022

**Citation:** Gavrilova A, Gavrilov G (2022) Assessment of morphological pharmacognostic characteristics of the content and label information of dried herbs marketed as food supplements in Bulgaria. Pharmacia 69(3): 865–872. https://doi.org/10.3897/pharmacia.69.e87549

## Abstract

According to the current food legislation in Bulgaria the dried herbs are classified under the large group of food supplements and their trade is allowed in pharmacies, drugstores and grocery stores. The aim of this study is to assess the morphological pharmacognostic characteristics of the content and the additional information on the labels of food supplements containing dried herbs in Bulgaria, in the light of key standard and regulatory documents related to the quality of herbal substances and food supplements. 91 herbal substances of 10 main groups were studied, which included 103 commercial products from 8 companies. The authenticity of the herbal substances was confirmed with macro-and microscopic tests. The macroscopic morphological indicators which were used as elements of trade-commodity analysis were changes in color, presence of other parts of the same or different plants, organic and mineral impurities, evidences of diseases and pest infestations. The phenological phase in which the herbal substances had been collected was determined also. We found that very small part of the studied products meet the standards for all of the selected criteria according to the considered documents which implies the need of strengthening control.

## Keywords

dried herbs, food supplements, morphological pharmacognostic characteristics, quality control

## Introduction

Medicinal plants are a national asset of great environmental, socio-economic and cultural importance. Bulgaria is one of the main exporters of high quality medicinal plants in Europe (Evstatieva et al. 2007). Much smaller part of the annual production of medicinal plants in the form of dried herbs is sold on the internal market. However, the consumers' demand for these products remains relatively constant over the years and its met by a short supply chain composed of small and medium-size manufacturers, distributors, pharmacies and drugstores. This sustainable interest in dried herb products can be attributed on one hand to the deep traditions of Bulgarian folk medicine and on the other to the still resonating in the society echo of a long-standing targeted policy for utilization of medicinal plant resources in the interest of public health, conducted between the 1960s and 1990s in Bulgaria (Hardalova et al. 1998). It should be mentioned also the ever-growing interest among consumers in alternative sources of pre-

vention and treatment of health issues, which appears to be a significant marketing factor at present (Glynn and Bhikha 2019). Currently, the local market is abundant of products containing dried herbs of single species or combinations of species, commonly known as „Herbs" and "Herbal teas" respectively. According to the Food Act (2020), these products are classified under the large group of food supplements and their trade is allowed in pharmacies, drugstores and grocery stores. The chain quality control of these herbal products from harvesting (from nature or culture) through primary processing, packaging, labeling, distribution and sale is divided between three ministries - Ministry of Environment and Water, Ministry of Agriculture, Food and Forestry, Ministry of Health and their substructures.

The aim of this study is to assess the morphological pharmacognostic characteristics of the content and the additional information on the labels of food supplements containing dried herbs in Bulgaria, in the light of key standard and regulatory documents related to the quality of herbal substances and food supplements.

## Materials and methods

91 herbal substances belonging to 10 main groups - Herba, Folium, Flos, Radix, Rhizoma, Cortex, Pericarpium, Semen, Fructus and Galbulus were studied. The substances were included in 103 commercial products from 8 companies which were purchased in 2019 from drugstores and pharmacies in Pleven, Bulgaria. The net weight of each product was between 40 and 50 g. All these products belong to the group of „food supplements" according to § 1, item 4 of the additional provisions of the Food Act (2020).

To determine the botanical taxa to which the studied dried herbs belong key reference sources were used (Jordanov 1963–1979; Valentine and Chater 1972; Webb 1972; Moore et al. 1976; Velčev 1982–1989; Kožuharov 1995; Delipavlov and Cheshmedzhiev 2003; Peev 2012). For all species whose herbal substances are included in European Pharmacopoeia the corresponding monographs were used to confirm their botanical affiliation and authenticity through macro- and microscopic tests. For the microscopic examination the herbal substances were grinded to crude powder and then reduced through a sieve number 355. Chloral hydrate was used as a clearing solution. Temporary microscopic slides were mounted using the fine fraction of the powdered herbal drugs and glycerine as media.

The macroscopic morphological indicators which were used as elements of trade-commodity analysis were changes in color, presence of other parts of the same or different plants, organic and mineral impurities, evidences of diseases and pest infestations. The phenological phase in which the herbal substances had been collected was determined also. All macroscopic observations were done by examining the content of the whole product packages.

The names of plant substances were cross-referred with Pharmacopoeia Europaea 10.0, Boeva et al. (1975), Assenov et al. (2019).

## Results and discussion

The studied products were obtained from plants referred to 47 families, 86 genera and 86 species.

In Suppl. material 1, column 3 are enlisted all investigated plant species given by the producers with authors' corrections of their botanical names where needed. In column 2 we indicated the affiliation to a botanical family of the enlisted species. In column 4 are given the correct names of the herbal substances based on the referenced literature. The data in columns 5 to 13 refer to the assessment of content and additional information on the labels of the tested products.

Column 5 represents the correspondence between the plant substances and the plant species from which they had been derived as originally labelled by the producers themselves. We found that in 4 commercial products (3.9%) the herbal substances originated from plant species other than the labelled ones in violation of the provisions of Ordinance №5 of 19 July 2004 on the requirements for the herbal procurement facilities and warehouses for herbs and the Food Act (2020). This is why in Suppl. material 1 the indices for these four species refering to columns 6 to 12 are marked as N/A (not available). In one product with a claim to be *Echinops ritro* L. we found inflorescences (secondary heads) from *E. microcephalus* Sibth. & Sm. The only species of this genus included in the appendix of the Medicinal plant Act (2000) is *E. spaerocephalus* L. Assenov and Nikolov (1988) considered *E. ritro* L. and *E. bannaticus* Rochel ex Schrader to be medicinal too. *E. microcephalus* differs clearly from the rest of the mentioned above species in the hairs at the base of the involucrium, which are few in number and five times shorter than the involucrium itself. In the other mentioned species the hairs are numerous and two-to-four times shorter than the involucrium (Delipavlov and Cheshmedzhiev 2003). It is worth mentioning that the commonly used herbal substance from the medicinal *Echinops* species are not the inflorescences, but the achenes, which are rich in quinoline alkaloids and have neuromuscular stimulating effect (Horn et al. 2008).

In other product with a claim to be *Equisetum arvense* L. we identified *E. telmateia* Ehrh. The main stems of *E. telmateia* are about 10–20 mm in diameter, whitish in colour, almost smooth, with 20–40 fine grooves. The sheaths are pale below, with dark teeth on top and hair-like, easily falling endings. The lowest internode of each branch is shorter than or equal to the adjacent sheath of the stem. In comparison *E. arvense* has sheaths that are green below and often with brown teeth on top. The lowest internode of each branch is longer than the adjacent sheath of the stem (Jordanov 1963; Tutin 1964; Delipavlov and Cheshmedzhiev 2003; Assenov

et al. 2019). According to Medicinal plant Act (2000) and European Pharmacopoeia 10.0 the eligible for use species of genus *Equisetum* is *E. arvense*. All other species of the genus are not allowed as plant substances due to insufficient *in vivo* toxicity studies and lack of consensus on effective and toxic dosage although they possess potent pharmacological properties (Boeing et al. 2021). In the other two cases, *Solidago gigantea* W. Aiton was observed instead of *Rhodiola rosea* L. and *Echium vulgare* L. instead of *Digitalis lanata* Ehrh. as both plant substances were thoroughly cross-referred with appropriate literature sources (Velčev 1982; Delipavlov and Cheshmedzhiev 2003; Barykina and Alyonkin 2016; European Pharmacopoeia 10.0). The plant substance that proved to be herba from *Solidago gigantea* had the following characteristic features - many rarely to densely pubescent stem particles corresponding to the upper part of the stems and few glabrous and smooth stem particles typical for the lower part of the stems. The upper leaf surface was green and generally glabrous (with sparse single hairs) though the lower leaf surface was greyish-green and pubescent, especially on the veins. Capitulum 3–6 mm in diameter, with single row of yellow ray florets about the same length as the involucre and yellow, tubular florets, slightly shorter, than the ray florets. The best part of the capitula were in late flowering stage (Fig. 1). For

**Figure 1.** Capitula of *Solidago gigantea* in the phaenological stage of late flowering.

comparison with the other two pharmacopoeial *Solidago* species: *Solidago canadensis* L. differes from *S. gigantea* in generally glabrous stems and equal in length ray- and tubular florets, also equal to the involucre bracts, while *Solidago virgaurea* L. has twice larger capitula in diameter and twice longer ray florets than the adjacent tubular florets. The plant substance that appeared to be herba of *E. vulgare* had grooved, hispidous stem fragments. The leaf blade fragments had moderate mixed strigose adaxial side and hispid abaxial side. In general the hairs are simple and glandular. Some simple hairs were sessile and others subtended by 'rosettes'. The flowers were zygomorphic, with whitish at the base straight tube and blue corolla lobes, different in shape and size. The 2-branched style and the stamens were longer than the corolla (Fig. 2).

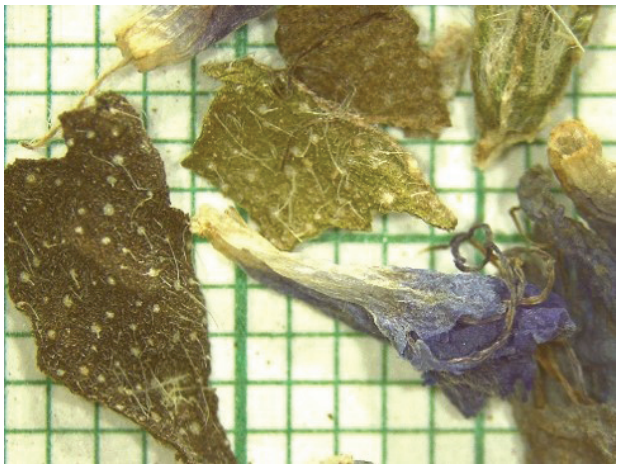

**Figure 2.** Leaf blade and flower segments of *Echium vulgare*.

In 33% of the studied products we found spelling errors in the Latin names of the herbal substances or their absence from the label information (Suppl. material 1, column 6). In many of these cases outdated nomenclature of the herbal substances was used according to the requirements of European Pharmacopoeia and the European Medicines Agency, e.g. Herba Achilleae (incorrect) - Achilleae herba (correct) (EMEA and CHMP 2010).

In regard with the authenticity of the herbal substances, in 9.71% of the cases the labelled substance did not correspond to packaged content (Suppl. material 1, column 7). One product contained whole secondary heads of *E. microcephalus* instead of the pharmacologically active achenes (Boeva et al. 1975; Horn et al. 2008). Other product containing Solidaginis herba was labelled incompletely and inaccurately only with the common Bulgarian name "zlatovrah" for golden root (*Rhodiola rosea* L.) which is famous for the pronounced adaprogenic properties of its rhizomes (Khanum et al. 2005; Panossian et al. 2010). We found flowering stems instead of just leaves in two products labeled as Convallariae folium and Malvae folium. Ordinance №2 / 20.01.2004 on the rules and requirements for the collection of herbs and genetic material from medicinal plants specifies further that the leaves of *Convallaria majalis* must be collected before flowering, and those from *Malva sylvestris* during flowering. For two products – Cichorii herba and Rutae herba there is strong reason to believe that the whole aerial parts had been collected instead of just the upper parts of the stems. In these two cases we found a significant share of basal stems parts with bigger size, stronger development of mechanical tissues and in Rutae herba also with presence of peridermis. In the products indicated as Calendulae flos and Cyani flos, were found the whole heads (Calendulae anthodium and Cyani anthodium) instead of the ray florets only. The plant substance designated as Rosae flos contained the whole flowers with sepals instead of just petals of *Rosa damascena*. We also found some differences between the

plant substance definitions of European Pharmacopoeia, which we mainly followed here and the guidelines for gathering herbs according to Ordinance №2 / 20.01.2004 on the rules and requirements for the collection of herbs and genetic material from medicinal plants currently in force. For the herbal substances of some large herbaceous plants the legal provisions of Ordinance №2 / 20.01.2004 appear to be stronger and require gathering only of the leafy tops of plants like *Verbena officinalis* and *Leonurus cardiaca* while European Pharmacopoeia allows the harvest of the whole aerial parts of these species.

Regarding the phenological phase at the time of harvest, about 25.2% of the investigated plant substances showed evidence that they had been collected later than the recommended period according to Ordinance №2 / 20.01.2004 (Suppl. material 1, column 8). In a product that contained Pini turiones, all turions were completely shattered. The reasons for this may be overdevelopment of the turions before processing, collecting of turions larger than 5 cm, which in the process of drying develop further and shatter or using inappropriate for this case drying method like sun drying (Assenov et al. 2019). The following substances had been collected during fruit bearing instead of flowering: Achilleae herba, Bursae-pastoris herba, Polygoni avicularis herba, Adonidis herba, Agrimoniae herba, Cichorii herba, Verbene herba, Euphrasiae herba, Polygoni hydropiperis herba, Rutae herba. In these herbal substances we did not find any fragments of petals, only fruits and in the case of *Cichorium intybus* – just empty receptacles. In the substance Robiniae flos we found that the best part of the flowers had formed immature legumes with withered sepals and some petals still attached. In the substance Crataegi folium cum flore predominated flowers in late blooming instead of initial flowering as evidenced by the advanced development of hypanthia and completely detached petals (Assenov et al. 2019). In late flowering instead of full flowering had been harvested the herbal substances Absinthii herba, Solidaginis herba, Meliloti herba, Centaurii herba, Veronicae herba, Calendulae flos, Cyani flos, Helichrysi flos and Lupuli flos. In the substance Coryandri fructus the presence of a large number of splitted in two fruits indicated that their collection occurred after the recommended stage of milk ripeness. In the herbal substance Rosae flos predominated fully opened flowers instead of flower buds.

In 35.9% of the tested products the color of the plant substances deviated visibly from the accepted standards (Suppl. material 1, column 9). According to the European Pharmacopoeia 10.0 and Bulgarian State Standard (according to Assenov et al. 2019), the amount of altered parts that had lost their natural color should be less than 2–3%, with small exceptions such as Asperulae herba, where the acceptable amount is up to 7%. In almost all of the observed cases the entire package content presented significant color change. The main biologically active compounds in the relevant substances belong to the following groups - essential oils, phenolic compounds, alkaloids, saponins, iridoids, cardiac glycosides, polysaccharides and mucilaginous substances, vitamins, amines and peptides. It is well known that the color loss after harvest of plant substances is due to enzymatic and non-enzymatic oxidative reactions which cause degradation of pigments as chlorophylls, anthocyanins, carotenoids, other phenolic compounds, essencial oils, carbochydrates, etc. The course of these oxidation processes, which take place in herbs mostly prior or during primary processing, is initiated by critical amounts of light and/or heat in presence of moisture and pH influence (Li et al. 2018; Calín-Sánchez et al. 2020; Thamkaew et al. 2021). The most significant loss of color and decrease in phytochemical content of herbal substances happens during processing and it is linked to the use of conventional drying techniques (Calín-Sánchez et al. 2020). An occasional development of microflora as bacteria (*Bacillus*, *Pseudomonas*, *Agrobacterium*, etc.) and molds (*Chaetomium*, *Fusarium*, *Aspergillus*, *Penicillium*, etc.) as a result of inappropriate harvesting, drying and/or packing of herbal materials, can cause additional selective metabolization of various biologically active compounds such as pectin, sugars, essential oils, phenolic compounds, alkaloids and glycosides by these organisms (Prusinowska and Śmigiełski 2015). Again, these processes are visually presented by color loss.

Impurities were found in 53.4% of the tested substances (Suppl. material 1, column 10) and their amount exceeded the permitted limits according to the European Pharmacopoeia 10.0 (2.8.2) and BDS standarts (according to Assenov et al. 2019). In the herbal substances that consisted of fruits, e.g. fruits of *Coriandrum sativum*, *Crataegus monogyna* and *Prunus spinosa*, the impurities were fruits of the same species with a damaged skin, altered colour or compromised by pests. Regarding the leaves as an herbal substance, impurities were associated with their discoloration, the presence of twigs, roots and fruits of the same or other plants, inorganic impurities such as soil and small stones. For two of the products containing leaves, we were able to identify impurities up to the level of a specific botanical species as follows: in the substance Uvae ursi folium there were leaves of *Vaccinium vitis-idaea* and *Pinus sylvestris*, and in the substance Rosmarini folium there were broken stalks of *Equisetum arvense*. In the case of Matricarie flos, the impurities were different parts of other grassland species like *Alopecurus pratensis*, *Bromus mollis*, *Bromus sterilis*, *Elymus repens*, *Melilotus* sp., *Erodium cicutarium*, *Papaver rhoeas*, *Capsella bursa-pastoris*, *Anthemis austriaca*. The impurities occurring in the substances of roots and rhizomes were mostly of inorganic nature and secondly underground parts of the same or other plant species, changed in colour. In the case of stems, flowers and inflorescences, the impurities were from altered colour parts of the same substances, other organ fragments of the same plants (highly lignified stems, immature or ripe fruits, etc.), fragments of other plants and inorganic impurities. In one of two tested products of Verbasci flos, obtained from different manufacturers, more than 70% of the content weight was due to the sepals and fruits of unidentifiable in that form *Verbascum* species

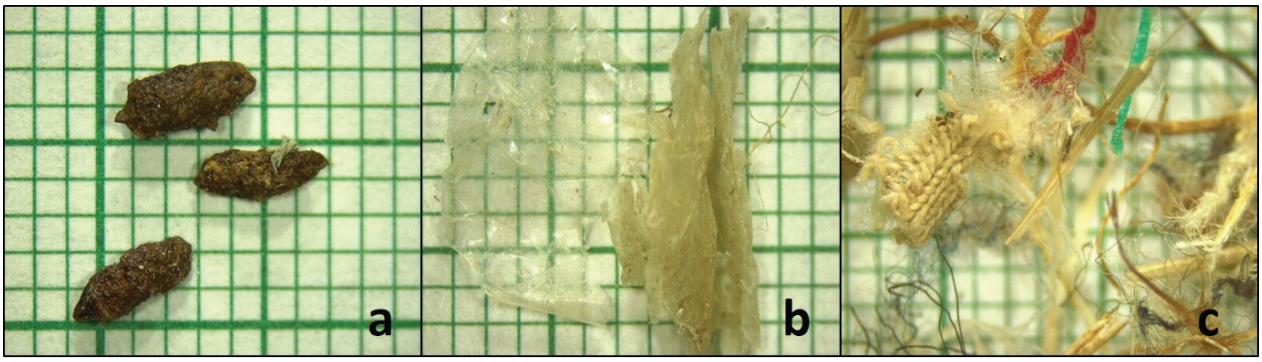


**Figure 3.** Impurities of mouse droppings (**a**), residues of plastics and polyethylene (**b**), textile fibres and fabrics (**c**).

instead of just petals. In the other product the petals prevailed but their colour was significantly darkened in up to 50% of the content. The presence of even small amounts of some other impurities, such as mouse droppings, residues of plastics, polyethylene, textile fibres and fabrics with various degree of degradation in some products (Fig. 3), is very worrying. This indicates that there are some cases of poor control over the collection sites of herbal substances, which possibly may fall even within areas of unregulated landfills, the hygiene prior or after the primary processing of raw plant materials till storage and packing, andthe presence of pests.

In 20.4% of the tested substances we found a deterioration in their quality as a result of the activity of pests in violation of the provisions of Ordinance №5 of 19 July 2004 on the requirements for the herbal procurement facilities and warehouses for herbs and the Food Act (2020) (Suppl. material 1, column 11). In 12% of cases, the damage was caused by *Lasioderma serricorne* (Fabricius, 1792) (cigarette beetle) and *Stegobium paniceum* (Linnaeus, 1758) (drugstore beetle) which worldwide distribution includes Bulgaria (Borowski and Zahradnik 2007; Tomov et al. 2009; Cvetkovska-Gjorgievskaet et al. 2019). The plant substances that were damaged to various degree were aerial parts (Solidaginis herba, Polygoni avicularis herba, Veronicae herba, Hyperici herba, Achilleae herba), flowers (Robiniae flos, Matricariae flos, Calendulae flos), leaves (Urticae folium) and dry dehiscent and indehiscent fruits respectively as Coriandri fructus and Silybi mariani fructus. Whole dead beetles, parts of them and a large amount of frass were found in the packings of the listed substances. Furthermore, the packings were damaged as covered with many small and rounded holes, drilled by the insects. These two beetles are one of the most serious pests of stored products worldwide and particularly often exploit dried materials of tobacco and herbs (Cabrera 2002; Edde et al. 2012; Cao et al. 2019). Kim et al. (2017) found that infestation with *L. serricorne* leads not only to damage of the product presentation and hygiene violation, but also to reduction of active ingredients of the medicinal herbs which in our opinion can be attributed also to the impact of *S. paniceum* also. The leaves of *Malva sylvestris* L. were highly infected by *Puccinia malvacearum* Bertero ex Mont. The leaves of this medicinal plant contain mucilage in ideoblasts or cavities (Classen and Blaschek 1998; Classen et al. 1998). *P. malvacearum* is an autoecious rust fungus that seems to stimulate mucilage production by the host plant but at the same time destroys large part of its tissues (Classen et al. 2001). The leaves of *Tussilago farfara* L. were infested by *Coleosporium tussilaginis* (Pers) Lev. in uredinial stage. The aecial stage of this rust is parasitic on two-needle pines (*Pinus* spp.) (Helfer 2013). This leads to the logical conclusion that the harvesters of coltsfoot leaves should avoid pine forests and their fringes as sourcing sites. Another rust species that commonly infests *T. farfara* and can be expected to appear in more detailed studies is *Puccinia poarum* Niels in its pycnial and aecial stages (De Nooij et al. 1995). In the herbal substances Verbenae herba and Cichorii herba were found mouse excrements. The substances Juniperi galbulus, Rosae pseudo-fructus, Crataegi fructus, Pruni spinosae fructus and Granati pericarpium showed signs of infestation by ascomycetes, larvae of moths and garden weevils – withered velvet powdery mycelia or scattered deposits of gum and frass on some fruit surfaces (Burks et al. 2012; Huseyin et al. 2015; Roques et al. 2017).

In 47.6% of all products, the labels contain texts directly stating or implying that the relevant herbal substances have a preventive or curative effect on human body against particular diseases. This is in open conflict with the Ministerial decree № 434 of 10 December 2021 for adoption of an ordinance on food supplements and Regulation (EU) No 1169/2011 of the European Parliament and of the Council on the provision of food information to consumers, both presenting the current EU policies in this regard.

17,5% of all products contain plants specified as poisonous according to Ordinance № 47 of 28 December 2004 on the requirements for food supplements, which was in force at the time of purchase of these products. According the current ordinance on food supplements adopted through Ministerial decree № 434 of 10 December 2021, the variety of herbal substances in Suppl. material 1 which are prohibited for use in food supplements remains almost the same with the exception of the herbal substances derived from *Tussilago farfara* and *Punica granatum*. These two species were included in a list of poisonous plants according to Ordinance №47 of


**Figure 4.** Evidences for infestation of dried herbs with cigarette and drugstore beetles: *Lasioderma serricorne* (Fabricius, 1792) (**a**), *Stegobium paniceum* (Linnaeus, 1758) (**b**), rounded holes, drilled by these insects in the substance of Coriandri fructus.

28 Desember 2004 but were left out of the guidelines of Ministerial decree №434 of 10 Decembre 2021 currently in force. For *Tussilago farfara* 10 pyrrolizidine alkaloids which can possibly cause hepatotoxic effects have been reported so far (Pabreiter 1992; Nedelcheva et al. 2015; Smyrska-Wieleba et al. 2017; Moreira et al. 2018; Kopp et al. 2020), yet currently there is no firm scientific statement on its safty (EMA/HMPC/893108/2011 Rev. 1; Chen et al. 2021). The species *Punica granatum* is much less studied for toxic compounds or such effects and the available data showed that the researched substances and extracts were safe (Jahromi et al. 2015; Wibowo et al. 2018). We think that the safety of these plants as ingredients of food supplements needs to be clarified in future studies.

## Conclusions

In conclusion, very small part of the studied products meet the standards for all of the selected criteria and the considered standard and normative documents which implies the need of strengthening control over dried herbs marketed as food supplements in Bulgaria.

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# Supplementary material 1

## Table S1

Authors: Anna Gavrilova, Genadi Gavrilov

Data type: Xlsx file.

Explanation note: Macroscopic pharmacognostic analysis of dried herbs in Bulgaria.



Link: https://doi.org/10.3897/pharmacia.@@.e87549.suppl1