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Evaluation of the Morphological and Pharmacognostic Features of Dried Herbs and Their Label Information in Bulgarian Food Supplement Products

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ABSTRACT

In Bulgaria, dried herbs are regulated as food supplements under current legislation, allowing their sale in pharmacies, drugstores, and grocery stores. This study aimed to evaluate the morphological pharmacognostic characteristics of dried herb content and the supplementary information provided on the labels of Bulgarian food supplements, in accordance with key standards and regulatory guidelines concerning herbal substance and food supplement quality. A total of 91 herbal substances from 10 major groups were examined, encompassing 103 commercial products produced by 8 different companies. The authenticity of the herbal materials was verified using both macro- and microscopic analyses. Macroscopic morphological parameters assessed as part of tradecommodity evaluation included color variations, presence of foreign plant parts, organic and mineral contaminants, and signs of disease or pest damage. Additionally, the phenological stage at which the herbs were harvested was determined. Results indicated that only a small fraction of the products fully complied with all selected quality criteria from the referenced standards, highlighting the need for more rigorous regulatory control.

Keywords: Dried herbs, Quality control, Food supplements, Morphological pharmacognostic characteristics

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Introduction

Medicinal plants constitute a valuable national resource in Bulgaria, with notable environmental, socio-economic, and cultural significance, and the country is recognized as a leading European exporter of high-quality medicinal plants [1]. Only a minor portion of the annual harvest of medicinal plants, in the form of dried herbs, is retained for domestic consumption. Despite this limited availability, demand among local consumers has remained relatively steady, supported by a short supply chain involving small- and medium-sized producers, distributors, and retail outlets such as pharmacies and drugstores. This persistent interest in dried herb products is rooted both in Bulgaria's long-standing folk medicine traditions and in the historical policies of the 1960s–1990s, which aimed to promote the use of medicinal plants for public health purposes [2]. In addition, contemporary consumer interest in alternative preventive and therapeutic approaches has emerged as a strong factor driving market demand [3]. The current market offers a wide variety of products, either consisting of single-species herbs or mixtures, marketed respectively as "Herbs" or "Herbal teas." Under the Food Act (2020) [4], these products fall under the category of food supplements, and their sale is permitted in pharmacies, drugstores, and grocery stores. Oversight of quality throughout the supply chain—from harvest (wild or cultivated) through primary processing, packaging, labeling, and distribution—is shared among the Ministry of Environment and Water, the Ministry of Agriculture, Food and Forestry, the Ministry of Health, and their respective agencies.

This study aimed to evaluate both the morphological and pharmacognostic features of dried herb content and the additional information presented on labels of food supplements in Bulgaria, following the requirements of relevant standards and regulatory documents governing herbal substances and food supplement quality.

Materials and Methods

A total of 91 herbal substances, categorized into ten main types—Herba, Folium, Flos, Radix, Rhizoma, Cortex, Pericarpium, Semen, Fructus, and Galbulus—were analyzed. These substances were included in 103 commercial products from eight different companies, purchased in 2019 from pharmacies and drugstores in Pleven, Bulgaria. Each product weighed between 40 and 50 g and was legally classified as a "food supplement" under §1, item 4 of the Food Act (2020) [4].

Identification of the plant materials relied on authoritative botanical references [5-12]. For species listed in the European Pharmacopoeia, monographs were used to confirm their botanical identity and authenticity through macro- and microscopic evaluation. For microscopic analysis, the herbs were ground, sieved through a 355-mesh, and treated with chloral hydrate. Temporary slides were prepared using the fine powder and glycerine as a mounting medium.

Macroscopic features used to assess trade and quality included color changes, presence of extraneous plant material (either from the same species or other species), organic or mineral contamination, and visible signs of disease or pest infestation. The phenological stage at the time of harvest was also recorded. All macroscopic observations were performed on the full content of the product packages. Plant names were verified against Pharmacopoeia [13-15].

Results and Discussion

The products analyzed derived from plants representing 47 families, 86 genera, and 86 species. Column 2 shows their botanical family affiliation, while column 4 provides the verified herbal substance names based on reference literature. Columns 5–13 summarize content assessment and label information.

Column 5 presents the match between the labeled plant species and the actual species present. In 4 products (3.9%), the herbal content did not match the label, violating Ordinance №5 (19 July 2004) [16] on herbal procurement and storage facilities, as well as the Food Act (2020) [4]. For these products, corresponding data in columns 6–12 are marked N/A. For instance, a product labeled as Echinops ritro L. was found to contain inflorescences of E. microcephalus Sibth. & Sm. Only E. spaerocephalus L. is included in the Medicinal Plants Act (2000) [17], while E. ritro L. and E. bannaticus Rochel ex Schrader are also recognized as medicinal [18]. E. microcephalus differs distinctly from other species in involucrum hair density and length [11]. Importantly, the medicinal part of Echinops species is typically the achenes, which contain quinoline alkaloids with neuromuscular stimulatory properties, rather than the inflorescences [19].

In one product marketed as Equisetum arvense L., the material was found to actually consist of E. telmateia Ehrh. The stems of E. telmateia are whitish, almost smooth, and measure 10–20 mm in diameter, featuring 20–40 fine longitudinal grooves. Its sheaths are pale at the base, with darker teeth at the tips, which are hair-like and easily shed. The lowest internode of each branch is equal to or shorter than the adjacent sheath. In contrast, E. arvense has greenish basal sheaths, often with brown-tipped teeth, and its lowest internode exceeds the length of the adjacent sheath [5, 11, 15, 20]. Per the Medicinal Plants Act (2000) [17] and European Pharmacopoeia 10.0 [13], only E. arvense is considered suitable for medicinal use; other Equisetum species are excluded due to insufficient in vivo toxicity data and unclear therapeutic/toxic dose ranges, despite their pharmacological potential [21].

Additionally, two other products contained unintended plant species. Material labeled as Rhodiola rosea L. was actually Solidago gigantea W. Aiton, while material labeled as Digitalis lanata Ehrh. was identified as Echium vulgare L., as confirmed through detailed comparison with authoritative literature [9, 11, 13].

The Solidago gigantea herba consisted of a mix of stem fragments, with the upper portions densely to sparsely pubescent and the lower parts mostly smooth. Leaves were green and mostly glabrous above, with sparse hairs, while the lower surface was grey-green and pubescent along veins. Flower heads (capitula) measured 3–6 mm in diameter, with a single row of yellow ray florets matching the involucre length, and yellow tubular florets slightly shorter; most capitula were in late flowering stage (**Figure 1**). For reference, Solidago canadensis L. differs by having predominantly glabrous stems and ray and tubular florets of equal length to the involucre, while Solidago virgaurea L. has much larger capitula and ray florets roughly twice as long as the tubular florets.

The Echium vulgare herba was represented by grooved, rough stems and leaves with mixed hairs—strigose on the upper surface and hispid on the lower—with both simple and glandular hair types, some sessile and others

emerging from rosettes. Flowers were zygomorphic, with a whitish basal tube and blue corolla lobes of variable shapes. The style was bifurcated, and the stamens extended beyond the corolla (Figure 2).



Figure 1. Flower heads (capitula) of Solidago gigantea observed during the late flowering phenological stage.

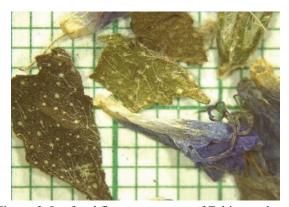


Figure 2. Leaf and flower structures of Echium vulgare.

Analysis of label information revealed that roughly one-third of the products (33%) contained mistakes in the Latin names of the herbs or omitted them entirely (Suppl. material 1, column 6). Many labels used outdated or nonstandard nomenclature, inconsistent with the guidelines of the European Pharmacopoeia and the European Medicines Agency—for example, "Herba Achilleae" instead of the correct "Achilleae herba" [22].

Regarding content verification, in 9.71% of cases, the labeled plant material did not correspond to the actual herb present (Suppl. material 1, column 7). One product labeled as Echinops ritro L. contained whole secondary heads of E. microcephalus rather than the pharmacologically active achenes [14, 19]. Another product identified as Solidaginis herba was inaccurately labeled only with the Bulgarian common name "zlatovrah," typically referring to Rhodiola rosea L. and its rhizomes, known for adaptogenic properties [23, 24]. In two products, leaves were harvested together with flowering stems rather than solely the leaves: Convallariae folium and Malvae folium. Ordinance No2 / 20.01.2004 [25] specifies that Convallaria majalis leaves must be collected before flowering, while Malva sylvestris leaves may be collected during flowering.

In several cases, the whole aerial portion of the plant appeared to have been harvested instead of just the upper stems. This was observed in products labeled as Cichorii herba and Rutae herba, where basal stem segments with thicker mechanical tissues, and in Rutae herba, periderm, were present. Similarly, Calendulae flos and Cyani flos contained complete flower heads instead of just the ray florets, and Rosae flos included entire flowers with sepals instead of only petals of Rosa damascena. Differences were also noted between the European Pharmacopoeia definitions and Ordinance №2 / 20.01.2004 [25] collection rules. For some large herbaceous species, the Ordinance is stricter, permitting harvest only of the leafy tops (e.g., Verbena officinalis and Leonurus cardiaca), whereas the Pharmacopoeia allows the entire aerial part to be collected.

Analysis of harvest timing indicated that approximately one-quarter (25.2%) of the plant materials were collected after the recommended phenological stage (Suppl. material 1, column 8). For instance, in a product containing Pini turiones, all turions were broken, likely due to over-maturity, harvesting specimens larger than 5 cm, or improper drying methods such as sun-drying [15]. Several herbs, including Achilleae herba, Bursae-pastoris

herba, Polygoni avicularis herba, Adonidis herba, Agrimoniae herba, Cichorii herba, Verbenae herba, Euphrasiae herba, Polygoni hydropiperis herba, and Rutae herba, were collected during fruiting instead of flowering, resulting in products containing mainly fruits and no petals. In Cichorium intybus, only empty receptacles were present. In Robiniae flos, flower samples had partially developed immature legumes with withered sepals and a few remaining petals. In Crataegi folium cum flore, flowers were predominantly at late bloom, indicated by advanced hypanthium development and fully detached petals [15]. Several other herbs—including Absinthii herba, Solidaginis herba, Meliloti herba, Centaurii herba, Veronicae herba, Calendulae flos, Cyani flos, Helichrysi flos, and Lupuli flos—were harvested at late flowering rather than at full bloom. In Coryandri fructus, multiple fruits were split in two, suggesting post-milk ripeness collection. In Rosae flos, fully opened flowers predominated instead of buds.

Approximately 35.9% of the analyzed products showed noticeable deviations in the color of the herbal materials compared to accepted standards (Suppl. material 1, column 9). According to European Pharmacopoeia 10.0 and Bulgarian State Standards [15], the proportion of plant parts that have lost their natural color should generally remain below 2-3%, with a few exceptions such as Asperulae herba, where up to 7% discoloration is tolerated. In most of the affected products, however, the discoloration affected the majority of the package contents. The main active constituents in these herbs include essential oils, phenolics, alkaloids, saponins, iridoids, cardiac glycosides, polysaccharides, mucilaginous compounds, vitamins, amines, and peptides. Color changes in harvested plant materials occur primarily due to enzymatic and non-enzymatic oxidation, which degrades chlorophyll, carotenoids, anthocyanins, other phenolic compounds, essential oils, carbohydrates, and related metabolites. These oxidative reactions are triggered by exposure to heat, light, moisture, and pH variations, occurring mainly before or during primary processing [26-28]. Traditional drying methods are particularly associated with significant color loss and reduction of phytochemical content [27]. Additionally, microbial growth—including bacteria such as Bacillus, Pseudomonas, Agrobacterium, and molds like Chaetomium, Fusarium, Aspergillus, and Penicillium—can further metabolize key bioactive compounds like pectins, sugars, essential oils, phenolics, alkaloids, and glycosides, with these processes also manifested visually as color deterioration [29].

Contamination with impurities was observed in 53.4% of the tested samples (Suppl. material 1, column 10), exceeding the limits established by European Pharmacopoeia 10.0 (2.8.2) [13] and Bulgarian State Standards [15]. In fruit-based herbs such as Coriandrum sativum, Crataegus monogyna, and Prunus spinosa, the impurities consisted of damaged, discolored, or pest-affected fruits of the same species. Leaf-based herbs contained impurities including discolored leaves, twigs, roots, or fruits from the same or other species, as well as inorganic matter like soil and small stones. In two cases, impurities were identified to the species level: Uvae ursi folium contained leaves of Vaccinium vitis-idaea and Pinus sylvestris, while Rosmarini folium included broken stems of Equisetum arvense. In Matricariae flos, contaminants were parts of other grassland species, including Alopecurus pratensis, Bromus mollis, Bromus sterilis, Elymus repens, Melilotus spp., Erodium cicutarium, Papaver rhoeas, Capsella bursa-pastoris, and Anthemis austriaca.

Herbs consisting of roots and rhizomes were mainly contaminated with inorganic material and portions of other underground plant parts with altered color. For stems, flowers, and inflorescences, impurities included discolored fragments of the same plant, lignified stems, immature or mature fruits, organs of other plant species, and inorganic debris. In one of two tested products of Verbasci flos from different manufacturers, over 70% of the content consisted of sepals and fruits of unidentified Verbascum species instead of petals, while in the other product, petals predominated, but up to 50% of their color was significantly darkened.

Even trace amounts of contaminants such as rodent droppings, plastic residues, polyethylene, textile fibers, and fabrics with varying degrees of degradation were found in some products (**Figure 3**). These observations indicate inadequate control over harvesting sites, potential collection from unregulated areas, insufficient hygiene during and after primary processing, and improper storage or packaging, in addition to pest contamination.

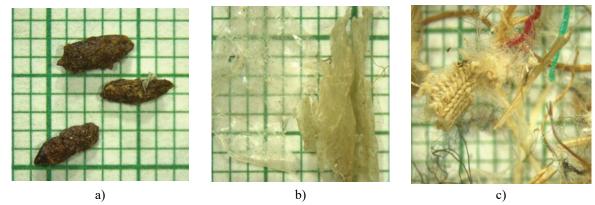


Figure 3. Examples of contamination detected in the herbal products: mouse droppings (a), plastic and polyethylene fragments (b), and textile fibers or fabric remnants (c).

Pest-related deterioration affected 20.4% of the products, representing noncompliance with Ordinance №5 of 19 July 2004 [16] and the Food Act (2020) [4] regarding proper herbal storage and handling (Suppl. material 1, column 11). In 12% of cases, damage was caused by Lasioderma serricorne (cigarette beetle) and Stegobium paniceum (drugstore beetle), which are globally distributed pests present in Bulgaria [30-32]. Affected plant parts included aerial stems (Solidaginis herba, Polygoni avicularis herba, Veronicae herba, Hyperici herba, Achilleae herba), flowers (Robiniae flos, Matricariae flos, Calendulae flos), leaves (Urticae folium), and dry fruits (Coriandri fructus, Silybi mariani fructus). Packaging contained dead beetles, insect fragments, and extensive frass, with many small, round holes drilled by the insects. These beetles are recognized as major pests in stored plant materials, particularly dried herbs and tobacco [33-35], and their activity can reduce levels of bioactive compounds in herbs [36].

Fungal infestations were also observed. Leaves of Malva sylvestris were heavily infected by Puccinia malvacearum, an autoecious rust that triggers increased mucilage production while simultaneously destroying large portions of leaf tissue [37, 38]. Tussilago farfara leaves were colonized by Coleosporium tussilaginis in the uredinial stage, which completes its aecial stage on two-needle pines (Pinus spp.) [39], highlighting the importance of avoiding pine forest areas during leaf collection. Another rust potentially present in coltsfoot is Puccinia poarum in pycnial and aecial stages [40].

Additional contamination included rodent droppings in Verbenae herba and Cichorii herba. Fruits from Juniperi galbulus, Rosae pseudo-fructus, Crataegi fructus, Pruni spinosae fructus, and Granati pericarpium showed evidence of ascomycete fungi, moth larvae, and garden weevils, with symptoms including powdery mycelia, dried residues, and deposits of frass or gum [41-43].

Label analysis revealed that 47.6% of products included statements implying preventive or therapeutic effects for specific human diseases. Such claims conflict with EU and national regulations, including Ministerial Decree №434 of 10 December 2021 [44] on food supplements and Regulation (EU) No 1169/2011 on food information for consumers.

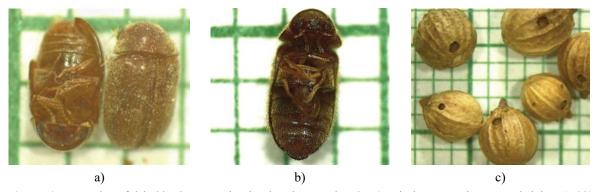


Figure 4. Examples of dried herb contamination by cigarette beetles (Lasioderma serricorne, Fabricius, 1792) and drugstore beetles (Stegobium paniceum, Linnaeus, 1758), including characteristic rounded perforations in Coriandri fructus.

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Examination of the products revealed that 17.5% contained plant species categorized as toxic under the previous Ordinance №47 of 28 December 2004 [45], which regulated food supplements at the time of acquisition. The current regulation, Ministerial Decree №434 of 10 December 2021[44], maintains restrictions on most of these species, but Tussilago farfara and Punica granatum are no longer listed as prohibited. T. farfara contains several pyrrolizidine alkaloids (up to ten identified) with potential liver toxicity [46-50], yet definitive safety data are still lacking (EMA/HMPC/893108/2011 Rev.1) [51, 52]. In contrast, P. granatum has been less investigated for toxic effects, but current studies indicate that its extracts and plant parts are generally safe. These findings suggest that additional studies are necessary to fully evaluate the safety of these species as components of food supplements.

Conclusion

The assessment revealed that only a minor portion of the products met all evaluated quality criteria and complied with relevant regulatory standards, underlining the urgent need for improved oversight and stricter quality control of dried herbs sold as food supplements in Bulgaria.

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Ethics Statement: None

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