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Extraction Methods Applied to Natural *Lamiaceae*-Derived Compounds: An Overview Based on Patents †

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Abstract: Secondary metabolites of plants are of major interest for pharmaceutical, cosmetic, and food applications. The extraction methods for these compounds must be optimized to achieve the best possible yield without altering the effectiveness of the targeted compounds. In this paper, we examine the methods of extraction of plant compounds, especially those applied to plants of the *Lamiaceae* family, renowned for their aromatic and medicinal roles. To do so, we consulted databases specialized in patent documentation, using appropriate keywords with the help of International Patent Classification (IPC) codes. Our results present the analysis of the 140 relevant documents selected. The first patent relating to our field of study was granted in the United States in 1998. It concerned a process for obtaining antioxidants from plant materials. The year 2020 saw the registration of the largest number of these documents (15). Most of the documents identified (66) were filed in China. The French company Naturex, which specializes in plant-based products, is the number one depositor in the field of plant extraction. The relevant patents selected describe processes using various extraction methods and agents, most of which are valid for many plants, while some focus on genera of *Lamiaceae*, such as *Salvia*, *Ziziphoria*, and *Clinopodium*.

Keywords: extraction; *Lamiaceae*; innovation; patent; plant compound

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1. Introduction

Extraction methods for natural plant-derived compounds are essential processes used to isolate bioactive molecules, flavors, fragrances, and other valuable substances from plants [1]. However, it is essential to note that these methods vary depending on the type of compounds, the plant source, and the equipment used [2].

There are a large number of medicinal and aromatic plants (MAPs), and various varieties are cultivated extensively for medicinal, ornamental, and culinary purposes, among them the *Lamiaceae* [3]. The *Lamiaceae* family comprises approximately 200 genera and around 3300 species, consisting of aromatic plants that are either annual or perennial herbs or low shrubs. This plant family finds significant representation in both the Mediterranean region and the British Isles. The flowers within this family are typically bisexual and exhibit zygomorphic characteristics. These flowers are often organized in clusters known as verticillasters [4].

The *Lamiaceae* genera include, among others, *Rosmarinus*, *Lavandula*, *Marrubium*, *Lamium*, *Salvia*, *Origanum*, *Thymus*, *Mentha*, and *Ocimum* [5]. Many members of the family are used as culinary or medicinal herbs, as sources of volatile oils, and, in some cases, for the preparation of constituents of volatile oils such as menthol and thymol [6]. Besides volatile oils, the *Lamiaceae* family encompasses a diverse array of compounds. These include, but are not limited to, diterpenoids and triterpenoids, flavonoids, saponins, tannins, alkaloids,

and iridoids, along with their glycosides, quinones, furanoids, cyclitols, and coumarin, as well as the sugars raffinose and stachyose [5,7].

The diversity of natural *Lamiaceae*-derived compounds implies the need to adapt extraction methods to achieve the best possible selectivity. Moreover, the extraction methods for these compounds must be optimized to obtain the best possible yield without altering the effectiveness of the targeted compounds [5,8].

In this work, we examine the extraction methods of plant compounds based on patents, which can serve as a reference for researchers interested in MAPs as well as phytotherapy and cosmetology. Our overview focuses especially on extraction methods applied to plants of the *Lamiaceae* family.

2. Resources and Methods

To identify the most commonly used extraction method related to natural *Lamiaceae*-derived compounds and the innovation underlying it, we examined four patent documentation databases:

1. Patentscope [9];
2. PatFT-AppFT database [10];
3. Espacenet Patent Search [11];
4. Lens Patent Data Set [12].

Different terms and keywords related to "extraction methods applied to natural *Lamiaceae*-derived compounds" have been used. Further, the International Patent Classification (IPC) codes have also been used to refine the results. The IPC is defined as "a code-based hierarchical system that separates all technological domains into sections, classes, subclasses, groups, and subgroups" [13,14]. In our case, the code A61k36/53 was employed. It concerns medicinal preparations of undetermined constitution containing material from plants or derivatives such as *Lamiaceae* [15,16]. Finally, the found patent documents have been filtered to include only patent applications and granted patents.

3. Results and Discussion

Following our research methodology and using the four cited databases, we found 140 patent documents between 1978 and 2023. There comprise 112 patent applications and 28 granted patents.

Figure 1 presents the evolution of patent documents (i.e., patent applications and granted patents) related to innovation in *Lamiaceae* component extraction as a function of publication date (year).

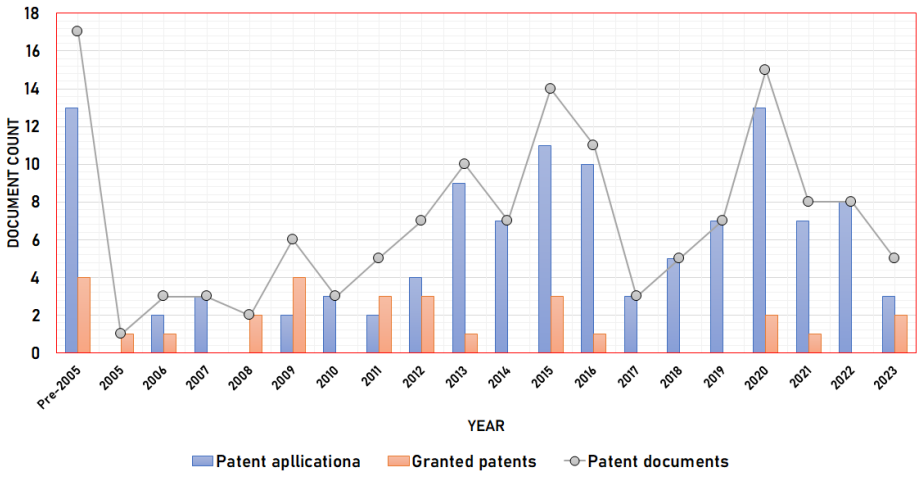

Figure 1. Evolution of patent documents (i.e., patent applications and granted patents) related to innovation in *Lamiaceae*-component extraction.

Based on our previous study, we define the publication date as "the date on which a patent document is published with an assigned publication number by a patent authority, thereby making it part of the state of the art" [1]. The first patent application in the field of plant extraction, in relation to Lamiaceae, was filed in 1977 (and published in 1978) in Japan [17]. On the other hand, the first patent was granted in 1998 in the United States for a process for obtaining antioxidants from plant materials [18]. Moreover, the number of patent applications filed has steadily increased in recent years, with a peak of 15 patent documents in 2020.

We previously defined patent applicant and patent jurisdiction, respectively, as "either a natural person or a legal entity that submits a patent application. In some cases, the applicant can also be the inventor, and there may be more than one applicant for a single patent application" and "the country or region where applicants can file a patent application through the appropriate patent office" [1].

For innovation in *Lamiaceae*-component extraction, Figure 2 presents the top 10 applicants and the top 10 jurisdictions. As a result, the French botanical company Naturex (Avignon, France) is the leading patent depository in this field of study. It specializes in the production and distribution of plant extracts and plant-based natural ingredients tailored for the food industry, serving various purposes such as flavoring, coloring, and preservation. The company boasts a global presence, with manufacturing facilities spanning, among others, Europe, the United States, Canada, Brazil, Morocco, and India. On the other hand, "Avignon Université" (Avignon, France) is leading the way in patenting the extraction methods applied to natural *Lamiaceae*-derived compounds.

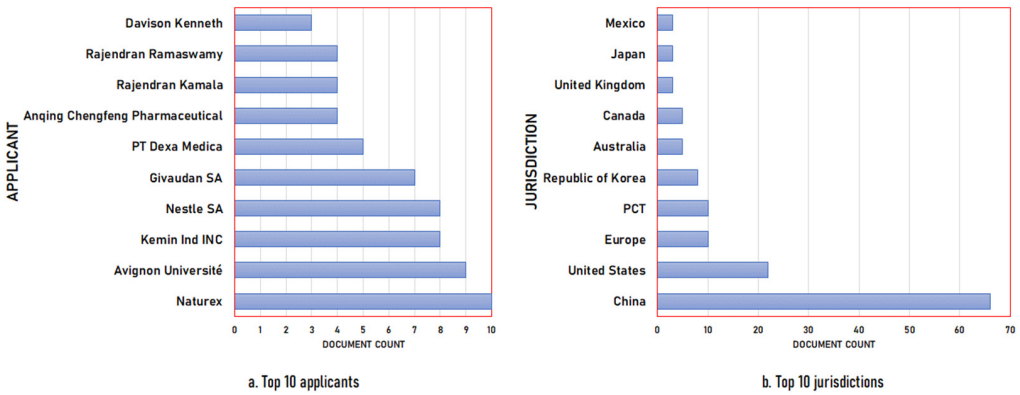

Figure 2. Patent information related to innovation in *Lamiaceae*-component extraction: (**a**) top 10 applicants; (**b**) top 10 jurisdictions.

China is the jurisdiction with the most patents in this area (i.e., 66 patent documents), followed by the United States, Europe's jurisdiction, and the Patent Cooperation Treaty (PCT) international system. This system, overseen by the World Intellectual Property Organization (WIPO), enables inventors to obtain protection for their inventions in many countries worldwide by filing a single patent application [19].

Through the analysis of the selected granted patents (Supplementary Materials, Table S1), the extraction methods applied to natural *Lamiaceae*-derived compounds are summarized in Figure 3. These extraction methods involve nine techniques classified by the number of granted patents appearing as fellows: decoction, maceration, supercritical fluid extraction, soxhlet extraction, ultrasound-assisted extraction, pressurized liquid extraction, microwave-assisted extraction, extraction by eutectic solvents, and mechanochemical-assisted extraction. Based on prior investigations, Table 1 outlines each extraction method according to its description, strengths, and weaknesses [20–22].

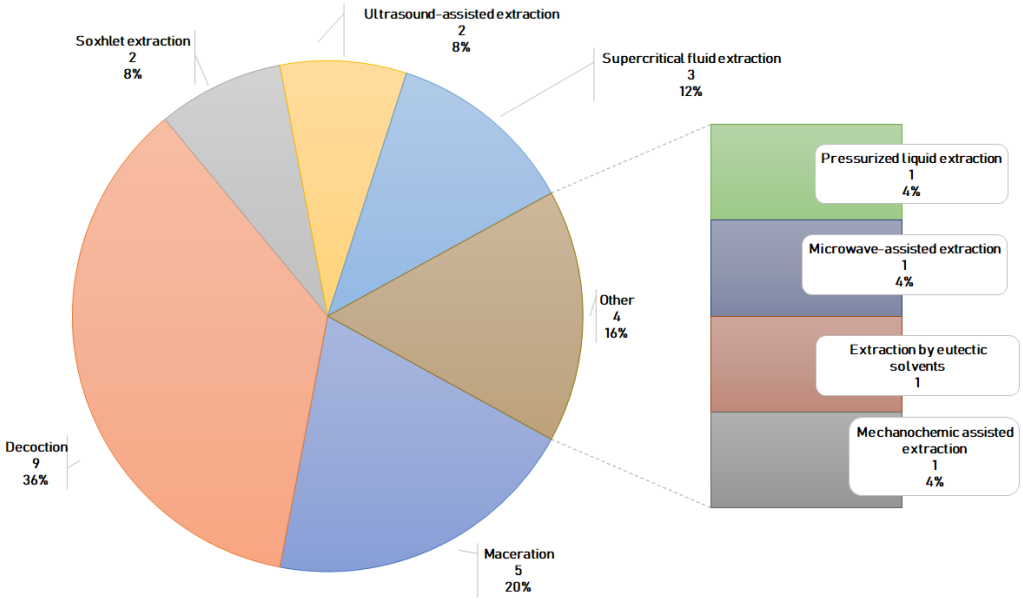

Figure 3. The extraction methods cited and used in the studied granted patents (Supplementary Materials, Table S1).

Table 1. A summary of plant extraction methods cited in selected granted patents * (the description of the methods is adapted from Jha and Sit, 2022 [20], Fan et al., 2022 [21], and Lefebvre et al., 2021 [22]).

Extraction Method	Description	Strengths	Weaknesses
Decoction	The plant is boiled in a solvent.	More effective than maceration for extracting water-soluble compounds.	Degradation of heat sensitive compounds.
Maceration	The plant is left to soak in a solvent for a certain period of time.	It is inexpensive and easy to implement.	Ineffective for extracting the most water-soluble compounds.
Supercritical fluid extraction	The solvent is heated to a supercritical state for selective extraction.	Selective and efficient extraction.	Expensive.
Soxhlet extraction	The solvent is heated and circulated around the plant.	Complete and efficient extraction.	Long and expensive.
Ultrasound-assisted extraction	Sound waves break down plant cells and facilitate the extraction of compounds.	Rapid and efficient extraction.	Expensive.
Pressurized liquid extraction	The solvent is pressurized and heated to a higher temperature.	Rapid and efficient extraction.	Expensive.
Microwave-assisted extraction	Microwaves heat the solvent for faster extraction.	Rapid and efficient extraction.	Degradation of heat sensitive compounds.
Extraction by eutectic solvents	The solvent is a mixture of two or more compounds that has a very low melting point.	More efficient than traditional extraction methods.	Safer because temperatures are lower.
Mechanochemical-assisted extraction	The solvent is used with mechanical energy to break down plant cells and facilitate the extraction of compounds.	More efficient and selective than traditional methods.	Less damaging to the plant material, which may limit the extraction of certain compounds.

* The detailed list of the selected granted patents are presented in Table S1 (Supplementary Materials).

Finally, the analysis of the selected granted patents ensures that the relevant inventions selected describe processes for obtaining different natural compounds using various extraction methods and agents, most of which are valid for many plants, while some focus on genera of *Lamiaceae*. In this regard, *Lamiaceae* genera involved in the extraction innovation in this study are specifically genera such as *Thymus*, *Mentha*, *Origanum*, *Salvia*, *Rabdosia*, *Clerodendranthus*, etc. (Figure 4).

Figure 4. *Lamiaceae* genera that are involved in the extraction innovation.

4. Conclusions and Outlook

In this patent study, we examined the methods of extraction of *Lamiaceae*-derived compounds renowned for their aromatic and medicinal roles. According to our research methodology, 140 patent documents were published between 1978 and 2023. These encompass 112 patent applications and 28 granted patents. The relevant patents selected describe processes using various extraction methods and agents, most of which are valid for many plants, while some focus on the genera of *Lamiaceae*. These extraction methods involved nine techniques, such as decoction, maceration, and supercritical fluid extraction. However, each method has its benefits and drawbacks, and scientists frequently choose the most appropriate technique based on their objectives and the resources at their disposal. Moreover, the selection of the extraction method depends especially on the specific compounds, the characteristics of the plant material, and the suitable final product.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/ASEC2023-15362/s1, Table S1: The detailed list of the selected granted patents related to extraction methods applied to natural *Lamiaceae*-derived compounds. References [23–42] are cited in the supplementary materials.

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