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METHODS OF EXTRACTING THE ACTIVE PRINCIPLES FROM MEDICINAL AND AROMATIC PLANTS – A REVIEW

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Abstract: Medicinal plants are gaining much interest recently because their use in ethno–medicine treating in common disease such as cold, fever and other medicinal claims are now supported with sound scientific evidences. A wide range of technologies is available for the extraction of active components and essential oils from medicinal and aromatic plants. The choice depends on the economic feasibility and suitability of the process to each particular situation. Bioactive compounds from medicinal plants were synthesized using effective extraction methods which have important roles in the pharmaceutical product development. In this paper will be presented some methods of extraction of active principles from medicinal and aromatic plants.
Keywords: medicinal plants, active components, bioactive compounds, extraction methods

INTRODUCTION

Plants were once considered as a daily food. Now, plants are popular used as a common source in medicinal agents, food additives, cosmeceuticals and nutraceuticals. Although, the medicinal properties of plants have gained attention, many research studies are still conducted to discover their values because the utilisation of synthetic drugs to heal or control most chronic diseases have caused several long–term effects. There is rising approach regarding the application of herbal medicinal plants in treating diseases with minimal or no aftereffects. Therefore, the extraction of bioactive compounds from herbal medicinal plants offers great potentials for new drug discoveries (Nur Amanina Abd Aziz et al., 2021).

These therapeutically useful medicinal compounds in plants are extracted or separated by using selective solvents through a standard procedure. Generally, the extraction techniques can be divided into two categories, namely classical technique and modern technique. The former technique faces several limitations, such as the use of excess solvents, time–consuming and a long heating time which could risk the degradation of bioactive compounds. In most cases, extraction by using these solvents was hazardous and toxic to human health and the environment.

Organic solvents release greenhouse gases into the environment, threatening humans, agriculture and microorganisms. Moreover, the usage of excess solvent produces a large amount of waste by–products. Contrary to the hazardous classical techniques, environmentally friendly extraction approaches like ‘green solvents’, ‘green processing’ and ‘green product’ are favoured.

Green extraction methods should be applied to encourage efficient and safe extraction method. Green extraction methods reduce energy consumption which allow the use of alternative solvents and renewable natural sources to produce a safe and high–quality product. Therefore, these modern extraction techniques are considered as green processing. These techniques reduce the usage of organic solvents, minimise bioactive compounds degradation in the sample and improve extraction efficiency (Nur Amanina Abd Aziz et al., 2021; Antigoni Oreopoulou et al., 2019).

The increased population led to a higher utilization of these plants, so their residues are proportional, with a huge amount of biomass generated as by–products, representing a growing market in the natural–based products. The general use of MAPs all over the world is not homogenic, due to different factors:

- in developed countries, even if the demand for natural treatments is high, profits of the growers and producers remain low because of the existing intermediaries which increase the price, as well as the lack of organization and networking by the poor collectors of medicinal plants from the wild;
- rigorous regulations and documentations requirements; and
- in less developed countries, there are poor traceability mechanisms from plant to population (Pruteanu A et al., 2014).

In addition to traditional medical applications of medicinal and aromatic plants, there is the possibility of using them in cosmetic products, feed or food additives and preservatives, or as a viable tool for biotechnological applications, such as the enhancement of secondary metabolites by genetic engineering (Radu Claudiu Fierascu et al., 2021).

Extraction, as the term is used pharmaceutically, involves the separation of medicinally active portions of plant or animal tissues from the inactive or inert components by using selective solvents in standard extraction procedures. The products so obtained from plants are relatively impure liquids, semisolids or powders intended only for oral or external use. These include classes of preparations known as decoctions, infusions, fluid extracts, tinctures, pilular (semisolid) extracts and powdered extracts. Such preparations popularly have been called galenicals, named after Galen, the second century Greek physician.

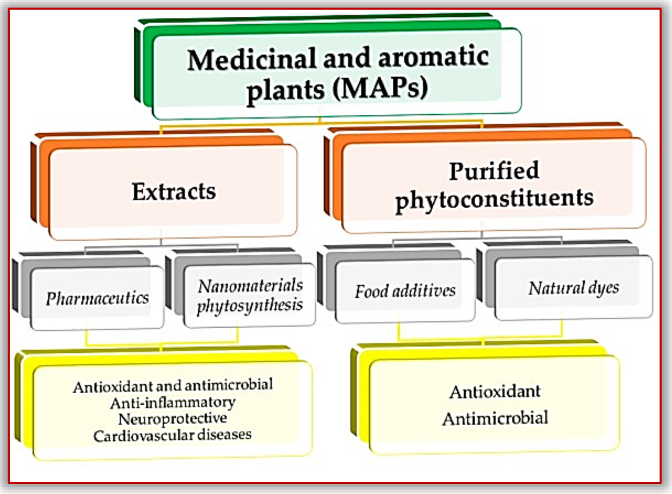

Figure 1. Some of the potential applications of medicinal and aromatic plants (Radu Claudiu Fierascu et al., 2021)

The purposes of standardized extraction procedures for crude drugs are to attain the therapeutically desired portion and to eliminate the inert material by treatment with a selective solvent known as menstruum. The extract thus obtained may be ready for use as a medicinal agent in the form of tinctures and fluid extracts, it may be further processed to be incorporated in any dosage form such as tablets or capsules, or it may be fractionated to isolate individual chemical entities such as ajmalicine, hyoscine and vincristine, which are modem drugs. Thus, standardization of extraction procedures contributes signifi cantly to the final quality of the herbal drug (S. S. Handa et al., 2008; Romulus Gruia, 2015).

An overview of the results recorded in recent years in the field of medicinal and aromatic plants and the analysis of statistical data show a clear increase in the interest of consumers and businesses to the green pharmacy.

The activity of production and marketing of medicinal plants grown from spontaneous flora has become a large economic activity, being a source of income for producers, traders, but also for various processors.

In Romania, the general framework for the production, processing and organization of the market for medicinal and aromatic plants, the relations between producers, processors and traders is established by the Law on Medicinal and Aromatic Plants (Law 491/2003), and Order 244/2005 regulates the processing, processing and the marketing of medicinal and aromatic plants used as such, partially processed or processed in the form of pre–dosed food supplements.

In the production of medicinal and aromatic plants the quality of the products is given by the content in active principles. The amount of active principles in the plant is conditioned by ecological factors, the zoning of the species, the cultivation technology, the biological value of the cultivar (population, variety, hybrid, etc.) and last but not least, the processing (processing) (Romulus Gruia, 2015; Ţuia Steluţa, 2021).

MATERIALS AND METHODS

Plant extracts are fluid soft or dry pharmaceutical / phytopharmaceutical preparations obtained by extracting plant products with different solvents. In recent years, emphasis has been placed on the pharmaceutical and therapeutic revaluation of herbal preparations through a good knowledge of the physico–chemical and therapeutic properties of the active principles in medicinal plants and by the development of extraction techniques and quality control means.

Extracts obtained from medicinal plants can be classified according to several criteria, as can be seen in the table below (Ţuia Steluţa, 2021).

Table 1. Extracts obtained from medicinal plants (Ţuia Steluţa, 2021)

Classification criterion	The name of the extract	Remarks
By the nature of the solvent	Extract aqueous Hydroalcoholic extract Extract oils Medicinal vinegars Medicinal wines	–
According to the method of obtaining	Selective extract Non–selective extract Extract obtained by pressing or centrifugation	Selective extracts cannot be considered to be entirely natural preparations, as they are hyperconcentrated and without identical patterns in nature.
After the preparation operation	Simple extract Successive extract Multiple extract	–
After the parts of the plant subjected to extraction	Partial extract Total extract	–
According to the humidity of the plant	Extract obtained from the dried plant Extract obtained from fresh plant	–

Solvent extraction is the most widely used type of extraction for bioactive plant compounds. This separation technique involves the extraction of components from a solid or semi–solid sample in a suitable solvent. In the extraction operation, the choice of solvent is made depending on the nature of the substance to be extracted and the nature of the raw material. The actual solubilization of bioactive compounds is achieved by treating the finely chopped plant with water, saline solutions, hydroalcoholic solutions, etc. The chemical nature of the optimal extraction medium, its molarity and pH, as well as the time required for optimal extraction are determined experimentally (Ţuia Steluţa, 2021).

In the preparation of extracts, in particular, the influence of the following factors must be taken into account:

≡ Nature of the solvent: solvents must dissolve and extract most of the active components in a high yield and contain as few inert materials as possible without therapeutic value; the most used solvents used in the plant extract industry are: water (for alkaloid salts, glycosides, sugars, proteins, enzymes, tannins, etc.), 50% or 70% alcohol (for biofertilizers, hydrocarbons, tannins, base alkaloids and salts of glycosides, resins, chlorophyll, etc.), ethyl ether (for base alkaloids, resins, biofertilizers, etc.), oil, wine, vinegar;
≡ Degree of crushing of the plant: the more advanced the plant product is brought to a degree of crushing, the larger the contact surface, so the extraction is complete; for aqueous extractive solutions it is recommended to grind according to the plant product;
≡ The ratio between the amount of plant and solvent: the Romanian Pharmacopoeia provides concentrations of up to 6% for aqueous extracts, 20% for most tinctures and 10% for tinctures prepared from plant products containing highly active substances;
≡ Contact time between plant and solvent: differs depending on the extraction technique applied, but also on the type of extract; for aqueous extracts it is 5–6 hours, and for alcoholic ones 6–10 days;
≡ Shaking effect: shaking shortens the time to obtain the extract;
≡ The temperature at which it is worked: it positively influences the extraction efficiency, due to the increased solubility of the hot active principles; The Romanian Pharmacopoeia provides for the extraction of thermostable principles, at a temperature of 90 – 100 ° C, in the case of infusions and decoctions;
≡ Separation of the mixture and how to recover the active compounds from the solid residue.

In the case of preparation of aqueous or hydroalcoholic extractive solutions by maceration, the degree of crushing plays a very important role. This correlated with the nature of the solvent used and the intensity of stirring determines the contact time for the extraction of soluble components until the concentration balance between the solid phase and the liquid phase is reached (Sukhdev Swami Handa, 2008; Ţuia Steluţa, 2021).

RESULTS

Extraction can be performed by batch processes (maceration, percolation, infusion, decoction, as well as new high–performance methods: accelerated solvent extraction, microwave–assisted extraction, supercritical fluid extraction) and continuous processes (continuous extraction with organic solvents, continuous percolation, Soxhlet extraction) (Ţuia Steluţa, 2021; Popova A, 2018).

— **Maceration**: consists in treating the crushed vegetable product with a required amount of solvent, keeping in contact for a certain period (macerated in water 8–12 hours), simultaneously with continuous or intermittent stirring and then separating the extractive solution from the residue by filtration or settling; in the case of macerations in other solutions (alcohol, oil, wine, vinegar), the maceration time increases, reaching a few weeks.

Maceration is applied especially in the case of extraction of easily cold–soluble and thermolabile principles. Maceration can be done:
≡ in the cold (17–22 ° C).
≡ hot (called digestion) at 40–60 ° C (Popova A, 2018; Ţuia Steluţa, 2021).

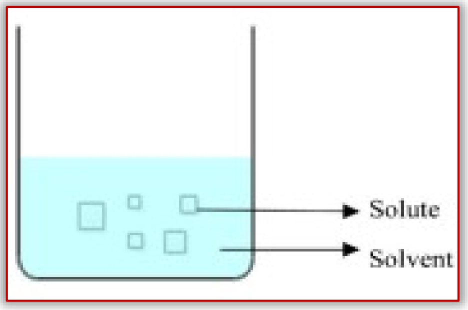

Figure 2. Example of maceration extraction method
(Nur Amanina Abd Aziz et al., 2021)

— **Percolation**: the process by which the active principles are extracted from plants, cold, using solvent in countercurrent. The process takes place as follows: before the solvent becomes saturated in the extracted active ingredients, it is displaced by another layer of solvent in which the plant product undergoes a short maceration and yields another part of the active ingredients. This phenomenon is continuous, each portion of solvent added coming into contact with the plant product until its complete depletion (Ţuia Steluţa, 2021).

Soxhlet extraction is a common conventional method used for extracting heat–stable compounds. The advantage of this method is that large amounts of drug can be extracted with a much smaller quantity of solvent. This is tremendously economic in terms of time, energy, and consequently financial inputs. The Soxhlet extractor consists of a distillation flask, an extractor, and a condenser. The solvent in the distillation flask is heated and the resulting vapor is condensed in the condenser. The condensed solvent from the condenser fills into the thimble holder containing the sample that needs to be extracted.

When the solution in the extractor reaches the overflow level, a siphon aspirates the solution of the thimble holder and unloads it back into the distillation flask, carrying dissolved solute into the bulk liquid. The solute is left in the distillation flask while the solvent is evaporated, condensed, and passed back into the sample solid bed.

This process is repeated 3–5 times or until a complete extraction is achieved (Popova A., 2018).

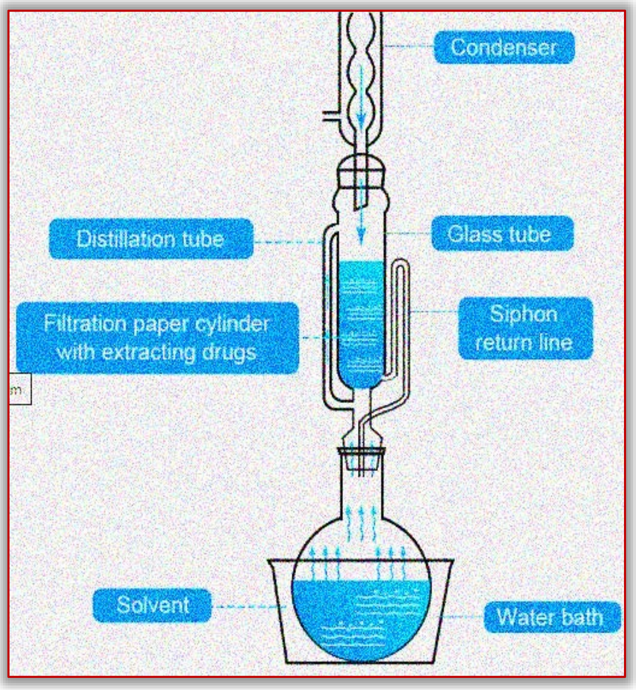

Figure 3. Soxhlet extractor (Ţuia Steluța, 2021; Popova A., 2018)

- **Infusion**: consists in wetting the crushed plant product with water, except for plant products containing biofertilizers which are moistened with a dilute alcohol solution; after about 5 minutes add the mass of water provided, heated to boiling and leave in contact for 30 minutes [5]. After the infusion time has elapsed, the solution is filtered. In general, the infusion is used in the case of plant parts that have thinner cell walls (flowers, leaves, herbaceous parts) (Ţuia Steluța, 2021).

Infusion and decoction use the same principle as maceration; both are soaked in the cold/boiled water. The maceration period for infusion is shorter, and the sample is boiled in specified volume of water (e.g., 1:4 or 1:16) for a defined time for decoction, however. An infusion is a dilute solution that contains readily soluble constituents prepared by short period of sample maceration (steeping) in cold/boiling water. Heat–sensitive compounds are recommended to be extracted by cold water.

Infusion (the folk method) is made using tablespoon of fresh/dried herbs per cup of boiled water. The aerial parts are mainly used and steeped for 2–10 min, covered. Common herbal infusions are aromatic plants, including mint, chamomile, lavender, and ginger. Nourishing herbal infusion (folk method) is produced for a minimum of 4 hours of steeping dried herbs in a 1:10 herb: water ratio. This should extract most of the minerals contained in the plant. Popular nourishing herbal infusions are made from U. dioica, Avena sativa, and Trifolium pretense (Danciu A et al., 2011; Popova A, 2018).

- **Decoction**: the preparation technique is similar to that of infusions: the chopped vegetable product will be soaked in 5 parts cold water; soak for 5 minutes and then add the remaining hot water to the required proportion (1% or 5%) after which it is heated on the water bath for 30 minutes (boiling). At the end, strain and wash the residue to the prescribed volume (medicinal plants with a high content of essential oils will be moistened with 50 ° C alcohol, then hot water will be added). In phytotherapy, the decoction is made in the case of plant organs (roots, rhizomes, bark, etc.) from which the principles are more difficult to extract (Ţuia Steluța, 2021; Popova A, 2018).
- **Alcoholic extraction by fermentation**: The active principles contained in some medicinal preparations are obtained by extraction in a fermentation process. The extraction procedure involves soaking the plant material either in the form of a decoction or in a ground state, for a certain period of time, during which the fermentation and generation of alcohol takes place in situ; this facilitates the extraction of the active substances contained in the plant material. The alcohol thus generated also has a preservative role. On an industrial scale, wooden vats, porcelain vessels or food grade stainless steel vessels are used (Ţuia Steluța, 2021; Popova A, 2018).
- **Continuous extraction with organic solvents**: The principle of extraction is simple. The components present in the crude raw material are extracted by dissolving in the liquid–solvent. The raw material is placed in a specially built extractor, and the solvent must be continuously recycled through the mass of plant material (Ţuia Steluta, 2021; Popova A, 2018).
- **Accelerated solvent extraction (ASE)** is a new extraction method, based on the use of high temperature and pressure to accelerate the dissolution kinetics and break the analytical interaction bonds (Ţuia Steluța, 2021; Popova A, 2018).

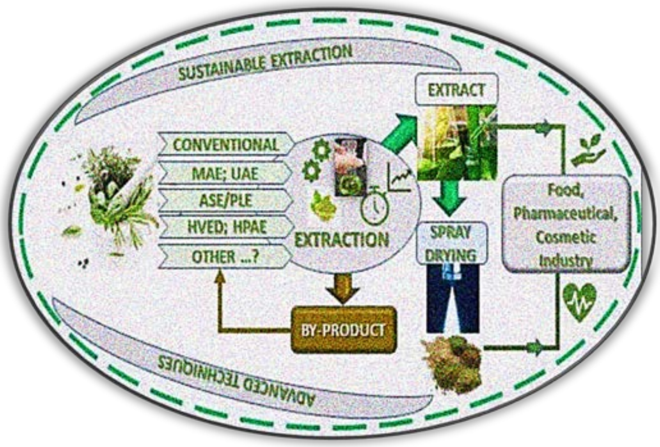

Figure 4. Sustainable extraction with advanced techniques (Popova A., 2018)

- **Microwave assisted extraction (MAE)** is suitable for the recovery of a vast array of compounds and is recognized as a versatile and efficient extraction technique of secondary plant metabolites. A lot of examples suggested that MAE has some considerable merits such as shorter extraction time, higher extraction yield, and less solvent consumption compared to conventional extraction methods. MAE utilizes microwave energy to facilitate partition of analytes from the sample matrix into the solvent. Microwave radiation interacts with dipoles of polar and polarizable materials (e.g., solvents and sample) causes heating near the surface of the materials and heat is transferred by conduction.

Dipole rotation of the molecules induced by microwave electromagnetic disrupts hydrogen bonding; enhances the migration of dissolved ions, and promotes solvent penetration into the matrix. In non–polar solvents, poor heating occurs as the energy is transferred by dielectric absorption only. MAE of plant secondary metabolites may be affected by a large variety of factors, such as power and frequency of microwave, duration of microwave radiation, moisture content and particle size of plant samples, type and concentration of solvent, ratio of solid to liquid, extraction temperature, extraction pressure, and number of extraction cycles. MAE was currently regarded as a robust alternative to traditional extraction techniques (Popova A, 2018; Azwanida NN, 2015).

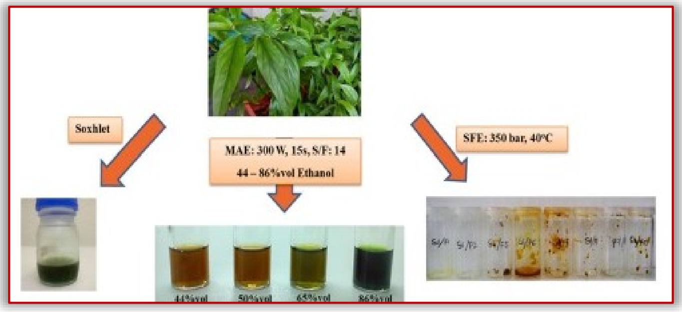

Figure 5. Extraction of phytocompounds from the medicinal plant Clinacanthus nutans Lindau by microwave–assisted extraction and supercritical carbon dioxide extraction (Popova A., 2018)

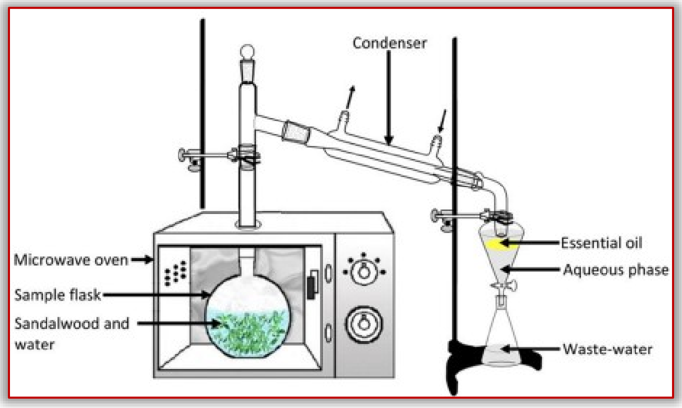

Figure 6. Microwave–assisted extraction (Nur Amanina Abd Aziz et al., 2021)

— **Ultrasound–assisted extraction (UAE)** is one of the most important techniques used for the extraction of valuable compounds from plant materials and is quite adaptable on a small or large scale (eg in the laboratory or on an industrial scale). Comparing this technique with others, such as microwave assisted extraction (MFA), the ultrasound device is cheaper and easier to handle (Ţuia Steluța, 2021; Popova A, 2018).

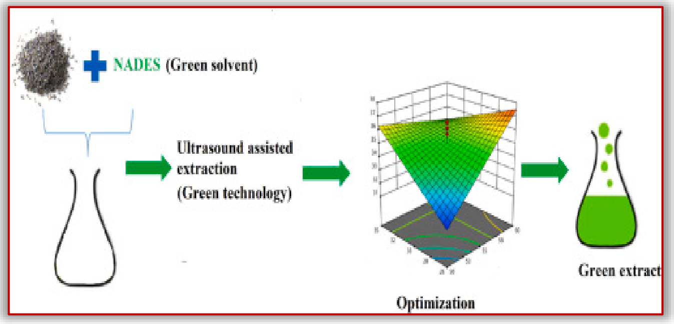

Figure 7. Ultrasound–assisted extraction of antioxidant phenolic compounds from Lavandula angustifolia flowers using natural deep eutectic solvents (Popova A., 2018)

— **Supercritical fluid extraction** has been developed in recent years for analytical use, as an alternative to conventional solvent extraction. In practice, more than 90% of supercritical fluid extractions are performed with CO2 for several practical reasons (Ţuia Steluța, 2021; Popova A, 2018).

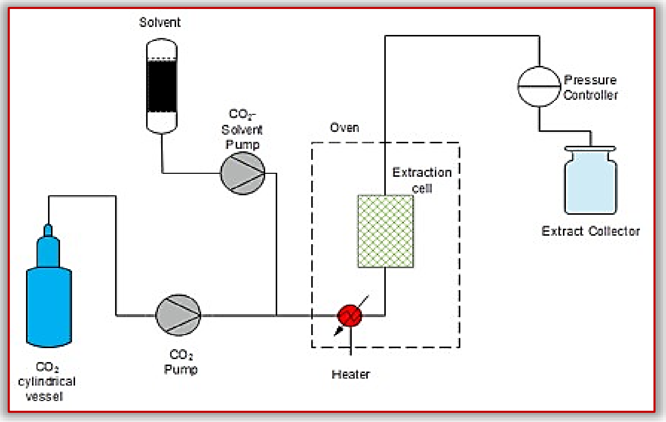

Figure 8. Supercritical fluid extraction (SFE) (Nur Amanina Abd Aziz et al., 2021)

CONCLUSIONS

All stages of extractions, from the pre–extraction and extraction are equally important in the study of medicinal plants. The sample preparation such as grinding and drying affected the efficiency and phytochemical constituents of the final extractions; that eventually have an effect on the final extracts. It can be concluded that, no universal extraction methods is the ideal method and each extraction procedures is unique to the plants.

The improvement of the extraction methods is essential for the most comprehensive obtaining of the compounds of bioactive substances from the vegetal resources, simultaneously with the isolation and standardization of each component substance of the extract.

The technologies regarding the extraction of the active principles and of conditioning of the vegetal extracts will allow to raise the qualitative level of the processing, with the possibility of obtaining innovative foods.

Advanced techniques for obtaining extracts of bioactive plant compounds offer wide applicability in the food, pharmaceutical, textile or cosmetic industries, meaning, lead to innovative foods such as functional foods, nutraceuticals, composite foods of excellence, and, can be used in the manufacturing process of certain drugs, natural food or industrial pigments, food or cosmetic flavors, etc.

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