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Review

APPLICATION OF SUPERCRITICAL FLUID EXTRACTION FOR THE SEPARATION OF NUTRACEUTICALS AND OTHER PHYTOCHEMICALS FROM PLANT MATERIAL

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In the present work, a literature review of the application of supercritical fluid extraction (SFE) for the isolation of nutraceuticals and some other phytochemicals up to December of 2012 is presented. The manuscript provides knowledge of SFE processes and possible applications of SFE for the extraction of bioactive compounds that serve as nutraceuticals. Compounds are classified into groups based on their chemical nature (carotenoids, flavonoids and other phenolic compounds, essential oils, lipids and fatty acids, and alkaloids and other bioactive phytochemicals), and they are reviewed in tabular form along with the plant material from which they were extracted using supercritical fluids.

Keywords: supercritical fluid extraction; nutraceuticals; phytochemicals; plant material

ПРИМЕНА НА СУПЕРКРИТИЧНАТА ФЛУИДНА ЕКСТРАКЦИЈА ЗА СЕПАРАЦИЈА НА НУТРИЕНТИ И ДРУГИ ФИТОХЕМИКАЛИИ ОД РАСТИТЕЛЕН МАТЕРИЈАЛ

Во овој труд е презентиран прегледот на литературата објавена до декември 2012 година, а се однесува на извлекување на нутриенти и некои фитохемикалии со суперкритичната флуидна екстракција (SFE). Презентирани се сознанијата за процесите на суперкритичната флуидна екстракција како можност за извлекување на биоактивни компоненти кои се користат како нутриенти. Соединенијата се класифицирани во групи врз основа на нивната хемиска природа (каротеноиди, флавоноиди и други фенолни соединенија, есенцијални масла, липиди, масни киселини, алкалоиди и други биоактивни фитохемикалии). Во трудов тие се презентирани во табели заедно со растителен материјал од кој се извлекувани со користење суперкритична флуидна екстракција.

Клучни зборови: суперкритична флуидна екстракција; нутриенти; фитохемикалии; растителен материјал

1. INTRODUCTION

In recent time, there is greater emphasis on the recovery of high value-added products by using sustainable technologies. One of the ways to achieve this is the application of sub- and supercritical fluids (ScF). ScF can be applied as solvents for precipitation and micronization (PGSS®, RESS, *etc.*), as reaction medium, as mobile phase for chromatography (supercritical fluid chromatography – SFC), and as solvent for extraction, which is the most investigated process. From an economic point of view, technologies involving elevated pressures require high investment costs for high–pressure equipment. Because of this, it is reasonable to apply supercritical fluid extraction (SFE) for the separation of components with high added value, such as nutraceuticals, pharmaceuticals, food additives, or components with high feed-to-solvent (F/S) extraction ratio.

SFE is a separation process where solid or liquid matter is processed with ScF in order to obtain soluble compounds from mixtures. ScF offers a variety of applications due to specific properties, which can be relatively easily adjusted with changing pressure and temperature. A fluid above critical temperature has gaslike viscosity, liquid-like density, and its diffusion magnitude of order is between the two fluid states.

An important factor that has to be considered is the mass transfer of the solute in the supercritical solvent. Mass transfer depends on the solubility of the solute in the given solvent. Different compounds have different solubilities at various operating conditions. In general, temperature and pressure have the biggest influence on the solubility of compounds in supercritical fluids. Temperature has two competing effects on solubility. First, increasing the temperature at constant pressure decreases the density of the solvent. Thus, solubility of the solute is decreased. On the other hand, by increasing the temperature at constant density, the vapor pressure of the solute is increased. Therefore a solute is more soluble in a supercritical fluid. Which effect will prevail is dependent on the properties of the system.

The effect of pressure is more direct. With increasing the pressure of a supercritical medium, higher densities are achieved; the higher the density of the medium, the higher the solubility of the solute [1]. The most common solvent used as a supercritical fluid is carbon dioxide (CO_2). When polar components are extracted and supercritical CO_2 (SC–CO_2) is used as a solvent, a polar modifier or co–solvent is mixed with SC–CO_2 to enhance solubility. Examples of such a modifier are methanol, ethanol, *etc*.

When a modifier is added, not only solubility but also viscosity and density are increased. Because of the increase in density and viscosity, the diffusivity in the mobile phase decreases, hence mass transfer is reduced.

To obtain the highest yields possible, the process has to be optimized. First, the influence of process parameters on the extraction has to be studied. Regarding the results, the optimal operating parameters are chosen. One way to determine the optimal parameters is with the use of response surface methodology (RSM). This method uses the multiple regression model (a polynomial second-order equation), from which optimum parameters are selected. This model is described as well in the work of Wang *et al.* [2], where optimization with RSM for SFE of essential oils from *Cyperus rotundus* is described.

Last but not the least, the extraction rates depend on the morphology of the material and the location of the solute in plant material. If the desired solute is on the surface of the material, generally extraction rates are high. However, when desired compounds are deeper in the material, it takes more time to extract them. In those cases, mass transfer depends on particle shape, size, and the porosity of the solid material. If the structure of the material is more complex and the desired compounds are deeper inside, a greater resistance for diffusion is expected [3–7]. Therefore, the preparation of a sample is very important for SFE of natural matter [8, 9]. Usually, a material has to be mechanically pretreated – *i.e.* mechanically processed by grinding, milling, cutting, *etc.* – to reduce mean particle size. As mentioned in this section, smaller particles provide faster extraction due to lower diffusion paths and less diffusion resistance. The importance of proper material pretreatment is presented in the work of Uquiche *et al.* [10], where kinetics of SFE of pretreated boldo was studied.

The aim of this work is to review investigations of the separation of compounds from natural matter with SFE performed in recent years. Several reviews have been published before. Among them were the reviews of Reverchon and De Marco [7], Pereira and Meireles [6], and Sovova and Stateva [11]. Hence there is no reason for repetition. The present work is focused on extractions of nutraceuticals and other phytochemicals from plant material and the collection of investigations in tabular form. Sections of this paper are divided into bigger groups (carotenoids, flavonoids and phenolic compounds, essential oils, lipid and fatty acids, and alkaloids and other phytochemicals). Compounds that belong to a certain group are listed in tabular form in the corresponding section. Key words used in the present work for searching the literature are listed in Table 1.

Table 1

Keywords used for searching the literature

Group	Subgroup	Keywords
Nutraceuticals	General	nutraceuticals, supercritical fluid extraction
	Carotenoids	carotenoids, supercritical fluid extraction
	Flavonoids	flavonoids, supercritical fluid extraction
	Phenolic compounds	phenolic, phenols, supercritical fluid extraction
	Essential oils	essential oils, supercritical fluid extraction
	Lipids, FFA	lipids, FFA, supercritical fluid extraction

2. SFE OF NATURAL MATTER: NUTRACEUTICALS

Nutraceuticals are substances that may be considered as food or part of a food and provide medical or health benefits, including the prevention and treatment of diseases [12]. Such products may range from isolated nutrients, dietary supplements and diets to genetically engineered “designer” food, herbal products, and processed food (such as cereals, soups, and beverages) [12].

In some aspects, functional food is related to nutraceuticals. Functional food provides health benefits over normal nutrition, and it is different from medical food or dietary supplements [12].

The nutraceuticals reviewed in this work are based on their chemical nature, classified into the following groups: carotenoids, flavonoids and other phenolic compounds, fatty acids and lipids, essential oils, and alkaloids and other phytochemicals.

2.1. *Carotenoids*

Carotenoids are divided into two subgroups: carotenes and oxygenized hydrocarbons xanthophylls. They consist of eight isoprenoid units joined in such a manner that their arrangement is reversed at the center of the molecule. All carotenoids may be formally derived from the acyclic $C_{40}H_{56}$ structure [13]. Trivial names are usually used for common carotenoids. The functional groups most frequently observed in the group of xanthophylls are hydroxy, methoxy, oxo, carboxy, and epoxy [13].

Carotenoids in food are recognized as antioxidants and pigments [14, 15]. As antioxidants, they affect human living tissues by preventing the oxidation of the molecules, inhibiting harmful microbiological activities, and protecting against cancer [16]. In Table 2, investigations of SFE of carotenoids from different plant material are listed.

Table 2

Application of SFE for the extraction of carotenoids from plants

Compound(s)	Plant material (Biological name)	Methods/solvents	Reference(s)
Astaxanthin	Microalgea (*Haematococcus pluvialis*)	SFE/CO_2 + ethanol	[23–26]
Astaxanthin	Microalgea (*Chlorella vulgaris*)	SFE/CO_2	[27]
Carotenes	Seabuckthorn (*Hippophae rhamnoids L.*)	SFE/CO_2	[28, 29]
Canthaxanthin	Microalgea (*Chlorella vulgaris*)	SFE/CO_2	[27]
Fucoxanthin	Microalgea (*Undaria pinnatifida*)	SFE/CO_2 + ethanol	[30]
Lutein	Daylily (*Hemerocallis disticha*)	SFE/CO_2	[31]
Lutein	Marigold (*Tagetes erecta*)	SFE/CO_2	[32–34]
Lutein	Spearmint (*Mentha spicata*)	SC–CO_2	[35]
Lutein	Pumpkin (*Curcurbita moschata*)	SE, SFE/CO_2	[36, 37]
Lutein	Stinging nettle (*Urtica dioica*)	SC–CO_2 + ethanol	[38]
Lutein	Microalgea (*Chlorella vulgaris*)	SFE/CO_2	[27, 39]
Lycopene	Fungal species (*Blakeslea trispora*)	SFE/CO_2	[40]
Lycopene	Hazelnut (*Corylus avellana L.*)	SFE/CO_2	[41]
Lycopene	Tomato (*Lycopersicum esculentum L.*)	SFE/CO_2 + ethanol, water, canola oil	[42, 43]
Lycopene	Tomato (*Lycopersicum esculentum L.*)	SFE/CO_2	[41, 44]
Lycopene	Tomato (*Lycopersicum esculentum L.*)	SFE/CO_2	[45, 46]
Lycopene	Tomato (*Lycopersicum esculentum L.*)	SE/acetone/water, SFE/CO_2	[16, 47–49]
Lycopene	Guava (*Psidium guajava*)	SFE/CO_2	[50]
Lycopene	Brazilian cherry (*Hymenaea courbaril*)	SFE/CO_2	[51]
Lycopene	Pumpkin (*Curcurbita moschata*)	SE, SFE/CO_2	[36, 37]
N.D.	Microalgea (*Chlorococcum littorale*)	SFE/CO_2	[52]
N.D.	Dog rose (*Rosa canina*)	SFE/CO_2 + ethanol	[53]
N.D.	lotus (*Nelumbo nucifera Gaertn*)	SFE/CO_2	[54]
N.D.	Persimmon peels (Japan)	SFE/CO_2 + ethanol	[55]
Xanthophylls	Paprika (*Capsicum annuum*)	SFE/CO_2	[56]
Zeaxanthin	Daylily (*Hemerocallis disticha*)	SFE/CO_2	[31]
Zeaxanthin	Microalgea (*Spirulina Pacifica*)	SFE/CO_2 + ethanol	[57]
Zeaxanthin	Microalgea (*Nannochloropsis oculata*)	SFE/CO_2	[58, 59]
Zeaxanthin	Microalgea (*Spirulina platensis*)	SFE/CO_2 + ethanol	[60]
Zeaxanthin	Microalgea (*Paracoccus zeaxanthinifaciens*)	SFE/CO_2 + ethanol	[61]
β-carotene	Paprika (*Capsicum annuum*)	SFE/CO_2	[56]
β-carotene	Red pepper (*Capsicum annum L.*)	SFE/CO_2	[62]
β-carotene	Palm oil (*Elaeis guineensis*)	SFE/CO_2	[63]
β-carotene	Fungal species (*Blakeslea trispora*)	SFE/CO_2	[40]
β-carotene	Tomato (*Lycopersicum esculentum L.*)	SFE/CO_2	[45, 46]
β-carotene	Microalgea (*Dunaliella bardawil*)	SFE/CO_2	[19]

β-carotene	Spearmint (*Mentha spicata*)	SFE/CO_2	[35]
β-carotene	Tomato (*Lycopersicum esculentum L.*)	SFE/CO_2	[16]
β-carotene	Lotus (*Nelumbo nucifera*)	SFE/CO_2	[64]
β-carotene	Brazilian cherry (*Hymenaea courbaril*)	SFE/CO_2	[51]
β-carotene	Microalgea (*Chlorella vulgaris*)	SFE/CO_2	[27]
β-carotene	Microalgea (*Dunaliella salina*)	SFE/CO_2	[27]
β-carotene	Microalgea (*Spirulina platensis*)	SFE/CO_2 + ethanol	[60]
β-carotene	Apricot (*Prunus armeniaca*)	SFE/CO_2	[65]
β-carotene	Carrot (*Daucus carota*)	SFE/CO_2	[66]
β-carotene	Paprika (*Capsicum annuum*)	SFE/CO_2; + DMP; + DEP; + TEOF	[8]
β-cryptoxanthin	Microalgea (*Spirulina pacifica*)	SFE/CO_2 + ethanol	[57]
β-carotene	Palm oil (*Elaeis guineensis*)	SFE/R134a	[67]

2.1.1. *Carotenes*

Palozza and Krinsky [17] reviewed carotenes and the biological antioxidant properties of *β*-carotene. The work of Palozza and Krinsky [17] also describes SFE and the influence of pressure and temperature on carotene extraction. It was concluded in most cases that by increasing pressure, hence increasing density, the solubility of carotenes in CO_2 generally also increases; therefore, higher extraction yields are achieved in less time at moderate temperatures. A bigger amount of extracted carotenes means bigger antioxidant potential. If extraction is carried out at a temperature that is too high, *β*-carotene could be decomposed; therefore less antioxidant potential is observed.

The extraction of *β*-carotene from material that contains too much water is less effective [8]. The material should be dried to achieve better extraction efficiency. Weathers *et al.* [8] suggested the use of freeze-drying in order to preserve thermolabile compounds.

Subra *et al.* [18] examined the influence of particle size and the influence of CO_2 and/or N_2O and their flow rates on the extraction of *β*-carotene from freeze–dried carrots. Subra *et al.* [18] observed that with smaller particles, higher yields were obtained, due to grinding, which destroyed the inner structures of particles. Furthermore, with higher solvent flow rates, higher extraction rates were achieved, due to an increased external mass transfer. Extractions carried out with N_2O were slightly faster than with CO_2 because of the higher solvent power of N_2O.

Gamlieli–Bonshtein *et al.* [19] investigated SFE for the separation of trans–cis isomers of *β*-carotene from concentrates containing several carotenoids. They concluded that SFE can be used to separate geometrical isomers of *β*-carotene based on their different solubilities in SC–CO_2. According to their investigations, it could be concluded that for SFE, an excessive research of solubility data is often required.

Baysal *et al.* [15] studied the extraction of *β*-carotene and lycopene from tomato wastes with CO_2. They investigated the influence of operating parameters (pressure, temperature, flow rate, extraction time, and co-solvent addition) on extraction yields. At the highest temperature, with a pressure of 30 MPa, yield was the highest; however, lycopene could not be totally recovered because of partial thermal degradation. In the presence of a co–solvent, the recovery of extracted compounds increased, because solubility increased [15]. A similar research was done by Doker *et al.* [20]. The modeling they performed [20] show that external and internal mass-transfer resistances are very important and control overall extraction rates.

Pressure is one of the most important parameters influencing the extraction and concentration of carotenes. By changing the pressure, the solubility of carotenes in SC-CO_2 is changed. When extraction is pressure-programmed, which means that pressure automatically or manually increases after a period of time, separate fractions of compounds can be collected. Lau *et al.* [21] investigated the pressure-programmed extraction of palm-pressed mesocarp fiber, where pressure was subsequently increased, first from 10 MPa to 20 MPa after the initial 3 h, then from 20 MPa to 30 MPa after 1 h, where it was maintained for 6 h. β-carotene (provitamin of vitamin E) and squalene were extracted at lower pressures (10 MPa) of SC-CO_2; therefore these compounds were collected mainly in the early stage, while other carotenes were collected in the following fractions at pressures above 20 MPa. Often co–solvents such as methanol, ethanol, or acetone are used to enhance solubility, but in some cases edible oils as co-solvents are used for the same purpose [22].

In the work of Ciurlia *et al.* [41], an innovative separation method is presented, where dried tomato and roasted hazelnuts are mixed. Oil extracted from hazelnut helps with the extraction of carotenes, acting as an actual co–solvent. This method is interesting because it requires less addition of other co-solvents.

Sanal *et al.* [65] investigated parameters that might influence extraction yields besides those previously mentioned in this section. The influence of drying procedure, sample amount, and influence of DMP could be obtained from their work [65].

In order to calculate extraction yields, extracts were characterized with several analytical methods, such as high-performance liquid chromatography (HPLC) and spectrophotometry in the UV/VIS region. In the literature [68–70], some other analytical methods are described.

2.1.2. *Xanthophylls*

Xanthophylls are a group of naturally occurring oxygenized carotenoids produced mainly by plants and microorganisms. They are applied as food additives and food colorants for antioxidant purposes and cancer prevention. Lutein, zeaxanthin, and cryptoxanthin are major xanthophyll carotenoids in the human plasma. Besides those, there are several other important xanthophylls, such as canthaxanthin, astaxanthin, neoxanthin, and violaxanthin [71].

Lutein and zeaxanthin are both present in the cells of the human eye [72]. Lutein not only serves as a pigment or colorant but also prevents diseases, such as ophthalmopathy [55]. Takahashi *et al.* [55] carried out the extraction of xanthophylls and other carotenoids from the Japanese persimmon with the addition of ethanol as modifier. It was observed that a higher concentration of the added modifier decreases the selectivity of the solvent for carotenoids, hence other compounds other than carotenoids are extracted. Like modifiers, mostly organic solvents are used, but vegetable oils can modify the mobile phase as well. Gao *et al.* [33] investigated the extraction of lutein from the marigold flower with CO_2 in the presence of vegetable oils as modifier. Results showed that the addition of edible oils to the mobile phase results to a higher amount of lutein extracted. Other sources of lutein are listed in Table 2 along with other carotenoids extracted from plants. Wu *et al.* [73] investigated SFE of lutein from cultivated *Chlorella pyrenoidosa*, and they observed that this species contain an amount of lutein comparable to that found in the marigold flower; hence this culture could be used as an alternative.

Among other xanthophylls, astaxanthin provides health benefits also. It contains two oxygenated groups on each ring structure, which are responsible for its enhanced antioxidant properties [74]. Astaxanthin can be obtained by SFE from pigments of *H. pluvialis* [24] and some other microalgae or sea fruits [27, 75–79].

2.2. *Flavonoids and other phenolic compounds*

Applications of SFE for the extraction of flavonoids and other phenolic compounds from plant material are presented in Table 3.

Table 3

Application of SFE for separation of phenolic compounds from plants

Compound(s)	Plant material (Biological name)	Methods/solvents	Reference(s)
Anthocyanins	Raspberry (*Rubus idaeus*)	SFE/CO_2	[109]
Anthocyanins	Blueberry (*Vaccinium corymbosum*)	SFE/CO_2	[109]
Anthocyanins	Cranberry (*Vaccinium oxycoccos*)	SFE/CO_2	[109]
Anthocyanins	Elderberry (*Sambucus nigra L.*)	SFE/CO_2	[110]
Apigenin	Chamomile (*Chamomilla recutita*)	SFE/CO_2 + ethanol	[94, 101, 102, 111]
Apigenin	Olive (*Olea europaea*)	SFE/CO_2	[112]
Apigenin	Strobilanthes crispus	SFE/CO_2 + ethanol	[113]
Apigenin	Marchantia convoluta	SFE/CO_2 + ethanol	[92, 114–116]
Apigenin	Shiyacha (*Adinandra nitida*)	SFE/CO_2 ; methanol	[117]
Apigenin	Spearmint (*Mentha spicata L.*)	SFE/CO_2	[118, 119]
Artepillin C	Brazilian propolis	SFE/CO_2	[120]
Artepillin C	Brazilian propolis	SFE; SE/CO_2 + ethyl acetate; ethyl acetate	[121, 122]
Artepillin C	Brazilian propolis	SFE; SE/CO_2 + ethanol; ethanol, ethyl acetate, chloroform, *n*-hexane, water, water + ethanol	[123]
Baicalin	Baikal skullcap (*Scutellaria baicalensis*)	SFE/CO_2 + methanol, + ethanol, + 1,2-propanediol	[98]
Baicalin	Blue skullcap (*Scutellaria lateriflora*)	SFE/CO_2 + ethanol, SHW	[97]
Baicalin	Skullcap (*Scutellariae radix*)	SE; SFE/CO_2 + methanol	[124]
Baicelein	Skullcap (*Scutellariae radix*)	SE; SFE/CO_2 + methanol	[124]
Camellianin A	Shiyacha (*Adinandra nitida*)	SFE/CO_2; methanol	[117]
Camellianin A	Macela (*Achyrocline satureioides*)	SFE/CO_2 + ethanol	[125]
Catechin	Tea (*Camellia sinensis*)	SFE/CO_2	[126]
Catechin	Rooibos (*Aspalathus linearis*)	SFE/CO_2	[126]
Catechin	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol	[127]
Catechin	Grape	SE; SFE/ethanol + water; CO_2, + ethanol	[83, 84, 87, 92, 93, 128–130]
Catechin	Spearmint (*Mentha spicata L.*)	SFE/CO_2	[118, 119]
Catechin	Olive (*Olea europaea*)	SE; SFE/ethanol; CO_2	[131]
Catechin	Pine tree	SFE/CO_2; SFE/CO_2 + ethanol	[132, 133]
Catechin	Berries	SFE/CO_2	[109]
Catechin	Marigold (*Calendula officinalis*)	SFE/CO_2 + ethanol	[101]

Catechin	Hawthorn (*Crataegus apathulata*)	SFE/CO_2 + ethanol	[101]
Catechin	Scented mayweed (*Matricaria recutita*)	SFE/CO_2 + ethanol	[101]
Catechin	Strobilanthes crispus	SFE/CO_2 + ethanol	[113]
Cinnamic acid	Olive oil residues (*Olea europaea*)	SFE; SE/CO_2; ethanol	[131]
Cinnamic acid	Macela (*Achyrocline satureioides*)	SFE/CO_2, CO_2 + ethanol	[125]
Cinnamic acid	Cherry (*Prunus avium*)	SFE/CO_2, CO_2 + ethanol	[134]
Cirsimaritin	Rosemary (*Rosmarinus officinalis*)	SFE/CO_2 + ethanol	[135]
Corilagin	Barbados nut (*Jatropha curcas*)	SFE/CO_2 + methanol	[136]
Coumaric acid	Brazilian propolis	SFE/CO_2	[120]
Coumaric acid	Olive oil residues (*Olea europaea*)	SFE; SE/CO_2; ethanol	[131]
Coumarins	Rice (*Oryza sativa*)	SFE/CO_2 + ethanol	[137]
Coumarins	Dodder (*Cuscuta reflexa*)	SFE/CO_2 + methanol	[138]
Coumarins	Wormwood (*Artemisia capillaris*)	SFE/CO_2	[139]
Coumarins	Emburana (*Torresea cearensis*)	SFE/CO_2	[140]
Coumarins	Sweet grass (*Hierochloe odorata*)	SE; MAE; SFE/organic solvents; CO_2	[141]
Coumarins	Dodder (*Cuscuta reflexa*)	SFE/CO_2 + methanol	[138]
Coumarins	Bai Zhi (*Angelica dahurica*)	SFE/CO_2 + ethanol	[142, 143]
Daidzein	Soybean (*Glycine max*)	SFE/CO_2 + methanol/water, + ethanol	[96, 105, 144]
Fucocoumarin	Bishop's flower (*Ammi majus*)	SFE/CO_2 + ethanol	[145]
Gallic acid	Rice (*Oryza sativa Linn.*)	SFE/CO_2 + ethanol	[137]
Gallic acid	Barbados nut (*Jatropha curcas*)	SFE/CO_2 + methanol	[136]
Galic acid	Macela (*Achyrocline satureioides*)	SFE/CO_2 + ethanol	[146]
Ellagic acid	Barbados nut (*Jatropha curcas*)	SFE/CO_2 + methanol	[136]
Genistein	Soybean (*Glycine max*)	SFE/CO_2 + methanol; water + ethanol	[96, 105, 144]
Genistein	Wild Cherry (*Prunus avium*)	SFE/CO_2	[134]
Genistein	Grape	ethanol/water; SFE/CO_2 + ethanol, + methanol	[86, 147, 148]
Genistein	Grape	SFE/CO_2; SFE/CO_2 + ethanol	[85, 87, 89, 128]
Genistein	Black currant (*Ribes nigrum*)	SE; SFE/methanol; CO_2 + methanol	[149]
Genistein	Black currant (*Ribes nigrum*)	SE; SFE/ethanol; CO_2	[150]
Genistein	Elderberry (*Sambucus*)	SFE/CO_2	[91]
Genistein	Sour cherry (*Prunus cerasus*)	SFE/CO_2	[151]
Glabridin	Licorice (*Glycyrrhiza glabra*)	SFE/CO_2	[152]

Isocoumarin	Coriander (*Coriandrum sativum*)	SFE/CO_2	[153]
Kaempferol	Strobilanthes crispus	SFE/CO_2 + ethanol	[113]
Kaempferol	Tea (*Camellia sinensis L.*)	SFE/CO_2	[126]
Kaempferol	Rooibos (*Aspalathus linearis*)	SFE/CO_2	[126]
Kaempferol	Indian gooseberry (*Phyllanthus emblica L.*)	SFE/CO_2 + methanol; methanol	[149]
Kaempferol	Black currant (*Ribes nigrum*)	SE; SFE/methanol; CO_2 + methanol	[149]
Kaempferol	Black currant (*Ribes nigrum*)	SE; SFE/ethanol; CO_2	[150]
Kampheride	Brazilian propolis	SFE/CO_2	[120]
Lignan	Five-flavor berry (*Schisandra chinensis*)	SFE/CO_2, SC–CO_2 + ethanol	[154]
Lignan	Five-flavor berry (*Schisandra chinensis*)	USE; SFE/CO_2, CO_2 + ethanol	[155]
Lignan	*Forsythia* species	SE; SFE/ethanol, methanol; CO_2, + methanol, + ethanol	[156]
Lignan	Flax (*Linum usitatissimum*)	SFE/CO_2 + ethanol	[157]
L–DOPA	Mucuna	SFE/CO_2	[158]
Myricetin	Crowberry (*Empetrum nigrum*)	SE, SFE/ethanol, CO_3	[159]
Myricetin	Tea (*Camellia sinensis L.*)	SFE/CO_2	[126]
Myricetin	Rooibos (*Aspalathus linearis*)	SFE/CO_2	[126]
Myricetin	Spearmint (*Mentha spicata L.*)	SFE/CO_2	[118, 119]
Naringenin	Aspen (*Populus tremula*)	PHWE	[160]
Naringenin	Spearmint (*Mentha spicata L.*)	SFE/CO_2	[118, 119]
Naringin	Grapefruit (*Citrus paradisi*)	SFE/CO_2 + ethanol	[161]
Orientin	Maypop (*Passiflora incarnata*)	SFE/CO_2 + ethanol, + methanol, + CHCl3	[82]
Orientin	Pigeon pea (*Cajanus cajan L.*)	SFE/CO_2	[162]
Orotinin	Patrinia villosa Juss.	SFE/CO_2 + ethanol	[163]
Osthole	Cnidium monnieri	SFE/CO2	[164]
Pinostrobin	Pigeon pea (*Cajanus cajan L.*)	SFE/CO_2	[162]
Polyphenols	Cocoa	SFE/CO_2 + ethanol	[165]
Quercetin	Crowberry (*Empetrum nigrum*)	SE, SFE/ethanol, CO_2	[159]
Quercetin	Tea (*Camellia sinensis L.*)	SFE/CO_2	[126]
Quercetin	Rooibos (*Aspalathus linearis*)	SFE/CO_2	[126]
Quercetin	Onion (*Allium cepa*)	SFE; SWE/CO_2 + ethanol; SFE/CO_2	[166, 167]
Quercetin	Hypericum	SFE/CO_2 + methanol	[100]
Quercetin	Grape	SFE/CO_2 + ethanol	[84]
Quercetin	Macela (*Achyrocline satureioides*)	SFE/CO_2 + ethanol	[146]
Resveratol	Grape	SFE/CO_2 + ethanol	[84]
Resveratol	Polygonum cuspidatum	SFE/CO_2 + ethanol	[168]
Resveratol	Hop (*Humulus lupulus*)	SFE/CO_2	[169]

Rhamnetin	Marigold (*Calendula officinalis*)	SFE/CO_2 + ethanol	[101]
Rhamnetin	Hawthorn (*Crataegus spathulata*)	SFE/CO_2 + ethanol	[101]
Rhamnetin	Scented mayweed (*Matricaria recutita*)	SFE/CO_2 + ethanol	[101]
Rutin	Tea (*Camellia sinensis L.*)	SFE/CO_2	[126]
Rutin	Rooibos (*Aspalathus linearis*)	SFE/CO_2	[126]
Rutin	Elderberry (*Sambucus nigra*)	ASE	[170]
Rutin	Olive (*Olea europaea*)	SFE/CO_2	[112]
Rutin	Spearmint (*Mentha spicata L.*)	SFE/CO_2	[118, 119]
Rutin	Strobilanthes crispus	SFE/CO_2 + ethanol	[113]
Rutin	Passionflower (*Passiflora incarnata*)	SFE/CO_2 + ethanol, + methanol, + $CHCl_3$	[82]
Scutellarein	Rosemary (*Rosmarinus officinalis*)	SFE/CO_2 + ethanol	[135]
Stilbenes	Pigeon pea (*Cajanus cajan L.*)	SFE/CO_2	[162]
Tannin	Canola (*Brassica napus L.*)	SE; SFE/hexane; CO_2, SC-CO_2 + ethanol	[171, 172]
Tannin	Stonebreaker (*Phyllanthus niruri*)	SE; SFE; PWHE/organic solvents; CO2; H2O	[173, 174]
Thymoquinone	Black cumin (*Nigella sativa*)	SFE/CO_2 + ethanol	[175]
Vitexin	Maypop (*Passiflora incarnata*)	SFE/CO_2 + ethanol, + methanol, + CHCl3	[82]
Vitexin	Pigeon pea (*Cajanus cajan L.*)	SFE/CO_2	[162]
Wogonin	Blue skullcap (*Scutellaria lateriflora*)	SFE; SubWE/CO_2 + ethanol; water	[97]
Wogonin	Skullcap (*Scutellariae radix*)	SE; SFE/CO_2 + methanol	[124]
Xanthone	Mangosteen (*Garcinia mangostana*)	SFE/CO_2 + ethanol	[176–178]
γ-oryzanol	Rice (*Oryza sativa*)	SFE/CO_2	[179–181]
N.D.	Bitter melon (*Momordica charantia*)	SFE/CO_2 + ethanol	[182]
N.D.	Pomelo (*Citrus grandis*)	SFE/CO_2 + ethanol	[183]

Flavonoids are phenolic compounds that serve as antioxidants and are found in plants as pigments [80]. There are many flavonoid classes; the most common are flavanes, flavones, flavanones, flavonols, isoflavonoids, and anthocyanins. Their structure is usually characterized by a C6–C3–C6 carbon skeleton with double bonds and attached oxygen functional groups [81]. One of the earliest studies in this field has been done by Moraes *et al.* [82], where the isolation of glycosylated flavonoids from *Passiflora edulis* has been carried out. One of potential sources of flavonoids is grape and its by-pruducts. Several authors researched the extraction of flavonoids from grape and grape by-products [83–93]. Results obtained from these researches can majorly contribute to grape-processing industries and wineries in terms of giving information on how to achieve a higher recovery of some flavonids extracted with SFE, besides those obtained in grape processing. Flavonoids are polar compounds, and because of this, there are only few studies on their separation with pure CO_2. Such example is the isolation of the moderately polar apigenin from chamomile, where the investigations showed that extraction with SC–CO_2 is more rapid than other conventional methods [94].

Because of the polarity of flavonoids, polar modifiers have to be added to SC–CO_2 to increase solubility. Numerous researches were performed, where the influence of modifiers was investigated [84, 90, 94–107]. These studied showed that the addition of a modifier generally increases the efficiency of extraction by increasing yields. However, too much co-solvent used is not economical, due to the higher amount of energy required for its removal.

As mentioned, mainly CO_2 is used for SFE, but other fluids could be used as well. Chiu *et al.* [99] investigated the extraction of flavonoids from ginkgo using CO_2, N_2O, and Freon 1,1,1,2-tetrafluoroethane (R134a). Each supercritical medium itself could not extract flavonoids from dried ginkgo leaves in this case. Successful extraction was achieved with the addition of ethanol as modifier [99].

Before undertaking an individual SFE process, often a sample preparation is required. Antolovich *et al.* [95] reviewed techniques in the preparation of samples for the extraction of phenolic compounds from fruits, then described the analytical methods for the determination of phenolic compounds present in the processed material. The particle size of the material played an important role. Liu *et al.* [108] investigated the influence of the particle size of *M. stigma* on extraction yields of flavonoids. When particle size is reduced, contact area is increased; hence yields got higher. Oppositely, when the particles are too small, this lowers the extraction yield, due to agglomeration [108].

Not only SFE but also pressurized hot–water extraction (PHWE) [184, 185] and accelerated solvent extraction (ASE) [170] could be applied in order to ensure sustainability. Ollanketo *et al.* [185] investigated PHWE of antioxidant compounds (among them were also flavonoids) from sage (*Salvia officinalis*). This method is promising and presents an alternative to conventional processes; however, the disadvantage is thermal degradation of thermolabile compounds.

Senorans *et al.* [186] and Simo *et al.* [187] carried out countercurrent supercritical fluid extraction (CC–SFE) of flavonoids from orange juice. After extraction, further separation of flavonoids with reversed-phase liquid chromatography (RP–LC) and/or micellar electrokinetic chromatography (MEKC) was performed in order to fractionate compounds. CC–SFE was performed in extraction column filled with spherical steel packing. This packing material provides a larger contact area, which allows more efficient separations [186]. This technique is promising for the separation of other nutraceuticals from liquid samples (*e.g.* extracts).

SFE was applied to sample preparation of *Rhodiola rosea*, and further analysis of flavonoids was performed by liquid chromatography in tandem with mass spectroscopy [188]. Flavonoids are the most often determined and identified by chromatographic methods such as thin layer chromatography (TLC) [189], RP–LC with C8- or C18-bonded silica column packing. Liquid chromatography could be coupled with mass spectroscopy (LC–MS). Those methods are usually applied with a diode array detector (DAD). High-performance liquid chromatography (HPLC) could also be coupled with electrospray ionization detection (ESI), or light–scattering detection (LSD). Another way of identifying compounds is gas chromatography (GC) coupled with mass spectroscopy (GC–MS). Detailed identification and determination methods of various phenolic compounds are described in the literature [135, 141, 190–192]. Klejdus *et al.* [193] investigated a new analytic approach by using supercritical fluids, *i.e.* solid–phase/supercritical fluid extraction (SPE/SFE), which provides a more efficient isolation and identification of simple phenols and phenolic acids as only by using CO_2. Crego *et al.* [194] proposed subcritical water extraction (SubWE) of phenolic compounds from rosemary in combination with capillary electrophoresis (CE). The water used in those processes contributed to the sustainability of said processes. The application of SubWE is useful for the isolation of more polar compounds, and extraction could be achieved faster in a less viscous extraction medium.

The utilization of renewable resources, *e.g.* waste from the forestry industry, is becoming more attractive. Therefore, Yesil-Celiktas *et al.* [133] proposed the recovery of some flavonoids from pine barks using SFE with CO_2.

Flavonoids, as known, help to prevent diseases with their antioxidant, antiviral, antimicrobial, and cancer prevention properties; therefore they could be used as medicines. Miao *et al.* [195] studied SFE of flavonoids from ginkgo biloba leaves with CO_2 and ethanol as co–solvent, coupled with micronization, in order to obtain smaller particles for better drug release in the body.

The future trend of SFE investigations is scaled up from an analytical scale to preparative or even an industrial scale. Such processes have to be optimized in order to achieve cost–effective operations. Therefore solvents shall be recycled. Often organic solvents are used as modifiers of SC-CO_2. In order to achieve successful recycling, evaporation of the modifier is required. The evaporation step consumes a great amount of energy. If an extract is gathered in a collective trap, filled with a co-solvent, the evaporative step could be avoided, which presents a more economical solution [88].

Although SFE serves mainly for the isolation of certain compounds from plant matrix, it could be used for the pretreatment of sample preparations for further processing. Xu *et al.* [196] observed that SFE could break cell walls in order to extract lysed bee pollen oils from rape bee pollen more efficiently. Compounds in the extract could be separated afterward with chromatographic methods. Supercritical fluid chromatography (SFC) is one of many chromatographic methods that were rarely used in the past, but lately it has become more attractive. Ramirez *et al.* [197], [198] studied rosemary extraction followed by SFC. The extracted compounds could be analysed withouth further sample preparation (online analysis) if analytical SFC is applied, or even fractionationated in the case of preparative SFC (prep SFC) [197].

Generally, the solubility of a compound in a solvent has to be known in order to optimize the extraction and fractionation of the solutes. The importance of solubility data could be observed in the work of Nunes *et al.* [199], where a study of the extraction of phenolic compounds (catechin, qurcetin-3-glycoside, coumaric acid, resveratol) from water-alcohol mixtures (such as wine) was conducted. At elevated pressures, water is poorly miscible with SC–CO_2; on the other hand, ethanol is completely miscible above supercritical conditions. Therefore two phases are formed when pressurized CO_2 is introduced into a water-ethanol mixture. Observations of Nunes *et al.* [199] confirmed that the ethanol-rich phase contains more phenolic compounds. This advantage could be used for the separation of flavonoids from alcoholic beverages, such as wine.

SCF can be applied as an alternative solution for the extraction of oils from biomass, rich with several phenolics, esters, and furan derivatives. Such extraction was investigated by Chumpoo and Prasassarakich [200], where supercritical ethanol in inert H_2 atmosphere was used for the extraction of oils from biomass. This study provides knowledge about supercritical ethanol extraction, where it's mandatory to use inert atmosphere to avoid combustion. Supercritical ethanol extraction is like SubWE, an appropriate alternative to SFE, where mainly modified CO_2 is used, because it can separate polar compounds more effectively. On the other hand, this method is not as economical as SFE with CO_2 because of the higher price of ethanol.

Flavonoids cover a wide range of phenolic compounds, but there are other phenolic compounds in the natural matrices that affect human health. They are grouped into the following categories (besides flavonoids): simple phenols, benzoic acid derivates, stilbenes, tannins, lignans, and lignins [201]. Some of those compounds were extracted by SFE–*e.g.* phenylacetic acids [137]; cinnamic acids, coumarins, isocoumarins [138–143, 145, 153, 164, 174, 202], and chromones; lignans [155–157]; tannins [171, 172]; xanthones [178] and stilbenes [64, 84, 106, 168, 169, 189]; and betacyanins [203]. In the work of Wang *et al.* [204], SFE combined with liquid-liquid extraction (LLE) for the separation of coumarins from *Angelica dahurica* is presented. First, SFE of material was performed, and the extract obtained was afterwards extracted by LLE. SFE extract was diluted in petroleum ether. The prepared sample was extracted first with water, then with a 60 % methanol/water solution with LLE. Selectivity and separation efficiency were better than with SFE alone [204]. Phenyllpropanoides, benzoic acid derivates, coumarins, and isocoumarins are very important from a pharmacological point of view, because they prevent diseases and have antioxidant, antimicrobial, anti-inflammatory, and cancer-preventive properties. Coumarins could also help in treating AIDS, among other properties.

2.3. *Essential oils*

Investigations of SFE of essential oils from natural matter are listed in Table 4.

Essential oils are volatile and liquid aroma components present in plant materials. Often they are poorly soluble in water and have characteristic odors. Therefore, they are used in food flavoring and perfumery.

Essential oils are mostly separated with hydrodistillation (HD), including steam distillation (SD) [205]. Many investigations and comparisons between conventional and newer separation processes (as SFE, PHWE, microwave–assisted hydrodistillation [MAHD], and solid-phase micro extraction [SPME]) were done [206–215]. Results obtained showed that conventional processes in general give higher global yields. On the other hand, newer processes, such as SFE, provide faster separation. The properties of a solvent in SFE could be manipulated by just adjusting operation parameters, and a wide range of applications is possible. However, SFE is not always the optimal solution, because the solubility of a desired component in supercritical media is not satisfactory in that case. Even more, the depressurization step of CO_2 and the renewed repressurization of SFE is a big disadvantage, because more energy is consumed. Carlson *et al.* [216] proposed the use of reverse-osmosis membranes to avoid instantaneous expansion and to lower operating costs.

Undesired racemization, where risky enantiomers are included in the racemic mixture, could be avoided [217]. First, the plant material is pretreated with SFE and then is extracted and further fractionated with SFC in chiral mode. This procedure is appropriate for the isolation of bioactive compounds, which could be added in food, which requires higher purity, or could be used as medicine, on the other hand.

The main application of SFE in the field of essential oils is the isolation of terpenes (sesquiterpenes) or terpenoids from plant material. When the presence of terpenes and terpenoids is not desired in the final products, the process is called deterpenation [218–220]. Mostly, CO_2 is applied for SFE of essential oils, but ethane could be a great alternative, because its solvent power for the extraction of essential oils is higher, which is desired in the deterpenation process [221].

Table 4

Application of SFE for the separation of phenolic compounds from plants

Compounds	Plant material (Biological name)	Method/solvent	Reference(s)
1,8-Cineole	Swamp mallet (*Eucalyptus spathulata*)	HD; SFE/CO_2	[222]
1,8-Cineole	Coolabah (*Eucalyptus microtheca*)	HD; SFE/CO_2	[222]
1,8-Cineole	French lavender (*Lavandula x intermedia Grosso*)	SFE/CO_2	[217]
1,8-Cineole	Lavandin abrial (*Lavandula x intermedia Abrial*)	SFE/CO_2	[217]
1,8-Cineole	Lavandin Puper (*Lavandula x intermedia Super*)	SFE/CO_2	[217]
1,8-Cineole	Sage (*Salvia officinalis L.*)	HD; SFE/CO_2	[223–226]
1,8-Cineole	French lavender (*Lavandula stoechas*)	HD; SubWE/H2O	[209]
1,8-Cineole	Khoa (*Satureja boliviana*)	SFE/CO_2 + ethanol	[146]
1,8-Cineole	Cotton lavender (*Santolina chamaecyparissus*)	HD; SFE/CO_2	[227]
1,8-Cineole	Sweet bay (*Laurus nobilis L.*)	SFE/CO_2	[228, 229]
1,8-Cineole	Saigon cinnamon (*Cinnamomum loureiroi*)	SFE/CO_2	[230]
1,8-Cineole	Clove basil (*Ocimum gratissimum*)	SFE/CO_2	[231]
1,8-Cineole	Argyle apple (*Eucalyptus cinerea*)	HD; SFE/CO_2	[232]
1,8-Cineole	Sweet gale (*Myrica gale L.*)	SFE/CO_2	[233]
1,8-Cineole	French lavender (*Lavandula stoechas*)	SFE/CO_2	[220]
1,8-Cineole	Peppermint (*Mentha piperita*)	SFE/CO_2	[234]
4-Nonanone	(*Capillipedium parviflorum*)	HD; SFE/CO_2	[235]
4-Nonanol	(*Capillipedium parviflorum*)	HD; SFE/CO_2	[235]
4-Terpineol	Black cumin (*Nigella sativa L.*)	MAE; SFE/CO_2	[202]
4-Undecanone	(*Capillipedium parviflorum*)	HD; SFE/CO_2	[235]
4-Undecanol	(*Capillipedium parviflorum*)	HD; SFE/CO_2	[235]
Artemisinin	Sweet wormwood (*Artemisia annua*)	SFE/CO_2+ ethanol	[236]
Bicyclogermacrene	Peruvian pepper (*Schinus molle*)	SFE/CO_2	[237]
Boldine	Boldo (*Peumus boldus*)	PHWE; SFE/CO_2	[208]
Cadinol	Marigold (*Calendula officinalis*)	SFE/CO_2	[238]
Camphor	Lavandin abrial (*Lavandula x intermedia Abrial*)	SFE/CO_2	[217]
Camphor	Sage (*Salvia officinalis*)	SFE/CO_2	[223]
Camphor	Sage (*Salvia officinalis L.*)	HD; SFE/CO_2	[223–226]
Camphor	French lavender (*Lavandula stoechas*)	HD; SubWE/H_2O	[209]
Camphor	Cotton lavender (*Santolina chamaecyparissus*)	HD; SFE/CO_2	[227]
Camphor	(*Thymus lotocephalus G. Lopez & R. Morales*)	SFE/CO_2	[239]
Camphor	Borage (*Borago officinalis*)	HD; SFE/CO_2 + methanol	[240]
Camphor	French lavender (*Lavandula stoechas*)	SFE/CO_2	[220]

Camphor	Lavender (*Lavandula angustifolia*)	SFE/CO_2	[241, 242]
Camphor	Lavender (*Lavandula viridis*)	HD; SFE/CO_2	[243]
Carnosic acid	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2 + ethanol	[197, 212, 225, 244–250]
Carnosic acid	Rosemary (*Rosmarinus officinalis L.*)	SubWE	[184, 194]
Carnosic acid	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2	[251–253]
Carnosic acid	Rosemary (*Rosmarinus officinalis L.*)	SFE; PLE/CO_2; H_2O, ethanol	[254]
Carnosol	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2 + ethanol	[197, 212, 225, 244–250]
Carnosol	Rosemary (*Rosmarinus officinalis L.*)	SubWE	[184, 194]
Carnosol	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2	[251–253]
Carnosol	Rosemary (*Rosmarinus officinalis L.*)	SFE; PLE/CO_2; water, ethanol	[254]
Carvacrol	Oregano (*Origanum vulgare*)	SFE/CO_2	[218, 255, 256]
Carvacrol	Summer savory (*Satureja hortensis*)	HD, SFE/CO_2 + methanol	[257]
Carvacrol	Winter savory (*Satureja montana*)	HD; SFE/CO_2	[258]
Carvacrol	Lippia origanoides	SFE/CO_2	[214]
Carvacrol	Black cumin (*Nigella sativa L.*)	MAE; SFE/CO_2	[202]
Carvone	Indian dill (*Anethum sowa*)	HD; SFE/CO_2	[259]
Carvone	Spearmint (*Mentha spicata*)	SE; SFE/hexane, DCM, ethanol; CO_2	[260]
Caryophyllene	Curry plant (*Helichrysum italicum*)	HD; SFE/CO_2	[261]
Caryophyllene	Khoa (*Satureja boliviana*)	SFE/CO_2 + ethanol	[262]
Caryophyllene	Oregano (*Origanum vulgare*)	SFE/CO_2	[218, 255, 256]
Caryophyllene	Oregano (*Origanum vulgare*)	SubWE	[263]
Caryophyllene	Peruvian pepper (*Schinus molle*)	SFE/CO_2	[237]
Caryophyllene	Indian gooseberry (*Phyllanthus emblica L.*)	SFE/CO_2	[149]
Caryophyllene	Madeira mahogany (*Persea indica*)	SFE/CO_2 + ethanol	[264]
Caryophyllene	Honeyherb (*Lippia dulcis*)	HD; SFE/CO_2	[265]
Caryophyllene	Sri Lanka cinnamon (*Cinnamomum zeylanicum*)	SFE/CO_2	[266]
Cedrene	Dragonhead (*Dracocophalum tanguticum*)	SFE/CO_2	[267]
Chrysanthenol acetate	Wormwood (*Artemisia absinthium L.*)	SFE/CO_2 + ethanol	[268]
Cinnamonaldehyde	Cinnamon (*Cinnamomum verum*)	HD; SFE/CO_2	[269, 270]
Cinnamonaldehyde	Saigon cinnamon (*Cinnamomum loureiroi*)	SFE/CO_2	[230]
Cinnamonaldehyde	Sri Lanka cinnamon (*Cinnamomum zeylanicum*)	SFE/CO_2	[266]
Citral	Lemongrass (*Cymbopogon citrates*)	SD; SFE/CO_2	[271]
Citronellol	Carqueja (*Baccharis genistelloides*)	SFE/CO_2	[272]
Copaene	Saigon Cinnamon (*Cinnamomum loureiroi*)	SFE/CO_2	[230]
Copaene	Honeyherb (*Lippia dulcis*)	HD; SFE/CO_2	[265]
Curcumene	Curry plant (*Helichrysum italicum*)	HD; SFE/CO_2	[261]

Cycloartenol	Carrot (*Daucus carota*)	SFE/CO_2	[273]
Dihydrocarvone	Indian dill (*Anethum sowa*)	HD; SFE/CO_2	[259]
Dillapiole	Indian dill (*Anethum sowa*)	HD; SFE/CO_2	[259]
Dillapiole	(*Diplotaenia cachrydifolia*)	SFE/CO_2 + ethanol	[274]
E-ocimenone	Marigold (*Tagetes minuta*)	SFE/CO_2	[223]
Epoxyocimene	Wormwood (*Artemisia absinthium L.*)	SFE/CO_2 + ethanol	[268]
Eugenol	Clove (*Eugenia caryophyllata*)	HD; SFE/CO_2	[275]
Eugenol	Clove (*Syzygium aromaticum*)	SFE/CO_2	[255, 276]
Eugenol	Clove basil (*Ocimum gratissimum*)	SFE/CO_2	[231]
Eugenol	Sri Lanka cinnamon (*Cinnamomum zeylanicum*)	SFE/CO_2	[266]
Eugenol	Purple nutsedge (*Cyperus rotundus*)	SFE/CO_2	[277]
Eugenyl acetate	Clove (*Syzygium aromaticum*)	SFE/CO_2	[255, 276]
E-β-ocimene	Southern cone marigold (*Tagetes minuta*)	SFE/CO_2	[223]
Farnesene	Annual herbs (*Nepeta persica*)	SFE/CO_2 + methanol; HD	[278]
Fenchon	French lavender (*Lavandula stoechas*)	HD; SubWE/H_2O	[209]
Fenchon	Lavender (*Lavandula angustifolia*)	SFE/CO_2	[241, 242]
Geranial	Moldavian dragonhead (*Dracocophalum moldavica L.*)	SFE/CO_2	[210]
Geraniol	Moldavian dragonhead (*Dracocophalum moldavica L.*)	SFE/CO_2	[210]
Geraniol	Scented geranium (*Pelargonium graveolens*)	HD; SFE/CO_2	[279]
Germacrone	Dragonhead (*Dracocophalum tanguticum*)	SFE/CO_2	[267]
Hentriacontano	Abajeru (*Chrysobalanus Icaco*)	SE; SFE/ethanol; CO_2	[280]
Hernandulcin	Honeyherb (*Lippia dulcis*)	HD; SFE/CO_2	[265]
Incensole	Boswellia (*Boswellia carterii*)	HD; SFE/CO_2	[281]
Incensole acetate	Boswellia (*Boswellia carterii*)	HD; SFE/CO_2	[281]
Isomenthyl acetate	Peppermint (*Metha piperita*)	SFE/CO_2	[223]
Khusimol	Vetiver (*Chrysopogon zizaniodes*)	SFE/CO_2 + ethanol	[282]
Limonene	(*Diplotaenia cachrydifolia*)	SFE/CO_2 + ethanol	[274, 283]
Limonene	Peruvian pepper (*Schinus molle*)	SFE/CO_2	[237]
Limonene	Oregano (*Origanum vulgare*)	SFE/CO_2	[218, 255, 256]
Limonene	Lemon (*Citrus* × *limon*)	SFE/CO_2	[219]
Limonene	Lemongrass (*Cymbopogon citrates*)	HD; SFE/CO_2	[271]
Limonene	Indian dill (*Anethum sowa*)	HD; SFE/CO_2	[259]
Limonene	Red clover (*Trifolium pratense*)	SFE/CO_2	[255, 284]
Limonene	Carqueja (*Baccharis genistelloides*)	SFE/CO_2	[272]
Linalool	Lavandin super (*Lavandula x intermedia Super*)	SFE/CO_2	[217]
Linalool	Levandin grosso (*Lavandula x intermedia Grosso*)	SFE/CO_2	[217]
Linalool	Lavandin abrial (*Lavandula x intermedia Abrial*)	SFE/CO_2	[217]

Linalool	French lavender (*Lavandula stoechas*)	SFE/CO$_2$	[209]
Linalool	Sweet bay (*Laurus nobilis L.*)	SFE/CO$_2$	[228, 229]
Linalool	(*Thymus lotocephalus G. Lopez & R. Morales*)	SFE/CO$_2$	[239]
Linalool	Carqueja (*Baccharis genistelloides*)	SFE/CO$_2$	[272]
Linalool	French lavender (*Lavandula stoechas*)	SFE/CO$_2$	[220]
Linalool	Lavender (*Lavandula angustifolia*)	SFE/CO$_2$	[241, 242]
Linalyl acetate	Lavender (*Lavandula angustifolia*)	SFE/CO$_2$	[241, 242]
Menthol	Peppermint (*Metha piperita*)	SFE/CO$_2$	[223]
Menthol	Peppermint (*Mentha piperita*)	SFE/CO$_2$	[189, 234, 285]
Menthone	Peppermint (*Mentha piperita*)	SFE/CO$_2$	[189, 234, 285]
Myrcene	Common juniper (*Juniperus communis L.*)	SFE/CO$_2$	[286]
Myrcene	Sage (*Salvia officinalis L.*)	HD; SFE/CO$_2$	[223–226]
Myrcene	Lemongrass (*Cymbopogon citrates*)	HD; SFE/CO$_2$	[271]
Myrcene	Surinam cherry (*Eugenia uniflora*)	SFE/CO$_2$	[51]
Myrtenol	French lavender (*Lavandula stoechas*)	HD; SubWE/H$_2$O	[209]
Myrtenol	Lavender (*Lavandula viridis*)	HD; SFE/CO$_2$	[243]
N.D.	Black pepper (*Piper nigrum*)	SFE/CO$_2$	[287, 288]
N.D.	Orange	SFE/CO$_2$; ethane	[221]
N.D.	Abise (*Pimpinella anisum*)	HD; SFE/CO$_2$	[289]
N.D.	Silver vine (*Actinida polygama*)	SFE/CO$_2$	[290]
Nanocosane	Abajeru (*Chrysobalanus icaco*)	SE; SFE/ethanol; CO$_2$	[280]
Nepetalactone	Annual herbs (*Nepeta persica*)	HD; SFE/CO$_2$ + methanol	[278]
Nerol	Moldavian dragonhead (*Dracocophalum moldavica L.*)	SFE/CO$_2$	[210]
Ocimene	Oregano (*Origanum vulgare*)	SFE/CO$_2$	[218, 255, 256]
Ocimene	Sage (*Salvia officinalis L.*)	HD; SFE/CO$_2$	[223–226]
Ocimene	Sweet bay (*Laurus nobilis L.*)	SFE/CO$_2$	[228, 229]
Ocimene	Carqueja (*Baccharis genistelloides*)	SFE/CO$_2$	[272]
Ocimene	Corn parsley (*Ridolfia segetum*)	SFE/CO$_2$	[291]
Octanol acetate	Boswellia (*Boswellia carterii*)	HD; SFE/CO$_2$	[281]
P-Cymene	Lippia origanoides	SFE/CO$_2$	[214]
P-Cymene	Khoa (*Satureja boliviana*)	SFE/CO$_2$ + ethanol	[262]
P-Cymene	Black cumin (*Nigella sativa L.*)	MAE; SFE/CO$_2$	[202]
Phyllocladene	Boswellia (*Boswellia carterii*)	HD; SFE/CO$_2$	[281]
Rosmanol	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO$_2$ + ethanol	[197, 212, 225, 244–250]
Rosmanol	Rosemary (*Rosmarinus officinalis L.*)	SubWE/H$_2$O	[184, 194]
Rosmanol	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO$_2$	[251, 253]
Rosmanol	Rosemary (*Rosmarinus officinalis L.*)	SFE; PLE/CO$_2$; water, ethanol	[254]
Rosmarinic acid	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO$_2$ + ethanol	[197, 212, 225, 244–250]

Rosmarinic acid	Rosemary (*Rosmarinus officinalis L.*)	SubWE/H_2O	[184, 194]
Rosmarinic acid	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2	[251, 253]
Rosmarinic acid	Rosemary (*Rosmarinus officinalis L.*)	SFE; PLE/CO_2; water, ethanol	[254]
Sabinene	Common juniper (*Juniperus communis L.*)	SFE	[286]
Sabinene	Sweet bay (*Laurus nobilis L.*)	SFE/CO_2	[228, 229]
Sabinene	Hyssop (*Hyssopus officinalis*).	SFE/CO_2 + methanol	[292]
Sabinene	Seseli (*Seseli bocconi*)	SFE/CO_2	[293]
Sabinene	Carrot (*Daucus carota*)	SFE/CO_2	[273]
Sabinene	Coral ginger (*Zingber corallinum*)	HD; SFE/CO_2	[294]
Salinene	Curry plant (*Helichrysum italicum*)	HD; SFE/CO_2	[261]
Santalol	Indian sandalwood (*Santal album*)	HD; SFE/CO_2	[281]
Scopoletin	Sweet wormwood (*Artemisia annua*)	SFE/CO_2 + ethanol	[236]
Sesquiterpenes	Sweetleaf (*Stevia rebaudiana*)	SFE/CO_2	[295]
Terpinen-4-ol	Coral ginger (*Zingber corallinum*)	HD; SFE/CO_2	[294]
Thymol	Oregano (*Origanum vulgare*)	SFE/CO_2	[218, 255, 256]
Thymol	Oregano (*Origanum vulgare*)	SubWE/H2O	[263]
Thymol	Summer savory (*Satureja hortensis*)	HD, SFE/CO_2 + methanol	[257]
Thymol	Khoa (*Satureja boliviana*)	SFE/CO_2 + ethanol	[262]
Thymol	Winter savory (*Satureja montana*)	SFE/CO_2, HD	[258]
Thymoquinone	Black cumin (*Nigella sativa L.*)	MAE; SFE/CO_2	[202]
Valerenic acid	Sage (*Salvia officinalis L.*)	HD; SFE/CO_2	[223–226]
Valerenic acid	Cotton lavender (*Santolina chamaecyparissus*)	HD; SFE/CO_2	[227]
Valerenic acid	Valerian (*Valeriana officinalis*)	SFE/CO_2	[296, 297]
Verbenone	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2 + ethanol	[164, 205, 225, 249]
Verbenone	Lavender (*Lavandula viridis*)	HD; SFE/CO_2	[243]
α-bergamotol	Indian sandalwood (*Santal album*)	HD; SFE/CO_2	[281]
α-cyperone	Surinam cherry (*Eugenia uniflora*)	SFE/CO_2	[51]
α-cyperone	Purple nutsedge (*Cyperus rotundus*)	SFE/CO_2	[277]
α-calacorene	(*Diplotaenia cachrydifolia*)	SFE/CO_2 + ethanol	[274]
α-humulene	Carrot (*Daucus carota*)	SFE/CO_2	[273]
α-pinnene	Borage (*Borago officinalis*)	HD; SFE/CO_2 + methanol	[240]
α-pinnene	Swamp mallet (*Eucalyptus spathulata*)	HD; SFE/CO_2	[222]
α-pinnene	Coolabah (*Eucalyptus microtheca*)	HD; SFE/CO_2	[222]
α-pinnene	Oregano (*Origanum vulgare*)	SFE/CO_2	[218, 255, 256]
α-pinnene	Rosemary (*Rosmarinus officinalis L.*)	SFE/CO_2 + ethanol	[225, 244]
α-pinnene	Cnidium monnieri	SFE/CO_2	[164]
α-pinnene	Common juniper (*Juniperus communis L.*)	SFE/CO_2	[286]
α-pinnene	Sage (*Salvia officinalis L.*)	HD; SFE/CO_2	[223–226]
α-pinnene	Sweet bay (*Laurus nobilis L.*)	SFE/CO_2	[228, 229]

α-pinnene	Red clover (*Trifolium pratense*)	SFE/CO$_2$	[255, 284]
α-pinnene	Carrot (*Daucus carota*)	SFE/CO$_2$	[273]
α-pinnene	Sweet gale (*Myrica gale L*)	SFE/CO$_2$	[233]
α-selinene	Guava (*Psidium guajava*)	SFE/CO$_2$ + ethanol, + isopropanol	[298]
α-thujene	Black cumin (*Nigella sativa L.*)	MAE; SFE/CO$_2$	[202]
α-thujone	Sage (*Salvia officinalis*)	SFE/CO$_2$	[223]
α-ylangene	Five-flavor berry (*Schisandra chinensis*)	SFE; HD; SFE/CO$_2$	[215]
β-cadinene	Saigon cinnamon (*Cinnamomum loureiroi*)	SFE/CO$_2$	[230]
β-cadinene	Marigold (*Calendula officinalis*)	SFE/CO$_2$	[238]
β-caryophyllene	Carrot (*Daucus carota*)	SFE/CO$_2$	[273]
β-caryophyllene	Spearmint (*Mentha spicata*)	SE; SFE/hexane, DCM, ethanol; CO$_2$	[260]
β-caryophyllene	Guava (*Psidium guajava*)	SFE/CO$_2$ + ethanol, + isopropanol	[298]
β-selinene	Guava (*Psidium guajava*)	SFE/CO$_2$ + ethanol, + isopropanol	[298]
β-selinene	Clove basil (*Ocimum gratissimum*)	SFE/CO$_2$	[231]
γ-terpinene	Oregano (*Origanum vulgare*)	HD; SFE/CO$_2$	[218]
γ-terpinene	Oregano (*Origanum vulgare*)	SubWE/H$_2$O	[263]
γ-terpinene	Cotton lavender (*Santolina chamaecyparissus*)	HD; SFE/CO$_2$	[227]
γ-terpinene	Winter savory (*Satureja montana*)	HD; SFE/CO$_2$	[258]
γ-terpinene	Summer Savory (*Satureja hortensis*)	HD, SFE/CO$_2$+ methanol	[257]
γ-terpinene	Black cumin (*Bunium persicum*)	HD; SFE/CO$_2$	[299]
δ-cadinene	Madeira mahogany (*Persea indica*)	SFE/CO$_2$ + ethanol	[264]
δ-cadinene	Honeyherb (*Lippia dulcis*)	HD; SFE/CO$_2$	[265]
τ-cadinol	Dragonhead (*Dracocophalum tanguticum*)	SFE/CO$_2$	[267]

2.4. *Extraction of lipids and fatty acids*

In Table 5, examples of SFE application for the separation of lipids and fatty acids from plant material are listed.

There are several fatty acids or their derivates that are interesting for investigation because of their functional potential. Precursors of conjugated linoleic acid, linoleic acid, and polyunsaturated fatty acid (PUFA) are intensively produced in plants. Plants primarily produce fatty acids in order to become triglycerides, which serve as stored energy (oils) as well as components of the cell membrane, *i.e.*, glycerophospholipids and glyceroglycolipids, which have roles similar to phospholipids in humans. Major fatty acids produced in plants are palmitic acid (16:0), oleic acid (18:1 ω-9), linoleic acid (18:2 ω-6), and α-linolenic acid (18:3 ω-3) [12]. Among fatty acids are essential fatty acids (ω-6 and ω-3), which are very important for human health as antioxidants and because they dissolve vitamins that do not dissolve in water. Those fatty acids in most cases have to be provided to the body through nutrition because the body cannot produce them.

Besides fatty acids, there are two other types of lipids in food products: first are structured lipids and the others are diglycerides. Structured lipids are triglycerides that have undergone hydrolysis and esterification, which resulted in triglycerides with a new combination of fatty acids. Diglycerides have been used as emulsifying agents in manufactured food products [12].

Phospholipids compose the main part of the cell membranes and are composed of diglycerides and a phosphate group, on which another organic compound is attached, such as choline [300], serine, and ethanolamine.

Lipids are mainly isolated with organic solvents, and most of them pollute the final product. To avoid further processing in order to separate organic solvents, supercritical fluid extraction could be used.

Separation is relatively easy to achieve because of the nonpolar nature of lipids. Several works have been published where the performance of conventional solvent extraction (SE) was compared to that of SFE. Even though SE generally gives higher yields than SFE, the use of organic solvents does not provide the desired selectivity; hence, other compounds besides fatty acids are extracted [301]. In order to have a faster and more selective extraction, SFE is the alternative to use.

The most common use of SFE is in the removal of oils (edible) from plant seeds and kernels. Oil-and-fat-processing industries encounter oil acidification due to the hydrolysis of the fatty acid chains and lipid oxidation, which is caused by an enzyme, heat, or moisture. In order to avoid hydrolysis, chemical and physical deacidification methods have been used in the industry. Some processes of deacidification could be performed by SFE [179, 302, 303]. When oils are removed from a solid material, extraction is usually semicontinuous. Alternatively, if bare oils are extracted and fractioned, the process is performed continuously in countercurrent mode. Such a process was performed by Al–Darmaki *et al.* [304] when they extracted squalene from a palm oil fatty–acid distillate. In the first set of experiments [304], pressure was changed and temperature remained constant. Afterward, a set of experiments followed, where temperature was changed and pressure was kept constant. At the highest pressure at constant temperature, recovery of squalene was the highest, while it was the lowest at the highest temperature at constant pressure [304].

Materials that contain a lot of oil extractables can be pretreated with SC–CO_2 [305]. Fatty acids and triglycerides dissolve CO_2 well under pressure. Meyer *et al.* [305] performed a CO_2 pretreatment of sunflower and St. John's wort. The material was exposed to compressed and heated CO_2 in a magnetic suspension balance (MSB). Absorbed CO_2 caused expansion of extractables in cells; during the fast decompression and desorption, the material was pulverized, and as consequence the material's contact area became bigger, and thus faster separation was achievable. In the case when a material does not contain many lipid extractables, this phenomenon did not occure [305].

SFE can be applied for the isolation of tocopherols and phytosterols from residues of vegetable-oil processing, which are deodorized – *i.e.* offensive odor is removed. Such residues could be obtained after the removal of oil from sunflower [1], soybean [306–309], olive [310], corn, canola, *etc.*

The use of SC–CO_2 for the extraction of essential fatty acids from plant materials has become very attractive. Such case is the isolation of ω–3 and other PUFA from plant materials rich in oils, such as flaxseed [311] or canola seed [312]. Other examples could be seen in Table 5, where applications of SFE for the isolation of fatty acids and other lipids from plant material are listed.

Not only SC–CO_2 could be used as a solvent for the extraction of oils and fats, but subcritical propane and supercritical sulphur hexafluoride (SF_6) could also be used as solvents in extraction processes. Ilic *et al.* [313] measured the phase behavior of sunflower and soybean oils in propane and SF_6.

Table 5

Application of SFE for the separation of lipid compounds from natural matter

Compound(s)	Plant material (Biological name)	Methods/solvents	Reference(s)
Campesterol	Carrot (*Daucus carota*)	SFE/CO_2	[273]
Campesterol	Red globe grape	SFE/CO_2 + ethanol	[314]
Campesterol	Victoria globe grape	SFE/CO_2 + ethanol	[314]
Cycloartenol	Carrot (*Daucus carota*)	SFE/CO_2	[273]
Ergosterol	Shitake (*Lentinula edodes*)	SE; SFE/*n*-hexane, DCM; CO_2	[315]
Erucic acid	Canola (*Brassica napus*)	SFE/CO_2	[307, 316]
Eicosenoic acid	Canola (*Brassica napus*)	SFE/CO_2	[307, 316]
Fatty acid (unsaturated)	Cuphea (*Cuphea lanceolata*)	SFE/CO_2	[317]
Fatty acid (unsaturated)	Passion fruit (*Passiflora*)	SFE/CO_2	[318]
Fatty acid (unsaturated)	Seabuckthorn (*Hippophae rhamnoides*)	SFE/CO_2	[196, 319]
Fatty acid (unsaturated)	Sikkim microula (*Microula sikkimenisis*)	SFE/CO_2	[320]
Fatty acid (unsaturated)	White pitaya (*Hylocereus undatus*)	SE; MAE; AEE; SFE/CO_2	[321]
Fatty acid (unsaturated)	Rice (*Oryza sativa*)	SFE/CO_2 + ethanol	[137]
Fatty acid (unsaturated)	African oil palm (*Elaais guinensis*)	SFE/CO_2	[322]
Hexadecanal	Lotus (*Nelumbo nucifera*)	SFE/CO_2	[64]
Linoleic acid	Broccoli (*Brassica oleracea*)	SFE/CO_2	[323]
Linoleic acid	Canola (*Brassica napus*)	SFE; SE/CO_2; *n*-hexane	[171, 324]
Linoleic acid	Canola (*Brassica napus*)	SFE/CO_2	[307, 316]
Linoleic acid	Canola (*Brassica napus*)	SFE/CO_2; propane	[325]
Linoleic acid	Canola (*Brassica napus*)	SFE/CO_2 + ethanol	[326]
Linoleic acid	Cantaloupe (*Cucumis melo var. cantalupensis*)	SFE/CO_2	[327]
Linoleic acid	Cork oak (*Quercus suber*)	SE; SFE/*n*-hexane; CO_2	[328]
Linoleic acid	Gardenia (*Gardenia jasminoides*)	SFE/CO_2	[329]
Linoleic acid	Holm oak (*Quercus rotundifolia*)	SFE/CO_2	[330]
Linoleic acid	Holm oak (*Quercus rotundifolia*)	SE; SFE/*n*-hexane; CO_2	[328]
Linoleic acid	Borage (*Borago officinalis*)	SFE/CO_2 + methanol	[240]
Linoleic acid	Kenaf (*Hibiscus cannabinus*)	SFE/CO_2	[331, 332]
Linoleic acid	Agaricus brasiliensis	SFE/CO_2 + ethanol	[333]
Linoleic acid	Olive (*Olea europaea*)	SFE/CO_2	[112]
Linoleic acid	Pea (*Pisum sativum*)	SFE/CO_2 + ethanol	[326]
Linoleic acid	Rosehip (*Rosa canina*)	SFE/CO_2	[78]
Linoleic acid	Siam pumpkin (*Cucurbita ficifolia*)	SFE; SE/CO_2	[334]
Linoleic acid	Soybean (*Glycine max*)	SFE/CO_2	[307, 335]
Linoleic acid	Soybean (*Glycine max*)	SFE/CO_2 + ethanol	[326]
Linoleic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]

Linoleic acid	Peach kernel (*Prunus persica*)	SE; HD; SFE/ethanol, water, DCM; CO_2, + ethanol	[337]
Linoleic acid	Shitake (*Lentinula edodes*)	SE; SFE/*n*-hexane, DCM; CO_2	[315]
Linoleic acid	Peach kernel (*Prunus persica*)	SFE/CO_2 + ethanol	[338]
Linoleic acid	Prickly pear (*Opuntia dillenii*)	SFE/CO_2	[318]
Linoleic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2; propane	[339]
Linoleic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2 + ethanol	[326]
Linoleic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2	[1]
Linoleic acid	Viper’s bugloss (*Echium amoenum*)	SFE/CO_2	[340]
Linoleic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
Linolenic acid	Broccoli (*Brassica oleracea*)	SFE/CO_2	[323]
Linolenic acid	Canola (*Brassica napus*)	SFE; SE/CO_2; *n*-hexane	[171, 324]
Linolenic acid	Canola (*Brassica napus*)	SFE/CO_2	[307, 316]
Linolenic acid	Canola (*Brassica napus*)	SFE/CO_2; propane	[325]
Linolenic acid	Canola (*Brassica napus*)	SFE/CO_2 + ethanol	[326]
Linolenic acid	Lotus (*Nelumbo nucifera Gaertn*)	SFE/CO_2	[54]
Linolenic acid	Olive (*olea europaea*)	SFE/CO_2	[112]
Linolenic acid	*Pea* (*Pisum sativum*)	SFE/CO_2 + ethanol	[326]
Linolenic acid	Prickly pear (*Opuntia dillenii*)	SFE/CO_2	[318]
Linolenic acid	Rosehip (*Rosa canina*)	SFE/CO_2	[78]
Linolenic acid	Soybean (*Glycine max*)	SFE/CO_2	[307, 335]
Linolenic acid	Soybean (*Glycine max*)	SFE/CO_2 + ethanol	[326]
Linolenic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
Linolenic acid	Carrot (*Daucus carota*)	SFE/CO_2	[273]
Linolenic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
N.D.	Almond (*Prunus dulcis*)	SFE/CO_2	[341]
N.D.	Apricot (*Prunus armeniaca*)	SFE/CO_2 + ethanol	[342]
N.D.	Guava (*Psidium guajava*)	SFE/CO_2 + ethanol	[343]
N.D.	Okra (*Hibiscus esculentus*)	SFE/CO_2	[344]
N.D.	Pomegranate (*Punica granatum*)	SFE/CO_2	[206]
N.D.	Sesame (*Sesamun indicum*)	SFE/propane, CO_2	[345]
N.D.	Sorghum (*Sorghum*)	SE; SFE/*n*-hexane; CO_2	[346]
N.D.	Yelleowhorn (*Xanthoceras sorbifolia*)	SFE/CO_2	[347]
Oleic acid	Canola (*Brassica napus*)	SFE; SE/CO_2; *n*-hexane	[171, 324]
Oleic acid	Canola (*Brassica napus*)	SFE/CO_2	[307, 316]
Oleic acid	Canola (*Brassica napus*)	SFE/CO_2; propane	[325]
Oleic acid	Canola (*Brassica napus*)	SFE/CO_2 + ethanol	[326]
Oleic acid	Cantaloupe (*Cucumis melo var. cantalupensis*)	SFE/CO_2	[327]
Oleic acid	Cork oak (*Quercus suber*)	SE; SFE/*n*-hexane; CO_2	[328]

Oleic acid	Holm oak (*Quercus rotundifolia*)	SE; SFE/*n*-hexane; CO_2	[328]
Oleic acid	Kenaf (*Hibiscus cannabinus*)	SFE/CO_2	[331, 332]
Oleic acid	*Pea* (*Pisum sativum*)	SFE/CO_2 + ethanol	[326]
Oleic acid	Peach kernel (*Prunus persica*)	SE; HD; SFE/ethanol, water, DCM; CO_2, + ethanol	[337]
Oleic acid	Peach kernel (*Prunus persica*)	SFE/CO_2 + ethanol	[338]
Oleic acid	Siam pumpkin (*Cucurbita ficifolia*)	SFE; SE/CO_2	[334]
Oleic acid	Soybean (*Glycine max*)	SFE/CO_2	[307, 335]
Oleic acid	Soybean (*Glycine max*)	SFE/CO_2 + ethanol	[326]
Oleic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2; propane	[339]
Oleic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2 + ethanol	[326]
Oleic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2	[1]
Oleic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
Palmitic acid	Cantaloupe (*Cucumis melo var. cantalupensis*)	SFE/CO_2	[327]
Palmitic acid	Borage (*Borago officinalis*)	SFE/CO_2 + methanol	[240]
Palmitic acid	Agaricus brasiliensis	SFE/CO_2 + ethanol	[333]
Palmitic acid	Gardenia (*Gardenia jasminoides*)	SFE/CO_2	[329]
Palmitic acid	Holm oak (*Quercus rotundifolia*)	SFE/CO_2	[330]
Palmitic acid	Prickly pear (*Opuntia dillenii*)	SFE/CO_2	[348]
Palmitic acid	Rosehip (*Rosa canina*)	SFE/CO_2	[78]
Palmitic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2; propane	[339]
Palmitic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2 + ethanol	[326]
Palmitic acid	Sunflower (*Helianthus annuus*)	SFE/CO_2	[1]
Palmitic acid	Siam pumpkin (*Cucurbita ficifolia*)	SFE; SFE/CO_2	[334]
Palmitic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
Palmitic acid	Broccoli (*Brassica oleracea*)	SFE/CO_2	[323]
Palmitic acid	Shitake (*Lentinula edodes*)	SE; SFE/*n*-hexane, DCM; CO_2	[315]
Phytosterol	Polygala (*Polygala cyparissias*)	SE; SFE/*n*-hexane, DCM; CO_2	[349]
Phytosterol	African oil palm (*Elaais guinensis*)	SFE/CO_2	[322]
Phytosterol	Pumkin (*Cucurbita pepo*)	SFE/CO_2	[350]
Phytosterol esters	Soybean (*Glycine max*)	SFE/CO_2	[306, 309, 335]
PUFA	Canola (*Brassica napus*)	SFE/CO_2	[312]
PUFA	Common walnut (*Juglans regia*)	SFE/CO_2	[351]
PUFA	Grape	SFE/CO_2; propane	[352]
PUFA	Microalgae (*Schizochytrium limacinum*)	SFE/CO_2 + ethanol	[353]
PUFA	Viper's bugloss (*Echium amoenum*)	SFE/CO_2	[340]
PUFA	Hemp (*Cannabis sativa L.*)	SE; SFE/CO_2	[354, 355]
Spilanthol	Jambu (*Spilanthes acmella*)	SFE/CO_2	[356]

Squalene	African oil palm (*Elaais guinensis*)	SFE/CO_2	[322]
Squalene	Canola (*Brassica napus*)	SFE/CO_2	[307]
Squalene	Olive (*olea europaea*)	SFE/CO_2	[310]
Squalene	Lotus (*Nelumbo nucifera Gaertn*)	SFE/CO_2	[54]
Squalene	Purple amaranthus (*Amaranthus paniculatus*)	SFE/CO_2	[357]
Stearic acid	Rosehip (*Rosa canina*)	SFE/CO_2	[78]
Stearic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
Stearic acid	Tea (*Camellia sinensis*)	SFE/CO_2 + ethanol; + methanol	[336]
Stearic acid	Cantaloupe (*Cucumis melo var. cantalupensis*)	SFE/CO_2	[327]
Sterols	Canola (*Brassica napus*)	SFE/CO_2	[307]
Sterols	Rice (*Oryza sativa*)	SFE/CO_2	[179–181, 358]
Stigmasterol	Red globe grape	SFE/CO_2 + ethanol	[314]
Stigmasterol	Victoria globe grape	SFE/CO_2 + ethanol	[314]
Stigmasterol	Carrot (*Daucus carota*)	SFE/CO_2	[273]
Stigmasterol	Lotus (*Nelumbo nucifera Gaertn*)	SFE/CO_2	[54]
Tocopherols	Seabuckthorn (*Hippophae rhamnoids*)	SFE/CO_2	[28]
Tocopherols	Common walnut (*Juglans regia*)	SFE/CO_2	[351]
Tocopherols	Kenaf (*Hibiscus cannabinus*)	SFE/CO_2	[331, 332]
Tocopherols	Olive (*olea europaea*)	SFE/CO_2	[112]
Tocopherols	Canola (*Brassica napus*)	SFE/CO_2	[307]
Tocopherols	Gardenia (*Gardenia jasminoides*)	SFE/CO_2	[329]
Tocopherols	Lotus (*Nelumbo nucifera*)	SFE/CO_2	[64]
Tocopherols	Rice (*Oryza sativa.*)	SFE/CO_2	[179–181, 358]
Tocopherols	Soybean (*Glycine max*)	SFE/CO_2	[307, 335]
Tocopherols	Sunflower (*Helianthus annuus*)	SFE/CO_2	[1]
Tocopherols	Red globe grape	SFE/CO_2 + ethanol	[314]
Tocopherols	Victoria globe grape	SFE/CO_2 + ethanol	[314]
Tocopherols	Seabuckthorn (*Hippophae rhamnoides L.*)	SFE/CO_2 + methanol	[29]
Tocotriols	Rice (*Oryza sativa*)	SFE/CO_2	[179–181, 358]
Triglycerides	Agarwood (*Aquilaria crassna*)	SFE/CO_2	[359]
Triglycerides	Cork oak (*Quercus suber*)	SE; SFE/*n*–hexane; CO_2	[328]
Triglycerides	Crude palm oil	SFE/CO_2	[360]
Triglycerides	Holm oak (*Quercus rotundifolia*)	SE; SFE/*n*–hexane; CO_2	[328]
Triglycerides	Barbados nut (*Jatropha curcas*)	SFE/CO_2	[361]
Triglycerides	Jelly fig (*Ficus awkeotsang M.*)	SFE/CO_2	[362]
Triterpenes	Polygala (*Polygala cyparissias*)	SE; SFE/*n*–hexane, DCM; CO_2	[349]
Triterpenes	Tasmanian blue gum (*Eucalyptus globulus*)	SFE/CO_2	[363]

Triterpenoid	Tasmanian blue gum (*Eucalyptus globulus*)	SFE/CO$_2$ + ethanol	[364]
Vitamin E	Grape*	PLE	[365]
Vitamin E	Soybean (*Glycine max*)	SFE/CO$_2$	[308, 335]
α-linolenic acid	Chia (*Salvia hispanica*)	SFE/CO$_2$	[366, 367]
α-linolenic acid	Jelly fig (*Ficus awkeotsang M.*)	SFE/CO$_2$	[362]
α-linolenic acid	Flax (*Linum usitatissimum*)	SFE/CO$_2$	[311]
α-tocopherol	African oil palm (*Elaais guinensis*)	SFE/CO$_2$	[322]
α-tocopherol	Almond (*Prunus dulcis*)	SFE/CO$_2$	[368]
α-tocopherol	Sunflower (*Helianthus annuus*)	SFE/CO$_2$; propane	[339]
α-tocopherol	Red pepper (*Capsicum annum L.*)	SFE/CO$_2$	[62]
β-sitosterol	Kenaf (*Hibiscus cannabinus*)	SFE/CO$_2$	[331, 332]
β-sitosterol	Laquat (*Eribotrya japonica*)	SFE/CO$_2$	[369]
β-sitosterol	Black alder (*Alnus glutinosa*)	SFE/CO$_2$ + ethanol	[370]
β-sitosterol	Lotus (*Nelumbo nucifera Gaertn*)	SFE/CO$_2$	[54]
β-sitosterol	Carrot (*Daucus carota*)	SFE/CO$_2$	[273]
β-sitosterol	Red globe grape	SFE/CO$_2$ + ethanol	[314]
β-sitosterol	Victoria globe grape	SFE/CO$_2$ + ethanol	[314]
γ-linolenic acid	Viper's bugloss (*Echium amoenum*)	SFE/CO$_2$	[340]
γ-linolenic acid	Borage (*Borago officinalis*)	SFE/CO$_2$ + methanol	[240]
γ-linolenic acid	Chia (*Salvia hispanica*)	SFE/CO$_2$	[366, 367]
γ-linolenic acid	Flax (*Linum usitatissimum*)	SFE/CO$_2$	[311]
γ-linolenic acid	Rice (*Oryza sativa*)	SFE/CO$_2$ + ethanol	[137]
γ-linolenic acid	Cyanobacteria (*Arthrospira maxima*)	SFE/CO$_2$	[27]
γ-oryzanol	Rice (*Oryza sativa*)	SFE/CO$_2$	[179–181, 358]

2.5. *Alkaloids and other bioactive phytochemicals*

In previous sections, nutraceuticals were discussed. Nutraceuticals improve food properties (flavor and taste – essential oils, color – carotenoids), and others act in preventing and treating diseases (antioxidants – carotenoids, phenolic compounds, essential oils *etc*.). Besides nutraceuticals, some other phytochemicals could also be extracted from plants. Even though a lot of nutraceuticals extracted with SFE act as antioxidants, they also provide other health-beneficial properties – anti-inflammatory, antimicrobial, cancer prevention, *etc.*

Garcia–Risco *et al.* [371] studied SFE of thymol, which poses antioxidant, analgesic (relieve pain), and anti-inflammatory properties. The extract was further fractionated with SFC in order to achieve a better separation.

As mentioned before, SFE could serve for a sample preparation. Pretreated samples are suitable for further preparation or analysis [372, 373].

Alkaloids can be separated with SFE as well. The most common application of SFE for the isolation of alkaloids is the removal of caffeine from coffee beans and tea leaves [374–379]. The process is called decaffeination and is one of the first SFE processes that were developed at an industrial scale. The product is decaffeinated coffee and tea [378]. There are also some other alkaloids with health–beneficial properties, such as boldine [208], sinomenine [380], indole

[381, 382], purine [383], tetrahydropalmatine [384], nicotine [385], isoquinolines [386], *etc.* [387] investigated safranol extraction with supercritical CO_2. Safranol exhibits antioxidant properties, acts against cancer cells, and serves as an antidepressant.

SFE of bioactive compounds from plant materials was also used for the recovery of evodiamine and rutacarpine from the *Evodia rutacarpa* [388] fruit, hyperforin and adhyperforin from *Hypericum perforatum L.* [389], and solanosol from tobacco leaves [390].

3. CONCLUSIONS

In the present work, recent investigations of SFE and its applications are presented. Most of the researches were performed on a laboratory scale. These researches give insight into important properties of SFE processes and provide the data required for a potential scale up to a preparative or even industrial scale. This paper not only shows properties of SFE processes but also describes the possibilities of SFE application for the separation of nutraceuticals and other bioactive compounds at an industrial scale. SFE is appropriate for the separation of high value-added products because of its high investment cost. But obtained extracts are free from organic solvents, contributing to sustainability. The future trend of SFE shall go in the direction of not only the extraction of certain compounds but also the purification and concentration of compounds. Therefore, other separation processes could be coupled with SFE. Among them is SFC, where the purification of several compounds is possible. To implement SFE in the industry, optimization studies are required in order to reduce operating costs. Hence, proper operating parameters should be selected using different optimization methods, such as response surface methodology. To reduce operating costs even more, the recirculation of solvent should be applied. Vessels where the extract would be collected should be under pressure to reduce the amount of energy required for renewed pressurization. Further investigations of the SFE process should include the application of SFE for industrial purposes, meaning coupling with other separation processes such as SFC and micronization processes, in order to purify and concentrate and to obtain smaller particles, which could be used effectively in the food or pharmaceutical industry.

Even though many investigations were done in this field, many possibilities of SFE application from plant material could be discovered in the future.

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ABBREVIATIONS

SFE	Supercritical fluid extraction
RSM	Response surface methodology
DMP	2,2-dimethoxypropane
HPLC	High-performance liquid chromatography
PHWE	Pressurized hot-water extraction
ASE	Accelerated solvent extraction
CC-SFE	Countercurrent supercritical fluid extraction
RP-LC	Reversed-phase liquid extraction
C8	Silica columns with 8-carbon chains on the surface
C18	Silica columns with 18-carbon chains on the surface
TLC	Thin layer chromatography
DAD	Diode array detector
MS	Mass spectroscopy
ESI	Electrospray ionization detector
ELSD	Evaporative light-scattering detector
GC-MS	Gas chromatography coupled with mass spectroscopy
SubWE	Subcritical water extraction
SE	Solvent extraction or Soxhlet extraction

CE	Capillary electrophoresis
HD	Hydrodistillation
SD	Steam distillation
PUFA	Polyunsaturated fatty acids
MSB	Magnetic suspension balance
SF_6	Sulphur hexafluoride
R134a	Freon 1,1,1,2-tetrafluoroethane
DEP	2,2-diethoxypropane
TEOF	Triethyl orthoformate
MAE	Microwave-assisted extraction
AEE	Aqueous enzymatic extraction

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