Chemical constituents of the pine extracts and their activities: A review

Ayari[a] M., Romdhane[a] M.,

[a] *Laboratoire de recherche Energie, Eau, Environnement et Procédés (LEEEP), Ecole Nationale d'Ingénieurs de Gabès, Université de Gabès, Route de Médenine, 6029 Gabès, Tunisie.*

Abstract:

Pines are very important economically. There is a diverse set of studies related to chemical composition of Pine extracts and their activities. This review discusses the different compounds of 4 species of pine; *Pinus halepensis, Pinus pinaster, Pinus pinea* and *Pinus brutia* which include Terpenoids, Phenolic, Resin and Fatty acids. Many activities are attributed to them, such as herbicidal, antifungal, antiradical, antimicrobial and many others. Chemical composition of some pine extracts and their activities are discussed herein, and structure–activity relationships are presented when appropriate

Keywords: *Pinus halepensis, Pinus pinea, Pinus pinaster*, *Pinus brutia*, Extraction, Bioactive compounds

[*] *Corresponding author e-mail address: mehrez.romdhane1965@gmail.com*

Introduction:

The genus Pinus belongs to the Pinaceae family and comprises about 250 species (Yang et *al*., 2010), which is found in most of the Northern Hemisphere (Kurose et *al*., 2006).This plant (Figure 1) is used in ethnomedical practice throughout the world. For example, the Indians use a boiled extract of the inner bark from *P. strobes* (White pine) as an astringent for diarrhea or in cough remedies. In 19th century North America, *P. sylvestris* (Scots pine) was employed as a diuretic and to induce perspiration and thus help break a fever. *P. brutia* is used in folk medicine in Turkey, and the antimicrobial activity of tar obtained from the roots and stems of *P. brutia* against Staphylococcus aureus, Streptococcus pyrogenes, Escherichia coli, and Candida albicans was reported (Sakagami et *al*., 1991; Unten et *al*., 1989). Previous studies on Pinus species reported the diterpenoids (Cheung et *al*, 1993; Fang et *al*., 1991), triterpenoids (Fang et *al*., 1991), flavonoids (Fang et *al*., 1988), and lignans (Yang et *al*., 2010).In this context, various parts of some pine species, such as bark, needle, cone and resin have been used as a folk medicine for rheumatism or as anti-inflammatory, antioxidant and antiseptic. In the last decades, modern science has shown increasing interest in folk medicine for a better understanding of the chemical composition of natural products and in finding alternative usages (Ahmed et *al*., 1969; Packer *al*., 1999; Kähkönen et *al*., 1999; Devaraja *al*., 2002; Villagomez et *al*., 2005; Willför *al*., 2009).

Also, it has been reported that Pinus plants have fatigue-relieving, anti-aging and anti-inflammatory activities (Watanabe et *al*., 1995) and that there are nutritional compounds in Pinus seeds (Isidorov et *al*., 2003).

To determine the chemical composition and the different activities, several extracts were studied. In particular, the essential oil extracted from its various parts by hydrodistillation. It possesses biological effects such as antifungal, (Amri et *al*.,2013, Abi-Ayad et *al*., 2011), antibacterial (Fekih et *al*., 2014, Hmamouchi et *al*.,2001, Mimoune et *al*., 2013, Sadou et *al*.,2015), herbicidal (Amri et *al*., 2013, Hanana et *al*.,2014), antioxidant (Tumen et *al*., 2017, Ustun et *al*., 2012 , Ulukanl et *al*., 2014, Djerrad et *al*., 2015) and anti-inflammatory activities (Tumen et *al*., 2017, Süntar et *al*., 2012).Pine oils are widely used as fragrances in cosmetics, as flavoring additives for food and beverages, as scenting agents in a variety of household products, and intermediates in the synthesis of other perfume chemicals (Ustun et *al*., 2006, Sezik et *al*., 2010).

Other extractions were aplicated on various parts of some pine species with a different solvent. In this context, (Tümen et *al*., 2017 and Cheikh-Rouhou et *al*., 2007) were studied the extracts of *pinus pinaster* (cone, needles and wood) and *pinus halepensis* (seeds) with the hexane respectively. Methanolic extracts of *pinus halepensis* (cones and seeds) and *pinus brutia* (barks, resins and cones) were studied (Dhibi et *al*., 2012 and Dıgrak et *al*., 1999) to determine their antiradical and antimicrobial activities respectively.

In this review, we summarized the most recent papers describing chemical constituents and some activities of various parts of four pine species (*pinus halepensis*, *pinus brutia*, *pinus pinea* and *pinus pinaster*) extracts.

Chemical constituents of pine extracts:

Pine extracts have different pharmacological activities and isolated compounds have been extensively studied. In this part we have summarized the studies that have been done on the composition of essential oils and extracts of *pinus halepensis, pinea, pinaster* and *brutia*.

First of all it should be mentioned that *pinus halepensis* is the largest and the most important representative of the genus Pinus that comprises 250 species, widely distributed all over the Mediterranean Basin (Dob et *al*., 2005). In Tunisia, this pinus occupies 200,000 ha of natural forests and more than 161,221 ha of artificial forests. Then we found *pinus pinea* with 20 922 ha in the artificial forests, *pinus pinaster* with 5153 ha in the natural forests and 750 ha of pine mix in the artificial forests (ORGANISATION DES NATIONS UNIES POUR L'ALIMENTATION ET L'AGRICULTURE, 2012).

The main extraction method used by these studies was hydrodistillation, followed by soxhlet. Hexane and acetone methanol were the most used by the papers, followed by methanol. The pharmacological activities attributed to these extracts are summarized in (Table 1).

Chemical constituents of pine essential oils

Pine oils are widely used as fragrances in cosmetics, flavoring additives for food and beverages, scenting agents in a variety of household products, and intermediates in the synthesis of perfume chemicals. They are also used for medicinal purposes in aromatherapy as carminative, rubefacient, emmenagogue, and abortifacient agents. Pines are among the most important forest trees in the Mediterranean region. There are numerous studies dealing with the essential oils of conifer species and especially those of pinus (Yang et *al*., 2010).

Among these studies are those of Amri et *al*., 2013 who have studied the extraction of essential oils from different parts of pinus.

The chemical composition of pine essential oils varied between organs. This was proved by (Amri et *al*., 2013 and Fekih et *al*., 2014) for the *P.halepensis* (cone, needles, stems, twigs and buds). Fifty eight compounds were identified in the oil from needles with (Z)-caryophyllene and b-pinene as main constituents; 57 in cone oil with a-pinene and (Z)-caryophyllene as main components; and only 27 in stem oil with apinene and b-myrcene as main components (Amri et *al*., 2013). For the *P.pinaster*, (Tümen et *al*., 2017) studied the difference in chemical composition of the essential oils of three parts (cone, needles and wood); in the wood, β-pinene, limonene, α-terpineol, and junipene were present in their highest amounts. By comparison, α -pinene, β-pinene, δ-3-carene, limonene, junipene, and trans-caryophyllene were found in cones. Major compounds in needles were identified as α -pinene, β-pinene, β-myrcene, transcaryophyllene, α-amorphene, rimuene, cupresseneabietatriene, and abietadiene. This differs from the results obtained by (Macchioni et *al*., 2003), who found the major compound of the *P.pinea* essential oils of all the three plant parts (needles, branches and cones) is limonene, in *P. halepensis* α-pinene and myrcene, in *P. pinaster* α-pinene and β-pinene.

In the other hand, the chemical composition of essential oils depends, also, to the pine species and this is provided inseveral studies. Twenty three compounds were identified in the essential oil from *P.halepnsis* needles with α -pinene and β-myrcene as main constituents; 21 in *P.brutia* oil with, β-pinene and α -pineneas main components; 19 in *P.pinaster* oil with,β- caryophyllene and α -pinene as main components; and only 12 in *P.pinea* oil with α –pinene as main component (Lahlou et *al*., 2008) and the same major compounds were found by (Hmamouchi et *al*., 2001).

Chemical constituents of pine extracts

With the extraction of essential oils from various parts of some pine species, we find extraction by different solvents. In the following section, the compounds of four pine extracts (*P.halepensis, P.pinea, P.brutia* and *P.pinaster*) are listed.

Essential fatty acids and their long-chain derivatives are important structural elements of cell membranes and are essential for the formation of new tissues (McKevith, 2005). According to Nasri et *al*., (2005), qualitatively, *P. pinea* and *P. halepensis* fatty acid seeds composition

is identical and therefore characteristic of the genus Pinus. For *P. halepensis* linoleic acid is the major fatty acid (56.06% of total fatty acids) followed by oleic (24.03%) and palmitic (5.23%) acids. For *P. pinea*, the same fatty acids are found with the proportions 47.28%, 36.56%, and 6.67%, respectively. Extracted fatty acids from both species are mainly unsaturated, respectively, 89.87% and 88.01%. *Pinus halepensis* cis-5 olefinic acids are more abundant (7.84% compared to 2.24%) (Nasri et *al*., 2005) .

Sixteen fatty acids were detected in the hexane seed extracts of *P.halepensis* by gas–liquid chromatography (Cheikh-Rouhou et *al*., 2006). Fatty acid composition of *P.halepensis* seed oil (Cheikh-Rouhou et *al*., 2006) showed that linoleic and oleic acids account for more than 76% of the total fatty acids which is in agreement with the results found by (Dhibi et *al*., 2012). *Pinus halepensis* seed oil is rich in unsaturated fatty acids, saturated fatty acids accounted for 16.8% of total fatty acids. Among them, the main saturated acids were palmitic, stearic and behenic, with minute amounts of arachidic, lignoceric, myristic and margaric (Cheikh-Rouhouet *al*., 2006).Then, this seeds oil was analysed for sterol composition by (Cheikh-Rouhouet *al*., 2008) and he found that β-Sitosterol and stigmasterol were the majors sterol.

The results of Kadri et *al*., (2015) show that the fatty acids present in the oils are mainly unsaturated from 70% for *P. pinaster* to 87% for *P. halepensis*. Excepted for *P. pinea*, the ratio of linoleic acid to oleic acid was more than 2:1, which is in agreement with the results found in literature on the studies of the lipid profiles of some pinus seeds (Andrey and Long, 1996; Nergiz and Domnez, 2004; Nasri et *al*., 2005).

Previous studies showed that the oils of pinus varieties contained oleic and linoleic acids at relatively high levels (Sagrero-Nieves, 1992; Wolff and Marpeau, 1997).

Flavonoids comprise an important group of plant secondary metabolites and can be classified in several subgroups (Ferreyra et *al*., 2012). These compounds are produced from the reaction between one molecule of cinnamoyl-CoA and three molecules of malonyl-CoA (Ferreyra et *al*., 2012).

Total phenol and flavonoid contents of the twigs and needles of *P. brutia*, *P. halepensis* and *P. pinea* extracts (acetone, ethyl acetate, and ethanol) were determined by Folin–Ciocalteau and AlCl3 reagents, respectively (Ustun et *al*., 2012).According to the results; the richest extracts concerning total phenol content was *P. halepensis* - needles - ethyl acetate (102.56 ± 5.47 mg). However; the extracts were found to have either no or low total flavonoid

amounts, while again *P. halepensis* - needles - ethyl acetate extract had the highest total flavonoid content (52.49 ± 3.57 mg) (Ustun et *al*., 2012)). Kilic et *al*., (2010) was interested in the polysaccharide (cellulose and polyoses) composition of *P. halepensis*, *P. pinea* and *P. brutia* cones. The results revealed that more polyoses than cellulose in the different cones and the highest amount was found in *P. pinea*.

The chemical variation of pine oils is due to several reasons, among these reasons we can quote geographical (Rezzi et *al*., 2001), seasonal (Isidorov et *al*., 2003), genotypic (Lazutka et *al*., 2001), and environmental (Kupcinskiene et *al*., 2008). In this context, Mohareb et *al*., 2017, studied the influence of altitude on the quantity and quality of the acetone extracts of *P. halepensis* (needles and bark). The results revealed differences in the yield of extracts and the chemical composition of different altitudes (125, 391 and 851m of sea level). Although the main components of all extracts are common, their percentages are different based on the tree species and the altitude level. These differences are the results of an adaptive process to particular ecologic conditions, which are in direct relation with the altitudes (Mohareb et *al*., 2017).

Pharmacological studies of Pine extracts

Many activities were detected from pine essential oils. In this review we summarized some of these activities (antifungal, herbicidal, anti-inflammatory, antioxidant, antibacterial, molluscicidal, antimicrobial, phytotoxic, insecticidal and antiradical) from different ancient studies in of *Pinus halepensis, pinea*, *brutia* and *pinaster*.

Antimicrobial activity

According to the articles studied in this review, the antimicrobial activities are the most extensively studied pharmacological activity of pinus essential oils extracts. Among these researches, we quote that of Fekih et *al*.,(2014) who found that the essential oil of *P.halepensis* needles, twigs and buds revealed an interesting antimicrobial effect against Lysteria monocytogenes, Enterococcus faecalis, Pseudomonas aeruginosa, Acinetobacter baumanii, Citrobacterfreundii and Klebsiella pneumoniae. For the same plant, the essential oils needles were tested in vitro by Sadou et *al*.2015 against three antibiotic resistant bacteria (Escherichia coli, Staphylococcus aureus and Pseudomonas aeruginosa) often responsible for nosocomial infections have showed antibacterial activity.

Hmamouchi et *al* .2001 studied needles essential oils of *P.halepensis*, *P.brutia*, *P. pinaster* and *P.pinea* and the examination of the antibacterial activity revealed that only P. pinaster and P. pinea oils exhibited a definite activity against all organisms tested. Compared to gentamicin, tetracycline and chloramphenicol, both oils showed a marked antiseptic activity. On the other hand, the results obtained by Mimoune et *al*.,(2013) relieved that the essential oils needles of *P. pinaster* posses a weak antibacterial effect against the microorganisms tested (S. aureus, B. subtilis and Escherichia coli) ,but did not present any activity against the fungalstrains (Erwiniaamylovora, Aspergillus niger and Aspergillus flavus).

For the resins, Ulukanliet *al*., 2014 showed that the essential oils of *P. brutia* and *P. pinea* grown in the Mediterranean region of Turkey had various levels of antimicrobial activities on Gram-positive, Gram-negative bacteria, and the yeast species, and hence, confirming the traditional uses of these resinous substances. Another part of *P.halepnsis* and *P.pinster* were studied by (Ghanmi et *al*., 2007): the gum. The turpentine was extracted and a biotest showed the efficient antibacterial and antifungal activity against Escherichia coli, Bacillus subtilis, Micrococcus luteus, Staphylococcus aureus, Penicilliumparasiticus and Aspergillus niger (Ghanmi et *al*., 2007).

The antibacterial and antifungal activities of chloroform, acetone and methanol extracts of several parts of *P. brutia* (barks, resins and cones) were studied by (Digrak et *al*.,1999).The extracts of several parts of the tree had no antifungal effect. The chloroform extract of the bark inhibited the growth of E. coli, with an inhibition zone of 8 mm. All the extracts tested inhibited the growth of all the other bacteria tested with zones of inhibition between 9–19 mm (Digrak et *al*., 1999). Four pathogenic bacteria (Agrobacterium tumefaciens, Erwiniacarotovora, Corynebacterium fascians and Pseudomonas solanacearum) and one fungal strain were used by Mohareb et *al*., 2017; to determine the antimicrobial activity of acetone extracts of *P. halepensis* (needles and bark) collected from three altitudes (125, 391 and 851 m). The extract obtained from the *P. halepensis* needles showed that Level I extract was the most active of the three levels of altitude with MIC values of 600, 840, 585 and 900mg/L against the growth of A. tumefaciens, E. carotovora, C. fascians and P. solanacearum, respectively. However, the level II extract showed the lowest activity with MIC of 735, 1200, 850 and 1300mg/L against the same bacteria, respectively. On the other hand, the extract obtained from the bark indicated that the altitude extract III was the most active among the three altitude levels (MIC=555, 600, 610 and 835mg/L against A. tumefaciens, E. carotovora, C. fascians and P. solanacearum, respectively).

On the other hand, the antifungal activitythe of extracts obtained from the leaves of *P. halepensis* had higher activity (EC50=119.20, 134.20, and 115.42mg/L) than the bark extracts (127.30, 154.71, and 139.70mg/L) at altitudes I, II, and III, respectively (Mohareb et *al*., 2017).

Antifungal activity of *P.halepensis* cone, needles and stems essential oils, were tested against several phytopathogenic strains by (Amri et *al*., 2013). The three samples of oils exhibited similar inhibition effects on the growth of fungi: F. avenaceum, F. culmorum, F. oxysporum, F. subglutinans, F. verticillioides, F. nygamai, Rhizoctonia sp., M. nivale, Alternaria sp. and B. sorokiniana (Amri et *al*., 2013).The study of (Abi-Ayad et *al*., 2011) show that spine essential oil of *P.halepensis* tree possesses antifungal activity against A. flavus, A. niger, F. oxysporum and R. stolonifer.

Antioxidant and anti-inflammatory activities

The acetone, ethyl acetate, and ethanol extracts and essential oils of the twigs and needles of *P. brutia*, *P. halepensis* and *P. pinea* along with pycnogenol (the bark extract of *P. pinaster*) were examined for their inhibitory effects against acetylcholinesterase (AChE) and butyrylcholinesterase (BChE) by (Ustun et *al*., 2012).Also, Their antioxidant activity was tested using 2,2-diphenyl-1-picrylhydrazyl (DPPH) and N,N-dimethyl-p-phenylendiamine (DMPD) radical scavenging, metal-chelation, and ferric-reducing antioxidant power (FRAP) assays (Ustun et *al*., 2012).According to this study, the best AChE and BChE inhibition was caused by the twig essential oil (83.91 ± 3.95%) and the needle ethanol extract (82.47 ± 5.57%) of *P. halepensis*, respectively(Ustun et *al*., 2012).Then for the *P.pinaster* cone, needles and wood, Tümen et *al*.,2017; investigated the antioxidant (DPPH photometric analysis, ABTS radical scavenging assay, FRAP assay and Non-site-specific hydroxyl radical (•OH) scavenging activity assay) and anti-inflammatory (Carrageenan-induced hind paw edema model, Acetic-Acid-Induced Increase in Capillary Permeability and TPA-induced mouse-ear edema) activities of the n-hexane and acetone extracts and essential oils. The essential oil from cones of *P. pinaster* revealed the highest activities, whereas other parts of the plant did not display any appreciable antioxidant, or anti-inflammatory effects (Tümen et *al*., 2017) .

This results was the same for the anti-inflammatory activity of essential oils of cones and needles of *P.halepensis*, *P.pinea* and *P.brutia* (Süntar et *al*., 2012). Antioxidant activities of *P. halepensis* essential oils, collected from ten different bioclimatic areas in Algeria have been determined by four tests namely DPPH, reducing power, -carotene bleaching and chelating ability on ferrous ions (Djerrad et *al*., 2015).

Other activities

The wound healing activity of essential oils extracted from cones and needles of *P.halepensis*, *P.pinea* and *P.brutia* were studied by (Süntar et *al*., 2012). And the same activity of the n-hexane and acetone extracts and essential oils of *P.pinaster* cone, needles and wood were tested by Tümen et *al*., 2017. The essential oils obtained from the cones of *P.pinea* and *P.halepensis* demonstrated the highest effects. On the other hand, the rest of the essential oils did not show any significant wound healing activities (Süntar et *al*., 2012). Other activity of essential oils from *P. halepensis*, *P. brutia*, *P. pinaster* and *P. pinea* were studied by Lahlou et *al*., 2003; the molluscicidal activity against Bulinustruncatus.

Concluding remarks:

This review presented compounds isolated from Pine extracts, including terpenoids, flavonoids, and fatty acids.

Cone was the most well-studied part of the plant, followed by extracts from the needle and seed. Hydro-distillation and soxhlet were the extraction methods with the ASE apparatus chosen by the most of studies, whereas hexane, acetone, methanol, water, acid hydrolysis and acid methanolysis, were used. Antioxidant activity was the most extensively studied of Pine extracts but many other activities are attributed to Pine extracts, such as Herbicidal, Antifungal, Wound healing, Antiradical and many others.

Several factors contribute to Pine importance including its worldwide geographic distribution, long history of use in folk medicine, food and cosmetics industry. Taken together, these factors stimulated an increasing interest in the isolation and identification of Pine compounds. The activities of some of the isolated compounds have not been investigated and, therefore, a comprehensive investigation of these compounds is still necessary. In this context, this species is a promising candidate as an alternative source of bioactive compounds.

Table 1. Chemical composition and activities of some extracts of *Pinus Halepensis*, *Pinus Pinea*,*Pinus Pinaster* and *Pinus Brutia*

Pinus	Tissue	Activity	Extraction method	Solvent	Compound detected	References
Halepensis	cone, needles and stems	antifungal and herbicidal	Hydrodistillation (3h)	_	Major compounds : α-pinene and (Z)-caryophyllene	Amri et *al*.,2013
Halepensis *Pinea* and *Pinaster*	cone, needles and branches	_	Hydrodistillation (2h)	_	Major compounds : α-pinene and myrcene Major compound :limonene Major compound s: α-pinene and β-pinene	Macchioni et *al*.,2002
Pinaster	cone, needles and wood	anti-inflammatory and antioxidant effects	Soxhlet Soxhlet Hydrodistillation (2h)	n-Hexane acetone _	_ _ Major compounds : α-pinene and β-pinene	Tümen et *al*.,2017
Halepensis *Pinea* and *Brutia*	cone	_	Hydrodistillation (3h)	_	Major compound : α-pinene Major compound :limonene Major compound : β-pinene	Tümen et *al*.,2010
Halepensis and *Pinea*	seeds	_	_	chloroform-methanol-NaCl (1%)	Major fatty acid : linoleic acid, oleic and palmitic	Nasri et *al*.,2005
Halepensis	spine	Antifungal _	Hydrodistillation (2h) cold solvent method	_	Major compounds :caryophyllene oxide, thumbergol and humulene oxide Major sterol compounds : β-Sitosterol and	Abi-Ayadet *al*.,2011

Halepensis	seeds			Hexane	campesterol	Cheikh-Rouhou et *al*.,2008
Halepensis Pinea and *Brutia*	twigs and needles	Antioxidant	Hydrodistillation (3h)	_ acetone, ethyl acetate ethanol	Major compounds: α-pinene and β-pinene Total phenol and flavonoid	Ustun et *al*.,2012
Halepensis	seeds	_	_	hexane	Major fatty acid : oleic and linoleic	Cheikh-Rouhou et *al*.,2006
Halepensis and *Pinaster*	seeds	_	Hydrodistillation (5h)	_	Major compound :limonene	Kadri et *al*.,2015
Halepensis Pinea and *Pinaster*	seeds	_	Soxhlet	chloroform/ methanol (2:1, v/v)	Fatty acids	Kadri et *al*.,2015
Halepensis	needles, twigs and buds	Antibacterial	Hydrodistillation (5h)	_	Major compounds :myrcene, α-pinene, E-β-caryophyllene, terpinolene, 2-phenyl ethyl isovalerate, terpinene-4-ol and sabinene	Fekih et *al*.,2014
	needles	Molluscicidal	Hydrodistillation (4h)	_		

Halepensis ,brutia, pinaster and *pinea*	needles	Antibacterial	Hydrodistillation (6h)	_	Major compounds :α-pinene, β-pinene and β–Myrcene	Lahlou et *al.*,2003
	resins	antimicrobial, phytotoxic, antioxidant, and insecticidal	Hydrodistillation (3h)	_	Major compounds :α-pinene, β-pinene and β–caryophyllene	Hmamouchi et *al.*,2001
Halepensis ,brutia, pinaster and *pinea*	needles		Hydrodistillation (3h)	_	Major compounds :α-pinene, β-pinene and caryophyllene	ULUKANLI et *al.*2014
Pinea and *Brutia*	needles	Herbicide and antifungal	Hydrodistillation (2h)	_	Major compounds : α-pinene and β-pinene	Hanana et *al.*,2014
Brutia	Needles	_	Hydrodistillation (2h)	_	Major compounds: β-caryophyllene, α-humulene and aromadendrene	Dob et al. (2005)
Halepensis	Needles	_	Hydrodistillation (4h)	_	Major compounds: β-caryophyllene, allo-aromadendrene and α-humulene.	Dob et al.(2005)
Pinaster	cones andneedles	Antibacterial	Hydrodistillation (6h)	_	Major compounds : β-caryophyllene and β-selinene	Mimoune et al.(2013)
Pinaster		wound healing and anti-inflammatory	_		_	Süntar et al.2012
Halepensis Pinea and	cones and		Soxhlet	Methanol chloroform,	Bioactive compounds : Total polyphenols, flavonoids, anthocyanins and carotenoids	Dhibi et al.2012

Brutia	seeds	Antiradical	AcceleratedSolventExtractor	acetone and methanol	_	
Halepensis	barks, resins and cones	Antimicrobial		acetone:water (95:5 v v−1)	Phenolic constituents	Digrak et al.1999
Brutia	cones		Hydrodistillation (3h)		major components : α-pinene, myrcene, p-cymene, (Z)-β-caryophyllene and caryophyllene oxide	Kilic et al.2010
Halepensis *Pinea* and *Brutia*	not mentioned	Antioxidant	Hydrodistillation (3h)	_	Major compounds : β–caryophyllene, α-pinene and β-myrcene	Djerrad et al.2015
Halepensis	needles	Antibacterial	Hydrodistillation (2h)	_	major components : α-pinene, β-pinene and germacrene D	Sadou et al.2015
Halepensis	needles	_	Hydrodistillation (3h)	_		Roussis et al. 1995
Halepensis *Pinea* and *Brutia*	cones	_	Hydrodistillation (5h)	_	Major compounds : α-pinene,myrcene, (E)-β-caryophyllene and caryophyllene oxide	

Halepensis	needles	–	Hydrodistillation (4h)		Major compounds : α-pinene, (E)-β-caryophyllene, germacrene D andabieta-7,13-diene	Nam et al.2014
Pinaster	barks	–	Hydrodistillation (2h)	–	Major compounds : α-pinene, β-caryophyllene and junipene	Ottavioli et al.2008
Pinaster	needles	–	Hydrodistillation	–	Major compounds : α-pinene,germacrene D and (E)-caryophyllene	Zolfaghari et al.2011
Pinaster	gem	Antibacterial and antifungal	–	–	Major compound : α-pinene	Petrakis et al.2001
Halepensis and *Pinaster*	needles and bark	Antimicrobial		Acetone	Major compounds: caryophyllene, α-caryophyllene and β-cubebene.	
Halepensis	Seeds	Antiradical	Soxhlet	petroleumether	Major compounds : D-glucose, 4,6-o-ethylidene, dibutyl phthalate and di-n-octyl phthalate	Ghanmi et al.2007
Halepensis	wood, cones, and needles	Antioxidant, wound healing and antiinflammatory	Hydrodistillation (4-5h)		Fattyacids Volatile compounds	Mohareb et *al*.,2017
Pinaster			Hydrodistillation (2h)		Volatile, lipophilic, and hydrophilic	Dhibi et *al*.,2012

	needles	Antimicrobial, insect larvicidal	Hydrodistillation (3h)		components	Tümen et *al*.,2018
Halepensis					Major terpene compounds: (E)-Caryophyllene, Thunbergol, Phenylethyl 3-methylbutanoate, α-Humulene and Myrcene	Mitić, et *al*.,2019
Pinea	needles, fruits and barks	Antibacterial and antioxidant	Masquelier modified method			Halloum et *al*.,2019
Pinea, *pinaster* and *halepensis*	Bark	Antioxidant, cytotoxicity, antiproliferative			Limonene, β-caryophyllene, Caryophyllene oxide and Guaiol	Gascón et *al*.,2018

Figure 1. Pine morphology (https://fr.depositphotos.com/226090882/stock-illustration-parts-plant-morphology-pine-tree.html)

Competing interests

We have no competing interests.

References

Abi-Ayad M., Abi-Ayad F. Z., Lazzouni H. A., Rebiahi S. A., Ziani_Cherif C. and Bessiere (2011), Chemical composition and antifungal activity of Aleppo pine essential oil. *Journal of Medicinal Plants Research* 5(22) pp5433-5436.

Ahmed SO Mohareb, Ibrahim EA Kherallah, Mohamed EI Badawy, Mohamed ZM Salem, Hameda A Yousef, (2017). Chemical composition and activity of bark and leaf extracts of pinus halepensis and oleaeuropaea grown in AL-Jabel AL-Akhdar region, Libya against some plant phytopathogens. *Journal of Applied Biotechnology & Bioengineering* 7;3(3), pp331–342.

Ahmed Z, Siddiqui M, Khan I (1969) Combined effects of diphenyliodonium chloride, pine oils and mustard oil soaps on certain microorganisms. *Appl Environ Microbiol* 17(6), pp857–860.

Amri, I., Hamrouni, L., Hanana, M., Gargouri, S., Fezzani, T., & Jamoussi, B. (2013). Chemical composition, physico-chemical properties, antifungal and herbicidal activities of Pinus halepensis Miller essential oils. *Biological Agriculture & Horticulture* 29(2), pp91–106.

Andrey, B. Imbs., & Long, Q. P. (1996). Fatty acids and triacylglcerols in seeds of pinaceae species. *Phytochemistry*, 42(4), pp1051–1053.

Cheikh-Rouhou S., Hentati B., Besbes S., Blecker C., Deroanne C., and Attia H. (2006).Chemical Composition and Lipid Fraction Characteristics of Aleppo Pine (PinushalepensisMill.) Seeds Cultivated in Tunisia. *Food Sci. Tech. Int.* 15(5), pp407–416.

Cheikh-Rouhou, S., Besbes, S., Lognay, G., Blecker, C., Deroanne, C., & Attia, H. (2008). Sterol composition of black cumin (Nigella sativa L.) and Aleppo pine (Pinus halepensis Mill.) seed oils. *Journal of Food Composition and Analysis* 21(2), pp162–168.

Cheung H., Toshio Miyase, Mark P. Lenguyen, and Mary A. Smal, (1993). Further Acidic Constituents and Neutral Components of Pinus massoniana Resin, *Tetrahedron* 49(36), pp7903-7915.

Devaraja S, Vega-Lópeza S, Kaula N, Schönlaub F, Rohdewaldb P, Jialala I (2002) Supplementation with a pine bark extract rich in polyphenols increases plasma antioxidant capacity and alters the plasma lipoprotein profile. *Lipids* 37(10), pp931–934.

Dhibi, M., Mechri, B., Brahmi, F., Skhiri, F., Alsaif, M. A., & Hammami, M. (2012). Fatty acid profiles, antioxidant compounds and antiradical properties of Pinus halepensis Mill. cones and seeds. *Journal of the Science of Food and Agriculture* 92(8), pp1702–1708.

Dığrak, M., İlçim, A., & Hakkı Alma, M. (1999).Antimicrobial activities of several parts of Pinus brutia, Juniperus oxycedrus, Abies cilicia, Cedrus libani and Pinus nigra. *Phytotherapy Research* 13(7), pp584–587.

Djerrad, Z., Kadik, L., & Djouahri, A. (2015). Chemical variability and antioxidant activities among Pinus halepensis Mill. essential oils provenances, depending on geographic variation and environmental conditions. *Industrial Crops and Products* 74, pp440–449.

Dob T., Berramdane T. and Chelghoum C. (2005). ANALYSIS OF ESSENTIAL OIL FROM THE NEEDLES OF Pinus pinaster GROWING IN ALGERIA. *Chemistry of Natural Compounds* 41, pp545–548.

Falcone Ferreyra, M. L., Rius, S. P., & Casati, P. (2012). Flavonoids: biosynthesis, biological functions, and biotechnological applications. *Frontiers in Plant Science* 3.

Fang JM, Lang CI, Lien W, Cheng YS (1991). Diterpenoid Acid from the Leaves of Armand Pine. *Phytochemistry* 30, pp2793-2795.

Fang JM, Su WC, Cheng YS (1988). Flavonoids and Stilbenes From Armand Pine. *Phytochemistry* 27, pp1395-1397.

Fang JM, Tsai WY, Cheng YS (1991).Serratene Triterpenes from Pinus armandii Bark. *Phytochemistry* 30, pp1333-1336.

Fekih, N., Allali, H., Merghache, S., Chaïb, F., Merghache, D., El Amine, M., Costa, J. (2014). Chemical composition and antibacterial activity of Pinus halepensis Miller growing in West Northern of Algeria. *Asian Pacific Journal of Tropical Disease* 4(2), pp97–103.

Gascón, S., Jiménez-Moreno, N., Jiménez, S., Quero, J., Rodríguez-Yoldi, M. J., & Ancín-Azpilicueta, C. (2018). Nutraceutical composition of three pine bark extracts and their antiproliferative effect on Caco-2 cells. *Journal of Functional Foods* 48, pp420–429..

Ghanmi, M., Satrani, B., Chaouch, A., Aafi, A., Abid, A. E., Ismaili, M. R., & Farah, A. (2007). Composition chimique et activité antimicrobienne de l'essence de térébenthine du pin maritime (Pinus pinaster) et du pin d'Alep (Pinus hale- pensis)du Maroc. *Acta Botanica Gallica* 154(2), pp293–300.

Hanana M, Bejia A., AMRI I., GARGOURI S., Jamoussi B., Hamrouni L. (2014). Activités biologiques des huiles essentielles de pins. *Journal of New Sciences* 4(3).

Halloum B. , Ashok K. Shakya, Zaha A. Elagbar, Rajashri R. Naik (2019), GC-MS Analysis and Biological activity of Essential Oil of Fruits, Needles and Bark of Pinus pinea grown wildly in Jordan. *Acta Poloniae Pharmaceutica - Drug Research* 76(5), pp825-831.

Hmamouchi M., Hamamouchi J., Zouhdi M.& Bessiere J.M (2001): Chemical and Antimicrobial Properties of Essential Oils of Five Moroccan Pinaceae, *Journal of Essential Oil Research* 13(4), pp298-302.

Isidorov VA, Vinogorova VT and Rafaowski K (2003). HS-SPME analysis of volatile organic compounds of coniferous needle litter. *Atmos Environ* 37, pp4645–4650 .

Kadri, N., Khettal, B., Aid, Y., Kherfellah, S., Sobhi, W., & Barragan-Montero, V. (2015). Some physicochemical characteristics of pinus (Pinus halepensis Mill., Pinus pinea L., Pinus pinaster and Pinus canariensis) seeds from North Algeria, their lipid profiles and volatile contents. *Food Chemistry* 188, pp184–192.

Kähkönen MP, Hopia AI, Vuorela HJ, Rauha JP, Pihlaja K, Kujala TS, Heinonen M (1999) Antioxidant activity of plant extracts containing phenolic compounds. *J Agric Food Chem* 47(10), pp3954–3962.

Kilic, A., Hafizoglu, H., Tümen, I., Dönmez, I. E., Sivrikaya, H., Sundberg, A., & Holmbom, B. (2010). Polysaccharides in cones of eleven coniferous species growing in Turkey. *Wood Science and Technology* 44(3), pp523–529

Kilic, A., Hafizoglu, H., Tümen, I., Dönmez, I. E., Sivrikaya, H., & Hemming, J. (2010). Phenolic extractives of cones and berries from Turkish coniferous species. European Journal of Wood and Wood Products 69(1), pp63–66.

Kurose, K., Okamura, D., & Yatagai, M. (2006).Composition of the essential oils from the leaves of ninePinus species and the cones of three ofPinus species. *Flavour and Fragrance Journal* 22(1), pp10–20.

Lahlou M.(2015). Composition and Molluscicidal Properties of Essential Oils of Five Moroccan Pinaceae. *Pharmaceutical Biology* 41(3), pp207-210·

Macchioni F., Cioni P. L., Flamini G., Morelli I., Maccioni S. and Ansaldi M. (2002). Chemical composition of essential oils from needles, branches and cones of Pinus pinea, P. halepensis, P. pinaster and P. nigra from central Italy. *Flavour and Fragrance Journal* 18 (2), pp139-143.

McKevith, B. (2005).Nutritional aspects of oil seeds. *Nutrition Bulletin* 30(1), pp13–26.

Mimoune N.A., Mimoune D.N, Yataghene A. (2013). Chemical composition and antimicrobial activity of the essential oils of Pinus pinaster. *Journal of Coastal Life Medicine* 1(1), pp55-59.

Mitić, Z. S., Jovanović, B., Jovanović, S. Č., Stojanović-Radić, Z. Z., Mihajilov-Krstev, T., Jovanović, N. M., Stojanović, G. S. (2019). Essential oils of Pinus halepensis and P. heldreichii: Chemical composition, antimicrobial and insect larvicidal activity. *Industrial Crops and Products* 140, 111702.

Nergiz, C., & Dönmez, İ. (2004). Chemical composition and nutritive value of Pinus pinea L. seeds. *Food Chemistry* 86(3), pp365–368.

Nam AM., Casanova J., Tomi F. and Bighelli A.(2014). Composition and Chemical Variability of Corsican Pinus halepensis Cone Oil. *Natural Product Communications* 9(9).

Nasri, N., Khaldi, A., Fady, B., & Triki, S. (2005).Fatty acids from seeds of Pinus pinea L.: Composition and population profiling. Phytochemistry 66(14), pp1729–1735.

Ottavioli, J., Bighelli, A., & Casanova, J. (2008). Diterpene-rich needle oil ofPinus pinaster Ait. from Corsica. *Flavour and Fragrance Journal* 23(2), pp121–125.

Packer L, Rimbach G, Virgili F (1999). Antioxidant activity and biological properties of a procyanidin rich extract from Pine. *Free RadicBiol Med* 27, pp704–724.

Panos V. Petrakis, Christina Tsitsimpikou, Olga Tzakou, Maria Couladis, ConstantinosVagias and VassiliosRoussis,(2001). Needle volatiles from five Pinus species growing in Greece. *FlavourFragr. J.* 16, pp249–252.

Roussis V., Petrakis PV., ORTIZ A. and Mazomenos B E. (1995). Volatile constituents of needles of five Pinus species grown in Greece. *Phytochemistry* 39(2), pp357–361.

Sadou N., Seridi R., Djahoudi A. & Hadef Y. (2015). Chemical composition and antibacterial activity of the needls essential oil of Pinus halepensis Mill. from north east of Algeria. *Rev. Sci. Technol., Synthèse* 30, pp33-39.

Sagrero-Nieves, L. (1992). Fatty acid composition of mexican pine nut (Pinuscembroides) oil from three seed coat phenotypes. *Journal of the Sciences of Food Agriculture* 59, pp413–414.

Sakagami H, Kawazoe Y, Komatsu N, Simpson A, Nonoyama M, Konno K, Yoshida T, Kuroiwa Y, Tanuma S (1991). Antitumor, antiviral and immunopotentiating activities of pine cone extracts: potential medicinal efficacy of natural and synthetic lignin-related materials (review). *Anticancer Res.* 11(2), pp881-888.

Sezic E., Ustun O., Demirci B., K.Husnu Can BASER (2010). Composition of the essential oils of Pinus nigra Arnold from Turkey. *Turk J Chem* 34, pp313-325.

Süntar, I., Tumen, I., Ustün, O., Keleş, H., & Küpeli Akkol, E. (2012).Appraisal on the wound healing and anti-inflammatory activities of the essential oils obtained from the cones and needles of Pinus species by in vivo and in vitro experimental models. *Journal of Ethnopharmacology* 139(2), 533–540.

Tumen, I., Hafizoglu, H., Kilic, A., Dönmez, I. E., Sivrikaya, H., & Reunanen, M. (2010). Yields and Constituents of Essential Oil from Cones of Pinaceae spp. Natively Grown in Turkey. *Molecules* 15(8), pp5797–5806.

Tümen, İ., Akkol, E. K., Taştan, H., Süntar, I., & Kurtca, M. (2018). Research on the antioxidant, wound healing, and anti-inflammatory activities and the phytochemical composition of maritime pine (Pinus pinaster Ait). *Journal of Ethnopharmacology* 211, pp235–246.

ULUKANLI, Z., KARABÖRKLÜ, S., BOZOK, F., ATES, B., ERDOGAN, S., CENET, M., & KARAASLAN, M. G. (2014). Chemical composition, antimicrobial, insecticidal, phytotoxic and antioxidant activities of Mediterranean Pinus brutia and Pinus pinea resin essential oils. *Chinese Journal of Natural Medicines* 12(12), pp901–910.

Unten S., Sakagami H., and Konno K. (1989). Stimulation of Granulocytic Cell lodination by Pine Cone Antitumor Substances. *Journal of Leukocyte Biology* 45, pp168-175.

Ustun, O., Senol, F. S., Kurkcuoglu, M., Orhan, I. E., Kartal, M., & Baser, K. H. C. (2012). Investigation on chemical composition, anticholinesterase and antioxidant activities of extracts and essential oils of Turkish Pinus species and pycnogenol. *Industrial Crops and Products* 38, pp115–123.

Ustün, O., Ozçelik, B., Akyön, Y., Abbasoglu, U., & Yesilada, E. (2006). Flavonoids with anti-Helicobacter pylori activity from Cistus laurifolius leaves. *Journal of Ethnopharmacology* 108(3), pp457–461.

Villagomez HZ., Peterson DM., Herrin L., Young RA. (2005). Antioxidant activity of different components of pine species. *Holzforschung* 59, pp156–162.

Watanabe K, Momose F, Handa H, Nagata K (1995). Interaction between influenza virus proteins and pine cone antitumor substance that inhibits the virus multiplication. *Biochemical and Biophysical Research Communications* 214(2), pp318–323.

Willför S, Ali M, Karonen M, Reunanen M, Arfan M, Harlamow R (2009). Extractives in bark of different conifer species growing in Pakistan. *Holzforschung* 63, pp551–558.

Wolff, R. L., & Marpeau, A. M. (1997). D5-olefinic acids in the edible seeds of nut pines (Pinuscembroides edulis) from the United States. *Journal of American Oil Chemists Society* 74(5), pp613–614.

Xu, R.-B., Yang, X., Wang, J., Zhao, H.-T., Lu, W.-H., Cui, J., Hu, X.-L. (2012). Chemical Composition and Antioxidant Activities of Three Polysaccharide Fractions from Pine Cones. *International Journal of Molecular Sciences* 13(12), pp14262–14277.

Yang X., Zhao H. T., Wang J. , Meng Q, Zhang H, Yao L., Zhang Y.C., Dong A. J., Ma Y , Wang Z. Y. , Xu D. C. and Ding Y. (2010). Chemical composition and antioxidant activity of essential oil of pine cones of Pinus armandii from the Southwest region of China . *Journal of Medicinal Plants Research* 4(16), pp1668-1672.

Zolfaghari, B., & Iravani, S. (2012). Essential Oil Constituents of the Bark of Pinus pinaster from Iran. *Journal of Essential Oil Bearing Plants* 15(3), pp348–351.

Arabian Journal of Medicinal and Aromatic Plants
www.ajmap.info **ISSN 2458-5920**