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Review

Diversity of Needle Terpenes Among *Pinus* Taxa

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Abstract: Essential oils are mixtures of volatile compounds often found in the leaves, wood, and fruits of coniferous trees and shrubs. The composition and abundance of individual oil components vary across different plant parts. Terpenes (monoterpenes, sesquiterpenes, diterpenes, triterpenes) dominate in the essential oils of many plants. They are the most abundant class of secondary metabolites, with plants containing over a hundred of them at varying concentrations. The terpene profile of certain species consists of a few dominant (abundant) components and numerous less abundant ones. It is believed that the biological activity of essential oil mostly depends on the dominant terpene components. In most of the analyzed *Pinus* species, the most abundant terpene compounds are α-pinene, β-pinene, δ-3-carene, β-caryophyllene, limonene/β-phellandrene, and germacrene D. In certain taxa, additional dominant compounds include α-cedrol, bornyl acetate, caryophyllene oxide, α-phellandrene, trimethylbicyclo [3.1.1]hept-2-ene, 2H-benzocyclohepten-2-one, phenylethyl butyrate, 4-epi-isocembrol, β-thujene, and thunbergol. Moreover, compounds with abundances exceeding 15% include methyl chavicol (=estragole), geranylene, myrcene, γ-muurolene, sabinene, and abieta-7,13-diene. It can be concluded that the terpene profiles of the needles of the analyzed pine taxa depend on the type of chromatographic columns, the method of obtaining essential oils, the origin of the trees (in natural habitat or artificial plantation), the age of the needles, the variety, and the season in which the needles were collected.

Keywords: needles; essential oil; *Pinus* taxa; variability; terpene profiles; terpene classes

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1. Introduction

Plant essential oils are mixtures of volatile compounds predominantly found in the wood, bark, buds, leaves, and cones of coniferous trees and shrubs. They are also found in the seeds, fruits, flowers, and roots of many plants [1]. The terpene content in plants is not high, reaching up to 1% [2], but it is highly variable. Terpenes (monoterpenes, sesquiterpenes, diterpenes, triterpenes) dominate in the essential oils of many plants. They are the most abundant class of secondary metabolites, with plants containing sometimes over a hundred of them at varying concentrations [3]. The terpene profile of certain species consists of a few dominant (abundant) components (mainly monoterpenes and sesquiterpenes) and numerous less abundant ones. The composition and abundance of individual oil components vary across different plant parts. External factors, such as industrial pollution [4] and various ecological factors, can influence the terpene profile of a plant [5,6].

Among the plants that produce essential oils, the genus *Pinus* stands out for its rich content of essential oils, which have been used since ancient times in traditional medicine, as well as raw materials in the pharmaceutical, cosmetic, perfumery, and food industries [7]. It is believed that the biological activity of essential oil mostly depends on the dominant terpene components, but the less abundant components are also important due to their synergistic effects with the dominant ones. According to current research, pine essential oils possess antiviral, antibacterial, antifungal, herbicidal, molluscicidal, nematocidal, and acaricidal activities, as well as toxicity towards insects and rodents. They also exhibit antioxidant, anti-inflammatory, analgesic, immunomodulatory, cytotoxic, antigenotoxic, dermatoprotective, the inhibition of acetylcholinesterase, and other properties [8,9]. In this way, pine essential oils have shown great potential in the prevention and treatment of various diseases, including cancer [10]. They have also been found to be effective as natural insecticides and can be used for storing agricultural products [7]. Furthermore, certain terpenes, which are often dominant compounds in pine essential oil (such as limonene and pinanes), are used in the production of perfumes and air fresheners [11]. Finally, studying a larger number of biological activities could help expand our understanding of the medicinal benefits and applications of pine essential oils in medicine and pharmacology, as well as their role in plant protection and ecological activities (such as biocides) [12].

According to Royal Botanic Gardens and Kew and Missouri Botanical Garden, there are around 126 recognized species of pines, 35 unresolved species, and many more synonyms [13]. The range of only one pine species crosses the equator (*Pinus merkusii*), on the island of Sumatra [13].

Pines are evergreen, monoecious, and resinous trees (rarely shrubs) with whorled branches, predominantly resinous buds, and shoots with needle-shaped leaves arranged in clusters, usually 2–5 (up to 8) together. In the cross-section, the needle is triangular or semicircular, with one vascular bundle (Haploxylon) or two vascular bundles (Diploxylon), and it usually has two or more resin canals [13].

All pines are classified into two subgenera based on needle morphology: *Pinus* (Diploxylon, hard pines) and *Strobus* (Haploxylon, soft pines) (Little and Critchfield 1969, after [14]). The subgenus *Pinus* is divided into two sections: the *Pinus* section, with the subsections *Pinus* and *Pinaster*, and the *Trifoliae* section, with the subsections *Contortae*, *Australes*, and *Ponderosae*. The subgenus *Strobus* is divided into two sections: the *Parrya* section, with the subsections *Balfourianae*, *Cembroides*, and *Nelsoniae*, and the *Quinquefoliae* section, with the subsections *Gerardianae*, *Krempfianae*, and *Strobus*. Morphological differences between subgenera, sections, and subsections are also supported by numerous molecular phylogenetic studies, as cited by Gernandt et al. [14] and later studies [15–20].

The aims of this review were to summarize references about the essential oils of pines, to correlate terpene profiles with their taxonomic status, as well as to compare terpene profiles with the variability of the type of chromatographic column, procedure of terpenes isolation, origin and variety of trees, and method of needles collection.

2. The Basis Properties of 104 *Pinus* Taxa

This review paper presents data for 104 taxa of the genus *Pinus* (66 species, 20 subspecies, 17 varieties, and 1 form), organized in alphabetical order (Table S1). Taxonomic positions of *Pinus* species were used according to Gernandt et al. [14]. Synonyms were used after Vidaković and Franjić [13] and Earle [21]. Natural areas were presented according to the maps of Critchfield and Little [22].

A total of 78 taxa are presented in the subgenus *Pinus*, with 48 in the *Pinus* section (including 36 taxa in the *Pinus* subsection and 12 taxa in the *Pinaster* subsection) and 30 in the *Trifoliae* section (including 15 taxa in the *Australes* subsection, 4 in the *Contortae* subsection, and 11 in the *Ponderosae* subsection). A total of 26 taxa are presented in the subgenus *Strobus*, with 4 in the *Parrya* section (2 taxa in the *Balfourianae* subsection and 2 taxa in the *Cembroides* subsection) and 22 in the *Quinquefoliae* section (2, 1, and 19 taxa in the *Gerardianae*, *Krempfianae*, and *Strobus* subsections, respectively). This review includes 15 endemic and 3 relict pines.

3. The Main Terpene Compounds of *Pinus* Taxa

The main (most abundant) terpene components in the needles of *Pinus* taxa are shown in Table S2.

From the data presented in Table S2, it is clear that the terpene profiles of the needles of the analyzed pine taxa depend on the type of chromatographic columns, the method of obtaining essential oils, the origin of the trees (in natural habitat or artificial plantation), the age of the needles, the variety, and the season in which the needles were collected.

In *Pinus albicaulis*, the terpene composition differs between the artificial plantation, where α-pinene dominates [23], and its natural distribution, where δ-3-carene predominates [24] (Table S2). In addition to the origin of the trees, these two oils were processed under different chromatographic conditions (column type) and using different techniques for obtaining essential oil: hydrodistillation and hydrodistillation with extraction. These two *P. albicaulis* oils are derived from needles collected in winter and summer, respectively.

In the needles of *P. aristata*, δ-3-carene dominates [23], while in *P. arizonica*, α-pinene and germacrene D are the dominant components (average of three locations) [25].

In the essential oil of *P. armandii*, α-pinene dominates in both autumn and winter conifers, regardless of whether it is from the native habitat [26], artificial plantation [23], or the method of oil extraction [27]. However, it seems that the terpene profile of needles collected in the summer [28] is significantly different, with β-caryophyllene being the most abundant. The headspace technique has determined that limonene/β-phellandrene dominate in *P. armandii* [29]. In the EO of *P. armandii* subsp. *xuannhaensis*, α-cedrol is the most abundant [30]. In the EO of *P. attenuata*, in addition to α-pinene [23], β-pinene [31] also dominates.

β-Pinene is also the most abundant in the essential oil of *P. balfouriana* ssp. *balfouriana* [32]. In the EO of *P. banksiana* obtained from summer needles, bornyl acetate is the most abundant [23].

In the needles of *P. brutia*, β-pinene also dominates [23,25,33–36]. Only in two artificial plantations [37,38] do α-pinene and α-terpineol, respectively, dominate. In *P. brutia* ssp. *eldarica*, oils obtained by different extraction and chromatography techniques differ [34,39]. In the oil of *P. brutia* var. *stankewiczii*, β-pinene is the most abundant, but germacrene D is also abundant [36]. In the case of *P. bungeana*, needle sampling was conducted over the summer, but the terpene profiles of the EOs differ due to habitat differences (natural and artificial plantations) and chromatographic conditions [23,27,40,41]. α-Pinene dominates only in the natural habitat [27].

In several samples of EOs from the endemic pine, *P. canariensis*, germacrene D is the dominant component, but the content of α-pinene is also quite high [23,25,36,42], and even dominant in an artificial plantation in Egypt [43].

In the EO of *P. caribaea* from artificial plantations, aromadendrene [37] and limonene [44] are the most abundant. In *P. caribaea* var. *bahamensis* and *P. caribaea* var. *caribaea*, the most abundant components are β-phellandrene and geranylene [45]. In *P. caribaea* var. *hondurensis*, limonene dominates [44].

In *P. cembra*, α-pinene dominates both during headspace extraction [29] and in oils from Romania [46], Slovakia [47], and Austria [48], while in the Botanical Garden and Arboretum, germacrene D, α-pinene [23], and α-cadinene [49] dominate, respectively.

In *P. cernua* needles, myrcene is dominant [50]. In *P. contorta* ssp. *contorta* and ssp. *murrayana* EOs, β-phellandrene dominates [23,51], while in ssp. *latifolia*, β-pinene and β-phellandrene are most abundant [23,52]. In artificial plantations of *P. coulteri*, β-caryophyllene [37], an unidentified compound [23], or multiple terpenes [53] dominate. In winter needles of the endemic *P. culminicola* and in the EO of *P. dalatensis*, α-pinene is the most abundant component [23,30]. In the needles of the endemic pine, *P. densata*, β-caryophyllene dominates [27].

In *P. densiflora* needles from an artificial plantation, collected in the autumn, α-pinene dominates [54], while in the artificial plantation from Korea, β-thujene is the most abundant [55]. The EO of *P. densiflora* from Korea is dominated by α-pinene [56] and camphene [57]. In the summer needles of the same species, camphene [57], α-humulene [58], and *trans*-cadina-1(6),4-diene [23] are the most abundant, while in the spring, α-pinene, sabinene [40], and α-myrcene [41] dominate. On average, throughout all four seasons, β-phellandrene and β-pinene are the most abundant [9].

In the EOs of *P. elliotti* from artificial plantations, the highest content of α-pinene is found during the summer [57], while germacrene D dominates in winter [23] and autumn [59]. In one report, geranylene is the dominant component [45].

In the natural habitat of the endemic pine *P. fenzeliana*, α-pinene dominates [60]. In the EO of *P. flexilis*, α-pinene predominantly dominates, regardless of whether it is from artificial plantations [23,61] or native habitats [51]. The only exception is the EO from Utah, where β-pinene and α-pinene dominate [24].

In the artificial plantation of *P. gerardiana*, α-pinene dominates [23].

In the EOs of *P. halepensis*, the dominant components are (E)-caryophyllene [23,25,36,62–71], α-pinene [33,34,72–76], caryophyllene [65,77,78], thunbergol [69], myrcene [69,79–81], caryophyllene-oxide [82], and β-pinene [35,83].

In the needles of *P. hartwegii*, β-pinene and caryophyllene oxide dominate [84].

In the needles of the tertiary relic and Balkan subendemic *P. heldreichii*, germacrene D [85–89] or limonene [23,31,68,86,90–93] dominates. In the essential oils of *P. heldreichii* var. *leucodermis* from Bosnia and Herzegovina [94] and Bulgaria [95], limonene dominates, while in the same variety from Italy [96], α-pinene is the most abundant.

In the needles of the endemic pine *P. henryi*, β-caryophyllene dominates [97]. In the needles of *P. jeffreyi* from artificial plantations, α-pinene dominates [23,98].

In the needles of *P. koraiensis*, limonene (headspace) [29], α-pinene [84], β-caryophyllene [27], and most frequently germacrene D [16,23,40] dominate. In the endemic pine *P. krempfii* from Vietnam, α-pinene dominates [99].

In the summer needles of *P. lambertiana*, β-pinene strongly dominates [24]. In the needles of *P. latteri* from China and Vietnam, caryophyllene was the most abundant [45].

In the needles of *P. massoniana*, various terpenes also dominate: α-pinene [23,45], β-caryophyllene [97], and α-thujene [27]. In the *massoniana* variety, α-pinene dominates [45]. In the needles of *P. merkusii*, β-caryophyllene dominates [84], while in the needles of *P. monophylla*, β-pinene dominates [23]. In *P. monticola*, three components have similar abundance: β-elemene, α-pinene, and β-pinene [23], or only β-pinene dominates [24,100].

Essential oils from the needles of *P. mugo* have been frequently studied, so the results vary. The most abundant terpene components are α-pinene [23,91,101–105], bornyl acetate [106], δ-3-carene [61,81,89,93,107,108], β-pinene [109], and limonene [110]. In the needles of *P. mugo* ssp. *mugo*, the most abundant component is δ-3-carene [108,111]. In the essential oil of *P. mugo* var. *prostrata*, the dominant terpene components are bornyl acetate and α-pinene, while in *P. mugo* var. *pumilio*, δ-3-carene and α-pinene dominate [23]. The EO of *P. mugo* ssp. *uncinata* contains the highest amounts of α-pinene and myrcene [29], or bornyl acetate [112]. In the needles of *P. muricata*, the most abundant component is germacrene D [23].

The most studied pine in terms of essential oils from needles is certainly *P. nigra*. In most studies, the most abundant component is α-pinene [25,35,36,54,79,81,92,93,113–116], with acetic acid [61] or germacrene D [36] being notably present as well. α-Pinene is also dominant in the needles of ssp. *banatica* [117,118], var. *calabrica* [96], ssp. *dalmatica* [118,119], ssp. *gocensis* [117], ssp. *laricio* [23,120], ssp. *nigra* [23,96,111,117,118,121], and ssp. *pallasiana* [117,118]. β-Pinene is dominant in ssp. *caramanica* [23] and, together with germacrene D, in ssp. *pallasiana* [23]. β-Pinene sometimes dominates alongside α-pinene [122]. Manoyl oxide is exceptionally abundant in ssp. *laricio* [120], and in one case, it is the most abundant component [123]. Caryophyllene oxide dominates in ssp. *laricio*, ssp. *nigra*, ssp. *pallasiana*, and ssp. *salzmannii* [124]. Germacrene D is the most abundant in some samples of ssp. *pallasiana* [89] and ssp. *salzmannii* [23]. The dominance of (E)-caryophyllene or hexadecanoic acid has also been observed in ssp. *mauritanica* [125].

In the endemic pine *P. occidentalis*, the dominant terpene is β-pinene [126].

The needles of *P. palustris* from an artificial plantation have α-terpineol as the dominant component [84]. In artificial plantations of *P. parviflora*, the most abundant components are α-pinene [84], bornyl acetate [40], or β-phellandrene [23]. In headspace samples, the most abundant components are limonene/β-phellandrene [29] and α-pinene [61]. In artificial plantations of *P. patula*, the most abundant components are caryophyllene oxide [84], α-pinene [127], and germacrene D [23].

Needles of the endemic-relict pine *P. peuce* have α-pinene as the most abundant EO component in its natural habitats [23,29,31,91,93,111,128–134].

In the needles of *P. pinaster* from natural habitats, α-pinene is the most abundant [31,79], while in artificial plantations, in addition to α-pinene [70,135], other abundant components include (E)-caryophyllene [33,136–138], abieta-7,13-diene [139], β-pinene [140], limonene [83], and isoabienol [23]. (E)-caryophyllene is also the most abundant in two varieties of *P. pinaster*: *maghrebiana* and *renoui* [141].

In the EO profiles of *P. pinea*, the most abundant terpenes are limonene [23,25,70,79,83,142–145], α-pinene [33,73], β-pinene [35,74], and guaiol [146]. In the needles of *P. ponderosa*, β-pinene predominates [23,84,147,148]. In var. *ponderosa*, β-pinene also predominates [51], while in var. *brachyptera*, α-pinene is dominant [149]. In the EOs of *P. pumila*, limonene/β-phellandrene [29], α-pinene [84], and δ-3-carene [23,150] predominate. β-Pinene predominates [151] in the needles of *P. pungens*.

In the EOs of *P. radiata*, β-pinene also predominates [23,31,53,152]. In the needles of *P. resinosa*, β-pinene [148] and caryophyllene [61] predominate. In *P. rigida*, the most abundant compounds are β-pinene [23,84], bornyl acetate [40], and 2H-benzocyclohepten-2-one [58]. In *P. roxburghii*, the most abundant terpenes are α-pinene [153], (E)-caryophyllene [23,154], and α-phellandrene [155].

In the needles of the endemic species *P. sabiniana*, α-pinene predominates [23,24,98], with phenylethyl butyrate being exceptionally present [53]. The commercial oil of *P. sibirica* also has the highest content of α-pinene [156]. In the endemic species *P. strobiformis* from an artificial plantation, the most abundant terpene is germacrene D [23]. In the EOs of *P. strobus*, in addition to δ-3-carene [29], α-pinene [23,27,40,148] and β-pinene [36,100] also predominate.

In the needles of *P. sylvestris*, α-pinene predominates, both in natural and artificial plantations, under different types of essential oil extraction and chromatographic column types [23,34,54,81,93,111,157–162]. In addition to α-pinene, there are reports where the most abundant compounds are manoyl oxide [101] and trimethylbicyclo [3.1.1]hept-2-ene [41]. In the needles of ssp. *scotica*, isoabienol is the most abundant [23], while in the needles of var. *mongolica*, α-pinene [163] and δ-cadinene [27] predominate.

In the EOs of *P. tabuliformis*, the most abundant compounds are β-caryophyllene [28,97], α-pinene [23,27], and trimethylbicyclo [3.1.1]hept-2-ene [41]. In var. *mukdensis*, β-caryophyllene [97] and α-pinene [27] predominate, while in the form *shekanensis* and the variety *umbraculifera*, β-caryophyllene [97] is the most abundant.

In the needles of *P. taeda*, α-pinene predominates [45]. In the needles of the endemic pines *P. taiwanensis* and *P. teocote*, α-pinene predominates [23]. In the EOs of *P. thunbergii*, the most abundant components are β-pinene [40,54], δ-3-carene [57], germacrene D [23,41], and 2H-benzocyclohepten-2-one [58]. In the needles of *P. torreyana* from artificial plantations, the most abundant compound is 4-epi-isocembrol [23], while in ssp. *insularis* and ssp. *torreyana*, thunbergol predominates [164].

In *P. uliginosa*, the profile of the most abundant terpene components consist of α-pinene, bornyl acetate, and δ-3-carene, which are of similar abundance [112].

The most abundant terpene components in the needles of *P. wallichiana* obtained using the headspace technique are limonene/β-phellandrene [29], while hydrodistillation reveals β-pinene [23,100,165]. In the needles of *Pinus wangii* subsp. *kwangtungensis*, β-pinene predominates [30].

In the EOs from the needles of *P. yunnanensis*, the most abundant components are β-caryophyllene [27] and β-pinene [45], while in its variety *tenuifolia*, α-pinene predominates [45].

Thus, for most of the analyzed species of the genus *Pinus*, the most abundant compounds are α-pinene (in 56 taxa, approximately 139 references), β-pinene (in 31 taxa, approximately 154 references), δ-3-carene (in 10 taxa, approximately 64 references), β-caryophyllene (in 23 taxa, approximately 196 references), limonene/β-phellandrene (in 15 taxa, approximately 112 references), and germacrene D (in 12 taxa, approximately 147 references).

For some taxa, the leading compounds in the terpene profile also include α-cedrol, bornyl acetate, caryophyllene-oxide, α-phellandrene, trimethylbicyclo [3.1.1]hept-2-ene, 2H-benzocyclohepten-2-one, phenylethyl butyrate, 4-epi-isocembrol, β-thujene, and thunbergol. Other compounds with an abundance greater than 15% include methyl chavicol (=estragole), geranylene, myrcene, γ-muurolene, sabinene, and abieta-7,13-diene.

The terpene profile depends on the type of essential oil extraction. The results using the headspace technique have already been proven to differ from those of other types of distillation in many species, including the needles of *P. armandii* from China (Yunnan) [26], where α-pinene, limonene, β-caryophyllene, and β-pinene are more abundant when using the headspace technique compared to hydrodistillation (Figure 1).

HD (Clevenger)

HS-SPME

Figure 1. Differences in the terpene profiles of *P. armandii* based on terpene isolation techniques. Data from Yang et al. [26] were used for the graphical representation.

In the needles of *P. halepensis* originating from Tunisia, there are drastic differences in terpene profiles when using HD and MAE (Microwave-Assisted Extraction) techniques [71] (Figure 2), which is also the case with *P. nigra* ssp. *mauritanica* [125] (Figure 3).

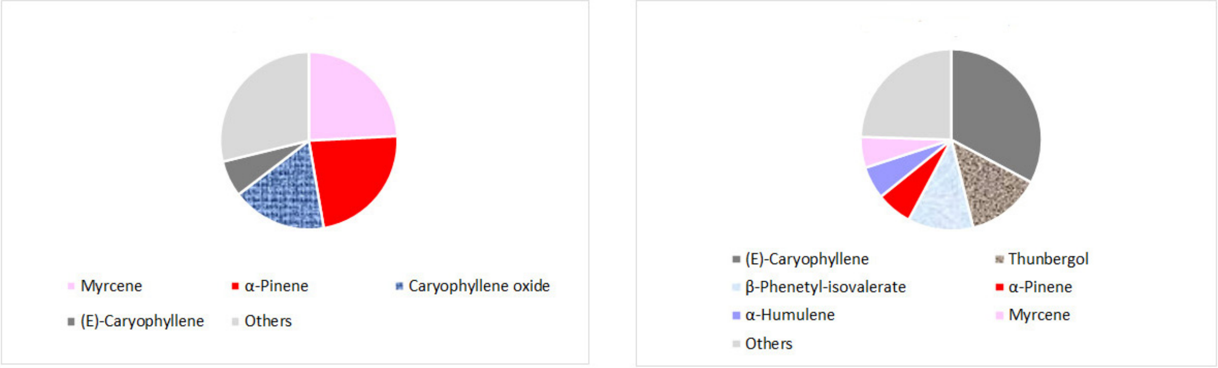
HD **MAE**

Figure 2. Differences in the terpene profiles of *P. halepensis* based on terpene isolation techniques. Data from Hamzaoui et al. [71] were used for the graphical representation.

HD (Clevenger) **HS-SPME**

Figure 3. Differences in the terpene profiles of *P. nigra* ssp. *mauritanica* based on terpene isolation techniques. Data from Adjaoud et al. [125] were used for the graphical representation.

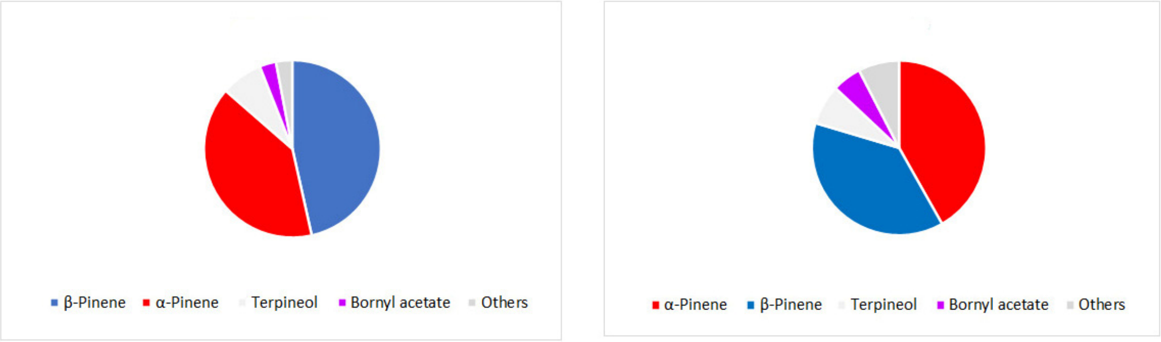
Additionally, there are noticeable differences in the terpene profiles of young (new) and last year's (one-year-old) needles of *P. halepensis* [69]. The highest content in young needles is myrcene (24.2%), while in last year's needles, β-caryophyllene predominates (32.8%) (Figure 4).



YOUNG NEEDLES **ONE-YEAR-OLD NEEDLES**

Figure 4. Differences in the terpene profiles of *P. halepensis* based on needle age. Data from Khouja et al. [69] were used for the graphical representation.

The differences between one-year-old and two-year-old needles of *P. nigra* are more evident in the abundance of the main components than in their profile [122]. In one-year-old needles, β-pinene is the most abundant, while in two-year-old needles, α-pinene predominates (Figure 5).



ONE-YEAR-OLD NEEDLES **TWO-YEAR-OLD NEEDLES**

Figure 5. Differences in the terpene profiles of *P. nigra* based on needle age. Data from Supuka et al. [122] were used for the graphical representation.

The content and composition of the main components also depend on the locations where the needles were sampled. In *P. pinea* [142], there are noticeable differences in the terpene profiles across locations, except for limonene, which is always dominant (Figure 6). A similar pattern is observed in the terpene profiles of two *P. pinea* locations in Libya [73], where α-pinene is dominant, but the composition of other abundant components differs significantly between the two locations (Figure 7).

Tunisia, Bečetur **Tunisia, Dar Čičou**

Tunisia, Džebel Essma **Tunisia, Džebel Hadada**

Tunisia, Oued El Bir **Tunisia, Zouara**

Figure 6. Differences in the terpene profiles of *P. pinea*—locations in Tunisia. Data from Nasri et al. [142] were used for the graphical representation.

Libiya, Werdama **Libiya, Al-Mansura**

Figure 7. Differences in the terpene profiles of *P. pinea*—locations in Libya. Data from Elkady et al. [73] were used for the graphical representation.

In *P. halepensis* from Libya, there are very significant differences in the profiles of two locations [73] (Figure 8).

Libya, Sidi Alhamry **Libya, Alaslab**

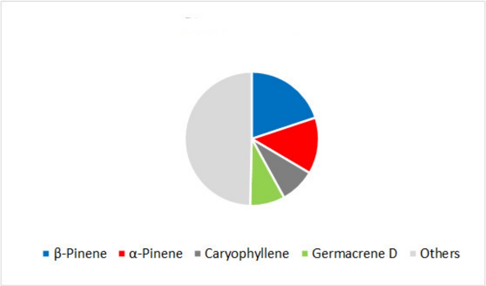
Figure 8. Differences in the terpene profiles of *P. halepensis*—locations in Libya. Data from Elkady et al. [73] were used for the graphical representation.

In *Pinus nigra* ssp. *laricio* from Corsica, there are also significant differences between locations [120] (Figure 9).

Corsica, Group I **Corsica, Group II**

Figure 9. Differences in the terpene profiles of *P. nigra* ssp. *laricio*—populations in Corsica. Data from Rezzi et al. [120] were used for the graphical representation.

The influence of altitude on the composition of terpene profiles has been confirmed in the needles of *P. halepensis* from Libya [63] (Figure 10). As the altitude increases (Level II and Level III), the dominance of β-pinene and α-pinene is replaced by caryophyllene.



Lybia, Level I

Lybia, Level II

Lybia, Level III

Figure 10. Differences in the terpene profiles of *P. halepensis* based on altitude. Data from Mohareb et al. [63] were used for the graphical representation.

The content and composition of the main terpene components depend on the season in which the needles were sampled. For example, in *Pinus densiflora* (Republic of Korea, artificial plantation) [16], 3-carene dominates in the needles sampled in February (28.6%) and then decreases to 1%–2% during the growing season (Figure 11). In contrast, germacrene D and β-pinene are present only during the growing season. β-pinene is most abundant in October (25.8%), while the abundance of germacrene D does not differ significantly between April and October (10.5%–11.9%).

February

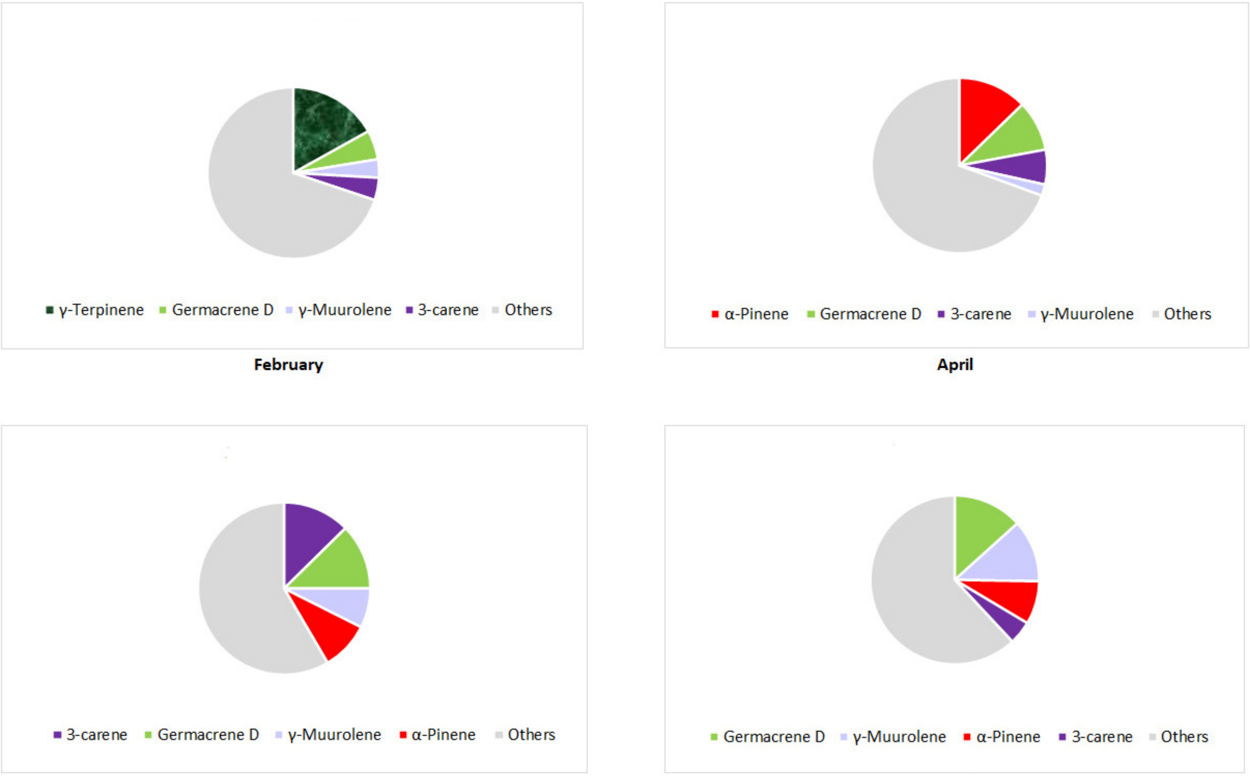
April

July

October

Figure 11. Differences in the terpene profiles of *P. densiflora* by season. Data from Kim et al. [9] were used for the graphical representation.

There are much greater differences in the composition of the main components by season, for example, in *P. koraiensis* (Figure 12) [16]. In February, γ-terpinene dominates (16.9%), in spring, α-pinene is the most abundant (12.7%), while in summer and autumn, germacrene D along with 3-carene (July) and γ-muurolene (October) are most abundant.



February

April

July

October

Figure 12. Differences in the terpene profiles of *P. koraiensis* across seasons. Graphical representation based on data from Kim et al. [9].

It appears that these differences cannot be observed at the population level, as is the case with *P. nigra* [116], where the same components are always dominant, in the same order of abundance: α-pinene, β-pinene, germacrene D, and β-caryophyllene. Their abundance is somewhat higher in spring and summer than in autumn and winter (Figure 13). α-Pinene, germacrene D, and β-caryophyllene are most abundant in spring, while β-pinene is highest in summer. Germacrene D is lowest in summer, β-pinene and β-caryophyllene are lowest in autumn, while α-pinene consistently decreases during the growing season and is lowest in winter (Figure 14).

Spring

Summer

Figure 13. *Cont.*

Autumn **Winter**

Figure 13. Differences in the terpene profiles of *P. nigra* by season—population level. Data from Sezik et al. [116] were used for the graphical representation.

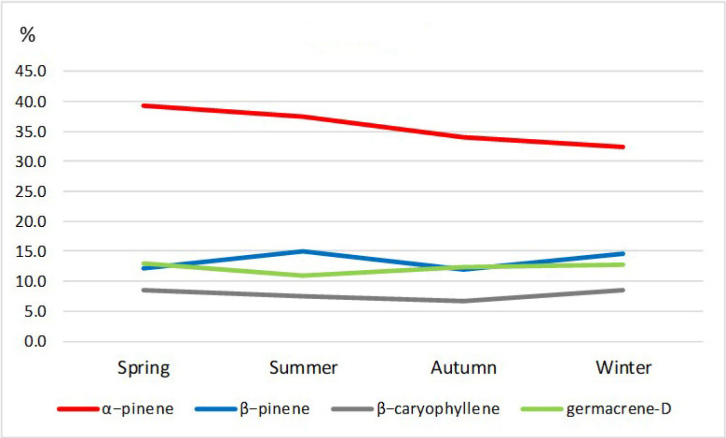


Figure 14. Dynamics of the increase and decrease in the abundance of four terpenes in *P. nigra* throughout the year. Data from Sezik et al. [116] were used for the calculations and graphical representation.

There is a degree of similarity in the terpene profiles of different varieties of *P. caribaea* [45] (Figure 15).

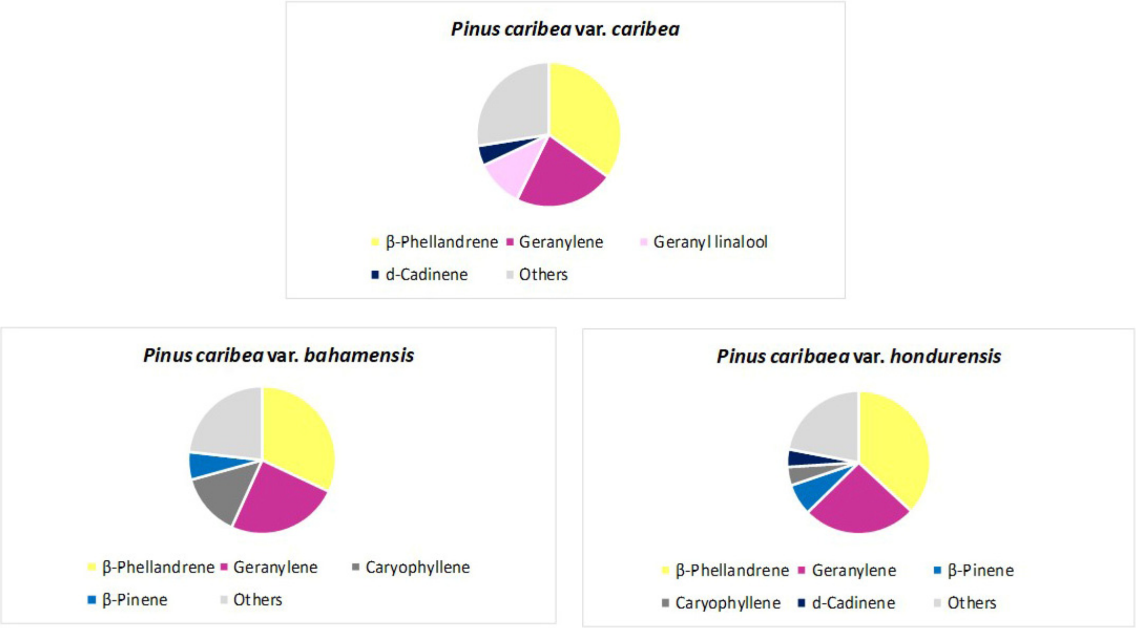


Figure 15. Similarities and differences in the terpene profiles of three *P. caribaea* varieties [45].

The terpene profile of *P. mugo* is more similar to that of the var. *pumilio* than that of var. *prostrata* [23] (Figure 16).

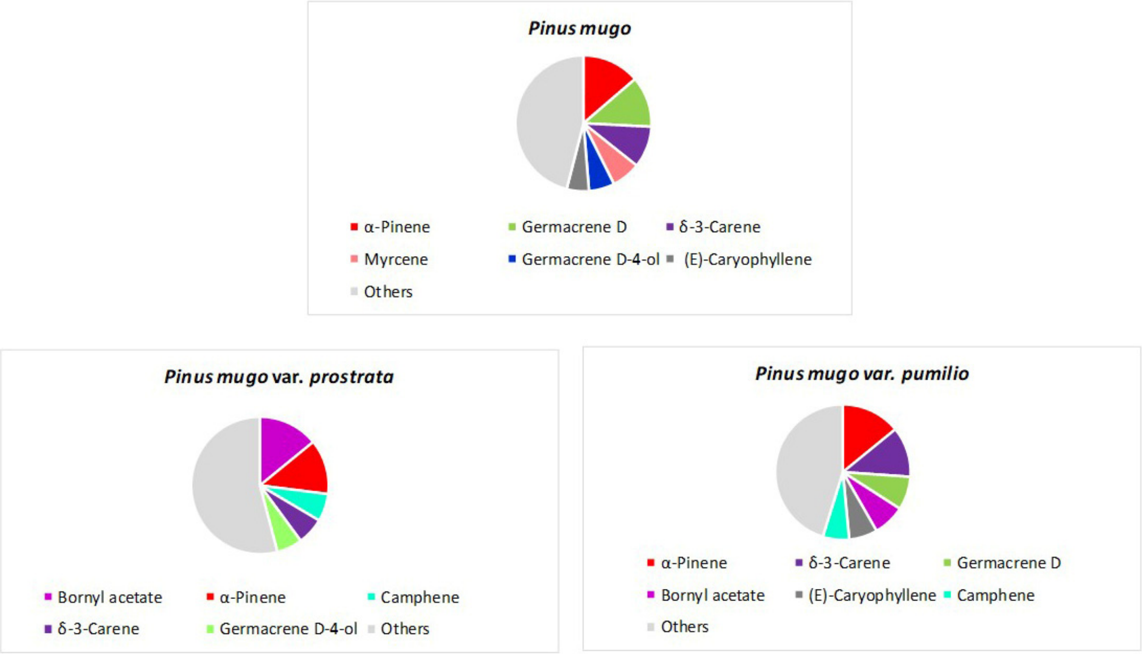


Figure 16. Similarities and differences in the terpene profiles of *P. mugo* and its two varieties according to the data from Ioannou et al. [23].

4. Conclusions

This review paper covers data for 104 taxa of the genus *Pinus* (66 species, 20 subspecies, 17 varieties, and 1 form), with 78 taxa in the subgenus *Pinus* and 26 taxa in the subgenus *Strobus*. The presented taxonomic classification, distribution, and abundance of the main components of EOs also apply to 15 endemic and 3 relict pine species. The most abundant compounds are α-pinene (56 taxa), β-pinene (31 taxa), δ-3-carene (10 taxa), β-caryophyllene (23 taxa), limonene/β-phellandrene (15 taxa), and germacrene D (12 taxa). In addition to these, the terpene profile is occasionally dominated by α-cedrol, bornyl acetate, caryophyllene oxide, α-phellandrene, trimethylbicyclo [3.1.1]hept-2-ene, 2H-benzocyclohepten-2-one, phenylethyl butyrate, 4-epi-isocembrol, β-thujene, and thunbergol. Furthermore, methyl chavicol (=estragole), geranylene, myrcene, γ-muurolene, sabinene, and abieta-7,13-diene also show significant abundance (>15%).

The terpene profile depends on the type of essential oil extraction (in *P. armandii*), techniques of chromatography (*P. halepensis* and *P. nigra* ssp. *mauritanica*), the needles' age (*P. nigra*), location (*P. pinea*), altitude (*P. halepensis*), season (*P. densiflora*, *P. koraiensis*), and variety (*P. caribaea*, *P. mugo*). Further investigations applied in a higher number of species could approve these statements.

Finally, we can conclude that studying a larger number of biological activities could help expand our understanding of the medicinal benefits and applications of pine essential oils in medicine and pharmacology, as well as their role in plant protection and ecological activities (such as biocides).

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/f16040623/s1, Table S1: Latin and English names and synonyms, taxonomy and natural area of examined *Pinus* taxa; Table S2: The main terpene compounds in the needles of *Pinus* taxa in correlation with type of chromatographic column, type of terpenes isolation, origin of trees, and season of needle connection. References [166–168] are cited in Supplementary Materials.

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