FURTHER FUROCLERODANES FROM TEUCRIUM GENUS

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Abstract - The review updates the results reported during the last four years on the chemistry of these diterpenoids.

The interest for the furoclerodane diterpenoids (neo-clerodane skeleton) occurring in Teucrium species (family Labiatae) seems to meet no pause. The main reason is their powerful insect antifeedant activity. The previous reviews are updated by the present report, concerning 19 new taxa (plus five reinvestigated) and 51 new natural products; in particular, some substances with transposed skeletons are attractive for the researcher. This genus is surely one of the richest sources of neo-clerodanes.

The new taxa and products will be reported mostly in chronological order, and for the new products the numbering used in the previous reviews will be continued.

A paper escaped the previous review and published in 1990 referred to Teucrium grisebachii growing in Argentina: two furoclerodanes were isolated: one is the new triacetylteumassilin (156), the other is 6-acetylteucjaponin B (20), claimed as a new derivative but in the fact previously known.

A reinvestigation of the extract of Teucrium polium collected in Armenia (subspecies not indicated) yielded two products named tepolin A and tepolin B; they were assigned the structures (157) and (158), whose more peculiar detail is the occurrence of not lactonized 12-OH and 9α-COOH.

Another reinvestigation was concerned with Teucrium pernyi. From this species, growing in South-East China, the teupernins A (115), B (105) and C (106) had been described by two chinese groups. In a second time, a product indicated as teupernin D (155) had been isolated. Another product was then extracted and also named teupernin D, but its structure, reported as 159, is quite different and shows an unprecedented carbomethoxy group on C-4. The nomenclature of these two products should be revised. In the same species also the known teucvidin (91), teuflin (93), montanin D (46) and teuscorodonin (68) were found.

Structurally more interesting are the nine furoclerodanes isolated from Teucrium brevifolium, growing in small areas of Greece and collected in the island of Karpathos. Teubrevin A (160) has a rearranged skeleton arising from cleavage of the C-5/C-10 bond and formation of a C-1/C-6 linkage; teubrevin B (161) is its 8β-OH derivative.

Dedicated to Professor Koji Nakanishi on the occasion of his 75th birthday.
Teubrevin C (162), obtained\textsuperscript{12} for the first time from a natural source, was found to be identical to the diacetyl derivative of teulepicin (44).\textsuperscript{2} Teubrevin D (163) is the 8β,10β-dihydroxy derivative of teubrevin C. The neo-clerodane absolute stereochemistry of 163 was established from its CD curve. The structure of teubrevin E was elucidated by careful NMR investigations,\textsuperscript{12} proving the occurrence of an eight-membered carbocycle, formed by carbon atoms 1-5, 19 and 9-10, fused at C-4, C-5 with a γ-lactone arising from C-18 and C-6, and bearing a spiro 20,12-γ-lactone; moreover, the carbon atoms 7, 8 and 17 have been lost. The quite novel structure of teubrevin E is therefore represented by 164. Extremely similar is the structure\textsuperscript{12} of teubrevin F (165): it differs from 164 only for the inverted relative configuration at C-9. The two products are therefore epimers at C-9. Also teubrevin G and teubrevin H are epimers at C-9: instead of the lactone carbonyl C-6, they bear a CH$_2$-CO-CH$_3$ side chain formed by C-7, C-8 and C-17, whereas the atoms C-18, C-4, C-5 and C-6 form a furan ring. The structures of these two unusual products are shown\textsuperscript{12} by 166 resp. 167. Even more complex is the structure\textsuperscript{12} of teubrevin I, solved by X-Ray diffraction analysis: it has not only the eight-membered carbocyclic ring, but also a substituted tetrahydropyrano-1,2-dione moiety, and a five-membered dioxa ring involving C-4, C-18, O, C-8, O. This fascinating molecular structure is represented\textsuperscript{12} by 168. Teubrevins E-I (164-168, respectively) are the first reported compounds having a rearranged neo-clerodane skeleton with an eight-membered ring. Their biogenesis may be rationalized by a mechanistic pathway\textsuperscript{12} starting from teubrevin D. The absolute stereochemistry of these products is the one depicted in their formulae, because all of them could be derived biogenetically from the proved neo-clerodane structure (163) of teubrevin D.

Eight furoclerodanes (teucrolivins A-H, 109-114, 118-119) had been reported previously\textsuperscript{13-15} from Teucrium oliverianum growing in Saudi Arabia. The plant was reinvestigated by another group: teucrolivins A-C (109, 110, 111) were found again, together with five new compounds (teucrolins A-E) and three diterpene artifacts.\textsuperscript{16} Teucrolin A (169) differs from teucrolivin A (109) by having an axial 3α-OAc group replacing the ketone carbonyl. Teucrolin B (170) has a free 6α-OH and a 12-OH group of unknown configuration. In teucrolin C (171) this hydroxy group is oxidized to ketone, whereas the 6α-OH is acetylated to 6α-OAc. Teucrolin D (172) is a tetranor-diterpene quite similar to teucrolivin F (112), from which differs only by the occurrence of 3α-OAc instead of the keto group. In teucrolin E (173) the 4,18-epoxide system is replaced by a 4-CH$_2$OH and an unusual oxygen bridge between C-4 and C-10, whose stereochemistry was not proved.\textsuperscript{17} The artifacts were: 12-O-methyl-teucrolin A (174), 12-O-methyl-teucrolivin A (175) and 12-O-ethyl-teucrolin A (176) arising from alkylation of 169 and 109 by MeOH used for the extraction and by EtOH contained in CHCl$_3$ used for partitioning. This easy alkylation at the tertiary hydroxy group on C-12 is very unusual.

A reinvestigation of Teucrium lamiifolium allowed the isolation\textsuperscript{18} of a glucoside, teulamioside (177); the glucose moiety is bonded to the 18β hydroxy group of an 18,19 hemiacetale system. Teulamioside is the second glucoside until now isolated from Teucrium species out of the 206 neo-clerodanes occurring in this genus. The already known\textsuperscript{2} teuspinin (62) and montanin E (51) were also detected. Teucrium trifidum, growing in southern Africa, yielded\textsuperscript{19} two neo-clerodanes. The first was the already known\textsuperscript{3} 4α,18-
epoxytafricanin A (116), whereas the second was new and assigned the name teutrifidin and the structure (178). It differs from 116 only for the occurrence of a 7β-OH group.

Five neo-clerodanes were extracted from Teucrium polium ssp. aurasianum growing in Algeria. One was the previously described teumicropodin (142). Another is 3-deacetylteumicropodin (179), whereas the three remaining are products of progressive deacetylation of teupyreinidin (29), i.e. 3,20-bis-deacetylteupyreinidin (180), 6,20-bisdeacetylteupyreinidin (31) and 3,6,20-trisdeacetylteupyreinidin (181). The authors claimed that the product formerly reported as 6,20-bisdeacetylteupyreinidin (31) was on the contrary the 3,20-bisdeacetyl derivative (180).

Two species originating from Australia were studied: Teucrium racemosum yielded the chlorine-containing teuracemin (182), together with the known teutrifidin (178), 4α,18-epoxytafricanin A (116) and 20-oxoteuflavin (117). Teucrium corymbosum gave the 19-nor-neo-clerodane teucorymin (183) and the three known 19-acetylnaphthalin (22), teucjaponin A (18) and 6-acetylteucjaponin B (20). Four new neo-clerodanes were found in Teucrium yemense, collected in Saudi Arabia. The first is 6β-acetyl-3β-hydroxyteucroxylepin (184), with the rare δ-lactone between C-20 and C-19. Teucryemin (185) and 19-acetylteucryemin (186) have the usual γ-lactone system between C-20 and C-12, with 12S configuration. On the contrary, teucryeminone (187) has the infrequent 12R configuration.

Very fascinating is the structure of teubetonin (188), a rearranged homo-neo-clerodane derivative isolated from Teucrium betonicum growing in the island of Madera (Portugal). Indeed, teubetonin has a C-18/C-19 chain bonded to C-4 and an unprecedented CH₂OH group on C-7. An hypothesis on its biogenesis proposes an aldol condensation of a 6-keto precursor and formaldehyde. The same species contained also six already known furoclerodanes: 19-acetylnaphthalin (22), teucvin (89), teucrin H₂ (76), teucrin E (72), 6β-hydroxyteuscordin (80) and 6α-hydroxyteuscordin (79).

Four new products have been isolated from Teucrium alyssifolium growing in Turkey. They are alysin A (189), alysin B (190), 3-deacetylassin B (191) and alysin C (192). All the products show an unusual linkage between C-8 and C-16 (indicated in the original paper as C-14): this rearrangement constitutes a new variant of the neo-clerodane skeleton.

Recently, two more furoclerodanes were extracted from the above species: alysin D (193) and alysin E (194). Both products show the unusual bond between C-8 and C-16, characteristic of this species.

Extraction of Teucrium chamaedrys ssp. syspirense, also growing in Turkey, yielded the 19-norditerpene syspirensin A (195) and the diterpene syspirensin B (196). In the latter product is remarkable the transformation of the furan ring into a chain with primary hydroxy groups on C-15 and C-16, and a C-13/C-14 double bond.

Also Teucrium sandrasicum occurs in Turkey. From its aerial parts three new neo-clerodanes were isolated: sandrasin A (197), 6-deacetylsandrasin A (198) and sandrasin B (199). The first and second products show a 10β-OH group, whereas sandrasin B has the epoxy group opened to give 4α-OH and 4β-CH₂OH, the hydroxy group on C-10 having 10α-OH configuration. Moreover, the three substances are reported to have the rare 12R configuration.

The investigation of Teucrium sandrasicum was resumed recently by another group: six new neo-clerodane diterpenoids were isolated, teusandrin A to teusandrin F. It is probable that teusandrin A and teusandrin B are identical with sandrasin A and 6-deacetylsandrasin A, previously reported: however, the
configuration at C-12 of teusandrin A and B is certainly 12S, and not 12R as suggested for sandrasin A and 6-deacetylsandrasin A. Therefore the structures of these four products are worthy to be confirmed.

Teusandrin C (200) has an oxetane ring in which the C-4, C-5 and C-19 carbons are involved, and a 4β-CH₂OH group. Teusandrin D (201) is identical apart from the lack of the 8β-OH group. Teusandrin E (202) shows a 4β,10β ether bridge, forming an oxetane ring that involves C-4, C-5 and C-10. Teusandrin F (203) differs from teusandrin E only by having an equatorial 6α-OH hydroxy group instead of the keto group. From the same species also two known diterpenoids were isolated: teucjaponin B (19) and 6-acetylteucjaponin B (20).

A new derivative, teucriasiatin (204) occurs in Teucrium asiaticum, collected in the island of Majorca, Spain: it shows an hemiacetalic 20α-OH group. It can be remembered that a previous paper had reported the occurrence of only the two already known neo-clerodanes auropolin (25) and teufolin (93). Quite recently, another product was isolated, teucrasiolide: its is remarkable for the occurrence of an until now unprecedented C-11/C-12 trans olefinic double bond, of a 7α-(20α-O-acetyl)hemiacetal bridge, and for the change of the furan ring into an α,β-unsaturated γ-lactone involving the C-13, C-14, C-15 and C-16 carbons and having an acetoxyl substituent at its γ-position. The product occurs as a mixture of epimers at C-15; owing to the poor stability of the crude product, it was isolated as its triacetyl derivative (205), the unstable precursor having no acetoxyl substituent.

Three neo-clerodanes were isolated from Teucrium nudicaule, collected in Northern Chile: two of them are the already described triacetylteumassilin (156) and 6-acetylteucjaponin B (20). The third is the new 12-epi-teupyreinin (206) showing the usual 12S configuration.

At the end, the stereostructure of montanin E (51) was confirmed by X-Ray diffraction.

Several other Teucrium taxa were examined during the last years, and many already known neo-clerodanes were isolated. Teucrium montanum ssp. pannonicum yielded auropolin (25) and montanin H (135). Teucrium alpestre (from Crete island, Greece) contained teupyrone (32), 3-acetylteumicropin (140), teumicropin (139) and 3-O-deacetylteupyrenone (141). Teucrium cuneifolium (from Crete island, Greece) gave only 3-O-deacetylteupyrenone (141). Teucrium divaricatum ssp. villosum (from Karpathos island, Greece) yielded teufolin (93), teuschordinone (82), teuflidin (95), montanin D (46), teucrin A (97), dihydroteugin (78), 6β-hydroxyteucriolin (80) and teugin (77). Teucrin A (97) was extracted also from a sample of the same species harvested near Istanbul. From Teucrium flavum ssp. hellenicum (from Greece) teucvidin (91), 12-epi-teucvidin (145) and teufuin (93) were isolated. Teucrium ravas-martinezii (from Spain) yielded 19-acetylgnaphalin (22) and 19-acetylteulepicin (45). Teucrium divaricatum ssp. divaricatum (from Greece) contained only teucrin A (97). Eventually, Teucrium haenseleri (from Portugal) was shown to contain 19-acetylgnaphalin (22), eriocephalin (13), isoeriocephalin (16) and 20-deacetyleriocephalin (14).

Also in these last years several interesting papers appeared concerning researches aiming at modifying the structures (rings and functional groups) of natural Teucrium furoclerodanes, on the purpose of obtaining semisynthetic, non-natural derivatives with increased antifeedant activity. The substrates used for these researches were eriocephalin (13), montanin C (8), teucrin P1 (24), teupolin III (50), 19-acetylgnaphalin (22), teucjaponin A (18), teucjaponin B (19), teucroxylepin (129), teucvidin (91), capitatin (5) and montanin E (51).
As far as we know, no researches either for total synthesis or for the biogenetic mechanism were attempted on the furoclerodanes from *Teucrium*.

The antifeedant activity of natural furoclerodanes and their derivatives was reported in some other papers.\(^{33,34,39,41,42,43}\)

Another field of interest arose from the casual discovery of the toxicity of teucrin A. The extract of *Teucrium chamaedrys*, its furoclerodane fraction, and teucrin A were found to cause irreversible liver necrosis on mice:\(^{44}\) several cases of heavy intoxication were reported in men when herbalist preparations of this species were used for weight control.\(^{45}\) It is possible that all the furoclerodanes from *Teucrium* are toxic.\(^{36}\)

Table 1 report the structures of the 51 new neo-clerodanes isolated since the third review.\(^3\) The symbol F = β-furyl.

Table 2 lists the 108 *Teucrium* taxa (species, subspecies, chemotypes) investigated till now, in alphabetical order, and the neo-clerodanes isolated from each taxon, indicated by the numbering adopted in the two previous\(^2,3\) and in the present review. The names of the taxa are those indicated in the original papers. However, some changes were reported\(^46\) for the systematic taxonomy of *Teucrium* species in recent years: *T. belion*\(^3\) and *T. polium* ssp. *belion*\(^3\) are now called *T. puechiae*; *T. polium* ssp. *pilosum*\(^2\) is now *T. decaisnei*; *T. scorodonia* ssp. *euganeum*\(^2\) is now *T. siculum*; *T. polium* ssp. *expansum*\(^3\) is *T. expansum*; *T. polium* ssp. *vincentinum*\(^3\) is *T. vincentinum*; *T. polium* ssp. *capitatum*\(^2\) is *T. capitatum*.
Table 1

\[
F = \beta \text{-furyl}
\]

[156] 6,12,19-triacetylteumassilin

\[R = \text{OH}
\]

\[T. \text{grisebachii}^4\]

\[C_{26}H_{36}O_8\]

[157] tefolin A

\[R = \text{OH}
\]

\[T. \text{polium}^6\]

\[C_{29}H_{26}O_7\]

[158] tefolin B

\[R = \text{H}
\]

\[T. \text{polium}^6\]

\[C_{29}H_{26}O_6\]

[159] "teupernin D"

\[T. \text{pernyi}^{10}\]

\[C_{21}H_{26}O_7\]

[160] teubrevin A

\[R = \text{H}
\]

\[T. \text{brevifolium}^{11}\]

\[C_{22}H_{24}O_8\]

[161] teubrevin B

\[R = \text{OH}
\]

\[T. \text{brevifolium}^{11}\]

\[C_{22}H_{24}O_9\]
[162] teubrevin C \( R = H \) 
\( T. \) brevifolium\(^{12}\) 
C\(_{24}\)H\(_{28}\)O\(_9\)

[163] teubrevin D \( R = \text{OH} \) 
\( T. \) brevifolium\(^{12}\) 
C\(_{24}\)H\(_{28}\)O\(_{11}\)

[164] teubrevin E 
\( T. \) brevifolium\(^{12}\) 
C\(_{19}\)H\(_{18}\)O\(_8\)

[165] teubrevin F 
\( T. \) brevifolium\(^{12}\) 
C\(_{19}\)H\(_{18}\)O\(_8\)

[166] teubrevin G 
\( T. \) brevifolium\(^{12}\) 
C\(_{22}\)H\(_{22}\)O\(_8\)

[167] teubrevin H 
\( T. \) brevifolium\(^{12}\) 
C\(_{22}\)H\(_{22}\)O\(_8\)
[168] teubrevin I
  $T.\text{brevifolium}^{12}$
  $\text{C}_{22}\text{H}_{24}\text{O}_{9}$

[169] teucrolin A
  $R = H$
  $\text{C}_{26}\text{H}_{34}\text{O}_{10}$
  $T.\text{oliverianum}^{16}$

[174] 12-O-methylteucrolin A
  $R = \text{Me}$
  $\text{C}_{27}\text{H}_{36}\text{O}_{10}$
  $T.\text{oliverianum}^{16}$

[176] 12-O-ethylteucrolin A
  $R = \text{Et}$
  $\text{C}_{28}\text{H}_{38}\text{O}_{10}$
  $T.\text{oliverianum}^{16}$

[170] teucrolin B
  $R_1 = H, \ R_2 = H, \text{OH}$
  $\text{C}_{24}\text{H}_{34}\text{O}_{8}$
  $T.\text{oliverianum}^{16}$

[171] teucrolin C
  $R_1 = \text{Ac}, \ R_2 = \text{O}$
  $\text{C}_{26}\text{H}_{34}\text{O}_{9}$
  $T.\text{oliverianum}^{16}$

[172] teucrolin D
  $\text{C}_{22}\text{H}_{30}\text{O}_{9}$
  $T.\text{oliverianum}^{16}$

[173] teucrolin E
  $T.\text{oliverianum}^{16, 17}$
  $\text{C}_{22}\text{H}_{30}\text{O}_{8}$
[175] 12-O-methylteucrolivin A  
*T. oliverianum*\(^{16, 17}\)  
\[\text{C}_{25}\text{H}_{32}\text{O}_9\]

[177] teulamioside  
*T. lamiiifolium*\(^{18}\)  
Gl = glucose  
\[\text{C}_{28}\text{H}_{38}\text{O}_{12}\]

[178] teutrifidin  
*T. trifidum*\(^{19}\)  
\[\text{C}_{22}\text{H}_{24}\text{O}_9\]

[179] 3-deacetylteumicropodin  
*T. polium ssp. aurasianum*\(^{20}\)  
\[\text{C}_{22}\text{H}_{28}\text{O}_8\]

[180] 3,20-bis-deacetylteupreinidin \(R = \text{Ac}\)  
*T. polium ssp. aurasianum*\(^{20}\)  
\[\text{C}_{24}\text{H}_{32}\text{O}_9\]

[181] 3,6,20-tris-deacetylteupreinidin \(R = \text{H}\)  
*T. polium ssp. aurasianum*\(^{20}\)  
\[\text{C}_{22}\text{H}_{30}\text{O}_8\]
[182] teucacemin
T. racemosum\textsuperscript{21}
C\textsubscript{22}H\textsubscript{25}O\textsubscript{9}Cl

[183] teucorymbin
T. corymbosum\textsuperscript{22}
C\textsubscript{21}H\textsubscript{24}O\textsubscript{6}

[184] 6β-acetyl-3β-hydroxyteucroxylep
T. yemense\textsuperscript{23}
C\textsubscript{22}H\textsubscript{28}O\textsubscript{8}

[185] teucryemin
R = H
T. yemense\textsuperscript{23}
C\textsubscript{22}H\textsubscript{28}O\textsubscript{8}

[186] 19-acetylteucryemin
R = Ac
T. yemense\textsuperscript{23}
C\textsubscript{24}H\textsubscript{30}O\textsubscript{9}

[187] teucryeminone
T. yemense\textsuperscript{23}
C\textsubscript{24}H\textsubscript{28}O\textsubscript{9}
[188] teubetonin
  T. betonicum$^{24}$
  $\text{C}_{21}\text{H}_{24}\text{O}_{7}$

[189] alysin A  \( R_1 = \text{OH} \quad R_2 = \text{Ac} \)
  T. alyssifolium$^{25}$
  $\text{C}_{24}\text{H}_{30}\text{O}_{9}$

[192] alysin C  \( R_1 = R_2 = \text{H} \)
  T. alyssifolium$^{25}$
  $\text{C}_{22}\text{H}_{28}\text{O}_{7}$

[190] alysin B  \( R = \text{Ac} \)
  T. alyssifolium$^{25}$
  $\text{C}_{26}\text{H}_{34}\text{O}_{11}$

[191] 3-deacetylalysin B  \( R = \text{H} \)
  T. alyssifolium$^{25}$
  $\text{C}_{24}\text{H}_{32}\text{O}_{10}$

[193] alysin D
  T. alyssifolium$^{26}$
  $\text{C}_{22}\text{H}_{28}\text{O}_{7}$

[194] alysin E
  T. alyssifolium$^{26}$
  $\text{C}_{24}\text{H}_{32}\text{O}_{9}$
[195] syspirensis A
*T. chamaedrys* ssp. *syspirensis*\(^{27}\)

\[
\text{C}_{19}\text{H}_{24}\text{O}_{6}
\]

[196] syspirensis B
*T. chamaedris* ssp. *syspirensis*\(^{27}\)

\[
\text{C}_{20}\text{H}_{36}\text{O}_{5}
\]

[197] sandrasin A
12R  \(R = \text{Ac}\)
teusandrin A 12S
*T. sandrasicum*\(^{28,29}\)

\[
\text{C}_{24}\text{H}_{30}\text{O}_{10}
\]

[198] 6-deacetylsandrasin A 12R  \(R = \text{H}\)
teusandrin B 12S
*T. sandrasicum*\(^{28,29}\)

\[
\text{C}_{22}\text{H}_{28}\text{O}_{9}
\]

[199] sandrasin B
*T. sandrasicum*\(^{28}\)

\[
\text{C}_{20}\text{H}_{26}\text{O}_{8}
\]
[200] teusandrin C
  \( T. \text{sandrasicum}\) \(^{29}\)
  \( R = \text{OH} \)
  \( C_{20}H_{26}O_{8} \)

[201] teusandrin D
  \( T. \text{sandrasicum}\) \(^{29}\)
  \( R = \text{H} \)
  \( C_{20}H_{26}O_{7} \)

[202] teusandrin E
  \( T. \text{sandrasicum}\) \(^{29}\)
  \( R = \text{O} \)
  \( C_{20}H_{24}O_{7} \)

[203] teusandrin F
  \( T. \text{sandrasicum}\) \(^{29}\)
  \( R = \alpha-\text{OH}, \beta-\text{H} \)
  \( C_{20}H_{26}O_{7} \)

[204] teucriasatin
  \( T. \text{asiaticum}\) \(^{30}\)
  \( C_{22}H_{28}O_{7} \)

[205] teucriasolide
  \( T. \text{asiaticum}\) \(^{32}\)
  \( C_{26}H_{30}O_{11} \)
Table 2

- **T. abutiloides** 8, 131, 132, 133
- **T. africanum** 52, 53
- **T. algarbiense** ----
- **T. alpestre** 32, 139, 140, 141
- **T. alyssifolium** 189, 190, 191, 192, 193, 194
- **T. apollinis** ----
- **T. asiaticum** 25, 93, 204, 205
- **T. barbeyanum** 74, 75, 97
- **T. belion** 13, 22, 97
- **T. betonicum** 22, 72, 76, 79, 80, 89, 188
- **T. bicolor** 8, 9, 76, 89, 90, 104, 121
- **T. bidentatum** 76, 82, 84, 93, 105
- **T. botrys** 46, 56, 61, 64, 80, 91
- **T. brevifolium** 160, 161, 162, 163, 164, 165, 166, 167, 168
- **T. buxifolium** 22, 45
- **T. canadense** 9, 54, 89, 91, 93, 131, 148, 149
- **T. carolipau** 4, 22
- **T. chamaedrys** (Bulgarian chemotype) 64, 72, 76, 78, 79, 93, 97
- **T. chamaedrys** (Italian chemotype) 74, 75, 89, 91, 93, 97
- **T. chamaedrys** (Moldavian chemotype) 72, 74, 75, 78, 97
- **T. chamaedrys** (Spanish chemotype) 47, 72, 76, 77, 78, 88, 93, 95, 96, 97, 98
- **T. chamaedrys ssp. syspirense** 195, 196
- **T. cartaginense** ssp. homoticum 13, 22
- **T. compactum** ----
- **T. corymbosum** 18, 20, 22, 183
- **T. cossonii** 135, 153, 154
- **T. creticum** 19, 22, 36, 138
- **T. cubense** 89
- T. cuneifolium
- T. cyprium
- T. cyrenaicum
- T. davaeanum
- T. decipiens
- T. divaricatum ssp. canescens
- T. divaricatum ssp. divaricatum
- T. divaricatum ssp. villosum
- T. eriocephalum
- T. flavum ssp. flavum
- T. flavum ssp. glaucum
- T. flavum ssp. hellenicum
- T. fragile
- T. fruticans
- T. gnaphalodes
- T. gracile
- T. grisebachii
- T. haenseleri
- T. heterophyllum
- T. hircanicum
- T. intricatum
- T. japonicum (Japanese chemotype)
- T. japonicum (Chinese chemotype)
- T. kotschyanum
- T. lamiifolium
- T. lanigerum
- T. lepicephalum
- T. leucocladum
- T. lucidum
- T. lusitanicum
- T. marum
- T. massiliense
- T. microphyllum
- T. micropodioides
- T. montanum ssp. montanum
- T. montanum ssp. pannonicum
- T. montanum ssp. skorpilii
- T. montbretii ssp. heliotropifolium
- T. montbretii ssp. montbretii

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- T. montbretii ssp. pamphilicum
- T. nudicaule
- T. odontites
- T. oliverianum
- T. oxylepis ssp. marianum
- T. pernyi
- T. pestalozzae
- T. polium (Armenian ssp.)
- T. polium (Bulgarian ssp.)
- T. polium (German ssp.)
- T. polium (Moldavian ssp.)
- T. polium ssp. album
- T. polium ssp. aurisanum
- T. polium ssp. aureum (Sicilian chemotype)
- T. polium ssp. aureum (Spanish chemotype)
- T. polium ssp. capitatum
- T. polium ssp. expansum
- T. polium ssp. pilosum
- T. polium ssp. vincentinum
- T. pseudochamaepytis
- T. pumilum ssp. carolipau
- T. pyrenaicum
- T. racemosum
- T. rivas-martinezii
- T. rotundifolium
- T. salviastrium
- T. sandrasicum
- T. scordioides
- T. scorodion (Bulgarian chemotype)
- T. scorodion (German chemotype)
- T. scorodonia ssp. euganeum
- T. scorodonia ssp. scorodonia (Italian chemotype)
- T. scorodonia ssp. scorodonia (Spanish chemotype)
- T. spinosum
- T. subsinuosum
- T. subspinosum
- T. subspinosum

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• *T. trifidum 116, 178
• *T. turredanum 13, 16, 22
• *T. viscidum ssp. miquelianum 89, 91, 93
• *T. webbianum 92, 95, 97
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