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# Chemical Compositions of the Needle Essential Oils Obtained from *Pinus brutia* Ten. Growing in Turkey

# Türkiye'de Yetişen *Pinus brutia* Ten.'den Elde Edilen İbre Uçucu Yağlarının Kimyasal Yapıları

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**Abstract**

There are 5 *Pinus* species growing in Turkey. The Turkish Ministry of Forestry is utilizing only 3 species of this genus in order to produce timber. These species are *P. brutia, P. nigra and P. sylvestris*. However, the Ministry does not make use of the other parts of these trees. The aim of this study was to evaluate the essential constituents of *Pinus brutia* as a non-wood forestry product. The composition of essential oils were analysed by means of GC and GC/MS. Components were assessed by using MS-Library Search. The essential oils showed high ratio of monoterpenoid hydrocarbon components.

Results of the present study revealed that *P. brutia* of Turkish origin belonged to so-called "the chemotype-B". 47 constituents were characterized in the essential oils of *P. brutia*, with α- pinene, β-pinene, germacrene D, $\Delta^3$-carene as main constituents.

**Keywords:** *Pinus brutia*, Pinaceae, essential oil, pinenes, chemotaxonomy.

## Introduction

Previous studies on Turkish *Pinus* species were aimed at improving the yield of turpentine but fewer studies were conducted on the essential oil content of these species. The essential oil of *Pinus brutia* has many, high-value commercial uses such as industrial and household cleaning products, disinfectants, solvents, fragrance, cosmetic technology, medicine and aromatherapy. Essential oils of the materials obtained from eight different localities were evaluated from the view point of chemotypes suggested by some investigators (Chalchat et al., 1995a, 1995b, Gomes da Silva et al., 2001, Kubeczka et al., 1987, Macchioni et al., 2003, Orav et al., 1996, Petrakis et al., 2000, 2001, Roussis et al., 1995, Simic et al., 1996). In addition to the essential oils, terpenes can be obtained from slash and milling waste. There are numerous studies dealing with the essential oils of conifer species and especially *Pinus* species. These studies generally

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discuss variations in chemical composition of the essential oils of *Pinus* species from geographical, seasonal, genotypic and environmental points of view (Bader et al., 2000, Barnola et al., 2000, Bojovic et al., 2005, Chalchat et al., 1994, 1995, Ghosn et al., 2006, Kaundun et al., 1997, Koukos et al., 2000, Kowal et al., 1979, Llusia et al., 2006, Mita et al., 2002, Nikolic et al., 2007, Pagula et al., 2006, Papadopoulou et al., 1996, Tazerouti et al., 1993, Velasquez et al., 2000, Venskutonis et al., 2000). Previous studies on the chemical composition of the essential oils obtained from Pine needles have been published (Afsharypuor et al., 2005, Dob et al., 2005a, Dob et al., 2006, Dob et al., 2005b, Duqesnoy et al., 2007, Hamamouchi et al., 2001, Henning et al., 1994, Isidorov et al., 2003, Kartnig et al., 1998, Koukos et al., 2001, Lahlou 2003, Mumm et al., 2004, Ochocka et al., 2002, Pfeifhofer 2000, Rezzi et al., 2001, Roussis et al., 2001, Shatar et al., 1996, Tsitsimpikou et al., 2001, Vidrich et al., 1999, Vidrich et al., 1996, Yatagai et al., 1997). We aimed at studying the effects of seasonal variation on the essential oil content of *Pinus brutia* leaves in Turkey.

## Material and Methods

*Plant material and hydrodistillation*

Young needles of *P. brutia* Ten. were collected from different regions in different seasons. The collecting sites and dates are given in Table 1.

The sample was hydrodistilled using a Clevenger type apparatus. The oil was collected for 3 hours after first drop of the distillate eluted from cold finger. The oils were kept in a refrigerator until the GC-MS analysis.

**Table 1.** Sites and dates for the collection of *P. brutia* needles

| BURSA | KÜTAHYA | MUĞLA | DENİZLİ | ANTALYA | İÇEL | SİNOP | SAMSUN |
|---|---|---|---|---|---|---|---|
| 20.09.1994 | 21.09.1994 | 6.6.1994 | 8.6.1994 | 9.6.1994 | 16.6.1994 | 4.8.1994 | 25.05.1994 |
| 25.12.1994 | 27.12.1994 | 07.09.1994 | 10.09.1994 | 16.09.1994 | 14.09.1994 | 07.11.1994 | 18.08.1994 |
| 31.03.1995 | 02.04.1995 | 14.12.1994 | 16.12.1994 | 24.12.1994 | 05.12.1994 | 20.02.1995 | 17.11.1994 |
| 27.07.1995 | 28.07.1995 | 28.02.1995 | 01.03.1995 | 09.03.1995 | 15.02.1995 | 25.05.1995 | 17.02.1995 |

*GC and GC-MS Analysis*

The oils were analysed by GC- MS using a Hewlett-Packard GCD system. Thermon 600T column (50m x 0.25mmϕ with 0.25 μm film thickness) was used with Nitrogen as carrier gas. GC oven temperature was kept at $70^0$ C for 10 min. and programmed to $180^0$ C at rate of $2^0$ C/ min. then kept constant at $180^0$ C for 30min. Split ratio adjusted at 60:1. The injector and detector temperatures were $250^0$ C. MS were taken at 70eV. Mass range was from *m/z* 10-400. A library search was carried out using the NBS/NIH/EPA Library and the BASER Library of Essential Oil Constituents. Relative percentage amounts were calculated from the FID results.

## Results and Discussion

The quantitative ratios of the needle oils are calculated upon the dried materials. The results are given in Table 2. Selected two GC/MS chromatograms are shown in Figures 1 and 2.

Table 2. Sites and the volatile oil ratios according to collection dates

| BURSA | | KÜTAHYA | | MUĞLA | | DENİZLİ | |
|---|---|---|---|---|---|---|---|
| Date | % | Date | % | Date | % | Date | % |
| 20.09.1994 | 0.8 | 21.09.1994 | 0.5 | 06.06.1994 | 1.5 | 08.06.1994 | 1.5 |
| 25.12.1994 | 0.8 | 27.12.1994 | 0.5 | 07.09.1994 | 1.1 | 10.09.1994 | 0.6 |
| 31.03.1995 | 0.9 | 02.04.1995 | 0.8 | 14.12.1994 | 1.1 | 16.12.1994 | 0.5 |
| 27.07.1995 | 1.1 | 28.07.1995 | 1.3 | 28.02.1995 | 0.7 | 01.03.1995 | 0.6 |

| ANTALYA | | İÇEL | | SİNOP | | SAMSUN | |
|---|---|---|---|---|---|---|---|
| Date | % | Date | % | Date | % | Date | % |
| 09.06.1994 | 1.0 | 16.06.1994 | 1.3 | 04.08.1994 | 0.3 | 25.05.1994 | 0.4 |
| 16.09.1994 | 0.9 | 14.09.1994 | 1.1 | 07.11.1994 | 0.6 | 18.08.1994 | 0.6 |
| 24.12.1994 | 0.3 | 05.12.1994 | 0.6 | 20.02.1995 | 0.3 | 17.11.1994 | 0.5 |
| 09.03.1995 | 0.8 | 15.02.1995 | 0.6 | 25.05.1995 | 0.6 | 17.02.1995 | 0.4 |

The ratio of the yield of oils in *P. brutia* was determined between 0.3-1.3%. As shown in Tables 3-6, 47 compounds were detected in the essential oils of *P.brutia*. α-Pinene, β-pinene, germacrene D, $\Delta^3$ carene were found as the main constituents. In addition to, β-caryophyllene, myrcene, $\Delta^3$-carene, α-terpinyl acetate, α-humulene and limonene were confirmed with notable ratios in the needle oils. The highest and lowest contents of main components are shown in Table 7.

**Table 3.** Results of analysis of *P. brutia* needle oils from İçel and Antalya

| | İçel | | | | Antalya | | | |
|---|---|---|---|---|---|---|---|---|
| Collection Dates/ | 16.6.1994 | 14.9.1994 | 5.12.1994 | 15.3.1995 | 9.6.1994 | 16.9.1994 | 24.12.1994 | 9.3.1995 |
| Compounds | % | % | % | % | % | % | % | % |
| Tricyclene | 0.04 | 0.04 | 0.04 | 0.1 | 0.03 | 0.04 | 0.1 | 0.03 |
| ***α-pinene*** | **27.7** | **27.5** | **30.5** | **22.0** | **11.0** | **28.2** | **32.1** | **18.7** |
| α-thujene | Trace | Trace | trace | trace | 0.02 | trace | trace | trace |
| Camphene | 0.5 | 0.5 | 0.5 | 1.2 | 0.2 | 0.7 | 0.8 | 0.3 |
| Hexenal | 0.03 | 0.02 | 0.01 | 0.02 | 0.1 | 0.02 | 0.01 | 0.01 |
| ***β-pinene*** | **48.9** | **41.0** | **34.5** | **23.7** | **24.5** | **37.7** | **38.1** | **40.4** |
| Sabinene | 0.2 | 0.1 | 0.1 | 0.1 | 0.2 | 0.04 | 0.1 | 0.2 |
| $\Delta^3$-carene | 2.8 | 1.4 | 1.1 | 2.1 | 11.5 | 1.8 | 1.7 | 1.1 |
| Myrcene+α-phellandrene | 1.6 | 1.2 | 1 | 0.6 | 8.9 | 1.5 | 1.3 | 0.9 |
| α-terpinene | 0.04 | 0.03 | 0.02 | 0.03 | 0.09 | 0.02 | 0.03 | 0.03 |
| Limonene | 1.3 | 0.8 | 0.6 | 0.9 | 0.6 | 0.8 | 0.9 | 1.4 |
| βphellandrene+1.8cineole | 0.8 | 0.7 | 0.6 | 0.8 | 0.5 | 0.8 | 0.8 | 0.8 |
| (Z)-β-ocimene | 0.04 | 0.03 | 0.03 | 0.03 | 0.1 | 0.04 | 0.04 | 0.01 |
| γ-terpinene | 0.1 | 0.1 | 0.04 | 0.1 | 0.2 | 0.1 | 0.1 | 0.1 |
| (E)-β-ocimene | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 0.8 | 1.0 | 0.3 |
| P-cymene | 0.1 | 0.1 | 0.04 | 0.1 | 0.2 | 0.1 | 0.1 | 0.03 |
| Isoterpinolene | 0.01 | --- | --- | 0.01 | 0.1 | ----- | ----- | ----- |
| Terpinolene | 0.3 | 0.2 | 0.2 | 0.4 | 0.8 | 0.2 | 0.3 | 0.2 |
| α-copaene | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| β-bourbonene | 0.1 | 0.1 | 0.1 | 0.3 | 0.2 | 0.1 | 0.04 | 0.1 |
| Linalool | 0.2 | 0.1 | 0.01 | 0.01 | 0.3 | 0.5 | 0.5 | 0.02 |
| linalyl acetate | 0.1 | 0.1 | 0.1 | 0.1 | 1.4 | 0.1 | 1.3 | 0.1 |
| Longifolene | 0.6 | 0.3 | 0.1 | 0.3 | 0.03 | 1.2 | 0.03 | 0.2 |
| bornyl acetate | 0.2 | 0.1 | 0.1 | 0.03 | 0.1 | 0.04 | 0.03 | 0.1 |
| β-elemene | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | 0.1 | 0.04 |
| β-caryophyllene | **3.0** | **5.9** | **6.9** | **8.2** | **6.8** | **6.8** | **6.6** | **10.9** |
| Myrtenal | 0.2 | 0.1 | 0.1 | 0.1 | 0.2 | 0.2 | 0.1 | 0.1 |
| trans-pinocarveol | 0.3 | 0.2 | 0.1 | 0.2 | 0.3 | 0.3 | 0.1 | 0.2 |
| α-humulene | 0.5 | 1.1 | 1.2 | 1.5 | 1.3 | 1.3 | 1.2 | 2.0 |
| (Z)-β-farnesene | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | 0.04 | 0.03 | 0.3 |
| α-terpineol | 0.2 | 0.3 | 0.3 | 0.8 | 0.8 | 0.2 | 0.1 | 0.5 |
| α-terpinyl acetate | 1.6 | 2.5 | 2.6 | 1.2 | 1.2 | 1.0 | 1.1 | 0.7 |
| germacrene- D | **6.1** | **9.4** | **11.2** | **17.0** | **10.3** | **3.6** | **2.3** | **12.7** |
| α-muurolene | 0.1 | 0.2 | 0.2 | 0.5 | 0.3 | 0.2 | 0.1 | 0.3 |
| Δ-cadinene | 0.4 | 0.6 | 0.8 | 1.7 | 1.3 | 0.5 | 0.3 | 1.2 |
| β-phenylethyl-2-methyl butyrate | 0.03 | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 | 0.03 | 0.01 |
| caryophyllene oxide | 0.8 | 0.4 | 0.2 | 0.2 | 0.1 | 0.1 | 0.1 | 0.1 |
| β-phenyethyl isovalerate | 0.01 | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 |
| methyl eugenol | 0.01 | 0.1 | 0.1 | 0.2 | 0.4 | 0.1 | 0.1 | 0.1 |
| T- cadinol | 0.2 | 0.1 | 0.1 | 0.3 | 0.3 | 0.1 | 0.1 | 0.1 |
| Trans-methyl eugenol | 0.1 | 0.1 | 0.1 | 0.3 | 0.4 | 0.3 | 0.4 | 0.1 |
| α-cadinol | Trace | 0.03 | 0.04 | trace | 0.1 | ----- | 0.1 | ----- |
| γ-cadinol | 0.2 | 0.1 | 0.1 | 0.2 | 0.1 | 0.02 | 0.1 | 0.1 |
| T- muurolol | 0.04 | 0.02 | 0.4 | 0.1 | 0.3 | 0.1 | 0.1 | 0.3 |
| (E.E)farnesol | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.04 | 0.1 |
| **TOTAL** | 99.8 | 96.2 | 94.9 | 86.4 | 86.1 | 90.2 | 92.8 | 95.1 |

Trace: <0.01%

**Table 4.** Results of analysis of *P. brutia* needle oils from Muğla and Denizli

| | Muğla | | | | Denizli | | | |
|---|---|---|---|---|---|---|---|---|
| Collection Dates/ | 6.6.1994 | 7.9.1994 | 14.12.1994 | 28.2.1995 | 8.6.1994 | 10.9.1994 | 16.12.1994 | 1.3.1995 |
| Compounds | % | % | % | % | % | % | % | % |
| tricyclene | 0.03 | 0.03 | 0.03 | 0.04 | 0.03 | 0.04 | 0.03 | 0.03 |
| ***α-pinene*** | **18.8** | **20.5** | **17.8** | **11.3** | **14.3** | **21.8** | **16.8** | **15.8** |
| α-thujene | Trace | Trace | trace | trace | ----- | trace | trace | Trace |
| camphene | 0.3 | 0.3 | 0.5 | 0.7 | 0.3 | 0.3 | 0.3 | 0.5 |
| Hexenal | 0.02 | 0.01 | 0.01 | ----- | ----- | 0.1 | 0.01 | 0.02 |
| ***β-pinene*** | **45.8** | **36.7** | **45.4** | **33.4** | **51.0** | **47.6** | **41.1** | **49.6** |
| sabinene | 0.2 | 0.2 | 0.2 | 0.6 | 0.1 | 0.2 | 0.1 | 0.1 |
| $\Delta^3$-carene | 1.0 | 1.8 | 1.0 | 1.7 | 1.1 | 0.5 | 0.3 | 0.1 |
| Myrcene+α-phellandrene | 3.6 | 2.3 | 1.8 | 0.6 | 1.9 | 1.4 | 1.2 | 1.5 |
| α-terpinene | 0.03 | 0.04 | 0.04 | 0.1 | 0.04 | 0.02 | 0.02 | 0.02 |
| limonene | 0.9 | 0.8 | 2.2 | 1.2 | 1.1 | 1.8 | 1.3 | 0.9 |
| βphellandrene+1.8cineole | 0.8 | 0.7 | 0.8 | 0.5 | 0.8 | 0.8 | 0.7 | 0.8 |
| (Z)-β-ocimene | 0.01 | 0.02 | 0.02 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| γ-terpinene | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.04 | 0.04 | 0.03 |
| (E)-β-ocimene | 1.5 | 0.8 | 1.0 | 0.4 | 0.4 | 1.1 | 0.7 | 0.2 |
| P-cymene | 0.03 | 0.1 | 0.1 | 0.2 | 0.04 | 0.04 | 0.03 | 0.03 |
| isoterpinolene | ---- | 0.01 | ----- | ----- | ----- | ---- | ---- | ---- |
| terpinolene | 0.3 | 0.4 | 0.3 | 0.7 | 0.4 | 0.2 | 0.1 | 0.2 |
| α-copaene | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| β-bourbonene | 0.1 | 0.2 | 0.2 | 0.7 | 0.3 | 0.1 | 0.1 | 0.2 |
| linalool | 0.1 | 0.1 | 0.4 | 0.6 | 0.3 | 0.1 | 0.2 | 0.02 |
| linalyl acetate | 0.1 | 0.1 | 0.4 | 0.2 | 0.1 | 0.2 | 0.2 | 0.3 |
| longifolene | 0.1 | 0.2 | 0.8 | 0.2 | 1.1 | ----- | ----- | ----- |
| bornyl acetate | 0.2 | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | 0.1 |
| β-elemene | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.04 | 0.1 | 0.1 |
| β-caryophyllene | 6.4 | 7.5 | 4.7 | 6.2 | **4.3** | **7.2** | **5.7** | **4.3** |
| myrtenal | 0.1 | 0.1 | 0.4 | 0.8 | 0.2 | 0.1 | 0.2 | 0.3 |
| trans-pinocarveol | 0.2 | 0.2 | 0.8 | 1.0 | 0.4 | 0.3 | 0.4 | 0.5 |
| α-humulene | 1.2 | 1.4 | 0.9 | 1.2 | 0.8 | 1.3 | 1.1 | 0.8 |
| (Z)-β-farnesene | 0.2 | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | 0.1 | 0.1 |
| α-terpineol | 0.3 | 0.6 | 0.4 | 1.0 | 0.4 | 0.2 | 0.3 | 0.3 |
| α-terpinyl acetate | 2.4 | 2.9 | 1.9 | 1.3 | 2.2 | 0.8 | 1.1 | 1.4 |
| germacrene- D | **7.0** | **12.2** | **8.2** | **15.2** | **9.9** | **7.3** | **7.5** | **11.0** |
| α-muurolene | 0.1 | 0.2 | 0.2 | 0.4 | 0.2 | 0.2 | 0.2 | 0.2 |
| Δ-cadinene | 0.6 | 1.3 | 0.8 | 1.9 | 0.8 | 0.6 | 0.6 | 0.7 |
| β-phenylethyl-2-methyl butyrate | 0.02 | trace | trace | trace | 0.6 | trace | 0.02 | 0.02 |
| caryophyllene oxide | 0.03 | 0.2 | 0.1 | 0.1 | 0.4 | 0.2 | 0.2 | 0.2 |
| β-phenyethyl isovalerate | 0.03 | 0.2 | 0.1 | 0.04 | 0.3 | 0.2 | 0.2 | 0.1 |
| methyl eugenol | 0.1 | 0.04 | 0.04 | 0.2 | 0.02 | 0.03 | 0.1 | 0.2 |
| T- cadinol | 0.1 | 0.2 | 0.2 | 0.4 | 0.2 | 0.1 | 0.2 | 0.2 |
| Trans-methyl eugenol | 0.9 | 0.8 | 0.2 | 0.3 | 0.2 | 0.2 | 0.2 | 0.2 |
| α-cadinol | Trace | trace | 0.2 | trace | ----- | ----- | 0.6 | ----- |
| γ-cadinol | 0.4 | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | ----- | 0.2 |
| T- muurolol | 0.3 | 0.01 | trace | 1.0 | 0.02 | 0.01 | 9.7 | 0.2 |
| (E.E)farnesol | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.04 | 0.1 | 0.1 |
| **TOTAL** | **94.7** | **93.9** | **92.8** | **85.2** | **95.2** | **95.7** | **92.2** | **91.8** |

Trace: <0.01%

**Table 5.** Results of analysis of *P. brutia* needle oils from Kütahya and Bursa

| | Kütahya | | | | Bursa | | | |
|---|---|---|---|---|---|---|---|---|
| Collection Dates/ | 21.9.1994 | 27.12.1994 | 2.4.1995 | 28.7.1995 | 20.9.1994 | 25.12.1994 | 31.3.1995 | 27.7.1995 |
| Compounds | % | % | % | % | % | % | % | % |
| tricyclene | 0.04 | 0.03 | 0.04 | 0.03 | 0.1 | 0.03 | 0.04 | 0.03 |
| ***α-pinene*** | **18.3** | **19.3** | **16.7** | **19.0** | **16.0** | **18.3** | **15.5** | **19.7** |
| α-thujene | Trace | trace | trace | trace | trace | trace | trace | trace |
| camphene | 0.4 | 0.3 | 0.4 | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 |
| hexenal | 0.02 | 0.03 | 0.03 | 0.02 | 0.04 | 0.03 | 0.03 | 0.02 |
| ***β-pinene*** | **48.4** | **46.6** | **52.0** | **51.3** | **44.5** | **47.4** | **50.3** | **47.0** |
| sabinene | 0.2 | 0.2 | 0.4 | 0.2 | 0.2 | 0.2 | 0.5 | 0.2 |
| $\Delta^3$-carene | 1.5 | 1.5 | 0.4 | 0.1 | 2.7 | 2.5 | 2.9 | 3.0 |
| Myrcene+α-phellandrene | 2.3 | 2.4 | 2.9 | 3.0 | 6.1 | 2.7 | 2.3 | 2.5 |
| α-terpinene | 0.03 | 0.03 | 0.1 | 0.02 | 0.03 | 0.04 | 0.1 | 0.1 |
| limonene | 1.0 | 2.0 | 2.3 | 2.8 | 0.7 | 1.2 | 2.3 | 1.5 |
| βphellandrene+1.8cineole | 0.9 | 0.7 | 0.8 | 0.9 | 0.8 | 0.8 | 0.8 | 0.9 |
| (Z)-β-ocimene | 0.04 | 0.1 | 0.04 | 0.03 | 0.02 | 0.1 | 0.1 | 0.2 |
| γ-terpinene | 0.1 | 0.1 | 0.1 | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 |
| (E)-β-ocimene | 0.3 | 0.5 | 0.5 | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 |
| P-cymene | 0.04 | 0.04 | 0.04 | 0.01 | 0.04 | 0.03 | 0.1 | 0.04 |
| isoterpinolene | ----- | trace | ----- | ----- | 0.01 | 0.01 | 0.02 | 0.02 |
| terpinolene | 0.3 | 0.3 | 0.4 | 0.3 | 0.4 | 0.4 | 0.8 | 0.5 |
| α-copaene | 0.1 | 0.1 | 0.1 | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 |
| β-bourbonene | 0.1 | 0.2 | 0.2 | 0.2 | 0.2 | 0.1 | 0.2 | 0.1 |
| linalool | 0.1 | 0.02 | 0.02 | 0.03 | 0.02 | 0.03 | 0.03 | 0.03 |
| linalyl acetate | 0.1 | trace | 0.2 | 0.04 | ----- | ----- | 0.1 | 0.1 |
| longifolene | 0.1 | 0.3 | 0.2 | 0.02 | 0.3 | 0.2 | 0.1 | ----- |
| bornyl acetate | 0.4 | 0.1 | 0.1 | 0.2 | 0.2 | 0.1 | 0.3 | 0.1 |
| β-elemene | 0.1 | 0.1 | 0.1 | 0.1 | 0.03 | 0.04 | 0.1 | 0.1 |
| β-caryophyllene | **4.9** | **4.4** | **3.8** | **3.7** | **4.0** | **4.6** | **3.8** | **4.9** |
| myrtenal | 0.1 | 0.1 | 0.3 | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 |
| trans-pinocarveol | 0.2 | 0.2 | 0.5 | 0.1 | 0.2 | 0.1 | 0.2 | 0.1 |
| α-humulene | 0.9 | 0.8 | 0.7 | 0.7 | 0.7 | 0.8 | 0.7 | 0.9 |
| (Z)-β-farnesene | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| α-terpineol | 0.3 | 0.3 | 0.4 | 0.3 | 0.3 | 0.3 | 0.4 | 0.3 |
| α-terpinyl acetate | 1.9 | 1.5 | 1.3 | 1.5 | 0.6 | 0.7 | 1.8 | 0.9 |
| germacrene- D | **10.4** | **10.9** | **7.6** | **10.8** | **13.6** | **12.5** | **9.5** | **10.7** |
| α-muurolene | 0.1 | 0.2 | 0.2 | 0.1 | 0.2 | 0.2 | 0.1 | 0.1 |
| Δ-cadinene | 0.8 | 0.9 | 0.8 | 0.7 | 0.9 | 0.8 | 0.9 | 0.7 |
| β-phenylethyl-2-methyl butyrate | 0.02 | 0.02 | 0.01 | 0.01 | 0.04 | 0.02 | 0.02 | ----- |
| caryophyllene oxide | 0.2 | 0.2 | 0.3 | 0.1 | 0.7 | 0.4 | 0.3 | 0.3 |
| β-phenyethyl isovalerate | 0.1 | 0.1 | 0.2 | 0.1 | 0.2 | 0.2 | 0.2 | 0.2 |
| methyl eugenol | 0.1 | 0.1 | 0.1 | 0.02 | 0.1 | 0.1 | 0.01 | 0.04 |
| T- cadinol | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| Trans-methyl eugenol | 0.6 | 0.2 | 0.1 | 0.3 | 1.0 | 0.7 | 0.3 | 0.1 |
| α-cadinol | trace | trace | 0.2 | trace | trace | trace | trace | ----- |
| γ-cadinol | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| T- muurolol | 0.4 | 0.3 | 0.3 | 0.2 | 0.02 | 0.02 | 0.01 | 0.01 |
| (E,E)farnesol | 0.1 | 0.1 | 0.03 | 0.1 | 0.1 | 0.1 | 0.1 | 0.02 |
| TOTAL | **96.2** | **95.6** | **95.2** | **98.1** | **96.3** | **97.0** | **96.3** | **96.6** |

Trace: <0.01%

**Table 6.** Results of analysis of *P. brutia* needle oils from Sinop and Samsun

| | Sinop | | | | Samsun | | | |
|---|---|---|---|---|---|---|---|---|
| Collection Dates/ | 4.8.1994 | 7.11.1994 | 20.2.1995 | 25.5.1995 | 25.5.1994 | 18.8.1994 | 17.11.1994 | 17.2.1995 |
| Compounds | % | % | % | % | % | % | % | % |
| Tricyclene | 0.04 | 0.03 | 0.03 | 0.03 | 0.1 | 0.03 | 0.04 | 0.2 |
| ***α-pinene*** | **18.2** | **13.3** | **14.2** | **15.0** | **21.8** | **16.7** | **18.5** | 33.5 |
| α-thujene | trace | trace | trace | trace | trace | trace | trace | ----- |
| Camphene | 0.6 | 0.3 | 0.3 | 0.2 | 0.6 | 0.4 | 0.4 | 1.0 |
| Hexenal | 0.02 | 0.03 | 0.02 | 0.03 | 0.02 | 0.02 | 0.03 | 0.04 |
| ***β-pinene*** | **38.5** | **43.9** | **44.4** | **39.1** | **22.5** | **31.1** | **40.6** | **8.6** |
| Sabinene | 0.1 | 0.2 | 0.2 | 0.2 | 0.1 | 0.2 | 0.1 | 0.02 |
| $\Delta^3$-carene | 3.3 | 2.8 | 2.7 | 3.6 | 1.5 | 4.0 | 1.3 | 0.1 |
| Myrcene+α-phellandrene | 1.4 | 1.1 | 1.3 | 1.0 | 0.9 | 1.4 | 0.8 | 0.6 |
| α-terpinene | 0.03 | 0.03 | 0.04 | 0.1 | 0.03 | 0.1 | 0.03 | 0.02 |
| Limonene | 2.3 | 1.2 | 1.6 | 2.3 | 1.5 | 2.8 | 1.1 | 0.5 |
| β-phellandrene+1.8cineole | 0.7 | 0.6 | 0.8 | 0.7 | 0.5 | 0.7 | 0.6 | 0.4 |
| (Z)-β-ocimene | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 0.02 |
| γ-terpinene | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.03 |
| (E)-β-ocimene | 0.1 | 0.2 | 0.4 | 0.7 | 0.7 | 1.2 | 0.1 | 0.4 |
| P-cymene | 0.1 | 0.1 | 0.04 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| Isoterpinolene | 0.01 | 0.01 | 0.01 | 0.02 | ----- | 0.01 | ----- | ----- |
| Terpinolene | 0.4 | 0.3 | 0.4 | 0.5 | 0.2 | 0.5 | 0.2 | 0.1 |
| α-copaene | 0.1 | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | 0.3 |
| β-bourbonene | 0.2 | 0.3 | 0.2 | 0.2 | 0.3 | 0.2 | 0.1 | 0.5 |
| Linalool | 0.1 | 0.04 | 0.02 | 0.01 | 0.1 | 0.2 | 0.03 | 0.1 |
| linalyl acetate | 0.5 | 0.1 | 0.1 | 0.3 | 0.3 | 0.6 | 0.1 | 0.5 |
| Longifolene | 0.02 | 0.1 | 0.2 | 0.02 | 0.4 | ----- | 0.2 | 0.1 |
| bornyl acetate | 0.1 | 0.2 | 0.1 | 0.1 | 0.4 | 0.1 | 0.2 | 1.0 |
| β-elemene | 0.1 | 0.1 | 0.1 | 0.04 | 0.1 | 0.1 | 0.1 | 0.02 |
| β-caryophyllene | **4.9** | **5.6** | **5.0** | **7.1** | **7.8** | **6.7** | **8.7** | **8.1** |
| Myrtenal | 0.1 | 0.1 | 0.1 | 0.1 | 0.5 | 0.1 | 0.1 | 0.7 |
| trans-pinocarveol | 0.2 | 0.2 | 0.1 | 0.1 | 0.5 | 0.2 | 0.2 | 0.9 |
| α-humulene | 0.9 | 1.0 | 0.9 | 1.2 | 1.4 | 1.2 | 1.5 | 1.5 |
| (Z)-β-farnesene | 0.2 | 0.2 | 0.1 | 0.2 | 0.2 | 0.3 | 0.1 | ----- |
| α-terpineol | 0.5 | 0.6 | 0.5 | 0.5 | 1.5 | 0.6 | 0.5 | 2.8 |
| α-terpinyl acetate | 1.2 | 2.4 | 2.2 | 1.8 | 1.9 | 2.9 | 2.8 | 0.9 |
| germacrene- D | **15.8** | **16.0** | **15.8** | **16.9** | **16.6** | **17.6** | **13.2** | **10.1** |
| α-muurolene | 0.3 | 0.2 | 0.2 | 0.2 | 0.7 | 0.4 | 0.2 | 1.0 |
| Δ-cadinene | 1.1 | 1.4 | 1.2 | 1.3 | 2.9 | 1.6 | 1.1 | 5.3 |
| β-phenylethyl-2-methyl butyrate | 0.02 | 0.01 | 0.03 | 0.03 | 0.04 | trace | 0.1 | 0.1 |
| caryophyllene oxide | 0.1 | 0.4 | 0.7 | 0.8 | 0.3 | 0.4 | 0.6 | 0.1 |
| β-phenyethyl isovalerate | 0.1 | 0.2 | 0.2 | 0.2 | 0.1 | 0.1 | 0.3 | 0.1 |
| methyl eugenol | 0.2 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.2 |
| T- cadinol | 0.2 | 0.2 | 0.2 | 0.1 | 0.2 | 0.2 | 0.2 | 0.3 |
| Trans-methyl eugenol | 0.2 | 0.2 | 0.2 | 0.2 | 0.3 | 0.4 | 0.3 | 0.3 |
| α-cadinol | ----- | 0.02 | 0.02 | 0.02 | ----- | ----- | trace | 0.03 |
| γ-cadinol | 0.1 | 0.1 | 0.1 | 0.1 | 0.2 | 0.2 | 0.1 | 0.1 |
| T- muurolol | trace | trace | trace | trace | 0.1 | 0.2 | 0.3 | 0.1 |
| (E,E)farnesol | 0.1 | ----- | 0.1 | 0.1 | 0.1 | 0.04 | 0.04 | 0.1 |
| **TOTAL** | **93.3** | **94.2** | **95.2** | **95.6** | **88.0** | **94.2** | **95.4** | **80.9** |

Trace: <0.01%

Essential oil contents of the needles were higher in spring and summer than those collected in other seasons. On the other hand, the oil content gradually decreased in fall and reached a minimum during winter. Oil contents were found to be lower in those collected in North Anatolia than those of the West and South Anatolia.

The needle oils were evaluated from the view point of chemotypes suggested by some investigators (Chalchat et al., 1995a, 1995b, Gomes da Silva et al., 2001, Kubeczka et al., 1987, Macchioni et al., 2003, Orav et al., 1996, Petrakis et al., 2000, 2001, Roussis et al., 1995, Simic et al., 1996). Results of the present study revealed that *P. brutia* of Turkish origin belonged to the so-called "chemotype-B" according to four main constituents (Roussis et al., 1995).

**Table 7.** Main constituents of *P.brutia* collected from different sites of Turkey.

| *Pinus brutia* | **α-pinene** | **β- pinene** | **$\Delta^3$-carene** | **β-caryophyllene** | **germacrene-D** |
|---|---|---|---|---|---|
| **Antalya** | 11.0-32.1 | 24.5-40.4 | 1.1-11.5 | 6.6-10.9 | 2.2-12.7 |
| **Bursa** | 15.5-19.7 | 44.5-50.3 | 2.5-3.0 | 3.8-4.9 | 9.5-13.6 |
| **Denizli** | 14.3-21.8 | 41.1-51.0 | 0.1-1.1 | 4.3-7.2 | 7.3-11.0 |
| **İçel** | 22.0-30.5 | 23.7-48.9 | 1.1-2.8 | 3.0-8.2 | 6.1-17.0 |
| **Kütahya** | 16.7-19.3 | 46.6-52.0 | 0.1-1.5 | 3.7-4.9 | 7.6-10.9 |
| **Muğla** | 11.3-20.5 | 33.4-45.8 | 1.0-1.8 | 4.7-7.5 | 7.0-15.2 |
| **Samsun** | 16.7-33.5 | 8.6-40.6 | 0.1-4.0 | 6.7-8.7 | 10.1-17.6 |
| **Sinop** | 13.3-18.2 | 38.5-44.4 | 2.7-3.6 | 4.9-7.1 | 15.8-16.9 |

Figures 1 and 2 show selected chromatograms of *P. brutia* collected from İçel at different date.

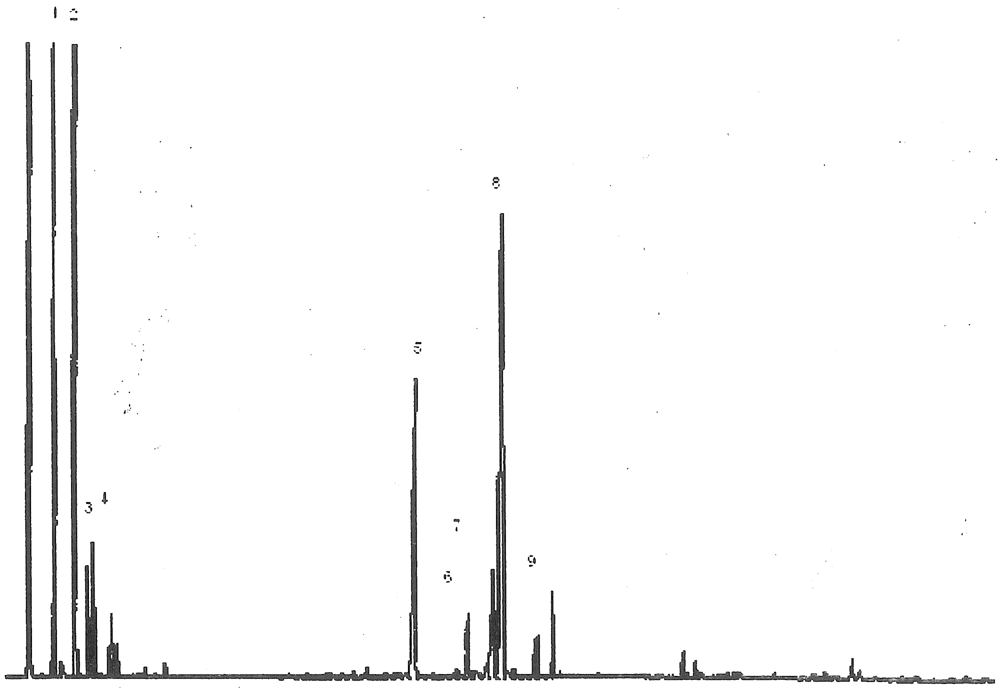

**Figure 1.** The chromatogram of P. brutia collected from İçel at 14.9.1994
1. α-pinene, 2.β-pinene , 3. $\Delta^3$ carene, 4. myrcene, 5.β-caryophyllene, 6. α-humulene, 7. α-terpinyl acetate, 8. germacrene D, 9. Δ-cadinene.

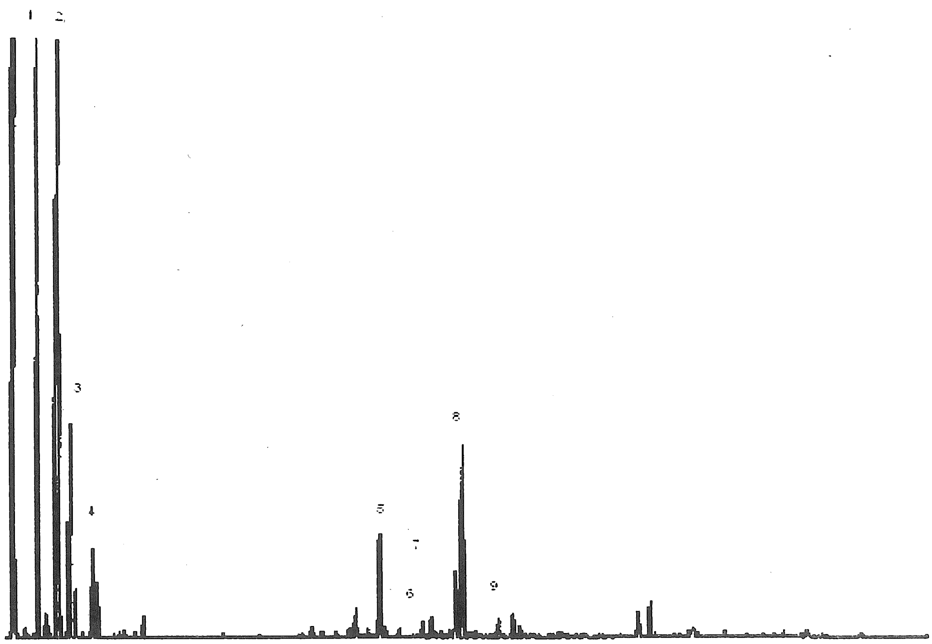

**Figure 2.** The chromatogram of P. brutia collected from İçel at15.2.1995
1. α-pinene, 2.β-pinene , 3. $\Delta^3$ carene, 4. myrcene, 5.β-caryophyllene, 6. α-humulene, 7. α-terpinyl acetate, 8. germacrene D, 9. Δ-cadinene.

The abundance of the essential oil peaks on chromatogram show the influence of the collection date on components.

### Özet

Türkiye'de doğal olarak 5 *Pinus* (çam) türü yetişmekte ve bunlardan sadece 3 tanesi T.C. Tarım ve Orman Bakanlığı tarafından kerestelik amaçla kullanılmaktadır. Bakanlık tarafından bu ağaçların diğer kısımları kullanılmamaktadır. Bu türler *P. brutia* (Kızılçam), *P.nigra* (Karaçam) ve *P. silvestris* (Sarıçam)'dir. Bu çalışmanın amacı *Pinus brutia*'dan elde edilen uçucu bileşiklerin orman tali ürünü olarak değerlendirilmesidir. Uçucu yağın bileşimi GC-GC/MS yöntemleriyle analiz edilmiştir. Bileşikler MS-Kütüphanesi kullanılarak tespit edilmiştir. Uçucu yağların yüksek oranda monoterpen hidrokarbon bileşikleri taşıdığı tespit edilmiştir.

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