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Seasonal Changes in the Concentrations of Terpenic Compounds Contained in the Leaves of Coniferous Trees

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This research tries to compare terpene compounds of five species of coniferous trees to find out which tree has relatively high–concentrated terpene compounds, and to check their concentration by season to find out which season the concentration is the highest. In addition, it tries to find out whether terpene compounds in the leaves are actually volatilized into the atmosphere. The results of the research show that there were no relatively high–concentrated terpene compounds in the leaves of *Pinus densiflora* and *Pinus koraiensis*, compared to the others. However, when the concentration of terpene compounds, which are volatilized in an airtight container, was checked, relatively high–concentrated terpene compounds were volatilized: 8 kinds of terpene compounds in the leaves of *Pinus densiflora*; 16 in *Pinus koraiensis*; 10 in *Abies holophylla*; 1 in *Chamaecyparis obtusa*; 2 in *Chamaecyparis pisifera*. And the concentration of terpene compounds by season was checked, the concentration of 21 terpene compounds was high in spring, excluding those of *Abies holophylla*, which showed high concentration in summer. The concentration of the terpene compounds by substance was generally high in spring and summer. Therefore, it is more effective to extract essential oil from coniferous trees in spring and summer.

Key words: Terpene compound, Essential oil, Softwood, Season

INTRODUCTION

As South Korea has become economically affluent and the people's average life expectancy has been extended, the people's interest in healthy lifestyles has become very high. In addition, the people's interest in comfortable environments and forests that can support a healthy lifestyle also increased and populations that go to mountains have been rapidly increasing. One important reason for going to forests as such is that the people believe that the volatile organic compounds (phytoncide) that volatilize from plants will enhance immune functions and health functions. Many studies on phytoncide have already been conducted.

The phytoncides that volatilize from *Pinus densiflora*, *Pinus koraiensis*, *Chamaecyparis obtusa*, and *Chamaecyparis pisifera* act on the central nervous system and thus are effective for stress relief (Na *et al*., 1998, 1999). In particular, the essential oil of *Chamaecyparis obtusa* leaves has been reported as having antibacterial effects (Lee *et al*., 2001), antifungal effects (Lee *et al*., 2001; Park *et al*., 2005; Gwak *et al*., 2006), antimicrobial effects (Kang *et al*., 2007), effects to reduce the concentration of immunoglobulin–E that causes atopy (Korea Forest Service 2014), and antioxidative effects (Park *et al*., 2008). In addition, it has also been reported as having effects to deodorize formaldehyde, which can play the role of a sick house syndrome–treating agent (Kim *et al*., 2009), hair growth promoting effects (Park *et al*., 2013), human stress index and fatigue index reducing effects (Shin *et al*., 2010), and human depression–relieving effects (Lee *et al*., 2011). As studies and mass communication promotions have been concentrated on *Chamaecyparis obtuse* as such, *Chamaecyparis obtuse* has been magnified as the most preferred afforestation tree species and a tendency to regard other tree species as relatively less valuable has been formed.

However, not only *Chamaecyparis obtuse* but also the essential oils of other coniferous trees, such as *Pinus koraiensis*, *Pinus densiflora*, *Chamaecyparis obtusa*, *Abies koreana* E.H. Wilson, A*bies holophylla*, and *Picea jezoensis* (S. et Z.) Carriere, were also found to be commonly containing α–pinene, camphene, myrcene, limonene, and bornyl acetate (Moon and Yoo, 2005). As such, terpenic compounds contained in coniferous trees' essential oils were reported as having biological properties, such as anti–inflammatory properties, bactericidal properties, appetite improving properties, tonic properties, circulation promoting properties, deodorizing properties, phlegm discharging properties, pesticidal properties, and tranquilizing properties (Mishra and Dubey, 1994).

Therefore, the present study was intended to extract seasonally essential oils from the leaves of five coniferous tree species, including *Chamaecyparis obtuse*, to compare changes in terpenic compounds, and to identify the correlations between leaf terpenic compound con-

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tents and the quantities of terpenic compounds volatized to the air.

MATERIALS AND METHODS

Determination of materials to be analyzed

As the purpose of the present study was to determine the terpenic compound contents in *Pinus densiflora*, *Pinus koraiensis*, *Abies holophylla*, *Chamaecyparis obtusa*, and *Chamaecyparis pisifera*, 21 kinds of terpenic compounds known to be useful to humans were selected as study subjects based on *Chamaecyparis obtuse*–related studies (Lee, 1999; Park *et al*., 2008; Kim *et al*., 2009; Kim *et al*., 2011; Korea Forest Service, 2014) and other coniferous tree–related studies (Kubeczka and Schultze, 1987; Moon and Yoo, 2005; Bakkali *et al*., 2008; Oh *et al*., 2013; Kim, 2014). In addition, for quantitative analyses, calibration curves for three concentrations of standard materials within the quantitation range were prepared (Table 1).

Essential oil extraction and analysis

Testing Materials

The tested tree species in the present study were coniferous trees comprising *Pinus densiflora*, *Pinus koraiensis*, *Abies holophylla*, *Chamaecyparis obtus*a, and *Chamaecyparis pisifera* and leaves were collected from three trees per species. As seasonal samples, *Chamaecyparis obtusa* samples were collected in Jagye–ri, Yonghwa–myeon, and Chungcheongbuk–do and *Pinus densiflora*, *Pinus koraiensis*, *Abies holophylla*, and *Chamaecyparis pisifera* samples were collected in the academic forest of Chungbuk National University in Songgye–ri, Hansu–myeon, Jecheon–si, and Chungcheongbuk–do on May 21, 2014 (spring), July 30, 2014 (summer), October 22, 2014 (autumn), and January 29, 2015 (winter).

Essential Oil Extraction and GC/MSD Analysis Using S.D.E Equipment

After sampling, the samples were put into zipper bags (30 cm × 35 cm, Clean Zipper Bag, Korea) and the zipper bags were sealed and put into ice boxes (ice box XX large number 3, Minseon General Trading Company, Korea) before being transported. Only green leaf parts of the sample were cut and ground with a mixer (HMF–3250S, Hanil Mixer, Korea). Then, 100 g of the sample and 1,000 mL of distilled water were put into a 3,000–mL distillation flask and heated. Thereafter, 100 mL of diethyl ether (case no. 60–29–7, Merck, Germany) and 100 mL of pentane (case no. 1009–66–0, Junsei, Japan) were put into a 1,000–mL flask for collection to extract essential oil components for 5 h using simultaneous steam distillation equipment (S.D.E). Anhydrous sodium sul-

Table 1. The Calibration curve of standard materials

Standards	For essential oil						For sampling of terpene emission	
	May		July		October			
	R^2	Standard curve	R^2	Standard curve	R^2	Standard curve	R^2	Standard curve
isoprene	0.9968	y = 59059x – 32784	0.9993	y = 42462x – 65510	0.9994	y = 67401x – 18789	0.9914	y = 3782.6x + 77842
bornyl acetate	0.9996	y = 112798x + 18528	0.9984	y = 128235x – 36249	0.9987	y = 186884x – 55125	0.9983	y = 8361.4x – 61394
camphene	1.0000	y = 149417x + 2592.6	0.999	y = 134396x – 46494	0.9999	y = 192209x – 16469	0.9984	y = 10928x – 51315
camphor	0.9996	y = 129189x + 10210	0.9985	y = 145631x – 46029	0.9991	y = 205249x – 43210	0.9975	y = 8788.9x – 64244
carene	0.9999	y = 147621x – 2562.2	0.9986	y = 137418x – 56277	0.9999	y = 193566x – 25540	0.9982	y = 13691x – 78189
eucalyptol	0.9999	y = 177460x + 4747	0.9986	y = 188003x – 65850	0.9995	y = 282325x – 39669	0.9981	y = 2746.3x – 10709
limonene	1.0000	y = 170970x – 6133.8	0.9985	y = 162438x – 69222	0.9998	y = 227434x – 26540	0.9976	y = 14357x – 109440
linalool	0.9994	y = 108335x – 25287	0.9991	y = 110433x – 25391	0.9976	y = 160771x – 75918	0.9968	y = 6228.5x – 69082
myrcene	0.9971	y = 22916x – 13221	0.9979	y = 19904x – 10683	0.9974	y = 30074x – 15580	0.9998	y = 1930.6x – 4988.7
p–cymene	0.9999	y = 196934x + 8310.2	0.9986	y = 200127x – 74063	0.9999	y = 283206x – 25644	0.9983	y = 31864x – 89386
sabinene	0.9999	y = 149276x – 10282	0.998	y = 137927x – 67930	0.9998	y = 188280x – 33518	ND	
terpineol	1.0000	y = 67024x – 2231.4	0.9975	y = 74167x – 22539	0.9983	y = 107343x – 33252	0.9970	y = 1718.9x – 16130
terpinolene	0.9998	y = 156450x – 3130.2	0.9981	y = 159006x – 73916	0.9995	y = 223082x – 56497	0.9980	y = 750.07x – 4781.6
α–phellandrene	0.9999	y = 171917x – 3171.4	0.9983	y = 160757x – 75084	0.9997	y = 223433x – 43083	0.9982	y = 20330x – 108049
α–pinene	1.0000	y = 165112x + 4413	0.999	y = 144316x – 50260	0.9997	y = 202918x – 4430.8	0.9986	y = 17440x – 78742
α–terpinene	0.9999	y = 176196x – 5102	0.9982	y = 170544x – 77255	0.9997	y = 235954x – 30308	0.9984	y = 11553x – 59690
β–pinene	1.0000	y = 209379x + 279.4	0.9987	y = 192011x – 74653	0.9999	y = 270414x – 25354	0.9977	y = 17589x – 127067
γ–terpinene	0.9999	y = 170646x – 900.8	0.9982	y = 167945x – 75394	0.9998	y = 239569x – 41997	0.9976	y = 16144x – 135152
cedrol	0.9993	y = 113371x + 13755	0.9975	y = 114465x – 45652	1.0000	y = 161346x – 208.6	0.9967	y = 6227.7x – 72966
transcaryophyllene	0.9992	y = 114682x + 29267	0.9983	y = 131980x – 61809	0.9975	y = 191379x – 96382	0.9983	y = 4144x – 30037
α–humulene	0.9995	y = 100661x – 943.6	0.9977	y = 112863x – 56548	0.9977	y = 160076x – 80522	0.9983	y = 11773x – 83379

Table 2. GC/MSD conditions

GC–MSD	GCMS–QP2010 Plus, Shimadzu, Japan				
Carrier Gas	Helium (99.999)				
Injector Temp.	220°C				
Interface Temp.	230°C				
Capillary	HP–INNOWAX				
column	(30m × 0.25mmI.D × 0.25μm, filmthickness)				
Column Flow	0.9 mL/min				
	Initial	Initial	Rate	Final	Final
Oven Program	Temp. (°C)	Time (min)	(°C/min.)	Temp. (°C)	Time (min)
	35	6	5	240	2
Post run	250 °C, 3 min				
Ion Source Temp.	250 °C				
EI voltage	70 eV				
Monitor Ion	m/z = 35 ~ 400				
Solvent Delay	1 min				
Monitering mode	SCAN mode				

fate (case no. 7757–82–6, Junsei, Japan) was put into the extracted essential oil and the essential oil was kept in a –2°C cold room for 24 h to agglutinate moisture. Thereafter, to remove the sodium sulfate that was holding in the moisture, the essential oil was pushed through a syringe (Kovax–syringe 50 mL, Korea Vaccine Co., Ltd., Korea) installed with a filtration filter (0.45 μm syringe filter, Sartorius, Germany) to separate the dehydrated essential oil. The dehydrated essential oil was put into a 500–mL round flask, the solvent in the essential oil was removed, the essential oil was being concentrated using a rotary decompression and compression system (CVE–3100, Eyela, Japan), and the essential oil was moved to a 15–mL conical tube. To collect the essential oil sticking to the wall of the 500–mL round flask, 5 mL of diethyl ether (case no. 60–29–7, Merck, Germany) and 5 mL of pentane (case No. 1009–66–0, Junsei, Japan) were put into the flask. The wall was washed by micro–vibrating the flask using an ultrasonic washer (SD–D300H, Seongdong Ultrasonic Co., Korea), the collected essential oil was put into the abovementioned 15–mL conical tube containing the essential oil, the essential oil was completely concentrated using a nitrogen evaporator (MG–2100, Eyela, Japan), and the weight of the concentrated essential oil was measured.

Thereafter, 100 μg of the essential oil was taken and put into a 100–mL volume flask and the flask was filled with dichloromethane (case no. 75–09–2, Sigma–Aldrich, USA) up to the scale mark to dilute the essential oil. Then, a certain quantity of the diluted essential oil was taken and used as a sample for GC/MSD analysis. GC/MSD (GCMS–QP2010 Plus, Shimadzu, Japan) analysis conditions were set as shown in (Table 2) to identify terpenic compounds.

Collection and Analysis of Terpenic Compounds Volatilizing from Coniferous Tree Leaves in Airtight Containers

To analyze the correlation between the terpenic compounds contained in the leaves and the terpenic compounds volatilizing through stomas, the sample was put into an airtight container and the air in the container was collected and analyzed. Out of the sample (leaves) collected on October 22, 5.0 g was put into a plastic airtight container (Cereal dispenser 3.9 L, Lock & Lock, China) and the air in the airtight container was collected with a sampling pump (MP–Σ30KN, Sibata, Japan) installed with a solid absorption tube (KT50601, Supelco, USA) filled with Tenax–TA. The sample that was put into the airtight container together with the sampling pump and was maintained in a 20°C constant temperature state and the sampling pump was set to collect 2 L of the air from 90 min after sealing at a flow rate of

Table 3. Thermal desorber conditions

Thermal Desorber	TurboMatrix ATD 350, Perkinelmer, USA
Column Flow	1 mL/min
Desorb Flow	50 mL/min
Temp.	Max. 320°C
Valve Temp.	250°C
Transferline Temp.	250°C
Cold trap	Tenax – TA
Low Temp.	–30°C
High Temp.	310°C
Rate	40°C/sec
Inlet Flow	off
Outlet Split ratio	100 : 1

Table 4. GC/MSD conditions

GC–MSD	GCMS–QP2010 Plus, Shimadzu, Japan				
Carrier Gas	Helium (99.999)				
Interface Temp.	230°C				
Capillary column	HP–INNOWAX (30m × 0.25mmI.D × 0.25μm, filmthickness)				
Column Flow	1 mL/min				
Oven Program	Initial	Initial	Rate	Final	Final
	Temp. (°C)	Time (min)	(°C/min.)	Temp. (°C)	Time (min)
	35	6	5	240	2
Post run	250°C, 3 min				
Ion Source Temp.	250°C				
EI voltage	70 eV				
Solvent Delay	1 min				
Monitering mode	SIM mode				

100 mL/min. The collected sample was kept in a –2°C freezer until it was analyzed. To analyze the terpenes absorbed by the solid absorption tube (KT50601, Supelco, USA) within 48 h, the conditions of an automatic thermal desorption system (TurboMatrix ATD 350, PerkinElmer, USA) were set as shown in (Table 3). In addition, the GC/MSD (GC/MSD–QP2010 Plus, Shimadzu, Japan) for analysis of the desorbed terpenes was set as shown in (Table 4) to analyze the terpenes.

RESULTS AND DISCUSSION

Relationship between terpenic compounds contained in Leaves and those that volatilize from the leaves

The essential oils contained in the leaves of the five species of coniferous trees were extracted seasonally and

Table 5. The terpenes content of essential oils from five conifer species

(Unit: ng/g)

Terpenes	Component	*Pinus densiflora*	*Pinus koraiensis*	*Abies holophylla*	*Chamaecyparis obtusa*	*Chamaecyparis pisifera*
hemiterpene	isoprene†	0.0±0.0a	0.0±0.0a	0.0±0.0a	0.1±0.1b	0.0±0.0a
monoterpenes	bornyl acetate†	474.3±761.4a	743.8±720.5a	7818.1±6402.7c	3684.2±4013.8ab	5445.1±5123.9bc
	camphene†	106.9±117.8a	414.6±404.8a	1827.4±1563.8b	132.3±117a	146.5±133.8a
	camphor†	0.3±0.2a	0.4±0.4a	0.6±0.4a	0.6±0.6b	3.9±4.2a
	carene†	5.8±4.7a	105.8±175.1a	4711.0±6456.1b	32.8±28.7a	4960.6±5339.8b
	eucalyptol†	0.1±0.1a	0.0±0.0a	0.6±0.6b	0.0±0.0a	0.1±0.1a
	limonene†	119.5±143.1a	436.8±476.8a	1309.9±1901.6a	3529.1±3349.5b	802.0±778.9a
	linalool†	12.0±10.7a	11.2±10.4a	195.8±235.7b	101.0±91.8a	62.8±49.8a
	myrcene†	403.4±315.6a	300.6±259.8a	3569.7±2477.5c	1738.2±1105.4b	2780.4±2389.5bc
	p–cymene†	3.4±4.1a	3.6±4.9a	13.9±18.1ab	45.4±44.8c	25.7±20b
	sabinene†	12.7±11.1a	12.6±11.9a	35.8±34.9a	5937.3±5429.2b	29.2±21.7a
	terpineol*	11.6±4.8a	7.3±4.6a	24.6±14.1b	67.6±21.2c	16.5±9.1ab
	terpinolene†	386.0±490.5a	419.9±469.8a	725.1±633.3bc	571.5±545.1a	1247.6±1005.8c
	α–phellandrene	7.0±6.1	38.8±45	24.5±26.2	38.1±39.1	19.1±12.8
	α–pinene†	635.9±605.8a	925.5±890.8a	1138.6±932.2a	559.4±499.3a	3332.0±3129.3c
	α–terpinene†	7.5±7.3a	38.0±42a	48.9±47.5a	254.7±230.2c	32.4±24.6a
	β–pinene	112.9±113.7	110.3±117.7	52.6±48	80.7±74.4	60.3±49.5
	γ–terpinene†	12.0±11.2a	14.1±11.7a	102.5±92a	1345.7±1227.1c	65.4±51.4a
sesquiterpenes	cedrol†	8.8±12.6a	3.4±3.4a	6.8±5.9a	1705.6±1602.2c	352.4±374a
	transcaryophyllene†	1342.0±1617.5a	808.5±688.4a	2576.8±2598.1c	222.7±208.6a	439.9±425a
	α–humulene†	255.6±291.5a	179.5±149.9a	820.3±983.9c	178.9±120.7a	18.0±17.8a

Note: Values are expressed as mean±SD (n=12),
Statistical analysis of data was analyzed using one way ANOVA (a, b, c: Duncan's test, *: $p<0.05$, †: $p<0.01$)

the concentrations of terpenic compounds were examined. According to the results, of the 21 kinds of terpenic compounds, 19 kinds, except for α–phellandrene and β–pinene, were different in concentrations among the tree species, as shown in (Table 5). The concentrations of terpenic compounds were relatively lower in the essential oils in the leaves of *Pinus densiflora* and *Pinus koraiensis* compared to *Abies holophylla*, *Chamaecyparis obtusa*, and *Chamaecyparis pisifera*. On the other hand, the concentrations of eight kinds of terpenic compounds (bornyl acetate, camphene, carene, eucalyptol, linalool, myrcene, trans–caryophyllene, and α–humulene) in *Abies holophylla* leaves, nine kinds of terpenic compounds (isoprene, camphor, limonene, p–cymene, sabinene, terpineol, α–terpinene, γ–terpinene, and cedrol,) in *Chamaecyparis obtuse* leaves, and three kinds of terpenic compounds (carene, terpinolene, and α–pinene) in *Chamaecyparis pisifera* leaves were relatively higher compared to other tree species.

However, according to the results of the examination of the concentrations of volatilizing terpenic compounds in the air collected from the airtight container containing coniferous tree leaves, out of 20 kinds of terpenic compounds, the concentrations of 19 kinds, except for terpinolene, were different among the tree species, as shown in (Table 6). Eight kinds of terpenic compounds (eucalyptol, linalool, myrcene, terpineol, α–pinene, β–pinene, and γ–terpinene) were volatilized relatively more from the leaves of *Pinus densiflora* among the five coniferous trees species, in addition to 16 kinds (bornyl acetate, camphene, camphor, eucalyptol, limonene, linalool, myrcene, p–cymene, terpineol, α–phellandrene, α–pinene, α–terpinene, β–pinene, cedrol, trans–caryophyllene, and α–humulene) from the leaves of *Pinus koraiensis*, 10 kinds (bornyl acetate, camphene, camphor, eucalyptol, linalool, myrcene, terpineol, cedrol, and α–humulene) from the leaves of *Abies holophylla*, one kind (γ–terpinene) from the leaves of *Chamaecyparis obtusa*, and two kinds (isoprene and cedrol) from the leaves of *Chamaecyparis pisifera*.

This means that as the emission tendencies of terpenes contained in trees vary with the physical properties, such as molecular weights, water solubility, and volatility of individual materials (Oh *et al*., 2013), even if the quantity of terpenic compounds in essential oil–contained leaves is large, the quantity of terpenic compounds volatilized is not proportional. Therefore, unless essential oils are to be extracted from the leaves, *Chamaecyparis obtusa* afforestation is unnecessary to be insisted, but tree species for afforestation should be selected considering various aspects of ecological environments.

Seasonal changes in the concentrations of terpenic compounds contained in the five species of coniferous trees

As shown in (Tables 7 and 8), the seasonal concen-

Table 6. The terpenes content from coniferous leaf in sealed container

(Unit: ng/g)

Terpenes	Component	*Pinus densiflora*	*Pinus koraiensis*	*Abies holophylla*	*Chamaecyparis obtusa*	*Chamaecyparis pisifera*
isoprene	isoprene†	246.2±50.2bc	210.0±49.7b	190.8±78.9ab	108.5±34.1ac	316.5±24.1c
monoterpenes	bornyl acetate*	181.5±83.8ab	386.7±217.2b	295.5±83.9b	67.0±12.2a	48.1±13.9a
	camphene†	632.5±292.0a	3228.9±1761.0b	3428.2±1212.0b	28.7±0.9a	45.7±11.5a
	camphor†	13.7±1.7bc	15.8±3.1c	16.3±3.1c	1.7±0.5a	8.9±5.1b
	carene†	183.0±37.9a	1610.8±2521.1a	4740.7±496.6b	20.8±5.4.0a	1781.7±145.8a
	eucalyptol†	8.2±1.1c	9.1±1.3c	9.7±1.4c	0.3±0.0a	4.5±3.7b
	limonene*	301.2±231.6a	2501±1809.1b	640.9±629.1a	139.7±26.1a	90.3±35.0a
	linalool†	29.5±2.4b	28.2±0.4b	34.3±7.8b	3.2±2.0a	18.9±16.0b
	myrcene†	19014±9799.6b	13813±7306.5b	21505.4±4881.5b	1290.3±85.0a	1894.7±832.1a
	p–cymene†	23.1±12.8bc	31.1±3.5c	13.9±2.1ab	6.6±1.1a	8.9±3.4a
	terpineol†	28±0.5b	27.9±0.2b	27.9±0.1b	0.0±0.0a	0.0±0.0a
	terpinolene	147.3±102.7	747.5±721	201.4±65.5	25.4±1.1	70.3±3.1
	α–phellandrene*	69.1±55.5ab	146.1±107.1b	27.5±5.2a	2.0±0.4a	10.7±7.4a
	α–pinene†	3948.4±470.1d	4265.9±458.8d	2843.2±876.7c	133.5±19.0a	1381.8±489.1b
	α–terpinene†	32.8±17.9a	177.1±103.3b	27.1±6.5a	9.6±2.4a	10.0±6.3a
	β–pinene	2974.7±1544.9b	1721.5±851.8b	190.9±101.5a	56.3±66.0a	34.8±22.7a
	γ–terpinene†	34.2±14.9ab	49.8±13.9bc	35.8±6.8ab	59.0±11.8c	15.3±5.9a
sesquiterpenes	cedrol†	28.2±0.2b	28.1±0.0b	28.2±0.1b	4.0±2.3a	19.0±16.2b
	transcaryophyllene†	142.7±31.5bc	184.8±101.1c	81.3±33.1ab	4.3±1.8a	16.5±12.7a
	α–humulene†	34.7±3.6b	38.6±10.6b	33.2±10.4b	2.6±1.5a	13.8±11.9a

Note: Values are expressed as mean±SD (n=3),
Statistical analysis of data was analyzed using one way ANOVA (a, b, c: Duncan's test, *: $p<0.05$, †: $p<0.01$)

Table 7. The seasonal variation of terpenes content from five species essential oils (1)

Pinus densiflora

Component	Spring		Summer		Fall		Winter	
	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%
isoprene	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
bornyl acetate*	1525.1±960.8b	16.3	270.3±77.3a	7.5	46.8±15.0a	3.2	54.6±42.0a	4.6
camphene**	274.7±100.8b	2.9	90.7±74.5a	2.5	28.5±16.7a	1.9	33.6±20.8a	2.8
camphor	0.1±0.0	0.0	0.2±0.0	0.0	0.3±0.1	0.0	0.3±0.2	0.0
carene**	9.2±2b	0.1	10.7±2b	0.3	0.6±0.1a	0.0	2.4±0.3a	0.2
eucalyptol	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
limonene**	326.1±141.6b	3.5	102.7±25.5a	2.8	27.0±14.2a	1.8	22.2±11.4a	1.9
linalool**	22.3±3.5b	0.2	21.0±6.6a	0.6	3.8±1.3a	0.3	0.9±0.1a	0.1
myrcene**	7.7±1.5a	0.1	285.0±168.8a	7.9	750.9±163b	50.6	569.8±117.8b	48.0
p–cymene**	1.5±1.2a	0.0	9.7±3b	0.3	0.6±0.1a	0.0	1.6±0.2a	0.1
sabinene**	27.6±1.3c	0.3	16.6±6.6b	0.5	3.8±1.0a	0.3	2.7±1.2a	0.2
terpineol**	10.4±0.2b	0.1	14.8±4.4ab	0.4	15.5±2.1b	1.0	5.3±1.5a	0.4
terpinolene**	1086.9±521.7b	11.6	317.9±43.0a	8.8	82.9±56.5a	5.6	56.0±25.1a	4.7
α–phellandrene**	8.1±1.7b	0.1	15.0±5.6b	0.4	2.0±0.2a	0.1	2.6±0.4ab	0.2
α–pinene**	1552.3±131.8b	16.5	586.5±397.2a	16.2	195.8±86.5a	13.2	208.9±88.1a	17.6
α–terpinene**	11.4±0.5b	0.1	16.0±6.3b	0.4	1.7±0.1a	0.1	0.8±0.4a	0.1
β–pinene**	284.7±38.6c	3.0	109.5±61.2b	3.0	28.8±10.5a	1.9	28.5±13.1a	2.4
γ–terpinene**	22.6±2.1b	0.2	21.3±8.4b	0.6	3.0±0.2a	0.2	0.8±0.3a	0.1
cedrol**	27.2±12.9b	0.3	5.6±2.1a	0.2	1.6±2.1a	0.1	0.8±0.8a	0.1
trans–caryophyllene**	3532.0±1800.6b	37.7	1431.1±184.9a	39.6	240.2±72.3a	16.2	164.5±27.9a	13.9
α–humulene**	650.1±304.3b	6.9	290.7±17.1a	8.0	50.7±13.9a	3.4	30.9±8.0a	2.6
total	9380.0	100.0	3615.3	100.0	1484.5	100.0	1187.2	100.0

Pinus koraiensis

Component	Spring		Summer		Fall		Winter	
	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%
isoprene	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
bornyl acetate*	1489.4±760.0b	17.7	1119.7±515.3b	17.2	166.7±61.2a	9.4	199.3±134.2a	12.4
camphene	844.9±456.2	10.1	533.8±401.9	8.2	152.3±61.0	8.6	127.4±43.3	7.9
camphor	0.1±0.0	0.0	0.3±0.1	0.0	0.3±0.1	0.0	0.8±0.6	0.0
carene	87.9±77.8	1.0	214.1±346.2	3.3	69.9±119.4	3.9	51.1±62.5	3.2
eucalyptol	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
limonene	898.0±592.4	10.7	643.8±419.5	9.9	110.9±48.1	6.2	94.5±51.5	5.9
linalool**	14.0±6.6ab	0.2	24.1±9.6b	0.4	3.9±0.7a	0.2	2.6±1.7a	0.2
myrcene	57.9±50.4	0.7	190.4±60.1	2.9	499.1±283.4	28.0	454.7±288.7	28.3
p–cymene**	1.1±0.9a	0.0	11.1±3.4b	0.2	0.8±0.1a	0.0	1.2±0.4a	0.1
sabinene	21.4±17.0	0.3	21.1±4.4	0.3	4.3±2.1	0.2	3.3±2.8	0.2
terpineol	5.5±4.0	0.1	8.6±7.9	0.1	8.2±3.9	0.5	6.7±3.4	0.4
terpinolene	783.8±575.8	9.3	679.4±528.9	10.4	124.5±86.1	7.0	91.8±52.6	5.7
α–phellandrene	75.6±66.8	0.9	58.5±39.3	0.9	10.5±6.4	0.6	10.5±8.4	0.7
α–pinene*	1914.3±1015.8b	22.8	1157.0±802.4ab	17.8	345.8±86.6a	19.4	284.9±119.6a	17.8
α–terpinene	73.4±60.1	0.9	58.1±33.9	0.9	10.5±5.8	0.6	9.7±8.6	0.6
β–pinene	229.2±157.9	2.7	148.3±93.9	2.3	33.8±10.1	1.9	29.9±19.0	1.9
γ–terpinene**	21.6±4.6b	0.3	27.7±4.9b	0.4	4.4±0.8a	0.2	2.5±1.0a	0.2
cedrol	7.5±4.1	0.1	3.9±0.7	0.1	0.2±0.1	0.0	1.7±1.1	0.1
trans–caryophyllene**	1547.6±389.8b	18.4	1306.8±331.8b	20.1	189.5±17.5a	10.6	189.8±52a	11.8
α–humulene**	330.5±84.6b	3.9	299.8±70.7b	4.6	45.5±4.3a	2.6	41.9±10.5a	2.6
total	8403.7	100.0	6506.5	100.0	1781.1	100.0	1604.3	100.0

Abies holophylla

Component	Spring		Summer		Fall		Winter	
	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%
isoprene	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
bornyl acetate**	14433.3±2744.9b	46.9	13140.8±1578.2b	27.7	1849.8±537.5a	15.7	1848.4±352.8a	18.5
camphene**	3390.9±1380.2b	11.0	2905.9±940b	6.1	510.4±78.3a	4.3	502.3±90.2a	5.0
camphor**	0.1±0.0b	0.0	0.6±0.2a	0.0	0.5±0.1a	0.0	1.0±0.2c	0.0
carene**	25.3±10.2a	0.1	14652.2±5201.3b	30.9	2198.5±163.4a	18.6	1967.8±481.5a	19.7
eucalyptol*	0.0±0.0a	0.0	0.8±0.3ab	0.0	1.1±0.8b	0.0	0.3±0.1a	0.0
limonene	2413.8±2404.7	7.8	2304.0±2719.7	4.9	240.2±106.3	2.0	281.6±289.1	2.8
linalool	286.2±294.1	0.9	423.6±224.9	0.9	42.5±17.9	0.4	30.7±41.7	0.3
myrcene**	20.2±7.4a	0.1	4321.2±1976.7b	9.1	5797.4±756.9b	49.2	4139.9±1264.6b	41.4
p–cymene**	4.7±1.1b	0.0	43.4±3.4a	0.1	3.9±3.7a	0.0	3.2±0.2a	0.0
sabinene**	44.6±16.9b	0.1	82.7±24.8c	0.2	10.4±2.5a	0.1	5.5±1.9a	0.1
terpineol	19.8±18.4	0.1	38.1±8.9	0.1	26.2±12.2	0.2	14.1±6.8	0.1
terpinolene**	1298.3±446.8b	4.2	1272.4±346.4b	2.7	175.3±28.6a	1.5	154.4±35.0a	1.5
α–phellandrene**	22.0±8.8a	0.1	64.3±16.3b	0.1	5.9±0.7a	0.1	5.7±0.6a	0.1
α–pinene**	1973.3±561.2b	6.4	1931.5±691.7b	4.1	341.7±79.4a	2.9	307.5±48.0a	3.1
α–terpinene**	60.2±25.2b	0.2	113.7±29.6c	0.2	12.8±2.3a	0.1	8.7±2.5a	0.1
β–pinene**	81.2±20.1b	0.3	102.5±50.7b	0.2	15.5±4.7a	0.1	11.0±2.5a	0.1
γ–terpinene**	161.9±60.9b	0.5	203.3±54.1b	0.4	24.6±3.6a	0.2	20.0±4.8a	0.2
cedrol*	11.2±5.8b	0.0	11.1±3.8b	0.0	0.3±0.2a	0.0	4.3±3.4ab	0.0
trans–caryophyllene*	5002.3±2257b	16.2	4377.2±2213.1b	9.2	405.3±58a	3.4	522.2±196.9a	5.2
α–humulene	1541.9±1135.3	5.0	1439.1±1146.4	3.0	128.1±16.1	1.1	171.7±124.6	1.7
total	30791.2	100.0	47428.4	100.0	11790.4	100.0	10000.3	100.0

Note: Values are expressed as mean±SD (n=3),
Statistical analysis of data was analyzed using one way ANOVA (a, b, c: Duncan's test, *: $p<0.05$, †: $p<0.01$)

Table 8. The seasonal variation of terpenes content from five species essential oils (2)

Chamaecyparis obtusa

Component	Spring		Summer		Fall		Winter	
	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%
isoprene†	0.0±0.0a	0.0	0.1±0.1ab	0.0	0.2±0.1b	0.0	0.0±0.0a	0.0
bornyl acetate	7140.9±5967.6	19.1	5944.0±1650.2	19.4	777.0±253.5	12.2	874.5±322	13.6
camphene†	235.7±77.9b	0.6	235.3±70.3b	0.8	28.5±7.1a	0.4	29.4±9.5a	0.5
camphor*	0.4±0.1a	0.0	0.4±0.1a	0.0	0.3±0.1a	0.0	1.3±0.7b	0.0
carene†	44.9±12.7b	0.1	68.7±19.6c	0.2	5.4±4.5a	0.1	12.0±4.2a	0.2
eucalyptol	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
limonene†	7805.8±1967.3c	20.9	5030.2±1373.4b	16.4	745.4±166.5a	11.7	534.6±171.9a	8.3
linalool†	190.5±57.6b	0.5	175.4±48.3b	0.6	23.2±3.1a	0.4	14.7±5.0a	0.2
myrcene†	36.2±9.9a	0.1	1917.6±335b	6.2	2503.2±383.2b	39.2	2495.4±573.8b	38.7
p–cymene†	63.4±28.6b	0.2	100.1±35.2b	0.3	7.2±1.1a	0.1	10.7±2.7a	0.2
sabinene†	11796.5±3589.4b	31.6	9571.2±3163.8b	31.2	1191.4±344.8a	18.7	1189.9±458.0a	18.5
terpineol	88.9±23.3	0.2	61.5±19.3	0.2	68.2±13.6	1.1	51.6±15.3	0.8
terpinolene†	1265.1±308.2c	3.4	822.5±226.1b	2.7	112.3±27.2a	1.8	85.7±29.4a	1.3
α–phellandrene†	39.1±11.1b	0.1	96.0±24.6c	0.3	8.5±1.3a	0.1	8.6±2.1a	0.1
α–pinene†	1006.8±341.2b	2.7	988.2±315.8b	3.2	120.9±29.3a	1.9	121.5±39.9a	1.9
α–terpinene†	496.0±117.4b	1.3	429.4±118.8b	1.4	48.3±11.0a	0.8	44.8±16.4a	0.7
β–pinene†	134.5±43.2b	0.4	158.7±45.4b	0.5	15.6±3.3a	0.2	14±4.9a	0.2
γ–terpinene†	2811.6±645b	7.5	2062.8±534.6b	6.7	253.1±59.7a	4.0	255.1±83.1a	4.0
cedrol†	3587.1±1209.7b	9.6	2470.2±1076.6b	8.0	368.4±151.4a	5.8	396.5±181.6a	6.2

trans–caryophyllene†	473.1±193c	1.3	297.5±136.9bc	1.0	73.6±22.4b	1.2	46.7±22.1a	0.7
α–humulene*	145.9±75.5a	0.4	277.4±26.1ab	0.9	32.4±5.6b	0.5	259.9±124.4b	4.0
total	37362.4	100.0	30707.2	100.0	6383.1	100.0	6446.9	100.0

Chamaecyparis pisifera

Component	Spring		Summer		Fall		Winter	
	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%	Mean±SD (ng/g)	%
isoprene	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0	0.0±0.0	0.0
bornyl acetate†	12696.7±3039.5c	45.5	6339.6±2225.3b	24.4	1167.6±281.8a	11.7	1576.5±393.1a	10.2
camphene†	342.6±101.3b	1.2	120.3±76.8a	0.5	40.2±11.2a	0.4	82.5±18.1a	0.5
camphor	3.0±3.7	0.0	2.4±2.9	0.0	5.1±4.5	0.1	5.0±6.7	0.0
carene*	27.8±1.4a	0.1	11857.3±6502.7b	45.6	3170.1±987a	31.8	4786.8±928.4a	30.9
eucalyptol†	0.1±0.0a	0.0	0.0±0.0a	0.0	0.1±0.1b	0.0	0.1±0b	0.0
limonene†	1951.8±570.4b	7.0	778.3±265.5a	3.0	215.7±34.5a	2.2	262.1±51.7a	1.7
linalool†	120.3±47.8c	0.4	83.0±33.5bc	0.3	18.9±6.2a	0.2	2 9.0±3.2ab	0.2
myrcene†	21.9±0.7a	0.1	1438.4±560.1b	5.5	3737.0±176c	37.5	5923.9±864.6d	38.3
p–cymene†	48.5±4.2b	0.2	38.2±12.4b	0.1	5.3±0.5a	0.1	10.5±3.9a	0.1
sabinene†	56.6±11.6a	0.2	38.0±13.5b	0.1	9.4±1.5a	0.1	12.4±3.3a	0.1
terpineol*	12.6±1.9a	0.0	8.6±1.9a	0.0	16.3±4.1a	0.2	28.4±10.2c	0.2
terpinolene†	2654.5±147.8a	9.5	1484.1±657.8b	5.7	358.5±80.5c	3.6	493.2±98.4a	3.2
α–phellandrene†	24.2±1.2b	0.1	34.9±11b	0.1	6.3±1.8a	0.1	10.9±2.2a	0.1
α–pinene†	7917.1±2665.4b	28.3	2546.6±1585a	9.8	953.0±243a	9.6	1911.2±453.1a	12.3
α–terpinene†	60.7±6.6b	0.2	46.9±19.3b	0.2	10.0±1.3a	0.1	11.9±1.5a	0.1
β–pinene†	130.6±24.8c	0.5	64.3±33.7b	0.2	16.9±3.9ab	0.2	29.4±5a	0.2
γ–terpinene†	132.4±9.5c	0.5	87.2±31.1b	0.3	19.2±3.1a	0.2	22.5±3.2a	0.1
cedrol	743.9±442.2	2.7	435.9±380.7	1.7	88.3±73.9	0.9	141.4±107.2	0.9
trans–caryophyllene*	964.6±442.3b	3.5	529.9±275.7ab	2.0	1 29.3±59.1a	1.3	135.6±97.1a	0.9
α–humulene†	16.7±12.7a	0.1	43.7±10.5b	0.2	6.7±0.5a	0.1	4.8±3.1a	0.0
total	27926.6	100.0	25977.6	100.0	9973.9	100.0	15478.1	100.0

Note: Values are expressed as mean±SD (n=3),
Statistical analysis of data was analyzed using one way ANOVA (a, b, c: Duncan's test, *: $p<0.05$, †: $p<0.01$)

trations of all the 21 kinds of terpenic compounds were the highest in summer in the case of *Abies holophylla* and in spring in the cases of *Pinus densiflora*, *Pinus koraiensis*, *Chamaecyparis obtusa*, and *Chamaecyparis pisifera* according to analyses. In addition, seasonal differences in the concentrations of terpenic compounds were also found among the kinds of terpenic compounds. Eighteen kinds of terpenic compounds in the case of *Pinus densiflora*, seven kinds in the case of *Pinus koraiensis*, 16 kinds in the case of *Abies holophylla*, 18 kinds in the case of *Chamaecyparis obtusa*, and 18 kinds in the case of *Chamaecyparis pisifera* showed significant seasonal differences in concentrations in the analysis. The concentrations of most of the various terpenic compounds contained in the leaves of the five species of coniferous trees were higher in spring and summer, except for the concentrations of myrcene, which showed a tendency to increase in autumn and winter.

The reason why the concentrations of terpenic compounds contained in coniferous trees are higher in summer or spring as such is judged to be that germination, growth, and development are the most active in spring (Kim, 2001) and that the time of biosynthesis of terpenic compounds is seasons in which physiological activities are vigorous (Song, 1995). The concentration of myrcene was shown to commonly increase as seasons changed from spring to summer, autumn, and winter in order of precedence in the essential oils extracted from the leaves of the five species of coniferous trees. This was consistent with the result of a previous study conducted by Son and Hwang (Son and Hwang, 1990), indicating that the concentration of myrcene contained in the leaves of *Pinus densiflora*, *Pinus rigida* Mill., and *Pinus thunbergii* Parl. increased until July, but not with the result indicating that the concentration decreased in October.

The correlations among various terpenic compounds that increased or decreased seasonally were analyzed and the results are as shown in (Tables 9 and 10). That is, the concentrations of terpenic compounds contained in the essential oils extracted from the leaves of five species of coniferous trees generally showed positive (+) correlations with each other, except for several terpenic compounds that showed negative (−) correlations with each other. In the essential oil extracted from the leaves of *Pinus densiflora*, myrcene showed negative (−) cor-

Table 9. The correlation among the terpenes (1)

		isoprene	bornyl acetate	camphene	camphor	carene	eucalyptol	limonene	linalool	myrcene	p–cymene	sabinene
isoprene	P. densiflora	1										
	P. koraiensis	1										
	A .holophylla	1										
	C. obtusa	1										
	C. pisifera	1										
bornyl	P. densiflora	ND										
acetate	P. koraiensis	ND	1									
	A .holophylla	ND	1									
	C. obtusa	–0.242	1									
	C. pisifera	ND	1									
camphene	P. densiflora	ND	0.925†	1								
	P. koraiensis	ND	0.839†	1								
	A .holophylla	ND	0.957†	1								
	C. obtusa	–0.239	0.898†	1								
	C. pisifera	ND	0.936†	1								
camphor	P. densiflora	ND	–0.486	–0.474	1							
	P. koraiensis	ND	–0.390	–0.424	1							
	A .holophylla	ND	–0.609	–0.542	1							
	C. obtusa	–0.463	–0.278	–0.346	1							
	C. pisifera	ND	–0.382	–0.290	1							
carene	P. densiflora	ND	0.438	0.564	–0.558	1						
	P. koraiensis	ND	–0.022	–0.060	–0.165	1						
	A .holophylla	ND	0.383	0.368	0.178	1						
	C. obtusa	–0.160	0.775†	0.936†	–0.289	1						
	C. pisifera	ND	–0.205	–0.236	.007	1						
eucalyptol	P. densiflora	ND	–0.285	–0.261	0.721†	–0.331	1					
	P. koraiensis	ND	ND	ND	ND	ND	1					
	A .holophylla	ND	–0.242	–0.247	0.086	0.369	1					
	C. obtusa	ND	ND	ND	ND	ND	1					
	C. pisifera	ND	–0.504	–0.315	–0.131	–0.401	1					
limonene	P. densiflora	ND	0.534	0.656*	–0.541	0.645*	–0.061	1				
	P. koraiensis	ND	0.973†	0.914†	–0.388	–0.085	ND	1				
	A .holophylla	ND	0.488	0.499	–0.142	0.295	–0.235	1				
	C. obtusa	–0.333	0.867†	0.948†	–0.362	0.791†	ND	1				
	C. pisifera	ND	0.976†	0.972†	–0.306	–0.253	–0.401	1				
linalool	P. densiflora	ND	0.524	0.660*	–0.619*	0.936†	–0.415	0.753†	1			
	P. koraiensis	ND	0.826†	0.640*	–0.313	0.018	ND	0.792†	1			
	A .holophylla	ND	0.715†	0.622*	–0.380	0.578*	0.025	0.053	1			
	C. obtusa	–0.186	0.718†	0.932†	–0.390	0.880†	ND	0.924†	1			
	C. pisifera	ND	0.961†	0.883†	–0.427	–0.035	–0.507	0.930†	1			
myrcene	P. densiflora	ND	–0.697*	–0.738†	0.703*	–0.790†	0.306	–0.742†	–0.742†	1		
	P. koraiensis	ND	–0.573	–0.599*	0.660*	0.035	ND	–0.556	–0.467	1		
	A .holophylla	ND	–0.577*	–0.520	0.472	0.393	0.726†	–0.418	–0.144	1		
	C. obtusa	0.353	–0.569	–0.619*	0.367	–0.388	ND	–0.809†	–0.691*	1		
	C. pisifera	ND	–0.828†	–0.663*	0.183	0.088	0.720†	–0.776†	–0.759†	1		
p–cymene	P. densiflora	ND	–0.103	0.070	–0.156	0.683*	–0.338	–0.031	0.562	–0.236	1	
	P. koraiensis	ND	0.430	0.299	–0.108	0.123	ND	0.407	0.847†	–0.260	1	
	A .holophylla	ND	0.519	0.426	0.036	0.893†	0.301	0.316	0.570	0.141	1	
	C. obtusa	–0.093	0.769†	0.936†	–0.286	0.987†	ND	0.793†	0.895†	–0.372	1	
	C. pisifera	ND	0.893†	0.826†	–0.198	0.168	–0.730†	0.869†	0.882†	–0.827†	1	
sabinene	P. densiflora	ND	0.721†	0.861†	–0.597*	0.844†	–0.307	0.865†	0.930†	–0.828†	0.317	1
	P. koraiensis	ND	0.812†	0.435	–0.353	0.213	ND	0.678*	0.738†	–0.420	0.470	1
	A .holophylla	ND	0.836†	0.830†	–0.290	0.799†	0.082	0.548	0.687*	–0.125	0.799†	1
	C. obtusa	–0.314	0.910†	0.986†	–0.349	0.870†	ND	0.981†	0.914†	–0.708†	0.867†	1
	C. pisifera	ND	0.944†	0.915†	–0.300	0.049	–0.592*	0.947†	0.930†	–0.811†	0.969†	1

Note: Values are expressed as Pearson's product moment correlation coefficient(n=3), *: $p<0.05$, †: $p<0.01$)

Table 10. The correlation among the terpenes (2)

		terpineol	terpinolene	α–phellandrene	α–pinene	α–terpinene	β–pinene	γ–terpinene	cedrol	trans –caryophyl-lene	α–humulene
terpineol	P. densiflora	1									
	P. koraiensis	1									
	A .holophylla	1									
	C. obtusa	1									
	C. pisifera	1									
terpinolene	P. densiflora	–0.006	1								
	P. koraiensis	0.413	1								
	A .holophylla	0.214	1								
	C. obtusa	0.670*	1								
	C. pisifera	–.0444	1								
α–phellandrene	P. densiflora	0.454	0.328	1							
	P. koraiensis	0.182	0.338	1							
	A .holophylla	0.532	0.776†	1							
	C. obtusa	0.200	0.629*	1							
	C. pisifera	–0.515	0.716†	1							
α–pinene	P. densiflora	0.077	0.862†	0.459	1						
	P. koraiensis	0.362	0.742†	0.848†	1						
	A .holophylla	0.263	0.970†	0.786†	1						
	C. obtusa	0.589*	0.953†	0.824†	1						
	C. pisifera	–0.304	0.857†	0.431	1						
α–terpinene	P. densiflora	0.448	0.448	0.962†	0.632*	1					
	P. koraiensis	0.151	0.348	0.998†	0.845†	1					
	A .holophylla	0.417	0.899†	0.961†	0.867†	1					
	C. obtusa	0.585*	0.978†	0.772†	0.990†	1					
	C. pisifera	–0.537	0.955†	0.838†	0.816†	1					
β–pinene	P. densiflora	0.078	0.913†	0.485	0.988†	0.642*	1				
	P. koraiensis	0.250	0.516	0.972†	0.948†	0.969†	1				
	A .holophylla	0.405	0.871†	0.854†	0.952†	0.861†	1				
	C. obtusa	0.511	0.907†	0.895†	0.990†	0.970†	1				
	C. pisifera	–0.397	0.955†	0.635*	0.963†	0.937†	1				
γ–terpinene	P. densiflora	0.342	0.650*	0.854†	0.801†	0.951†	0.821†	1			
	P. koraiensis	0.105	0.612*	0.753†	0.719†	0.782†	0.751†	1			
	A .holophylla	0.319	0.983†	0.876†	0.955†	0.962†	0.893†	1			
	C. obtusa	0.630*	0.996†	0.689*	0.971†	0.992†	0.936†	1			
	C. pisifera	–0.516	0.986†	0.776†	0.849†	0.986†	0.957†	1			
cedrol	P. densiflora	–0.050	0.854†	0.187	0.860†	0.381	0.844†	0.566	1		
	P. koraiensis	–0.082	0.434	0.853†	0.828†	0.866†	0.886†	0.697*	1		
	A .holophylla	–0.008	0.0853†	0.693*	0.899†	0.763†	0.867†	0.833†	1		
	C. obtusa	0.730**	0.972†	0.650*	0.947†	0.951†	0.909†	0.967†	1		
	C. pisifera	–0.331	0.806†	0.604*	0.667*	0.768†	0.749†	0.757†	1		
trans –caryophyllene	P. densiflora	0.050	0.970†	0.449	0.841†	0.545	0.911†	0.730†	0.740†		
	P. koraiensis	0.129	0.747†	0.814†	0.897†	0.832†	0.874†	0.916†	0.808†	1	
	A .holophylla	0.250	0.733†	0.604*	0.836†	0.632*	0.889†	0.707*	0.808†	1	
	C. obtusa	0.682*	0.889†	0.540	0.827†	0.842†	0.794†	0.873†	0.945†	1	
	C. pisifera	–0.379	0.910†	0.654*	0.663*	0.813†	0.787†	0.850†	0.908†	1	
α–humulene	P. densiflora	0.062	0.971†	0.473	0.848†	0.574	0.915†	0.755†	0.759†	1	1
	P. koraiensis	0.145	0.760†	0.797†	0.883†	0.817†	0.856†	0.929†	0.781†	0.998†	1
	A .holophylla	0.227	0.589*	0.541	0.734†	0.524	0.842†	0.574	0.753†	0.998†	1
	C. obtusa	–0.181	0.205	0.495	0.359	0.300	0.395	0.252	0.232	0.973†	1
	C. pisifera	–0.548	0.427	0.861†	0.205	0.655*	0.393	0.554	0.275	0.088	1

Note: Values are expressed as Pearson's product moment correlation coefficient(n=3), *: $p<0.05$, †: $p<0.01$)

relations with bornyl acetate, camphene, carene, limonene, linalool, α–pinene, α–terpinene, β–pinene, and γ–terpinene, camphor showed negative (−) correlations with γ–terpinene, and myrcene and camphor showed positive (+) correlations with each other. In the essential oil extracted from the leaves of *Pinus koraien-*

sis, myrcene showed negative (−) correlations with camphene, terpinolene, α–pinene, γ–terpinene, trans–caryophyllene, and α–humulene and in the essential oil extracted from the leaves of *Abies holophylla*, myrcene showed negative (−) correlations with bornyl acetate and trans–caryophyllene and camphor showed negative (−) correlations with bornyl acetate. In the essential oil extracted from the leaves of *Chamaecyparis obtuse*, myrcene showed negative (−) correlations with camphene, limonene, linalool, sabinene, terpinolene, α–pinene, α–terpinene, γ–terpinene, cedrol, and trans–caryophyllene. In the essential oil extracted from the leaves of *Chamaecyparis pisifera*, myrcene showed negative (−) correlations with bornyl acetate, camphene, limonene, linalool, p–cymene, sabinene, terpinolene, α–phellandrene, α–pinene, α–terpinene, β–pinene, γ–terpinene, cedrol, and trans–caryophyllene, eucalyptol showed negative (−) correlations with p–cymene, sabinene, α–phellandrene, α–terpinene, γ–terpinene, trans–caryophyllene, and α–humulene, and myrcene and eucalyptol showed positive (+) correlations with each other.

Zafra and García–Peregrín (1976) reported that in the essential oil extracted from the leaves of *Pinus syvestris*, the contents of sabinene and camphene were inversely proportional to each other, but in the present study, the results of analyses of *Pinus densiflora* and *Pinus koraiensis*, which are in the same genus as *Pinus syvestris*, were different.

The reason why the results of the present study are partially inconsistent with the previous studies is thought to be that the tree species studied were different. Although the genus was the same, individuals have genetic differences from each other, even if they are in the same species, and terpenic compounds are subject to genetic influences (Pauly and Rudloff, 1971; Franklin, 1976). In addition, even the same individual contains different terpenic compounds among the branches or locations where samples were collected (Rudloff, 1972; Rudloff and Granat, 1982; Sim and Ahn, 1989) and the contents of terpenic compounds are different among growth and development environments, as well (Wilkinson *et al*., 1971; Son and Hwang, 1990). In addition, various enzymes involved in the biosynthesis of essential oil in mevalonate pathways, which are biosynthesis metabolism paths, are affected by seasonal changes, temperatures, and photosynthesis (Abad Farooqi *et al*., 1993), and the damage by diseases and insects or pests suffered during growth and development can also be a cause of the differences (Wilkinson, 1980; Harris *et al*., 1983).

Because changes in the concentrations of terpenic compounds are affected by diverse factors as such, continued studies are necessary to establish correlations. However, if essential oils are to be extracted from the leaves of coniferous trees and used in aromatherapy, flavoring agents, or deodorizing agents, according to the results of studies thus far, extracting essential oils in spring and summer when the concentrations of terpenic compounds are high is judged to be most effective.

CONCLUSTIONS

The present study was conducted to compare the contents of various terpenic compounds contained in essential oils extracted from the leaves of five species of coniferous trees to identify tree species that contain relatively high concentrations of terpenic compounds and determine seasonal changes in the concentrations of the terpenic compounds. In addition, the present study was also intended to compare terpenic compounds contained in the essential oils and those that are volatilized from the leaves through stomas to determine whether the terpenic compounds contained in the leaves actually volatilize to the air.

According to the results, first, the concentrations of terpenic compounds were relatively lower in essential oils extracted from the leaves of *Pinus densiflora* and *Pinus koraiensis* compared to *Abies holophylla*, *Chamaecyparis obtusa*, and *Chamaecyparis pisifera*. In addition, the concentrations of eight kinds of terpenic compounds in the leaves of *Abies holophylla*, nine kinds in the leaves of *Chamaecyparis obtuse*, and three kinds in the leaves of *Chamaecyparis pisifera* were particularly higher compared to other tree species. However, as for the concentrations of useful terpenic compounds volatilized through stomas in airtight containers, eights kinds in the leaves of *Pinus densiflora*, 16 kinds in the leaves of *Pinus koraiensis*, 10 kinds in the leaves of *Abies holophylla*, one kind in the leaves of *Chamaecyparis obtuse*, and two kinds in the leaves of *Chamaecyparis pisifera* showed relatively higher concentrations compared to other tree species. Therefore, woodland walks or activities for forest healing in *Pinus densiflora*, *Pinus koraiensis*, or *Abies holophylla* forests are judged very effective.

Second, the seasonal total quantities of the 21 kinds of useful terpenic compounds contained in the essential oils extracted from the five species of coniferous trees were large in summer in the case of *Abies holophylla* and in spring in the cases of *Pinus densiflora*, *Pinus koraiensis*, *Chamaecyparis obtusa*, and *Chamaecyparis pisifera*. In addition, the concentrations of various kinds of terpenic compounds by kind were found to be high in spring and summer through analyses. However, the concentration of myrcene was shown to commonly increase as seasons changed from spring to summer, autumn, and winter in order of precedence in the essential oils extracted from the leaves of the five species of coniferous trees and showed negative (−) correlations with most terpenic compounds. Therefore, if essential oils are to be extracted from the leaves of coniferous trees and utilized in forest healing, extracting essential oils in spring and summer when the concentrations of terpenic compounds are high is judged most effective.

AUTHOR CONTRIBUTIONS

C. S. SHIN designed the study and wrote the paper. J. W. LEE and P. S. YEON performed the chemical analy-

sis. C. S. SHIN and J. Y. CHA performed data analysis. P. S. YEON and S. OHGA designed the study, supervised the work. All authors assisted in editing the manuscript and approved the final version.

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