Chemical Characteristics of Heartwood and Sapwood of Red-Heart Chinese Fir (*Cunninghamia lanceolata*)

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Abstract

The chemical composition of wood is an important factor affecting the properties and utilization of wood. To compare the difference in chemical compositions between heartwood and sapwood of red-heart Chinese fir (*Cunninghamia lanceolata*), three graded wood, dominant, average, and overtopped trees were selected from the Chenshan Forest Station of Anfu County in Jiangxi Province. Their chemical composition parameters were determined according to international standards. Our results indicate that sapwood contains on average a higher moisture content than that of heartwood, constituting 9.4 and 8.6 percent, respectively. The pH values of wood present acidic and are higher in sapwood. Cellulose is abundant in both wood tissues; the heartwood content (52.0%) is higher than that of sapwood (48.6%) on average. Furthermore, the lignin in heartwood is slightly less than that of sapwood. Hemicellulose content is similar in heartwood and sapwood (23.4% vs. 23.1%), on average. All kinds of extractives in heartwood are substantially richer. Approximately three times more benzene–ethanol extractives are in heartwood than sapwood. This suggests that there is a considerable variation of chemical constituents among the graded woods ($P < 0.05$). The pH values are both significantly correlated with the contents of the four extractives in heartwood and sapwood ($P < 0.05$). The longitudinal variation of chemical compositions is different along the direction of tree height in heartwood and sapwood. Understanding the chemical heterogeneity of wood is vital for wood product manufacturing as well as for wood property improvement.

Chinese fir (*Cunninghamia lanceolata* (Lamb.) Hook), which is one of the most important native tree species in China, exhibits excellent timber quality and fast growth (Tian et al. 2011a, 2011b). Because *C. lanceolata* is also an important commercial species appreciated for its high-quality timber, it plays an important role in wood production practice of plantations (Hu et al. 2018, Wen et al. 2018). Red-heart Chinese fir originates in Jiangxi Province and is well known for the beautiful color of its heartwood, the xylem part that lies close to the pith, which is called ''red-heart'' (Fan et al. 2015, Liao et al. 2018). Compared with other *C. lanceolata* cultivars, this species has great

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resistance, strong adaptability, excellent material quality, and a fast growth rate (Yan 2013). As a result, the unique red-heart Chinese fir variety is regarded as one of the best timbers, with excellent weather and pathogen resistance for use in construction, furniture, and decorative applications. Because of the growing number of construction projects and the improvement of living standards across China, red-heart Chinese fir has broad market prospects, and it is in increasing demand (Li 2008). Plantations where red-heart Chinese fir are cultivated have expanded rapidly in recent years and now account for a large proportion of total *C. lanceolata* plantings (Wen et al. 2018). However, the availability of high-quality timber from this conifer remains limited, since its current population is restricted to a narrow area (Wen et al. 2018). Therefore, several strategies for the industrialized development and protection of red-heart Chinese fir have been proposed (Yang and Zeng 2003).

Recent developments in red-heart Chinese fir studies have led to a renewed interest in rapid propagation, growth, afforestation, and genetic improvements. These studies aimed to increase the end-use value of red-heart Chinese fir, thereby strengthening the sustainability of the species. Recently, many studies have been conducted with the themes of tissue culture (Cheng et al. 2015, Yang 2015, Qin et al. 2017, Han et al. 2018), genetic variation (Zeng et al. 2001, Zhang 2009), DNA extraction (Wu et al. 2012), and the rhizosphere effect (Yin 2014; Liu et al. 2016, 2017). However, the chemical composition of red-heart Chinese fir has received scant attention in the research literature.

The chemical composition of wood is a key factor in the properties and use of wood (Nie and Lin 1998). The contents of the major chemical components (i.e., cellulose, hemicellulose, lignin, and extractions) significantly affect the physical and mechanical properties of lignocellulosic materials (Hung et al. 2017). Wood extractives, which are important components of wood, are distributed within the structure of wood (e.g., in the conduit, ray, resin channel, and axial parenchyma). Although the overall content of wood extractives is small, their influence on wood cannot be underestimated. The extractives are closely related to the color, odor, acidity, and alkali resistance of the wood, which can affect the physical properties and processing performance of the wood (Song 2012). Moreover, the extractives endow wood with ever-changing wood color and properties, which provide a good basis for wood identification.

Knowing the chemical composition of wood is vital for wood selection, wood processing, wood property improvement, wood identification, and other applications (Chen 1995). Therefore, the specific objectives of this study were to (1) determine and compare the chemical composition of heartwood and sapwood from graded wood of red-heart Chinese fir, (2) examine the correlations among the chemical composition of heartwood and sapwood, and (3) analyze the variation in the chemical composition of heartwood and sapwood along the directions of the trunk height and radial diameter.

Materials and Methods

Study sites

The study was undertaken at the Chenshan Forestry Station (27°14′N, 114°19′E) in Anfu County, Jiangxi Province. The elevation there ranges from 140 to 150 m above sea level, and the region experiences the typical humid characteristics of subtropical regions. The mean annual temperature in the region ranges from 5.9°C (January) to 28.9°C (July), averaging 17.7°C, and annual mean precipitation and rainfall days are 1,663 mm and 166 days, respectively. The average annual sunshine duration is 1,649 hours, and the annual frost-free period is 279 days (from 1970 to 2017). The soil type is acidic, with pH 4.27 to 4.73. There are *Sapium sebiferum*, *Vernicia fordii*, *Phyllostachys edulis*, and *Choerospondias axillaris* in the understory vegetation layer.

Sampling and sample preparation

All sample trees were obtained in September 2017. We selected and cut down three graded wood, with no obvious disease and good growth, including dominant, average, and overtopped trees (Table 1). Discs, thicknesses approximately 5 cm, were taken at distances of 0, 1.3, 3.6, 5.6, 7.6, and 9.6 m from the base of each trunk. Samples were brought back to the laboratory and placed in a ventilated and shaded area. After all discs were fully air-dried, the sapwood and heartwood were separated from each disc and were splintered, dried outdoors, and ground in a Wiley cutting mill (Model 4 Bench, 115 V, 50/60 Hz). The wood meal obtained was classified based on the mesh opening. Chemical analyses were performed with the sawdust fraction containing particles from 40 mesh (425 μm) to 60 mesh (250 μm) according to the T 257 cm-85 standard (Technical Association for Pulp and Paper Industry [TAPPI] 2000). Samples were stored in 10-mL glass bottles in dry conditions before analysis.

Determination of chemical composition

The moisture content was the ratio of the mass lost to the original mass of the sample (1 to 2 g) when the sample was oven-dried at 105°C ± 2°C (Boxun, GZX-9030 MBE, 220 V ± 10 V, 50 Hz; China) to constant weight, according to GB/T 2677.2-2011 (Standardization Administration of the People's Republic of China [SAC] 2011). The pH values were measured with the pH meter (PXS-270, 220 ± 22 V, 50 ± 1 Hz; China), according to GB/T 6043-2009 (SAC 2009).

The ash content was determined in accordance with TAPPI T211om-85 (1991) by incinerating samples of wood meal (approximately 2 g) in porcelain crucibles in a furnace (TAISITE, SX-4-10, 220 V, 50 Hz; China) with sufficient ventilation to enable complete incineration. The temperature was first increased to 300°C to calcinate the sample and then increased to 500°C. The temperature was held at 500°C for 2 hours to ensure complete incineration. The results were given in weight-percentage dry matter. The extractives contained in the wood meal (2 g) were determined according to TAPPI T-207om-88 (1988a), TAPPI T-212om-88 (1988b), and TAPPI T-204om-88 (1988c).

The conventional method for determining the cellulose content of wood is to remove the lignin from the sample of the tree with sodium chlorite when pH is 4 to 5. The content of holocellulose obtained is the total amount of cellulose, partial hemicellulose, and lignin retained in the whole part of the plant. Therefore, the measured holocellulose is a mixture, which does not separate cellulose, hemicellulose, and lignin, and its content does not really reflect the cellulose content in the sample but is a general nutrient determined under compulsory regulation (Feng et al. 1994).

Table 1.—Characteristics of the sampled red-heart Chinese fir.[a]

Graded wood	BD/cm	DBH/cm	DMH/cm	H/m	C/m	T/year
Dominant tree	29.6	24.0	19.4	16.9	2.5	25
Average tree	23.7	17.4	14.0	16.1	1.5	25
Overtopped tree	13.8	11.0	7.8	12.3	1.4	25

[a] BD = basal diameter; DBH = diameter at breast height; DMH = diameter at middle height; H = tree height; C = crown width; T = tree age.

In this study, we used the methodology described by Goering and Van Soest (1970) to determine the contents of cellulose, hemicellulose, and lignin. This method made up for the defects of the conventional method; additionally, it was easy to operate and can be carried out in a standard laboratory (Feng et al. 1994, Chen and Guo 2011).

Statistical analysis

To verify the significance of differences between the individual analyzed parameters, analysis of variance and Tukey's multiple comparison test ($P < 0.05$) were performed using the two wood tissues (heartwood and sapwood) and the different chemical components in each tree as criteria. Analogously, Pearson correlation analysis determined the relationship of chemical components in heartwood and sapwood. All statistical analyses used Excel and R 3.3.3 software.

Results and Discussion

Chemical compositions of heartwood and sapwood

The chemical compositions of heartwood and sapwood from the graded wood of Chenshan red-heart Chinese fir are given in Table 2. Considerable chemical differences are observed between heartwood and sapwood.

The moisture content of sapwood was higher than that of heartwood, with 9.44 and 8.63 percent on average, respectively. This was consistent with the research of Mertoğluelmas (2003), in which the moisture content decreased during the transformation process of sapwood into heartwood in *Pinus* species. Hygroscopic properties of wood cell walls differ between sapwood and heartwood, which may be one of the reasons for the different moisture contents of heartwood and sapwood (Song 2012). Wood environmental regulation ability, dimensional stability, and physical and mechanical properties are related to the cells' moisture content. A large number of published studies have discussed and analyzed the changes in the viscoelastic energy of wood under different moisture contents, providing a reliable basis for the study of viscoelastic properties of different polymers in the cell walls (Kelley et al. 1987, Lenth and Kamke 2001, Jiang and Lu 2008).

Most wood is slightly acidic, while a few species are slightly alkaline (Li and Xiang 1963). Our results showed that the pH values varied from 4.69 to 5.78 in heartwood and sapwood, reflecting acidic characteristics. The pH values of sapwood were higher than those of heartwood. The amount of ash was very small in both heartwood and sapwood, below 1 percent, but there was a great difference between heartwood and sapwood, which were 0.30 and 0.16 percent on average, respectively. The above results were consistent with previous studies (Fan et al. 2015).

As previously reported, cellulose was abundant in both wood tissues, with a slightly higher content in sapwood than in heartwood (Timell 1986, Shupe et al. 1997, Fengel and Wegener 2003). Our results showed that the sapwood and heartwood cellulose were 48.62 and 51.98 percent on average, respectively. Only small differences in hemicellulose content have been observed between heartwood (23.39%) and sapwood (23.10%) on average (Table 2), which were consistent with the findings of Holmbom et al. (2000). In addition, heartwood contained slightly less lignin than sapwood, contrary to the study results on Norwegian spruce (Bertaud and Holmbom 2004). On the one hand, all samples in this experiment were obtained from an area of mature wood; thus the influence of young wood was avoided. On the other hand, the analysis and detection methods used in the current study differed from those of Bertaud and Holmbom (2004). And different sampling times may also cause the growth differences.

All types of extractives were substantially more abundant in heartwood than in sapwood, as Mertoğluelmas (2003) and Miranda et al. (2017) reported. The benzene–ethanol extractives of heartwood (4.14% to 5.26%) were almost two times greater than that of sapwood (1.42% to 1.71%). The heartwood of red-heart Chinese fir is redder and darker than the sapwood, and these characteristics may be related to the benzene–ethanol extractive contents (Fan et al. 2001). Furthermore, the heartwood may contain a large number of color-developing components that are easily soluble in

Table 2.—Summative chemical compositions of the sapwood and heartwood of red-heart Chinese fir.[a]

Items	Dominant tree		Average tree		Overtopped tree	
	Heartwood	Sapwood	Heartwood	Sapwood	Heartwood	Sapwood
Moisture content (%)	7.70 ± 0.46 A a	8.98 ± 0.61 A b	8.44 ± 0.81 AB a	9.56 ± 0.70 A b	9.76 ± 0.48 C a	9.79 ± 0.17 A a
Ash (%)	0.18 ± 0.03 A a	0.31 ± 0.07 A b	0.15 ± 0.07 A a	0.29 ± 0.07 A b	0.15 ± 0.05 A a	0.30 ± 0.07 A b
pH	4.71 ± 0.10 A a	5.35 ± 0.13 A b	4.69 ± 0.29 AB a	5.40 ± 0.15 AB b	5.66 ± 0.12 C a	5.78 ± 0.05 C b
Cold water (%)	3.16 ± 0.98 A a	3.04 ± 1.46 A a	3.14 ± 1.07 A a	1.82 ± 0.73 A b	2.18 ± 0.44 A a	1.67 ± 0.50 A a
Hot water (%)	5.80 ± 0.30 A a	4.52 ± 0.59 A b	5.52 ± 1.38 AB a	3.81 ± 1.07 A b	4.23 ± 0.59 B a	3.41 ± 0.22 A b
1% NaOH (%)	13.89 ± 0.94 A a	11.79 ± 2.46 A a	12.53 ± 2.45 AB a	8.49 ± 1.94 B b	9.56 ± 1.36 C a	7.27 ± 1.21 BC b
Benzene–ethanol (%)	5.26 ± 0.69 A a	1.47 ± 0.47 A b	5.30 ± 1.40 A a	1.71 ± 0.64 A b	4.14 ± 0.53 A a	1.42 ± 0.43 A b
Cellulose (%)	49.23 ± 0.51 A a	52.78 ± 0.67 A b	48.67 ± 0.81 A a	52.04 ± 0.76 AB b	47.96 ± 0.33 A a	51.11 ± 0.43 B b
Hemicellulose (%)	20.66 ± 0.95 A a	21.66 ± 1.05 A a	21.94 ± 1.65 AB a	20.83 ± 1.82 AB a	27.57 ± 4.93 C a	26.82 ± 3.66 C a
Lignin (%)	33.81 ± 1.06 A a	34.16 ± 1.06 A a	32.65 ± 1.41 AB a	33.0 ± 1.27 AB a	31.18 ± 0.67 B a	31.53 ± 0.67 B a

[a] Values are mean ± standard deviation. Different lowercase letters indicate significant difference among heartwood and sapwood ($P < 0.05$). Different uppercase letters indicate significant difference among graded wood ($P < 0.05$).

organic solvents, leading to the deepening of the color of the heartwood.

In comparison with other coniferous species, the wood extractives were generally enriched in heartwood, although the extent of enrichment varied depending on the species (Lu and Qin 1993, Mertoğluelmas 2003).

Overall, the variations of chemical constitution showed significant differences within two tissues between the graded wood ($P < 0.05$; Table 2). Chemical composition variations were largely observed between dominant and overtopped trees in the heartwood and sapwood. The moisture content had the strongest difference between the dominant and overtopped trees ($P < 0.01$); there were significant differences in 1 percent NaOH extractives content, hemicellulose content, and lignin content ($P < 0.05$). This indicated that the same species have significant differences of chemical constituents among individuals.

Correlations between the chemical components of heartwood and sapwood

The correlations between the contents of chemical components in heartwood and sapwood from graded wood are shown in the Tables 3 and 4.

Significant differences ($P < 0.05$) were registered between the chemical compositions of heartwood from graded wood (Table 3). The moisture content in heartwood had a positive relationship with pH ($P < 0.01$) and negatively correlated with the contents of hot water extractives, cold water extractives, 1 percent NaOH extractives, and benzene–ethanol extractives ($P < 0.05$). The ash content in heartwood was positively correlated with hot water extractives ($P < 0.05$; Table 3). The pH values were both significantly correlated with the contents of the four extractives in heartwood and sapwood ($P < 0.05$), as previously noted (Liang and Luo 2004). To some extent, this indicated that the wood acidity or alkalinity was influenced by the content of extractives. The data from the study of Fan et al. (2015) suggested that the pH of red-heart Chinese fir was related to the content of 1 percent NaOH extractives.

The acid–base properties and extractives content were both important chemical properties in wood. It is of great significance for wood processing and utilization to study the acid-base properties, extract contents, and their relationship. Li and Xiang (1963) reported that when wood was stored in a moist environment, its acidity gradually increased, especially as the temperature increased. This was due to the hydrolysis of ethyl phthalein on wood hemicellulose to form acetic acid. This revealed that the pH value may be related to the hemicellulose content of wood. Our results showed that the pH values of heartwood and sapwood were significantly positively correlated with hemicellulose content ($P < 0.05$). Further work should be undertaken to investigate the specific reason.

All extractives exhibited significant positive correlations in heartwood ($P < 0.05$). Only the contents of cold water extractives in sapwood had a positive relationship with the contents of benzene–ethanol extractives ($P < 0.05$). The cellulose contents in heartwood were positively related with hot water extractives and benzene–ethanol extractives ($P < 0.05$), while they were negatively correlated with lignin content ($P < 0.01$). The hemicellulose content in heartwood

Table 3.—Pearson's correlation between the chemical components of heartwood.

Parameter[a]	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}
X_1	1									
X_2	−0.437	1								
X_3	0.747**	−0.174	1							
X_4	−0.701**	0.560*	−0.452	1						
X_5	−0.704**	0.434	−0.622**	0.683**	1					
X_6	−0.809**	0.411	−0.749**	0.726**	0.897**	1				
X_7	−0.477*	0.171	−0.500*	0.532*	0.914**	0.771**	1			
X_8	−0.037	0.200	0.058	0.516*	0.360	0.264	0.482*	1		
X_9	0.723**	−0.401	0.647**	−0.453	−0.685**	−0.777*	−0.605**	−0.080	1	
X_{10}	−0.339	0.066	−0.450	−0.179	0.005	0.184	−0.158	−0.688**	−0.487*	1

[a] * = $P < 0.05$, ** = $P < 0.01$. X_1 = moisture content; X_2 = ash; X_3 = pH; X_4 = hot water extractives; X_5 = cold water extractives; X_6 = 1% NaOH extractives; X_7 = benzene–ethanol extractives; X_8 = cellulose; X_9 = hemicellulose; X_{10} = lignin.

Table 4.—Pearson's correlation between the chemical components of sapwood.

Parameter[a]	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}
X_1	1									
X_2	0.086	1								
X_3	0.417	−0.239	1							
X_4	−0.271	0.369	−0.475*	1						
X_5	−0.335	−0.052	−0.548*	0.603**	1					
X_6	−0.362	0.262	−0.612**	0.901**	0.787**	1				
X_7	0.198	0.017	−0.322	0.159	0.481*	0.244	1			
X_8	−0.244	−0.244	0.192	−0.322	−0.302	−0.410	0.093	1		
X_9	0.299	−0.236	0.573*	−0.269	−0.249	−0.333	−0.149	0.076	1	
X_{10}	−0.441	0.455	−0.482*	0.655**	0.551*	0.644**	0.372	−0.047	−0.505*	1

[a] * = $P < 0.05$, ** = $P < 0.01$. X_1 = moisture content; X_2 = ash; X_3 = pH; X_4 = hot water extractives; X_5 = cold water extractives; X_6 = 1% NaOH extractives; X_7 = benzene–ethanol extractives; X_8 = cellulose; X_9 = hemicellulose; X_{10} = lignin.

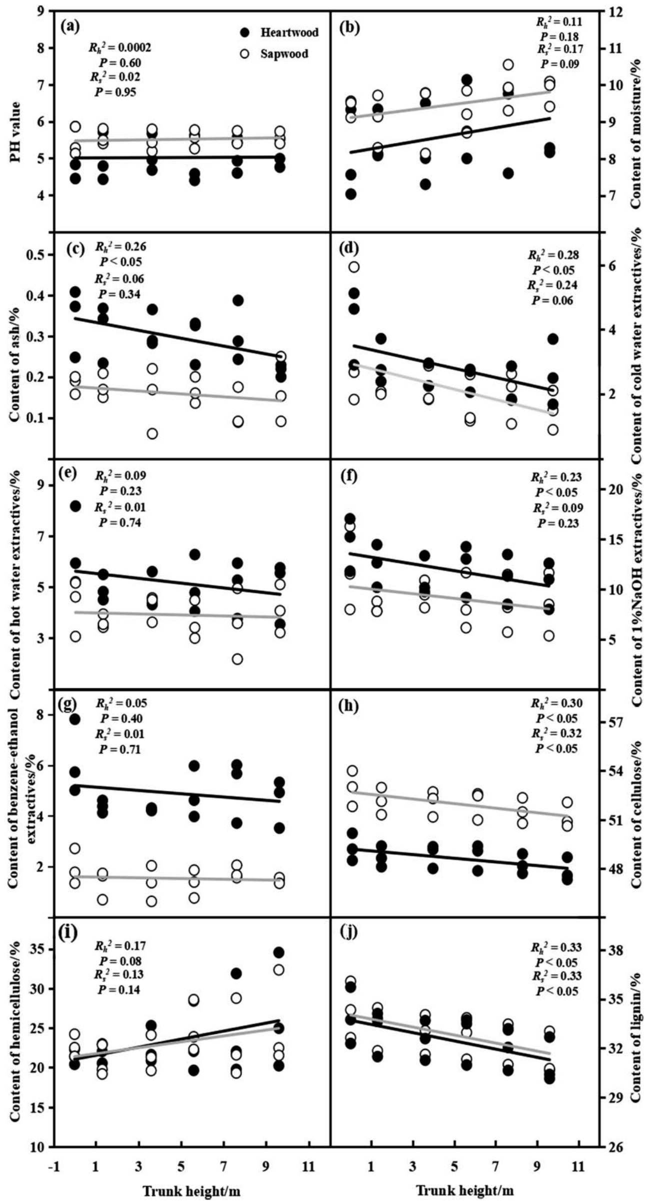

Figure 1.—Variation in the contents of 10 chemical constituents of heartwood and sapwood from graded wood with trunk height. For comparison, all data are presented as the average with standard deviation. (a) through (j) represent the pattern of variation of different chemical compositions with tree height, respectively.

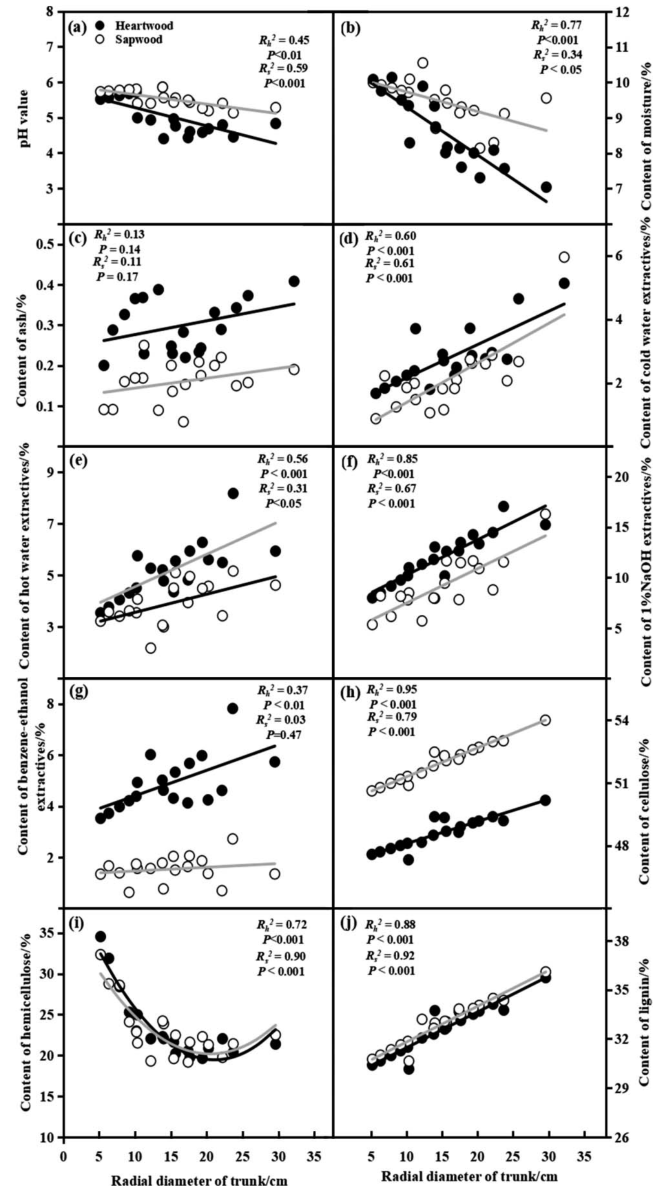

Figure 2.—Variation in the contents of 10 chemical constituents of heartwood and sapwood from graded wood with trunk radial diameter. For comparison, all data are presented as the average with standard deviation. (a) through (j) represent the pattern of variation of different chemical compositions with trunk radial diameter, respectively.

was positively correlated with moisture content ($P < 0.01$), while it was negatively correlated with cold water extractives, 1 percent NaOH extractives, lignin content, and benzene–ethanol extractives ($P < 0.05$). The lignin content of sapwood was positively correlated with cold water extractives, hot water extractives, and 1 percent NaOH extractives ($P < 0.05$), while it was negatively correlated with pH and hemicellulose content ($P < 0.05$). Although the current study was based on a small sample of trees, the findings suggested that each chemical component did not exist independently; instead, the chemical components interacted with each other. Further studies that take these interactions into account are needed.

The relationship between chemical composition, trunk height, and trunk radial diameter

In this study, we analyzed the longitudinal variation of chemical composition based on trunk height and trunk radial diameter in red-heart Chinese fir. With trunk height gradually increasing, the cellulose and lignin contents showed obvious decreases ($P < 0.05$), but the pH and moisture contents, as well as hemicellulose contents, did not significantly increase ($P > 0.05$; Fig. 1). The hot water extractives and benzene–ethanol extractives contents showed weak correlations with trunk height in two tissues. There were significant negative correlations between trunk height and the contents of ash, cold water extractives, and 1 percent NaOH extractives in heartwood ($P < 0.05$).

As the radial diameter of the trunk increased, the pH values and moisture contents obviously decreased (Figs. 2a and 2b), and the hemicellulose content first decreased and then increased in heartwood and sapwood (Fig. 2i). In contrast, the other chemical components increased with increasing trunk radial diameter in two tissues (Fig. 2). Except for the ash content ($R_h^2 = 0.13$, $P = 0.14$; $R_s^2 = 0.11$, $P = 0.17$; Fig. 2c), the correlations between the contents of all analyzed chemical constituents in heartwood and sapwood were significant with trunk radial diameter ($P < 0.05$). There were significant differences among the correlations between chemical components and trunk height and radial diameter. Our results were consistent with previous studies (Zeng 1998, Liang and Luo 2004).

The contents of the four extractives from heartwood and sapwood showed downward trends along the longitudinal direction of the trunk (Zhou et al. 1999, Fan et al. 2015). In other words, the extractives contents were negatively correlated with the distance from the root to the tree in the longitudinal direction. This can be explained by the physiological process by which wood absorbs water, nutrients, and other living substances from the environment through the roots and transfers them to various parts of the trunk through structures such as ducts (Zhai 2017). The contents of cellulose, hemicellulose, and lignin, which were the three major components of cell walls, varied in the longitudinal direction along tree height. The contents of cellulose and lignin decreased gradually with increasing trunk height, while the content of hemicellulose increased, consistent with the results of Li et al. (2007).

The chemical compositions differed with the change in tree heights in heartwood and sapwood. This vertical difference affects physical and mechanical properties in wood and reflects the heterogeneity of wood materials (Qin et al. 2004). Therefore, it is necessary to study the variation in the chemical components and how these variations affect the physical and mechanical properties of the wood, which will help to explain the causes of changes in wood properties in theory (Zobel and Buijtenen 1989, Hung et al. 2017).

Conclusions

This project was undertaken to compare the chemical composition differences of heartwood and sapwood from Chenshan red-heart Chinese fir and to reveal the patterns of chemical variation within this species. The results show that sapwood contains a higher moisture content than heartwood, and that pH values are slightly higher in sapwood than in heartwood, although both have acidic pH values. Cellulose is abundant in both wood tissues, although the cellulose content is higher in sapwood than in heartwood. Furthermore, heartwood contains slightly less lignin than sapwood. The total amount of hemicellulose is nearly the same in sapwood and heartwood. All kinds of extractives are substantially more abundant in heartwood than in sapwood. Significant differences in the contents of chemical constituents are observed between the dominant, average, and overtopped tree samples. The contents of the various chemical constituents of heartwood and sapwood are closely related to each other. The longitudinal variation of chemical composition of heartwood and sapwood in different grades of wood along the direction of tree height is similar. These results contribute to the literature about red-heartwood Chinese fir and provide a basis for development and use of red-heart Chinese fir.

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