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Dynamic distribution and biological storage analysis of procyanidins, arabinogalactan and dihydroquercetin in different parts of *Larix gmelinii*

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ABSTRACT

Every year, a large number of construction scrap wood and industrial processing residues are generated, not only caused heavy pressure on the environment, also the natural biological active ingredient in larch wood is wasted. In order to solve this urgent problem, the dynamic distribution and biological storage analysis of procyanidins, arabinogalactan and dihydroquercetin extracted by ultrasound-assisted extraction (UAE) in different parts of larch wood was studied. The results show that pith and sapwood have the highest and lowest moisture content, respectively. Moisture content also exhibited an increase from outer to inner heartwood. However, moisture content tended to decrease from 13.11 to 6.99% with increasing latitude. Most PCs were exclusively distributed in the bark and sapwood. The PC content of bark was about 10–20 times higher than that in sapwood, which increased with latitude increasing in bark. The highest PC content was 601.94 mg/g at 14 m height. AG content in the outer heartwood and sapwood was higher, and was lower in pith. Bark contained almost no AG. AG content was increased with the latitude increasing, and the highest content of AG was found at 6 m in sapwood. The peak of DHQ content in outer heartwood was 31.58 mg/g at the ground 0 m, where it was lower in mid- and inner heartwood than that in outer heartwood, and was lowest in bark (0.08 mg/g). Two peaks of DHQ content were observed with increasing latitude (at 0 and 6 m, respectively).

1. Introduction

Larix gmelinii (syn. *L. dahurica*; Pinacese), a species of larch (Gernandt and Liston, 1999), is endemic to the forests of northeastern China, an area with a cool temperate climate in the northern hemisphere. *L. gmelinii* is dominant in the boreal forests of Russia and Canada (Gros-Louis et al., 2005). In the northeast region in China, because of the clear and elegant texture, strong natural corrosion resistance, larch wood as main building materials, a large number of construction scrap wood and industrial processing residues are generated every year, not only caused heavy pressure on the environment, also the natural biological active ingredients in larch wood are wasted. Therefore, it is more important to investigate the distribution of active ingredients in larch wood, and that can create the enormous economic benefits for the secondary utilization of the scrap wood. It is also an urgent problem of industrial and building field for a long time. Previous research has shown that the active compounds in bark of *Larix gmelinii* were procyanidins (PCs) (Yang et al., 2011), and the active compounds in xylem were saccharides (mainly arabinogalactan, AG) (Huang et al., 2007) and dihydroquercetin (DHQ) (Xu and Bao, 2010).

PCs as condensed tannins are condensed by different numbers of (+)-catechin and/or (+)-epicatechin and/or gallic acid units linked mainly through C4-C6 or C4-C8 bonds. PCs constitute a class of oligomeric and polymeric polyphenols with flavan-3-ols as monomeric building blocks (Hellenbrand et al., 2015) (Fig. 1a). *Larix gmelinii* bark polyphenols have been reported to contain oligomeric procyanidins (OPC) consisting of 2–6 flavanol units, together with polymeric procyanidins (PPC), consisting of more than six flavanol units. PCs have attracted attention in the fields of pharmacology and food chemistry. Such as used for to stabilize substances and

Abbreviations: PCs, procyanidins; AG, arabinogalactan; DHQ, dihydroquercetin.

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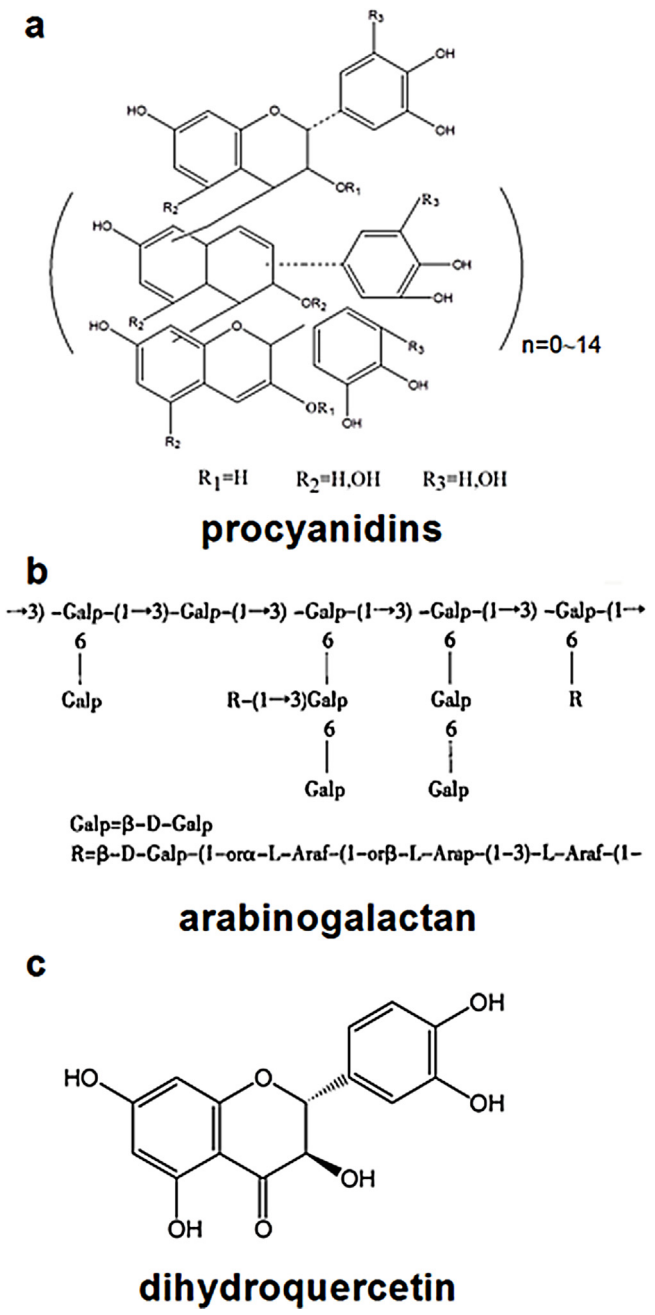

Fig. 1. Chemical structure of procyanidins (PCs), arabinogalactan (AG), and dihydroquercetin (DHQ) in *Larix gmelinii*.

counteract the oxidation processes of colorants and fats as well as to improve the taste of food (Li and Deinzer 2009; Malien-Aubert et al., 2009), and also used for their antioxidative, antimicrobial, antiviral, antimutagenic and wound healing properties. Moreover, researchers have documented significant effects of PCs related to promoting hair-growth (Takahashi et al., 1999), possessing anti-allergy effects (Kanda et al., 1998) having inhibitory activity against enzymes and receptors (Yanagida et al., 2000), to improve the functioning of vascular and cardiac systems (Holt et al., 2012; Arranz et al., 2013).

AG and DHQ are active compounds in larch xylem but have usually been considered to be wastes during the processes of pulping and papermaking, but they also have many pharmacological effects.

AG makes up about 15–30% of the woody tissue of the larch wood (Medvedeva et al., 2003). Larch AG is a highly branched polysaccharide consisting of β-D-galactopyranose, α-L-arabinofuranose and β-L-arabinopyranose residues (Ernest and Darning, 1997) (Fig. 1b). In 1974, the US Food and Drug Administration generally recognized AG as being safe for human consumption (Goellner et al., 2011), and used commercially as a dietary supplement (Esther et al., 2011) or a food ingredient because of its water-soluble and non-viscous properties (Marett and Slavin, 2004). Moreover, many pharmacological effects of larch AG have been reported, such as anti-inflammatory (Medvedev et al., 2002), gastro-protective (Medvedev et al., 2002), membranetropic (Josephson et al. Patent), and immune-modulating activities and effects (Peng et al., 2016). Additionally, AG can be converted into more valuable products through a sugar platform (Ernest and Darning, 1997). Therefore, AG has been recognized as a multi-purpose natural product with great economic potential and environmental value, which has attracted increasing attention by researchers.

DHQ is a type of flavanone alcohol compound that is present in *Larix gmelinii* (An et al., 2008), and is also known as taxifolin (2-(3,4-dihydroxyphenyl)-2,3-dihydro-3,5,7-trihydroxy-4H-benzopyran-4-one) and vitamin P. The chemical structure was shown in Fig. 1c. DHQ has many biological activities, including antioxidant in cerebral ischemia (Plotnikov et al., 2000), asbestosis (Kostyuk and Potapovich 1998; Kostyuk et al., 2001), experimental hepatitis in rats (Teselkin et al., 2000), and also in an *in vitro* model of osteoarthritis (Jovanovic et al., 2002), in protecting cells against oxidative stress (Maroziene et al., 2000), as well as having anti-radiation (Sugihara et al., 1999), anti-viral (Chu et al., 1992), anti-tumor (Kawaii et al., 1999) and scavenging free radical properties (Trouillas et al., 2004). The antioxidant properties of DHQ can be comparable or superior to many synthetic or natural antioxidants because of its rich content of phenolic hydroxyl groups. DHQ was also found to exhibit unique properties effectively inhibiting cellular melanogenesis while concurrently increasing tyrosinase protein levels (An et al., 2008). Moreover, it is not toxic to the fetuses, and is teratogenic, mutagenic or allergenic.

The ultrasound-assisted extraction (UAE) method was used to obtain the PCs, AG, and DHQ in larch wood. UAE has been employed in preparing bioactive compounds from different plant materials in recent years, and has been proved to be effective; it contains essential oils (Kubilay Tekina et al., 2015), phenolic acid (Yang and Wei, 2016), polysaccharides (Yan et al., 2011), flavonoids (Guamán-Balcázar et al., 2016), lignans (Ma et al., 2011), and alkaloids (Ma et al., 2012) and so on. In particular, UAE has been also employed in extracting anthocyanins and PCs (José Luis Pasquel Reátegui et al., 2014), AG and DHQ (Ma et al., 2014) from plant materials.

Therefore, the primary aim of the present study is to determine of the content of PCs, AG, and DHQ in the different parts of larch wood with UAE, and to analyze the dynamic distribution conditions and biological storage related to PCs, AG, and DHQ in *Larix gmelinii*, in order to facilitate efficient use of scrap wood.

2. Materials and methods

2.1. Materials

2.1.1. Wood materials

Larix gmelinii trees approximately 18 m tall with about 12.0 cm diameter at breast height (DBH) were selected from the MaoEr Mountain Experimental Forest Farm (MaoEr Mountain Forest Park), a teaching, scientific research, and practice base of Northeast Forestry University (Greater Khingan Mountains area, Heilongjiang, China). Samples of bark and xylem were collected at every 2 m in height with a thickness of about 4 cm (If we sampled every 2 m, the number of samples was no less than 7, to ensure the accuracy of

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