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Ethyl lactate as a potential green solvent to extract hydrophilic (polar) and lipophilic (non-polar) phytonutrients simultaneously from fruit and vegetable by-products

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ABSTRACT

Phytonutrients extracted from natural resources are receiving much attention among researchers due to their highly antioxidative characteristics which prevent several degenerative diseases including cardiovascular diseases and cancers. These nutraceutical compounds can be used in food, pharmaceutical and cosmetic products as natural antioxidants, preservatives, colourants and functional foods. Huge volume of food wastes are generated from the processing industry and these low-value food residues are rich in various phytonutrients worth recovering. This approach of valorisation reduces the generation of food wastes and is cost-effective considering the cheap feedstock, reduced waste management expenses and high market value of extracted compounds. In light of the health and safety risks posed by commonly used organic extraction solvents derived from the petrochemical industry, there is a need to recover the phytonutrients using green, sustainable and efficient solvents that are safe for human consumption. This work discusses ethyl lactate as a safe, green, efficient and potentially cheap solvent to recover phytonutrients from fruit and vegetable by-products. Ethyl lactate is compared with other organic solvents commonly used from the aspects of safety, environmental impacts and efficiency. Current challenges when employing ethyl lactate are also discussed.

1. Introduction

As the Greek word '*phyto*' refers to plant, phytonutrients (or phytochemicals) are bioactive compounds found in plants. They provide plants with protection against adverse environmental and external treats such as radiation, heat, pathogens and insects. More than 25,000 of phytonutrients have been identified and the six important phytonutrients listed by WedMD are carotenoids, ellagic acid, flavanoids, resveratrol, glucosinolates and phytoestrogens (WedMD, 2016). Three of these (ellegic acid, flavanoids and resveratrol) are categorised under phenolic compounds. Phytonutrients can be obtained from a variety of fruits, vegetables, whole grains, legumes, nuts, seeds, herbs and spices. Most of the phytonutrients act as antioxidants and cancer-prevention agents which protect the body against damage from free-radicals. As compared to their synthetic counterparts, these natural antioxidants are favoured among consumers for use in food processing in order to improve the nutritional values as well as to lengthen the shelf life of the products. They are superior in preventing lipid peroxidation and acting as oxygen radical scavengers. The extracted phytonutrients is not just limited to the food industry and can also be integrated into pharmaceutics and cosmetics.

Though many attempts of extraction have been carried out to obtain the phytonutrients from various plants and herbs species, these highly valuable compounds can be recovered from industrial food residues or by-products. The presence of natural phytonutrients in plant foods is under-utilised due to destruction at extreme conditions during food processing. Furthermore, much of the food matrix containing these nutrients is removed and wasted due to undesirable palatability, texture, colour, flavour, taste and aroma. These large amounts of food wastes pose certain degree of

Abbreviations: GRAS, generally recognized as safe; PLA, polylactic acid; PLE, pressurized liquid extraction; SNAP, Significant New Alternatives Policy Program; USEPA, US Environmental Protection Agency; USFDA, US Food and Drug Administration

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environmental damage due to the presence of organic compounds which increase the biochemical and chemical oxygen demands in wastewater. The phenolic compounds are also reported to inhibit seeds germination (Negro et al., 2003). Due to high concentration of these phytonutrients in fruit and vegetable wastes from the industries, they represent potential alternative, sustainable and low-cost sources from which valuable nutraceutical compounds can be obtained. Extracting phytonutrients from fruit and vegetable by-products reduces the impact of food wastes to the environment whilst recovering high demand nutrients from natural and low value resources.

Nonetheless, the majority of commonly used organic extraction solvents are produced from the petrochemical industry. They are highly volatile, flammable, corrosive, carcinogenic and toxic. In light of this, there is a need to recover the phytonutrients using green, sustainable and efficient solvents that are safe for human consumption. Ethyl lactate (ethyl 2-hydroxypropanoate) which is an ester of lactic acid originating from the fermentation of carbohydrates feedstock is one such solvent. The solvent is environmentally benign as it is readily biodegradable to carbon dioxide and water. As an effective solvent, ethyl lactate is capable of dissolving in both aqueous (polar) and hydrocarbon (non-polar) mediums. Hence, it has the potential to recover compounds with a wide range of polarity without the presence of co-solvent. In 2005, the US Food and Drug Administration (USFDA) approved the use of ethyl lactate in food and pharmaceutical products, and it is generally recognized as safe (GRAS) solvent (US Food and Drug Administration, 2005). This work discusses ethyl lactate as a safe, green, efficient and potentially cheap solvent to recover phytonutrients from fruit and vegetable by-products. The phytonutrients of focus in this discussion are phenolic compounds and carotenoids. They are selected to represent polar and non-polar compounds, respectively. In the discussion, ethyl lactate is compared with other organic solvents commonly used from the aspects of safety, environmental impacts and efficiency. Current challenges when employing ethyl lactate are also presented.

2. Phenolics and carotenoids phytonutrients

The functional compounds extracted from fruit and vegetable by-products are classified into (1) insoluble (e.g. fibres), (2) water-soluble (e.g. phenolics) and (3) lipid-soluble (e.g. carotenoids) according to Schieber et al. (2001). Phenolic compounds and carotenoids are most abundant, and are commonly found in various fruit and vegetable wastes. Therefore, these two groups of compounds are targeted to represent polar and non-polar phytonutrients, respectively.

Phenolic compounds are naturally occurring organic compounds, which are commonly found in plants. They are the most abundant secondary metabolites in plants containing at least one benzene ring, with one or more hydroxyl groups. The structures range from simple molecule to complex high molecular mass polymer (Balasundram et al., 2006). Phenolic compounds are broadly classified into flavonoids, phenolic acids, tannins, stilbenes and lignans (Ignat et al., 2011). Flavonoids constitute half of the phenolic compounds and are characterised by the hydroxylation pattern around the central C-ring. They possess a high number of electron-donor hydroxyl groups and double bonds, which improve the overall antioxidant activity (Harborne and Williams, 2000; Tsuda et al., 1994). Most researchers concluded that phenolic compounds in wine are responsible in preventing heart disease. This is an effect frequently described as “the French paradox” (Zagklis and Paraskeva, 2015). Phenolic compounds also act as antioxidants and anti-inflammatory factors. They possess anticarcinogenic, antiviral, antibacterial, antiradical, antidiabetic properties (Ehrenkranz et al., 2005; Makarova et al., 2015; Palma and Taylor, 1999; Porto et al., 2014) and are protective against antherosclerosis, brain dysfunction and hypertension (Ignat et al., 2011; Sivaprakasapillai et al., 2009). They are also reported to protect high-density lipoprotein against oxidation (Meyer et al., 1997). In the industries, phenolic compounds could be used as natural preservatives, colourants and in the production of paints, paper and cosmetics (Bosso et al., 2016).

There are more than 600 known natural carotenoids and more than 100 can be found in fruits and vegetables. Carotenoids are classified into two groups of C_{40} hydrocarbon: carotenes and xanthophylls. They consist of an extensive system of conjugated double bonds, which absorbs light and imparts characteristic colours. In plants, harmful rays will be absorbed by the conjugated double bonds to protect the chloroplasts (Yahia and Paz, 2010). Being highly unsaturated with many conjugated double bonds, carotenoids are unstable towards isomerisation and oxidation. Carotenes such as lycopene and α-carotene are the major carotenes found in tomatoes and carrots, respectively. Xanthophyll β-crytoxanthin is prevalent in orange-flesh fruits such as peaches and papayas. Similar to phenolic compounds, carotenoids are natural antioxidants to fight against several degenerative diseases including cardiovascular diseases and cancers. They inhibit low-density lipoprotein oxidation and exhibit anti-inflammatory activity (Rafi et al., 2007; Rao and Agarwal, 1999). The antioxidant activity of lycopene is twice that of β-carotene and ten times higher than of α-tocopherol (Agarwal and Rao, 2000; Di Mascio et al., 1989; Urbonaviciene et al., 2012). Due to the distinctive colour of these compounds, they are frequently used in the food industry as nutritious food colourants as well as natural preservatives.

3. Phytonutrients from fruit and vegetable by-products

There is significant potential for the extraction of phytonutrients from fruit and vegetable by-products based on reported data in the literature. For fruits, 80% of the grapes harvested are used for wine making (Mazza and Miniati, 1993) while 20% of the total weight of the grapes is discarded as pomace (Rondeau et al., 2013; Schieber and Stintzing, 2001). As much as 5–9 million tonnes per year of wastes is produced by the wine industry to be used as cattle feed or soil conditioning (Meyer et al., 1998; Schieber et al., 2001). Pomace (or marc) is one of the richest sources of polyphenols because they are more concentrated in the skins or peels (Łata et al., 2009; Makris et al., 2007; Vrhovsek et al., 2004). Approximately 70% of the grape phenolics remained in the pomace (Mazza, 1995). The major phenolic compounds in grape pomace are flavan-3-ols, catechin, epicatechin and gallic acid (Lafka et al., 2007; Tsanova-Savova et al., 2005).

Approximately 70% of apples are marketed as fresh fruits while 30% are processed into juices, ciders and jams (Fromm et al., 2013). Approximately 30% of apple pomace and 10% of pomace sludge are produced from the juice-making industry (Dhillon et al., 2011). Up to 90% of the antioxidant activity is lost after juicing (Candrawinata et al., 2015) while 58% of the phytonutrients are lost into the apple pomace (Guyot et al., 2003). Only 3–10% of the antioxidant activity of the fruit is extracted into apple juice (Van Der Sluis et al., 2002). Dihydrochalcones, flavonols, flavanols and phenolic acids have been reported in apple pomace (Bhushan et al., 2008). Apple pomace can be used as cattle feed, substrate for ethanol and citric acid production, enzyme and pectin recovery for use as gelling agent, stabilizer and source of dietary fibre (García et al., 2009; Lavelli and Corti, 2011; Lohani and Muthukumarappan, 2015).

Banana peels constitute up to 30% of the ripe fruits. Both