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ORIGINAL PAPER

# Effect of Subcritical Water on the Extraction of Bioactive Compounds from Carrot Leaves

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## Abstract

Carrot leaves, which are generally considered as agricultural residue, are rich in bioactive compounds, such as polyphenols. This study investigates the extraction of polyphenols and luteolin (flavonoid) from freeze-dried and ground carrot leaves ($d < 100$ μm) using subcritical water (SCW). Water at elevated temperatures and at high pressure (40 bar) could behave as low-polar solvent to enhance extraction of organic compounds. SCW was investigated at different temperatures (110–230 °C), time (0–114 min), and solid-liquid ratio (15 and 35 g/L). Accordingly, it was revealed that total phenolic content (TPC) from carrot leaves using SCW has an increasing trend with temperature and resulted in 42.83 ± 1.85 mg per g of dry weight in gallic acid equivalent at 210 °C/113.5 min. However, luteolin content using SCW extraction behaved differently, where increase of temperature adversely affected its content. Hot water extraction studies revealed the presence of optimum luteolin content (0.768 ± 0.009-mg/g dry weight) at 120 °C for 10 min. In conclusion, it was shown that carrot leaves are a promising feedstock to extract polyphenols that has high content of luteolin.

**Keywords** Subcritical water · Carrot leaves · Polyphenol · Luteolin

## Introduction

Carrot or *Daucus carota* (Umbelliferae family), generally orange in color, was originated in Western Asia and then was introduced to New Zealand in the 1700s by the Europeans. It has become one of the top ten economically grown vegetable crops in the world (Simon et al. 2008). Carrot is consumed as a "root vegetable," which means carrot leaves are not accepted as an edible part by consumers because of its specific bitter and astringent taste. Commonly, the leaves are treated as an agricultural waste, which is separated after harvesting and used as animal feed or compost.

Bioactive compounds are present in both the carrot root and leaves (Kähkönen et al. 1999; Kaur and Kapoor 2002; Warman and Havard 1997). Carrot leaves are abundant in compounds, such as carotenoid, chlorophyll, vitamins, a variety of trace elements, and polyphenolic compounds, including luteolin and apigenin (Almeida et al. 2009; Bowman and Simon 2013; Leite et al. 2011). Kähkönen et al. (1999) reported that total phenolic compound (TPC) in carrot leaves is 7.4 mg of gallic acid equivalence (GAE) per g, over ten times more than that from fresh carrots (0.6-GAE mg/g dry weight).

The application of plant polyphenols in the food, cosmetic, and pharmaceutical industries is growing very rapidly. It is used as antioxidants, food pigments, nutrition supplements, and antiaging ingredients in a number of consumer products (Díaz-García et al. 2015; Sun et al. 2015; Xiao et al. 2011).

Polyphenols are a generic term for a class of compounds, which have one or more phenolic hydroxyl groups. They are plant secondary metabolites, which widely exist in the plant skin, root, fruit, and leaves. According to the nature of their carbon backbones, polyphenols are subdivided into four main classes: flavonoids, phenolic acids, the less common stilbenes, and lignans. Among them, flavonoids are the most abundant polyphenols in our daily intake of fruits and vegetables (Scalbert and Williamson 2000). Luteolin is a type of flavonoid having a chemical structure consisting of two benzene rings linked by a three-carbon chain that forms a closed pyran ring (Peng and Yan 2009). The structure of luteolin is as shown in Fig. 1.

Luteolin possess many biochemical properties like antioxidant, anti-inflammatory, and antibacterial activity due to the

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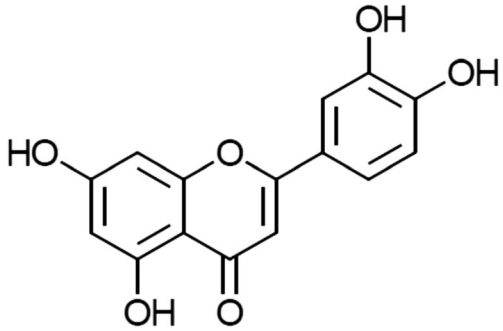


**Fig. 1** Chemical structure of luteolin

configuration, substitution, and total number of hydroxyl groups present in its structure (Chen et al. 2014; Roy et al. 2015; Wang and Xie 2010). It has been confirmed to play a role in the treatment of cancers, HIV-1, and also in the inhibition of Alzheimer's disease (Sawmiller et al. 2014).

Many researchers have worked on extracting polyphenolic compounds using solvent extraction from potential vegetables, fruits, leaves, oilseeds, roots, spices, and herbs. Generally, hot water, acetone, ethanol, and methanol are used as solvent extraction chemicals in industry (Babbar et al. 2011; Rababah et al. 2004; Turkmen et al. 2006). The yield of TPC depends on the species and organ of plants, extraction time, temperature, and type of solvents. Normally, TPC increases with the increase of extraction temperature and time in plant matter (Babbar et al. 2011; Rababah et al. 2004).

Solvent extraction is an operation with a low technical threshold, but on the downside, it takes a long time and/or demands high operating temperatures. For example, polyphenols from tea was extracted after 9 h of treatment with organic solvent, while it took 3 h 80 °C to extract bioactive compounds from ginseng (Turkmen et al. 2006; KyoungAh et al. 2013). In addition, it is reported that solvent extraction under high temperature for a long period of time may lead to degradation of heat sensitive polyphenols (Sólyom et al. 2014).

Subcritical water (SCW) extraction is one of the novel technologies that have been recognized as a "green technology." It is an extraction method using water at elevated temper atures between 100 and 374 °C with high pressure to maintain water in the liquid phase (Vergara-Salinas et al. 2012). At these conditions, the polarity (dielectric constant) of water decreases and behaves as a solvent showing enhanced extractability of polyphenols including flavonoids (Carr et al. 2011; Teo et al. 2010; Anekpankul et al. 2007). Vergara-Salinas et al. (2012) mentioned that SCW could be also effective in hydrolysis of lignocellulosic material, which is usually overlooked and thereby contribute to the release of phenolic cell wall-associated compounds. The effect of SCW on polyphenol extraction from different plant matters has been studied by many researchers (Chainukool et al. 2014; Ju and Howard 2005; Ko et al. 2011; KyoungAh et al. 2013; He et al. 2012). However, no work has been done on extraction of polyphenols and luteolin from carrot leaves using SCW. Therefore, this study was an attempt to explore the potential of using SCW technology to extract polyphenols and luteolin from carrot leaves. Different extraction conditions were investigated and the effect of important parameters of SCW were analyzed and compared.

## Materials and Methods

### Sample Preparation

Fresh carrots with intact leaves were purchased locally, washed with cold water, and was stored at 4 °C. Then, it was dried using a freeze dryer (BENCHTOP, VirTis). Dried samples were crushed using a coffee grinder (EM0405, Sunbeam) and graded into three ranges of particle size (< 100 μm, 100–200 μm, and 1 cm). Freeze-dried samples were stored in sealed containers for a few days at ambient temperature until extraction was carried out.

### Subcritical Water Extraction

SCW extraction was performed using a Parr Reactor (Parr Instrument Company, USA, Model 4540). Carrot leaf sample of particle size $d < 100$ μm was mixed with 500-mL deionized water to obtain either 15- or 35-g/L mixtures and was sealed in the treatment chamber. The stirring speed was maintained at 500 rpm, while the initial pressure was set at 40 bar. Nitrogen gas was used as protective gas and also to provide the initial pressure in order to keep water in liquid form. Extraction temperatures were set at 110, 180, 210, and 230 °C with sampling times of 0, 10, 30, 60, and 90 min. After each extraction, solid phase and liquid phase were separated and the filtrates were stored at − 20 °C until further analysis.

### Hot Water Extraction

Hot water extraction was carried out for carrot leaf sample of particle size $d < 100$ μm with concentration of 15 g/L using the autoclave (SS-325, TOMY). 1.5-g sample powder was mixed with 100-mL deionized water and heated to set temperatures of 85, 100, 110, or 120 °C, while sampling at 0, 5, 10, and 20 min.

### Total Phenolic Content

Total phenolic content (TPC) was determined by spectrophotometry using gallic acid as the standard according to the method adapted by Vázquez et al. (2015) with some modifications. A 20 μL of Folin-Ciocalteu reagent (Sigma Aldrich), 10 μL of sample solution, 130 μL of $Na_2CO_3$, and 140 μL of de-ionized water were added into each well of a 96-well plate.

The mixtures were incubated for 1 h, and the absorbance at 760-nm wavelength was measured using the micro-plate reader (EnSpire, PerkinElmer). The concentration of TPC in samples was expressed as GAE mg/g of dry weight of carrot leaves.

### Analysis of Luteolin

The analysis of luteolin in carrot leaf extract was adapted from the methods referred by Bhagat et al. (2014) and Rajasekaran et al. (2014) with modification. Shimadzu LC-20AT HPLC unit equipped with Lab Solutions software for the data acquisition together with a ZORBAX Eclipse C18 column (4.6 mm × 150 mm, 5 μm) was used for the sample separation and identification. The mobile phase consisted of 0.2% acetic acid in water (A) and HPLC grade methanol (B). A gradient elution program was used (0–3 min 5% B, 3–5 min 5–30% B, 5–30 min 30–80% B, 30–35 min 80% B, and 35–36 min 80–100% B). The sample injection volume was 30 μL. The chromatogram peaks were detected at 350-nm wavelength at a residence time of 27.7 min using a UV-detector (SPD-20A). Each sample was analyzed using HPLC in duplicates.

### Statistical Analysis

All experiments were carried out in triplicates. Microsoft Excel 2013 was used to perform the statistical analysis. Mean values were compared at confidence level of 95% ($p \leq 0.05$). The variability of the data was expressed as standard deviation, which was presented as error bar on results reported in figures.

## Results and Discussion

### Effect of Particle Size on Extraction

A preliminary study was conducted to investigate the effect of particle size using hot water extraction at 85 °C with three different particle sizes. Figure 2a, b shows the TPC and luteolin content obtained when the experiments were conducted at different times. As expected, TPC and luteolin content increased with the decrease of particle size significantly ($p \leq 0.05$) giving the highest value of 15.47 ± 0.59 GAE mg/g and 0.049 ± 0.001 mg/g, respectively, at the end of 20 min for particle size of $d < 100$ μm. This showed that size reduction of plant matter before extraction increased surface area, which in turn enhanced mass transfer of bioactive from plant material to the solvent. Mukhopadhyay et al. (2006) also showed that particle size of black cohosh (*Cimicifuga racemosa*) was an important parameter that influenced the rate of pressurized liquid extraction. Further, many previous researchers also mentioned that the use of a small particle size leads to higher extraction yields (Çam and Hışıl 2010; Mukhopadhyay et al. 2006).

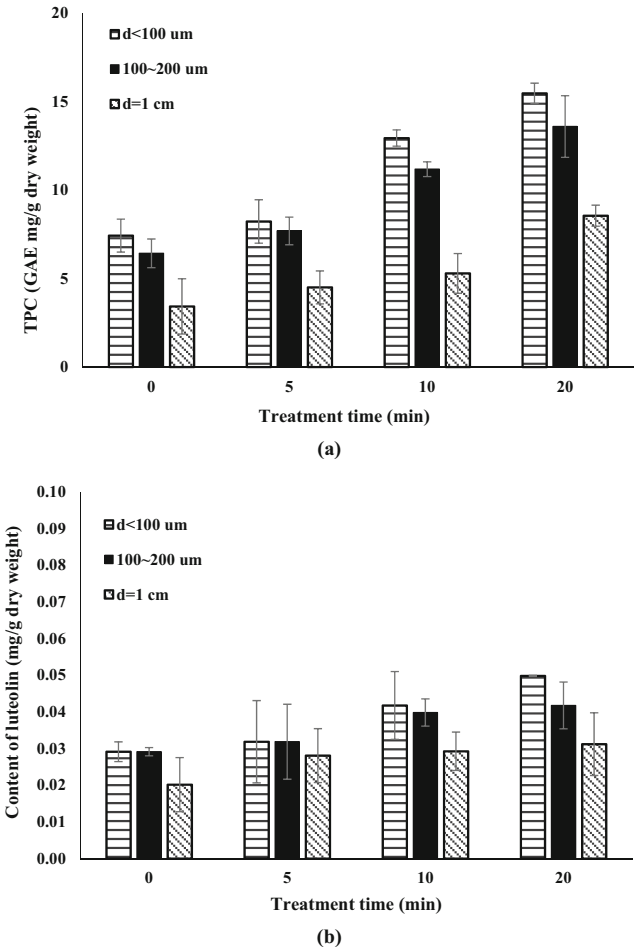


**Fig. 2** Effect of particle size on **a** TPC and **b** the luteolin content in the extract based on dry mass of carrot leaves at 85 °C

### SCW Extraction Studies

#### Influence of Solid-Liquid Ratio and Treatment Time

The effect of solid-liquid ratio in SCW extraction was studied using two different concentrations 35 and 15 g/L of carrot leaf suspension at 180 °C. The results presented in Fig. 3a show the concentration of polyphenol obtained with SCW extraction operating at different time intervals. TPC after 110 min of extraction at 35 and 15 g/L was 28.31 ± 0.49 and 30.62 ± 1.61-GAE mg/g dry weight, respectively. One-way ANOVA displayed that the solid-liquid ratio did not influence TPC in a significant manner ($p > 0.05$). The results are also supported by Silva et al. (2007) who studied the effect of solid-liquid ratio on TPC from *Inga edulis* leaves and found that the change of TPC was less affected by concentration of raw material when varied from 12.5 to 50 g/L. In another study, the impact of solid-liquid ratio on TPC extracted from grape seeds has also been

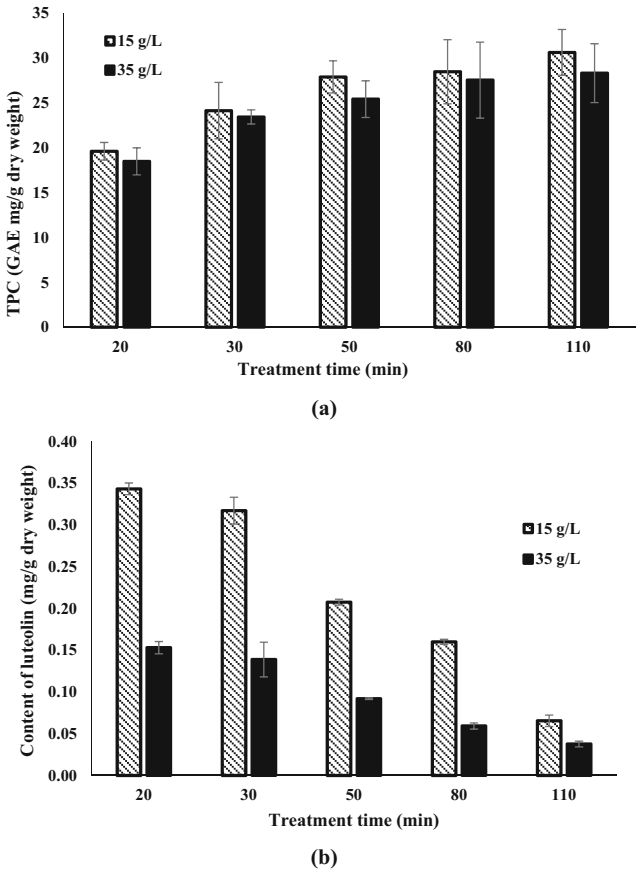


**Fig. 3** **a** TPC and **b** the luteolin content in the extract at solid-liquid ratios of 15 and 35 g/L when treated with SCW extraction at 180 °C

confirmed to be substantially unnoticed within a range of 25 to 50 g/L (Bucić-Kojić et al. 2007). The insignificant influence of solid-liquid ratio on extraction concentration of polyphenol from plant materials may be due to the high solubility of polyphenols in the extraction solvent (Silva et al. 2007).

During the same treatment, the variation in luteolin content with solid-liquid ratio and time is as shown in Fig. 3b. In contrast to polyphenols, the luteolin content that was extractable has reduced with the increase of solid/liquid ratio significantly ($p \leq 0.05$). At a solid-liquid ratio of 15 g/L, the bioactive compounds have more opportunity to diffuse and contact with the solvent than at 35 g/L. Han and Row (2011) showed that extraction of luteolin from celery using ultrasonication had an optimum solid-liquid ratio. Casazza et al. (2011) also confirmed this fact with grape seed extraction, mentioning that more phenolic compounds have the tendency to permeate into the solvent when a higher amount of solvent was used. Therefore, in the present study, higher luteolin content of 0.343 ± 0.015 mg/g was obtained in 15-g/L suspension, while it was only 0.153 ± 0.013 mg/g for 35 g/L. Figure 3b also shows a significant decrease in the content of luteolin with time for both 15 and 35 g/L confirming the tendency to degradation. Accordingly, the highest luteolin content of 0.343 ± 0.015 mg/g was obtained at 20 min of SCW treatment time, while it has reduced considerably to 0.065 ± 0.003 mg/g after 110 min when the solid-liquid ratio was 15 g/L. These studies show that a raw material concentration of 15 g/L was appropriate in subsequent studies of extraction of polyphenol and luteolin with SCW from carrot leaves.

## Influence of Temperature and Treatment Time

Figure 4a shows TPC extracted from carrot leaves at different temperatures and treatment times. TPC at the experimented temperatures of 110 to 210 °C increased initially and reached a stable value with time. Further increase of temperature has enhanced the extration of phenolic compounds to statistically significant levels ($p \leq 0.05$). The highest TPC of 42.83 ± 1.85-GAE mg/g dry weight was obtained at 210 °C when the treatment time was 114 min. Moreover, a treatment temperature of 230 °C gave a similar TPC of 41.79 ± 0.20 GAE mg/g at a treatment time of 84 min but shows a tendency to degrade at prolong treatment times. Several other investigators too considered that extraction temperature and treatment time are two significantly influencing factors that contribute to the efficiency of SCW extraction (Ju and Howard 2005; Kumar et al. 2011). Chainukool et al. (2014) reported that the amount of TPC extracted from barks of *Shorea roxburghii* increased from 0.063- to 0.3-mg/g dry weight with the increase of temperature (from 100 to 190 °C) when treated for 360 min. In another work, He et al. (2012) analyzed TPC from pomegranate seeds after 30 min of extraction at different treatment temperatures. They reported that TPC increased from 4.39-mg/g dry weight at 80 °C to 48.55-mg/g dry weight at 220 °C and subsequently reduced with the increase of temperature. Vergara-Salinas et al. (2013) reported that the amount of TPC extracted from grape pomace up to 150 °C using SCW was five times more than that collected at 50 °C. Other researchers mentioned that the range of the optimal extraction temperature of TPC with SCW extraction was from 100 to 220 °C (Kumar et al. 2011; KyoungAh et al. 2013).

Contrarily to polyphenols, the increase of temperature displayed a negative impact on the amount of luteolin extracted. As shown in Fig. 4b, the content of luteolin in the extract decreased as temperature increased. The decrease of luteolin content was statistically significant ($p \leq 0.05$) when the temperature was increased from 110 to 180 °C and kept reducing with further increase of temperature to 210 °C. In addition, luteolin content was decreased with treatment time at all temperatures.

As mentioned previously, the solubility of luteolin in water is less than that in organic solvent, such as ethanol and methanol. Peng and Yan (2009) reported that the solubility of luteolin increased with the percentage of ethanol in ethanol water mixture as solvent. This explains the fact that the closer the dielectric constant (polarity) of subcritical solvent to ethanol, the higher the efficiency of SCW extraction of luteolin. The dielectric constant of ethanol and methanol are 25 and 31, respectively, and

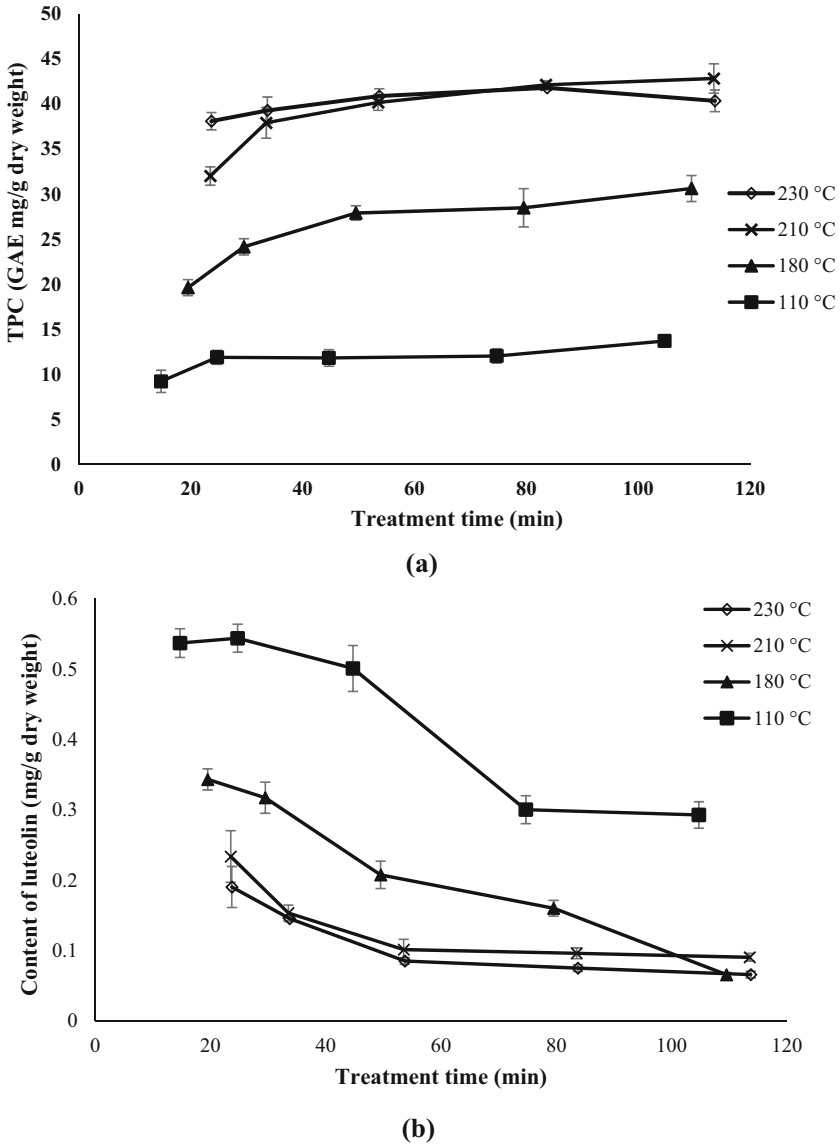


**Fig. 4** **a** TPC and **b** the luteolin content in SCW extracts based on dry mass of carrot leaves at different temperatures with treatment time for a solid-liquid ratio of 15 g/L

the polarity of water is similar to these organic solvent at temperature from 200 to 250 °C (Wohlfarth 2008; Yang et al. 1998). Therefore, theoretically, extraction efficiency of luteolin should increase up to 250 °C. However, Ko et al. (2014) cited that polycyclic aromatic hydrocarbons like flavonoids degrade rapidly at high temperatures of 250 °C during long term heating over 30 min in pressurized hot water. Further, they mentioned that the optimum extraction conditions with SCW extraction is dependent on the molecular structure and the most suitable extraction temperature for flavonoids with an —OH side chain is 170 °C. On the other hand, Murakami et al. (2004) reported the effects of thermal treatment on polyphenol flavonoids rutin (quercetin-3-glucoside), luteolin-7-glucoside, and luteolin at 100 and 180 °C. They stated that luteolin was more stable at lower temperature (110 °C) than rutin and luteolin-7-glucoside. There was only less than 10% reduction of luteolin after heating at 100 °C for 180 min. However, luteolin was not stable at higher temperature (180 °C), where they noted a sharp decrease in luteolin with time. Moreover, in a recent study, Chaaban et al. (2017) investigated the kinetics of pure luteolin in aqueous solution and found that at 110 °C; about 30 and 80% of luteolin was degraded after being heated for 20 and 120 min, respectively. In addition to that the degradation kinetics of luteolin was much faster at 130 °C than at 110 °C. Therefore, investigations of Chaaban et al. (2017) and Murakami et al. (2004) on pure solutions of flavanoids complimented this study and confirmed the decrease in luteolin when being heated at high temperatures for a long period of time.

Thus, from these studies, it is seen that TPC is increased with temperature but the content of luteolin is decreased after a specific treatment time and temperature. It could be further stated that even though SCW extraction could give an effect comparable to organic solvents, the degradation of luteolin becomes the dominant factor at temperatures higher than

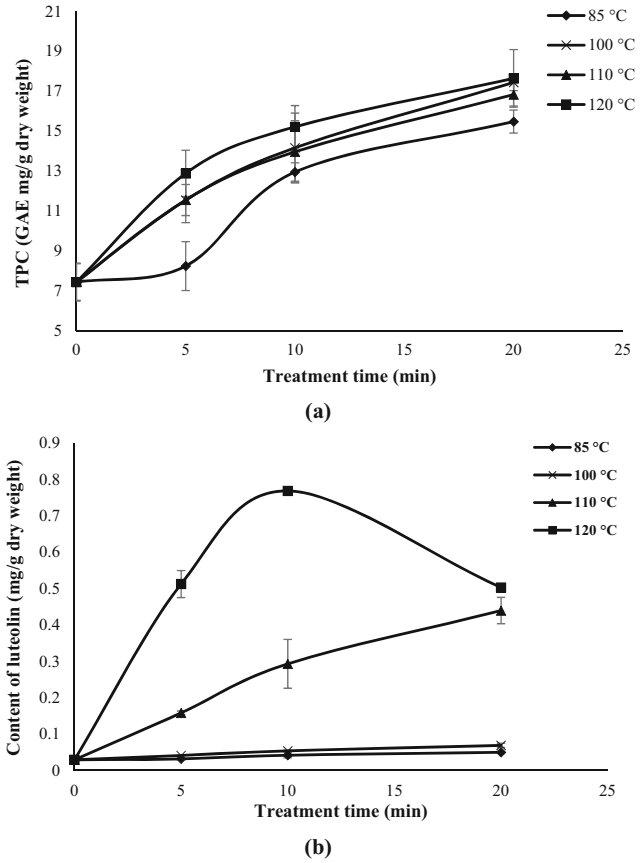


**Fig. 5** **a** TPC and **b** the luteolin content in hot water extraction based on dry mass of carrot leaves at different temperatures with treatment time for a solid-liquid ratio of 15 g/L

180 °C. These reports indicate that possibly, there is a time-temperature combination between 100 and 180 °C and $t < 20$ min that gives maximum yield of luteolin.

## Hot Water Extraction Studies

Hot water extraction studies were carried out to investigate further the important effect of temperature on polyphenols and luteolin content of carrot leaf extract. Figure 5a, b shows the content of polyphenol and luteolin in the extract with treatment time obtained at four different temperatures using hot water. At 85 °C, there was no apparent rise in TPC in the first 5 min. A relatively obvious increase occurred at the next 5 min, where TPC increased from 8.23 ± 1.15 to 12.94 ± 0.47-GAE mg/g dry weight and continued increasing to 15.47 ± 0.58-GAE mg/g dry weight at 20 min. Further, a statistically insignificant ($p > 0.05$) increase in TPC with time was observed for the treatment processes at 100, 110, and 120 °C. This means that there is only a slight effect on TPC at treatment temperatures lower than 120 °C in hot water extraction. If compared with SCW extracts, TPC obtained after 20 min were 19.61 ± 0.35, 32.01 ± 0.37, and 38.10 ± 0.74 GAE mg/g at 180, 210, and 230 °C, respectively, which was much higher than that resulted with hot water extraction. Obviously, hot water extraction is not an effective extraction method to obtain polyphenols within a short treatment time compared to methods like SCW extraction. For instance, it has been shown by Vergara-Salinas et al. (2013) and He et al. (2012) that using SCW extraction, TPC from grape pomace and pomegranate seeds could be increased five and ten times, respectively.

In terms of luteolin, there was practically no change at 85 and 100 °C, where the content was less than 0.1 mg/g after 20 min. However, a significant increase ($p \leq 0.05$) in luteolin extraction was observed above 100 °C when the extraction time was 10 min. In more details, at 110 °C, the luteolin content kept increasing from 0.029 ± 0.003-mg/g dry weight at 0 min to 0.439 ± 0.036-mg/g dry weight at 20 min, which was over 15 times. However, when the temperature increased to 120 °C, the trend displayed an optimum compared to that experienced at other temperatures. It increased rapidly to 0.768 ± 0.009-mg/g dry weight at the first 10 min but decreased to 0.503 ± 0.011-mg/g dry weight at the end of experimental period of 20 min. It affirms that luteolin degrades at temperatures above 120 °C with extended heating time beyond 10 min. These studies further confirm the behavior of luteolin at higher temperature as experienced by Chaaban et al. (2017).

## Comparison of Extraction Yield from Different Feedstock

A comparison of TPC and luteolin content obtained from this study with the outcomes of previous researchers using different agricultural residues is as shown in Tables 1 and 2.

It is being noted that the TPC obtained with SCW extraction at 180 °C for 90 min in an acidic environment (pH = 4) from mango peel (50.3-GAE mg/g dry weight) was higher

**Table 1** TPC from different types of agricultural residues using SCW extraction

| Raw material | Extraction condition | | | TPC (GAE mg/g dry weight) | Reference |
|---|---|---|---|---|---|
| | Treatment time (min) | Temperature (°C) | Treatment pressure (MPa) | | |
| Carrot leaves | 113.5 | 210 | 8 | 42.8 | This study |
| Mango peel | 90 | 180 | – | 50.3 | Tunchaiyaphum et al. (2013) |
| Onion peel | 10 | 165 | 3.45 | 25.0 | KyoungAh et al. (2011) |
| Winery waste | 100 | 140 | 11.6 | 31.7 | Aliakbarian et al. (2012) |

**Table 2** Content of luteolin from different types of agricultural residues and extraction methods

| Raw material | Extraction method | Extraction condition | | | Content of luteolin (mg/g dry weight) | Reference |
|---|---|---|---|---|---|---|
| | | Time | Temperature (°C) | Treatment pressure (MPa) | | |
| Carrot leaves | Hot water | 10 min | 120 | 0.25 | 0.768 | This study |
| Peanut hulls | Solvent extraction (ethanol) | 120 min | 25 | 0.1 | 0.566 | Pang et al. (2014) |
| Olive leaves | Solvent extraction (methanol) | 120 min | 40 | 0.1 | 0.277 | Škerget et al. 2005 |
| Pigeon pea leaves | Enzyme assisted extraction | 18 h | 35 | 0.1 | 0.268 | Fu et al. (2008)] |
| Sweet potato leaves | Refluxed extraction (60% aqueous methanol with 1.2-M HCl) | 120 min | 90 | 0.1 | 0.04 | Chu et al. (2000) |

than that of TPC obtained from carrot leaves (42.83 ± 1.85-GAE mg/g dry weight). Apart from mango peels, the content of polyphenols from carrot leaves with SCW extraction is higher than TPC extracted from other agricultural residues. TPC of carrot leaves with SCW extraction is over 35 and 70% higher than the content of polyphenols extracted from winery waste and onion peel (Aliakbarian et al. 2012; KyoungAh et al. 2011). Therefore, carrot leaves which is presently considered as an agricultural residue could be utilized as a potential feedstock for polyphenol production.

Further, Table 2 gives a comparison of luteolin extraction from different agricultural residues and extraction methods. When compared to other agricultural residues like peanut hulls, olive leaves, pigeon pea leaves, and sweet potato leaves, carrot leaves have given a higher luteolin content in the extracts at comparatively shorter time (Chu et al. 2000; Fu et al. 2008; Pang et al. 2014; Škerget et al. 2005). On the other hand, there is plant matter rich in luteolin like *Reseda luteola* and Dandelion flowers that contain 8.6 and 25 mg/g, respectively (Cerrato et al. 2002; Hu and Kitts 2004). Moreover, Ko et al. (2014) reported that luteolin content from carrot root was 2.506 mg/g with SCW extraction at 190 °C for 15 min, which is three times that obtained from carrot leaves as reported in this study. This study reported degradation of luteolin from carrot leaves at 120 °C, which was also supported by Murakami et al. (2004) that luteolin degrades at 180 °C. The discrepancy in luteolin withstand ability to different temperatures could be due to the difference in cellular matrix in the leaves and the roots of carrots, even though it is the same plant matter. Although there are other plant matters that are rich in luteolin than that from carrot leaves, the leaves still could be considered as a potential feedstock for luteolin production, since it is an agricultural residue and is considered non-edible.

## Conclusion

In this study, we have investigated the use of SCW extraction of polyphenols and luteolin from carrot leaves, which is considered as an agricultural residue and found in abundance in a New Zealand context. It was shown that with the increase of temperature of water at subcritical conditions, there is an increase in the extraction of TPC, while the content of luteolin in the extract shows a significant decrease. Further, hot water extraction studies confirmed the presence of an optimum temperature-time combination that gave maximum content of luteolin confirming previous studies showing thermal degradation of luteolin. It was also shown that TPC and luteolin content in carrot leaves are comparable to other agricultural residues that have been investigated so far.

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