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## Optimization of Extraction of Polyphenols from Syrian Sumac (*Rhus coriaria* L.) and Chinese Sumac (*Rhus typhina* L.) Fruits

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**Abstract:** Sumac is the common name for *Rhus* genus and various species have been used for medicinal and food purposes. In this study, a solid-liquid extraction method was used to extract polyphenols from Syrian sumac (*Rhus coriaria* L.) and Chinese sumac (*Rhus typhina* L.) fruits. Various experimental conditions such as ethanol concentration in water (0-100%, v/v), extraction time (1-9 h), extraction temperature (20-60°C), particle size (0.5-2.5 mm) and solvent to sumac ratio (5:1-25:1 mL g<sup>-1</sup>) were investigated to optimize the extraction. The amount of polyphenols was measured by Folin-Ciocalteu procedure to monitor the efficiency of extraction. The optimal extraction conditions were found to be similar with both sumac species except for the extraction time, with Syrian sumac requiring 1 h and Chinese sumac 5 h; ethanol concentration (20%, v/v); extraction temperature (40°C); particle size (1.0 mm) and solvent to sumac ratio (15:1 mL g<sup>-1</sup>). In addition, extraction time and ethanol concentration were the most significant processing parameters with Syrian and Chinese sumac fruits, respectively. Under optimal conditions, the polyphenols content was 159.32 and 150.68 mg gallic acid equivalent per gram of extract with Syrian and Chinese sumac fruits, respectively. The present findings showed that polyphenols can be extracted from sumac fruits using the solid-liquid extraction method which is efficient, safe and less costly.

**Key words:** Sumac fruits, *Rhus coriaria*, *Rhus typhina*, polyphenols, solid-liquid extraction

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### INTRODUCTION

Sumac is the common name for a genus (*Rhus*) that contains over 250 individual species of flowering plants in the family Anacardiaceae (USDA, 2007). In general, sumac can grow in non-agriculturally viable regions and various species have been used by indigenous cultures for medicinal and other purposes, suggesting potential for commercializing the bioactivity of this plant without competing for food production land uses (Wyk and Wink, 2004). The fruits of sumac (drupes) are of red color and contain a seed.

*Rhus coriaria* grows wild in Turkey, Iran, Palestine, Jordan and Syria, where the ground fruit is commonly used as a condiment and sprinkled over kebabs (grilled meat) and salad as well as over the boiled broad beans. In addition, it is also a principal ingredient of za'atar, the

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popular spice mixture of dried and ground leaves of *Origanum syriacum*, powdered seed coats of *R. coriaria* as acidulant and responsible for the typical red color, roasted sesame seeds, salt and olive oil (Lukas *et al.*, 2009). Treatment of diarrhea is reported as the main medicinal use of this species in Jordan (Lev and Amar, 2002). In different historical records from the area of Bilad Al-Sham (a historical geographical term by former Arab rulers that included significant parts of present-day Syria, Lebanon, Israel, Palestine and Jordan), *R. coriaria* was used against stomach, intestine and eye diseases, animals bites and poisons, hemorrhoids, sexual diseases and pains (Lev, 2002).

*Rhus typhina* originated in North America is used to make a beverage termed Sumac-ade or Rhus juice prepared from its fruits (Peterson, 1977) and serves also as a traditional medicine having pharmacological functions such as antihemorrhoidal, antiseptic, diuretic, stomachic and tonic (Foster and Duke, 1990; Moerman, 1998). It was introduced to China in 1959 by the Botanical Garden, Institute of Botany, Chinese Academy of Sciences (Pan and You, 1994) and was identified as a main forestation species in Beijing municipality (Wang *et al.*, 2008). It is frequently used for rehabilitation of degraded lands in most mountain areas of North China. Besides, the plant also has some ornamental value because of its brilliant red foliage in autumn.

The fruit extracts of *R. coriaria* have been reported to contain high levels of polyphenols such as gallic acid, anthocyanins and hydrolysable tannins (Kosar *et al.*, 2007) and possess strong antimicrobial and antioxidant activities (Nasar-Abbas and Halkman, 2004; Fazeli *et al.*, 2007; Candan and Sokmen, 2004; Bozan *et al.*, 2003). In addition, gallic acid and gallotannin were identified in the leaves of *R. typhina* (Frohlich *et al.*, 2002; Werner *et al.*, 2004). Furthermore, earlier study showed that the fruits of *R. coriaria* and *R. typhina* are rich in oleic and linoleic acids, vitamins as well as minerals (Kossah *et al.*, 2009).

To the best of our knowledge, there are no available reports on the optimization of polyphenols extraction from either Syrian sumac (*R. coriaria*) or Chinese sumac (*R. typhina*) fruits. Therefore, the aim of this study was to optimize and compare the extraction of polyphenols from Syrian and Chinese sumac fruits using different extraction parameters such as ethanol concentration in water, extraction time, extraction temperature, particle size and solvent to sumac ratio.

## MATERIALS AND METHODS

### Plant Materials

Ripened and dried fruits of *R. coriaria* were brought from Syria (Lattakia), while those of *R. typhina* were obtained from China (Lanzhou) in Autumn 2008.

### Reagents and Chemicals

Folin-Ciocalteu's phenol reagent (2N) was purchased from Sigma (Sigma-Aldrich). Gallic acid was purchased from Chinese National Institute for the Control of Pharmaceutical and Biological Products. Ethanol, methanol and other chemicals were obtained from Shanghai Chemical Co., (Shanghai, China) and were of analytical grade.

### Experimental Procedure

Sumac fruits were ground in a household flour mill (Tianjin, China) and were separated into particles of different size using sieves then extracted with aqueous ethanol at different temperatures for different times with occasional stirring. After extraction, the mixture was filtered and the obtained extract was concentrated with a rotary evaporator (SBW-1, Shanghai Shenbo Instrument Co., China) under reduced pressure in a water bath at 45°C.

The extraction parameters such as ethanol concentration in water (0-100%, v/v), extraction time (1-9 h), extraction temperature (20-60°C), particle size (0.5-2.5 mm) and solvent to sumac ratio (5:1-25:1 mL g<sup>-1</sup>) were systematically varied as shown in Fig. 1-5.

#### Determination of Polyphenols Content

The amount of polyphenols in Syrian Sumac Fruit (SSF) and Chinese Sumac Fruit (CSF) extracts was determined according to the Folin-Ciocalteu procedure (Gamez-Meza *et al.*, 1999) with some modifications. A solution was prepared from each extract at a concentration of 1 mg mL<sup>-1</sup> in 50% (v/v) aqueous methanol. Five hundreds microliters of extract solution were placed into test tubes and then 100 µL Folin-Ciocalteu's phenol reagent solution (50%, v/v, in water) and 7.5 mL sodium carbonate solution (2%, w/v, in water) were added. The contents of tubes were mixed thoroughly and allowed to stand in the dark for 1.5 h at room temperature and the absorbance was measured at 750 nm. Gallic acid was used as a standard for the calibration curve and the results were expressed as Gallic Acid Equivalents (GAE) in mg g<sup>-1</sup> extract.

#### Statistical Analysis

All experiments were run in duplicates and the resulting data were subjected to the Analysis of Variance (ANOVA) using the SAS System for Windows, Version 8.0 (SAS, 1999). Duncan's multiple-range test was used to compare means at a significance level of 5%.

### RESULTS AND DISCUSSION

#### Effect of Ethanol Concentration in Water

The effect of ethanol concentration in water on the extraction of polyphenols from sumac fruits is given in Fig. 1. In fact, results obtained with water alone were lower than those with ethanol. This fact is in correlation with the polarity of extraction solvent and the solubility of phenolic compounds in it (Canadanović-Brunet *et al.*, 2006; Turkmen *et al.*,

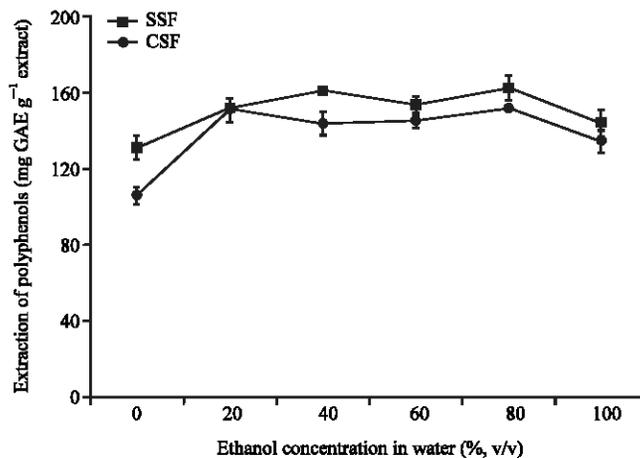


Fig. 1: Effect of ethanol concentration in water on the extraction of polyphenols from sumac fruits. SSF: Syrian Sumac Fruit; CSF: Chinese Sumac Fruit. Extraction conditions: extraction time, 3 h; extraction temperature, 40°C; particle size, 1 mm; solvent to sumac ratio, 15:1 mL g<sup>-1</sup>

2006). Results from this study are in accordance with those of Yu *et al.* (2005) and Zahida *et al.* (2006), where 80% aqueous ethanol was more efficient than water for extracting phenolics from peanut skin and astragaloside from *Thesium chinense* Turcz, respectively. Increasing ethanol concentration from 20 to 100% had no significant ( $p > 0.05$ ) effect on the extraction of polyphenols which reached the maximum at a concentration of 80% (v/v). Moreover, no significant difference was found between the amounts of polyphenols from either SSF or CSF using 20 and 80% ethanol. Thus, 20% (v/v) was regarded as the optimal ethanol concentration, which was used throughout other experiments.

#### Effect of Extraction Time

The effect of time on the extraction of polyphenols from sumac fruits is shown in Fig. 2. Results showed that the extraction of polyphenols increased with the increase of time up to 5 h. The maximum extraction of polyphenols from SSF and/or CSF was obtained in 5 h. Longer extraction time decreased the content of polyphenols, possibly because of some loss of phenolic compounds via oxidation and these products might polymerize into insoluble compounds (Shi *et al.*, 2003). No significant difference ( $p > 0.05$ ) was observed between the amounts of polyphenols extracted from SSF for 1 and 5 h, whereas those extracted from CSF were found to be significantly different ( $p < 0.05$ ). Therefore, 1 and 5 h were considered as optimal times for the extraction of polyphenols from SSF and CSF, respectively.

#### Effect of Extraction Temperature

Figure 3 shows the impact of temperature on the extraction of polyphenols from sumac fruits. A marked increase of polyphenols was observed over the extraction temperature ranging from 20 to 40°C. The amounts of polyphenols reached the maximum at 40°C, but a further temperature increase to 60°C resulted in a rapid decrease of the extraction, especially with CSF. This may be due to the degradation of some phenolic compounds during extraction at high temperature coupled with a relatively longer extraction time (5 h) as mentioned earlier. An extraction temperature of 40°C was chosen as optimal for the extraction of polyphenols from both SSF and CSF.

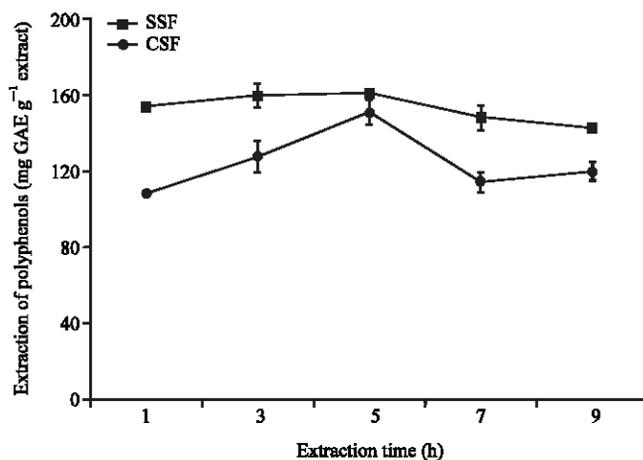


Fig. 2: Effect of time on the extraction of polyphenols from sumac fruits. SSF: Syrian Sumac Fruit; CSF: Chinese Sumac Fruit. Extraction conditions: Ethanol concentration, 20% (v/v); extraction temperature, 40°C; particle size, 1 mm; solvent to sumac ratio, 15:1 mL g<sup>-1</sup>

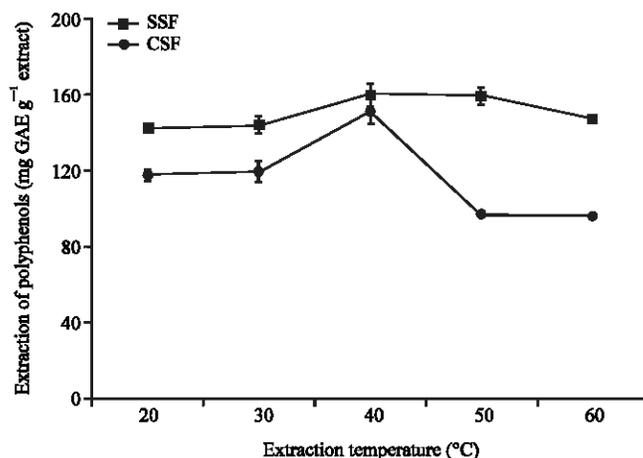


Fig. 3: Effect of temperature on the extraction of polyphenols from sumac fruits. SSF: Syrian Sumac Fruit; CSF: Chinese Sumac Fruit. Extraction conditions: Ethanol concentration, 20% (v/v); extraction time, 1 h (SSF) and 5 h (CSF); particle size, 1 mm; solvent to sumac ratio, 15:1 mL g<sup>-1</sup>

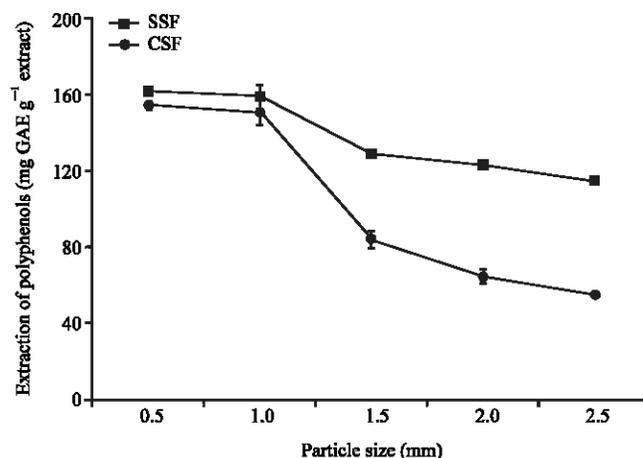


Fig. 4: Effect of particle size on the extraction of polyphenols from sumac fruits. SSF: Syrian Sumac Fruit; CSF: Chinese Sumac Fruit. Extraction conditions: ethanol concentration, 20% (v/v); extraction time, 1 h (SSF) and 5 h (CSF); extraction temperature, 40°C; solvent to sumac ratio, 15:1 mL g<sup>-1</sup>

#### Effect of Particle Size

The effect of particle size on the extraction of polyphenols from sumac fruits is given in Fig. 4. When the particle size increased from 0.5 to 2.5 mm, the extraction of polyphenols decreased and a similar observation was made by Pinelo *et al.* (2007). However, the contents of polyphenols obtained using 0.5 and 1.0 mm as sumac particle size were not significantly different ( $p > 0.05$ ). Since small particles (0.5 mm) resulted in processing difficulties such as dust and heat generation during grinding and blocked filters during extraction, 1.0 mm was selected as the optimal particle size to be used for the extraction of polyphenols from both

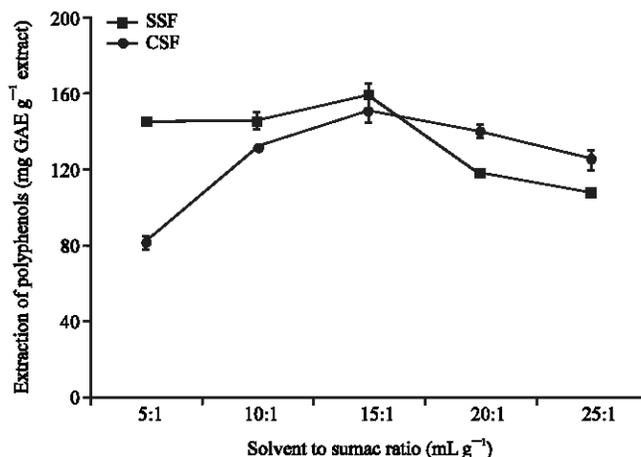


Fig. 5: Effect of solvent to sumac ratio on the extraction of polyphenols from sumac fruits. SSF: Syrian Sumac Fruit; CSF: Chinese Sumac Fruit. Extraction conditions: Ethanol concentration, 20% (v/v); extraction time, 1 h (SSF) and 5 h (CSF); extraction temperature, 40°C; particle size, 1 mm

SSF and CSF. Particle size of 1.0 mm has also been reported to give full recovery of phenolics and essential oil from sage leaves when ethanol-water mixtures were used as extraction solvent (Durling *et al.*, 2007).

#### Effect of Solvent to Sumac Ratio

The effect of solvent to sumac ratio on the extraction of polyphenols from sumac fruits is shown in Fig. 5. Increasing solvent to sumac ratio up to 15:1 mL g<sup>-1</sup> caused an increase in the extraction of polyphenols. However, higher solvent to sumac ratios led to lower extraction of polyphenols. This may be due to the fact that more solvent provides more dissolved oxygen, which causes oxidation. Similarly, solvent to sumac ratios as low as 5:1 and 10:1 mL g<sup>-1</sup> created difficulties in the filtration process, due to some solvent being absorbed by the dry sumac material. Hence, 15:1 mL g<sup>-1</sup> was judged to be the optimal solvent to sumac ratio for the extraction of polyphenols from either SSF or CSF.

### CONCLUSIONS

Optimization of polyphenols extraction from SSF and CSF was carried out using ethanol-water as solvent. Optimal extraction conditions were found to be similar for both sumac materials: ethanol concentration in water (20%, v/v), extraction temperature (40°C), particle size (1.0 mm) and solvent to sumac ratio (15:1 mL g<sup>-1</sup>). However, the optimal extraction time with CSF was 5 h, while being only 1 h with SSF. Of all the extraction parameters studied in this work, extraction time and ethanol concentration in water had the most significant effects on the extraction of polyphenols from SSF and CSF, respectively.

To present knowledge, this is the first report demonstrating that Syrian sumac (*Rhus coriaria*) and Chinese sumac (*Rhus typhina*) are good sources of polyphenols, which may be used in the food and pharmaceutical industries due to their antioxidant and antimicrobial properties. Further investigations are needed to ascertain the antioxidant and antimicrobial activities of extracts obtained from these sumac species.

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