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# Optimisation of the aqueous extraction conditions of phenols from meadowsweet (*Filipendula ulmaria* L.) for incorporation into beverages

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## ABSTRACT

Meadowsweet was extracted in water at a range of temperatures (60–100 °C), and the total phenols, tannins, quercetin, salicylic acid content and colour were analysed. The extraction of total phenols followed pseudo first-order kinetics, the rate constant (*k*) increased from 0.09 ± 0.02 min<sup>-1</sup> to 0.44 ± 0.09 min<sup>-1</sup>, as the temperature increased from 60 to 100 °C. An increase in temperature from 60 to 100 °C increased the concentration of total phenols extracted from 39 ± 2 to 63 ± 3 mg g<sup>-1</sup> gallic acid equivalents, although it did not significantly affect the proportion of tannin and non-tannin fractions. The extraction of quercetin and salicyclic acid from meadowsweet also followed pseudo first-order kinetics, the rate constant of both compounds increasing with an increase in temperature up until 90 °C. Therefore, the aqueous extraction of meadowsweet at temperatures at or above 90 °C for 15 min yields extracts high in phenols, which may be added to beverages.

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# 1. Introduction

Meadowsweet (Filipendula ulmaria L.) is a perennial herb indigenous to Europe. It has been used traditionally as an anti-inflammatory for joint and rheumatic pain. Recent research supports the use of meadowsweet extracts as anti-inflammatory agents (Halkes et al., 1997; Jaciulevičiūtė, Keturkienė, & Leonavičienė, 2001; Tunón, Olavsdotter, & Bohlin, 1995). These anti-inflammatory properties are partly due to the phenolic content, which is made up of tannins (mainly rugosin-D), salicylates (salicylaldehyde, methyl salicylate) and flavonoids (Blumental, Goldberg, & Brinckmann, 2000). Previous studies have reported that methanolic extracts of meadowsweet have a total phenolic content between 7.2 (Proestos, Boziaris, Kapsokefalou, & Komaitis, 2008) and 26.8 mg/g d.w. (Kahkonen et al., 1999). Flavonoids have been extracted from meadowsweet leaves using hot aqueous ethanol (70%) (Krasnov, Raldugin, Shilova, & Avdeeva, 2006) and also using methanol in a Soxhlet apparatus (Papp et al., 2004). They have also been extracted from meadowsweet flowers using hydro-alcoholic solutions and these extracts were found to contain up to 6% total flavonoids (Lamaison, Carnat, & Petitjean-Freytet, 1991). For both leaves and flowers the main flavonoids present were derivatives of quercetin, including spiraeoside, rutin, quercitrin and quercetin-3-O-β-glucuronide (Lamaison et al., 1991; Papp et al., 2004).

Although meadowsweet has been studied from a pharmacological and analytical point of view, overall there have been few publications relating to this medicinal herb. Traditionally, meadowsweet has been taken as an aqueous decoction or liquid extract (Mills & Bone, 2000). Therefore, it would be important to study the effect of time and temperature on the aqueous extraction of phenolic compounds (including quercetin and salicylic acid) from meadowsweet. The effect of extraction time and temperature on the phenolic content of green tea (Labbé, Tremblay, & Bazinet, 2006; Price & Spitzer, 1994), black tea (Price & Spiro, 1985) and rooibos tea (Jaganyi & Wheeler, 2003) has previously been studied. However, as yet there has been no work published on optimising the extraction conditions for meadowsweet, to maximise the phenolic content of its aqueous extracts.

To produce extracts for incorporation into a beverage it is important not only to maximise the total phenolic content but also to minimise the content of tannins, as they have astringent properties, which may not be desirable in a beverage. Another factor which may influence the quality of a beverage is colour, as appearance is one of the most important characteristics of a food (Hutchings, 1999). Therefore, it would be important to study both the effect of time and temperature on the proportion of tannins and on the colour of meadowsweet extracts. Also, other authors have established that pH of the solvent has an effect on the extraction of both green tea (Kim, Park, Lee, & Han, 1999) and black tea (Spiro & Price, 1987); therefore a change in pH of the extracting solvent may also have an effect on meadowsweet extracts.





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The objectives of this study were to investigate the effect of extraction conditions on the total phenols, the tannin and non-tannin fractions, the bioactives (salicylic acid, quercetin) and the colour of meadowsweet extracts, in order to produce a high quality aqueous extract rich in phenols. The study also seeks to establish the effect that extraction pH has on the phenolic constituents of meadowsweet extracts. Overall, the aim of this study was to establish the optimum extraction conditions, in order to produce foodgrade meadowsweet extracts rich in phenols and with minimum detrimental tannin fraction and colour, so as to prepare ingredients that can be directly incorporated into flavoured waters and/or fruit drinks.

## 2. Materials and methods

## 2.1. Materials

The dried aerial parts of meadowsweet were purchased from The Organic Herb Trading Company, Somerset, UK. Laboratory grade gallic acid, cinchonine hemisulphate, ethanol, formaldehyde and HPLC-grade acetonitrile, phosphoric acid and methanol were purchased from Sigma–Aldrich (St. Louis, MO). The HPLC standards quercetin and salicylic acid were also obtained from Sigma–Aldrich (St. Louis, MO). The Folin–Ciocalteu reagent, ethanol and sodium carbonate were purchased from Merck (Darmstadt, Germany). Laboratory grade hydrochloric acid was obtained from VWR International Ltd. (Poole, UK).

# 2.2. Extraction

Meadowsweet (2.5 g) was steeped in 100 ml of distilled water at 60, 70, 80, 90 and 100 °C, for a number of time points at each temperature (n > 10). After heating, the extract was filtered using Whatman No. 1 filter paper under vacuum and cooled immediately on ice.

Extractions at pH 3.9 and 6.4 were carried out at 80  $^{\circ}$ C, using citric acid and citrate to prepare buffers with an ionic strength of 0.2 M.

## 2.3. Total phenols

The total phenolic content of the extracts was determined by the Folin–Ciocalteu method, according to Singleton and Rossi (1965). Briefly, the extract (0.2 ml) was mixed with 0.5 ml of Folin–Ciocalteu reagent, 1.5 ml of 20% sodium carbonate solution and 7.8 ml of distilled water. The samples were left to stand for 2 h at room temperature and the absorbance at 760 nm was read on a UV–visible spectrophotometer (UV-1240, Shimadzu, Milton Keynes, UK). The results were expressed as mg gallic acid equivalents (mg GAE)/g dry weight of plant material.

## 2.4. Tannin separation

Tannin separation and estimation was determined using the method of Peri and Pompei (1971). For separation of the tannin from the non-tannin fraction 1 ml of meadowsweet extract was mixed with 1 ml of 0.5% cinchonine hemisulphate solution. The mixture was vortexed and then centrifuged in a MSE Micro Centaur bench centrifuge (MSE Scientific Instruments, Crawley, UK) at 5668g for 5 min. The supernatant was used to calculate the non-tannin fraction and the pellet was redissolved in a solution containing 1 ml of ethanol and 1 ml of HCl (10%) to calculate the total tannin fraction. After separation all fractions were quantified using the Folin–Ciocalteu procedure and calculated as gallic acid equivalents.

#### 2.5. Colour

The Hunter  $L^*$  (lightness),  $a^*$  (red–green) and  $b^*$  (yellow–blue) values of the extracts were measured using a Minolta colorimeter (Model No. CR-300, Minolta Ltd., Milton Keynes, UK). The colorimeter was calibrated for internal light (D65) using a white calibration plate before carrying out colour measurements. Colour measurements were carried out in triplicate. Hue angle ( $h^\circ$ ) expresses the colour difference and is defined as follows: red–purple = 0°, yellow = 90°, bluish-green = 180°, and blue = 270°. Hue angle was calculated using the following equation (McGuire, 1992):

$$h^{\circ} = \arctan \frac{b^{*}}{a^{*}}$$
(1)

#### 2.6. HPLC analysis

Meadowsweet extracts were hydrolysed using a modification of the method by Hertog, Hollman, and Venema (1992). Briefly, 4.5 ml of meadowsweet extract, 4.5 ml of methanol and 1 ml of HCl (35%) were mixed and heated at 90 °C under reflux for 2 h. After heating the samples were cooled in an ice-bath and then filtered through Whatman No. 1 filter paper. The filtrate was then filtered through a 0.2-µm membrane filter and 10 µl was injected directly onto the HPLC column. HPLC separation was carried out using an Agilent 1200 HPLC system (Agilent Technologies, Santa Clara, CA), in combination with an Agilent Zorbax Eclipse XDB- $C_{18}$  (150 mm  $\times$  4.6 mm i.d.; 5  $\mu$ m particle size) column with a  $C_{18}$ guard column (Phenomenex, Macclesfield, UK). The solvents used were 0.025 M phosphoric acid (A) and acetonitrile (B). The separations were performed at 30 °C by gradient elution at a flow rate of 1 ml/min. UV detection was set at 210 nm. The following gradient was used: 0-15 min, from 20% to 40% B; 15-20 min, 20% B. Identification was based on retention times by comparison with a commercial standard.

#### 2.7. Data analysis

Extraction reaction rate constants (k) and equilibrium concentrations ( $C_{\infty}$ ) were calculated using non-linear regression in Sigma-Plot version 10( $^{\odot}$ 2004, Systat Software Inc., San Jose, CA) according to a pseudo first-order equation (Eq. (2)).

$$C = C_{\infty}(1 - \exp(-kt)) \tag{2}$$

The  $r^2$  value was above 0.91 in all cases, indicating a good data fit to the model. Differences among treatments were determined using a student's *t*-test (two-tailed); results with  $p \leq 0.05$  were considered significantly different.

## 3. Results and discussion

#### 3.1. Extraction of total phenols

The extraction of total phenols from meadowsweet followed pseudo first-order kinetics (Fig. 1). At all temperatures the concentration of total phenols rises rapidly initially before reaching an equilibrium concentration ( $C_{\infty}$ ). This is in agreement with previous reports, which studied the aqueous extraction of phenols from various plants (Harbourne, Jacquier, & O'Riordan, 2009; Jaganyi & Wheeler, 2003; Price & Spiro, 1985; Price & Spitzer, 1994). The rate constant (k) increased from 0.09 ± 0.02 to 0.44 ± 0.09 min<sup>-1</sup>, as the temperature increased from 60 to 100 °C (Table 1). The maximum total phenols extracted ( $C_{\infty}$ ) also increased as the temperature increased from 60 to 100 °C. It is evident from Fig. 1 that between 80



**Fig. 1.** Aqueous extraction of total phenols from meadowsweet as a function of time at temperatures of 60 ( $\bullet$ ), 70 ( $\bigcirc$ ), 80 ( $\lor$ ), 90 ( $\triangle$ ) and 100 ( $\blacksquare$ ) °C (note: broken lines represent the behaviour predicted by the pseudo first-order kinetic model).

and 90 °C there was a significant ( $p \leq 0.05$ ) increase in the maximum total phenols from  $48 \pm 1$  to  $61 \pm 2$  mg/g d.w. There was no further increase in the maximum total phenols extracted between 90 and 100 °C (Table 1). A major increase of total phenols at high temperatures (90-100 °C) compared to low temperatures (20-40 °C) has been previously reported in both extracts of Phyllanthus amarus (Lim & Murtijava, 2007) and rooibos tea (Joubert, 1990). This increase has been attributed to (i) the high temperatures causing the cellular constituents in the plant to break down, resulting in the release of cell wall or bound phenolics (Lim & Murtijava, 2007), (ii) an increase in the solubility of some of the phenols (Joubert, 1990; Lim & Murtijava, 2007) or (iii) the hydrolysis of tannins into simple phenols (Lim & Murtijava, 2007). As mentioned previously, for meadowsweet there was no further increase in the total phenol content in the extracts between 90 and 100 °C, so an increase in the solubility of the phenolic compounds does not seem to be the main reason for the increase. Therefore, the effect of temperature on the extraction of tannins and non-tannins was studied to investigate if high temperatures of extraction cause the tannins to break down into simple phenols in meadowsweet extracts.

### 3.2. Extraction of tannins

The effect of temperature on the proportion of tannins and nontannins in meadowsweet extracts is presented in Fig. 2. The extraction duration at each temperature was chosen when maximum total phenols were extracted. There was no significant difference in the proportion of tannins and non-tannins extracted from meadowsweet in the temperature range 60–100 °C ( $p \ge 0.05$ ). Therefore, the increase in total phenols in meadowsweet does not seem to be due to the breakdown of tannins to simple phenols.



**Fig. 2.** Effect of extraction temperature (60–100 °C) on the tannin ( $\square$ ) and non-tannin ( $\square$ ) concentration in aqueous meadowsweet extracts (note: the value is the average of three replicates and the error bars represent the standard error).

Hence, it is likely that temperatures  $\ge 90$  °C caused the breakdown of the plant cells, thus causing more phenols to be extracted.

To get a better understanding of the extraction of the phenolic compounds, the extraction kinetics of tannins and non-tannins were also studied at both 80 and 100 °C and the results are presented in Table 2. The extraction of both tannin and non-tannin phenols were modelled using the pseudo first-order kinetic model. At a temperature of 80 °C the reaction rate constant (k) for nontannins was 0.36  $\pm$  0.07 min<sup>-1</sup> and for tannins was 0.2  $\pm$  0.02 min<sup>-1</sup> (Table 2). When the temperature was increased to 100 °C the rate increased to  $0.65 \pm 0.06$  and  $0.46 \pm 0.04$  min<sup>-1</sup> for non-tannins and tannins, respectively. As expected the k and the  $C_{\infty}$  values for both fractions increased as the temperature increased from 80 to 100 °C. Also, at both temperatures the rate constant for non-tannins was significantly higher than for tannins ( $p \le 0.05$ ); the slower release of the tannin fraction was possibly because they may have been bound to other cellular constituents. The rate constant for tannins was not significantly different from that of total phenols  $(p \ge 0.05)$ . This is in agreement with previous work on the extraction of tannins from Cassia occidentalis seeds at 100 °C using a firstorder kinetic model, which established that the extraction rate constant for total phenols (0.08 min<sup>-1</sup>) and for tannins  $(0.09 \text{ min}^{-1})$  were comparable (Medoua & Mbofung, 2007).

Overall, temperature did not have a significant effect on the proportion of tannins and non-tannins extracted but an increase in temperature resulted in faster extraction of both tannin and non-tannin fractions from meadowsweet. The non-tannin fraction was extracted faster than both the tannins and total phenols from meadowsweet at both temperatures studied. Therefore, the extraction of tannins is likely to be the rate-limiting step in the extraction of total phenols from meadowsweet.

#### Table 1

Extraction rate constants (k) and equilibrium concentrations ( $C_{\infty}$ ) of compounds extracted in aqueous meadowsweet extracts.

Temperature (°C)	Total phenols		Quercetin		Salicylic acid		Hue-angle		Lightness	
	$k (\min^{-1})$	$C_{\infty} (mg/g)$	$k (\min^{-1})$	$C_{\infty} (\mathrm{mg/g})$	$k (\min^{-1})$	$C_{\infty}$ (mg/g)	$k (\min^{-1})$	$h_{\infty}$ (°)	$k (\min^{-1})$	$L_{\infty}$
60	$0.09 \pm 0.02^{\circ}$	39 ± 2 <sup>c</sup>	$0.19 \pm 0.04^{\circ}$	$2.2 \pm 0.1^{d}$	$0.15 \pm 0.04^{\circ}$	$0.39 \pm 0.0^{b}$	$0.7 \pm 0.2^{b}$	51 ± 3 <sup>a</sup>	$0.41 \pm 0.08^{\circ}$	$49 \pm 2^{a}$
70	$0.15 \pm 0.04^{cb}$	45 ± 3 <sup>cb</sup>	$0.25 \pm 0.04^{\circ}$	$2.7 \pm 0.1^{\circ}$	$0.22 \pm 0.04^{\circ}$	$0.44 \pm 0.01^{a}$	$0.97 \pm 0.1^{b}$	$44 \pm 1^{b}$	$0.56 \pm 0.23^{\circ}$	$50 \pm 2^{a}$
80	$0.2 \pm 0.01^{b}$	48 ± 1 <sup>b</sup>	$0.34 \pm 0.0^{b}$	$3.07 \pm 0.06^{b}$	$0.35 \pm 0.0^{b}$	$0.43 \pm 0.01^{ab}$	$1.7 \pm 0.4^{a}$	$36 \pm 4^{b}$	0.603 ± 0.1 <sup>c</sup>	$48 \pm 1^{a}$
90	$0.36 \pm 0.06^{a}$	$61 \pm 2^{a}$	$0.91 \pm 0.04^{a}$	$3.22 \pm 0.03^{a}$	$0.68 \pm 0.18^{a}$	$0.44 \pm 0.02^{a}$	$2.2 \pm 0.3^{a}$	25 ± 2 <sup>c</sup>	$1.5 \pm 0.2^{b}$	$48 \pm 1^{a}$
100	$0.44 \pm 0.09^{a}$	$63 \pm 3^{a}$	$0.97 \pm 0.1^{a}$	$3.3 \pm 0.1^{a}$	$0.69 \pm 0.08^{a}$	$0.46 \pm 0.01^{a}$	$2.4 \pm 0.2^{a}$	$18 \pm 1^d$	$2.3 \pm 0.2^{a}$	47.1 ± 0.3

Mean values ± standard deviation represented by the same letters within the same column are not significantly different at  $p \ge 0.05$ .

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Extraction rate constants (k) and equilibrium concentrations ( $C_{\infty}$ ) of meadowsweet tannin and non-tannin phenolics extracted at temperatures of 80 and 100 °C. Temperature (°C)  $\frac{80 °C}{k ~(min^{-1})} C_{\infty} ~(mg/g ~d.w.)} \frac{100 °C}{k ~(min^{-1})} C_{\infty} ~(mg/g ~d.w.)}$ 

Mean values ± standard deviation represented by the same letters within the same column are not significantly different at  $p \ge 0.05$ .

#### 3.3. Extraction of bioactives from meadowsweet

Table 2

The extraction of guercetin and salicylic acid from meadowsweet also followed pseudo first-order kinetics. The rate constant (k) for the extraction of salicylic acid from meadowsweet increased from  $0.15 \pm 0.02$  to  $0.68 \pm 0.18 \text{ min}^{-1}$  at 60 °C and 90 °C, respectively; between 90 and 100 °C the rate constant was not significantly different ( $p \ge 0.05$ ) (Table 1). The *k* values for the extraction kinetics of non-tannins at 80  $(0.36 \pm 0.07 \text{ min}^{-1})$  and 100 °C (0.65  $\pm$  0.06 min<sup>-1</sup>) were comparable to those reported for salicylic acid (Table 1). The *k* value for the extraction of quercetin from meadowsweet increased from  $0.19 \pm 0.04$ to  $0.91 \pm 0.04 \text{ min}^{-1}$  as the temperature increased from 60 to 90 °C; as for salicylic acid the rate constant did not increase significantly between 90 and 100 °C ( $p \ge 0.05$ ). Again, the *k* values for non-tannins and guercetin at 80 °C were similar however the rate constant at 100 °C for quercetin was significantly ( $p \le 0.05$ ) higher than for non-tannins (Table 1). Perhaps an increase in extraction temperature above 90 °C not only causes a disruption in the cells, resulting in faster extraction, but also may have increased the solubility of quercetin.

The maximum salicylic acid ( $C_{\infty}$ ) extracted increased with an increase in temperature from 60 to 70 °C, but above 70 °C there was no further increase seen with increasing temperature up to 100 °C. Salicylic acid is quite soluble in water; therefore, it is reasonable that the maximum salicylic acid was extracted from mead-owsweet at 70 °C. Unlike salicylic acid, the maximum quercetin extracted ( $C_{\infty}$ ) significantly increased from 2.2 ± 0.1 to 3.22 ± 0.03 mg/g d.w. with increasing temperature from 60 to 90 °C ( $p \leq 0.05$ ). There was no further increase in quercetin with an increase in temperature to 100 °C (Table 1). Similarly, Joubert (1990) reported an increase in the extraction of flavonoids from rooibos tea with an increase in temperature from 23 to 90 °C, although temperatures up to 100 °C were not investigated in this study.

**Fig. 3.** Effect of extraction time on the lightness of meadowsweet extracts at temperatures of 80 ( $\blacksquare$ ), 90 ( $\triangle$ ) and 100 ( $\square$ ) °C.

Overall, the rate constant (k) for the total phenols increased gradually as the temperature increased from 60 to 100 °C whereas the k values for both guercetin and salicylic acid increased dramatically between 80 and 90 °C (Table 1), although increase in k value was higher for quercetin than for salicylic acid. Therefore, an increase in extraction temperature from 80 to 90 °C has an effect on the rate of extraction of both compounds but high temperature has a bigger influence on the extraction rate of quercetin than salicylic acid. Also, there was a dramatic increase in maximum total phenols extracted as the temperature increased from 80 to 90 °C, whereas quercetin content increased progressively, with an increase in temperature up to 90 °C after which temperature it levelled off. Unlike the other compounds the maximum salicylic acid was extracted at 70 °C. Extraction temperature (60–100 °C) did not affect the proportion of tannins and non-tannins in the extracts.

Therefore, temperatures at or above 90 °C for 15 min may be used to extract the maximum concentration of total phenols  $(57 \pm 2 \text{ mg/g} \text{ d.w.})$  and plant bioactives  $(3.30 \pm 0.02 \text{ mg/g} \text{ d.w.})$ quercetin and  $0.40 \pm 0.01 \text{ mg/g} \text{ d.w.}$  salicylic acid), without having any adverse effects on the tannin concentration of meadowsweet extracts. These actual content values of total phenols, salicylic acid, and quercetin at the optimum extraction conditions (90 °C, 15 min) compare very favourably with data from the literature (Kahkonen et al., 1999; Lamaison et al., 1991), considering the published data are usually based on alcoholic extraction systems and are therefore not suitable for food-grade preparation of extracts destined to be incorporated into beverages.

#### 3.4. Colour

The evolution of colour (lightness and hue angle) with time at all temperatures was also fitted to the pseudo first-order kinetic model and the results are presented in Table 1. It is clear that light-







p

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Table 3

Extraction rate constant	(k) and equilibrium conc	centrations ( $C_{\infty}$ ) of meado	owsweet phenolics extracte	d at 80 °C at pH 3.9 and 6.	4.		
рН	Total phenols		Quercetin		Salicylic acid		
	$k ({ m min}^{-1})$	$C_{\infty}$ (mg/g)	$k ({ m min}^{-1})$	$C_{\infty}$ (mg/g)	$k ({\rm min}^{-1})$	$C_{\infty} (\mathrm{mg/g})$	
3.9	$0.21 \pm 0.04^{a}$	43 ± 2 <sup>c</sup>	$0.26 \pm 0.04^{ab}$	$2.2 \pm 0.1^{b}$	$0.24 \pm 0.04^{b}$	$0.43 \pm 0.02$	
Control (pH 5.6)	$0.2 \pm 0.01^{a}$	$48 \pm 1^{b}$	$0.34 \pm 0.03^{a}$	$3.07 \pm 0.06^{a}$	$0.35 \pm 0.01^{b}$	$0.43 \pm 0.01$	
6.4	$0.16 \pm 0.03^{a}$	$57 \pm 2^{a}$	$0.17 \pm 0.07^{b}$	$2.7 \pm 0.4^{ab}$	$0.5 \pm 0.1^{a}$	$0.42 \pm 0.02$	

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Mean values  $\pm$  standard deviation represented by the same letters within the same column are not significantly different at  $p \ge 0.05$ 

ness decreases initially with time before reaching an equilibrium value  $(L_{\infty})$  (Fig. 3); the evolution of hue angle with time also followed the same trend. This is in agreement with a previous study carried out which studied the evolution of both lightness and hue angle with time at temperatures from 57 to 100 °C in chamomile extracts (Harbourne et al., 2009). The rate of development of colour (both lightness and hue) increased with an increase in temperature (Table 1). Both developed faster than the rate of phenolic extraction. The *k* values for hue angle were higher than those for lightness, indicating the extract became redder faster than it darkened. The equilibrium lightness  $(L_{\infty})$  did not significantly  $(p \ge 0.05)$  change with increasing temperature from 60 to 100 °C (Table 1); this is in agreement with previous work on the colour of black tea (Liang & Xu, 2003), which found no change in the lightness of the extracts between 60 and 90 °C. The equilibrium hue angle  $(H_{\infty})$  decreased with increasing temperature, indicating that the extract became slightly redder with increasing temperature.

### 3.5. Extraction pH

The extraction of phenolic compounds, with aqueous solutions of citrate and citric acid, at various pH values (3.9 and 6.4) were fitted to the pseudo first-order kinetic model (Fig. 4). The rate constant was not significantly different for meadowsweet extracted at pH 3.9, 6.4 or in water (pH 5.6) ( $p \ge 0.05$ ). The maximum total phenols extracted from meadowsweet increased from  $43 \pm 2$  to  $57 \pm 2 \text{ mg/g}$  d.w. with an increase to pH 6.4 (Table 3). Kim et al. (1999) studied the effect of extraction pH on green tea but unlike in meadowsweet they found the total catechin content of green tea infusions did not significantly change when the green tea was extracted at pH 4, 5 or 6, but when the extraction pH was increased to 7 the total catechin content decreased significantly. Also, Spiro and Price (1987) studied the effect of pH on theaflavins in black tea; they found that increasing the pH to 6.8 had no significant effect on the theaflavin content in black tea but decreasing the pH caused an increase in theaflavin content.

Unfortunately the tannin separation method (Peri & Pompei, 1971) did not allow for the measurement of the tannin and nontannin fractions in the buffered solutions; however, the active ingredients (salicylic acid and guercetin) were measured to determine the effect of extraction pH on these compounds. The rate constant for salicylic acid was lower at pH 3.9 in comparison to the control; however, the rate constant for the sample extracted at pH 6.4 was higher than that of the unbuffered solution. Extraction at both pH 3.9 or 6.4 caused a decrease in the rate constant (k) for quercetin (Table 3) in comparison with the control sample  $(0.34 \pm 0.03 \text{ mg/g d.w.})$ . There was no significant difference in the maximum salicylic acid extracted at pH 3.9, pH 6.4 or in the control sample. However, extraction at both pH 3.9 and 6.4 caused a slight decrease in the maximum quercetin extracted, in comparison with the meadowsweet extracted in water (Table 3), possibly because quercetin may be slightly less soluble in saline solutions.

Although a higher extraction pH seems to increase the total phenol content of meadowsweet extracts, it has no significant effect on the extraction of salicylic acid or quercetin. Therefore, lowering the pH of the extraction solvent does not seem to have any major advantage over extracting meadowsweet in water.

# 4. Conclusion

The aqueous extraction of meadowsweet at temperatures above 90 °C yields extracts rich in phenols, guercetin and salicylic acid, with good colour and a relatively low proportion of tannins in the extracts. Altering the pH of the extraction solvent between 3.9 and 6.4 did not have any major advantage over aqueous extraction. Therefore, the aqueous extraction of meadowsweet at temperatures at or above 90 °C for 15 min yields extracts, which may have potential for inclusion in beverages.

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#### References

- Blumental, M., Goldberg, A., & Brinckmann, J. (Eds.). (2000). Herbal medicine: Expanded commission E monographs (pp. 253-256). Massachusetts: Integrative Medicine Communications.
- Halkes, S. B. A., Beukelman, C. J., Kroes, B. H., van den Berg, A. J. J., Labadie, R. P., & van Dijk, H. (1997). In vitro immunomodulatory activity of Filipendula ulmaria. Phytotherapy Research, 11, 518-520.
- Harbourne, N., Jacquier, J. C., & O'Riordan, D. (2009). Optimisation of the extraction and processing conditions of chamomile (Matricaria chamomilla L.). Food Chemistry, 115, 15-19.
- Hertog, M. G. L., Hollman, P. C. H., & Venema, D. P. (1992). Optimization of a quantitative HPLC determination of potentially anticarcinogenic flavonoids in vegetables and fruits. Journal of Agricultural and Food Chemistry, 40(9), 1591 - 1598
- Hutchings, J. B. (1999). Food color and appearance. Maryland: Aspen Publisher (pp. 12-21).
- Jaciulevičiūtė, L., Keturkienė, A., & Leonavičienė, L. (2001). Relation between zinc and magnesium content in liver of rats with adjuvant arthritis and treatment with the tincture of Filipendula ulmaria (L.) Maxim. Acta medica Lituanica, 8(4), 253-256.
- Jaganyi, D., & Wheeler, P. J. (2003). Rooibos tea: Equilibrium and extraction kinetics of aspalathin. Food Chemistry, 83, 121-126.
- Joubert, E. (1990). Effect of batch extraction conditions on extraction of polyphenols from rooibos tea (Aspalathus linearis). International Journal of Food Science and Technology, 25, 339-343.
- Kahkonen, M. P., Hopia, A. I., Vuorela, H. J., Rauha, J. P., Pihlaja, K., Kujala, T. S., et al. (1999). Antioxidant activity of plant extracts containing phenolic compounds. Journal of Agricultural and Food Chemistry, 47(10), 3954-3962.
- Kim, S. H., Park, J. D., Lee, L. S., & Han, D. S. (1999). Effect of pH on green tea extraction. Korean Journal of Food Science and Technology, 31(4), 1024-1028.
- Krasnov, E. A., Raldugin, V. A., Shilova, I. V., & Avdeeva, E. Y. (2006). Phenolic compounds from Filipendula ulmaria. Chemistry of Natural Compounds, 42(2), 148-151.
- Labbé, D., Tremblay, A., & Bazinet, L. (2006). Effect of brewing temperature and duration on green catechin solubilization: Basis for production of EGC and EGCG-enriched fractions. Separation and Purification Technology, 49, 1-9.
- Lamaison, J. L., Carnat, A., & Petitjean-Freytet, C. (1991). Main flavonoid contents of commercial samples of Filipendula ulmaria (L.,) Maxim. Plantes Médicinales et Phytothérapie, 25, 1-5.
- Ling, Y., & Xu, Y. (2003). Effect of extraction temperature on cream and extractability of back tea [*Camellia sinensis* (L.) O. Kuntze]. International Journal of Food Science and Technology, 38, 37-45.
- Lim, Y. Y., & Murtijaya, J. (2007). Antioxidant properties of Phyllanthus amarus extracts as affected by different drying methods. LWT, 40, 1664-1669.

McGuire, R. G. (1992). Reporting of objective colour measurements. *Hortscience*, 27, 1254–1255.

- Medoua, G. N., & Mbofung, C. M. F. (2007). Kinetic studies of some physico-chemical substances during roasting and preparation of beverages made by *Cassia* occidentalis seeds. *LWT – Food Science and Technology*, 40(4), 730–736.
- Mills, S., & Bone, K. (2000). Principles and practice of phytotherapy: Modern herbal medicine. London: Churchill livingstone.
- Papp, I., Apati, P., Andrasek, V., Blazovics, A., Balazs, A., Kursinszki, L., et al. (2004). LC–MS analysis of antioxidant plant phenoloids. *Chromatographia*, 60, S93–S100.
- Peri, C., & Pompei, C. (1971). Estimation of different phenolic groups in vegetable extracts. *Phytochemistry*, 10, 2187–2189.
- Price, W. E., & Spiro, M. (1985). Kinetics and equilibria of tea infusion: Rates of extraction of theaflavin, caffeine and theobromine from several whole teas and sieved fractions. *Journal of the Science of Food and Agriculture*, 36, 1309–1314.
- Price, W. E., & Spitzer, J. C. (1994). The kinetics of extraction of individual flavonols and caffeine from a Japanese green tea (Sen Cha Uji Tsuyu) as a function of temperature. *Food Chemistry*, 50, 19–23.

- Proestos, C., Boziaris, I. S., Kapsokefalou, M., & Komaitis, M. (2008). Natural antioxidant constituents from selected aromatic plants and their antimicrobial activity against selected pathogenic microorganisms. *Food Technology and Biotechnology*, 46(2), 151–156.
- Singleton, V. L., & Rossi, J. A. (1965). Colorimetry of total phenolics with phosphomolybdic-phosphotungstic acid reagents. *American Journal of Enology* and Viticulture, 16, 44–158.
- Spiro, M., & Price, W. E. (1987). Kinetics and equilibria of tea infusion-part 6: The effects of salts and of pH on the concentrations and partition constants of theaflavins and caffeine in kapchorua pekoe fannings. *Food Chemistry*, 24, 51–61.
- Tunón, H., Olavsdotter, C., & Bohlin, L. (1995). Evaluation of anti-inflammatory activity of some Swedish medicinal plants. Inhibition of prostaglandin biosynthesis and PAF-induced exostosis. *Journal of Ethnopharmacology*, 48, 61–67.