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Supercritical fluid extraction technology for essential oil extraction of *Murraya exotica*

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Abstract

Supercritical fluid extraction (SCFE) is an effective method for extracting the bioactive compounds from planting materials as it is more selective and causes no thermal degradation of components. SCFE gives oil of superior quality because extracts are obtained without using any chemical solvents. Three treatment conditions of temperature and pressure i.e. T_1= 35°C/75 bar, T_2=40°C/80 bar and T_3=45°C/90 bar were employed in this study. Whereas, treatment T_2 was found to be superior in getting better yield and quality of essential oil while producing 0.18% concrete oil and 0.008% absolute oil then rest of treatments. Whereas, extraction done at 45°C and 90 bar pressure (T_3) gave the second highest concrete (0.17%) and absolute (0.006%) oil yield and T_1 (35°C/75 bar) treatment resulted in the lowest concrete (0.15%) and (0.005%) oil yield. Moreover, temperature had more influence on the essential oil extraction of *M. exotica* through supercritical fluid extraction technique.

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Introduction

Murraya exotica commonly known as orange jasmine, belongs to family Rutaceae. These plants are adapted to wide range of conditions and can be grown from nearly sea level to an elevation of 1500 m. This plant grows well under bright sun and is moderately intolerant of shade, though it produces few flowers growing well under partial shade. Natively it was found in China, India, Sri Lanka, northeastern Australia and Taiwan (Parrotta, 2001). *Murraya exotica* possess fragrant white flowers in the form of small clusters that are produced at the end of branches. These flowers are highly aromatic and contain an adequate amount of essential oil (Naseem *et al*., 2015).

Essential oils are the volatile products extracted from odoriferous plants. Such oils are known to be essential, as they were thought to represent the characteristic essence of odor and flavor. These oils are extremely volatiles and can be isolated by using different extraction techniques (Younis *et al*., 2007; Younis *et al*., 2011). Where, we can extract minute quantity of essential oil from different flowers and other aromatic parts of plants (Lis-Balchin and Deans, 1997; Younis *et al*., 2009; Akram *et al*., 2014).

Supercritical fluid extraction method (SCFE) was firstly developed in 1960 (Lin *et al*., 1999). Extraction of the volatile oils by means of the supercritical fluid extraction method (SCFE) has been the subject of substantial interest (Younis *et al*., 2011). In this method essential oil extraction from natural plant materials is mainly done by using the carbon dioxide (CO_2) as a solvent (Lang and Wai, 2001; Diaz-Maroto *et al.,* 2002; Hu *et al*., 2007). The liquid like solvating power and gas-like transport qualities of supercritical fluids make them chiefly suitable for extracting chemical compounds from plant tissues with a high grade of recovery in a short period of time. (Wheeler and McNally, 1989; Caredda *et al*., 2002).

As compared to conventional methods Viz. Steam distillation, Hydro-distillation, and Solvent extraction (Younis *et al*., 2008), SCFE is more convenient and superior because it is more selective and extracts obtained have no added solvent that gives oil of superior quality (Carlson *et al*., 2001; Wood *et al*., 2006).The main objective of this study was to identify the suitable extraction conditions of SCFE for essential oil extraction of *Murraya exotica*

Materials and methods

The study was done at Rosa Lab, Institute of Horticultural Sciences, University of Agriculture Faisalabad. Fresh flowers were collectedarly in the morning. Prior to placement in SCFE apparatus (DEVEN, Supercritical Pvt. Ltd, India), fresh weights were taken and petals were spread in a tray under shade at room temperature in order to remove the excessive moisture. Twenty kilograms of M. exotica petals were used for extraction in a single batch of SCFE. Three treatment conditions with three replications were used to get reproducible results. Treatments plan are T_1= 35°C/75 Bar,T_2= 40°C/80 Bar and T_3= 45°C/90 Bar. Yield attributes are "Yield of concrete oil (%)" and "Yield of absolute oil (%)". Physical attributes are color of absolute oil, refractive index, congealing point, optical rotation, and specific gravity. Chemical attributes are acid number, ester number.

Supercritical fluid extraction (SCFE)

The *M. exotica* petals were filled in the extraction vessel after weighing of 20kg weight. The pressure was maintained through a gas booster, censored by pressure-transductor and a pneumatic control-valve. The samples were placed in a separation-vessel at specific temperature and pressure range maintained through a connected computer having SCFE software. The oil was separated by changing the CO_2 phase and absolute oil was collected in a flask.

The percentage of concrete oil was calculated by using equation;

$$\text{Percentage of concrete oil} = \frac{\text{Weight of concrete oil}}{\text{Weight of petals}} x100$$

The absolute oil percentage was calculated by using formula;

$$\text{Percentage of absolute oil} = \frac{\text{Weight of absolute oil}}{\text{Weight of petals}} x100$$

Physio-chemical analysis of Murraya exotica essential oil

This part of study was carried out at Department of Chemistry, University of Agriculture, Faisalabad and Department of Chemistry, Forman Christian College Lahore. Color of absolute oil was determined by spectrophotometric method (Paquot, 1986a). The refractive index of the essential oil was measured at 25°C by using Abbe's refractometer (Paquot, 1986b).Congealing point of the oil was determined by placingminute amount of oil in the capillary tube that was further suspended into a larger tube having a thermometer. The maximum temperature at which solidification occurred was noted as the congealing point of the oil. To get value for optical rotation, polarimeter-tube (10 ml) was used. The essential oil's specific gravity was determined by using gravity-bottle.

The acid number was determined through indicator method (Paquot, 1986b). Then acid number was calculated by the formula,

$$\text{Acid number} = \frac{56.1\,(\text{Number of c.c of 0.1 KOH})}{\text{Weight of the sample (g)}}$$

The ester content was calculated by:

Ester number $= \frac{28.05\,(a)}{s}$, Where, a = number of c.c. of 0.5 normal HCI used in the saponification; s = Weight of sample in g.

Statistical analysis

Experiment was laid out according to Completely Randomized Design (CRD).The data were subjected to statistical analysis using analysis of variance technique at 5% probability (Steel *et al.*, 1997) by using Statistica Software.

Results and discussion

The main purpose of the present work was to investigate the combined effects of experimental parameters such as pressure and temperature, on the extraction yield and quality of the essential oil of *M. exotica*. According to our best knowledge no work has been done before on the supercritical fluid extraction of *M. exotica* essential oil.

Extraction of essential oil

Solvent strength of supercritical fluid can be manipulated by changing pressure (P) and/or temperature (T), to achieve a remarkable high selectivity of analysts at suitable conditions of temperature and pressure (Liza *et al.*, 2009; Reverchon *et al.*, 1993). The special note of this process for selective extraction of soluble compounds from a raw material is the usage of gases above their critical points (Reverchon, 1997; Diaz-Maroto *et al.*, 2002).

A number of research articles related to supercritical fluid extraction from natural planting material appeared in the literature (Anitescu *et al.*, 1997; Reis-Vasco *et al.*, 1999; Omidbaigi *et al.*, 2003;; Sarrazin *et al.*, 2000; Leal *et al.*, 2008; Ueno *et al.*, 2008; Liu *et al.*, 2009).

In this study three different conditions of temperature and pressure i.e. 35ºC temperature and 75 bar pressure (T_1), 40ºC temperature and 80 bar pressure (T_2) and 45ºC and 90 bar pressure (T_3) were used to estimate the best for the essential oil extraction.

Analysis of variance (Table 1) showed the significant variation among different treatment conditions. It was observed that the treatment T_2 (40ºC temperature and 80 bar pressure) gave the highest oil yield for both concrete (0.18%) and absolute oil (0.008%).

Table 1. Mean Square values from analysis of variance for Concrete and absolute oil % of *Murraya exotica* essential oil extracted through different conditions (temperature and pressure) of SCFE.

Source	DF	Concrete oil %	Absolute oil %
Replication	2	0.00744	0.00002
Treatment	2	0.0641*	0.000973*
Error	4	0.000111	0.0000583
Total	8		

The extraction done at 45ºC and 90 bar pressure (T_3) gave the second highest concrete (0.17%) and absolute (0.006%) oil yield, whereas T_1 treatment resulted in the lowest concrete (0.15%) and (0.005%) of absolute oil yield (Table 2). It is obvious from the results that by increasing the treatment conditions of temperature and pressure from T_1 (35ºC/ 75 bar) to T_2 (40ºC/ 80 bar) oil yield also increased, further increase in temperature and pressure to T_3 (45ºC/90 bar) causes decrease in yield of oil.

Table 2. Concrete and absolute oil percentage of *Murraya exotica* essential oil extracted through SCFE under different temperature and pressure conditions.

Treatments	Concrete oil %	Absolute oil %
T1	0.15 c	0.005 c
T2	0.18 a	0.008 a
T3	0.17 b	0.006 b

The literature reported by Liza *et al.* (2009); Rezaei and Temelli (2000); Wang *et al.* (2008) clearly indicates the positive effect of pressure in increasing the density of supercritical CO_2 that ultimately enhances the extraction efficiency. The decrease in oil yield in T_3 might be the result of increase in temperature that dominates over the solute vapor pressure and decreases the solvating power of CO_2, caused by reduction in its density. The decrease in oil yield by increasing the extraction temperature was also supported by the work of Nilufer *et al.*, 2009; Gopalakrishnan *et al.* (1990); Lou *et al.* (2003).

Physio-chemicalproperties of essential oil

Qualitative analysis was done by analyzing physio-chemical properties of the essential oil. Different extraction techniques and conditions produced oils with varied physiochemical properties(Joy *et al.*, 2001).Color is an important physical parameter used for the qualitative analysis of the essential oils.

Highly significant effect of various extraction treatments was noted on the color of absolute oil of *M. exotica*. The extraction done at 35°C and 75 bar pressure (T_1) resulted in light yellow color of the oil, whereas the pale yellow color was obtained in other treatments i.e. T_2 (40°C and 80 bar pressure) and T_3 (45°C and 90 bar pressure) (Table 3).

Table 3. Physio-chemical properties of *Murraya exotica* essential oil extracted through SCFE under different temperature and pressure conditions.

Treatments	T1	T2	T3
Color	Light yellow	Pale yellow	Pale yellow
Refractive index	1.46 at 25°C	1.48 at 25°C	1.47at 25°C
Congealing Point	20°C	22°C	21°C
Optical Rotation	+4.1 at 25°C	+4.4 at 25°C	+4.3at 25°C
Specific Gravity	0.866	0.871	0.868
Acid Number	1.48	1.55	1.51
Ester Number	135	143	139

In qualitative analysis oil color is considered an important physical parameter. Change in color of the essential oil can be the outcome of burning of chemical constituents of the oil due to high temperature (Assis *et al.*, 2000; Wenqiang *et al.*, 2007).

Refraction of light while passing from the essential oil is calculated as the refractive index of the oil. It was observed that refractive index of the oil was affected by the extraction treatments. The oil extracted at 40°C and 80 bar pressure (T_2) gave the highest refractive index i.e. 1.48. The refractive index of the oil extracted at 45°C and 90 bar pressure (T_3) was found to be 1.47, whereas the lowest value (1.46) was obtained from T1 treatment (Table 3).

Different extraction treatments showed variation in congealing point. It was found highest (22°C) in the oil extracted through T_2 (40°C and 80 bar pressure) and was followed by T_3 (45°C and 90 bar pressure) which gave a temperature of 21°C, whereas the lowest congealing point (20°C) was obtained by extracting oil using T1 (35°C and 75 bar pressure) treatment (Table 3).

The degree at which light is bend in a direction of left or right while passing through the oil is measured as optical rotation. Optical rotation of *Murraya exotica* oil was highly influenced by different SCFE treatments. The oil obtained by using 40°C and 80 bar pressure (T_2) exhibited highest value of +4.4 for this property whereas, the extraction performed at 45°C and 90 bar pressure (T_3) gave the second highes value (+4.3) than T_1 treatment (35°C and 75 bar) which gave the lowest value (+4.1) for optical rotation (Table 3).Specific gravity of essential oil of *M. exotica* was also affected by different SCFE treatments. The highest value (0.871) was obtained from the oil extracted using T_2 treatment followed by 0.868 in the oil obtained through T_3 treatment, whereas the lowest value (0.866) was recorded using T_1 treatment (Table 3).

Acid value which is an indicator of free fatty acid content due to enzymatic activity in the samples (Mohammed and Hamza. 2008) was also analyzed and it was found that different extraction treatments varied significantly for acid number in the oil.

The highest acids (1.55) were found in the oil extracted at 40ºC and 80 bar pressure (T_2) followed by T_3 (45ºC and 90 bar pressure) that gives 1.51 whereas the oil obtained at 35ºC and 75 bar pressure (T_1) exhibited the lowest number (1.48) of acids (Table 3). The oil extracted by using treatment T_2 maintained the highest esters (143), followed by T_3 that yielded second highest number of esters in oil (139) while the treatment T_1 resulted in the lowest number of esters (135) in the oil (Table 3).*M. exotica* essential oil has very pleasant, sweet odor like citrus, whereas the strongest odor was found in the supercritical fluid extracted oil having high number of esters (143).

Physical (refractive index, congealing point, optical rotation and specific gravity) and chemical (acid number and ester number) characteristics are the measure of the purity and quality of the essential oils. In this experiment analysis of the oils proves the superiority of the T_2 over other treatments as variation in the physiochemical characters of the can be the result of composition of essential oils greatly effects by extraction treatments (Vijayalakshmi *et al.*, 2010).

Conclusion

Based on this discussion it was cleared that the supercritical fluid extraction is an effective method for extracting the bioactive compounds from planting materials whereas treatment T_2 was superior in getting better yield and quality of essential oil then rest of treatments. Moreover, temperature had more influence on the essential oil extraction of *M. exotica* through supercritical fluid extraction technique.

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