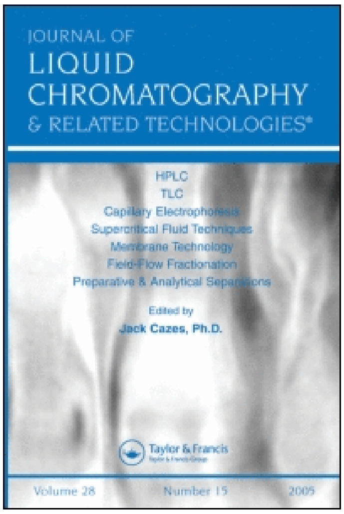

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Comparative Evaluation of SFE and Steam Distillation Methods on the Yield and Composition of Essential Oil Extracted from Spearmint (*Mentha spicata*)

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Abstract: A supercritical fluid extraction (SFE) process was optimized to obtain high quality mint oil. The effect of oven and freeze drying techniques on the quality of mint oil was investigated. The flavor principles in the SFE extract were compared with those obtained by a conventional steam distillation method. Then highest extraction yield was obtained at SFE conditions (50°C and 350 bar), however, high quality oil was obtained at 30°C and 150 bar. The compositional quality of SFE extract was found to be superior as compared to that of a steam distillation process. The extraction yield and compositional quality of oil from mint samples obtained from different regions of UAE and other countries were reported.

Keywords: Essential oils, Extraction, Spearmint, Steam distillation, Supercritical fluid extraction

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INTRODUCTION

Spices have been widely used for centuries to flavor foods, as well as to preserve them. Large quantities of spices are also used in medicine, pharmaceutical, perfumery, cosmetics, and several other industries. Spearmint (*Mentha spicata*), popularly known as mint, is considered as one of the most important spices throughout the world. The essential oils obtained from different varieties of mint are widely used as a flavoring in food, cosmetic, and pharmaceutical industries. Locally grown mint is extensively used in the UAE, but no data is available on its composition.

Presently, mint is extensively grown in the UAE, which makes it important to study its quality.

Conventional solvent extraction and steam distillation have been used for the extraction of mint oil. Solvent extraction requires several hours for complete extraction and the extracted products may contain toxic solvent residues. In addition, solvent extraction is likely to involve losses of more volatile compounds during removal of the solvent. Steam distillation is highly nonselective and may result in the removal of undesirable components from the plant. In addition, steam distillation requires high energy consumption.

In recent years, supercritical fluid extraction (SFE) has become an alternative to the conventional extraction techniques. This is mainly because the dissolving power of supercritical fluids (SCFs) can be adjusted by simply changing the pressure and temperature. Supercritical carbon dioxide (SC CO_2) has been the most commonly used solvent in the food and pharmaceutical industries, since it is nontoxic, nonflammable, chemically stable, inexpensive, environmentally acceptable, and easily separated from the extract. In addition, supercritical conditions of CO_2 are readily attained (critical temperature: 31.05°C and critical pressure: 72.8 atm). With this technology it is possible to extract heat sensitive compounds and avoid any toxic solvent residues in the product.[1,2] A number of workers have used the SFE process to obtain essential oils from different species such as mint, oregano, basil, thyme, parsley, chamomile, sage, and other medicinal plants.[3–7] In a review article, Lang and Wai[8] discussed the recent developments in the SFE of herbs. Graspar et. al.[9] proposed models for the extraction of essential oils from herbs using compressed carbon dioxide. A comparison of performances of SC CO_2 extraction with steam distillation, hydrodistillation, and solvent extraction of different medicinal herbs and plant leaves indicates that the overall yield and composition of extracts obtained by SC CO_2 extraction are similar, or better than, that of the conventional methods.[4,10] However, extraction with SC CO_2 has several advantages over the traditional methods of hydrodistillation, steam distillation, and solvent extraction, because CO_2 does not alter the delicate balance of components in natural products. In addition, the lower temperature in the SFE process is an advantage for the separation of thermally vulnerable materials.

Essential oils from different varieties and sources of mint (eg., Spanish, Romanian, Italian, Turkish) have been extracted and analyzed by different researchers using different techniques.[11–14] However, no data is available on the essential oils of the native mint grown in UAE. The main objective of this project is to obtain the yield and composition of essential oils extracted from a UAE mint using the SC CO_2 extraction method.

Supercritical CO_2 extracts from different varieties of mint (native and imported) have been prepared at different conditions (temperature, pressure, CO_2 flow rate, CO_2 volume) and analyzed using chromatographic and spectroscopic studies. This study will demonstrate the potential use of SC CO_2 for the extraction of essential oils from mint. This new process will be faster, safer, and more efficient in selectively extracting volatile compounds from mint with less energy consumption and environmental impact than conventional solvent extraction and steam distillation processes.

EXPERIMENTAL

Collection and Drying of Samples

Fresh mint grown in Al Ain and Abu Dhabi areas was procured from the local market. The mint was cleaned under running water and spread on a sieve to drain excess water and divided into three equal portions. Two portions were subjected, separately, to freeze drying and oven drying. The third portion was ground to a fine paste for extraction as a fresh sample. The cleaned samples were subjected to freeze drying (in a Labconco freeze dryer at $-40^{\circ}C$), as well as air oven drying (at $35^{\circ}C$) over night. The water loss during drying was calculated. The dried samples were powdered prior to extraction. Dried mint samples (India, France, and Syria) were also procured from different department stores. The residual water content of all samples was analyzed by an immiscible solvent distillation method using toluene. The water content of the samples was used for calculating yield, as well as composition on a dry basis.

Extraction of Mint Oil

Supercritical Fluid Extraction

The environmentally friendly supercritical fluid technology (CO_2, 99.995% pure), studied earlier by the author,[15–19] was used for the extraction of mint. The experimental apparatus consisted of a 260 mL capacity syringe pump and controller system (ISCO 260D), and an ISCO series 2000 SCF extraction system (SFX 220) consisting of a dual chamber extraction module with two 10 mL stainless steel vessels. Temperature and pressure within the vessels were measured and could be independently adjusted.

The 10 mL stainless steel cell (diameter 1.5 cm) was filled with a known quantity (about 2 g) of dried mint. The cell was pressurized and heated to the desired pressure and temperature and kept for about 15 min to reach equilibrium. A known volume of SC CO_2 was passed through the cell at a desired flow rate ranging from 1 to 3 mL/min. The extract was collected in about 4 mL ethanol after depressurization of the gas. The lines were flushed with 5 mL ethanol to collect remnants of extract in the lines. The collected sample was diluted to 10 mL and subjected to gas chromatographic (GC) analysis. The percent extraction yield was calculated from the weight loss of the extraction vessel after the extraction process and results are expressed on the dried sample basis.

Steam Distillation

A known quantity (about 2 g of dried mint, about 10 g of wet mint) was weighed into the distillation vessel and connected to the steam generator (VELP Scientific, UDK 142). The condenser of the steam distillation vessel was continuously cooled using a cooler circulator attached to the steam generator. The distillation vessel was subjected with steam at 80% efficiency, and about 900 mL of distillate was collected into a 1 L separatory funnel containing about 50 mL of ethyl acetate or cyclohexane for 10, 15, and 20 min. The steam distillate obtained was extracted with 3×30 mL portions of the same solvent, the combined solvent layer was dried over anhydrous sodium sulphate and evaporated in a rotary evaporator (at 35°C) under vacuum. The concentrated extract was quantitatively transferred into a 10 mL volumetric flask, made up to volume, and subjected to GC analysis.

Characterization of Mint Oil

GC/MS Characterization of Flavor Principles in Mint Extract

The extracts obtained from SFE, as well as the steam distillation process, were subjected to GC/MS analysis on a Varian Saturn 2000 ion-trap GC/MS. The conditions were as follows: GC Column: CP-Sil 8 CB (30 m $\times$ 0.32 mm i.d., 1 μm film thickness). Injector Temp: 220°C. Column Temp.: 80°C-10 min. −10°C/min. −220°C-6 min. −20°C/min. −300°C-20 min. Ion Trap Temp.: 170°C.

The flavor principles namely α-pinene, limonene, cineole, linalool, menthol, menthone, methylacetate, carvone, dihydrocarvone, and azulene were identified and quantified. Other compounds, such as pregnanedione, 5-isopropenyl-2-mehtylcyclopeny-1-enecarboxaldehyde, 3,6-dimethyl-phen-1, 4-diol, 1-(4-methoxymethyl-2,6-dimethylphenyl)ethanol, and sitosterol were also identified using NIST Mass Spectral library.

GC Quantitation of Flavor Principles

The flavor principles were quantified on a Varian 3800 GC with flame ionization detector (FID). The GC conditions were as follows: GC Column: CP-Sil 8 CB (30 m × 0.32 mm i.d., 1 μm film thickness). Injector Temp: 220°C. Column Temp.: 80°C −10 min. −10°C/min. −220°C-6 min. −20°C/min. −300°C-20 min. FID Temp: 280°C.

Analytical standards of the flavor principles were obtained from Sigma/Aldrich. A calibration curve was constructed by injecting standard solutions in duplicate and plotting average area under the peak against the concentration of standard. The concentration of each flavor principle in the extract was deduced from the calibration curve using the average areas from duplicate injections.

RESULTS AND DISCUSSION

A combination of temperature, pressure, volume, and flow rate of CO_2 was studied for obtaining optimum yield and composition of mint oil, using oven dried mint of Al Ain (Table 1). Under the conditions, supercritical CO_2 density ranged from 0.70 to 0.97 g/mL and the total mass ratio of CO_2/sample in different runs were found to be in the range of 34.9–89.6. Maximum yield of 10.9% was obtained at a temperature, pressure, and CO_2 volume combination of 50°C, 350 bar, and 150 mL. Increasing CO_2 volume to 200 mL did not increase the yield. The extraction yield slightly increased with the increase in CO_2 flow rate. The higher yield obtained under high pressure and temperature conditions may be due to the extraction of other non-volatile principles, such as lipids and pigments. A combination of temperature, pressure, volume, and flow rate of CO_2 (50°C, 350 bar, and 100 mL at 1 mL/min) was selected for further studies to save liquid CO_2. The average yield (average of three runs) obtained under the selected conditions are given in Table 1 and Figure 1. Fresh mint obtained in Al Ain and Abu Dhabi emirates gave comparable yield. Imported dry mint (from 3 different countries) gave the yields ranging from 5.70–7.77% w/w. The low extract yield of dry imported mint samples (France and India) may be attributed to differences in the variety, geographical conditions, and loss of volatile components during long storage periods.

Figure 2 shows the influence of pressure on the yield of local mint extracted by the SFE process. At a constant temperature, higher pressures led to greater extraction yields. For example, raising the pressure from 150 to 350 bar increased the yield by about 48.5% at 30°C, 18.6% at 40°C, and 17.6% at 50°C. This was expected since an increase in pressure leads to an increase in CO_2 density, resulting in a higher solubility of solutes in SC CO_2.

Table 1. Experimental conditions of SFE process for the extraction of essential oils from mint

Mint variety	Water content (%w/w)	T (°C)	P (bar)	CO_2 volume (mL)	CO_2 flow rate (mL/min)	CO_2 density (g/mL)	m_{CO2}/m_{sample} (g/g)	Yield (%w/w) on dry basis
Al-Ain(Oven-dried)	9.00	30	150	100	1	0.847	42.08	4.13 ± 0.12^a
		30	250	100	1	0.922	45.74	5.28
		30	350	100	1	0.970	47.87	6.13 ± 0.19^a
		40	150	100	1	0.780	38.64	5.96 ± 0.21^a
		40	250	100	1	0.879	43.80	6.98
		40	350	100	1	0.935	46.18	7.07
		50	150	100	1	0.700	34.86	6.74
		50	250	100	1	0.834	41.12	7.47
		50	350	100	1	0.899	44.51	7.93 ± 0.29^a
		50	350	150	1	0.899	67.13	10.90
		50	350	200	1	0.899	89.57	10.91
		50	350	100	2	0.899	44.77	8.37
		50	350	100	3	0.899	44.63	8.61
Al Ain (Freeze dried)	7.90	50	350	100	1	0.899	44.90	5.51
Abu Dhabi (Freeze dried)	7.95	50	350	100	1	0.899	44.84	5.47
Abu Dhabi (Oven-dried)	8.92	50	350	100	1	0.899	44.86	7.60
Dry mint (France)	6.80	50	350	100	1	0.899	44.59	6.26
Dry mint (India)	6.85	50	350	100	1	0.899	44.33	5.70
Dry mint (Syria)	6.75	50	350	100	1	0.899	44.65	7.77

[a]Average ± Std. Dev.

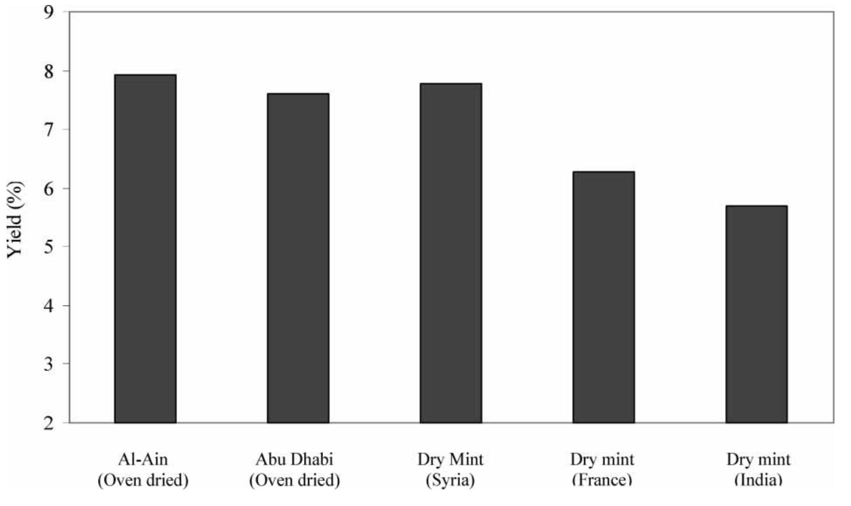

Figure 1. SFE yield of dry mint obtained from different regions of UAE and other Countries, P = 350 bar, T = 50°C, CO_2 volume = 100 mL, CO_2 flow rate = 1 mL/min.

The influence of temperature on the yield of local mint extracted by the SFE process is presented in Figure 3. At all pressures, when the temperature was increased, the yield increased. For example, increasing the temperature from 30 to 40°C increased the yield from 4.13 to 5.96 (44.4%) at 150 bar, while the yield increases by 13.1% when the temperature was increased from 40 to 50°C at the same pressure. Similar trends were observed at the other pressures.

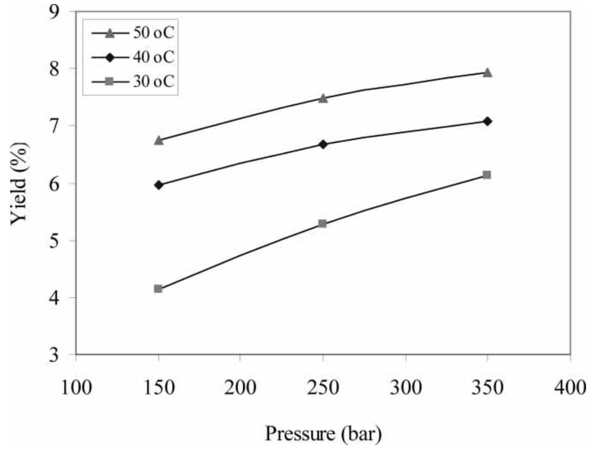

Figure 2. Effect of pressure on the yield of local mint extract, CO_2 volume = 100 mL, CO_2 flow rate = 1 mL/min.

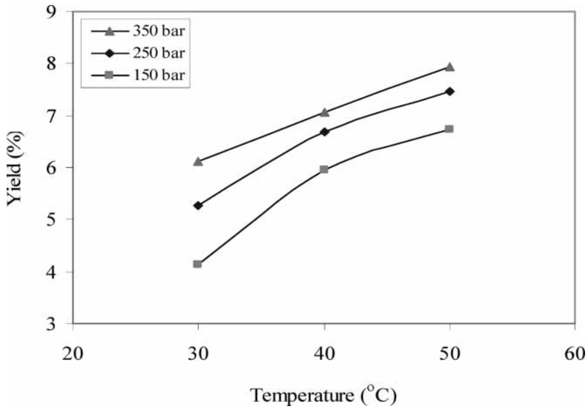

Figure 3. Effect of temperature on the yield of local mint extract, CO_2 volume = 100 mL, CO_2 flow rate = 1 mL/min.

Figure 4 shows the relation between the extraction yield and the density of CO_2. As can be seen on the figure, at a constant temperature, the yield increases linearly with density ($r^2 > 99.5\%$). The increase in yield with an increase in CO_2 density is a result of the increase in the solvent power of CO_2 at higher densities.

Table 2 provides the compositional data (flavor principles) of mint oil obtained with the SFE process. Average values were reported for selected runs (shown in Table 1). The best compositional quality of the oil was achieved under the SFE conditions of temperature (30°C), pressure (150 bar), and CO_2 volume (100 mL, 1 mL/min).

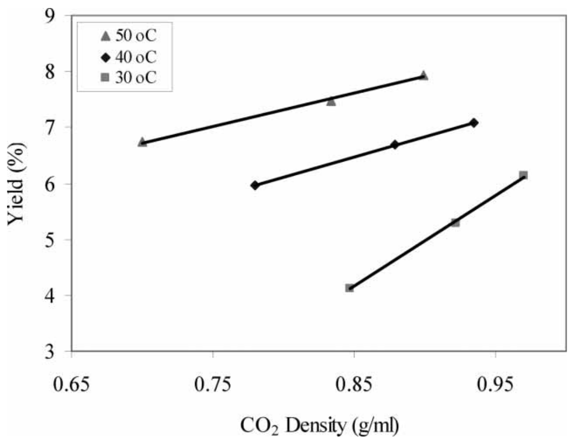

Figure 4. Effect of CO_2 density on the yield of local mint extract, CO_2 volume = 100 mL, CO_2 flow rate = 1 mL/min.

Table 2. Characterization of flavor principles in mint samples (SFE process)

Sample	T (°C)	P (bar)	CO_2 mL, Flow rate (mL/min)	Composition (μg/g, on dry basis)									
				α-pinene	Limonene	Cineole	Linalool	Menthone	Menthol	Dihydrocarvone	Carvone	Menthyl acetate	Azulene
Al Ain (Oven dried)	30	150	100, 1	48.2	2453	95.6	33.4	0.00	22.5	27.8	11443	0.00	0.00
	30	250	100, 1	26.3	1374	62.3	18.6	0.00	14.5	17.5	7413	0.00	0.00
	30	350	100, 1	26.7	1200	50.8	15.0	0.00	12.3	13.7	5898	0.00	0.00
	40	150	100, 1	28.1	1354	66.2	26.9	0.00	21.9	23.3	11200	0.00	0.00
	40	250	100, 1	22.3	953	44.3	14.7	0.00	12.2	13.8	5342	0.00	0.00
	40	350	100, 1	22.3	877	37.2	10.6	0.00	0.00	0.00	4125	0.00	0.00
	50	150	100, 1	24.1	925	40.8	12.9	0.00	0.00	0.00	4788	0.00	0.00
	50	250	100, 1	22.9	921	48.3	12.9	0.00	0.00	0.00	4659	0.00	0.00
	50	350	100, 1	14.8	730	38.0	10.6	0.00	0.00	0.00	4082	0.00	0.00
	50	350	150, 1	7.22	662	55.0	17.8	0.00	14.9	14.7	6620	0.00	0.00
	50	350	200, 1	10.9	1067	91.2	33.5	0.00	28.0	28.0	12303	0.00	0.00
	50	350	100, 2	15.9	722	39.1	13.7	0.00	9.8	0.0	5104	0.00	0.00
	50	350	100, 3	20.5	732	39.3	14.5	0.00	12.8	0.00	5656	0.00	0.00
Al Ain (Freeze dried)	50	350	100, 1	7.48	275	0.00	0.00	0.00	0.00	140	2090	0.00	0.00
Al Ain (Fresh)	50	350	100, 1	0.00	349	131	55.7	0.00	0.00	67.8	14969	0.00	0.00
Abu Dhabi (Oven dried)	50	350	100, 1	32.5	1614	73.5	27.5	0.00	26.4	31.5	10949	0.00	0.00
Abu Dhabi (Freeze dried)	50	350	100, 1	9.92	466	11.7	0.00	0.00	0.00	0.00	3219	0.00	0.00
Abu Dhabi (Fresh)	50	350	100, 1	0.00	326	65.9	48.2	0.00	0.00	0.00	12456	0.00	0.00
Dry Mint (France)	50	350	100, 1	7.49	255	124	0.00	0.00	36.8	587	4404	17.1	0.00
Dry Mint (India)	50	350	100, 1	9.12	103	167	0.00	0.00	0.00	103	1431	14.3	0.00
Dry mint (Syria)	50	350	100, 1	22.3	671	428	0.00	0.00	25.4	141	7877	31.9	0.00

Table 3 provides compositional data obtained by a steam distillation process. Preliminary results in the steam distillation process indicated that ethylacetate is a better extraction solvent than cyclohexane, hence, ethyl acetate was selected as the extraction solvent in all steam distillation experiments. The recovery of flavor principles with 15 and 20 min distillation time was comparable. Distillation time of 10 min gave lower recovery; hence, 15 min distillation time was selected for the steam distillation of samples.

The major flavoring principles, namely carvone and limonene, are used as marker compounds to compare the extraction efficiency of both SFE and steam distillation processes. The effect of temperature and pressure on the extraction of carvone and limonene are shown in Figure 5. The concentration of carvone and limonene decreased with increase in pressure and temperature. Although, the steam distillation process yielded higher concentration of a few flavor principles, higher concentration of major flavoring compounds were obtained in the SFE process, indicating that the quality of the SFE extract is superior as compared to that of the steam distillation process. Results showed that carvone, α-pinene, limonene, and linalool were significantly higher in the SFE extracts of local mint than that of the imported mint, but the contents of cineole and methyl acetate were higher in the imported mint than in the local mint. Results also indicate that menthone and azulene were not present in any SFE or steam distillation extracts and methyl acetate was only available in the SFE extracts of imported mint, while linalool was only present in the SFE extracts of local mint.

The quality of mint extract obtained from oven dried samples was found to be superior, as compared to that of freeze dried samples, which is in agreement with the findings by other researchers.[11] Results indicated that carvone, α-pinene, limonene, and linalool were significantly higher in local mint as compared to that of imported mint, but the contents of cineole and methyl acetate were higher in the imported mint than in the local mint. The differences in extraction yield and composition of imported mint, as compared to that of local mint samples, may be attributed to the differences in the variety, geographical conditions, and loss of volatile components during long storage periods in imported samples.

CONCLUSIONS

In this study, supercritical fluid technology was used to extract essential oils from local UAE mint, as well as dry imported mint from other countries. Extraction yield and composition of the extracted oils were obtained. The essential oils of the native mint grown in UAE were extracted using SC CO_2, and analyzed for the first time. Effects of temperature, pressure, CO_2 volume, and CO_2 flow rate on the yield and composition of the extracts were investigated. Extraction yield increased with pressure, temperature,

Table 3. Characterization of mint samples (steam distillation process)

Sample	Composition (μg/g, on dry basis)									
	α-pinene	Limonene	Cineole	Linalool	Menthone	Menthol	Dihydrocarvone	Carvone	Menthyl acetate	Azulene
Al Ain (Oven dried)	0.0	510	54.1	35.0	0.0	28.2	21.4	10780	0.0	0.0
Al Ain (Freeze dried)	0.0	69.6	0.00	0.00	0.00	0.00	159	2753	0.00	0.00
Al Ain (Fresh)	0.0	209	27.2	40.8	0.0	36.7	39.3	11504	0.0	0.0
Abu Dhabi (Oven dried)	0.0	104	63.5	26.2	0.0	24.6	24.4	8759	0.0	0.0
Abu Dhabi (Freeze dried)	0.0	92	26.1	21.7	0.00	22.0	18.8	8134	0.00	0.00
Abu Dhabi (Fresh)	0.0	195	35.6	35.3	0.0	26.5	34.8	11488	0.0	0.0
Dry Mint (France)	0.0	17.9	25.3	0.0	0.0	33.0	132	1815	0.0	0.0
Dry Mint (India)	0.0	11.7	118	0.0	0.0	0.0	28.2	1481	0.0	0.0
Dry Mint (Syria)	0.0	181	59.1	30.3	0.0	24.3	19.1	9110	0.0	0.0

Extraction conditions: steam (80%), distillation time (15 min) and receiving solvent (ethyl acetate).

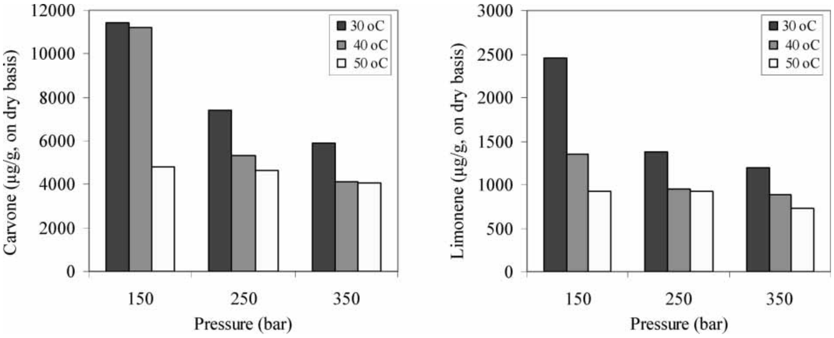

Figure 5. Effect of temperature and pressure on the composition (carvone and limonene) of mint extract (oven dried).

and CO_2 density. Linear relations were observed between extraction yield and CO_2 density. Moreover, extraction yield increased with CO_2 volume, but was nearly constant when the CO_2 flow rate was varied from 1 to 3 mL/min. Although extraction yield increased with increase in pressure and temperature, lower pressure and temperature resulted in obtaining better a quality of mint oil with respect to its composition. Therefore, in order to maximize the extraction of essential oils, the extraction process should be operated at the lower pressure and temperature of 150 bar and 30°C. The quality of extract obtained under SFE conditions was found to be superior as compared to that of the steam distillation process. The superior quality of locally grown mint can be attributed to its higher yield, as well as high concentration of major flavor principles, as compared to that of dry imported mint.

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