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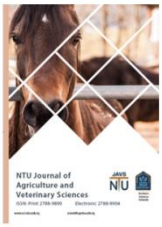

The effect of using some extraction techniques on the physical and chemical properties of essential oils of citrus peels

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Physical and chemical properties.

A B S T R A C T

The aim of the current study was to determine how extraction techniques affected the physical and chemical properties of the essential oils from citrus peels, specifically C.bitter orange and C.lemon. The results of the study were summarized as follows:

1- The extraction method using microwave-assisted hydro-distillation without solvents was the best in terms of yield, time, and energy savings compared to the extraction method by pre-treatment with ultrasound and then hydro-distillation and hydro-distillation extraction .

2- The essential oil extracted from dried bitter orange peels gave the best extraction yield using microwave-assisted hydro-distillation without solvents, reaching 4.21%, followed by lemon, which reached 2.21%.

3- No significant effect was observed between the extraction methods used and the physical properties. While observed, there is a slight significant effect between the extraction methods used and the chemical properties of the C. bitter orange peel essential oil.

4-It is noted that there is a slight significant effect between the extraction methods used and the physical and chemical properties of the C. Limon peel essential oil.

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Introduction

Citrus fruits are among the most effective medicinal plants in traditional medicine and the most important grown and produced worldwide[1]due to their high nutritional value, as they contain high levels of bioactive compounds such as phenols, flavonoids, vitamins, especially vitamin C and carotenoids, and essential oils [2]. Their consumption results in the loss of a large amount of components such as peels, seeds, and pulp, which together constitute 50% of the weight of citrus fruits, which makes them a challenge from an environmental perspective [3]. Essential oils are one of the most important products of secondary metabolism. They are a complex crude mixture consisting of about 400 aromatic compounds, which are usually volatile and have a nicearoma [4]. Although extracting these oils seems relatively easy, the quality and quantity of the essential oil as well as its chemical composition may vary greatly depending on the extraction method used. The extraction technique used in extracting citrus fruits varies depending on several factors, including: the type of part used in the oil The plant (peels, flowers, and leaves) and the final yield required, in addition to the time and cost [5], Essential oils are extracted from whole plants, different parts of plants, or from more than one part of the same plant, and it is often the part that carries most of the plant's odor, such as flowers, seeds, leaves, peels, fruits, and roots[6]. There are several techniques for extracting essential oils from citrus peels, including traditional techniques such as hydro-distillation (HD), cold expression, and solvent extraction [7]. However, many disadvantages may accompanied traditional extraction techniques, such as the loss of some volatile compounds, low extraction efficiency in addition to the long time and high energy accompanying the process, and the deterioration of unsaturated compounds or ester compounds through thermal effects or hydrolysis [8], prompted researchers over the past years to develop innovative sustainable technologies that can enhance environmental protection by reducing carbon dioxide emissions, reducing energy consumption, and improving extraction efficiency according to the concept of "green chemistry" [9]. Among these modern technologies are ultrasound-assisted extraction (UAE), microwave-assisted extraction (MAE), and supercritical fluid extraction (SFE) [10]. The aim of the current study was to determine how extraction techniques affected the physical and chemical properties of the essential oils from citrus peels, specifically C.bitter orange and C.lemon.

Materials & Methods

The study was carried out on samples of local citrus fruits (citrus, bitter oranges, and lemons), which were obtained from the local markets of the city of Mosul and Nineveh Governorate in mid-January for the 2021–2022 season. The fruits are carefully peeled with the help of a sharp knife to avoid any damage to the sebaceous glands, and the peels are cut into small pieces. They are spread in a single layer on a tray and dried in the oven at a temperature of 45–60 °C until the weight is stable. The drying process is coupled with hot air circulation to facilitate drying and avoid fungal growth [11], and then the dried citrus peels used in the study are crushed. Immediately before extraction to avoid loss of essential oil.

Extraction of citrus peel essential oil

Essential oils were extracted from citrus peels according to the following methods:

Hydro-distillation extraction apparatus and procedure

submit an amount of 50 grams of freshly groun d dry citrus peels in a 500-ml glass beaker (Clavanger **apparatus**) and add an amount of distilled water to it until the sample is completely submerged, and the extraction process continues for 3 hours[12]. The oil is collected and dried using anhydrous sodium sulfate. After that, the essential oils are kept at -4°C in tiny, dark glass tubes that are carefully closed.. This process was repeated under the same conditions several times.

Solvent-free microwave extraction apparatus and procedure

Solvent-free microwave extraction was performed in a microwave oven modified to fit a Clevenger apparatus. This oven had a frequency of 2450 MHz and a maximum delivered power of 700 W[13]. The temperature was adjusted using an external infrared thermometer (IR thermometer).

The extraction was done by submit an amount of 50 g of dry citrus peels in a 1-liter glass beaker after soaking them in a ratio of 1:2 (distilled water: weight of the sample) for 30 minutes. The beaker was then placed in a microwave oven, and the citrus peels were heated using energy. At a constant power of 700 watts at medium power for 30 minutes, the oil extract is separated and traces of moisture are removed using anhydrous sodium sulfate. The resulting essential oil is stored in an airtight container at a temperature of -4°C until the time of analysis.

extraction by hydro-distillation Assisted Pre-treatment by ultrasound apparatus and procedure

Submit 50 g of freshly grated dry citrus peels in a 500-ml spherical flask. Add an amount of distilled water to it until the sample is completely submerged. Then the spherical flask is covered and placed in an ultrasonic water bath device (ultrasound cleaning bath-powersonic 405). prepared by the Korean company Daihan Labtech and under extraction conditions ((at a frequency of 50 KHz with a power of 350 W and at a constant temperature of 25 °C, the length of the extraction period is 30 minutes) and after ultrasonic treatment, the resulting mixture is submitted to hydro-distillation using a Clevenger type device in order to extract essential oils[14]. The oil extract is separated from moisture using anhydrous sodium sulfate. The resulting essential oil is stored in an airtight container at a temperature of -4°C until the time of analysis.

Percentage of yield from extracting essential oils

The percentage of yield from extracting essential oils was calculated according to[15], as the yield of extracts is the ratio between the mass of the dry extract obtained and the mass of the dry plant material used for extraction, multiplied by 100.

Evaluation of physical properties

The refractive index was estimated using the Abbe 60/DR refractometer prepared by the World Health Organization (British origin) as stated [16]. Density of citrus peel essential oil in water was determined by the procedure given by [17].

Evaluation of chemical properties

Chemical properties such as acid value [18], peroxide value[19]. saponification value[20]. Ester value [21] .was estimated in the citrus peel essential oil.

statistical analysis

The results were analyzed statistically using the Social Program Statistics System (SPSS) according to the ANOVA test, and the arithmetic means were compared using the Duncan test to compare the means at the level ($P<0.05$) [22] .

Results & Discussion

The effect of different extraction methods on the percentage of yield.

It is noted from Table (1) that there are significant differences between the average values of % of the amount extracted from the different plant sources used in the study at the probability level (0.05). The percentage of the proceeds of extraction using the hydro-distillation (HD) extraction technique for both bitter orange and lemon peels was 3.63 and 1.86% (based on dry weight), respectively. While the percentage of proceeds of extraction using the hydro-distillation technique after pre-treatment with ultrasonic waves for both the bitter orange and lemon was 3.72 and 1.98%, respectively, and with regard to the hydro-distillation technique with the help of a microwave without solvents, the percentage yield for extraction of bitter orange, mandarin, and lemon was 4.21 and 2.21%, respectively. From this, it is clear that the hydro-distillation technique with the help of a microwave without solvents gave the highest percentage of returns, and that bitter orange peels had the highest percentage of returns amounting to 4.21%, followed by lemons with 2.21%, respectively. These obtained results may agree or differ with what a number of researchers found, as [23] found that the percentage of revenue from lemon peels using the HD extraction technique was (1.25%). [24] also stated that the percentage of revenue from lemon peels using the SFM+HD technique amounted to (2.5%) on a dry weight basis, while [25] stated that the percentage of revenue for both bitter orange and lemon using the HD extraction technique was 1.36 and 2.3%, respectively. while these percentages increased with the SFMA technique +HD, reaching for both bitter orange and lemon (4.5 and 6.3%), respectively [26] stated that treating citrus fruits, specifically sweet lemon, with ultrasound, followed by extraction using a microwave with the help of HD, has a significant impact on the production of essential oils from sweet lemon peel, as The percentage of rent ranged from 0.84 to 1.06%, and these differences between the results can be explained by the fact that the rent of essential oils is affected by several factors, including those related to the plant (species, cultivar, genetic composition, soil composition, geographical origin, etc.). On extraction conditions (extraction time, part of the plant used, temperature, and extraction method, etc) [27].

The effect of different extraction methods on the physical and chemical properties of bitter orange essential oil

The data in Table 2 shows that, at the probability level of 0.05, there are no significant effect was observed between the extraction methods used and the physical properties. While observed, there is a slight significant effect between the extraction methods used and the chemical properties of the C. bitter orange peel essential oil.. According to the table 2, the HD extraction method had the greatest refractive index, at 1.472. SFM+HD and ultrasonic pretreatment+HD came in second and third, with 1.468 and 1.467, respectively. This is consistent with what [28] reported. Likewise, the HD extraction technique gave the highest density, followed by the SFM+HD and ultrasonic pretreatment+HD techniques, with values ranging from 0.857 to 0.855 to 0.850 g/ml, respectively. These readings were close to what was mentioned by [29], as they indicated. The researchers found

that the density of bitter orange oil ranged between 0.848 and 0.849 grams/mL. Regarding the chemical properties, we find that the pH number for the HD, ultrasonic pretreatment + HD, and SFM + HD extraction methods is 7.923, 7.871, and 7.898 (mg KOH/gm oil), respectively, and the peroxide number for the three extraction methods mentioned above reached 55.220, 55.110, and 55.150 (mEq of oxygen/1000 gm of oil), respectively, while the saponification number was 168.330, 167.910, and 168.120 (mg KOH/gm oil), respectively, and the ester number was 160.407, 160.039, and 160.012 (mg KOH/gm oil), respectively. These results were similar to those reported by [30].

The effect of different extraction methods on the physical and chemical properties of lemon essential oil

The data in Table 3 shows that, at the probability level of 0.05, It is noted that there is a slight significant effect between the extraction methods used and the physical and chemical properties of the C.Limon peel essential oil. It is apparent from the data that the HD extraction method yielded the greatest refractive index of 1.475. Ultrasonic pretreatment+HD and SFM+HD followed with rates of 1.472 and 1.471, respectively. The HD extraction technique also gave the highest density, followed by the SFM+HD and ultrasonic pre-treatment+HD techniques, with values ranging from (0.852 - 0.845 - 0.842 g/ml), respectively. This is consistent with[31]. similarly, with the essential oil recovered from oregano, [32] found no significant differences between the HD and the SFM+HD extraction methods. These results were Comparable to what was reported by [33],who indicated that the values of the acidity number, saponification number, and ester were 6.77, 143.34, and 132.66 mg KOH/g oil, respectively. In addition, a study conducted by [34]. indicated that the acidity value and saponification value of lemon reached 8.68 and 158.42 (mg KOH/g of oil), respectively, while the peroxide value was 53.76 (millioxygen equivalent/1000 g of oil).

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Table 1.%yield extracting essential oils from citrus fruits

Average	Extraction Yield Extraction methods			Plant source
	C*	B*	A*	
3.85[a]	4.21[a]	3.72[b]	3.63[c]	C.bitter orange
2.01[b]	2.21[d]	1.98[ed]	1.86[e]	C.lemon
	3.36[a]	3.07[b]	2.94[ab]	Average

***** Dissimilar letters differ significantly at the level of 0.05.

*A= Hydro-distillation extraction

*B= extraction by hydro-distillation Assisted Pre-treatment by ultrasound

*C= Solvent-free microwave extraction

Table 2. The effect of different extraction methods on the physical and chemical properties of C. bitter orange peel essential oil.

Physical and chemical properties	Extraction methods		
	A*	B*	C*
physical properties			
refractive index	1.472[a]	1.467[a]	1.468[a]
Density	0.857[a]	0.850[a]	0.855[a]
chemical properties			
acid value	7. 923[a]	7. 871[c]	7. 898[ab]
peroxide valu	55.220[a]	55.110[c]	55.150[b]
saponification value	168.330[a]	167.910[c]	168.120[b]
Ester value	160.407[a]	160.039[b]	160.012[c]

***** Dissimilar letters differ significantly at the level of 0.05.

*A= Hydro-distillation extraction

*B= extraction by hydro-distillation Assisted Pre-treatment by ultrasound

*C= Solvent-free microwave extraction

Table3. The effect of different extraction methods on the physical and C.Limon peel essential oil.

Physical and chemical properties	Extraction methods		
	A*	B*	C*
physical properties			
refractive index	1.475[a]	1.472[a]	1.471[a]
Density	0.852[a]	0.842[b]	0.845[ab]
chemical properties			
acid value	7. 41[c]	7. 53[a]	7. 49[b]
peroxide valu	53.88[b]	54.13[a]	53.87[b]
saponification value	159.12[b]	159.93[a]	159.21[b]
Ester value	151.71[a]	152.4[a]	151.72[a]

***** Dissimilar letters differ significantly at the level of 0.05.

*A= Hydro-distillation extraction

*B= extraction by hydro-distillation Assisted Pre-treatment by ultrasound

*C= Solvent-free microwave extraction