Essential Oil Extraction for All: A Flexible and Modular System for Citrus Biomass Waste

Edson E. Armenta, Marcos A. Coronado, José R. Ayala,* José A. León, and Daniela Montes

The essential oil industry is a growing sector that generates 5.41 billion USD annually worldwide. Essential oils are widely used in medicine, agriculture, and perfumery. Although there are available systems in the market for domestic essential oil extraction, replacing the entire equipment in case of repair or malfunction can be costly. To address this problem, a pilot-scale essential oil extractor system was developed that operates through hydrodistillation. This system was used to process various citrus wastes such as green and yellow lemons, oranges, grapefruits, and *Eucalyptus globulus*. A factorial design was performed, and the best conditions were used to extract other biomass residues. GC-MS analysis revealed that the primary compound for orange, grapefruit, and green lemon essential oils is D-imonene, with 95.4%, 95.5%, and 49.2%, respectively. For yellow lemon the primary compound appeared to be D-limonene with 73.0% content, though the GC/MS data were less clear, and for eucalyptus, it is eucalyptol with 71.0%. The estimated production costs were 0.01 USD/mL, 0.04 USD/mL, 0.06 USD/mL, 0.07 USD/mL, and 0.15 USD/mL for orange, grapefruit, green lemon, yellow lemon, and eucalyptus essential oils, respectively. Therefore, the developed system is a competitive option for pilot-scale essential oil extraction.

DOI: 10.15376/biores.18.3.4977-4993

Keywords: Citrus; Essential oil; Eucalyptus; Extraction system

Contact information: Universidad Autónoma de Baja California, Instituto de Ingeniería, Blvd. Benito Juárez y Calle de la Normal S/N, Col. Insurgentes Este, 21280, Mexicali, Baja California, México;
** Corresponding author: ramon.ayala@uabc.edu.mx*

INTRODUCTION

Plants have been widely used in medicine, agriculture, and perfumes throughout history, but in recent years interest in essential oils (EOs) has increased noticeably due to their properties and applications in the food, pharmaceutical, and cosmetic industries, as well as having antioxidant and biocide activities (Maes *et al.* 2019; Manyako *et al.* 2022). Essential oils (EOs) are synthesized in aromatic plants as secondary metabolites and are distributed in multiple plant segments such as shells, bark, leaves, seeds, roots, stems, flowers, peels, and fruits. EOs are heterogeneous mixtures of terpenes, sesquiterpenes, acids, esters, phenols, and lactones (Aziz *et al.* 2018; Bailão *et al.* 2022; Jaramillo-Colorado *et al.* 2022). EOs are composed mainly of terpenes but also contain other chemical compounds (Maes *et al.* 2019).

The quality of essential oil may vary considerably, depending on factors such as geographical origin, plant variety, extraction method, and storage (Uysal *et al.* 2011). It can be determined by analyzing its organoleptic, physical, and chemical properties. The chemical composition of essential oil is usually the primary parameter used to assess its

quality, and GC-MS is the most widely used technique for this purpose (Yadav 2022).

In 2020, the global trade value of essential oils reached $5.41 billion USD. Orange essential oils (OEO) alone accounted for a trade value of $440 million during the same time (Observatory of Economic Complexity 2023). In 2021, worldwide orange production reached 75.57 million t (United Nations 2017). It has been estimated that 34% of the world's orange production is allocated for juice production, while 45% of the by-product generated during the process consists of orange peels (Velasco *et al*. 2017).

Mexico was one of the top exporters of OEO in 2020, ranking just below Brazil, the United States of America, and Germany (OEC 2023). Additionally, in 2021, Mexico became the world's fourth-largest producer of oranges (FAO 2023). The juice industry takes advantage of all the byproducts and waste streams as a strategy to cover operating costs. Therefore, it is not possible to take further advantage of the waste generated in this industry. However, there are also commercial establishments that produce and sell juices directly to customers. These businesses are not accounted for in industrial sector statistics. Nevertheless, they generate waste in the form of peels with pulp and seeds, which presents an attractive opportunity for revaluation. This waste represents an opportunity to develop efficient, cost-effective, and profitable technological processes that can transform it into raw material to produce high-value-added products.

Hydrodistillation is a common method for extracting essential oils from citrus fruits (Weng *et al.* 2019). The method is utilized at a laboratory, pilot, and industrial scales. At the laboratory scale, the equipment's easy installation and intuitive use make it an ideal option for didactic purposes, recreational activities, and preliminary experiments. At this scale, the Clevenger is the most widely used device in essential oil extractions (Kant and Kumar 2022). It has a processing capacity of 100 to 2,000 mL. At the pilot scale, processing capacity ranges from 20 to 500 L, and the equipment used is typically constructed with commercial steel, stainless steel, or copper. The work's primary objective is to achieve optimal distillation parameters and determine the technical and economic feasibility of producing essential oils and floral water. This processing scale makes it possible to produce important quantities of essential oils for commercial purposes (Sanchez *et al.* 2022; Rodríguez *et al.* 2012). The industrial scale is typically geared towards distillation processes with feed capacities exceeding 500 L. Its primary objective is to obtain EOs and floral water of high value, maximum quality, and efficient production batches (Rodríguez *et al.* 2012).

Small, integrated systems are available on the market for essential oil extraction. These systems are domestic in scale, and their production and yields are like laboratory-scale ones. One disadvantage of these systems is their lack of repairability and rigidity. In the event of system failure, the entire system must be replaced. This issue represents an opportunity for designing and developing modular essential oil extraction systems that are flexible, easy to maintain, and scalable for increased essential oil production.

In this work, an essential oil extraction system was developed that operates through pilot-scale hydrodistillation. Citrus wastes from green lemon (*Citrus aurantifolia*), yellow lemon (*Citrus limon*), orange (*Citrus sinensis*), grapefruit (*Citrus paradisi*), and eucalyptus leaves (*Eucalyptus globulus*) were processed. The extraction system was designed for portability, flexibility, ease of use, and maintenance. The essential oil quality was evaluated by gas chromatography coupled with mass spectrometry. The system's performance was evaluated based on operation yields, production costs, and energy requirements.

EXPERIMENTAL

Design and Installation of the Essential Oil Extraction System

The essential oil extraction system was developed using the hydrodistillation technique. The extraction process involved stages of size reduction, heating, an extractor tank, gas and steam piping, a condenser, and a decanter (da Costa *et al.* 2022). The equipment was specifically designed to process peel, and a support was placed inside the extractor to simulate steam distillation for leaves. Modularity, portability, flexibility, and easy maintenance were prioritized during the development of the system.

A Retsch GM 300 mill with a 5 L capacity container was utilized during the size reduction stage. A conventional blender can replace this equipment, repeating the process as needed to obtain the desired volume. The significance of this stage lies in the fact that when extracting essential oils from peels, volatile compounds are located between the peel and the fruit's albedo. Therefore, mechanical support is necessary to break the peel and release the essential oil, favoring the mass transfer process between the peel and steam.

The heating source used was a liquefied petroleum (LP) gas stove with a double-ring burner. It was chosen for its versatility and independence from electrical power. For the extractor equipment, a 20 L aluminum hermetic container was considered, which includes a pressure gauge and a relief valve. A 2/3-inch outlet was adapted to this extractor to connect a food-grade gas hose, which was insulated and connected to a condenser.

The stainless steel condenser was designed as a single-pass unit with two concentric tubes and two outlets for 3/8-inch hoses. Public water served as a cooling medium directly connected between the water outlet and the condenser, operating in countercurrent. A 250 mL florentine was placed at the end of the exchanger to continuously separate the essential oil from the water, functioning as a decanter and EO accumulator. Drinking water was used for the essential oil extraction. The extraction system can be observed in Fig. 1.

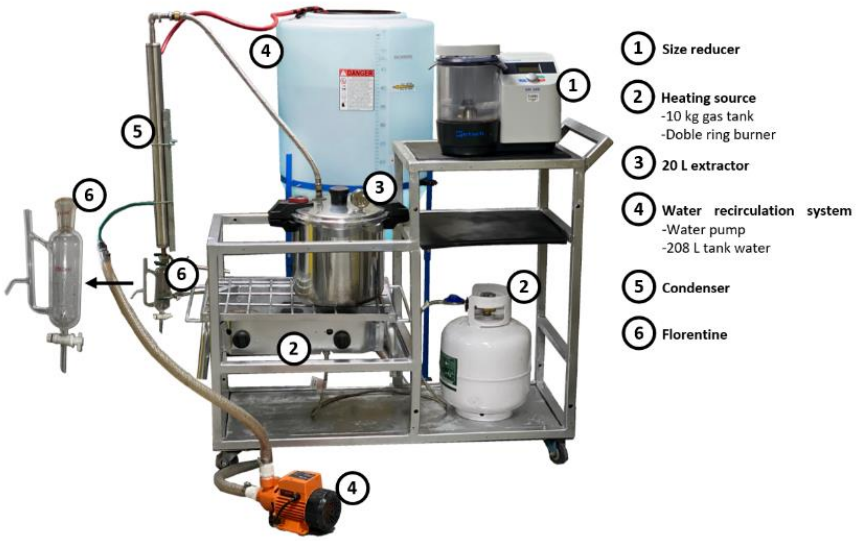

Fig. 1. Essential oil extraction system

Factorial Design Applied to the Essential Oil Extraction from Orange Waste

To characterize the extraction system and determine its optimal operating levels, a 23 factorial design was specifically developed for orange waste (OW) (Ghasemi *et al.* 2021). OW, which includes orange peels, pulp, and seeds, is easily obtainable from various

commercial sources and is currently collected at no cost. Due to the size and complexity of the other biomass wastes, they were not included in the factorial design. Using OW as a representative biomass waste allows for easier management at pilot and industrial scales, takes advantage of the moisture content in the pulp, and reduces pretreatment costs. The key operating conditions identified for OW can also be applied to other biomass wastes.

In the experimental design, the orange peels processed included the peel, pulp, and seeds. These were collected from a commercial establishment that specializes in producing and selling juices.

Factor 1 in the extraction process was the ratio of OW to water volume, with values of 1.7 and 1.9 L per kg of OW (Yumnam *et al.* 2023). This factor primarily affects the mass transfer between the orange EO and water. Increasing the water volume also increases the amount of EO produced up to a certain point, after which the volume of EO plateaus. In other words, additional water no longer increases the volume of EO (Ayala *et al.* 2017).

Factor 2 involved the reduction of OW size (Karanicola *et al.* 2021; Wei *et al.* 2023). This size reduction creates more contact between the plant material and the water, facilitating EO extraction. Grinding times of 5 and 10 s were selected. This operation was performed with a fraction of the drinking water used in factor 1. The grinding times can vary depending on the mill's power used. A conventional blender, for example, can take up to 1 min. The ground waste should have a uniform and fine appearance to allow the passage of bubbles.

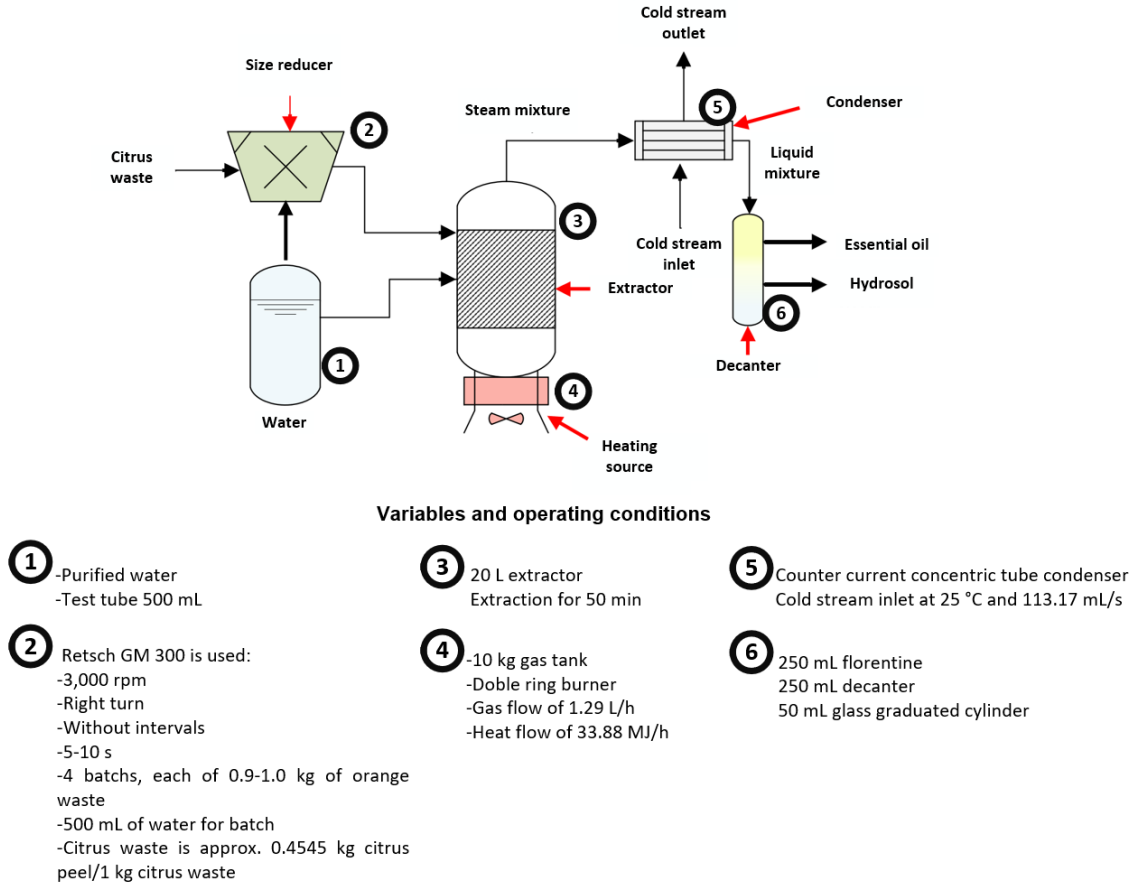

Fig. 2. Stages and parameters in the operation of EO extraction

Without having a direct way to measure pressure and temperature inside the hydrodistillation vessel, factor 3 was the amount of plant material, defined as 3.5 and 4.0 kg. The input mass is a critical factor in experimental design due to its direct impact on the pressure within the extraction vessel. Pressure, in turn, increases or decreases the mixture's boiling point. Hence making the amount of plant material a crucial factor for extraction yield.

The response variable was the volume of EO in mL obtained in the extraction process. OW was acquired from the same supplier on the day of extraction at the same time to minimize variability in the experimental design. The supplier was a juice sales establishment. After extraction, the EO was separated from the floral water using a funnel decanter. The volume was measured using a 50 mL graduated cylinder immediately after extraction and separation from the floral water to prevent losses due to volatilization and hydrolysis. The extraction method remains constant in the parameters of the other process stages and the system operated under batch conditions. These parameters are described in Fig. 2.

Gas Chromatography-Mass Spectrophotometry Analysis

After selecting the operating conditions obtained in the factorial design, EO was extracted from orange, green lemon, yellow lemon, grapefruit, and eucalyptus leaves wastes. Gas chromatography coupled to mass spectrometry of the different EO was performed at the University of Cartagena, Colombia.

The EO was analyzed using an Agilent Technologies GC-MS system model 7890A Network GC coupled to a mass selective detector model 5975 equipped with a split/split-less injection port (230 °C, split ratio 20:1). The mass spectra were obtained by electron-impact ionization at 70 eV energy. The GC elements were an HP-5MS capillary column (30 m × 0.25 mm id × 0.25 μm df) with 5% phenyl-poly (methyl siloxane), and a stationary phase was used for the separation of mixtures. The oven started at 50 °C for 2 min, and its temperature increased at a rate of 5 °C/min, up to 250 °C. The carrier gas was helium, with an inlet pressure at the head of the column of 12.667 psi at a rate of 1.172 mL/min at 50 °C. The samples were prepared by diluting 20 microliters of EO in 980 microliters of dichloromethane. From the resulting solution, 1 microliter was taken for injection. The MS range used was *m*/*z* 30 to 600. The results were compared with the NIST library ver. 2.0 from 2008 (Jaramillo *et al.* 2022).

Cost Analysis for EO Extraction

The authors constructed a metal structure to mount most of the components. Once the entire system was assembled, and essential oil samples were obtained, a water recirculation system was added. This system included a 208 L water storage tank and a Trupper peripheral water pump with a maximum capacity of 42 L/min and a maximum height of 45 m. The recirculation system reduced water consumption per extraction, and the environmental impact caused by water misuse, lowered the cost of extracting essential oil per mL, and increased the equipment's independence from a fixed installation.

Operating costs were determined by considering the necessary inputs for the experimental runs, which included water and energy (Cruz-Sánchez *et al.* 2022; León *et al.* 2022). The plant material was donated from various sources, so no cost was associated with it. Citrus waste was donated from a local juice business and eucalyptus leaves were gathered from the eucalyptus trees inside the university campus.

The cost of energy inputs was calculated based on the energy requirements of the extraction process stages. The size reduction stage required energy, and the extraction stage needed heat. The Retsch GM300 equipment for size reduction has a power range of 1,100-3,000 W and 13 A and is used for 5 to 10 s. The heat source operated on LP gas at a flow rate of 1.29 L/h.

The cost of water was determined based on the price per m^3 of commercial water. Two scenarios were considered: before and after the installation of the water recirculation system. Potable water was used to feed the extraction equipment, and the amount of water required varied from 5.95 to 7.6 L depending on the amount of plant material used. The equations used to calculate the costs are described in Eqs. 1, 2, and 3.

$$TCN_{EO} = \left(\frac{nP_{mill}Et_{mill}}{3600}\right)_{grinding} + (W_{ex}V_{ex})_{Extractor} + \left(Gt_{op}\right)_{Heater} + \left(\frac{W_c}{Qt_{cold}}\right)_{condenser} \quad (1)$$

$$TCN_{REO} = \left(\frac{nP_{mill}Et_{mill}}{3600}\right)_{grinding} + (W_{ex}V_{ex})_{Extractor} + \left(Gt_{op}\right)_{Heater} + \left(\frac{P_{pump}Et_{pump}}{3600}\right)_{pump} \quad (2)$$

$$Y_{EO} = \frac{TC_{EO}}{V_{EO}} \quad (3)$$

where TCN_{EO} represents the total essential oil extraction cost of a given plant without water recirculation (USD), and TC_{REO} the total essential oil extraction cost of a given plant with water recirculation (USD). The variable n denotes the number of grinding batches, P_{mill} the mill power (kWh), E the electricity cost (USD/kWh), t_{mill} the grinding time (s,) W_{ex} the purified water price (USD/L), V_{ex} the extractor volume (L), G the gas price (USD/h), t_{op} operation time (h), W_c the public water price (USD/m^3), Q water flow (m^3/s), t_{cold} the running time of the condenser (S), P_{pump} the pump power (kWh), t_{pump} the pump operation time (s), $Y_{EO,}$ essential oil volume price (USD/mL) and V_{EO} the essential oil volume (mL).

Each essential oil extraction cost was determined, and an average cost was calculated for multiple extractions of the same plant material. The inputs and electrical energy costs were based on commercial tariffs, with commercial water costing 19.55 USD/m^3 and potable water costing 0.02 USD/L. The commercial electricity tariff was 0.2 USD/kWh, and the cost of LP gas was estimated at 0.66 USD/L.

RESULTS AND DISCUSSION

Extractor System for Essential Oils

The essential oil extraction system mounted on the steel structure is shown in Fig. 1. According to the characteristics of the plant material, it may be necessary to use a mill. The plant material is loaded into the extraction equipment, and potable water is added according to the appropriate ratio. The lid is closed, and the burner is turned on. After 5 min of flame, the recirculation pump is turned on to maintain the condenser at the appropriate temperature. The first condensate drop occurs 20 min after the burner is turned on. With these steps, the extraction process takes approximately 50 min from the burner ignition. This operation time was established based upon prior observation on the EO extraction in the extractor system. After 50 min of operation the amount of EO extracted is not significant.

Design of Experiments Results

Table 1 shows the results of OW essential oil extractions for each of the factorial design runs. The response variables were the volume of essential oil and the extraction yield. The volumes were low for a grinding time of 5 s, a little less than 50% of those obtained with 10 s of grinding. Additionally, in the 5-second grinding treatments, the oil had a yellowish appearance, unlike the essential oil obtained in the 10-second treatments, which had a colorless appearance.

Table 1. Factorial Design for Essential Oil Extraction from OW

Run	Orange waste in kg (A)	Grinding time in s (B)	Water/waste ratio (C)	EO volume In mL	Yield in mL/kg
1	3.5	5	1.7	30	8.6
2	3.5	5	1.7	30	8.6
3	3.5	5	1.9	65	18.6
4	3.5	5	1.9	58	16.6
5	3.5	10	1.7	32	9.1
6	3.5	10	1.7	27	7.7
7	3.5	10	1.9	64	18.3
8	3.5	10	1.9	58	16.6
9	4	5	1.7	33	8.3
10	4	5	1.7	38	9.5
11	4	5	1.9	65	16.3
12	4	5	1.9	69	17.3
13	4	10	1.7	30	7.5
14	4	10	1.7	30	7.5
15	4	10	1.9	67	16.8
16	4	10	1.9	68	17.0

The conditions for obtaining the highest amount of essential oil were found to be a grinding time of 10 s, a water-to-waste ratio of 1.9 L per kg, and 4 kg of processed waste. Conversely, the lowest amount of essential oil was obtained with a ratio of 1.9 L of water per kg of OW, 5 s of grinding, and 3.5 kg of waste.

Table 2. ANOVA Analysis for the OW Essential Oil Extractions

Source	EO volume					EO Yield				
	DF	Adj SS	Adj MS	F-Value	P-Value	DF	Adj SS	Adj MS	F-Value	P-Value
Model	7	4,477	639.57	67.32	0	7	313.728	44.818	61.79	0
Orange waste A	1	81	81	8.53	0.019	1	1	1	1.38	0.274
Grinding time B	1	9	9	0.95	0.359	1	0.59	0.59	0.81	0.394
Water/waste relation C	1	4,356	4,356	458.53	0	1	310.641	310.641	428.3	0
AB	1	4	4	0.42	0.535	1	0.232	0.232	0.32	0.587
AC	1	9	9	0.95	0.359	1	0.141	0.141	0.19	0.671
BC	1	9	9	0.95	0.359	1	0.562	0.562	0.78	0.404
ABC	1	9	9	0.95	0.359	1	0.563	0.563	0.78	0.404
Error	8	76	9.5			8	5.802	0.725		
Total	15	4,553				15	319.531			

The highest yield was achieved by processing 3.5 kg of waste with 5 s of grinding and a 1.9 water-to-waste ratio. On the other hand, the lowest yield was obtained by processing 4 kg of OW with 10 s of grinding and a ratio of 1.7. Generally, yields obtained with 10 s of grinding were comparable to the highest yields reported in the literature (Manyako *et al.* 2022). The ANOVA result for the extraction of essential oil from OW is shown in Table 2.

According to Table 2, only factors A and C were significant when evaluating the volume of essential oil obtained as the response variable. Only factor C was significant when analyzed with yield as the response variable in the factorial design. In both cases factor C, defined as the water/waste ratio, was the most important factor in the extraction. None of the interactions resulted in a P-value <0.05. Figure 3 shows the simultaneous analysis of the response variables.

Figures 3a and 3b were created to identify the operational range in which a volume of more than 55 mL of EO can be obtained with a yield exceeding 15 mL/kg. This region is depicted in a white region in Figs. 3a and 3b, allowing for flexibility in the operating conditions. Moreover, the optimization point for these conditions occurs using 4 kg of OW, with 10 s of grinding and a ratio of 1.9. These conditions ensure the maximum volume of EO with the highest yield, as per the factorial design data.

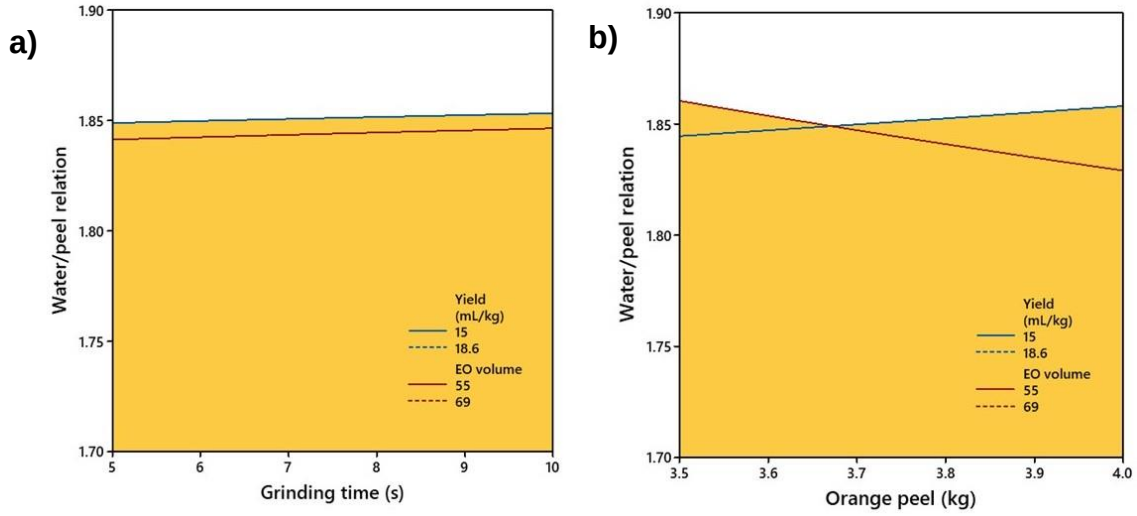

Fig. 3. Simultaneous response variable analysis, a) factor A constant, b) factor B constant

Essential Oils Gas Chromatography-Mass Spectrophotometry Results

Table 3 shows the compounds identified by GC-MS for the essential oils of orange, grapefruit, green lemon, yellow lemon, and eucalyptus leaves wastes. The essential oils of orange, grapefruit, and green lemon are mainly composed of limonene. According to the default output, 1,5-dimethyl-1,5-cyclooctadiene was reported as the most representative compound in the essential oil of yellow lemon, though this is due to similar *m*/*z* output, this compound was tentatively assigned as D-limonene. Eucalyptol was found to be the major component in the essential oil of eucalyptus leaf. These representative compounds were detected with a retention time between 10.9 to 11.1 min.

The values obtained in the chromatograms coincide and fall within the range of published literature. The content of D-Limonene in orange EO ranges from 71.26% to above 90% (Moemenbellah-Fard *et al.* 2020; Li Yan *et al.* 2022).

Table 3. GC-MS for the Essential Oils

Peak number	RT (min)	Compound	CAS	% Composition				
				OEO	GEO	GLEO	YLEO	EEO
1	3.1773	Glyodin acetate	000556-22-9	-	-	0.3405	-	-
2	3.2074	(Z)-9-Hydroxy-2,4-dimethyl-non-7-enoic acid lactone	1000144-65-1	-	-	0.1353	-	-
3	3.3204	Desmethyldoxepin	001225-56-5	-	-	1.2602	-	-
4	6.9374	β-Thujene	028634-89-1	-	-	0.6088	0.3495	-
5	7.1333	L-α-Pinene	007785-26-4	-	-	2.6444	-	1.2372
6	7.1485	D-(+)-α-pinene	007785-70-8	0.7994	0.7494	-	1.8689	-
7	7.6382	Camphene	000079-92-5	-	-	0.1714	0.0684	0.0551
8	8.6330	Sabinene	003387-41-5	0.2292	0.1491	-	-	-
9	8.7008	β-Pinene	000127-91-3	-	0.0174	18.7433	7.9132	-
10	9.3035	2,3-Dehydro-1,8-cineole	092760-25-3	-	-	0.0411	-	-
11	9.4166	β-Myrcene	000123-35-3	2.1961	1.9881	1.0515	1.6305	0.0609
12	9.8309	α-Phellandrene	000099-83-2	0.0681	-	0.0616	-	-
13	9.8536	α-Thujene	002867-05-2	-	0.0454	-	0.0909	-
14	9.9215	Octanal	000124-13-0	0.1294	0.1239	-	-	-
15	10.0797	(+)-3-Carene	000498-15-7	0.2155	-	-	-	-
16	10.3509	α-Terpinene	000099-86-5	-	-	0.0997	0.391	-
17	10.3736	4-(2-Methylamino)ethyl)pyridine	055496-55-4	0.0217	-	-	-	-
18	10.7126	o-Cymene	000527-84-4	-	-	6.254	-	-
19	10.7278	p-Cymene	000099-87-6	-	0.0668	-	0.583	-
20	10.7352	4-Ethyl-m-xylene	000874-41-9	-	-	-	-	18.9446
21	10.9689	1,5-dimethyl-1,5-Cyclooctadiene*	003760-14-3	-	-	-	73.0405	-
22	11.0216	D-Limonene	005989-27-5	95.4047	95.5393	49.1568	-	-
23	11.0140	Eucalyptol	000470-82-6	-	-	-	-	71.0404
24	11.3983	trans-β-Ocimene	003779-61-1	-	-	0.1177	0.0665	0.204
25	11.8203	β-Ocimene	013877-91-3	-	0.1307	0.2572	-	-
26	11.8355	cis-Ocimene	003338-55-4	-	-	-	0.0799	0.0992
27	12.205	γ-Terpinene	000099-85-4	-	0.0542	4.8014	10.3351	2.7201
28	13.493	Terpinolene	000586-62-9	0.0429	-	0.5317	0.604	-
29	14.1713	Linalool	000078-70-6	-	-	0.2871	-	-

30	14.1789	cyclofenchene	000488-97-1	0.3714	-	-	0.0857	-
31	15.505	cis-Limonene oxide	013837-75-7	-	-	0.1175	-	-
32	15.716	trans-Limonene oxide	004959-35-7	-	-	0.0946	-	-
33	15.7236	trans-Pinocarveol	000547-61-5	-	-	-	-	0.2662
34	16.0928	2-Methyl-1-nonene-3-yne	070058-00-3	-	-	0.0794	-	-
35	16.7936	Pinocarvone	030460-92-5	-	-	-	-	0.0956
36	17.0045	endo-Borneol	000507-70-0	-	-	0.0827	-	-
37	17.517	Terpinen-4-ol	000562-74-3	0.0306	0.0385	1.4375	0.5517	0.9139
38	18.0218	m-Cymen-8-ol	005208-37-7	-	-	0.1183	-	-
39	18.0897	trans-p-mentha-1(7),8-dien-2-ol	1000374-16-7	-	-	-	-	0.2575
40	18.1801	Terpineol	1000411-59-6	0.0463	-	1.4712	-	0.5491
41	18.1877	α-Terpineol	000098-55-5	-	-	-	0.5622	-
42	18.210	L-α-Terpineol	010482-56-1	-	0.0458	-	-	-
43	18.964	Decanal	000112-31-2	0.0749	0.1468	0.169	-	-
44	19.2727	3-Carene	013466-78-9	-	-	0.0835	-	-
45	19.3028	Sabinol	003310-02-9	-	-	-	-	0.1567
46	19.9358	cis-p-mentha-1(7),8-dien-2-ol	1000374-16-8	-	-	-	-	0.2401
47	20.4859	3,7-dimethyl-2,6-Octadienal	000106-26-3	-	-	0.8086	-	-
48	21.2469	(+/-)-Lavandulol	000498-16-8	-	-	0.1067	-	-
49	21.8498	Citral	005392-40-5	-	-	1.0754	0.0857	-
50	22.9725	Thymol	000089-83-8	-	-	-	-	0.418
51	23.3343	3-Methyl-4-isopropylphenol	003228-02-2	-	-	-	-	0.1753
52	23.6206	3-Methyl-4-isopropylphenol	003228-02-2	-	-	-	-	0.1806
53	24.9167	2-Acetoxy-1,8-cineole	057709-95-2	-	-	-	-	0.1057
54	25.1653	Hexestrol	000084-16-2	-	-	0.0661	-	-
55	26.0092	Neryl acetate	000141-12-8	-	-	0.1058	0.5865	-
56	26.243	α-Copaene	1000360-33-0	-	0.1053	-	-	-
57	26.8381	Lavandulyl acetate	025905-14-0	-	-	0.2227	-	-
58	26.899	β-Copaene	018252-44-3	-	0.0714	-	-	-
59	28.021	Phenylephrine	000059-42-7	0.0198	-	-	-	-
60	28.036	Caryophyllene	000087-44-5	-	0.3109	1.0245	0.1673	-
61	28.8199	trans-α-Bergamotene	013474-59-4	-	-	1.2988	-	-

62	28.8276	cis-α-Bergamotene	018252-46-5	-	-	-	0.2739	-
63	29.4303	1,4,7,-Cycloundecatriene, 1,5,9,9-tetramethyl-, Z,Z,Z-	1000062-61-9	-	-	0.1472	-	-
64	29.807	(-)-β-Santalene	025532-78-9	-	-	0.118	-	-
65	30.7791	α-Selinene	000473-13-2	-	-	0.3892	-	-
66	31.6155	cis-α-Bisabolene	029837-07-8	-	-	0.124	-	-
67	31.8416	β-Bisabolene	000495-61-4	-	-	2.4758	-	-
68	32.377	δ-Cadinene	000483-76-1	-	0.116	-	-	-
69	33.5521	Germacrene B	015423-57-1	-	-	0.3288	-	-
70	34.5694	Caryophyllene oxide	001139-30-6	-	-	0.1771	-	-
71	34.6523	(-)-Globulol	000489-41-8	-	-	-	-	0.727
72	34.9462	γ-Selinene	000515-17-3	-	-	-	-	0.147
73	36.4456	Isospathulenol	088395-46-4	-	-	0.1271	-	-
		Others		0.35	0.301	1.1868	0.6656	1.4058

OEO: Orange Essential Oil, GEO: Grapefruit Essential Oil, GLEO: Green Lemon Essential Oil, YLEO: Yellow Lemon Essential Oil, EEO: Eucalyptus Essential Oil.
*Possible D-Limonene due to similar retention time.

Table 4. Extraction System Comparative

EO	Present Work		Literature			
	Main compound in the EO (%)	EO yield (mL/kg)	Main compound in the EO (%)	EO yield (mL/kg)	Operation Conditions	Reference
Orange	D-Limonene (95.40)	16.8	Limonene (75.3)	8.35	Laboratory scale, steam distillation	Manyako *et al.* 2022
			Decanal (12)	5.9	Pilot plant, steam distillation	Manyako *et al.* 2022
Yellow Lemon	1,5-dimethyl-1,5-Cyclooctadiene (73.04)*	7.74	NA	3.39	Pilot plant, 60 min operation, 4 kg of yellow lemon peel, hydrodistillation	León *et al.* 2020
			NA	5.13-5.15	Pilot plant, 100 min operation, 8 kg and 12 kg of yellow lemon peel, hydrodistillation	León *et al.* 2020
Green Lemon	D-Limonene (49.15)	8.11	Citronellol (10.67)	7.7	Laboratory scale, 4 h, 0.5 kg of green lemon peel, hydrodistillation	Suresh *et al.* 2021
Grapefruit	D-Limonene (95.53)	6.75	NA	5.42	Pilot plant, 60 min operation, 16.636 kg of grapefruit peel, steam distillation	Justiniano-Medina *et al.* 2022
			Limonene (92-96)	2.56	Pilot plant, 0.5 kg of dry grapefruit peel, Twin-screw struder	Trujillo-Juárez *et al.* 2021
			Limonene (88.6)	4.4	Laboratory scale, 180 min operation, 0.25 kg of grapefruit, hydrodistillation	Uysal *et al.* 2011
Eucalyptus	Eucalyptol (71.04)	7 .55	Eucalyptol (85.4)	0.72	Laboratory scale, 180 min operation, 0.5 kg of eucalyptus leaves, hydrodistillation	Torrenegra *et al.* 2019
			Eucalyptol (89.9)	1.21	Laboratory scale, 180 min operation, 0.5 kg of eucalyptus leaves, microware assisted hydrodistillation	Torrenegra *et al.* 2019

NA: Not Available in the study
*Possible D-Limonene due to similar retention time

In grapefruit EO, D-Limonene content has been recorded from 81.86% to 96% (Uysal *et al.* 2011; Trujillo-Juárez *et al.* 2021; Li Yan *et al.* 2022). For green lemon EO, D-Limonene content has been recorded between 47.24% and 61.8% (Ashmawy *et al.* 2019; Moemenbellah-Fard *et al.* 2020). The most abundant compound in yellow lemon EO was found to be 1,5-dimethyl-1,5-Cyclooctadiene, comprising 73.04% of the oil's composition. Other studies have identified Geranial and D-Limonene as the primary compounds in yellow lemon EO, with concentrations ranging from 29% to 44.3% for Geranial (Marongiu *et al.* 2006) and from 48.28% to 71% for D-Limonene (Cardoso *et al.* 2022; Khang *et al.* 2022). Neither Geranial nor D-Limonene were reported in the results presented in Table 3. However, it is worth noting that the retention time of 1,5-dimethyl-1,5-Cyclooctadiene is very similar to that of D-Limonene (10.9689 min and 11.0216 min respectively), suggesting that the reported concentration of 73.04% could be attributed to D-Limonene as indicated by previous literature (Khang *et al.* 2022). Eucalyptol values in eucalyptus leaf EO vary from 15.1% to 89.9% (Torrenegra *et al.* 2019; Moemenbellah-Fard *et al.* 2020; Khedhri *et al.* 2022). The variations found in this EO are attributed to the extraction method, process scale, and eucalyptus tree variety.

When scaling a laboratory process to a pilot one, certain changes occur in the operating conditions. Those can impact the concentrations of essential oils. Table 4 presents a comparison of some systems for essential oil extraction and their operating level.

The compositions of the essential oils obtained in the extractor system are generally within the range reported in the literature for laboratory and pilot scales. It should be noted that a longer time is required to obtain the EO in pilot-scale equipment. The observed increase in processing time may be attributed to differences in certain process parameters when scaling up from laboratory to pilot scale. At the laboratory level, the hydrodistillation process typically operates at temperatures between 100-110 °C. While this operating temperature was achieved at the pilot scale, the amount of energy supplied per unit of residue was not equivalent to that used at the laboratory scale. As a result, the net heat applied during pilot scale processing was lower than that used at the laboratory level, leading to an increase in operating time.

Essential Oil Extraction Cost

Table 5 presents the production cost information obtained by solving Eqs. 1 through 3 for the optimal extraction conditions of the various plant materials. The cost presented in Table 5 does not include investment costs, it is only the operating cost of the installed system. Adding a water recirculation system can reduce the cost of essential oil production by 75.4% on average, making it a crucial element in the process. The system lowers operating costs and enables operation in regions with limited water resources. The most cost-effective extraction was achieved with orange essential oil, costing only 0.01 USD per mL.

Table 5. EO Production Cost

Essential Oil	YEO without recirculation (USD/mL)	YEO with recirculation (USD/mL)
Orange	0.05	0.01
Grapefruit	0.14	0.04
Green lemon	0.25	0.06
Yellow lemon	0.29	0.07
Eucalyptus	0.62	0.15

There is a difference in the extraction costs of essential oils despite using the same conditions in all cases. This may be attributed to the availability of essential oil, which varies among wastes. For instance, orange peels have the highest availability of essential oil, while eucalyptus leaves have the lowest. By redesigning the extraction system to operate at larger scales, it may be possible to further reduce the production cost of essential oil. It can be achieved by taking advantage of the availability and utilization of additional energy streams as well as the potential to generate other valuable by-products. When extracting essential oil from citrus fruits, it is crucial to reduce the size of the fruit to allow the essential oil to flow freely through the interface of the peel and albedo into the water. The volumes of essential oil produced were up to 20 times compared to laboratory extractions using a standard Kimble Kem-Kit Taper 19/22. The comparison was based on the volume of essential oil produced rather than the ratio of essential oil per kg of processed waste. Additionally, the chromatography of essential oils demonstrates that the compound profile, including their concentrations, is analogous to those obtained at the laboratory level. These results make the developed system a pilot-level alternative capable of extracting high-quality essential oil. Due to its flexibility and portability, the developed extractor system can operate in remote areas and under water scarcity conditions by maintaining a recirculation system. The costs associated with EO extraction by the developed system are competitive.

CONCLUSIONS

1. Most of the pilot-scale essential oil extraction system was constructed using existing market elements, with only the condenser and the support structure being custom-designed and fabricated in a mechanical workshop. It makes the system easy to replicate without adding unnecessary complexity. Furthermore, the system can be adapted to the specific needs of any region where it may be utilized.
2. According to the experimental design, the best operating conditions for OW were 4 kg waste with 10 s of grinding and a water-to-waste ratio of 1.9. These conditions were used in all the extractions in the system.
3. One critical aspect of pilot-scale production is the identification of representative compounds in the essential oil. The concentrations of D-limonene and eucalyptol, two of the most important compounds, were within the acceptable range, producing essential oils of comparable quality to those obtained on a laboratory scale.
4. The operating costs of the system are low, providing an affordable opportunity for individuals or businesses interested in implementing an essential oil extraction system for personal or commercial use.
5. To implement a similar extraction system, it is recommended to begin by conducting laboratory tests to identify key extraction parameters and methods. The next step is to select materials and equipment that are available on the market and are easy to repair and maintain. Once a prototype has been constructed, its performance can be evaluated through exploratory testing and experimental design. It is also important to select appropriate analytical methods that are relevant to the specific case. By following these steps, it is possible to develop a customized extraction system that meets the specific applications needs.

ACKNOWLEDGMENTS

The authors thank the National Council on Science and Technology (CONACYT) and the Engineering Institute of the Universidad Autónoma de Baja California for its support in the development of this work.

REFERENCES CITED

Agriculture Organization of the United Nations statistics division 2021 Crops and livestock products, (https://www.fao.org/faostat/en/#data/QCL), Accessed 2 February 2023.

Ashmawy, A., Mostafa, N., and Eldahshan, O. (2019). "GC/MS analysis and molecular profiling of lemon volatile oil against breast cancer," *Journal of Essential Oil-Bearing Plants* 22(4), 903-916. DOI: 10.1080/0972060x.2019.1667877

Ayala, J. R., Montero, G., Campbell, H. E., García, C., Coronado, M. A., León, J. A., Sagaste, C. A., and Pérez, L. J. (2017). "Extraction and characterization of orange peel essential oil from Mexico and United States of America," *Journal of Essential Oil-bearing Plants* 20(4), 897-914. DOI: 10.1080/0972060x.2017.1364173

Aziz, Z. A., Ahmad, A., Setapar, S. H. M., Karakucuk, A., Azim, M. M., Lokhat, D., Rafatullah, M., Kamal, M. A., and Ashraf, G. M. (2018). "Essential oils: Extraction techniques, pharmaceutical and therapeutic potential-a review," *Current Drug Metabolism* 19(13), 1100-1110. DOI: 10.2174/1389200219666180723144850

Bailão, E. F. L. C., Pereira, D. G., Romano, C. A., Paz, A. T. de S., Silva, T. M. e., Paula, J. R. de, Gomes, C. M., and Borges, L. L. (2022). "Larvicidal effect of the *Citrus limettioides* peel essential oil on *Aedes aegypti*," *Suid-Afrikaanse Tydskrif Vir Plantkunde [South African Journal of Botany]* 144, 257-260. DOI: 10.1016/j.sajb.2021.09.013

Cruz-Sánchez, E., García-Vargas, J. M., Gracia, I., Rodríguez, J. F., and García, M. T. (2022). "Pilot-plant-scale extraction of antioxidant compounds from lavender: Experimental data and methodology for an economic assessment," *Processes* 10(12), 2708.

da Costa, J. S., Maranduba, H. L., de Sousa Castro, S., de Almeida Neto, J. A., and Rodrigues, L. B. (2022). "Environmental performance of orange citrus waste as raw material for pectin and essential oil production," *Food and Bioproducts Processing*, 135, 165-177. DOI: 10.1016/j.fbp.2022.07.008

Ghasemi, J., Moradi, M., Karparvarfard, S. H., Golmakani, M. T., and Mousavi Khaneghah, A. (2021). "Thin layer drying kinetics of lemon verbena leaves: A quality assessment and mathematical modeling," *Quality Assurance and Safety of Crops & Foods* 13(1), 59-72. DOI: 10.15586/qas.v13i1.835

Jaramillo-Colorado, B. E., Vega-Díaz, R., and Pino-Benítez, C. N. (2022). "Volatile chemical composition of Colombian *Plectranthus amboinicus* (Lour.) Spreng essential oil and its biocidal action against *Tribolium castaneum* (Herbst)," *Revista Colombiana de Ciencias Hortícolas* 16(1), e13413. DOI: 10.17584/rcch.2022v16i1.13413

Justiniano-Medina, A., Arrieta-Conde, J., Torres-Taipe, A. L., Cruz-Anchiraico, J. A., and Huamanchahua, D. (2022). "Design of a steam distillation of essential oils from grapefruit peel with Raspberry Pi B+ using Telegram Connection," in: *IEEE 13th*

Annual Ubiquitous Computing, Electronics & Mobile Communication Conference (UEMCON), pp. 0132-0137.

Kant, R., and Kumar, A. (2022). "Review on essential oil extraction from aromatic and medicinal plants: Techniques, performance and economic analysis," *Sustainable Chemistry and Pharmacy* 30, article 100829. DOI: 10.1016/j.scp.2022.100829

Karanicola, P., Patsalou, M., Stergiou, P.-Y., Kavallieratou, A., Evripidou, N., Christou, P., Panagiotou, G., Damianou, C., Papamichael, E. M., and Koutinas, M. (2021). "Ultrasound-assisted dilute acid hydrolysis for production of essential oils, pectin and bacterial cellulose via a citrus processing waste biorefinery," *Bioresource Technology* 342, article 126010. DOI: 10.1016/j.biortech.2021.126010

Khedhri, S., Polito, F., Caputo, L., Manna, F., Khammassi, M., Hamrouni, L., Amri, I., Nazzaro, F., De Feo, V., and Fratianni, F. (2022). "Chemical composition, phytotoxic and antibiofilm activity of seven Eucalyptus species from Tunisia," *Molecules (Basel, Switzerland)* 27(23), 8227. DOI: 10.3390/molecules27238227

León, C., Jordán, E. P., Salazar, K., Castellanos, E. X., and Salazar, F. W. (2020). "Extraction system for the industrial use of essential oil of the subtle lemon (*Citrus aurantifolia*)," *Journal of Physics. Conference Series* 1432(1), article 012044. DOI: 10.1088/1742-6596/1432/1/012044

Li, Y., Liu, S., Zhao, C., Zhang, Z., Nie, D., Tang, W., and Li, Y. (2022). "The chemical composition and antibacterial and antioxidant activities of five citrus essential oils," *Molecules (Basel, Switzerland)* 27(20), article 7044. DOI: 10.3390/molecules27207044

Maes, C., Bouquillon S., and Fauconnier, M. (2019). "Encapsulation of essential oil for the development of biosourced pesticides with controlled release: A review," *Molecules (Basel, Switzerland)* 24(14), article 2539. DOI: 10.3390/molecules24142539

Manyako, K. E., Chiyanzu, I., Mulopo, J., and Abdulsalam, J. (2022). "Pilot-scale evaluation of concentrating solar thermal technology for essential oil extraction and comparison with conventional heating sources for use in Agro-based industrial applications," *ACS Omega* 7(24), 20477-20485. DOI: 10.1021/acsomega.1c06879

Marongiu, B., Piras, A., Porcedda, S., and Tuveri, E. (2006). "Comparative analysis of the oil and supercritical CO2 extract of *Cymbopogon citratus* Stapf," *Natural Product Research* 20(05), 455-459. DOI: 10.1080/14786410500277837

Moemenbellah-Fard, M. D., Shahriari-Namadi, M., Kelidari, H. R., Nejad, Z. B., Ghasemi, H., and Osanloo, M. (2020). "Chemical composition and repellent activity of nine medicinal essential oils against *Anopheles stephensi*, the main malaria vector," *International Journal of Tropical Insect Science* 41(2), 1325-1332. DOI: 10.1007/s42690-020-00325-2

Rodríguez, M., Alcaraz, L., and Real, S. (2012). "Procedimientos para la extracción de aceites esenciales en plantas aromáticas," Proyecto SAGARPA-CONACYT 126183, Available at https://cibnor.repositorioinstitucional.mx/jspui/bitstream/1001/540/1/rodriguez_m.pdf

Suresh, A., Velusamy, S., Ayyasamy, S., and Rathinasamy, M. (2021). "Techniques for essential oil extraction from kaffir lime and its application in health care products—A review," *Flavour and Fragrance Journal* 36(1), 5-21. DOI: 10.1002/ffj.3626

"The observatory of economic complexity," (https://oec.world/en/profile/hs/essential-oils-of-orange), Accessed on 2 February 2023.

Torrenegra Alarcón, M., Granados Conde, C., and León Mendez, G. (2019). "Extracción, caracterización y actividad antioxidante del aceite esencial de *Eucalyptus globulus* Labill," *Revista Cubana de Farmacia* 52(1).

Trujillo-Juárez, L. G., Hernández-Meléndez, Ó., Gimeno, M., Gracia-Fadrique, J., and Bárzana, E. (2021). "Extraction of essential oil from waste grapefruit peel using a pilot-scale twin-screw extruder," *ACS Food Science & Technology* 1(7), 1198-1205. DOI: 10.1021/acsfoodscitech.1c00098

Uysal, B., Sozmen, F., Aktas, O., Oksal, B. S., and Kose, E. O. (2011). "Essential oil composition and antibacterial activity of the grapefruit (*Citrus paradisi* L.) peel essential oils obtained by solvent-free microwave extraction: Comparison with hydrodistillation: Essential oil composition and antibacterial activity of the grapefruit," *International Journal of Food Science & Technology* 46(7), 1455-1461. DOI: 10.1111/j.1365-2621.2011.02640.x

Velasco, D., Senit, J., Santos, T., Yustos, P., Santos, V., Ladero, M., and Torre, I. D. la. (2017). "Optimization of the enzymatic saccharification process of milled orange wastes," *Fermentation* 3(3), 37. DOI: 10.3390/fermentation3030037

Wei, L., Yu, X., Li, H., Zhu, M., Pu, D., Lu, Q., Bao, Y., and Zu, Y. (2023). "Optimization of solvent-free microwave extraction of essential oil from the fresh peel of *Citrus medica* L. var. *arcodactylis* Swingle by response surface methodology, chemical composition and activity characterization," *Scientia Horticulturae* 309, article 111663. DOI: 10.1016/j.scienta.2022.111663

Weng, J., Wei, M.-M., Wu, S.-J., Liu, Y.-Q., Li, S.-R., Ye, Y.-Y., Wang, M., and Wang, D. (2019). "High-value utilization of citrus peel: Efficient extraction of essential oil and preparation of activated carbon," *BioResources* 14(2), 3899-3913. DOI: 10.15376/biores.14.2.3899-3913

Yadav, S. K. (2022). "Physiochemical properties of essential oils and applications," in: *Essential Oils - Advances in Extractions and Biological Applications*, 1st Ed., D. M. Santana and A. E. H. De Aguilar (eds.), Intechopen, London, UK, pp. 1-18 (2022).

Yumnam, M., Marak, P. R., Gupta, A. K., Rather, M. A., and Mishra, P. (2023). "Effect of pomelo peel essential oil on the storage stability of a few selected varieties of freshwater fish," *Journal of Agriculture and Food Research* 11, article 100472. DOI: 10.1016/j.jafr.2022.100472

Article submitted: April 26, 2023; Peer review completed: May 13, 2023; Revised version received: May 17, 2023; Accepted: May 20, 2023; Published: May 26, 2023.
DOI: 10.15376/biores.18.3.4977-4993