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Effect of Supercritical Fluid Extraction Process on Chemical Composition of *Polianthes tuberosa* Flower Extracts

Javier C. Fragoso-Jiménez [1], Ernesto Tapia-Campos [1], Mirna Estarron-Espinosa [1], Rodrigo Barba-Gonzalez [1], Ma. Claudia Castañeda-Saucedo [2] and Gustavo A. Castillo-Herrera [1,*]

[1] Centro de Investigación y Asistencia en Tecnología y Diseño del Estado de Jalisco A.C., Guadalajara C.P. 44270, Mexico; jafragoso_al@ciatej.edu.mx (J.C.F.-J.); etapia@ciatej.mx (E.T.-C.); mestarron@ciatej.mx (M.E.-E.); rbarba@ciatej.mx (R.B.-G.)

[2] Centro Universitario del Sur (CUSUR), Universidad de Guadalajara, Ciudad Guzmán C.P. 49000, Mexico; claudia.saucedo@cusur.udg.mx

* Correspondence: gcastillo@ciatej.mx; Tel.: + 52(33)-3345-5200 (ext. 1950)

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Abstract: Supercritical fluid extracts from flowers of *Polianthes tuberosa* var. *double* were ob tained using carbon dioxide as a solvent. Yield extract obtained was 2.5%. The effects of the pressure process (18 MPa, 28 MPa, and 38 MPa) and temperature process (313 K, 323 K, and 333 K) on the volatile composition of tuberose flowers extracts were evaluated, and a significant variation in chemical composition was found. Characteristic compounds of tuberose as methyl isoeugenol, benzyl benzoate, methyl anthranilate, pentacosene, and heptacosene were obtained mainly at 18 MPa and 333 K process conditions, and could be used in the perfume or fragrance industry. Components such as geraniol, farnesol, and methyl eugenol were also obtained, these extracts could be used in the development of cosmeceutical products. This work allowed to identification of the chemical composition profile and evaluation of the changes in tuberose extracts due to the extraction process.

Keywords: *Polianthes tuberosa*; supercritical fluid extraction; chemical composition

1. Introduction

Tuberose (*Polianthes tuberosa* var. *double*) is a plant endemic to México and belonging to the Asparagaceae family. Tuberose is used globally as an ornamental flower because of its size and sweet fragrance [1]. Due to its odor, extracts from tuberose flowers have high value in the perfume industry [2]. Furthermore, there are reports mentioning that extracts from tuberose flowers contain bioactive compounds which show antimicrobial or insecticidal activity [3,4], giving a special interest value to tuberose extracts.

Conventional extraction methods have been used to obtain tuberose extracts; these methods are mainly focused on the recovery of essential oil, achieving low yields of extracts and also degrading compounds because of the high temperatures used during distillation or solvent extraction [2,5,6]. Although there is another extraction method used to obtain volatile compounds from flowers, known as enfleurage, heating is also required, and compounds are also degraded during the extraction process, besides which a distillation is needed for recovery of compounds from the fat used as a solvent, which requires long extraction times.

A technology used to obtain volatile compounds from flowers alongside traditional methods is supercritical fluid extraction (SFE). In particular, the use of carbon dioxide for supercritical extraction has been shown to be an effective method for obtaining volatile compounds from flowers, because

carbon dioxide has non-polar behavior and low critical parameters, which makes it suitable for volatile compounds. Additionally, the use of supercritical carbon dioxide could obtain different compounds, modifying solvation capability as a function of extraction process conditions. There are another green technologies, such as ultrasound-assisted extraction which requires the use of solvents, and is considered an optimization of soxhlet or maceration extraction. Otherwise, ionic liquid-based extraction requires a lot of water consumption and ionic salts, besides which this technology has shown a low recovery of metabolites [7,8]. SFE has been used to obtain fatty acids, such as linoleic, oleic, and palmitic acid, that could also be used in the cosmetic industry, like mango kernel butter and cacao butter [9–11]. During extraction, volatile compounds are also obtained, and extracts could be used in the perfume or cosmetic industries [12,13]. SFE is considered a free solvent and an efficient extraction method for obtaining extracts from sensible materials such as flowers [14–16].

Moreover, the quality of supercritical extracts has been compared to enfleurage [2], and because of its benefits, SFE also has been used for recovery of volatile compounds from different flowers, such as lavender, jasmine, and geranium [17–19]. SFE also has been used for extraction of tuberose flowers; studies report low yield extracts, and few chemical compounds have been identified in extracts [2,4,20].

In addition, none of these studies studied the process conditions of particle size, humidity content, and carbon dioxide flow rate, which all affect the yield of extracts. Neither have any reported optimization of pressure and temperature processes, even when these factors affect chemical composition.

Therefore, the aim of this work was to study how the main parameters in the supercritical fluid process affect the chemical composition profile of *Polianthes tuberosa* flower extracts, and how these process parameters act, suggesting different potential applications of the extraction because of the different compositions obtained at different process conditions.

2. Materials and Methods

2.1. Tuberose Cultivation

Bulbs of this cultivar were acquired from tuberose producers of the Cuachichinola region in Morelos, México, and then were established in a greenhouse in a Research Centre for Technology and Assistance in Design of Jalisco State (CIATEJ) at 20° 42′ 03.9″ N, 103° 28′ 24.5″ W coordinates, handling the culture to generate flowers. Once blossomed during August–September, flowers were harvested and vacuum packed, and stored at 193 K until the dehydration process.

Before supercritical extraction, flowers were dehydrated in a convection dryer (San-Son, Edo. de México, México) at 298 K until they reached 10% ± 1 moisture content. Moisture was measured in duplicate in a AND MF-50 humidity analyzer (A&D Technologies, Wood Dale IL, US), and dried flowers were ground in an IKA MF 10.1 Cutting-grinding head mill (IKA, Wilmington, NC, US) and sieved in a Rot-Tap RX-29 (Ohio, US), using for extraction milled flowers that passed through 40 mesh.

2.2. Supercritical Fluid Extraction

A Thar Technologies-Waters®-SFE-500 supercritical fluid extractor (Thar Process, Pittsburgh, PA, US) was used. Tuberose flowers weighing 100 g were placed into the extraction vessel, and the effects of three pressure levels, 18, 28, and 38 megapascals (MPa) and three temperature levels, 313, 323, and 333 K were evaluated for extraction yield and chemical composition of the extract. A constant flow of 10 g/min CO_2 (medical grade, INFRA^{TM}, Guadalajara, México) was established with bottom extraction vessel feeding, total extraction time was 3 h [4,12,20]. Extracts obtained in collector vessel were kept in amber flasks and stored at 253 K until gas chromatography analysis, and a diagram of the process is displayed in Figure 1.

A mixed factorial 3^2 design was used, allowing evaluation of linear effects and interactions between the pressure and temperature processes, and analysis of effects and quadratic curvature

factors. Experiments were done in duplicate and statistical analysis was performed using Statgraphics Centurion XVI software Version, 16, Warrenton, V., US.

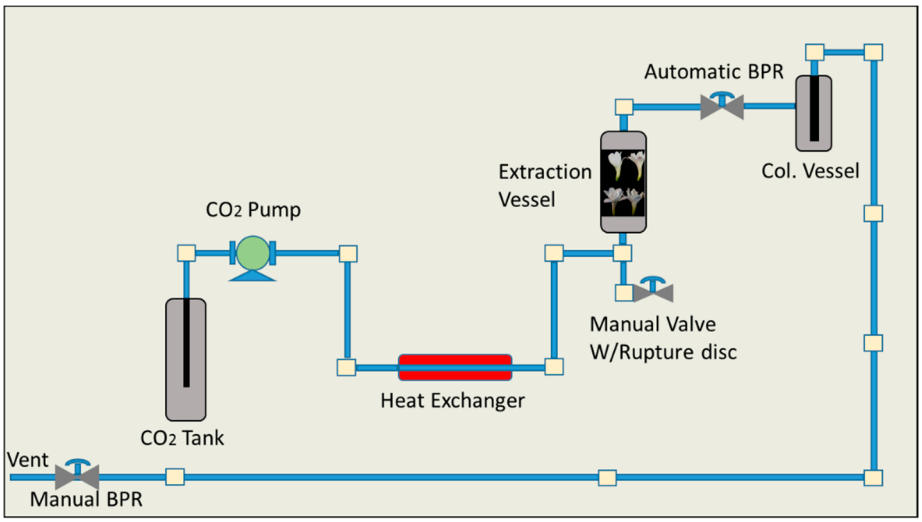

Figure 1. Diagram of the extraction process by supercritical CO_2.

2.3. Gas Chromatography–Mass Spectrometry Analysis

Analysis of extracts was carried out in a gas chromatograph Hewlett Packard 5890 Series II (Palo Alto, CA, US) coupled to a Hewlett Packard 5972A mass selective detector (Palo Alto, CA, US). Separation was carried out in an INNOWAX capillary column of 60 m × 0.25 mm × 0.25 µm (J & W Scientific, Folsom, CA, USA). Injector and detector temperatures were 503 K and 523 K, respectively. Injection volume of diluted extract in petroleum ether (1:1 mg/mL) was 1 µL (split ratio 1:50), and the initial oven temperature was 323 K, increasing to 513 K at a rate of 5 K/min and held for 60 min; carrier gas was helium at a flow rate of 0.7 mL/min. The mass spectrometer quadrupole type was operated with an ionization voltage of 70 eV (EI). Mass spectra and reconstructed ion currents (TIC) were obtained by automatic scanning from m/z 20–450 at 0.81 scan/s. Chemical compounds were identified by comparing mass spectra with those provided by the NIST05.L database, as well as from retention times of some standard compounds (Sigma-Aldrich, >95%, State of Mexico, Mexico) injected under identical analytical conditions, and verification of elution order by Kovats Indices from literature [21]. The quantification was based on the relative percentage area from the automatic integration of detected peaks.

3. Results and Discussion

3.1. Supercritical Fluid Extraction

Tuberose flowers yield extracts obtained at different pressure and temperature processes are shown in Table 1, and were different and higher than yields reported by Gosh et al. [4], who reported a supercritical yield extraction of 0.5%. Yield extracts were even higher than those reported by Rakthaworn et al. [2], who evaluated different extraction methods and reported yields of tuberose flowers extracts obtained by cold palm wax enfleurage, hot palm oil enfleurage, and solvent extraction with hexane and petroleum ether, obtaining yields of 0.3137%, 6.5808%, 0.0279%, and 0.0182% respectively. However, in the 6.58% yield, Rakthaworn et al. [2] made an accumulative extraction increasing tuberose flower quantity, so it is not comparable to the yield extracts reported in this work.

The higher yields obtained in this work could be due to different factors such as a higher pressure process, which affects the solubility of non-polar compounds. We ensured careful post-harvest handling and drying conditions of the tuberose flowers to avoid compound degradation, also the particle size used for extraction was controlled to improve the mass transfer. These factors were

reported as important factors for jasmine and lavender extraction [19,22]. However, these factors were not evaluated in this work, because the aim of the research was to study how the main parameters that affect carbon dioxide solubility in the supercritical fluid process impacts the chemical composition profile of *Polianthes tuberosa* extracts. It is known that factors as time, particle size, and carbon dioxide flow are factors that affect recovery efficiency or yield extraction, but not the solubility carbon dioxide.

Table 1. Extract yield of *P. tuberosa* flowers and experimental design.

Experiment	Pressure (MPa)	Temperature (K)	Extract Yield (%)
1	18	333.15	0.73
2	28	323.15	2.43
3	18	313.15	1.11
4	28	323.15	2.28
5	38	313.15	1.49
6	38	333.15	1.75
7	18	333.15	1.11
8	28	323.15	2.52
9	18	313.15	1.06
10	28	323.15	2.54
11	38	313.15	2.42
12	38	333.15	2

In supercritical extract yield of tuberose flowers, the pressure process was statistically significant (p-value < 0.05), meanwhile, extract yield was not affected by the temperature process. Higher tuberose extract yield was obtained at 28 MPa and 323 K, and curvature effect of the pressure process was observed (Figure 2). Extraction yield can be described by the following model, which demonstrates the effect of the curvature of design with an adjusted R^2 of 88.07%:

$$\% \text{ Extraction Yield} = -2.64556 + 0.527856 \cdot P - 0.012075 \cdot T - 0.0098375 \cdot P^2 + 0.0002125 \cdot P \cdot T \quad (1)$$

where P = Pressure (MPa), T = Temperature (K).

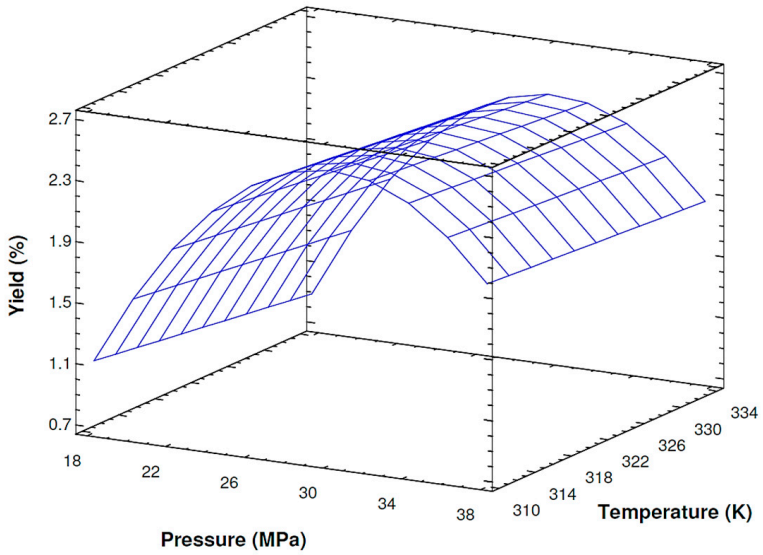

Figure 2. Response surface for extraction process over extraction yield.

Tuberose flower extracts present similar behavior to that observed by Liu et al. [23] in *Opuntia dillenii* seeds, and by Yu et al. [24] in grapefruit seeds, both using SFE. Additionally, in both studies temperature had no significant influence over extraction yield. Similar behavior was also reported by Aladić et al. [25] in supercritical extraction of *Cannabis sativa* L. seeds.

Finally, an efficient recovery of volatile compounds from tuberose flowers was effected by means of carbon dioxide in supercritical conditions with 3 h extraction time.

3.2. Chemical Composition in Tuberose Flowers

Analysis of tuberose flowers extracts allowed identification of 37 compounds, mainly volatile compounds as terpenes, hydrocarbons, alcohols, and esters, as shown in Table 2. The characteristic aroma of tuberose flowers is given by compounds as methyl isoeugenol, methyl anthranilate, pentacosane, benzyl benzoate, and pentacosane which are shown in Figure 3 [26].

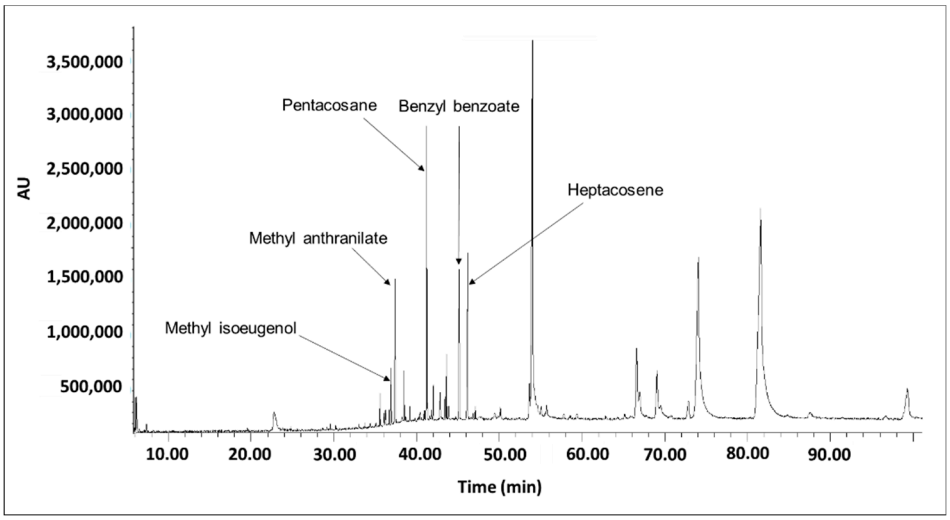

Figure 3. Chromatogram showing characteristic compounds for supercritical tuberose extract.

Furthermore, chemical composition was affected by the pressure and temperature processes during supercritical extraction. The main volatile compounds from tuberose flowers were found predominantly at lower pressure extraction (18 MPa), while common compounds in supercritical plant extracts, as terpenes, alcohols, and some fatty acids were found in almost all extracts at different extraction pressures. Similar behavior was reported by Ghosh et al. [4] and Gomes et al. [18].

Characteristic compounds of tuberose flowers that were affected by extraction process are methyl isoeugenol, pentacosane, methyl antralinate, benzyl benzoate, and heptacosene, and the effect of process parameters can be observed in Figure 4, giving these extracts a greater potential use in the perfume industry. Meanwhile, compounds as methyl eugenol, geraniol, 9,12,15-octadecatrienonic acid, and farnesol were not affected by the extraction process, allowing them to be used in cosmeceutical products, mainly due to the bioactivities these compounds have been reported to possess.

Analysis of tuberose flower extracts resulted in four similar compounds reported by Ghosh et al. [4] in supercritical extracts, eight similar compounds were identified by Ahmadian et al. [6], three similar compounds were reported by Bin et al. [12], and seven compounds were reported by Reverchon [13], who only studied tuberose essential oils. Additionally, it was also possible to identify compounds such as benzyl alcohol and benzoic acid in tuberose flower extracts.

Due to the diversity found in the chemical composition of supercritical fluid extracts, it is important to analyze the potential of extracts depending on process conditions. Extracts obtained at 18 MPa contain methyl eugenol, isoeugenol, and benzyl benzoate, which could be used as antimicrobial agents, according to Gosh et al. [4] who evaluated the antimicrobial activity of tuberose extracts against *Staphylococcus aureus*, *Pseudomonas aeruginosa*, *Escherichia coli*, *Helicobacter pylori*, *Vibrio cholera*, and *Candida albicans*. Otherwise, Anu et al. [27] also reported antimicrobial activity of tuberose essential oil against *Kleibsiella pneumoniae*, *P. aeruginosa*, *Proteus mirabilis*, *Escherichia coli*, *Staphylococcus aureus*, and *Bacillus subtilis*.

Table 2. The chemical composition of *P. tuberosa* flower extracts at different extraction conditions.

	Process Conditions		38 MPa/333.15 K		38 MPa/313.15 K		28 MPa/323.15 K		28 MPa/323.15 K		18 MPa/333.15 K		18 MPa/313.15 K		
	Experiment [a]		6	12	5	11	2	8	4	10	1	7	3	9	
Compound	**KIR [g]**	**R.T. (min) [b]**	**% AREA**												**ID**
Geraniol	1781	28.7	0.08	0.07	0.07	0.09	0.07	0.05	0.04	0.04	0.06	0.07	0.07	0.07	MS, STD, KI
Benzyl alcohol	1889	29.6	0.11	0.12	0.09	0.09	0.11	0.09	0.10	0.09	0.14	0.17	0.13	0.12	MS, STD, KI
Methyl eugenol [c,d]	2028	32.3	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.02	0.02	0.02	MS, STD, KI
Caprylic acid	2039	33.1	-	-	0.07	0.07	0.07	0.05	0.05	0.05	0.06	0.08	0.07	0.06	MS, STD, KI
Heneicosane	2100	33.7	0.09	0.10	0.05	0.07	0.08	0.08	0.08	0.07	0.09	0.08	0.09	0.09	MS, STD, KI
Methyl isoeugenol [d,f]	2185	35.6	0.36	0.39	0.36	0.33	0.47	0.35	0.35	0.31	0.48	0.54	0.52	0.54	MS, STD, KI
Methyl palmitate	2218	36.1	0.22	0.24	0.18	0.20	0.22	0.18	0.18	0.15	0.23	0.23	0.23	0.19	MS, STD, KI
δ-Decalactone [f]	2193	36.2	0.26	0.22	0.32	0.30	0.34	0.26	0.26	0.23	0.36	0.40	0.36	0.37	MS, KI
Ethyl palmitate	2250	36.7	2.56	2.70	2.98	3.54	2.66	2.74	2.15	1.96	2.21	2.37	4.10	4.06	MS, STD, KI
Methyl anthranilate [d,f]	2232	36.9	0.49	0.53	0.33	0.31	0.57	0.49	0.42	0.41	0.56	0.62	0.52	0.45	MS, KI
Tricosane	2300	37.4	1.56	1.14	2.32	2.21	2.20	2.04	1.78	1.66	2.40	2.73	2.15	1.65	MS, KI
Farnesol [d,f]	2356	38.5	0.90	0.94	1.11	1.09	1.05	0.86	0.79	0.71	1.04	1.21	1.12	0.88	MS, STD, KI
Isoeugenol [d,f]	2352	38.7	0.06	0.06	0.06	0.05	0.09	0.07	0.07	0.07	0.08	0.09	0.07	0.07	MS, STD, KI
Tetracosane	2400	39.0	0.41	0.38	0.37	0.34	0.34	0.37	0.38	0.34	0.42	0.34	0.40	0.38	MS, STD, KI
Methyl stereate	2422	39.8	0.06	0.06	-	-	-	-	-	-	0.06	0.06	0.07	0.05	MS, STD, KI
Ethyl stereate	2464	40.4	0.16	0.17	0.23	0.21	0.19	0.21	0.09	0.09	0.11	0.11	0.25	0.22	MS, STD, KI
Benzoic acid [c,e]	2448	40.4	0.36	0.36	0.40	0.37	0.40	0.35	0.33	0.32	0.38	0.33	0.27	0.28	MS, KI
Ethyl oleate	2493	40.9	0.13	0.14	0.16	0.15	0.15	0.16	0.11	0.11	0.14	0.15	0.19	0.18	MS, STD, KI
Pentacosane [c,d]	2500	41.2	6.54	7.19	6.52	6.15	7.05	7.63	7.76	6.97	9.93	7.52	10.97	11.06	MS, STD, KI
Methyl linoleate [f]	2509	41.4	0.13	0.14	0.13	0.13	0.12	0.12	0.04	0.04	0.32	0.27	0.06	0.06	MS, STD, KI
Ethyl linoleate	2536	42.0	0.26	0.24	0.38	0.36	0.31	0.35	0.32	0.28	0.36	0.35	0.48	0.44	MS, STD, KI
Methyl linolenate	2583	42.8	0.21	0.21	0.21	0.19	0.18	0.20	0.19	0.19	0.25	0.23	0.28	0.28	MS, STD, KI
Hexacosane	2600	43.4	0.62	0.57	0.41	0.47	0.49	0.55	0.60	0.55	0.63	0.50	0.72	0.78	MS, STD, KI
Ethyl linolenate	2613	43.6	0.53	0.52	0.77	0.75	0.58	0.65	0.66	0.59	0.80	0.78	1.06	1.03	MS, STD, KI
Benzyl benzoate [c,d,e,f]	2655	45.1	6.19	6.71	5.53	5.41	6.33	5.40	5.17	4.76	6.67	7.39	7.17	7.89	MS, STD, KI
Heptacosene	2688	46.1	5.09	5.44	5.47	5.03	5.87	6.58	6.52	5.85	7.68	6.01	8.29	7.26	MS, STD, KI
Heptacosane	2700	46.9	0.15	0.11	-	-	0.11	0.11	0.14	0.13	0.14	0.13	0.16	0.14	MS, STD, KI
Octacosene	2794	47.6	0.36	0.27	0.34	0.31	0.27	0.28	0.27	0.27	0.33	0.28	0.36	0.39	MS, STD, KI
Octacosane	2800	49.2	0.29	0.24	0.25	0.23	0.25	0.28	0.31	0.45	0.38	0.31	0.38	0.41	MS, STD, KI
Benzyl salicylate [f]	2810	50.2	1.14	1.24	1.12	1.03	1.09	0.97	0.95	0.90	1.21	1.33	1.34	1.44	MS, STD, KI
Nonacosane	2900	53.5	1.76	1.83	0.42	0.69	2.02	2.22	2.51	2.19	2.52	1.99	2.69	2.64	MS, STD, KI
Palmitic acid	2930	53.9	12.85	13.44	16.10	14.87	12.38	13.83	12.94	12.67	13.24	11.91	12.79	13.93	MS, STD, KI
Stearic acid	3090	66.6	4.08	4.29	4.91	4.62	3.59	4.12	4.67	4.60	4.79	3.57	3.82	4.00	MS, STD, KI
Oleic acid	3157	69.0	2.00	2.13	2.46	2.25	1.90	2.15	2.48	2.38	2.56	1.99	2.19	2.34	MS, STD, KI
Linoleic acid [e]	3168	73.9	8.82	9.34	11.90	11.05	8.46	9.21	11.42	11.37	10.40	10.40	10.37	10.08	MS, STD, KI
9,12,15-Octadecatrienonic acid	3554	80.5	33.93	31.25	28.97	30.76	33.97	31.18	28.32	31.98	22.48	29.71	17.73	17.39	MS, KI
n-Hexatriacontano	3600	99.1	4.65	4.58	2.28	3.56	3.11	2.99	5.32	5.21	3.56	3.37	5.11	5.60	MS, STD, KI
Number of Identified Compounds			36	36	35	35	36	36	36	36	37	37	37	37	

[a] Experiment: Number of experiments in experimental design. [b] R.T.: Retention time. Compounds reported in *Polianthes tuberosa*: [c] Gosh et al. [4], [d] Reverchon et al. [20], [e] Bin et al. [14], [f] Ahmadian et al. [6]. [g] KIR: Kovats Index Reference based on NIST [21], MS: Mass spectrometry, STD: Standard compounds, ID: Identification.

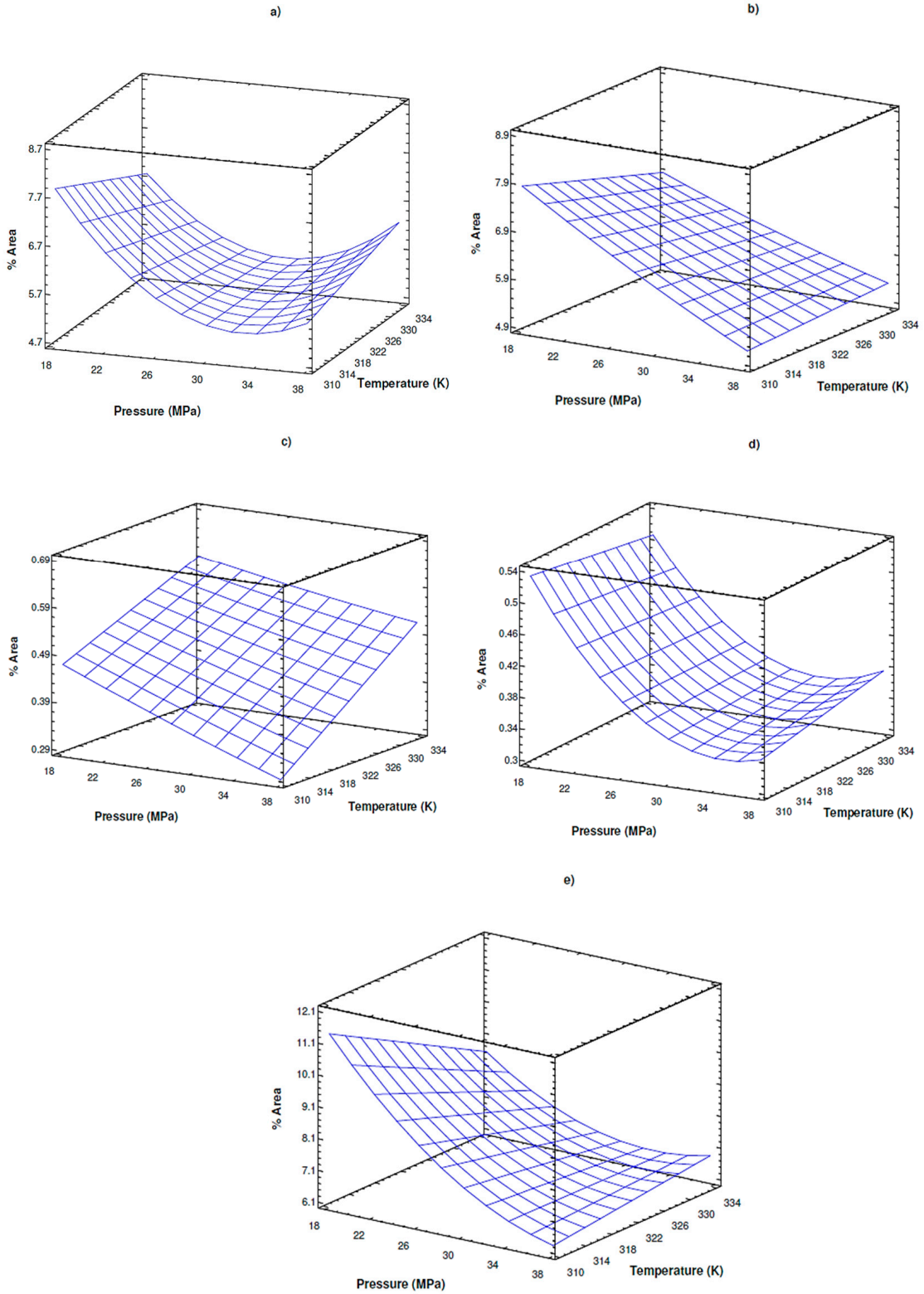

Figure 4. Effect of pressure and temperature on characteristic tuberose flower compounds: (**a**) benzyl benzoate, (**b**) heptacosene, (**c**) methyl anthranilate, (**d**) methyl isoeugenol, (**e**) pentacosane.

Extracts obtained at 18 MPa also contain compounds such as geraniol, methyl isoeugenol, farnesol, and δ-decalactone, compounds that are preferred in the fragrance industry, as mentioned for jasmine and rose geranium oil, which also contain these compounds [18,28]. Meanwhile, extracts with 9,12,15-Octadecatrienonic acid, obtained at 38 MPa, could be used in the cosmetic industry.

4. Conclusions

This study found that volatile compounds and extraction yield obtained from tuberose flowers in this research could have potential uses in perfume or cosmeceutical products. The chemical profile of extracts was dependent on the process conditions, mainly the pressure process. With the accomplishment of this research, it was possible to identify changes in the chemical composition profile of *Polianthes tuberosa* supercritical extracts, and also how to focus extraction process conditions to maximize compounds like methyl isoeugenol, methyl anthranilate, pentacosene, and heptacosene in tuberose flower extracts, which have high industry value.

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