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The effect of the drying process on the SFE optimization of bio-compounds of mango (*Mangifera indica* L.) and pineapple (*Ananas comosus*) peels

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Abstract The effect of vacuum and freeze drying on the optimization of the supercritical fluid extraction (SFE) process of mango and pineapple peels to obtain bioactive extracts was evaluated. The influence of several SFE parameters was investigated using a central composite design (CCD) with four central points, to estimate the effect of three independent variables (co-solvent flow rate, pressure and temperature) on the yield, antioxidant activity, and content of bioactive compounds. It was found that vacuum drying was more effective for mango peels (ethanol flow) and freeze-drying for pineapple peels (ethanol flow and temperature). Using the multiple response methodology by the desirability approach, the optimum SFE conditions for mango peel were found at a co-solvent flow rate of 13.65%, temperature of 36.76 °C, and pressure of 294.82 bar, whereas the optimal conditions for pineapple peel were found at a co-solvent flow rate of 12.39%, temperature of 43 °C, and pressure of 100 bar. According to these results, an adequate combination of the drying method and SFE in optimal conditions makes possible the integral use of these fruits to obtain valuable extracts with high potential to be used in various industries.

Keywords: Agro-industrial residues, Drying methods, Extraction optimization, Response surface methodology, Supercritical fluid extraction

Introduction

The worldwide growth of fruit consumption is due in part to increased agro-based industries that allow an increasing number of processed products to emerge, such as juices, jams, nectars and concentrates (Berardini *et al*., 2005; Upadhyay *et al*., 2010). This increase has, in turn, been generating a high quantity of tons of wastes and by-products that are mostly composed of stalks, peels, seeds and bagasse, becoming a serious environmental problem for disposal. Mango

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(*Mangifera indica* L.) and pineapple (*Ananas comosus*) are some of the most popular fruits throughout the world, with a global production of 50.64 and 29.93 million metric tons respectively (FAO, 2019). After processing these fruits, peels alone contribute to 7-24% and 29–40% of the mango and pineapple total weight, respectively (Jawad *et al*., 2013; Shakya and Agarwal, 2019). Several authors have reported that these by-products are an important source of nutrients and bioactive compounds; they could have potential uses as raw materials that can be converted into value-added products (Ferreira *et al*., 2019; Saisung and Theerakulkait, 2011).

Several solvents and extraction techniques have been studied to obtain bioactive compounds from different plant matrices; however, the conventional methods show several limitations, including long extraction time and high temperatures that accelerate the degradation of the products of interest and use of expensive, and toxic organic solvents, which are deleterious both for human health and the environment (Carvalho *et al*., 2005; Ameer *et al*., 2017). Emerging and more efficient techniques have been recommended to improve the extraction of these compounds. Supercritical fluid extraction (SFE) is a technique that allows a much faster isolation of bioactive compounds than the conventional extraction methods. It has been widely used to add value to plant and food byproducts generated during processing. Moreover, it has been reported that SFE extracts have larger antioxidant activity than the obtained by extraction with organic solvent (Herrero *et al*., 2010). Optimizing the SFE process is important for obtaining the target compounds. The use of the optimum values of pressure, temperature, time, type and percentage of modifiers, solvent flow rate, etc., could significantly enhance the process performance and the quality of the extract (Carvalho *et al*., 2005; Herrero *et al*., 2010), likewise, a very important factor that should be taken into account is the drying method used in the raw material. During water evaporation, physical, chemical, structural and biological properties are altered, affecting not only the quality of dry products but also the physicochemical properties and efficacy of extracts (Li *et al*., 2017; Li *et al*., 2019).

Freeze drying (FD) or lyophilization is a well-known method based on the dehydration by sublimation of a frozen product. This method allows obtaining high-quality products, by not exposing the product to high temperatures and the virtual absence of oxygen in the air during the process. The main disadvantage of FD is its high operating cost and the relatively long time required to obtain the desired final moisture content (Karam *et al*., 2016; Chen *et al*., 2017). In addition, some studies have shown adverse effects on compounds of nutritional value and antioxidant activity by the FD method (Tetteh *et al*., 2019). Vacuum drying (VD), on the other hand, is an important dehydration method, suitable for heat

sensitive fruits and vegetables. In this method, the temperature can be controlled at relatively low levels, the heat is provided by conduction or radiation, and the oxidation of food compounds can be reduced by the removal of oxygen during drying. However, it is still costly for consideration at large production scales (Aghbashlo *et al*., 2013; Argyropoulos and Müller, 2014).

Several studies have shown that the different drying methods and conditions would affect the physicochemical properties and bioactive activities in a sample (Dorta *et al*., 2012; Routray *et al*., 2014; Li *et al*., 2017). Evaluation of the influence of the drying method on the extraction process can reveal significant information when the final product can have applicability as a functional or nutraceutical food ingredient. The aim of the present study was to investigate the effects of the selected drying methods (FD and VD) on the SFE process and optimize the SFE-CO_2 process of mango and pineapple peels. In order to accomplish those goals, the effects of three different conditions (temperature, pressure and percentage of ethanol as modifier) on yield of SFE process, antioxidant activity and the metabolites extraction such as gallic acid, ellagic acid, quercetin and mangiferin were investigated. Operational parameter ranges were determined according to the available literature data.

Materials and methods

Raw material

Mango (*Mangifera indica* L. var. Tommy Atkins) and pineapple (*Ananas comosus*) peels were obtained from fruits purchased at a local market in Medellín, Colombia, during the summer. The peels were separated from the pulp and allowed to mature, at room temperature (25 °C) until reaching 12-14 °Brix (complete maturity or consumption phase) for optimal extraction.

Drying treatments

Two drying methods were evaluated: VD at 40 °C and 10 mbar using a Memmert vacuum dryer (Germany) and FD where samples were frozen at −40 °C with a controlled heating rate of 0.05 °C/min and a heating plate at 30 °C (Labconco, USA). Both drying processes aimed to achieve a final moisture content of ≤7% (w/w). Dried samples were ground cryogenically (Ika MF 10 B S1) with liquid N_2 and sieved to ensure uniform particle sizes (80-270 mesh).

Supercritical fluid extraction (SFE)

The dried samples (3 g) were subjected to supercritical fluid extraction using an MV-10 ASFE System (Waters Corporation, USA) controlled by Chromo Scope™ software. Samples were extracted at varying temperatures (30, 40, 50 °C), pressures (100, 200, 300 bar), and ethanol co-solvent concentrations (5, 10, 15%). The $ScCO_2$ flow rate was maintained at 13.33 ml/min, with a dynamic extraction time of 75 min. Extracts were stored at 4 °C for further analysis.

Determination of global yield (X_0)

Extracts were evaporated at 40 °C to remove residual ethanol. The global yield (X_0) was calculated using the formula:

$$X_0 = \left(\frac{M_{extract}}{M_{sample}}\right) \times 100$$

Where $M_{extract}$ is the mass of the extract, and M_{sample} is the dry mass of the raw material.

Analytical determinations

All analyses were performed in triplicate for each drying and extract condition.

Moisture Content: Determined using AOAC (2000) method 934.01 at 105 °C until a constant weight was achieved.

Scanning Electron Microscopy (SEM): The morphological characteristics of peels were examined using SEM (JEOL-JSM 6490LV, USA) after gold coating. Images were taken at 200X and 1000X magnification.

DPPH and ABTS Methods: The antioxidant activities were assessed using the DPPH and ABTS assays, with results expressed as Trolox equivalent antioxidant capacity (TEAC) in µmol Trolox/g dry matter (DM).

Total Phenolic Content (TPC): Measured using the Folin-Ciocalteu method and expressed as mg gallic acid equivalent (GAE) per g DM.

Quantification by HPLC

Phenolic compounds—gallic acid (GA), ellagic acid (EA), mangiferin (M), and quercetin (Q)—were quantified using HPLC (Shimadzu Prominence UFLC System, Japan). The separation process was performed on a C-18 Zorbax Eclipse

Plus Agilent column (5 µm, 150 x 4.6 mm). The column oven temperature was maintained at 40 °C, with an injection volume of 10 µL and a flow rate of 1 ml/min following the gradient. The mobile phase consisted of a mixture of water with 1% acetic acid (A) and acetonitrile with 1% acetic acid (B). The optimized gradient program was as follows: 0 min, 0% B; 14 min, 90% B; 17 min, 0% B; 20 min, 0% B. Spectral measurements were taken in the range of 254–360 nm Results were based on standard solutions calibration curves (2 to 100 µg).

Experimental design and statistical analysis

A face-centered central composite design (CCD) with four central points was used to assess the effects of temperature (A), pressure (B), and co-solvent concentration (C) on the extraction process. Statistical analysis was performed using Statgraphics Centurion software, with a significance level set at $\alpha = 0.05$.

Optimization

The desirability function approach, implemented in Statgraphics Centurion, was used to optimize the extraction process by identifying the optimal combination of temperature, pressure, and co-solvent concentration for maximizing extraction yield and antioxidant activity.

Results

Effect of drying on yield, antioxidant capacity, and extraction of phytochemicals

The dried peel samples were used for the extraction of nutrients and antioxidants by supercritical fluid extraction (SFE). The experimental design for drying and optimization of the by-product extraction process is presented in Table 1. In general, better results were obtained for mango peel dried under vacuum compared to the lyophilized one. The opposite was observed in pineapple, where higher values were obtained in the lyophilized peel except in the yield. For a complete morphology study, the impact of the VD versus FD technique was analyzed using scanning electron micrographs (SEM).

Table 1. Experimental design for drying and optimization of SEF extraction of by-products using a central composite design (CCD) with four central points

Peel	Drying method	Symbol	Factors (Variables)	Ranges and levels		
				−1	0	1
Mango Pineapple	Vacuum Freeze -drying	A	Co-solvent (%)	5	10	15
		B	Temperature (°C)	30	40	50
		C	Pressure (bar)	100	200	300

Effect of drying methods on microstructure

The scanning electron microscopy (SEM) images of mango and pineapple peel (10–100 μm) dried by VD and FD are presented in Figure 1. The surface of mango peel treated with the VD method (MP-V) was observed to be rough, more compact in structure, and had numerous pores compared to the FD-treated sample (MP-F), which had a smoother surface. This structural difference allows the solvent to penetrate more easily, providing a greater surface for mass transfer and resulting in more efficient extraction of bioactive compounds. Pineapple peel treated with FD (PP-F) exhibited a distinct porous structure with few collapsed areas, while the vacuum-dried pineapple peel (PP-V) had a smooth surface and exhibited irregular particles.

Yield

After drying using VD and FD, the dried peels were subjected to cryogenic grinding, sieving, and extraction by the supercritical fluid extraction (SFE) method. The experimental results for SFE of MP-V (mango peel vacuum-dried), MP-F (mango peel freeze-dried), PP-V (pineapple peel vacuum-dried), and PP-F (pineapple peel freeze-dried) are summarized in Table 2. The yield (X_0) varied in the range of 8.54±1.27% to 38.83±0.77% for MP-V and 3.17±0.14% to 27.23±0.52% for MP-F. In both samples, the co-solvent flow had a significant effect ($p < 0.05$) (Tables 3 and 4), with X_0 being directly proportional to the co-solvent percentage. The maximum yield was reached at 100 bar, while the lowest was obtained at 200 bar. For pineapple, X_0 values ranged from 5.84±0.26% to 34.83±1.20% for PP-V, significantly affected by the co-solvent flow, whereas PP-F values were lower, ranging from 3.81±0.96% to 25.92±1.19%, influenced by both co-solvent flow and temperature ($p < 0.05$).

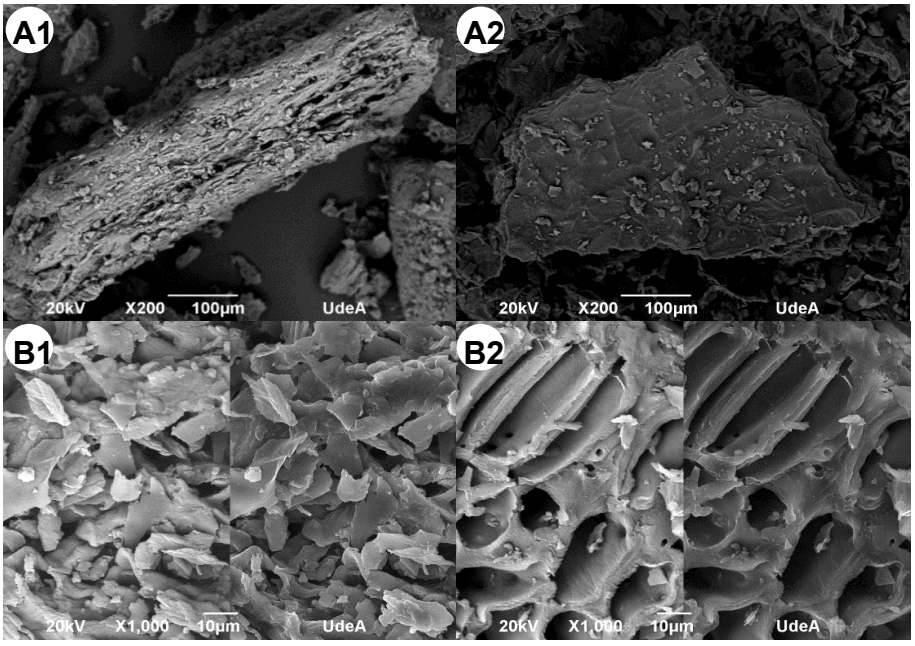

Figure 1. Scanning electron micrographs of peel dried. (A) Mango: A1: Vacuum drying; A2: Freeze drying (100µm, X200, 20kV). (B) Pineapple: B1: Vacuum drying; B2: Freeze drying (10µm, X1000, 20kV)

Antioxidant properties

ABTS Method. The antioxidant capacity of mango peel ranged from 64.43±2.65 to 981.51±13.57 TEAC, while pineapple peel exhibited lower values, ranging from 10.29±0.63 to 100.67±2.81 TEAC. The highest antioxidant capacity was observed in mango peel dried by vacuum (MP-V), followed by MP-F, PP-F, and PP-V. None of the variables had a significant effect on all samples analyzed ($p > 0.05$).

DPPH Method. The values obtained in terms of Trolox equivalent were lower than those from the ABTS assay, but the overall trend regarding the evaluated variables was consistent across both methods for the two samples. The peel extracts from the four treatments showed considerable differences in DPPH radical-scavenging activities at their maximum values. The highest value (1580.30±11.58 TEAC) was observed in MP-V samples, followed by MP-F with 400.67±21.39 TEAC, PP-F with 115.56±1.21 TEAC, and the lowest in PP-V with 55.0±1.21 TEAC. High values were observed under medium conditions for mango (10% EtOH, 40 °C, and 200 bar) and low conditions for pineapple (5% EtOH, 30 °C, and 100 bar). None of the parameters had a statistically significant effect in the tests performed ($p > 0.05$).

Table 2. Effect of different drying methods and extraction conditions on the evaluation parameters of mango and pineapple peel

	Yield (%)	DPPH (TEAC)	ABTS (TEAC)	Total phenolics (GAE)	Galic acid (μg/g)	Manguiferin (μg/g)	Ellagic ac. (μg/g)	Quercetin (μg/g)
	Vacuum dried mango peel – MP-V							
Min.	8.54 ± 1.27	48.80 ± 2.71	64.43 ± 2.65	5.95 ± 0.25	363.85 ± 1.22	509.30 ± 0.44	60.63 ± 0.12	501.94 ± 2.30
Max.	38.83 ± 0.77	1580.30 ± 11.58	981.51 ± 13.57	90.25 ± 0.48	1990.22 ± 0.83	83092.99 ± 0.27	6317.54 ± 3.56	637.75 ± 1.45
	Freeze dried mango peel – MP-F							
Min.	3.17 ± 0.14	42.85 ± 0.88	157.67 ± 6.33	17.67 ± 0.24	712.81 ± 0.36	1331.00 ± 0.21	73.04 ± 0.62	641.79 ± 0.27
Max.	27.23 ± 0.52	400.67 ± 21.39	474.97 ± 10.34	43.31 ± 0.67	1998.23 ± 0.52	90589.85 ± 3.76	3851.16 ± 0.34	2073.94 ± 0.92
	Vacuum dried pineapple peel – PP-V							
Min.	5.84 ± 0.26	18.91 ± 0,01	10.29 ± 0.63	4.89 ± 0.18	ND	ND	6.30 ± 0.24	ND
Max.	34.83 ± 1.20	55.00 ± 1.62	44.42 ± 1.99	9.22 ± 0.03	ND	ND	41.46 ± 0.38	ND
	Freeze dried pineapple peel – PP-F							
Min.	13.81 ± 0.96	63.55 ± 2.99	58.94 ± 2.07	12.11 ± 0.16	ND	ND	68.94 ± 0.07	ND
Max.	25.92 ± 1.19	115.56 ± 1.21	100.67 ± 2.81	18.46 ± 0.24	ND	ND	173.04 ± 1.19	ND

Total Phenolics. The total phenolic contents for MP-V and MP-F were 5.95±0.25 to 90.25±0.48 mg GAE/g DM and 17.67±0.24 to 43.31±0.67 mg GAE/g DM, respectively (Table 2). The co-solvent had a statistically significant effect on MP-F ($p < 0.05$). The best extraction conditions for mango peel were 15% EtOH, 30 °C, and 200 bar, while the lowest results were obtained at 10% EtOH, 50 °C, and 100 bar. For pineapple, freeze-dried peel retained more polyphenols than vacuum-dried peel, with values of 12.11±0.16 to 18.46±0.24 mg GAE/g DM and 4.89±0.24−9.22±0.03 mg GAE/g DM, respectively. The highest phenolic contents were obtained with 5% EtOH, 50 °C, and 100 bar, while the lowest results were obtained with 10% EtOH, 40 °C, and 200 bar.

HPLC analysis

The bioactive profile of flavonols, xanthones, and phenolic acids in fruit peels is shown in Table 2. Ellagic acid ranged from 60.63±0.12 to 6317.54±3.56 μg/g DM for mango peel and 6.30±0.24−173.04±1.19 μg/g DM for pineapple peel. The highest ellagic acid content was found in MP-V, while the lowest was in PP-V. Only the working pressure affected MP-F extraction ($p < 0.05$). The highest results were achieved under maximum conditions for mango, while for pineapple, optimal conditions were 5% EtOH, 50 °C, and 100 bar.

Gallic acid (GA) content was superior in MP-F samples, ranging from 712.81 to 1998.23 μg/g DM. Specifically, low temperatures (30 °C) favored its extraction, while low pressures decreased it. Therefore, the best GA extraction conditions for mango were 10% co-solvent, 30 °C, and 300 bar, with MP-F affected by pressure ($p < 0.05$). Similarly, mangiferin was detected at concentrations of 509.30±0.44 to 90589.85±3.76 μg/g DM, with MP-F extraction influenced by the percentage of ethanol ($p < 0.05$). In this case, high pressures and a greater amount of co-solvent favored extraction. In contrast, quercetin was extracted in greater quantities in freeze-dried samples (641.79±0.27 to 2073.94±0.92 μg/g DM), with optimal extraction conditions at 5% EtOH, 30 °C, and 200 bar. Moreover, the co-solvent had a statistically significant effect ($p < 0.05$).

Table 3. Statistical analysis (ANOVA) for MP-V in SFE

Variable	Main effects			Interaction and quadratic effects						
	A	B	C	AA	AB	AC	BB	BC	CC	R^2
Yield (%)	0.0059*	0.2669	0.8085	0.8589	0.6743	0.3276	0.2595	0.5627	0.7268	0.72
DPPH (TEAC)	0.1494	0.2682	0.1696	0.1402	0.3784	0.4907	0.3971	0.1378	0.2736	0.66
ABTS (TEAC)	0.1909	0.2429	0.1919	0.1277	0.4453	0.6372	0.5124	0.1938	0.1904	0.63
Total phenols (GAE)	0.1335	0.2550	0.2529	0.1008	0.7100	0.6451	0.7337	0.2131	0.1284	0.62
Manguiferin (μg/g)	0.4014	0.5988	0.4339*	0.6450	0.3388	0.9464	0.4414	0.2486	0.8076	0.75
Gallic acid (μg/g)	0.7251	0.8542	0.2274	0.8484	0.8239	0.8947	0.3340	0.2764	0.5350	0.64
Ellagic acid (μg/g)	0.3711	0.3280	0.2842	0.9585	0.9411	0.6855	0.2582	0.2249	0.5445	0.73
Quercetin (μg/g)	0.0221*	0.9160	0.1044*	0.7329	0.5186	0.3334	0.0904	0.8220	0.1530	0.69

A: co-solvent flow (ethanol); B: temperature; C: pressure

*Statistical significance at $p < 0.05$.

Table 4. Statistical analysis (ANOVA) for PP-F in SFE

Variable	Main effects			Interaction and quadratic effects						
	A	B	C	AA	AB	AC	BB	BC	CC	R^2
Yield (%)	0.0003*	0.0006*	0.8230*	0.0644	0.5862	0.6355	0.7656	0.2718	0.0409*	0.91
DPPH (TEAC)	0.4061	0.2626	0.2393	0.0713	0.5798	0.1396	0.8094	0.4854	0.9373	0.64
ABTS (TEAC)	0.1332	0.2022	0.1475	0.1106	0.2836	0.1632	0.2870	0.9208	0.6222	0.66
Total phenols (GAE)	0.2067	0.2054	0.1536	0.2154	0.5753	0.2379	0.6212	0.7434	0.8348	0.58
Ellagic acid (μg/g)	0.4098	0.1354	0.3041	0.1771	0.4996	0.0522	0.7034	0.8049	0.8814	0.63

A: co-solvent flow (ethanol); B: temperature; C: pressure
* Statistical significance at $p < 0.05$.

Optimization of supercritical fluid extraction process

Numerical optimization was used to find the factor combination that optimizes multiple responses, maximizing the desirability function (Table 5).

For mango peel, the optimum conditions with a desirability of 0.977 were a co-solvent flow rate of 13.65%, a temperature of 36.76 °C, and a pressure of 294.82 bar. Quercetin was excluded from optimization due to its distinct response to high co-solvent, pressure, and low temperature. For pineapple peel, optimization was based on yield and ellagic acid, as they retain statistical independence and improve extract quality. The optimum conditions were a co-solvent flow rate of 12.39%, a temperature of 43 °C, and a pressure of 100 bar, with a desirability of 0.909. The test was performed in triplicate, and Table 5 shows good agreement between experimentally obtained and predicted values.

Table 5. Experimental validation of SFE processing at optimum conditions

Variable	Mango peel		Pineapple peel	
	Experimental	Predicted	Experimental	Predicted
Yield (%)	23.55 ± 0.13	23.96	24.19 ± 2.03	24.20
DPPH (TEAC)	804.39 ± 4.52	805.56	N.E.	N.E.
ABTS (TEAC)	519.65 ± 6.56	520.54	N.E.	N.E.
Total phenols (GAE)	46.83 ± 0.09	48.10	N.E.	N.E.
Manguiferin (μg/g)	383.91 ± 4.31	379.78	N.E.	N.E.
Gallic acid (μg/g)	10.76 ± 0.97	10.70	N.E.	N.E.
Ellagic acid (μg/g)	25.05 ± 1.48	25.12	1.55 ± 0.03	1.54

N.E.: Not evaluated

Discussion

The temperature used in the VD process for mango was not very high, and the exposure time was short compared to the FD process. This likely preserved the phenolic compounds to a greater extent. In contrast, for pineapple, the tissue may not have deteriorated enough to produce a cellular rupture, which would have allowed more phenolic compounds to be released. Additionally, the lesser thickness of mango peel, compared to pineapple peel, could make it more susceptible to being affected by FD due to faster heat transfer, leading to the loss and oxidation of phenolic compounds over time. A collapse could also occur, generated when the solid matrix of the food can no longer support its weight. This phenomenon has been reported by other authors and occurs when certain operating variables are not well configured, causing severe structural changes such as loss of porosity, among others (Hossain *et al*., 2010; Karam *et al*., 2016). As is known, the quality of dry products changes during the drying process depending on the conditions selected, the food matrix, and the method used. By choosing the latter correctly, the porosity of the final product can be controlled, which is an important factor in obtaining a dry product with excellent characteristics.

Additionally, the rough and porous structure of the MP-V samples could be attributed to the vacuum environment, which causes an internal and external pressure difference, accelerating moisture migration from the sample to the periphery (Kantrong *et al*., 2014; Wang *et al*., 2017). This structural characteristic may explain the higher antioxidant capacity values observed in the VD-treated mango peel. Similarly, Ye *et al*. (2019) found higher flavonoid and phenolic compound content in vacuum-dried A. roxburghii samples compared to lyophilized samples. Therefore, VD may represent the most suitable drying method for mango peel.

Conversely, for pineapple peel, the FD method resulted in a crunchy porous structure, maintaining the original features due to the rapid freezing and sublimation process (Chen *et al*., 2020). In contrast, the PP-V samples were considerably ruptured, likely due to the applied negative pressure causing significant shrinkage (Hossain *et al*., 2010; Xu *et al*., 2020). These findings suggest that the FD method and its processing conditions affect the stability and extractability of bioactive compounds to a lesser extent than the VD method, resulting in better outcomes. It is important to emphasize that cryogenic grinding is crucial for preserving and facilitating the release of bio compounds during the subsequent extraction stage.

The yield results for mango and pineapple peels align with previous studies. Dos Santos *et al*. (2013) reported a yield range of 0.69% - 1.16% using SFE for mango leaves, while Souza (2015) found values between 11.6 ± 0.6% -

37.1 ± 0.7% for mango peel extracted by different methods, with SFE yielding 3.7±0.1% using CO_2 + 2.5% EtOH and 4.0±0.1% with CO_2 + 5.0% EtOH. For pineapple, Oliveira *et al*. (2009) achieved a yield of 30.2% in methanolic extracts of pineapple residues, and Kalaiselvi *et al*. (2012) reported yields of 2.40% in ethanol and 2.12% in water for *Ananas comosus* peel. The highest X_0 for all tests was obtained under the conditions of 100 bar, 50 °C, and 15% ethanol. Although not significant, pressure had a negative effect on yield, consistent with findings by Prado *et al*. (2013) for extraction pressures of 10 MPa, which can be attributed to the variation in solubility of solutes in the fluid phase due to changes in pressure and temperature (Silva *et al*., 2009).

Furthermore, the antioxidant capacity measured by the ABTS method improved in mango peel with increases in ethanol flow rate and pressure, likely due to increased solubility of compounds in the $ScCO_2$ - EtOH solution as pressure increased. Conversely, in pineapple peel, the ethanol flow rate had a negative effect. This difference can be attributed to the extraction capacity of $ScCO_2$ and ethanol, which depends on the nature of the biological matrix and the selectivity of the solvents for the compounds in the matrix. It is possible that the solute-matrix diffusion process was not favored in pineapple, reducing the release of active sites and thus better extraction of compounds (Garcia-Mendoza *et al*., 2015; Meneses *et al*., 2015).

Moreover, for mango extracts, intermediate values (10% EtOH and 40 °C) resulted in the lowest antioxidant capacity. Sogi *et al*. (2013) observed an ABTS activity of 197 μM Trolox equivalent (TE)/g extract in freeze-dried mango peel of the Tommy Atkins variety, while Madalageri *et al*. (2015) reported 24.76–24.81 mg TE/g DM in methanol extraction from peel of three mango varieties. Similarly, Martínez *et al*. (2012) reported antioxidant activities of 1.7±0.20 μM TE/g and 7.7±0.90 μM TE/g in ethanol and methanol: acetone extracts, respectively, from pineapple co-products. The values obtained in the present study for antioxidant capacity as TE were within the range of these reported results.

On the other hand, DPPH radical scavenging activity results varied from 36.5 to 52.0 TEAC in dried "Tommy Atkins" mango flesh (Le, 2012) and from 60.6–63.4 mg TE/g in ripe mango peels to 72.4–75.3 mg TE/g in raw peels (Tokas *et al*., 2020). Additionally, Martínez *et al*. (2012) reported values of 1.7±0.19 μM TE/g in ethanolic extract and 4.8±0.10 μM TE/g in methanol: acetone extract for pineapple co-products. Our results are consistent and even superior to those found in these investigations. According to Palafox-Carlos *et al*. (2012), polyphenols exhibit high cellular activity during the ripening process due to the metabolization of various biomolecules. The state of maturity of the samples, with possible oxidation of the compounds by enzymatic reactions

common to this process, could explain the low antioxidant activity values obtained by the DPPH method.

Overall, the results of antioxidant activity in this study were above or within previously reported ranges. For mango, previous studies reported 117±13.5 and 2.4±0.3 mg GAE/g in mango kernel and flesh, respectively, and 1.68–13.28 mg GAE/g dry weight in different mango peel extracts (Soong and Barlow, 2004; Souza, 2015; Liu *et al*., 2017). For pineapple, the results showed that freeze-dried peel retained more polyphenols than vacuum-dried peel, consistent with Kuskoski *et al*. (2006) and Oliveira *et al*. (2009), but lower than those reported by Da Silva *et al*. (2014).

The extraction variables followed the same trend with respect to antioxidant capacity for the extracts analyzed. The influence of solvent power, which increases with pressure, likely explains the behavior where pressure and ethanol favored extraction. High pressure applied on ethanol enhances mass transfer of solutes to the solvent, improving cell permeability and diffusion of secondary metabolites (Mustafa and Turner, 2011; Luthria, 2012). However, Osorio-Tobón *et al*. (2014) reported a negligible effect of increased pressure without the proper combination of other factors such as temperature.

Overall, the evaluated parameters indicated that ethanol flow rate and pressure had a generally positive effect on extraction, allowing the recovery of greater amounts of these compounds. The choice of solvents plays a crucial role due to differences in polarity, which can enhance solubility through specific solvent-matrix interactions (Paula *et al*., 2013). The polar nature of polyphenols allowed them to be extracted mainly with ethanol as a co-solvent, while supercritical CO_2 was used to obtain a polyphenolic fraction of lower polarity (Silva *et al*., 2009). For polyphenols like gallic acid, which have a simple structure, the ethanol flow rate had a negative effect due to their high polarity and affinity for solvents like water.

Furthermore, the extraction conditions significantly influenced the solubility of mangiferin, ellagic acid, and quercetin, given their low solubility in CO_2 due to high polarity. However, quercetin showed a negative effect with ethanol flux, as reported in previous studies (Sánchez-Mesa *et al*., 2020).

The optimization process effectively identified conditions that enhance the extraction of desired bio-compounds while minimizing unwanted co-extractions. The high desirability scores indicate that the chosen conditions are well-suited for maximizing the efficiency and quality of the extracts. For mango peel, the exclusion of quercetin from the optimization highlights the complexity of optimizing multiple bioactive compounds simultaneously, as different compounds may respond differently to the same extraction conditions. In the case of pineapple peel, the focus on yield and ellagic acid reflects the need to balance

the extraction of specific compounds with overall extract quality. The successful alignment of experimental and predicted values underscores the robustness of the optimization approach used.

Based on the findings of this study, it is possible to conclude that VC and FD had an impact on yields and the antioxidant and functional properties of the extracts obtained by SFE. Freeze-drying induced more degradation in mango peel, reducing the content of extracted phytochemicals. On the contrary, this drying method allowed a better extraction of the compounds of interest when applied to pineapple peel. A first conclusion is that vacuum drying could be a suitable method for drying mango peel, while freeze-drying seems to be more appropriate for pineapple peel. SFE-CO_2 optimization by RSM based on CCD was effective to evaluate the effects of pressure, temperature, and co-solvent flow rate on yield and extraction of bioactive compounds from the evaluated by-products.

The optimized parameters to maximize extract quality within the experimental domain were 13% ethanol, 45.50 °C, and 300 bar for mango, and 12.39% ethanol, 43 °C, and 100 bar for pineapple. The contribution of each variable of the SFE to total yields, in vitro antioxidant capacity, and phytochemical composition evaluated could be ranked in the following order: ethanol flow rate > pressure > temperature. The results of this work confirmed that optimizing extraction conditions is a critical step to recover the major part of the antioxidative active constituents from studied byproducts. Fruit peels are generally discarded as waste, in this aspect SFE offers a better alternative by allowing to take full advantage of the whole fruit, transforming the peels into value-added products, which can be used among others, as an ingredient to produce functional foods for human consumption and therefore generate an additional source of income from agricultural waste.

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