Water Science & Technology

IWA PUBLISHING

Water Science & Technology Vol 88 No 9, 2400 doi: 10.2166/wst.2023.346

Optimization of phenolic compounds extraction from olive mill wastewater using response surface methodology

Elnaz Shahkouhmahali[a,*] and Jalal Mohamadzadeh[b]

[a] Department of Food Science & Technology, Golestan Institute of High Education, Gorgan, Iran
[b] Agricultural Engineering Research Department, Golestan Agricultural and Natural Resources Research Center, AREEO, Gorgan, Iran
*Corresponding author. E-mail: elenaz.shahkoohmahaly@gmail.com

ABSTRACT

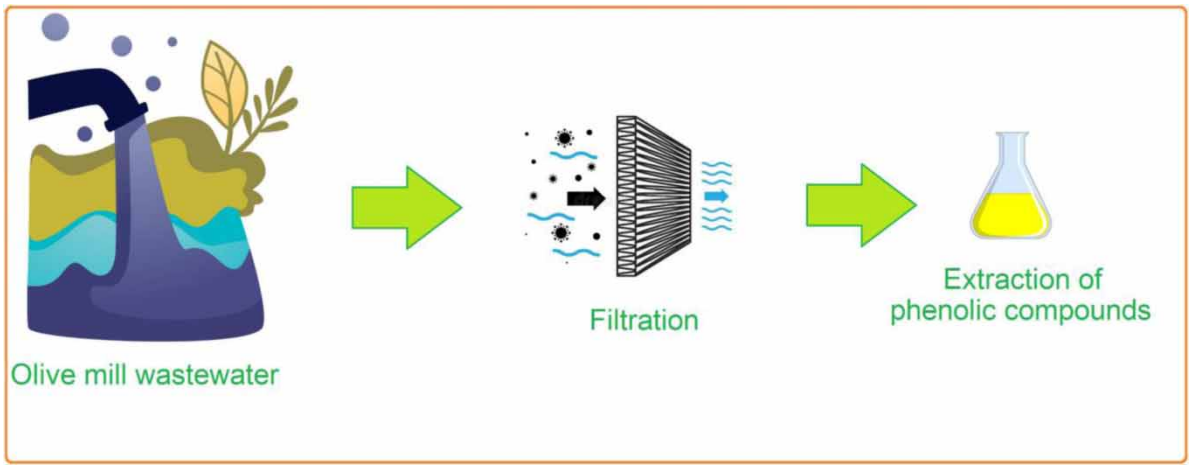
Large amounts of pomace are produced during the extraction of olive oil. This is because only 21% of an olive's weight is made up of oil, while 79% includes water, bark and pulp. This project extracted total phenolic compounds (TPCs) from olive mill wastewater as efficiently as possible. The TPCs were analyzed and the total antioxidant activity (TAA) was evaluated by spectrophotometry. Data were evaluated using the response surface method (RSM). The largest TAA and TPC were discovered in extracting using 80% ethanol at 25 °C, and 2.5 bar with pH = 4. The highest amount of TPCs was 11.614 mg of gallic acid per 100 mL, and a value of 71.06% was reported for TAA. The results of the quadratic model showed that R^2 is equal to 0.937, because it has a larger coefficient and the pH factor had the least effect. The temperature factor had the greatest impact on the extraction of TPC and TAA, and the mutual temperature and pH impacts affected the extraction positively. As a result, it can be concluded that the RSM was a useful tool for assessing the ideal circumstances for phenolic component extraction.

Key words: antioxidant activity, membrane methodology, optimization of extraction, polyphenolic compounds, response surface method

HIGHLIGHTS

- Producing phenolic extracts from olive byproducts is very important as a new product.
- A membrane can be used to recycle food components with high added value.
- Different ultrafiltration and nanofiltration are used.
- Optimal use of harmful compounds in the wastewater of olive oil factories that are used in the pharmaceutical and food industries. The advantages are lower cost and easier access and stronger filtration.

GRAPHICAL ABSTRACT



1. INTRODUCTION

In Europe, with 95% of the global edible oil production, olive oil wastes are considered an environmental problem (IOOC 2001). Different methods of olive oil extraction produce waste with varying capacities and properties. Over the past few decades, the olive sector has also implemented the continuous centrifugation extraction process in addition to the conventional hydraulic press method (Bhatnagar *et al.* 2014). Pomace and waste fluids from an olive mill are produced during extraction. The centrifugal extraction technique can be employed with each of the separate three- or two-phase systems. By generating just one pomace with nearly 80% humidity and consisting of peel, pulp, and stone, the two-phase centrifugation process prevents the growth of waste streams in olive mills. More consideration should be given to the experiments and analytical measures that are made (Böhmer-Maas *et al.* 2020). Many variables, including the composition of solvent, extraction time, temperature, solvent/solid ratio, and the pressure of extraction, can have a significant impact on extraction efficacy (Wettasinghe & Shahidi 1999).

The response surface method (RSM), initially proposed by Box & Wilson (1951), made it possible to evaluate the impacts of different process variables and their interactions on response variables. RSM can be defined as a set of mathematical and statistical methods contributing effectively to creating, enhancing, and optimizing processes. The model's suitability has been checked using the lack of fit, the coefficient of determination (R^2), and the F test result. A solid pomace is formed within the three-phase centrifugation mechanism, including the fruit pulp, rind, and stone, having 25–50% moisture and 5–7% olive oil. Furthermore, as water is added within the three-phase centrifugation mechanism, a greater volume of olive mill wastewater (OMW) is formed than in the traditional method. The OMW produced in this system is primarily composed of water (83–94%), organic matter (4–16%), and mineral salt (0.4–2.5%) (Alu'datt *et al.* 2010). Olive pomace is used in organic fertilizers and animal supplementary foods (Innangi *et al.* 2017), as well as the preservation of food productions, including French fries.

The use of phenolic extract in food or medicine has the potential to improve human health. It can also be used as a natural food preservative and to keep oils, cosmetics, and pharmaceuticals from spoiling. The proper isolation and recycling of these chemicals are critical. Membrane separation technologies are increasingly being used in the food business, and this is one of the most important uses of these technologies. Membrane techniques can be used to recycle high-value food ingredients such as total phenolic compounds (TPCs) from olive oil effluents. The extraction of highly valuable chemicals from this widely created byproduct adds to its valorization (Benito-Román *et al.* 2020). The olive oil wastewater treatment process uses a variety of ultrafiltration and nanofiltration membranes, including laboratory-made and commercial membranes. Various pretreatment procedures, such as acidification and sedimentation, are utilized as principal pretreatments, with pH adjustment and coagulation serving as supplemental pretreatments, to limit the harmful effect of sediment on the membrane (Zirehpour & Rahimpour 2016).

The purpose of this article is to provide an overview of the methods for recycling usable materials and their uses in various sectors. The TPCs derived from the effluent related to olive oil factories can be used as antibiotics in the pharmaceutical and food industries, and an RSM is also effective in the optimization of the variables of the phenolic compounds extraction procedure to benefit from the industry's optimal conditions.

2. MATERIALS AND METHODOLOGY

2.1. Preparation of samples and chemicals

The wastewater of 'Kolonaki' olive variety was collected from the olive oil factory in Golestan, Iran. After correcting the pH with citric acid or hydrochloric acid from the Kimia chemistry company, the sample (olive wastewater) passed through an 80 mm filter to separate the suspended solids (Table 1). The Folin-Ciocalteu from Merc Company, DPPH (2-diphenyl-1-picryl-hydrazyl) from Merc Company, and Na_2CO_3 from Kimia Chemistry Company were also used.

2.2. Empirical design

Response surface methodology is a collection of mathematical and statistical techniques whose purpose is to analyze, by an empirical model, problems as the one posed. The extraction process was optimized using RSM. The study also benefited from the central composition design (CCD). The independent variables were pH concentration, temperature, and pressure. Each variable that was to be optimized had three levels of encoding (-1, 0, and $+1$). Twenty randomized experiments with three replicates were applied in this study. The TPC and the total antioxidant activity (TAA) were chosen as the dependent variables

Table 1 | Basic characteristics of OMW

Parameter	Value	Unit
Lipids	8.02 ± 0.0	% w/w
Total solids	7.5 ± 0.6	% w/w
Sugar	3.2 ± 0.7	% w/w
Density	5.03 ± 0.1	g/mL
TPC	2,304 ± 14	mg/L
pH	13.1 ± 0.5	–

(responses) (Table 2). Each condition was tested three times, with the mean values given as measured responses. TPC and TAA values were achieved using the specified optimal circumstances.

2.3. Membrane filtration technique

After preparation, the effluent entered the membrane process. Using membrane equipment available in Golestan Technical and Engineering Research Department (1 – Steel feed tank equipped with heater, 2 – Drain valve, 3 – Pump, 4 – flowmeter, 5 – Pressure gauge and 6 – Membrane section: microfiltration (three Module, 0.24 m^2, with sizes of 100, 50, and 5 μm) and in the spiral ultrafiltration stage of polyethylene sulfone, 1.6 m^2, and 20–80 kDa, with three pressure treatments (two to three times), temperature (20–30 °C), and pH (4.5–5.5). Separating TPC was performed, after which the phenolic extract underwent concentration in a vacuum and dried under a vacuum oven (Garcia-Castello *et al.* 2010).

Table 2 | Results obtained for different treatments designed with the response surface method

Standard order	pH	Temperature	Pressure	TPC	TAA
1	4	25	2.5	11.46	71.06
2	3.5	25	2.5	9.125	64.17
3	3.5	30	3	8.656	54.64
4	4	25	2	8.876	56.282
5	4.5	25	2.5	10.216	60.47
6	4	25	2.5	11.101	69.3
7	4.5	20	3	7.816	50.117
8	4	30	2.5	9.996	68.07
9	3.5	20	2	6.056	41.74
10	4.5	30	3	9.036	63.23
11	4	25	2.5	10.041	69.7
12	4	25	2.5	11.416	70.2
13	4	25	3	10.465	62.3
14	4.5	30	2	8.536	54.24
15	3.5	30	2	7.976	52.85
16	4.5	20	2	7.096	47.026
17	4	25	2.5	11.314	69.8
18	3.5	20	3	7.536	46.13
19	4	25	2.5	10.812	63.81
20	4	20	2.5	9.976	60.63

2.4. Determining TPC

Measurement of TPC was conducted utilizing the photometric Folin-Ciocalteu test modified by Swan & Hillis. In a mixing bowl, a mixture of 20 mL ethanolic extract of mill wastewater and 1.16 mL deionized water was prepared, followed by the addition of 100 mL of Folin-Ciocalteu reagent to the aforesaid solution and letting it rest for 6 min in a dark room. The addition of 300 mL of Na_2CO_3 to the aforesaid solution took place after the reagent had time to take effect. For 30 min, the aforesaid solution was deposited in a hot water bath at 40 °C. In the control solution, 80% ethanol was used instead of ethanolic extract, and the rest of the methods were the same as before. To calibrate the spectrophotometer, a control sample was analyzed. The spectrophotometer was calibrated at 765 nm with a control solution to measure total phenol compounds. The absorption at 765 nm of the ethanolic extract was evaluated. Known gallic acid concentrations at a range of 0–200 mg/L were considered to define the standard curve (Mashayekhi & Atashi 2015).

2.5. Determining TAA

The extraction method is the same as the one for ethanol extraction, including the addition of 1 mL of the prepared ethanolic extract and 1 mL of the diuretic reagent to the test tube. The control was prepared with 80% ethanol instead of ethanolic extract, and the rest of the methods were the same as the other samples. To ensure that the radical scavenging effect was effective, the test tubes containing the solution were held in the dark for 30 min. The absorbance was measured at 517 nm using a spectrophotometer. The gadget was calibrated with percent ethanol before reading the control sample and the rest of the samples for measurement. The final method for estimating TAA was used to calculate the final numbers (Mashayekhi & Atashi 2015).

$$\text{TAA(total antioxidant activity)} = \frac{A_c - A_s}{A_c} \times 100$$

where A_c is the absorption rate of the control sample and A_s is the absorption rate of each sample.

2.6. Statistical analysis

ANOVA was performed for TPC and TAA using Statistica 6.0 at a confidence level of 95% ($p < 0.05$). The model's suitability was checked using the lack of fit, R^2, and the F test results of ANOVA. The means were compared at a 5% level of significance using the Tukey test. The 3D response surface plots showed the link between the independent factors and the response variables.

3. RESULTS AND DISCUSSION

According to the results of a quadratic model (Table 3), it was shown that R^2 is equal to 0.937, because it has a larger coefficient, and the pH factor had the least effect. The effects of independent factors on the TPC amounts indicate that increasing the temperature decreased the TPC extraction efficiency. The analysis of variance revealed that the linear expressions of time, temperature, and pH significantly influenced the quantity of phenolic chemicals. The interactions, on the other hand, seemed to have no effect on the extraction rate.

3.1. Effects of temperature on TPC and TAA

The p-value, considering all values <0.05 significant, can be used to determine which effects are thought to be important. The possibility of seeing a statistical test result over or equal to what was discovered is known as the p-value. The significance of the coefficients is assessed using this value, and the bigger the importance of the coefficient of variation, the smaller the p-value. The TPCs have also been linked significantly by several authors to DPPH scavenging action (Cuvelier *et al.* 1996; Hagerman *et al.* 1998; Bakkalbaşı *et al.* 2005). The greater possibility rate at higher temperatures is due to TPC binding to proteins and polysaccharides, as well as increased TPC solubility. This can boost mass transfer and increase the extraction speed of TPCs (Gan & Latiff 2011).

The highest amount of TPC and TAA compounds was recorded at pH = 4 and pressure = 2.5 bar at temperature = 25, and this amount can be seen in Table 2, which is equal to 11.46 mL and 71.06%. According to Table 3, the temperature factor had the most positive effects on the TPC and TAA extraction, and the mutual effects of temperature and pressure, as well as temperature and pH, did not affect the extraction efficiency significantly.

Table 3 | Evaluation of analysis of variance of total phenolic compounds

Source	Sum of squares	DF	Mean square	*F*-value	*p*-value
TPC					
Model	43.91	9	4.88	16.44	<0.0001
pH	1.12	1	1.12	3.78	0.0804
Temperature	3.27	1	3.27	11.03	0.0077
Pressure	2.47	1	2.47	8.32	0.0163
Tem*pH	0.0181	1	0.0181	0.0608	0.8102
pH*pressure	0.1105	1	0.1105	0.3722	0.5554
Tem*pressure	0.1301	1	0.1301	0.4383	0.5229
pH^2	3.69	1	3069	12,045	0.0055
$Temperature^2$	1.96	1	1.96	6059	0.0280
$Pressure^2$	3.69	1	3.69	12.45	0.0055
Residual	2.97	10	0.2967		
Lack of fit	1033	5	0.2669	0.8172	0.5849
Pure error	1.63	5	0.3266		
TAA					
Model	1453.58	9	161.51	20.24	<0.0001
pH	24.13	1	24.13	3.02	0.1127
Temperature	224.36	1	224.36	28.11	0.0003
Pressure	58.85	1	58.85	7.37	0.0217
pH*temperature	0.0661	1	0.0661	0.0083	0.9293
pH*pressure	4.38	1	4.38	0.5491	0.4757
Temperature*pressure	1.38	1	1.38	0.1725	0.6866
pH^2	82.54	1	82.54	10.34	0.0092
$Temperature^2$	32.70	1	32.70	4.10	0.0705
$Pressure^2$	199.03	1	199.03	24.94	0.0005
Residual	79.81	10	7.98		
Lack of fit	45.97	5	9.19	1.36	0.3724
Pure error	33.84	5	6.77		

All numbers with p-value <0.0001 are significant. In this table temperature has significant effect on our total phenol and antioxidant activity.

Increased phenolic compounds' extraction efficiency was caused due to temperature raising, as evidenced by the effects of independent factors on the phenolic compounds' quantity. The results of the analysis of variance confirmed the strong association between the linear expressions of time, temperature, and pH and the concentration of phenolic chemicals. The interactions had no appreciable impact on the extraction rate (Figures 1(a) and 1(b)), though. The interaction of phenolic chemicals with proteins and polysaccharides caused an increase in extraction rate at higher temperatures (Figures 1(c) and 1(d)). This is because of phenolic compounds' higher solubility, which might enhance mass transfer and boost the extraction rate of the TPCs at greater temperatures come after extraction (Gan & Latiff 2011).

High temperatures may promote the simultaneous breakdown of antioxidants that were previously mobilized at lower temperatures while mobilizing some antioxidants. It was also mentioned that at high temperatures, the extraction rate of thermally stable antioxidants is larger than the breakdown rate of less soluble antioxidants. The comparatively greater antioxidant capacities of extracts made at higher temperatures have been used to support the increase in the extract of total antioxidants activity in high temperatures. Increasing the temperature may help with extraction by making phenolic chemicals more soluble in the solvent. Increased extraction rate and shorter extraction times may be two of the main effects of higher extraction temperatures (Cacace & Mazza 2002).

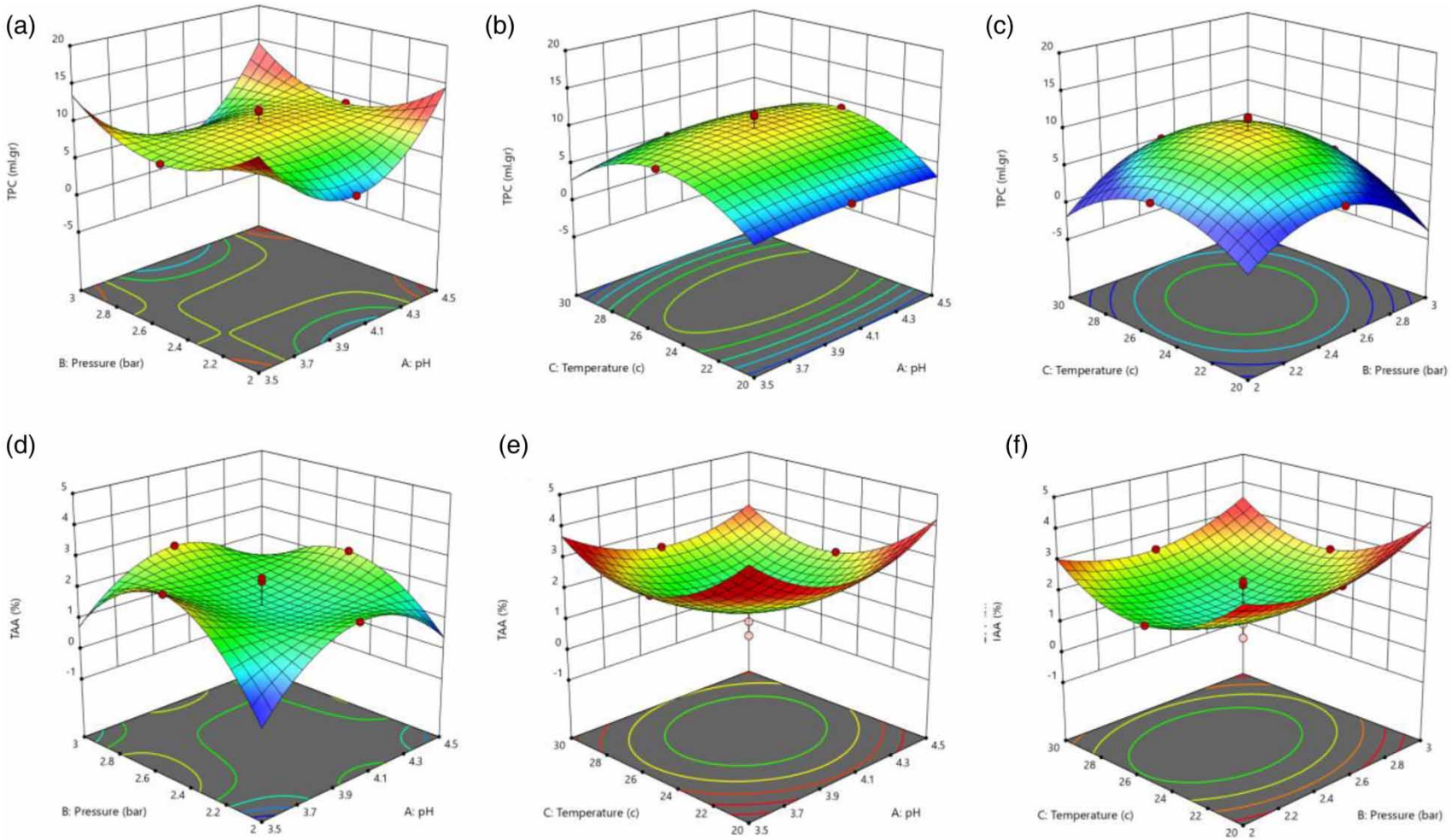


Figure 1 | Three-dimensional response surface plots and corresponding contour plots showing the effect of (a,d) pressure and pH; (b,e) temperature and pH; (c,f) temperature and pressure on TPC and TAA of OMW.

3.2. Effects of pressure on TPC and TAA

Figure 1(c) shows the positive impacts of temperature and pressure on the TPC extraction yield, which is in agreement with Luque-Rodríguez *et al.* (2007). Based on Table 3, the pressure factor did not affect TPC and TAA extraction significantly, and the mutual impacts of the pressure factor, temperature, pressure, and pH did not include significant changes (Figures 1(c) and 1(d)), but the maximum extraction was observed at pressure = 2.5 bar.

High pressures present in supercritical fluid extraction (SFE) cause cell disruption and reduce particle size. SFE achieves appropriateness for a wide range of objectives by combining ideal levels of pressure and temperature, the best particle diameter, and run duration (Pimentel-Moral *et al.* 2019). However, the higher pressure required in SFE necessitates proficiency and specialized equipment (such as a pressurization unit, gas storage system, as well as pressure sample vessel), limiting availability and driving up the cost of scaling (Panja 2018), which in agreement with the result of this study (Figure 1(b)).

Luque-Rodríguez *et al.* (2007) verified the benefits of pressure and temperature on TPC extraction yield. In this study, it was also shown that a precipitate forms after the subcritical water extraction, once the extracts are cooled to room temperature, and that if eliminated, a sizable portion of the bioactive substances dissolved at the high temperature is not taken into account. Re-dissolving this precipitate is, therefore, an essential step in order to precisely identify and measure the bioactive chemicals recovered under high-temperature conditions. A solvent with a polarity similar to that of water at a high temperature is produced by mixing ethanol with the subcritical water extract after it has been cooled.

3.3. Effects of pH on TPC and TAA

According to Table 3, the pH factor and its mutual effects did not have a noticeable effect on the extraction, the maximum amount of extraction was at pH = 4, and the mutual effect of temperature and pH simultaneously increased the extraction efficiency (Figure 1(b) and 1(e)).

Acidic foods (such as pomegranate peel at 6 M HCl and 40 °C within a 2 h period; (Sun *et al.* 2021), basic foods (such as red cabbage and Brussels sprouts at 4 M NaOH and 80 °C within 30- and 45-min periods), and other foods must all undergo a hydrolytic pretreatment (Gonzales *et al.* 2015) or enzymatic digestion (Angeloni *et al.* 2018). The measurement of TPCs and antioxidant properties was carried out using a spectrophotometric device, and the final data were analyzed with RSM software in a study that focused on optimizing the phenolic compounds' extraction from olive pomace utilizing methanolic extraction (Böhmer-Maas *et al.* 2020). The best conditions for extracting TPC were 40% methanol, 70 °C, and 180 min, whereas the best conditions for extracting antioxidants were 40% methanol, 45 °C, as well as 180 min. This confirms the findings of our study (Tsao 2010).

However, the polyphenols' extraction frequently takes place under low levels of pH as such substances take the neutral state in acidic environments, which is the most suitable for being solubilized (Figure 1(a) and 1(d)). However, excessive acidity might hinder extraction since the hydrolysis of simple (acyl) glycosides may affect the profile of native polyphenols (Ahmadian Koucharsarai *et al.* 2016). In addition, pH is crucial for releasing non-extractable polyphenols, which stay bound to matrix structural components in their natural state. In these cases, a hydrolytic pretreatment is needed.

The findings indicated that the optimum extraction efficiency of antioxidant compounds is obtained when the extraction duration is 104.29 min, the extraction temperature is 66.31 °C, and the percentage of ethanol is 58.96. The quantity of phenolic components under these conditions is 1,134 mg per 100 g of dry petal, flavonoids, 44.85 mg per gram of dry petal, anthocyanin, 35,840. 13 mg per 100 g of dried petal) were obtained, which, according to Figure 1 contradicts with our findings.

3.4 Optimizing extraction conditions

Collecting the greatest amount of TPCs and antioxidant qualities were considered as the objectives of studies in statistical analysis during the process of extracting TPC and antioxidant properties. Numerical optimization has been used to determine the best operating conditions. The optimization goals, response levels, and independent variables were first developed for this purpose. The utility function technique has been used to find the appropriate answers. The amount of phenolic compounds was 11.614 mg of gallic acid per 100 mL, and the TAA of the extract equaled 71.06%, based on the results of the optimum conditions for TPC extraction and antioxidant properties at 25 °C, 2.5 bar pressure, and 4.5 pH.

4. CONCLUSION

This paper mainly aimed to use the response surface technique to create optimal conditions to extract TPCs and antioxidant characteristics in relation to independent variables such as pH (4.5–5.5), temperature (20–30 °C), and pressure (2–3 bar). After analyzing certain data from phenolic compound extraction and free radical inhibition, it was found that the best conditions for phenolic compound extraction and antioxidant properties were 25 °C, pH = 4, and 2.5 bar. Producing a phenolic extract from a newly discovered substance that possesses the following features could be highly valuable. Technology-assisted extraction typically takes place under intense pressure and temperature (moderate to high), which cuts down on the amount of time and consumables (with powerful solvents) needed and produces very little waste. In brief, technically assisted extraction enhances quality standards in three important dimensions; decreased solvent volume, short working durations, and energy savings. The obtained foundational accomplishments offer environmental advantages that will significantly aid the industrialization of the techniques used in the laboratory. Although the use of membrane filtration technique to remove phenolic compounds from OMW was effective in this study, its use on an industrial scale faces challenges such as high cost, complex technology, and high maintenance cost.

STATEMENT OF NOVELTY

Producing phenolic extracts from olive byproducts can be very important as a new product. One of the separation technologies that has found increasing use in the food industry today is membrane separation processes, which is one of the important applications of the processes a membrane can be used to recycle food components with high added value, such as phenolic compounds, from the effluent of the olive oil extraction process. Different ultrafiltration and nanofiltration membranes, including membranes made on a laboratory scale and commercial membranes, are used in the olive oil effluent treatment process. This project has been carried out due to the optimal use of harmful and toxic compounds in the wastewater of olive oil factories, which are extracted using the membrane process of phenolic compounds in the wastewater and are used in the pharmaceutical and food industries. The advantage of using the membrane method is lower cost and easier access and stronger filtration.

ACKNOWLEDGEMENTS

The authors express their deep and sincere thanks to the Department of Agriculture Engineering Research Center special Dr Jalal Mohamadzadeh for funding the research.

FUNDING

The Department of Agriculture Engineering Research Center of Golestan provided financial support for the current work. A.D. obtained research support from Company D.

AUTHOR CONTRIBUTION

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Elnaz Shahkouhmahali, Elia Shahkouhmahali, Amin Mehrvar and Jalal Mohamadzadeh. The first draft of the manuscript was written by Elnaz Shahkouhmahali and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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First received 31 July 2023; accepted in revised form 20 October 2023. Available online 31 October 2023