processes

MDPI

Article

Response Surface Methodology: An Optimal Design for Maximising the Efficiency of Microwave-Assisted Extraction of Total Phenolic Compounds from *Coriandrum sativum* Leaves

Soraya Hihat [1,2], Noureddine Touati [2,3,*], Abdelhakim Sellal [3,4] and Khodir Madani [1,5]

[1] Laboratoire de Biomathématiques, Biophysique, Biochimie, et Scientométrie, Faculté des Sciences de la Nature et de la Vie, Université de Bejaia, Bejaia 06000, Algeria; soraya.hihat@univ-bba.dz (S.H.); khodir.madani@univ-bejaia.dz (K.M.)
[2] Département des Sciences Alimentaires, Faculté des Sciences de la Nature et de la Vie et des Sciences de la Terre et de l'Univers, Université Mohamed el Bachir el Ibrahimi, Bordj Bou Arreridj 34030, Algeria
[3] Laboratoire Santé et Environnement, Faculté des Sciences de la Nature et de la Vie et des Sciences de la Terre et de l'Univers, Université Mohamed el Bachir el Ibrahimi, Bordj Bou Arreridj 34030, Algeria; sellalhak@yahoo.fr
[4] Département de Biochimie, Faculté des Sciences de la Nature et de la Vie, Université Ferhat Abas Sétif 1, Sétif 19000, Algeria
[5] Centre National de Recherche en Technologies Agroalimentaires, Route de Targa-Ouzemour, Bejaia 06000, Algeria
* Correspondence: n.touati@univ-bba.dz; Tel.: +213-791-568-864

Abstract: The optimization of total phenolic compounds (TPC) extraction yield and maximization of total antioxidant capacity (TAC) from coriander leaves were investigated using response surface methodology. The extraction of TPC was carried out using microwave-assisted extraction. A Box-Behnken design was used to study the effects of the three independent variables, solvent concentration (ethanol/water 20–80%), microwave power (100–500 watt) and irradiation time (30–150 s) on the response. A second-order polynomial model was used to predict the reaction. The regression analysis showed that about 99% of the variations could be explained by the models. The predicted values were 50.97 GAE/g dw and 5.75 mg GAE/g dw for TPC and TAC, respectively. The reaction surface analysis showed that the optimum extraction parameters that maximized the extraction of antioxidants yield were 52.62% ethanol, 452.12 watt and 150 s. Under optimal conditions, the experimental values for TPC and TAC were 49.63 ± 0.93 mg GAE/g dw and 5.55 ± 0.07 mg GAE/g dw, respectively. The experimental values are in agreement with the predicted values, indicating the suitability of the model used and the success of the response surface methodology in optimizing the extraction conditions.

Keywords: *Coriandrum sativum*; microwave-assisted extraction; response surface methodology; total antioxidant capacity; total phenolic compounds

check for updates

Citation: Hihat, S.; Touati, N.; Sellal, A.; Madani, K. Response Surface Methodology: An Optimal Design for Maximising the Efficiency of Microwave-Assisted Extraction of Total Phenolic Compounds from *Coriandrum sativum* Leaves. *Processes* **2024**, *12*, 1031. https://doi.org/10.3390/pr12051031

Academic Editor: Agnieszka Zgoła-Grześkowiak

Received: 1 April 2024
Revised: 12 May 2024
Accepted: 14 May 2024
Published: 19 May 2024

Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

Polyphenols are bioactive compounds found abundantly in various plant-based foods that have antioxidant properties [1,2]. They are known for their potential health benefits, including reducing inflammation and protecting against certain chronic diseases such cardiovascular diseases, cancer, diabetes, and neurodegenerative disorders [3–6]. Polyphenols also play a crucial role in food preservation due to their antioxidant and antimicrobial activities. When used in food preservation, polyphenols help to prevent oxidation and spoilage by inhibiting the growth of microorganisms. This can extend the shelf life of food products and maintain their quality for longer periods of time [7].

Coriander, also known as cilantro, is a versatile herb that has a distinctive taste and aroma. It is widely used in different cuisines around the world and can be used in various forms, including leaves and seeds [1]. Cilantro leaves are often used as a garnish or ingredient in dishes like salsa, guacamole, and curries, while the seeds can be ground into a powder or used whole in dishes like pickles, sausages, and curries. In addition to its culinary uses, coriander has numerous health benefits that make it a valuable addition to any diet. Coriander leaves are a good source of antioxidants, specifically polyphenols, that can protect the body against damage caused by free radicals. According to a study conducted by Ashika et al. [8], coriander leaves contain vanillic, p-coumaric, cisferulic, and trans-ferulic acids, which are responsible for the antioxidant properties of the plant. Additionally, Scandar et al. [9] found that the leaves and stems of coriander are the most nutritious and beneficial parts of the plant. Nhut et al. [10] found that coriander leaves contain rutin, a type of flavonoid that has antioxidant, anti-inflammatory, and anti-cancer properties. The study also found that the total flavonoids content, the compound considered as one of the secondary plant metabolites, demonstrates the effectiveness of biological roles such as inhibiting plasma platelet aggregation, histamine release, and antiviral activity.

Extraction is the first and most important step in the recovery and purification of bioactive compounds from plant materials. Among the various novel and sophisticated extraction techniques that have been introduced and studied to improve efficiency, microwave-assisted extraction (MAE) has garnered significant attention in recent years for its efficient extraction of bioactive compounds from various plant sources [11,12]. MAE offers advantages such as reduced extraction times, higher yields, and lower environmental impact compared to traditional methods like Soxhlet extraction and maceration [13]. The success of MAE relies on factors like solvent choice, exposure time, temperature, and equipment specifications [14]. Additionally, the unique heating mechanism of microwaves allows for quicker extraction processes and better preservation of thermolabile compounds compared to conventional heating methods [15,16]. Furthermore, MAE has been recognized as an environmentally friendly technique that minimizes energy consumption and solvent usage, making it ideal for extracting bioactive compounds from agro-industrial waste [17].

Response surface methodology is a statistical and mathematical technique for modelling and optimizing process parameters [18]. Response surface methodology is highly useful in various fields such as engineering design optimization, chemical and biological process optimization, and natural product extraction [19]. It provides a systematic approach to relate the input variables to the responses and is a cost-effective and efficient method for optimizing experimental processes. The importance of response surface methodology lies in its ability to approximate the relationship between input and output variables, which supports the optimization of complex systems and reveals synergistic effects of process parameters. The methodology includes the design of experiments, the analysis of results and the creation of models to improve the results in different applications. The planes used in the response surface methodology study are quadratic planes such as the Box-Behnken planes or the central composite planes (Box-Wilson). Response modelling is performed using regression techniques that allow a response to be linked to a set of factors [20].

The aim of this study is to enhance the extraction efficiency of antioxidants from coriander leaves using microwave-assisted extraction. This will be achieved by modeling and optimizing the extraction conditions, which include solvent concentration, microwave power, and irradiation time. Response surface methodology will be utilized to determine the optimal conditions for extracting total phenolic compounds and total antioxidant capacity.

2. Materials and Methods

2.1. Chemical Reagents

DPPH reagent (2,2-diphenyl-1-picrylhydrazyl) and Sodium carbonate (Na_2CO_3) were purchased from Sigma–Aldrich (Darmstadt, Germany), Folin–Ciocalteu phenol reagent ($H_3PW_{12}O_{40}$) and gallic acid from Biochem, Chemopharma (Montreal, QC, Canada). Ethanol (Sigma–Aldrich, $\geq$99.8% (GS)) of analytical grade of purity were used. All chemicals and solvents used were of analytical grade.

2.2. Sample Preparation

In this study, coriander plant (*Coriander sativum*) was procured from a local market located in the Bejaia region of Northern Algeria. Leaves were separated from the stem and washed with tap water followed by distilled water, and left to dry for approximately 48 h at room temperature in a ventilated and dark room. The dried sample was then ground to obtain a fine powder with a diameter of less than 250 µm using a grinder (A11 basic grinder from Ika, Staufen in Germany). The powder was stored at −20 °C before analysis. The moisture content was determined by constant weight at 105 °C and was found to be 5.0 ± 0.5%. The water activity (Aw) was measured using a Hygro Palm Aw instrument (Rotronic AG Bassersdorf, Switzerland) and was found to be 0.18 ± 0.2 at a temperature of 20.6 °C.

2.3. Experimental Work

To optimize the microwave-assisted extraction procedure, a series of single-factor experiments were conducted to determine the effects of individual process parameters. This approach was chosen to minimize the total experimental work required while still providing valuable insights for improving the procedure. Through this method, the impact of each parameter on the process could be assessed and the most significant factors could be identified. This allowed for a more targeted and efficient optimization process (Table 1). The variable was kept constant (50%, 500 watt and 150 s for ethanol concentration, microwave power and irradiation time, respectively) when it was not studied.

Table 1. Results of single-factor experiments for microwave-assisted extraction from coriander leaves.

Solvent Concentration			Microwave Power			Irradiation Time		
%	TPC	TAC	watt	TPC	TAC	s	TPC	TAC
10	37.85 ± 0.24 [c]	2.92 ± 0.11 [c]	100	36.43 ± 0.65 [bc]	3.25 ± 0.13 [bc]	30	38.66 ± 0.14 [c]	3.06 ± 0.13 [c]
20	39.35 ± 0.34 [ab]	3.11 ± 0.09 [ab]	300	40.97 ± 0.93 [a]	3.89 ± 0.08 [a]	90	43.08 ± 0.27 [a]	4.03 ± 0.07 [a]
50	42.91 ± 0.48 [a]	3.91 ± 0.21 [a]	500	38.46 ± 0.29 [b]	2.48 ± 0.01 [b]	150	40.98 ± 0.75 [b]	3.59 ± 0.05 [b]
80	40.80 ± 0.82 [b]	3.50 ± 0.19 [b]	700	37.89 ± 0.83 [bc]	3.33 ± 0.15 [ab]	210	39.71 ± 0.53 [bc]	3.26 ± 0.14 [ab]
100	38.65 ± 0.59 [c]	2.89 ± 0.04 [c]	900	35.96 ± 0.47 [c]	2.98 ± 0.09 [bc]	270	38.88 ± 0.71 [c]	2.92 ± 0.04 [c]

Results are reported as mean ± SD. Same letters in the same column refer to mean not statistically different according to ANOVA test. TPC: total phenolic compounds (mg GAE/g dw); TAC: total antioxidant capacity (mg GAE/g dw); GAE: gallic acid equivalent; dw: dry weight of leaves.

In both the single-factor trials and the consecutive response surface methodology optimizations, the focus was on evaluating the extraction yield of total phenolic compounds and the maximization of total antioxidant capacity.

Following that, an optimization of the processes was carried out utilizing a response surface methodology approach based on a Box-Behnken design to refine the conditions (Table 2).

Table 2. Independent variables affecting the microwave-assisted extraction.

Independent Variables	Factor Levels		
	−1	0	+1
x1: Solvent concentration (%)	20	50	80
x2: Microwave power (watt)	100	300	500
x3: Irradiation time (s)	30	90	150

The experimental design applied was a three-level three-factor Box-Behnken design, and the required number of experiments (*N*) was determined using the formula outlined in Equation (1):

$$N = 2k(k-1) + C_0 \tag{1}$$

where k is the number of factors and C_0 is the number of central points (3).

To analyze the data, a regression analysis was conducted to fit a second-order polynomial equation (quadratic model) based on the general equation (Equation (2)) in order to predict the optimal conditions for the extraction process.

$$y = a_0 + \sum_{i=1}^{3} a_i x_i + \sum_{i=1}^{3} a_{ii} x_i^2 + \sum_{i=1}^{3} \sum_{j=1}^{3} a_{ij} x_i x_{j\ (i \neq j)} \quad (2)$$

where y represents the response function; a_0 is a constant coefficient; a_i, a_{ii} and a_{ij} are the coefficients of the linear, quadratic and interactive terms, respectively, and x_i and x_j represent the coded independent variables.

The factor levels were coded as −1 (low), 0 (central point or middle) and 1 (high). The variables were coded according to the following equation (Equation (3)):

$$x_i = \frac{X_i - X_0}{\Delta X} \quad (3)$$

where x_i represents a new variable (dimensionless) encoded from the original variable X_i. X_0 represents the reference value or initial value, and ΔX represents the increment or discretization step.

After conducting the analysis of variance, the regression coefficients for the individual linear, quadratic, and interaction terms were determined. To illustrate the impacts of independent variables and their interactions, three-dimensional surface plots were created from the fitted polynomial equation using the regression coefficients.

To confirm the reliability of the model, further extraction experiments were conducted at the predicted optimal conditions determined by the response surface methodology. The experimental results obtained were then compared to the values forecasted by the regression model.

2.4. Microwave-Assisted Extraction Process

For the optimization of the microwave-assisted extraction process, the following parameters influencing the extraction process were selected: solvent concentration, microwave power and irradiation time. A quantity (1 g) of coriander leaf powder was placed in a 250 mL bottom flask containing ethanol-water quantities. The suspension was extracted at different concentrations of solvent, microwave power and irradiation time. The extracts were separated by centrifugation at 3000 rpm (NF 200, Nüve, Turkey) for 10 min and stored at 4 °C until use. Fifteen tests were performed before the optimum was determined. An extraction was then performed under the optimum conditions determined using response surface methodology. The total phenolic compounds and total antioxidant capacity were considered in the response surface methodology optimization and model validation tests.

2.5. Response Parameters

2.5.1. Determination of the Total phenolic Compounds

The total phenolic compounds (TPC) of the plant samples was determined using the Folin-Ciocalteu reagent as reported by Singleton and Rossi [21]. A volume of 0.1 mL of the extract was mixed with 0.8 mL Folin-Ciocalteu reagent (10% *v*/*v*) and 0.4 mL sodium carbonate (7.5% *w*/*v*). The absorbance was measured at 720 nm (UV/Vis spectrophotometer, Biotech Engineering Management Co., Ltd., Nicosia, Cyprus) after 60 min of incubation at room temperature against a blank (made as reported for the sample but with 0.1 mL of sample solvent). The results were expressed as milligrams of gallic acid equivalent per gram of dry weight (mg GAE/g dw) by referring to a calibration curve.

2.5.2. Evaluation of the Total Antioxidant Capacity

The total antioxidant capacity (TAC) of the plant samples was evaluated using the DPPH radical method as reported by Brand-William [22]. A volume of 0.2 mL of the extract was mixed with 1 mL of methanolic DPPH solution (60 μM). The absorbance was measured at 515 nm after 30 min of incubation at room temperature against a blank (made as reported for the sample but with 0.2 mL of sample solvent). The results were expressed as milligrams of gallic acid equivalent per gram of dry weight (mg GAE/g dw) by referring to a calibration curve.

2.6. Statistical Analysis

All extraction trial and subsequent analyses were performed in triplicate, and the results are reported as means ± standard deviation (SD). The impact of each factor on the TPC yield and TAC in the single-factor experiment for microwave-assisted extraction was evaluated through ANOVA and Tukey's post hoc test at a 95% confidence level to determine their statistical significance. The data collected from the Box-Behnken design experiments for microwave-assisted extraction were subjected to ANOVA to assess the significance and appropriateness of the model for the response variable. A significance level of $p < 0.05$, $p < 0.01$ and $p < 0.001$ was considered as indicating significant, highly and very highly significant results, respectively. The JMP (Version 17.0, SAS, Cary, NC, USA) software was utilized to analyze all the experimental results and create the Box-Behnken design.

3. Results and Discussion

3.1. Effect of Single Factors

The influence of different extraction parameters on TPC yield and TAC was investigated using a method that varied one parameter at a time. The factors examined were ethanol concentration, microwave power, and irradiation time.

Choosing the right extraction solvent is crucial for determining the quantity and quality of the extracted phenolic compounds. Acetone, ethanol, and methanol are widely used solvents for extracting phenolic compounds from plants [23]; nevertheless, given the toxicity concerns associated with methanol and acetone, which are not suitable for food applications, ethanol solvent was chosen instead. Ethanol offers several advantages over other solvents, including higher extraction efficiency, environmental friendliness, and lower cost. The efficiency of extraction is significantly influenced by the concentration of ethanol, as reported in the literature [24]. Therefore, the impact of varying ethanol concentration was evaluated to optimize the extraction efficiency. Based on Table 1, the value of TPC and TAC ranged from 37.85 ± 0.24 to 42.91 ± 0.48 mg GAE/g dw and 2.92 ± 0.11 to 3.91 ± 0.21 mg GAE/g dw, respectively. As can be seen, the TPC and TAC increased significantly from the concentration of 10 to 50%, and then decreased to achieve a value of 38.65 ± 0.59 mg GAE/g dw and 2.89 ± 0.04 mg GAE/g dw, respectively. A similar tendency was reported for the extraction of total polyphenols from other plant material [25–27]. The effect of ethanol concentration on the extraction of antioxidants can be attributed to polarity changes. As the ethanol concentration in the solvent increases, its polarity decreases, which enhances the extraction of less polar components [28]. Moreover, the increase in ethanol concentration also promotes the breakdown of cell membranes, which in turn enhances the solvent's ability to penetrate the solid matrix during the extraction process [29,30]. At higher ethanol concentrations, the resulting polarity becomes unsuitable for extracting antioxidants from coriander leaves, making it less effective for this purpose. Given these findings, the concentration range of 20–80% was chosen for the response surface methodology trials.

The selection of microwave power is crucial in determining the efficiency and yield of the phenolic compounds extraction process. Higher microwave power, which leads to the increase in the temperature, can accelerate the extraction by disrupting hydrogen bonds, enhancing solvent penetration into the matrix, and facilitating the release of target compounds [31]; however, it is important to note that there is a limit to this effect and beyond a certain point, increasing the microwave power may not lead to any further improvements in the extraction yield as higher microwave power levels can induce degradation of phenolic compounds due to elevated temperatures [32]. As shown in Table 1, the value of TPC yield and TAC ranged from 35.96 ± 0.47 to 40.97 ± 0.93 mg GAE/g dw and 2.98 ± 0.09 to 3.89 ± 0.08 mg GAE/g dw, respectively. The variation of microwave power over the range of 100–900 watt caused the increment in TPC yield and TAC which was achieved at 300 watt followed by a significant decrease. Our result was lower than those reported in the literature [33]. Based on these results, the microwave power range 100–500 watt was selected for the response surface methodology trials.

According to the Table 1, the value of TPC and TAC ranged from 38.66 ± 0.14 to 43.08 ± 0.27 mg GAE/g dw and 2.92 ± 0.04 to 4.03 ± 0.07 mg GAE/g dw, respectively. As can be seen, the TPC and TAC increased significantly from the irradiation time of 30 to 90 s, and then decreased to achieve the value of 38.88 ± 0.71 mg GAE/g dw and 2.92 ± 0.04 mg GAE/g dw, respectively, after an extended extraction time. Our result was higher than that reported in the literature [12]. Based on these results, the irradiation time range 30–150 s was selected for the response surface methodology trials.

3.2. Optimization by Response Surface Methodology

3.2.1. Construction of the Experimental Plan

The optimization of the antioxidants extraction from coriander leaves by response surface methodology is operated using the Box-Behnken model based on the maximization of TPC extraction yield and TAC as response variables. Three parameters (independent variables), solvent concentration (ethanol 20–80%), microwave power (100–500 watt) and irradiation time (30–150 s) were investigated. The ranges (the lower and upper ends) of each independent variable were determined based on the result of the single factor effect from preliminary work (Section 2.2). The factors levels, observed and predicted values of TPC and TAC were summarized in Table 3.

Table 3. Box-Behnken design matrix, experimental and predicted values of total phenolic compounds (TPC) and total antioxidant capacity (TAC).

Run	Variable Levels			TPC (mg GAE/g dw)		TAC (mg GAE/g dw)	
	x_1	x_2	x_3	Observed Value	Predicted Value	Observed Value	Predicted Value
1	20 (−)	100 (−)	90 (0)	24.34	26.72	2.15	3.08
2	20 (−)	500 (+)	90 (0)	33.33	35.96	3.30	4.38
3	80 (+)	100 (−)	90 (0)	38.84	40.22	3.86	4.79
4	80 (+)	500 (+)	90 (0)	39.41	41.04	3.57	4.64
5	50 (0)	100 (−)	30 (−)	40.79	42.54	3.72	4.87
6	50 (0)	100 (−)	150 (+)	41.28	43.78	4.15	5.15
7	50 (0)	500 (+)	30 (−)	45.18	46.68	4.42	5.42
8	50 (0)	500 (+)	150 (+)	47.45	49.70	4.90	5.75
9	20 (−)	300 (0)	30 (−)	32.91	34.79	3.08	4.00
10	80 (+)	300 (0)	30 (−)	41.73	44.61	3.89	4.82
11	20 (−)	300 (0)	150 (+)	36.32	37.45	3.06	4.13
12	80 (+)	300 (0)	150 (+)	44.08	46.21	4.22	5.30
13	50 (0)	300 (0)	90 (0)	47.81	49.48	4.27	5.20
14	50 (0)	300 (0)	90 (0)	46.81	49.48	4.22	5.20
15	50 (0)	300 (0)	90 (0)	47.81	49.48	4.12	5.20

x_1, solvent concentration; x_2, microwave power; x_3, irradiation time; GAE: gallic acid equivalent; dw: dry weight of leaves.

The amount of TPC and the maximization of TAC from *Coriandrum sativum* leaf extract using the MAE method ranged from 26.34 to 49.70 mg GAE/g dw and 3.08 to 5.75 mg GAE/g dw, respectively. The highest yield in TPC and TAC was observed under extraction conditions of 50% (v/v) solvent concentration, 500 watt microwave power, and an irradiation time of 150 s. Hihat et al. [1], who investigated the effect of oven and microwave drying on the total polyphenols and antioxidant capacity of coriander leaves, reported values of 48.44 mg GAE/g dw and 82.21%, respectively.

3.2.2. Analysis of the Model

To assess the significance of the model, an ANOVA analysis was conducted. Table 4 shows the ANOVA results for the effects of solvent concentration, microwave power and irradiation time on TPC and TAC, relative to the dry weight of coriander leaves.

Table 4. Analysis of model variance and lack of fit for total phenolic compounds (TPC) and total antioxidant capacity (TAC) of coriander leaves.

Source	DF	Sum of Squares	Mean Square	F Ratio	Prob. > F
TPC (mg GAE/g dw)					
Model	9	619.553	68.839	87.880	<0.0001 *
Error	5	3.916	0.783		
Corrected total	14	623.470			
Lack of fit	3	3.250	1.083	3.250	0.2441
Pure error	2	0.666	0.333		
Total error	5	3.916			
R^2	0.994				
R^2 adjusted	0.982				
TAC (mg GAE/g dw)					
Model	9	6.401	0.711	35.498	0.0005 *
Error	5	0.100	0.020		
Corrected total	14	6.502			
Lack of fit	3	0.088	0.029	5.058	0.1695
Pure error	2	0.011	0.005		
Total error	5	0.100			
R^2	0.984				
R^2 adjusted	0.956				

* Statistically significant values ($p < 0.05$).

The coefficients of determination (R^2) for the TPC and TAC models are 0.994 and 0.984, respectively, indicating that only a very small percentage (0.06 and 0.16%, respectively) of the total variation remains unexplained by these models. In addition, the adjusted coefficients of determination (R^2 adj) for the TPC and TAC models are 0.982 and 0.956, respectively, indicating close agreement between the experimental and predicted values. The lack of fit test is used to determine if the model is appropriate for describing the experimental data or if another model should be selected. The lack of fit test values for TPC and TAC are 0.2441 and 0.1695, respectively, which are higher than 0.05 and not significant compared to the pure error. This suggests that the current model is sufficient to fit the experimental data.

The regression coefficients for the intercept, the linear, quadratic and interaction terms of the models were calculated using the least squares method and presented in Table 5. The ANOVA of regression coefficient showed a linear response based on the p-value of solvent concentrations (x_1), which were highly significant ($p < 0.0001$), followed by microwave power (x_2) and irradiation time (x_3) for both TPC and TAC. In the interaction between variables, only solvent concentration-microwave power (x_1x_2) had a significant effect on TPC ($p < 0.0051$) and TAC ($p < 0.0038$). While in the quadratic model, only solvent concentrations (${x_1}^2$) and microwave power (${x_3}^2$) were significant for TPC ($p < 0.0001$ and $p < 0.0002$, respectively), and solvent concentration (${x_1}^2$) and irradiation time (${x_3}^2$) for TAC ($p < 0.0001$ and $p < 0.0314$, respectively). The mathematical equations correlating the TPC and TAC (Equation (4) and Equation (5), respectively) with process variables are given below in terms of coded factors excluding non-significant terms.

$$TPC = 49.476 + 4.645x_1 + 2.515x_2 + 1.065x_3 - 2.105x_1x_2 - 9.205x_1^2 - 4.290x_2^2 \quad (4)$$

$$TAC = 5.203 + 0.493x_1 + 0.288x_2 + 0.152x_3 - 0.360x_1x_2 - 0.859x_1^2 + 0.218x_3^2 \quad (5)$$

Table 5. Regression coefficient, standard error, and Student's *t*-test results of response surface of TPC and TAC referred to dry weight of coriander leaves.

Parameter	Estimate	Standard Error	t Ratio	Prob > |t|
TPC (mg GAE/g dw)				
Intercept	49.476	0.510	96.83	<0.0001 *
Solvent concentration (x_1)	4.645	0.312	14.84	<0.0001 *
Microwave power (x_2)	2.515	0.312	8.04	0.0005 *
Irradiation time (x_3)	1.065	0.312	3.40	0.0192 *
x_1*x_2	−2.105	0.442	−4.76	0.0051 *
x_1*x_3	−0.265	0.442	−0.60	0.5754
x_2*x_3	0.445	0.442	1.01	0.3608
x_1*x_1	−9.205	0.460	−19.99	<0.0001 *
x_2*x_2	−4.290	0.460	−9.32	0.0002 *
x_3*x_3	0.489	0.460	1.06	0.3368
TAC (mg GAE/g dw)				
Intercept	5.203	0.081	63.67	<0.0001 *
Solvent concentration (x_1)	0.493	0.050	9.87	0.0002 *
Microwave power (x_2)	0.288	0.050	5.77	0.0022 *
Irradiation time (x_3)	0.152	0.050	3.05	0.0285 *
x_1*x_2	−0.360	0.070	−5.09	0.0038 *
x_1*x_3	0.087	0.070	1.24	0.2713
x_2*x_3	0.012	0.070	0.18	0.8667
x_1*x_1	−0.859	0.073	−11.66	<0.0001 *
x_2*x_2	−0.124	0.073	−1.69	0.1527
x_3*x_3	0.218	0.073	2.96	0.0314 *

* Values statistically significant at $p < 0.05$.

3.2.3. Analysis of Plots Describing Factors Effect

The most effective method for illustrating the impact of an independent variable on the yield of TPC extraction and TAC is to generate response surface plots using a model. This involves manipulating two variables within the experimental range being studied, while holding the remaining variable at its central level (0 level) [34]. Figure 1 demonstrates the influence of solvent concentration, microwave power, and irradiation time on the yield of TPC extraction and TAC from coriander leaves.

As can be seen in Figure 1a, the increase in solvent concentration (x_1) and microwave power (x_2) resulted in an augmented yield of TPC and TAC. However, higher solvent concentration (x_1) and microwave power (x_2) had a negative effect on the extraction, probably due to the decrease in polarity of the solvent [35] and/or the degradation of antioxidants [36]. Moreover, analysis of the surface plots showed that there were optimal levels of concentration of TPC and TAC, which is due to the quadratic effect of the solvent concentration (${x_1}^2$) as corroborated in Table 5. Additionally, the impact of solvent concentration was observed to be statistically significant in interaction with microwave power (x_1x_2) (p value 0.0051 and 0.0038 for TPC and TAC, respectively). This finding was in agreement with the study which reported the significance of ethanol percentage on the yield extraction of total polyphenols from *Pistacia lentiscus* [33]. Polarity plays a crucial role in extracting antioxidants from various plant and marine sources. Several studies have shown that the polarity of the solvent affects the antioxidant activity of the extracts. By increasing the ethanol concentration in the solvent, the cell membrane breaks down, allowing the solvent to more easily penetrate the solid matrix [29]. Moreover, increasing the ethanol concentration in the solvent reduces its polarity, which enhances the extraction of less polar components [28]. However, using a very high ethanol concentration is not suitable for extracting TPC from plant material.

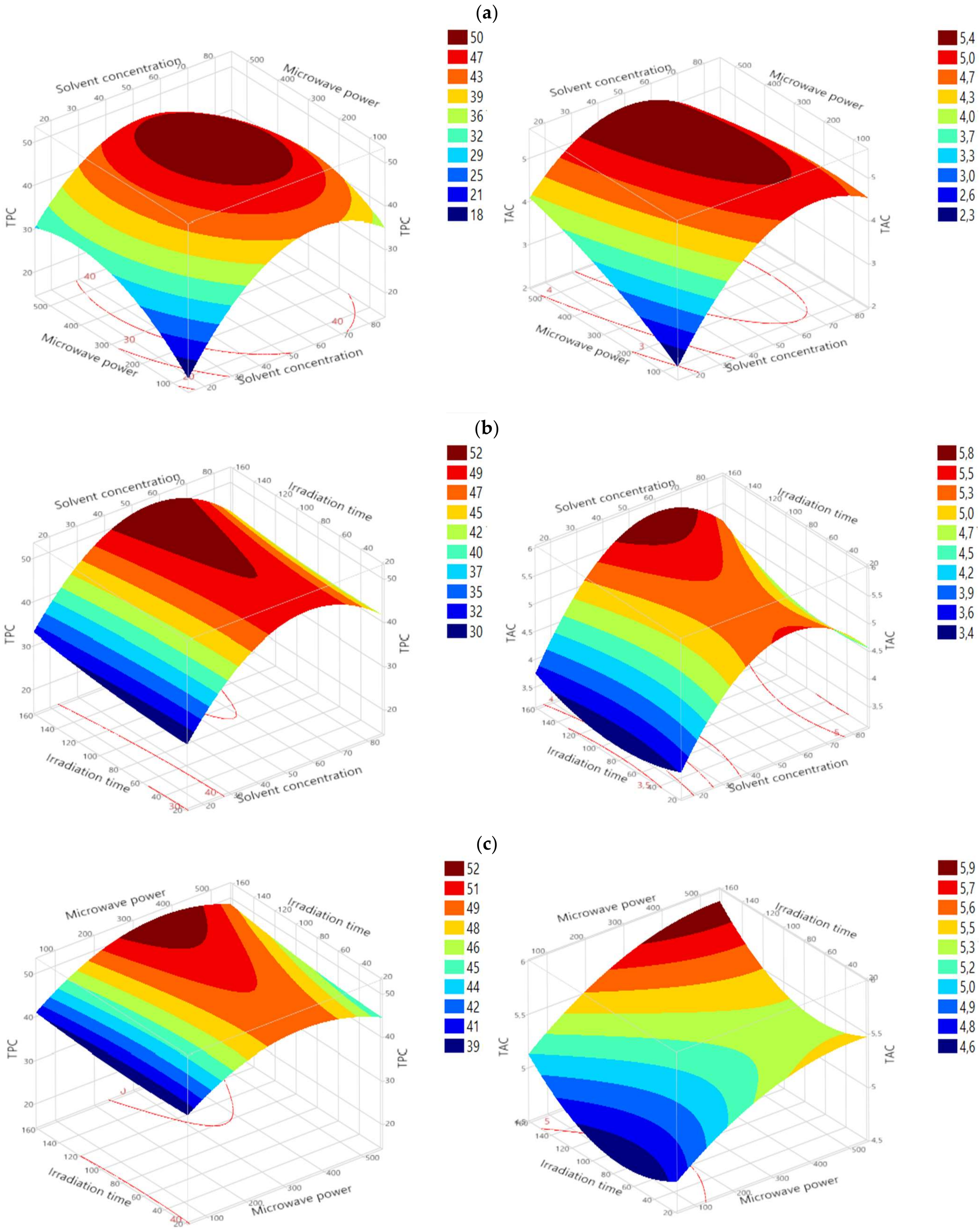

Figure 1. Effects of solvent concentration and microwave power (**a**), solvent concentration and irradiation time (**b**), and microwave power and irradiation time (**c**) on TPC (mg GAE/g dw) and TAC (mg GAE/g dw).

According to Figure 1b, the yield of TPC and TAC is affected by the increase in solvent concentration (x_1) and irradiation time (x_3). It was observed that both factors independently influenced the extraction of TPC and TAC. Additionally, analyzing the surface plots revealed that there were optimal concentration levels for TPC and TAC, which is attributed to the quadratic effect of irradiation time (x_3^2) as supported by Table 5. The impact of solvent concentration in interaction with irradiation time (x_1x_3) was found to be not significant (*p* value 0.5754 and 0.2713 for TPC and TAC, respectively). An elongated extraction time allows for prolonged contact between the plant material and the solvent, facilitating the transfer of phenolic compounds into the solvent. This can lead to increased extraction yields as more compounds have the opportunity to be released from the plant matrix [37].

Based on the Figure 1c, increasing microwave power (x_2) and irradiation time (x_3) resulted in higher yields of TPC and TAC. In other words, both microwave power (x_2) and irradiation time (x_3) simultaneously influenced TPC and TAC. However, it was observed that microwave power had a greater impact on TPC extraction than irradiation time. It was reported that the higher absorption of microwave energy led to increased temperatures within the sample, causing cell rupture and facilitating the release of antioxidant compounds [38]; however, excessive power may also degrade sensitive compounds [39]. Further, many studies have reported a decline in the levels of recovered TPC when using microwave-assisted extraction for long periods. Zhang et al. [40] observed that the yields of phenolic compounds of A. blazei increased significantly from 1 to 5 min during MAE, followed by a slight decrease.

3.2.4. Validation of the Model

To assess the predictive power of the model, the optimal conditions were determined based on maximum desirability. The optimal conditions for achieving the highest TPC and maximum TAC were found to be 52.62% ethanol concentration, 452.12 watt microwave power, and 150 s irradiation time. Under these conditions, the experimental values for TPC and TAC were 49.63 ± 0.93 mg GAE/g dw and 5.55 ± 0.07 mg GAE/g dw, respectively. These experimental results were in good agreement with the predicted values for TPC and TAC, which were 50.97 and 5.75 mg GAE/g dw, respectively. Zeković et al. [12] investigated coriander seeds and reported optimal microwave-assisted extraction conditions with an ethanol concentration of 63%, an extraction time of 19 min, and an irradiation power of 570 watt to simultaneously maximize total polyphenols yield and increase antioxidant activity. Their predicted values for TPC and antioxidant activity (IC_{50}) were 311.23 mg GAE/100 g and 0.0315 mg/mL, respectively.

4. Conclusions

The response surface methodology was employed to explore the individual and interactive effects of three variables, namely solvent concentration, microwave power and irradiation time, with the objective of optimizing the microwave-assisted extraction of TPC and the maximization of TAC from *Coriander sativum*. The high correlation of the mathematical model indicates that a quadratic polynomial model can be used for modelling the solid–liquid extraction of TPC and TAC. From the response surface plots, all three investigated factors significantly affected the TPC extraction yield and the maximization of TAC. The experimental values (49.63 ± 0.93 mg GAE/g dw and 5.55 ± 0.07 mg GAE/g dw for TPC and TAC, respectively) agreed with the predicted values (50.97 GAE/g dw and 5.75 mg GAE/g dw for TPC and TAC, respectively) and clearly showed the suitability of the developed quadratic models. These results confirm the predictability of the model for TPC from coriander leaves and TAC under the experimental conditions used (52.62% aqueous ethanol as solvent, 452.12 watt as microwave power and 150 s as irradiation time). This optimized process, which is simple, fast, efficient and non-denaturing, can be used for the extraction of substances of interest in both research and industrial settings.

Author Contributions: Conceptualization, S.H. and N.T.; Methodology, S.H. and N.T.; Software, S.H. and N.T.; Validation, S.H. and N.T.; Formal analysis, S.H. and N.T.; Investigation, S.H. and N.T.; Writing—original draft, S.H., N.T. and A.S.; Visualization, S.H. and N.T.; Supervision, K.M.; Project administration, K.M. All authors have read and agreed to the published version of the manuscript.

Funding: This work was financed by the Algerian Ministry of Higher Education and Scientific Research.

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Hihat, S.; Remini, H.; Madani, K. Effect of oven and microwave drying on phenolic compounds and antioxidant capacity of coriander leaves. *Int. Food Res. J.* **2017**, *24*, 503–509.
2. Touati, N.; Barba, F.J.; Louaileche, H.; Frigola, A.; Esteve, M.J. Effect of storage time and temperature on the quality of fruit nectars: Determination of nutritional loss indicators. *J. Food Qual.* **2016**, *39*, 209–217. [CrossRef]
3. Marinova, D.; Ribarova, F.; Atanassova, M. Total phenolics and total flavonoids in bulgarian fruits and vegetables. *J. Univ. Chem. Technol. Metal.* **2005**, *40*, 255–260.
4. Kang, M.H.; Lee, J.S.; Kim, H.Y.; Kwon, S.; Choi, Y.S.; Chung, H.R.; Kwak, T.K.; Cho, Y.H. Selecting items of a food behavior checklist for development of Nutrition Quotient (NQ) for children. *Korean J. Nutr.* **2012**, *45*, 372–389. [CrossRef]
5. Rubio, L.; Motilva, M.J.; Romero, M.P. Recent advances in biologically active compounds in herbs and spices: A review of the most effective antioxidant and anti-inflammatory active principle. *Food Sci. Nut.* **2013**, *53*, 943–953. [CrossRef] [PubMed]
6. Forzato, C.; Vida, V.; Berti, F. Biosensors and Sensing Systems for Rapid Analysis of Phenolic Compounds from Plants: A Comprehensive Review. *Biosensors* **2020**, *10*, 105. [CrossRef] [PubMed]
7. Sakhraoui, A.; Touati, N.; Hihat, S. Effect of Time and Temperature Storage on the Quality of unpasteurized Prickly Pear Juice Enriched with Hydro-soluble Opuntia ficus indica seeds Extract. *Turk. J. Agric. Food Sci. Technol.* **2023**, *11*, 1817–1824. [CrossRef]
8. Ashika, B.D.; Chitrali, L.R.; Naresh, S.; Sunil, K.S.; Akki, S.; Balasubramanian, S. Phytochemical studies on the methanolic extract of coriander sativum leaves-an in vitro approach. *Eur. J. Biomed. Pharm. Sci.* **2018**, *5*, 494–500.
9. Scandar, S.; Zadra, C.; Marcotullio, M.C. Coriander (*Coriandrum sativum*) polyphenols and their nutraceutical value against obesity and metabolic syndrome. *Molecules* **2023**, *28*, 4187. [CrossRef]
10. Nhut, P.T.; Quyen, N.T.N.; Truc, T.T.; Minh, L.V.; An, T.N.T.; Anh, N.H.T. Preliminary study on phytochemical, phenolic content, flavonoids and antioxidant activity of Coriandrum Sativum l. originating in Vietnam. *IOP Conf. Ser. Mater. Sci. Eng.* **2020**, *991*, 012022. [CrossRef]
11. Putnik, P.; Lorenzo, J.M.; Barba, F.J.; Roohinejad, S.; Jambrak, A.R.; Granato, D.; Montesano, D.; Kovačević, D.B. Novel Food Processing and Extraction Technologies of High-Added Value Compounds from Plant Materials. *Foods* **2018**, *7*, 106. [CrossRef]
12. Zeković, Z.; Vladić, J.; Vidović, S.; Adamović, D.; Pavlić, B. Optimization of microwave-assisted extraction (MAE) of coriander phenolic antioxidants–response surface methodology approach. *J. Sci. Food Agric.* **2016**, *13*, 4613–4622. [CrossRef]
13. López-Salazar, H.; Camacho-Díaz, B.H.; Ocampo, M.L.A.; Jiménez-Aparicio, A.R. Microwave-assisted extraction of functional compounds from plants: A Review. *Bioresources* **2023**, *18*, 6614–6638. [CrossRef]
14. Das, A.; Basak, S.; Chakrabartty, S.; Dhibar, M. Microwave: An ecologically innovative, green extraction technology. *Curr. Anal. Chem.* **2022**, *18*, 858–866. [CrossRef]
15. Dean, J.R. *Extraction Techniques for Environmental Analysis: Microwave-Assisted Extraction*; John Wiley & Sons Ltd.: Hoboken, NJ, USA, 2022; pp. 205–217.
16. Verma, A.; Alsayadi, G.M.H.; Choudhary, Y.; Sandal, P.; Kurmi, B.D. Microwave-assisted processes and their applications. *Pharmaspire* **2022**, *14*, 59–64. [CrossRef]
17. Arellano, C.A.; Corpuz, A.; Nguyen, L.T. Microwave-Assisted Extraction for the Valorization of Agro-Industrial Waste. In *Valorization of Agro-Industrial Byproducts*, 1st ed.; Anal, A.K., Panesar, S., Eds.; ImprintCRC Press: Boca Raton, FL, USA, 2022; pp. 31–48. [CrossRef]
18. Bouallegue, A.; Casilloc, A.; Chaari, F.; Cimini, D.; Corsaro, M.M.; Bachoual, R.; Ellouz-Chaabouni, S. Statistical optimization of levan: Influence of the parameter on levan structure and angiotensin I-converting enzyme inhibitory. *Int. J. Biol. Macromol.* **2020**, *158*, 945–952. [CrossRef]
19. Vesa, I.; Spraggon, M.; Fattah, I.M.R. Response surface methodology (RSM) for optimizing engine performance and emissions fueled with biofuel: Review of RSM for sustainability energy transition. *Results Eng.* **2023**, *18*, 101213. [CrossRef]
20. Myers, R.H.; Montgomery, D.C.; Anderson-Cook, C.M. *Response Surface Methodology: Process and Product Optimization Using Designed Experiments*, 3rd ed.; John Wiley & Sons: Hoboken, NJ, USA, 2009; ISBN 978-1-118-91601-8.
21. Singleton, V.L.; Rossi, J.A.J.R. Colorimetry of total phenolics with phosphomolybdic phosphotungstic acid reagents. *Am. J. Enol. Vitic.* **1965**, *16*, 144–158. [CrossRef]
22. Brand-Williams, W.; Cuvelier, M.E.; Berset, C. Use of a free radical method to evaluate antioxidant activity. *Food Sci. Technol.* **1995**, *28*, 25–30. [CrossRef]
23. Boeing, J.S.; Barizão, É.O.; Silva, B.S.; Montanher, P.F.; Almeida, V.D.C.; Visentainer, J.V. Evaluation of solvent effect on the extraction of phenolic compounds and antioxidant capacities from the berries: Application of principal component analysis. *Chem. Cent. J.* **2014**, *8*, 48. [CrossRef]
24. Jiménez-Moreno, N.; Volpe, F.; Moler, J.A.; Esparza, I.; Ancín-Azpilicueta, C. Impact of Extraction Conditions on the Phenolic Composition and Antioxidant Capacity of Grape Stem Extracts. *Antioxidants* **2019**, *8*, 597. [CrossRef] [PubMed]

25. Spigno, G.; Tramelli, L.; De Faveri, D.M. Effects of extraction time, temperature and solvent on concentration and antioxidant activity of grape marc phenolics. *J. Food Eng.* **2007**, *81*, 200–208. [CrossRef]
26. Li, H.; Deng, Z.; Wu, T.; Liu, R.; Loewen, S.; Tsao, R. Microwave-assisted extraction of phenolics with maximal antioxidant activities in tomatoes. *Food Chem.* **2012**, *130*, 928–936. [CrossRef]
27. Pan, X.; Niu, G.; Liu, H. Microwave-assisted extraction of tea polyphenols and tea caffeine from green tea leaves. *Chem. Eng. Process.* **2003**, *42*, 129–133. [CrossRef]
28. Cheok, C.Y.; Chin, N.L.; Yusof, Y.A.; Talib, R.A.; Law, C.L. Optimization of total phenolic content extracted from *Garcinia mangostana Linn*. hull using response surface methodology versus artificial neural network. *Ind. Crops Prod.* **2012**, *40*, 247–253. [CrossRef]
29. Vatai, T.; Škerget, M.; Knez, Ž. Extraction of phenolic compounds from elder berry and different grape marc varieties using organic solvents and/or supercritical carbon dioxide. *J. Food Eng.* **2009**, *90*, 246–254. [CrossRef]
30. Zhang, S.; Chen, R.; Wu, H.; Wang, C. Ginsenoside extraction from *Panax quinquefolium* L. (American ginseng) root by using ultrahigh pressure. *J. Pharm. Biomed. Anal.* **2006**, *41*, 57–63. [CrossRef] [PubMed]
31. Zeng, S.; Wang, B.; Lv, W.; Wu, Y. Effects of microwave power and hot air temperature on the physicochemical properties of dried ginger (*Zingiber officinale*) using microwave hot-air rolling drying. *Food Chem.* **2023**, *404*, 134741. [CrossRef]
32. Zahoor, I.; Khan, M.A. Microwave assisted convective drying of bitter gourd: Drying kinetics and effect on ascorbic acid, total phenolics and antioxidant activity. *J. Food Meas. Charact.* **2019**, *13*, 2481–2490. [CrossRef]
33. Dahmoune, F.; Spigno, G.; Moussi, K.; Remini, H.; Cherbal, A.; Madani, K. *Pistacia lentiscus* leaves as a source of phenolic compounds: Microwave-assisted extraction optimized and compared with ultrasound-assisted and conventional solvent extraction. *Ind. Crops. Prod.* **2014**, *61*, 31–40. [CrossRef]
34. Hayat, K.; Hussain, S.; Abbas, S.; Farooq, U.; Ding, B.; Xia, S. Optimized microwave-assisted extraction of phenolic acids from citrus mandarin peels and evaluation of antioxidant activity *in vitro*. *Separ. Puri. Technol.* **2009**, *70*, 63–70. [CrossRef]
35. Zhang, Z.S.; Li, D.; Wang, L.J.; Ozkan, N.; Chen, X.D.; Mao, Z.H.; Yang, H.Z. Optimization of ethanol-water extraction of lignans from flaxseed. *Sep. Purif. Technol.* **2007**, *57*, 17–24. [CrossRef]
36. Cheng, Y.; Xue, F.; Yang, Y. Hot water extraction of antioxidants from tea leaves—Optimization of brewing conditions for preparing antioxidant-rich tea drinks. *Molecules* **2023**, *28*, 3030. [CrossRef]
37. Jha, A.K.; Sit, N. Extraction of bioactive compounds from plant materials using combination of various novel methods: A review. *Trends Food Sci. Technol.* **2022**, *119*, 579–591. [CrossRef]
38. García-Baños, B.; Reinosa, J.J.; Peñaranda-Foix, F.L. Temperature assessment of microwave-enhanced heating processes. *Sci. Rep.* **2019**, *9*, 10809. [CrossRef] [PubMed]
39. Bachir Bey, M.; Meziant, L.; Benchikh, Y.; Louaileche, H. Deployment of response surface methodology to optimize recovery of dried dark fig (*Ficus carica* L., var. Azenjar) total phenolic compounds and antioxidant activity. *Int. Food Res. J.* **2014**, *21*, 1477–1482. [CrossRef]
40. Zhang, Z.; Lv, G.; Pan, H.; Fan, L. Optimisation of the microwave-assisted extraction process for six phenolic compounds in *Agaricus blazei* murrill. *Int. J. Food Sci. Technol.* **2012**, *47*, 24–31. [CrossRef]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.