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Valorization of plant extracts from local raw materials to obtain antioxidant products

SUMMARY OF THE DOCTORAL THESIS

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CUPRINS

INTRODUCTION

Polyphenols are a class of bioactive compounds of plant origin, known for their antioxidant, anti-inflammatory and antimicrobial properties (Vega, 2025). In the last two decades, the interest in investigating polyphenols has been demonstrated by numerous works related to the extraction and separation of polyphenolic compounds (Mojzer, 2016).

The main method of obtaining bioactive compounds, which are extremely necessary in the food and pharmaceutical industries, is extraction from plant sources (Wani, 2023). The extraction of polyphenols from less valuable sources, such as corn or flax seeds, or from food residues, such as grape marc, represents a sustainable strategy, which allows the recovery of some bioactive compounds, reducing waste and environmental impact.

Blackthorn (*Prunus spinosa* fruits) have various pharmacological properties, such as diuretic, anti-inflammatory, spasmolytic, etc. Despite all these beneficial properties, due to the astringent taste, food products from blackthorn are difficult to find on the market. Regarding the extraction of polyphenols, blackthorn, as a raw material, has not been the subject of many studies, although the literature presents data showing the remarkable antioxidant properties of blackthorn extracts (Oancea, 2024). Also, data are presented that showed the antibacterial effect of the aqueous extract of blackthorn on the strain of *Pseudomonas sp.* (Gegiu, 2015). In addition, the ethanolic extract shows antimicrobial activity against *Staphylococcus aureus*, *Escherichia coli*, *Pseudomonas aeruginosa*, *Salmonella abony* and *Candida albicans* (Velickovic, 2014).

Grape marc, another suitable raw material for the extraction of polyphenols, is a by-product of the wine industry that still contains compounds that can be exploited. The management of waste, such as grape marc, an area of significant environmental concern, and its use for obtaining polyphenol extraction can have significant economic advantages (Wani, 2023). The phenolic composition of grape marc varies greatly depending on the grape variety, climatic conditions, soil type, degree of ripeness and the winemaking process (Georgiev, 2014). The literature attributes the antioxidant activity of grapes to grape marc, 90% of this effect being due to the presence of anthocyanins (Yilmaz, 2004).

Also, there is not much data in the literature regarding the isolation and identification of polyphenolic compounds from flax seeds and their associated properties (Wang, 2024). From the available data, flax seed extract contributes to the reduction of cholesterol levels (Shim, 2015) and exhibits a strong antibacterial effect on *Escherichia coli*, *Staphylococcus aureus*, *Salmonella*, *Pseudomonas fluorescens* and *Listeria monocytogenes* (Wang, 2024).

In this context, the present study is relevant, as it addresses the challenges of efficient extraction of polyphenols from less exploited plant materials, considering the specific characteristics of each method used. At the same time, it proposes the optimization of extraction parameters to obtain maximum yield and the implementation of sustainable methods, with reduced consumption of toxic solvents and minimal impact on the environment.

This research falls within the field of chemical engineering, having direct implications in the optimization of extraction processes of bioactive compounds and in the development of pharmaceutical and food products.

Thus, the general objective of the experimental study in this paper was to optimize the methods of extraction of polyphenols from three plant materials (blackthorn, grape marc and flax seeds) in order to obtain natural extracts with applicability in the pharmaceutical and food industries.

The general objective of the study results in the following specific objectives:

- Optimization of the classical extraction of polyphenols from blackthorn.
- Optimization of the ultrasound-assisted extraction of polyphenols from blackthorn.
- Obtaining and using polyphenol extracts from grape marc in order to obtain lotions with a photoprotective effect.
- Combined use of microwave and ultrasound extraction technologies to increase the extraction yield of polyphenols from flax seeds.

Therefore, this work makes significant contributions in the field of polyphenol extraction, proposing solutions for optimizing extraction processes, implementing sustainable extraction methods and using extracts in order to obtain lotions with a photoprotective effect. The results can be applied in industry, contributing to the development of new pharmaceutical and food products.

I. ORIGINAL CONTRIBUTIONS

I.1. Objectives of the study

The aim of the study presented in this paper was to optimize the methods of extraction of polyphenols from three plant materials (blackthorn, grape marc and flax seeds) in order to obtain natural extracts with applicability in the pharmaceutical and food industries.

The main objectives of the experimental study are the following:

O1: Optimization of the classical extraction of polyphenols from blackthorn.

O2: Optimization of the ultrasound-assisted extraction of polyphenols from blackthorn.

O3: Obtaining and using polyphenol extracts from grape marc in order to obtain lotions with a photoprotective effect.

O4: Combined use of microwave and ultrasound extraction technologies to increase the extraction yield of polyphenols from flax seeds.

I.2. Materials and methods used

To obtain polyphenolic extracts from plant raw materials (blackthorn/grape marc/flax seeds), both conventional and unconventional methods were used (ultrasound extraction, microwave extraction, combined ultrasound and microwave extraction), using solvents such as ethanol, water or acetone.

After obtaining the extracts, they were analyzed by spectrophotometric methods: Folin-Ciocâlteu analysis, antioxidant activity analysis by the CUPRAC method, spectrophotometric analysis of anthocyanin content and determination of total polyphenol content using the OD280 index. Also, the determination of the chemical profile was performed by HPLC analysis and high-resolution mass spectrometry.

To obtain lotions with a photoprotective effect, the extracts obtained from the grape marc were mixed with a commercial lotion base. The photoprotective effect of the extracts was determined, and then the photoprotective effect of the lotions containing the extracts was analyzed by the COLIPA method.

All experimental measurements were performed in triplicate, and the results are presented as mean ± standard deviation (SD). A one-way ANOVA post hoc test with Tukey's HSD test was applied to assess whether the process factors had a significant effect ($p < 0.05$). The values of the dependent variables obtained at different levels of the extraction factors were processed using principal component analysis (PCA). The Pearson correlation coefficient (r) was used to assess the strength of linear correlations between the dependent variables. The response surface method was also used to quantify the effects of the process factors. In addition, the following experimental designs were selected: Plackett-Burman Design and Central Composite Design.

I.3. Results and discussions regarding the optimization of the conventional extraction process of polyphenols from *Prunus spinosa* fruits

This chapter aimed to establish the optimal conditions for the classical extraction of polyphenols from *Prunus spinosa* L. fruits. The effects of the following extraction parameters were studied: ethanol concentration in the extraction solvent (c_{et}), extraction temperature (t) and extraction time (τ). The response factors of the process were: total phenolic content (*TPC*), total anthocyanin content (*TAC*), antioxidant capacity (*AC*), protocatechuic acid content (*PA*), caffeic acid content (*CA*), vanillic acid content (*VA*), rutin hydrate content (*RH*) and quercetin content (*Q*). The results of this chapter were published in the article "*Effects of Extraction Process Factors on the Composition and Antioxidant Activity of Blackthorn (Prunus spinosa L.) Fruit Extracts*" (Drăghici-Popa, 2023).

According to the results, the maximum mean value of TPC (37.23 mg GAE/g DM) was obtained at the following optimal levels of process factors: c_{et} = 50%, τ = 30 min, and t = 60 °C. The values of TPC obtained in this study, i.e., 4.50 - 37.40 mg GAE/g DM (1.39 -11.59 mg GAE/g fresh fruit), were consistent with those reported in the related literature (Opriş, 2021).

The effects of process factors (c_{et}, τ and t) on total polyphenol content were quantified using the response surface regression model described with equation (*1*), where the regression coefficients were obtained based on experimental data.

$$TPC_{pred} = -24.18 + 0.782c_{et} - 0.008c_{et}^2 + 0.237\tau - 0.001\tau^2 + 1.208t - 0.010t^2 \qquad (1)$$

The statistical values characteristic of the regression model indicate a good agreement between the experimental and predicted data.

A desirability function, $d(Y)$, is defined by Equation (2) (Dima, 2020), where L_Y = 4.71 mg GAE/g DM and U_Y = 37.23 mg GAE/g DM are the lower and upper limits of process response. Under these optimal conditions, the process response is $Y_{opt} = TPC_{pred,opt}$ = 35.92 mg GAE/g DM and desirability function is $d(Y_{opt})$ = 0.96

$$d(Y)=\begin{cases}0 & \text{if } Y<L_Y\\ \dfrac{Y-L_Y}{U_Y-L_Y} & \text{if } L_Y\le Y\le U_Y\\ 1 & \text{if } Y>U_Y\end{cases} \tag{2}$$

The specific color of P. spinosa fruits indicates a high content of anthocyanins (Tahirović, 2018), as confirmed in the present study. The values of *TAC* obtained in this study, i.e., 0.03 - 0.42 mg C3GE/g DM (0.01 - 0.13 mg C3GE/g fresh fruit), were consistent with those reported in the literature. The anthocyanin content obtained by Tahirovic et al. by UAE with 50% ethanol (0.97 mg C3GE/g DM) highlights the strong influence of ultrasounds on the extraction of anthocyanins (Tahirović, 2018).

The effects of process factors (c_{et}, τ, and t) on total anthocyanin content were quantified using response surface regression model described by Equation (*3*), where the regression coefficients were estimated based on the experimental data.

$$TAC_{pred}=\left(-0.28+7.82c_{et}-0.08{c_{et}}^2+2.37\tau-0.01\tau^2+12.1t-0.10t^2\right)\times10^{-3} \tag{3}$$

Regarding *AC*, the effect of c_{et} was similar to those on *TPC* and *TAC*. Moreover, *AC* (19.49 - 68.04 µmol TE/g DM, 6.04 - 21.09 µmol TE/g fresh fruit) was very strongly correlated with *TPC* (r = 0.997) and *TAC* (r = 0.988). The highest mean value of *AC* at t = 30 °C (56.82 µmol TE/g DM) was achieved for c_{et} = 50% and the lowest mean value (20.30 µmol TE/g DM) for c_{et} = 100%. For c_{et} = 50%, the mean value of *AC* at t = 60 °C (67.36 µmol TE/g DM) was significantly higher (by 19%) than that obtained at t = 30 °C. Tahirović et al. demonstrated that antioxidant activity increased with decreasing ethanol concentration in the solvent mixture (Tahirović, 2018).

Using HPLC-PDA analysis of extracts obtained from *Prunus spinosa* fruits, five polyphenols were identified out of the twenty-three standard substances (table II.1), five in the extracts prepared at 30 °C and, respectively, four in the one obtained at 60 °C.

Tabelul I.1. HPLC-PDA polyphenolic profile of blackthorn extracts obtained at an extraction time of 30 min.

Sample	c_{et} (%)	t (°C)	*PA* (mg/100g DM)	*CA* (mg/100g DM)	*VA* (mg/100g DM)	*RH* (mg/100g DM)	*Q* (mg/100g DM)
P12	50	60	4.47 ± 0.06 a	2.96 ± 0.02 d	2.65 ± 0.11 a	1.87 ± 0.00 b	-
P1	50	30	3.97 ± 0.02 b	3.34 ± 0.02 a	2.37 ± 0.03 b	3.42 ± 0.17 a	0.26 ± 0.00 d
P2	66.67	30	3.83 ± 0.02 c	3.15 ± 0.03 b	2.55 ± 0.02 a	3.43 ± 0.33 a	0.83 ± 0.01 b
P3	75	30	3.75 ± 0.03 d	3.04 ± 0.02 c	2.51±0.01 ab	3.56 ± 0.02 a	1.02 ± 0.01 a
P4	100	30	2.08 ± 0.03 e	1.06 ± 0.01 e	1.46 ± 0.10 c	1.96 ± 0.09 b	0.68 ± 0.01 c

(cet) volume percentage of ethanol in the extraction solvent; (t) extraction temperature; (PA) protocatechuic acid content; (CA) caffeic acid content; (VA) vanillic acid content; (RH) rutin hydrate content; (Q) quercetin content; (DM) dry matter content; different letters in the same column indicate a significant difference ($p < 0.05$).

The FT-ICR-MS method was applied for further identification of the phenolic compounds quantified by HPLC-PDA. The phenolic compounds were successfully identified by comparing the predicted with the measured mass-to-charge ratio (m/z) values.

PCA was applied to evaluate the effects of the extraction conditions (c_{et} and *t*) on *TPC*, *TAC*, *AC*, *PA*, *CA*, *VA*, *RH*, and *Q* in the extract samples obtained at $\tau = 30$ min. PCA results highlighted that only the eigenvalues corresponding to PC1 (5.84) and PC2 (1.89) were > 1 and they explained 96.6% (73.0% + 23.6%) of the total variance.

The results suggest the following:

• depending on significant levels of factor loadings, the most important variables are *TPC*, *TAC*, *AC*, *PA*, *CA*, and *VA* for PC1 as well as *RH* and *Q* for PC2;

• extract samples P1 (c_{et} = 50% and t = 30 °C) and P12 (c_{et} = 50% and t = 60 °C) had higher values of *TPC*, *TAC*, *AC*, *PA*, *CA*, and *VA* than samples P4 (c_{et} = 100% and t = 30 °C) (discrimination on PC1);

• extract samples P2 (c_{et} = 66.67% and t = 30 °C) and P3 (c_{et} = 75% and t = 30 °C) had higher values of *RH* and *Q* than samples P4 (c_{et} = 100% and t = 30 °C) and P12 (c_{et} = 50% and t = 60 °C) (discrimination on PC2);

• *TPC*, *TAC*, *AC*, *PA*, *CA*, and *VA* were strongly directly correlated ($0.722 \leq r \leq 0.995$); *TPC*, *TAC*, and *AC* were inversely correlated with *Q* ($-0.748 \leq r \leq -0.634$); *RH* was directly correlated with *CA* ($r = 0.642$) and *Q* ($r = 0.515$).

This study highlighted that optimal extraction of some polyphenols requires special experimental conditions depending on their chemical and thermal stability, as well as molecular mass.

I.4. Results and discussions regarding the optimization of the ultrasound-assisted extraction process of polyphenols from pigeon peas

This chapter presents the study on the optimization of the ultrasound-assisted extraction process of polyphenolic compounds from *Prunus spinosa* fruits, using different aqueous ethanol solutions as green solvent. Using Plackett–Burman Design (PBD), six independent variables were studied: solvent/plant material ratio, ethanol concentration, extraction temperature, solvent pH, extraction amplitude and time. After identifying the relevant variables, i.e. solvent/plant material ratio, ethanol concentration and extraction temperature, the process was optimized by applying a Central Composite Design (CCD). The results of this chapter were published in the article "Optimization of Ultrasound-Assisted Extraction of Phenolic Compounds from Romanian Blackthorn (Prunus spinosa L.) Fruits" (Drăghici-Popa, 2025a).

According to the Plackett–Burman 6-factor design, 12 experiments were performed at 2 levels for each process factor, i.e. R_{LS} (5 cm^3/g and 15 cm^3/g), c_{et} (30% and 70%), t (30 °C and 70 °C), pH (2 and 7), A (30% and 70%) and τ (5 min and 15 min). The levels of the dimensionless are specified in figure II.1 and table II.2

Run	x_1	x_2	x_3	x_4	x_5	x_6
1	1	-1	1	-1	-1	-1
2	1	1	-1	1	-1	-1
3	-1	1	1	-1	1	-1
4	1	-1	1	1	-1	1
5	1	1	-1	1	1	-1
6	1	1	1	-1	1	1
7	-1	1	1	1	-1	1
8	-1	-1	1	1	1	-1
9	-1	-1	-1	1	1	1
10	1	-1	-1	-1	1	1
11	-1	1	-1	-1	-1	1
12	-1	-1	-1	-1	-1	-1

Fig. I.1. Levels of dimensionless factors in the PBD with 6 factors and 12 experimental runs

Tabelul I.2. Mean experimental values of extraction process responses ($y_{j,m}$, j = 1…3) and related values of regression coefficients (a_{ij}, i = 0…6, j = 1…3), coefficient of determination (R_j^2), adjusted coefficient of determination ($R_{j.adj}^2$), F statistic (F_j), and p_j-value for F_j at different levels of dimensionless process factors (x_i, i = 1…6) in PBD.

Run	x_1	x_2	x_3	x_4	x_5	x_6	$y_{1,m} = TPC_m$ (mg GAE/g DM)	$y_{2,m} = TAC_m$ (mg C3GE/g DM)	$y_{3,m} = AC_m$ (mg TE/g DM)
1	1	-1	1	-1	-1	-1	13.30	0.436	11.52
2	1	1	-1	1	-1	-1	8.160	0.134	5.343
3	-1	1	1	-1	1	-1	7.058	0.260	8.001
4	1	-1	1	1	-1	1	14.89	0.450	13.35
5	1	1	-1	1	1	-1	10.23	0.186	8.036
6	1	1	1	-1	1	1	11.72	0.226	9.611
7	-1	1	1	1	-1	1	7.316	0.160	8.290
8	-1	-1	1	1	1	-1	9.157	0.279	9.937
9	-1	-1	-1	1	1	1	8.543	0.364	9.883
10	1	-1	-1	-1	1	1	13.21	0.301	10.83
11	-1	1	-1	-1	-1	1	6.279	0.093	6.077
12	-1	-1	-1	-1	-1	-1	8.074	0.185	7.338
			j				1	2	3
			a_{0j}				**9.829**	**0.256**	**9.018**
			a_{1j}				**2.091**	0.033	**0.763**
			a_{2j}				**-1.367**	**-0.080**	**-1.458**
			a_{3j}				**0.744**	0.046	**1.099**
			a_{4j}				-0.112	0.006	0.122
			a_{5j}				0.159	0.013	0.366
			a_{6j}				0.498	0.010	0.656
			R_j^2				0.972	0.807	0.935
			$R_{j,adj}^2$				0.939	0.575	0.856
			F_j				29.13	3.476	11.91
			p_j				0.001	0.096	0.008

Statistically significant regression coefficients are highlighted in bold.

The factors that significantly affected the extraction process were R_{LS}, c_{et}, and t. Thus, these factors were further optimized.

Optimization of extraction process factors, aiming at maximizing the process response variables, i.e., *TPCpr*, *TACpr*, and *ACpr*, was based on the desirability function (d) approach (Drăghici-Popa, 2023). The optimal levels of dimensionless factors were $X_{1,opt}$, $X_{2,opt}$ and $X_{3,opt}$,, corresponding to $R_{LS,opt}$ = 15.1 cm^3/g, $c_{et,opt}$ = 33.2%, and t_{opt} = 66.8 °C, and the value of d under optimal process conditions was 0.963. The values of the process response variables predicted by Equation (4) at $X_{1,opt}$, $X_{2,opt}$ and $X_{3,opt}$, i.e. $Y_{j,pr,opt}$ (j = 1…3),, are specified in and Table II.3.

$$Y_{j,pr} = b_{0j} + \sum_{i=1}^{3} b_{ij} X_i + \sum_{i=1}^{3} b_{iij} X_i^2 + \sum_{\substack{i.k=1 \\ k>i}}^{3} b_{ikj} X_{ij} X_{kj}, \quad j = 1...3 \tag{4}$$

Tabelul I.3. Predicted and experimental values of extraction response variables under optimal process condi-tions.

j	**Response Variable**		**Optimal Value**		**Percentage Prediction Error**
	Symbol	**Units**	**Predicted** $Y_{j,pr,opt}$	**Experimental** $Y_{j,m,opt} \pm SD_j$	ε_j **(%)**
1	*TPC*	mg GAE/g DM	15.13	14.45 ± 0.718	-4.71
2	*TAC*	mg C3GE/g DM	0.589	0.405 ± 0.057	-1.90
3	*AC*	mg TE/g DM	14.71	16.75 ± 1.144	3.35

In the extract obtained at optimal levels of extraction process factors, six polyphenols were quantified: protocatechuic acid (6.83 mg/100 g DM), neochlorogenic acid (4.88 mg/100 g DM), chlorogenic acid (1.93 mg/100 g DM), caffeic acid (1.51 mg/100 g DM), vanillic acid (3.70 mg/100 g DM) and rutin hydrate (1.84 mg/100 g DM).

I.5. Results and discussions regarding the obtaining and use of polyphenolic extracts from grape marc in order to obtain lotions with a photoprotective effect

This chapter presents the use of polyphenolic extracts from grape marc to obtain sunscreen creams. Classical extractions were performed in ethanol and acetone solutions using grape marc from different grape varieties (Merlot, Burgund, Fetească Neagră, Isabella). The extracts obtained were analyzed to determine the total polyphenol content, antioxidant activity and sun protection factor (SPF). Sunscreen creams were prepared using the extract with the highest potential photoprotective effect determined.

Results presented in this chapter were published in the article "*Cosmetic Products with Potential Photoprotective Effects Based on Natural Compounds Extracted from Waste of the Winemaking Industry*" (Drăghici-Popa, 2024).

The experimental data obtained revealed that the extraction of polyphenolic compounds from grape marc is dependent on the type of marc and the type of solvent used for extraction. Thus, the highest extraction yields were obtained for grape marc from Feteasca Neagră, followed by grape marc from Merlot, Burgund and Isabella. The solvent that ensures the best extraction in all cases is 70% ethanol. Ethanol ensures a better extraction yield compared to acetone. The addition of water has a beneficial effect on both solvents, increasing the amount of extracted polyphenolic compounds. The best extraction yield is obtained in the case of 70% ethanol extraction of Feteasca Neagră grape marc (189 mg polyphenolic extract/g DM).

Analysis of the extracts by determining the OD280 index shows that the best results are obtained using 70% ethanol, followed by 100% ethanol. The grape pomace varieties that lead to a high OD280 index are: Feteasca Neagră (65.84 mg GAE/g DM, using 70% ethanol and 56.95 mg GAE/g DM using 100% ethanol) and Merlot (47.59 mg GAE/g DM, using 70% ethanol). Extraction with 100% or 70% acetone leads to low OD280 index values for all types of grape marc.

The analysis of the extracts obtained shows that the highest polyphenol content is obtained in the case of 70% acetone extraction of Feteasca Neagră pomace (238.62 mg GAE/g DM) followed by 70% ethanol extraction of Feteasca Neagră pomace.

Also, the evaluation of antioxidant activity (AC) by the CUPRAC method highlights the fact that Feteasca Neagră pomace has a high content of polyphenolic compounds with high antioxidant activity.

The data analysis shows that the best results, given the presence of compounds capable of absorbing UV radiation (290-320 nm), are obtained when the extraction is performed with 70% ethanol. In addition, the variety that contains these compounds in the highest quantity is the Merlot variety, followed by Feteasca Neagră and Burgund. The sun protection factor estimated by the Mansur method for 70% ethanol extraction from the Merlot variety is 7.83. In addition, for lotions obtained by adding polyphenolic extract, the protection factor increases with increasing polyphenol concentration, the maximum $SPF_{in\ vitro}$ value (14.07) being obtained when 50% aqueous polyphenolic extract is added to the lotion base. For ethanolic extract, the $SPF_{in\ vitro}$ value for the 50% concentration is 11.46. In all cases, creams obtained with ethanolic extracts show lower in vitro SPF values than creams obtained with aqueous extracts at the same concentrations.

I.6. Results and discussions regarding the study of the impact of green methods on the extraction of polyphenols from flax seeds

This chapter presents the integrated use of microwave and ultrasound extraction technologies to increase the extraction yield of polyphenols from flax seeds. Flax seeds were pretreated with microwaves before being subjected to ultrasound-assisted extraction. The impact of different parameters (ethanol concentration in the extraction solvent, extraction temperature and extraction time) on the extraction efficiency was also examined.

The article "Extraction of bioactive compounds from flaxseed using sequential green technology: microwave and ultrasound", which contains the results of this chapter, is accepted for publication in Scientific Bulletin, Series B (Drăghici-Popa, 2025b).

The obtained data showed a significant increase ($p<0.05$) in TPC and AC for the extract made with an ethanol concentration of 80% compared to 50% and 96%, regardless of the type of flax seeds used (microwave-pretreated flax seeds and ultrasound-assisted extraction – denoted ***MW+US***, untreated flax seeds and ultrasound-assisted extraction – denoted ***US***).

All unconventional methods (***MW+US***, ***US*** and microwave-pretreated flaxseed and conventional extraction – denoted ***MW***) led to better results compared to conventional extraction (untreated flaxseed and conventional extraction – denoted ***Conv***). In addition, extraction of microwave-pretreated seeds led to higher yields compared to extraction from untreated seeds, regardless of ethanol concentration.

Also, ANOVA analysis showed a non-significant increase in TPC in the temperature range from 30 to 60 °C, regardless of the type of flaxseed used (***MW+US*** and ***US***).

In addition, both TPC and AC increased significantly in the first 15 minutes (according to ANOVA analysis, $p<0.05$), regardless of the type of flaxseed used (***MW+US*** and ***US***). After 15 min of extraction, a slight decrease in polyphenol content occurs.

Therefore, the combined ***MW+US*** extraction (TPC = 7.72 mg GAE/g DM; AC = 10.25 mg TE/g DM) outperformed the individual methods, yielding a 30% improvement over ***MW***, a 24% increase over ***US***, and a 60% improvement over ***Conv***. This strategy is not only more efficient but also more environmentally friendly than traditional methods. The resulting polyphenol-rich extracts have potential applications in pharmaceuticals, food, and other industries as alternative sources of antioxidants.

II. Conclusions

In this work, the paper presented a study on the valorization of plants or plant residues to obtain polyphenol extracts because this class of compounds presents numerous beneficial properties: antioxidants, anti-inflammatory, antimicrobial, etc.

The paper addresses the challenges of efficient extraction of polyphenols from less valorized plant materials, considering the specific characteristics of each method, thus using, the extraction of polyphenols from cornelian cherry, flax seeds, and ostrich pomace was achieved. The extraction of bioactive compounds from less valorized plants or from food residues, a sustainable strategy, which allows the recovery of bioactive compounds, reducing waste and environmental impact.

Finally, I mention that a novelty element of the present research consists in the development and optimization of conventional and unconventional methods for the extraction of bioactive compounds from cornelian cherry, an unused plant material that is part of the spontaneous flora. Also, another new element consists in the possibility of valorizing some waste from the industrial industry in a less addressed direction, that of cosmetic products. Although the use of polyphenols from various sources in the production of cosmetic products is a widely addressed field at the moment, the production of lotions with a photoprotective effect is less studied. It was also aimed to find extraction options that target those compounds that have good and very good absorption in the UVB and UVA range, taking into account the fact that most polyphenols absorb mainly below 300 nm. The method of investigating the photoprotective effect aimed at finding simple, efficient and cheap methods that would allow the rapid characterization of the extracts obtained for use in sunscreen products. Given the compositional variety of grape marc and the multitude of factors that influence it, the results of the study offer the possibility of an analysis and a rapid decision regarding the direction of valorization of these wastes from the wine industry.

In addition, another element of originality was the combination of two advanced extraction techniques to increase the yield of polyphenols extracted from flax seeds. To date, studies have examined microwave pretreatment of flax seeds or ultrasonic extraction of polyphenols from flax seeds, the use of microunit and pretreatment of polyphenols for the extracted section of polyphenols has not been investigated. in.

Therefore, this work makes significant contributions in the field of polyphenol extraction, proposing solutions for optimizing the extraction process. The results obtained in this work can be applied in industry, contributing to the development of pharmaceutical and food products.

Dissemination of results

List of published works

- **Drăghici-Popa, A.-M.**; Boscornea, A. C.; Brezoiu, A.-M.; Tomas, S, .T.; Pârvulescu, O. C.; Stan, R. Effects of Extraction Process Factors on the Composition and Antioxidant Activity of Blackthorn (*Prunus spinosa* L.) Fruit Extracts. *Antioxidants* 2023, 12, 1897. https://doi.org/10.3390/ **(**IF at the date of publication – 6, ISI)
- **Drăghici-Popa, A.-M.**; Buliga, D.-I.; Popa, I.; Tomas, S.T.; Stan, R.; Boscornea, A.C. Cosmetic Products with Potential Photoprotective Effects Based on Natural Compounds Extracted from Waste of the Winemaking Industry. *Molecules* 2024, 29, 2775. https://doi.org/10.3390/molecules29122775 (IF at the date of publication – 4.6, ISI)
- **Drăghici-Popa, A. M**., Pârvulescu, O. C., Stan, R., Brezoiu, A. M. Optimization of Ultrasound-Assisted Extraction of Phenolic Compounds from Romanian Blackthorn (Prunus spinosa L.) Fruits. *Antioxidants*, 2025, 14(6), 680. https://doi.org/10.3390/antiox14060680 (IF at the date of publication – 6.6, ISI)
- **Drăghici-Popa, A.-M**.; Boscornea, A.C.; Călinescu, I.; Gavrilă, A. I.; POPA, I. Extraction of Bioactive Compounds from Flaxseed Using Sequential Green Technology: Microwave and Ultrasound, *Scientific Bulletin,* Series B, 2025 (IF at the date of publication **–** 0.3, ISI)

International conferences

- RICCCE 23, **Ana-Maria Drăghici-Popa**, Adina Ionuța Gavrilă, Aurelian Cristian Boscornea, Ioana Popa, “Bioactive compounds extraction using sequential green technology: ultrasound and microwave”, Romania, 2024.
- RICCCE 23, Ioana Popa, **Ana-Maria Drăghici-Popa**, Adina Ionuța Gavrilă, Diana-Ioana Buliga, Aurelian Cristian Boscornea, “A promising solvent, fatty acid ethyl esters, for ultrasound-assisted extraction of liposoluble bioactive compounds”, Romania, 2024.
- GPMB 2024, Ioana Popa, **Ana-Maria Drăghici-Popa**, Adina Ionuța Gavrilă, Aurelian Cristian Boscornea, “Combined microwave and ultrasound technique as an efficient extraction method of bioactive compounds from flaxseed”, Italia, 2024.
- SICHEM, **Ana-Maria Drăghici-Popa**, Oana Cristina Pârvulescu, Aurelian Cristian Boscornea, Raluca Stan, Ana-Maria Brezoiu, Ștefan Theodor Tomas, “Superior valorization of forgotten berries – *Prunus spinosa* L. (blackthorn) ”, Romania 2024.
- PRIOCHEM XIXth Edition, **Ana-Maria Drăghici-Popa**, Diana-Ioana Buliga, Ioana Popa, Stefan Tomas, Aurelian Cristian Boscornea, “Photoprotective cosmetics products with polyphenolic extract from grape marc”, Romania, 2023.
- PRIOCHEM XVIIIth Edition, Diana-Ioana Buliga, Ioana Popa, Aurelian Cristian Boscornea, **Ana-Maria Drăghici-Popa**, București, “New cosmetic formulations obtained by applying integrated and sustainable bioeconomy approaches” (BBB PP-47) - PRIOCHEM-BOOK of ABSTRACTS-2022, Edited by: INCDCP - ICECHIM Bucharest, Romania, 2022

Bibliography

1. Dima, A. D., Pârvulescu, O. C., Mateescu, C., Dobre, T. (2020). Optimization of substrate composition in anaerobic co-digestion of agricultural waste using central composite design. *Biomass Bioenerg, 138*, 105602. doi:https://doi.org/10.1016/j.biombioe.2020.105602
2. Drăghici-Popa, A. M., Boscornea, A. C., Brezoiu, A. M., Tomas, S. T., Parvulescu, O. C., Stan, R. (2023). Effects of Extraction Process Factors on the Composition and Antioxidant Activity of Blackthorn (Prunus spinosa L.) Fruit Extracts. *Antioxidants, 12*(10). doi:10.3390/antiox12101897
3. Drăghici-Popa, A. M., Buliga, D. I., Popa, I., Tomas, S. T., Stan, R., Boscornea, A. C. (2024). Cosmetic Products with Potential Photoprotective Effects Based on Natural Compounds Extracted from Waste of the Winemaking Industry. *Molecules, 29*(12). doi:10.3390/molecules29122775
4. Drăghici-Popa, A. M., Pârvulescu, O. C., Stan, R., Brezoiu, A. M. (2025a). Optimization of Ultrasound-Assisted Extraction of Phenolic Compounds from Romanian Blackthorn (Prunus spinosa L.) Fruits. *Antioxidants*, 14(6), 680, doi.org/10.3390/antiox14060680
5. Drăghici-Popa, A.-M.; Boscronea, A.C.; Călinescu, I.; Gavrilă, A. I.; POPA, I. (2025b) Extraction of Bioactive Compounds from Flaxseed Using Sequential Green Technology: Microwave and Ultrasound, *Scientific Bulletin, Series B*
6. Gegiu, G., Branza, A. D., Bucur, L., Branza, A. D., Grigorian, M., Tache, T. Badea, V. (2015). Contributions to the antimicrobial and antifungal study of the aqueous extract of Prunus spinosa L. *FARMACIA, 63*.
7. Georgiev, V., Ananga, A. Tsolova, V. (2014). Recent advances and uses of grape flavonoids as nutraceuticals. *Nutrients, 6*(1), 391-415. doi:10.3390/nu6010391
8. Mojzer, E. B., Knez Hrncic, M., Skerget, M., Knez, Z., Bren, U. (2016). Polyphenols: Extraction Methods, Antioxidative Action, Bioavailability and Anticarcinogenic Effects. *Molecules, 21*(7). doi:10.3390/molecules21070901
9. Oancea, A.-G., Saracila, M., Vlaicu, P. A., Varzaru, I., Untea, A. E., Dragomir, C. (2024). Assessment of the Antioxidant Potential of Blackthorns and Hawthorns: Comparative Analysis and Potential Use in Ruminants' Nutrition. *Separations, 11*(9). doi:10.3390/separations11090275
10. Opriş, O., Soran, M.-L., Lung, I., Stegarescu, A., Guţoiu, S., Podea, R., Podea, P. (2021). Optimization of extraction conditions of polyphenols, antioxidant capacity and sun protection factor from Prunus spinosa fruits. Application in sunscreen formulation. *Journal of the Iranian Chemical Society, 18*(10), 2625-2636. doi:10.1007/s13738-021-02217-9
11. Shim, Y. Y., Gui, B., Wang, Y., Reaney, M. J. T. (2015). Flaxseed (Linum usitatissimum L.) oil processing and selected products. *Trends in Food Science Technology, 43*(2), 162-177. doi:10.1016/j.tifs.2015.03.001
12. Tahirović, A., Bašić, N., Čopra-Janićijević, A. (2018). Effect of solvents on phenolic compounds extraction and antioxidant activity of Prunus spinosa L. fruits. *Bulletin of the Chemists Technologists of Bosnia Herzegovina, 50*.
13. Vega, E. N., Gonzalez-Zamorano, L., Cebadera, E., Barros, L., Pires, Tcsp, Molina, A. K., da Silveira, T. F. F., Vidal-Diez de Ulzurrun, G., Tardio, J., Camara, M., Fernandez-Ruiz, V.,

Morales, P. (2025). Natural Food Colorant Obtained from Wild Berberis vulgaris L. by Ultrasound-Assisted Extraction: Optimization and Characterization. *Foods, 14*(2). doi:10.3390/foods14020183

14. Velickovic, J., Kostic, D., Stojanovic, G., Mitic, S., Mitic, M., Randjelovic, S., Djordjevic, A. (2014). Phenolic composition, antioxidant and antimicrobial activity of the extracts from Prunus spinosa L. fruit. *Hemijska industrija, 68*(3), 297-303. doi:10.2298/hemind130312054v
15. Wang, N., Liu, X., Ma, Y., Huang, X., Song, L., Guo, H., Sun, X., Sun, X., Hai, D., Zhao, P., Shen, Y. (2024). Identification of polyphenol extracts from flaxseed and study on its bacteriostatic mechanism. *Food Bioscience, 58*. doi:10.1016/j.fbio.2024.103618
16. Wani, T. A., Majid, D., Dar, B. N., Makroo, H. A., Allai, F. M. (2023). Utilization of novel techniques in extraction of polyphenols from grape pomace and their therapeutic potential: a review. *Journal of Food Measurement and Characterization, 17*(5), 5412-5425. doi:10.1007/s11694-023-02040-1
17. Yilmaz, Y., Toledo, R. T. (2004). Major flavonoids in grape seeds and skins: antioxidant capacity of catechin, epicatechin, and gallic acid. *Journal of agricultural and food chemistry, 52*(2), 255-260.