 https://doi.org/10.3311/PPch.20264

Periodica Polytechnica Chemical Engineering, 66(4), pp. 660–674, 2022

High-temperature Thin-layer Drying Kinetic of Cultivated and Wild Algerian Olive Leaves

Modeling and Effect on Oleuropein and Chlorophyll Contents

Aissa Boukhiar[1,2*], Salem Benamara[2], Yougourthane Bouchal[3], Kahina Touderte[2], Siham Messouidi[2]

[1] Research Laboratory in Food Technology, Faculty of Technology, University of Boumerdes, Frantz Fanon city, Boumerdes 35000, Algeria
[2] Department of Process Engineering, Faculty of Technology, University of Boumerdes, Frantz Fanon city, Boumerdes 35000, Algeria
[3] Department of Food Phytochemistry, Instituto de la Grasa (Spanish National Research Council, CSIC), Ctra. de Utrera km. 1, Pablo de Olavide University Campus, Building 46, 41013 Seville, Spain
* Corresponding author, e-mail: a.boukhiar@univ-boumerdes.dz

Received: 01 April 2022, Accepted: 20 June 2022, Published online: 30 August 2022

Abstract

Olive leaves (OLs) are well known for being rich in oleuropein, their main bioactive molecule which has recently been attracting great interest from the scientific community due to its antiviral properties, including Covid-19 disease. Furthermore, the high-temperature/short-time drying process has found applications for various plants and food processing, which might be also implemented for the drying of OLs. This study focuses on: 1. the mathematical modeling of thin-layer high-temperature-drying (HTD) kinetic of olive (*var. Chemlal and Oleaster*) leaves, and 2. the determination of HTD effect on some physicochemical properties (oleuropein, chlorophylls, and CIELab color parameters) of the dried olive leaves (DOLs). For this, four drying temperatures (100, 120, 140, and 160 °C) were applied. For comparison purposes, low-temperature DOL samples were also prepared. The obtained data have shown that among the tens tested mathematical models, the Midilli et al. model describes more correctly experimental data for all drying temperatures and for both olive leaf varieties (R^2 = 0.9960, SEE = 0.0085, RMSE = 0.0165 and χ^2 = 0.0006). Moreover, the results show that the HTD at 120 and 160 °C does not differ from freeze-drying in terms of oleuropein retention ($p < 0.05$), highlighting the technological interest in the high-temperature/short-time drying process. Considering the biological value of oleuropein, in particular its antiviral activity, the study deserves further investigation in order to elucidate certain questions such as the storability of DOLs, and their valorization as fortification ingredient in food and pharmaceutical formulations, evaluation in vitro of their biological activities, etc.

Keywords

high-temperature-drying, dried olive leaves, mathematical modeling, oleuropein, chlorophylls

1 Introduction

Olive trees (*Olea europaea*) are widespread in the countries of the Mediterranean basin: Algeria, Tunisia, Spain, Italy, Greece, and others. Depending on the variety, their fruits (olives) can be consumed as it is after debittering [1–3], and/or used for oil extraction [4, 5]. Concerning olive oil, it is well known to be the backbone of the Mediterranean diet [6–8].

The olive leaves (OLs) represent an important biomass generally used for animal feeding or burnt periodically on the fields. However, many scientific studies reveal their high richness in bioactive compounds known for their antioxidant, antimicrobial and antiviral properties, among others: oleuropein and derivatives (hydroxytyrosol, dimethyl-oleuropein, dihydro-oleuropéine, etc.), verbascoside, catechin, and rutin. All these considerations make the use of OLs very promising for food, pharmaceutical, and cosmetic applications [9–12]. In traditional medicine, they have been used since antiquity for the treatment of various physiological dysfunctions of the human body, such as fever, hypertension, malaria, diabetes, etc. [13–16].

With the advent of the Covid-19 pandemic, consumers, as well as scientists, have shown an unprecedented interest

Cite this article as: Boukhiar, A., Benamara, S., Bouchal, Y., Touderte, K., Messouidi, S. "High-temperature Thin-layer Drying Kinetic of Cultivated and Wild Algerian Olive Leaves: Modeling and Effect on Oleuropein and Chlorophyll Contents", Periodica Polytechnica Chemical Engineering, 66(4), pp. 660–674, 2022. https://doi.org/10.3311/PPch.20264

in medicinal plants, especially those with antiviral properties. In this context, oleuropein (oleuropeosides), the main compound of the olive leaves (but also present in the fruit and oil but in smaller quantities) with a concentration of up to 200 mg/g dry OL powder in some cases [17], has received special attention due to its anti-Covid-19 activity [18–24].

To facilitate the conservation and preparation of herbal teas and infusions, the dry form is preferred. In fact, the drying techniques for medicinal plants dehydration are numerous, but usually, solar drying (with or without direct exposure to solar radiation) and at low temperature (<100 °C) are the most commonly used conservation methods, especially on a small scale. However, although this method is not expensive, it has the disadvantage of being time-consuming and induces considerable losses of bioactive compounds, oleuropein in particular [12, 25]. This is mainly related to enzymatic alteration reactions (oxidation and/or hydrolysis) [12, 17]. This is what we have also observed in the case of black olives hot-air-dried in the temperature range of 25–75 °C [3]. Moreover, several authors have reported that drying at high temperatures (>100 °C) and for a short time maximizes the content of bioactive compounds in OLs and increases the antioxidant activity of the final product [14, 25].

From a technological point of view, there are a few differences between high-temperature-drying and roasting processes. The purpose of drying is to remove water, while the main objective of roasting is to improve the sensory properties (color, aroma, and taste) [26, 27]. In the present manuscript, the two terminologies (high-temperature-drying and roasting) are used interchangeably.

According to some previous studies, the high-temperature-drying of OLs allows both rapid reduction of the water activity and the inhibition of the enzymes present in the plant matrix [12, 25, 28]. Add to that, this treatment is very effective in preserving bioactive compounds. However, as far as we know, the kinetic of high-temperature-drying of OLs is not reported in the literature.

This present study focuses on:

1. the mathematical modeling of thin-layer high-temperature-drying (HTD) of olive (*var. Chemlal and Oleaster*) leaves, and
2. the determination of HTD effect on some physicochemical properties (oleuropein, chlorophylls, and CIELab color parameters).

For this, four drying temperatures (100, 120, 140, and 160 °C) were applied. For comparison purposes, low-temperature dried olive leave samples were also prepared.

2 Material and methods

2.1 Plant material

The OLs from cultivated olive trees (*Chemlal* variety) (COL) and wild olive trees (*Oleaster*) (WOL) were hand-picked during the period of March-April 2015 from an olive grove in the Bouira region (Northern Algeria). After sorting, samples were maintained at 4 °C until use. In all cases, the samples were processed in less than three days.

2.2 Physical properties

The determination of different dimensions of studied OLs was performed on 20 randomly selected fresh leaves of each variety. Their linear dimensions (length, width, and thickness) and weight were determined by using a caliper (accuracy of 0.01 mm) and electronic balance (accuracy of 0.0001 g), respectively.

The water content of the fresh OLs was gravimetrically determined at 105 °C according to the procedure described by Idoui and Bouchefra [29].

2.3 Drying kinetics

The Thin-layer drying experiments were conducted at different temperatures (100, 120, 140, and 160 °C) using a laboratory static-oven dryer (Memmert, Germany). During the drying process, the samples were weighed periodically until the difference between two successive weighings is lower than 0.001 g (equilibrium state). There were three replications of each temperature for both COL and WOL.

To study the drying kinetics, the variation of moisture ratio (MR) versus time was analyzed by following the weight loss (water loss) moisture ratio (MR) was calculated using Eq. (1):

$$\mathrm{MR} = \frac{M_t - M_{eq}}{M_0 - M_{eq}} = \frac{W_t - W_{eq}}{W_0 - W_{eq}}, \tag{1}$$

where M_0, M_t, and M_{eq} were the initial, at time t and equilibrium water content of OL sample, respectively, W_0, W_t, and W_{eq} were the initial, at time t and equilibrium weight of OL sample, respectively.

The water content at any time (M_t) can be deduced as follows:

$$M_t = M_0 - \left(W_0 - W_t\right). \tag{2}$$

The experimental data were analyzed by applying ten mathematical models widely applied in food drying operations (Table 1) [30–36]. Data analysis was performed by non-linear regression method with statistical software, Statistica 6.0. The goodness of fit of the tested models to the experimental data was evaluated by five error

Table 1 Mathematical models applied to the thin-layer drying kinetic

N°	Model name	Equation	References
1	Wang and Singh	$MR = 1 + a\,t + b\,t^2$	[30]
2	Newton	$MR = \exp(-k\,t)$	[31]
3	Henderson and Pabis	$MR = a\exp(-k\,t)$	[32]
4	Logarithmic	$MR = a\exp(-k\,t) + c$	[33]
5	Page	$MR = \exp(-k\,t^n)$	[34]
6	Modified Page	$MR = \exp(-(k\,t)^n)$	[34]
7	Diffusion approach	$MR = a\exp(-k\,t) + (1-a)\exp(-k\,b\,t)$	[33]
8	Verma et al.	$MR = a\exp(-k\,t) + (1-a)\exp(-g\,t)$	[35]
9	Midilli et al.	$MR = a\exp(-k\,t^n) + b\,t$	[36]
10	Demir et al.	$MR = a\exp(-kt)^n + b$	[34]

MR: moisture ratio; t: time; k: drying rate constant; a, b, c, g, n: constants

parameters [37]: R^2 (coefficient of determination) and SEE (Standard Error of Estimate) were obtained directly from the Statistica 6.0 software. RMSE (root means squared error), SSE (sum squared error), and χ^2 (reduced Chi-square) were calculated using the following formula:

$$\mathrm{RMSE} = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\mathrm{MR}_{exp,i} - \mathrm{MR}_{pre,i}\right)^2} \tag{3}$$

$$\mathrm{SSE} = \sqrt{\sum_{i=1}^{N}\left(\mathrm{MR}_{exp,i} - \mathrm{MR}_{pre,i}\right)^2} \tag{4}$$

$$X^2 = \frac{\sum_{i=1}^{N}\left(\mathrm{MR}_{exp,i} - \mathrm{MR}_{pre,i}\right)^2}{N - z}, \tag{5}$$

where $\mathrm{MR}_{pre,i}$: predicted by the model MR, $\mathrm{MR}_{exp,i}$: experimental MR, N: number of data points, and z: number of model constants.

A model is considered more adequate when R^2 values are higher (closer to 1) and SEE, RMSE, SSE, and X^2 values are lower (closer to 0).

2.4 Effective diffusivity coefficient and activation energy

The diffusivity coefficient governs the drying rate in a homogeneous and isotropic solid. This coefficient is affected by several parameters, in particular the water content, temperature, and physical properties of the matrix [36]. In a real product, the diffusivity coefficient takes into account various water delivery mechanisms (molecular diffusion, capillary flow, etc.) and it is then called the "effective diffusion coefficient". Effective diffusivity coefficients for each drying temperature were determined from the slope of the straight line, obtained by plotting ln(MR) against drying time [38].

An analytical solution of Fick's second law equation for an infinite slab (Eq. (6)) was used to estimate the apparent moisture diffusivity of the OLs from the high-temperature-drying kinetics [39].

$$\ln \mathrm{MR} = \ln\left(\frac{8}{\pi^2}\right) - \left(\frac{\pi^2 D_{eff}}{(e/2)^2}\right)t \tag{6}$$

Where D_{eff}, e, and t are the effective diffusivity (m^2/s), thickness of the olive leaf (m), and the drying time (min), respectively.

By plotting ln(MR) against drying time, a straight line is obtained and the effective moisture diffusivity is calculated as

$$\mathrm{Slope} = -\left(\frac{\pi^2 D_{eff}}{(e/2)^2}\right) \Rightarrow D_{eff} = -\frac{(e/2)^2}{\pi^2}\mathrm{Slope}\,. \tag{7}$$

The activation energy for the water diffusion during high-temperature-drying of OLs was determined based on Arrhenius-type equation [38]:

$$\ln D_{eff} = \ln D_0 - \frac{E_a}{R_g}\frac{1}{T_a}\,. \tag{8}$$

By plotting $\ln D_{eff}$ against $1/T_a$, a straight line is obtained and the activation energy is calculated as

$$\mathrm{Slope} = -\frac{E_a}{R_g} \Rightarrow E_a = -\frac{\mathrm{Slope}}{R_g}, \tag{9}$$

where D_0, E_a, R_g, and T_a are the constant equivalent to the diffusivity at infinitely high temperature (m^2/s) pre-exponential factor (or Arrhenius constant), activation energy (kJ/mol), universal gas constant (8.314 J/mol K) and absolute temperature (K), respectively.

2.5 Determination of physical chemical properties of DOLs

For comparison purposes, the OLs of both studied varieties were processed by two other drying methods:

1. blanching followed by drying at 60 °C and
2. freeze-drying.

Briefly, the blanched OLs were pretreated by soaking in boiling water for 2.5 min and then dried using a Memmert static oven at 60 °C until a constant weight was reached. Furthermore, the freeze-dried samples were prepared by using Cryodos-50 Telstar laboratory freeze dryer (temperature/vacuum pressure of −45 °C/0.44 mbars) without any pretreatment. The obtained samples were taken here as controls.

The prepared dried olive leaves (DOLs) samples are coded according to Table 2. They were analyzed for oleuropein content, chlorophylls content, and CIELab color indices. The leaf samples resulting from the different drying methods were powdered using a commercial grinder. Then, the obtained powders were stored at −20 °C until further analysis.

2.5.1 Oleuropein content

The oleuropein content was determined by spectrophotometry according to Amiot et al. [40] method as described by Hurtado et al. [41] (with some minor modifications). About 0.1 g (W) of leaf powder (DOL) was macerated in 20 mL of distilled water for 3 hours, at room temperature (~25 °C) and sheltered from light. After filtration, using Whatman filter paper grade 4, the absorbance (A) of the extract was measured, using a UV-V is spectrophotometer (Jasco) at two wavelengths of 280 nm (oleuropein + verbascoside) and 330 nm (verbascoside).

The oleuropein content was calculated according to Eq. (10):

$$\text{Oleuropein}\left(\text{g}/100\ \text{g DOL}\right) = \frac{A_{280} - 0.9A_{330}}{75W} FV\,, \qquad (10)$$

where V: volume of solution (mL), and F: dilution factor.

Table 2 Coding of DOLs samples

Drying method	COL	WOL
Drying 100 °C	C-100 °C	O-100 °C
Drying 120 °C	C-120 °C	O-120 °C
Drying 140 °C	C-140 °C	O-140 °C
Drying 160 °C	C-160 °C	O-160 °C
Lyophilizing	C-lyophilized	O-lyophilized
Blanching/drying 60 °C	C-blanched	O-blanched

2.5.2 Chlorophylls content

Determination of chlorophyll a and chlorophyll b content of DOL was carried out according to Huang et al. [42]. Briefly, about 0.1–0.2 g (W) of DOL was extracted by 50 mL (V) of 80% (v/v) acetone for 2 min and filtered. The absorption values of the filtrate were determined spectrophotometrically at two wavelengths of 663 nm (chlorophyll a) and 645 nm (chlorophyll b). The chlorophyll a and the chlorophyll b content in olive leave samples are calculated according to the following formulas:

$$C_a\left(\text{mg/g DOL}\right) = \frac{12.7A_{663} - 2.95A_{645}}{1000W} V\,,$$

$$C_b\left(\text{mg/g DOL}\right) = \frac{22.9A_{645} - 4.67A_{663}}{1000W} V\,,$$

with A_{663}: absorbance at 663 nm, A_{645}: absorbance at 645 nm, W: weight of sample extracted (g), V: final volume (mL) of extract.

The total chlorophylls (a + b) is given by the formula:

$$C_t\left(\text{mg/g DOL}\right) = C_a + C_b\,.$$

2.5.3 Color determination

The color measurements were quantified by using a Minolta color reader (CR10, Japan). The L^*, a^*, and b^* values were determined for each sample (DOL): lyophilized, blanched/dried, and dried (100, 120, 140, and 160 °C). The color values are expressed as: L^* ranging from 0 (dark) to 100 (white), a^* ranging from −60 (red) to +60 (green), and b^* ranging from −60 (blue) to +60 (yellow).

2.6 Statistical analysis

All analyses were done in triplicate and results were expressed as mean ±SD. Data were analyzed for differences between means using one-way analysis of variance (ANOVA) and Tukey's post-hoc test, with statistical significance when $p < 0{,}05$, using Xlstat 2014 software. In addition, a multivariate statistical analysis focused on principal component analysis (PCA) and hierarchical analysis clustering (HAC) was performed using the same software, to extract linear relationships among the variables studied, and to compare obtained DL samples based on their dissimilarities. In order to facilitate the reading of the graphs, the parameters taken into account in this study were codified as shown in Table 3.

Table 3 Coding of parameters

Parameter	Code
Oleuropein content	*Oleur*
Chlorophyll a	*Chl a*
Chlorophyll b	*Chl b*
Total chlorophylls	*Chl tot*
Color indices *L*-value	*L**
Color indices *a*-value	*a**
Color indices *b*-value	*b**
Drying time	Time
Weight loss	WL

2.7 Uncertainty analysis

Experimental error and uncertainty can be caused by several factors (instrument specification, instrument calibration, measurement condition, etc.) [43]. In the present study, the designated operating range and uncertainties of used instruments based on the manufacturer's data are given in Table 4.

Uncertainty analysis is an important tool to provide the quality of measurements. In the present study, uncertainty analysis was performed according to Monte-Carlo method using LNE-MCM software (version 2017) from the French National Metrology laboratory (LNE). The uncertainties of the principal determined parameters were: ±0.58% (moisture ratio), ±2.75% (oleuropein content) and ±0.51% (chlorophylls tot).

3 Results and discussion

3.1 Physical properties and water content of fresh olive leaves

The physical properties (length, width, thickness, and weight) as well as the water content of fresh leaves of COL and WOL are reported in Table 5.

From Table 5, it is easy to see the differences in lengths, thicknesses and water contents between COL and WOL leaves ($p < 0.05$). This result confirms the possibility of using the physical properties of the leaves to distinguish the oleaster from the olive varieties [44].

Table 4 Uncertainties in measurement of studied parameters

Instrument	Range	Uncertainties
Electronic balance (Sartorious CP 224S)	0 to 220 (g)	±0.001 g
Static heating oven (Mmmert unb400)	20 to 220 (°C)	±0.5 °C
UV-V spectrometer (Jasco V530)	−2.0 to 3.0 (*Abs*)	±0.002 *Abs*(0 to 0.5 *Abs*) ±0.004 *Abs*(0.5 to 1 *Abs*)
Glass vial	50 (mL)	±0.06 mL
	100 (mL)	±0.05 mL

Table 5 Physical characteristics and water content of fresh olive leaves

Parameter	COL	WOL
Length (cm)	5.71 ± 0.90^a	3.66 ± 0.73^b
Width (cm)	0.78 ± 0.19^a	1.06 ± 0.45^a
Thickness (mm)	0.51 ± 0.10^a	0.45 ± 0.16^b
Weight (g)	0.164 ± 0.074^a	0.157 ± 0.058^a
Water content (g/100g, wb*)	54.70 ± 0.71^a	43.47 ± 1.52^b

Different superscripts in the same column indicate significant difference at $p < 0.05$.
* wb: wet basis

Regarding the water content, the obtained values are comparable to those (from 46.24 to 49.75%) reported by Boudhrioua et al. [39] for Tunisian olive varieties (*Chemlali*, *Chetoui*, *Chemchali*, and *Zarrazi*).

Because of its relatively high-water content, OLs cannot be stored for a long period, which makes necessary a conservation treatment, drying at high temperatures, among others.

3.2 Drying kinetic

The thin-layer drying kinetic curves of COL (Fig. 1 (a)) and WOL (Fig. 1 (b)) at different temperatures are shown in Fig. 1. From these data, a higher drying temperature resulted in a significantly lower drying time. Thus, passing from 100 to 160 °C may reduce the drying time by ~3.5 times.

On the other hand, it should be noted that although the water content of the WOL is the lowest, compared to that of COL, the time required to reach equilibrium is longer. For his part, Hata [12] reported a drying time of 72 h when OLs are dried at 25 °C or freeze-dried. It is 18 h for temperatures between 50 and 65 °C, and 3 h for 70 °C.

The results of the thin-layer modeling of OLs are reported in Table 6. As can be seen, except Wang and Sing model, all other tested models seem to be appropriate for describing the thin-layer drying curves of COL and WOL. However, the Midilli et al. model provides the best fit to the experimental data (with mean values of $R^2 = 0.996$, SEE = 0.0085, RMSE = 0.0165, SSE = 0.0006, and $\chi^2 = 0.0697$). This finding is in concordance with previous studies: thin-layer infrared drying of wet olive husk at temperatures between 80 and 140 °C [34], thin-layer microwave drying of celery leaves [45], thin layer drying of sour cherry in a solar dryer [46], etc.

The obtained parameter values of the Midilli et al. model are presented in Table 7.

3.3 Effective moisture diffusivity and activation energy

The values of effective moisture diffusivity (D_{eff}) and activation energy (E_a) deduced from the linearized Arrhenius equation-type of OLs are recapitulated in Table 8.

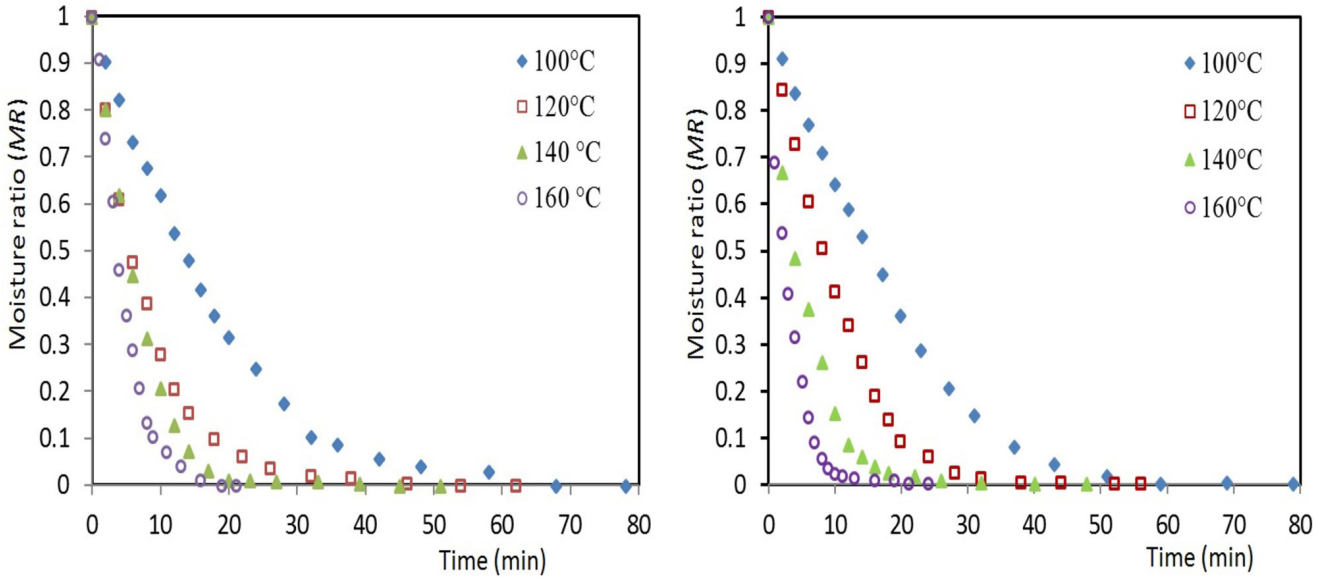

Fig. 1 Thin-layer drying curves versus temperature: COL (a), WOL (b)

Table 6 Thin-layer modeling results

		COL				WOL				
Model name	Error parameters	100 °C	120 °C	140 °C	160 °C	100 °C	120 °C	140 °C	160 °C	Mean*
1. Wang and Singh	R^2	0.9730	0.7703	0.8228	0.9780	0.9778	0.9453	0.8439	0.7954	0.8883
	SEE	0.0545	0.3402	0.2748	0.0358	0.0507	0.0968	0.3634	0.2792	0.1869
	RMSE	0.0522	0.1458	0.1310	0.0489	0.0503	0.0733	0.1421	0.1282	0.0965
	χ^2	0.0030	0.0243	0.0196	0.0028	0.0028	0.0060	0.0227	0.0186	0.0125
	SSE	0.2334	0.5833	0.5242	0.1892	0.2251	0.3111	0.6028	0.5284	0.3997
2. Newton	R^2	0.9919	0.9987	0.9883	0.9810	0.9859	0.9901	0.9698	0.9960	0.9877
	SEE	0.0163	0.0019	0.0182	0.0309	0.0321	0.0175	0.0704	0.0055	0.0241
	RMSE	0.0285	0.0110	0.0337	0.0454	0.0401	0.0312	0.0625	0.0179	0.0338
	χ^2	0.0009	0.0001	0.0012	0.0022	0.0017	0.0010	0.0041	0.0003	0.0014
	SSE	0.1277	0.0440	0.1349	0.1759	0.1793	0.1325	0.2653	0.0739	0.1417
3. Henderson and Pabis	R^2	0.9937	0.9989	0.9906	0.9875	0.9890	0.9919	0.9711	0.9961	0.9899
	SEE	0.0127	0.0016	0.0146	0.0203	0.0252	0.0143	0.0673	0.0054	0.0202
	RMSE	0.0252	0.0099	0.0302	0.0368	0.0355	0.0282	0.0612	0.0178	0.0306
	χ^2	0.0007	0.0001	0.0010	0.0016	0.0014	0.0009	0.0042	0.0004	0.0013
	SSE	0.1129	0.0396	0.1207	0.1425	0.1586	0.1196	0.2595	0.0734	0.1284
4. Logarithmic	R^2	0.9959	0.9990	0.9921	0.9906	0.9930	0.9943	0.9723	0.9964	0.9917
	SEE	0.0083	0.0015	0.0122	0.0153	0.0161	0.0100	0.0645	0.0049	0.0166
	RMSE	0.0204	0.0098	0.0276	0.0309	0.0283	0.0236	0.0598	0.0171	0.0272
	χ^2	0.0005	0.0001	0.0009	0.0013	0.0009	0.0007	0.0043	0.0004	0.0011
	SSE	0.0910	0.0393	0.1105	0.1237	0.1267	0.1001	0.2539	0.0703	0.1144
5. Page	R^2	0.9983	0.9994	0.9994	0.9991	0.9976	0.9981	0.9782	0.9960	0.9958
	SEE	0.0034	0.0009	0.0009	0.0015	0.0056	0.0033	0.0508	0.0054	0.0090
	RMSE	0.0131	0.0076	0.0075	0.0098	0.0167	0.0136	0.0531	0.0178	0.0174
	χ^2	0.0002	0.0001	0.0001	0.0001	0.0003	0.0002	0.0032	0.0004	0.0006
	SSE	0.0586	0.0306	0.0301	0.0381	0.0746	0.0577	0.2253	0.0736	0.0736

Table 6 Thin-layer modeling results (continued)

		COL				WOL				
Model name	Error parameters	100 °C	120 °C	140 °C	160 °C	100 °C	120 °C	140 °C	160 °C	Mean*
6. Modified Page	R^2	0.9983	0.9994	0.9994	0.9991	0.9976	0.9981	0.9782	0.9960	0.9958
	SEE	0.0034	0.0009	0.0009	0.0015	0.0056	0.0033	0.0508	0.0054	0.0090
	RMSE	0.0131	0.0076	0.0075	0.0098	0.0167	0.0136	0.0531	0.0178	0.0174
	χ^2	0.0002	0.0001	0.0001	0.0001	0.0003	0.0002	0.0032	0.0004	0.0006
	SSE	0.0586	0.0306	0.0301	0.0381	0.0746	0.0577	0.2253	0.0736	0.0736
7. Diffusion approach	R^2	0.9937	0.9989	0.9906	0.9875	0.9890	0.9919	0.9711	0.9961	0.9899
	SEE	0.0127	0.0016	0.0146	0.0203	0.0252	0.0143	0.0673	0.0054	0.0202
	RMSE	0.0252	0.0099	0.0302	0.0356	0.0355	0.0282	0.0612	0.0178	0.0305
	χ^2	0.0007	0.0001	0.0011	0.0017	0.0015	0.0010	0.0045	0.0004	0.0014
	SSE	0.1129	0.0396	0.1207	0.1425	0.1586	0.1196	0.2595	0.0734	0.1284
8. Verma et al.	R^2	0.9987	0.9989	0.9993	0.9993	0.9940	0.9957	0.9740	0.9960	0.9945
	SEE	0.0027	0.0016	0.0011	0.0011	0.0138	0.0076	0.0606	0.0055	0.0118
	RMSE	0.0116	0.0099	0.0082	0.0084	0.0262	0.0205	0.0580	0.0179	0.0201
	χ^2	0.0002	0.0001	0.0001	0.0001	0.0008	0.0005	0.0040	0.0004	0.0008
	SSE	0.0517	0.0395	0.0329	0.0337	0.1173	0.0870	0.2462	0.0739	0.0853
9. Midilli et al.	R^2	0.9986	0.9994	0.9995	0.9992	0.9984	0.9984	0.9783	0.9963	0.9960
	SEE	0.0028	0.0009	0.0008	0.0013	0.0037	0.0028	0.0506	0.0051	0.0085
	RMSE	0.0119	0.0075	0.0072	0.0091	0.0135	0.0124	0.0530	0.0173	0.0165
	χ^2	0.0002	0.0001	0.0001	0.0001	0.0002	0.0002	0.0036	0.0004	0.0006
	SSE	0.0532	0.0299	0.0289	0.0365	0.0605	0.0526	0.2249	0.0714	0.0697
10. Demir et al.	R^2	0.9959	0.9990	0.9921	0.9906	0.9930	0.9943	0.9723	0.9964	0.9917
	SEE	0.0083	0.0015	0.0122	0.0153	0.0161	0.0100	0.0645	0.0049	0.0166
	RMSE	0.0204	0.0098	0.0276	0.0319	0.0283	0.0236	0.0598	0.0171	0.0273
	χ^2	0.0005	0.0001	0.0010	0.0014	0.0010	0.0007	0.0046	0.0004	0.0012
	SSE	0.0910	0.0393	0.1105	0.1237	0.1267	0.1001	0.2539	0.0703	0.1144

* Mean value for all drying temperatures for both varieties

Table 7 Constants and coefficients of Midilli et al. model

		COL				WOL			
Model name	Constants	100 °C	120 °C	140 °C	160 °C	100 °C	120 °C	140 °C	160 °C
Midilli et al.	a	0.9810	1.0022	0.9921	1.0071	0.9692	0.9812	0.9907	0.9857
	k	0.0301	0.1101	0.0775	0.1183	0.0190	0.0507	0.0929	0.3005
	n	1.2100	1.0632	1.3110	1.3414	1.3254	1.2471	1.2880	1.0256
	b	0.0000	0.0001	0.0000	0.0002	−0.0001	−0.0001	0.0001	−0.0002

Table 8 Effective moisture diffusivity and activation energy of *COL* and *WOL*

Variety	T (°C)	D_{eff} (× 10^{12}, m^2/s)	R^2	E_a (kJ/mol)	D_0 (× 10^7, m^2/s)	R^2
COL	100	6.364	0.958	440.113	1.250	0.970
	120	12.092	0.988			
	140	19.730	0.944			
	160	24.185	0.944			
WOL	100	6.162	0.902	444.888	1.209	0.991
	120	9.756	0.956			
	140	14.376	0.980			
	160	25.159	0.936			

Effective moisture diffusivity of the OLs varies, respectively, from 6.36 × 10^{-12} to 2.418 × 10^{-11} and from 6.162 × 10^{-12} to 2.52 × 10^{-11} m^2/s for COL and WOL. Globally, increasing the temperature from 100 to 160 °C increases the effective moisture diffusivity by three times for both varieties, confirming the considerable temperature effect on drying kinetics.

The graph giving $\ln(D_{eff})$ as a function of $1/T_a$ is a straight line in the range of studied temperatures of both varieties, indicating Arrhenius type dependence between D_{eff} and T_a (Fig. 2). The obtained activation energy values are 440.11 kJ/mol (COL) and 444.89 kJ/mol (WOL).

3.4 Physical chemical properties of DOLs

3.4.1 Oleuropein content

To begin, it must be remembered that the Oleuropein (Fig. 3) is an ester (hydroxytyrosol + elenolic acid) belonging to the family of secoiridoid polyphenols [13]. This molecule and its derivatives are the main bioactive compounds of the olive tree products: leaves, fruits, twigs, and oil, and are responsible for therapeutic and preventive virtues, widely described in the scientific literature [47, 48]. Oleuropein and some of its derivatives are the main compounds responsible for the bitterness of leaves, olives, and even oil. It should also be noted that this molecule is water-soluble and heat resistant [12].

The oleuropein content at the end of the processing of DOLs are shown in Table 9.

The oleuropein content in freeze-dried leaves of the COL (7.71 g/100 g) is comparable to that found in WOL (7.87 g/100 g) ($p < 0.05$). It should be recalled that the extraction rate could be sensibly enhanced if the extraction was performed-with hydroalcoholic solutions [12]. In all cases, our results are comparable to those (6 to 9 g/100 g of dry matter) reported by Achat et al. [47] but lower than those (9–13% of Powdered Leaves/of dry matter) communicated by Savournin et al. [49] concerning the OLs of 14 Tunisian (*Bid el Haman*, *Chemlali*, *and Meski*) and French (*Aglandau, Cailletier, Cayet Rouge, Cayon, Grossanne, Lucques, Picholine, Picholine Noire, Tanche, Verdale de l'Herault, Verdale Picholine hybrid*) varieties, the latter authors having used an aqueous-methanolic extraction. Globally, the differences in the oleuropein composition can be attributed to various factors including extraction and quantifying method [25, 50].

It is well known that freeze-drying can ensure better preservation of the raw material properties and that its applications on a large scale are limited due to its high cost. Therefore, it is more reserved for the drying of thermo-sensitive substances. Compared to freeze-dried DOLs, HTD procedures have induced a significant decrease in the oleuropein content of OLs ($p < 0.05$) whatever the variety and the preparation conditions, except for DOLs obtained at 160 °C, where the drying time is shorter indicating the technological interest of high-temperature/short-time drying process. For both olive varieties, the lowest contents

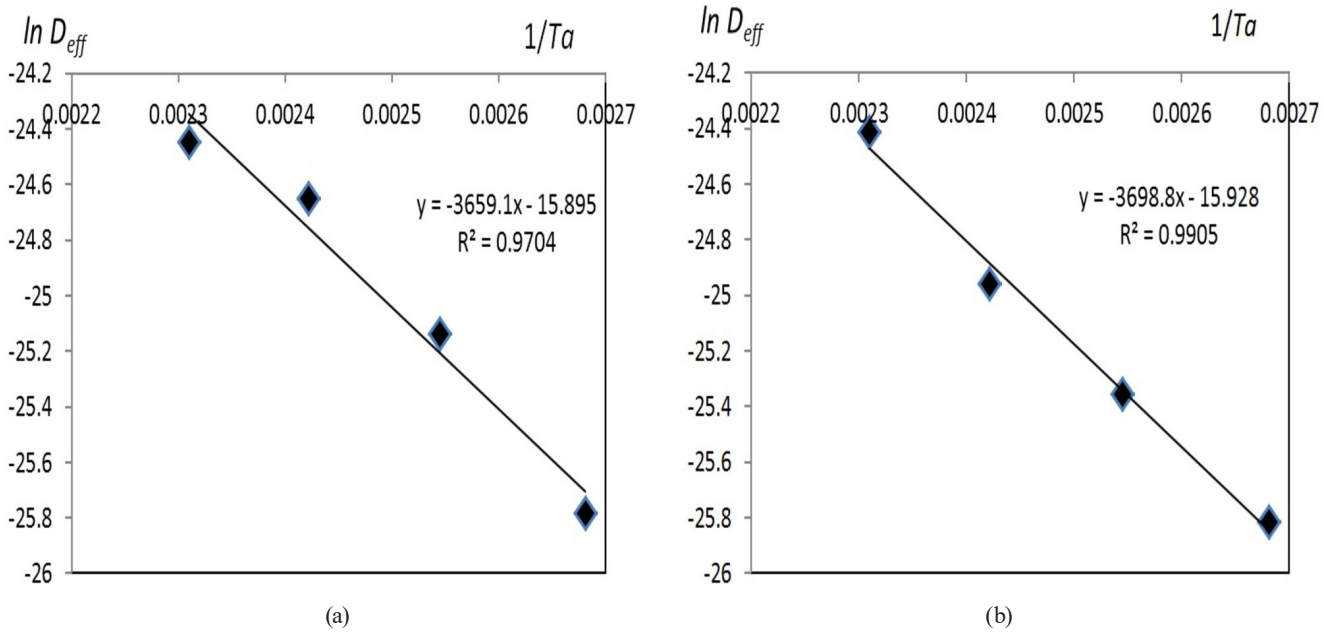

Fig. 2 Arrhenius type relationship between moisture diffusivity (D_{eff}) and absolute temperature (*Ta*) of COL (a) and WOL (b)

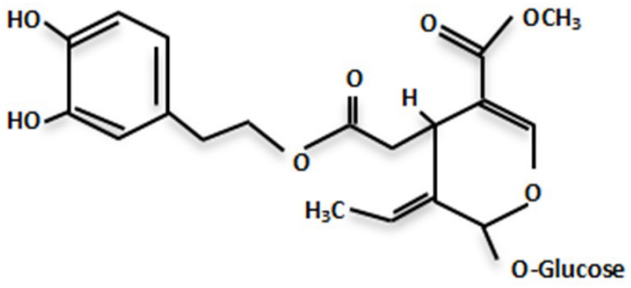

Fig. 3 Oleuropein

Table 9 Oleuropein content of studied DOLs

Preparation method	Oleuropein content (g/100g DOL)	
	COL	WOL
Lyophilized	7.71 ± 1.26^{a}	7.87 ± 0.56^{a}
Blanched/dried 60 °C	5.77 ± 0.35^{b}	$5.47 \pm 0.05^{c,d}$
Dried 100 °C	3.97 ± 0.2^{c}	5.00 ± 0.35^{d}
Dried 120 °C	$6.32 \pm 0.07^{a,b}$	$5.33 \pm 0.20^{c,d}$
Dried 140 °C	5.90 ± 0.20^{b}	$6.26 \pm 0.45^{b,c}$
Dried 160 °C	$7.17 \pm 0.07^{a,b}$	$7.15 \pm 0.46^{a,b}$

Different superscripts in the same column indicate significant difference at $p < 0.05$.

are recorded during the longest drying time at low temperatures, in particular at 100 °C, and this for both varieties. This decrease is of the order of 36 to 50% compared to freeze-dried DOLs. On the other hand, except for the DOLs obtained at 120 and 160 °C (case of WOL), there is no significant difference between samples dried at the temperatures 120, 140 and 160 °C.

In our opinion, the fluctuations observed in the effect of temperature on the oleuropein content could only be explained by taking into account the effect of the time-temperature couple, in particular the activation energy linked to the degradation reaction of this molecule. In addition, the much lower value (3.97 g/100 g DOL), obtained at 100 °C in the case of COL, is probably also related to the initial water content and the thickness of leaves which are relatively higher in this variety.

Indeed, several authors have already highlighted the thermoresistance of oleuropein and its vulnerability to hydrolytic and oxidative enzymes [12, 51, 52]. In the case of olive fruits of the *Manzanilla* variety (green-yellow color on the surface), Garcia et al. [52] have found good correlation between oleuropein content and enzymatic browning. In addition, the chemical and enzymatic degradation reactions of oleuropein lead principally to an improvement in content of hydroxytyrosol, whose virtues on the human body are widely also described in the scientific literature, namely antimicrobial, hypoglycemic, hypolipidemic, hypocholesterolic and antioxidant [13, 28].

Regarding the effect of temperature on the intensity of oleuropein degradation, there are various opinions in the literature. Al Juhaimi et al. [50] highlighted the heat sensitivity of polyphenols of OLs, among which oleuropein. Ahmad-Qasem et al. [25] have underlined the interest of HTD in increasing the rate of water removal from the plant matrix. These same authors have recalled the direct and/or indirect (by reducing water activity) thermal inactivation of polyphenol oxidase. Mostly, the complete inactivation of this enzyme is observed at higher temperatures exceeding 70 °C [53].

Concerning the blanched DOLs, for which the raw leaves were pretreated in a boiling water bath before drying at 60 °C in order to denature the endogenous enzymes, the obtained results revealed a significant ($p < 0.05$) decrease in oleuropein content (25–30%). This diminution may be related to the release of oleuropein from the plant matrix into the soaking media during the blanching step.

Hata [12] has reported that drying of unblanched OLs at temperatures below 100 °C leads to significant losses of oleuropein up to 70%. This decrease is even more important when the drying is done at low temperatures as also highlighted by Boukhiar et al. [3] concerning the drying of black olive fruits at temperatures ranging from 25 to 75 °C.

For valorization purposes of OLs, as a potential source of oleuropein, recent scientific studies show that it is possible to use them, as-is or after extraction, to enrich various products for human consumption: olive oil [47], table olives [54], coffee [55], date powder tablets [56], etc.

3.4.2 Chlorophylls content

Numerous recent scientific studies show that chlorophylls contribute to the health benefits of medicinal plants [57]. However, their presence in the oil is undesirable because of its pro-oxidant effect in the presence of light [58].

The chlorophyll contents (*a*, *b*, and total) of processed DOLs are given in Table 10. From these results, the chlorophyll b content of all considered samples is about two times higher than that of chlorophyll a, regardless of variety and preparation method.

Moreover, as expected for both varieties, the chlorophyll a and b contents of the lyophilized DOLs were statistically higher ($p < 0.05$) than those of the other dried samples, indicating the degradation effect of hot-air drying on the pigments.

Table 10 Chlorophylls in studied DOLs

Preparation method	COL			WOL		
	Chlorophyll a (mg/100 g)	Chlorophyll b (mg/100 g)	Chlorophylls tot (mg/100 g)	Chlorophyll a (mg/100 g)	Chlorophyll b (mg/100 g)	Chlorophylls tot (mg/100 g)
Lyophilized	22.47 ± 0.34^{a}	40.02 ± 0.60^{a}	62.49 ± 0.95^{a}	18.43 ± 1.06^{a}	32.85 ± 1.89^{a}	51.28 ± 2.95^{a}
Blanched/dried 60 °C	15.68 ± 1.21^{b}	23.38 ± 1.73^{b}	39.06 ± 2.94^{b}	14.30 ± 1.57^{b}	25.50 ± 2.80^{b}	39.80 ± 4.36^{b}
Dried 100 °C	12.97 ± 0.54^{b}	23.12 ± 0.96^{b}	36.08 ± 1.49^{b}	6.20 ± 1.32^{d}	9.32 ± 1.97^{d}	15.52 ± 3.29^{d}
Dried 120 °C	8.77 ± 0.19^{c}	13.32 ± 0.26^{c}	22.08 ± 0.22^{c}	10.73 ± 0.43^{c}	19.18 ± 0.76^{c}	29.90 ± 1.19^{c}
Dried 140 °C	8.95 ± 1.52^{c}	15.95 ± 2.71^{c}	24.90 ± 4.23^{c}	$9.04 \pm 1.24^{c,d}$	$14.26 \pm 2.86^{c,d}$	$23.30 \pm 3.96^{c,d}$
Dried 160 °C	9.40 ± 1.43^{c}	13.77 ± 2.05^{c}	23.17 ± 3.47^{c}	10.46 ± 0.62^{c}	15.40 ± 0.90^{c}	25.86 ± 1.51^{c}

Different superscripts in the same column indicate significant difference at $p < 0.05$.

In fact, the effect of temperature on the degradation of chemical and biochemical compounds, including chlorophylls, is well described by the Arrhenius equation but this dependence is a function of the nature of the molecule and also the surrounding environment. Theoretically, it is also well known that an increasing temperature of 10 °C implies an increase in the alteration rate of 2–3 times.

In addition, it should be remembered that the role played by enzymes (peroxidase and chlorophyllase) in the deterioration of chlorophylls is important. In this context, Khaushal et al. [59] recommended a chemical alkali blanching using sodium bicarbonate (0.1%) for 10 seconds in *Colocasia Schott* leaves.

As mentioned previously, the physical and chemical changes induced by pyrolysis reactions during roasting depend directly on the treatment conditions (temperature and duration in particular) and material characteristics [27]. Concerning the chlorophylls, these reactions induce their conversion into pheophytins and pyropheophytins [60].

In the case of blanched samples, the decrease in chlorophyll contents can be attributed, in addition to the effect of enzymes and pyrolysis reactions, to the release of pigments in the soaking media as previously explained for oleuropein.

3.4.3 Color measurements

As known, color is a key quality factor of numerous food and non-food materials. It is generally considered to be the most decisive parameter for consumer acceptability and a useful tool for monitoring many food processes (roasting, drying, baking, etc.).

CIELab color parameters (L^*, a^*, and b^* values) of the prepared DOLs are presented in Table 11. As can be seen, there is a noticeable increase in browning degrees with increasing drying time and/or temperature.

Compared to the freeze-dried leaves whose color characteristics are closer to those of fresh OLs, drying induced significant decreases ($p < 0.05$) in L^* and a^* values for both leaf varieties indicating the effect of browning with a loss of green color. From our point of view, these changes could be associated with non-enzymatic browning reactions (also called Maillard reaction), caramelization, and degradation of chlorophylls (bright green color) into pheophytins and pyropheophytins (yellow-brown color) as already mentioned in the literature about OLs [60–62]. It is also worth noting that the products of the Maillard reaction would have antioxidant properties as reported by Lin et al. [62] for almond kernels. Regarding the b^* values, its variations are rather difficult to explain, depending, however, on both the variety and the operating conditions of preparation.

Table 11 Color parameters indices of studied DOLs

Preparation method	COL			WOL		
	L^* value	a^* value	b^* value	L^* value	a^* value	b^* value
Lyophilized	52.52 ± 0.66^{a}	-1.36 ± 0.70^{c}	$26.84 \pm 1.18^{b,c}$	54.42 ± 0.98^{a}	-3.60 ± 2.31^{b}	30.76 ± 1.17^{a}
Blanched/dried 60 °C	$46.22 \pm 1.93^{c,d}$	1.10 ± 0.90^{b}	28.52 ± 0.65^{a}	46.12 ± 1.67^{d}	0.72 ± 0.58^{a}	$29.34 \pm 1.06^{a,b}$
Dried 100 °C	46.02 ± 0.86^{d}	3.20 ± 0.39^{a}	23.20 ± 0.61^{d}	49.40 ± 1.25^{b}	2.16 ± 1.15^{a}	$30.65 \pm 1.14^{a,b}$
Dried 120 °C	$46.98 \pm 0.42^{b,c,d}$	3.24 ± 1.31^{a}	25.72 ± 0.92^{c}	$48.80 \pm 0.48^{b,c}$	0.76 ± 0.54^{a}	$28.90 \pm 0.38^{b,c}$
Dried 140 °C	$48.22 \pm 1.28^{b,c}$	$2.08 \pm 0.81^{a,b}$	$26.10 \pm 0.44^{b,c}$	$46.64 \pm 1.27^{c,d}$	-0.16 ± 0.68^{a}	27.14 ± 0.65^{c}
Dried 160 °C	48.74 ± 0.09^{b}	0.80 ± 0.44^{b}	$27.32 \pm 0.57^{a,b}$	$48.24 \pm 0.78^{b,c,d}$	2.08 ± 1.02^{a}	$30.22 \pm 0.65^{a,b}$

Different superscripts in the same column indicate significant difference at $p < 0.05$.

3.5 Multivariate statistical analysis

The results of PCA are presented in Figs. 4 and 5. The obtained results reveal that 72.18% of the total variability is explained by the first two components (*F*1: 49.24% and *F*2: 22.93%), while 16.64% of the variability is explained by the third component (*F*3).

From Fig. 4, it is easy to see that *Oleur*, *Chl a*, *Chl b*, *Chl tot*, and *L** are positively correlated with *F1* but negatively with *a**. On the other hand, *b** and WL are correlated with *F*2, while Time is correlated with *F*3. Moreover, the calculated Pearson correlation coefficient shows the existence of strong correlations ($R > 0.98$) between *Chl a*, *Chl b*, and *Chl tot*. For the oleuropein content (*Oleur*), it is positively correlated with *L** and negatively with Time and *a**. Concerning the color parameters, *a** is negatively correlated with *Oleur*, *Chl a*, *Chl b*, *Chl tot*, and *L**. In addition, *L** is positively correlated with *Oleur*, whereas *b** is correlated only with WL. At last, the Time parameter is not correlated with WL (best shown by the biplot of *F*2 versus *F*3 components, not presented here).

From Fig. 5, it is clearly observed that blanched, dried at 120 and at 140 °C DOL, of both olive varieties, present relatively similar characteristics because they are clustered together.

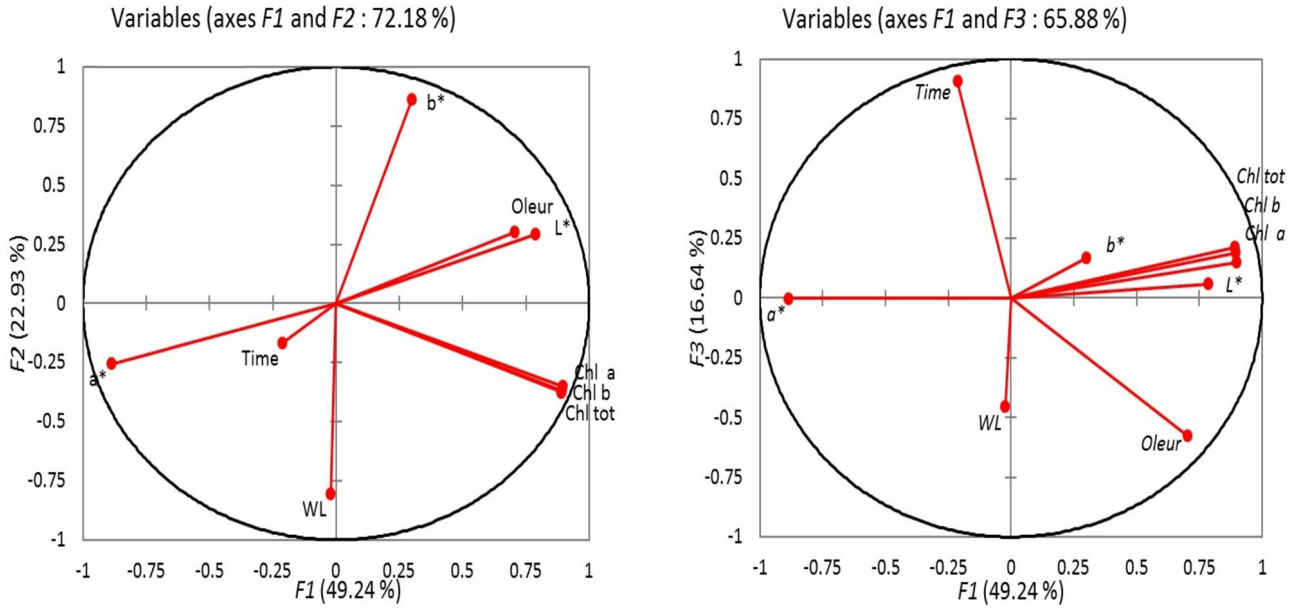

Fig. 4 Correlation plot between variables of DOLs

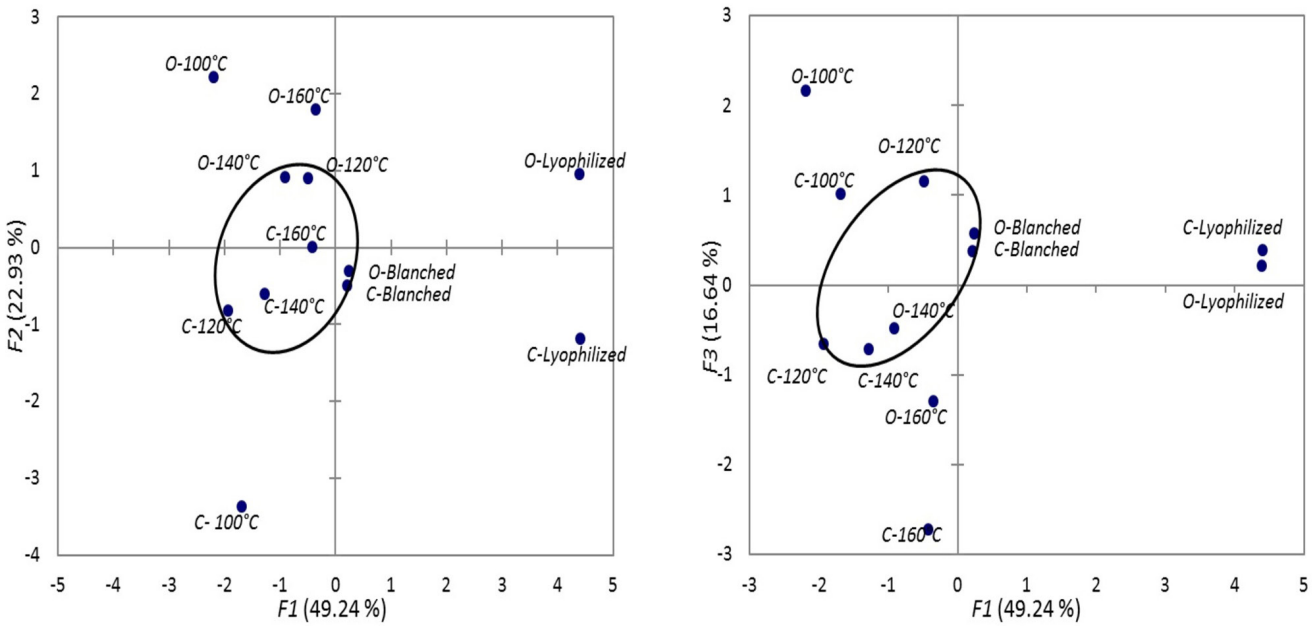

Fig. 5 Clustering of DOLs using PCA

As a complement to PCA, the dissimilarity dendrogram that was determined by hierarchical ascending classification (HAC), with Euclidean distance and Ward criterion, allows good visualization of the links between the different DOLs studied (Fig. 6).

It should be noted here that the two variables *Chl a* and *Chl b* are not taken into account, as they are strongly correlated with the *Chl tot*. In addition to the latter parameter, the variables considered here were *Oleur*, *L**, *a**, *b**, Time, and WL. Moreover, the missing values were replaced by the mean values (case of Time and WL of lyophilized and blanched DOLs).

It is clear at first sight that the varietal effect is negligible compared to the preparation method (Fig. 6).

Thus, four classes can be distinguishable:

- Class 1: DOLs dried at 160 °C;
- Class 2: DOLs lyophilized;
- Class 3: DOLs blanched, DOLs dried at 120 and at 140 °C;
- Class 4: DOLs dried at 100 °C.

These findings are in agreement with the PCA results presented above concerning:

1. the similarities of DOLs blanched, dried at 120 and at 140 °C, and
2. the neglecting of the varietal effect.

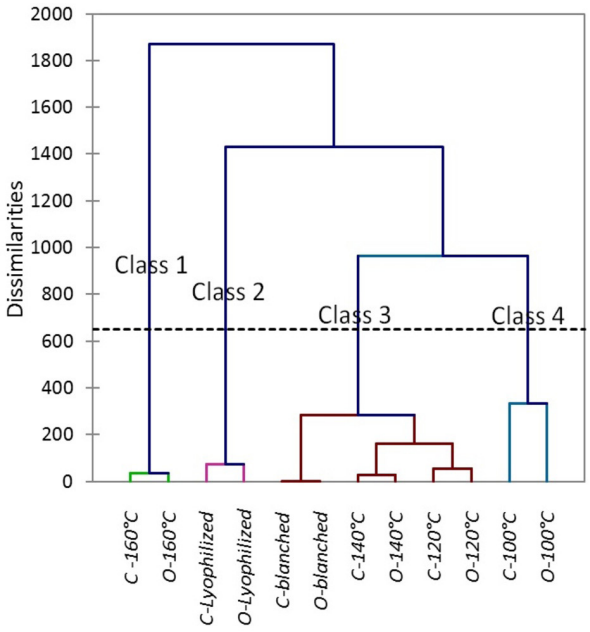

Fig. 6 Dendrogram of HAC Clusters

4 Conclusion

This study focuses on:

1. the mathematical modeling of high temperature thin layer drying (HTD) of olive (*var. Chemlal and Oleaster*) leaves, and
2. determination of HTD effect on some physicochemical properties (oleuropein, chlorophylls, and CIELab color parameters).

The obtained data have shown that among the tens tested mathematical models, that of Midilli et al. describes more correctly experimental data for all drying temperatures and for the both varieties. Moreover, the results show that the HTD at temperatures between 120 and 160 °C does not differ from freeze-drying in terms of oleuropein retention ($p < 0.05$), highlighting the technological interest of high-temperature/short-time drying process.

Considering the biological value of oleuropein, in particular its antiviral activity, the study deserves further investigation in order to elucidate certain questions such as the storability of DOL, their valorization as fortification ingredient in food and pharmaceutical formulations, evaluation in vitro of their biological activities, etc.

Acknowledgments

The authors would like to thank the following for their contributions to this work: Ait-Atmane Mouhoub for the language verification of the manuscript; Dr. Haderbache Latifa and Benantar Salah-Eddine for their helping in uncertainty analysis.

References

[1] Marsilio, V., Lanza, B., Pozzi, N. "Progress in table olive debittering: degradation *in vitro* of oleuropein and its derivatives by *Lactobacillus plantarum*", Journal of the American Oil Chemists' Society, 73(5), pp. 593–597, 1996. https://doi.org/10.1007/BF02518113

[2] Ramírez, E., Medina, E., García, P., Brenes, M., Romero, C. "Optimization of the natural debittering of table olives", LWT, 77, pp. 308–313, 2017. https://doi.org/10.1016/j.lwt.2016.11.071

[3] Boukhiar, A., Kechadi, K., Abdellaoui, R., Iguergaziz, N., Guemmane, M., Benamara, S. "Drying ability of whole black olive (*Olea europaea* L.) fruits in Kabylian region (North-East Algeria)", Indian Journal of Traditional Knowledge, 16(1), pp. 89–94, 2017. [online] http://nopr.niscpr.res.in/bitstream/123456789/37019/1/IJTK%2016%281%29%2089-94.pdf [Accessed: 01 June 2018]

[4] Uceda, M., Jiménez, A., Beltrán, G. "Olive oil extraction and quality", Grasas y Aceites, 57(1), pp. 25–31, 2006. https://doi.org/10.3989/gya.2006.v57.i1.19

[5] Tamborrino, A., Romaniello, R., Caponio, F., Squeo, G., Leone, A. "Combined industrial olive oil extraction plant using ultrasounds, microwave, and heat exchange: Impact on olive oil quality and yield", Journal of Food Engineering, 245, pp. 124–130, 2019. https://doi.org/10.1016/j.jfoodeng.2018.10.019

[6] Marongui, B., Özcan, M. M., Rosa, A., Assunta Dessi, M., Piras, A., Al Juhaimi, F. "Monitoring of the fatty acid compositions of some olive oils", La Rivista Italiana Delle Sostanze Grasse, 92(1), pp. 39–42, 2015. [online] https://iris.unica.it/handle/11584/144435 [Accessed: 25 May 2022]

[7] Juhaimi, F., Ghafoor, K., Adiamo, O. Q., Babiker, E. E. "Phenolic compounds and sterol contents of olive (*Olea europaea* L.) oils obtained from different varieties", Pakistan Journal of Botany, 49(1), pp. 169–172, 2017. [online] https://inis.iaea.org/search/searchsinglerecord.aspx?recordsFor=SingleRecord&RN=48093420 [Accessed: 25 May 2022]

[8] Özcan, M. M., Al Juhaimi, F., Uslu, N., Ghafoor, K., Ahmed, I. A. M., Babiker, E. E. "The Effect of Olive Varieties on Fatty Acid Composition and Tocopherol Contents of Cold Pressed Virgin Olive Oils", Journal of Oleo Science, 68(4), pp. 307–310, 2019. https://doi.org/10.5650/jos.ess18251

[9] Kesente, M., Kavetsou, E., Roussaki, M., Blidi, S., Loupassaki, S., Chanioti, S., Siamandoura, P., Stamatogianni, C., Philippou, E., Papaspyrides, C., Vouyiouka, S., Detsi, A. "Encapsulation of Olive Leaves Extracts in Biodegradable PLA Nanoparticles for Use in Cosmetic Formulation", Bioengineering, 4(3), 75, 2017. https://doi.org/10.3390/bioengineering4030075

[10] Rahmanian, N., Jafari, S. M., Wani, T. A. "Bioactive profile, dehydration, extraction and application of the bioactive components of olive leaves", Trends in Food Science & Technology, 42(2), pp. 150–172, 2015. https://doi.org/10.1016/j.tifs.2014.12.009

[11] Goldsmith, C. D., Vuong, Q. V., Stathopoulos, C. E., Roach, P. D., Scarlett, C. J. "Optimization of the Aqueous Extraction of Phenolic Compounds from Olive Leaves", Antioxidants, 3(4), pp. 700–712, 2014. https://doi.org/10.3390/antiox3040700

[12] Hata, S. "Effect of drying temperature on the oleuropein content of olive (*Olea europea* L.) leaves", Food Preservation Science, 30(4), pp. 191–193, 2004. https://doi.org/10.5891/jafps.30.191

[13] Özcan, M. M., Matthäus, B. "A review: benefit and bioactive properties of olive (*Olea europaea* L.) leaves", European Food Research and Technology, 243(1), pp. 89–99, 2017. https://doi.org/10.1007/s00217-016-2726-9

[14] Kamran, M., Hamlin, A. S., Scott, C. J., Obied, H. K. "Drying at high temperature for a short time maximizes the recovery of olive leaf biophenols", Industrial Crops and Products, 78, pp. 29–38, 2015. https://doi.org/10.1016/j.indcrop.2015.10.031

[15] Ghedira, K. "L'olivier" (Olivetree), Phytothérapie, 6(2), pp. 83–89, 2008. https://doi.org/10.1007/s10298-008-0294-2

[16] Akbaş, Ü. G., Uslu, N., Al Juhaimi, F., Özcan, M. M., Ghafoor, K., Babiker, E. E., Jamiu, F. G., Hussain, S. "The effect of drying on phenolic compound, antioxidant activity, and mineral contents of leaves of different olive varieties", Journal of Food Processing and Preservation, 42(5), e13606, 2018. https://doi.org/10.1111/jfpp.13606

[17] Şahin, S., Elhussein, E., Bilgin, M., Lorenzo, J. M., Barba, F. J., Roohinejad, S. "Effect of drying method on oleuropein, total phenolic content, flavonoid content, and antioxidant activity of olive (*Olea europaea*) leaf", Journal of Food Processing and Preservation, 42(5), e13604, 2018. https://doi.org/10.1111/jfpp.13604

[18] Sharma, T., Zaman, M. N., Rashid, S., Santoshi, S. "In Silico Analysis of Plant-Derived Medicinal Compounds Against Spike Protein of SARS-CoV-2 and Ace2", In: International Conference on Innovative Computing and Communications, Delhi, India, 2022, pp. 299–313. ISBN 978-981-16-2596-1 https://doi.org/10.1007/978-981-16-2597-8_25

[19] Ünlü, A. E. "Green and Non-conventional Extraction of Bioactive Compounds from Olive Leaves: Screening of Novel Natural Deep Eutectic Solvents and Investigation of Process Parameters", Waste and Biomass Valorization, 12(10), pp. 5329–5346, 2021. https://doi.org/10.1007/s12649-021-01411-3

[20] Vijayan, R., Gourinath, S. "Structure-based inhibitor screening of natural products against NSP15 of SARS-CoV-2 revealed Thymopentin and Oleuropein as potent inhibitors", Journal of Proteins and Proteomics, 12(2), pp. 71–80, 2021. https://doi.org/10.1007/s42485-021-00059-w

[21] İstifli, E. S. "In Silico Screening of the Phenolic Compound Oleuropein and Its Hydrolysis Product 3-Hydroxytyrosol Against Certain Structural and Non-Structural Proteins of SARS-CoV-2", Türk Tarım ve Doğa Bilimleri Dergisi, 8(3), pp. 824–833, 2021. https://doi.org/10.30910/turkjans.953603

[22] Kiani, A. K., Dhuli, K., Anpilogov, K., Bressan, S., Dautaj, A., Dundar, M., Beccari, T., Ergoren, M. C., Bertelli, M. "Natural compounds as inhibitors of SARS-CoV-2 endocytosis: A promising approach against COVID-19", Acta Biomedica, 91(13), e2020008, 2020. https://doi.org/10.23750/abm.v91i13-S.10520

[23] Sun, Z., Ostrikov, K. K. "Future antiviral surfaces: Lessons from COVID-19 pandemic", Sustainable Materials and Technologies, 25, e00203, 2020. https://doi.org/10.1016/j.susmat.2020.e00203

[24] Khaerunnisa, S., Kurniawan, H., Awaluddin, R., Suhartati, S., Soetjipto, S. "Potential Inhibitor of COVID-19 Main Protease (M^{pro}) from Several Medicinal Plant Compounds by Molecular Docking Study", Preprints, 2020, 2020030226, 2020. https://doi.org/10.20944/preprints202003.0226.v1

[25] Ahmad-Qasem, M. H., Barrajón-Catalán, E., Micol, V., Mulet, A., García-Pérez, J. V. "Influence of freezing and dehydration of olive leaves (var. Serrana) on extract composition and antioxidant potential", Food Research International, 50(1), pp. 189–196, 2013. https://doi.org/10.1016/j.foodres.2012.10.028

[26] Fadri, R. A., Sayuti, K., Nazir, N., Suliansyah, I. "The Effect of Temperature and Roasting Duration on Physical Characteristics and Sensory Quality Of Singgalang Arabica Coffee (*Coffea arabica*) Agam Regency", Journal of Applied Agricultural Science and Technology, 3(2), pp. 189–201, 2019. https://doi.org/10.32530/jaast.v3i2.117

[27] Perrone, D., Donangelo, R., Donangelo, C. M., Farah, A. "Modeling Weight Loss and Chlorogenic Acids Content in Coffee during Roasting", Journal of Agricultural and Food Chemistry, 58(23), pp. 12238–12243, 2010. https://doi.org/10.1021/jf102110u

[28] Ahmad-Qasem, M. H., Ahmad-Qasem, B. H., Barrajón-Catalán, E., Micol, V., Cárcel, J. A., García-Pérez, J. V. "Drying and storage of olive leaf extracts. Influence on polyphenols stability", Industrial Crops and Products, 79, pp. 232–239, 2016. https://doi.org/10.1016/j.indcrop.2015.11.006

[29] Idoui, T., Bouchefra, A. "The Black Olive Fruits of Jijelian Sigoise Variety (Eastern Algeria): Quality Evaluation for Possible Use as Table Olives and Pesticides Research", The Online Journal of Science and Technology, 4(1), pp. 45–52, 2014. [online] https://dergipark.org.tr/en/download/article-file/210333 [Accessed: 02 March 2015]

[30] Wang, C., Singh, R. P. "Use of variable equilibrium moisture content in modeling rice drying", Transactions of American Society of Agricultural Engineers, 11(6), pp. 668–672, 1978.

[31] Yaldýz, O., Ertekýn, C. "Thin layer solar drying of some vegetables", Drying Technology, 19(3–4), pp. 583–597, 2001. https://doi.org/10.1081/DRT-100103936

[32] Henderson, S. M., Pabis, S. "Grain Drying Theory I. Temperature Effect on Drying Coefficient", Journal of Agricultural Engineering Research, 6(3), pp. 169–174, 1961.

[33] Celma, A. R., Rojas, S., Lopez-Rodriguez, F. "Mathematical modelling of thin-layer infrared drying of wet olive husk", Chemical Engineering and Processing: Process Intensification, 47(9–10), pp. 1810–1818, 2008. https://doi.org/10.1016/j.cep.2007.10.003

[34] Demir, V., Gunhan, T., Yagcioglu, A. K. "Mathematical modelling of convection drying of green table olives", Biosystems Engineering, 98(1), pp. 47–53, 2007. https://doi.org/10.1016/j.biosystemseng.2007.06.011

[35] Verma, L. R., Bucklin, R. A., Endan, J. B., Wratten, F. T. "Effects of Drying Air Parameters on Rice Drying Models", Transactions of the ASAE, 28(1), pp. 0296–0301,1985. https://doi.org/10.13031/2013.32245

[36] Midilli, A., Kucuk, H., Yapar, Z. "A new model for single-layer drying", Drying Technology, 20(7), pp. 1503–1513, 2002. https://doi.org/10.1081/DRT-120005864

[37] Kahyaoglu, T., Kaya, S. "Modeling of moisture, color and texture changes in sesame seeds during the conventional roasting", Journal of Food Engineering, 75(2), pp. 167–177, 2006. https://doi.org/10.1016/j.jfoodeng.2005.04.011

[38] Ngcobo, M. E. K., Pathare, P. B., Delele, M. A., Chen, L., Opara, U. L. "Moisture diffusivity of table grape stems during low temperature storage conditions", Biosystems Engineering, 115(3), pp. 346–353, 2013. https://doi.org/10.1016/j.biosystemseng.2013.03.013

[39] Boudhrioua, N., Bahloul, N., Slimen, I. B., Kechaou, N. "Comparison on the total phenol contents and the color of fresh and infrared dried olive leaves", Industrial Crops and Products, 29(2–3), pp. 412–419, 2009. https://doi.org/10.1016/j.indcrop.2008.08.001

[40] Amiot, M. J., Tacchini, M., Fleuriet, A., Macheix, J. J. "Le processus technologique de désamérisation des olives: caractérisation des fruits avant et pendant le traitement alcalin", Sciences des Aliments, 10(3), pp. 619–631, 1990. [online] https://hal.inrae.fr/hal-02715273 [Accessed: 01 March 2015]

[41] Hurtado, A., Reguant, C., Bordons, A., Rozès, N. "Influence of fruit ripeness and salt concentration on the microbial processing of *Arbequina* table olives", Food Microbiology, 26(8), pp. 827–833, 2009. https://doi.org/10.1016/j.fm.2009.05.010

[42] Huang, Y., Sheng, J., Yang, F., Hu, Q. "Effect of enzyme inactivation by microwave and oven heating on preservation quality of green tea", Journal of Food Engineering, 78(2), pp. 687–692, 2007. https://doi.org/10.1016/j.jfoodeng.2005.11.007

[43] Gunhan, T., Demir, V., Hancioglu, E., Hepbasli, A. "Mathematical modelling of drying of bay leaves", Energy Conversion and Management, 46(11–12), pp. 1667–1679, 2005. https://doi.org/10.1016/j.enconman.2004.10.001

[44] Hannachi, H., Breton, C., Msallem, M., Ben El Hadj, S., El Gazzah, M., Berville, A. "Are olive cultivars distinguishable from oleaster trees based on morphology of drupes and pits, oil composition and microsatellite polymorphisms?", Acta Botanica Gallica, 155(4), pp. 531–545, 2008. https://doi.org/10.1080/12538078.2008.10516132

[45] Demirhan, E., Özbek, B. "Thin-layer drying characteristics and modeling of celery leaves undergoing microwave treatment", Chemical Engineering Communications, 198(7), pp. 957–975, 2011. https://doi.org/10.1080/00986445.2011.545298

[46] Akpinar, E. K., Bicer, Y. "Modelling of thin layer drying kinetics of sour cherry in a solar dryer and under open sun", Journal of Scientific and Industrial Research, 66(9), pp. 764–771, 2007. [online] https://www.semanticscholar.org/paper/Modelling-of-thin-layer-drying-kinetics-of-sour-in-Akpinar-B%C3%AD%C3%A7er/fb554319aee27395ac2fc39402f528ec6cb528e1 [Accessed: 25 January 2017]

[47] Achat, S., Tomao, V., Madani, K., Chibane, M., Elmaataoui, M., Dangles, O., Chemat, F. "Direct enrichment of olive oil in oleuropein by ultrasound-assisted maceration at laboratory and pilot plant scale", Ultrasonics Sonochemistry, 19(4), pp. 777–786, 2012. https://doi.org/10.1016/j.ultsonch.2011.12.006

[48] Bulotta, S., Celano, M., Lepore, S. M., Montalcini, T., Pujia, A., Russo, D. "Beneficial effects of the olive oil phenolic components oleuropein and hydroxytyrosol: focus on protection against cardiovascular and metabolic diseases", Journal of Translational Medicine, 12(1), pp. 1–9, 2014. https://doi.org/10.1186/s12967-014-0219-9

[49] Savournin, C., Baghdikian, B., Elias, R., Dargouth-Kesraoui, F., Boukef, K., Balansard, G. "Rapid High-Performance Liquid Chromatography Analysis for the Quantitative Determination of Oleuropein in *Olea europaea* Leaves", Journal of Agricultural and Food Chemistry, 49(2), pp. 618–621, 2001. https://doi.org/10.1021/jf000596+

[50] Al Juhaimi, F., Özcan, M. M., Uslu, N., Ghafoor, K., Babikler, E. E., Adiamo, O. Q., Alsawmahi, O. N. "The effects of conventional heating on phenolic compounds and antioxidant activities of olive leaves", Journal of Food Science and Technology, 55(10), pp. 4204–4211, 2018.
https://doi.org/10.1007/s13197-018-3356-y

[51] Goupy, P., Fleuriet, A., Amiot, M.-J., Macheix, J.-J. "Enzymic Browning, Oleuropein Content, and Diphenol Oxidase Activity in Olive Cultivars (*Olea europaea* L.)", Journal of Agricultural and Food Chemistry, 39(1), pp. 92–95, 1991.
https://doi.org/10.1021/jf00001a017

[52] García, A., Romero, C., Medina, E., García, P., de Castro, A., Brenes, M. "Debittering of Olives by Polyphenol Oxidation", Journal of Agricultural and Food Chemistry, 56(24), pp. 11862–11867, 2008.
https://doi.org/10.1021/jf802967y

[53] Iqbal, A., Murtaza, A., Hu, W., Ahmad, I., Ahmed, A., Xu, X. "Activation and inactivation mechanisms of polyphenol oxidase during thermal and non-thermal methods of food processing", Food and Bioproducts Processing, 117, pp. 170–182, 2019.
https://doi.org/10.1016/j.fbp.2019.07.006

[54] Lalas, S., Athanasiadis, V., Gortzi, O., Bounitsi, M., Giovanoudis, I., Tsaknis, J., Bogiatzis, F. "Enrichment of table olives with polyphenols extracted from olive leaves", Food Chemistry, 127(4), pp. 1521–1525, 2011.
https://doi.org/10.1016/j.foodchem.2011.02.009

[55] Boukhiar, A., Iguergaziz, N., Moussi, D., Zeggane, K., Benamara, S. "A New Food-Nonfood Fortification: Olive Leaves as Ingredient in Coffee Beverage", presented at World Academy of Science, Engineering and Technology (WASET), Paris, France, Oct., 07-08, 2013.

[56] Iguergaziz, N., Benamara, S., Boukhiar, A., Djallouli, F.-Z., Guebrili, A., Angar, N.-E., Bitam, A. "Release characteristics of paracetamol and oleuropein from *Mech-Degla* date fruit tablets enriched and non-enriched with freeze-dried olive leaf extract", Chemical Engineering Communications, 206(4), pp. 524–534, 2019.
https://doi.org/10.1080/00986445.2018.1505615

[57] Kizhedath, A., Suneetha, V. "Estimation of chlorophyll content in common household medicinal leaves and their utilization to avail health benefits of chlorophyll", Journal of Pharmacy Research, 4(5), pp. 1412–1413, 2011. [online] https://www.researchgate.net/profile/Arathi-Kizhedath/publication/299499753_Estimation_of_chlorophyll_content_in_common_household_medicinal_leaves_and_their_utilization_to_avail_health_benefits_of_chlorophyll/links/58da67ea45851578dfb6bcd8/Estimation-of-chlorophyll-content-in-common-household-medicinal-leaves-and-their-utilization-to-avail-health-benefits-of-chlorophyll.pdf [Accessed: 05 March 2022]

[58] Giuliani, A., Cerretani, L., Cichelli, A. "Chlorophylls in Olive and in Olive Oil: Chemistry and Occurrences", Critical Reviews in Food Science and Nutrition, 51(7), pp. 678–690, 2011.
https://doi.org/10.1080/10408391003768199

[59] Kaushal, M., Sharma, K. D., Attri, S. "Effect of blanching on nutritional quality of dehydrated colocasia, *Colocasia esculenta* (L.) Schott leaves", Indian Journal of Natural Products and Resources, 4(2), pp. 161–164, 2013. [online] http://nopr.niscpr.res.in/handle/123456789/19898 [Accessed: 05 May 2020]

[60] Pumilia, G., Cichon, M. J., Cooperstone, J. L., Giuffrida, D., Dugo, G., Schwartz, S. J. "Changes in chlorophylls, chlorophyll degradation products and lutein in pistachio kernels (*Pistacia vera* L.) during roasting", Food Research International, 65, pp. 193–198, 2014.
https://doi.org/10.1016/j.foodres.2014.05.047

[61] Bahloul, N., Boudhrioua, N., Kouhila, M., Kechaou, N. "Effect of convective solar drying on colour, total phenols and radical scavenging activity of olive leaves (*Olea europaea* L.)", International Journal of Food Science & Technology, 44(12), pp. 2561–2567, 2009.
https://doi.org/10.1111/j.1365-2621.2009.02084.x

[62] Lin, J.-T., Liu, S.-C., Hu, C.-C., Shyu, Y.-S., Hsu, C.-Y., Yang, D.-J. "Effects of roasting temperature and duration on fatty acid composition, phenolic composition, Maillard reaction degree and antioxidant attribute of almond (*Prunus dulcis*) kernel", Food Chemistry, 190, pp. 520–528, 2016.
https://doi.org/10.1016/j.foodchem.2015.06.004