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Karim Assami, Smain Chemat, Brahim Y. Meklati, Farid Chemat. Ultrasound-Assisted Aromatisation with Condiments as an Enabling Technique for Olive Oil Flavouring and Shelf Life Enhancement. *Food Analytical Methods*, 2016, 9 (4), pp.982-990. 10.1007/s12161-015-0273-9 . hal-01351180

HAL Id: hal-01351180

<https://hal.science/hal-01351180v1>

Submitted on 27 May 2020

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Ultrasound-Assisted Aromatisation with Condiments as an Enabling Technique for Olive Oil Flavouring and Shelf Life Enhancement

Karim Assami^{1,2} · Smain Chemat¹ · Brahim Y. Meklati² · Farid Chemat³

Abstract Olive oil flavour represents the most important factor that influences customer's perception for quality. In this study, an innovative aromatisation approach that enhanced quality and shelf life of olive oil is tested by means of in situ ultrasound-assisted extraction of *Carum carvi* L. seeds in olive oil. After the flavouring process, carvone and limonene, representing 99 % of caraway essential oil, were identified in virgin olive oil as revealed by headspace solid-phase microextraction gas chromatograph coupled to mass spectrometer (HS-SPME gas chromatograph (GC)/mass spectrometer (MS)) analyses. Furthermore, kinetic studies indicate that flavouring rate in ultrasound-assisted system is faster than conventional aromatisation producing very stable aromatised oil with no changes in quality. This fact is attributed to ultrasound contribution to mass transfer intensification of essential oil components from seeds to the bulk medium, confirmed by scanning electron micrograph (SEM) micrographs in which cell damage and exudation of its main content are illustrated. However, it is important to note that ultrasound-treated samples produce higher degrees of conjugated dienes and conjugated trienes compared to conventional maceration. Nevertheless, this increase is not alarming and remains within

permitted values of the European Community and the International Olive Oil Council (IOOC). Ultrasound-flavouring system stands as a fast alternative aromatisation method, which allows on top shelf life enhancement and olive oil quality preservation.

Keywords Ultrasound · Flavouring · Olive oil · *Carum carvi* L. · Oxidative stability · Shelf life

Introduction

Olive oil represents, nowadays, a major source for fatty acids in Mediterranean diet regimen (Visioli and Galli 2002), mainly attributed to assertive nutritional and health benefits (Luaces et al. 2003). In Algeria, olive oil market has always played an important role in agriculture and food industry with an annual production of 45,000 t spread over 100,000 olive producers according to the International Olive Oil Council data (IOOC International Olive Oil Council 2009).

Generally, olive oil recovery from olive fruits (*Olea europaea* L.) is mainly performed via mechanical and physical methods such as hydraulic or cold pressing; then, a separation step from water by decanters is necessary before the filtration stage. In this situation, three different grades of olive oil suitable for consumption can be distinguished (extra virgin, virgin and ordinary) according to their acid value (or free acidity), expressed as grams of oleic acid equivalent per 100 g of oil. According to the International Olive Oil Council (IOOC) (IOOC International Olive Oil Council 2015), these grades have an acid value (AV) respectively of <0.8 % (extra virgin), 0.8 % < AV < 2.0 % (virgin) and 2.0 % < AV < 3.3 % (ordinary).

Olive oil consists of fatty acids (Ollivier et al. 2003), sterols (Jimenez de Bias and del Valle Gonzalez 1996), aroma

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(Angerosa et al. 2004) and phenolics (Tripoli et al. 2005). Among these aroma compounds, distinctive sensorial properties can be attributed to the complex mixture of more than 100 volatile compounds (Morales et al. 1994). Commercial olive oils presenting off-flavours and a bitter taste are less attractive from a consumer perspective. However, this bitter taste attributed to glycosides is considered as a positive nutritional characteristic according to IOOC trade standard (IOOC International Olive Oil Council 2015).

Continuous efforts to meet client needs have been initiated by industries to look for specific olive oil products profiling an added value. Vegetable oil enriched with essential oil has made a significant breakthrough and was realised through direct topping (Damechki et al. 2001; Gambacorta et al. 2007). However, it is believed that the latter process induces an increase in oil acidity, thus altering its stability and calling for an extra refinery setup. Other methods such as infusion have been tested to aromatise oil with vegetables, fruits, herbs or flowers (Moldão-Martins et al. 2004). Traditionally, herb-infused olive oil is particularly used with crusty bread in Italy, whereas olive oil is infused in Spain with hot red peppers. Conventional flavouring of olive oil needs a simple setup but requires large storage tanks and long period of 4 to 6 weeks of maceration. In this respect, an ultrasound-assisted aromatisation method is proposed and compared to a conventional mixing, in an attempt to accelerate significantly the process without altering olive oil quality.

This technology offers several advantages over conventional techniques such as yield improvement and time reduction. For instance, Wei et al. (2008) reported a yield increase of more than 20 % for rapeseed oil recovery using ultrasound over Soxhlet extraction technique. Furthermore, Luque-Garcia and Luque de Castro (2004) achieved great recovery of total fat from sunflower seeds with Soxhlet extraction assisted by ultrasound. Low solvent consumption associated with acceleration of heat and mass transfer as a result of positive cavitation effect is highly appreciated in the recovery of high-value compounds such as essential oils, anthocyanins and beta-carotene from food waste (Galanakis 2012). Nonetheless, non-standardised methodologies and the absence of defined control parameters represent the main problems encountered during industrial process development (Galanakis 2013).

Largely used condiment, caraway fruits (*Carum carvi* L.) are selected for aromatisation of olive oil by means of conventional maceration versus ultrasound-assisted extraction. It belongs to the Umbelliferae family and characterised by a sweet and a slight peppery aroma fulfilled by its essential oil, mainly composed of antioxidant compounds such as carvone and limonene (Elmastas et al. 2006; Roberto et al. 2010). For this study, the comparison of olive oil aromatisation with caraway seeds is achieved by means of ultrasound and maceration. Olive oil quality is assessed through analyses of major

compounds transferred from flaked carvi fruits into olive oil, free fatty acids yield, peroxide value, fatty acid composition and oxidation degree. Finally, the influence of aromatisation mechanism is studied by collecting scanning electron micrographs of caraway seeds' residue before and after processing.

Materials and Methods

Materials

Caraway seeds (*C. carvi* L.) and olive oil branded as virgin oil were purchased from a local supermarket (Algiers, Algeria). Reference compounds (S)-(+)-carvone and (R)-(+)-limonene of 98.0 % purity were purchased from Sigma-Aldrich (Germany). All reagents were of analytical grades, and solvents of HPLC grades were supplied by Merck (Darmstadt, Germany).

Isolation of Caraway Essential Oil by Hydrodistillation

For comparison purposes, isolation and characterisation of *C. carvi* L. essential oil are performed in order to evaluate in terms of quality and quantity the transfer yield of major compounds from flaked seeds into olive oil. In this case, hydrodistillation procedure is used in which 150 g of caraway seeds was finely ground using an electric grinder (Ika, Germany) at 20,000 rpm over 20 s. The flaked seeds were immediately submitted for hydrodistillation in a Clevenger-type system (Clevenger 1928; ISO 1987) and extracted with 1.5 L of distilled water over 3 h (i.e. until no more essential oil was obtained). The essential oil was collected and analysed with gas chromatograph (GC)/mass spectrometer (MS) as described in earlier studies (Assami et al. 2012).

Aromatisation Procedures

In this section, two procedures were studied: conventional aromatisation and ultrasound-assisted aromatisation as explained on Fig. 1 that illustrates several stages involved in olive oil aromatisation by *C. carvi* L.

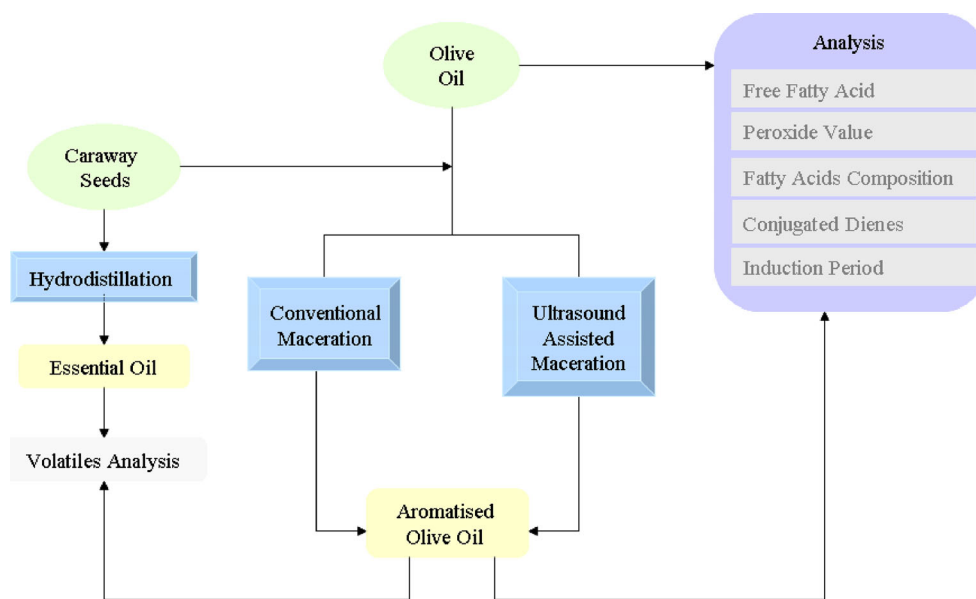
Conventional Aromatisation

In the conventional system, 150 g of crushed caraway seeds was added to 1 L of virgin olive oil; this ratio has been selected according to literature, which recommends, for spices, the addition of 0.5 to 20 % relative to olive oil weight (Van Boom et al. 2002). The mixture was placed into a 2-L double jacketed reactor into which cooling water circulates in order to keep the mixture at a constant temperature of 25 °C. The mixture was homogenised using a rotating pale at a speed of 1000 rpm and left for several hours, with intermediate

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Assami, K. (Auteur de correspondance), Chemat, S., Meklati, B. Y., Chemat, F. (2016).
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Fig. 1 Stages of olive oil aromatisation by *Carum carvi* L. using conventional maceration or ultrasound-assisted maceration



samplings for aromatisation kinetics purposes. Our preliminary assays indicated that beyond 6 h (720 min), very little mass transfer is observed, which led us to establish that 6 h was the optimal infusion time. The final aromatised mixture was filtered through a coffee filter in order to remove traces of crushed seeds, and samples were kept at ambient temperature until further analysis. All experiments were repeated in triplicates, and mean values are reported.

Ultrasound-Assisted Aromatisation

An ultrasonic system (REUS, France) operating at a frequency of 25 kHz is used (Fig. 2). The ultrasonic reactor consists of a double-mantle stainless steel jug into which cooling water

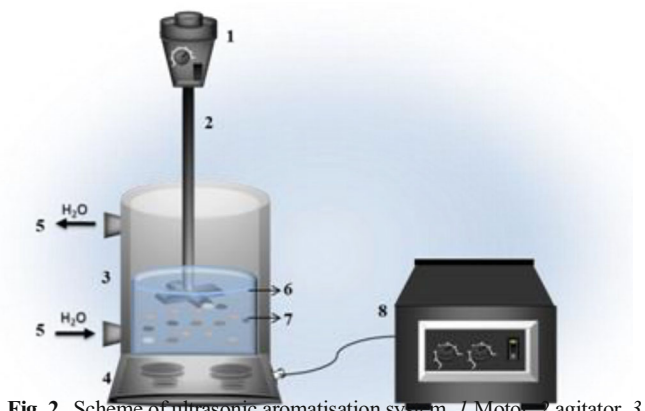


Fig. 2 Scheme of ultrasonic aromatisation system. 1 Motor, 2 agitator, 3 double mantle, 4 transducers, 5 cooling system, 6 olive oil, 7 crushed caraway seeds, 8 ultrasound generator.

circulates to keep the mixture at constant temperature of 25 °C. Similarly, 150 g of crushed caraway seeds was placed into the reactor together with 1 L of olive oil and the mixture was homogenised during ultrasound processing using a rotating pale at a speed of 1000 rpm.

In an attempt to determine the real power output converted into ultrasound, power at different points is recorded to estimate ultrasound system efficiency. In this case, consumed power C_p at the socket (measured using a Watt meter), generated power G_p (using equipment conversion tables) and dissipated power into heat D_p (measuring temperature change of the medium by means of a thermocouple and applying first law of thermodynamics' equation) are registered. The results indicate that increasing generated power G_p (i.e. consumed power as well) engenders relatively a notable increase in temperature of the medium, attributed to the multiplication of interactions and friction forces as a response to high olive oil viscosity and resistance of ultrasound penetration.

At a fixed power, generator efficiency is evaluated to reach 77 % while converter efficiency is estimated to be 51.6 %. Therefore, the global ultrasound system efficiency may reach a maximum of 39.3 %. In this situation, temperature increased during time at a linear pattern for which dissipated power D_p is estimated at 1.36 W/mL of olive oil.

Furthermore, groundwork trials using ultrasound indicated that saturation is achieved between 20 and 30 min. Therefore, an aromatisation period of 30 min is selected. The collected mixture was filtered through a coffee filter in order to remove traces of crushed seeds, and samples were kept at ambient temperature until further analysis. Experiments were performed in triplicates, and mean values are reported.

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Analytical Procedures

For the analytical procedures, a comparison was performed between three samples, namely olive oil before aromatisation (OO), conventional aromatised olive oil (CAOO) and ultrasound aromatised olive oil (UAOO).

Physico-Chemical Parameters

In this part, a physico-chemical assessment of oil, before and after aromatisation, is conducted to see changes on free fatty acid (FFA) yield, peroxide value (PV) and absorption coefficients in UV (at 232 and 270 nm). These analyses were conducted following analytical protocols described in the European Regulations (EUC 2013). Experiments were performed in triplicates, and mean values are reported with standard deviation.

Fatty Acid Composition

Before injection, olive oil samples are dissolved first in *n*-heptane (0.12 g/2 mL) and then trans-methylated with a cold solution of KOH (2 M) (1 mL) (Galtier et al. 2007). The resulting fatty acid methyl esters (FAMES) were analysed on an Agilent 6890 GC coupled to a 5973A MS using fused silica capillary column with polar phase, Stabilwax, consisting of Carbowax™ PEG (60 m×0.25 mm internal diameter (i.d.)×0.25- μ m film thickness). The oven temperature program is performed as follows: 13 min at 200 °C, then increasing from 200 to 230 °C at 6 °C/min, and finally held 17 min at 230 °C. The injection port and ionising source were kept at 250 and 230 °C, respectively. One-microliter samples are injected using a split ratio of 70:1. Helium is used as carrier gas with a flow of 0.7 mL/min using an electronic impact ionisation mode at ionisation energy of 70 eV.

Volatile Analysis

Essential oil's components that are believed to have infused in olive oil are detected using a headspace solid-phase microextraction gas chromatograph coupled to mass spectrometer (HS-SPME-GC/MS) as described earlier by Veillet et al. (2010). In this situation, 4±0.032 g of olive oil is inserted in a 20-mL vial preincubated at 40 °C for 15 min. The volatile compounds contained into the vial's headspace are left for 30 min to be absorbed on a SPME fiber (75- μ m carboxen-polydimethylsiloxane) under magnetic stirring of the oil.

The resulting volatiles are automatically injected into Agilent 6890 GC coupled to a 5973A MS detector using fused silica capillary column with polar phase, Stabilwax, consisting of Carbowax™ PEG (60 m×0.25 mm i.d.×0.25- μ m film thickness). The initial oven temperature is held at 45 °C for 3 min. Then, it is increased by 3 °C/min to 250 °C and held for

5 min at this temperature. The injector temperature is set at 250 °C, and desorption time of the fiber into the injection port is fixed at 5 min in a splitless mode. Helium is used as carrier gas with a flow of 0.7 mL/min. The ionising source is kept at 230 °C, and the ionisation mode used is electronic impact (EI) at ionisation energy of 70 eV. Component identification is confirmed by comparison of mass spectral fragmentation patterns with those stored in the MS data bank (Wiley 7) and compared with published data (Ferhat et al. 2010; Assami et al. 2012).

Accelerated Oxidation Tests

Measurement of the induction period (IP) is performed using the well-established Rancimat method using Rancimat model 734 (Metrohm, Herisau, Switzerland) operating at 120 °C (Tura et al. 2007). A dry air flow of 20 L/h was passed through an oil sample of 3±0.001 g. The volatile oxidation products, arising from the oxidation test, were dissolved in cold Milli-Q water (60 mL) causing an increase in the electrical conductivity. The concept is based on conductivity change of the distillate collected from an oil sample that was submitted to an accelerated oxidation at a prefixed temperature. This conductivity change is attributed to the production of formic and other carboxylic acids as a consequence of forced oxidation of secondary products. These experiments were performed in triplicate, and the mean value is reported (Table 1). In this case, the time taken to reach an inflection point at the induction curve is considered for oil stability index (OSI) calculations.

Statistical Analysis

One-way analysis of variance (ANOVA) was used using Statgraphics V[®] software (Statistical Graphics Corp., Rockville, MD). Each measurement was replicated three times, and a Tukey multiple range test method was used to point out deviation from the mean value. In this case, trends were considered significant only where means of compared parameters are different at $P<0.05$ significance level.

Scanning Electron Micrographs

In an attempt to explain extraction mechanism, after each extraction, seeds' residues are sampled for scanning electron micrographs. The specimens were plunged in liquid nitrogen and then cut with a cold knife. The sectioned particles were fixed on a specimen holder with aluminium tape and then sputtered with gold in sputter coater. All the specimens were examined using FEI-Philips XL30 scanning electron microscope under standard vacuum conditions.

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Table 1 Physicochemical parameters and fatty acid composition of studied olive oil

Parameters	OO	CAOO	UAOO
FFA (% of oleic acid)	0.46±0.02	0.48±0.03	0.53±0.02*
PV (meq O ₂ kg ⁻¹)	2.1±0.4	2.7±0.2	2.4±0.4
K 232	1.69±0.05	1.76±0.01	1.99±0.05
K 270	0.14±0.02	0.16±0.02	0.19±0.03
Induction period (IP) (h)	3.45±0.5	4.53±0.5	6.39±1*
Fatty acid composition (%)			
Palmitic acid	17.78±0.58	18.39±0.57	18.14±0.57
Palmitoleic acid	0.10±0.02	0.10±0.03	0.10±0.02
Margaric acid	1.77±0.04	1.89±0.04	1.68±0.03
Stearic acid	1.57±0.04	1.59±0.06	1.57±0.01
Oleic acid	63.96±1.33	62.47±1.08	62.36±0.87
Linoleic acid	10.26±0.16	10.24±0.13	9.79±0.13
Linolenic acid	0.55±0.03	0.53±0.01	0.51±0.01
Arachidic acid	0.28±0.06	0.36±0.02	0.35±0.05
Saturated fatty acids (SFA)	25.13±0.26	26.66±0.26	27.24±0.27
Monounsaturated fatty acids (MUFA)	64.06±0.92	62.57±0.74	62.46±0.60
Polyunsaturated fatty acids (PUFA)	10.81±0.09	10.77±0.08	10.30±0.08
Oleic/linoleic ratio	6.23	6.10	6.36

P value was determined by one-way analysis of variance (ANOVA) with Tukey multiple range test method

OO olive oil, CAOO conventional aromatized olive oil, UAOO ultrasound aromatized olive oil

**P*<0.05 significance level

Results and Discussion

Physico-Chemical Analysis

For the three samples, olive oil before aromatisation (OO), conventional aromatised olive oil (CAOO) and ultrasound aromatised olive oil (UAOO), the free fatty acid (FFA) values reported on Table 1 remained below the limit (0.8 g of oleic acid per 100 g) required for virgin olive oil by the IOOC (IOOC International Olive Oil Council 2015). In this situation, FFA values ranged from 0.46 % for non-aromatised oil (OO) to 0.53 % for the oil aromatised by ultrasound (UAOO). In addition, the results indicate an increase on peroxide value (PV) of oils aromatised conventionally (CAOO) with 2.7 meq O₂kg⁻¹ in comparison to un-aromatised oil; however, this increase is far lower than the upper limit of 20 meq O₂ kg⁻¹, permitted by the IOOC. These results suggest that processing did not alter oils that underwent aromatisation.

In another part, the absorbance measurements indicate a slight increase for ultrasound-treated samples in K232 (conjugated dienes) and K270 (conjugated trienes) compared to conventional maceration. Nevertheless, this increase attributed eventually to the production of conjugated dienes and other

secondary oxidation compounds is not alarming and remains within the EC permitted values of 2.5 for K232 and 0.25 for K270 (EUC 2013).

We hypothesise that this increase is mainly attributed to rapid temperature increase as a consequence of high frictions forces as a response to high olive oil viscosity during ultrasound treatment as evidenced by ultrasound equipment efficiency findings reported above.

Despite a statistically significant increase in FFA and K232 data observed for UAOO relative to CAOO and OO (*p*<0.05), still, these values remain below the IOOC limits (2015). This

Table 2 Caraway essential oil composition analysed by GC/MS

No.	Compound ^a	R.I. ^b	R.I. ^c	HD (%)
	Monoterpenes			46.00
1	Pinene <alpha->	929	1024	0.04
2	Sabinene	968	1122	0.03
3	Myrcene <beta->	989	1163	0.35
4	Limonene	1031	1212	45.49
5	Ocimene <beta->	1046	1250	0.01
6	Terpinene <gamma->	1054	1246	0.09
	Oxygenated monoterpenes			53.43
7	Linalool	1086	1535	0.02
8	Limonene oxide <cis->	1127	1445	0.01
9	Limonene oxide <trans->	1132	1458	0.15
10	Dihydrocarvone <cis->	1188	1620	0.01
11	Isoanethole	1190	1627	0.02
12	Dihydrocarvone <trans->	1195	1639	0.01
13	Carveol	1226	1869	0.14
14	Carvone	1218	1763	52.86
15	Perillal	1267	1804	0.15
16	Anethole	1273	1842	0.06
	Sesquiterpenes			0.08
17	Elemene <beta->	1383	1599	0.02
18	Caryophyllene <trans->	1409	1611	0.04
19	Germacrene <d->	1471	1734	0.01
	Oxygenated sesquiterpenes			0.01
20	Caryophyllene oxide	1575	2006	0.01
	Other oxygenated compounds			0.01
21	Dihydrocarveol acetate	1318	1810	0.01
	Extraction time (min.)			180
	Yield (%)			1.86
	Total oxygenated compounds (%)			53.45
	Total non-oxygenated compounds (%)			46.08

^a Essential oil compounds sorted by chemical families and percentages calculated by GC-MS on non-polar HP5MS™ capillary column

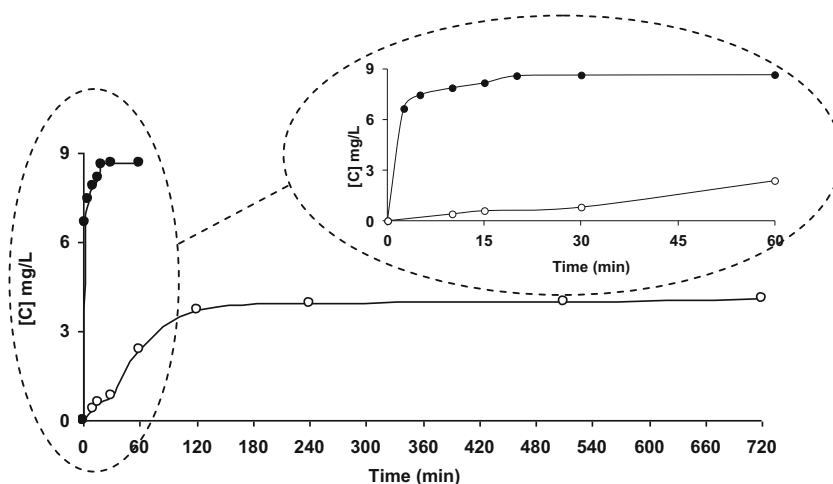
^b Retention index relative to C₅-C₂₈ *n*-alkanes, calculated on non-polar HP5MS™ capillary column

^c Retention index relative to C₅-C₂₈ *n*-alkanes, calculated on polar Stabilwax™ PEG capillary column

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Fig. 3 Kinetics of Carvone recovery using ultrasound-assisted maceration (*closed circles*) versus to conventional maceration (*open circles*)



finding is another evidence in favour of ultrasound-assisted aromatisation; it is clear that ultrasound did not affect olive oil quality.

Accelerated Oxidation Tests

The results reported on Table 1 show that the induction period (IP) varies according to each olive oil sample. UAOO samples represent the highest IP value with 6.39 h followed by CAOO (4.45 h) and, finally, the untreated sample OO (3.45 h). Statistically speaking, the IP values for UAOO samples were significantly ($p < 0.05$) higher than CAOO and OO. Generally, oxidative stabilities of oils are attributed to the combination of fatty acid composition and the effect of various pro-antioxidants present in the olive oil (Hrnčirik and Fritsche 2005). It is important to highlight that caraway essential oil presents valuable antioxidant activities (Samojlik et al. 2010), which may explain, in part, noticeable increases in IP of aromatised olive oil, particularly for UAOO, since essential oil yield using ultrasound is more important compared to CAOO sample. These results suggest that ultrasound-

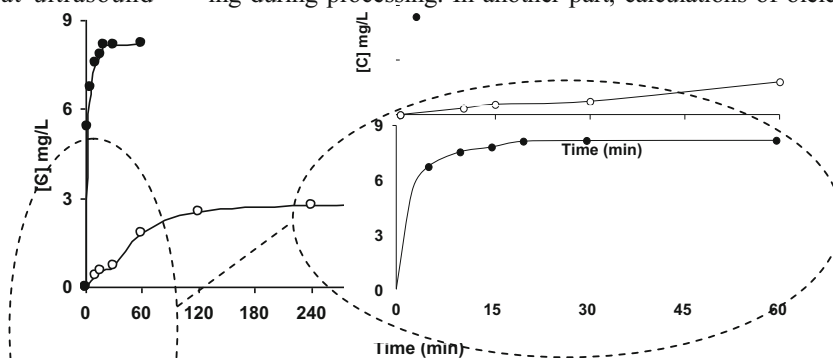
assisted treatment grants an added value for samples by increasing their IPs, i.e. shelf life time, so treatment by means of ultrasound contributes to a longer preservation period of packaged foods.

Fatty Acid Composition

The composition of fatty acids is reported on Table 1. The major fatty acids present in raw olive oil are as follows: oleic acid (C18:1) 64 %, palmitic acid (C16:0) 17.8 %, linoleic acid (C18:2) 10.3 % and stearic acid (C18:0) with 1.6 %.

As shown in Table 1, total monounsaturated fatty acids (MUFA) include the largest fatty acid group in the studied olive oil ranging, respectively, for UAOO and OO samples (62.46–64.06 %), followed by saturated fatty acids (27.24–25.13 %) and then polyunsaturated fatty acids (10.30–10.81 %). In this context, slight difference in fatty acid composition is observed, indicating that aromatisation procedure did not affect fatty acid composition. In addition, MUFA value remains intact which indicates that no oxidation was happening during processing. In another part, calculations of oleic/

Fig. 4 Kinetics of limonene recovery using ultrasound-assisted maceration (*closed circles*) versus to conventional maceration (*open circles*)



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linoleic acid ratios (O/L) for non-treated, conventional and ultrasonic treated olive oils give, respectively, 6.23, 6.10 and 6.36 which correlate well with an olive oil characterised as stable portraying a long shelf life as reported by Aparicio et al. (1999) and Rotondi et al. (2004).

Essential Oil Composition

For aromatisation purposes, caraway seeds were extracted using hydrodistillation giving a yield of 1.86 % (w/w). According to our previous studies (Assami et al. 2012), 21 volatile secondary metabolites were identified and grouped into five classes. In this study, oxygenated monoterpenes represent around 54 % of total oil versus 46 % of non-oxygenated compounds (Table 2). The major compounds were identified as carvone 52.86 % and limonene 45.49 % of essential oil.

Comparison of Aromatisation Methods

According to the previous section, carvone and limonene represent the major compounds; therefore, these two compounds were selected as indicators of the aromatisation level of olive oil samples. First of all, calibration curves for both compounds were established in order to estimate their concentrations in aromatised olive oils. In this case, different concentrations of carvone and limonene standards were prepared in olive oil and analysed by HS-SPME-GC/MS.

Moreover, samples of olive oils were taken at different periods of maceration in order to capture the efficiency of the aromatisation process. In Figs. 3 and 4, a significant difference between ultrasound-assisted and conventional maceration is observed during all the processing period. For ultrasound, it appears that only 10 min is required to reach equilibrium of 90 % essential oil recovery then reaching its maximum value at 30 min with 8.75 and 8.27 mg/L for carvone and limonene, respectively. In parallel, conventional procedure failed to meet the same rate recovering only 0.81 mg/L for carvone and 0.73 mg/L for limonene. Even after 12 h of conventional processing, still, only 4.21 and 2.81 mg/L, respectively, for carvone and limonene could be achieved. These values remain very low compared to those found under ultrasonic treatment. These results are in accordance with the work of Veillet et al. (2010) on ultrasound-assisted aromatisation of olive oil with basil where they reported that maximum linalool and eugenol yields could be reached in only 30 min under ultrasound and are two times higher than those obtained at conventional maceration for 72 h.

This fact is attributed to the positive effect of ultrasound on mass transfer intensification through rapid exudation of essential oil components from seeds into the bulk medium, well reported in earlier studies by Vinatoru et al. (1997) and Assami et al. (2012).

Morphology Changes at Seeds' Residues

In this section, scanning electron micrograph (SEM) micrographs have facilitated the illustration of modifications provoked by each extraction process. In Fig. 5, the micrograph of untreated seeds (A) is juxtaposed to those of seed samples treated with ultrasound (B) and those processed under conventional heating (C). In the vertical cut of untreated seeds, glands are unaffected keeping their initial intact cell form. However, the ultrasound-assisted extraction process produced major changes on the cell structure in which cell perforations are observed in some parts and total destruction of cell walls and exploded glands are noticeable in other parts of seeds' residue (Fig. 5b). Veillet et al. (2010) reported the same

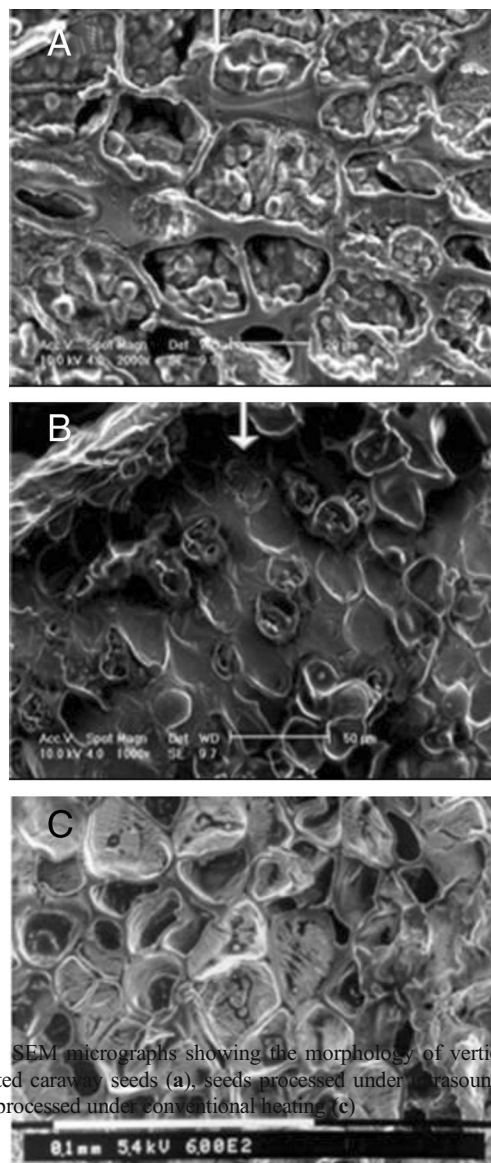


Fig. 5 SEM micrographs showing the morphology of vertical cut in untreated caraway seeds (a), seeds processed under ultrasound (b) and seeds processed under conventional heating (c)

observation of exploded glands, arguing that essential oil glands located at the surface of the leaves are directly exposed to the intense mechanical stress induced by cavitation bubbles. For conventional processing seeds' residue (Fig. 5c), cell walls appeared intact, but certain dryness in some cells is noticeable, apparently a synonym of exudation at low pace for part of seed content into the bulk medium. Since kinetic studies ascertained a fast liberation of gland content under ultrasound treatment, this micrograph may explain why ultrasound-assisted extraction is faster than the conventional one (Figs. 3 and 4). In this respect, severe localised thermal stresses at the surface of cell walls, allegedly generated by cavitation phenomena, are suggested to be responsible for such damage. This finding corroborates with earlier results which confirms the strong mechanical effect of ultrasound (Chemat and Esveld 2013).

Conclusion

The findings supported the potential borne by ultrasound treatment in the aromatisation of vegetable oils with condiments in an attempt to enhance quality and shelf life of olive oil.

The application of HS-SPME-GC/MS for the analysis of aromatised virgin olive oil by crushed caraway seeds allowed the detection of significant differences in carvone and limonene proportions in which a faster flavouring rate is registered for ultrasound-assisted system versus the conventional one. Samples treated under ultrasound produced very stable aromatised oil with no changes in quality as revealed by olive oil purity, FFA composition and degree of oxidation analysis. In this part, ultrasound-flavouring process was able, in few minutes, to achieve high yields versus several hours for conventional method. This fact is attributed to ultrasound contribution to mass transfer intensification of essential oil components from seeds to the bulk medium, confirmed by SEM micrographs in which cell damage and exudation of its main content are illustrated.

The results indicate also that IP of olive oil has increased following aromatisation, particularly for ultrasound-treated samples, which is an indication of better quality and extended shelf life granting, therefore, an added value label for ultrasound samples.

Compliance with Ethics Requirements

Ethical Approval This article does not contain any studies with human or animal subjects by any of the authors.

Conflict of Interest Karim Assami declares that he has no conflict of interest. Smain Chemat declares that he has no conflict of interest. Brahim Y. Meklati declares that he has no conflict of interest. Farid Chemat declares that he has no conflict of interest.

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Assami, K. (Auteur de correspondance), Chemat, S., Meklati, B. Y., Chemat, F. (2016) Ultrasound-Assisted Aromatisation with Condiments as an Enabling Technique for Olive Oil Flavouring and Shelf Life Enhancement. *Food Analytical Methods*, 9 (4), 982-990. DOI : 10.1007/s12161-015-0273-9

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