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A sound approach: Exploring a rapid and non-destructive ultrasonic pulse echo system for vegetable oils characterization

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ABSTRACT

A rapid and non-destructive ultrasonic pulse echo system was developed for vegetable oils characterization. To understand the differences in the ultrasonic properties of the oils, physical traits, such as their viscosity and density, were related to the ultrasonic data. In turn, these physical traits were correlated with the fatty acid compositions of the oils. Eighty oil samples, including 30 extra virgin olive oil (EVOO), 15 refined olive oil, 15 pomace olive oil, 10 rapeseed oil, 5 sunflower oil and 5 peanut oil samples, were analysed for their sound properties, viscosities, densities and fatty acid compositions. It was observed that the ultrasonic velocity of EVOO decreased linearly with increase in temperature, the temperature coefficient of ultrasonic velocity in EVOO was $-2.92\,m{\cdot}s^{-1}{\cdot}°C^{-1}$. The ultrasonic velocity of EVOO (1453 ± 2 m/s) differed significantly from those of pomace olive oil and the oils of other botanical origin, but not from the velocity of refined olive oil. Ultrasonic velocity was positively correlated with the density and negatively correlated with the viscosity of the oils. The higher density and lower viscosity of the oils were in turn related to a higher unsaturation degree of the oils. Hence, oils with a higher proportion of unsaturated fat present higher densities and lower viscosities, which resulted in higher ultrasonic velocity values. Ultrasonic measurements allow rapid, non-destructive analysis, and this first application for characterization of these oils is promising.

1. Introduction

Extra virgin olive oil (EVOO) extracted from fresh olive fruits using traditional cold pressing methods is the highest quality olive oil available commercially. Because of its high nutritional value, premium organoleptic quality and high price, it turns out to be an attractive target for food fraud. EVOO adulteration is considered a common fraudulent practice in the olive oil industry and these adulterations include the addition of lower grade olive oils (refined olive oil (ROO) and pomace olive oil (POO)), and other vegetable oils (rapeseed oil (RSO), sunflower oil (SFO) and peanut oil (PNO)) (De Oliveira & Catharino, 2015).

In order to ensure food safety and assess EVOO, the European Commission (2013) and International Olive Council (2016) released official methods on the characteristics of olive oil and on the relevant methods of analysis. Moreover, a large number of potential techniques for EVOO characterization have been reported (Bajoub, Bendini, Fernandez-Gutierrez, & Carrasco-Pancorbo, 2018), which can be divided into chromatography (Yan, Oey, van Leeuwen, & van Ruth, 2018), mass spectrometry (Marone et al., 2017), vibrational spectroscopy (Wang, Sun, Zhang, & Liu, 2016), nuclear magnetic resonance (Fragaki, Spyros, Siragakis, Salivaras, & Dais, 2005), DNA-based techniques (Pasqualone et al., 2016) and other techniques (Chiavaro, Vittadini, Rodriguez-Estrada, Cerretani, & Bendini, 2008). Rapid, inexpensive and non-invasive analytical approaches for EVOO characterization are continually being sought (Persuric, Saftic, Masek, & Pavelic, 2018; Squeo, Grassi, Paradiso, Alarnprese, & Caponio, 2019), whereas the sound properties, which could be interesting markers for rapid and non-destructive analysis, have hardly received any attention to date. Sound (0–20 kHz) measurements have been applied as an effective approach for salt profiling and identification (van Ruth et al., 2019). Analyses using the ultrasound (> 20 kHz) range could also be promising for food characterization. Ultrasonic spectroscopy is a development of the pulse-echo technique which uses broadband (0.5–10 MHz) ultrasound and analyses the spectra of the echo pulses

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(Brown, 1973). It is a promising technique for rapid and non-destructive analyses. The ultrasonic waves generated from an ultrasonic transducer, propagate through the oil samples. Thus, measurement of the characteristics of these ultrasonic waves may then show differences which relate to properties of vegetable oils.

Measurement of the ultrasonic velocity (speed of sound) is the basis of most ultrasonic techniques used to evaluate the properties of foods (McClements, 1997). Furthermore, ultrasonic velocity has been correlated to the rheological properties of vegetable oils (Gladwell, Javanaud, Peers, & Rahalkar, 1985) and the density of oils (Sankarappa, Kumar, & Ahmad, 2005). Measurements of ultrasonic velocity have been employed to characterize edible oils (Alouache, Khechena, Lecheb, & Boutkedjirt, 2015; Alouache, Laux, Hamitouche, Bachari, & Boutkedjirt, 2018), such as for solid fat content analysis (Singh, McClements, & Marangoni, 2004), recycled edible oils evaluation (Ali & Ahmad, 2018), and frying oil degradation assessments (Benedito, Garcia-Perez, Dobarganes, & Mulet, 2007; Benedito, Mulet, Velasco, & Dobarganes, 2002). Moreover, researchers have previously reported a potential correlation between ultrasonic velocities of olive oil and their chemical composition (Kumari, Yadav, & Singh, 2017; McClements & Povey, 1988), which has been proposed as an alternative method for compositional analyses. Therefore, ultrasonic measurements could be an alternative and promising approach for rapid and non-destructive assessment of the integrity of oils. Considering the vulnerability of olive oil to fraud, olive oil would be an interesting target to explore this type of approach.

In this study, a pulse-echo ultrasonic system was employed to explore the characterization of the vegetable oils, and to examine the underlying causes for the ultrasonic velocity differences between the oils. The detailed objectives of this study are 1) to evaluate a developed ultrasonic velocimetry measurement system for the vegetable oils characterization; 2) to investigate the correlation between ultrasonic measurements and the viscosity and density of the oils; 3) to relate the viscosity and density of the oils to their fatty acid compositions.

2. Materials and methods

2.1. Samples

Eighty vegetable oil samples were collected, and their authenticity was confirmed by official methods, including fatty acid compositional fingerprinting, and spectrophotometric tests measuring extinction coefficients (K232, K268 and ΔK) (International Olive Council, 2015). They were also evaluated for their monochloropropanediol esters contents (Yan et al., 2018). They included olive oil samples: 30 EVOO, 15 ROO and 15 POO samples, as well as oils of other botanical origins which are often found as adulterants: 10 RSO, 5 SFO and 5 PNO samples. The information of the 80 oil samples is provided as supplementary material (Supplementary Table S1). Prior to analysis, samples were stored in capped bottles, which were kept in the dark at room temperature until analysis.

2.2. Ultrasonic velocity analysis

The ultrasonic measurements were carried out using a purpose-built pulse-echo system. This pulse-echo system, shown schematically in Fig. 1a, is composed of a sample platform (Fig. 1b), an immersion transducer (diameter 12.54 mm, Panametrics-NDT, Olympus NDT U.K. Ltd., Rotherham, South Yorkshire, UK) with a central frequency of 5 MHz, a computer controlled pulse generator/receiver (Panametrics-NDT Model 5800, Olympus NDT U.K. Ltd., Rotherham, South Yorkshire, UK), an oscilloscope (Tektronix TDS 210, Tektronix UK Ltd., Bracknell, UK) and a computer (Dell, Texas, US).

As can be seen in Fig. 1b, a special sample platform which is fixed on a stainless steel base plate was custom fabricated and consisted of a transparent plastic tube (2), a solid stainless steel cylinder (3) and a micrometer (4) (fixed on one side of a vertical stainless steel bracket). A good seal between the plastic cylinder and the solid stainless steel cylinder is managed by rubber rings to avoid leakage, and the generated volume is used as a sample cell (63 mm internal diameter). The transducer (1) is placed in a holder fixed to the micrometer. The distance between the transducer surface and the bottom of the sample cell can be measured with the micrometer (0.01 mm), with an integrated spring in the micrometer maintaining a constant position force. The sample platform was placed into a temperature-controlled water bath to maintain the sample temperature (23.5 °C ± 0.1 °C). When recording a measurement, the face of the transducer had to be placed below the oil surface as shown in Fig. 1b, and any air bubbles trapped in the oil on the transducer face were manually removed prior to testing by swiping over the surface of the transducer with finger.

The principle of the pulse echo system is similar that reported by other researchers (Alouache, Boutkedjirt, & Laux, 2016; Awad, Moharram, Shaltout, Asker, & Youssef, 2012) but presents some differences. The computer controlled pulse generator/receiver is used to produce/generate monopolar electrical pulses, which are converted to ultrasonic pulses by the transducer. The pulse generator/receiver was operated in pulse echo mode with a pulse repetition frequency of 1 kHz, 25 μJ pulse energy with 20 dB of input attenuation, and 20 dB of receiver amplification. The generated ultrasonic pulse propagates through the vegetable oils from the transducer until it reflects from the stainless steel cylinder face (solid reflector), back to the same transducer, which acts as a receiver and converts the returned ultrasonic pulses into electrical signals that are displayed on the oscilloscope. Subsequently, the ultrasonic signal from each sample was transferred to the computer via a general-purpose interface bus interface with the oscilloscope. The signal acquisition was repeated with different separations between the transducer face and the stainless steel base using a Matlab (R2015b, The Math Works Inc., Natick, MA, USA) program.

The ultrasonic velocity can be calculated by analysing the times of the echoes received at the oscilloscope. As shown in Fig. 1a, the distance between the transducer and the bottom of the sample cell (L) is equal to half the distance propagated by the ultrasonic pulse. Six measurements were carried out under six consecutive distances (10, 12, 14, 16, 18 and 20 mm) for each sample, and the amount of the oils used under each measurement distance are 31.17, 37.41, 43.64, 49.88, 56.11 and 62.34 ml, respectively. The ultrasonic velocity (V) can be thus calculated by two measurements under two consecutive distances using Eq. (1):

$$V = \frac{2(L_2 - L_1)}{T_2 - T_1}, \tag{1}$$

where L_1 is the lower distance of one-way echo path (m); L_2 is the consecutive next distance of one-way echo path (m), which is larger than L_1; T_1 is the time of receiving the first reflected signal (s) under the distance of L_1; T_2 is the time of receiving the first reflected signal (s) under the distance of L_2; V is the propagation velocity of the ultrasound in samples (m/s).

Since six distances were measured for each sample, five velocity values were calculated. The final velocity of each sample was the average of the five velocity values. This device was calibrated by measuring the ultrasonic velocity of water (1499 ± 3 m/s at 26 °C) (Engineering ToolBox, 2004).

2.3. Viscosity analysis

The apparent viscosity of the oils was measured using a Haake Roto Visco 1 rotational viscometer (Thermo Fisher Scientific, GmbH, Karlsruhe, Germany), equipped with a Z41 concentric cylinder and Z43 cup. Each sample (10 ml) was loaded into the gap between the cylinder and the cup, before being allowed to equilibrate at 20 °C for 1 min before analysis. In the first step, the sample was subjected to a shear rate ramp from 0 to 200 s^{-1} over 2 min, and then held at 200 s^{-1} for

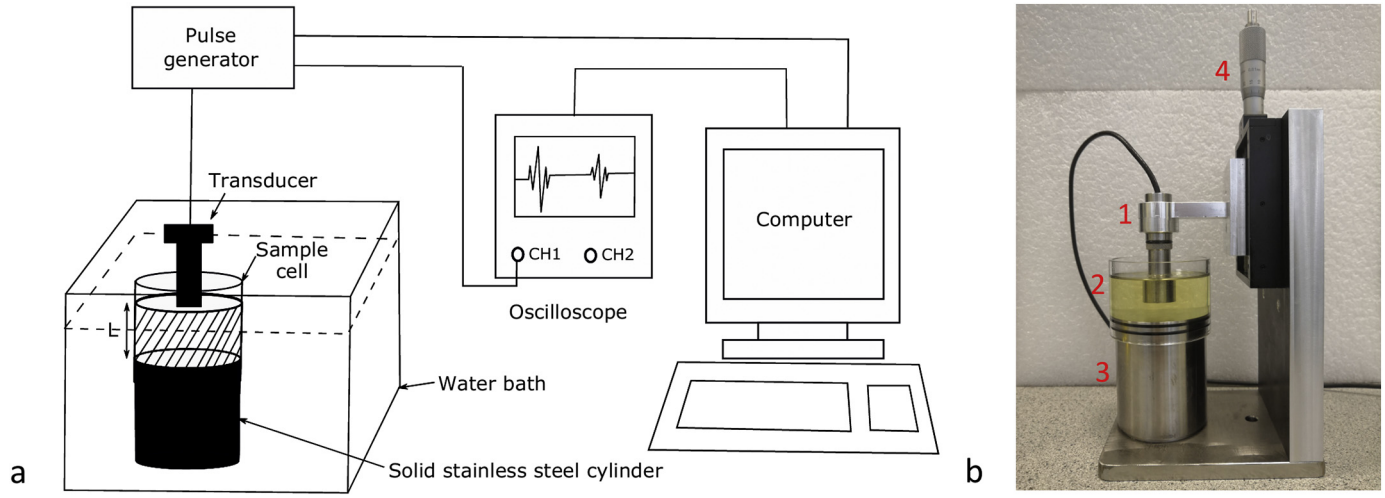


Fig. 1. a Schematic diagram of ultrasonic pulse-echo system. L, length of the half-way wave path in oil samples. b Sample platform of the pulse-echo system. (1) transducer; (2) transparent plastic cylinder; (3) solid stainless steel cylinder; (4) micrometer.

2 min before returning shear rate from 200 to 0 s^{-1} over 2 min. The test was carried out at 20.0 ± 0.1 °C, and the temperature was controlled by a digital recirculating water bath. The average viscosity was taken at a shear rate of 200 s^{-1}. The equation of the viscosity (see Eq. (2)) was used to describe the rheological properties of the samples:

$$\eta = \frac{\tau}{\gamma}, \tag{2}$$

where η is the viscosity (Pa.s), τ is the shear stress (Pa), γ is the shear rate (s^{-1}). Duplicate measurements of all samples were conducted.

2.4. Density analysis

The density values were calculated using a gravimetric method. The flask and the 25 ml glass pipette were weighed together (m_1) by electronic lab balance (± 0.001 g), then the 25 ml (ν) oil sample was pipetted into the flask by the glass pipette. Subsequently, the flask, the glass pipette and the oil were weighed together (m_2). The density of the oil was obtained by using Eq. (3):

$$\rho = \frac{m_2 - m_1}{\nu} \times 1000, \tag{3}$$

where ρ is the density of the sample (kg/m^3), m_1 is the total weight of the empty flask and the glass pipette (g), m_2 is the total weight of the oil sample, the flask and the glass pipette (g), and ν is the accurate volume of the sample (ml). The tests were carried out at room temperature (20.0 ± 0.1 °C). Duplicate measurements were conducted for all samples.

2.5. Measurement of fatty acids

2.5.1. Fatty acid methyl esters preparation

The 100 µl of the internal standard solution (50 mg tritridecanoin (C13:0) and 50 mg methyl undecanoate (C11:0) dissolved by 10 ml pentane) was pipetted into a gas chromatography (GC) vial, and the solution was evaporated to dry under a stream of nitrogen. Then, one droplet of each oil sample (approximately 10–20 mg) was transferred into the prepared GC vial and the vial was closed with a magnetic cap. Then, the samples were placed in the autosampler (Gerstell MPS, GmbH, Germany). Subsequently, an automatic boron trifluoride transmethylation procedure was conducted for the sample preparation as reported in International Organization for Standardization (2017).

2.5.2. Fatty acid methyl ester analysis

The gas chromatographic analysis of fatty acids (FAs) was carried out according to International Organization for Standardization (2015). The Agilent HP7890A GC (Agilent Technologies, Inc., Wilmington, DE, USA) was equipped with a 100 m × 0.25 mm × 0.2 µm film thickness fused silica capillary column (Varian, Palo Alto, CA) coupled to a flame ionization detector. Working condition: initial column temperature, 120 °C; final column temperature, 240 °C; heating ramp, 4.0 °C/min; hold time, 7 min at 240 °C; run time, 37 min; carrier, hydrogen; constant flow, 1.0 ml/min; injection, 1 µl; split, 1:100; injector temperature, 250 °C; detector temperature, 250 °C.

Individual fatty acid methyl esters (FAMEs) were identified by comparison with external standards. The internal standard C13:0 was employed for the quantification of individual FAMEs. In addition, the performance of transesterification was evaluated by internal standards C13:0 and C11:0. Blanks were performed prior to analysing each batch of samples for the stability test of the machine.

Iodine values (IV, g I/100 g) and Saponification numbers (SN, mg KOH/g) of 80 vegetable oils were calculated from their FA compositions as described by Kalayasiri, Jeyashoke, and Krisnangkura (1996). Duplicate measurements of all samples were conducted.

2.6. Statistical analysis

Means and standard deviations were calculated for each sample and oil group. Non-parametric Kruskal-Wallis tests were applied for group comparisons due to non-normality nature of the data. The pairwise comparisons were carried out by Mann-Whitney *U* tests ($p < 0.05$). All of the above data analysis methods were performed by SPSS statistic 23 (IBM, Chicago, IL, USA).

The correlations between the ultrasonic velocity, viscosity and density, as well as between these physical characteristics and the FAs compositional data were assessed by computing Pearson correlation coefficients (r). Paired samples *t*-tests were applied to assess significance of the correlation coefficients. All calculations were performed by scripts developed in R 3.3.3 (R Foundation for Statistical Computing, Vienna, Austria).

Partial least squares (PLS) regressions with leave-one-out cross validation were carried out, using Pirouette 4.5 (Infometrix, USA) to create predictive models based on FAs data. The FAs dataset was preprocessed in three ways, including auto-scaling, mean-centering, and range-scaling. The performance of models was evaluated by coefficient of determination for cross-validation (r^2) and standard error of cross-validation (SEV), since SEV is the best single estimate of the prediction capability of these kinds of models (Fernandez-Cabanas, Polvillo, Rodriguez-Acuna, Botella, & Horcada, 2011).

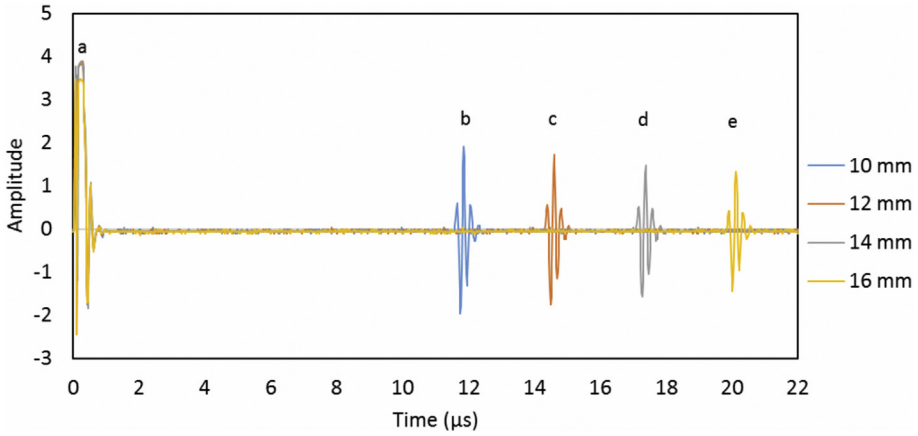


Fig. 2. Plot of the first propagation signal of extra virgin olive oil analysed at four distances. a refers to the transmitter pulse, b, c, d, e refer to the first propagation signal at the distance of 10, 12, 14 and 16 mm, respectively.

3. Results and discussion

3.1. Pulse echo system – ultrasonic velocity measurements

3.1.1. Development of the measurement system

In terms of the sample platform, oil was placed in a sample cell of 63 mm internal diameter with adjustable height from 0 to 50 mm, which illustrate the maximum amount of oil required for the measurement is < 150 ml. The amount of the oils used under each measurement distance are 31.17 ml (10 mm), 37.41 ml (12 mm), 43.64 ml (14 mm), 49.88 ml (16 mm), 56.11 ml (18 mm) and 62.34 ml (20 mm). Moreover, the measurement time for each sample at a certain distance from generating signal to receiving and recording the signal is < 30 s, and this could be reduced with automation of the transducer positioning.

Fig. 2 shows the variation in ultrasonic propagation delay of EVOO under four different distances (10 mm, 12 mm, 14 mm and 16 mm). The first signal (a) in Fig. 2 is the transmitter pulse without propagating through the oil sample and is electronic cross-talk between the pulser and the amplifier. Thus, the first propagation signal is the second one (b), the time delay of receiving the first propagation signal (the time when the signal reaches its peak) increases when the distance increases, i.e., from 11.862 μs (b) to 14.614 μs (c), 17.364 μs (d) and 20.116 μs (e). Because of the difficulty of picking out the peak of the first signal, the velocity of ultrasound cannot be calculated using the time and distance directly, but applying the relative time between two distances is an alternative way for velocity calculation. In this case, the mean value of velocity of EVOO is 1453.84 ± 0.47 m/s.

3.1.2. Initial evaluation of the measurement system

Previous studies reported that the ultrasonic properties may vary when the measurement temperature is changed (Benedito et al., 2002; Sankarappa et al., 2005). The variation of the ultrasonic velocity with temperature in EVOO was measured over a temperature range from 18 to 36 °C, using an ultrasonic frequency of 5 MHz. The signal was recorded at 2 °C intervals and the temperature was controlled by a water bath. As shown in Fig. 3, the velocity was observed to be decreasing with the increase of temperature in the applied temperature range, which results in a velocity decrease from 1470 to 1416 m/s. Sankarappa et al. (2005) explained that velocity changes with temperature are attributed to changes in intermolecular distance with temperature.

Fig. 3 reveals that the correlation coefficient (R^2) is > 0.99, therefore, ultrasonic velocity is linearly related to the temperature of the olive oil over the temperature range studied. This is in agreement with other observations made in olive oil (Benedito et al., 2002) and other oils (sunflower oil and refined groundnut oil) (Sankarappa et al., 2005).

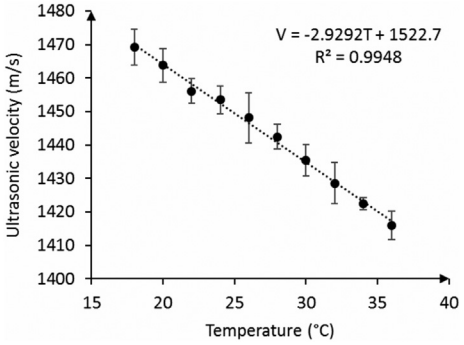


Fig. 3. Variation of ultrasonic velocity with temperature for extra virgin olive oil at the ultrasonic frequency of 5 MHz.

However, a previous study also reported that velocity decreased non-linearly with increasing temperature in coconut oil, castor oil and unrefined Kardi (safflower) oil (Sankarappa et al., 2005). Moreover, the temperature coefficient for ultrasonic velocity in EVOO is $-2.93\ m{\cdot}s^{-1}{\cdot}°C^{-1}$. This slope is close to the value reported by Benedito et al. (2002) for olive oil (ranges from -3.37 to $-3.54\ m{\cdot}s^{-1}{\cdot}°C^{-1}$) and McClements and Povey (1988) ($-3.28\ m{\cdot}s^{-1}{\cdot}°C^{-1}$). This illustrates that the assay accuracy of this pulse echo system is high. Since the ultrasonic measurements are temperature dependent, all 80 samples were measured at a constant temperature (23.5 °C ± 0.1 °C) in the subsequent study to obtain repetitive and reliable results.

3.2. Ultrasonic velocity of the different oils

The 80 vegetable oils, including 30 EVOO, 15 ROO, 15 POO, 10 RSO, 5 SFO and 5 PNO, were subjected to ultrasonic velocity measurements using the developed pulse echo system, and the results are presented in Table 1. As can be seen in Table 1, the ultrasonic velocities of the six categories are in the range of 1450 to 1462 m/s and significant differences between groups were observed (Kruskal-Wallis test, $p < 0.05$). Specifically, a significant difference exists between EVOO (1453 ± 2 m/s) and the other vegetable oils (RSO (1460 ± 1 m/s), SFO (1461 ± 1 m/s), and PNO (1458 ± 1 m/s)), which also can be noticed in Table 1. Furthermore, a significant difference in mean values between EVOO (1453 ± 2 m/s) and POO (1456 ± 1 m/s) was established. This result may be explained by the fact that POO is extracted from olive residue with the help of chemicals and/or high temperatures, which results in a lower quality oil compared with EVOO, with different characteristics (Gunstone, 2011). However, there is no significant difference between EVOO (1453 ± 2 m/s) and ROO

Table 1
Averages and standard deviations (SD) of ultrasonic velocity, viscosity and density of 80 oil samples, including 30 extra virgin olive oils (EVOO), 15 refined olive oils (ROO), 15 pomace oils (POO), 10 rapeseed oils (RSO), 5 sunflower oils (SFO) and 5 peanut oils (PNO). Different letters (a, b, c, d) indicate significant differences between oil groups (Kruskal-Wallis and Mann-Whitney *U* tests, $p < 0.05$).

Type	Ultrasonic velocity (m/s)	Viscosity (mPa.s)	Density (kg/m^3)
EVOO (n = 30)	1455 ± 2	83.6 ± 0.4	913 ± 0
	1456 ± 1	82.9 ± 0.3	911 ± 0
	1455 ± 0	83.4 ± 0.2	912 ± 0
	1454 ± 2	85.5 ± 0.1	911 ± 1
	1454 ± 2	82.5 ± 0.2	912 ± 0
	1455 ± 3	82.8 ± 0.1	909 ± 0
	1452 ± 1	83.4 ± 0.2	911 ± 0
	1456 ± 3	84.2 ± 0.2	909 ± 0
	1453 ± 2	84.5 ± 0.3	912 ± 1
	1452 ± 0	81.4 ± 0.2	911 ± 1
	1455 ± 0	84.1 ± 0.2	912 ± 0
	1453 ± 2	82.1 ± 0.4	913 ± 1
	1453 ± 3	83.8 ± 0.2	913 ± 1
	1454 ± 1	83.0 ± 0.3	914 ± 1
	1456 ± 3	86.4 ± 0.4	913 ± 1
	1454 ± 1	82.8 ± 0.2	909 ± 1
	1453 ± 2	83.5 ± 0.3	911 ± 0
	1452 ± 1	85.5 ± 0.2	911 ± 1
	1452 ± 1	82.7 ± 0.4	913 ± 0
	1452 ± 2	84.3 ± 0.2	909 ± 0
	1452 ± 2	83.3 ± 0.4	912 ± 1
	1450 ± 1	82.0 ± 0.2	912 ± 0
	1455 ± 3	81.9 ± 0.3	911 ± 0
	1453 ± 4	83.6 ± 0.4	916 ± 1
	1453 ± 2	84.3 ± 0.2	912 ± 1
	1452 ± 2	83.0 ± 0.3	913 ± 0
	1454 ± 1	82.6 ± 0.2	911 ± 0
	1452 ± 0	82.2 ± 0.4	914 ± 1
	1448 ± 1	82.4 ± 0.3	914 ± 0
	1454 ± 3	83.3 ± 0.2	912 ± 0
Mean ± SD	1453 ± 2^{c}	83.3 ± 1.2^{b}	912 ± 2^{d}
ROO (n = 15)	1455 ± 2	87.1 ± 0.2	913 ± 0
	1453 ± 2	86.6 ± 0.2	912 ± 1
	1454 ± 1	87.0 ± 0.3	913 ± 1
	1454 ± 4	86.4 ± 0.4	913 ± 0
	1455 ± 2	84.7 ± 0.4	913 ± 1
	1456 ± 0	85.6 ± 0.3	914 ± 1
	1457 ± 3	85.5 ± 0.2	913 ± 1
	1454 ± 3	86.1 ± 0.3	913 ± 0
	1454 ± 2	88.0 ± 0.3	913 ± 0
	1455 ± 1	86.0 ± 0.1	912 ± 0
	1456 ± 4	86.0 ± 0.2	913 ± 0
	1455 ± 3	86.9 ± 0.1	913 ± 0
	1454 ± 2	86.5 ± 0.2	913 ± 1
	1456 ± 4	85.8 ± 0.2	910 ± 0
	1453 ± 2	85.7 ± 0.2	912 ± 0
Mean ± SD	1455 ± 1^{bc}	86.3 ± 0.8^{a}	913 ± 1^{cd}
POO (n = 15)	1457 ± 1	88.0 ± 0.2	911 ± 0
	1456 ± 1	85.8 ± 0.2	912 ± 1
	1457 ± 2	85.8 ± 0.3	911 ± 1
	1456 ± 3	86.1 ± 0.3	915 ± 0
	1456 ± 2	90.4 ± 0.3	914 ± 1
	1455 ± 2	87.4 ± 0.3	913 ± 0
	1457 ± 8	87.6 ± 0.3	916 ± 0
	1455 ± 3	84.7 ± 0.3	915 ± 1
	1457 ± 1	85.5 ± 0.3	916 ± 0
	1457 ± 2	86.2 ± 0.3	917 ± 1
	1455 ± 1	86.2 ± 0.3	917 ± 1
	1454 ± 1	85.7 ± 0.2	914 ± 0
	1456 ± 2	86.7 ± 0.2	916 ± 0
	1458 ± 2	85.7 ± 0.2	914 ± 1
	1457 ± 2	84.9 ± 0.1	918 ± 1
Mean ± SD	1456 ± 1^{b}	86.4 ± 1.4^{a}	915 ± 2^{bc}
RSO (n = 10)	1461 ± 3	72.1 ± 0.2	920 ± 1
	1461 ± 1	72.2 ± 0.3	921 ± 4
	1460 ± 2	73.4 ± 0.2	922 ± 3
	1459 ± 2	70.6 ± 0.1	915 ± 1
	1460 ± 3	71.9 ± 0.2	919 ± 1
	1462 ± 2	65.1 ± 0.3	920 ± 1
	1462 ± 3	75.5 ± 0.2	917 ± 1
	1461 ± 3	74.9 ± 0.2	918 ± 1
	1459 ± 2	74.8 ± 0.2	917 ± 1
	1459 ± 3	80.2 ± 0.3	915 ± 0
Mean ± SD	1460 ± 1^{a}	73.1 ± 3.9^{c}	919 ± 3^{a}
SFO (n = 5)	1461 ± 1	64.8 ± 0.1	921 ± 1
	1460 ± 2	67.1 ± 0.2	922 ± 0
	1461 ± 5	65.8 ± 0.2	921 ± 1
	1462 ± 4	66.5 ± 0.2	921 ± 1
	1460 ± 4	72.2 ± 0.2	920 ± 0
Mean ± SD	1461 ± 1^{a}	67.3 ± 2.9^{d}	921 ± 1^{a}
PNO (n = 5)	1458 ± 5	87.6 ± 0.2	916 ± 1
	1457 ± 3	87.5 ± 0.2	916 ± 0
	1458 ± 2	86.8 ± 0.2	916 ± 0
	1457 ± 4	84.2 ± 0.2	915 ± 0
	1458 ± 5	87.6 ± 0.3	915 ± 0
Mean ± SD	1458 ± 1^{b}	86.7 ± 1.5^{a}	915 ± 1^{b}

(1455 ± 1 m/s). This may due to the fact that EVOO and ROO are primarily a mixture of triacylglycerols with a similar pattern, with only 0.5–1.5% FAs, mono- and diacylglycerols, and non-glyceridic constituents (Gunstone, 2011). Thus, differences in composition are relatively small.

Three vegetable oils (RSO, SFO and PNO) present higher velocity values compared with the olive oils with average values of 1460 ± 1 m/s, 1461 ± 1 m/s and 1458 ± 1 m/s, respectively. Meanwhile, the velocities of SFO and RSO are higher than the velocity of PNO, this trend is in accordance with earlier observations reported by Coupland and McClements (1997).

In the following sections we explore underlying causes for the differences established.

3.3. Viscosity and density of the different oils

All samples were subjected to viscosity and density measurements, the results of which are presented in Table 1, along with the significant differences between oil groups. The viscosity of EVOO (83.3 ± 1.2 mPa.s) is significantly higher than that of RSO (73.1 ± 3.9 mPa.s) and SFO (67.3 ± 2.9 mPa.s), and is significantly lower than that of PNO (86.7 ± 1.5 mPa.s). Furthermore, the viscosity of EVOO (83.3 ± 1.2 mPa.s) is significantly lower than ROO (86.3 ± 0.8 mPa.s) and POO (86.4 ± 1.4 mPa.s). This may be due to the high temperature during the refining process, which leads to the formation of long-chain FAs (polymers), which results in the increase of viscosity (Kreps et al., 2017).

As shown in Table 1, the three olive oils (EVOO, ROO, POO), with viscosities ranging from 81 to 88 mPa.s, are more viscous than RSO (73.1 ± 3.9 mPa.s) and SFO (67.3 ± 2.9 mPa.s). This may be due to the fact that the majority of bonds in olive oil are single bonds with a "zig-zag" configuration, which cause higher viscosities than other bonds (Schaschke, Allio, & Holmberg, 2006). However, the viscosity of PNO (86.7 ± 1.5 mPa.s) does not significantly differ from ROO (86.3 ± 0.8 mPa.s) and POO (86.4 ± 1.4 mPa.s), which is possibly due to the broad similarity in composition.

The density values of EVOO (912 ± 2 kg/m^3) differ significantly from those of the other three vegetable oils (RSO, SFO, and PNO; Table 1). These differences are mainly due to differences in chemical composition (Kalogianni, Karapantsios, & Miller, 2011). Furthermore, a significant difference between density values of EVOO (912 ± 2 kg/m^3) and POO (915 ± 2 kg/m^3) is observed, but no significant

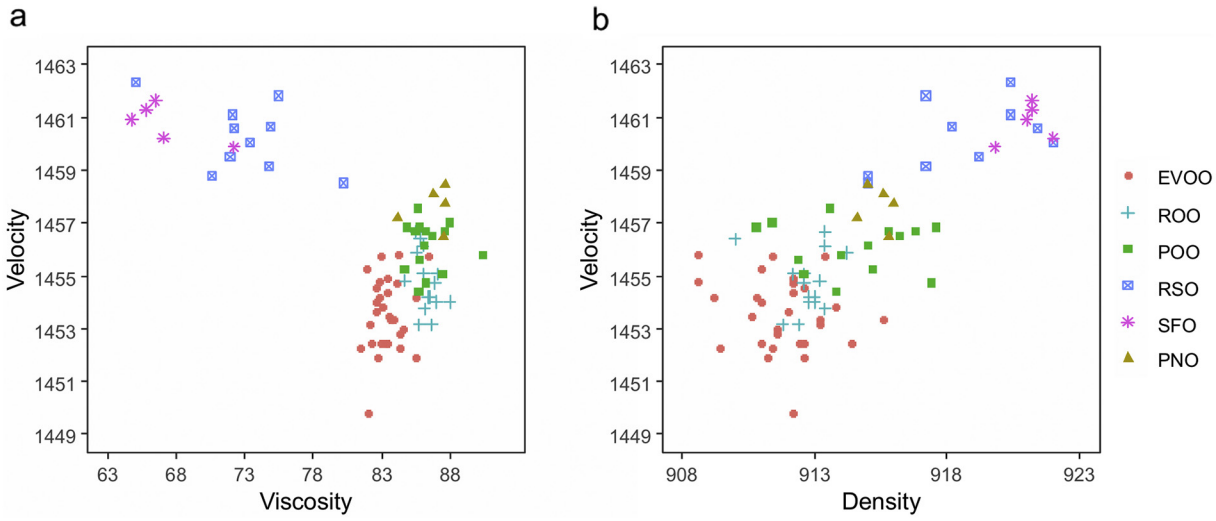


Fig. 4. Scatter plots of ultrasonic velocity versus a) viscosity and b) density data of 30 extra virgin olive oils (EVOO), 30 other olive oil grade samples (15 refined olive oils (ROO) and 15 pomace oils (POO)), and 20 oils of other botanical origins (10 rapeseed oils (RSO), 5 sunflower oils (SFO) and 5 peanut oils (PNO)).

difference between EVOO (912 ± 2 kg/m^3) and ROO (913 ± 1 kg/m^3). It is evident that the density of EVOO is lowest for all olive oils, and this is in agreement with the previous study reported that unrefined oils have lower densities than their refined counterparts (Sankarappa et al., 2005).

3.4. Correlation of the ultrasonic velocity, viscosity and density of the oils

In order to explore the correlation of the ultrasonic velocity, viscosity and density in vegetable oils, the relationship between velocity data and the viscosity and density data are shown in Fig. 4a and b, respectively. The EVOO is located in the lower right hand corner of the velocity/viscosity plot (Fig. 4a) and fully separated from the oils of other botanical origins (RSO, SFO and PNO). EVOO presents somewhat lower viscosity and velocity values than the lower grade olive oils (ROO and POO). Subsequently, the correlation coefficient (r) was calculated for the ultrasonic velocity and viscosity. A significant negative correlation between the velocity and the viscosity ($r = -0.64$, $p < 0.05$) was observed, which is in agreement with previous research (Alouache et al., 2015; Alouache et al., 2016).

Comparing the velocity and density values, it appears that the EVOO is located in the lower left side corner of the corresponding plot (Fig. 4b). EVOO can be discriminated from RSO, SFO and PNO using velocity and density values, but EVOO has a slight overlap with POO and presents considerable overlap with ROO. A significant positive correlation ($r = 0.75$, $p < 0.05$) between the velocity and density was observed.

The results above reveal that the intermolecular structure may be responsible for the velocity differences. Further exploration of correlation of ultrasonic velocity, viscosity, density and intermolecular composition would be useful.

Table 2
Averages and standard deviations of relative fatty acid composition (FA), iodine values (IV), and saponification numbers (SN) of six types of edible oils. The significant differences between oil groups are indicated by superscript letters (Kruskal-Wallis and Mann-Whitney *U* tests, $p < 0.05$). EVOO, extra virgin olive oil; ROO, refined olive oil; POO, pomace olive oil; RSO, rapeseed oil; SFO, sunflower oil; PNO, peanut oil; SFA, sum of saturated fatty acids; MUFA, sum of monounsaturated fatty acids; PUFA, sum of polyunsaturated fatty acids.

Chemical properties (g/100 g)	EVOO (n = 30)	ROO (n = 15)	POO (n = 15)	RSO (n = 10)	SFO (n = 5)	PNO (n = 5)	P value
C14:0	0.05 ± 0.00^{e}	0.02 ± 0.00^{e}	0.02 ± 0.00^{d}	0.05 ± 0.00^{b}	0.08 ± 0.05^{a}	0.03 ± 0.00^{c}	< 0.05
C15:0	0.05 ± 0.00^{d}	0.05 ± 0.00^{d}	0.00 ± 0.00^{e}	0.02 ± 0.00^{a}	0.02 ± 0.00^{b}	0.05 ± 0.00^{c}	< 0.05
C16:0	12.64 ± 1.58^{a}	11.97 ± 1.02^{ab}	11.60 ± 0.46^{b}	4.51 ± 0.36^{d}	6.96 ± 0.71^{c}	8.48 ± 1.29^{c}	< 0.05
C17:0	0.08 ± 0.04^{a}	0.08 ± 0.02^{a}	0.08 ± 0.05^{a}	0.07 ± 0.05^{a}	0.04 ± 0.05^{b}	0.08 ± 0.05^{a}	< 0.05
C18:0	2.63 ± 0.43^{b}	3.20 ± 0.56^{a}	2.86 ± 0.21^{b}	1.56 ± 0.06^{c}	3.26 ± 0.05^{a}	2.61 ± 0.55^{b}	< 0.05
C20:0	0.50 ± 0.04^{b}	0.43 ± 0.04^{d}	0.47 ± 0.02^{c}	0.18 ± 0.04^{f}	0.27 ± 0.06^{e}	1.56 ± 0.20^{a}	< 0.05
C22:0	0.15 ± 0.02^{e}	0.14 ± 0.03^{e}	0.20 ± 0.05^{d}	0.36 ± 0.05^{c}	0.76 ± 0.06^{b}	3.12 ± 0.33^{a}	< 0.05
C24:0	0.05 ± 0.03^{e}	0.04 ± 0.03^{e}	0.09 ± 0.00^{d}	0.12 ± 0.05^{c}	0.26 ± 0.045^{a}	2.07 ± 0.17^{b}	< 0.05
SFA	16.07 ± 1.49^{ab}	15.88 ± 0.70^{ab}	15.34 ± 0.40^{b}	6.94 ± 0.39^{d}	11.64 ± 0.60^{c}	17.96 ± 1.94^{a}	< 0.05
C16:1n9	0.13 ± 0.09^{a}	0.12 ± 0.05^{a}	0.12 ± 0.05^{a}	0.04 ± 0.00^{c}	0.03 ± 0.05^{d}	0.05 ± 0.00^{b}	< 0.05
C16:1n7	0.88 ± 0.32^{a}	0.99 ± 0.26^{a}	0.82 ± 0.06^{a}	0.20 ± 0.05^{b}	0.16 ± 0.08^{bc}	0.09 ± 0.02^{c}	< 0.05
C17:1n7	0.14 ± 0.07^{a}	0.12 ± 0.02^{a}	0.13 ± 0.02^{a}	0.06 ± 0.05^{b}	0.04 ± 0.05^{c}	0.06 ± 0.05^{b}	< 0.05
C18:1n9	71.68 ± 3.47^{a}	71.44 ± 2.57^{a}	69.45 ± 1.06^{b}	59.92 ± 4.72^{c}	27.95 ± 2.69^{d}	62.61 ± 4.35^{c}	< 0.05
C18:1n7	2.01 ± 0.57^{c}	2.11 ± 0.39^{c}	2.42 ± 0.17^{b}	2.68 ± 0.20^{a}	0.80 ± 0.13^{d}	0.72 ± 0.11^{d}	< 0.05
C20:1n9	0.36 ± 0.06^{b}	0.30 ± 0.04^{c}	0.35 ± 0.02^{b}	1.36 ± 0.04^{a}	0.23 ± 0.02^{d}	1.84 ± 0.42^{a}	< 0.05
C22:1n9	0.00 ± 0.00^{c}	0.00 ± 0.00^{c}	0.00 ± 0.00^{b}	0.20 ± 0.14^{a}	0.00 ± 0.00^{c}	0.12 ± 0.13^{ab}	< 0.05
MUFA	75.20 ± 2.80^{a}	75.07 ± 2.15^{a}	73.29 ± 1.02^{b}	64.45 ± 4.67^{c}	29.20 ± 2.88^{d}	65.50 ± 4.58^{c}	< 0.05
C18:2n6	7.71 ± 1.72^{d}	8.26 ± 1.65^{d}	10.09 ± 0.98^{c}	18.67 ± 2.34^{b}	58.56 ± 3.29^{a}	16.09 ± 3.22^{b}	< 0.05
C18:3n3	0.66 ± 0.05^{b}	0.58 ± 0.05^{c}	0.64 ± 0.04^{b}	9.05 ± 1.20^{a}	0.18 ± 0.04^{d}	0.00 ± 0.00^{e}	< 0.05
PUFA	8.38 ± 1.73^{e}	8.84 ± 1.65^{e}	10.73 ± 1.01^{d}	27.72 ± 4.29^{b}	58.73 ± 3.29^{a}	16.09 ± 3.22^{c}	< 0.05
IV (g I/100 g)	83.5 ± 1.5^{d}	84.1 ± 1.2^{d}	86.0 ± 1.1^{c}	116.4 ± 5.5^{b}	132.8 ± 3.4^{a}	87.9 ± 2.5^{c}	< 0.05
SN (mg KOH/g)	200.2 ± 0.5^{a}	200.4 ± 0.4^{a}	199.4 ± 0.3^{b}	197.3 ± 0.9^{c}	199.2 ± 0.3^{b}	196.7 ± 0.3^{d}	< 0.05

3.5. Fatty acid composition of the oils

The 80 oils were analysed for their FA compositions, the results of which are listed in Table 2. The raw data of the fatty acid methyl ester analysis are shown in supplementary material (Supplementary Table S2). All 17 FAs showed significant differences ($p < 0.05$) between the six oil categories, as well as for the sum of (un)saturated fatty acids (SFA, MUFA and PUFA). The three most abundant FAs in the six oil categories are C16:0, C18:1n9 and C18:2n6, which account for > 85% of total FAs.

In terms of a comparison of EVOO with all other oils, EVOO differed significantly from the other three vegetable oils (RSO, SFO and PNO) for 14 out of 17 FAs. Since lower grade olive oils are also extracted from olive fruit, they present smaller differences compared to EVOO regarding the FAs composition than compared to other vegetable oils. Nevertheless, EVOO differed significantly from the two lower grade olive oils (ROO and POO) for 4 out of 17 and 10 out of 17 FAs, respectively. Moreover, the three olive oils present significantly higher concentrations of MUFA and lower concentrations of PUFA than the other botanical origin oils.

Since FAs vary in terms of chain length as well as number of double bonds, the values of IV and SN were calculated based on FAs data and are presented in Table 2. The IV expresses the degree of unsaturation (Stavarache, Vinatoru, & Maeda, 2007). The IVs of EVOO and ROO do not differ significantly, but they do differ significantly with those of all other oils. The IVs of RSO (116.4 ± 5.5 g I/100 g) and SFO (132.8 ± 3.4 g I/100 g) are significantly higher than those of the other oils (in the range of 81–92 g I/100 g), which indicates a higher unsaturation degree of those two oils. In contrast, the IV of PNO (87.9 ± 2.5 g I/100 g) does not differ significantly from the IV of POO (86.0 ± 1.1 g I/100 g).

The value of SN is related to the chain length of FAs, a higher SN implies a short carbon chain length (Stavarache et al., 2007). No significant differences were determined between the SN values of EVOO (200.2 ± 0.5 mg KOH/g) and ROO (200.4 ± 0.4 mg KOH/g), but the SN value of POO (199.4 ± 0.3 mg KOH/g) is significantly lower than that of the other two olive oils. This result is generally consistent with a previous study (Kreps et al., 2017), which reported that high temperature treatment of oils may contribute to polymerization and formation of long-chain FAs, and this results in decreased SN values. Another possible reason is that POO contains more waxes (long chain fatty acid ester with long chain alcohol, C40-C46) than EVOO as mentioned in the IOC standard (International Olive Council, 2016). The SN values of the three olive oils are significantly higher (in the range of 198.6–201.3 mg KOH/g) than those of RSO (197.3 ± 0.9 mg KOH/g) and PNO (196.7 ± 0.3 mg KOH/g), which implies that olive oils contain shorter chain length FAs than RSO and PNO. EVOO can be separated from nearly all other types of oil studied here, except ROO, whereas an overlap can be noticed between EVOO and POO at a lower extent, based on IV and SN values in Supplementary Fig. S1.

Taken together, these results provide important insights into the FAs characteristics of six oil categories.

3.6. Relationships between ultrasonic velocity, viscosity, density and fatty acid composition

Fig. 5 presents the correlation between velocity, viscosity and density on the one hand and FAs, IV and SN on the other hand. It can be seen that viscosity correlated significantly ($p < 0.05$) with the accumulated saturated and unsaturated fatty acid contents (SFA, MUFA and PUFA). The r values of viscosity and PUFA, and viscosity and MUFA are −0.87 and 0.78, respectively. This is in agreement with studies of Santos, Santos, and Souza (2005), which indicated that the viscosity of vegetable oils is more related to the presence of polyunsaturated chains than to monounsaturated chains in an oil/fat mixture. Moreover, viscosity readings correlate more strongly with C18:1n9 and C18:2n6 ($r = 0.79$ and -0.78, respectively) than with C16:0 ($r = 0.69$). Results also indicate that the long-chain compounds correlate more strongly with the viscosity. In addition, the position of fatty acids in triacylglycerol may also influence oil viscosity according to the previous study (Snouber et al., 2019). It is reported that thermal triggered oxidation induces the formation of polar triglyceride oligopolymers, which results in an increase in viscosity.

According to a previous study (Rodenbush, Hsieh, & Viswanath, 1999), the relationship between viscosity (η) and the ratio of IV over SN can be expressed by Eq. (4):

$$\log \eta = (-1.4 + 1.25(IV/SN)) + (500 - 375(IV/SN)) /((T + 140) - 85(IV/SN)) \quad (4)$$

A significant negative correlation ($r = -0.93$, $p < 0.05$) between viscosity and IV is presented in Fig. 5, which confirms that a higher unsaturation degree results in a lower viscosity (Rodenbush et al., 1999; Santos et al., 2005; Schaschke et al., 2006). The presence of double bonds prevents the same level of intermolecular contact, resulting in an increased capability of the fluid to flow (Abramovic & Klofutar, 1998; Schaschke et al., 2006). Furthermore, viscosity and SN show a weak but significant positive correlation ($r = 0.36$, $p < 0.05$). This result indicates that the long-chain FAs result in a lower viscosity. However, the result is in contradiction with previous studies (Geller & Goodrum, 2000; Rodenbush et al., 1999). In some studies, scientists worked with pure triglycerides (Eiteman & Goodrum, 1993; Geller & Goodrum, 2000). However, natural vegetable oils are complex mixtures of many triglycerides with different chain lengths.

Density correlates significantly ($p < 0.05$) with three abundant FAs (Fig. 5). It correlates also significantly with the sum of (un)saturated FAs: density/SFA ($r = -0.61$), density/MUFA ($r = -0.72$), and density/PUFA ($r = 0.78$). Generally, the density is expected to correlate with both IV and SN (Rodenbush et al., 1999), according to the Lund Eq. (5):

$$sg\ (15/15°C) = 0.8475 + 0.00030\ SN + 0.00014\ IV, \quad (5)$$

where sg is the specific gravity of the vegetable oil at 15 °C.

The significant positive correlation between density and IV ($r = 0.81$, $p < 0.05$; Fig. 5) confirms that density increases with increasing degree of unsaturation. A significant negative correlation ($r = -0.56$, $p < 0.05$) between density and SN was determined, which means that density increases with the decreasing saponification number, as well as with increasing chain length of FAs.

Those results demonstrate that the FA composition plays an important role when it comes to the viscosity and density of oils. Moreover, the differences in viscosity and density are more strongly related to the unsaturation degree than to the chain length of FAs. According to the correlation results in Section 3.6, a higher unsaturation degree of oils results in a lower viscosity and higher density. Moreover, a higher ultrasonic velocity is related to a lower viscosity, and a higher density of oils (Section 3.4). Consequently, a higher unsaturation degree of oils indirectly results in a higher ultrasonic velocity. This is also supported by the significant positive correlation between velocity and IV ($r = 0.80$, $p < 0.05$; Fig. 5).

3.7. Prediction of the velocity from the oils' compositions

PLS regression was applied to determine the capability of FAs for prediction of the ultrasonic velocities. The data were pre-processed involving auto-scaling, mean-centering, and range-scaling, and models with the three types of pre-processing were compared. The prediction capabilities of the three corresponding regression models are presented in Table 3.

In general, a low SEV and high r^2 present better prediction ability of a model (Fernandez-Cabanas et al., 2011). For these three variants of

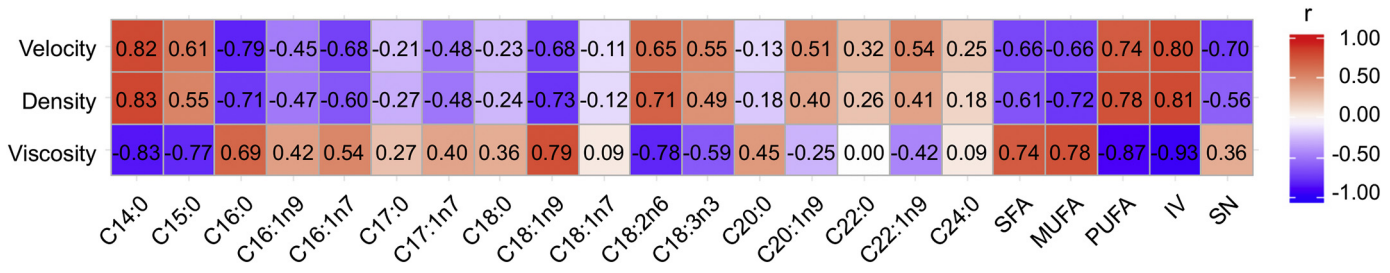


Fig. 5. Pearson correlation coefficients for the correlation of the compositions of the oils and their physical properties. Red refers to positive correlation; blue refers to negative correlation. The correlation coefficients in each spot, described the strength of the correlation between two variables. The larger the absolute value of correlation coefficient, the darker the colour. Correlation coefficients < -0.22 and $> +0.22$ are significant (Paired samples *t*-test, $p < 0.05$). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Performance characteristics of PLS regression models predicting the ultrasonic velocity by fatty acid compositions of 80 vegetable oil samples with leave-one-out cross validation and different pre-processing methods.

Pre-processing	Factor	SEC	R^2	SEV	r^2
Mean-centering	1	1.00	0.93	1.01	0.93
Auto-scaling	3	1.01	0.93	1.07	0.92
Range-scaling	4	1.10	0.92	1.20	0.90

Factor, optimal factor of each model; SEC, standard error of calibration; R^2, correlation coefficient of calibration; SEV, standard error of cross-validation; r^2, correlation coefficient of cross-validation.

PLS regression, best prediction ability is observed with mean-centering pre-processing, although the prediction ability of the three models varies only to a limited extent. The best model presents an r^2 of 0.93 and SEV of 1.01. This regression model is shown in Supplementary Fig. S2. Therefore, this model allows a reasonable estimation, and an indication of the expected velocity for various vegetable oils from their FA compositions.

4. Conclusions and outlook

This study reveals that the developed ultrasonic measurement system is a rapid and non-destructive technique which can be applied for the characterization of vegetable oils. The ultrasonic measurements are temperature dependent, and the temperature coefficient of the velocity in EVOO is $-2.92\,m{\cdot}s^{-1}{\cdot}°C^{-1}$. The ultrasonic velocity of EVOO (1453 ± 2 m/s, at 23.5 °C) differed significantly from POO and other vegetable oils, except for ROO. The differences in ultrasonic velocity are primarily due to the level of unsaturation. Prediction of the ultrasonic velocity from the FA composition is promising.

Since the ultrasonic technique is rapid and non-destructive, it is an interesting approach to be included in the EVOO characterization toolbox. In addition, simultaneous measurement of the ultrasonic attenuation and temperature coefficients of vegetable oils can offer further valuable information of the ultrasonic properties, which could be another method for the characterization of vegetable oils.

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