



# Authentication of pure and adulterated edible oils using non-destructive ultrasound

A. Jiménez<sup>a</sup>, M. Rufo<sup>a</sup>, J.M. Paniagua<sup>a</sup>, A. González-Mohino<sup>b,\*</sup>, L.S. Olegario<sup>b</sup>

<sup>a</sup> Department of Applied Physics, Research Institute of Meat and Meat Products, School of Technology, Universidad de Extremadura, Avenida de la Universidad s/n, 10003 Cáceres, Spain

<sup>b</sup> Department of Food Technology, Research Institute of Meat and Meat Products, Universidad de Extremadura, Avenida de la Universidad s/n, 10003 Cáceres, Spain

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## ABSTRACT

At present, the quality of edible oil is evaluated using traditional analysis techniques that are generally destructive. Therefore, efforts are being made to find alternative methods with non-destructive techniques such as Ultrasound. This work aims to confirm the feasibility of non-destructive ultrasonic inspection to characterise and detect fraudulent practices in olive oil due to adulteration with two other edible vegetable oils (sunflower and corn). For this purpose, pulsed ultrasonic signals with a frequency of 2.25 MHz have been used. The samples of pure olive oil were adulterated with the other two in variable percentages between 20% and 80%. Moreover, the viscosity and density values were measured. Both these physicochemical and acoustic parameters were obtained at 24 °C and 30 °C and linearly correlated with each other. The results indicate the sensitivity of the method at all levels of adulteration studied. The responses obtained through the parameters related to the components of velocity, attenuation, and frequency of the ultrasonic waves are complementary to each other. This allows concluding that the classification of pure and adulterated oil samples is possible through non-destructive ultrasonic inspection.

## 1. Introduction

Edible oils have economical and nutritional benefits. Although there is a wide range of types on the market, there are premium varieties such as olive oil (Salah & Nofal, 2021). Adulteration in food matrices is a frequent problem in the current food industry (Tan et al., 2021). In particular, adulteration of edible oils is considered a threat to public health (Karami et al., 2020) due to reduced nutritional quality, greater susceptibility to oxidation and the possibility of forming unhealthy substances (Lozano-Castellón et al., 2022). One of the most common fraudulent practices is the substitution of expensive ingredients for cheaper ones, such that higher-priced and valuable oils are mixed with lower-priced and valued oils (Tan et al., 2021). Olive oil, for example, is one of the food products most likely to be subject to adulteration today, which can involve mixing it with other cheaper oils (e.g., refined oils of corn, sunflower, soybean, canola, palm), diluting its quality, flavour, and nutritional value (Aslam et al., 2023; Lozano-Castellón et al., 2022; Moore et al., 2012). This fact is particularly worrying not only for the health of the final consumer but also for the production and marketing companies themselves, in the sense that it not only reduces the health

benefits but also the characteristic organoleptic qualities of a certain type of oil (Vanstone et al., 2018). These fraudulent practices are also favoured by the similar visual appearance of the different types of edible oils (Ben-Ayed et al., 2013), e.g., viscosity or thickness, translucent colour, clarity and brightness. However, less frequently, color adulteration techniques are also applied, with the use of chlorophyll and  $\beta$ -carotene (Wang & Wan, 2021). Given that adulterations are becoming more and more sophisticated, their detection requires specific techniques.

Parallel to the new legal requirements aimed at mitigating food fraud, food safety certification systems, such as BRCGS Global Standard for Food Safety (BRCGS, 2022) or IFS International Food Standard (IFS, 2023), have also introduced requirements related to this issue into their schemes. In this way, one of its measures is to review and adjust the controls and monitoring requirements of raw materials. Until now, conventional techniques applied to oil quality control have been expensive and complex and require great specialisation. Thus, the most of methods used to characterise the authenticity of oils have destructive nature (Salah & Nofal, 2021), which, in addition to being tedious and slow, involve the destruction or use of a large amount of sample.

\* Corresponding author at: Universidad de Extremadura (IPROCAR), Avda Ciencia s/n, 10003 Cáceres, Spain.

E-mail address: [albertogj@unex.es](mailto:albertogj@unex.es) (A. González-Mohino).

Therefore, it is necessary to search for techniques that do not mean the destruction of samples and can be adapted to the food industry. Among these emerging techniques are image analysis applied for virgin olive oil (Alamprese et al., 2021), and spectroscopic methodologies such as near-infrared spectroscopy (NIR) applied for palm and coriander oils (Kaufmann et al., 2019, 2022), nuclear magnetic resonance (NMR), mid-infrared (MIR), and Raman techniques applied for extra virgin olive oil (Maestrello et al., 2022; Mendes et al., 2015), and visible spectroscopy applied for virgin olive oil (Ferreiro-González et al., 2017). Spectroscopic methods are fast and non-destructive and require little or no preparation and do not use solvents or produce waste. The main disadvantages of these methods may be the inability to detect trace compounds, the need to build a model for chemometric prediction with intensive analytical work (Aslam et al., 2023). In addition to these techniques, ultrasound inspection is postulated as a suitable tool for successfully characterisation of different types of edible oils, as previous works has demonstrated (Alouache et al., 2018; Jiménez et al., 2022; Yan et al., 2019). All these proposed methodologies offer advantages such as faster analyses, sensitive, lower costs, easy handling, and automation in routine analyses, and without residues (Arendse et al., 2021) thus complying with the principles of green and white chemistry. In addition, the methods based on these techniques have the great advantage that they can be easily converted into methods that use portable devices, so that analyses can be carried out in situ, without the need to transport the samples to the laboratory (da Silva Medeiros et al., 2023; García Martín, 2022). However, in complex matrices, they provide a huge amount of information that sometimes makes the use of traditional chemometric techniques insufficient. It must also be pointed out that the validity of ultrasonic techniques in the aforementioned characterization of oils and detection of fraudulent mixtures is usually based mainly on the velocity of propagation of acoustic waves and, less frequently, on their attenuation (Alouache et al., 2015; Azman & Abd Hamid, 2017). For this reason, the recent inclusion of other ultrasonic parameters in this objective, such as those linked to the frequency components transmitted by the waves (Jiménez et al., 2022), is appreciated, since they allow refining and broadening the potential conclusions related to the purity of the samples. Thus, the greater or lesser attenuation of the ultrasonic waves when they propagate through the oil, varies not only as a function of the type of oil but also of the frequencies of the ultrasonic waves transmitted. Additionally, the physicochemical characterization of the different types of oil is of utmost importance to distinguish them. Among these parameters, two of the most recommended are density and viscosity (Reglamento, 1991). For this reason, an analysis of the density and viscosity of the oil samples studied in this work is also carried out, with the aim of carrying out a correlation study of the results from these physicochemical parameters with the ultrasonic ones.

The first objective of this work is to acoustically characterise three edible oils (olive, sunflower, corn) and different mixtures with the olive oil at two measured temperatures (24 °C and 30 °C), thus detecting the adulteration of the most expensive of them, olive oil, with different proportions of the others two. The acoustic parameters analysed are ultrasound pulse velocity (UPV), frequency variables obtained from the Fast Fourier Transform (FFT), and attenuation. The final purpose of this contribution is to complement and complete the studies carried out by Jimenez et al. (2022), expanding the capacity of non-destructive ultrasound inspection as a feasible tool to discriminate blends between these three varieties of oil and authenticate the purity of each of them separately.

## 2. Material and methods

### 2.1. Samples

Three types of edible oils were bought from the brands 'Coosur' (olive) and 'Coosol' (sunflower and corn) (Jaén, Spain), were bought in a

supermarket. Six one-litre samples of each type were purchased, all of them corresponding to different lots. Eleven batches were prepared in triplicate (33 samples in total), three of which corresponded to pure samples: i) Olive oil; ii) Sunflower oil; iii) Corn oil; and the rest by mixing these three main samples in different volumetric ratios: iv) OS (80–20) (80% Olive oil and 20% Sunflower oil); v) OS (60–40) (60% Olive oil and 40% Sunflower oil); vi) OS (40–60) (40% Olive oil and 60% Sunflower oil); vii) OS (20–80) (20% Olive oil and 80% Sunflower oil); viii) OC (80–20) (80% Olive oil and 20% Corn oil); ix) OC (60–40) (60% Olive oil and 40% Corn oil); x) OC (40–60) (40% Olive oil and 60% Corn oil); and xi) OC (20–80) (20% Olive oil and 80% Corn oil). Batches were prepared on the same opening day as the six one-litre samples. The 33 samples were inspected ultrasonically the next day. Consequently, it is to be expected that the oxidation of the samples prior to their ultrasonic inspection would be minimal.

### 2.2. Density and viscosity

As described by Sahasrabudhe et al. (2017), viscosity and density properties are suitable parameters to discriminate different types of oils. Also, as reported in previous contributions, viscosity ( $\eta$ ) and density ( $\rho$ ) are considered significant parameters in edible oils, which depend on the temperature ( $T$ ), showing their influence by the following model eqs. (1), (2) (Ghosh et al., 2017)

$$\eta = \eta_0 \cdot e^{\frac{B}{T}} \quad (1)$$

$$\rho = A \cdot T + \rho_0 \quad (2)$$

where  $A$ ,  $\eta_0$ ,  $\rho_0$ , and  $B$ , are constant characteristics of the edible oil sample.

These parameters were evaluated in triplicate and at two temperatures, 24 °C, and 30 °C, employing an incubator FTC E90 (VELP Scientifica Srl, Italy). The temperatures were chosen since they were common storage temperatures and also to observe statistical differences between the considered parameters. Viscosity was assessed utilizing a Selecta STS-2011 viscometer (J. P. SELECTA, Spain). The selected spindle was the TL5 with APM adapter at 20 rpm in 5 ml of sample. On the other hand, the density was measured using a glass and normalized density meter DIN 12791 (Laboquimia, Spain).

### 2.3. Ultrasound inspection

The ultrasound inspection was similar to the procedure followed in previous works (Jiménez et al., 2022). From each of the 33 prepared samples, an aliquot of 150 ml was introduced into a Nessler tube with 200 ml capacity. Each aliquot was heated to 30 °C in a thermostatic bath (JP SELECTA, Spain). Ultrasonic tests were performed starting at this temperature and, after a short wait while cooling to room temperature, at again 24 °C. Thus, a total of 66 ultrasonic tests were performed. The immersion technique in pulse-echo (PE) was the mode selected to inspect the samples. The piezoelectric transducer selected was the Olympus Model V305-SU with a nominal frequency of 2.25 MHz (main characteristics described in Supplementary Material 1 (SM1)). The transducer was located at the top of the Nessler tube, submerged approximately 1.5 cm in oil. The separation distance between the surface of the transducer and the bottom of the Nessler tube, which acts as a reflecting mirror for the ultrasonic waves, was 158.00 mm. This distance ensures that the measurements were made in the far field (Fraunhofer region) setup since when working in pulse-echo, the ultrasonic waves travel a minimum distance from when they are emitted until they are received of  $158.00 \times 2 = 316.00$  mm, which is much greater than the near field length (Fresnel region) indicated in table SM1. This question is not trivial when it comes to evaluating parameters linked to the attenuation and frequency of the received signal, so that, in our case, it can be ensured that wave interference phenomena do not intervene in the

evaluation of these parameters (Kinsler et al., 1999).

The Panametrics-NDT Model 5077PR Pulser-Receiver was used to emit and receive the signals from the piezoelectric transducer. This equipment was connected to a KEYSIGHT InfiniiVision DSO-X 3032A oscilloscope, in charge of registering and saving the A-scans with 10.000 points in csv format. [Supplementary Material 2 \(SM2\)](#) shows the setup used for these measurements. From each of the 66 ultrasonic tests carried out, 3 A-scans were recorded, in each of which the aim was to collect different aspects of the signal and, therefore, obtained with a different time interval ( $TI$ ) on the oscilloscope. In particular: (i)  $TI = 2000 \mu\text{s}$ , which included the trigger pulse and the first seven full echoes; (ii)  $TI = 500 \mu\text{s}$ , which included the first two full echoes; and (iii)  $TI = 250 \mu\text{s}$ , which included only the first full echo. In total, the study has encompassed the treatment of 198 A-scans. As an example, [Supplementary Material 3 \(SM3\)](#) shows three characteristic A-scans corresponding to one of the inspected aliquots.

As indicated, the ultrasonic parameters studied were: propagation velocity ( $UPV$ ); the frequency values for which 25%, 50%, and 75% ( $FFT_{25}$ ,  $FFT_{50}$ , and  $FFT_{75}$ , respectively) of the entire frequency spectrum obtained in the FFT are received; and the attenuation ( $\alpha$ ).

In particular, as previous contributions reported (Jiménez et al., 2022; Rubalya Valentina et al., 2013), the relationship between velocity ( $UPV$ ) and temperature ( $T$ ) is linear. Therefore, from equations (1) and (2) it can be unequivocally deduced that the relationship between velocity and density must also be linear and that the relationship between them must satisfy equation (3):

$$\frac{1}{UPV} = C \ln \eta + D \quad (3)$$

### 2.3.1 Determination of ultrasonic parameters

The procedures followed to calculate the aforementioned ultrasonic parameters are practically identical to those shown in previous works (Jiménez et al., 2022), so this information was notably summarised, highlighting only those aspects that differ from those indicated there. Specifically, the main novelty lies in the nominal frequency of the transducer used in this work, 2.25 MHz, compared to that used in the previous work, 1.0 MHz. In this way, when a higher inspection frequency is used, the waves are more sensitive to small alterations of the samples, as the wavelength is now on the order of 0.64 mm compared to 1.45 mm in the previous work.

Regarding  $UPV$ , two methods were employed, both using the A-scans with  $TI = 2000 \mu\text{s}$ : (i) performing least squares fit the equation of a straight line of the known distance travelled by the signal versus the time of flight for each of the seven echoes. The slope of the fit corresponds to the propagation velocity  $UPV_{lr}$ . (ii) From the so-called cepstrum method, or what is the same, apply the FFT to the FFT in turn of the corresponding A-scan, which always shows a characteristic periodic excitation from the multiple echoes caused by the successive reflections between the bottom of the Nessler tube and the surface of the transducer. This second FFT directly displays the time elapsed between these excitations (time of flight between two consecutive echoes) which, together with the known distance travelled by the ultrasonic wave between them, allows the velocity  $UPV_c$  to be calculated directly. The errors associated with the determinations of  $UPV_{lr}$  and  $UPV_c$  are on the order of 0.20% and 0.15%, respectively.

The determination of the  $FFT_{25}$ ,  $FFT_{50}$ , and  $FFT_{75}$  is based on the fact that the ultrasonic frequencies transmitted mainly in the samples do not coincide exactly with those nominally emitted by the transducer, so it makes more sense to consider those actually transmitted, which are those that appear in the associated FFT at each A-scan. From this FFT, the accumulated frequency periodogram is constructed and the values of the 25th, 50th, and 75th percentiles of the frequencies of the received signals are evaluated. However, it is necessary to establish a caveat as a consequence of the Nyquist theorem, which establishes the minimum sampling frequency  $f'$  necessary to carry out a correct study of any

frequency signal  $f$  in the frequency domain, as shown in Eq. (4) (Zhong & Zhang, 2021):

$$f' \geq 2f \quad (4)$$

Thus, when  $TI = 2000 \mu\text{s}$ ,  $f' = 10000 / (2000 \cdot 10^{-6}) = 5 \cdot 10^6 \text{ Hz}$ , which, although greater than  $2 \cdot 2.25 \cdot 10^6 = 4.5 \cdot 10^6 \text{ Hz}$ , is too close to it. For this reason, to calculate the frequency parameters, only the A-scans obtained with  $TI = 250 \mu\text{s}$  ( $FFT_{25} 1$ ,  $FFT_{50} 1$ , and  $FFT_{75} 1$ ) and  $TI = 500 \mu\text{s}$  ( $FFT_{25} 1-2$ ,  $FFT_{50} 1-2$ , and  $FFT_{75} 1-2$ ) were chosen, whose values of  $f'$  are  $4 \cdot 10^7 \text{ Hz}$  and  $2 \cdot 10^7 \text{ Hz}$ , respectively.

Finally, the attenuation coefficient  $\alpha$  (in neper/m) was calculated as Eq. (4):

$$\alpha = \frac{1}{2d} \ln \left( \frac{A_i}{A_j} \right) \quad (4)$$

where  $A_i$  and  $A_j$  are the (peak-to-peak) amplitudes of echoes  $i$  and  $j$ , respectively, and  $2d$  represents the space covered by the ultrasound wave between them. Seven echoes of the A-scan corresponding to  $TI = 2000 \mu\text{s}$  were considered, and  $\alpha$  was determined from the slope of the plot of  $\ln(A_i/A_j)$  vs.  $2d$  (Koc & Ozer, 2008).

### 2.4. Data analysis

Significant differences between the eleven batches (Olive, Sunflower, Corn, OS (80–20), OS (60–40), OS (40–60), OS (20–80), OC (80–20), OC (60–40), OC (40–60), OC (20–80)) were studied. The normality of the parameters considered was performed with the Shapiro-Wilk test. Depending on the normality, the Student-T test ( $p \leq 0.05$ ) or U Mann-Whitney test ( $p \leq 0.05$ ), or ANOVA ( $p \leq 0.05$ ) for measurements comparison between all batches tested with Tukey average test ( $p \leq 0.05$ ), was applied using the IBM SPSS v.22 (IBM Co., U.S.A) statistic software package. The correlation coefficient (Pearson's R) among viscosity and density, and ultrasound parameters, was performed by applying the XLSTAT software package (Addinsoft, France).

## 3. Results and discussion

### 3.1. Density and viscosity results

Mean values ( $\pm$ SD) of viscosity and density results for the eleven batches are displayed in [Table 1](#). As can be observed, both parameters are highly influenced by the temperature since they are emulsion systems (Rukke & Schüller, 2017). Regarding viscosity results, all batches showed significant differences between the temperatures considered, 24 °C (higher values) and 30 °C (lower values). The mean values of viscosity decreased when the temperature increased, as expected (Sahasrabudhe et al., 2017). Now considering the batches studied and starting with those corresponding to pure oils, olive oils were the ones that presented the highest viscosity values, followed by corn oils and finally sunflower oils. In general, the results shown are consistent with those indicated in the bibliography (Azman & Abd Hamid, 2017; Jiménez et al., 2022). The three pure varieties are perfectly distinguishable from each other at any of the two temperatures. When considering the remaining batches corresponding to samples of two mixed varieties, their viscosity values obtained are between the values of each of them in a pure state, approximately proportional to their presence in the mixture. Thus, as the proportion of sunflower oil increases and that of olive oil decreases, the viscosity also decreases. And identically for the olive-corn mixture. It is noteworthy that pure olive oil constitutes a single category for both temperatures with the adulteration proportions of corn and sunflower studied in this work. In fact, only when the proportion of olives is the minimum studied (20%), the viscosity values are similar to those of the cheapest pure oils. For all other proportions of olive oil, the categories are perfectly distinguishable, as long as we consider adulteration separately with sunflower or corn. If not, there are

**Table 1**

Viscosity ( $\eta$ ) and density ( $\rho$ ) results (means  $\pm$  SD)\* for the eleven batches (Olive, OS (80–20), OS (60–40), OS (40–60), OS (20–80), Sunflower, OC (80–20), OC (60–40), OC (40–60), OC (20–80), Corn) for 24 and 30 °C.

	$\eta_{24}$ (cP)	$\eta_{30}$ (cP)	$\rho_{24}$ (kg/m <sup>3</sup> )	$\rho_{30}$ (kg/m <sup>3</sup> )
Olive	70.1 $\pm$ 0.5 <sup>hB</sup>	63.6 $\pm$ 1.1 <sup>fA</sup>	910 $\pm$ 0.4 <sup>aA</sup>	907 $\pm$ 0.7 <sup>aB</sup>
OS (80–20)	67.2 $\pm$ 0.2 <sup>dB</sup>	58.8 $\pm$ 0.6 <sup>dA</sup>	910 $\pm$ 0.3 <sup>aA</sup>	907 $\pm$ 0.6 <sup>aB</sup>
OS (60–40)	62.5 $\pm$ 0.4 <sup>dB</sup>	55.8 $\pm$ 0.3 <sup>cA</sup>	911 $\pm$ 0.5 <sup>bB</sup>	908 $\pm$ 0.2 <sup>bA</sup>
OS (40–60)	60.1 $\pm$ 0.3 <sup>bB</sup>	53.5 $\pm$ 0.7 <sup>aA</sup>	912 $\pm$ 0.5 <sup>cB</sup>	909 $\pm$ 0.5 <sup>cA</sup>
OS (20–80)	59.6 $\pm$ 0.7 <sup>abB</sup>	52.9 $\pm$ 0.4 <sup>aA</sup>	914 $\pm$ 0.2 <sup>dB</sup>	911 $\pm$ 0.3 <sup>dA</sup>
Sunflower	58.3 $\pm$ 0.6 <sup>aB</sup>	52.7 $\pm$ 0.5 <sup>aA</sup>	915 $\pm$ 0.3 <sup>dB</sup>	912 $\pm$ 0.6 <sup>eA</sup>
OC (80–20)	68.1 $\pm$ 0.5 <sup>dB</sup>	60.3 $\pm$ 0.6 <sup>eA</sup>	911 $\pm$ 0.7 <sup>bb</sup>	908 $\pm$ 0.1 <sup>bA</sup>
OC (60–40)	66.1 $\pm$ 0.6 <sup>dB</sup>	58.7 $\pm$ 0.4 <sup>dA</sup>	912 $\pm$ 0.3 <sup>cB</sup>	910 $\pm$ 0.8 <sup>dA</sup>
OC (40–60)	63.6 $\pm$ 0.8 <sup>dB</sup>	56.4 $\pm$ 0.5 <sup>cA</sup>	914 $\pm$ 0.8 <sup>dB</sup>	912 $\pm$ 0.7 <sup>eA</sup>
OC (20–80)	61.2 $\pm$ 0.7 <sup>cB</sup>	54.3 $\pm$ 0.3 <sup>bA</sup>	916 $\pm$ 0.5 <sup>bB</sup>	914 $\pm$ 0.3 <sup>fA</sup>
Corn	61.6 $\pm$ 0.7 <sup>cB</sup>	54.1 $\pm$ 0.7 <sup>bA</sup>	917 $\pm$ 0.6 <sup>dB</sup>	915 $\pm$ 0.4 <sup>gA</sup>

\* Different superscript lowercase letters in each column mean the significant differences between oil type. Capital letters mean temperature effect and lowercase letters mean type effect. ( $p \leq 0.05$ ).

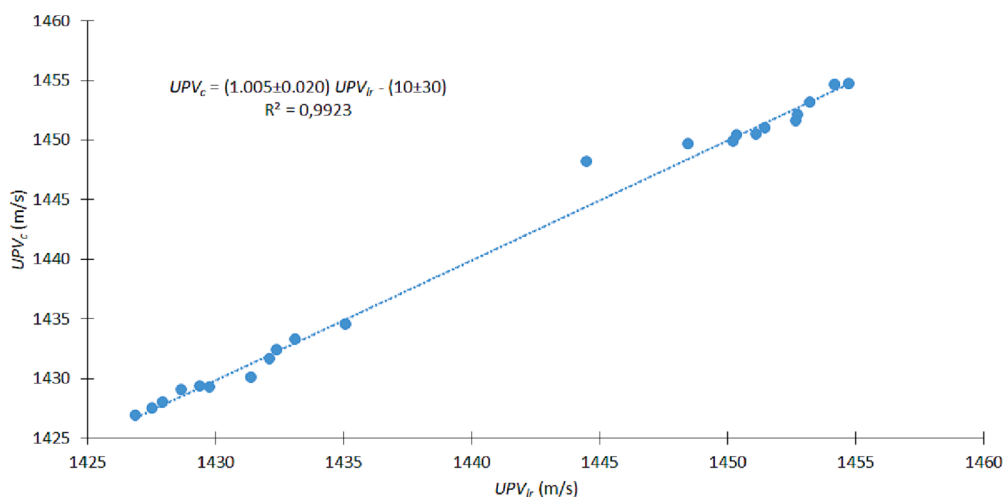
indistinguishable batches, such as OS (60–40) and OC (40–60).

Concerning density mean values, the temperature was also highly influential, showing significant results among 24 °C (higher values) and 30 °C (lower values). The corn oils presented higher density values than the ten remaining batches, so it is significantly different from both the pure batches (sunflower and corn oils) and the mixed ones. Considering the pure oils, after those of corn, were those of sunflower and finally those of olive. The values obtained are also consistent with others found in the literature (Alouache et al., 2018; Azman & Abd Hamid, 2017). The three batches are perfectly distinguishable from each other based on the density at any of the two temperatures. And again, when considering the remaining batches corresponding to samples of two mixed varieties, the density values obtained are situated between the values of each of them in a pure state, in approximate proportion to their presence in the mixture. Thus, as the proportion of corn oil increases and that of olive oil decreases, the density also increases. The same applies to for the olive-sunflower mixture. However, the adulteration of olive oil with 20% sunflower oil is not detected by density at either of the two temperatures. Instead, regarding the combinations of olive and corn oil, all the batches prepared can be classified into different categories. But again, if olive oil adulterations with sunflower and corn are considered together, numerous batches are indistinguishable from one another.

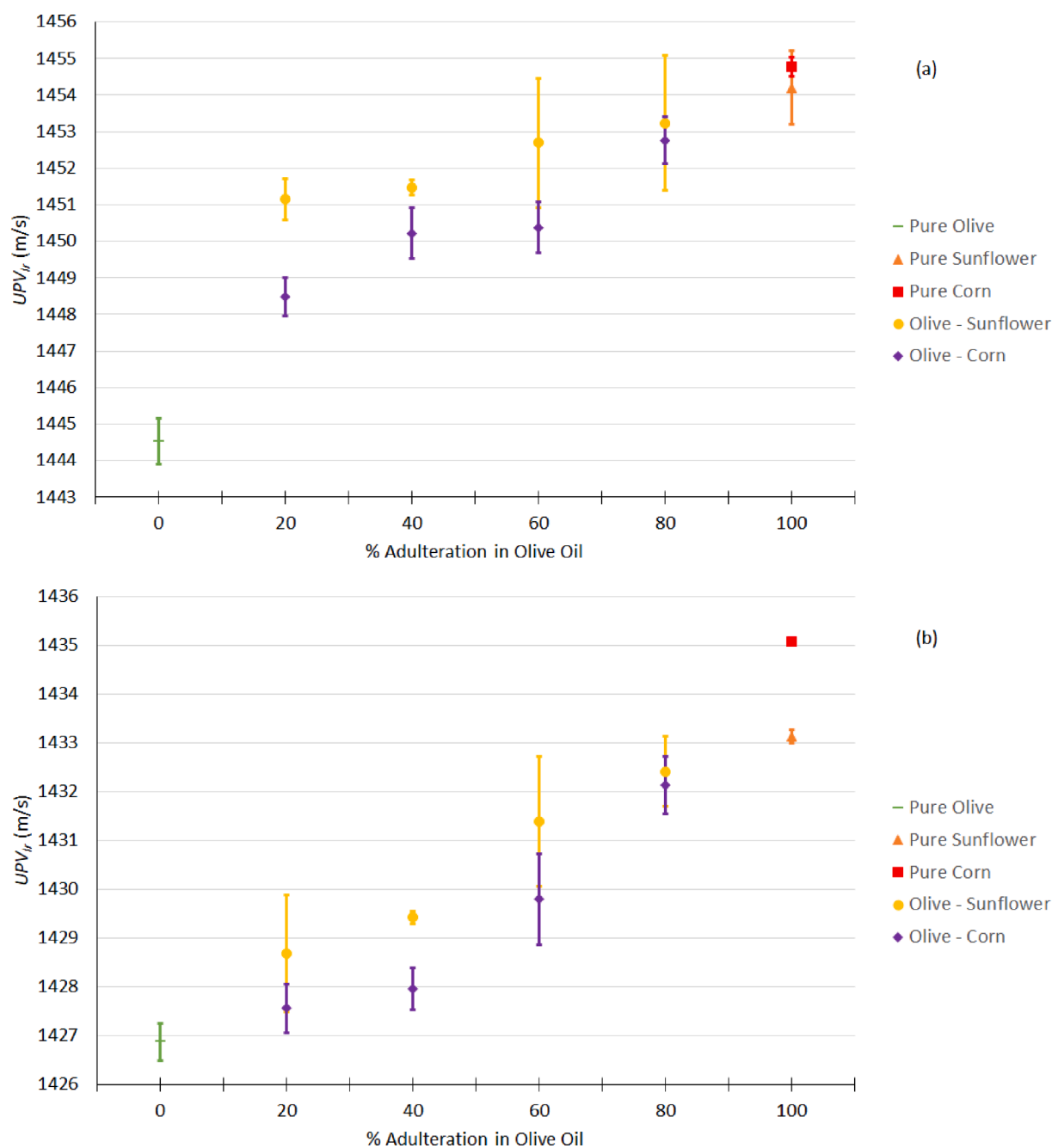
### 3.2. Ultrasound inspection results

Fig. 1 shows the relationship between the propagation velocities obtained by both methods ( $UPV_{Ir}$  and  $UPV_c$ ) in the samples inspected at

both temperatures. As can be seen, there is a highly significant linear relationship between the two, with a slope that, considering the margins of error, encompasses the value 1. Because of carrying out the ultrasonic inspection at two different measurement temperatures, the expression of the results was concentrated in two regions of the graph. This same behaviour was obtained by previous works (Jiménez et al., 2022) for transducers of different frequencies (1 MHz) at different temperatures (24 °C and 29 °C), being the correlations obtained perfectly valid. Intermediate experimental points can be obtained for ultrasonic inspection within the studied temperature range, which can help future prediction studies. This result, in addition to certifying the validity of the measurement procedures followed in both methods, makes it possible to carry out the following studies involving velocity from only one of them, either  $UPV_{Ir}$  or  $UPV_c$ . Therefore, Fig. 2 shows the  $UPV_{Ir}$  values for both temperatures in the adulteration batches of olive oil with sunflower and corn. As it was indicated, the results are practically identical considering  $UPV_c$ . Both the velocity values obtained for the pure samples at each temperature and their evolution are consistent with those found in the literature, as cited in Jiménez et al. (2022). It should only be noted that because the frequency of inspection (2.25 MHz) in this work is higher than that used by Jiménez et al. (2022) (1 MHz), the speed values are slightly lower. This is logical, as the frequency is higher, the wavelength of the ultrasound used now is shorter and, consequently, the signal is likely to take longer to travel identical distances or, equivalently, to travel at a lower speed. In this way, the velocities showed the same behaviour as density or viscosity, in the sense that their values are greater at 24 °C than at 30 °C. The lower speed value of olive oil



**Fig. 1.** Evolution of the ultrasonic velocity values ( $UPV_{Ir}$  and  $UPV_c$ ) obtained by both procedures in the samples inspected at both temperatures and linear regression fit between them.



**Fig. 2.** Evolution of ultrasonic velocity values  $UPV_{lr}$  versus percentage of adulteration in olive oil with sunflower and corn oils at (a)  $T = 24\text{ }^{\circ}\text{C}$  and (b)  $T = 30\text{ }^{\circ}\text{C}$ . Error bars indicate standard deviation.

compared to sunflower and corn oil at both temperatures also seems clear. Between these last two, the speeds are closer, being slightly higher in the case of corn, particularly noticeable at  $30\text{ }^{\circ}\text{C}$ .

Regarding the adulterated samples, it can be observed that at each temperature the velocity values obtained are distributed between the extreme values of the corresponding pure oils more or less proportionally to the percentages of adulteration. In this way, and taking as an example the adulteration of olive oil with sunflower oil, the velocity of propagation of the samples gradually increases with the percentage of sunflower oil present. And identically with corn one. In addition, at  $24\text{ }^{\circ}\text{C}$ , the velocity of pure olive oil is perfectly distinguishable from those adulterated with any percentage of sunflower or corn if we consider the associated errors, including the minimum (20%) and therefore, it is possible to detect any blended adulteration (density did not allow this

discrimination). The same can be stated at  $30\text{ }^{\circ}\text{C}$  with the exception that the velocities of pure olive oil and that adulterated with 20% corn overlap each other. Also noteworthy is the fact that all the adulteration percentages with corn oil are distinguishable from each other at  $24\text{ }^{\circ}\text{C}$ , except those corresponding to 40% and 60%, whose values overlap. This also includes pure corn oil, whose velocity is clearly higher than that of olive oil, mostly adulterated with 80% corn oil. The same behaviour is observed at  $30\text{ }^{\circ}\text{C}$ , except that in this case the olive adulterations with 20% and 40% corn are the only ones whose velocity values overlap each other. Regarding the adulteration of olive oil with sunflower oil, overlaps between velocity values for the different percentages are more frequent. Therefore, at  $24\text{ }^{\circ}\text{C}$ , the velocity values in samples adulterated with more than 60%, including pure sunflower oil, are very similar to each other. And identically the adulterated ones with percentages of

20% and 40%. Although to a lesser extent, similar considerations can be established for these adulterations at 30 °C. Finally, it should be indicated that, in general, the adulterations of olive oil with sunflower have shown velocity values slightly higher than those carried out with corn, although the error margins obtained only allow this difference to be clearly established when the percentage of adulteration was 40% at either of the two study temperatures or 20% for the one carried out at 24 °C.

As was the case with the velocities, the 25th, 50th and 75th percentiles of the frequency components obtained by both procedures ( $FFT_{25,50,75} 1$  and  $FFT_{25,50,75} 1-2$ ) are very significantly related to each other, as can be deduced from Fig. 3. In this case, although the values of the slope practically coincide with the unit, the large negative y-intercept values for all three fits suggest that for the frequency ranges shown, the frequency components obtained for echoes 1 and 2 together are slightly lower than those obtained from the first echo alone. This result was expected given the higher attenuation that higher frequency waves experience when propagating through any medium, so that the frequency components received when studying a longer path of the waves (echoes 1 and 2) are lower than those received on a shorter path (echo 1). This result, in addition to certifying the validity of the measurement procedures followed in both methods, again allows the following studies to be carried out based on the frequency components of only one of them, either  $FFT_{25,50,75} 1$  or  $FFT_{25,50,75} 1-2$ . Thus, Table 2 shows the result of the frequency parameters obtained from the first two echoes in the eleven batches. As it has explained, the results obtained from echo 1 alone are practically the same. As can be seen, pure olive oil transmits clearly lower frequency components than sunflower and corn oil, which are particularly similar at 24 °C. This result basically coincides with those of previous studies (Jiménez et al., 2022), which, although they were carried out with a 1 MHz frequency transducer, also point to a lower value of the frequency components transmitted by pure olive oil at 25 °C and 29 °C compared to those transmitted by those of sunflower and corn.

When the adulterated olive samples are studied, it is observed how, once again, the frequency components are distributed among the extreme values of the pure samples in the same order as the percentage of adulteration of sunflower or corn, although without being exactly proportional. In addition, another thing that draws attention is the fact that the number of categories established in the classification decreases as higher frequency components are considered. This fact leads us to think that when studying the adulteration of olive oil with other oils, it is

convenient to pay attention to the transmitted frequency components of lower value. In this way, in the adulteration of olive oil with sunflower oil, pure olive oil cannot be clearly distinguished until the mixture does not have a sunflower proportion greater than 40%, as can be deduced from the categorization established by the minor frequency components, in our case  $FFT_{25} 1-2$  both at 24 °C and 30 °C. On the other hand, when its adulteration with corn oil is studied, it can be detected since the mixture has a corn proportion of 20%. However, for the latter, it is more difficult to clearly distinguish what percentage of corn adulteration it has, especially if it is attempted to be carried out according to the higher frequency components, that is,  $FFT_{75} 1-2$ . In addition, the study of the lowest frequency components at 30 °C does make it possible to distinguish pure sunflower or corn oils from those that contain 20% olive oil, which was impossible based on the viscosity data. At this point, it is necessary to highlight the complementarity of the results offered by the velocity and frequency studies. Thus, for example, frequency studies are able to distinguish the adulteration of olive oil with corn since the percentage is 20%, something that speed studies are unable to do at 30 °C. On the contrary, those of speed distinguish the adulteration with sunflowers from 20%, while those of frequency are incapable.

Finally, it should be noted that, with the same percentage of adulteration of olive oil with sunflower or corn, the former are the ones that present lower frequency components at either of the two study temperatures. This fact is distinguishable when the percentages of adulteration are low, that is, 20% and 40%.

Table 2 also displays the parameters based on attenuation at both temperatures. Firstly it must be pointed out that these values are much higher than those obtained in previous works (Jiménez et al., 2022). This fact is a consequence of the higher frequency used in this work (2.25 MHz) compared to previous one (1 MHz). In this way, when using higher frequencies, it was expected that the attenuation would increase notably, as it has happened. And higher attenuation values are also obtained at 30 °C than at 24 °C, as occurred in the aforementioned previous work, with the exception that in the latter the temperatures were 25 °C and 29 °C as indicated there. The cause of this result must be found in the fact that a greater friction (greater viscosity) would be equivalent to a greater binding force between the particles, so it would be transmitted from one particle to another with less energy loss and, therefore, with less attenuation.

Regarding the batches of pure oils, the attenuation was significantly different between the type of olive and those of sunflower and corn at both temperatures, obtaining significantly lower values in pure olive oil.

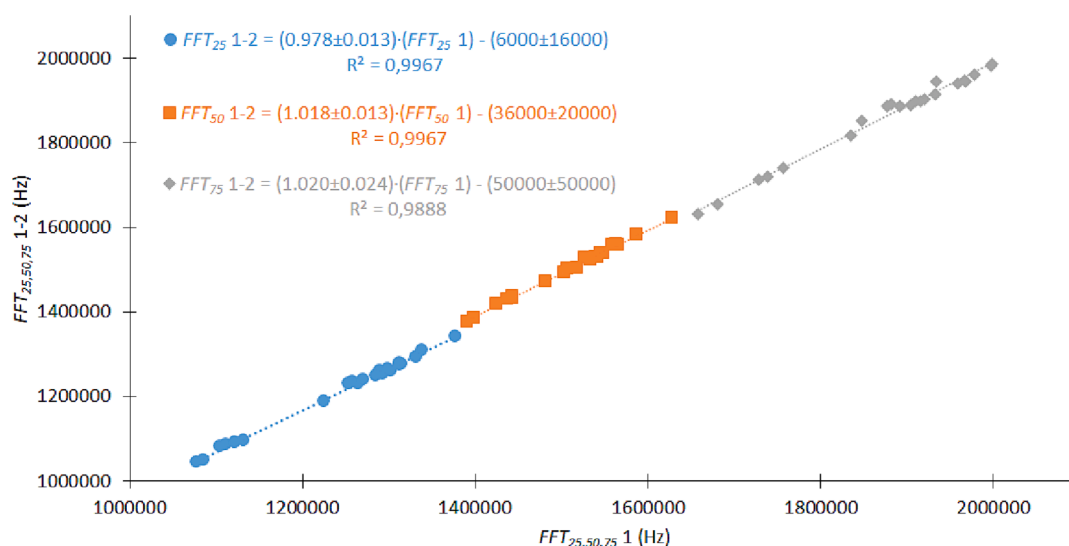


Fig. 3. Evolution of the 25th, 50th, and 75th percentiles of the frequency components ( $FFT_{25,50,75} 1$  and  $FFT_{25,50,75} 1-2$ ) obtained by both procedures in the samples inspected at both temperatures and linear regression fits between them.

**Table 2**

$FFT_{xx}$ ,  $1-2_T$  corresponding to the first two echoes at temperatures  $T = 24\text{ }^\circ\text{C}$  or  $30\text{ }^\circ\text{C}$  and attenuation  $\alpha_T$  at temperatures  $T = 24\text{ }^\circ\text{C}$  or  $30\text{ }^\circ\text{C}$  results (mean values  $\pm$  SD)\* for the eleven batches (Olive, OS (80–20), OS (60–40), OS (40–60), OS (20–80), Sunflower, OC (80–20), OC (60–40), OC (40–60), OC (20–80), Corn).

	Olive	Sunflower	Corn	OS (80–20)	OS (60–40)	OS (40–60)	OS (20–80)	OC (80–20)	OC (60–40)	OC (40–60)	OC (20–80)
$FFT_{25}$ 1–2 <sub>24</sub>	1045 $\pm$ 14 <sup>a</sup>	1279 $\pm$ 25 <sup>c</sup>	1263 $\pm$ 40 <sup>de</sup>	1049 $\pm$ 9 <sup>ab</sup>	1090 $\pm$ 11 <sup>b</sup>	1229 $\pm$ 17 <sup>cd</sup>	1260 $\pm$ 13 <sup>de</sup>	1187 $\pm$ 25 <sup>c</sup>	1229 $\pm$ 12 <sup>cd</sup>	1238 $\pm$ 10 <sup>de</sup>	1249 $\pm$ 2 <sup>de</sup>
$FFT_{50}$ 1–2 <sub>24</sub>	1377 $\pm$ 13 <sup>a</sup>	1557 $\pm$ 14 <sup>e</sup>	1538 $\pm$ 32 <sup>de</sup>	1386 $\pm$ 18 <sup>a</sup>	1435 $\pm$ 14 <sup>b</sup>	1493 $\pm$ 17 <sup>cd</sup>	1529 $\pm$ 20 <sup>de</sup>	1472 $\pm$ 15 <sup>c</sup>	1504 $\pm$ 23 <sup>cde</sup>	1528 $\pm$ 19 <sup>de</sup>	1538 $\pm$ 18 <sup>e</sup>
$FFT_{75}$ 1–2 <sub>24</sub>	1631 $\pm$ 58 <sup>a</sup>	1960 $\pm$ 101 <sup>c</sup>	1946 $\pm$ 48 <sup>c</sup>	1720 $\pm$ 84 <sup>ab</sup>	1741 $\pm$ 55 <sup>ab</sup>	1852 $\pm$ 79 <sup>bc</sup>	1944 $\pm$ 52 <sup>c</sup>	1887 $\pm$ 69 <sup>c</sup>	1887 $\pm$ 80 <sup>bc</sup>	1888 $\pm$ 54 <sup>c</sup>	1903 $\pm$ 77 <sup>c</sup>
$\alpha_{24}$	37.3 $\pm$ 0.7 <sup>a</sup>	46.8 $\pm$ 0.9 <sup>d</sup>	45.4 $\pm$ 1.0 <sup>cd</sup>	38.7 $\pm$ 1.7 <sup>a</sup>	40.3 $\pm$ 1.6 <sup>ab</sup>	42.4 $\pm$ 0.9 <sup>bc</sup>	44.8 $\pm$ 0.9 <sup>cd</sup>	43.2 $\pm$ 0.9 <sup>bc</sup>	43.6 $\pm$ 0.7 <sup>c</sup>	43.9 $\pm$ 0.8 <sup>cd</sup>	44.4 $\pm$ 0.8 <sup>cd</sup>
$FFT_{25}$ 1–2 <sub>30</sub>	1080 $\pm$ 7 <sup>a</sup>	1342 $\pm$ 6 <sup>g</sup>	1310 $\pm$ 15 <sup>f</sup>	1085 $\pm$ 17 <sup>ab</sup>	1095 $\pm$ 6 <sup>b</sup>	1275 $\pm$ 10 <sup>def</sup>	1292 $\pm$ 9 <sup>f</sup>	1234 $\pm$ 8 <sup>c</sup>	1253 $\pm$ 12 <sup>cd</sup>	1261 $\pm$ 9 <sup>de</sup>	1273 $\pm$ 5 <sup>e</sup>
$FFT_{50}$ 1–2 <sub>30</sub>	1418 $\pm$ 14 <sup>a</sup>	1621 $\pm$ 18 <sup>d</sup>	1582 $\pm$ 14 <sup>c</sup>	1430 $\pm$ 26 <sup>a</sup>	1432 $\pm$ 20 <sup>a</sup>	1530 $\pm$ 13 <sup>bc</sup>	1556 $\pm$ 14 <sup>c</sup>	1502 $\pm$ 14 <sup>b</sup>	1523 $\pm$ 20 <sup>b</sup>	1529 $\pm$ 11 <sup>b</sup>	1558 $\pm$ 14 <sup>c</sup>
$FFT_{75}$ 1–2 <sub>30</sub>	1655 $\pm$ 75 <sup>a</sup>	1982 $\pm$ 42 <sup>d</sup>	1986 $\pm$ 59 <sup>d</sup>	1713 $\pm$ 46 <sup>ab</sup>	1817 $\pm$ 59 <sup>bc</sup>	1898 $\pm$ 55 <sup>cd</sup>	1944 $\pm$ 64 <sup>d</sup>	1891 $\pm$ 78 <sup>cd</sup>	1897 $\pm$ 98 <sup>cd</sup>	1914 $\pm$ 68 <sup>cd</sup>	1939 $\pm$ 100 <sup>cd</sup>
$\alpha_{30}$	38.9 $\pm$ 0.7 <sup>a</sup>	48.4 $\pm$ 2.7 <sup>d</sup>	45.9 $\pm$ 1.7 <sup>cd</sup>	39.5 $\pm$ 0.7 <sup>a</sup>	40.8 $\pm$ 0.9 <sup>b</sup>	43.4 $\pm$ 0.7 <sup>c</sup>	47.3 $\pm$ 2.1 <sup>d</sup>	44.6 $\pm$ 2.2 <sup>cd</sup>	45.1 $\pm$ 1.6 <sup>cd</sup>	45.2 $\pm$ 1.6 <sup>cd</sup>	45.5 $\pm$ 1.7 <sup>cd</sup>

\* Different superscript letters on each row mean significant differences between oil ( $p \leq 0.05$ ).

Regarding mixed batches, it can be seen how the adulteration of olive oil with corn is detectable at both temperatures from the lowest percentage (20%). In contrast, this does not occur with the one carried out with sunflower, which is detectable from 40% only at  $30\text{ }^\circ\text{C}$ . Although of less economic interest, the adulterations of corn or sunflower oil with olive oil are not detectable from the attenuation values obtained. In particular, the attenuation values of the batches that have any percentage of corn oil at any temperature are practically indistinguishable from each other if it is considered the margins of error. The same does not occur with the sunflower ones. Thus, at  $24\text{ }^\circ\text{C}$ , the sunflower type was different with all blended batches of OS, except OS (20–80). Even at  $30\text{ }^\circ\text{C}$  ultrasonic attenuation can discriminate the degree of mixing of any of the OS batches. This last discrimination, for example, could not be made when the ultrasonic parameters were velocity or frequency components or even viscosity, thus demonstrating the complementarity of all the ultrasonic parameters included in this work when categorising pure and adulterated oil samples.

Finally, the results obtained highlight the sensitivity of different ultrasonic parameters in the inspection of oils and demonstrate how their behavior varies in relation to different samples. This indicates that the variability of the samples influences the results obtained. To better understand the association of these results with different physical characteristics, such as density and viscosity, a correlation study was performed.

### 3.3. Correlation study

Table 3 displays the linear correlation coefficients among the acoustic and non-acoustic parameters obtained in the eleven batches at  $24\text{ }^\circ\text{C}$  and  $30\text{ }^\circ\text{C}$ . All correlations between physicochemical ( $\eta$  and  $\rho$ ) and ultrasonic (UPV,  $FFT_{25}$ ,  $FFT_{50}$ ,  $FFT_{75}$ , and  $\alpha$ ) parameters were statistically significant, maintaining positive and negative correlations regardless of the measurement temperature.

As far as propagation velocity is concerned, the significant positive linear relationships of UPV with  $\rho$  and of  $1/UPV$  with  $\ln \eta$  were expected based on what is indicated in the methodology (Section 2.3). Furthermore, for the range of temperatures and viscosities obtained in our work,

if UPV is considered instead of its inverse and/or  $\eta$  instead of  $\ln \eta$ , the linear fits also work perfectly. Regarding the frequency components, the linear correlations found with density (positive) and with viscosity (negative) suggest that the denser and less viscous samples are those that favour the presence of higher frequency components in the oil samples. Finally, the significant correlations of attenuation with density (positive) and with viscosity (negative) indicate the greater acoustic attenuation experienced by ultrasonic waves in denser or less viscous samples. This fact results in what was stated above, that is, higher viscosity values (or lower density) imply greater friction or greater bonding between the particles so that the loss of energy (attenuation) when the wave passes from some particles to others is less. All these results were similar to those reported by Jimenez et al. (2022), now extended to the new batches of blended oils considered in the present work.

Significant correlations between the acoustic and physicochemical variables could allow the building of predictive models that provide a deeper understanding of the relationships between these parameters. Further literature contributions should explore this ultrasonic prediction nature of edible oils. In addition, in view of the results, the ultrasounds are adapted to the requirements described by Salah & Nofal (2021) being an accurate, simple, and cost-effective technique tool for evaluation and adulterant detection of edible oils which is highly demanded by consumers.

## 4. Conclusions

Ultrasonic inspection allowed to distinguish and categorise the three types of pure oils (olive, sunflower, and corn) studied in this work at two different measurement temperatures ( $24\text{ }^\circ\text{C}$  and  $30\text{ }^\circ\text{C}$ ). To a great extent, it also allows for discrimination of the adulteration of olive oil with various percentages of the other two types. For this aim, it is necessary to consider not only the values of ultrasonic speed, but also those of attenuation and those of other parameters linked to the frequency components, since all of them offer complementary results that, together, make such a classification possible. In addition, all ultrasonic parameters studied at either of the two temperatures have a significant linear correlation with viscosity and density in all batches (pure and

**Table 3**

Coefficients\* of linear correlations among non-acoustic and acoustic parameters at 24 and  $30\text{ }^\circ\text{C}$ .

	$\eta_{24}$	$\ln \eta_{24}$	$\rho_{24}$		$\eta_{30}$	$\ln \eta_{30}$	$\rho_{30}$
$UPV_{lr24}$	<b>-0.90</b>	<b>-0.89</b>	<b>0.72</b>	$UPV_{lr30}$	<b>-0.85</b>	<b>-0.85</b>	<b>0.82</b>
$UPV_{c24}$	<b>-0.87</b>	<b>-0.87</b>	<b>0.81</b>	$UPV_{c30}$	<b>-0.82</b>	<b>-0.83</b>	<b>0.80</b>
$1/UPV_{lr24}$	<b>0.90</b>	<b>0.89</b>	–	$1/UPV_{lr30}$	<b>0.85</b>	<b>0.86</b>	–
$1/UPV_{c24}$	<b>0.87</b>	<b>0.87</b>	–	$1/UPV_{c30}$	<b>0.82</b>	<b>0.83</b>	–
$FFT_{25}$ 1–2 <sub>24</sub>	<b>-0.72</b>	<b>-0.72</b>	<b>0.83</b>	$FFT_{25}$ 1–2 <sub>30</sub>	<b>-0.70</b>	<b>-0.70</b>	<b>0.78</b>
$FFT_{50}$ 1–2 <sub>24</sub>	<b>-0.76</b>	<b>-0.75</b>	<b>0.87</b>	$FFT_{50}$ 1–2 <sub>30</sub>	<b>-0.73</b>	<b>-0.73</b>	<b>0.82</b>
$FFT_{75}$ 1–2 <sub>24</sub>	<b>-0.70</b>	<b>-0.69</b>	<b>0.84</b>	$FFT_{75}$ 1–2 <sub>30</sub>	<b>-0.80</b>	<b>-0.79</b>	<b>0.83</b>
$FFT_{25}$ 1–2 <sub>24</sub>	<b>-0.72</b>	<b>-0.72</b>	<b>0.82</b>	$FFT_{25}$ 1–2 <sub>30</sub>	<b>-0.68</b>	<b>-0.68</b>	<b>0.78</b>
$FFT_{50}$ 1–2 <sub>24</sub>	<b>-0.76</b>	<b>-0.76</b>	<b>0.87</b>	$FFT_{50}$ 1–2 <sub>30</sub>	<b>-0.72</b>	<b>-0.72</b>	<b>0.83</b>
$FFT_{75}$ 1–2 <sub>24</sub>	<b>-0.69</b>	<b>-0.68</b>	<b>0.79</b>	$FFT_{75}$ 1–2 <sub>30</sub>	<b>-0.77</b>	<b>-0.76</b>	<b>0.81</b>
$\alpha_{24}$	<b>-0.72</b>	<b>-0.72</b>	<b>0.83</b>	$\alpha_{30}$	<b>-0.66</b>	<b>-0.66</b>	<b>0.75</b>

\* Values in bold mean statistical significance ( $p < 0.05$ ).

blended) of oil. This predictive nature should be explored in further work.

All this results in the suitability of non-destructive ultrasonic inspection as an alternative method to traditional techniques for evaluating edible oils. The latter involve, on the one hand, the destruction of the product and, on the other, they generate chemical residues that must be treated later. In addition, the time needed for a complete destructive analysis is high and carries with it an economic cost, to which the high price of the samples of this type of product is added. Instead, the low-frequency ultrasound inspection is postulated as a feasible, non-destructive and cost-effective technique, with a predictable nature, being, therefore, a suitable tool for authenticating edible oils, and detecting fraudulent practices.

### CRediT authorship contribution statement

**A. Jiménez:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Data curation, Investigation, Writing – original draft, Writing – review & editing, Supervision, Funding acquisition. **M. Rufo:** Formal analysis, Investigation, Data curation, Writing – review & editing, Validation, Resources, Funding acquisition, Supervision. **J.M. Paniagua:** Investigation, Validation, Data curation, Writing – review & editing, Resources, Funding acquisition, Supervision. **A. González-Mohino:** Conceptualization, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing. **L.S. Olegario:** Investigation, Formal analysis, Data curation, Validation, Writing – original draft, Writing – review & editing, Supervision.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.foodchem.2023.136820>.

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