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Impact of sound attenuation on ultrasound-driven yield improvements during olive oil extraction

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ABSTRACT

High frequency ultrasound can enhance olive oil extractability industrially. However, the ultrasound attenuation phenomena and their implications on extractability, are not well understood. This work aims at evaluating the ultrasound attenuation effects on the oil extraction efficiency, while providing deeper insights into the physics behind the ultrasound extraction in a heterogeneous medium. Olives were collected and processed both in Italy and Uruguay during their respective harvest seasons. Sound pressure distribution was characterized in a high frequency ultrasound reactor, carrying 3 kg of water or paste, by using an indirect contact hydrophone device at 0.4 MHz or 2 MHz. A through-transmission ultrasonic technique was applied to determine attenuation profiles and coefficients in paste at the central frequency of each transducer, with various paste to water ratios and reactor sizes. Other ultrasound improvements on extractability were evaluated including reduction of malaxation time (10, 30 min), sonication time (2.5, 5 min) and power level (174, 280 W) without water addition and in a reactor with a 14.5 cm transducer to wall distance. However, no sound pressure levels in paste were detectable beyond 9 cm from the transducer at both frequencies. Among the various effects evaluated, an emission frequency of 0.4 MHz better improved extractability compared to 2 MHz. The attenuation profiles corroborated these findings with attenuation coefficients of 3.9 and 5.3 dB/cm measured near the respective frequencies. Improvements in oil extractability due to increasing sonication time and power level were significant ($p < 0.05$) also when sonicating beyond 14.5 cm and without water addition. Oil extractability improvements were observed even when sound pressure was undetectable beyond 9 cm from the transducer, suggesting that the standing wave oil trapping effect is not the governing mechanism for separation in high attenuation media for large scale systems.

1. Introduction

Traditional ultrasound assisted extraction systems operate in the 20–100 kHz range. Recently, the use of an extended frequency range, covering from 400 kHz to 2 MHz has been tested in different applications. The operation in this high frequency range is also denominated as megasonic in this application field. High frequency ultrasound has been recently studied as a novel technology for the improvement of oil recovery and milk fat creaming acceleration [1,2]. Exposing oil bearing biomass such palm, olive, avocado and coconut to high frequency ultrasound (megasonics) has shown to generate improvements in the oil separation processes [3–14]. At industrial or semi-industrial scale evaluations of oil extraction were performed by Juliano et al. for palm oil and olive paste [8,14]. They showed improvements in oil recovery by up 3.2% (g oil/100 g oil in paste) after applying 585 kHz for up to 15 min and up to 220 kJ/kg to the olive oil paste, with up to 30% water addition to the paste. A later study [10] has corroborated improvements at semi-industrial plant scale (350 kg/h) by applying 400 + 600 kHz treatments with a residence time of 35 min and 21 kJ/kg.

According to discussions in previous reports, high frequency ultrasound may affect oil extractability by several mechanisms: (a) a standing wave oil droplet trapping effect, (b) semi-stable or stable cavitation bubbles that create rubbing effects in oil-bearing matter [2,14], (c) acoustic steaming and (d) internal heating. The standing wave mechanism is based on the action of forces known as radiation forces, which promote the formation of a standing wave sound field causing

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particles suspended in a fluid to move to either nodal or antinodal pressure planes. This stationary wave standing field is generated by the constructive interference of sound waves generated by the transducer and the waves that are reflected by an opposite solid surface (e.g., vessel wall) [1]. These nodal and antinodal planes are located at a length that is a whole multiple of the half wavelength of the propagated sound [2–17], and particles migrate to these planes of the standing wave field depending on the compressibility and density relationships of the particles in the fluid. Acoustic streaming is permitted by a gradient in the velocity field and this gradient arises from various mechanisms, such as the spatial attenuation of the sound wave in the medium due to absorption of the energy, to friction between a vibrating element and the surrounding medium, or due to scattering of sound waves [1]. Further detail of the physical principles that govern acoustic particle/droplet separation and the mathematical modelling techniques developed to understand, predict, and design acoustic separation processes, with particular emphasis on acoustic streaming are covered elsewhere [1,14].

It was reported that the most suitable ultrasonic frequencies for use in separation applications range from 0.4 MHz to 2 MHz. Free radicals are also developed in this frequency regions with a maximum on 0.8 MHz. An additional increase in the frequency beyond 0.8 MHz decreases the sonochemical performance of such products, since the energy released by the collapse of the cavitation bubble becomes small [18]. Beyond 1 MHz it is described that cavitation is more stable and less transitory cavitation occurs, with larger amounts of smaller cavitation bubbles being formed and at 2 MHz the free radical activity is undetectable [19]. Such bubble formation mechanism may impact in oil extractability effects during aqueous-based vegetable-oil extraction processes. Previous research has shown that the chemical and sensory parameters for extra-virgin olive oil quality were preserved after ultrasound treatment, while increasing the presence of phenolic compounds in the oil [20].

It is important to understand the attenuation effects in oil bearing materials as well as the frequency effects on oil recovery. Understanding attenuation may inform the selection of the transducer to wall distance, which is a key parameter in the design of a larger vessel. However, there are very few references on the ultrasound frequency effects on olive oil recovery. Chanamai and Mc Clements [19] describe the attenuation effects on pure edible oils including olive oil, corn oil and sunflower, but no previous reports have addressed attenuation in olive paste. It is worth mentioning, that the olive oil extraction process involves the crushing of the olives, followed by a shear mixing or malaxation step, and a centrifugation step to separate oil, water, and solids. The ultrasound intervention studied in previous papers deals with either pre- or post-malaxation treatments of olive paste to enhance oil separation [8,21].

The aim of this work was to evaluate the effects of sound attenuation on olive oil extractability to further provide understanding on ultrasound oil separation mechanisms and the implications in reactor design. High frequency attenuation effects on extractability are compared without water, at increasing water to paste ratio, or by decreasing the distance between wall and transducer. Ultrasound effects on processing improvements were investigated including: high frequency selection, malaxation time reduction, sonication time, and ultrasound power input, without water addition and by using a worst case scenario for paste attenuation at maximum transducer to wall distance.

2. Materials and methods

2.1. Olive sampling

Olive fruits (*Olea europaea L.*) of the Coratina variety were harvested in olive groves near Andria, Apulia province, Italy in the 2016 crop season. The Coratina cultivar is one of the most popular olive cultivars in Italy [22]. The olives were brought to the laboratory immediately after harvesting in 30 kg boxes. Sampling was carried out during the month of November. The olive maturation index was determined using a representative sample according to the method proposed by the International Olive Oil Council [23], based on the evaluation of the skin and pulp colour. Other trials carried out in Uruguay, utilized the olive Arbequina variety from Canelones and Lavalleja departments in Uruguay.

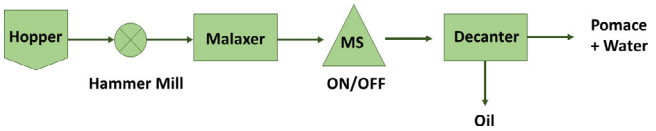

Fig. 1. Process flow diagram of olive oil extraction with or without a megasonic intervention post malaxation.

2.2. Olive oil extraction method

Olive oil industrial extraction was carried out with a standard Abencor laboratory separation equipment [24–26], as follows: (1) crushing 3 kg of olives in a hammer mill, (2) malaxing of the paste at 30 °C for 30 min, (3) separation of the oil from the other phases by decanter centrifugation (Fig. 1). Ultrasound intervention was selected post malaxation and before the centrifugation step. As reported earlier [10], oil droplets are better dispersed into the aqueous phase after malaxation step and therefore show better separation for ultrasound intervention. Discussions on the mechanisms of olive oil droplet coalescence during malaxation can be found elsewhere [5,11]. Olive oil obtained using the traditional process with no ultrasound intervention is used as reference and named “control” through the text. The control sample consisted of the paste waiting for 5 min at the same temperature conditions without sonication before the centrifugation step.

2.3. Ultrasound reactor

The equipment used in this study (Fig. 2a) is composed by a high frequency transducer plate and the corresponding electrical power generator (SONOSYS Ultraschallsysteme GmbH, NeuenBurg, Germany). Plate transducers are vertically positioned inside a metallic container (Fig. 2b). Two types of transducers were used for comparisons, operating at 0.4 and 2 MHz, both with a power draw of 280 W when set at 100% output. The megasonic vessel was a stainless steel container with a volume of 4 L (approx. 3 kg) with internal dimensions of 130 × 165 × 190 mm. The distance between the transducer plate to opposite wall can be varied to test the penetration of the ultrasound field inside the vessel (Fig. 2b).

2.4. Experimental design

This work includes two types of trials: (a) characterisation of attenuation profiles and coefficients on olive paste including crushed solid pits and (b) the influence of various process parameters on extractability at extended transducer to wall distance. Attenuation trials were conducted on the ultrasound reactor itself. Table 1 shows the experimental design to evaluate effects of selected ultrasound processing options in the olive oil extraction process and its impact on oil extractability. Experiment 1 evaluates the effects of the ultrasound frequency on extractability. At lower frequencies cavitation is more intense, however it has been shown that attenuation in liquid media increases with frequency [13]. In experiment 2, the size of the reactor is changed by the transducer to wall distance. This experiment evaluates the penetration of the ultrasonic field and its effect on extractability. Experiment 2 was carried out keeping the specific energy constant so that the transducer to wall-distance (TWD) and frequency effects could be compared irrespective of the mass and energy conditions, i.e., when

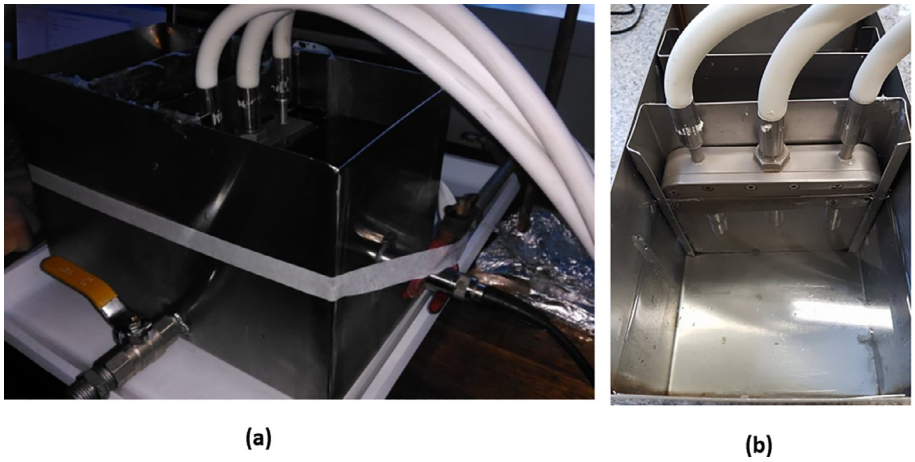

Fig. 2. High frequency ultrasound equipment: (a) pulse-echo ultrasound broad-band reactor, adapted to be used in through-transmission mode for the attenuation experiments; (b) ultrasound reactor with adjustable transducer to wall distance.

the distance was changed form 14.5 cm to 9, the mass of the paste was modified from 3 to 1.5 kg. Experiment 5 varied the power and specific energy, while sonication time remained constant. In experiment 3 the objective is to determine if the malaxation time can be reduced. The reduction on malaxation time when ultrasound is applied can be a process improvement even in the case of similar oil extraction. The ultrasound application time and the power level are also evaluated in experiment 4 and 5 to determine their effect on the extractability without water addition. Fixed parameters for each experiment are shown in detail in Table 1. For experiment 1, ultrasound frequencies 0.4 and 2 MHz were compared with and without the addition of water during malaxation, highlighting the attenuation effect for different mixes. The attenuation effect is also evaluated with the distance between the transducer and the opposite wall (experiment 2), for these two different distances from transducer to reflecting wall, 9 and 14.5 cm. Experiment 3 was carried out to evaluate if malaxation time can be reduced after an ultrasound intervention. The olive oil process efficiency can be improved if oil extractability is maintained after ultrasound treatment at reduced malaxation time. A reduction of 30 min to 10 min malaxation was evaluated in the non-ultrasound and ultrasound assisted process. Experiment 4 considered further reducing ultrasound exposure from 5 to 2.5 min beyond the values studied by Juliano et al [8], with unripe olives and without water addition. Power level effects using other varieties from those studied by Juliano et al [8] are tested in Experiment 5. Further reduction of power input will inform the selection of the number of transducers required in a large scale ultrasound vessel.

Table 1
Experimental design.

Experiment	Olive batch origin	Treatment effect	Variable	Fixed parameters	Design
1	1-Italy 2-Uruguay	High frequency effect × water addition	(0, 0.4, 2) MHz × (no water added, % water added)	30 min malaxation 5 min sonication 3500 rpm centrifugation TWD = 14.5 cm 117 kJ/kg	Comparison of means (triplicate)
2	2-Uruguay	Transducer to wall distance × frequency	(9, 14.5) cm (0, 0.4, 2) MHz	no water added 30 min malaxation 5 min sonication (134, 280) W 117 kJ/kg	Factorial 2 × 3 (triplicate)
3	1-Italy	Malaxation time	(10, 30) min × (MS OFF, MS ON)	5 min sonication 0.4 MHz 280 W 117 kJ/kg no water added TWD = 14.5 cm	Factorial 2 × 2 (triplicate)
4	1-Italy	Sonication time	(2.5, 5) min × (MS OFF, MS ON)	30 min malaxation 0.4 MHz 280 W 117 kJ/kg no water added TWD = 14.5 cm	Factorial 2 × 2 (triplicate)
5	1-Italy 2-Uruguay	Power level	(174, 280) W × (MS OFF, MS ON)	30 min malaxation 0.4 MHz no water added TWD = 14.5 cm 5 min sonication	Factorial 2 × 2 (triplicate)

*TWD (transducer to wall distance).

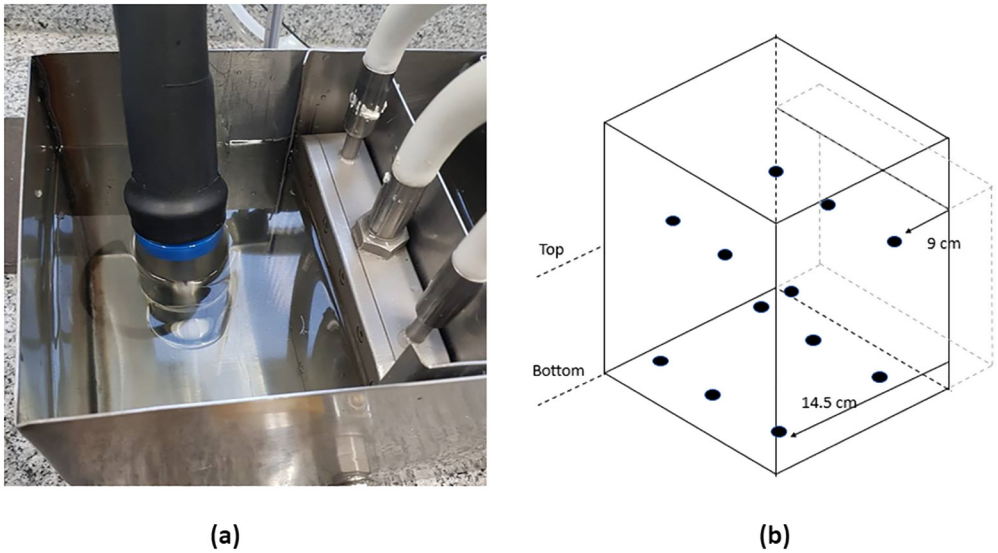

Fig. 3. Sound pressure measurement system in water and paste: (a) hydrophone device including a protective glass membrane for direct measurement on paste; (b) measurement points across the ultrasound reactor.

2.5. Ultrasound reactor characterization

The ultrasound distribution in the reactor was measured both in water and olive paste with crushed pits in the megasonic reactor (Fig. 3) as described further below. These measurements also informed attenuation effects. Attenuation effects were also evaluated by using a pulse-echo broad-band ultrasonic technique in a laboratory cell.

2.5.1. Indirect hydrophone measuring system

Sound Pressure Levels (SPL) during full power operation at 0.4 and 2 MHz frequency (Fig. 3a) were measured in water and paste using a hydrophone device (Fig. 3a) encompassing a needle hydrophone (HNR_1000 S/N: 1761, Onda, Sunnyvale, CA, United States of America, external amplifier Onda AH2010 S/N: 1589; Nom. Gain: 20 dB) with a protective glass membrane for direct measurement on paste. Hydrophones are very sensitive to the presence of oil and protein material and therefore manufacturers do not recommend direct contact with food materials. Although hydrophones have a directive response, they give qualitative information about the pressure field inside the reactor.

Sound measurements were carried out at twelve specific points in the reactor as shown in Fig. 3b. In the experiment 2, the measurements were carried out at two transducers to wall distances (9 and 14.5 cm), and two different depths into the vessel (close to top and bottom). This information gives a qualitative sound distribution map across the reactor (Fig. 3b). The peak-to-peak voltages were recorded three times at each measurement point with an oscilloscope (TDS 1002, Tektronix, Beaverton, Oregon, United States of America) and the mean value was converted into sound pressure level (SPL) using Eq. (1). Trials were carried out at room temperature with a maximum sonication time of 5 min.

$$SPL[dB] = 20\log(Vrms) - EOC \quad (1)$$

where Vrms is the root mean square voltage (V) determined from the peak-to-peak values measured, and EOC (End of Cable) is the hydrophone sensitivity (dB V/μPa) and was obtained from the hydrophone calibration chart supplied by the manufacturer. Besides measuring sound pressure levels, the oscilloscope can also confirm the frequency of the ultrasonic field. Sound pressure was measured at various points across in the paste by using transducer to wall distances of 9 and 14.5 cm, used in experiment 2.

2.5.2. Quantitative determination of the attenuation coefficients by pulse echo broadband

To determine the attenuation coefficient and its dependence with frequency, an experiment using a broadband transducer (1 MHz central frequency) is performed in a laboratory cell. In this experiment, a pulse-echo response is taken and used to estimate the attenuation (Fig. 4). Spectra of ultrasonic waves travelling through media of interest were measured for calculating the attenuation coefficient α(f) at frequencies in the power transducers' bandwidth. This experiment starts by testing distilled water. Gradually olive paste was added and manually agitated to have olive-solid concentrations ranging 0.0–2.4% w/w solids/total liquid in paste. A pulse-echo ultrasonic technique was driven with a 1 MHz ultrasonic transducer.

The signal (Fig. 7a) consists of one pulse, generated by the transducer when being excited by a single broadband pulse. This signal travels through the fluid and is reflexed at the aluminium plane, undoing the path to return to the transducer. Therefore, the frequency spectrum can be computed by using the Fast Fourier Transform (FFT). This computation allows the determination of the energy backscattered for each frequency over the transducer bandwidth. Finally, the

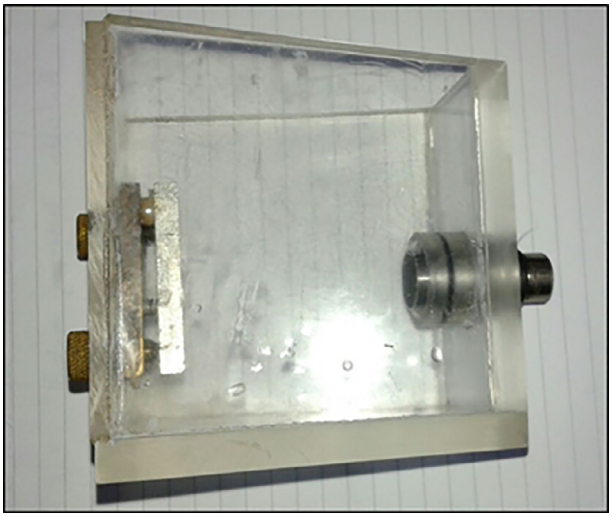

Fig. 4. Pulse-echo ultrasound laboratory cell used in broad-band ultrasonic technique for the attenuation experiments.

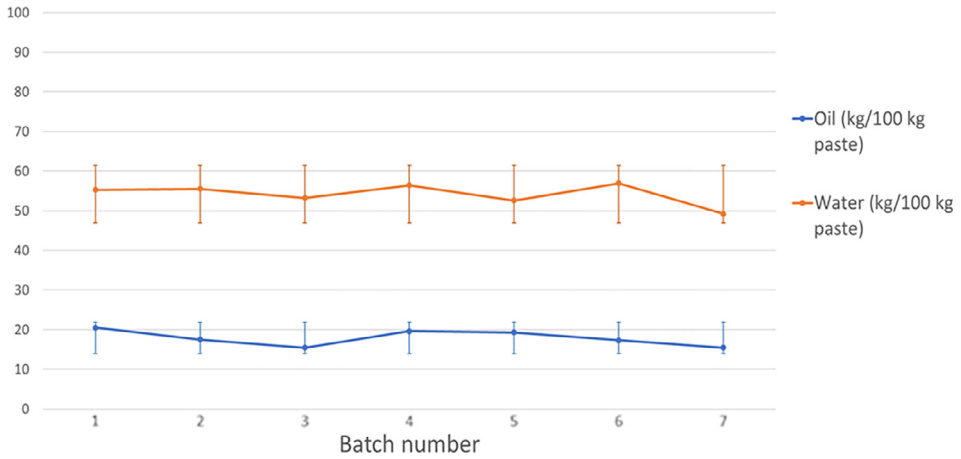

Fig. 5. Oil and water content variability in all tested olive batches. Analysis held on paste sampled before malaxation.

attenuation coefficient is computed, using distilled water as reference for calculations. Fig. 7b

The results are presented at two representative frequencies, 0.6 and 1.4 MHz are selected, since their signal to noise ratio (SNR) was considerably better than for the operating frequencies of 0.4 and 2 MHz in the megasonic reactor employed for extractability trials. For the attenuation coefficient calculation, a plane wave propagation with exponential decay due to attenuation was assumed [27].

$$y(x, t) = A_0 e^{-\alpha(f)x} e^{j\omega t} \tag{2}$$

In the Fourier decomposition, the pulsed signal can be understood as a sum of sinusoidal components like in Eq. (2). Considering only the spatial term in the pulse echo response, Eq. (3) R is obtained.

$$y(x) = R_{12} A_0 e^{-\alpha(f)2L} \tag{3}$$

Where A_0 is the initial amplitude, α(f) the attenuation coefficient for the frequency f, L the length of the chamber and R_{12} the reflection coefficient between the liquid and the reflective plane (aluminium).

The ratio between the amplitudes of the reflected signal travelling through the fluid (f) and water (w) can be written as

$$\frac{y_w}{y_f} = \frac{R_{w,al} A_0 e^{-\alpha_w(f)2L}}{R_{f,al} A_0 e^{-\alpha_f(f)2L}} \tag{4}$$

Assuming that the initial amplitude and the reflection coefficients are similar, with α in cm^{-1}, the ratio can be approximated by

$$\frac{y_w}{y_f} = e^{(\alpha_f(f) - \alpha_w(f))2L} \tag{5}$$

The attenuation coefficient in dB/cm can be obtained after some algebraic manipulation:

$$\alpha_f(f) = \frac{20}{2L} log\left(\frac{y_w}{y_f}\right) + \alpha_w(f) \tag{6}$$

According to Kino [27], the attenuation coefficient for distilled water is approximated by

$$\alpha_w(f) = 0.002 f^2 \tag{7}$$

2.6. Paste composition analysis

The total moisture and oil contents in the paste were determined on 30 g of sample. First, the moisture in the sample was calculated by dehydrating the sample at 103 ± 0.5 °C until reaching constant weight. For oil analysis, the sample was extracted with hexane in an automatic extractor (Randall 148, Velp Scientifica, Milan, Italy) following the analytical technique described by Cherubini et al. [26]. The sample porous container was immersed directly in the boiling solvent. The sample was then subjected to washing at 139 °C for 40 min; the sample container was removed from the solvent and reflux washed. The final part of the process which was conducted at 139 °C for 30 min was solvent recovery. Results were expressed as percentage of oil on wet and dry matter.

The extractability (E) is the percentage of oil extracted with respect to total oil content determined from the processed olives. The E was calculated using the following equation:

$$E = \frac{W_{oil}}{W_{total\ oil}} 100 \tag{8}$$

where Woil is the mass of the extracted oil (kg), and Wtotal oil is the oil mass in the batch of processed olives (kg).

For comparison between ultrasound application and control, Net Ultrasound Extractability (NUE) was calculated from Eq. (9) to evaluate the difference with respect to non-ultrasound (ultrasound – control) on an oil in paste basis.

$$NUE = E(ultrasound) - E(control) \tag{9}$$

2.7. Statistics

Statistical differences on the tested variables, including megasonic improvement on NUE, were investigated through analysis of variance (ANOVA) by using Tukey's HSD, (95% confidence) and performed using the XL-Stat 2012 software (Addinsoft, NY, USA).

3. Results and discussion

3.1. Olive fruit composition

In general, little variation in oil and water content was observed across the batches loaded for each run irrespective of olive origin, indicating that the selected batch of pooled olives was homogenous (Fig. 5). Low variation coefficients per treatment, for both water and oil content, were detected ranging between 2.0% and 2.7% for both the water and oil content, respectively. Therefore, it is expected that oil extractability was not affected due to batch conditions.

3.2. Megasonic reactor characterization

Fig. 6 shows the sound pressure levels measured at bottom and top sections of the megasonic reactor at 9 cm and 14.5 cm, respectively, both in water and in paste. Fig. 6a and b show results at 0.4 and 2 MHz, respectively. As expected, in pure water sound pressure is detected across the reactor and pressure levels increased at higher transducer

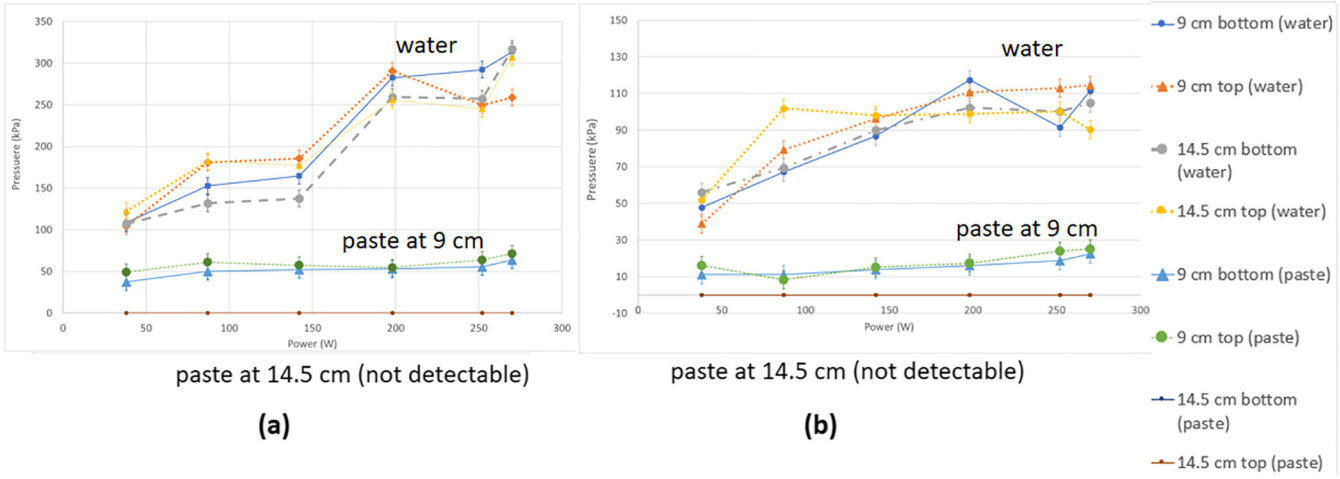

Fig. 6. Sound pressure level measurement in water and paste at both 9 cm and 14.5 cm transducer to wall distance at selected power levels using (a) 0.4 MHz and (b) 2 MHz transducers.

power at both frequencies. This behaviour does not depend on the transducer to wall distance. In pure water, due to the low attenuation, a stationary pattern is formed with the incident and the reflected waves. Sound pressure distribution depends on the relationship between the wave length and the reactor size and not on the size itself. However, such increase was not observed when measuring sound in paste due to attenuation effects. The average sound pressure level obtained in water at 0.4 MHz (100–320 kPa) was greater than at 2 MHz (47–120 kPa) when applying 117 kJ/kg of electrical power in both cases. These results are dependent on the efficiency in the transducers emission, but are consistent with the usual assumption that attenuation is proportional to the square of the frequency, for example findings reported by Trujillo et al. [13].

Sound pressure measured in olive paste was detectable at 9 cm but no sound pressure was detectable at 14.5 cm from the transducer source at both frequencies. Attenuation was also seen at 9 cm where paste provided much lower sound pressure levels than water, (30–70) kPa and (10–25) kPa at 0.4 MHz and 2 MHz, respectively. Olive oil paste has a very heterogeneous composition, including olive skin, pit and oil droplets, and water so the ultrasound waves traveling through are absorbed by the solid components. According to Chamanai et al. [19], attenuation also increases with ultrasound frequency in oil, while ultrasound was more attenuated at 2 MHz than at 0.4 MHz. Even though there are differences between attenuation of oil and water [27], the main contribution to the attenuation is due to the solids. As explained before, in viscous media the attenuation of the acoustic waves is proportional to the square of the frequency. However, when the liquid has solids in suspension, the dominant phenomena to produce attenuation, is the scattering.

Further to these experiments, a pulse-echo broadband measurement technique was employed on the megasonic reactor. This experiment allows the evaluation of the effects of olive paste solids on sound attenuation in the reactor scale avoiding the stationary pattern formed in the continuous wave. Fig. 7 corroborates the decrease of sound detection at increasing olive paste solids in liquid in the reactor system. Results are expressed in two forms, first the sound detection through the signal energy decay (Fig. 7a) and second, through the alpha (attenuation coefficient) (Fig. 7b). In the experiment, the paste-water ratio is increased gradually to identify the effect of the concentration in the attenuation of the ultrasound signal. The receiver was only able to detect sound in paste up to 2.0% solid paste in liquid as the signal became increasingly weakened with the solid content. The attenuation coefficient plots also verify the greater sound attenuation for increasing frequency at the selected paste solids (Fig. 7b). Table 2 shows the attenuation coefficients calculated from Eq. (7) at frequencies 0.6 and 1.4 MHz. The attenuation values of olive paste have not been reported earlier, while there are reports only edible oils [19]. The attenuation profiles corroborated these findings with attenuation coefficients of 3.9 and 5.3 dB/cm, for 0.6 and 1.4 MHz, respectively (Table 2). Oil extractability was neither improved by water addition during the treatment nor by decreasing the reactor size.

As it can be observed from the results of the through transmission technique presented in Table 3 and Fig. 8, attenuation coefficient at 0.4 MHz becomes high when increasing the solid content. For 2 MHz, the signal was undetectable for high paste concentration. Considering the paste dissolution in such a great volume (3 L aprox for the transducer to wall distance of 14.5 cm), pits sedimentation begins immediately after they are added to the system, so for every determination the system was homogenized by stirring, adding variability to this essay. This is why the determination is considered qualitative, given the fact that for a more accurate determination, it would be advisable to employ a laboratory setup.

3.3. Attenuation effects on olive oil extractability

The effects of sound attenuation were also studied by obtaining values on olive oil extractability at transducer to wall distances 9 and 145 cm, respectively, at both 0.4 and 2 MHz. This experiment was adjusted so that the same specific power and energy (Es = 117 kJ/kg, considering solid content) was applied when using greater paste masses of 1.5 kg and 3 kg so that transducers are covered by paste both at 9 and 14.5 cm transducer to wall distance, respectively.

Firstly, oil extractabilities of the control (non-ultrasound) samples at 9 and 14.5 cm were significantly similar ($p > 0.05$, Table 4, experiment 2), indicating consistent results between experiments by using the same raw material. When ultrasound was applied either at 0.4 or 2 MHz, an improvement in oil extractability was observed at both distances as seen by positive NUE values (Eq. 9). However, NUE was significantly greater ($p < 0.05$) when using a larger transducer to wall distance of 14.5 cm than at 9 cm for the transducer operating at 0.4 MHz.

Ultrasound effects on NUE may be explained due to the presence of larger amount of cavitating bubbles distributed across a larger volume of biomass. Cavitation at high frequencies (0.4–2 MHz) has proven to be more stable than at lower frequencies (20–100 kHz), given that bubbles do not have sufficient time to increase in size to create strong unstable implosions [28]. As discussed in previous papers [8,10], effects on high frequency ultrasound olive oil extractability enhancement were suggested to be due to (a) oil droplet trapping mechanisms in standing waves and/or (b) microstreaming oil removal effects due to stable

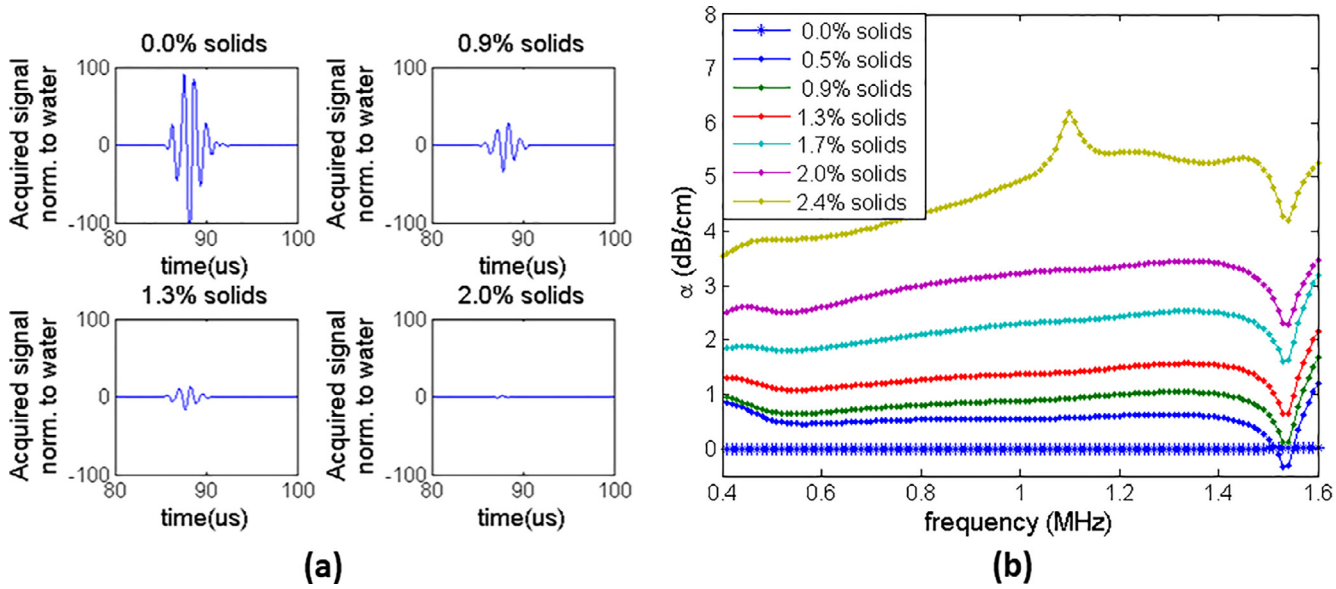

Fig. 7. Pulse-echo measurement: (a) response obtained at selected solid content with the 1 MHz transducer (b) Attenuation coefficient of the response at [0.4 1.4] range, at selected olive solid content (w/w, solids/[oil + water]).

Table 2
Attenuation coefficient at various solid concentrations at both 0.6 MHz and 1.4 MHz.

Paste solids to liquid concentration (total oil and water basis, %, w/w)	αf at 0.6 MHz (dB/cm)	αf at 1.4 MHz (dB/cm)
0.0	0.00072	0.00390
0.5	0.46	0.58
0.9	0.67	1.01
1.3	1.11	1.54
1.7	1.85	2.50
2.0	2.61	3.42
2.4	3.90	5.28

Table 3
Attenuation coefficient at various solid concentrations at both 0.4 MHz and 2 MHz.

Paste solids to liquid concentration (total oil and water basis, %, w/w)	αf at 0.4 MHz (dB/cm)	αf at 2.0 MHz (dB/cm)
0.0	0.0003	0.0080
1.1	0.58	0.38
2.1	0.93	1.0
2.8	2.19	0.58

cavitation bubbles imparting oil "rubbing" effects in localised regions across the olive paste, regarding the potential presence of localised heating, even though no temperature increase was seen in the bulk of the olive paste. Results above, indicate that a standing wave oil trapping mechanism is less likely to have an effect due to the large extent of attenuation in this heterogeneous media. However, given that greater oil ultrasound yield improvements are seen when using greater volumes and distances at the same specific power at 0.4 MHz, it is expected that 0.4 MHz ultrasound penetrates across the biomass but the residual undetected ultrasound suffices the creation of cavitation bubbles across the whole 3 kg sample. Effects of increasing attenuation and scattering mentioned above at 2 MHz are also reflected on the lower (non-significant) results seen at 14.5 cm, compared to 9 cm ($p > 0.05$, Table 4, experiment 2).

3.4. Water addition effects

Water was added in paste (1:3 ratio) during malaxation to evaluate if attenuation improvements in oil extractability were achieved. Table 4 (experiment 1) shows that extractability improvements occurred both with and without water addition at both 0.4 MHz and 2 MHz, as shown by positive NUE values. However, when comparing NUE values with and without water addition, ultrasound oil improvement on oil extractability was not significant upon water addition neither at 0.4 MHz nor at 2 MHz. Even though Figs. 6 and 7a showed that additional water content decreases attenuation, this did not impact on extractability improvements due to ultrasound. The olives solid content, with no water addition, was 12% (w/w, solids/[water + oil]). When water was added the percentage solids to liquid content was 9% (w/w, solids/water + oil), which is much greater than the values considered in the attenuation studies (Fig. 8).

The fact that no NUE improvements are seen due to water addition may also indicate that level of solids achieved after dilution (9%) will still affect the standing wave oil trapping mechanism due to attenuation effects. However, the more stable cavitation siphoning bubble mechanisms combined with potential localised heating prevails, promoting further oil release. Previous studies [8] evaluated the effects of high frequency ultrasound improvement with water addition showing benefits of the technology, therefore evaluating ultrasound power and time effects with 30% water addition in paste. This former study, however did not evaluate high frequency effects.

3.5. Frequency effects

Experiments 1 and 2 (Table 4) show the result of both experiences in Italy and Uruguay with three comparisons at 0.4 MHz and 2 MHz being carried out. For all trials, NUE was significantly higher ($P < 0.05$) at 0.4 MHz than at 2 MHz and similar differences between NUE values were obtained with and without water addition. As discussed earlier, Fig. 6 also showed that attenuation is greater at 2 MHz suggesting that megasound operating at 0.4 MHz lead to a better performance and higher oil extractability. Leone et al. (2017) [10] have shown the reduction of the paste viscosity after treatments of 0.4 + 0.6 MHz and 10 kJ/kg. Earlier works has demonstrated that a linear relationship exists between viscosity reduction and olive oil yield increase [29]. Therefore, based on yield results, it can be inferred that the paste

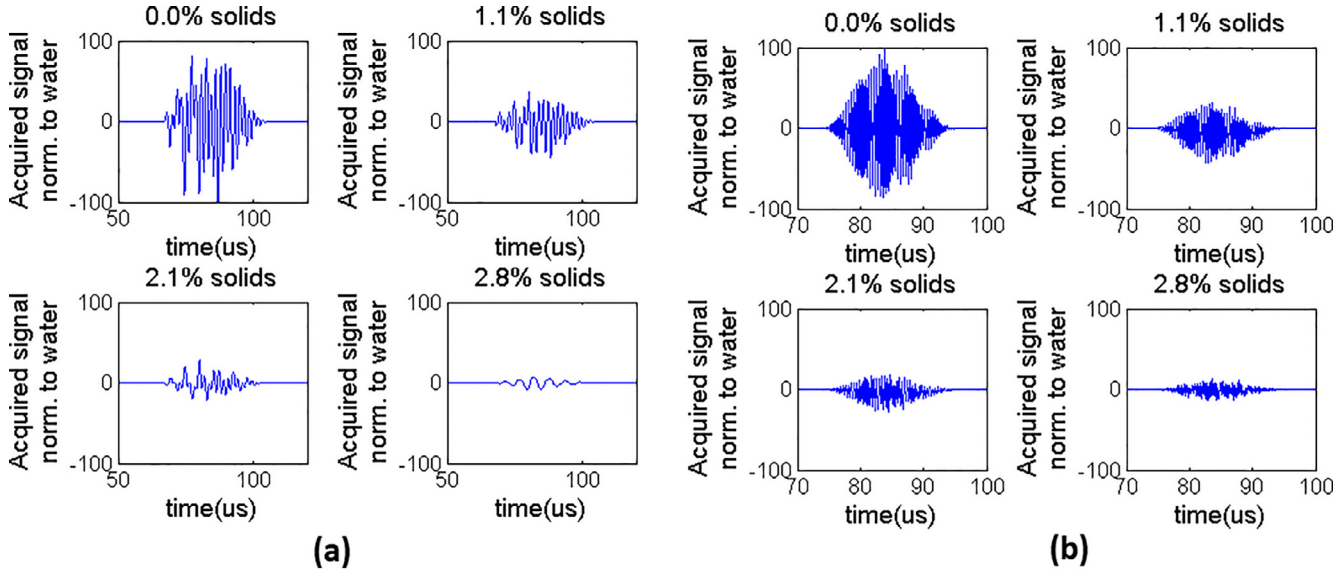

Fig. 8. Through-transmission broadband measurement: response obtained at selected solid content (a) with the 0.4 MHz transducer (b) with the 2 MHz transducer.

Table 4
Oil extractability in non-ultrasound and ultrasound treated olive paste. Corresponding experiment numbers are indicated and each batch is labelled. Different letters indicate significant differences with respect to the control ($p < 0.05$) in selected batches.

Exp	Batch	Country	M. I.	Malaxation time	water addition**	Frecuency (MHz)	Sonication time (min) α	Power (W)	Es (kJ/kg)	Sound pressure in paste (kPa)	Transducer wall distance (cm)	Oil Extractability (%)*	NUE***	p-value
1	1	Italy	2.5	30	no	0	5	0	0		14.5	72.1		0.003
		Italy	2.5	30	no	0.4	5	280	252	N/D	14.5	82.3	**10.2 a**	
		Italy	2.5	30	no	2	5	280	252		14.5	74.8	**2.7 b**	
	2	Uruguay	1.0	30	yes	0	5	0	0		14.5	78.5		0.021
		Uruguay	1.0	30	yes	0.4	5	280	252		14.5	88.1	**9.6 a**	
		Uruguay	1.0	30	yes	2	5	280	252		14.5	82.0	**3.5 b**	
		Uruguay	1.0	30	no	0	5	0	0	N/D	14.5	78.5		
		Uruguay	1.0	30	no	0.4	5	280	252		14.5	88.9	**10.4 a**	
		Uruguay	1.0	30	no	2	5	280	252		14.5	80.5	**2.0 b**	
2	3	Uruguay	1.0	30	no	0	5	0	0	0	9	58.1		0.0076
		Uruguay	1.0	30	no	0.4	5	174	252	70	9	60.8	**2.7 b**	
		Uruguay	1.0	30	no	2	5	174	252	22	9	60.1	**2.0 b**	
		Uruguay	1.0	30	no	0	5	0	0	0	14.5	58.7		
		Uruguay	1.0	30	no	0.4	5	280	252	N/D	14.5	65.2	**6.5 a**	
		Uruguay	1.0	30	no	2	5	280	252	N/D	14.5	59.7	**1.0 b**	
3	4	Italy	1.5	10	no	0	5	0	0		14.5	70.9		0.028
		Italy	1.5	10	no	0.4	5	280	252		14.5	72.6	**1.7 b**	
		Italy	1.5	30	no	0	5	0	0	N/D	14.5	81.1		
		Italy	1.5	30	no	0.4	5	280	252		14.5	89.0	**7.9 a**	
4	5	Italy	1.5	30	no	0	2.5	0	0		14.5	83.3		0.043
		Italy	1.5	30	no	0.4	2.5	280	252	N/D	14.5	85.3	**2.0 b**	
		Italy	1.5	30	no	0	5	0	0		14.5	85.6		
		Italy	1.5	30	no	0.4	5	280	252		14.5	91.4	**5.8 a**	
5	6	italy	1.3	30	no	0	5	0	0		14.5	81.6		0.173
		Italy	1.3	30	no	0.4	5	174	157		14.5	83.0	**1.4 a**	
		italy	1.3	30	no	0	5	0	0	N/D	14.5	71.7		
		Italy	1.3	30	no	0.4	5	280	252		14.5	79.8	**8.1 a**	
	7	Uruguay	3.0	30	no	0	5	0	0		14.5	71.9		0.460
		Uruguay	3.0	30	no	0.4	5	174	157		14.5	79.4	**7.5 a**	
		Uruguay	3.0	30	no	0	5	0	0	N/D	14.5	77.6		
		Uruguay	3.0	30	no	0.4	5	280	252		14.5	82.2	**4.6 a**	

α the control paste refers to 0 MHz, when the paste was untreated for an equivalent time with no sonication.
Es = Specific energy.
N/D = non detected.
* M.I. = Maturity Index.
** water added in a ratio water:paste (1:3).
*** NUE (difference with respect to non-ultrasound control (ultrasound – control) on an oil in paste basis.

viscosity was lower at 0.4 kHz than at 2 MHz. Higher frequencies such as 2 MHz create larger number of smaller bubbles than at 0.4 MHz. Larger number of bubbles may have better localised effects; however, microstreaming might not be powerful enough, given the fact that solids attenuated more the higher frequencies. Due to a better performance in oil extractability of 0.4 MHz compared to 2 MHz, the next sections only provide comparisons at 0.4 MHz.

3.6. Malaxation time reduction

Experiment 3 was carried out to test if ultrasound would assist in reducing malaxation time from 30 min to 10 min without water addition. Results provided in an earlier publication [8] were only demonstrated with water addition of 30% of the paste weight. In this case, significantly higher NUE was obtained with longer malaxation time ($p < 0.05$). A reduced malaxation time led to lower amounts of oil released, and therefore less free oil for ultrasound to separate. Due to lesser amount of free oil present at lower malaxation times, high frequency ultrasound may not assist reducing malaxation time. Among literature [13,30–31], have shown an effect of viscosity reduction due to malaxation and so high frequency ultrasound may be better suited to penetrate less viscous materials.

3.7. Ultrasound energy reduction

The installation of ultrasound reactors needs to consider not only the capital costs but also the operating costs in terms of power demands. Furthermore, reduction of residence times is important for the large flow rates occurring in olive oil processes, which deserve studying the minimum obtainable ultrasound application time.

Table 4, experiment 4, shows that extractability improvements could be achieved even at a sonication time of 2.5 min (NUE = 5.8%), even though a 5 min ultrasound provided significantly better oil recovery. The time effects were also demonstrated elsewhere [8,20] by water addition with up to 5 min treatment. Results suggest reducing ultrasound treatment to a few minutes while maintaining a high specific energy (117 kJ/kg).

Experiment 5, evaluated the effects of specific power reduction without water addition. In this case, there was no significant difference ($P > 0.05$) in the results observed when applying 73 kJ/kg or 117 kJ/kg in two separate experiments carried out in Italy and Uruguay. Even though this contradicts results reported by Juliano et al. [8], after water addition in paste, this effect may disappear when no water is added. Fig. 5 shows that even though sound pressure levels increase with power in water, they do not necessarily increase in paste. Therefore, the same effects are obtained when using both low and high specific power levels, suggesting that lower specific power is required for olive oil extraction. Industrial data reported by Leone et al. [10] at 300 kg/h without water addition utilise a specific energy of 20 kJ/kg obtaining NUE values of 2.3. These NUE values are lower compared to the NUE obtained in the present work (2.7–10.4). It is important to evaluate ultrasound power reduction through a cost benefit analysis since industrial transducers are relative expensive; therefore, decreasing power requirements will decrease both capital and operating costs of the industrial reactor.

4. Conclusions

Oil extractability improvements in paste still occurred even when high levels of attenuation exist to undetectable pressure levels measured with the current device in certain areas of the megasonic reactor. Therefore, the standing wave oil droplet trapping mechanism is unlikely to be taking place in the olive paste. Such mechanism relies on the formation of a stationary pressure pattern across the vessel, which is unlikely to occur due to the scattering effects observed. Nevertheless, other ultrasound phenomena may take place, such as microstreaming effects and stable non-emulsifying cavitation, which contribute to additional oil recovery. The megasonic reactor performance on oil extractability was better at 0.4 MHz than 2 MHz, where attenuation is reduced. Reduction of malaxation time from 30 to 10 min by application of a 0.4 MHz megasonic treatment, could not be achieved in the current system. The present work has provided insights into the mechanism of ultrasound application on olive oil extraction based on attenuation studies, as well as further information on the megasonic system performance without water addition, which supports the future design of industrial megasonic reactors.

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