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Green Extraction of Date Palm Fruits via Ultrasonic-Assisted Approach: Optimizations and Antioxidant Enrichments

Hisham Mohamed [1,2,*], Mohamed Al-Hajhoj [3], Mohamed Al-Saikhan [3], Nashi Alqahtani [4], Mohammad Zayed [5], Mahmoud Moawad [6], Waleed Alsenaien [1] and Maged E. Mohamed [7,*]

1 Date Palm Research Center of Excellence, King Faisal University, Al-Ahsa P.O. Box 400, Hofuf 31982, Saudi Arabia; walsenaien@kfu.edu.sa
2 Agricultural Research Center, Ministry of Agricultural, Giza 58312, Egypt
3 Arid Lands Agriculture Department, Faculty of Agriculture & Food Sciences, King Faisal University, KSA, Hofuf 31982, Saudi Arabia; malhajhoj@kfu.edu.sa (M.A.-H.); msaikhan@kfu.edu.sa (M.A.-S.)
4 Food Science and Technology Department, Faculty of Agriculture & Food Sciences, King Faisal University, Al-Ahsa P.O. Box 400, Hofuf 31982, Saudi Arabia; nalqahtani@kfu.edu.sa
5 Quantitative Methods Department, Faculty of Business Management, King Faisal University, Al-Ahsa P.O. Box 400, Hofuf 31982, Saudi Arabia; mzayed@kfu.edu.sa
6 Pathology Department, National Cancer Institute, Cairo University, Giza 12613, Egypt; mahmoud.moawad@nci.edu.eg
7 Pharmaceutical Science Department, Faculty of Clinical Pharmacy, King Faisal University, Al-Ahsa P.O. Box 400, Hofuf 31982, Saudi Arabia
* Correspondence: hamohammed@kfu.edu.sa (H.M.); memohamed@kfu.edu.sa (M.E.M.); Tel.: +966-054-633-1974 (H.M.); +966-54-299-0226 (M.E.M.)

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Abstract: Background: Green extraction involves using green solvents, such as water, to reduce energy consumption, avoid health and environmental hazards and induce the quality and quantity of the extract. Date palm fruits are a vital source of food and medicinal activities, as they contain a high diversity of phytochemicals, mainly phenolic and flavonoid compounds. The main aim of this study is to investigate the use of water as a green solvent, when assisted by different ultrasonic frequencies, in the extraction of four different cultivars of date palm fruits, by evaluating the phenolic and flavonoid composition as well as the antioxidant capacity of the extract. Methods: Four date palm fruits' cultivars (Agwa, Anbarah, Khalas, and Reziz) were extracted using conventional methods (by water and ethanol) and by ultrasonic means, using two frequencies, 28 and 40 kHz, and applying temperatures (30, 45, and 60 °C), also measuring extraction times (20, 40, 60 min.). Response surface methodology was used for the statistical analysis, applying three factors (temperature, time, and ultrasonic frequency), four responses (total phenolic content, total flavonoid content, FRAP, and ABTS), and four cultivars (categories). Results: Conventional water extraction obtained minimal phenolic and flavonoid compounds (up to 52% of ethanol extraction). This percent improved to reach 60% when heat was utilized. The application of ultrasonic frequencies significantly enhanced the extraction of phenolics/flavonoids and the antioxidant ability of the extract to nearly 90% and 80%, respectively. The use of 40 kHz ultrasonic power managed to extract more phenolic and flavonoid components; however, the antioxidant capacities of the extract were less than when the 28 kHz power was utilized. Agwa and Khalas demonstrated themselves to be the best cultivars for ultrasonic-assisted extraction, depending on the results of the optimized responses. Conclusion: This study could be implemented in the industry to produce date palm fruits' enriched extracts with phenolic and flavonoid components and/or antioxidants.

Keywords: phenolic; flavonoid; ABTS; FRAP; response surface methodology; RSM

1. Introduction

Date palm (*Phoenix dactylifera* L., Family Arecaceae) is an essential fruit crop in Saudi Arabia. The Kingdom retains around 31 million palms, producing approximately

1.5 million tons of date fruits of different cultivars annually [1]. Saudi Arabia dominates a diverse range of date cultivars, including Khalas and Reziz, which are particularly popular in the Kingdom's Eastern Province [2]. Date palm fruits offer an abundant food and energy supply for a growing population, enhancing food security [3] and health [4]. The antioxidant activity of ripe date palm fruits is relatively high because of their patterns of phytochemicals, such as polyphenols and vitamins [3,5]. Polyphenols and flavonoids have potent antioxidant activity and play an essential role in date palm fruit antioxidant activity [6–9]. Therefore, one of the methods to express the antioxidant power of date fruits is to determine the total polyphenol content (TPC) and total flavonoid content (TFC) [10].

Although date palm fruits can be consumed fresh or dried, there is a high add-value in the processing of the fruit to produce various concentrates (spread, syrup, and liquid sugar), as well as fermented date products (wine, alcohol, vinegar, and organic acids) [11,12]. However, processing date palm fruits is not feasible until the fruit is extracted. There are many methods of extraction, which fall into conventional and unconventional methods. The major drawbacks of conventional extraction approaches (e.g., percolation, maceration, and reflux) are the large volume of solvents needed, time-consumption, low amounts of bioactive compounds extracted, and non-environment friendly solvents [13].

Consequently, the term "green extraction" has arisen, identifying the discovery and design of extraction methodologies to moderate energy consumption, allow alternative green solvents, and ensure a safe and high-quality extract/product [14]. Green extraction operates several innovative and unconventional techniques, such as microwave-, ultrasonic-, supercritical fluid-assisted extractions, and/or encourages using eco-friendly solvents or solvent combinations [15]. Ultrasonic-assisted extraction (UAE) is one of the unconventional green methods currently applied to extract natural products. UAE allows the minimization of processing time, maximization of products' quantity and quality, and ensures the safety of food and natural products [16,17]. Likewise, the UAE technique allows the extraction process of thermolabile phytochemicals deep in the tissue; its equipment is simple, energy-saving, and economically cheap [18].

This study aims to develop an UAE method to extract different cultivars of date palm fruits using water as a green solvent. The study also extends to investigate various factors affecting the extractions of the phenolic compounds and flavonoids as leading indicators for the antioxidant activity of the extracted concentrate; therefore, allowing the optimization of the developed method.

2. Materials and Methods

2.1. Date Palm Fruits Materials

Four cultivars of date palm fruits, Ajwa (Ag), Anbarah (An), Khalas (Kh), and Reziz (Rz), were freshly collected from original date palm orchards; Date Palm Research Center of Excellence, King Faisal University, Al-Ahsa, KSA in 2021. The obtained fruits were in a complete ripening and mature stage (i.e., Tamar stage), without any physical or insect damage or fungal infections according to the scale recorded by Abdulhadi [19]. The palms and fruits were identified and authenticated by the plant taxonomist at Date Palm Research Centre of Excellence, KFU, KSA.

2.2. Preparation of Date Palm Fruit Extracts via Conventional Extraction

Conventional ethanolic or water extractions were carried out using the procedure described by Siddeeg et al. [20] with modification. The 100 g date palm fruits from different cultivars were pitted and sliced (dimensions 1.0 × 1.0 × 0.3 cm approximately) and then placed in 600 mL ethanol–water (4:1 (v/v)) or distilled water for 12 h. The extracts were filtered and then centrifuged (2500× g for 10 min). The extracts were evaporated under reduced pressure, and residues were kept at 4 °C until use.

2.3. Determination of Total Phenolic Content (TPC)

TPC in date palm extracts was determined by Folin-Ciocalteau colorimetric assay according to Ainsworth and Gillespie [21] and Al-Farsi, et al. [4] using Gallic acid (G7384, Merck, Darmstadt, Germany) as a standard. In brief, 30 µL aliquot of the diluted sample or serial set of gallic acid (Standard, 25, 50, 75, and 100 g/mL in methanol) was added to 150 µL of Folin–Ciocalteau (1.09001, Merck, Darmstadt, Germany) reagent, previously diluted with distilled water to 10-fold. After mixing the contents for 5 min, 120 µL of Na_2CO_3 (6%) was added. The obtained blue-colored mixture was incubated for 30 min/40 °C. Absorbance measurements were taken at 765 nm using a microplate reader (Shimadzu, Kyoto, Japan). For each measurement, triplicate samples were generated, and values of TPC were calculated as gallic acid equivalents (GAE: mg/100 g fresh sample)

2.4. Determination of Total Flavonoid Content (TFC)

TFC was determined by aluminum chloride colorimetric assay according to Ouahida et al. [22] and Hatamnia, et al. [23]. In brief, 40 µL of the aliquot diluted sample or Quercetin (standard, Q4951, Merck, Darmstadt, Germany, serial dilution, 0.25, 0.5, 0.75, and 1.0 mg/mL) was diluted with distilled water up to 100 µL, then 5 µL of sodium nitrate solution (5%) was added, incubated for 5 min, then 10 µL $AlCl_3$ solution (10%) was added. Finally, after 6 min, 50 µL of 1 M NaOH and 100 µL of distilled water were added to the mixture. Then immediately, the absorbance measurements took place at 510 nm using a microplate reader (Shimadzu, Kyoto, Japan). For each measure, triplicate samples were generated, and the values of TFC were calculated as Quercetin equivalents (QE: mg/100 g fresh sample)

2.5. Ferric Reducing Antioxidant Power Assay (FRAP)

Measurement of the reduction of the ferric 2,4,6-tripyridyl-s-triazine complex (Fe^{3+}-TPTZ) to its ferrous form (Fe^{2+}-TPTZ) in the presence of antioxidants, were performed according to Paz, et al. [24] with minor modifications. A fresh FRAP working solution was prepared by mixing 10 mL 300 mmol/L acetate buffer (pH = 3.6), 1.0 mL 10 mmol/L TPTZ solution (T1253, Merck, Darmstadt, Germany) in 40 mmol/L HCl, and 1.0 mL 20 mmol/L $FeCl_3$ solution. The reaction was performed by adding 20 µL of sample or ethanol (Blank) or L-Ascorbic acid (Standard, A92902, Merck, Darmstadt, Germany) or sample to 180 µL of the FRAP working solution in a 96-well microplate for 15 min at 37 °C. Absorbance was recorded at 593 nm against a blank using a microplate reader (Shimadzu, Kyoto, Japan). The results were expressed as ascorbic acid equivalent (AE: mmol/100 g fresh sample).

2.6. Radical Scavenging Antioxidant Capacity by ABTS

ABTS (2,2′-azinobis-3-ethylbenothiazoline-6-sulphonic acid-diammonium salt, ABTS, A9941, Merck, Darmstadt, Germany) radicals were used to estimate the scavenging ability of the date palm fruits' extracts according to Thoo et al. [25]. The ABTS solution was prepared by adding 10 mL of 7 mM ABTS solution to 10 m1 of 2.45 mM $K_2S_2O_8$ solution and kept in the dark for 12-16 h at room temperature. The ABTS solution was adjusted to an absorbance of 0.7 ± 0.02 at 734 nm with ethanol using a spectrophotometer (Thermo Spectronic Genesys 20, T165762, 4001/4) before usage. Next, 1.95 mL of adjusted ABTS solution was added to 0.5 mL of the sample or standard (L-Ascorbic acid (Standard, A92902, Merck, Darmstadt, Germany) solution and vortexed using (Vortex mixer, Fisher Scientific, USA), and then incubated in the dark at 30 °C for 5 min. Absorbance was measured at 734 nm against methanol (blank) using a microplate reader (Shimadzu, Kyoto, Japan). The results were computed as mmol Ascorbic Eq/100 fresh sample. All samples' radical scavenging assays were represented as a percentage of scavenging ABTS radicals (or percent inhibition of absorbance) using the equation:

$$\text{Inhibition (\%) or ABTS scavenging effect (\%)} = (A0 - A1/A0) \times 100 \quad (1)$$

where: A0 is the control absorbance, and A1 is the absorbance of the sample. 100% distilled water was used as a control.

2.7. Green Extraction of Date Palm Fruits Using UAE Techniques

2.7.1. The Experimental Design

Response surface methodology (RSM) is a statistical technique that involves many variants in a complex model of calculations to allow an optimization process. The method was carried out to investigate the effects of independent variables, "Factors", on the recovery of antioxidants in extracted samples, expressed as dependent variables, "Responses". The experimental design and statistical analysis were generated using Design-Expert statistical software (Stat-Ease Inc., Minneapolis, software version 11.1.2.0, MN, 55413 USA). A randomized central composite design was used as a type of response surface analysis.

The experimental design of the UAE method used four "factors", two of them were continuous factors; extraction "temperature" (included 3-levels, 30, 45, and 60 °C), and extraction "time" (included 3-levels, 20, 40, 60 min). The third factor was a non-continuous one; the "frequency" of the ultrasonic used (included two levels, 20 and 40 Khz). The fourth factor was the category factor; the type of date palm fruit "cultivars" (included four categories, Ag, An, Kh, and Rz). The experimental design model allowed relating all the factors with the different four "responses"; TPC, TFC, FRAP, and ABTS. Data were statistically analyzed using analysis of variance (ANOVA). The best model to fit all responses was the quadratic model. The *p*-value for all responses was less than 0.0001, and R^2 and adjusted R^2 values of not less than 0.88 and 0.87, respectively. Design-Expert software was used to perform 3D surface plots through the RSM model to optimize all independent variables (Factors) influence on each dependent variable (Responses). Optimization was calculated using desirability methods, with all calculated desirability values ranging from 0.4 to 0.7. In numerical optimization, the goal criteria were adjusted to "minimize" in case of factors (time and temperature) and to "maximize" in case of responses (TPC, TFC, FRAP, and ABTS). All the generated statistical models were validated by comparing the actual experimental and predicted values of responses after the generation of the regression equation.

2.7.2. Date Palm Fruit Sample Preparation for UAE

Date fruits of the previously mentioned cultivars were pitted and sliced (dimensions 1.0 × 1.0 × 0.3 cm approximately), and then 100 g of each date palm cultivar was mixed with 200 mL of deionized distilled water.

2.7.3. UAE Method

The UAE method was carried out using a digitally controlled ultrasonic cleaning bath (Branson CPXH 5800E 9.51 l) [26]. Each sample was placed in a 50 mL falcon tube and partially immersed into the ultrasonic bath, with the bottom of the tubes approximately 2.0 cm above the tub. The ultrasonic water bath was adjusted to the required temperature (Factor 2, Table 1), then the ultrasonic equipment was adjusted to the required frequency (20 or 40 Khz, Factor 1, Table 1). The time was adjusted as required (Factor 3, Table 1), and the experiment was allowed to run. Then, tubes were centrifuged at 2500× *g* for 10 min at 5.0 °C. The supernatant was decanted in a clean test tube and stored at 4 °C. All samples were diluted with distilled water to 1:20 ratio before assessing all dependent variables (Responses), as previously mentioned above.

Table 1. Design of experiment by RSM. The number and description of factors and responses implemented.

Independent "Factors"	Unit	The Variable Type	Level of Variation		
			−1	0	+1
Frequency [a]	Khz	Non-Continuous	28	–	40

Table 1. *Cont.*

Independent "Factors"	Unit	The Variable Type	Level of Variation			
			−1	0	+1	
Temperature [b]	°C	Continuous	30	45	60	
Time	min.	Continuous	20	40	60	
Cultivar	Type	Category	Agwa	Anbarah	Khalas	Reziz
Dependent "Response"		**Abbreviation**	**Unit**			
Total phenolic content		TPC	GAE: mg/100 g fresh sample			
Total flavonoid content		TFC	QE: mg/100 g fresh sample			
Ferric reducing antioxidant power asssy		FRAP	AE: mmol/100 g fresh sample			
Radical scavenging antioxidant capacity by ABTS		ABTS	Scavenging effect %			

[a]: Frequency of each treatment I = 28 kHz, II = 40 kHz. [b]: The temperature of the extracted sample was measured at the end of the exposure time of each treatment.

3. Results

3.1. Analysis of Different Cultivars of Date Palm Fruits' Ethanol and Water Conventional Extract

Table 2 illustrates the TPC, TFC, FRAP, and ABTS values of the four date Palm Fruits' cultivars using conventional maceration techniques. The results illustrated the low extractive power of water (as a solvent) related to ethanol. For example, water extracted only 23.8%, 27.4%, 23.0%, and 26.0% of the TPC extracted by ethanol in Ag, An, Kh, and Rz cultivars, respectively. Similarly, water extraction represented 38.1%, 40.9%, 37.5%, and 43.0% of the ethanol-extracted TFC in the four cultivars, respectively, Table 2. The antioxidant power of the extract was reduced when water was used as a solvent. For instance, the extract reducing powers (represented by FRAP values) were lessened to 41.7, 41.5, 50.0, and 52.0% of ethanol extract in Ag, An, Kh, and Rz cultivars, respectively. In the same way, the extract scavenging power (represented by ABTS values) was reduced to 33.3, 44.5, 46.1, and 34.0% of that of ethanol extract in the same four cultivars, respectively.

Table 2. Conventional ethanol and water extraction results for the date palm fruit cultivars under investigation; Agwa, Anbarah, Khlas, and Reziz. The four different responses were analyzed; total phenolic content (TPC), total flavonoid content (TFC), Ferric Reducing Antioxidant Power assay (FRAP), and ABTS scavenging effect (ABTS).

Extraction Solvent	Responses	Date Palm Cultivars			
		Agwa	Anbarah	Khalas	Reziz
Ethanol:Distilled water (4:1)	Total phenolics (GAE: mg/100 g fresh weight)	874.65 ± 12.7	923.86 ± 20.1	824.59 ± 14.4	771.41 ± 64.6
	Total flavonoids (QE: mg/100 g fresh weight)	42.45 ± 3.5	61.21 ± 20.9	48.60 ± 19.6	52.08 ± 4.3
	FRAP (AE: mmol/100 g fresh sample)	1120 ± 54.3	1042 ± 89.3	944.13 ± 14.4	1023.43 ± 59.3
	ABTS (Scavenging effect %)	78.23 ± 4.9	74.12 ± 3.2	65.98 ± 5.2	72.62 ± 6.9
Distilled water	Total phenolics (GAE: mg/100 g fresh weight)	232.12 ± 15.2	253.76 ± 14.6	190.10 ± 21.4	201.24 ± 15.5
	Total flavonoids (QE: mg/100 g fresh weight)	16.43 ± 1.3	25.37 ± 3.6	18.93 ± 12.3	19.44 ± 14.1
	FRAP (AE: mmol/100 g fresh sample)	468.12 ± 23.5	433.24 ± 76.1	472.62 ± 9.2	532.45 ± 23.5
	ABTS (Scavenging effect %)	26.31 ± 2.2	33.24 ± 2.4	30.05 ± 1.4	25.15 ± 2.3

Represented data are Mean + Standard Error "SE"; all values are mean for three replicates.

Furthermore, the results of ethanol extraction exhibited descending order values of TPC (An > Ag > Kh > Rz), with An of the highest total phenolic concentration (923.86 ± 20.1 GAE: mg/100 g fresh weight). For TFC, again, An had the highest flavonoid concentration (61.21 ± 20.9 QE: mg/100 g fresh weight). However, the arrangement changed to An > Kh > Rz > Ag. This arrangement changed again when antioxidant powers are considered; for example, in FRAP, the arrangement was Ag > An > Rz > Kh, and in ABTS, the arrangement was Ag > An > Rz > Kh. These arrangement changes meant that the antioxidant power of the extract depended only partially on phenolic and flavonoid contents. Other classes of phytochemicals could be involved in such antioxidant activity.

Moreover, some of these arrangements changed when water was used for extraction; for example, in TPC, the arrangement was An > Ag > Rz > Kh. This change in the sequence illustrated that water as a solvent is not universal (cf. ethanol). Still, water, preferably, extracts certain classes of chemicals and leaves others, which is a problem if water were be used in green extraction methods.

3.2. Production of Date Palm Fruits' Extract Applying the UAE Approach

Date palm fruits' extract was prepared following a technique that combines temperature, extraction time, and frequency of ultrasonic as multi-factors. Water was used as a solvent for the green extractions. These multi-factors were selected as independent variables because they demonstrate a significant effect on extracting flavonoid and related phenolic compounds from date palm fruit flesh.

3.2.1. UAE of Agwa Cultivar of Date Palm Fruit

Figure 1 represents the interactions between temperature and time of the extraction process with, and without, application of two ultrasonic frequencies (28 and 40 Khz) for the Agwa cultivar to obtain the response variables (TPC, TFC, FRAP, and ABTS). Rising temperatures from 30 to 60 °C through 20 to 60 min extraction time enhanced the TPC, TFC, FRAP, and ABTS values. The increase in temperature and time (60 °C and 60 min) increased TPC by 1.9 folds from conventional water extraction (Table 1) without ultrasonic application. However, the application of 28 Khz and 40 Khz frequencies of ultrasonic increased the TPC extraction by 3.2 and 4 folds from conventional water extraction. Similarly, increase in temperature and time increased TFC by 1.5, 2.25, and 3.9 folds when 0, 28, and 40 kHz Ultrasonic frequencies were used. FRAP activity increased by 1.3, 2.7, and 2.3 folds for antioxidant capabilities when 0, 28, and 40 Khz ultrasonic frequencies were used. The ABTS scavenging activity showed a similar pattern to FRAP, and increased by 2.2, 3.3, and 2.2 folds when 0, 28, and 40 Khz Ultrasonic frequencies were used. It is worth noticing that the antioxidant power of the extracts increased when applying just temperature and time maxima (a nearly two-fold increase); however, these folds increased more with the application of the 28 kHz frequency of Ultrasonic. Yet the application of the 40 kHz did not significantly increase the antioxidant power (i.e., FRAP and ABTS values were reduced at the higher frequency), although it significantly increased the TPC and TFC.

3.2.2. UAE of the Anbarah Cultivar of Date Palm Fruit

Figure 2 illustrates the interactions between two factors (temperature and time) with each of the four responses (TPC, TFC, FRAP, and ABTS) with, and without, the application of two frequencies of ultrasonic (28 and 40 kHz) for the Anbarah cultivar. Increasing the contact extraction time (from 20 to 60 min) and the temperature (from 30 to 60 °C) augmented all response values. TPC was elevated by 1.6 folds from conventional water extraction (Table 1) after application of temperature and time upper limits and without ultrasonic application. However, the utilization of 28 Khz and 40 Khz frequencies of ultrasonic improved the TPC extraction by 2 and 2.6 folds from conventional water extraction. Comparably, the increase in temperature and time increased the TFC by 1.34, 1.6, and 2.12 folds when 0 Khz, 28 Khz, and 40 Khz ultrasonic frequencies were used. FRAP activity intensified by 1.5, 2.74, and 1.98 folds for antioxidant capabilities when applied at 0, 28,

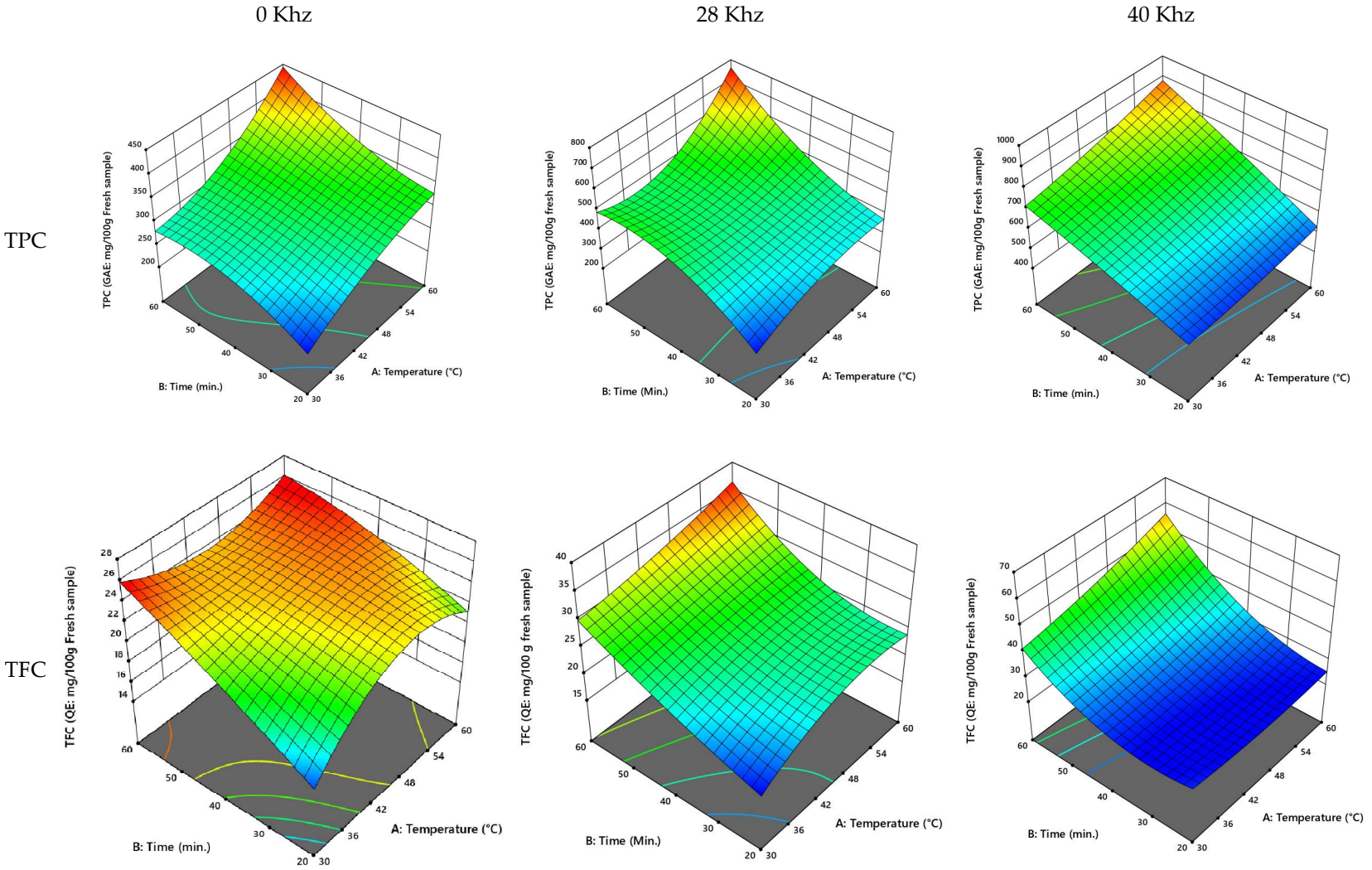
and 40 kHz ultrasonic frequencies. ABTS scavenging activity showed a similar pattern to FRAP, with an enhancement by 1.5, 2.12, and 1.78 folds when 0, 28 and 40 kHz ultrasonic frequencies were used. The antioxidant power of the Anbarah extracts increased due to the application of both 28 Khz frequency and 40 Khz ultrasonic frequencies (cf. Agwa cultivar extracts).

3.2.3. UAE of the Khalase Cultivar of Date Palm Fruit

Figure 3 demonstrates the effect of temperature and time on TPC, TFC, FRAP, and ABTS with, and without, applying two ultrasonic frequencies (28 and 40 Khz, factor) for the Khalas cultivar. The passing of extraction time (from 20 to 60 min) improved all response values. However, this was not always the case with temperature elevation (from 30 to 60 C). The escalation of temperature and time to reach 60 °C and 60 min. amplified TPC by two folds in relation to the conventional water extraction (Table 1) without ultrasonic application. However, the 28 and 40 Khz frequencies of ultrasonic application intensified the TPC extraction by 3.52 and 4 folds from conventional water extraction. The 4-fold increase with 40 Khz frequency happened when the temperature was 45 °C; rise in temperature beyond that significantly decreased the amount of TPC, Figure 3. Relatedly, temperature and time escalation boosted TFC by 1.23, 1.38, and 1.76 folds when 0, 28, and 40 kHz ultrasonic frequencies were used. An increase of 1.77 folds occurred with the 40 Khz frequency when the temperature was 45 °C, and rise in temperature beyond this value significantly decreased the fold increase of TFC, Figure 3. FRAP activity increased by 1.36, 1.99, and 1.75 folds for antioxidant capacities when using 0, 28, and 40 kHz ultrasonic frequencies. The ABTS scavenging activity shows a comparable pattern to FRAP and intensified by 1.51, 1.97, and 1.37 folds when 0, 28, and 40 kHz Ultrasonic frequencies were used. The antioxidant power of the extracts increased when applying the highest values of temperature and time (60 °C and 60 min, nearly 1.5 folds and 2 folds increase, respectively). Antioxidant power was intensified upon using the 28 kHz frequency of ultrasonic more than for the application of the 40 Khz frequency.

3.2.4. UAE of the Reziz Cultivar of Date Palm Fruit

The interaction of the three factors (temperature, time, and ultrasonic frequency) in the Reziz cultivar extraction process and their effect on all the responses (TPC, TFC, FRAP, and ABTS) are illustrated in Figure 4. In general, the increase in the extraction time (from 20 to 60 min) improved all response values. Nevertheless, temperature manipulation did not result in the same outcome. The temperature-time augmentation, without ultrasonic application, boosted TPC by 1.97 folds relative to conventional water extraction (Table 1). Using 28 and 40 kHz frequencies of ultrasonic intensified TPC extraction by 2.7 and 3.06 folds, respectively. On the other hand, elevation in temperature and time increased TFC by 1.23, 1.60, and 2.38 folds when 0, 28, and 40 kHz ultrasonic frequencies were used. The 1.23 folds and 1.60 folds increase with 0 Khz and 28 Khz frequencies occurred when temperature was 45 °C. However, rise of temperature beyond this value did not significantly influence the concentration of TFC values, Figure 4. FRAP activity increased by 1.18, 2.07, and 1.59 folds when 0, 28, and 40 kHz ultrasonic frequencies were used. Similarly, ABTS scavenging activity increased by 1.77, 2.74, and 1.88 folds when 0, 28, and 40 kHz ultrasonic frequencies were used. Interestingly, the antioxidant power of the extracts increased when applying just temperature and time maximum values (60 °C and 60 min, nearly 1.77 fold increase (ABTS only). However, this fold increase occurred with the application of the 28 Khz frequency of ultrasonic, more than with the application of the 40 kHz; although, this pattern was not followed in TPC and TFC extraction values.



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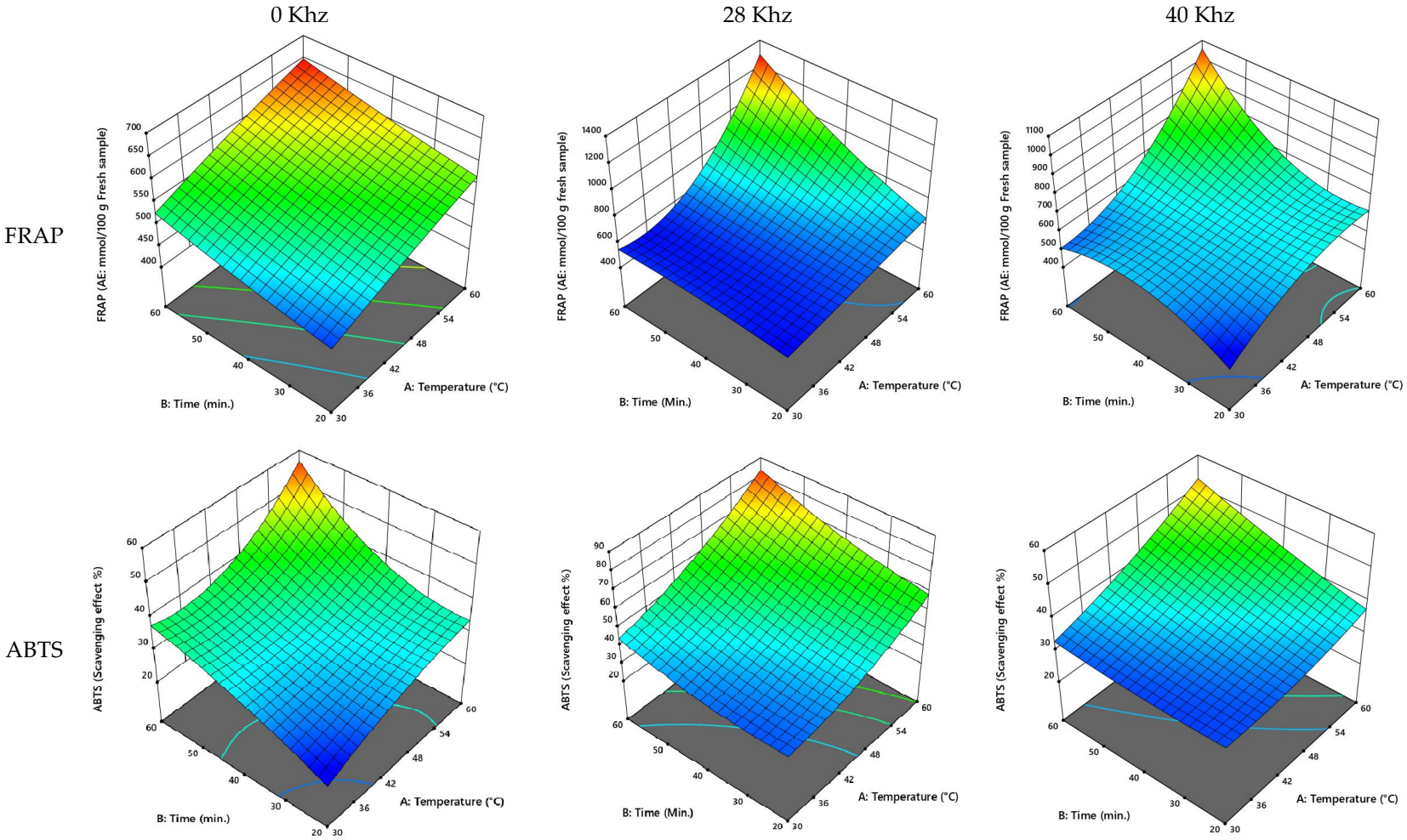
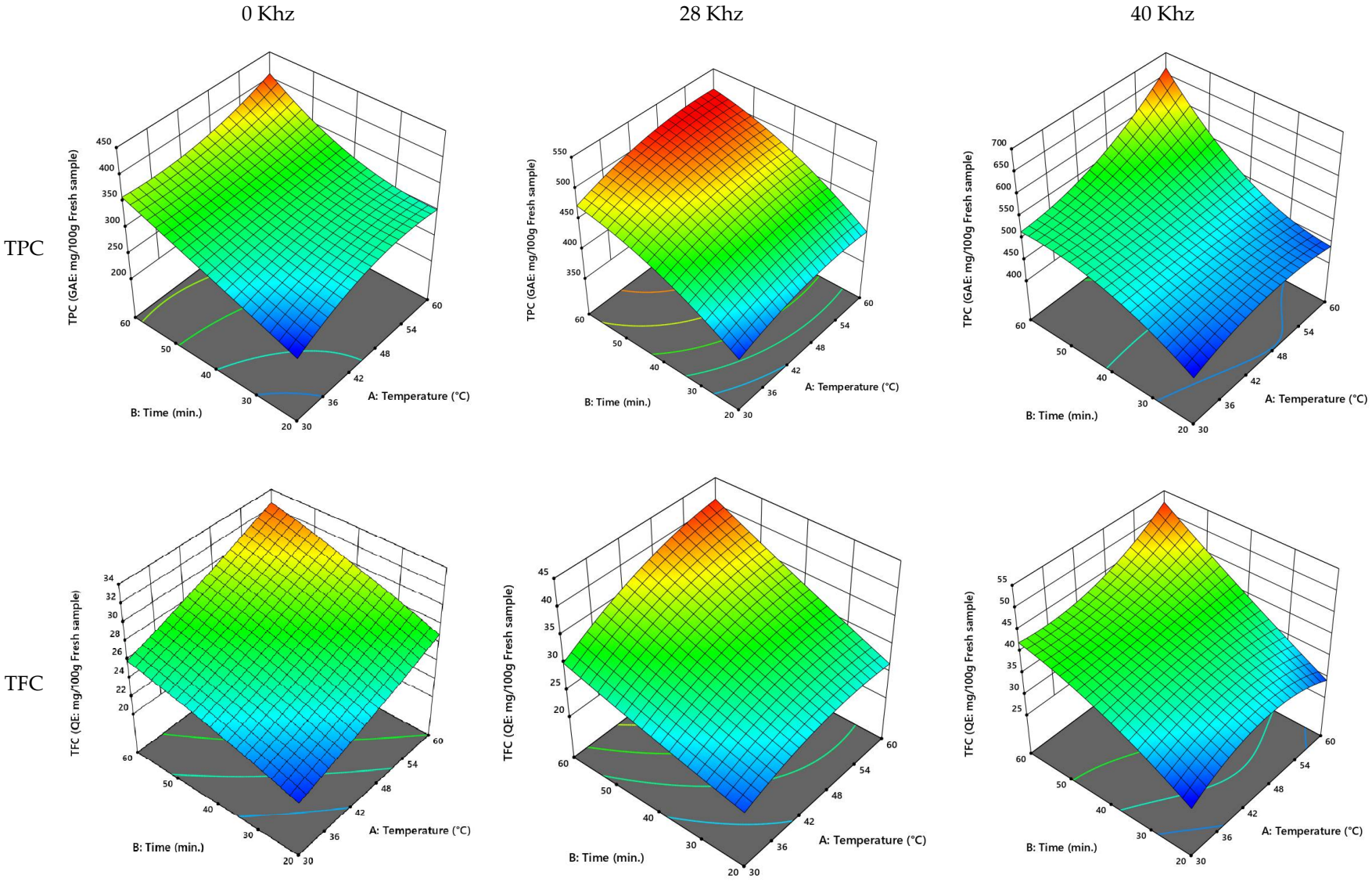


Figure 1. 3D response surface plot for response variables (TPC, TFC, FRAP, and ABTS) as a function of UAE with frequency 0, 28, and 40 kHz in Agwa date palm fruits' cultivar.



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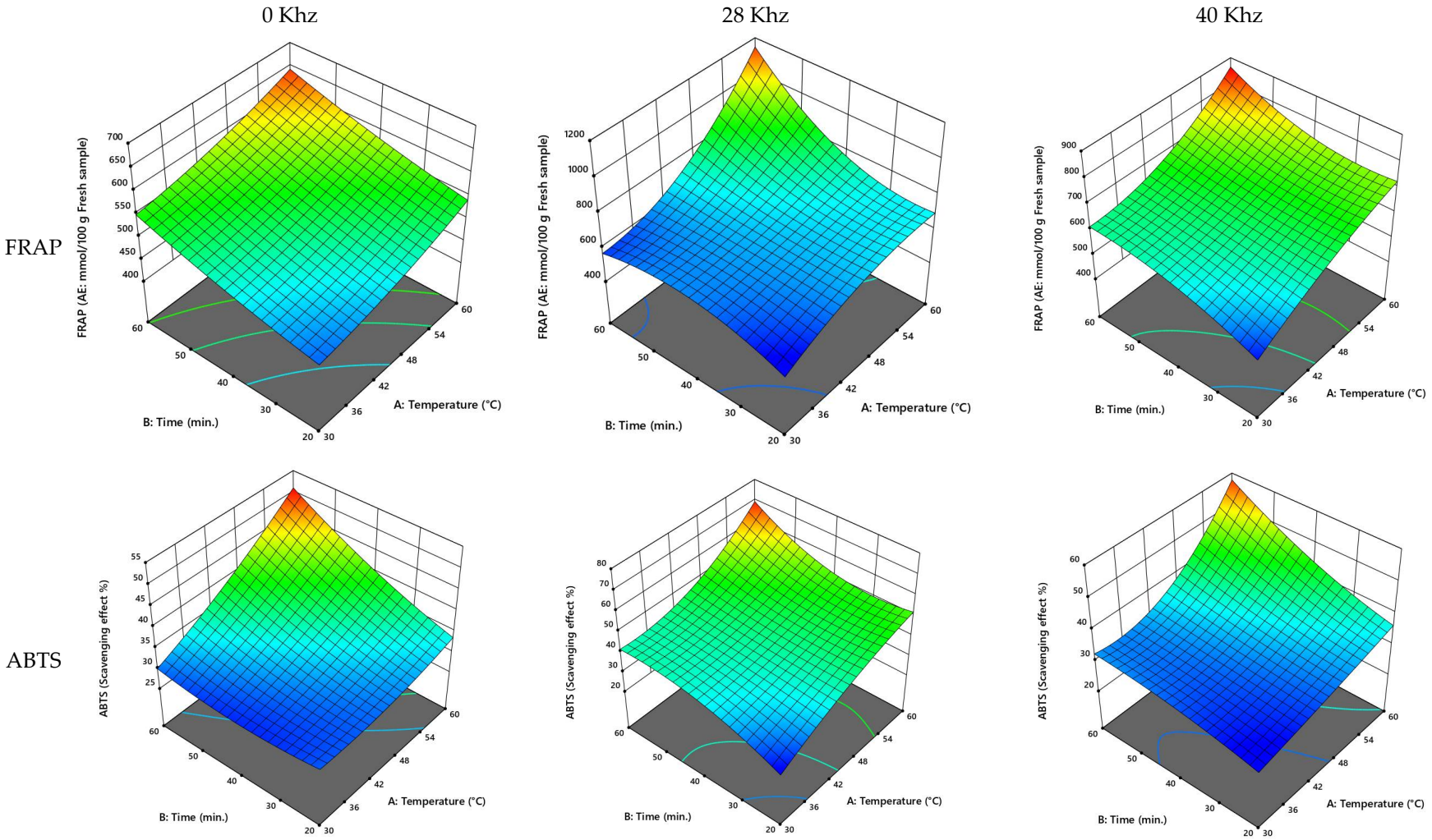

Figure 2. 3D response surface plot for response variables (TPC, TFC, FRAP, and ABTS) as a function of UAE with frequency 0, 28, and 40 kHz in Anbarah date palm fruit cultivar.

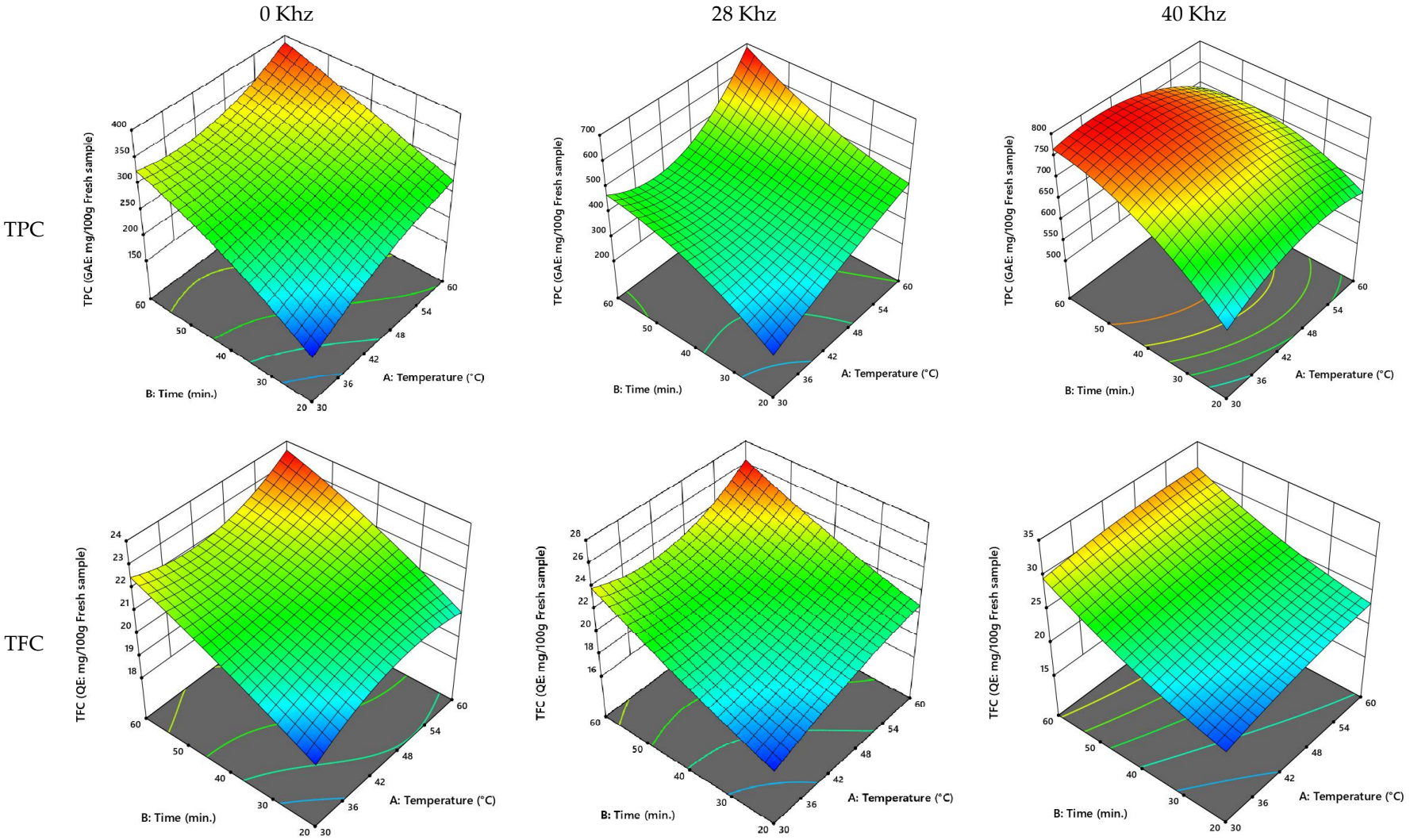

Figure 3. *Cont.*

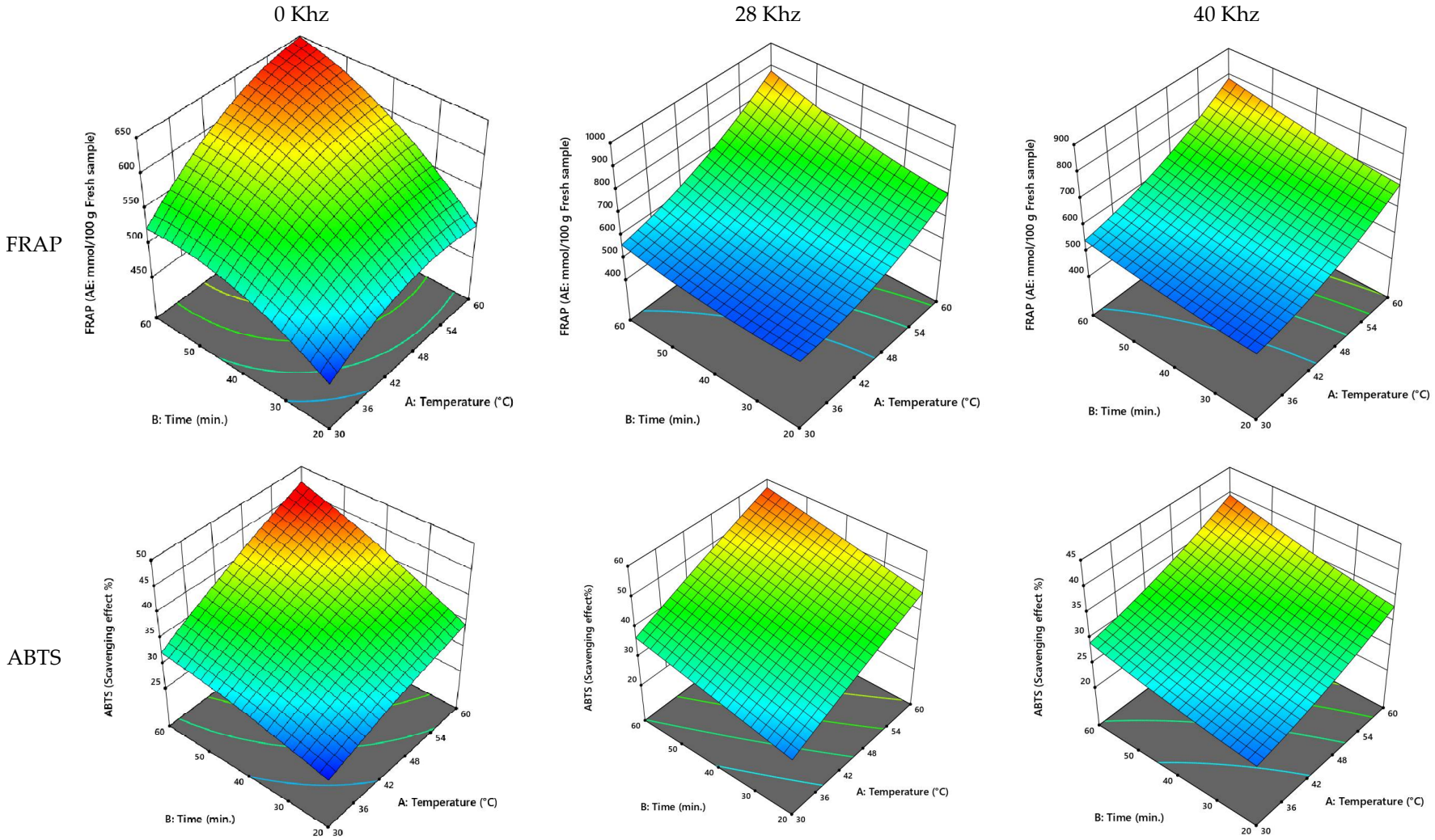

Figure 3. 3D response surface plot for response variables (TPC, TFC, FRAP, and ABTS) as a function of UAE with frequency 0, 28, and 40 kHz in Khalase date palm fruit cultivar.

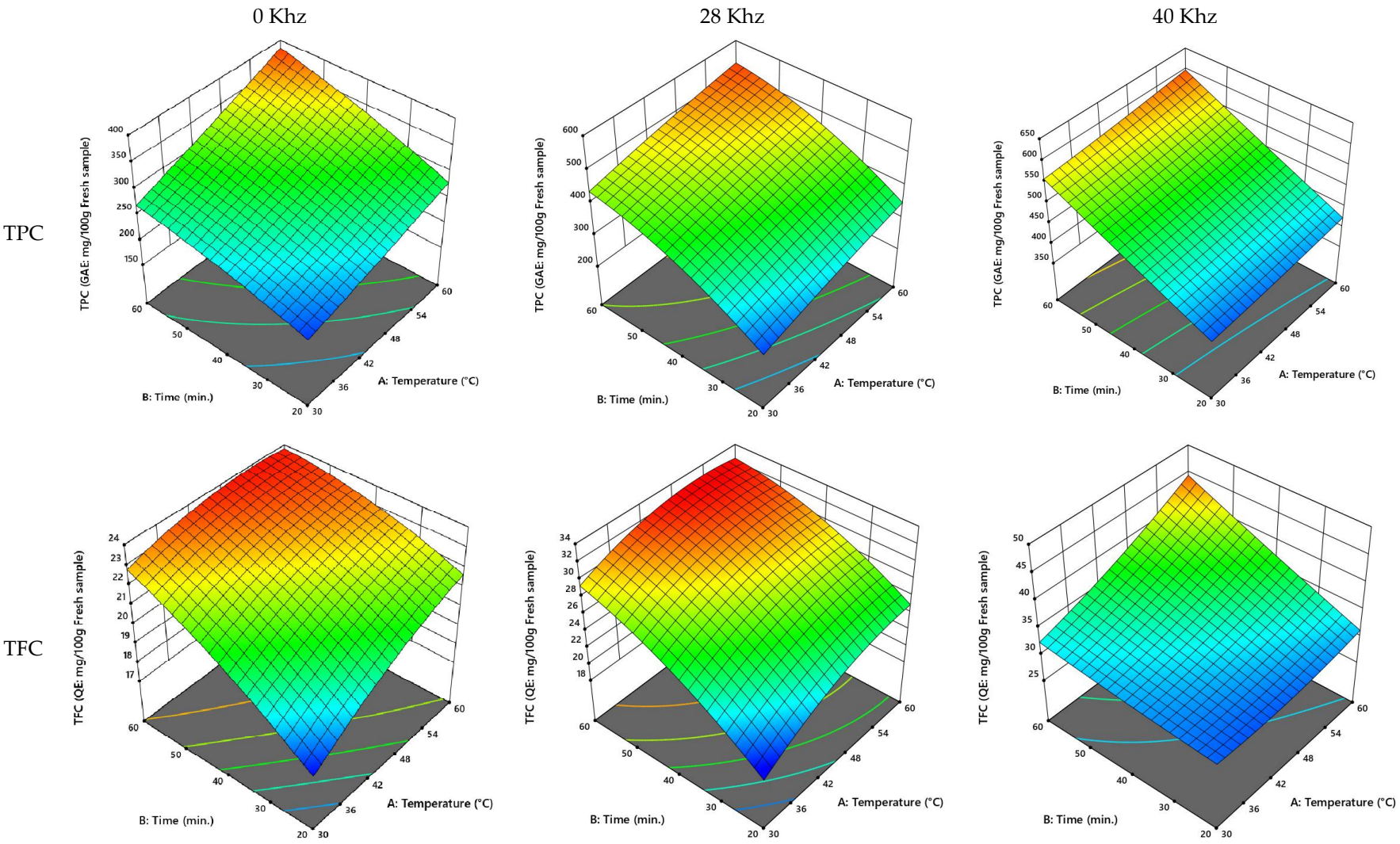

Figure 4. *Cont.*

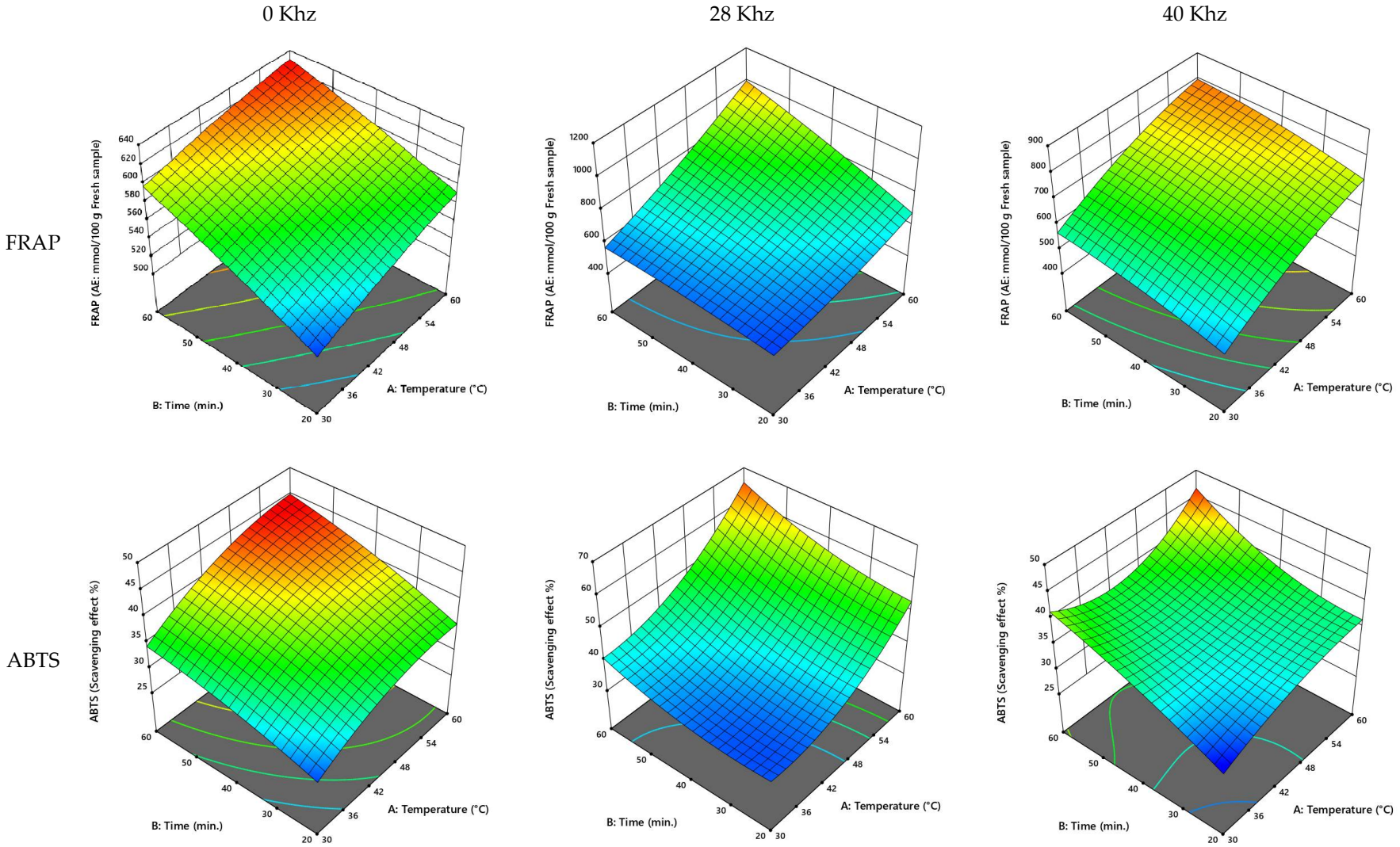

Figure 4. 3D response surface plot for response variables (TPC, TFC, FRAP, and ABTS) as a function of UAE with frequency 0, 28, and 40 kHz in Reziz date palm fruit cultivar.

3.2.5. UAE Process Optimization

Table 3 discusses the calculated optimized extraction procedures to produce the maximum of each response, using the minimum of the factors. In the Agwa cultivar, the optimized calculation stated that using the frequency of 40 kHz, at 53.53 °C and 53.20 min, a maximum of 787.59 (GAE: mg/100 g Fresh sample), 43.45 (QE: mg/100 g Fresh sample), 755.97 (AE: mmol/100 g Fresh sample), and 44.97 (Scavenging effect %) of TPC, TFC, FRAP, and ABTS, respectively, were obtained, which represented a fold-increase of 3.32, 2.64, 1.60, and 1.73 for the four responses, respectively, relative to conventional water extraction (Table 1). In the Anbarah cultivar, and applying the same optimizing approach, the frequency of 40 Khz, in a temperature of 53.95 °C and 51.08 min, a maximum of 554.83 (GAE: mg/100 g Fresh sample) TPC, 44.54 (QE: mg/100 g Fresh sample) TFC, 730.97 (AE: mmol/100 g Fresh sample) FRAP activity, and 42.99 ABTS scavenging effect percentage could be reached. These values corresponded to increases of 2.18, 1.78, 1.68 and 1.3 folds in the four responses, respectively, compared to conventional water extraction. The application of 40 kHz frequency, with a temperature of 48.9 °C and a time 45.25 min, allowed the achieving of 753.46 (TPC; GAE: mg/100 g Fresh sample), 27.10 (TFC; QE: mg/100 g Fresh sample), 630.28 (FRACP activity; AE: mmol/100 g Fresh sample), and 32.37 (ABTS scavenging effect percentage) in the Khalas cultivar, which accounted for a fold-increase of 3.96, 1.42, 1.33, and 1.0 for the four responses, respectively, compared to conventional water extraction. In the Reziz cultivar, optimization stated the use of 40 Khz ultrasonic frequency with 50.188 °C and 48.162 min produced an utmost TPC of 532.19 (GAE: mg/100 g Fresh sample), a TFC 36.677 (QE: mg/100 g Fresh sample), a FRAP activity of 735.736 (AE: mmol/100 g Fresh sample), and an ABTS scavenging effect of 37.45%. These values illustrated a fold increase of 2.64, 1.89, 1.38, and 1.50 for the four responses, compared to conventional water extraction.

Table 3. Ideal predicted values of the factors (UAE frequency, temperature, and time) to get the maximum responses (TPC, TFC, FRAP, and ABTS) for each of the Date palm fruits' cultivars (Agwa, Anbarah, Khalas, and Reziz) according to the surface plot analysis (see Figures 1–4).

Cultivar	Power (kHz)	Temperature (°C)	Time (min)	TPC (GAE: mg/100 g Fresh Sample)	TFC (QE: mg/100 g Fresh Sample)	FRAP (AE: mmol/100 g Fresh Sample)	ABTS (Scavenging Effect %)
Ajwa	0	50.36	42.87	332.57	24.25	576.64	34.62
	28	54.78	51.12	553.25	30.44	896.50	66.93
	40	53.53	53.20	787.59	43.45	755.97	44.97
Anbarah	0	51.61	49.51	346.33	29.47	577.17	39.58
	28	51.657	48.873	499.439	35.985	764.903	49.376
	40	53.950	51.081	554.827	44.536	730.965	42.994
Khalase	0	47.10	44.14	306.20	21.74	598.92	38.64
	28	53.35	43.52	470.22	22.44	699.67	48.78
	40	48.91	45.25	753.46	27.10	630.28	32.37
Reziz	0	47.13	44.76	307.29	22.88	597.05	40.33
	28	51.82	45.56	472.44	31.06	788.53	47.79
	40	50.19	48.16	532.19	36.68	735.74	37.45

Interestingly, the optimization results, seen in Table 3, demonstrated that the antioxidant power parameters (FRAP and ABTS) were more augmented with the application of 28 kHz ultrasonic frequency in all cultivars and reached 1.89 and 2.53 folds in Agwa; 1.5 and 1.75 folds in Anbarah; 1.48, 1.62 folds in Khlase; and 1.48, 1.89 folds in Reziz, for FRAP and ABTS, respectively. Although the shift from 40 to 28 kHz allowed a significant increase in antioxidant power for some cultivars, e.g., for Agwa, ABTS elevated from 1.73 to 2.53 folds (46% increase), and for Khlase; ABTS upgraded from 1 to 1.62 folds (37.5% increase), many of the elevations were not significantly high. Furthermore, the TPC and TFC were less than those extracted with 40 Khz frequency for all the cultivars, Table 3.

4. Discussion

Green extraction techniques apply innovative technology to allow less, or no, organic solvent usage to diminish health and environmental influences and/or enlarge the yield of needed and required phytochemicals [16]. Conventional extraction methods, e.g., heat reflux, maceration, Soxhlet extraction, and others, require the use of large volumes of solvent with an extended extraction time. Most green extraction methods utilize water as a solvent of extraction, due to its advantages of being safe for health and the environment, being highly available, and of low cost. However, water is a polar solvent and suffers many setbacks regarding its extractive powers for many natural compounds, especially regarding lipophilic properties [27]. Furthermore, water endures high heat capacity, making its application for extraction a high-energy process [28]. Therefore, as a green solvent, the utilization of water for extraction should be accompanied by advanced physical methods to overcome some of these disadvantages. These physical systems are the basis of many green extraction methods, e.g., microwave-assisted, ultrasound-assisted, pulsed electric field extractions, and supercritical fluid extractions.

Date palm fruits are a rich source of phenolic and flavonoid-type antioxidants (1–2%) [5,9,12,29]. These compounds are the main reason for date palm fruits' medicinal and pharmacological activities. Many of these compounds are slightly soluble in water, especially non-glycosidic phenolic compounds and flavonoid aglycones [16,27].

Regarding the results of this study, the use of water only as an extraction solvent provided a low extractive value for TPC (23 to 27.4%) and TPC (38 to 43%) compared to ethanol extraction for all date palm fruits' cultivars. Moreover, water extraction reduced the antioxidant power of the extract; for example, FRAP values were 41 to 52%, and ABTS values were 33–46% of the ethanol extracts' values for all cultivars. These values indicate the necessity to use a physical aiding method to improve the extractive values of water for date palm fruits' phenolic components, flavonoids, and other antioxidant materials.

Ultrasonic-assessed extraction (UAE) is a promising technology to reduce processing time and energy and maximize food products' quality, quantity, and safety. This methodology can considerably boost the extraction of intracellular components from complex phytochemical matrices [18,30–32]. UAE of date palm fruits has been infrequently discussed in some studies in order to evaluate the effect of ultrasonic application on the production of various medicinally effective phytochemicals, including phenolic and flavonoid compounds [17,33,34]. For example, Almusallam et al. [34] examined the effect of lactic acid and sucrose as solvents; when various ultrasonic frequencies were applied to extract from two Algerian cultivars of date palm fruits, and they were able to recover up to 1393.5 GAE mg/100 g of TPC. In another study, Fatiha et al. [17] and co-workers extracted 215.47 to 246.61 GAE mg/100 g of TPC from one Algerian cultivar of date palm fruit using different alcohol-based solvents and solvent combinations and applying different ultrasonic frequencies. Very few studies have applied pure water for the extraction of date palm fruits.

As a result, the UAE approach was implemented to extract different cultivars of date palm fruits and to enhance the phenolic and flavonoid contents of the extract, hence the antioxidant power. After optimization using the RSM, Table 4 was produced to relate the optimized values of all responses (Table 3) to the importance of conventional ethanol and water extractions (Table 2). Analysis of Table 4 reveals that when temperature and time cascades were applied in the absence of ultrasonic frequencies (i.e., 0 Khz, Table 4), a minor fold increase from conventional water extraction reached a maximum of 1.6 folds (Rz ABTS value). Additionally, the percentage achievement from conventional ethanol extraction did not exceed 63% (Kh, FRAP value), and the majority of the percentages were around 30 to 45%. Optimizations of the UAE methodology toward the maximum extraction responses and minimum factors resulted in the production of 532 to 787 GAE: mg/100 g fresh sample TFC from the four cultivars. Ag and Kh cultivars produced the best value of TPC with 3.39 folds and 3.96 folds increase over conventional water extraction and achieved 90% and 91% of the conventional ethanol extraction, respectively, Table 4. Although An cultivar achieved top values for TFC (44.53 QE: mg/100 g Fresh sample), an Ag cultivar scored

the best fold increase over conventional water extraction (2.64) and the best-achieved percentage from conventional ethanol extraction (102%), Table 4. The fold increase in antioxidant power assessment (FRAP and ABTS) did not exceed two-folds in most cases; however, the percentage of achievement in ethanol conventional extraction was high and reached 80% (FRAP, Agwa, 28 kHz) and 74% (ABTS, Khalas, 28 kHz), Table 4. It can be noticed that the antioxidant fold increase or percentage achieved as the result of increase in ultrasonic power parameters were reduced to nearly reach the values of non-ultrasonic usage (0 kHz), Table 4. However, TPC and TFC values were not affected. This reduction could be attributed to the destruction of valuable antioxidant compounds by applying heat and ultrasonic high frequency, or more favorable extraction toward classes of compounds with no, or less, antioxidant activity. However, further investigations are needed to prove either or both of the hypotheses as no studies could be retrieved to support these hypotheses or further explain these results.

Table 4. Fold and percentage changes of the optimized UAE responses (TPC, TFC, FRAP, and ABTS) in relation to the conventical extraction methods (ethanol and water). This table is relating Tables 2 and 3.

Response	Ultrasonic Power		Agwa	Anbarah	Khalas	Reziz
TPC	0 kHz	Fold increase from water conventional extraction	1.43	1.36	1.61	1.53
		Percentage achievement from ethanol conventional extraction	38.02	37.49	37.13	39.83
	28 kHz	Fold increase from water conventional extraction	2.38	1.97	2.47	2.35
		Percentage achievement from ethanol conventional extraction	63.25	54.06	57.02	61.24
	40 kHz	Fold increase from water conventional extraction	3.39	2.19	3.96	2.64
		Percentage achievement from ethanol conventional extraction	90.05	60.06	91.37	68.99
TFC	0 kHz	Fold increase from water conventional extraction	1.48	1.16	1.15	1.18
		Percentage achievement from ethanol conventional extraction	57.12	48.14	44.73	43.92
	28 kHz	Fold increase from water conventional extraction	1.85	1.42	1.19	1.60
		Percentage achievement from ethanol conventional extraction	71.71	36.66	46.17	59.63
	40 kHz	Fold increase from water conventional extraction	2.64	1.76	1.43	1.89
		Percentage achievement from ethanol conventional extraction	102.37	72.76	55.76	70.42
FRAP	0 kHz	Fold increase from water conventional extraction	1.23	1.33	1.27	1.12
		Percentage achievement from ethanol conventional extraction	51.49	55.39	63.44	58.34
	28 kHz	Fold increase from water conventional extraction	1.92	1.77	1.48	1.48
		Percentage achievement from ethanol conventional extraction	80.04	73.41	74.11	77.05
	40 kHz	Fold increase from water conventional extraction	1.61	1.69	1.33	1.38
		Percentage achievement from ethanol conventional extraction	67.50	70.15	66.76	71.89
ABTS	0 kHz	Fold increase from water conventional extraction	1.32	1.19	1.29	1.60
		Percentage achievement from ethanol conventional extraction	44.25	53.41	58.56	55.53
	28 kHz	Fold increase from water conventional extraction	2.54	1.49	1.62	1.90
		Percentage achievement from ethanol conventional extraction	63.12	66.62	73.94	65.80
	40 kHz	Fold increase from water conventional extraction	1.71	1.29	1.08	1.49
		Percentage achievement from ethanol conventional extraction	57.49	58.01	49.06	51.56

From Table 4, it can be concluded that UAE is a good extraction technique for the four cultivars of date palm fruits, especially the Agwa and Khalase cultivars, as similar patterns of TPC, TFC, FRAP, and ABTS to those of ethanol extraction could be produced using water as a solvent and applying ultrasonic by both frequencies used.

5. Conclusions

The UAE method, using water as a solvent, was developed and optimized in this study toward the high production of total phenolic and flavonoid moieties and to induce high antioxidant capacity from four different date palm fruits' cultivars. Conventional water extraction methods failed to extract phenolic and flavonoid compounds from the fruits' cultivar relative to that of ethanol extraction (up to 43%). Similarly, water extracts

produced minimal antioxidant capabilities, calculated as FRAP and ABTS activities, when related to ethanol extraction (maximum 52%). Applying only heat and time factors slightly improved the extraction process to reach up to 60% of ethanol extraction values in both compounds and antioxidant capacities. Application of ultrasonic in two frequencies (28 and 40 kHz) succeeded in improving water extractive powers to mainly 75–90% of the ethanol extract capacity for phenolic and flavonoid extraction and 65 to 80% for antioxidant capabilities. The use of 28 Khz ultrasonic power improved the antioxidant ability of the extract. However, 40 kHz ultrasonic power managed to extract more phenolic and flavonoid components. Based on their optimized response results, the best cultivars to be extracted by UAE were the Agwa and Khalase cultivars. This study can be applied in the date palm fruit industry to produce extracts more enriched with phenolic and flavonoid components and/or empowered by more antioxidant functions.

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