

ORIGINAL ARTICLE

A sustainable valorization of industrial tomato seeds (cv Rio Grande): Sequential recovery of a valuable oil and optimized extraction of antioxidants by microwaves

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Abstract

Tomato seeds are a major waste of tomato paste manufacturing. In this study, an interesting yield of oil was obtained (21.69%) from the seeds of an industrial tomato by-product (Rio Grande variety), after a Soxhlet extraction. The fatty acid profile of the extracted oil was established by gas chromatography and the tomato seed oil (TSO) was rich in unsaturated fatty acids (79.83%). After that, microwave-assisted extraction (MAE) was used to recover phenolic compounds from the defatted tomato seeds (DTS). The effect of three independent variables (microwave power, irradiation time, and solvent fraction) on the total phenolic content (TPC) and the antioxidant activity (AA) was estimated by using response surface methodology (RSM). The approach of desirability was applied to define the best operative conditions for the simultaneous extraction of TPC and AA which resulted in 700 W for microwave, 70 s for irradiation time, and water as extracting solvent.

Practical applications

By adopting a sustainable development approach, agro-industrial manufactories could earn the challenge of turning their waste into a source of additional income while also saving on disposal costs. In this study, a process by exhaustion following a smart and green approach was designed to recover added-value nutrients from an industrial tomato byproduct. First, tomato seeds were used as a feedstock to extract a multi-useful oil (edible, biofuel, cosmetic, etc.). Then, the residue was extracted again with hydro-ethanol as a safe and green solvent, by using microwaves as a green and cheap technology to optimize the extraction of antioxidants. In addition, computational tools (RSM and desirability approach) were applied for optimization and modeling of the operative conditions.

Abbreviations: AA, antioxidant activity; Adeq Precision, adequate precision; Adj R^2 , adjusted R^2 ; CCD, central composite design; CV, coefficient of variation; DPPH, 1,1-diphenyl-2-picrylhydrazyl; DTS, defatted tomato seeds; FA, fatty acids; FAMES, fatty acid methyl esters; FAO, food and agriculture organization of the United Nations; ISO, international organization for standardization; MAE, microwave-assisted extraction; RSM, response surface methodology; TP, total phenols; TPC, total phenolic content; TS, tomato seeds; TSO, tomato seed oil.

1 | INTRODUCTION

In recent years, restrictions about the use of synthetic additives in food products have significantly increased since they have been closely linked to consumers' health damages like allergies and carcinogenesis (Jimenez-Lopez et al., 2020). Therefore, natural compounds like phenolics have gained a great interest thanks to their benefits including human health promoting (Cosme et al., 2020) and a potential application as natural ingredients or to be included in food's bioactive packaging (Arroyo et al., 2019). In this trend, fruit and vegetable wastes which are mainly residues of agriculture and agro-industry (stems, peels, kernels, and seeds) contain very high added-value nutrients comprising natural nutraceuticals (Kowalska et al., 2017) and unconventional oils which are useful both for edible or industrial purposes like food, pharmaceutical, cosmetics, and biofuel manufactures (Alves et al., 2021; Srivastava et al., 2021; Ying et al., 2018). In addition, the valorization of the residues would improve the industrial process both economically and environmentally, in sync with the new concept of a circular and sustainable bioeconomy (Caballero & Soto, 2019; Jimenez-Lopez et al., 2020).

Tomato (*Solanum lycopersicum* L.) is one of the most important worldwide vegetable crops with nearly an annual harvesting of 181 million tons (FAO, 2019). About 25% of the fresh crop is consumed by processing industries and these operations generate about 8.5 million tons of solid wastes per year, mainly composed of peels, seeds, and a minor residual fraction of the pulp (Coelho et al., 2019; Knoblich et al., 2005; Szabo et al., 2018). There is a lack of statistics about the outcome of these byproducts; only little amounts are principally used for animal feed or as fertilizers, whereas a major part of the remaining quantity is wasted and could induce an intricate disposal problem for the industry. Actually, they constitute a very interesting resource of valued compounds like carotenoids (especially lycopene) which are well-known to promote feed and health of organism (Coelho et al., 2019; Knoblich et al., 2005; Strati & Oreopoulou, 2014; Szabo et al., 2018). Indeed, tomato-derived byproducts may be attractive for both food, pharmaceutical, and cosmetic industries and can find a sustainable use of their added-value molecules (Navarro-González et al., 2011; Szabo et al., 2018). Tomato seeds (TS) are an important but underestimated fraction of the tomato pomace comparing to the peels. Indeed, TS show a high content of a valuable oil. This latter is rich in unsaturated fatty acids, especially linoleic and oleic acids (Botineştean et al., 2015; Eller et al., 2010; Yilmaz et al., 2015), and has physicochemical characteristics to be useful as edible (Giuffrè et al., 2017) and as a biofuel (Giuffrè et al., 2016; Sivasubramanian et al., 2020). Moreover, it has been demonstrated to have some biological activities like antioxidative and antimicrobial (Mechmeche et al., 2018) and antihyperlipidemic (He et al., 2020). It could also be used as an ingredient in cosmetic's formulations, thanks to its protective effect against hazardous radiation rays like gamma radiation (Ezz et al., 2018).

Rio Grande tomatoes are very large, meaty, pear-shaped tomatoes that mature to a deep red. These tomatoes are perfect for making tomato paste, sauce, and juice. Therefore, this variety is widely

used in the tomato-processing industry. Oil from seeds of this industrial variety was characterized for the first time in this paper and its fatty acid profile was established by GC-FID.

To the best of our knowledge, few papers broached total phenolic content in tomato seeds (TS). In general, the already-accomplished works have compared phenolic content of each fraction of fruit (Chandra & Ramalingam, 2011; Chandra et al., 2012; Toor & Savage, 2005). Furthermore, seeds can represent up to 60% of the tomato pomace and so constitute the major component of the solid waste (Azabou et al., 2020). That wherefore tomato seeds could be an attractive reservoir of polyphenols for which an efficient recovery needs to use optimized tools and procedures. Nevertheless, in all the above-mentioned studies, extraction was performed by conventional methods that require big volumes of solvents and can be unsafe both for the environment and the operators' health. In addition, the conventional extraction techniques are time-consuming with low yields (Rahimpour & Taghian Dinani, 2018). Hence, to develop new emerging technologies appears to be crucial for an efficient recovering of high added-value compounds from matrixes (Alexandre et al., 2018).

Microwave-assisted extraction (MAE) is one of the eco-extraction techniques which gives better extraction yield and enhances the quality of extracts while decreasing the extraction time and the solvent consumption in comparison to conventional extraction methods. Moreover, another important advantage of the MAE is its applicability as well as in a laboratory and on a pilot and industrial scale (Llompert et al., 2019; Vinatoru et al., 2017). Thus, MAE constitutes a sustainable and economical device to recycle agri-food waste into new resources and so that permits to the manufacturers a simultaneous generation of new gain and avoiding disposal annoyances (Sharma et al., 2021).

This study aims to recover valued nutrients from a byproduct of tomato manufacturing by (1) extraction of a good nutritional oil from the tomato seeds; (2) optimization of microwave-assisted extraction of antioxidants from the defatted tomato seeds (DTS) by using the response surface methodology (RSM); and (3) a multi-response optimization of total phenolic content and antioxidant activity was performed by applying the desirability approach to determine the optimal conditions of extraction. Likewise, a comparison of efficiency between the MAE optimized extract and conventional extraction was investigated.

2 | MATERIALS AND METHODS

2.1 | Reagents

The compounds 1,1-diphenyl-2-picrylhydrazyl (DPPH), sodium carbonate, gallic acid, and Folin-Ciocalteu phenol reagent were purchased from Sigma-Aldrich (Germany). Fatty acid methyl esters (FAMES) standards were purchased from SUPELCO (USA). All solvents used were of analytical grade and purchased from Prolabo (France).

2.2 | Preparation of samples

Tomato pomace, a solid waste from the processing of the Rio Grande variety, was collected from a local tomato-processing manufacturer (Sarl Conserverie AMOUR) located in Blida (Algeria), in August 2019. The producer sows this variety of tomato for local farmers and collects their crops. The moisture content of the fresh pomace (a mix of skins and seeds) was determined by the oven method at 105°C to obtain a constant weight and was found to be 71.5 ± 1.2 g water per 100 g of byproduct. The tomato-processing waste was air dried at room temperature for 96 hr until obtaining a constant weight and a moisture content of 8.2 ± 0.1 g water per 100 g of the sample. After that, seeds were mechanically separated from peels and dried again in an oven at 45°C for 24 hr to eliminate any traces of the remaining moisture. The water activity of sample was determined by the WA meter apparatus (Aqualab Pawkit from Meter Food, USA). The water activity of the powder of the tomato seeds was 0.55 ± 0.06 at 25°C. All analyses were carried out in triplicate.

Finally, the dried by-product was ground with an electrical grinder (model A11 Basic from IKA, Germany) and the powder obtained was passed through a 500 µm sieve and stored in sealed polyethylene bags at -18°C until further analysis.

2.3 | Tomato seed oil extraction and characterization

Oil content of tomato seeds was recovered according to the standardized method ISO 659 of International Organization for Standardization (ISO) (2009). Briefly, 25 g of TS powder was extracted with 250 ml of *n*-hexane, by Soxhlet apparatus (model: Kex250 with 230 VAC/50–60 Hz voltage/frequency, 450 W power consumption, and dimensions in cm (W × D × H), approx. 23 × 33 × 80 from Behr, Germany), during 8h. When the extraction was ended, the solvent was released with a vacuum rotary evaporator and the distillation flask was cooled to ambient temperature in a desiccator and weighed to the nearest milligram. This last procedure was repeated until a difference between two consecutive weights was smaller than 5 mg. Five extractions were performed, and tomato seed oil (TSO) yield was expressed in mean ± SD. Its value was calculated as follows:

$$Y (\%) = \frac{mf}{ms} \times 100, \quad (1)$$

Y is the yield of TSO, m_s is the mass of the sample, and m_f is the mass of TSO.

The identification and quantification of the TSO fatty acids were explored. For this aim, the methylation of the fatty acids was carried out according to ISO 12966-2:2017 (ISO, 2017). Then, the identification of the methyl esters was elaborated by gas chromatography according to ISO 12966-4:2015 (ISO, 2015). Analysis was carried out by using a gas chromatograph (6890 Network GC System from Agilent Technologies, USA) equipped with a split/splitless injector

and a flame ionization detector and fitted with a capillary column DB 23 Agilent 122-2362 (60 m × 0.25 mm internal diameter × 0.25 µm film thickness). The carrier gas was H₂ with a debit of one mL/min and a pressure of 14.84 psi. One microliter of the sample was injected in split mode with a ratio of 1:50. Initially, the oven temperature was maintained at 130°C for 1 min, afterward increased from 130 to 170°C at a rate of 6.5°C/min, then from 170° to 215°C at 2.75°C/min, and held at this temperature for 12 min; finally, it was quickly (40°C/min) increased to 230°C and held isotherm for 3 min. Injector and detector temperatures were set at 250 and 270°C, respectively.

2.4 | RSM experimental design

As a part of the MAE optimization procedure, and to limit the total experimental work, the influences of the process parameters were separately investigated in single-factor experiments; each variable not studied was kept constant. Optimal extraction conditions were determined using response surface methodology (RSM) and central composite design (CCD) was chosen to investigate the effects of three independent variables: microwave power, irradiation time, and solvent fraction at three levels on the dependent variables total phenolic content (TPC) yield and antioxidant activity (AA). CCD uses the method of least squares regression to fit the data to a quadratic model.

The quadratic model for each response was as follows:

$$y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \sum \beta_{ij} X_i X_j, \quad (2)$$

where y is the predicted response, β_0 is a constant, β_i is the linear coefficient, β_{ii} is the quadratic coefficient, β_{ij} is the interaction coefficient of variables i and j , and X_i and X_j are independent variables. The software uses this quadratic model to build response surfaces. Statistical significance of the model and model parameters was determined at the 5% probability level ($p = .05$). Three-dimensional response surface plots were generated by keeping one independent variable at its zero level and plotting this against two factors (independent variables).

To verify the adequacy of the models, additional extraction trials were carried out at the optimal conditions predicted with the RSM and the obtained experimental data were compared to the values predicted by the regression model. The efficiency of the MAE was compared to conventional extraction methods based on TPC and antioxidant activity (according to DPPH assay) which were measured only on the extracts obtained under the optimum conditions selected by RSM.

2.5 | Microwave-assisted extraction

A domestic microwave oven (NN-S674MF from Samsung, Malaysia) with cavity dimensions of 22.5 cm × 37.5 cm × 38.6 cm and 2,450 kHz working frequency was used. The apparatus was equipped with a digital control system for irradiation time and microwave power (the

latter is, linearly, adjustable from 100 to 900 W). The oven was modified in order to condensate into the sample the vapors generated during extraction giving a constant sample volume.

For the extraction, a quantity of one gram of the defatted tomato seeds sample was placed in a round-bottom flask with volume of 30 ml of solvent. The flask was placed in a microwave oven and connected to a cold-water condenser. After irradiation, the flask was taken out and cooled in ice water bath. The solution was filtered through Whatman filter paper No.1 under vacuum.

2.6 | Conventional extractions

Extractions by maceration and stirring were carried out in order to compare with MAE. One gram of sample was extracted with 30 ml of hydroalcoholic solvent (40:60 ethanol:water) for 30, 60, and 120 min by stirring (with the help of a magnetic stirrer) at 30°C and for 1, 6, and 24 hr by maceration in a water bath kept at a temperature of 40°C. After each extraction, the mixture was filtered through Whatman filter paper No.1 under vacuum. Temperature and solvent fraction parameters were previously optimized. All extractions were repeated at least three times.

2.7 | Total phenolic compounds content

The content of TPC was determined by the Folin-Ciocalteu method as reported by Velioglu et al. (1998). Briefly, one milliliter of Folin-Ciocalteu reagent (diluted ten times by water) was mixed with 100 μ l of the extract. After 5 min, 1 ml of an aqueous solution of sodium carbonate (6%) was added and the mixture was kept for 60 min at room temperature. Absorbance was measured at 755 nm and concentrations were calculated using an ethanolic solution of gallic acid as standard.

2.8 | Antioxidant activity

The antioxidant activity of the extracts was estimated by the DPPH method, according to the procedure described by Haddadi-Guemghar et al. (2014). An aliquot of 0.5 ml of sample solution was mixed with 1.5 ml of ethanolic solution of DPPH (0.2 mM). The reaction mixture was incubated for 30 min in the dark at room temperature. The absorbance of the resulting solution was measured at 515 nm with a spectrophotometer (model UV1800 from Shimadzu, Japan). Ethanol instead of sample solution was used as a control. DPPH scavenging capacity of the tested samples was measured as a decrease in the absorbance and was calculated using the following equation:

$$\text{Scavenging activity(\%)} = \frac{A_c - A_s}{A_c} \times 100, \quad (3)$$

where A_c and A_s are the absorbances at 515 nm of the control and sample, respectively.

2.9 | Statistical analysis

Each extraction trial and all the analyses were carried out in triplicate and all the data in this work are reported as means $\pm$ SD. The influence of each factor on TPC yield in the single-factor experiment for the MAE was statistically assessed by ANOVA and Tukey's post hoc test with 95% confidence level and RSM was performed using the Design Expert software (Version 11.1.0.1. Stat-Ease, Inc. Minneapolis, MN 55413, USA). Regression and variance analyses were confirmed, and Derringer's desirability function was elaborated using the JMP Pro software (version 13.0.0, SAS institute Inc, USA).

The desirability function approach is based on using mathematical transformations to convert a multi-response problem into a single response one. First, each response is transformed into an individual desirability function d_i , with a range of values between 0.00 and 1.00 that measures how desirable it is the response of interest. A zero level means that performances of the corresponding response are unacceptable, whereas a desirability of one refers to the most satisfactory level of the response's performance (Del Castillo et al., 1996; Derrien et al., 2017). Individual desirability functions for modeling TPC and AA of DTS were calculated, as below, by using the Equation (4):

$$d_i = \begin{cases} 1 & \text{if } y_i \geq y_{i \max} \\ \left[\frac{y_i - y_{i \min}}{y_{i \max} - y_{i \min}} \right]^r & \text{if } y_{i \min} \leq y_i \leq y_{i \max}, \\ 0 & \text{if } y_i \leq y_{i \min} \end{cases} \quad (4)$$

where d_i is the individual response desirability; y_i is the response values, $y_{i \min}$ and $y_{i \max}$ are, respectively, the lower and upper limits (acceptable values) for response i , and r is a weight used to determine the scale of desirability.

After the individual desirability function is calculated for each of the n responses of interest, the composite (overall) desirability is defined as the weighted geometric mean (D) of the individual desirabilities d_i (from 1 to n); pre-defined weights $w_1, w_2, \dots, w_n$ for each of the characteristics are specified, and the aim is then to determine the factor settings impact that increases the composite desirability function D (Del Castillo et al., 1996; Derrien et al., 2017; Goethals & Cho, 2012). The Equation (5) allows the computing of the overall desirability of the two response TPC and AA of the DTS.

$$D = \{d_1^{w_1} \times d_2^{w_2} \times d_n^{w_n}\}^{1/\sum_{i=1}^n w_i} \quad (5)$$

3 | RESULTS AND DISCUSSION

3.1 | TS oil content

TSO extraction yield (expressed in percentage of dry weight) was $21.6\% \pm 1.5\%$. This latter was in accordance with the result reported by Lazos et al. (1998) with 21.8%. However, it was lower than those reported by Giannelos et al. (2005) who extracted TSO by Soxhlet,

using *n*-hexane as a solvent and obtained high yields between 32.1% and 36.9%. In fact, TSO yield could be affected by several factors like method and solvent extraction (Botineştean et al., 2015; Eller et al., 2010), cultivar (Giuffrè & Capocasale, 2015), and factors' condition of extraction (Botineştean et al., 2015; Shao et al., 2012).

In addition to the importance of extraction yields, the physico-chemical composition of an oil is very determinant to statute on its value. Particularly, the fatty acid (FA) composition is a very crucial parameter to define the quality of an oil and its eventual use. The chromatogram of FAMES illustrated in Figure 1 shows the detection of fourteen fatty acids (FA). Unsaturated FA represented, approximately, 4/5 parts of the total FA composition. Linoleic acid C18:2 was the predominant with 52.99%, followed by oleic acid C18:1 (23.5%) and the distinctive saturated FA, with a representative amount, was the palmitic acid C16:0 with 13.81%. It was also noticed the detection of a very appreciable amount (2.18%) of linolenic acid C18:3 (omega 3) which is, with the linoleic acid (omega 6), the essential fatty acid to the human metabolism and they must be provided by diet. The full TSO fatty acid composition indicated in Table 1 agreed with the results reported by Giannelos et al. (2005). Even more interestingly, Yilmaz et al. (2015) observed that roasting treatment of tomato seeds did not significantly affect the FA composition of their cold-pressed oil. Additionally, Lazos et al. (1998) reported that refining of TSO did not alter the FA composition. These findings indicate that processing operations do not affect FA profile of TSO, which makes of the byproduct (TS) an attractive source of a good quality oil.

3.2 | Preliminary study of TPC recovery optimization

As above-mentioned (Section 2.4), the effects of influential parameters of extraction: microwave power, irradiation time,

solvent type, and solvent-to-solid ratio were systematically studied in a single factor impact in order to determine the appropriate experimental ranges and to limit the number of factors to be considered during the optimization process. The results are shown in Table 2.

3.3 | Effect of solvent type

The solubility of the extract of interest, the interaction between solvent and plant matrix, and the solvent dielectric constant are important factors, which dictate solvent choice for MAE. Furthermore, the capacity of the extracting solvent to absorb and transmit the microwaves will, mainly, determine the efficiency of MAE (Doulabi et al., 2020; Mandal & Mandal, 2010).

Different types of solvents (80% methanol, 80% ethanol, and 80% acetone (v/v)) were used to investigate the best solvent for the recovery of TPC when the other extraction conditions were set as follows: microwave power 500 W, extraction time 60 s, and ratio of liquid to solid 20 ml/g (Table 2). Type of solvent influenced significantly the TPC yield, and aqueous ethanol was the best one. Due to that, it was selected as the solvent for the RSM trials.

3.4 | Effect of microwave power

Different microwave powers were set at 200, 400, 600, and 800 W to investigate its influence on the recovery of TPC when the other extraction conditions were set as follows: extraction time 60 s, 50% ethanol, and ratio of liquid to solid 20 ml/g. Table 2 shows that the extraction of TPC was greatly influenced by the microwave power. The extraction increased with the increase of microwave power. The range 300–900 W was selected for the RSM study, whereas the 800 W power was used for the last single-factor trials.

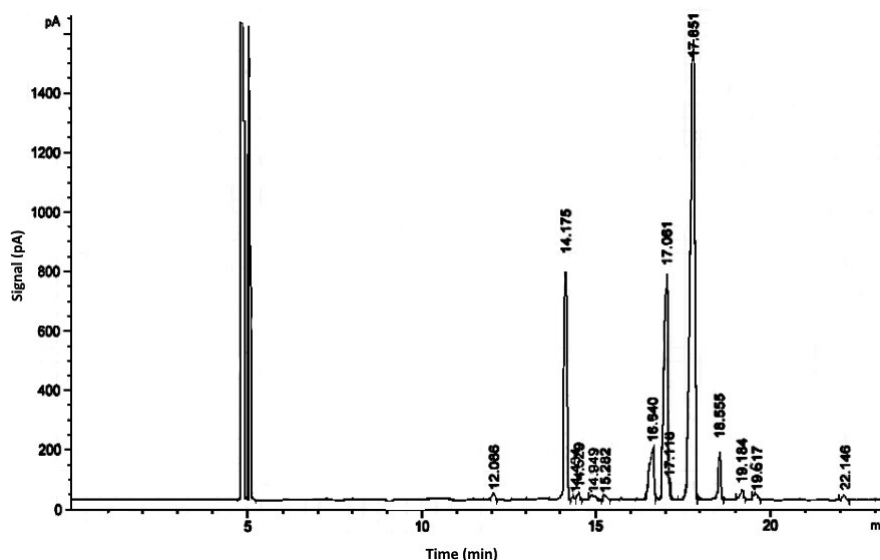


FIGURE 1 GC/FID chromatogram of tomato seed oil FAMES

TABLE 1 Fatty acid composition (FAMES) of tomato seeds oil

Fatty acid	Chemical structure	Retention time (min)	Fraction (%)
Myristic acid	C14:0	12.086	0.14
Palmitic acid	C16:0	14.175	13.81
Palmitoleic acid	C16:1cis ω9	14.434	0.07
Palmitoleic acid	C16:1cis ω7	14.529	0.31
Margaric (heptadecanoic) acid	C17:0	14.949	0.15
Margaroleic (cis-9-heptadecenoic) acid	C17:1	15.282	0.13
Stearic acid	C18:0	16.640	5.53
Oleic acid	C18:1cis ω9	17.061	23.50
Oleic acid	C18:1cis ω7	17.116	0.54
Linoleic acid	C18:2	17.851	52.99
Linolenic acid	C18:3	18.555	2.18
Arachidic (eicosanoic) acid	C20:0	19.184	0.42
Gadoleic (eicosenoic) acid	C20:1	19.617	0.11
Behenic acid	C22:0	22.146	0.12
SFA	–	–	20.17
USFA	–	–	79.83
MUFA	–	–	24.66
PUFA	–	–	55.17
USFA/SFA	–	–	3.96
PUFA/SFA	–	–	2.73

Abbreviations: MUFA, monounsaturated fatty acids; PUFA, polyunsaturated fatty acids; SFA, saturated fatty acids; USFA, unsaturated fatty acids.

3.5 | Effect of irradiation time

The irradiation time is another important factor that influences the extraction process of MAE. A long duration of extraction can increase the recovering of analytes but thermo labile components can be degraded (Chan et al., 2011; Doulabi et al., 2020). The recovery of TPC as affected by extraction times (20, 40, 60, 80, 100, and 120 s) is shown in Table 2, when the other factors (microwave power, ethanol proportion, and liquid-to-solid ratio) were kept at 800 W, 50% ethanol:water (v/v), and 20 ml/g. The results indicate that the recovery of TPC increased with the increase of MAE time. The recovery could reach its maximum after 60 s and after 120 s of MAE. Based on these results, 60 s was selected for the next single-factor trials, whereas the range 20–100 s was selected for the RSM trials.

3.6 | Effect of liquid-to-solid ratio

The solvent volume must always be enough to ensure that the entire sample is immersed during the extraction process. Generally, there is an increased extraction performance with a higher volume of solvent in conventional extraction techniques (Mandal &

Mandal, 2010). However, that may give a lower yield in MAE because higher volumes of solvent absorb more microwaves energy and can significantly attenuate the energy received by the matrix of interest that plausibly impacts the heating efficiency and the transfer of analytes to the medium (Doulabi et al., 2020). The recovery of TPC as affected by different ratios of liquid to solid (20, 30, 40, and 50 ml/g) can be seen in Table 2, when the other three factors: microwave power, extraction time, and ethanol proportion were fixed at 800 W, 60 s, and 50% ethanol:water (v/v). The result implied that the recovery of TPC was enhanced to the critical value of 296.45 ± 11.75 mg GAE/100 g at the ratio of 30 ml/g. Thus, the ratio of 30 ml/g was selected for the RSM optimization.

3.7 | RSM optimization of microwave-assisted antioxidants extraction

The experimental domain was defined by considering preliminary experiments. Each factor was evaluated at three levels at the following intervals: 300–900 W for microwave power, 20–100s for extraction time, and 0%–80% for ethanol fraction. However, the liquid-to-solid ratio was fixed in all experiments at 30 ml/g. The extraction efficiency of the microwave process was estimated by measuring TPC and the antioxidant activity of tomato seeds extracts.

3.8 | Fitting the model

Table 3 lists the analysis of variance (ANOVA) for the fitted quadratic polynomial models of TPC and antioxidant activity. *F*-test suggested that models have a very high model *F*-value and a very low *p*-value ($p < .0001$), indicating these models were highly significant. The lack of fit measures the failure of the model to represent the data in the experimental domain at points, which are not included in the regression. As shown in Table 3, *p*-values of the lack of fit of all responses were superior to .05, which implied that they were not significantly relative to the pure error and indicated that the model equations were adequate for predicting the yield of TPC and antioxidant activity under any combination of values of the variables.

To evaluate the goodness of the fit of the models, different descriptive statistical analysis such as the determination coefficient (R^2), the adjusted determination coefficient (Adj R^2), the predicted determination coefficient (Pred R^2), adequate precision (Adeq Precision), and coefficient of variance (CV) were used (Table 3). The R^2 values for TPC and antioxidant activity were .9699 and .9515, respectively, which implied that the sample variation was statistically significant at 96.99% and 95.15% for TPC and antioxidant activity. In both of the two responses, only less than 5% approximatively of the total variance could not be explained by the models. In other terms, a high value of the determination coefficient is close to indicate a high degree of correlation between the observed and predicted data. The Adj R^2 and the Pred R^2 of both TPC and antioxidant activity were also satisfactory to confirm the significance of the models. The Pred

TABLE 2 Results of single-factor experiments for microwave-assisted extraction

Solvent (500 W; 60 s; 20 ml/g) ¹		Microwave power (50% Ethanol; 60 s; 20 ml/g) ¹		Irradiation time (800 W; 50% Ethanol; 20 ml/g) ¹		Solvent/sample ratio (800W; 60 s; 50% Ethanol) ¹	
Type	TPC yield (mg GAE/100 g)	W	TPC yield (mg GAE/100 g)	s	TPC yield (mg GAE/100 g)	ml/g	TPC yield (mg GAE/100 g)
80% Methanol	208.23 ± 4.76 ^b	200	176.65 ± 2.22 ^d	20	229.55 ± 6.43 ^b	20	223.61 ± 15.72 ^c
80% Ethanol	253.16 ± 7.12 ^a	400	213.75 ± 3.47 ^c	40	227.14 ± 7.91 ^b	30	296.45 ± 11.75 ^a
80% Acetone	181.53 ± 3.84 ^c	600	245.75 ± 4.31 ^b	60	254.73 ± 8.89 ^a	40	291.73 ± 6.65 ^a
		800	273.53 ± 7.84 ^a	80	186.55 ± 6.38 ^c	50	273.14 ± 5.17 ^b
				100	228.97 ± 5.9 ^b		
				120	257.34 ± 9.02 ^a		

Note: Results are reported as means ± SD. Different letters in the same column refer to a significant difference according to ANOVA and Tukey's test ($p < .05$). TPC, total phenols yield referred to dry weight (dw) of tomato seeds.

Abbreviation: GAE, gallic acid equivalents.

¹Refers to fixed parameters in each trial series.

TABLE 3 ANOVA and regression analysis for the effect of microwave power, irradiation time, and ethanol fraction on TPC and antioxidant activity using a quadratic response surface model

Source	TPC		Antioxidant activity	
	F-value	p-value	F-value	p-value
Model	35.78	<.0001	21.81	<.0001
A-Microwave power	3.39	.0954	14.51	.0034
B-Irradiation time	41.01	<.0001	80.25	<.0001
C-Ethanol fraction	34.47	.0002	0.4828	.5030
AB	0.5619	.4707	8.31	.0163
AC	6.64	.0276	8.40	.0159
BC	1.40	.2639	14.64	.0033
A ²	36.24	.0001	4.13	.0695
B ²	11.31	.0072	25.47	.0005
C ²	19.61	.0013	45.42	<.0001
Lack of fit	0.8109	.5881	1.18	.4308
R ²	.9699		.9515	
Adjusted R ²	.9428		.9079	
Predicted R ²	.8245		.7129	
Adeq precision	19.1317		18.7911	
C.V. %	7.50		3.43	

R² were in reasonable agreement with the Adj. R²; the difference between the two parameters was less than .20 in both TPC and antioxidant activity models. The Adeq Precision measures the signal-to-noise ratio in which the value greater than 4 indicates an adequate signal and so the model could be used to navigate the design. The models TPC and AA indicated high values of adequate precision (19.13 and 18.79, respectively), which means both of them could be used for optimization.

Coefficient of variation (CV) describes the extent to which the data were dispersed. In general, a small value of CV gives a better reproducibility, and a high CV indicates that variation in the mean value is high and does not satisfactorily develop an adequate response model. As a rule, the CV should not be greater than 10%. The CV for TPC and antioxidant activity was within the acceptable range (Liyana-Pathirana & Shahidi, 2005).

3.9 | Significance of the variables

The significance of each coefficient was determined using the *p*-value in Table 3. The *p*-value is used as a tool to check the significance of each coefficient and the interaction strength between each independent variable. The *p* values less than .05 indicate that model terms are significant. In this case, irradiation time, ethanol fraction, and quadratic terms of microwave power were the major factors affecting the extraction of TPC followed by quadratic terms of ethanol fraction and irradiation time. It should be noted that interaction of microwave power with ethanol fraction also had a significant influence on TPC extraction that could be explained by dielectric properties of the extracting solvent and its capacity to absorb microwave energy using mechanisms of ionic conduction and dipole rotation. Thus, polar solvents, which are more absorbing microwaves energy than less polar ones, enhanced TPC extraction yields (Chan et al., 2011; Olalere & Gan, 2020).

Irradiation time and its quadratic terms besides to quadratic terms of ethanol fraction were the factors, mostly, affecting the antioxidant activity. Furthermore, it can be seen that ethanol fraction (C) did not influence the antioxidant activity but its interaction with other factors and its quadratic terms did it. A negative interaction was shown between microwave power and ethanol fraction, which means that at the low microwave power the best extraction of antioxidants was obtained by 80% ethanol; however, at high microwave power, the best extraction of antioxidants was obtained by water. This result suggests that the heat generated by microwaves

increases the coagulation of proteins by high concentration of ethanol and so we have obtained a best antioxidant activity with 80% ethanol at low microwave powers and a best antioxidant activity with water at high microwave powers.

After neglecting all the non-significant terms ($p > .05$), the fitted quadratic models for TPC and antioxidant activity in coded variables are given in Equations (6) and (7), respectively,

$$\begin{aligned} \text{TPC} = & +259.69 + 29.66B - 18.22C \\ & + 13.52AC - 53.78A^2 - 30.07B^2 - 39.76C^2 \end{aligned} \quad (6)$$

$$\begin{aligned} \text{AA} = & +65.10 + 2.80A + 6.58B + 2.37AB \\ & - 2.38AC + 3.14BC - 7.07B^2 + 9.44C^2 \end{aligned} \quad (7)$$

where A, B, and C are the coded terms for the independent variables: microwave power, irradiation time, and ethanol fraction, respectively. Square letters are the quadratic terms and the product of two letters are the interaction terms between the independent variables.

3.9.1 | Analysis of response surface

The 3D response surface is the graphical representation of a regression equation. It provides a method to visualize the relationship between responses and experimental levels of each variable and the type of interactions between two test variables (Yolmeh & Jafari, 2017). The plots were generated by plotting the response using the z-axis against two independent variables while keeping the other independent variable at zero level.

Figure 2a is a response surface plot showing the effect of microwave power and irradiation time on the TPC yield at the fixed ethanol fraction of 40%. The microwave power was shown as a negative quadratic effect on the yield ($p < .01$). The yield of extraction increased up to about 600 W followed by a decline with further increase of microwave power. The accelerated extraction of TPC using microwave power can be correlated with the direct effects of microwave energy on phyto-molecules by

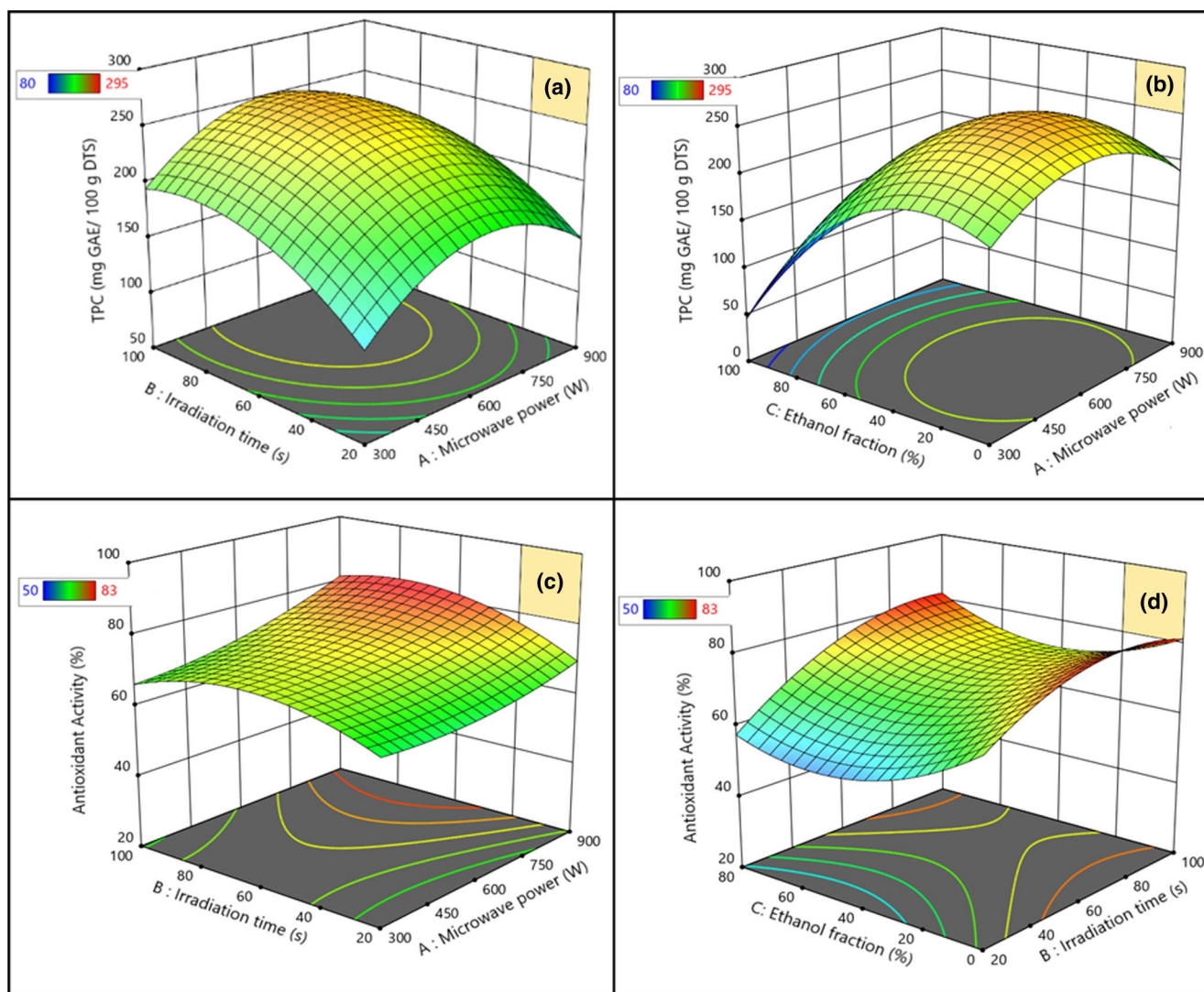


FIGURE 2 3D surface plot of TPC yields as a function of (a) microwave power and irradiation time at ethanol fraction of 40%; (b) of microwave power and ethanol fraction at irradiation time of 60 s; and (c) of the antioxidant activity as a function of microwave power and ethanol fraction at irradiation time of 60 s

ionic conduction and dipole rotation, which produces power dissipated in a volumetric fashion inside the solvent and plant material that then generates molecular movement and heating. It is known that the temperature of the extraction medium increases with increased microwave power. These increased temperatures result in improved extraction efficiencies, as desorption of analytes from active sites in the matrix will increase; however, very high temperatures may cause degradation of analytes. For a long extraction time, the negative quadratic effect was also significant ($p = .0072$). This was reflected in the plateau of the TPC yield for the extraction time over 80s. A further increase in extraction time resulted in little change for the yield of TPC. The irradiation time was the factor mostly affecting the extraction of TPC from tomato seeds by microwaves. The increase of the irradiation time from 20 to 80 s, over a microwave power of 600 W, improved TPC yield up to 25%.

The effect of polarity of solvent on yield of TPC is presented in Figure 2b. The yield increased significantly with an increase of ethanol concentration up to 31% and then declined (Figure 2b). The optimum was obtained with low concentration of ethanol. The negative effect of ethanol at high levels was probably due to the coagulation of proteins, which make a resistance to phenolic compounds diffusion. Similar results were found by Yang et al. (2009) who obtained a best extraction of crocin from *Gardenia jasminoides* Ellis fruits by low concentration of ethanol. The same phenomenon was recently observed by Doulabi et al. (2020) when varying ethanol concentration to extract phenolic compounds from eggplant byproduct and a negative correlation was remarked between TPC recovery and ethanol-to-water ratio.

Figure 2c is a response surface plot showing the effect of microwave power and irradiation time on the antioxidant activity at the fixed ethanol fraction of 40%. The microwave power was shown as a linear effect on the antioxidant activity ($p = .0034$). The yield of extraction increased with the increasing of microwave power. The irradiation time exhibited significant linear and negative quadratic effects on the antioxidant activity (Table 3 and Figure 2c). The increase of the irradiation time from 20 to 76 s, over a microwave power of 900 W and water, improved antioxidant activity up to 16%.

3.9.2 | Optimal extraction conditions

A quadratic model was successfully used to describe the extraction of antioxidants from DTS by microwaves, and we optimized three experimental variables for maximal extraction of phenolic compounds and

antioxidant activity. The results of the optimization are summarized in Table 4. When comparing the optimal conditions based on TPC to those obtained for antioxidant activity, it was found that the optima of irradiation time were closely related. However, there was a difference in the microwave power and the ethanol concentration required for optimal extraction of TPC and antioxidant activity. It could mean that after exposure to high microwaves energy, other bioactives molecules (very polar and non thermolabile) like sugars and polar amino acids were extracted with water and contributed actively to the AA of the extract.

Simultaneous optimization, using the desirability function (Figure 3), indicated that the optimum conditions for extraction of antioxidants and phenolics resulted in $A = 711$ W, $B = 70$ s, and $C =$ ethanol 0%, with a desirability value of 0.75. At this point, the investigated responses were theoretically calculated (with Equations 6 and 7) as TPC: 231.37 mg GAE/100 g DW and antioxidant activity: 78%. These results make in evidence the advantage of applying simultaneous optimization because when optimizing only TPC, a high value was found for this variable, but the value obtained under these conditions for antioxidant activity was not as good as when optimizing only antioxidant activity. In the same way, when optimizing only antioxidant activity, a high value was found for this variable, but the value obtained under these conditions for TPC was lower than that found in the previous analysis. In this regard, the simultaneous optimization achieves a compromise finding good values for both variables that are being optimized.

3.9.3 | Validation of the models

The optimized conditions obtained by RSM were used to validate the predictive model of extraction for TPC and antioxidant activity from tomato seeds. Table 4 shows that experimental values are reasonably close to the predicted values, confirming the validity and the adequacy of the predicted models. The experimental data were within 95% confidence interval of predicted values.

3.9.4 | Comparison between MAE and conventional extraction methods

The efficiency of TPC extraction and antioxidant activity (DPPH test) using MAE was compared to solid/liquid extraction by maceration and by stirring. The conditions of different techniques and their results are summarized in Table 5. The use of microwaves in the extraction of

TABLE 4 Optimum conditions for the MAE of TPC (mg GAE/100 g DW) and antioxidant activity (%) from tomato seeds^a

	Microwave power (W)	Irradiation time (s)	Ethanol fraction (%)	Prediction	Experimental
TPC (mg GAE/100 g)	600	78.90	32.41	268.47	265.31 ± 7.87
Antioxidant activity (%)	900	76.34	0	84.27	82.97 ± 2.71

^aAll values represent means ± SD.

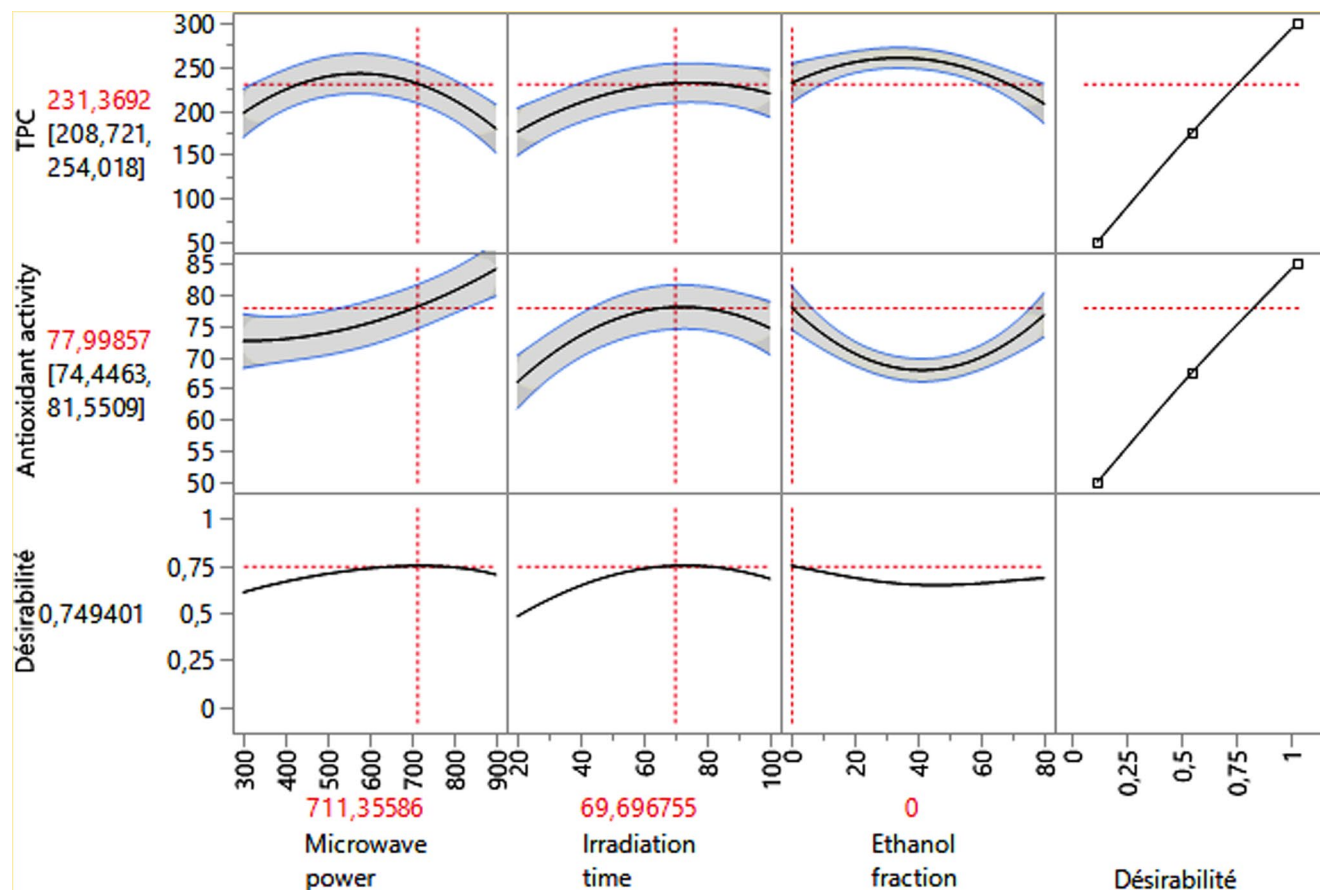


FIGURE 3 Profiles for predicted values and desirability function

TABLE 5 Comparison between microwaves-assisted extraction and conventional extraction

	Extraction methods	Irradiation time	Ethanol fraction	TPC (mg GAE/100 gDW)	AA (%)
TPC (mg GAE/100 g)	MAE	74 s	0% ethanol	266.9 ± 10.7 ^a	82.3 ± 4.8 ^a
	Maceration	2 hr	40% ethanol	171.3 ± 2.2 ^c	16.9 ± 0.7 ^d
		6 hr	40% ethanol	166.1 ± 2.3 ^d	14.0 ± 2.5 ^d
		24 hr	40% ethanol	145.6 ± 4.9 ^e	14.7 ± 1.1 ^d
	Stirring	0.5 hr	40% ethanol	171 ± 11.2 ^c	31.7 ± 4.7 ^c
		1 hr	40% ethanol	181.6 ± 9.7 ^b	48.2 ± 2.0 ^b
		2 hr	40% ethanol	181.1 ± 13.6 ^b	43.4 ± 1.8 ^b

Note: Results are reported as means ± SD. Different letters in the same column refer to mean statistically different according to ANOVA and Tukey's test ($p < .05$). TPC, total phenols yield referred to dry weight (dw) of tomato seeds.

Abbreviation: GAE, gallic acid equivalents.

phenolic compounds from tomato seeds increased significantly the antioxidant activity ($p < .05$) and reduced efficiently the extraction time to only 79 s. The latter was far less than those of conventional methods. Indeed, the MAE method was able to extract nearly 24% more TPC from tomato seeds in only 1/30 of the time duration required for the solid/liquid stirring extraction (Table 5). Many studies reported that MAE applied to several materials reduces significantly extraction time compared to conventional extraction methods (Djemaa-Landri et al., 2020; Haddadi-Guemghar et al., 2014; Hamzah & Leo, 2015).

4 | CONCLUSION

To conclude, tomato seeds are an interesting byproduct that contains, among other nutrients, a good value oil with a promising added-value to food and pharmacological industries. Even more interestingly, after its deoiling, the meal is still rich in other bioactive compounds like polyphenols. In this regard, the use of innovative methods like microwave ensured the ease and rapidity of extraction in comparison to conventional methods and the yield was better in

all cases. The results obtained indicate that industrial tomato by-products contain significant amounts of phenolic compounds, which are known to exert antioxidant activity. Therefore, in the perspective to be in line with the tendency of a sustainable approach to recycle/reuse agro-food wastes, on the one hand and to limit the use of synthetic ingredients, on the other hand. Tomato seeds are natural resources with value-adding constituents that could be used as natural antioxidants for the formulation of functional foods or serve as additives in food systems to increase their shelf life.

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CONFLICT OF INTEREST

The authors have declared no conflicts of interest for this article.

AUTHOR CONTRIBUTIONS

Toufik Ouatmani: Investigation; Writing-original draft. **Hayate Haddadi:** Methodology; Supervision; Writing-review & editing. **Lila Boulekbache-Makhlouf:** Validation; Visualization. **Djamila Terki:** Investigation. **Azzedine Maouche:** Validation. **Khodir Madani:** Formal analysis; Visualization.

DATA AVAILABILITY STATEMENT

Data openly available in a public repository that issues datasets with DOIs.

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