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Research Article

Optimization of Extraction Parameters by Response Surface Methodology in Handling Tea Extract From Fibrous Tea Waste

Mustafa Mortas[1*], Nour Awad[2]

[1*] Ondokuz Mayıs University, Faculty of Engineering, Food Engineering Department, Samsun, Turkey (ORCID: 0000-0002-0316-7768) mustafa.mortas@omu.edu.tr
[2] Ondokuz Mayıs University, Faculty of Engineering, Food Engineering Department, Samsun, Turkey (ORCID: 0000-0003-3441-9080) awadnur5@gmail.com

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Abstract

In Turkey, the extraction parameters of tea extract from the fibrous tea waste left after harvesting the tea crop is gaining an interest in the last few years. The handled tea waste extract can be used for ice tea and teabag production as a flavor and color enhancer. Therefore, the industry needs to implement a more efficient and cost-effective extraction protocol. At this point, optimizing the extraction parameters is the main goal of the study. Eight different response factors (Extract yield-EY, color measurements *L*, a*, b**, C (Chroma) and h (hue angle), total phenolic compounds-TTC, antioxidant activity by 2,2-diphenyl-1-picryl hydrazil radical (DPPH)- assay) were used with three of process parameters (tea waste/water ratio, temperature and time) and its predictive power has been demonstrated. In the formed model, the mean values for eight different responses as EY, *L*, a*, b**, C, h, TPC, DPPH were 2.37±0.83%, 1.42±0.35, 0.77±0.51, 6.64±0.41, 96.42±5.75 mg gallic acid/100g, 49.99±21.74 mg Trolox/g, respectively. The prediction model using the available responses was determined to be suitable only for L* and TPC. As a result of the modeling, for the optimum estimation point, tea waste/water ratio, temperature, and time parameters were determined 8%, 94.95°C, and 60 minutes, respectively. Besides, the desirability of the model was calculated by 0.839. Thus, the estimation of tea waste color and functionality parameters may aid in low-cost tea extract production. However, the large amount of generated tea waste could be used to produce both cold tea and teabags.
Keywords: Tea waste, Tea Extraction, Response Surface Methodology

Lifli Çay Atıklarından Çay Özütü Eldesinde Yanıt Yüzey Metodolojisi ile Ekstraksiyon Parametrelerinin Optimizasyonu

Öz

Türkiye'de yüksek miktarlarda çay üretimine bağlı olarak ortaya çıkan lifli çay atığının, ekstrakiyon işlemi ile çay özütü eldesindeki extraksiyon parametreleri son yıllarda önem kazanmaktadır. Elde edilen çay özütü ise buzlu çay üretiminde ve poşet çay üretiminde renk ve tad destekleyicisi olarak kullanılmaktadır. Bundan dolayı daha verimli ve maliyeti düşük bir ekstraksiyonun uygulanması endüstri açısından önemlidir. Bu noktada ise ekstraksiyon paramaterelerinin optimize edilmesi, çalışmanın ana hedefini oluşturmaktadır. Söz konusu parametrelerinden 3 tanesi (çay atığı/su oranı, sıcaklık ve süre) Yanıt Yüzey Metodolojisi kullanılarak sekiz farklı yanıt faktörü (Ekstrakt verimi-EY, L*, a*, b*, C (Chroma) ve h (hue angle), toplam fenolik madde-TPC, antioksidan aktivite 2,2-diphenyl-1-picryl hydrazil radical (DPPH) üzerinde modellenerek tahmin gücü ortaya konulmuştur. Oluşturulan 20 deneme deseni özelinde parametrelerin değer aralıkları çay atığı/su oranı, sıcaklık ve süre için sırasıyla 0.5-10%, 50-95°C, 1-50 dakika şeklinde belirlenmiştir. Oluşturulan modelde sekiz farklı yanıt özelinde ortalama değerler ise EY, L*, a*, b*, C, h, TPC, DPPH için sırasıyla 2.37±0.83%, 1.42±0.35, 0.77±0.51, 6.64±0.41, 96.42±5.75, 49.99±21.74 mg gallic acid /100g ve 35.89±17.82 mgTrolox/g şeklinde bulunmuştur. Mevcut yanıtların kullanıldığı tahminleme modeli sadece L* ve TPC için uygun olarak tespit edilmiştir. Modelleme sonucunda ise optimum tahminleme noktası (R^2) için çay atığı/su oranı 8%, sıcaklık, 94.95°C, süre 60 dakika olarak tespit edilmiş ve istenirliği 0.839 olarak hesaplanmıştır. Böylece çay atığının ilgili değişkenler özelinde elde edilecek renk ve fonksiyonelliğinin tahmin edilmesi ile birlikte daha az maliyet ve sürede çay özütü elde edilebilecektir. Böylece çay atığı rengi ve işlevsellik parametrelerinin tahmini, düşük

[*] Corresponding Author: Ondokuz Mayıs University, Faculty of Engineering, Food Engineering Department, Samsun, Turkey, ORCID: 0000-0002-0316-7768, mustafa.mortas@omu.edu.tr

maliyetli çay ekstresi üretimine yardımcı olabilir. Bununla birlikte, ortaya çıkan büyük miktardaki çay atığı hem soğuk çay hem de poşet çay üretimi için kullanılabilmektedir.
Anahtar Kelimeler: Çay atığı, Çay Ekstraksiyonu, Yanıt Yüzey Metodolojisi.

1. Introduction

Tea (*Camellia sinensis*), which has a growing habit of consumption, is one of the most consumed beverages in the world today (Groosman, 2011). Although coffee consumption has become popular until the beginning of the 20th century in Turkey (Tozlu, 2014), tea was known in the early 1600s (Ozdemir and Gokalp, 1989; Tekeli, 1976). On the other hand, the first serious attempt of the cultivation of the tea plant in Turkey was in 1888 (Kacar, 1987), and after that Turkey became one of most tea consuming countries all over the world (Tozlu, 2014). In 2015, the amount of tea production in the world was 5,305,000 tons (Zhao et al., 2018). As well as in the same year, the consumption of tea per capita was 3.2 kg in Turkey and the high production of tea meets the mentioned high consumption (1.327.934 tons) in different cities as Rize (%68), Trabzon (%20), Artvin (%10), Giresun (%2) and Ordu located in the Black Sea Region (TUIK,2016).

During the processing of tea, the mixture comprising of main four components 1) tea fiber, which is separated by electrostatic separators in dried black tea production, 2) tea litter originating from leaf stalk, 3) leaf veins that cannot turn into tea, 4) thick leaves is called 'tea waste' (Demir, 2015). Although this volume of waste is about 3-5 percent compared to the weight of wet leaves, due to the non-standard collection of green tea in the Eastern Black Sea Region, it rises to 17-18 percent. (Kacar, 1987). However, it's estimated that Black Sea tea factories produce about 40,000 tons of tea wastes every year. Since those large amounts of tea wastes might cause environmental problems when dumped in the environment or landfilled, nowadays, there is a new perspective of getting economic benefits from those waste and prevent pollution (Tutus et al. 2015)

Recently the researchers investigated some novel methods for the processing of tea waste into various non-food materials. For example, it has been reported that using tea waste could be effective in increasing the hardness of concrete by filling the gaps (Kara, 2018), organic tea wast as a growing media for vegetable cultivation (Karatas and Buyukdinc, 2017), for paper production (Kazaskeroglu, 2012), for the production of sound isolation materials (Kaya and Dalgar, 2017), cleaning absorber materials (Cubuk et al., 2014), biofuel in pellet form (Yolcu, 2019; Bilgin et al., 2016), gel film (Tasar, 2018), compost material (Asık, 2012), activated carbon (Gundogdu, 2010), sound absorber materials (Ekici et al., 2012), tea gel materials (Oskuei, 2009), and for the preparation of microcrystalline cellulose (Zhao et al. 2018). While tea has high production volumes, there are very limited studies relating to tea waste as a food component. For instance, the possibility of tea waste for producing valuable chemicals, especially caffeine and catechins, was evaluated by Serdar et al., (2017).

Tutus et al. (2015) investigated the chemical composition (%) of tea wastes collected from tea factories operating in the Black Sea region of Turkey. Data showed high holocellulose (60.8±1.14), cellulose (29.42±0.57), and lignin (36.94±0.34) contents. However, lignin works as an adhesive that binds cellulose together in the fiber. When the lignin content is low, the fiber becomes stronger and harder to break. Thus, tea waste can be used as supporting material for ice tea and tea-bag production. In literature, the studies are limited to constant parameters. In a published study, the preparation of tea extract from tea waste has been carried out at five different sample amounts in the range of 7-25%, five different temperatures in the range of 25-95 °C, and with constant temperature extraction at 25°C (Safi, 2018). In another research, tea extraction has occurred at 4°C for 6 hours with 4 grams tea/50 mL water ratio (Demir, 2011).

The aim of this study was to investigate the optimum extraction parameters points for fibrous tea waste. In our study, the effect of temperature, duration time, and tea waste sample/water ratio on the final extraction process has been estimated by surface response methodology.

2. Material and Method

2.1. Material

The tea waste samples for this study have been obtained from a tea company in Rize/Turkey. It was collected after the separation (sieving) procedure during the tea production process. The photograph of the tea waste taken (Nikon Coolpix L820) can be easily seen at Figure 1.

Figure 1. Sample of tea fibrous waste

2.2. Methods

2.2.1. Moisture Content

For determining moisture content in the tea waste, 2 grams of tea waste sample is weighed and dried in the oven until a constant scale is reached at 103°C (Gürses and Artık, 1987).

*2.1.2.*pH Measurements

The pH measurements for all extracted tea samples were carried out in triplicate using a pH meter (Toledo AG, China), calibrated at (pH=4.0 and pH=7.0).

*2.1.3.*Extract Yield

The model was taken following the parameters determined in the pattern, 15 mL of the samples obtained from the extraction was transferred to the pre-tared petri dishes and then dried at 65°C until the weighing was fixed in the oven. Extract yield was calculated on a dry matter (Hanay, 2011).

*2.1.4.*Color Measurements

The color values of the samples were measured with the color measurement device (Minolta Chroma Meter, CR-400, Osaka, Japan). *L* * (darkness-lightness), *a* * (green-redness), *b* * (blue-yellowness) parameters of the samples were measured.

*2.1.5.*Total Soluble solids (TSS-°Brix)

Brix measurements were carried out on all the samples treated with different extraction parameters. Before taking the

measurements, the refractometer surface was cleaned with cotton and then calibrated with distilled water.

2.1.6. Determination of total phenolic content (TPC)

The TPC of tea extracts was measured using the Folin-Ciocalteu method as described by (Zannou and Koca 2020) with slight modification. Briefly, an aliquot of 150 µL of the diluted extract was mixed 750 mL of 10% Folin-Ciocalteu reagent. The mixture was shaken for 1 min before adding 600 mL of 7.5% sodium carbonate solution. The mixture was shaken again and placed in the dark for two hr before reading the absorbance. The absorbance was read at 760 nm using a UV-spectrophotometer (Agilent Technologies, USA), and the TPC was calculated from a calibration curve using gallic acid as a standard. The results were given as mg gallic acid equivalent (GAE) g^{-1} DW (Dry weight) (Zannou and Koca 2020).

2.1.7. Determination of the DPPH (2,2-diphenyl-1-picryl hydrazil radical) radical scavenging activity

The DPPH radical scavenging was determined using a modified method as described by (Zannou and Koca, 2020). Briefly, an aliquot of a 50 µL sample was added with a 1 mL DPPH solution (0.06 mM in 80% methanol). The mixture was shaken and left to stand in the dark for one hr until the reaction was completed. After that, the absorbance at 517 nm was recorded. The DPPH solution was used as control. The reduction ratio of DPPH was determined with the following equation:

$$\text{Reduction\%}=\text{Ac-AsAc}\times 100 \quad (1)$$

where Ac = Absorbance of control and As = Absorbance of extract. The DPPH radical scavenging activity in each extract was calculated from a calibration curve using Trolox as a standard. The results were given as mg Trolox equivalent (TE) g^{-1} dw (Dry weight).

2.1.8. Response Surface Methodology

The extracting conditions for tea waste were optimized with central composite design (CCD) using the responses of the sample/water ratio, extraction temperature, and time.

To facilitate mathematical operations in the model pattern, actual values were coded;

$$x_i = \frac{z_i - 0.5\,(z_{i.max}+z_{i.min})}{0.5\,(z_{imax}+z_{imin})} \quad (2)$$

calculated by the mentioned equation. The z-real value, x-coded values in the equation, min. and max. expressions show the minimum and maximum values. Besides, the Central Composite Rotatable Design, which is used in the experimental design to reveal combinations of extraction factors, is rotatable, enabling equal estimation in all directions with the α values selected for optimum point detection (Montgomery, 2008). In the model design, the endpoints can be divided into -1, + 1, -a, and + a, while 0 is the center point (Table.1). Thus, the answers required by the system are obtained with 20 different combinations, including minimum and maximum regions in the model pattern (Table 2).

In the model pattern of the water extraction process applied to tea, the sample/water ratio varies between 1-10%, extraction temperature between 50-95 °C, and extraction time between 1-60 minutes. Optimum production variables were estimated using the desirability method using Design-Expert (Version 7.0, Minneapolis, USA). Regression analysis, response surface graphics, and contour lines were also prepared with the same program.

2.1.9. Statistical Analyses

Tea waste extraction parameters design was selected as Central Response Surface Design (CCRD), and the optimum points were determined with Design Expert 7.0 (State-Ease, Minneapolis, USA) (Trial Version). Multiple comparison tests were performed with the Duncan test based on a 95% confidence interval using the SPSS package program (SPSS 16.0), and analyses were carried out in triplicate for each replicate.

3. Results and Discussion

3.1. Moisture Content

To estimate the amount of moisture content obtained from tea waste, dry matter/moisture analysis was carried out to be used in the calculations. The range of the moisture content of the dried black tea was stated as 1.5-7.0% (Safi, 2018). Besides, the moisture content of the tea waste was found in other researches as 4,61% (Yakupoglu and Peksen, 2011), 8.4%, (Yilmaz et al., 2016), 6.54% (Kırbaslar et al., 2001). In this study, the moisture content of the fibrous tea waste sample was found as 5.51%, and this value is similar to other tea waste samples at other researches. This value is important to be used in the calculation of the extraction yield.

3.2. pH

The pH is another indicator of the extraction parameter's effects. Liang (2001) stated that the pH effect on the solid extraction yield of tea was significant in increasing instant tea yield. Additionally, they claimed that the behavior of tea extracted at extreme pH levels might be of great interest yield. The low pH levels can ensure stability for green tea compounds like catechins, purine alkaloids, flavonoids, and phenolic acids (Jang et al., 2014). Zimmerman et al. (2011) found that low pH level is a decisive factor rather than adding chemicals for extractions of some compounds from green tea. The pH value of green tea extract was found slightly acidic as 5.92-6.22 by Das and Eun (2018). We found that similar acidic values at the trials. At the end of the trials, the pH values of the samples were found in a range between 4.25 (0,5-sample/water ratio, 72.5°C, 30 minutes) and 5.46 (8-sample/water ratio, 60°C, 50 minutes) (Table 4). Similar pH values were measured at a similar sample weight. When the sample ratio and extraction time parameters were getting higher and longer, the values decreased.

3.3. Extraction Yield

Extraction yield can be recommended as an indicator for water-soluble solid contents of the tea, and it is a criteria for the quality of dried black tea (Salman et al., 2019). While tea contains approximately 300-450 g/kg extractable solids, instant tea contains about 200 g/kg tea in the production scale (Chen, 1979). According to ISO-9768 for tea standards, this value should be 32% based on dry matter (Salman et al., 2019). Many studies on Turkish tea show that extract yield values are lower (Ozdemir et al., 1993). In literature, the researchers have already performed some studies on tea extraction and discussed different conditions. Torun et al., (2008) used the extraction

parameters as 60-80°C temperature, 0.5-300 minutes and only tea sample: water ratio 2:98 g/mL and they found the highest extraction value at 75°C and 80°C. In another study, extraction efficiencies of the samples were calculated as at 92-94°C during 15 minutes, and in addition to that, other trials at the same temperature during 20 minutes extraction were found as 31.63-36.05%. Extraction time for tea was not statistically important in the study, and this finding was supported by Salman et al., (2019). They stated the values were valid for Turkish type brewing infusions. Dincer et al., (2008) were investigated time and temperature effects on tea infusion. For this purposes, the researchers selected the parameters for temperature as (60, 65, 70, 75, 80 °C) and for time as (0.5, 1, 1.5, 2, 3, 5, 10, 15, 20 minutes). They found that increasing the temperature and time parameters for infusion have an effect on the extract yield (Salman et al., 2019). The main reason for the extraction yield increasing with temperature is that water-soluble compounds transfer from tea to the brewing water getting higher at high temperatures (Suteerapataranon et al., 2008). Although temperature and time parameters have an effect on extraction yield for both tea and tea waste, tea harvesting conditions are important for extraction yield (Salman et al., 2019). Hanay et al. (2011) stated that the extraction time has a direct positive effect on the extraction yield. A total of 24.14% increasing in extraction yield was found from 3 minutes to 30 minutes at the mentioned study. Golukcu et al. (2014) found a negative relationship between time and temperature parameters at the mountain tea infusion process.

Extraction yield is undesirable to be less than 32% in teas, but it is normal to be less than that value for tea waste. In literature, Bindes et al. (2019) found fewer values in their study. Consequently, the extraction yield of fibrous tea waste was not higher than black tea. It can be used for supporting material for teabag consumption in terms of aroma and color. The extraction steps must be carried out with the most extraction yield and with the lowest production cost. At one phase, water-based extraction, water-soluble dry matter amount is directly related to extraction yield. When the amount of the tea extract produced from tea waste in terms of extraction yield is high, it has a positive economic effect on ice tea and teabag. In literature, some tea extraction studies were performed by the researchers. Still, our research was different from the others in terms of tea waste extraction yield by using Response Surface Methodology.

In our study, the lowest extraction yield (6.63±1.29%) was found at 0.5% (g/mL) sample ratio at 72.5°C and 30 minutes during extraction, whereas the highest extraction yield (83.21±2.67%) was found at 10 % (g/mL) at 72.5°C and 30 minutes during extraction (Table 4). As seen in Table 6, the mean value and standard deviation of the extraction yield response was calculated as 2.37±0.83. The extraction yield's coefficient of variation (CV) value, a mean-dependent measure of the residual variation in experimental data, was found 34.95%. At that point, 'adeq precision' measures the signal to noise ratio, and it is desirable that the ratio greater than 4. The adeq precision value of the response was calculated as 5.698, and the value was greater than the limit value as 4. The regression coefficient of the model was found 0.6519, and the p-value of the model was found as 0.1345. As a result, low values of R^2 and Adj-R^2 indicates deviation from the mean value. According to the mentioned statistical parameters, the extraction yield response for the extraction model was not statistically important for the model (0.05<p). The obtained result was normal because the tea waste material has a fibrous and heterogeneous structure since it leaves different fiber and high cellulose content on the sieve during the separation process.

3.4. Total Soluble Solids (TSS)

The TSS is as important as the extraction yield for tea waste infusion, and it is another parameter that should be measured. Both of the parameters are important for the extraction process, and they are directly related to each other. Total soluble solids of the different Indian black tea were determined at the range of 18-30 (Khanum et al., 2017). For green tea, total soluble solids values were found at the range of 5.60 and 7.55 (Ozunlu and Ergezer, 2019). In our study, the extraction with a sample/water ratio of 10% and a temperature of 72.5°C and a duration of 30 minutes reached the highest Brix value (2.15±0.07), while the extraction with a sample/water ratio of 0.5% and a temperature of 72.5°C and a duration of 30 minutes was reached lowest Brix value (0.10 ± 0.00). The main reason for finding different values is related to many factors such as tea preparation technique, temperature, time, and concentrations of other added ingredients that affect the Brix value (Ozunlu and Ergezer, 2019).

In our study, the lowest total soluble solid value (0.30±0.00 Brix) was found at 0.5% (g/mL) sample ratio at 72.5°C and 30 minutes during extraction, besides that the highest total soluble solid value (2.15±0.07 %) was found at 10 % (g/mL) at 72.5°C and 30 minutes during extraction (Table 4).

For the TSS response, the mean value and standard deviation of the TSS response were calculated as 1.19±0.46. The coefficient of variation (CV) value of the response, a mean-dependent measure of the residual variation in experimental data, was found 38.65%. At that point, 'adeq precision' measures the signal to noise ratio, and it is desirable that the ratio greater than 4. The adeq precision value of the response was calculated as 4.634, and the value was greater than the limit value as 4. Regression coefficient of the model was found 0.5646, and the p-value of the model was found as 0.2879. As a result, low values of R^2 and Adj-R^2 indicates a deviation from the mean value. According to the mentioned statistical parameters, the total soluble solid response for the extraction model was not statistically important for the model (0.05<p) (Table 6).

3.5. Color Measurement

The *L**- lightness, *a**- redness-greenness, *b**- yellowness-blueness, C (Chroma), and h (hue angle) color parameters have been estimated. The color parameters are important to indicate the effects of different extraction conditions on the color. The color properties of the tea waste were similar to black tea ones. However, this property can support teabag production theory from these tea wastes. Golukcu et al. (2013) found the *L**, *a**, *b**, C and h parameters of mountain tea infusions as 21.16-24.21, 1.46-2.85, 5.16-7.52, 5.60-7.67 and 61.69-78.43, respectively. Whereas, *a** values have been increased as extraction temperature increasing. Some studies concern a relation between extraction yield and color parameters (Golukcu et al., 2013; Dincer et al., 2008)

The color parameters of the tea waste extract were investigated in terms of *L**, *a**, *b**, C and h values at the study. According to our results, the lowest value for *L** (23.03±2.03) was at 2.5% (g/mL) sample ratio at 60°C and 50 minutes, for *a** (-2.26±0.01) and for C (5.56±0.13) at 0.5% (g/mL) sample

ratio at 72.5°C and 30 minutes, for *b** (5.54±0.14) at 5% (g/mL) sample ratio at 95°C and 30 minutes, for h (89.51±1.73) at 5% (g/mL) sample ratio at 72.5°C and 30 minutes. On another hand, the highest values for *L** (25.39±0.01) at 0.5% (g/mL) sample ratio at 72.5°C and 30 minutes for *a** (-0.11±0.01) at 8% (g/mL) sample ratio at 85°C and 50 minutes, for b (7.59±0.89) at 8% (g/mL) sample ratio at 60°C and 50 minutes, for C (7.75±0.46) for h at 5% (g/mL) sample ratio at 72.5°C and 60 minutes (Table 4).

According to color measurement results, mean values and standard deviations of the *L**, a*, b*, C and h parameters as -24.48±1.42, 0.51±0.71, 6.64±0.41, 6.76±0.69, and 96.42±5.75, respectively. The coefficient of variation (CV) values of the responses, a mean-dependent measure of the residual variation in experimental data, were found 1.42%, 66.83%, 6.21%, 10.15%, and 5.97%, respectively. Also, 'adeq precision' values of the responses should be greater than four, and the values were calculated for L, a, b, C and h responses as 9.763, 4.870, 6.383, 2.299 and 2.344. As a result, low values of R^2 and Adj-R^2 indicates a deviation from the mean value. According to the mentioned statistical parameters, color measurement responses were not statistically important, except L response (0.05<p) (Table 6). All of the statistical parameters of the *L** response were statistically important for the model. According to the model, the equation of the response with the extraction parameters as sample/water ratio, extraction temperature and extraction time was given as;

$$L=+24.58-0.18X_1-0.22*X_2+0.22X_3+0.036*X_1*X_2-0.39X_1*X_3-0.32X_2*X_3-0.31*X_1^2+0.092*X_2^2+0.076*X_3^2 \quad (3)$$

X_1 :Sample/water ratio (%), X_2 : Extraction temperature (°C), X_3:Extraction time (minutes)

3.6. Total phenolic content (TPC)

It can be stated that there is an increase in product functionality (total phenolic substance, antioxidant activity) with increasing temperature and time of extraction (Golukcu et al., 2014), and similar results were found by Hanay (2011). Thus, there are many published researches that discussed the extracted phenolic compounds of tea at different parameters (water: tea ratio, temperature and minutes) (Bindes et al., 2019; Sousa et al., 2016; Kumar et al., 2012; Zhang et al., 2012; Rou et al., 2011). Golukcu et al. (2013) were found phenolic compounds of the tea as 195.81 mg/kg for 70°C and 211.64 mg/kg for 100 °C. According to the study, extracted phenolic content was increased concerning increasing time at every temperature stages. Bindes et al. (2019), Hanay (2011), and (Horzic et al., 2009) stated similar foundings as the time and temperature parameters directly affect the phenolic compounds' extraction.

The lowest total phenolic content value (21.87±2.17) was found at 2.5% (g/mL) sample ratio at 60°C and 12.5 minutes of extraction, whereas the highest total phenolic content value (139.12±4.11) was achieved at 5% (g/mL) at 72.5°C and 30 minutes of extraction conditions (Table 4).

For the TPC response, the mean value and standard deviation of the TPC response were calculated as 49.99±21.74. The coefficient of variation (CV) value of the response, a mean-dependent measure of the residual variation in experimental data, was found as 43.49%. At that point, 'adeq precision' measures the signal to noise ratio, and it is desirable that the ratio greater than 4. The adeq precision value of the response was calculated as 6.599, and the value was greater than the limit value as 4. The regression coefficient of the model was found 0.7846, and the p-value of the model was found at 0.0200. As a result, low values of R^2 and Adj-R^2 indicates a deviation from the mean value. Moreover, the lack of fit value of the response was determined as 0.2132, and the value was not statistically important (Table 6). Therefore, the situation is desirable for a model, and the equation of the TPC value can determine the following equation;

$$TPC=+36.37-5.66*X_1-16.36*X_2-12.90X_3-0.012\ X_1*\ X_2-7.44\ X_1*\ X_3+30.38\ X_2*\ X_3+3.15\ X_1^2\ +14.35\ X_2^2+2.44\ X_3^2 \quad (4)$$

X_1 :Sample/water ratio (%), X_2 : Extraction temperature (°C), X_3:Extraction time (minutes)

The parameters of the extraction for the TPC response were shown in Figure .3.

3.7.DPPH

The ORAC, TEAC, FRAP, and DPPH methods can be used for antioxidant activity determination in foods (Huang et al., 2005), and in this study, the DPPH assay have been applied. In our study, the lowest antioxidant activity (10.55±0.24 mg Trolox/ g) was found at 2.5% (g/mL) sample ratio at 60°C and 12.5 minutes during extraction, whereas the highest value (57.48±0.33 mg Trolox/ g) was achieved at 2.5 % (g/mL) at 85°C and 50 minutes during extraction (Table 4).

For the DPPH response, the mean value and standard deviation of the DPPH response were calculated as 35.89±17.82 mg Trolox/ g) (Table 4). It was stated that the functionality of the product (tea) is getting higher with time and temperature increasing. (Golukcu et al., 2013; Golukcu et al., 2014) found the total phenolic compounds of tea as 195.81 mg/kg for 70°C and 211.64 mg/kg for 100°C. According to the study, extracted phenolic content was increased for increasing time at every temperature stages. Tekeli et al., (1976) found IC_{50} value as 0.07 mg/mL and 0.123 mg/mL at Sideritis species (mountain tea) as *Sideritis phrygia* and *Sideritis bilgerana*, respectively. In another mountain tea study, different solvents (methanol, ethyl acetate, acetone) were used and calculated the IC_{50} range as 50.9 66.2 µg/ml, respectively (Erkan et al., 2011). In another study, temperature and DPPH value relation was investigated, and the DPPH values at different extraction temperatures (70°C-80°C-90°C-100°C) evaluated as 0.56 mg/mg, 0.48 mg/mg, 0.49 mg/mg, 0.44 mg/mg, respectively Golukcu et al. (2013). The time and temperature parameters have direct effect on the extraction and similar results between TPC and DPPH (Hanay, 2011). Contrastly, the both parameters had no direct relation between each other Golukcu et al. (2013). However, DPPH value in tea can be change with tea differentiation, extraction parameters, tea harvesting time and type, etc. Golukcu et al. (2013).

The mean value of the DPPH was found at 35.89±17.82 mg Trolox/ g (Table 6). The coefficient of variation (CV), R^2 and Adj-R^2 values were not suitable for the model in addition to that lack of fit and model p-value were not suitable for the prediction model (Table 6).

3.8. Optimization

When looking at the interaction of temperature, time, and sample ratio of the model in question, not all responses of the model are statistically important (0.05<p). The extract from the tea waste can be used for supporting teabag or ice tea production. For this reason, it should be not only the color but also functionality supported, and some extraction parameters

Preferable to be high (a*, EY, TPC, DPPH) and other ones (L*, b*, C, h) are better with lower values.

As seen in Table 6, model-p, lack of fit, R^2, Adj-R^2, CV(%), and PRESS factors were used for model predictions. The regression coefficient (R^2) shows a deviation from the mean, and it should be closer to 1 value. The R^2 ve Adj-R^2 are significant for a model and used for prediction (Myers and Montgomery, 1995), and the higher values of parameters show closer to mean values. Adeq. Precision value is a parameter for the capability of model prediction, and it should be greater than 4 (Myers and Montgomery, 1995). Another factor for a good model is a variation of coefficient (CV), and it is expected higher value.

At tea waste extraction, the highest desirability values of the three optimization points were selected as 0.839, 0.824, and 0.807. The highest desirability one (0.839) has an 8% tea waste /water ratio, 94.95°C temperature, and 60.00 minutes extraction parameters. The second highest desirability was 0.824 at 6.10% tea waste /water ratio, 95.00°C temperature and 60.00 minutes, and other optimum points were 0.807 at 0.50% tea waste /water ratio, 59.82°C temperature and 1.00 minute (Table 3).

4. Conclusions and Recommendations

However, during the present study, eight parameters (*L*, a*,b*,* EY, C, h, TSS, TPC, and DPPH) have been investigated after the extraction of tea waste by applying the response surface methodology (RSM) approach. The obtained data indicated that the *L** and TPC responses were suitable for model prediction based on tea waste/water ratio, temperature, and time parameters of the extraction process. Meanwhile, all the ranges of the measured eight parameters were very inclusive, and it differed from the other similar researches. The *L** and TPC responses were suitable for the prediction of models, and the responses were very important for tea waste extraction.

Whereas, the extraction studies of the tea waste are limited; some published reports have discussed different procedures for tea extraction or infusion. In a study where different extraction methods were carried out for the extraction of caffeine and catechins from tea samples, the most efficient approach was determined to be hot water extraction (40 minutes at 80 °C). In addition to applying temperature and time parameters, the sample amount/water ratio was 10 grams/300 mL (Demir, 2015). In another study where the two-step hot water extraction method was also used using the same sample amount, the parameters of 10 minutes at 50°C for the first step and 10 minutes at 80°C for the second step were selected (Bazinet et al. 2007). In the same study, high-temperature pretreatment hot water extraction was also tried, and single temperature and duration parameters were applied with 10 g tea sample/300 mL boiling water for 3 minutes at 100°C and, 40 minutes at 80°C (Demir, 2015). In the study for investigating the effect of brewing on the antioxidant capacity of 21 green tea samples; Samples were processed for 5 minutes at 80°C in 200 mL of water (Sharpe et al., 2016). For the extraction of caffeine and catechins in white tea research, the components obtained by selecting five temperature parameters in the range of 60-98 °C and five duration in the range of 3-15 minutes were correlated with sensory analysis (Perez-Burillo, 2018). In another research conducted by Salman et al. (2019), the extraction of tea samples was carried out in 3 different times (15, 20, and 25 minutes) by the traditional brewing method, where they used 2.83 grams sample / 250 mL pure water ratio

In another extraction process using methanol and ethyl acetate, 4 grams of tea sample was treated with 50 mL of solution at 4 °C (Kelesoglu, 2012). As well as, 5 grams tea waste material was extracted with 100 mL distilled water at 80°C for 40 min. by Khan et al. (2018). In another study conducted by Langey-Evans (2000), they aimed to measure the antioxidant capacity of green tea samples with the FRAP method. For this purpose, the temperature parameters were selected in the range of 20-90°C, and the time parameters in the range of 15 seconds-15 minutes. In another study on the antioxidant capacity of green tea and brewing and water extraction, the temperature parameters were selected as the lowest 60°C, the highest 95°C, and the time as 5-300 minutes (Jin et al., 2019). In another extraction process, 1.6 grams of green tea sample was applied with 110 mL of water at three different temperatures in the range of 75-95°C and eight different times in the range of 1-45 minutes (Saklar et al., 2015).

In addition to single extraction, it is stated that when the second and third extraction processes were applied for 3 minutes and temperatures at 80°C, the antioxidant capacity of tea increases by 25% (Komes et al., 2010). In another study, 2.25 grams of tea samples were extracted in 180 mL of water for 2 minutes, and extraction yields were investigated (Petersona et al., 2004). Another study applied ethanol using the Box-Behnken experimental design for classical extraction performed classical extraction; the time was studied in three different times between 15-45 minutes, three different temperatures between 20-60°C and three different ethanol concentrations between 40-80% and the effects on the extraction of antioxidant compounds were investigated (Bostanci, 2016). In a study on the extraction of catechin gallate and epicatechin gallate components from tea, process temperatures in addition to tea types and particle sizes were studied in the range of 50-100°C (Yaday et al., 2018). In addition to green tea and black tea, there are studies in which extraction processes are applied in other tea varieties. Insoluble mountain tea production, absorbance measurements were made up to 350 minutes, with extraction conditions 1% sample/water amount, 60-80° C, five temperatures, and 0.5-20 minutes intervals, as well as nine different times. As a result, the temperature of 70°C was determined as the critical temperature. Besides, the extraction balance was achieved at 150 minutes at 60-65°C, 100 minutes at 70°C and 50 minutes at 75-80 °C (Dincer et al., 2008).

Upon what have been stated above, the extraction process of tea and tea waste can be carried out in different ways following many procedures, and according to our followed one the obtained results suggest high importance of this extract and the possibility of utilizing it for ice tea and tea-bag production. The advantage of these models is their lower process cost and effects on supporting tea color and functionality, but still more future researches are recommended.

Table 1. Model Design Levels

Bağımsız Değişkenler	-a	-1	0	+1	+a
X_1 Sample/water ratio (%)	0	2.5	5	8	10
X_2 Extraction temperature (°C)	50	60	72.5	85	95
X_3 Extraction time (minutes)	1	12.5	30	50	60

Table 2. Trial Plan for Three Factors Central Composite Rotatable Design

Trial Number	Sample/water ratio (%)	Extraction temperature (°C)	Extraction time (minutes)
1	2.5	85	50
2	5	72.5	30
3	5	72.5	30
4	0.5	72.5	30
5	5	72.5	1
6	8	60	12.5
7	8	85	50
8	5	72.5	60
9	8	85	12.5
10	5	95	30
11	10	72.5	30
12	5	72.5	30
13	5	50	30
14	5	72.5	30
15	5	72.5	30
16	5	72.5	30
17	2.5	60	50
18	2.5	85	12.5
19	2.5	60	12.5
20	8	60	50

Table 3. The optimum points of the responses

No	Tea waste /water ratio (%)	Temp (°C)	Time (min.)	EY	*L**	*a**	*b**	C	h	TSS	TPC	DPPH	Desirability
1	8.00	94.95	60.00	2.71	23.12	0.47	4.96	5.24	94.14	1.58	105.54	78.85	0.839
2	6.10	95.00	60.00	2.47	23.91	0.49	5.33	5.59	95.92	2.11	115.36	73.69	0.824
3	0.50	59.82	1.00	2.46	22.58	0.16	4.76	5.81	93.89	1.09	139.12	68.66	0.807

Table 4. The results of the experiment for the responses

No	Sample/water ratio (%)	Temp (°C)	Time (Min.)	TSS	EY	pH	L*	a*	b*	C	h	TPC (mg gallic acid /100g)	DPPH (mgTrolox/ g)
1	2.5	85	50	0.95±0.21cde	4.84±0.15a	4.70±0.02de	23.99±0.02ab	-0.75±0.07c	6.04±0.03ef	6.74±0.19abcd	90.01±1.91g	32.58±1.27jk	57.48±0.33a
2	5	72.5	30	1.15±0.07c	4.24±0.03b	4.68±0.00def	24.85±0.44ab	-0.66±0.01c	6.73±0.03bcde	6.27±0.15abcd	90.00±1.50g	45.48±1.88fg	53.34±1.07ab
3	5	72.5	30	1.15±0.07c	2.16±0.05de	4.66±0.01ef	24.69±0.16ab	-0.63±0.07c	6.62±0.22bcde	7.04±0.01abcd	99.67±0.75cd	24.22±0.57lj	15.68±0.55i
4	0.5	72.5	30	0.30±0.00f	2.67±0.03d	5.46±0.00a	25.39±0.01a	-2.26±0.01g	6.02±0.12ef	5.56±0.13d	95.19±0.80def	90.99±2.04c	50.50±1.46bc
5	5	72.5	1	0.70±0.14def	4.02±0.01bc	4.72±0.01de	24.28±0.06ab	-1.18±0.09de	6.94±0.01bcd	7.02±0.03abcd	91.62±1.06fg	65.09±2.71e	23.38±0.21h
6	8	60	12.5	1.55±0.35b	2.28±0.02de	4.63±0.02f g	25.53±0.47a	-0.75±0.16c	7.71±0.44a	7.36±0.48abc	98.92±2.76cde	41.05±1.11ghi	21.99±1.37h
7	8	85	50	2.00±0.00a	1.14±0.01f	4.61±0.01fg	24.37±0.30ab	-0.11±0.01a	6.27±0.15cde	7.19±0.01abc	104.30±1.49b	35.64±1.74ij	20.31±1.01hi
8	5	72.5	60	1.10±0.00cd	3.63±0.24c	4.65±0.01ef	24.26±0.03ab	-0.55±0.03bc	6.35±0.06cde	7.75±0.46a	95.58±0.85def	34.93±2.04ij	32.50±0.49gh
9	8	85	12.5	1.90±0.00ab	0.39± 0.07g	4.66±0.01ef	25.06±0.15a	-0.20±0.12ab	7.02±0.03bc	6.51±0.18abcd	110.59±0.39a	50.91±3.00f	5.08±0.18k
10	5	95	30	0.85±0.07cde	3.65±0.02c	4.64±0.01ef	23.79±0.06ab	-0.50±0.06abc	5.54±0.14f	6.18±0.07abcd	94.72±1.07def	39.13±2.22ghij	57.04±1.55a
11	10	72.5	30	2.15±0.07a	3.64±0.10c	4.56±0.01g	24.62±0.23ab	-0.16±0.00ab	6.64±0.04bcde	6.40±0.02abcd	94.79±0.00def	83.06±0.71d	43.10±2.38de
12	5	72.5	30	1.05±0.07cd	3.99±0.15bc	4.75±0.03cd	24.33±0.00ab	-0.52±0.01abc	6.48±0.19bcde	7.59±0.89ab	89.51±1.73g	35.85±2.13ij	39.06±1.75ef
13	5	50	30	0.80±0.00cd	2.61±0.02d	4.89±0.06b	24.59±0.29ab	-1.45±0.33ef	7.12±0.01ab	6.33±0.21bcd	94.66±0.58ef	24.81±0.89l	41.14±2.11de
14	5	72.5	30	1.15±0.07cd	0.99±0.12f	4.67±0.01ef	24.35±0.09ab	-0.64±0.18c	6.32±0.16cde	6.71±0.34abcd	98.60±2.59cde	125.02±3.85b	52.36±3.03ab
15	5	72.5	30	1.10±0.14def	2.52±0.02de	4.72±0.03de	24.08±0.21ab	-0.51±0.04abc	6.31±0.21cde	7.65±1.63ab	95.34±0.33def	35.47±2.43ij	34.02±0.97fg
16	5	72.5	30	1.05±0.07cd	1.45±0.01f	4.68±0.01def	23.77±0.35ab	-0.51±0.12abc	6.19±0.09ef	6.08±0.02bcd	97.12±0.73cde	139.12±4.11a	45.82±2.92cd
17	2.5	60	50	0.70±0.14def	2.41±0.04de	4.81±0.01c	23.03±2.03b	-1.13±0.26de	7.26±0.53ab	6.51±0.18abcd	94.66±0.21ef	26.74±2.30kl	28.63±0.81h
18	2.5	85	12.5	0.55±0.21ef	2.01±0.24e	4.94±0.01b	24.66±0.16ab	-0.99±0.24cd	6.63±0.38bcde	7.27±0.07abc	101.52±2.63bc	24.85±0.76l	45.54±2.55cd
19	2.5	60	12.5	0.35±0.07f	2.49±0.07de	4.60±0.00fg	24.82±0.11ab	-1.77±0.17f	6.96±0.06bcd	6.77±0.04abc	95.56±0.16def	21.87±2.17l	10.55±0.24j
20	8	60	50	1.95±0.07a	2.52±0.05de	4.25±0.07h	25.19±0.96a	-0.11±0.01a	7.59±0.89cde	6.35±0.17abcd	95.76±1.45def	22.92± 1.09l	40.32±3.08de

a-c: There is no difference between interactions with the same letter at same column

Table 5. The ANOVA table for effects of quadratic and interactions on tea waste extraction parameters

Source	Df	EY (%)		TSS (Brix)		L		a		b		C		h		TPC (mg gallic acid/100g)		DPPH (mgTrolox/ g)	
		Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value	Sum of Square	p-value
Model	9	12.82	0.1345	2.74	0.2879	5.55	**0.0089***	3.32	0.3035	3.92	0.0791	1.79	0.8951	149.85	0.8420	17211.11	**0.0200***	1489.45	0.8293
X_1	1	0.91	0.2767	0.29	0.2691	0.43	0.0892	0.12	0.5137	2.48	0.9703	0.35	0.4069	8.56	0.6221	437.32	0.3587	4.13	0.9114
X_2	1	5.66	0.0165	7.41	0.9854	0.65	0.0421	0.81	0.1095	0.13	0.4013	0.32	0.4307	42.47	0.2837	3655.75	0.0194	147.21	0.5113
X_3	1	0.05	0.7889	0.13	0.4567	0.68	0.0389	0.02	0.7898	0.87	0.0470	3.96	0.9287	0.30	0.9259	2271.58	0.0531	103.18	0.5812
$X_1 X_2$	1	0.36	0.4842	0.08	0.5523	0.011	0.7739	7.20	0.1095	0.03	0.6472	0.12	0.6177	0.67	0.8889	1.25	0.9987	11.45	0.8532
$X_1 X_3$	1	0.87	0.2856	0.50	0.1552	1.21	0.0101	4.05	0.7898	1.13	0.0273	0.06	0.7259	0.078	0.9622	443.42	0.3555	3.42	0.9194
$X_2 X_3$	1	1.51	0.1678	0.02	0.7648	0.80	0.0277	0.12	0.8722	0.73	0.0656	0.52	0.3181	5.49	0.6922	7381.13	0.0027	310.38	0.3461
X_1^2	1	1.64	0.1532	0.043	0.6613	1.41	0.0066	0.17	0.9039	7.86	0.8340	9.25	0.9891	70.47	0.1752	143.40	0.5938	249.49	0.3961
X_2^2	1	1.47	0.1737	0.026	0.7328	0.12	0.3369	0.14	0.5155	1.02	0.0345	0.29	0.4499	15.88	0.5042	2967.50	0.0311	679.62	0.1741
X_3^2	1	0.31	05139	1.61	0.0202	0.084	0.4233	1.88	0.4436	6.63	0.9846	0.15	0.5868	5.21	0.6998	85.94	0.6788	122.27	0.5487
Residual	10	6.84		2.11		0.12		2.64		1.70		4.71		330.88		4725.06		3174.13	
Lack of fit	5	3.37	0.5131	0.31	0.9631	0.11	0.5437	0.93	0.7375	1.24	0.1501	3.51	0.1327	95.37	0.8281	3215.22	0.2132	1817.86	0.3779
Pure error	5	3.47		1.81		0.13		1.71		0.46		1.20		235.51		1509.85		1356.27	
Total	19	19.66		4.86		6.75		5.97		5.62		6.50		480.73		21936.17		4663.57	

X_1: Tea waste/water ratio; X_2: Temperature; X_3: Time

Table 6. Statistical compatibility for responses for tea waste extraction

Statistical Data	Responses								
	EY (%)	TSS (Brix)	L*	a*	b*	C	h	TPC (mg gallic acid/100g)	DPPH (mgTrolox/ g)
Mean	2.37	1.19	0.35	-0.77	6.64	6.76	96.42	21.74	35.89
Standart Deviation	0.83	0.46	1.42	0.51	0.41	0.69	5.75	49.99	17.82
CV (%)	34.95	38.65	24.48	66.83	6.21	10.15	5.97	43.49	49.64
PRESS	30.57	5.13	5.27	9.64	10.13	28.66	1063.49	27075.13	15940.33
R^2	0.6519	0.5646	0.8213	0.5572	0.6978	0.2753	0.3117	0.7846	0.3194
Adj-R^2	0.3387	0.1728	0.6605	0.1587	0.4259	-0.3770	-0.3078	0.5907	-0.2932
Pred- R^2	-0.5546	-0.0553	0.2191	-0.6166	-0.8026	-3.4077	-1.2122	-0.2343	-2.4181
Adeq. Precision	5.698	4.634	9.763	4.870	6.383	2.299	2.344	6.599	1.980
Model p-value	0.1345	0.2879	0.0089*	0.3035	0.0791	0.8951	0.8420	0.0200*	0.8293
Lack of fit	0.5131	0.9631	0.5437	0.7375	0.1501	0.1327	0.8281	0.2132	0.3779

Figure 2. Response surface plots showing the effects of extraction temperature and time (X_1), extraction time and the ratio of raw material to water (X_2), extraction temperature and the ratio of water/tea waste material (X_3) on the L response for the prediction models

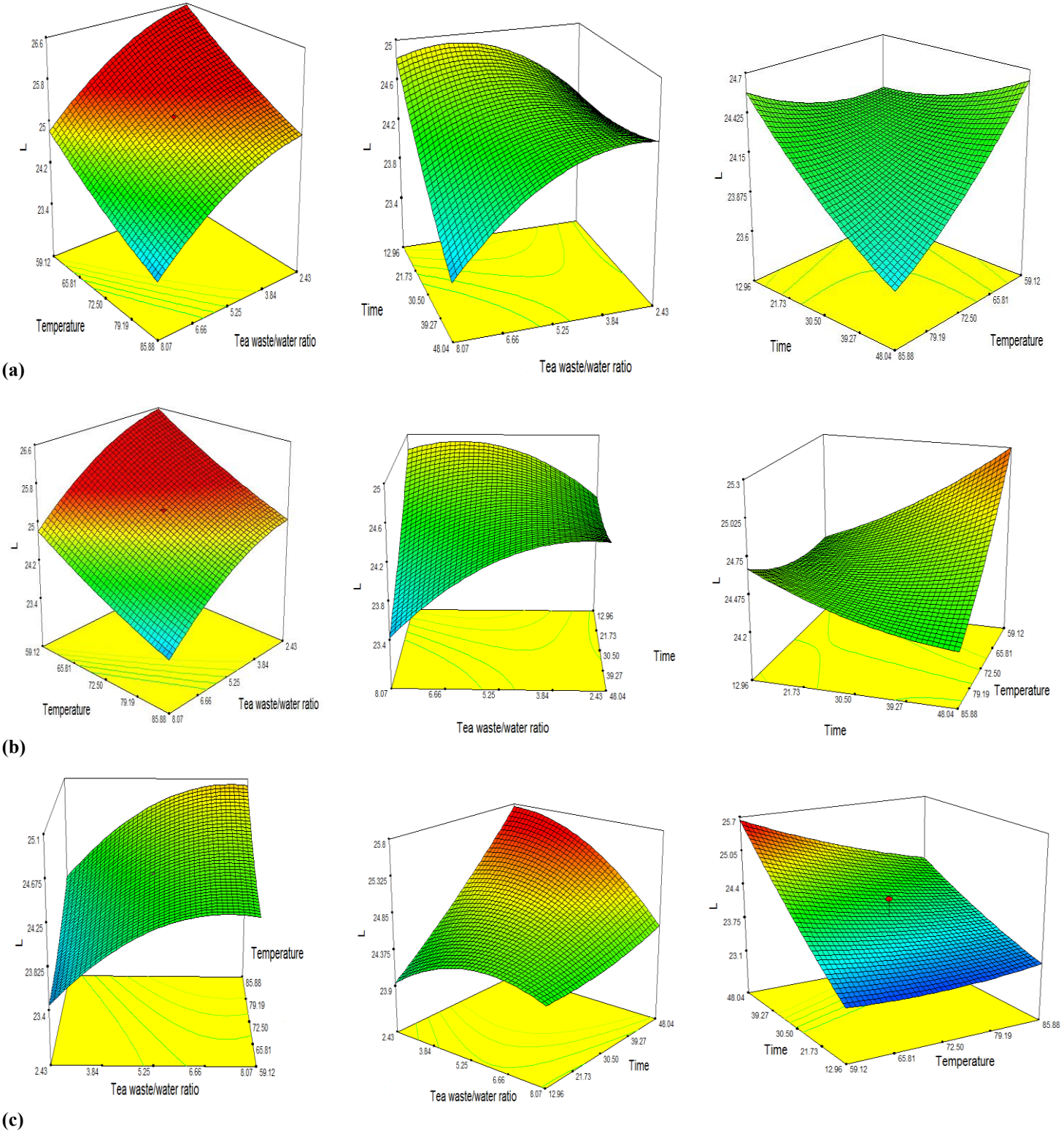
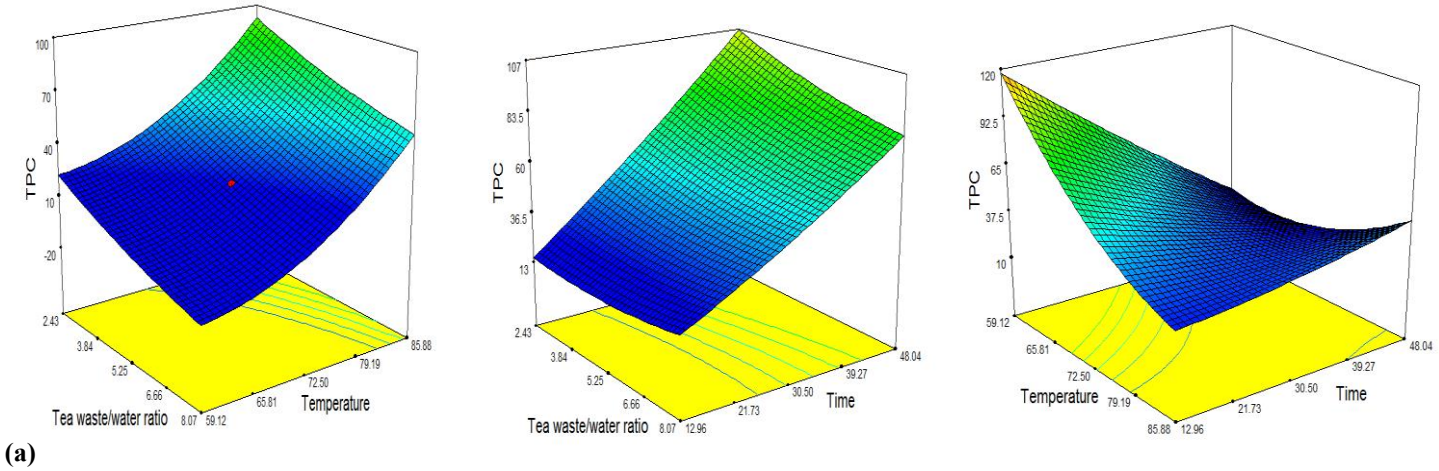
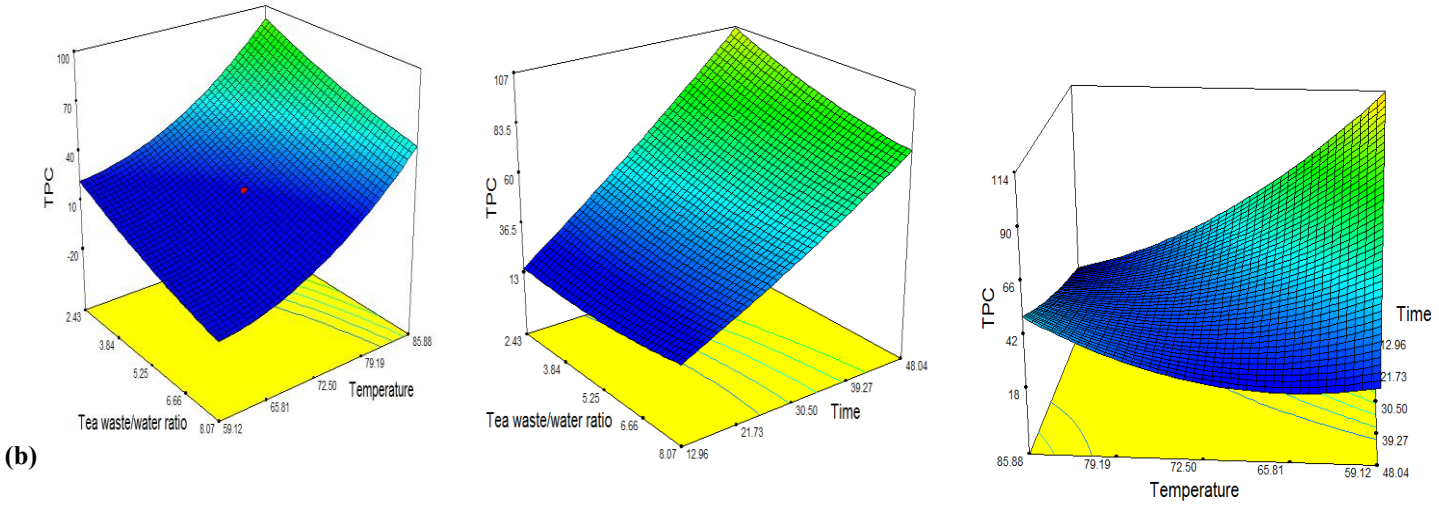


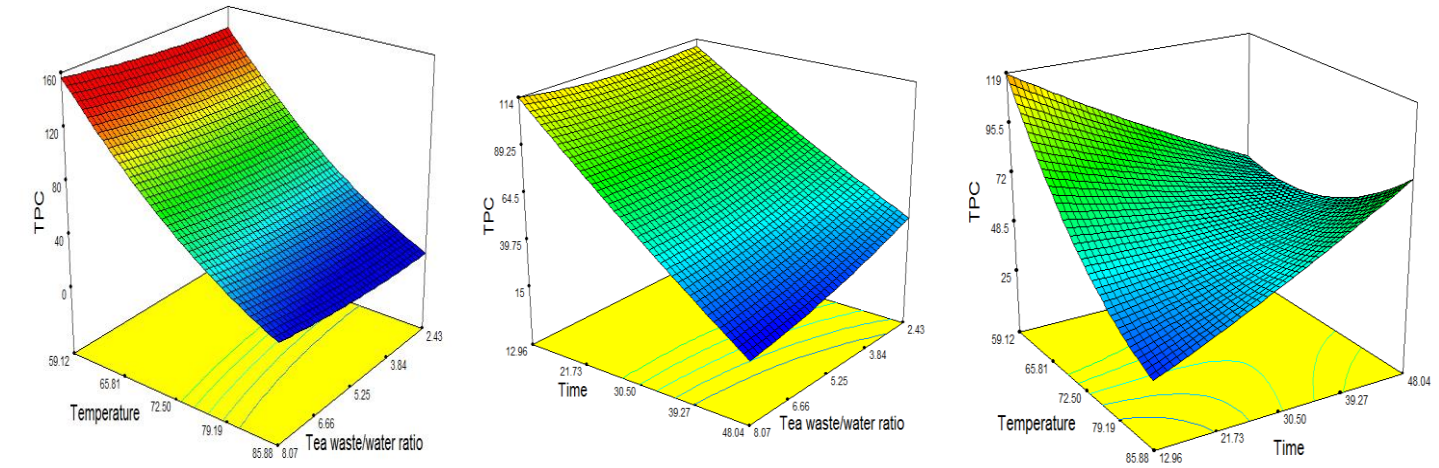
Figure 3. Response surface plots showing the effects of extraction temperature and time (X_1), extraction time and the ratio of raw material to water (X_2), extraction temperature and the ratio of water/tea waste material (X_3), on the TPC response for the prediction models



(a)



(b)



(c)

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