



Effect of water quality parameters on solar water disinfection: a statistical experiment design approach

M. Mansoor Ahammed*, Shilpa Dave, Abhilash T. Nair

Civil Engineering Department, S.V. National Institute of Technology, Surat 395007, India, Tel. +91 98258 75162; email: mansoorahammed@gmail.com

Received 14 April 2014; Accepted 14 June 2014

ABSTRACT

With a view to evaluating the effects and interactions of three influential factors, turbidity, pH and dissolved oxygen (DO), on the efficiency of solar water disinfection process, response surface methodology (RSM) based on Box–Behnken design was employed. Total coliform (TC) removal and heterotrophic plate count (HPC) removal were used as response factors. The RSM models were developed based on the experimental results of four-hour solar exposure in polyethylene terephthalate bottles. The measured and predicted removals were close to each other. Though the HPC removal and TC removal showed similar trends, the HPC removal was much lower (40–66%) compared to TC removal (68–97%) for the conditions studied. The results showed that turbidity up to certain levels (10–20 NTU) did not adversely impact the bacterial removal. In the range studied (6.0–9.5), pH showed little influence on the removals. While bacterial inactivation showed significant reduction at low DO levels, increasing DO beyond a certain level (5–6 mg/L) did not show any beneficial effect on bacterial inactivation. The study thus demonstrated the usefulness of RSM in modelling and analysing solar disinfection process.

Keywords: Heterotrophic bacteria; Household water treatment; Response surface methodology (RSM); Total coliforms

1. Introduction

The World Health Organization (WHO) has estimated that about 900 million people worldwide lack access to a safe and reliable source of drinking water [1]. Even people who have access to “improved” water supplies, such as household connections, public standpipes and boreholes, may not have microbiologically safe water as these sources are often contaminated with pathogens that cause infectious diseases such as cholera, enteric fever, dysentery and hepatitis,

which contribute significantly to the global burden of disease and death [2]. Further, the lack of on-site disinfection technologies, mainly in rural areas, limit widespread treatment of water supply. Despite major efforts to deliver safe drinking water to the world’s population, the reality is that water supplies delivering safe water will not be available to all people in the near future. The United Nations, as part of its Millennium Development Goals, has set a target of halving the proportion of people without safe drinking water by 2015.

There is considerable evidence that simple, low-cost domestic methods are capable of improving the

*Corresponding author.

microbial quality of household-stored water and reducing the diarrhoeal diseases in the populations. This has initiated a great interest in household treatment technologies. They refer to a range of treatment methods which treat water at the point of consumption rather than at source. A number of such methods based on filtration, flocculation, chlorination and UV radiation have been found to be effective in improving microbiological quality of water, and are approved by the WHO [2,3]. One of the most promising household treatment methods is the solar disinfection (SODIS). Solar radiation has been known as an effective antimicrobial agent for long and has been used for treatment of contaminated water for many years. Water disinfection using the SODIS process relies on the synergistic effect of mild thermal heating and solar UV radiation [4].

Batch-process SODIS involves storing microbiologically contaminated drinking water in transparent containers such as plastic bags or plastic or glass bottles. These are placed in direct sunlight for periods up to 8 h before consumption [5]. Solar water disinfection can be used to address the problem of microbiological contamination of drinking water in regions that receive intense solar radiation, and a large proportion of the world's population that lacks access to drinking water lives in areas which receive consistently intense solar radiation [6]. Solar water disinfection has been demonstrated to be effective against a range of bacteria and other micro-organisms like protozoa, fungi and viruses using laboratory isolates as well as naturally contaminated water [4,5,7–11]. Health impact assessment studies of SODIS have reported significant reduction in incidence of diarrhoea among SODIS users [12]. SODIS is currently in daily use by more than 4.5 million people in more than 50 countries across the developing world [13].

The antimicrobial effectiveness of sunlight is influenced by a number of factors such as source water characteristics, intensity of solar radiation, duration of exposure and type of container. Water quality parameters such as turbidity, pH, dissolved oxygen (DO) and concentration of different ionic species have been reported to be influencing the efficiency of SODIS [14–18]. Several studies have been reported on the influence of these variables. Most of these studies were conducted using one-factor-at-a-time approach which estimates the influence of a single variable while keeping all other variables at a fixed condition. The major disadvantage of this technique is that it cannot estimate interactive effects among the variables and thus cannot depict the complete effects of the parameters on the process [19,20]. It also requires large number of tests to be conducted. On the other

hand, statistically designed experiments are economical, and valid conclusions can be drawn with a small number of experiments. Response surface methodology (RSM) is one such efficient technique for modelling and analysing effects of multiple variables and their responses. It is used for designing experiments, building models, evaluating the effects of several variables and obtaining the optimum conditions for responses with a limited number of planned experiments [21]. The different types of RSM designs include central composite design, three-level factorial design, Box–Behnken design and D-optimal design [21]. Box–Behnken design, a modified central composite experimental design, is an efficient, rotatable and economical design. A comparison between Box–Behnken design and other RSM designs has shown that it is more efficient than other designs and requires fewer experiments [22]. Also, Box–Behnken design is more suitable for evaluating quadratic response surfaces in cases when predicting the response at the extreme levels is not the aim of the model [23].

Recently, RSM has been used for studying and optimising different processes used in water and wastewater treatment such as coagulation–flocculation [24–26], advanced oxidation processes [27], chlorination [28] and anaerobic sulphate removal [23]. To date, no study has reported modelling solar water disinfection process using RSM. In the present study, RSM with Box–Behnken design was used to investigate the effect of three important water quality parameters on bacterial inactivation as well as to determine the interactions of these parameters during batch-process SODIS in polyethylene terephthalate (PET) bottles. The three variables studied were turbidity, pH and DO, and the response factors were total coliform (TC) removal and heterotrophic plate count (HPC) removal.

2. Materials and methods

2.1. Materials

All SODIS tests were conducted in commercially available one-litre PET bottles of approximate diameter 8.3 cm. Bottles were washed first with alcohol and then several times with sterilised water prior to the tests. Groundwater from a dugwell near S.V. National Institute of Technology, Surat, India, was used as test water in all tests. A single batch of water was used in all the tests to ensure uniformity and the characteristics of the well water are presented in Table 1. Since the concentration of indicator organisms in the water was low, wastewater obtained from the Surat Municipal Corporation wastewater treatment plant was spiked to the well water, after cloth filtration, at the

Table 1
Characteristics of the well water used for the experiment

Parameter	Value
Turbidity (NTU)	1.2
pH	7.2
Conductivity ($\mu\text{S}/\text{cm}$)	1,380
Total hardness (mg/L as CaCO_3)	120
Alkalinity (mg/L as CaCO_3)	96
Chlorides (mg/L)	82
Sulphates (mg/L)	64
DO (mg/L)	1.6
Temperature ($^\circ\text{C}$)	24
Total dissolved solids (mg/L)	760
Total coliforms (MPN/100 mL)	220 (1.1×10^4)*
Faecal coliforms (MPN/100 mL)	11 (1.7×10^3)*
HPC (CFU/mL)	4.7×10^3 (1.73×10^4)*

*Values in parentheses indicate concentration after spiking with wastewater.

rate of 1 mL per 10 L to obtain a TC concentration of about 10^4 MPN/100 mL in the test water. Required turbidity was obtained by adding autoclave-sterilised kaolin. Preliminary test was conducted to determine the amount of kaolin needed for the test water to bring the turbidity to the required levels. DO in the water sample was increased to the required levels by aerating it with household aquarium pumps. Water pH was adjusted to the desired values by adding 0.1 N NaOH or H_2SO_4 .

2.2. Design of experiments, analysis and model fitting

A Box–Behnken statistical experimental design was used to investigate the effects of the three independent variables namely turbidity, pH and DO. The responses (dependent variables) studied were TC removal and HPC removal. The total number of runs required in Box–Behnken design is defined as $N = 2k(k - 1) + C_o$ where k is the number of variables studied and C_o is the number of central points [29,30]. In the present study, 15 runs were conducted, with three replicates at the centre of the design for estimation of pure error sum of squares [21,27]. Experimental data were fitted to a second-order polynomial model:

$$y = b_0 + \sum_{i=1}^k b_i x_i + \sum_{i=1}^k b_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{j=i+1}^k b_{ij} x_i x_j \quad (1)$$

where y is the predicted response used as dependent variable, x_i and x_j are the independent variables, b_0 is the constant coefficient, b_i is the coefficient that

determines the influence of variable i in the response, b_{ij} is the coefficient that determines the effect of interaction between variables i and j , b_{ii} is the parameter that determines the shape of the curve (quadratic effect) and k is the number of variables studied [27,31].

For each variable, selection of the minimum and maximum values is important for successful application of the model. The minimum and maximum values for the variables were selected as explained later in this section. Normalisation of variables is necessary before performing a regression analysis so that the responses are affected more evenly and the units of the variables become irrelevant [19]. The coded values for the independent variables were determined by the following equation [32,33]

$$x_i = \frac{z_i - z_0}{\Delta z_i} \quad (2)$$

where x_i is the dimensionless coded value of independent variable, z_i is the uncoded value of the i th independent variable, z_0 is the uncoded i th independent variable at the centre point and Δz_i is the step change value between the low level (−1) and high level (+1).

For turbidity, the low level (−1) was fixed as the initial turbidity of the well water (1.2 NTU) while high level (+1) was fixed as 54 NTU, as it is well known that at high turbidity, the solar inactivation is greatly reduced. For pH, the low and high levels were set as 6.0 and 9.5 for typical drinking water sources. The lower and higher values for DO were 1.6 and 7.0 mg/L, respectively, based on the initial DO value of well water and a typical high value for natural waters. The coded and actual values of the chosen independent variables used in the experiments are given Table 2.

The regression analysis of the experimental data was prepared using the software Design Expert 8.0 (Stat-Ease Corporation, Minneapolis, USA). This software was also used for obtaining the three-dimensional surface plots and contour plots of the response models.

2.3. Experimental procedure

The PET bottles were exposed to the sunlight at the rooftop of the Civil Engineering Department of SV National Institute of Technology, Surat, India, during February 2012 when sunlight intensity was moderate. The bottles were placed in duplicate and were exposed to natural sunlight from 9:30 am onwards for four hours. Since the main objective of the study was to investigate the influence of water quality

Table 2

Design matrix in coded and uncoded units, along with the observed and predicted responses

Uncoded and coded levels of variables			TC removal (%) (y_1)		HPC removal (%) (y_2)	
Turbidity (NTU) (x_1)	pH (x_2)	DO (mg/L) (x_3)	Observed	Predicted	Observed	Predicted
27.6 (0)	6.00 (-1)	1.6 (-1)	88	88.2	60	58.0
27.6 (0)	9.50 (+1)	1.6 (-1)	86	85.7	58	59.0
27.6 (0)	6.00 (-1)	7.0 (+1)	97	93.2	65	64.0
27.6 (0)	9.50 (+1)	7.0 (+1)	90	90.7	66	65.0
1.2 (-1)	6.00 (-1)	4.3 (0)	90	93.7	65	63.8
54.0 (+1)	6.00 (-1)	4.3 (0)	83	79.7	45	46.3
1.2 (-1)	9.50 (+1)	4.3 (0)	89	91.2	65	64.8
54.0 (+1)	9.50 (+1)	4.3 (0)	83	77.2	50	47.3
1.2 (-1)	7.75 (0)	1.6 (-1)	90	86.0	60	60.5
54.0 (+1)	7.75 (0)	1.6 (-1)	68	72.0	40	40.5
1.2 (-1)	7.75 (0)	7.0 (+1)	93	91.0	63	63.9
54.0 (+1)	7.75 (0)	7.0 (+1)	72	77.0	48	48.9
27.6 (0)	7.75 (0)	4.3 (0)	91	93.4	62	63.6
27.6 (0)	7.75 (0)	4.3 (0)	92	93.4	62	63.6
27.6 (0)	7.75 (0)	4.3 (0)	94	93.4	64	63.6

parameters on SODIS, it was necessary to ensure that complete inactivation of indicator organisms did not occur during the exposure period. Preliminary tests showed that during the exposure period of four hours complete inactivation of TC or HPC did not occur in any of the samples, and consequently four hours was chosen as exposure time for the experimental design study. The mean solar intensity during this 4-h exposure period was 512 W/m². Care was taken to ensure uniform exposure for all the bottles throughout the experimental duration. Transparent PET bottles containing water samples placed indoors served as control. Control bottles indicated a maximum reduction of about 4% for both the indicators tested. Samples were collected after the exposure period for microbiological analysis. DO in the bottles was also measured after the exposure period. Solar radiation was measured using a pyranometer (National Instruments Limited, Kolkata, India) connected to a data logger. The spectral range of the pyranometer used was 300–3,000 nm. Water and ambient temperatures were recorded using a digital thermometer.

2.4. Analyses

The concentrations of TCs and faecal coliforms were estimated by multiple-tube fermentation method (most probable number method). The result is expressed as most probable number per 100 mL (MPN/100 mL). Lauryl Tryptose Broth was used for the presumptive phase of total and faecal coliforms, and brilliant green

bile broth and EC medium were used for confirmation phase of total and faecal coliforms, respectively. The samples were collected and a dilution series was performed. The dilutions needed for plating and MPN test were estimated based on expected removal. Heterotrophic bacteria were enumerated using the pour plate method. For this, 0.5 mL of water sample (or diluted water sample) was poured into a petridish. Five to six millilitres of plate count agar was poured over this and mixed well. The petridishes were incubated at 37°C for 48 h for development of colonies. The result is expressed as colony-forming units per mL (CFU/mL). All the tests were conducted in accordance with the techniques described by *Standard Methods* [34]. Turbidity was measured using a turbidimeter (Hach 2100P). pH was determined using a pH meter (Hanna pH 209) and DO was monitored using a DO meter (ESICO, India).

3. Results and discussion

3.1. RSM models and their validation

Experimental results of TC removal and HPC removal in different PET bottles after four-hour exposure to solar radiation are presented in Table 2. The observed removals were used to compute the models using second-order polynomial as represented by Eq. (1). The models for TC and HPC removals in terms of coded factors were determined as:

$$\text{TC removal (\%)} = 93.38 - 7.00x_1 - 1.25x_2 + 2.5x_3 - 7.92x_1^2 - 3.92x_3^2 \quad (3)$$

$$\text{HPC removal (\%)} = 63.62 - 8.75x_1 + 0.50x_2 + 3.00x_3 + 1.25x_1x_3 - 8.08x_1^2 - 2.08x_3^2 \quad (4)$$

where x_1 is the turbidity, x_2 is the pH and x_3 is the DO.

In these models, statistically, non-significant square terms and interactive terms were removed according to the significant levels selected ($\text{prob} > F > 0.2$). The analysis of variance (ANOVA) results for TC and HPC removals are given in Tables 3 and 4, respectively, and were used for analysis of the model. It can be observed from the ANOVA tables that for TC removal, turbidity (x_1), DO (x_3) and square terms x_1^2 and x_3^2 were significant. For HPC removal, turbidity (x_1), DO (x_3), square terms x_1^2 and x_3^2 and the interactive term x_1 and x_3 were significant.

The adequacy and significance of a model are generally checked by model F values, probability values ($P > F$) and adequate precision [27,28,32]. A model is significant at 95% confidence interval if the F -test has a probability value ($P > F$) below 0.05. For the present models, these values were 0.0027 and < 0.0001 , respectively, for TC and HPC removals, showing the significance of the models. For lack of fit, a large value of $P > F$, possibly > 0.05 , is preferred, as it measures the model failure in representing the data points in the experimental domain [26,35]. In the present case, these values were 0.1066 and 0.2877, respectively, for TC and HPC removals, further implying that lack of fit of the model is insignificant. Adequate precision (AP) values are used to estimate the discrimination between

the range of predicted values at the design points and the average predictor error [27,35], and a model is considered adequate if the AP value is higher than four. For TC and HPC removals, AP values obtained were 8.47 and 20.38, respectively, which confirm that the models can be used for predicting bacterial inactivation.

The goodness of fit of the models was checked by coefficient of determination (R^2). A high R^2 value, close to 1, is desirable to ensure a satisfactory adjustment of the model to the experimental data. Also, a reasonable agreement with adjusted R^2 (R^2_{adj}) is necessary [25]. The values of R^2 (0.832 for TC removal and 0.974 for HPC removal) in this study indicate that only 16.8% (TC removal) and 2.6% (HPC removal) of the variability in the responses were not explained by the models. Further, the values of R^2_{adj} (0.74 for TC removal and 0.95 for HPC removal) were also high, indicating high significance of the models. If the model contains many terms and the sample size is not large, R^2_{adj} may be significantly lower than R^2 [27,36].

Fig. 1 shows the normal probability plots of the studentized residuals. The residuals are normally distributed if the points on the plot follow a straight line. As Fig. 1(a) and (b) illustrates, the assumption of normality is satisfied for both TC removal and HPC removal. For the model to be reliable, the response should be predicted with reasonable accuracy by the model equation. Fig. 2(a) and (b) present the relationship between observed and predicted values of TC and HPC removals. The statistical significance of the models was further evident from these figures as observed and predicted values were in good agreement with each other for both TC and HPC removals.

Table 3
ANOVA test for TC removal (%)

Source	Sum of squares	Degree of freedom	Mean square	F value	Prob > F
Model	729.7410	5	145.9482	8.9239	0.0027
Turbidity (x_1)	392.0000	1	392.0000	23.9686	0.0009
pH (x_2)	12.5000	1	12.5000	0.7643	0.4047
Oxygen (x_3)	50.0000	1	50.0000	3.0572	0.1143
x_1^2	233.1648	1	233.1648	14.2567	0.0044
x_3^2	57.1648	1	57.1648	3.4953	0.0944
Residual	147.1923	9	16.3547		
Lack of Fit	142.5256	7	20.3608	8.7261	0.1066
Pure Error	4.6667	2	2.3333		
Cor Total	876.9333	14			

Note: $R^2 = 0.83215$; $R^2_{\text{adj}} = 0.7389$; Adequate precision = 8.47363.

Table 4
ANOVA test for HPC removal (%)

Source	Sum of squares	Degree of freedom	Mean square	F value	Prob > F
Model	943.4577	6	157.2429	50.4341	< 0.0001
Turbidity (x_1)	612.5000	1	612.5000	196.4534	< 0.0001
pH (x_2)	2.0000	1	2.0000	0.6415	0.4463
Oxygen (x_3)	72.0000	1	72.0000	23.0933	0.0013
($x_1 x_3$)	6.2500	1	6.2500	2.0046	0.1946
x_1^2	242.3077	1	242.3077	77.7178	< 0.0001
x_3^2	16.0220	1	16.0220	5.1389	0.0531
Residual	24.9423	8	3.1178		
Lack of fit	22.2756	6	3.7126	2.7845	0.2877
Pure error	2.6667	2	1.3333		
Cor total	968.4000	14			

Note: $R^2 = 0.97424$; $R^2_{\text{adj}} = 0.95493$; Adequate precision = 20.3752.

3.2. Analysis of the results

In order to understand the effects of independent variables and their interactive effects, 3-D plots and their corresponding contour plots were generated based on the models developed. These plots are represented as a function two variables at a time, keeping the third variable at a fixed level (zero level). Figs. 3 and 4 show the response surfaces for TC removal and HPC removal, respectively. It is evident from Figs. 3 (a), (c), and 4(a) and (c) that there is little effect of pH on the removal in the range of pH studied (6.0–9.5). It can also be seen from Figs. 3(a), (b), 4(a) and (b) that the maximum TC and HPC removals occurred when turbidity of water was in the range 10–20 NTU, and not at the lowest turbidity values. Effect of DO on TC and HPC removals as illustrated in Figs. 3(b), (c), and 4(b) and (c) indicates that up to a certain DO (~5–6 mg/L), the removals increased with increase in DO. No beneficial effect of DO was evident beyond this level.

It may be noted that though the TC and HPC inactivation followed similar trends with respect to effect of the three variables studied, HPC removal was much lower compared with TC removal at all conditions, indicating that heterotrophic bacteria are more resistant to solar inactivation compared to TC. Higher resistance of heterotrophic bacteria to solar inactivation has been reported by previous researchers as well [37,38]. Amin and Han [38] reported the relative removal of micro-organisms as $\text{HPC} < \text{TCs} < \text{faecal coliforms/Escherichia coli}$ during SODIS. It should be mentioned that HPC is not generally monitored in the case of drinking water. However, in the present study, this parameter was included to compare its inactivation with coliforms. Generally, this indicator is used to establish the effectiveness of different treatment processes in a water treatment plant.

It is interesting to note that the highest TC and HPC removals did not occur at the lowest turbidity levels, but within a turbidity range of 10–20 NTU. Though turbidity reduces penetration of solar radiation, thus reducing the solar inactivation, several studies have indicated that moderate turbidity does not affect microbial inactivation rates during SODIS [15,37–39]. Amin and Han [38] showed that up to 20 NTU, there was no significant effect on solar inactivation of *E. coli* and TC. Samples of moderate turbidity might not have caused a significant reduction of solar penetration. In the present study, it was observed that the mean temperatures were 1.7 and 2.2°C higher in bottles with 27.6 and 54.0 NTU water samples, respectively, compared to bottles with 1.2-NTU turbidity (data not shown), which presumably enhanced the temperature-induced inactivation. This increase in temperature could be due to absorbance of radiation by turbid particles. In the case of high turbid water (such as the 54-NTU water in the present study), these advantages may not be enough to compensate for the reduction of solar penetration through it [15].

The present study found little effect of initial pH in the range investigated (pH 6.0–9.5). A few studies have reported the effect of pH on inactivation of micro-organisms during solar exposure. Alrousan et al. [18] who studied the effect of initial pH on the photocatalytic inactivation of *E. coli* in distilled water reported no marked effect of pH on inactivation within the pH range of 5.5–8.5. However, significant cell death was observed below 5.5 and above 8.5 pH units. Amin and Han [38] reported 10–20% increase in TC and *E. coli* inactivation when the initial pH was 5.0 compared with neutral pH values. However, their results differ from that of Rincoln and Pulgarin [40] who reported that initial pH values between 4 and 9

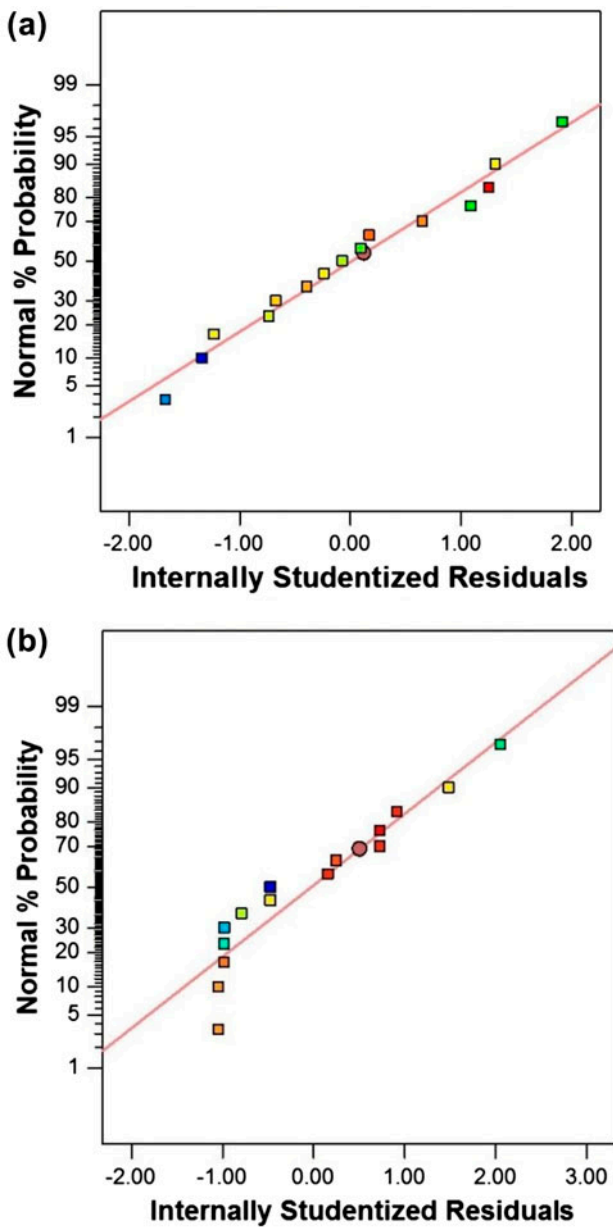


Fig. 1. Normal probability plot of internally studentised residuals for (a) TC removal and (b) HPC removal.

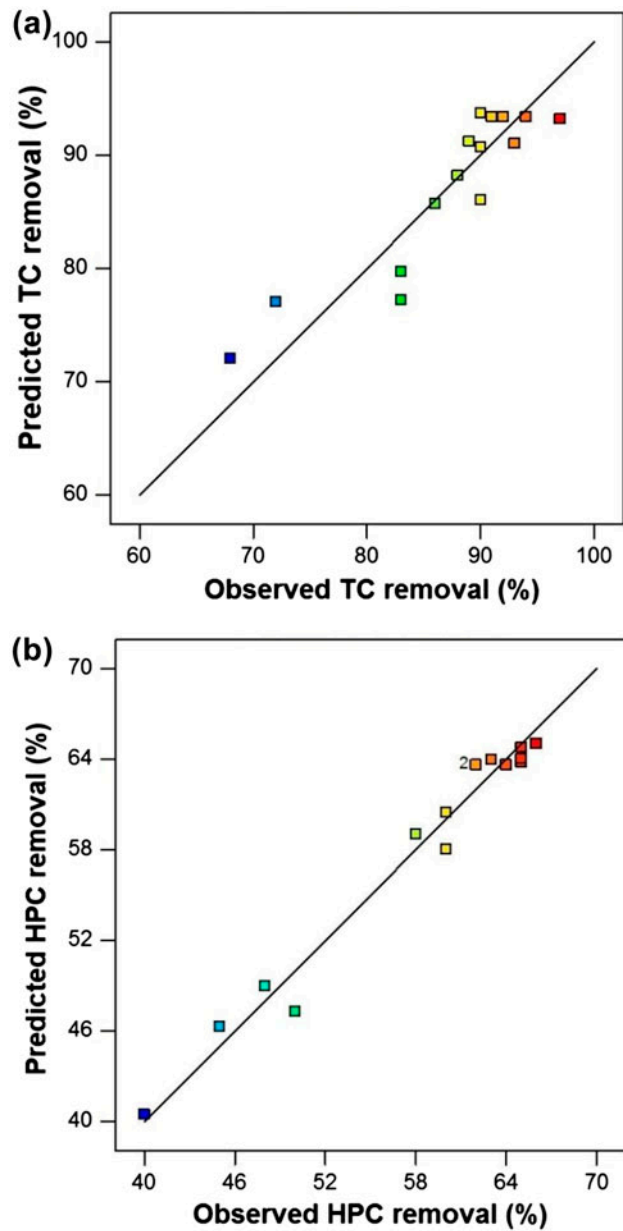


Fig. 2. Plot of predicted vs. observed values for (a) TC removal and (b) HPC removal.

did not affect *E. coli* inactivation rates. These differences could be due to variations in experimental conditions used in different studies, such as characteristics of water, like concentration of dissolved and suspended species, and solar exposure conditions. In the present study, a pH range of 6.0–9.5 was used considering the pH range of natural waters. At much higher and lower pH values, inactivation of micro-organisms increases due to additional stress to the cells [41,42].

Though it is known that DO is essential for the photoinactivation of micro-organisms due to generation of reactive oxygen species [43], and photoinactivation rates drop drastically in the absence of oxygen [14], systematic evaluation of effect of initial DO concentration during SODIS is missing in the literature. Contradictory observations on the effect of agitation during solar exposure which affects the DO levels have been reported. While Reed [14] reported that intermittent agitation of bottles during solar exposure

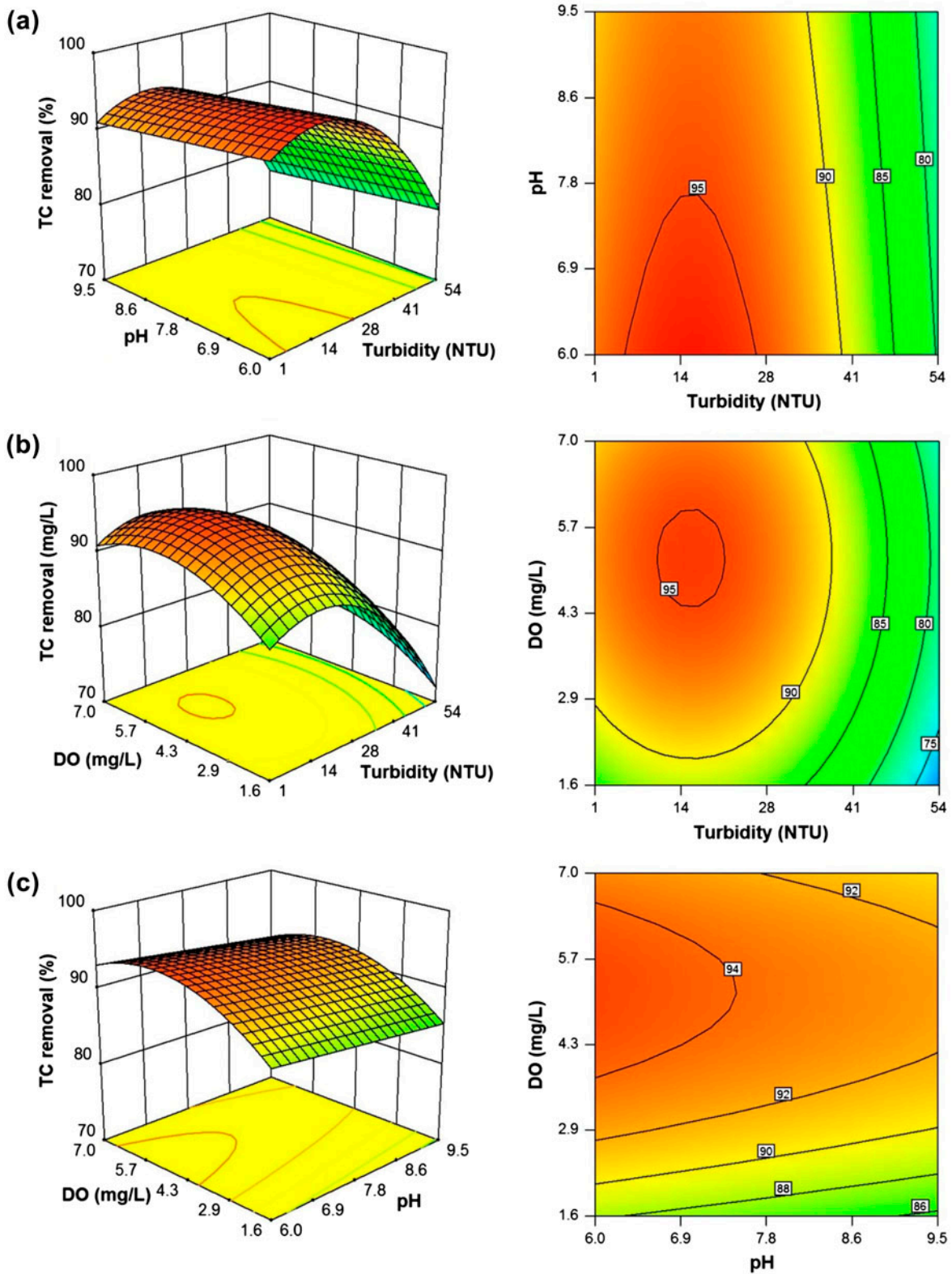


Fig. 3. Response surfaces and their corresponding contour plots for TC removal as a function of (a) turbidity and pH at DO 4.3 mg/L (b) turbidity and DO at pH 7.75 and (c) pH and DO at turbidity 27.6 NTU.

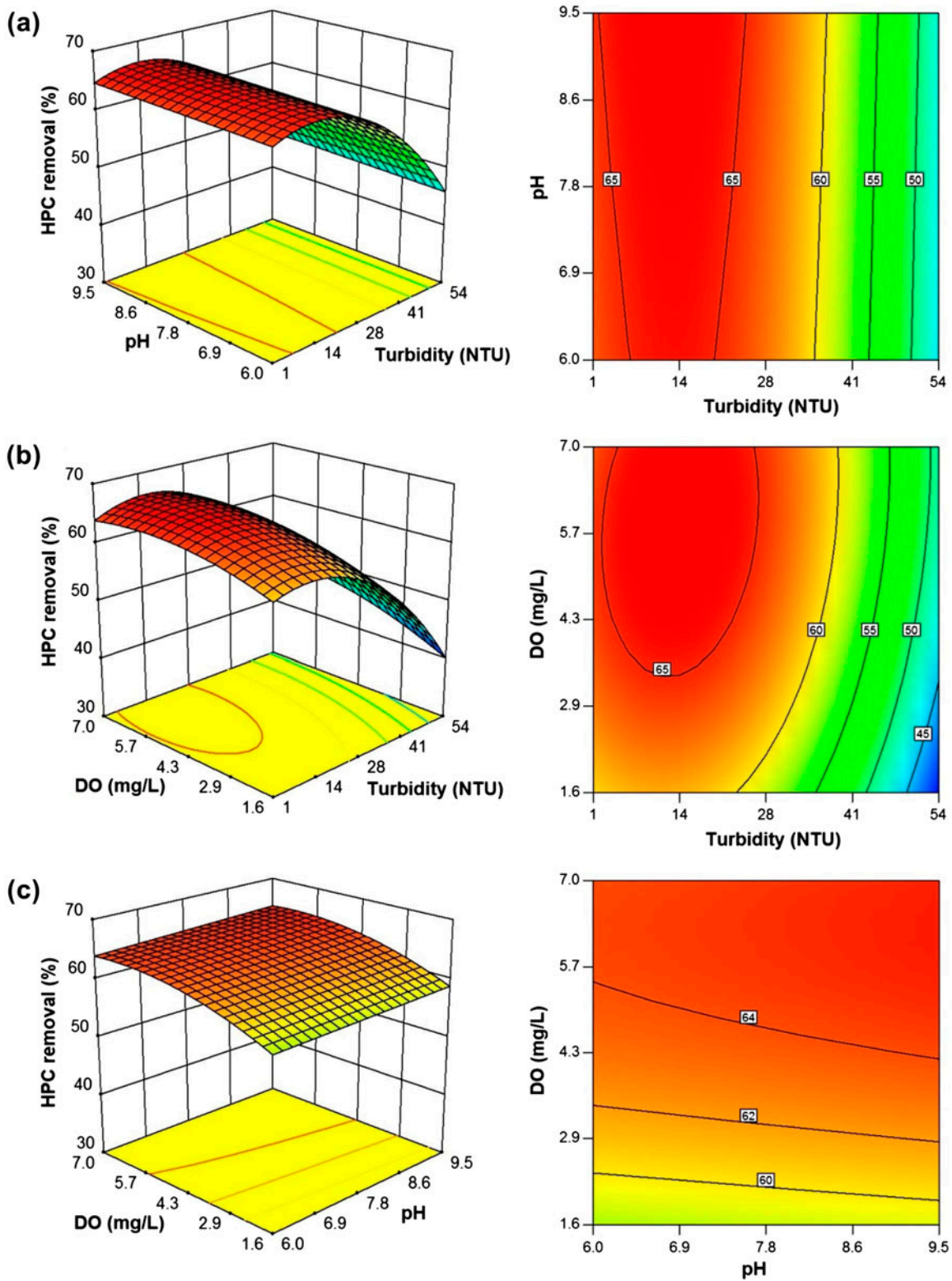


Fig. 4. Response surfaces and their corresponding contour plots for HPC removal as a function of (a) turbidity and pH at DO 4.3 mg/L (b) turbidity and DO at pH 7.75 and (c) pH and DO at turbidity 27.6 NTU.

was beneficial, Kehoe et al. [15] found no advantage of agitation of bottles during exposure. The beneficial effect of increased DO levels on the inactivation of micro-organisms is evident from the present study. However, increasing DO beyond a particular level (5–6 mg/L) did not result in higher removal of TC and HPC. This level, however, would depend on the characteristics of the water. It may be interesting to note that even at low DO levels used in the study (1.6 mg/L), bacterial inactivation occurred. This is presumably due to the relatively low initial concentration of bacteria used in this study. It was found that solar exposure for four hours reduced the DO in different bottles by 0.8–1.2 mg/L. It shows that some DO was present after the four-hour exposure period even in bottles with low initial DO value of 1.6 mg/L. These aspects need further investigation.

Using RSM models, overlay plots can be generated to obtain the optimum conditions for removal of both HPC and TC for arbitrarily selected values. Fig. 5 illustrates one such overlay plot. The shaded area was generated when the pH was set at 7.0 and it shows the region where TC removal would be greater than 95% and HPC removal greater than 65%. For example, 95% TC removal and 65% HPC removal would be achieved when turbidity and DO are 15 NTU and 5.5 mg/L, respectively. In order to confirm the results of the model studies, an additional confirmation test using three replicates was conducted under these conditions. Though this test was conducted on a different day, the average solar intensity for the four-hour

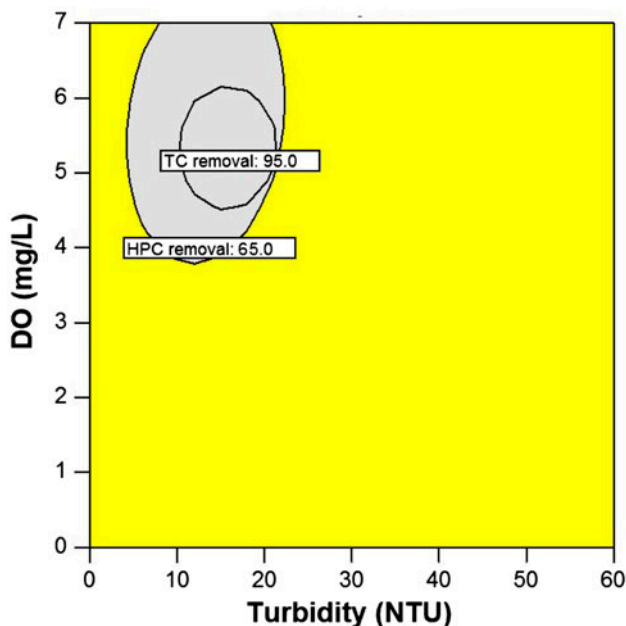


Fig. 5. Overlay plot for optimum conditions (pH 7.0).

exposure period (529 W/m^2) was very close to the value achieved in the experimental design study (512 W/m^2). The results showed TC and HPC removals of 97 and 63%, respectively, which were close to the values predicted by the regression models. This further demonstrates the usefulness of RSM in modeling solar water disinfection process.

It may be noted that only three variables influencing the solar water disinfection efficiency were included in this study. Other important parameters, such as solar intensity, exposure time and initial concentration of organisms, should also be included in future studies using RSM, which would clearly demonstrate the influence of these parameters and their interactions in microbial inactivation during SODIS. Since solar intensity cannot be varied as required if direct solar exposure is used, some of these tests will have to be conducted in the laboratory under controlled conditions.

4. Conclusions

In this study, RSM was used to investigate the effect of three water quality parameters, turbidity, pH and DO, on solar water disinfection process. Response models were developed for the removal of HPC and TC, and the predicted values were in good agreement with the observed values. The models indicated that up to certain level (10–20 NTU), turbidity of water did not adversely impact the bacterial removal. Initial water pH did not show much influence on the bacterial inactivation in the range of pH studied (6.0–9.5). DO impacted the removal with lower removal at low DO values. Increasing DO above certain levels (5–6 mg/L), however, did not result in increased bacterial removal. The RSM, thus, provided a useful approach for predicting the influence of different variables and their interactions on microbial inactivation during SODIS. More studies should be conducted by including more variables that influence the SODIS process.

Acknowledgment

The research was funded by the Department of Science and Technology, Government of India through a grant (Diary No.100/IFD/4130/2008–2009) under Water Technology Initiative.

References

- [1] WHO, UNICEF, Progress on Sanitation and Drinking Water, 2010 Update, World Health Organization and United Nations Children's Fund, Geneva, 2010.

- [2] M.D. Sobsey, C.E. Stauber, L.M. Casanova, J.M. Brown, M.A. Elliott, Point of use household drinking water filtration: A practical, effective solution for providing sustained access to safe drinking water in the developing world, *Environ. Sci. Technol.* 42 (2008) 4261–4267.
- [3] WHO, Combating Waterborne Disease at the Household Level, World Health Organisation, Geneva, 2007.
- [4] C. Navntoft, E. Ubomba-Jaswa, K.G. McGuigan, P. Fernández-Ibáñez, Effectiveness of solar disinfection using batch reactors with non-imaging aluminum reflectors under real conditions: Natural well-water and solar light, *J. Photochem. Photobiol., B* 93 (2008) 155–161.
- [5] M. Boyle, C. Sichel, P. Fernandez-Ibanez, G.B. Arias-Quiroz, M. Iriarte-Puna, A. Mercado, E. Ubomba-Jaswa, K.G. McGuigan, Bactericidal effect of solar disinfection under real sunlight condition, *Appl. Environ. Microbiol.* 74 (2008) 2997–3001.
- [6] E.G. Mbonimpa, B. Vadheim, E.R. Blatchley III, Continuous-flow solar UVB disinfection reactor for drinking water, *Water Res.* 46 (2012) 2344–2354.
- [7] M. Wegelin, S. Canonica, K. Mechsner, T. Fleischmann, F. Pesaro, A. Metzler, Solar water disinfection: Scope of the process and analysis of radiation experiments, *J. Water Supply Res. Technol.—Aqua* 43 (1994) 154–169.
- [8] B. Sommer, A. Marino, Y. Solarte, M.L. Salas, C. Dierolf, C. Valiente, D. Mora, R. Rechsteiner, P. Settler, W. Wirojanagud, H. Ajarmeh, A. Al-Assan, M. Wegelin, SODIS—An emerging water treatment process, *J. Water Supply Res. Technol.—Aqua* 46 (1997) 127–137.
- [9] L.F. Caslake, D.J. Connolly, V. Menon, C.M. Duncanson, R. Rojas, J. Tavakoli, Disinfection of contaminated water by using solar irradiation, *Appl. Environ. Microbiol.* 70 (2004) 1145–1151.
- [10] M. Berney, H.-U. Weilenmann, A. Simonetti, T. Egli, Efficacy of solar disinfection of *Escherichia coli*, *Shigella flexneri*, *Salmonella typhimurium* & *Vibrio cholerae*, *J. Appl. Microbiol.* 101 (2006) 828–836.
- [11] K.G. McGuigan, F. Mendez-Hermida, J.A. Castro-Hermida, E. Ares-Mazas, S.C. Kehoe, M. Boyle, Batch solar disinfection (SODIS) inactivates oocysts of *Cryptosporidium parvum* and cysts of *Giardia muris* in drinking water, *Appl. Microbiol.* 101 (2006) 453–463.
- [12] A. Rose, S. Roy, V. Abraham, G. Holmgren, K. George, V. Balraj, S. Abraham, J. Muliylil, A. Joseph, G. Kang, Solar disinfection of water for diarrhoeal prevention in Southern India, *Arch. Dis. Childhood* 91 (2006) 139–141.
- [13] K.G. McGuigan, R.M. Conroy, H. Mosler, M. Preez, E. Ubomba-Jaswa, P. Fernandez-Ibáñez, Solar water disinfection (SODIS): A review from bench-top to roof-top, *J. Hazard. Mater.* 235–236 (2012) 29–46.
- [14] R.H. Reed, Solar inactivation of faecal bacteria in water: The critical role of oxygen, *Lett. Appl. Microbiol.* 24 (1997) 276–280.
- [15] S.C. Kehoe, T.M. Joyce, P. Ibrahim, J.B. Gillespie, R.A. Shahr, K.G. McGuigan, Effect of agitation, turbidity, aluminum foil reflectors and container volume on the inactivation efficiency of batch-process solar disinfection, *Water Res.* 35 (2001) 1061–1065.
- [16] A. Martín-Domínguez, M.T. Alarcón-Herrera, I.R. Martín-Domínguez, A. González-Herrera, Efficiency in the disinfection of water for human consumption in rural communities using solar radiation, *Sol. Energy* 78 (2005) 31–40.
- [17] A.G. Rincón, C. Pulgarin, Solar photolytic photocatalytic disinfection of water at laboratory and field scale—Effect of the chemical composition of water and study of the postirradiation events, *J. Sol. Energy Eng.* 129 (2007) 100–110.
- [18] D.M.A. Alrousan, P.S.M. Dunlop, T.A. McMurray, A. Byrne, Photocatalytic inactivation of *E. coli* in surface water using immobilised nanoparticle TiO₂ films, *Water Res.* 43 (2009) 47–54.
- [19] D. Bas, I.H. Boyaci, Modeling and optimization I: Usability of response surface methodology, *J. Food Eng.* 78 (2007) 836–845.
- [20] A.T. Nair, A.R. Makwana, M.M. Ahammed, The use of response surface methodology for modelling and analysis of water and wastewater treatment processes: A review, *Water Sci. Technol.* 69 (2014) 464–478.
- [21] D.C. Montgomery, Design and Analysis of Experiments, seventh ed., Wiley India Pvt. Ltd., New Delhi, 2012.
- [22] F. Ay, E.C. Catalkaya, F. Kargi, A statistical experiment design approach for advanced oxidation of Direct Red azo-dye by photo-Fenton treatment, *J. Hazard. Mater.* 162 (2009) 230–236.
- [23] C. Moon, R. Singh, S.R. Chaganti, J.A. Lalman, Modeling sulfate removal by inhibited mesophilic mixed anaerobic communities using a statistical approach, *Water Res.* 47 (2013) 2341–2351.
- [24] S. Ghafari, H.A. Aziz, M.I. Isa, A.K. Zinatizadeh, Application of response surface methodology (RSM) to optimize coagulation flocculation treatment of leachate using poly aluminum chloride (PAC) and alum, *J. Hazard. Mater.* 163 (2009) 650–656.
- [25] S. Sadri Moghaddam, M.R. Alavi Moghaddam, M. Arami, Coagulation/flocculation process for dye removal using sludge from water treatment plant: Optimization through response surface methodology, *J. Hazard. Mater.* 175 (2010) 651–657.
- [26] M. Zainal-Abideen, A. Aris, F. Yusof, Z. Abdul-Majid, A. Selamat, S.I. Omar, Optimizing the coagulation process in a drinking water treatment plant—Comparison between traditional and statistical experimental design jar tests, *Water Sci. Technol.* 65 (2012) 496–503.
- [27] H. Zhang, Y. Li, X. Wu, Statistical experiment design approach for the treatment of landfill leachate by photoelectro-Fenton process, *J. Environ. Eng.* 138 (2012) 278–285.
- [28] M. Umar, H.A. Aziz, M.S. Yusoff, Assessing the chlorine disinfection of landfill leachate and optimization by response surface methodology (RSM), *Desalination* 274 (2011) 278–283.
- [29] S.L.C. Ferreira, R.E. Bruns, H.S. Ferreira, G.D. Matos, J.M. David, G.C. Brandão, E.G.P. da Silva, L.A. Portugal, P.S. dos Reis, A.S. Souza, W.N.L. dos Santos, Box-Behnken design: An alternative for the optimization of analytical methods, *Anal. Chim. Acta* 597 (2007) 179–186.
- [30] M.A. Bezerra, R.E. Santelli, E.P. Oliveira, L.S. Villar, L.A. Escalera, Response surface methodology (RSM) as a tool for optimization in analytical chemistry, *Talanta* 76 (2008) 965–977.

- [31] M.J.K. Bashir, M.H. Isa, S.R.M. Kutty, Z.B. Awang, H.A. Aziz, S. Mohajeri, I.H. Farooqi, Landfill leachate treatment by electrochemical oxidation, *Waste Manage.* 29 (2009) 2534–2541.
- [32] S. Mohajeri, H.A. Aziz, M.H. Isa, M.A. Zahed, M.N. Adlan, Statistical optimization of process parameters for landfill leachate treatment using electro-Fenton technique, 176 (2010) 749–758.
- [33] H. Zhang, Y.L. Li, X.G. Wu, Y.J. Zhang, D.B. Zhang, Application of response surface methodology to the treatment landfill leachate in a three-dimensional electrochemical reactor, *Waste Manage.* 30 (2010) 2096–2102.
- [34] American Public Health Association, Standards Methods for the Examination of Water and Wastewater, 20th ed, American Public Health Association/American Water Works Association/Water Pollution Control Federation, Washington, DC, 1998.
- [35] M.J. Anderson, P.J. Whitcomb, RSM Simplified, Productivity Press, New York, NY, 2005.
- [36] H.-L. Liu, Y.-W. Lan, Y.-C. Cheng, Optimal production of sulphuric acid by *Thiobacillus thiooxidans* using response surface methodology, *Process Biochem.* 39 (2004) 1953–1961.
- [37] V. Meera, M.M. Ahammed, Solar disinfection for household treatment of roof-harvested rainwater, *Water Sci. Technol.: Water Supply* 8 (2008) 153–160.
- [38] M.T. Amin, M.Y. Han, Roof-harvested rainwater for potable purposes: Application of solar collector disinfection (SOCO-DIS), *Water Res.* 43 (2009) 5225–5235.
- [39] E. Ubomba-Jaswa, P.F. Fernández-Ibáñez, C. Navntoft, M.I. Polo-López, K.V. McGuigan, Investigating the microbial inactivation efficiency of a 25 L batch solar disinfection (SODIS) reactor enhanced with a compound parabolic collector (CPC) for household use, *J. Chem. Technol. Biotechnol.* 85 (2010) 1028–1037.
- [40] A.G. Rincón, C. Pulgarin, Field solar *E. coli* inactivation in the absence and presence of TiO₂: Is UV solar dose an appropriate parameter for standardization of water solar disinfection? *Sol. Energy* 77 (2004) 635–648.
- [41] M.B. Fisher, C.R. Keenan, K.L. Nelson, B.M. Voelker, Speeding up solar disinfection (SODIS): Effects of hydrogen peroxide, temperature, pH and copper plus ascorbate on the photoinactivation of *E. coli*, *J. Water Health* 6 (2008) 35–51.
- [42] M.T. Amin, M.Y. Han, Improvement of solar based rainwater disinfection by using lemon and vinegar as catalysts, *Desalination* 276 (2011) 416–424.
- [43] T.P. Curtis, D.D. Mara, S.S. Silva, Influence of pH, oxygen, and humic substances on ability of sunlight to damage fecal coliforms in waste stabilization pond water, *Appl. Environ. Microbiol.* 58 (1992) 1335–1343.