A new empirical approach for modelling fate and transport of Chromium bioaccumulation in irrigated crops: A water-food-pollution nexus

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ABSTRACT

Discharge of chromium (Cr) into receiving water bodies is a serious problem in water resources worldwide that inevitably gets taken up by agricultural crops and hence threatens both the environment and human health. This study investigates the fate and transport modelling of Cr discharged into the Kashaf River by leather industries in Mashhad city, Iran and the bio magnification effects on agricultural crops irrigated by the river. The accumulative concentration of Cr in tomato in the present case study from the time of planting until harvest day shows an increasing trend of up to 126 µg/L. The sensitivity analysis illustrates that the accumulated chromium ions in tomato are affected by time in growth cycle, chromium dosage in water, and total hardness of water more than any other factors. This study adopts an empirical approach by developing statistical modelling for bio-accumulated Cr in tomato during the growth period and evaluates different 3D mathematical distribution such as Polynomial, Interpolant, and Lowest models. The results demonstrate Polynomial with x and y more than four-degree model has the best efficiency for the measurement of accumulated chromium ion in tomato as per qualitative factors. The outputs in this study can be viewed in the context of water-food-pollution nexus and how the pollution discharged from the industry into the water resources can have a major impact on the safety of food that is dependent on irrigation from freshwater resources.

KEYWORDS

Bio-magnification; chromium; footprint; water-food-pollution nexus; empirical modelling.

1. INTRODUCTION

Chromium is one of the heavy metals that can be released in water sources and lead to the occurrence of many immunological and epidemiological problems [1-3]. Meanwhile, chromium ions can accumulate in plant tissue and increase the risk of human disease following consumption [4-6]. However, with the increase in the amount of chromium pollutants in water sources, bioaccumulation raises in the tissue of plant organisms and food accordingly [7]. This can lead to cancer and increased health problems [8]. On the other hand, measuring chromium ion contamination *in situ* in real time, is a difficult and expensive task. This is where statistical and mathematical modelling add value, by making it possible to predict the amount of pollution, which can then enable appropriate decision-making for managing this challenge [6-8].

Various industries can lead to the release of chromium in water such as leather, electroplating, dyeing, and steel industries. The conceptual map shown in Fig. 1 illustrates this [9]. Furthermore, industrial activities and resulting waste discharged into water resources can be one of the main sources of pollution in the environment. More importantly, the operation of industrial wastewater treatment plants in developing economies is often overlooked due to the lack of attention and absence of proper enforcements from environmental organisations which can lead to the propagation of such a pollution [8-11]. More specifically, agricultural activities are carried out with the untreated sewage discharged into the rivers, and this leads to the occurrence of problems such as bioaccumulation [12]. In the bio-magnification process, the pollutant concentration in the plant irrigated by contaminated water is much higher, and similarly, the amount amassed in the body of the organism that uses the plant for nutrition is higher. In fact, in the food chain, the more pollution is transferred to higher lifeforms, increasing the health risk, several times [10-12].

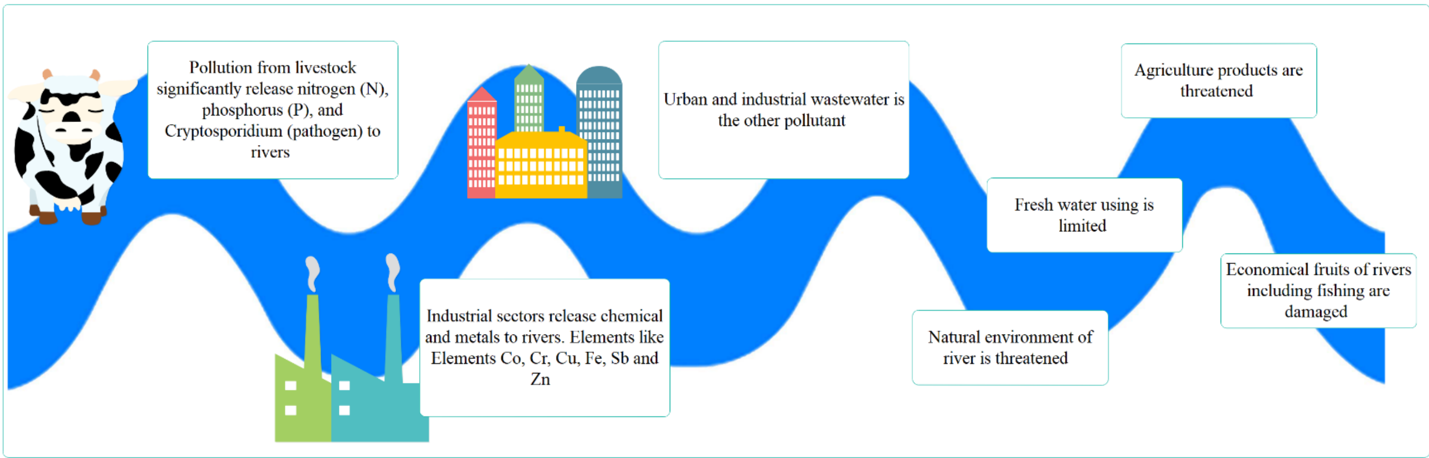


Fig. 1. The interaction of pollution from human activities discharged into a river with the threats to the environment and human demands.

Bio-magnification in tomato products, which is the subject case study herewith, is beginning to receive attention from the scientific community; and hence recent research works are summarised in Table 1.

Table 1. A summary of the literature review for bioaccumulation studies in tomato plants

Research target	Researchers	Reference
Experimental practices for exact determination of diclofenac, sulfamethoxazole, and trimethoprim bioaccumulation in tomato plants during long-term irrigation with wastewater resources	Christou et al. (2017)	[13]
Experimental evaluation of sludge and sludge biochar effects on heavy metals, bioaccumulations during growth process of cherry tomato	HOSSAIN et al. (2015)	[14]
Experimental assessment of nickel bio-magnification in tomato plants with focusing on human health risk reduction	Correia et al. (2018)	[15]
Appraisal of pharmaceuticals bioaccumulation in tomato plants by experimental practices	Boer et al. (2018)	[16]
Practical evaluation of perfluoroalkyl substances bioaccumulation in tomato which is irrigated by groundwater resources	Bao et al. (2020)	[17]
Fate and transport assessment of lead and cadmium in tomato by experimental practices	Khan et al. (2016)	[18]

This body of work shows that different contaminations have been investigated mainly on experimental practices. However, the chromium bioaccumulation process is rarely considered and analysed in detail. In addition, the chromium bioaccumulation statistical modelling is not strongly considered by previous investigations. Hence, this study aims to develop empirical models by adopting a statistical approach for estimating the bioaccumulation of chromium in tomato and conduct a sensitivity analysis to determine the key factors in the food-water nexus. This study therefore aims to (1) measure and evaluate an assessment of bio-accumulated chromium ions in tomato plants through irrigation process in a year with experimental practices, (2) conduct sensitivity analysis of effective factors of the bioaccumulation process including water temperature, pH, Total Dissolved Solid (TDS), Total Hardness (TH), chromium concentration in water, and Sodium Adsorption Ratio (SAR) using Historical data assessment in Response Surface Methodology,(3) develop statistical modelling based on the effective features after conducting sensitive analysis and (4) find out the best models for estimation of chromium concentration

2. METHODOLOGY

2.1. CASE STUDY

The subject case study is the agricultural tomato lands downstream of the Kashf River in Mashhad city located in the east of Iran. The Kashaf River basin is a part of Qaraqom district in the southwest of Qaraqom. Four areas of study include Mashhad, Sengbast, Narimani and Aghaderband, the Kashaf River basin, which is equivalent to 76,161 Km^2, between 58^o 22’ and 60^o 10’ longitude. The Kashaf River as the main drainage of this water basin is based in the valley located between Hezar-Masjid and Binalud mountains. The hydraulic and hydrological specifications of the Kashaf River is demonstrated in Fig. 2 along with the various industrial wastewater discharge into the Kashaf River shown in Fig. 3.

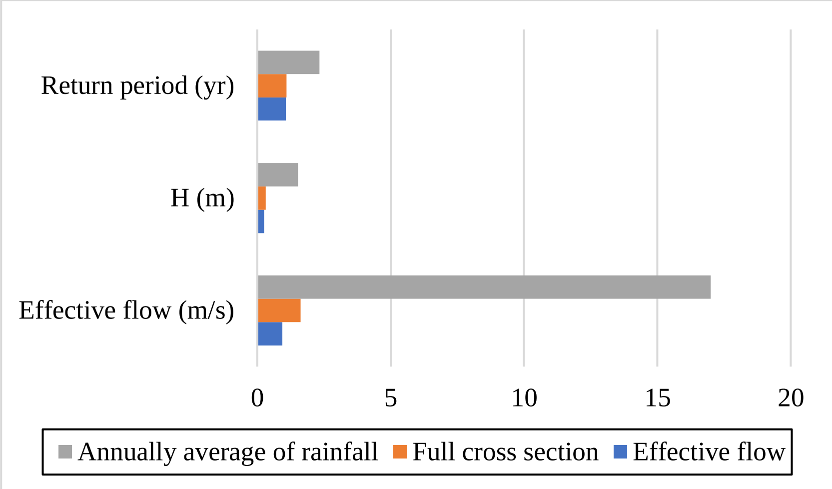


Fig. 2. Hydraulic and hydrological specifications of the Kashaf River

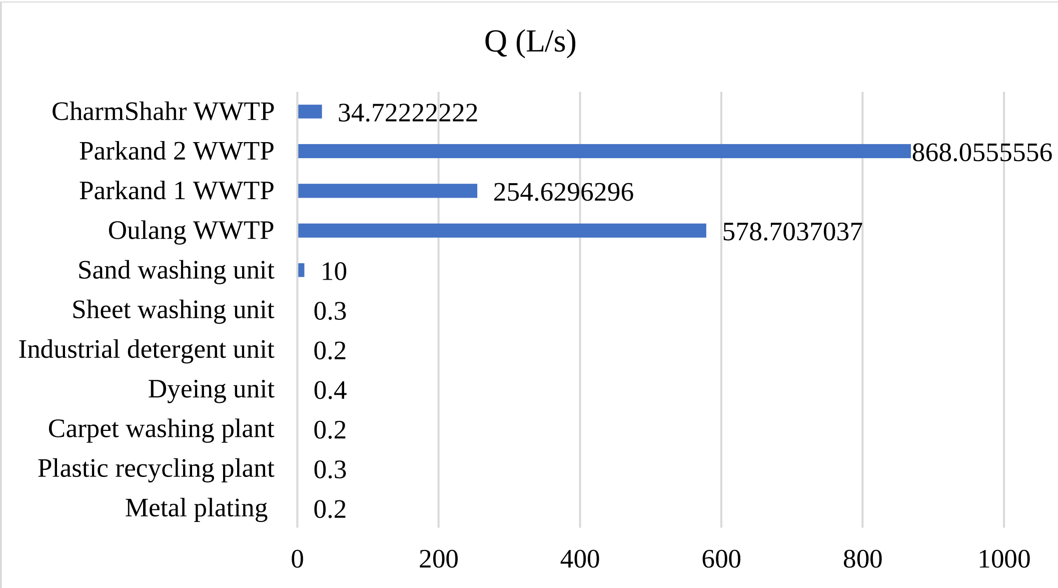


Fig. 3. List of the different industrial discharge into the Kashaf River

2.2. EXPERIMENTAL MEASUREMENTS

This study conducted experimental measurement for chromium, pH, TH, temperature, TDS, and SAR based on standard methods summarised in Table 2; this was in order to substantiate the mathematical model. It should be noted that all consumed materials and chemical compounds are provided by the instruments with highest standards and latest technologies in the market as outlined in the Table.

Table 2. The applied devices and protocols in this study

Measurement item	Instruments	Protocols	Reference
Measurement of chromium in water samples	of Perkin Elmer Analyst 700 Atomic Absorption Spectroscopy (AAS) and specific 357.9 nm lamp	Acidification by 1 mol L-1 HNO3 and AAS	[19]
Measurement of chromium in tomatoes	of Perkin Elmer Analyst 700 Atomic Absorption Spectroscopy (AAS) and specific 357.9 nm lamp	Standard methods for water and wastewater examinations	[20]
pH	pH and EC meter: 878, Metrohm AG, Switzerland	Standard methods for water and wastewater examinations	[20]
TDS	pH and EC meter: 878, Metrohm AG, Switzerland	Standard methods for water and wastewater examinations	[20]
TH	Hach 145300 Total Hardness Test Kit, Model 5-B	Standard methods for water and wastewater examinations	[20]
SAR	HI 38078 Sodium Adsorption Ratio (SAR) Test Kit	Soil chemistry methods	[21]

2.3. NUMERICAL ANALYSIS

This study conducts a sensitive analysis through the application of historical data analysis in Design Expert Software version 7 [22-26] and develops mathematical modelling using CFtool box in MATLAB 2018b software [27-30]. The best fitting condition is also implemented by using try and error practices in the data distribution appraisal step. To that end, the effects of each feature are first evaluated on bioaccumulation of chromium, and it is then modelled by the most important factors.

3. RESULTS AND DISCUSSION

3.1.EXPERIMENTAL OUTPUTS

The results of measuring some water and soil quality indicators during the tomato growth period are shown in Fig. 4. Different factors have some fluctuation during the growth of the plant and are widely distributed in the environment. Meanwhile, the amount of chromium bioaccumulation as shown in Fig. 4d increases during the growth period. The concentration of chromium ions in water also varies according to the different quality conditions of the effluent of the Charmshahr treatment plant as the main source of chromium discharge. From this perspective, we are unable to consider a specific trend for chromium concentration in water, because the factor reported is directly related to the operation of the industrial wastewater treatment plant and the hydraulic conditions of the river. However, the tolerance of pH and SAR vary between 7-8 and 2-3, respectively and hence the experimental factors have no considerable fluctuations on these factors.

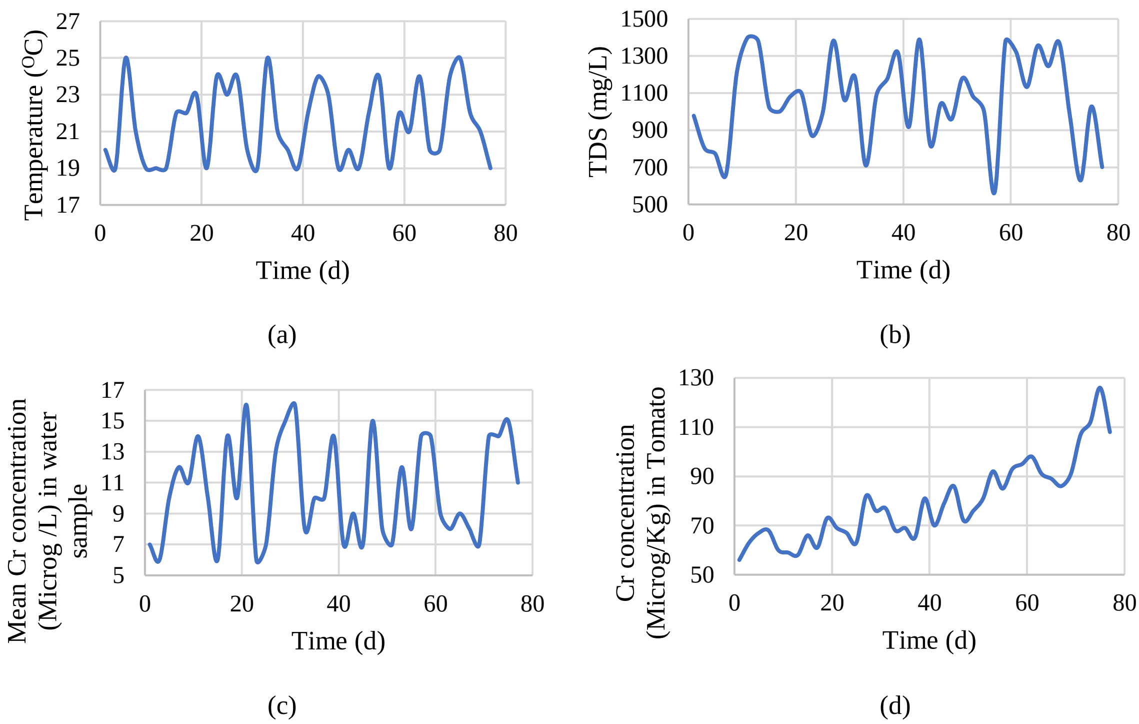


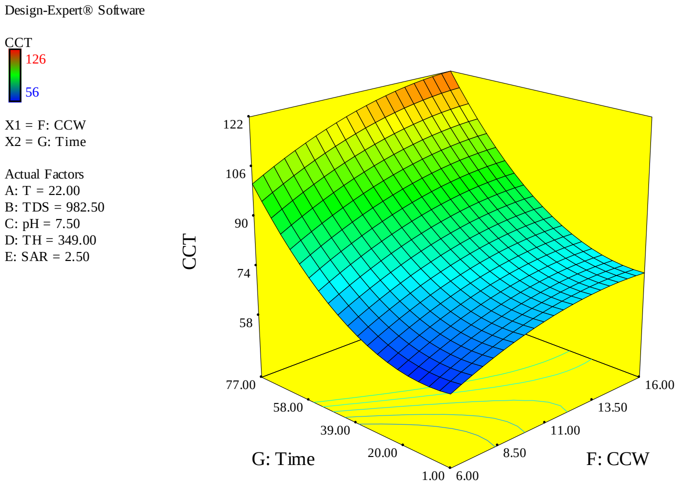
Fig. 4. The time series changes of (a) temperature, (b) TDS, (c) Chromium Concentration in Water (CCW), and (d) Chromium Concentration in Tomato (CCT).

3.2.SENSITIVITY ANALYSIS

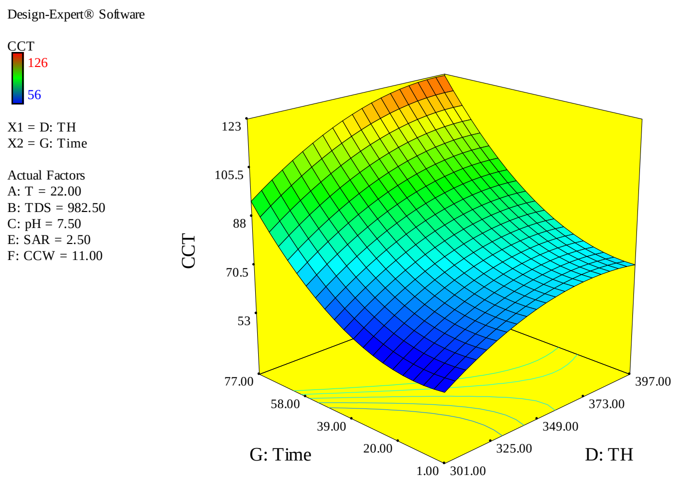
This study used ANOVA method in historical data analysis of response surface methodology to analyse the sensitivity of factors affecting chromium ion bioaccumulation (Table 3). Considering the table and evaluating P-value and F-value, it can be concluded that among all the factors, time, TH and CCW have the greatest impact on CCT. The binary sensitivity analysis of basic parameters is also shown in Fig. 5. It can be seen in Figs. 5a-b that the changes in the slope of the Time line are more than TH/CCW and this indicates the importance of this index compared to the other two cases. As time progresses during the growth period, the amount of accumulated chromium increases. However, the comparison of other two cases (TH and CCW) in Fig. 5c, it can be concluded that the amount of bioaccumulation of chromium in tomato is more sensitive to the concentration of pollution in water, which is also consistent with basic concepts.

Table 3. The outputs of ANOVA method in historical data analysis of response surface methodology.

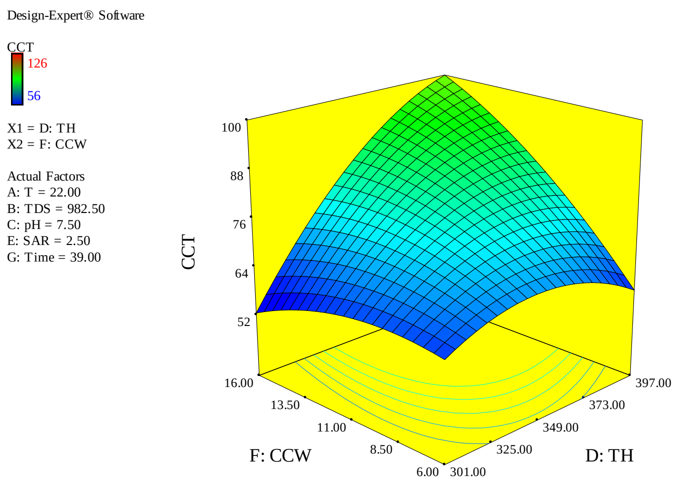
Source	Sum of Squares	Mean Square	F-Value	p-value
T	15.84674	15.84674	0.731152	0.4316
TDS	7.025597	7.025597	0.324154	0.5937
pH	27.67387	27.67387	1.276844	0.3098
TH	47.23769	47.23769	2.179499	0.1999
SAR	5.288823	5.288823	0.244021	0.6423
CCW	91.69992	91.69992	4.230942	0.0948
Time	800.9247	800.9247	36.95385	0.0017



(a)



(b)



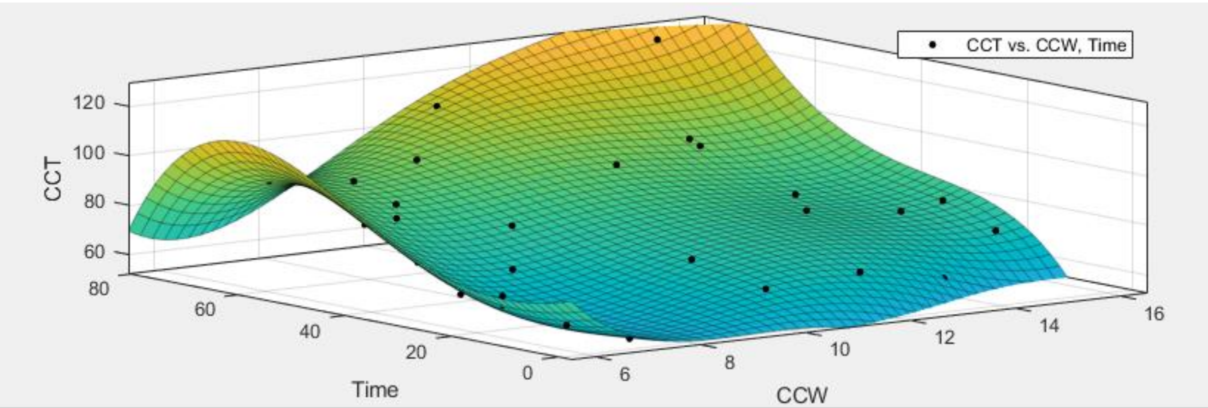
(c)

Fig. 5. The dual sensitivity analysis of (a) CCW-Time, (b) TH-Time, and (c) CCW-TH.

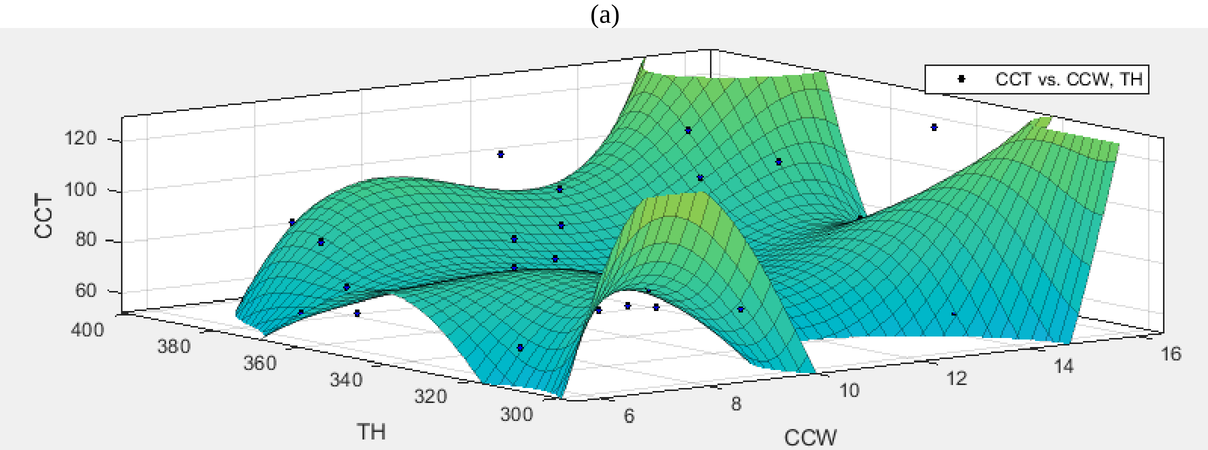
3.3.STATISTICAL MODELLING

This study adopts a trial-and-error approach to develop the empirical equations. As a result, it was shown that polynomial equations are the most appropriate in predicting CCT with effective variables. Although the ANOVA method shows the most significant factors for prediction of CCT are CCW and Time based on Fig. 6a and Eq. 1, it is evident that the CCT can be predicted based on CCW and Time with a correlation coefficient of 0.92.

On the other hand, the estimation of CCT is infeasible based on CCW and TH due to low regression coefficient during curve fitting process (0.51) that can also be found in Fig. 6b and Eq. 2. However, the Time-TH model for estimation of CCT is valid and acceptable with higher correlation coefficient (i.e. greater than 0.91) due to the importance of Time in this target (Fig. 6c and Eq. 3).



(a)



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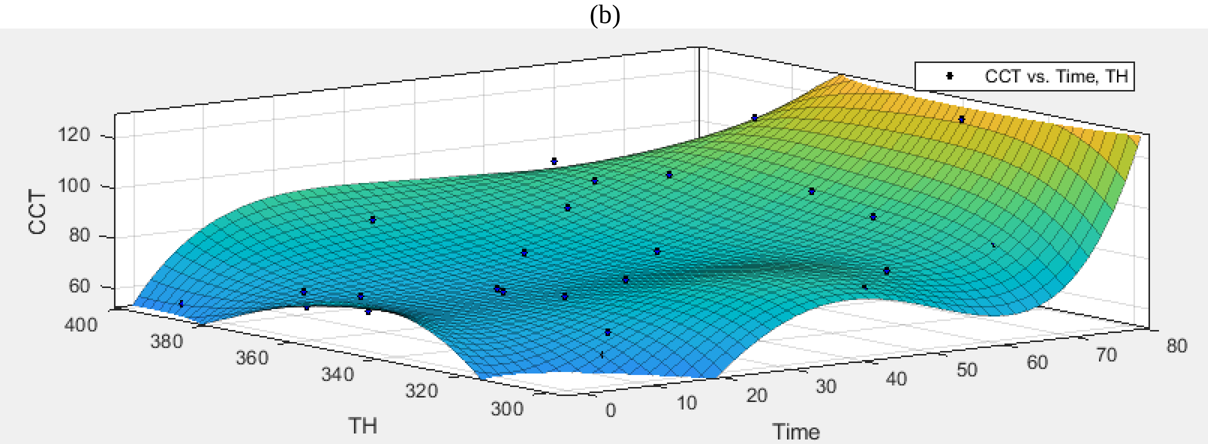

(c)

Fig. 6. The statistical models of (a) CCW-Time, (b) TH-Time, and (c) CCW-TH vs CCT.

(1)

$$f(x,y) = P_{00} + P_{10}X + P_{01}Y + P_{20}X^2 + P_{11}XY + P_{02}Y^2 + P_{30}X^3 + P_{21}X^2Y + P_{12}XY^2 + P_{03}Y^3 + P_{40}X^4 + P_{31}X^3Y + P_{22}X^2Y^2 + P_{13}XY^3 + P_{04}Y^4$$

Where f(x,y), X and Y represent CCT, CCW, and TH respectively.

Coefficients (with 95% confidence bounds):

P_{00} = 636.1	P_{10} = -227.6	P_{01} = -0.641	P_{20} = 32.2
P_{11} = -0.1382	P_{02} = 0.08948	P_{30} = -1.93	P_{21} = 0.02458
P_{12} = -0.009191	P_{03} = -0.000537	P_{40} = 0.04074	P_{31} = 0.0005519
P_{22} = -0.0004125	P_{13} = 0.0001588	P_{04} = -7.257e-06	

Goodness of fit:
SSE: 746.9
R-square: 0.9274
Adjusted R-square: 0.8851
RMSE: 5.579 µg/Kg

(2)

$$f(x,y) = P_{00} + P_{10}X + P_{01}Y + P_{20}X^2 + P_{11}XY + P_{02}Y^2 + P_{30}X^3 + P_{21}X^2Y + P_{12}XY^2 + P_{03}Y^3 + P_{40}X^4 + P_{31}X^3Y + P_{22}X^2Y^2 + P_{13}XY^3 + P_{04}Y^4 + P_{50}X^5 + P_{41}X^4Y + P_{32}X^3Y^2 + P_{23}X^2Y^3 + P_{14}XY^4 + P_{05}Y^5$$

Where f(x,y), X and Y represent CCT, CCW, and Time respectively.

Coefficients (with 95% confidence bounds):

P_{00} = 2.383e+06	P_{10} = -1.125e+04	P_{01} = -3.441e+04	P_{20} = -2155
P_{11} = 245.5	P_{02} = 196.8	p_{30} = 68.56	P_{21} = 12.27
P_{12} = -1.372	P_{03} = -0.559	P_{40} = 0.4304	P_{31} = -0.4358
P_{22} = -0.01568	P_{13} = 0.002849	P_{04} = 0.000791	P_{50} = -0.01337
P_{41} = 0.0006998	P_{32} = 0.0005782	P_{23} = -2.226e-06	P_{14} = -1.953e-06
P_{05} = -4.474e-07			

Goodness of fit:
SSE: 4985
R-square: 0.5157
Adjusted R-square: -0.02244
RMSE: 16.64 µg/Kg

(3)

$$f(x,y) = P_{00} + P_{10}X + P_{01}Y + P_{20}X\hat{}2 + P_{11}XY + P_{02}Y\hat{}2 + P_{30}X\hat{}3 + P_{21}X\hat{}2Y + P_{12}XY^2 + P_{03}Y^3 + P_{40}X^4 + P_{31}X^3Y + P_{22}X^2Y^2 + P_{13}XY^3 + P_{04}Y^4 + P_{41}X^4Y + P_{32}X^3Y^2 + P_{23}X^2Y^3 + P_{14}XY^4 + P_{05}Y^5$$

Where f(x,y), X and Y represent CCT, TH, and Time respectively.

Coefficients (with 95% confidence bounds):

P_{00} = -5814	P_{10} = -356.3	P_{01} = 0.2155	P_{20} = -1.642
P_{11} = 5.321	P_{02} = 0.3614	P_{30} = -0.008268	P_{21} = 0.0104
P_{12} = -0.02705	P_{03} = -0.001766	P_{40} = 0.0001645	P_{31} = -2.022e-05
P_{22} = -1.072e-05	P_{13} = 5.716e-05	P_{04} = 3.189e-06	P_{41} = -4.46e-07
P_{32} = 1.167e-07	P_{23} = -1.474e-08	P_{14} = -4.29e-08	P_{05} = -2.014e-09

Goodness of fit:
SSE: 920.8
R-square: 0.9105
Adjusted R-square: 0.8211
RMSE: 6.962 µg/Kg

4. CONCLUSION

One of the most important risks to the environment and human health is related to biological magnification. However, the food-water nexus is currently significantly overshadowed by this phenomenon. In developing economies, due to the increase in industrialisation, the release of heavy metals in water sources, biological magnification resulting from uncontrolled irrigation, and finally increasing the risk health problems can be a major challenge. This study first measured the concentration of chromium as in the case of tomato products during growth period and specifications of water samples including pH, SAR, TH, Time of growth, and CCW used for irrigation. After evaluation of fluctuations in the time series of variables, the sensitive analysis is conducted to determine the most significant features in bioaccumulation process of chromium ions in tomato. The results demonstrated that Time of growth, CCW, and TH have the most effects on CCT and with the application of fourth degree polynomial regression equations, the CCT can be predicted based on CCW-Time. It is recommended that future studies focus on the evaluation of more than one heavy metal biomagnification in tomato due to evaluation of competitive effects. Technologies such as microbial fuel cells can be used to monitor and treat hexavalent chromium, thus reducing the carcinogenic risk and therefore will be employed to enable real time monitoring, which in conjunction with machine learning for assessment and estimation of bioaccumulation process can be an effective tool for combating this challenge.

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