



Determination of arsenic removal efficiency by ferric ions using response surface methodology

Meltem Bilici Baskan*, Aysegul Pala

Department of Environmental Engineering, Dokuz Eylul University, Buca, Izmir, Turkey

ARTICLE INFO

Article history:

Received 28 July 2008

Accepted 25 November 2008

Available online 11 December 2008

Keywords:

Arsenic removal

Iron salts

Coagulation

Box–Behnken design

ABSTRACT

Arsenic contamination of drinking water is a serious problem in many parts of the world. The precipitation/coprecipitation method was used for arsenic removal from drinking water by ferric chloride, ferric sulfate and ferrous sulfate as coagulant. A Box–Behnken statistical experiment design method was used to investigate the effects of major operating variables such as initial arsenate concentration ($10\text{--}1000\ \mu\text{g L}^{-1}$), coagulant dose ($0.5\text{--}60\ \text{mg L}^{-1}$) and pH ($4\text{--}9$) were investigated. Experimental data were used for determination of the response functions coefficients. Predicted values of arsenate removal obtained using the response functions were in good agreement with the experimental data. Fe(III) ions were more effective and economic than Fe(II) ion due to required lower coagulant dose and pH. In the low initial arsenate concentrations, the highest arsenate removal efficiency was required high ferric chloride and ferric sulfate dose of 50 and 40 mg L^{-1} , while in the high initial arsenate concentrations, the highest arsenate removal efficiency was provided at low ferric chloride and ferric sulfate dose of 37 and 32 mg L^{-1} , respectively. This study showed that Box–Behnken design and response surface methodology was reliable and effective in determining the optimum conditions for arsenic removal by coagulation and flocculation.

© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Inorganic arsenic (trivalent arsenic [As(III)] and pentavalent arsenic [As(V)]) is mostly found in natural waters of many parts of the world and has been serious problem for its toxicity [1,2]. High arsenic drinking water areas are reported all over the world including Bengal Delta Plain (West Bengal, India and Bangladesh) [3–6], United States [7–10], China [11], and New Zealand [12]. India and Bangladesh especially, population over 450 million are at risk of arsenic poisoning [13]. Effects of arsenic exposure via drinking water include various type of skin lesions, neurological effects, peripheral vascular disease, cardiovascular disease, respiratory disease, skin and other cancers (bladder, lung, liver), and immune system [14–27]. The Environmental Protection Agency (EPA) reduced the maximum contaminant level (MCL) for arsenic in drinking water from 50 to $10\ \mu\text{g L}^{-1}$ due to the international crisis about arsenic contamination and associated health effects on humans [28]. According to the last edition of the World Health Organization (WHO) Guidelines for Drinking-Water Quality (1993), $10\ \mu\text{g L}^{-1}$ was established as a provisional guideline value for arsenic [29].

Natural water sources contained much higher levels of arsenic ($20\text{--}3000\ \mu\text{g L}^{-1}$) than maximum contaminated level set in Turkey national drinking water standards ($10\ \mu\text{g L}^{-1}$) were determined in western Turkey. High levels of arsenic in the natural waters were considered to be associated with the dissolution of some minerals in the colemanite boron formations [30–32]. Because of the boron deposits and volcanic formation in the Aegean Region, it is forecasted that drinking water sources in this region may have high concentration of arsenic.

A variety of treatment processes has been developed for arsenic removal from water. The USEPA has identified seven best available technologies (BATs), which are given in Table 1. EPA determined these technologies to be the BATs for the removal of arsenic in drinking water based on a demonstration of efficacy under field conditions taking cost into consideration. All of these BATs are for arsenate (As(V)). Arsenate is relatively easy to remove from water, since it bears a negative charge in natural waters above pH 2.2, and is electrostatically attracted to the positive charge on metal hydroxide surfaces [33]. Under reducing conditions at pH less than about pH 9.2, the uncharged arsenite (As(III)) species will predominate [1]. Therefore As(III) is less efficiently removed than As(V), so pre-oxidation is necessary for better removal [34]. This work presents an evaluation of coagulation with ferric ions for arsenate removal at different initial arsenate concentrations.

It has been indicated that coagulation is not only effective for the removal of turbidity and color but also effective for the removal

* Corresponding author. Tel.: +90 232 4127144 fax: +90 232 4531134.
E-mail address: meltem.bilici@deu.edu.tr (M.B. Baskan).

Table 1
Best available technologies and their arsenic removal efficiencies [33].

Treatment technology	Maximum removal (%)
Activated alumina	95
Coagulation/filtration	95
Ion exchange	95
Lime softening	90
Reverse osmosis	>95
Electrodialysis	85
Oxidation/filtration	80

of soluble contaminants such as arsenic [35,36]. In this process, fine particles in water first aggregate into coagulate because added ferric or aluminum ions strongly reduce the absolute values of zeta potentials of particles. Then, arsenic ions (arsenate or arsenite) precipitate with the ferric or aluminum ions on the coagulates, and thus concentrate in the coagulates [36]. Maximization of the coagulation process efficiency, thus, arsenic removal by optimization of the other operating conditions, such as chemical concentrations, operational pH, and initial arsenic concentration, therefore, is very important. For this aim, the Box–Behnken experimental design method was used to find optimum removal conditions. Box–Behnken experimental design is a response surface methodology used for analysis the experimental design data in order to be correlated to the independent variables [37].

The major objective of this study is to investigate removal efficiencies of arsenate from tap water by precipitation/coprecipitation with ferric chloride, ferric sulfate, and ferrous sulfate as coagulant. A Box–Behnken statistical experiment design method was used to investigate the effects of important operating parameters such as initial arsenic concentrations, pH, and coagulant doses on percent removal of arsenate and to find the combination of variables resulting in maximum arsenic removal efficiency.

2. Materials and methods

2.1. Reagents

Characterization of tap water used in the study is listed in Table 2. All the chemicals were of reagent grade or better and were used without further purification. Water samples has been provided with adding of sodium arsenate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) as As(V) source (purchased from Sigma). Synthetic contaminated water of 2 mg L^{-1} concentration was spiked with As(V) was prepared from tap water. In the experimental studies, this stock arsenic solution was used after diluted until desire concentration. For the coagulation experiments all solutions were prepared with distilled water and all glassware was previously acid-washed. Ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) and ferric sulfate ($\text{Fe}_2(\text{SO}_4)_3 \cdot 5\text{H}_2\text{O}$) used as source of Fe(III) and ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) used as source of Fe(II), were analytical grade and purchased from Merck or Carlo Erba. An amount of 500–5000 mg L^{-1} Fe(III) and Fe(II) stock solutions were prepared for further dilution to obtain a solution of desired con-

Table 2
Characterization of tap water used for experiment.

Components	Concentrations
pH	8.2
Turbidity (NTU)	0.1
Chloride (mg L^{-1})	46
Nitrate (mg L^{-1})	3
Iron (mg L^{-1})	0.0343
Manganese (mg L^{-1})	0.0141
Sodium (mg L^{-1})	23
Conductivity ($\mu\text{S/cm}$)	463
Sulfate (mg L^{-1})	36

centrations. Hydrochloric acid (HCl, 37%) and sodium hydroxide (NaOH) were used to adjust pH.

2.2. Experimental procedure

Coagulation experiments were conducted using the standard jar test apparatus. A series of jar tests was performed using the tap water has various As(V) concentrations. Coagulation was carried out with ferric chloride, ferric sulfate, and ferrous sulfate. The coagulant was added to each 1 L jar containing the sample water with rapid mixing at 120 ± 2 rpm. After 3 min of rapid mix, 30 min of slow mixing at 45 ± 2 rpm was provided, followed by at 30 min of settling. Prior to addition of coagulant, the sample water pH was adjusted by adding HCl or NaOH. At the end of the settling period, water samples were taken from the supernatants, filtered by $0.45\text{-}\mu\text{m}$ pore size membrane filter, and stored for analysis.

2.3. Analytical methods

The determination of arsenic in the influent and effluent aqueous solutions was performed by the hydride generation procedure coupled with inductively coupled plasma-atomic emission spectrometry (HG-ICP-AES), also known as inductively coupled plasma-optical emission spectrometry (HG-ICP-OES) (Optima 2100 DV). The sodium borohydride reduction procedure instantaneously converts As^{+3} to arsine gas; however, at room temperature, the reduction of As^{+5} to arsine occurs relatively slow. Therefore, a total arsenic determination requires a prereductant such as KI to convert all arsenic to the +3 oxidation state prior to the arsine formation step [38,39]. Water (20 ml) was first mixed with 2 ml mixture of KI (5%) and ascorbic acid (5%) and 6 ml HCl (10%) for 30 min at dark place to reduce As(V) to As(III). Then, 10 ml solution was taken for the analysis of As(III) concentrations.

2.4. Experimental design and statistical analysis

The Box–Behnken experimental design method was used to determine the effects of major operating variables on arsenic removal and to find the combination of variables resulting in maximum arsenic removal efficiency. The Box–Behnken design is a response surface methodology, which is a collection of mathematical and statistical techniques that are useful for the modelling and analysis of problems in which a response of interest is influenced by several variables and the objective is to optimize this response [40]. This optimization process involves a series of steps: identify the problem to be solved, determine the factors and levels that affect the response variable, performing the statistically designed experiments, and data analysis [41].

For precipitation/coprecipitation process, three important operating parameters such as initial arsenic concentration, coagulant dose, and pH were chosen as the independent variables and designated as X_1 , X_2 , and X_3 , respectively. Initial arsenic concentration (X_1) was changed between 10 and 1000 $\mu\text{g L}^{-1}$, coagulant dose was (X_2) varied between 0.5 and 60 mg L^{-1} , and pH (X_3) was ranged from 4 to 9. As presented in Table 3, the experimental design involved three parameters (X_1 , X_2 and X_3), each at three levels, coded -1 , 0, and $+1$ for low, middle and high concentrations, respectively.

Table 3
Levels of each factor for Box–Behnken.

Independent factors	Units	Symbol	Coded levels		
			-1	0	$+1$
Initial As concentration	$\mu\text{g L}^{-1}$	X_1	10	505	1000
Coagulant Dose	mg L^{-1}	X_2	0.5	30.25	60
pH	–	X_3	4	6.5	9

Table 4
Experimental data points used in Box–Behnken experimental design.

Run	Initial As concentration ($\mu\text{g L}^{-1}$)	Coagulant dose (mg L^{-1})	pH
1	1000	30.25	4
2	10	60	6.5
3	505	30.25	6.5
4	1000	30.25	9
5	505	30.25	6.5
6	505	30.25	6.5
7	505	30.25	6.5
8	1000	0.5	6.5
9	505	0.5	9
10	10	30.25	9
11	505	30.25	6.5
12	10	0.5	6.5
13	505	0.5	4
14	10	30.25	4
15	505	60	9
16	505	60	4
17	1000	60	6.5

Experimental points for Box–Behnken statistical design are shown in Table 4. The centre point in the design was repeated five times for estimation of errors. In the correlating of the arsenic removal efficiency (Y) with other independent variables (X_1 – X_3), following response surface function was utilized

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{23}X_2X_3 + b_{11}X_1^2 + b_{22}X_2^2 + b_{33}X_3^2 \quad (1)$$

where Y is the predicted response surface function (percent As(V) removal), b_0 is the model constant, b_1 – b_3 linear coefficients, b_{12} , b_{13} , and b_{23} the cross product coefficients, and b_{11} , b_{22} , and b_{33} are the quadratic coefficients. The software Design Expert (Version 7.0.0, Stat-Ease Inc., Minneapolis, USA) and Statistica 5.0 were used for experimental design, determination of the coefficients and data analysis.

3. Results and discussion

Removal of arsenate (As(V)) from drinking water was investigated by precipitation/coprecipitation using different coagulants. The effects of important operating variables on percent As(V) removal was investigated by using Box–Behnken statistical experimental design. The coefficients of the response function (Eq. (1)) for arsenate removal efficiencies were obtained using experimental data and presented in Table 5. Predicted values of percent As(V) removal were determined by the response functions with the obtained coefficients. Table 6 presents a comparison of experimental and predicted values using different coagulants for percent removal of As(V). As(V) removal efficiencies were varied between 0 and 98.10 for ferric chloride and ferric sulfate, 0 and 94.06 for ferrous sulfate. The correlation coefficients (R^2) between the observed and predicted values were obtained as 0.96, 0.97, and 0.92 for ferric chloride, ferric sulfate, and ferrous sulfate, respectively. Predicted and experimental values of As(V) removal were in good agreement as shown in Table 6.

Table 7
Analysis of variance (ANOVA) of response surface quadratic model for used coagulants.

Source	FeCl ₃			Fe ₂ (SO ₄) ₃			FeSO ₄		
	Mean squares	F value	p-Value	Mean squares	F value	p-Value	Mean squares	F value	p-Value
Model	3427.97	20.61	0.0003	3416.07	28.31	0.0001	996.94	9.29	0.0038
Residual	166.32			120.67			107.28		
Lack of fit	385.78	223.17	<0.0001	280.19	273.1	<0.0001	245.41	66.71	0.0007
Pure error	1.73			1.03			3.68		
	$R^2 = 0.9636$; adjusted $R^2 = 0.9169$			$R^2 = 0.9733$; adjusted $R^2 = 0.9389$			$R^2 = 0.9228$; adjusted $R^2 = 0.8235$		

p-Value less than 0.0500 indicate model terms are significant.

Table 5
Coefficients for the quadratic model.

Coefficients	Values		
	FeCl ₃	Fe ₂ (SO ₄) ₃	FeSO ₄
b_0	-234.484	-205.18388	103.364
b_1	0.032565	0.075553	0.034332
b_2	1.107735	1.07981	-0.73751
b_3	76.82133	63.37558	-34.6963
b_{12}	0.000138	0.00100331	-0.00007639
b_{13}	0.007051	0.002600606	0.004949
b_{23}	0.320807	0.29687	0.221647
b_{11}	-0.00007098	-0.0000939231	-0.00003590
b_{22}	-0.03398	-0.043741	-0.00445
b_{33}	-6.14048	-4.73016	2.618

Table 6
Observed and predicted As(V) removal efficiency.

No.	As(V) removal efficiency (%)					
	FeCl ₃		Fe ₂ (SO ₄) ₃		FeSO ₄	
	Observed	Predicted	Observed	Predicted	Observed	Predicted
1	10.00	9.73	10.4	23.60	20.70	22.82
2	90.00	75.52	30.00	31.53	0.00	15.24
3	94.85	94.50	94.46	96.00	30.69	28.91
4	84.90	78.48	93.30	90.95	85.20	77.78
5	95.64	94.50	96.44	96.00	30.89	28.91
6	95.25	94.50	95.84	96.00	26.34	28.91
7	92.28	94.50	96.04	96.00	28.71	28.91
8	0.00	14.49	9.00	7.47	24.60	19.36
9	0.00	0.00	6.34	10.22	21.78	34.44
10	50.00	50.28	70.00	56.80	30.00	37.88
11	94.46	94.50	97.23	96.00	27.92	28.91
12	0.00	7.79	0.00	9.32	0.00	0.00
13	2.57	0.00	5.15	0	21.58	24.70
14	10.00	16.43	0.00	2.35	0.00	7.42
15	97.23	100.00	94.46	100	94.06	90.94
16	4.36	12.42	4.95	1.07	27.92	15.26
17	98.10	90.32	98.10	88.78	30.10	40.64

Table 7 shows the analysis of variance results of the response surface quadratic model for ferric chloride, ferric sulfate, and coprecipitation. It is evident from Table 7 that the predicted responses fit the well with those of the experimentally obtained responses. The coefficients of determination (R^2) values are 0.9636, 0.9733 and 0.9228 which show that the equations are highly reliable. The p values less than 0.05 indicate that the model is statistically significant. The models for three types of coagulant were found to be adequate for prediction within the range of variable chosen.

3.1. Effect of pH

In order to determine the effect of pH on arsenate removal efficiency at an initial arsenate concentration of $500 \mu\text{g L}^{-1}$, some experiments were executed. Fig. 1(a–c) depicts the variation of percent arsenate removal with the ferric chloride, ferric sulfate and ferrous sulfate dose at different pH and a constant initial arsenate concentration of $500 \mu\text{g L}^{-1}$.

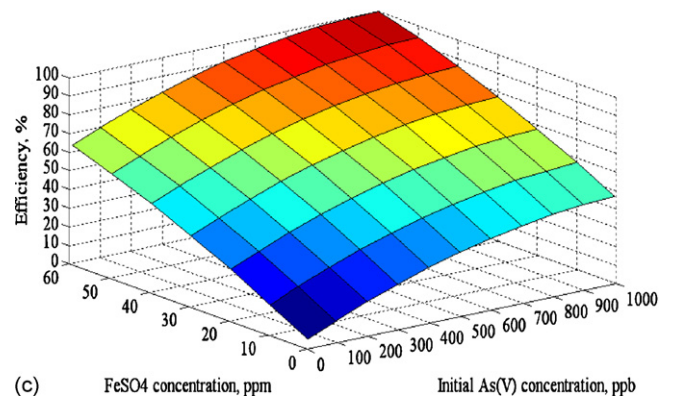
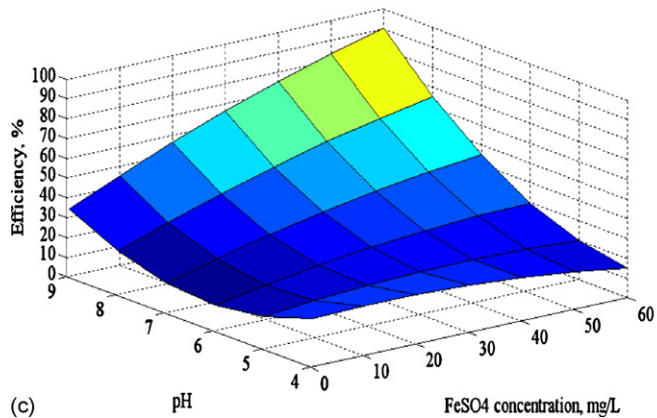
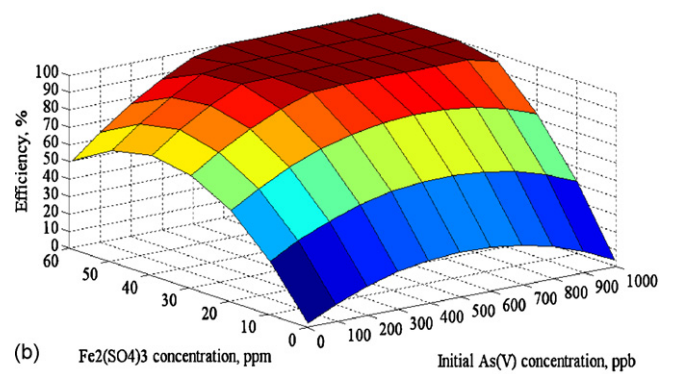
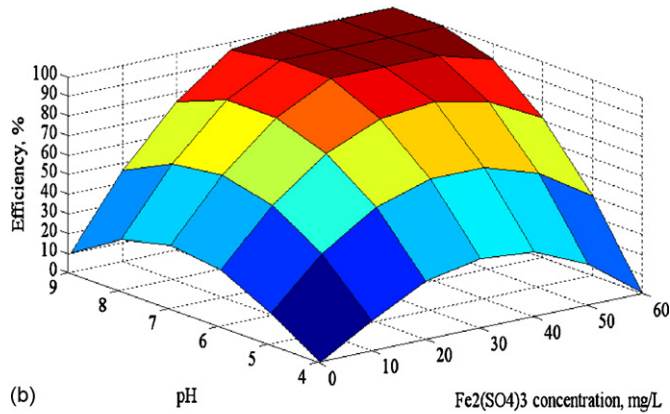
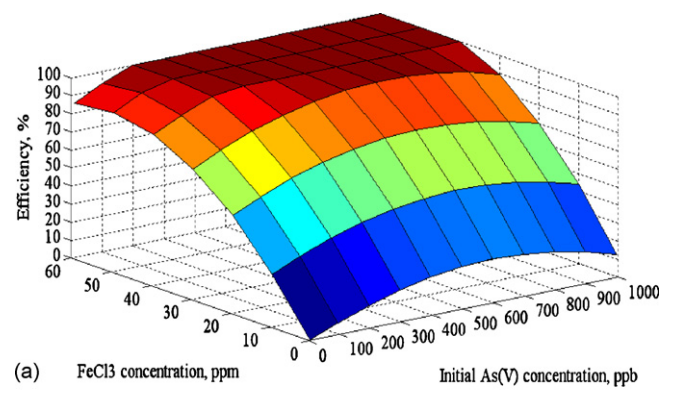
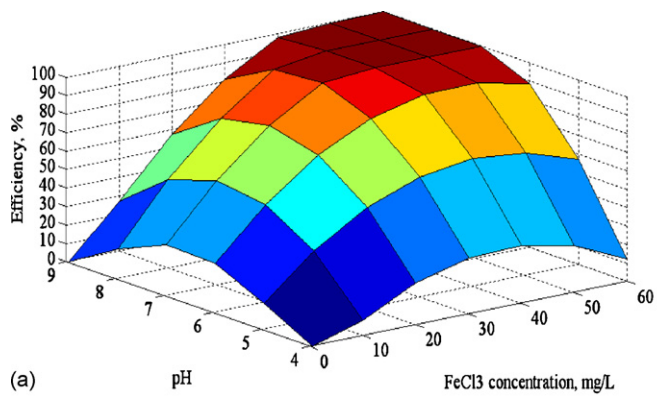


Fig. 1. Response surface plots showing effect of pH on arsenate removal efficiency with (a) ferric chloride and a constant initial arsenate concentration of $500 \mu\text{g L}^{-1}$; (b) ferric sulfate and a constant initial arsenate concentration of $500 \mu\text{g L}^{-1}$; (c) ferrous sulfate and a constant initial arsenate concentration of $500 \mu\text{g L}^{-1}$.

Percent arsenate removal increased with increasing pH content up to nearly 7.5, 8, and 9 for ferric chloride, ferric sulfate and ferrous sulfate, respectively. As it can be seen, in the high acidic and high alkaline pH for Fe(III) caused lower efficiencies. However, ferrous sulfate was effective in pH of 9 and form ferrous hydroxide in the alkaline range. The arsenate removal efficiency decreased at neutral and acidic pH values.

3.2. Effects of coagulants and initial As(V) concentrations

According to results of Box–Behnken experimental design method for ferrous sulfate (as a source of Fe(II)), ferric chloride and ferric sulfate (as a source of Fe(III)), optimum pH values for maximum arsenate removal were found as 7.5, 8 and 9. Fig. 2(a–c) shows the variations of percent arsenate removals with initial arsenate concentrations at different FeCl_3 , $\text{Fe}_2(\text{SO}_4)_3$ and FeSO_4 concentrations and constant pH of 7.5, 8 and 9, respectively.

Fig. 2. Three dimensional graph of variations of percent arsenate removals with initial arsenate concentrations at (a) different FeCl_3 concentrations and constant pH of 7.5; (b) different $\text{Fe}_2(\text{SO}_4)_3$ concentrations and constant pH of 8; (c) different FeSO_4 concentrations and constant pH of 9.

Pentavalent arsenic exists in anionic form of H_2AsO_4^- , HAsO_4^{2-} or AsO_4^{3-} above the pH of 2. Thus, addition of iron coagulants to water could facilitate the conversion of soluble arsenic species to insoluble reaction products [43]. These products might form through three major steps: (i) precipitation in forms of $\text{Fe}(\text{AsO}_4)$ solid; (ii) coprecipitation where soluble arsenic species were incorporated into a growing hydroxide phase via inclusion, occlusion, or adsorption; and (iii) adsorption involving the formation of surface complexes between soluble arsenic and the solid hydroxide surface site. In terms of arsenic removal from drinking water, however, precipitation may not contribute significantly toward the overall performance. This is because thermodynamics analysis on Fe and As has suggested that $\text{Fe}(\text{AsO}_4)$ solid could be easily formed in arsenic contaminated drinking water source [42,43].

It was found in this study that the addition of coagulant caused a substantial increase in the arsenate removal because the addition of coagulants facilitate the removal arsenate from the aqueous stream,

Table 8
Obtained highest arsenate removal efficiencies in the optimum pH and the optimum coagulant dose.

Coagulant type	Optimum pH	Initial As(V) concentrations					
		10 $\mu\text{g L}^{-1}$		500 $\mu\text{g L}^{-1}$		1000 $\mu\text{g L}^{-1}$	
		Dose (mg L^{-1})	Efficiency (%)	Dose (mg L^{-1})	Efficiency (%)	Dose (mg L^{-1})	Efficiency (%)
FeCl ₃	7.5	50	88	31	100	37	100
Fe ₂ (SO ₄) ₃	8	40	70	28	100	32	100
FeSO ₄	9	60	63	60	91	58	100

Table 9
Comparison of the experimental and predicted percent arsenate removal for different experimental points.

Coagulant type	As(V) concentration ($\mu\text{g L}^{-1}$)	Coagulant dose (mg L^{-1})	pH	Efficiency (%)	
				Observed	Predicted
FeCl ₃	50	30	7.5	80	75
Fe ₂ (SO ₄) ₃	50	40	8	72	74
FeSO ₄	50	60	9	72	66

by converting the soluble As(V) species into insoluble products. But above a certain ferric chloride and ferric sulfate dosage, the increase in arsenate removal was not significant. FeCl₃ dose above 40 mg L^{-1} and Fe₂(SO₄)₃ dose above 30 mg L^{-1} slightly increased percent arsenate removal. For ferrous sulfate, maximum arsenate removal efficiency was obtained at maximum coagulant dose. Percent arsenate removal increased with increasing coagulant concentration as a result of increasing amount of coagulant with increasing rate or kinetics of particle aggregation and floc formation. Obtained results at optimum pH values of coagulants are in good agreement with the values given in literatures [5,10,11,37].

For the initial arsenate concentration of 10, 500 and 1000 $\mu\text{g L}^{-1}$, obtained highest arsenate removal efficiencies in the optimum pH and the optimum coagulant dose are shown in Table 8. As it can be seen Fe(III) ions are more effective and economic than Fe(II) ion due to required lower coagulant dose and pH.

In the low initial arsenate concentrations, the highest arsenate removal efficiency was required high coagulant dose because of the difficulty in inducing collision between the colloids. However in the high initial arsenate concentrations, the highest arsenate removal efficiency was provided at low coagulant dose. The coagulant required was relatively small due to higher collision probabilities of the colloids in the high arsenate concentrations.

Three experiments different from Box–Behnken experimental design points were carried out in order to test reliability of the response functions predictions. The results of experiments and Box–Behnken response functions predictions were compared in Table 9. The three experiments were within the range of independent variables but different from design points of Box–Behnken. As it can be seen, response functions predictions were in good agreement with the experimental results. Therefore, Box–Behnken statistical design method was reliable and effective in determining the optimum conditions.

4. Conclusions

Precipitation/coprecipitation method was used for arsenate (As(V)) removal from tap water. Ferric chloride and ferric sulfate were used as source of Fe(III) and ferrous sulfate was used as source of Fe(II). A Box–Behnken statistical experiment design was used to determine the effects of initial arsenate concentration, coagulant dose and pH on arsenate removal efficiency. Arsenate concentration, coagulant dose and pH were chosen as independent variables. The objective functions were the arsenate removal efficiencies for three types of coagulants. A Box–Behnken statistical experiment design and response surface methodology were effective in determining the optimum conditions for arsenate removal by

coagulation–flocculation method using ferric chloride, ferric sulfate and ferrous sulfate. Therefore it could be employed to determine the optimum conditions for arsenate removal while minimising the number of experiments required.

Experimental results demonstrated that the Fe(III) ions are more effective for arsenate removal. Optimum pH values for maximum arsenate removal for ferric chloride, ferric sulfate and ferrous sulfate were found as 7.5, 8 and 9. At the constant pH of 7.5, 8 and 9, percent arsenate removal was increased with addition of coagulant as a result of floc formation and surface area. Decreasing initial arsenate concentration caused increases in required coagulant dose due to collision between the colloids. At the initial arsenate concentration of 10 $\mu\text{g L}^{-1}$, percent arsenate removal was reached a maximum level at FeCl₃, Fe₂(SO₄)₃ and FeSO₄ concentrations of 50, 40 and 60 mg L^{-1} , respectively. For the initial arsenate concentration of 500 and 1000 $\mu\text{g L}^{-1}$, percent arsenate removal efficiencies were obtained 100% at the FeCl₃ concentrations of 31 and 37 mg L^{-1} , Fe₂(SO₄)₃ concentrations of 28 and 32 mg L^{-1} and FeSO₄ concentrations of 60 and 58 mg L^{-1} . It was found that in almost all cases, the arsenic concentration can be reducing down to 10 $\mu\text{g L}^{-1}$ from initial concentrations of 1000 $\mu\text{g L}^{-1}$.

Acknowledgements

This study was supported by the Izmir Environmental Protection Foundation and Scientific Research Projects of the Dokuz Eylul University, Izmir, Turkey under grant number of 2005.KB.FEN.003.

References

- [1] P.L. Smedley, D.G. Kinniburgh, A review of the source, behaviour and distribution of arsenic in natural waters, *Appl. Geochem.* 17 (2002) 517–568.
- [2] C.M. Steinmaus, Y. Yuan, A.H. Smith, The temporal stability of arsenic concentrations in well water in western Nevada, *Environ. Res.* 99 (2005) 164–168.
- [3] C.F. Harvey, K.N. Ashfaq, W. Yu, A.B.M. Badruzzaman, M.A. Ali, P.M. Oates, H.A. Michael, R.B. Neumann, R. Beckie, S. Islam, M.F. Ahmed, Groundwater dynamics and arsenic contamination in Bangladesh, *Chem. Geol.* 228 (2006) 112–136.
- [4] R. Bhattacharyya, D. Chatterjee, B. Nath, J. Jana, G. Jacks, M. Vahter, High arsenic groundwater: mobilization, metabolism and mitigation—an overview in the Bengal Delta Plain, *Mol. Cell. Biochem.* 253 (2003) 347–355.
- [5] S.R. Wickramasinghe, B. Han, J. Zimbron, Z. Shen, M.N. Karim, Arsenic removal by coagulation and filtration: comparison of groundwaters from the United States and Bangladesh, *Desalination* 169 (2004) 231–244.
- [6] P. Mondal, C.B. Majumder, B. Mohanty, Laboratory based approaches for arsenic remediation from contaminated water: recent developments, *J. Hazard. Mater.* B137 (2006) 464–479.
- [7] A.M. Sancha, Review of coagulation technology for removal of arsenic: case of Chile, *J. Health Popul. Nutr.* 24 (2006) 267–272.
- [8] S. Wang, C.N. Mulligan, Occurrence of arsenic contamination in Canada: sources, behaviour and distribution, *Sci. Tot. Environ.* 366 (2006) 701–721.
- [9] S.S. Farias, V.A. Casa, C. Vazquez, L. Ferpozzi, G.N. Pucci, I.M. Cohen, Natural contamination with arsenic and other trace elements in groundwaters of Argentina Pampean Plain, *Sci. Tot. Environ.* 309 (2003) 187–199.

- [10] B. Han, T. Runnells, J. Zimbron, R. Wickramasinghe, Arsenic removal from drinking water by flocculation and microfiltration, *Desalination* 145 (2002) 293–298.
- [11] T. Yuan, Q. Luo, J. Hu, S. Ong, W. Ng, A study on arsenic removal from household drinking water, *J. Environ. Sci. Health A38* (2003) 1731–1744.
- [12] J. Gregor, Arsenic removal during conventional aluminium-based drinking water treatment, *Water Res.* 35 (2001) 1659–1664.
- [13] D. Chakraborti, S.C. Mukherjee, S. Pati, M.K. Sengupta, M.M. Rahman, U.K. Chowdhury, D. Lodh, C.R. Chanda, A.K. Chakraborti, G.K. Basu, Arsenic groundwater contamination in Middle Ganga Plain, Bihar, India: a future danger, *Environ. Health Perspect.* 111 (2003) 1194–1201.
- [14] A.A. Duker, E.J.M. Carranza, M. Hale, Arsenic geochemistry and health, *Environ. Int.* 31 (2005) 631–641.
- [15] T. Yoshida, H. Yamauchi, G.F. Sun, Chronic health effects in people exposed to arsenic via the drinking water: dose-response relationships in review, *Toxicol. Appl. Pharmacol.* 198 (2004) 243–252.
- [16] V.M. Rodriguez, M.E.J. Capdeville, M. Giordano, The effects of arsenic exposure on the nervous system, *Toxicol. Lett.* 145 (2003) 1–18.
- [17] Y. Xia, J. Liu, An overview on chronic arsenism via drinking water in PR China, *Toxicology* 198 (2004) 25–29.
- [18] M.M. Rahman, M.K. Ahamed, U.K. Chowdhury, A. Hossain, B. Das, D. Lodh, K.C. Saha, S. Pati, I. Kaies, A.K. Barua, D. Chakraborti, The magnitude of arsenic contamination in groundwater and its health effects to the inhabitants of the Jalangi—one of the 85 arsenic affected blocks in West Bengal, India, *Sci. Tot. Environ.* 338 (2005) 189–200.
- [19] S.C. Mukherjee, M.M. Rahman, U.K. Chowdhury, M.K. Sengupta, D. Lodh, C.R. Chanda, K.C. Saha, D. Chakraborti, Neuropathy in arsenic toxicity from groundwater arsenic contamination in West Bengal, India, *J. Environ. Sci. Health* 1 (2003) 165–183.
- [20] M. Ali, S.A. Tarafdar, Arsenic in drinking water and in scalp hair by EDXRF: a major recent health hazard in Bangladesh, *J. Radioanal. Nucl. Chem.* 256 (2003) 297–305.
- [21] D.N.G. Mazumder, Chronic arsenic toxicity: clinical features, epidemiology, and treatment: experience in West Bengal, *J. Environ. Sci. Health* 1 (2003) 141–163.
- [22] S.C. Mukherjee, K.C. Saha, S. Pati, R.N. Dutta, M.M. Rahman, M.K. Sengupta, S. Ahamed, D. Lodh, B. Das, M.A. Hossain, B. Nayak, A. Mukherjee, D. Chakraborti, S.K. Dulta, S.K. Palit, I. Kaies, A.K. Barua, K.A. Asad, Murshidabad-one of the nine groundwater arsenic-affected districts of West Bengal, India. Part II: dermatological, neurological, and obstetric findings, *Clin. Toxicol.* 43 (2005) 835–848.
- [23] O.S. Ehrenstein, D.N.G. Mazumder, Y. Yuan, S. Samanta, J. Balmes, A. Sil, N. Ghosh, M. Hira-Smith, R. Haque, R. Purushothamam, S. Lahiri, S. Das, A.H. Smith, Decrements in lung function related to arsenic in drinking water in West Bengal, India, *Am. J. Epidemiol.* 162 (2005) 533–541.
- [24] R.R. Shrestha, M.P. Shrestha, N.P. Upadhyay, R. Pradhan, R. Khadka, A. Maskey, M. Maharjan, S. Tuladhar, B.M. Dahal, K. Shrestka, Groundwater arsenic contamination, its health impact and mitigation program in Nepal, *J. Environ. Sci. Health* 1 (2003) 185–200.
- [25] A. Kelepertis, D. Alexakis, K. Skordas, Arsenic, antimony and other toxic elements in the drinking water of eastern Thessaly in Greece and its possible effects on human health, *Environ. Geol.* 50 (2006) 76–84.
- [26] M.R. Karagas, T.A. Stukel, T.D. Tosteson, Assessment of cancer risk and environmental levels of arsenic in New Hampshire, *Int. J. Hygiene Environ. Health* 205 (2002) 85–94.
- [27] D.D. Caceres, P. Pino, N. Montesinos, E. Atalah, H. Amigo, D. Loomis, Exposure to inorganic arsenic in drinking water and total urinary arsenic concentration in a Chilean population, *Environ. Res.* 98 (2005) 151–159.
- [28] Y. Lee, I. Um, J. Yoon, Arsenic(III) oxidation by iron(VI) (ferrate) and subsequent removal of arsenic(V) by iron(III) coagulation, *Environ. Sci. Technol.* 37 (2003) 5750–5756.
- [29] WHO, Guidelines for drinking-water quality, World Health Organization, Geneva, 1993.
- [30] M. Çolak, Ü. Gemic, G. Tarcan, The effects of colemanite deposits on the arsenic concentrations of soil and groundwater in Iğdekoy-Emet, Kutahya, Turkey, *Water Air Soil Pollut.* 149 (2003) 127–143.
- [31] M. Çöl, C. Çöl, Arsenic concentrations in the surface, well and drinking waters of the Hisarcik, Turkey, area, *Hum. Econ. Risk Assess.* 10 (2004) 461–465.
- [32] Ü. Gemic, G. Tarcan, Hydrogeological and hydrogeochemical features of the Heybeli spa, Afyon, Turkey: arsenic and the other contaminants in the thermal waters, *Bull. Environ. Contam. Toxicol.* 72 (2004) 1107–1114.
- [33] R. Johnston, H. Heijnen, P. Wurzel, Safe water technology, in: United Nations Synthesis Report on Arsenic in Drinking Water, World Health Organization, Geneva, 2001 (Chapter 6).
- [34] M. Fujimoto, The removal of arsenic from drinking water by carbon adsorption, Master of Science, Department of Civil and Environmental Engineering, Michigan State University, 2001.
- [35] M. Kang, H. Chen, Y. Sato, T. Kamei, Y. Magara, Rapid and economical indicator for evaluating arsenic removal with minimum aluminum residual during coagulation process, *Water Res.* 37 (2003) 4599–4604.
- [36] S. Song, A. Lopez-Valdivieso, D.J. Hernandez-Campos, C. Peng, M.G. Monroy-Fernandez, Arsenic removal from high-arsenic water by enhanced coagulation with ferric ions and coarse calcite, *Water Res.* 40 (2006) 364–372.
- [37] R. Kammoun, B. Naili, S. Bejar, Application of a statistical design to the optimization of parameters and culture medium for a-amylase production by *Aspergillus oryzae* CBS 819.72 grown on gruel (wheat grinding by-product), *Bioresour. Technol.* 99 (2008) 5602–5609.
- [38] T. Guerin, N. Molenat, A. Astruc, R. Pinel, Arsenic speciation in some environmental samples: a comparative study of HG-GC-QFAAS and HPLC-ICP-MS methods, *App. Organomet. Chem.* 14 (2000) 401–410.
- [39] A. Eaton, H.C. Wang, J. Nothington, Analytical Chemistry of Drinking Water, AWWA Research Foundation, 1998.
- [40] D. Montgomery, Design and Analysis of Experiments, third ed., John Wiley & Sons, NY, 1991.
- [41] M. Sullivan, Statistics Informed Decisions Using Data, Pearson Education, New Jersey, 2004.
- [42] M. Edwards, Chemistry of arsenic removal during coagulation and Fe–Mn oxidation, *J. Am. Water Works. Assoc.* 86 (1994) 79–90.
- [43] L.S. McNeill, M. Edwards, Predicting As removal during metal hydroxide precipitation, *J. Am. Water Works. Assoc.* 89 (1997) 75–86.