

Predictive model for drinking water treatment technology design – the efficiency of arsenic removal by in-situ formed ferric-hydroxide

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Received 2009-07-03, accepted 2009-10-05

Abstract

Coagulation by ferric-salt is a widely used technique for arsenic removal. Jar tests with synthetic water were conducted in order to investigate the combined effect of the raw water characteristics on the removal efficiency of in-situ formed ferric-hydroxide. Multiple linear regression analysis was carried to establish a robust model, which is able to estimate the residual arsenic concentration provided that the raw water contained 50–60 µg/L initial arsenic concentration. The estimation was based on the following variables: PO_4-P concentration, final pH, SiO_2 concentration and Fe^{3+} dose. The major factors influencing efficacy were the silicate concentration and applied coagulant dosage. The model was verified by data from 24 additional experiments, and the predicted and measured residual arsenic concentrations showed good agreement ($R^2 = 0.87$).

Keywords

Arsenic removal · Coagulation · Drinking water · Jar-test · Linear regression analysis · Silicate

Acknowledgement

The research work was supported by the Economic Competitiveness Operative Program (EU Co-funding system, project number: GVOP 3.1.1.-2004-05-0186/3.0). The authors would like to thank to HAJDÚVÍZ Co. for the arsenic analysis and to Péter Tomka (environmental engineering student at the Budapest University of Technology and Economics) for his kind assistance in the laboratory experiments.

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1 Introduction

Studies on long-term human exposure have shown that arsenic in drinking water is associated with liver, lung, kidney, bladder cancers and skin cancers as well [21]. The main source of arsenic in drinking water is arsenic-rich aquifer rocks in which the water is stored. It may also occur because of mining or industrial activities in some areas [19]. Arsenic poisoning due to excessive exposure to natural and anthropogenic arsenic in drinking water has been reported in Bangladesh, Argentina, China, Taiwan, Thailand, India, Mexico, USA, Ghana, Hungary, Romania, United Kingdom, Chile, New Zealand and Vietnam [16].

Inorganic arsenic can occur in the environment in several forms but in natural waters it is mostly found as trivalent arsenite (As(III)) or pentavalent arsenate (As(V)). Organic arsenic species, abundant in seafood, are very much less harmful to health, and are readily eliminated by the body [19].

In Hungary arsenic is found in deep confined aquifers of exclusively natural origin. As a consequence of the reductive conditions in these layers, arsenic is mostly found as arsenite (As(III)). Since the arsenic removal techniques are usually more efficient for arsenate, pre-oxidation is required prior to arsenic removal.

In 2001 new drinking water standards were introduced in Hungary, which lowered the earlier enforced 50 µg/L arsenic standard to 10 µg/L, which thereby corresponds to the 98/83 EU Directive. This new regulation affects around 1.5 million consumers at about 400 settlements [15]. At several water treatment plants, new technology has to be installed, or the existing one should be modified in order to meet the new maximum allowable concentration.

The conventional coagulation-flocculation method is capable of decreasing the arsenic concentration ([1, 4, 20]). The technology consists of three steps: the oxidation of arsenite (As(III)) to arsenate (As(V)), the conversion of soluble arsenate to insoluble form by addition of metal salts (usually iron(III) or aluminum(III)), and then the removal of the solid particles from the water by sedimentation, rapid sand filtration or microfiltration [5, 6, 20].

The liquid/solid transition of arsenic is carried out through co-precipitation and adsorption [3], and there are several water quality parameters, which effect this transition process, e.g., type and dosage of coagulant, oxidation state of arsenic, type and dosage of oxidant, pH [14], organic material content [8], phosphorous [7, 12], carbonate/bicarbonate [7] and silicate concentration [11, 18] of the water to be treated.

Inorganic carbon (H_2CO_3 , HCO_3^- and CO_3^{2-}) is found in all natural waters. Due to the geochemical conditions in Hungary, total inorganic carbon concentration is often around 10 mM. When iron or aluminum coagulants are added to the water – due to hydrolyses – metal-hydroxides are formed if the buffering capacity of the water is high enough. Therefore the carbonate/bicarbonate system plays an essential role in the coagulation process [17] and moreover the inorganic carbon concentration influences the pH after coagulation. The relatively high buffering capacity of the Hungarian subsurface waters ensures the complete transformation of the metal ions to metal-hydroxides [10]. On the other hand, research work by Holm [7] has demonstrated that 1 mM and 10 mM inorganic carbon concentration already has adverse effect on arsenic removal, since there is a competition between carbonate/bicarbonate and arsenic for the adsorption sites of ferric-hydroxide.

The Hungarian subsurface water sources may contain PO_4^{3-} ions as well, up to 0.8–1.0 mg/L concentration. Since the structure of arsenate and phosphate ions is similar, and they have similar sorption behavior [2], there is a competition between these ions for the adsorption sites at ferric-hydroxide [7, 12]. Moreover, the PO_4^{3-} concentrations in groundwater can be 10–100 times greater than As. Holm [7] found that there was significantly less As(V) sorption at all pH values for systems containing ~ 0.9 mg/L PO_4^{3-} compared to PO_4^{3-} -free systems.

Research work has been demonstrated that silicate can also significantly inhibit arsenic removal with ferric hydroxides [11, 13, 18]. According to Liu et al. [11] the adsorption and surface complexation of silicate on ferric hydroxide results in decreased ζ potential. More significant ζ potential decrease was observed at elevated pH conditions, however the presence of Ca^{2+} could buffer the adverse effects of silicate to some extent. Decreasing ζ potential increases the repulsive forces between the ferric hydroxide precipitates, which inhibits further particle agglomeration and therefore these small ferric-hydroxide colloids are able to pass through 0.45 μm pore-size membrane filters. Moreover, arsenate could not be associated with these small flocs.

Natural organic matter (NOM) also has significant effect on the arsenic removal processes [8]. There was even a difference of one order of magnitude in the required coagulant dosages (to reach the 10 $\mu\text{g/L}$ standard arsenic concentration) when the results of low ($\text{COD}_{\text{Mn}} \sim 1$ mg/L) and high ($\text{COD}_{\text{Mn}} \sim 13$ mg/L) organic content waters were compared [8, 9].

In spite of the large number of water quality parameters affecting arsenic removal efficiency, operating experts and design

engineers often discuss about the needed arsenic:metal ratio in order to achieve the 10 $\mu\text{g/L}$ concentration value, or about arsenic removal efficiency for a given type of coagulant. In order to point out the deficiency of such general considerations (often recommendations) the goal of the present research was to build up a very simple and robust model, which is able to predict the residual arsenic concentration as a function of the raw water quality and coagulant dose. Another aim was to define the most important raw water characteristics with respect to technological optimization.

Based on literature data [8] and also previous experiments [9] it is assumed that one of the most important influencing factors is the concentration of NOM. However, the important role of NOM is not addressed in this study. The main focus points of the present research were the phosphate, silicate and inorganic carbon concentration, pH and coagulant dose.

In Hungary arsenic is mostly found in the trivalent (As(III)) form, therefore the first step is pre-oxidation if the coagulation technology is applied for arsenic removal. In this study the different types of oxidants are not evaluated, the experiments were carried out with the pentavalent (As(V)) form, which means that in practice the results are applicable only after the oxidation process.

2 Methods and materials

2.1 Materials and reagents

All reagents used (except the Na_2SiO_3 solution) were of analytical grade. Ferric-salt solution was prepared from $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and nitric-acid was added to the solution in order to prevent the hydrolysis of Fe(III). Na_2SiO_3 stock solution containing 3.60 g/L SiO_2 was freshly prepared every day. Arsenate stock solution containing 50 mg/L As was prepared from $\text{Na}_2\text{HASO}_4 \cdot 7\text{H}_2\text{O}$. NaHCO_3 solution of 0.50 mol/L concentration was prepared and used. 50 mg/L $\text{PO}_4\text{-P}$ stock solution was prepared from KH_2PO_4 .

2.2 Experimental methods

Laboratory experiments were conducted with different types of model water made of de-ionized water or tap water of Budapest. The temperature was 22–27 $^\circ\text{C}$ during the laboratory experiments. In model systems made by de-ionized water the ionic strength was set to 0.005 M by addition of KCl. Jar-tests were performed in the following way: the water was introduced into 1 L beakers, and coagulant was added with rapid mixing (400 rpm) for 1 minute, followed by slow mixing (20 rpm) for 10 minutes and settling for 20 minutes. To evaluate the efficiency of arsenic removal, samples were taken from the clarified water, and then they were filtered through 0.45 μm pore size membrane. Arsenic concentration was measured from the membrane-filtered water.

2.3 Analytical methods

The arsenic concentration was measured by atomic absorption method (Perkin-Elmer Analyst 800 atomic absorption spectrophotometer).

3 Results and discussion

3.1 Linear regression analysis

First, the individual influencing mechanisms of several water quality parameters (pH, alkalinity, phosphate, silicate concentration and coagulant dose) were studied. Above 1 mmol/L NaHCO_3 , the adverse effect of bicarbonate was observed under the studied conditions, and resulted in higher remaining arsenic concentration. Also, 0.15 mg/L phosphate already had adverse effect on arsenic removal efficiency. At all pH values (7.5–7.8) and coagulant dosages (0.84–3 mg/L Fe) tested, the negative effect of phosphate on arsenic removal was evident. Silicate was also found to be an important influencing factor: as a result of silicate in the water, small ferric-hydroxide colloids were formed, which were able to go through the 0.45 μm pore-size membrane. By increasing the coagulant dose, the target arsenic concentration was finally achieved, however compared to the results with silicate-free systems, around 2.5–3.5 times higher ferric salt dose was needed to reach this value (in case of 30–50 mg/L SiO_2 content). The influence of silicate depended on the pH as well: at higher pH values the adverse effect of silicate was more significant.

After evaluating the effect of these components individually, overall analysis was carried out to see the aggregate effect of them. Multiple linear regression analysis was applied to determine which factors have the greatest influence on the arsenic removal efficiency and to build up a model to predict arsenic removal efficiency. Only data from experiments carried out with synthetic water made of de-ionized water were used. Only in-situ floc formation was studied, therefore the data from the experiments with pre-formed flocs were excluded. Moreover, results of experiments with low alkalinity (< 0.125 mmol/L NaHCO_3) were excluded, since under these circumstances the hydroxide formation is not sufficient enough. Finally 125 datasets were available for the analysis, and the following variables were applied: $\text{PO}_4\text{-P}$ concentration (0.0–0.66 mg/L), NaHCO_3 concentration (0.125–10.0 mmol/L), final pH values (pH = 5.7–8.3), SiO_2 (0–60 mg/L), Fe dose (0.50–8.20 mg Fe/L). The initial arsenic concentration was excluded as a variable, since its variation was low during the experiments (50–60 $\mu\text{g/L}$).

By carrying out the first regression analysis, it has been shown that NaHCO_3 has basically no effect on the remaining arsenic concentration. This indicates that HCO_3^- has indirect effect on arsenic removal through affecting the pH after the treatment. Higher HCO_3^- concentration results in higher final pH values, which means higher remaining arsenic concentrations. Therefore in the second analysis the NaHCO_3 concentration was excluded as a variable. The multiple linear regression analysis re-

sulted in the following equation for estimating the residual dissolved arsenic concentration:

$$\text{ResidualAs}[\mu\text{g/L}] = -94.44 + 31.14 \cdot \text{PO}_4 - \text{P}[\text{mg/L}] + 14.71 \cdot \text{pH} + 0.55 \cdot \text{SiO}_2[\text{mg/L}] - 5.80 \cdot \text{Fe}[\text{mg/L}] \quad (1)$$

The formula reflects the earlier findings: higher residual arsenic with increasing $\text{PO}_4\text{-P}$ and SiO_2 concentration, better removal efficiency at lower pH values and with higher coagulant dosage.

The difference between the calculated and measured values was determined for the 125 data (Fig. 1). From the frequency of the error, the 90% and 95% confidence intervals were calculated and they are presented on Fig. 1.

The total correlation (ρ_{As}) and partial correlation values (ρ_{Si} , ρ_{PO_4} , ρ_{pH} , ρ_{Fe}) are as follows:

- $\rho_{\text{As}} = 0.86$
- $\rho_{\text{Si}} = 0.79$
- $\rho_{\text{PO}_4} = 0.52$
- $\rho_{\text{pH}} = 0.59$
- $\rho_{\text{Fe}} = -0.70$

The relatively high total correlation value (0.86) indicates that there is a strong correlation between the remaining arsenic concentration and the studied water quality parameters. The two most important influencing factors were found to be the silicate concentration ($\rho_{\text{Si}} = 0.79$) and the applied coagulant dosage ($\rho_{\text{Fe}} = -0.70$) since these two variables had the highest partial correlation value.

According to Liu et al. [11] the presence of Ca^{2+} could buffer the adverse effects of silicate to some extent. Further research is needed to investigate this buffering effect in detail and to work out how to build this information into the model.

3.2 Model verification

The investigated formula was verified by 24 independent additional experiments, in which different types of initial water qualities were applied (synthetic waters from de-ionized water). The calculated and measured remaining arsenic concentration values were in good agreement ($R^2 = 0.87$; Fig. 2).

3.3 Defining the final pH

From the chemical point of view, the final pH value is more important than the initial pH, however in practice, the initial pH is the known parameter. In the suggested model the final pH is a variable, though this value depends on the alkalinity, initial pH and coagulant dose. Based on these parameters, the final pH can be defined by preliminary experiments, by a water quality modeling software or by equilibrium calculations. In this paper the calculation method is presented briefly.

The alkalinity of the water mostly consists of carbon alkalinity; albeit silicate and phosphate content also contributes to the buffering capacity of the water.

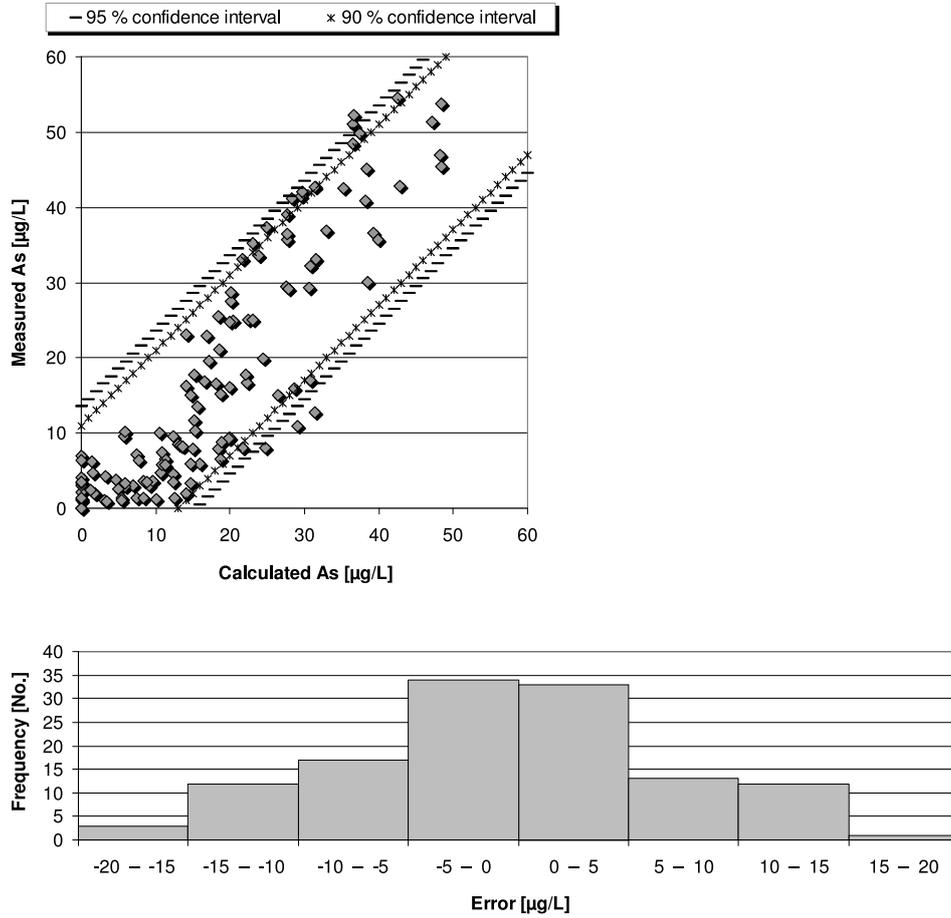


Fig. 1. Difference between the modeled and measured arsenic concentration values

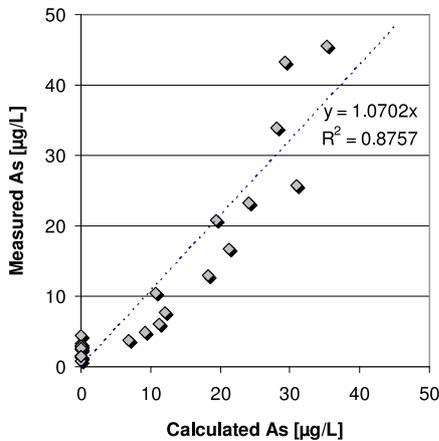


Fig. 2. Comparison of the modeled and measured arsenic concentration values – the verification process

The percentage of H_2CO_3 , HCO_3^- and CO_3^{2-} can be calculated as a function of pH from the following equations and equilibrium constants.



If pH_1 , $[H_2CO_3]_1$, $[HCO_3^-]_1$ and $[CO_3^{2-}]_1$ denote the initial pH and initial inorganic carbon concentration values and pH_2 , $[H_2CO_3]_2$, $[HCO_3^-]_2$ and $[CO_3^{2-}]_2$ indicate the values after the coagulation process, the task is to define pH_2 as a function of the

known parameters: pH_1 , $[H_2CO_3]_1$, $[HCO_3^-]_1$, $[CO_3^{2-}]_1$ and the applied coagulant dose.

During coagulation, the dosed Fe(III) transforms to Fe(III)-hydroxide and due to this process, H^+ ions are released into the water. If the transformation is complete, 1 mole Fe(III) releases 3 moles H^+ ion into the water. The released H^+ ions change the pH and the concentration values of the different inorganic carbon forms. If $\Delta[CO_3^{2-}]$, $\Delta[H_2CO_3]$ and $\Delta[H^+]$ denote the changes, the equations are as follows:

$$\Delta[CO_3^{2-}] = [CO_3^{2-}]_2 - [CO_3^{2-}]_1 \quad (4)$$

$$\Delta[H_2CO_3] = [H_2CO_3]_2 - [H_2CO_3]_1 \quad (5)$$

$$\Delta[H^+] = [H^+]_2 - [H^+]_1 \quad (6)$$

If $[Fe-H^+]$ denote the moles of H^+ ions released to the water (which is three times of the dosed coagulant in case of complete hydrolysis), then the capability of changing the different inorganic carbon concentration values can be described by the following equation:

$$[Fe - H^+] = \Delta[H^+] + \Delta[H_2CO_3] + \Delta[CO_3^{2-}] \quad (7)$$

Combining Eq. (7) with Eqs. (4)-(6):

$$[Fe - H^+] = [H^+]_2 - [H^+]_1 + [H_2CO_3]_2 - [H_2CO_3]_1 + [CO_3^{2-}]_2 - [CO_3^{2-}]_1 \quad (8)$$

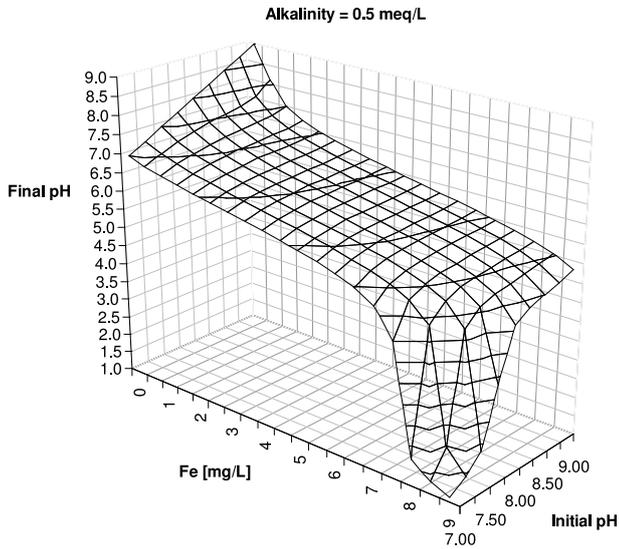


Fig. 3. Final pH as a function of initial pH and coagulant dose (alkalinity = 0.5 meq/L)

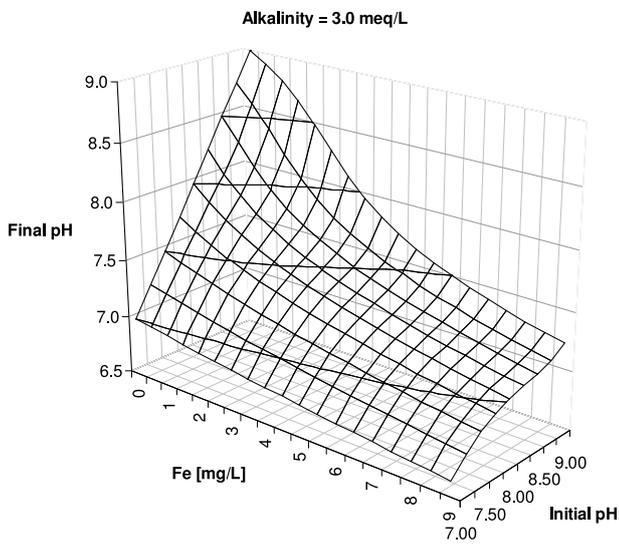


Fig. 4. Final pH as a function of initial pH and coagulant dose (alkalinity = 3.0 meq/L)

Without going into the details the combination of Eqs. (4) – (8) with the equilibrium Eqs. (2)–(3) results in the following equation:

$$0 = [\text{Fe} - \text{H}^+] - [\text{H}^+]_2 + [\text{H}^+]_1 + [\text{H}_2\text{CO}_3]_1 - \frac{[\text{NaHCO}_3]}{1 + \frac{k_1}{[\text{H}^+]_2} + \frac{k_1 \cdot k_2}{[\text{H}^+]_2^2}} - [\text{CO}_3^{2-}]_1 + \frac{k_1 \cdot k_2 \cdot [\text{NaHCO}_3]}{1 + \frac{k_1}{[\text{H}^+]_2} + \frac{k_1 \cdot k_2}{[\text{H}^+]_2^2}} + \frac{[\text{H}^+]_2^2}{[\text{H}^+]_2^2} \quad (9)$$

Where $[\text{NaHCO}_3]$ indicates the total concentration of the different inorganic carbon forms:

$$[\text{NaHCO}_3] = [\text{H}_2\text{CO}_3]_2 + [\text{HCO}_3^-]_2 + [\text{CO}_3^{2-}]_2 \quad (10)$$

From Eq. 9 $[\text{H}^+]_2$ can be calculated, which can be easily converted to pH_2 . In this study only two graphs are presented: from

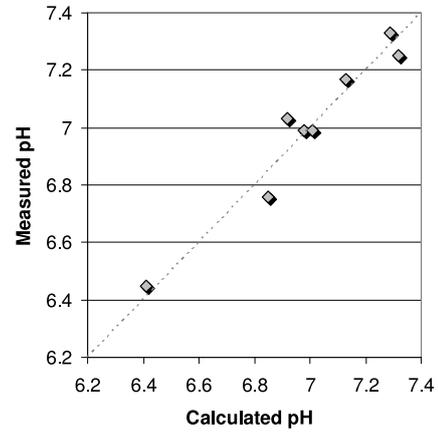


Fig. 5. Comparison of the calculated and measured final pH values

Figs. 3–4 the final pH can be determined as a function of Fe dose and initial pH, in case of 0.5 meq/L and 3.0 meq/L alkalinity. It is important to note that due to the geochemical conditions in Hungary, total inorganic carbon concentration is relatively high (usually higher than 3 meq/L and it often reaches even 10 meq/L), which means that the difference between the initial and final pH values are not that significant if the coagulation technology is applied.

Theoretically silicate and phosphate can also contribute to the alkalinity of the water. However, in the pH range usually applied in drinking water treatment (7.0–9.0) the buffering capability of silicate is not that significant. Phosphate could also contribute to alkalinity, but the typical concentration values are much lower than the typical inorganic carbon concentration values (usually there is a difference of two orders of magnitudes); therefore the buffering ability of phosphate is negligible. Additional laboratory experiments were carried out in order to compare the calculated and measured pH with model systems containing inorganic carbon, phosphate and silicate. The measured and calculated pH values were in good agreement, which means that the presented calculation method can sufficiently define the final pH value (Fig. 5).

3.4 Effect of silicate

In the experiments carried out with silicate-rich systems it was observed that the water could be filtered easily through the 0.45 μm pore-size membrane. However, as the amount of filtrate increased, the filtration process slowed down significantly. It was also noticed that the small ferric-hydroxide colloids (which are formed due to the presence of silicate) are able to form a layer (“cake”) on the membrane, which has additional filtration effect causing significantly lower effluent concentrations after that a certain amount of water already passed through the filter. This phenomenon resulted in high variation in the arsenic concentration of the filtrate. To enhance the comparability of the results, the volume of sample to be filtered through the 0.45 μm pore-size membrane was fixed to 120 mL. The first 20 mL of the filtrate was abandoned to ensure the data being reliable.

Linear regression analysis was carried out by excluding the

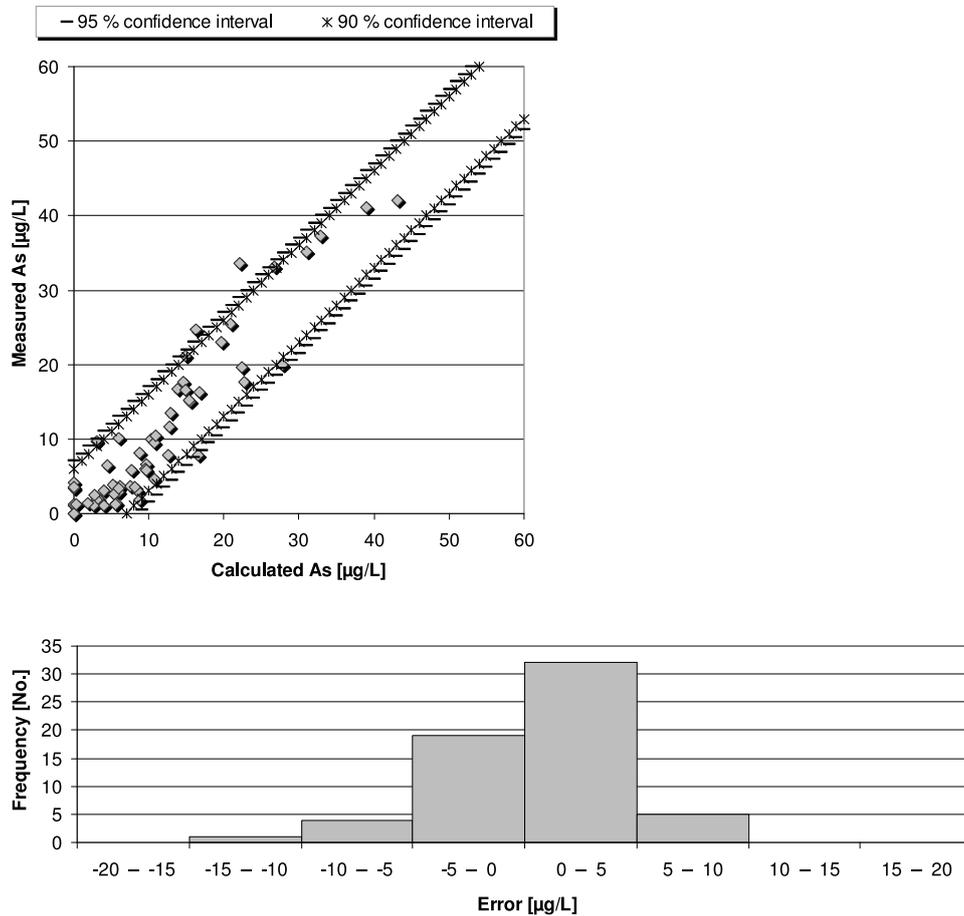


Fig. 6. Difference between the modeled and measured arsenic concentration values in silicate-free waters

results with silicate-rich systems in order to see whether the model gives better estimation. 61 datasets were available for the analysis, which resulted in the following equation:

$$\text{ResidualAs}[\mu\text{g/L}] = -60.36 + 57.10 \cdot \text{PO}_4 - \text{P}[\text{mg/L}] + 10.61 \cdot \text{pH} - 10.10 \cdot \text{Fe}[\text{mg/L}] \quad (11)$$

The total correlation was 0.92, which indicates strong correlation between the variables and the residual arsenic concentration.

The difference between the calculated and measured values was determined for the 61 datasets (Fig. 6). From the frequency of the error, the 90% and 95% confidence intervals were calculated and they are presented at Fig. 6. The confidence intervals were narrower, and the difference between the modeled and measured residual arsenic concentration values was smaller compared to the previous case (when the dataset included the silicate-rich waters as well).

These results indicate that in silicate-free systems the model can predict the residual arsenic concentration better. The reason is that if the water contains silicate, the size of the formed particles is mainly between 0.2 and 0.45 µm, which causes high variation in the arsenic concentration in the 0.45 µm filtrate. On the other hand silicate is one of the most important influencing factors when the coagulation technology is applied for arsenic removal (see Eq. (1)), therefore if the water contains silicate, the

silicate concentration must be included in the model as a variable.

3.5 Practical applications of the model

The linear regression model proved to be a robust tool to predict the remaining arsenic concentration when the ferric-chloride coagulation technology was applied for arsenic removal and the initial arsenic concentration was around 50–60 µg/L. Since the final pH is built into the model as a variable, the first step is to define the final pH for a given coagulant dose, alkalinity and initial pH. After that the second step is to predict the remaining arsenic concentration by using Eq. (1). Since the model does not consider the concentration of NOM (which is an important influencing factor) it is only feasible in case of low organic carbon (< 2 mg/L COD_{Mn}) content. When the organic content is higher, the model overestimates the arsenic removal efficiency.

Since aim of the model is to give preliminary estimate, it cannot be applied directly to design the treatment technology. Nevertheless, it is a good starting point for planning the laboratory experiments, which can further give good basis for pilot experiments. The results of the pilot-scale experiments are directly adaptable for planning the full-scale treatment plants. It means that the technology planning (scale-up) should consist of the following three steps: modeling, laboratory experiments and then pilot experiments with the real water.

4 Summary

Data from experiments with de-ionized water system (containing arsenic, phosphate, silicate and inorganic carbon) were merged and multiple linear regression analysis was carried out in order to build up a robust model to estimate the residual arsenic concentration if the raw water contains 50 – 60 µg/L initial arsenate concentration. The estimation was based on the following variables: PO₄-P concentration, final pH, SiO₂ concentration and Fe dose. When the presented model is applied, the first step is the calculation of the final pH value, which depends on the initial pH, alkalinity and the applied coagulant dose.

The investigated formula was verified by 24 independent additional experiments, in which different types of initial water qualities were applied (synthetic waters from de-ionized water). The calculated and measured remaining arsenic concentration values were in good agreement ($R^2 = 0.87$). The most important influencing factors proved to be the silicate concentration and applied coagulant dosage.

The presented model is the first step in the design process, since it is a very simple method to make estimation about the efficiency of the technology. Further research is needed however, to investigate the role of NOM on arsenic removal efficiency and also to study the effect of Ca²⁺ in silicate-rich systems.

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