Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.959064

56 (2015) 2105–2113
November

Arsenic preoxidation and its removal from groundwater using iron coagulants

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Received 9 January 2014; Accepted 23 August 2014

ABSTRACT

Recent studies revealed the similar chemism and appearance of heightened arsenic concentrations (over 200 μg/L) in groundwaters of the Pannonian basin (Hungary, Croatia, Serbia, and Romania) due to the geological composition. These are representing significant health issues since those groundwaters are the main source of drinking water for the population in these areas. The consumption of arsenic-contaminated water causes chronic poisoning, which manifests as various toxic and carcinogenic effects in humans. Due to similar physicochemical characteristics of Pannonian groundwaters, naturally arsenic-contaminated groundwater from well field "Vinogradi" in the area of Osijek town was used to remove arsenic using different types of iron coagulants: $FeSO_4$, $Fe_2(SO_4)_3$, $FeCl_3$, and oxidants: $KMnO_4$ and H_2O_2. During the experiments, up to 97% arsenic was removed from used groundwater (average initial arsenic concentration of 204 μg/L). This paper attempts to contribute to long-term quality improvement of drinking water in the area of Pannonian basin, since the coagulation and flocculation is a simple and affordable drinking water treatment usually applied in water treatment plants in most countries of the Pannonian basin.

Keywords: Drinking water; Arsenic removal; Arsenic preoxidation; Iron removal; Coagulation and flocculation

1. Introduction

Within Europe, many countries have aquifers with levels of arsenic above the EU drinking water limit of 10 μg/L, requiring some form of remediation before waters reach consumers [1,2]. Estimates suggest that nearly one million people in Europe are exposed to naturally occurring arsenic in drinking waters at levels above 10 μg/L, which WHO guidelines and EU directive set as maximum acceptable concentration [1,3] and most of them are inhabitants of the Pannonian basin. Recentlyly, different studies reported arsenic concentrations in groundwater that has been used for

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human consumption, namely up to 210 μg/L in south Hungary [2,4,5], 240 μg/L in western Romania [2], 350 μg/L in northern Serbia [4,6–9], and 610 μg/L in eastern Croatia [10]. The biggest well field in the area of eastern Croatia has an average arsenic concentration of around 200 μg/L [11–13].

Arsenic has been considered as a strong poisonous chemical due to its odorless and tasteless nature [14]. Ingestion of heightened arsenic concentration via drinking water has been linked to cardiovascular diseases, reproductive problems and neurological disorders, lower birthweight, infant mortality, and occurrence of skin, liver, lung, kidney, and bladder cancer [3,14,15].

Groundwater of eastern Croatia is characterized by high water hardness and increased concentrations of inorganic contaminants such as iron, manganese, ammonium, organic matter, and arsenic that ranges from 10 to 610 μg/L [10,16]. Elevated concentrations of arsenic in groundwater of eastern Croatia are due to the geological composition of soil [17]. It is well known that arsenite (As(III)) and arsenate (As(V)) are common arsenic species in the environment, and As (III) is more predominant in groundwater [2–4].

There are several techniques for arsenic removal from drinking water and their efficiency is mostly dependent on the arsenic valence. Arsenic is most effectively removed or stabilized in the pentavalent arsenate form [15,18]. Pentavalent arsenic exists in the anionic form as $H_2AsO_4^-$, $HAsO_4^{2-}$, or AsO_4^{3-}, above pH 2 [4], and the main task of water treatment for arsenic removal is to convert arsenic(III) to arsenic(V) species [15]. Most of these techniques include the following processes: flocculation with microfiltration, adsorption on natural or synthetic materials, ion exchange, microfiltration, ultrafiltration, nanofiltration, reverse osmosis, and electrodialysis, which are predominantly based on arsenic preoxidation [19].

One of the most used treatment processes for arsenic removal from drinking water is coagulation with iron and ferric salts during which positively charged cationic coagulants (usually iron-based coagulants) reduce the negative charge of the colloids, causing the formation of larger particles and their aggregation. Addition of polymer further enhances the formation of the flocs in which the soluble As species are precipitated/co-precipitated and later removed by precipitation and/or filtration [19–21].

The aim of this study was to investigate the effect of arsenic preoxidation on arsenic removal from naturally arsenic-contaminated groundwater from Osijek area using two types of oxidant in various concentrations followed by the addition of three iron-based coagulants with the purpose to determine optimal type and concentration of oxidant and coagulant as function of pH. Experiments were conducted using laboratory jar test to contribute to the development of new cost-effective technique for arsenic removal from raw arsenic-contaminated water and achievement of maximal arsenic concentration of 10 μg/L imposed by Croatian Regulations of parameters compliance and analysis methods of water for human consumption [22] and European Community Directive guidelines [1]. Since the raw water used is also characterized by high total iron concentrations, the coagulation efficiency on iron removal was also evaluated.

2. Materials and methods

Groundwater is the most important source of drinking water in the area of eastern Croatia. The purpose of this study was to determine the optimal conditions for arsenic removal from groundwater by varying the following process parameters: pH, oxidant type and concentration ($KMnO_4$/H_2O_2), and type of iron-based coagulant ($FeCl_3$/$Fe_2(SO_4)_3$/$FeSO_4$).

Aerated groundwater from "Vinogradi" well field was used for laboratory experiments and it was collected from the drinking water treatment plant at Osijek, in eastern Croatia. The raw water characteristics are described in detail in our previous study [13], which revealed that As(III) is the predominant species. Average values of the main raw water parameters are summarized in Table 1. Different combinations of preoxidant and coagulant type were examined within pH values from 4.5 to 7.5. Before the addition of oxidant and iron-based coagulant, pH of each liter was adjusted using 0.1 M HCl or 0.1 M NaOH, and experiments were performed using four stirrers Biblock Scientific, Floculateur 10405 jar test apparatus according to the jar test standard method as follows:

Table 1
Average values of some parameters of aerated groundwater from Osijek area

pH	7.5
Conductivity (μS/cm)	853
Turbidity (NTU)	5.9
As (μg/L)	204
Fe (mg/L)	1.0
Cl^- (mg/L)	6
SO_4^{2-} (mg/L)	4
Ca^{2+} (mg/L)	75
Mg^{2+} (mg/L)	30
Mn (mg/L)	97.5
Hardness (mg$CaCO_3$/L)	290

appropriate amount of tested oxidant followed by appropriate amount of iron-based coagulant was added to each sample. The samples were flash mixed at 120 rpm for 10 s followed by 10 min of slow mixing at 30 rpm. After mixing period, the samples were allowed to settle for 15 min and then filtered through a 0.45 μm pore-sized membrane filter. Before and after the jar test, pH value, total arsenic, total iron, sulfate, and chloride concentrations were measured.

Three preoxidation and coagulation protocols were tested:

Protocol 1: *Coagulation by* $FeCl_3$*:* the first set of groundwater samples with different pH (from 4.5 to 7.5) was treated with $KMnO_4$ concentrations of 5, 10, and 15 g/L, and second set with H_2O_2 concentrations of 34, 85, and 170 mg/L. After oxidation, coagulant $FeCl_3$ was added in doses to obtain total iron concentration of 4 mg/L. A dose of iron-based coagulant was calculated based on initial total iron concentration in the taken sample.

Protocol 2: *Coagulation by* $Fe_2(SO_4)_3$*:* in a third set of groundwater samples with different pH (from 4.5 to 7.5), oxidant $KMnO_4$ was dosed in concentrations of 5, 10, and 15 g/L, while in the fourth set, H_2O_2 was dosed in concentrations of 34, 85, and 170 mg/L. After oxidation, the coagulant $Fe_2(SO_4)_3$ was added in each sample in concentration up to 4 mg/L Fe.

Protocol 3: *Coagulation by* $FeSO_4$*:* In groundwater samples with adjusted pH, 5, 10, and 15 g/L $KMnO_4$ or 34, 85, and 170 mg/L H_2O_2 were dosed followed by the addition of ferrous sulfate up to 4 mg/L Fe.

Ferric chloride ($FeCl_3$) was obtained from J.T. Baker-A Division of Mallinckrodt Baker, Inc., ferric sulfate ($Fe_2(SO_4)_3$) was obtained from Kemika d.d., and ferrous sulfate ($FeSO_4$) was obtained from Acros Organics. The oxidants, potassium permanganate ($KMnO_4$) and hydrogen peroxide (H_2O_2), were obtained from Kemika d.d. All used iron salts were dissolved in deionized water as 40% solutions.

The total arsenic concentrations in groundwater samples were analyzed before every protocol and they varied between 186.4 and 214.3 μg/L for arsenic. Therefore, the processes performances were expressed as percentages of arsenic removal that were calculated by Eq. (1).

$$\text{removal}\,(\%) = (1 - C_f/C_g) \times 100 \qquad (1)$$

where C_g and C_f are the arsenic concentrations in the untreated groundwater and in the filtrate obtained after sedimentation, respectively.

To assure the accuracy, reliability, and reproducibility of the obtained results, all batch tests were performed in triplicate and mean values of the obtained results were only reported. Total As (As(tot)) was determined by an USEPA anodic stripping voltammetry (ASV) (Computrace 757 VA, Methrom) method (USEPA, 1996). The accuracy of the method was evaluated by spiking the samples with known amounts of arsenic by the use of internal standards and by analyzing standard reference materials and blanks. The limit of detection (LOD) for As(tot) was calculated three times the standard deviation of the blank and was 0.11 μg/L. The relative standard deviation of replicate measurements was 0.106 μg/L using a 2 μg/L arsenic solution.

Sulfate concentrations in initial and final samples were determined using USEPA SulfaVer 4 method, while chloride concentrations were determined using ion chromatography (ICS-3000 Ion Chromatography System, Dionex).

The pH values of the samples were determined by a SevenEasy pH meter (Mettler, Toledo). Total Fe (Fe(tot)) concentrations were determined using a Hach spectrophotometer (DR/2000) and prepackaged reagents.

3. Results and discussion

3.1. Effect of $KMnO_4$ and iron coagulants dosing on arsenic removal

In this study, the effect of pH, type, and dose of oxidant and coagulant on arsenic removal from groundwater are investigated. Figs. 1 and 2 show the results of arsenic removal obtained during protocol 1, 2, and 3, i.e. when preoxidant ($KMnO_4$ or H_2O_2) followed by iron coagulant ($FeCl_3$, $Fe_2(SO_4)_3$ or $FeSO_4$) is dosed in a manner that initial total iron concentrations are 4 mg/L.

Fig. 1 shows the combined effects of initial pH and iron coagulant type on the total arsenic removal when $KMnO_4$ is used as a preoxidant with different concentrations, while Fig. 2 shows the combined effects at the same condition when H_2O_2 is used as arsenic preoxidant in various concentrations. Using $KMnO_4$ followed by $FeCl_3$ (Fig. 1(a)), it can be observed that arsenic removal increases with increasing $KMnO_4$ concentrations from 5 to 15 g/L, especially in the condition of higher pH value, while at a lower pH value of 4.5, all the results present the same value (over 90%). The lowest residual arsenic concentration of 12.1 μg/L was achieved when arsenic preoxidation was conducted with the addition of 15 g/L $KMnO_4$ at pH 7.5. Using $KMnO_4$ followed by $Fe_2(SO_4)_3$ (Fig. 1(b)), it can be observed that the achieved residual arsenic concentrations were between 12.2 and 18.9 μg/L, whereas

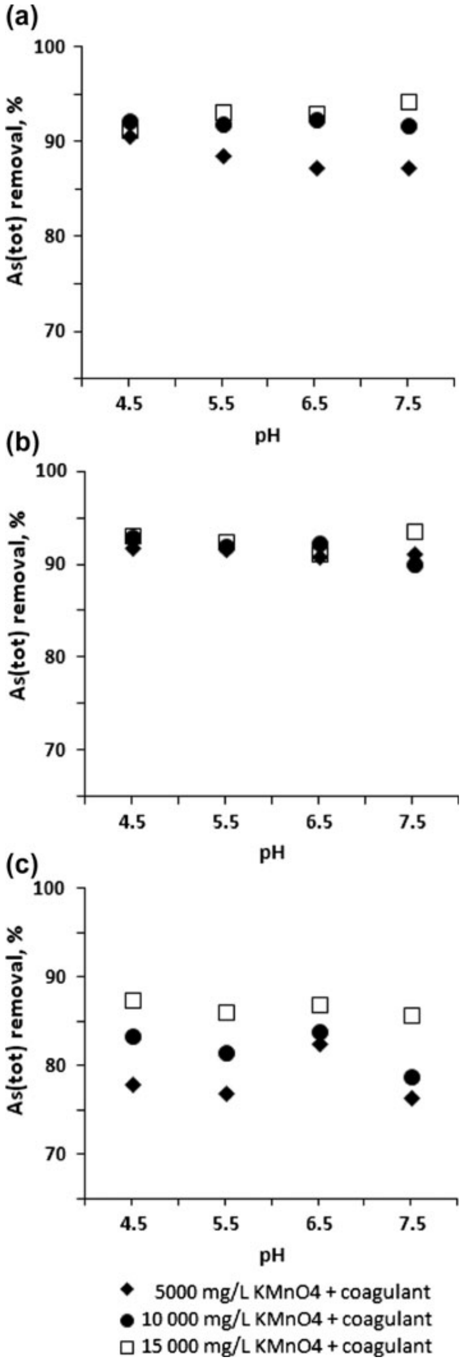

Fig. 1. Effect of pH, coagulant type (a) $FeCl_3$, (b) $Fe_2(SO_4)_3$, and (c) $FeSO_4$, and $KMnO_4$ dosage on total arsenic removal.

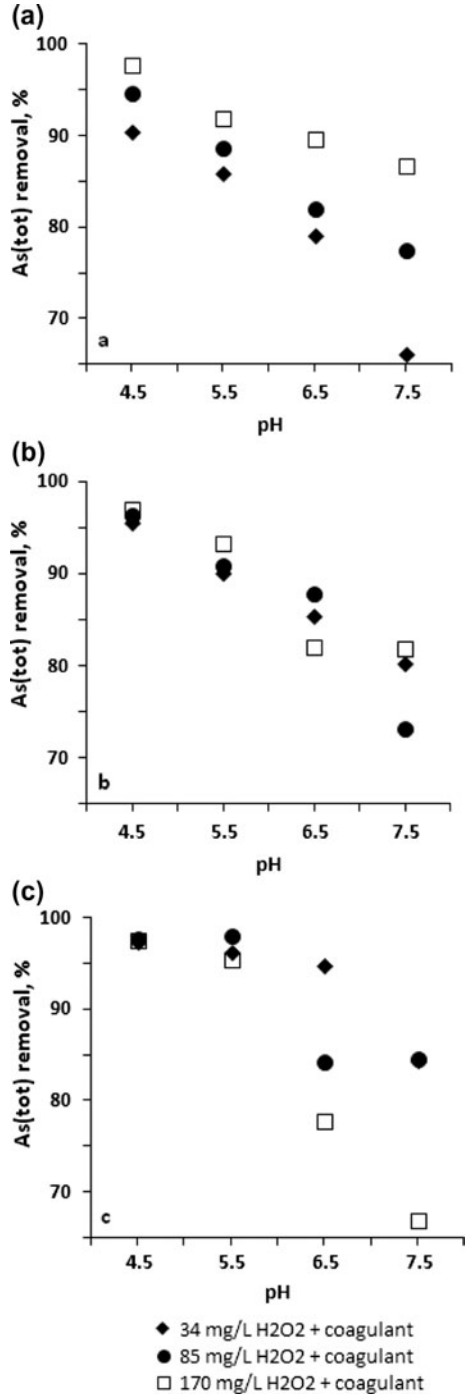

Fig. 2. Effect of pH, coagulant type (a) $FeCl_3$, (b) Fe_2SO_4, and (c) $FeSO_4$, and H_2O_2 dosage on total arsenic removal.

using $KMnO_4$ followed by $FeSO_4$ (Fig. 1(c)), residual arsenic concentrations were between 23.2 and 42.9 μg/L. Comparing the arsenic removals presented in Fig. 1(b), when ferric sulfate in combination with $KMnO_4$ was used, with the results of protocols 1 and 3 (Fig. 1(a) and (c)), it can be observed that the application of the above-mentioned combination resulted with highest percentages of arsenic removal at all tested pH values, while the strongest impact of $KMnO_4$ concentration on arsenic removal was noted when it was used in combination with $FeSO_4$ (Fig. 1(c)).

Because of its ability to oxidize a variety of organic and inorganic chemicals, effectiveness over a wide range of pH, $KMnO_4$ is one of the most commonly used oxidizing agent for hazardous contaminant that may occur in natural waters [23]. Influence of potassium permanganate and ferric chloride dosing on arsenic removal from model solutions and arsenic-contaminated groundwater was recently studied by Bordoloi et al. [24]. They reported that in the presence of initial iron ions between 1 and 5 mg/L, the optimum dose of $KMnO_4$ was 4 mg/L, and that higher initial iron concentrations in raw water demand higher $KMnO_4$ dosages. Our results show that, despite using different $KMnO_4$ concentrations, the type of iron-based coagulant strongly influenced the final arsenic concentration since over 85% of arsenic was removed when $FeCl_3$ and $(Fe_2SO_4)_3$ are used, while usage of $FeSO_4$ after arsenic preoxidation with $KMnO_4$ provided much lower percentage of arsenic removal from water.

Results of our study are also in accordance with results of the study conducted by Donmez et al. [21]. They also investigated arsenic removal from drinking water by coagulation using high concentrations of ferric chloride and ferrous sulfate (10 mg/L each one) and quoted that ferric chloride was used as a source of Fe(III) and ferrous sulfate was used as a source of Fe(II) in the process of floc formation. Results of their study confirmed that Fe(III) ions are more effective for arsenate removal, since Fe(III) coagulant, quickly and easily, formed insoluble $Fe(AsO_4)$ in the coagulation process [20,25].

3.2. Effect of H_2O_2 and iron coagulants dosing on arsenic removal

Fig. 2 shows the effect of pH value and dosage of different coagulants ($FeCl_3$, $Fe_2(SO_4)_3$, or $FeSO_4$) when various concentrations of H_2O_2 were used as arsenic preoxidant. It can be observed that the most effective arsenic removals (over 90%) are obtained with all tested iron coagulants and H_2O_2 dosage at lower pH value (4.5). Increasing the pH values decreases arsenic removal, especially using $FeCl_3$ and $Fe_2(SO_4)_3$. Comparing all the results of Fig. 2, it can be also observed that arsenic removal, generally, but not always, slightly increases when higher concentrations of H_2O_2 are used. Significant deviations of final results can only be noted at Fig. 2(c) when H_2O_2 is dosed followed by $FeSO_4$. In this protocol, the highest (over 98% of arsenic removal, i.e. residual arsenic of 4 μg/L) and lowest (residual arsenic 70.1 μg/L, i.e. 67% of its removal) percentages of arsenic removal are obtained. It seems that bivalent iron(II) has greater efficiency with hydrogen peroxide, while trivalent iron-based coagulants ($Fe_2(SO_4)_3$ and $FeCl_3$) increase arsenic removal with potassium permanganate (Fig. 1).

Dong et al. using H_2O_2 as preoxidant, followed by the addition of coagulants, also reported the effectiveness of arsenic removal, especially in acidic to neutral pH range [26]. Arsenic removal efficiency was increased with increasing concentration of H_2O_2, while at pH values close to neutral, the process efficiency was significantly lower. Those results are in accordance with the fact that H_2O_2 has low stability at higher pH and it is self-decomposed to oxygen and water at great extends at neutral pH [27], as well as with the fact that at ambient pH, $FeSO_4$ forms soluble $Fe(OH)_2$ that instantaneously causes significant reduction in iron precipitation; and hence, in arsenic removal [20,26]. It was also found that at neutral pH, single production of hydroxyl radicals from H_2O_2 results in lower process efficiency at higher pH values [28].

3.3. Final iron concentrations

High iron concentration in drinking water causes undesirable taste and yellowish color. Before it can be removed, iron has to be oxidized to a state in which it can form insoluble complexes, i.e. ferrous iron (Fe(II)) converts to ferric iron (Fe(III)), which readily forms the insoluble iron hydroxide complex $Fe(OH)_3$. Effectiveness of iron oxidation using $KMnO_4$ has been reported [29]. Since the groundwater used in this study contains high iron concentrations, final iron concentrations in treated groundwater were monitored. Obtained efficiencies of iron removal are presented in Figs. 3 and 4.

As shown in Fig. 3, when $KMnO_4$ was used as arsenic preoxidant, in most cases, residual iron concentrations were far below the regulation limit of 200 μg/L [22], especially in pH range near to neutral (from 6.5 to 7.5). Deviations of final iron concentrations are only noted among the results of *protocol 3* when $KMnO_4$ and $FeSO_4$ were used, and when the pH was below 6, extremely high residual iron concentrations were measured (from 3.16 to 4.36 mg/L).

As shown in Fig. 4, the initial pH values significantly influenced the iron precipitation in all protocols when H_2O_2 was used as preoxidant, particularly at H_2O_2 concentration of 170 mg/L. At pH 4.5 and 5.5, no iron was removed when $FeSO_4$ was used as coagulant and similar result was also obtained when ferric chloride was added. When H_2O_2 was added to groundwater samples with higher initial pH values (6.5 and 7.5), over 90% of iron was removed regardless of hydrogen peroxide concentration.

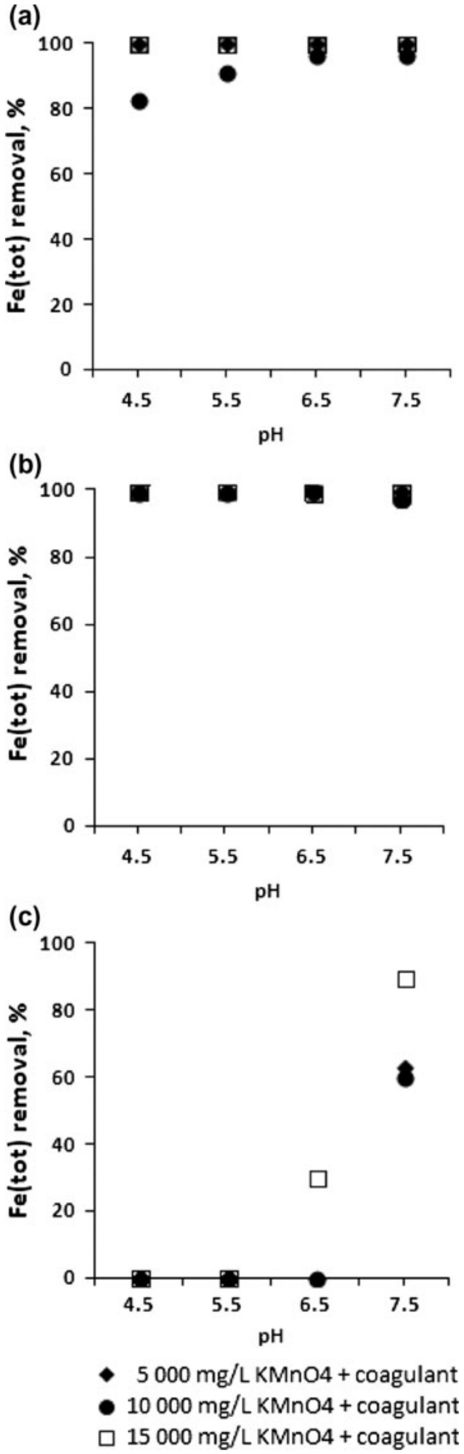

Fig. 3. Effect of pH, coagulant type (a) $FeCl_3$, (b) Fe_2SO_4, and (c) $FeSO_4$, and $KMnO_4$ dosage on Fe removal.

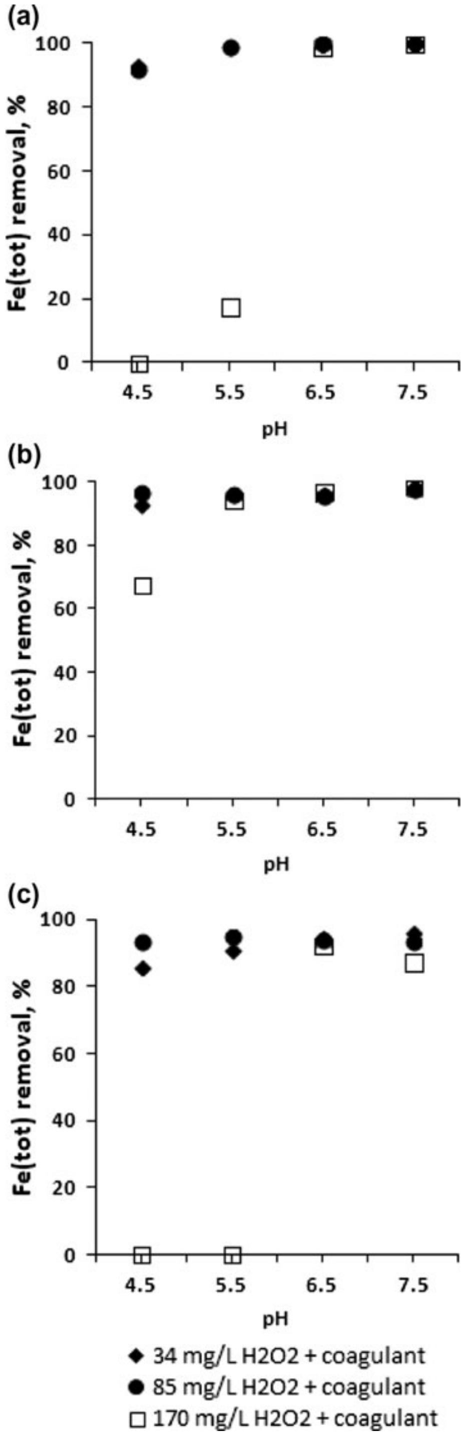

Fig. 4. Effect of pH, coagulant type (a) $FeCl_3$, (b) Fe_2SO_4, and (c) $FeSO_4$, and H_2O_2 dosage on Fe removal.

3.4. Final chloride and sulfate concentrations

As noted above, during this study, the chloride and/or sulfate ion concentrations are determined with the aim to define the impact of the addition of iron-based coagulant in water samples on their concentrations, i.e. when ferric chloride was used, chloride concentrations were determined and when

Table 2
Final chloride concentrations in treated water samples

		Final chloride concentration (mg/L)					
pH	Coagulant	5 g/L $KMnO_4$	10 g/L $KMnO_4$	15 g/L $KMnO_4$	34 mg/L H_2O_2	85 mg/L H_2O_2	170 mg/L H_2O_2
4.5	$FeCl_3$	37.2	36.8	31	34.46	32.2	17.4
5.5	$FeCl_3$	34.4	21.8	26.8	33.6	35.4	16.6
6.5	$FeCl_3$	21.8	19.4	19	28.4	26.4	17.8
7.5	$FeCl_3$	7.4	11.2	7.6	12.4	15.4	5.6

Table 3
Final sulfate concentrations in treated water samples

		Final sulfate concentration (mg/L)					
pH	Coagulant	5 g/L $KMnO_4$	10 g/L $KMnO_4$	15 g/L $KMnO_4$	34 mg/L H_2O_2	85 mg/L H_2O_2	170 mg/L H_2O_2
4.5	$Fe_2(SO_4)_3$	15.49	13.29	13.09	22	19	20
	$FeSO_4$	19	18	18	12	10	11
5.5	$Fe_2(SO_4)_3$	15.82	15.21	14.49	23	22	23
	$FeSO_4$	20	21	17	13	12	11
6.5	$Fe_2(SO_4)_3$	17.21	14.68	16.75	23	23	23
	$FeSO_4$	23	22	14	13	10	9
7.5	$Fe_2(SO_4)_3$	16.79	18.91	12.19	20	20	21
	$FeSO_4$	17	18	10	10	10	9

ferrous or ferric sulfate were used, sulfate concentrations were analyzed. Initial concentrations of chloride in all experiments were between 4 and 8 mg/L, while initial sulfate concentrations were between 2 and 4 mg/L. The obtained results presented in Tables 2 and 3 show that the addition of all three iron-based coagulants increases the chloride or sulfate concentrations in treated samples. When $KMnO_4$ followed by $FeCl_3$ are used, chloride concentrations were ranking from 7.4 to 37.2 mg/L and when H_2O_2 and $FeCl_3$ were dosed, chloride concentrations were between 5.6 and 35.4 mg/L. In both sets of experiments, the lowest chloride concentrations were obtained at highest oxidant concentration and at higher pH value (7.5). Sulfate concentrations were also elevated with regard to initial concentrations and when combination of $KMnO_4$ and $Fe_2(SO_4)_3$ was used, sulfate concentrations were ranking between 18 and 23 mg/L. When H_2O_2 and $Fe_2(SO_4)_3$ were added, sulfate concentrations were ranking between 19 and 23 mg/L. In experiments with $Fe_2(SO_4)_3$, no relationship among oxidant and coagulant dosages with final sulfate concentrations was observed. The results obtained were similar to the results described when $FeSO_4$ was dosed in combination with two used oxidants, i.e. final sulfate concentrations were between 18 and 23 mg/L when $KMnO_4$ was used and between 9 and 13 mg/L when H_2O_2 was used for preoxidation.

4. Conclusions

Previous studies revealed similar chemism and frequent occurrence of inorganic arsenic in groundwaters of the Pannonian basin, which usually exceeded a concentration of 200 μg/L, with the most often detected arsenic species reduced trivalent arsenic (As (III)) form. So, effective arsenic removal demands arsenic preoxidation. In order to reduce final arsenic concentration, naturally arsenic-contaminated groundwater from Osijek area is treated using two preoxidant ($KMnO_4$ and H_2O_2) in combinations with three iron-based coagulants ($FeCl_3$, $Fe_2(SO_4)_3$, and $FeSO_4$). Ferric chloride and ferric sulfate are used as a source of Fe (III) and ferrous sulfate is used as a source of Fe(II). The following conclusions have been drawn from this study:

Arsenic removal is more efficient when iron-based coagulants with Fe(III) ions in combination with potassium permanganate are used, while iron-based coagulant with Fe(II) ions shows more efficiency when hydrogen peroxide as arsenic preoxidant is used.

pH value did not have stronger impact on arsenic removal when $KMnO_4$ is used as preoxidant, since, within tested pH range, 87.3–94.4% of arsenic is removed using $FeCl_3$, 90.3–93.3% of arsenic is removed when $Fe_2(SO_4)_3$ is used, and 76.4–87.5% of arsenic is removed when $FeSO_4$ is used.

Application of H_2O_2 as preoxidant shows more pH dependence and in all protocols, over 90% of arsenic removal is only obtained at pH 4.5. Increasing in pH significantly reduces arsenic removal and at average natural pH of used groundwater (7.5), highest arsenic removals (86.8% with $FeCl_3$, 82.0 with $Fe_2(SO_4)_3$, and 84.7% with $FeSO_4$) are mostly obtained when highest H_2O_2 concentrations are used.

Acknowledgment

The authors are thankful to Mr Samuel O'Sullivan from AsureQuality, New Zeland, for the help during manuscript handling.

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