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REMOVAL OF ARSENIC AND FLUORIDE FROM DRINKING WATER WITH CAKE ALUM AND A POLYMERIC ANIONIC FLOCCULENT

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SUMMARY: The combined use of cake alum and a polymeric anionic flocculent (PAF) for removal of arsenic (As) and fluoride (F) from drinking water has been evaluated in water from two wells at Meoqui City, Chihuahua, Mexico. Field data revealed that As and F concentrations may, by this method, be reduced up to 99% and 77%, respectively. The addition of small amounts of PAF greatly facilitated sedimentation of the precipitate. Sodium hydroxide (8% NaOH solution) was used to adjust the pH near to an optimal 7.1. The efficiency of F removal depended on the amount of cake alum employed and varied with pH, whereas As removal required only the presence of cake alum. Adsorption of inorganic contaminants, which helps precipitation of metal hydroxide solids, was likely to be the dominant mechanism for F and As removal.

Keywords: Arsenic removal; Cake alum; Drinking water; Fluoride removal; Groundwater; Polymeric anionic flocculent.

INTRODUCTION

Groundwater contamination by arsenic (As) and by fluoride (F) can result from the natural dissolution of minerals from subterranean strata.[1-4] These inorganic contaminants in drinking water are known to cause serious health problems when the maximum contaminant levels (MCL) exceed 0.05 (recently lowered to 0.01) mg/L for As and 1.5 mg/L for F.[5-9]

Two efficient methods for removal of As and F from water are: a) addition of a given material (usually cake alum and/or ferric chloride) to the water during the softening or coagulation processes; and b) an ion-exchange process or an adsorption process. For the first method, the amount of cake alum required is related to pH and initial F concentration, and these factors need to be considered to avoid high operating costs.[10-15] At high cake alum concentrations, the constituent metal of the coagulant precipitates as an amorphous hydroxide floc in which colloidal particles containing As and F become entrapped.[16-17]

The aim of this research was to evaluate the removal of As and F from groundwater by a mixture of cake alum (aluminum sulfate octadecahydrate) and a polymeric anionic flocculent (PAF) and to obtain data for a cost-effective groundwater treatment to comply with the maximum contaminant level (MCL) for As and F in drinking water.

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MATERIALS AND METHODS

The groundwater used in this study was obtained from two wells at Meoqui City, Chihuahua, México, which is located 1945 km north of Mexico City and 448 km south of El Paso, Texas. The quality of the groundwater is given in Table 1.

Table 1. Groundwater quality before treatment

Chemical parameters	Well No. 1	Well No. 2
Fluoride (mg/L)	5.9	4.8
Arsenic (mg/L)	0.134	0.075
Aluminum (mg/L)	0.048	0.062
Conductivity (µmho/cm)	620	580
Turbidity (ntu)	1.4	1.1
Sulfate (mg/L)	105	134
Total Alkalinity (mg/L)	121	146
Total Hardness (mg/L)	24.5	58.3
pH	7.2	7.4

Laboratory treatment tests were carried out using glass beakers to study the effect of the combined use of cake alum/PAF for removal of F and As in groundwater. For the tests, six 1-L beakers were placed on a six-paddle stirrer unit (Phipps and Bird, Richmond, VA) and filled with the groundwater to be tested. Different amounts of cake alum (200 mesh) were added to each beaker, followed by mixing at 100 rpm for 1 min, 40 rpm for 15 min, and finally quiescent settling for 15 min. The treated groundwater was then gravity filtered through Whatman No. 2 filter paper (12.5 cm diameter).

Cake alum and sodium hydroxide (see below) were industrial grade. The PAF was prepared from acrylamide, CH_2=CHCONH$_2$ (Cinetica Quimica Co., Monterrey N.L. Mexico). Addition of 1 mg/L of PAF was found to be the most effective amount for removal efficiency. Total As concentration [As(III) + As(V)] was determined by hydride generation-flame atomic absorption spectrometry (HG-FAAS), equipped with an electrodeless discharge lamp and coupled to a GBC Avanta Σ-hydride generator (GBC Avanta, Dandenong Victoria, Australia) according to Standard Method 3114B. The F concentration was measured following the SM 4500-F procedure outlined in Standards Methods.[18] For this determination, a specific ion meter (Orion, model 720A, Boston, MA) equipped with both F and reference electrodes was employed. Calibration was done over the range of 0.1–10 mg/L.

From the laboratory results, a pilot-scale coagulation-filtration process was then investigated. The treatment system was designed for two volumetric flow rates: 5 and 10 Lps (liters per second). The initial stage of the process consisted of mixing 450 mg of cake alum, 1 mg of PAF, and 1.1 mL of 8% NaOH solution per liter of groundwater to be treated. In all cases, the pH was kept between 6.8 and 7.0. For the 5 Lps flow rate, the coagulation process required about 3 hr in an inclined plate settler, followed by filtration through two beds of silica sand and two beds of activated carbon. For the 10 Lps flow rate, after the settling stage, filtration was done through four sand beds and four activated carbon beds. Three replicates were performed for each of these field treatment tests.

The percent efficiency of removal was calculated by dividing the difference between the F or As concentration in the inflow and the outflow stream by the corresponding inflow concentration.

RESULTS AND DISCUSSION

In Table 2 the laboratory test results show the effect of cake alum dosage on the corresponding residual F concentrations in water from Wells No. 1 and 2. These tests indicate that an increase in the amount of cake alum resulted in a further decrease in residual F in the water from both wells. Addition of 1 mg of the polymeric anionic flocculent (PAF) facilitated the flocculation and sedimentation processes. As expected, an increase in the amount of cake alum also resulted in a rapid decrease in pH, which was raised by addition of NaOH. Contrary to what has been reported,[19] significant F removal (76–77%) was obtained even at low pH (4.6–4.8) as shown in Table 2.

Regarding pH control, although lab testing indicated that the minimum F solubility occurred in a pH range of 6.1–6.4, the sludge floc formed around pH 7.1 was much more stable and presented better settling characteristics. Therefore, the target pH was raised to 7.1 with 8% NaOH solution, which still an acceptable pH for F precipitation. A large portion of the sludge is probably formed by coagulation of sulfate with the complexed F in the aluminum hydroxide floc. Coincidentally, an increase in sulfate concentration was observed in the effluent streams for both wells, as recorded in Table 3. Some hydroxylated aluminum complex formed in the precipitation of alum has been suggested to combine with precipitates or to remove F along with the flocculated material by adsorption of F ions into the diffuse layer of the flocculent**.**[17]

Table 2. Laboratory tests for F removal efficiencies in one-liter batches of groundwater from Wells No. 1 and No. 2

Well No.	Cake Alum (mg/L)	[Ff] (mg/L)	% F[a] Reduction	pH[b]
1	340	2.9	51	5.5
	360	2.5	58	5.3
	380	2.4	59	5.1
	400	2.0	66	4.9
	420	1.7	71	4.8
	440	1.6	73	4.7
	450	1.4	76	4.6
2	300	1.7	65	6.2
	320	1.7	65	6.0
	340	1.6	67	5.8
	360	1.5	69	5.4
	380	1.4	71	5.1
	400	1.3	73	4.9
	420	1.1	77	4.8

[a] Rounded average of three replicates. [b] pH obtained during the batch test without addition of NaOH.

Table 3. Groundwater quality after treatment, treatment costs and comparison with MCL values

Chemical and Design Parameters	Well No. 1[a]	Well No. 2[b]	MCL Mexico	MCL USA
Fluoride (mg/L)	1.4	1.1	1.5	4.0
Arsenic (mg/L)	$<$ 0.001	$<$ 0.001	0.05	0.05
Aluminum (mg/L)	0.07	0.08	0.2	0.05-0.2*
Conductivity (µmho/cm)	750	730	-	-
Turbidity (ntu)	1.5	1.2	5.0	-
Sulfate (mg/L)	241	230	400	400-500†
Total Alkalinity (mg/L)	113	124	-	-
Total Hardness (mg/L)	25.1	56.3	500	-
PH	7.0‡	7.2‡	6.5-8.5	6.5-8.5*
Flow in designed system (Lps)	5	10		
Cost (US dollars)	121,250	169,600		

[a] Cake alum concentration of 450 mg/L. [b] Cake alum concentration of 420 mg/L. *Secondary Drinking Water Standards. †Proposed. ‡pH obtained after treatment (adding 8% NaOH to ajust the pH around 7.0).

It is also interesting that the conductivity and sulfate concentration increased in comparison to the initial groundwater composition (see Tables 1 and 3). From the standpoint of environmental legislation, it is worth stressing that these physicochemical parameters are below the MCL values established in Mexico and USA. Although F and Al comply with Mexican regulations, an experimental study has shown that an aluminum fluoride (AlF_3) concentration of 0.5 ppm, corresponding to a fluoride ion concentration of about 1 mg/L in the drinking water of rats was associated with an increase in morbidity and mortality, with a one-year administration of aluminum fluoride producing distinct morphological alterations in the brain, including effects on neurons and the cerebrovasculature.[20] It has also been reported that fluoride levels below MCL values can have long-term human toxic effects, including dental fluorosis, decrease in thyroid function, and disturbances of other soft tissues.[5-9, 21]

Although not shown in Table 2, but as seen in Table 3, excellent removal of As (98–99%) was achieved during all treatment conditions for F removal. These high As removal efficiencies remained constant for the water from both wells as the cake alum concentration was increased to 340–450 mg/L and 300–420 mg/L, respectively. From the treatment parameters of pH and cake alum dosage determined in the test experiments, the improvement in As removal appears to be directly attributable to a reduction in soluble Al.[16]

As a result of the increased sludge production, a filter press was found to be a cost effective treatment tool. Cost estimates for the current coagulation-filtration treatment system with groundwater pH adjusted to 7.1 were calculated for wells No. 1 and 2, as shown in Table 3. Operation and maintenance costs per cubic meter treated water were also estimated and compared with other treatment systems like activated alumina and reverse osmosis, as shown in Table 4. Clearly, the proposed treatment system is an attractive economical option.

TABLE 4. Estimated operation and maintenance costs per cubic meter of treated water from wells No. 1 and 2

Well No.	Design flow (Lps)	Activated alumina (US $/m^3)	Reverse osmosis (US $/m^3)	Proposed system (US $/m^3)
1	5	0.54	0.59	0.38
2	10	0.41	0.40	0.32

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