

Determination of Optimum Coagulants (Ferric Chloride and Alum) for Arsenic and Turbidity Removal by Coagulation

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Abstract

The Raw water from Deer Creek (DC) reservoir and Little Cottonwood Creek (LCC) reservoir in the Utah, USA were collected for jar test experiments. This study examined the removal of arsenic and turbidity by means of coagulation and flocculation processes using of aluminum sulfate and ferric chloride as coagulants for 13 jar tests. The jar tests were performed to determine the optimal pH range, alum concentration, ferric chloride concentration and polymer concentration for arsenic and turbidity removal. The results showed that a comparison was made between alum and ferric chloride as coagulant. Removal efficiency of arsenic and turbidity for alum (16 mg/L) of up to 79.6% and 90.3% at pH 6.5 respectively were observed. Removal efficiency of arsenic and turbidity for ferric chloride (8 mg/L) of up to 59.5% at pH 8 and 90.6% at pH 8 respectively were observed. Optimum arsenic and turbidity removal for alum dosages were achieved with a 25 mg/L and 16 mg/L respectively. Optimum arsenic and turbidity removal for ferric chloride dosages were achieved with a 20 mg/L and 8 mg/L respectively. In terms of minimizing the arsenic and turbidity levels, the optimum pH ranges were 6.5 and 8 for alum and ferric chloride respectively. When a dosage of 2 mg/L of potassium permanganate and 8 mg/L of ferric chloride were employed, potassium permanganate can improve arsenic removal, but not turbidity removal.

Key Words : Arsenic, Coagulation, Drinking water treatment, Flocculation, pH control

1. Introduction

Arsenic is a carcinogenic metalloid found in ground and surface water. Arsenic contamination in drinking water leads to detrimental effects on human health. It has been regulated in drinking water by U.S.EPA at a maximum contaminant level (MCL) of 0.05 mg/L since 1977, but a value of 0.01 mg/L has been accepted recently because of the fatal toxicity arsenic on human health(Lee et al., 2003).

Arsenic will increase the risk of skin cancer, liver,

kidney, and bladder when ingested and of lung cancer when inhaled. Chicago Tribune reported in February of 1999 that as many as 200,000 people are dying annually of arsenic poisoning from consuming contaminated well water in Bangladesh and India. Industrial effluents are a major source of arsenic to environment. Arsenic is also widely used in pigments and dyes, preservatives of animal hides and wood, pulp and paper production. Possible arsenic contaminations could originate from mine tailings, smelting of metals, and burning of fossil fuels. Water is one of the major of transport of arsenic in the environment(Thirunavukkarasu et al., 2002; Chen et al., 1994; Gregor, 2001).

Arsenic in its soluble form generally occurs in its +3 and +5 oxidation states. Both organic (methylated

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arsenic) and inorganic forms of arsenic can be detected in natural water system. Organic arsenic occurs at concentrations less than 1 g/L and is not of major significance in drinking water treatment (Edwards, 1994). The pentavalent arsenate species are predominant and stable in the oxygen-rich aerobic environments, whereas the trivalent arsenite species are predominant in reducing anaerobic environments such as ground water (Ghosh and Yuan, 1987). The pH determines the predominant arsenate and arsenite species. The toxicity scale of arsenic decreases in the order: arsine (AsH_3) > inorganic As(III) > organic As(III) > inorganic As(V) > organic As(V) > element As. The toxicity of As(III) is about ten times that of As(V) (Lee et al., Gupta and Chen, 1978; Pontius et al., 1994). Oxidation of As(III) to As(V) must be applied because technologies perform most effectively when treating arsenic in the form of As(V). In the pH range of 4 to 10, the predominant As(III) compound is neutral in charge, while the As(V) species are negatively charged. Removal efficiencies for As(III) are poor compared to removal As(V) by any of the technologies evaluated due to the negative charge (Chen et al., 1999; Pallier et al., 2010). Data on oxidants indicate that chlorine, ferric chloride, and potassium permanganate are effective in oxidizing As(III) to As(V). However, pre-oxidation with chlorine may create undesirable by-products. Ozone and hydrogen peroxides should oxidize As(III) to As(V), but there is no data available on performance. While the atomic adsorption spectrophotometer (AAS) can detect total arsenic, it can not speciate. If As(III) is found to predominate, then pre-oxidation should be considered to convert As(III) to As(V).

The goal of this work was to demonstrate the efficiency of chemical coagulation and flocculation for the removal of arsenic and turbidity from reservoir water.

2. Materials and Methods

All inorganic analyses were performed at Metropolitan

Water District Salt Lake (MWDSL's) Environmental Laboratory in the Salt Lake City, Utah, USA. Jar tests were performed using 2-L square jars with a six-place jar testing apparatus consisting of rectangular paddle impellers (Model 7790-400, PHIPPS & BIRD STIRRER, Richmond, VA). Arsenic concentrations were analyzed on a graphite furnace atomic absorption spectrophotometer (Model Zeeman 5100, Perkin Elmer). A turbidimeter (Model 2100AN, Hach Company, Loveland, Colo.) was used to measure the turbidity. It was calibrated daily by using standards of 0-2, 0-20, and 0-200 NTU. A pH meter (Model S310, Orion Research) which was calibrated daily by using pH 4.0 and pH 7.0 buffer solutions was used. Total and calcium hardness were measured by the ethylenediaminetetraacetic acid (EDTA) titrimetric method. Alkalinity was conducted by the titration method was measured using a calibrated thermometer. Both Deer Creek (DC) reservoir water and Little Cottonwood Creek (LCC) water in the Utah, USA were collected for jar test experiments.

Raw water quality characteristics of pH, alkalinity, turbidity, hardness, and temperature were measured by standard methods. Table 1 shows characterization of raw DC reservoir water. Table 2 shows characterization of raw LCC reservoir water. Alum and Ferric chloride were used as primary coagulants and cationic polymer was used as a coagulant aid. 0.1M NaOH and 0.1M H_2SO_4 solutions were used to adjust pH conditions. For each jar test, each jar was filled with 2L of raw water measured with a graduate cylinder, and the initial temperature was recorded. Then the coagulant and coagulant aid were added into all jars simultaneously. After that, the test solutions were rapidly mixed for 0.5 minutes at 300 rpm, gently stirred for 4 minutes at 45 rpm, 8 minutes at 35 rpm, 8 minutes at 15 rpm and then allowed to settle for 140 minutes. Potassium permanganate was added and mixed (200 rpm) for 30 minutes prior to adding coagulant and coagulant aid. Coagulation pH was measured

Table 1. Raw DC reservoir water characteristics

Jar Tests	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11	#12	#13
pH	7.72	7.56	7.74	7.63	7.81	7.61	7.72	7.57	7.61	7.72	7.72	7.58	7.72
*Alkalinity	133	133	129	129	127	127	131	131	132	130	126	135	131
Turbidity (NTU)	8.69	3.25	9.04	1.39	8.38	4.62	2.03	2.32	2.04	7.75	5.22	8.03	2.03
Temperature (°C)	18.5	21.2	18.2	21.1	18.4	19.7	21.3	21.0	21.0	20.0	19.2	19.8	21.3
*Total Hardness	198	168	168	168	172	166	168	171	174	174	172	186	168
*Ca ²⁺ Hardness	137	137	136	128	146	134	139	135	135	148	136	140	139

* Unit is mg/L as CaCO₃

Table 2. Raw LCC reservoir water characteristics

Jar Tests	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11	#12	#13
pH	7.62	7.97	7.76	7.95	7.97	7.90	8.10	7.89	7.96	7.75	7.99	7.71	8.10
*Alkalinity	47	60	55	56	44	54	49	48	49	48	51	48	49
Turbidity (NTU)	0.79	0.43	33.04	3.50	0.70	0.33	0.30	0.27	0.45	7.72	1.74	1.66	0.30
Temperature (°C)	18.5	20.9	19.9	20.5	17.9	19.1	20.9	20.8	21.0	22.4	19.5	22.0	20.9
*Total Hardness	74	89	114	88	68	75	65	69	69	70	72	72	65
*Ca ²⁺ Hardness	51	67	82	60	52	60	55	55	55	58	58	54	55

* Unit is mg/L as CaCO₃

Table 3. Mixed raw DC and LCC reservoirs water characteristics

Jar Tests	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11	#12	#13
pH	7.98	7.91	7.81	7.69	7.80	7.91	7.87	7.85	7.89	7.95	7.96	7.66	7.87
*Alkalinity	92	114	89	109	84	90	115	112	69	108	67	90	115
Turbidity (NTU)	3.53	2.95	21.22	2.71	3.68	2.46	1.93	1.84	1.04	6.41	3.78	4.84	1.93
Temperature (°C)	18.2	21.4	19.7	21.6	18.7	20.1	21.5	22.0	21.0	21.2	20.5	21.1	21.5
*Total Hardness	123	141	116	115	116	122	153	151	89	145	107	112	153
*Ca ²⁺ Hardness	93	115	94	158	90	96	115	111	80	117	73	87	115

* Unit is mg/L as CaCO₃

Table 4. Purposes and conditions for each jar tests

Jar Tests	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11	#12	#13	
Purpose	Optimum pH				Optimum Alum Dose				Optimum FeCl ₃ Dose					
pH	**F				8.22	8.19	8.75	8.79	8.79	8.79	8.88	8.19		
*Alum	16	16	0	0	0	**F				0	0			
*Ferric Chloride	0	0	8	8	8	0	0	0	0	0	0	**F		
*Cationic Polymer	0.5	0.5	0.5	0.5	0.5	0.5	0.25	0	0	0.5	0.5	0.5	0.25	
*Potassium Permanganate	0	0	0	0	2	0	0	0	0	0	0	0	0	

* Unit is mg/L

** Find out the optimum conditions for each jar tests

after rapid mixing. Table 3 shows characterization of mixed of raw DC and LCC reservoirs water. Table 4 shows purposes and conditions for each jar tests.

3. Results and Discussions

3.1. Optimization of pH on arsenic and turbidity removal

Jar tests 1 and 2 were performed to determine the optimal pH range for turbidity and arsenic removal, using alum as coagulant. The difference between jar test 1 and jar test 2 were ratios of mixed water. Raw water was mixed with 50% of DC and 50% of LCC for jar test 1. Raw water was mixed with 75% of DC and 25% of LCC for jar test 2. Raw water would be analyzed for concentrations of arsenic prior to spiking. Initial arsenic concentrations in jar test 1 and jar test 2 were 81.70 g/L and 128.50 g/L respectively. Alum and cationic polymer concentrations were held constant at 16 mg/L and 0.5 mg/L, respectively, and

the pH was varied to determine the effect of pH on arsenic and turbidity removal. A total of 6 pH conditions, 6.5, 7.0, 7.5, 8.0, 8.5, and 9.0 were applied in this jar test. For water with pH is higher than 7, the pH decreased upon addition of coagulant. Samples were collected for arsenic and turbidity removal after 140 minutes of sedimentation. Poor turbidity removal was observed at pH is higher than 8.5. Optimal turbidity removal and arsenic removal occurred at pH is higher than 7.5 when alum was used as a coagulant. In jar test 1, turbidity and arsenic removal percents at pH 6.5 were 89.8% and 73.9% respectively. In jar test 2, turbidity and arsenic removal percents at pH 6.5 were 90.3% and 79.6% respectively. Turbidity and arsenic removal decreased with increasing alkalinity and pH. Fig.1 shows removal of turbidity and arsenic by using alum and polymer as a function of pH in jar test 1 and 2.

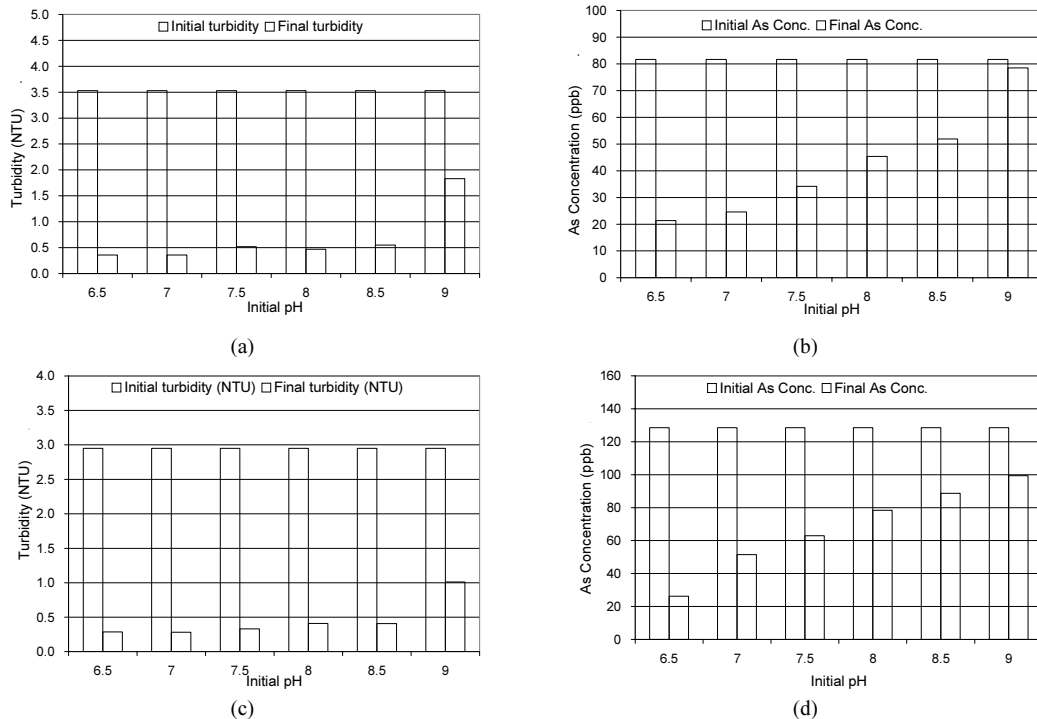


Fig. 1. Removal of turbidity and arsenic by using alum and polymer as a function of pH in jar test 1 (a, b) and 2 (c, d).

Jar test 3 and 4 were performed to determine the optimal pH range for turbidity and arsenic removal, using ferric chloride as a coagulant. Again, the ratios of mixed water were varied between jar test 3 and jar test 4. Raw water was mixed with 50% of DC and 50% of LCC for jar test 3. Raw water was mixed with 75% of DC and 25% of LCC for jar test 4. Raw water would be analyzed for arsenic concentration. Initial arsenic concentrations in jar test 3 and jar test 4 were 121.30 g/L and 104.50 g/L respectively. Ferric chloride and cationic polymer concentrations of 8 mg/L and 0.5 mg/L, respectively, were held constant in each jar to determine the effect of pH on arsenic and turbidity removal. Again, a total of 6 pH conditions, 6.5, 7.0, 7.5, 8.0, 8.5, and 9.0 were applied in this jar test. Addition of coagulants to water that pH is higher than 7.5 lowered the pH. In water with pH is lower than

7.5, the coagulants slightly raised the pH. Samples were collected for arsenic and turbidity removal after 140 minutes of sedimentation. In jar test 3, the highest arsenic removal percent at pH 9.0 was 50.6%. The highest turbidity removal percent at pH 8.5 was 92.5%. In jar test 4, the highest arsenic removal percent at pH 8.0 was 59.5%. The highest turbidity removal percent at pH 9.0 was 83.8%. Poor turbidity removal was observed at pH is lower than 7. Optimal turbidity removal occurred at pH is higher than 7.0 using ferric chloride as a coagulant. The turbidity removal increased with higher alkalinity. Alkalinity and pH do not have an effect on arsenic removal. Fig. 2 indicates removal of turbidity and arsenic by using ferric chloride and polymer as a function of pH in jar test 3 (a, b) and 4 (c, d).

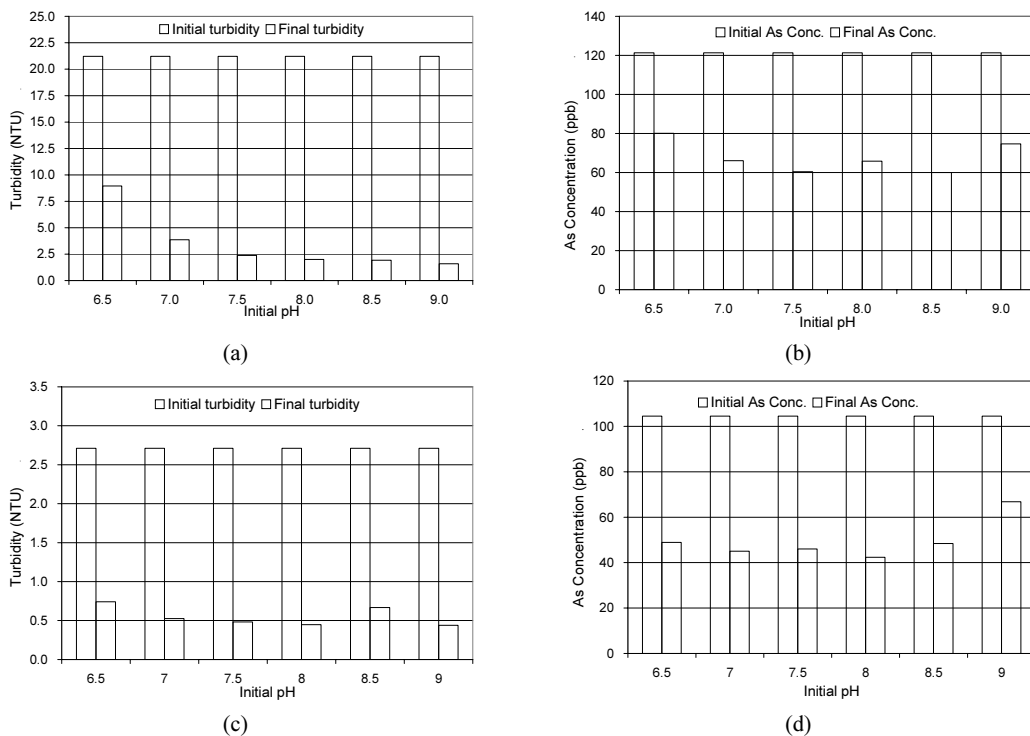


Fig. 2. Removal of turbidity and arsenic by using ferric chloride and polymer as a function of pH in jar test 3(a, b) and 4(c, d).

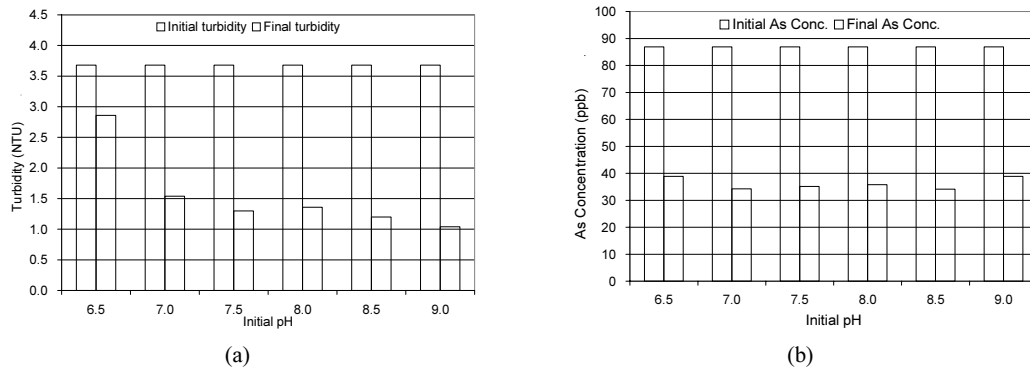


Fig. 3. Removal of turbidity (a) and arsenic (b) by using ferric chloride and potassium permanganate as a function of pH in jar test 5.

Oxidation of As (III) to As (V) must occur for effective arsenic removal by current water treatment processes. Jar test 5 was performed to determine the amount of potassium permanganate required to oxidize As (III) to As (V). Jar test 5 was carried out under the same conditions as jar test 3, except dosages of potassium permanganate were added. A dosage of 2 mg/L of potassium permanganate was employed. This dosage is an order of magnitude greater than the standard operating dosage. Optimal dosage of potassium permanganate was not determined yet for arsenic removal. The trend of turbidity removal was the same as that for jar test 3. Removal percent of arsenic in jar test 5 was around 15% higher than in jar test 3. Therefore, in future studies, optimum dosage of potassium permanganate should be researched for arsenic removal. Fig. 3 indicates removal of turbidity (a) and arsenic (b) by using ferric chloride, polymer and potassium permanganate as a function of pH in jar test 5.

3.2. Optimization of alum dose on arsenic and turbidity removal

Jar test 6 was performed to determine the optimal alum dose for turbidity and arsenic removal. Raw water was mixed with 50% of DC and 50% of LCC for this test. Mixed raw water pH was 7.91, which was adjusted to 8.22. Alum was added in increments of

12, 14, 16, 18, 20, and 22 mg/L to solutions containing 131.9 g/L of arsenic. The turbidity removal somewhat increased with increasing alum dosage and decreasing pH. Arsenic removal rapidly increased with increasing alum dosage and decreasing pH. Optimum dosage of alum for turbidity removal is 16 mg/L and arsenic removal increased upon addition of alum coagulant.

Jar test 7 was performed to determine the optimal alum dose for turbidity and arsenic removal. Raw water was mixed with 84% of DC and 16% of LCC. Mixed raw water pH was 7.87, which was adjusted to 8.19. Alum was added in increments of 5, 10, 15, 20, 25, and 30 mg/L to solutions containing 71.5 g/L of arsenic. The turbidity removal increased with increasing alum dosage and decreasing pH. The minimum dose of alum for removing turbidity is 10 mg/L. Arsenic removal increased with increasing alum dose. Optimum dose of alum for removal arsenic is 25 mg/L because 30 mg/L of alum dose was worse than 25 mg/L for arsenic removal. Fig. 4 indicates removal of turbidity and arsenic as a function of alum dose in jar test 6 (a, b) and 7 (c, d).

Jar test 8 was performed to determine turbidity removal by alum without the assistance of cationic polymer. Raw water was mixed with 75% of DC and 25% of LCC. The alkalinity of the mixed water was

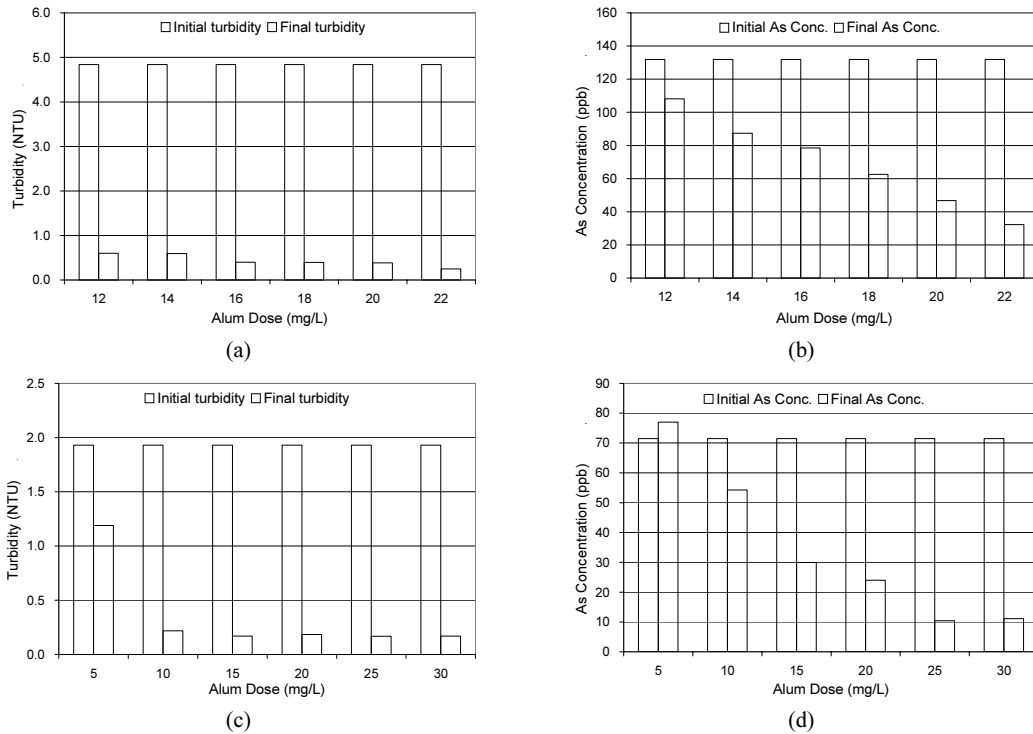


Fig. 4. Removal of turbidity and arsenic as a function of alum dose in jar test 6(a, b) and 7(c, d).

112 mg/L. An initial pH of 8.75 was measured in water sampled at the aeration basin. Mixed raw water pH was 7.85, which was adjusted to 8.75. Alum was added in increments of 10, 12, 14, 16, 18, and 20 mg/L to the test solutions. For alum dosages are higher than 14 mg/L, the turbidity was lower than 0.5 NTU. Jar test 9 was performed in the same manner as jar test 8 for the same purpose; however, the ratios of mixed water were reversed. Raw water was mixed with 25% of DC and 75% of LCC. The alkalinity of the mixed water was 69 mg/L. An initial pH of 8.79 was measured in water sampled at the aeration basin. Mixed raw water pH was 7.89, which was adjusted to 8.79. Alum was added in increments of 10, 12, 14, 16, 18, and 20 mg/L to the test solutions. The turbidity removal was higher for water with higher alkalinity using only alum as a coagulant.

Jar test 10 was performed to determine the optimal

alum dose for turbidity removal. Raw water was mixed with 75% of DC and 25% of LCC. The alkalinity of the mixed water was 108 mg/L. An initial pH of 8.75 was measured in water sampled at the aeration basin. Mixed raw water pH was 7.95, which was adjusted to 8.75. Alum was added in increments of 10, 12, 14, 16, 18, and 20 mg/L to the test solutions. The turbidity removal increased with increasing alum dosage and decreasing pH. Jar test 11 was performed to determine the optimal alum dose for turbidity removal. Raw water was mixed with 25% of DC and 75% of LCC. The alkalinity of the mixed water was 67 mg/L. An initial pH of 8.79 was measured in water sampled at the aeration basin. Mixed raw water pH was 7.96, which was adjusted to 8.79. Alum was added in increments of 10, 12, 14, 16, 18, and 20 mg/L to the test solutions. The turbidity removal increased with increasing alum dosage

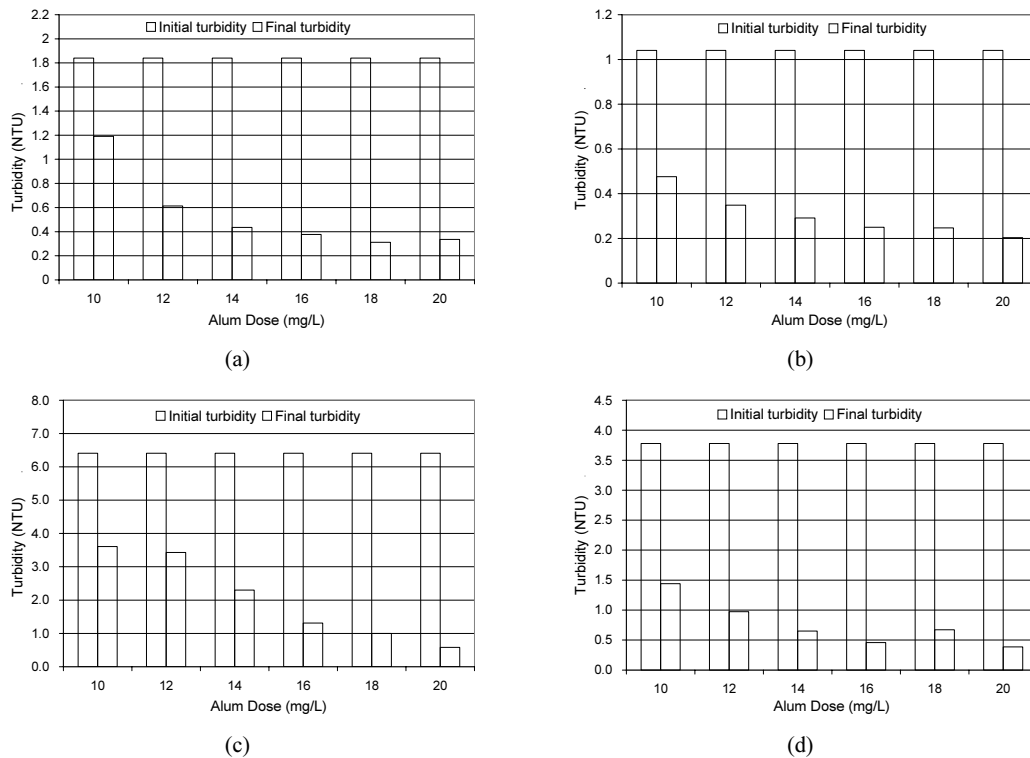


Fig. 5. Removal of turbidity as a function of alum dose in jar test 8(a), 9(b), 10(c) and 11(d).

and decreasing pH. Fig. 5 shows removal of turbidity as a function of alum dose in jar test 8 (a), 9 (b), 10 (c) and 11 (d).

3.3. Optimization of ferric chloride dose on arsenic and turbidity removal

Jar test 12 was performed to determine the optimal ferric chloride dose for turbidity and arsenic removal. Raw water was mixed with 50% of DC and 50% of LCC for jar test 12. An initial pH of 8.89 was measured in water sampled at the aeration basin. Mixed raw water pH was 7.66, which was adjusted to 8.89. Incremental amounts of ferric chloride, which were 6, 8, 10, 12, 14, and 16 mg/L were added to solutions containing 109.7 g/L of arsenic. The turbidity removal slightly increased with increasing ferric chloride dosage and decreasing pH. Arsenic removal rapidly increased with increasing ferric chloride dosage

and decreasing pH. Optimum dosage of ferric chloride for turbidity removal is 8 mg/L and arsenic removal increased upon addition of ferric chloride coagulant.

Jar test 13 was performed to determine the optimal ferric chloride dose for turbidity and arsenic removal. Raw water was mixed with 84% of DC and 16% of LCC. An initial pH of 8.19 was measured in water sampled at the aeration basin. Mixed raw water pH was 7.87, which was adjusted to 8.19. Ferric chloride was added in increments of 4, 8, 12, 16, 20, and 24 mg/L to solutions containing 71.30 g/L of arsenic. The turbidity removals increased with increasing ferric chloride dosage and decreasing pH. The minimum dose of ferric chloride for removing turbidity is 8 mg/L. The arsenic removal increased with increasing ferric chloride dose. Optimum dose of ferric chloride

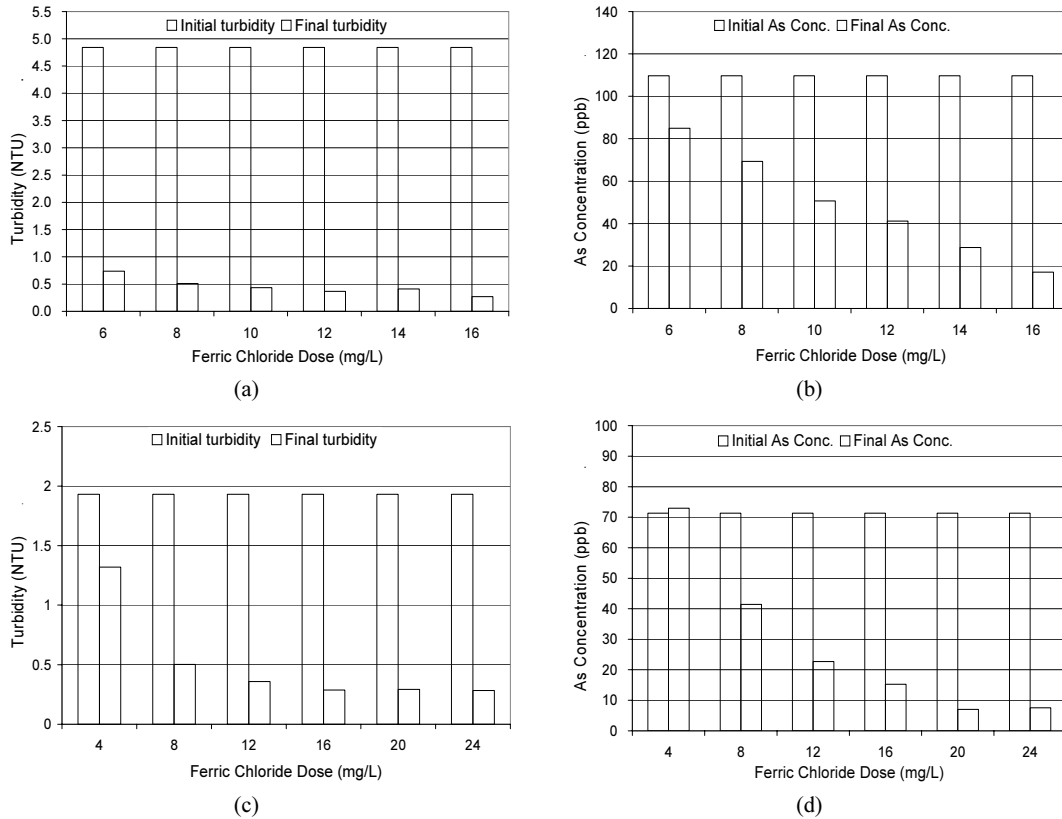


Fig. 6. Removal of turbidity and arsenic as a function of ferric chloride dose in jar test 12(a, b) and 13(c, d).

for removal arsenic is 20 mg/L because 24 mg/L of ferric chloride dose was worse than 20 mg/L for arsenic removal. Fig. 6 indicates removal of turbidity and arsenic as a function of ferric chloride dose in jar test 12 (a, b) and 13 (c, d).

4. Conclusions

The selection of an appropriate method for arsenic depends on a number of factors such as influent concentration of arsenic, water composition and pH which were investigated in this research. All factors were investigated in this research allowed for an effective removal of arsenic and turbidity from reservoir water.

The optimum pH ranges for arsenic and turbidity

removal were 6.5 and 8 for alum and ferric chloride coagulation respectively. The optimum dosages for arsenic and turbidity removal by alum coagulation were 25 mg/L and 16 mg/L. The optimum dosages for arsenic and turbidity removal by ferric chloride coagulation were 20 mg/L and 16 mg/L. Arsenic and turbidity removal decreased with increasing alkalinity and pH. Potassium permanganate can improve arsenic removal, but not turbidity removal. The optimum dosage of alum for turbidity removal is 16 mg/L, and arsenic removal increased upon addition of a coagulant of alum. When alum and ferric chloride were used as the coagulant, turbidity or arsenic removal did not depend on the added amount of cationic polymer.

Acknowledgements

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References

- Chen, H., Frey, M. M., Clifford, D., McNeil, L. S., Edwards, M., 1999, Arsenic treatment considerations, *J. of American Water Works Association*, 91, 74-85.
- Chen, S. L., Dzenk, S. R., Yang, M. H., 1994, Arsenic species in ground waters of the black-foot diseases area, Taiwan, *J. of Environ. Sci. Technol.*, 28, 877-881.
- Edwards, M., 1994, Chemistry of arsenic removal during coagulation and Fe-Mn Oxidation, *J. of American Water Works Association*, 86, 64-78.
- Ghosh, M. M., Yuan, J. R., 1987, Adsorption of inorganic arsenic and organo arsenicals on hydrous oxides, *Environmental Progress*, 6, 150-157.
- Gregor, J. E., 2001, Arsenic removal during conventional aluminum-based drinking-water treatment, *Wat. Res.*, 35, 1659-1664.
- Gupta, S. K., Chen, K. Y., 1978, Arsenic removal by adsorption, *J. of Water Pollution Control Federation*, 50, 493-506.
- Lee, Y., Um, I. H., Yoon, J., 2003, Arsenic (III) Oxidation by Iron (VI)(Ferrate) and Subsequent Removal of Arsenic (V) by Iron (III) Coagulation, *J. of Environ. Sci. Technol.*, 37, 5750-5756.
- Pallier, V., Feuillade-Cathalifaud, G., Serpaud, B., Bollinger, J., 2010, Effect of organic matter on arsenic removal during coagulation/flocculation treatment, *J. of Colloid and Interface Science*, 342, 26-32.
- Pontius, F. W., Brown, K. G., Chen, C. J., 1994, Health implications of arsenic in drinking water, *J. of American Water Works Association*, 86, 52-63.
- Thirunavukkarasu, O. S., Virarghavan, T., Subramanian, K. S., Tanjore, S., 2002, Organic arsenic removal from drinking water, *J. of Urban Water*, 4, 415-421.