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Reuse of greywater: effect of coagulant

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ABSTRACT

The objective of the present study was to (1) examine the effect of coagulants on greywater (GW) characteristics and (2) assess the potential of treated GW for reuse. Since the pH plays an important role in the amount of coagulant required, the effect of coagulants was studied under different pH conditions (pH 5.5, 6.5, 7.5 and 8.5) with the help of jar tests. Ferric chloride ($FeCl_3$) and polyaluminium chloride (PAC) coagulants were tested on light GW. Study revealed that, the optimum coagulant dose was decreased with a decrease in GW pH, indicating high coagulant demand at high pH. Turbidity removal was between 93 and 98% in $FeCl_3$ treatment and 97–98% in PAC treatment. In $FeCl_3$ treatment, mean biochemical oxygen demand (BOD_5) removal ranged from 50 to 59%, and *Escherichia coli* removal was above 91%. In the PAC treatment, mean BOD_5 removal ranged from 42 to 62%, and *E. coli* removal was above 92%. Total coliforms and faecal coliforms removal was above 99% in both the coagulant treatments. The present study shows that both ($FeCl_3$ and PAC) treatments satisfy almost all the reuse standards for discharge into land for irrigation, and industrial cooling in India.

Keywords: Greywater; Ferric chloride; Polyaluminium chloride; Jar test; Optimum dose; Reuse

1. Introduction

Water is a natural resource vital for the survival of all the species on earth. Around one billion people on earth are without reliable supplies of water, and more than two billion people lack basic sanitation. Growing populations, changing diets, increased urban, agricultural and industrial water demands increase the gap between water availability and water demand [1]. It is estimated that, by 2025, around 1.8 billion people will face absolute water scarcity (i.e. water availability < 500 m^3/person/year) and 67% of the world population could face water stress condition (i.e. water availability is below 1,700 m^3/year) [2]. As per Census carried out in 2011, India's population is over 1,200 million and around 31% of the population is living in urban areas. Total utilizable water availability in the country is below 1,000 m^3/person/year (water scarcity condition). Moreover, distribution of water resources in India is highly uneven over space and time [3].

Options to cope with water scarcity can be broadly grouped in supply enhancement and demand management. Reuse of wastewater is one of the options of supply enhancement [4]. Greywater (GW) is a wastewater from kitchen, bath and laundry, excluding wastewater from toilets [5,6]. Light greywater (LGW) is the GW from bathroom, showers, tubs and clothes

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washing machines sources. Dark greywater includes more contaminated waste from laundry facilities, dishwashers and kitchen sinks [7]. In a household, the proportion of GW flow is around 50–80% of the total wastewater flow [8,9]. Hence, GW reuse can be an effective measure for saving water on the domestic level and reducing load on wastewater treatment plant. Separating GW from blackwater (BW) reduces the danger posed by pathogens [9]. Domestic water consumption can be reduced up to 50% and achieves nearly “zero emission” when BW and GW is treated separately [10]. The reuse of grey water for non-potable water applications is a potential solution for water-deprived regions worldwide. GW is not suitable for direct use, but can be useful for non-potable reuse such as irrigation and toilet flushing [6].

Negative electric charges on the finely dispersed particles (colloids) in the wastewater repel them from each other. Coagulation and flocculation assists in removal of colloids. Coagulation can neutralize the negative charge and flocculation enhances the floc formation process, which increases the floc size and its mass. This leads to rapid settling of the suspended and colloidal particles. At present, investigations on GW using ferric chloride ($FeCl_3$) and polyaluminium chloride (PAC) are quite limited [11,12]. Friedler [13] examined LGW using ferric chloride coagulant by jar test. Parameters were analysed at optimum condition, but did not include some important reuse parameters like total suspended solids (TSS), biochemical oxygen demand (BOD_5), total coliforms (TC), faecal coliforms (FC), *Escherichia coli*, etc. Friedler and Alfiya [14] investigated LGW from office premises using ferric chloride by jar test, but did not include microbiological parameters. In another study, Antonopoulou et al. [15] examined the performance of alum and ferric chloride on shower, hand basin and kitchen GW; but, turbidity, BOD_5, TC, FC and *E. coli* were not included in the study. Moreover, the optimum coagulant dosages were not reported.

PAC is another coagulant which is used in water and wastewater treatment. But, its performance in treating LGW, and the potential of treated GW for reuse under different pH conditions is not documented so far [12]. PAC has a higher coagulant efficiency and relative low cost compared to other conventional coagulants. Less sludge is produced compared to alum at an equivalent dose and works effectively over a broader pH range of 5.0–8.0 [16–18]. Role of pH is important in case of coagulation/flocculation; and, an optimum dose (OD) is dependent on pH of GW [19,20]. As a whole, coagulation/flocculation studies documented so far lack in the study under variable pH conditions, optimizing the coagulant dosage (in terms of Fe or Al) and monitoring parameters from the reuse point of view [11,12]. Therefore, there is a need to assess performance of ferric chloride and PAC in treating GW from the reuse point of view including the reuse parameters which were not attended by the previous researchers.

The objective of the present study was to (1) examine the effect of coagulants ($FeCl_3$ and PAC) on GW characteristics (under different pH conditions) and (2) assess the potential of treated GW for reuse (irrigation, construction and industrial cooling). GW parameters were monitored at OD conditions.

2. Materials and methods

2.1. GW collection

GW was collected from a student hostel of capacity 400 located at Sardar Vallabhbhai National Institute of Technology, Surat, India. The hostel has a separate collection system for BW and GW. The GW comprised water from hand basins, showers and bathrooms. The GW collection pipe was cut and GW was diverted to the collecting tank (CT). GW was homogeneously mixed in the CT, prior to collection of samples. Samples were collected at around 10 am in the morning (four samples/week); around 60 L GW was collected for the experimental purpose and the remaining volume was discarded. The CT was washed each time before use with clean potable water to avoid any carryover of contaminants. The experiments were started within 30–45 min from the time of collection of GW.

2.2. Coagulants used

From literature review it was revealed that the investigations on GW using $FeCl_3$ and PAC were quite limited. Therefore, $FeCl_3$ and PAC were used in the study. $FeCl_3$ contained 30.97% Fe and PAC contained 15.07% Al by weight. Coagulant stock was prepared using dry coagulant and distilled water (DW) at least 1 d in advance to ensure proper dissolving in water. The stock was prepared regularly by dissolving 5 g coagulant in 250 mL DW, representing 1 mL of stock solution equal to 20 mg/L of the coagulant.

2.3. Experiments

A six-paddle stirrer (with rotating blades) jar test apparatus (DBK Instruments-DBK FLOCCULATOR) was used in GW coagulation/flocculation. Six jars of capacity 1 L each were used in the jar test. NaOH (1N) or H_2SO_4 (1N) was used to adjust the pH of raw GW. In the present study, raw GW pH was observed as

7.89 ± 0.35. For each sample four jar tests were conducted by adjusting pH to 8.5, 7.5, 6.5 and 5.5 and ODs were found. GW was poured into a 7 L capacity plastic bucket and its pH was adjusted to 8.5 by adding NaOH (1 N) to raw GW. H_2SO_4 (1 N) was used to lower the pH in case of adjusting pH to 7.5, 6.5 and 5.5. The pH-adjusted raw GW was then filled in the jars and the coagulant dose was added, in increasing order, to jar number 2–6. First jar did not contain any coagulant dose and was considered as "zero dose". GW in jar was subjected to rapid mixing at 120 rpm for 90 s. It was, then, immediately subjected to slow mixing at 30 rpm for 15 min. The flocs formed in the jars were allowed to settle for next 45 min without any disturbance. After the settling period was over, around 50 mL supernatant was collected from each jar and its residual turbidity was measured. The dose corresponding to the least turbidity was considered as OD. The available literature lacks in obtaining optimum coagulant dose of $FeCl_3$ and PAC in treating GW. Therefore, we had concentrated on finding OD and subsequently parameters were analysed at optimum dosage.

Next, the supernatant from the jar corresponding to the OD were collected in the sampling bottles and moved to the refrigerator (stored at 4˚C). Samples were analysed within 2–24 h (except the analysis of metals and ground elements). The entire jar test was repeated if an OD was not appearing. The treated samples corresponding to initial adjusted raw GW pH equal to 8.5, 7.5, 6.5 and 5.5 are referred as TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. Duration of the study was from December 2012 to June 2013.

2.4. Analytical procedures

The pH was measured using a digital pH meter (Hanna Instruments- pH 209); turbidity was measured using a digital Nephelo turbidity meter (Systronics-Turbidity Meter 132); and electrical conductivity (EC_{25}) was measured using a digital conductivity meter (Systronics- Conductivity total dissolved solids (TDS) Meter 308). Standardization of the instruments was checked each time before sample analysis for ascertaining their proper working condition. Solids were measured by drying the samples in hot air oven. Oil and grease (O&G) was analysed by partition-gravimetric method using petroleum ether (40˚C/60˚C). Phosphates (PO_4-P) were analysed by stannous chloride method. Dissolved oxygen in BOD_5 test was analysed as per the Winkler method with azide modification. Chemical oxygen demand (COD) was analysed by the open reflux method using potassium dichromate ($K_2Cr_2O_7$) as an oxidizing agent.

TC and FC were analysed by multiple-tube fermentation technique (five tubes, three dilutions). Presumptive test for coliforms was performed by using McConkey broth (incubation 35 ± 0.5˚C for 24 h). Further, TC was confirmed by using brilliant green bile broth (incubation 35 ± 0.5˚C for 24 h) and FC was confirmed by using EC broth (incubation 44.5 ± 0.2˚C for 24 h). Results were expressed as MPN/100 mL. *E. coli* was analysed by pour plate method using EMB agar (incubation 37˚C for 24 h) and results were expressed as colony forming units (cfu) per 100 mL.

Ca, Mg, Na, B, As, Fe and Al were analysed using inductively coupled plasma atomic emission spectroscopy at Sophisticated Analytical Instrument Facility laboratory, Indian Institute of Technology Bombay. All the parameters were analysed as per standard methods [21,22]. Reagent/laboratory grade chemicals were used in the study.

2.5. Statistical analysis

The results were analysed using descriptive and multivariate statistics using Excel 2007 and SYSTAT (Sigmaplot 10).

3. Results and discussion

Characteristics of the raw GW analysed in the present study are presented in Table 1. Most of the characteristics' levels observed in the present study are in the range of literature cited. The pH of the raw GW used in this study was slightly higher than that of literature cited. Since, fresh GW was used in the study and was tested immediately; so the change in pH was not possible. It was observed that pH was decreased when GW was stored for more than 12 h.

In the present study, BOD_5 and COD ranged from 130 ± 52 and 290 ± 76 mg/L, respectively; and COD/BOD_5 ratio varied from 1.41–3.16 (2.3 ± 0.44). In the literature reported, COD/BOD_5 ratio varied from 1.33 to 2.9 for bath, 1.52 to 2.8 for a shower and 1.88–3.6 in case of wash basin GW [24,26]. Abu Ghunmi et al. [27] reported COD/BOD_5 ratio as 2.33 for combined bath, laundry and washbasin GW. Wastewater with COD/BOD_5 ratio above two is not easily treatable by biological means. Biological process needs a minimum BOD_5:N:P ratio of 100:5:1 for complete BOD_5 removal under aerobic conditions [28]. Since, GW does not include urine; it is expected to be deficient in N. Similarly, most of the phosphorus originates from detergents used in washing and will only be present if the laundry GW is included [26]. Biological treatment can be used efficiently for collective wastewater

Table 1
Raw GW characteristics in the present study and literature data from different countries

	Present study			Literature data		
	n	Range	Avg ± SD	Australia [8]	France [23]	Israel [24,25]
GW sources		B,S,W	B,S,W	B	B,S,W	B,S,W
pH	27	7.13–8.53	7.89 ± 0.35	6.4–8.1	7.34–7.71	7–7.43
Turbidity (NTU)	27	30–175	83.78 ± 32	60–240	35.3–462@	
Temperature (^{0}C)	27	23.7–31.8	27 ± 2			
EC_{25} (μS/cm)	27	504–1,381	660 ± 171	82–250	358–627	1,130
Total solids (mg/L)	22	426–1,184	638 ± 146			770–1,090
TDS (mg/L)	22	320–992	469 ± 143			
TSS (mg/L)	22	82–256	167 ± 51	48–120	37–360.5	78–303
O&G (mg/L)	20	24–122	68 ± 29	37–78		7.2–164
Alkalinity (mg/L)	25	80–368	275 ± 58	24–43		
NH_3-N (mg/L)	19	0.84–10.6	3 ± 2	<0.1–15		0.39–1.2
PO_4-P (mg/L)	8	0.41–0.97	0.7 ± 0.2			4.56–15
BOD_5 (mg/L)	22	60–300	130 ± 52	76–200	78–670	44–424
COD (mg/L)	22	164–424	290 ± 76		100–795	230–645
TC (MPN/100 mL)	18	4.6E3–9.3E6	1.7E6 ± 3.4E6	500–2.4E7	4.9E6–4.7E9	
FC (MPN/100 mL)	18	2.6E3–9E4	2.2E4 ± 2.7E4	170–3.3E3	2.3E4–2.0E6	3.5E3–4E6
E. coli (cfu/100 mL)	18	45–7.8E3	1.4E3 ± 2E3			
Ca (mg/L)	8	1.23–56.79	17.5 ± 19	3.5–7.9		
Mg (mg/L)	8	10.23–83.54	25.8 ± 23.7	1.4–2.3		
Na (mg/L)	8	40.89–257.19	104.5 ± 64.82	7.4–18		112–151
SAR	8	2.26–6.34	3.87 ± 1.46			
B (mg/L)	8	0.024–0.29	0.14 ± 0.09			0.31–0.44
As (mg/L)	8	<0.01	–	0.001		
Fe (mg/L)	8	0.006–0.344	0.07 ± 0.12	0.34–1.1		
Al (mg/L)	8	0.002–0.821	0.21 ± 0.27	<1		

Notes: n, number of samples; Avg, average; SD, standard deviation; B, bath; S, shower; W, washbasin; and @, Turbidity (FNU).

treatment under supervision of trained staff, but it would be difficult to treat GW in single households where the inhabitants have no specific skills to treat wastewater [15]. Thus, high COD/BOD_5 ratio and nutrient deficiency of the GW indicates enough scope for physicochemical treatment in the present study.

The ratio of total dissolved solids (TDS) to EC_{25} of raw GW was observed as 0.7 ± 0.09. EC_{25} was observed equal to 1.189 times TDS plus 109 ($R^2 = 0.82$, $p < 0.05$). Both, EC_{25} and TDS, play an important role when water is reused for irrigation purpose. EC_{25} is a surrogate measure of TDS. It is a measure of salinity hazard, which relates to the total soluble salt content in the solution. High EC_{25} water disables plants to compete with ions in the soil solution for water. The higher the EC_{25}, the less water is available to plants, which ultimately reduces crop yield [29].

3.1. Effect of coagulant dose on physical and chemical characteristics

GW characteristics before and after treatment by ferric chloride and PAC are presented in Tables 2 and 3, respectively. The mean OD was observed as 57 ± 11, 55 ± 11, 49 ± 12 and 30 ± 6 mg-Fe/L, and 38.6 ± 9.3, 34.1 ± 7.8, 33.2 ± 8.1 and 29.8 ± 9.5 mg-Al/L for TGW8.5, TGW7.5, TGW6.5, and TGW5.5, respectively. At pH 5.5 OD required in both the coagulants is almost the same by mass. As the pH increased from 5.5 to 8.5, the amount of Fe and Al required also increased. More Fe was required by mass, compared to Al at same pH. However, conversion to molar concentrations indicates that more Al is required than Fe. For instance, in TGW6.5 mean Fe dose required was 49 mg-Fe/L (0.86 mM) and mean Al dose required was 33.2 mg/L (1.23 mM) indicating that proportionally more PAC was required per unit volume of treatment. Considering coagulant required at optimum dosage, the present cost of coagulant required (in India) to treat one million litre GW works out to be US$ 96, 92, 82 and 50 in case of ferric chloride; and that of PAC works out to be US$ 71, 62, 61 and 55 for TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. In similar study on LGW, Pidou et al. [30] observed an optimum ferric sulphate dose of 44 mg-Fe/L for pH 4.5 and an optimum alum dose of 24, 28 and 32 mg-Al/L for pH values of 4.5, 6 and 7,

Table 2
Mean GW characteristics (Avg ± SD) after $FeCl_3$ treatment under different pH conditions

			Treated greywater			
	n	Raw greywater	TGW8.5	TGW7.5	TGW6.5	TGW5.5
pH	13	7.84 ± 0.38	5.83 ± 0.8	5.72 ± 0.65	5.24 ± 0.72	4 ± 0.63
Turbidity (NTU)	13	78.4 ± 39	1.9 ± 2.5	2 ± 1.9	5.5 ± 6.9	5.5 ± 5.4
EC_{25} (μS/cm)	13	662 ± 113	801 ± 136	782 ± 112	797 ± 123	856 ± 146
TDS (mg/L)	11	473 ± 101	530 ± 101	529 ± 105	526 ± 111	548 ± 81
TSS (mg/L)	11	164 ± 53	31 ± 28	20 ± 17	40 ± 39	31 ± 20
O&G (mg/L)	11	57 ± 31	22 ± 20	22 ± 21	28 ± 24	30 ± 13
Alkalinity (mg/L)	12	278 ± 70	133 ± 66	93 ± 47	67 ± 32	40 ± 17
NH_3-N (mg/L)	11	3.2 ± 1.7	1.6 ± 0.7	1.7 ± 0.8	2.2 ± 0.9	2.4 ± 1.1
BOD_5 (mg/L)	11	115 ± 23	55 ± 22	46 ± 27	47 ± 22	58 ± 28
COD (mg/L)	11	251 ± 45	125 ± 42	105 ± 53	92 ± 52	110 ± 64
TC (MPN/100 mL)	9	2.2E6 ± 3.9E6	2.1E4 ± 2.2E4	1.5E3 ± 1.3E3	1.3E3 ± 1.1E3	1.2E4 ± 1.1E4
FC (MPN/100 mL)	9	2.1E4 ± 2.6E4	189 ± 131	138 ± 159	76 ± 83	196 ± 144
E. coli (cfu/100 mL)	9	1,467 ± 1,678	133 ± 248	86 ± 188	52 ± 90	138 ± 273
SAR	4	4.4 ± 1.91	3.76 ± 1.2	2.96 ± 0.64	2.52 ± 0.32	2.27 ± 0.43
B (mg/L)	4	0.19 ± 0.09	0.15 ± 0.05	0.13 ± 0.06	0.12 ± 0.08	0.1 ± 0.08
As (mg/L)	4	<0.01	<0.01	<0.01	<0.01	<0.01

Table 3
Mean GW characteristics (Avg ± SD) after PAC treatment under different pH conditions

			Treated greywater			
	n	Raw greywater	TGW8.5	TGW7.5	TGW6.5	TGW5.5
pH	14	7.9 ± 0.3	6.4 ± 0.5	6.3 ± 0.5	5.9 ± 0.3	4.6 ± 0.4
Turbidity (NTU)	14	88.8 ± 24	2.1 ± 3.2	2.9 ± 4.4	2.4 ± 2.6	3 ± 2.3
EC_{25} (μS/cm)	14	658 ± 215	756 ± 216	710 ± 212	719 ± 209	760 ± 217
TDS (mg/L)	11	464 ± 180	485 ± 159	469 ± 155	471 ± 153	503 ± 159
TSS (mg/L)	11	169 ± 52	31 ± 41	45 ± 51	40 ± 36	41 ± 35
O&G (mg/L)	9	82 ± 18	26 ± 16	22 ± 19	30 ± 24	41 ± 25
Alkalinity (mg/L)	13	272 ± 46	162 ± 39	147 ± 35	98 ± 26	50 ± 23
NH_3-N (mg/L)	8	3.4 ± 3.1	2 ± 2.5	1.9 ± 2.9	1.7 ± 2.7	1.6 ± 2.7
BOD_5 (mg/L)	11	145 ± 68	52 ± 28	65 ± 39	52 ± 37	83 ± 48
COD (mg/L)	11	329 ± 83	120 ± 56	165 ± 85	137 ± 102	186 ± 100
TC (MPN/100 mL)	9	1.3E6 ± 2.9E6	5.6E3 ± 5.7E3	1.2E3 ± 2.5E3	340 ± 755	361 ± 119
FC (MPN/100 mL)	9	2.3E4 ± 2.9E4	196 ± 173	124 ± 103	106 ± 161	88 ± 232
E. coli (cfu/100 mL)	9	1,389 ± 2,447	113 ± 103	25 ± 30	16 ± 35	23 ± 61
SAR	4	3.34 ± 0.76	2.94 ± 1.17	3.23 ± 0.95	2.96 ± 0.84	2.65 ± 0.9
B (mg/L)	4	0.13 ± 0.07	0.10 ± 0.04	0.1 ± 0.01	0.1 ± 0.02	0.09 ± 0.02
As (mg/L)	4	<0.01	<0.01	<0.01	<0.01	<0.01

Note: *n*, number of samples; Avg, average; and SD, standard deviation.

respectively. In the present study, also, OD was reduced as per decrease in pH (Fig. 1).

It was observed that the pH was dropped after addition of coagulant (Fig. 2). Drop in pH was more in case of ferric chloride dose comparing to PAC dose. Ferric ions act as "Bronsted acids" donating a proton to the solution, which depress the pH [15]. In addition, when ferric chloride is added to water, carbon dioxide gas is liberated. This CO_2 then reacts with water producing carbonic acid (H_2CO_3). Hence, the pH of ferric chloride-coagulated water dropped. In coagulation by Fe ions, drop in pH from 6 to 5 raises charge neutralizing capacity 2.43 times; and drop from 6 to 4 raises it 2.81 times as that of pH 6 [31]. This indicates that coagulation should proceed in acidic pH, which demands lower Fe dose.

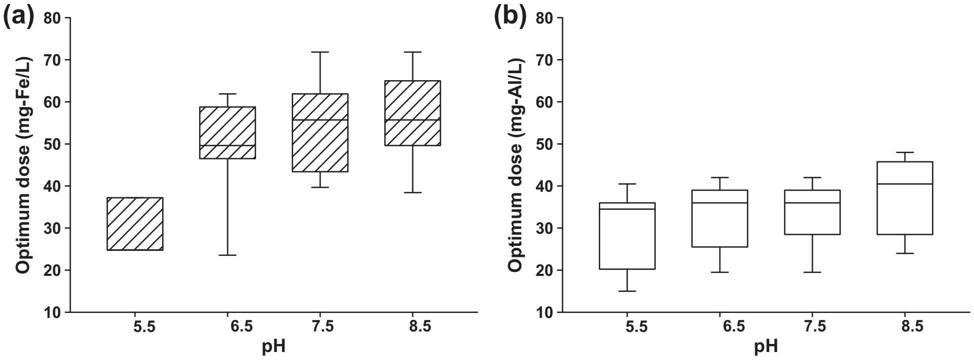

Fig. 1. Effect of pH on optimum coagulant dose. (a) Ferric chloride and (b) PAC.

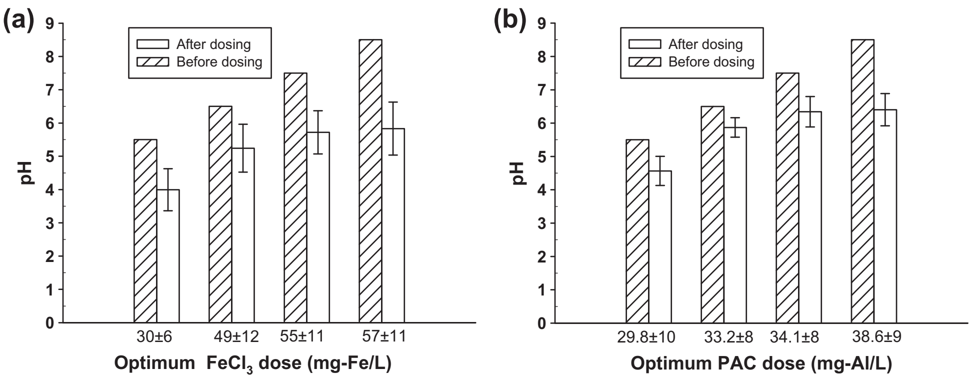

Fig. 2. Effect of coagulant dose on pH. (a) Ferric chloride and (b) PAC.

In case of PAC, pH drops due to release of H^+ ions in the hydrolysis reaction. Eq. (1) shows a typical hydrolysis reaction of PAC ($Al_2(OH)_3Cl_3$):

$$\begin{aligned} Al_2(OH)_3Cl_3 &\rightarrow Al_2(OH)_3^{3+} + 3Cl^- + 3H_2O \\ &\rightarrow 2Al(OH)_3 + 3H^+ + 3Cl^- \end{aligned} \quad (1)$$

When an aluminium-based coagulant such as PAC is added to water, the metal ion is hydrolyzed to form aluminium hydroxide floc as well as hydrogen ions. The H^+ ions will react with the alkalinity of the water and in the process, decrease the pH of the water. Hence, the pH of PAC-coagulated water drops [32].

Mean turbidity levels in $FeCl_3$-treated GW were dropped from 78.4 NTU to 1.9 (removal 98%), 2 (removal 98%), 5.5 (removal 93%) and 5.5 NTU (removal 93%) in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively Fig. 3(a). In similar studies on light GW, Friedler et al. [13] reported turbidity removal from 34 to 2 NTU (removal 94%) at an optimum $FeCl_3$ dose of 40–50 mg/L. In another study on light GW, Friedler and Alfiya [14] reported turbidity removal from 46 NTU to 5.7 NTU (removal 88%) at the OD of 22 mg-Fe/L. In the present study, turbidity removal was equal to or higher than that of literature referred above. However, mean ODs ranged from 30 to 57 mg-Fe/L, which was slightly higher than that reported in the literature. The higher dose required in the present study may be due to variation in raw GW and less rapid mixing. In the present study, the average raw GW turbidity was 78 NTU and rapid mixing was 120 rpm for 90 s. Whereas, Friedler [24] adopted rapid mixing of 100 rpm for 300 s while coagulating raw GW with turbidity 34 NTU. In another study, Friedler and Alfiya [14] adopted rapid mixing of 100 rpm for 300 s for raw GW having turbidity 46 NTU.

In PAC treatment, mean turbidity levels of treated GW were dropped from 88.8 NTU to 2.1 (removal 98%), 2.9 (removal 97%), 2.4 (removal 97%) and 3 NTU (removal 97%) in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively Fig. 3(b). Mean pH of treated GW ranged from 4.6 to 7.9. This indicates that the PAC used was effective for turbidity removal over a pH range of 4.6–7.9. While investigating dye removal from wastewater, Klimuk et al. [16] observed that the

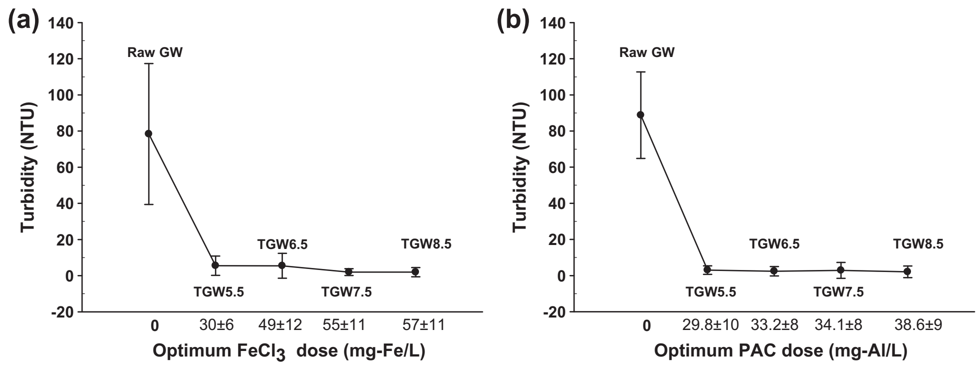

Fig. 3. Effect of coagulant dose on turbidity. (a) Ferric chloride and (b) PAC.

pH had no influence on turbidity removal and similar results were observed in the present study. This indicates that the PAC works equally well over a raw GW pH range of 5.5–8.5. PAC is found better to $FeCl_3$ in turbidity removal at optimum dosage. Moreover, the OD of PAC is quite low compared to $FeCl_3$ (refer Fig. 1).

The properties of the water, particularly pH, alkalinity and temperature are important in coagulation [19,20]. In the present study, a good correlation of iron as well as aluminium dose was observed with treated GW pH, turbidity removal and alkalinity consumed, under all the four pH conditions (Table 4). Correlation coefficient measures the linear association between two variables and takes values between −1 and + 1. Values near zero indicate lack of relationship. Values near + 1 indicate a strong positive relationship; whereas, values near −1 indicate a strong negative relationship. For instance, in the present study, coagulant dose and treated GW pH are negatively correlated in both the coagulants under all the four pH conditions. These correlation coefficients indicate enough scope for further regression analysis among the parameters. Table 5 indicates coefficients (Y_o, c1, c2 and c3) obtained in regression analysis corresponding to the Eq. (2) at the 95% confidence level applicable in the temperature range 23–31˚C:

$$OD = Y_o + c1 \times pH + c2 \times Tr + c3 \times Ac \quad (2)$$

where OD = optimum dose (mg-Fe/L or mg-Al/L); pH = pH of treated GW; Tr = turbidity removal (%); and Ac = alkalinity consumed (%). The above eqution can be helpful for initial trial in the jar test (i.e. adding Ferric chlroride or PAC dose) by targeting % turbidity removal and alkalinity consumption for a particular pH.

Mean TSS concentrations in $FeCl_3$-treated GW were reduced from 164 mg/L to 31 (removal 81%), 20 (removal 88%), 40 (removal 76%) and 31 mg/L (removal 81%) for TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively Fig. 4(a). Friedler and Alfiya [14] reported TSS removal from 70 to 7.4 mg/L (removal 89%). Antonopoulou et al. [15] observed that TSS concentration of 263 ± 103 mg/L was removed by 73% in investigation of light GW by $FeCl_3$ at dose 60 mg/L. In the present study, TSS removal was observed in the range of literature cited. TSS removal was better for TGW7.5 comparing to other three pHs. In the present study, in case of $FeCl_3$ coagulant, as per pH and TSS removal it is evident that charge neutralization and sweep flocculation mechanisms might have played better role in samples TGW8.5 and TGW7.5. Moreover, in sample TGW6.5 and TGW5.5 only charge neutral-

Table 4
Correlation coefficients

	$FeCl_3$ treatment				PAC treatment			
	TGW8.5	TGW7.5	TGW6.5	TGW5.5	TGW8.5	TGW7.5	TGW6.5	TGW5.5
Dose	1	1	1	1	1	1	1	1
Treated GW pH	–0.663	–0.585	–0.807	–0.522	–0.822	–0.850	–0.871	–0.722
Turbidity removal (%)	0.324	–0.161	0.303	–0.918	0.749	0.699	0.653	0.544
Alkalinity consumed (%)	0.499	0.714	0.646	–0.237	0.645	0.536	0.570	0.216

Table 5
Regression analysis

	$FeCl_3$ treatment				PAC treatment			
	TGW8.5	TGW7.5	TGW6.5	TGW5.5	TGW8.5	TGW7.5	TGW6.5	TGW5.5
Y_o	–134.840	288.769	186.254	87.029	188.795	6.689	229.746	135.042
c1	–9.974	3.027	–12.193	–0.018	–11.773	–9.475	–23.645	–19.927
c2	2.528	–3.103	–0.896	–0.561	–0.871	0.820	–0.780	–0.299
c3	0.010	0.676	0.168	–0.099	0.279	0.154	0.272	0.151
Multiple *R*	0.853	0.924	0.863	0.932	0.94	0.96	0.96	0.93
R Square	0.727	0.853	0.744	0.868	0.87	0.92	0.91	0.87
Significance *F*	0.022	0.038	0.033	0.031	0.0016	0.0001	0.0004	0.0017

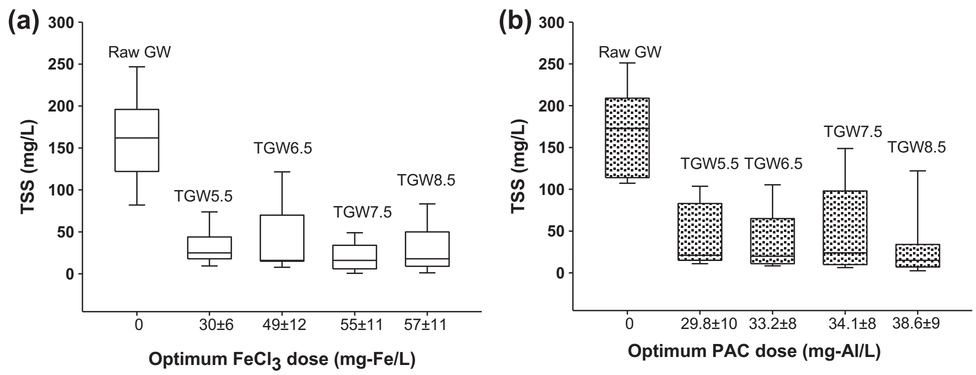

Fig. 4. Effect of coagulant dose on TSS. (a) Ferric chloride and (b) PAC.

ization may be dominating. Treated GW pH was around 6 in first two pH and it was around 5 and 4 in case of TGW6.5 and TGW5.5, respectively. Though the rise in pH reduces neutralizing capacity and affects the TSS removal, a presence of bicarbonate alkalinity is required to form the $Fe(OH)_3$ flocs. Around 0.92 mg/L alkalinity is required per 1 mg/L $FeCl_3$ for complete coagulation reaction [28].

Mean TSS concentrations in PAC-treated GW were reduced from 169 mg/L to 31 (removal 82%), 45 (removal 73%), 40 (removal 76%) and 41 mg/L (removal 73%) for TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively Fig. 4(b). At higher pH, a higher PAC dose is required and the colloids are entrapped in the aluminium hydroxide flocs. The colloids are, then, removed by enmeshment in the amorphous $Al(OH)_3$flocs [16]. In the present study, as per TSS removal, corresponding PAC dose and resultant pH, it is evident that charge neutralization and sweep flocculation mechanisms might have played a better role in the sample TGW8.5 [33]. Whereas, in sample TGW5.5, due to lack of alkalinity only charge neutralization may be dominating.

Mean BOD_5 concentration of 115 ± 23 mg/L was reduced after $FeCl_3$ treatment to 55 ± 22 (removal 52%), 46 ± 27 (removal 58%), 47 ± 22 (removal 59%) and 58 ± 28 mg/L (removal 50%) Fig. 5(a), and COD concentration of 251 ± 45 mg/L was reduced to 125 ± 42 (removal 50%), 105 ± 53 (removal 56%), 92 ± 52 (removal 62%) and 110 ± 64 mg/L (removal 56%) for TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. Friedler and Alfiya [14] reported BOD_5 removal from 103 ± 2.1 to 50 ± 8.6 mg/L (removal 51%) and COD removal from 180 ± 61 to 80 ± 76 mg/L (removal 56%) at a dose of 22 mg-Fe/L. In another study on light GW, Antonopoulou et al. [15] observed COD concentration of 845 ± 167 mg/L was removed by 59% at $FeCl_3$ dose of 60 mg/L. In the present study, BOD_5 removal was similar to that cited in literature; whereas, COD removal in TGW8.5 and TGW 5.5 was slightly lower as compared to the removal reported by Antonopoulou et al. [15]. However, it should be noted that, they had adopted rapid mixing at 100 rpm for 5 min and slow mixing at 25 rpm for 30 min, which was quite higher than that in the present study. BOD_5 removal was significant ($p < 0.05$) under all the four pH conditions examined.

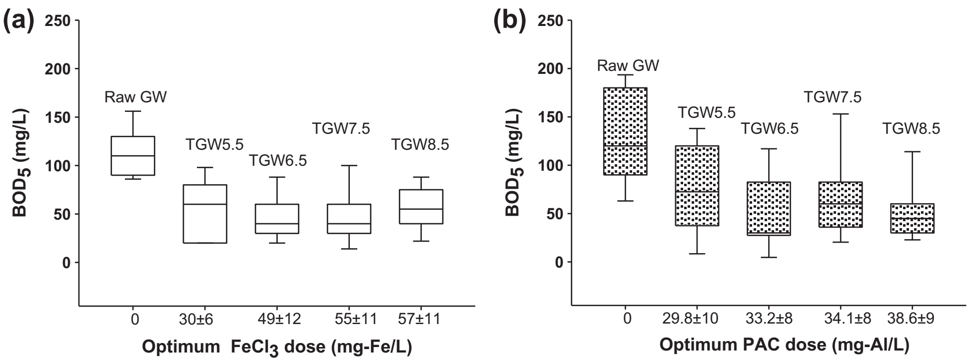

Fig. 5. Effect of coagulant dose on BOD_5. (a) Ferric chloride and (b) PAC.

In PAC treatment, mean BOD_5 concentration of 145 ± 68 mg/L was reduced after treatment to 52 ± 28 (removal 60%), 65 ± 39 (removal 54%), 52 ± 37 (removal 62%) and 83 ± 48 mg/L (removal 42%) Fig. 5(b), and COD concentration of 329 ± 83 mg/L was reduced to 120 ± 56 (removal 62%), 165 ± 85 (removal 50%), 137 ± 102 (removal 59%) and 186 ± 100 mg/L (removal 44%) for TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. BOD_5 concentrations before and after the PAC treatment differ significantly under all the four pH conditions at the 5% level of significance ($p < 0.05$).

In the present study, mean residual levels of the other parameters viz. turbidity, TSS, BOD_5 and COD were low in the GW8.5 samples. The pH of samples TGW8.5 was 6.4 ± 0.5 after treatment. Depending on water temperature and above pH 6–6.5, the system is oversaturated with $Al(OH)_3$. The solution gets theoretically oversaturated with amorphous $Al(OH)_3$, for pH greater than 6.2 at 20˚C. This causes a decrease in the soluble aluminium polymer concentration as aluminium hydroxide precipitates [19]. According to the present study, pH 6.4 may be an optimum pH for treating LGW by using PAC, which is in agreement with the literature cited [19,31].

In $FeCl_3$ treatment, O&G concentration of 57 mg/L was reduced to 22 (removal 61%), 22 (removal 61%), 28 (removal 51%) and 30 mg/L (removal 47%); and ammonia nitrogen (NH_3-N) concentration of 3.2 mg/L was reduced to 1.6 (removal 50%), 1.7 (removal 47%), 2.2 (removal 31%) and 2.4 mg/L (removal 25%) in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. Similarly, in PAC treatment, O&G concentration of 82 mg/L was reduced to 26 (removal 68%), 22 (removal 73%), 30 (removal 63%) and 41 (removal 50%), and NH_3-N concentration of 3.4 mg/L was reduced to 2 (removal 41%), 1.9 (removal 44%), 1.7 (removal 50%) and 1.6 (removal 53%) in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. Performance of PAC was found better comparing to $FeCl_3$ in O&G and NH_3-N removal.

3.2. Effect of coagulant dose on microbiological characteristics

In $FeCl_3$ treatment, mean TC count of 2.2E6 MPN/100 mL was reduced to 2.1E4, 1.5E3, 1.3E3 and 1.2E4 MPN/100 mL; mean FC count of 2.1E4 MPN/100 mL was reduced to 189, 138, 76 and 196 MPN/100 mL in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. Similarly, in PAC treatment mean TC count of 1.3E6 MPN/100 mL was reduced to 5.6E3, 1.2E3, 340 and 361 MPN/100 mL; mean FC count of 2.3E4 MPN/100 mL was reduced to 196, 124, 106 and 88 MPN/100 mL in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. Both TC and FC removal was above 99% in both the coagulant treatments.

Mean *E. coli* count of 1,467 cfu/100 mL was reduced by $FeCl_3$ treatment to 133 (removal 91%), 86 (removal 94%), 52 (removal 96%) and 138 cfu/100 mL (removal 91%); and *E. coli* count of 1,389 cfu/100 mL was reduced by PAC treatment to 113 (removal 92%), 25 (removal 98%), 16 (removal 99%) and 23 cfu/100 mL (removal 98%) in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. In a similar study on LGW, TC count of 56,500 was reduced to < 1 MPN/100 mL (removal > 99%) at OD of 44 mg-Fe/L and 24 mg-Al/L using ferric sulphate and alum, respectively, at pH 4.5 [30]. Similarly, corresponding *E. coli* removal was from 6,490 MPN/100 mL to < 1 MPN/100 mL by both the coagulants. In the present study, FC and *E. coli* removal was significant ($p < 0.05$) for both the coagulants ($FeCl_3$ and PAC) under all the four pH conditions. Both the coagulants ($FeCl_3$ and PAC) were effective in pathogen removal.

3.3. Effect of coagulant dose on metals and ground elements

Concentrations of Na^+, Ca^{2+}, Mg^{2+} and Boron observed before and after $FeCl_3$ treatment are presented in Table 2 and those after PAC treatment are presented in Table 3. These parameters play a critical role when water is used for irrigation purpose. The rate of water infiltration into soil reduces when the Na^+ concentration is high relative to Ca^{2+} and Mg^{2+}. This situation is termed as "sodicity" and results in the decrease in water movement through soil. Therefore, the actively growing plant roots may not be able to get adequate water, despite pooling of water on the soil surface after irrigation [25,29].

Most commonly, sodicity of water is expressed as sodium adsorption ratio (SAR). In the present study, in $FeCl_3$ treatment, mean SAR of the raw GW was reduced from 4.4 to 3.76, 2.96, 2.52 and 2.27, and in PAC treatment from 3.34 to 2.94, 3.23, 2.96 and 2.65 in TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. The mean percentage SAR removal by $FeCl_3$ treatment was better compared to PAC treatment (Fig. 6).

Boron (B) is another important element for water reuse in irrigation and is essential for plant growth. Boron results in toxicity when its concentration exceeds 0.5 mg/L [29]. Concentrations of B and As in raw GW itself were quite low. However, B removal from 13 to 51% was observed in $FeCl_3$ treatment and from 23 to 31% in PAC treatment. Major sources of B and Arsenic (As) at household level are faeces, urine, kitchen wastewater and laundry waste. Since, GW used in the present study did not include those sources, levels of B and As were expected to be low.

3.4. Compliance of treated GW with reuse guidelines and standards

USEPA [34] standards for restricted access area irrigation (areas where public access is prohibited) specifies pH 6–9, TSS ≤ 30 mg/L, BOD ≤ 30 mg/L, FC ≤ 200 MPN/100 mL and Chlorine(Cl_2)≥1 mg/L. The standards for construction (soil compaction, dust control, washing aggregate and making concrete) are

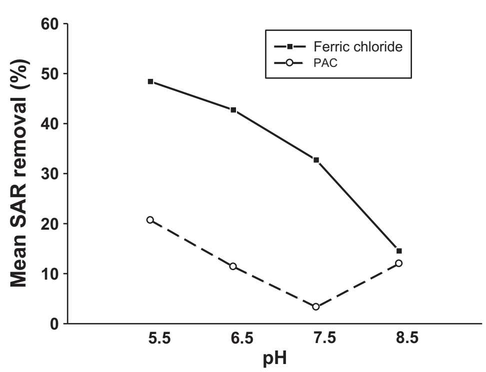

Fig. 6. SAR removal at optimum coagulant dose under different pH conditions.

same as that of restricted access area irrigation, without any restriction on pH. In $FeCl_3$ treatment, mean TSS concentration in TGW7.5 was 20 mg/L and was suitable for irrigation. The median TSS concentrations were observed 18, 16, 16 and 25 mg/L for TGW8.5, TGW7.5, TGW6.5 and TGW5.5, respectively. However, according to median concentrations TSS resulted suitable for both irrigation and construction, under all the four pH conditions. Residual BOD_5 concentrations were not according to reuse standards in any of the pH conditions (see Table 2). FC counts satisfied the irrigation and construction standards under all the four pH conditions (see Table 2). Chlorination of coagulated water is required to maintain minimum residual chlorine of 1 mg/L as per USEPA [34]. In PAC treatment, mean pH (except TGW5.5), TSS and FC of the treated samples satisfy the standards for restricted access area irrigation, under all the four pH conditions (see Table 3). All the treated samples satisfy restricted as well as unrestricted irrigation standards (*E. coli* < 1,000) and are safe from *E. coli* point of view [5] in case of both the coagulants used (see Tables 2 and 3). Though the treated GW is safe for irrigation from *E. coli* point of view, it is recommended that, its intrusion to surface water sources or ground water table should not be allowed to avoid any health consequences likely to arise.

In India, CPCB [35] standards for effluent discharge into land for irrigation prescribes pH 5.5–9.0, TSS < 200 mg/L, O&G < 10 mg/L, BOD < 100 mg/L and As < 0.2 mg/L. In $FeCl_3$ treatment, the pH of treated TGW8.5 and TGW7.5 was within these standards. Mean TSS and BOD_5 concentrations were within these limits under all the four pH conditions examined (see Table 2). O&G concentrations in the treated GW were slightly above standards referred (see Table 2). Pre-treatment of GW such as providing an O&G trap may reduce the O&G concentrations within limit. In PAC treatment, the mean concentrations of pH (except TGW5.5), BOD_5 and TSS of all the treated samples were within these limits (see Table 3). Arsenic level in raw GW was low (< 0.01 mg/L) and within CPCB limits. Another standard, in India, for irrigation, and industrial cooling prescribes EC_{25} < 2,250 μS/cm, SAR < 26 and Boron < 2 mg/L [36]. The GW EC_{25}, SAR and B were well within these limits (see Table 3). Both the coagulants satisfy the criteria for irrigation, and industrial cooling (except pH; in case of TGW5.5) as per CPCB [36].

4. Conclusions

The present study reveals that the role of pH is important and optimum coagulant ($FeCl_3$ or PAC) dose depends on GW pH. Optimum coagulant dose decreased with a decrease in GW pH, indicating high coagulant demand at high pH. The influence of pH is more on $FeCl_3$ dosage comparing to PAC dosage. Ferric chloride was found effective in the removal of turbidity under raw GW pH 7.5–8.5. PAC works equally well over a wide range of raw GW pH from 5.5 to 8.5. Treated GW quality was safe for restricted as well as unrestricted irrigation from FC and *E. coli* point of view under all the four pH conditions in both ($FeCl_3$ and PAC) treatments. GW treated with ferric chloride and PAC can be used for discharge into land for irrigation and industrial cooling in India (after removal of O&G). During the present study, pH of raw GW was never observed 5.5 or below. Therefore, investigations on this pH can be discarded in the future. Most of the times pH was in between 7 and 8, and so, further study can be concentrated in this range.

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