

The response of typical South African raw waters to enhanced coagulation

S. P. Dlamini, J. Haarhoff, B. B. Mamba and S. Van Staden

ABSTRACT

Drinking water treatment plants in South Africa rely almost entirely on surface water sources, which are often compromised due to high return flows and indirect reuse. The typical treatment plants focus on the removal of physical and microbial contaminants which include turbidity, colour, chemical compounds and microorganisms. A relatively new concern to this list is natural organic matter (NOM) which has become a major concern in potable water treatment due to its recent regulation. In this study, eight different raw water samples from the various water types found in the country were seasonally collected and treated for the removal UV absorbance at a wavelength of 254 nm (UV_{254}) using enhanced coagulation (EC). The efficacy of EC, which can be employed as a practical technology in the removal of both turbidity and NOM, was evaluated in remaining UV_{254} from these raw water sources. Jar tests were conducted, with ferric chloride used as the coagulant (due to its extensive use as a coagulant in the water treatment industry in South Africa) and specific pH values (initial water pH, 7.0, 6.0, 5.5, 5.0 and 4.5) were chosen as target values guiding the six different coagulant dosages for the jar tests. The pH of the low-alkalinity (<60 mg/L $CaCO_3$) raw waters were adjusted and raised by the addition of sodium carbonate. The response parameters of the tests were turbidity (NTU), pH and UV_{254} . Algorithms for finding the optimum coagulant dosage for UV_{254} removal were developed and consistently applied to all the results. Results showed large variations in the nature of NOM across the country from specific ultraviolet absorbance values. From the UV_{254} values, the concentrations of NOM also varied greatly geographically than temporally. The general trend observed in the EC results suggested that the pH should always be dropped to between 4.5 and 7.0 to lower the amounts of UV_{254} and turbidity to reasonable levels.

Key words | alkalinity, enhanced coagulation (EC), ferric chloride, natural organic matter (NOM), turbidity, UV absorbance

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INTRODUCTION

Natural organic matter (NOM) is a chemically complex and heterogeneous mixture of organic material broken down from decaying plant and animal origins. The character and amount of NOM are in many instances considered important for the determination of coagulant dosage and coagulation efficiency (Hwang *et al.* 2000). Various characterisation techniques have been formulated, and more are still under development, to give more understanding to the nature of NOM itself and its effect on water treatment processes. NOM is crudely quantified as total organic carbon

(TOC) which is the combination of particulate organic carbon and largely dissolved organic carbon (DOC). The TOC and DOC are generally measured using an organic carbon analyser, which in most cases is automated. The principle governing its usage requires the performance of multiple analytical steps done sequentially (Nkambule 2008). This makes this quantification method not suitable for small water treatment utilities due to the high costs and operational complexity that comes with it. One of the simple techniques is the measurement of surrogate

parameters, such as a single UV absorbance at a wavelength of 254 nm (UV_{254}). This technique has been widely accepted by water treatment operators as parameters to assess the performance of a plant with regard to NOM removal.

Conventionally, the drive to remove NOM from potable water would be the desire to remove colour from public water supplies. However, more problems in drinking water treatment associated with NOM have been identified. These include taste and odour, its tendency to foul membranes, interference with the removal of other contaminants and its potential to contribute to corrosion and slime growth in distribution systems. Moreover, it causes high demands for coagulants and disinfectants (Rizzo *et al.* 2005; Harrington *et al.* 1997). In recent years, both the number and stringency of drinking water regulations have been increased world wide. The current South African National Standards (SANS 241 2011) have stipulated the requirements of treated water NOM (10 mg/L TOC), which could become more stringent under future revisions. This has brought forth the need to employ and/or improve various treatment techniques for the reduction of NOM to meet the requirements.

This study investigates the efficacy of enhanced coagulation (EC), a viable option, in the removal of these organic substances from typical source waters of South Africa. EC is an extension of an existing treatment process 'chemical coagulation', which has conventionally been used with metallic salts to remove only particles (turbidity and pathogens) from water. Enhanced coagulation, a process to meet regulatory standards, ensures maximal removal of particles and NOM with the usage of coagulants such as aluminium and iron salts (Edzwald & Haarhoff 2012; Xie *et al.* 2011). The iron salt is used in this study for its common application in South Africa (SA). The removal efficiency of EC, however, depends on the physical and chemical properties of the water, as well as the operating conditions (Ghasri *et al.* 2005). Optimum conditions for the removal of both turbidity and NOM sometimes differ, with the generally higher dosage required for NOM removal as the controlling factor. However, turbidity removal at these conditions must still be achieved (Gao & Yue 2005).

In this study, a consistent and reproducible, yet simple jar test procedure was developed to find the optimum

coagulation conditions for the removal of UV_{254} . Batch tests on all samples were then performed to evaluate the removal efficiency of UV_{254} and turbidity at the optimum conditions of dosage and pH. The findings reported in this study, therefore, are based on bench-scale tests performed at ambient temperature (15–23 °C).

MATERIALS AND METHODS

Site selection and sample collection

Eight sites for sample collection were selected through a semi-random process based on the main surface water types encountered in SA. Five distinctively different raw water types were identified from the perspective of NOM removal. These included the following. (1) Oligotrophic water from Vaal Dam, a reservoir which supplies the largest water supply authority in the country. The NOM in this water type is characterised of surface water from a reasonably well protected highveld catchment area with minimal return flows. (2) Water emanating from warmer areas with high population densities, with a fairly high NOM load. (3) Coloured water from the south-western coast, very high in colour, humic and fulvic acid. (4) Eutrophic water from the highveld, with typically high NOM loads. The eutrophication is being driven by large return flows and agricultural runoff. (5) Treated sewage effluents, which dominate the NOM character in many streams and rivers in SA during periods of low flow. A random selection was made to ensure a representation of these water types, as well as a balanced spatial representation.

A total of 24 samples were collected at these sites over a period of 8 months (July 2010–February 2011), at about 3 month intervals representing winter, spring and summer (1, 2 and 3 respectively). As a measure to minimize errors, the samples were given codes derived from the first letter of the location name it was collected, and a numeric adjective denoting the season. The raw waters were collected into 25l containers before any pre-treatment, and were transported to the laboratory as quickly as possible and were stored in the dark at 4 °C to reduce biological activity until testing/experiment.

Analytical methods

The pH and temperature ($^{\circ}\text{C}$) were measured using a HANNA HI 98130 combo water proof pH, EC/TDS and temperature meter. Turbidity, in nephelometric turbidity units (NTU), was measured using a HACH 2100 turbidity meter. DOC concentrations in raw water samples were measured using a total organic carbon analyser (Teledyne Tekmar, TOC fusion). UV absorbance at 254 nm wavelength was measured using an ULTROSPEC II: UV/Vis spectrophotometer (Model 80-2091-73, Biochrom, UK) with 1 and 5 cm cuvette cells. DOC and UV_{254} samples were filtered through non-sterile 33 mm MILLEX-HV MILLIPORE 0.45 μm filter units. The specific ultraviolet absorbance (SUVA) was expressed as a ratio between UV_{254} and DOC values. Alkalinity (mg CaCO_3/L) and calcium hardness (mg Ca/L) were determined using protocols 403 and 311 C, respectively, outlined in [Standard Methods \(16th edition, 1985\)](#).

Reagents

Hydrochloric acid (HCl) 0.1 and 0.02 N were used for alkalinity titrations. Standard EDTA (0.01 M), Eriochrome Blue Black R indicator (stable form) and sodium hydroxide (NaOH), 1 and 8 N were used for the calcium titration.

The metal coagulant used was ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) and it was prepared from reagent grade granular ferric chloride in the laboratory. Standardized sodium carbonate (Na_2CO_3) was used to increase the alkalinity of the extremely low alkalinity samples.

Jar testing

An FC6S jar test apparatus (VELP SCIENTIFICA) was used for coagulation and flocculation experiments in 1 L round beakers containing 900 ml samples of water. The standard jar test protocol used for all jar testing had a 2 min rapid mix at 200 rpm after coagulant addition, followed by a 10 min flocculation at 30 rpm and a 30 min settling at 0 rpm. During jar tests, paddles were completely submersed in the sample. Settled water samples were collected from the centre of each jar using 50 ml pipettes with a 90° bend at the tip.

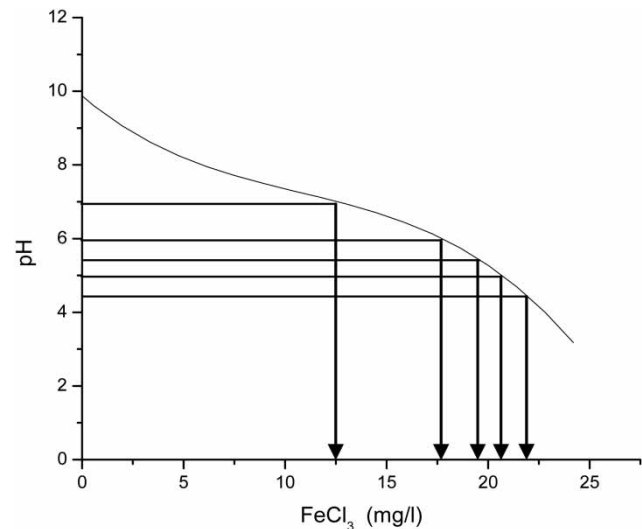


Figure 1 | Coagulant titration curve with ferric chloride (alkalinity 16 mg CaCO_3/L) after the addition of sodium carbonate.

To determine the amount of coagulant to be added in the different jars during jar testing, titration curves either with a 0.1 N concentration of HCl acid or with the coagulant itself were developed. [Figure 1](#) shows an example of a coagulant titration curve with ferric measured as FeCl_3 . Dosages corresponding to specific pHs (7.0, 6.0, 5.5, 5.0 and 4.5) were interpolated from similar curves as shown, with the first beaker acting as the control hence no coagulant added in it. However, since the practical goal of most water treatment plants in practice is to maintain an alkalinity greater than 40 mg CaCO_3/L and a calcium carbonate precipitation potential (CCPP) ranging between 3 and 6 mg/L, EC should be performed with a reasonably high alkalinity (approximately 60 mg CaCO_3/L). If the alkalinity of the water was low, theoretical amounts of base [Na_2CO_3 obtained from a STASOFT4 model] were added in to reach the 'minimum' alkalinity level. Titrations with either 0.1 N HCl or ferric were done again as with the high alkalinity waters.

Batch testing

These tests were done subsequently to jar testing. The same coagulation, flocculation and settling times and rates as for jar testing were used. To determine the optimised coagulation dosage and its resultant (optimum) pH, jar tests results were used. UV_{254} curves plotted against pH-specific dosage, as

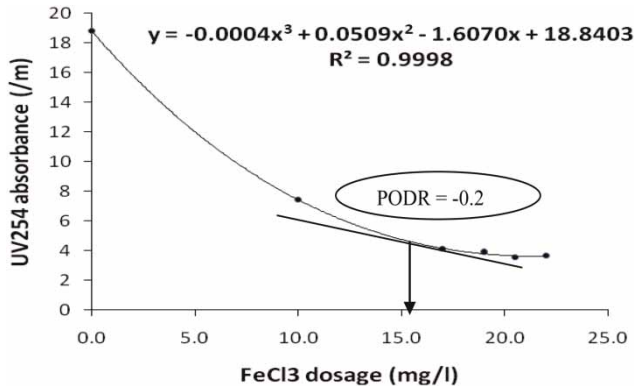


Figure 2 | Effect of coagulant dose on UV₂₅₄ removal (UV₂₅₄ reduction rate of 2 m⁻¹ per increasing 10 mg/L of ferric chloride).

shown in Figure 2, were used to develop three criteria for choosing the optimal coagulation conditions of dosage and pH. Most water treatment utilities from the literature, aim at UV₂₅₄ values of between 4 and 7 per metre (m⁻¹), the first criterion would then be aiming at reducing the water's UV to an absolute 6 m⁻¹ value. Some water sources, not only in SA, have low initial UVs close to or below this absolute value, the second criterion would then be reducing 65% of the initial UV₂₅₄ value from the basis that any removal of UV₂₅₄ is better than no removal at all. This percent removal, however, was arbitrarily chosen. The highest coagulant dosage corresponding to any of the two criteria would be chosen as optimal and it would be interpolated from the curve. The last criterion would be based on the point of diminishing returns (PODR), a concept from the US Environmental Protection Agency (USEPA 1999) enhanced coagulation/precipitative guidance manual. The dosage corresponding to the curve's tangential slope of -0.2 would be used as the 'break-even point' or the ceiling limit. Thus, if it was less than one or both of the other dosages, it then became optimal.

RESULTS AND DISCUSSION

Raw water quality profiles

Table 1 shows the bulk water properties of all the raw water samples used in this study. The first four raw water quality profiles are for waters classified as having low alkalinity (<60 mg CaCO₃/L) ranging from as low as 3 to 59 mg CaCO₃/L. The last four samples are for waters classified

Table 1 | Characteristics of raw waters examined

Sample ID	pH	Alk. (mg/L)	Ca ⁺⁺ (mg/L)	UV ₂₅₄ (m ⁻¹)	DOC (mg/L)	SUVA (L/mg m)	Turbidity (NTU)
P1	6.2	3.3	5.1	31	7.3	4.2	0.8
P2	7.6	12	5.6	42	9.2	4.6	0.5
P3	5.7	3.0	4.8	76	12	6.3	3.4
L1	7.5	16	14	17	7.1	2.3	3.0
L3	7.1	19	11	14	6.9	2.0	1.3
W1	8.1	54	26	7.3	4.6	1.6	1.2
W2	8.4	58	16	6.0	3.2	1.9	1.0
W3	8.7	58	17	6.1	3.2	1.9	0.9
V1	7.9	59	22	20	8.3	2.4	74
V2	8.0	58	17	18	8.0	2.3	56
V3	8.1	53	15	30	9.6	3.2	88
O1	8.2	93	50	15	5.8	2.6	1.5
O2	8.7	99	43	16	7.0	2.3	1.8
O3	8.5	88	39	15	6.7	2.3	2.6
M1	9.0	152	74	16	6.1	2.6	6.5
M2	8.7	136	75	18	7.5	2.4	2.6
M3	7.8	67	31	36	10	3.5	85
R1	8.3	121	41	16	5.9	2.8	1.0
R2	9.3	136	44	19	9.6	2.0	11
R3	7.8	97	38	24	8.1	2.9	1.6
U1	8.4	131	28	15	6.2	2.4	17
U2	8.5	129	29	17	7.2	2.3	9.2
U3	8.8	107	34	19	8.0	2.4	24

as moderate to high alkalinity (>60 mg CaCO₃/L) ranging from 67 to 152 mg CaCO₃/L. NOM surrogate values, UV₂₅₄ and DOC, ranged from 6 to 76 m⁻¹, and 3 to 12 mg/L respectively.

The corresponding SUVA values varied from 1.6 to 6.3 L/mg m. In general, SUVA gives the aromaticity or the hydrophobicity of NOM in a water body (Matilainen *et al.* 2010). Edzwald & Tobiasson (1999) suggested that samples with SUVA values: less than 2 L/mg m have NOM mainly composed of a high fraction of non-humic substances with a low UV₂₅₄; between 2 and 4 L/mg m have a mixture of aquatic humic and non-humic substances with a medium UV₂₅₄; and greater than 4 L/mg m have a high fraction of aromatic and hydrophobic matter with a high UV₂₅₄. All the P samples had SUVA values greater than 4 L/mg m, indicating that the DOC of the water is composed of a high

fraction of aquatic humic matter, which makes it amenable to coagulation. At the other extreme, the W samples had SUVA values less than 2 L/mg m, indicating that the DOC of the water is mainly composed of a high fraction of non-humic matter, hence low percent removals of UV₂₅₄ would be expected. Otherwise the SUVA of the rest of the waters ranged between 2 and 4 L/mg m, thus moderate to relatively high UV₂₅₄ percentage removals would be expected. The turbidity of the waters also showed large differences ranging from about 1 to 88 NTU.

Jar test results

Simple analytical techniques, including the single wavelength UV₂₅₄ measurement and colour (UV₄₅₆) are instrumental in monitoring the NOM content of water,

and are widely accepted by water treatment operators as parameters to assess the performance of water treatment plants. The measurement of these parameters is quick, and enables easy characterization of samples, and the spectrophotometers for both laboratory and field instruments are widely available and affordable (Yan *et al.* 2006). UV₂₅₄ measurements, in particular, provide a quick and precise indication of the organic carbon content of raw and treated water as TOC or DOC (Stephenson & Duff 1996). Moreover, various researchers have investigated the relationship and found good correlations between the UV₂₅₄ and DOC of raw and treated waters indicating that perhaps the treated water UV₂₅₄ could also be used as a reliable surrogate parameter of NOM and to predict the treated water DOC (Edzwald & Haarhoff 2012; Chow *et al.* 2007; Garcia & Moreno 2006).

Table 2 | Jar testing results (ferric chloride dosage required to meet EC criteria)

Sample ID	Na ₂ CO ₃ (mg/L)	FeCl ₃ (mg/L)	Criterion	pH	UV ₂₅₄ (m ⁻¹)	UV ₂₅₄ removed (%)
P1	61	16	Absolute target	5.2	6.0	81
P2	51	14	Absolute target	5.4	6.0	86
P3	61	17	Absolute target	5.0	6.0	92
L1	47	12	Absolute target	7.1	6.0	68
L3	50	16	PODR	6.1	5.1	65
W1	–	7.9	PODR	6.9	3.2	33
W2	–	6.7	PODR	7.0	3.0	29
W3	–	8.6	PODR	6.7	3.3	35
V1	–	15	Absolute target	6.5	6.0	76
V2	–	14	Absolute target	6.2	6.0	73
V3	8.0	16	Absolute target	6.1	6.0	79
O1	–	29	PODR	5.9	6.7	56
O2	–	30	PODR	6.0	6.2	61
O3	–	22	PODR	6.2	7.5	50
M1	–	23	PODR	7.2	7.8	46
M2	–	29	PODR	6.3	7.6	50
M3	–	19	Absolute target	6.0	6.0	72
R1	–	23	PODR	6.6	7.5	54
R2	–	38	Absolute target	5.6	6.0	68
R3	–	29	Absolute target	5.9	6.0	75
U1	–	22	PODR	6.8	6.9	47
U2	–	23	PODR	6.6	6.9	53
U3	–	31	PODR	6.1	6.0	68

Table 2 shows a summary of jar testing results on all the water samples. The three criteria were used simultaneously to interpolate the optimal dosages and their resultant pHs for batch tests. The residual UV_{254} values were also predicted with their equivalent percent removals. The optimal coagulant dosage in about half of the cases corresponded to the absolute 6 m^{-1} target and in the other half, the PODR was the controlling factor. The 65% removal criterion seemed not to control the optimal dosage. This means that the dosage corresponding to it was not, in any case, higher than the dosage corresponding to an absolute 6 m^{-1} target. If the same concept is to be employed, perhaps higher percentage values are to be used. The theoretical or predicted values of coagulant dosage from criteria were generally lower in the low alkalinity waters as compared to the high-alkalinity waters. In terms of UV_{254} the predicted percentage removals showed that in general, low alkalinity waters were more amenable to coagulation than high alkalinity waters. This was also true with the same source waters collected at different times. Clear examples are the M3, R3 and U3 samples, which had low alkalinities as compared to the other M, R and U samples. From the optimum coagulant dosages, these tests predicted matching 'optimum' coagulation pH values ranging from about 4.5 to 7.0. Theoretically, these 'optimum' conditions of coagulant dosage and pH are dependent on the initial UV_{254} , DOC and the bulk water properties like alkalinity and hardness.

Batch test results

All samples were analysed as described before and Table 3 gives a summary of the results. The overall settled water quality after these tests to improve UV-absorbing compounds removal also resulted in reasonable reduction of turbidity. As a consequence, meeting criteria for enhanced coagulation does not compromise the removal of turbidity.

For 21 of 23 sites, the residual turbidity was reduced to levels lower than 1.7 NTU irrespective of the initial or raw water turbidity. However, in two low-alkalinity cases (P1 and P2), the measured turbidity after the optimised dosage of UV_{254} removal was more than raw water turbidity. This could be because of the very low starting turbidity values of the samples which are 0.8 and 0.5 NTU respectively.

Table 3 | Batch testing results (values measured after the tests)

Sample ID	pH	UV_{254} (m^{-1})	UV_{254} removed (%)	Turbidity (NTU)
P1	4.9	7.9	72	4.3
P2	5.0	4.1	90	1.7
P3	4.7	4.4	94	1.3
L1	5.8	5.7	66	0.7
L3	5.8	5.2	63	0.8
W1	6.5	4.6	33	0.5
W2	6.6	4.7	29	0.5
W3	6.5	4.6	35	0.3
V1	6.1	6.0	76	1.0
V2	6.1	6.0	73	0.9
V3	5.9	6.4	79	1.7
O1	5.7	6.4	58	1.2
O2	5.8	7.4	54	1.1
O3	6.1	7.7	49	0.8
M1	6.5	7.8	46	1.1
M2	6.4	8.7	50	1.0
M3	6.0	7.3	72	2.3
R1	6.5	7.6	53	0.8
R2	6.0	7.3	61	0.4
R3	5.9	6.8	71	0.7
U1	6.5	7.8	57	0.7
U2	6.5	7.8	60	0.8
U3	5.9	6.0	69	0.8

Nevertheless, in all cases the residual turbidity was low enough for subsequent treatment steps like filtration.

The residual UV_{254} of all samples, expressed as the percentage removal, showed dependence on raw water SUVA regardless of water type and source as shown in Figure 3. A pattern from a plot of SUVA and UV_{254} percent removal values, though not linear in these results, is illustrated and is confirmed by a logarithmic function through the points giving a correlation coefficient of 0.6.

Similar observations were also made with pH as in Figure 4, where UV_{254} removal percentage increased with decreasing pH in the range between 4.5 and 7.0. This suggests that UV_{254} removal of 60% or higher will only be attained if the coagulation pH is 6 or lower, within the given range. To the contrary, as illustrated by Figure 5, the

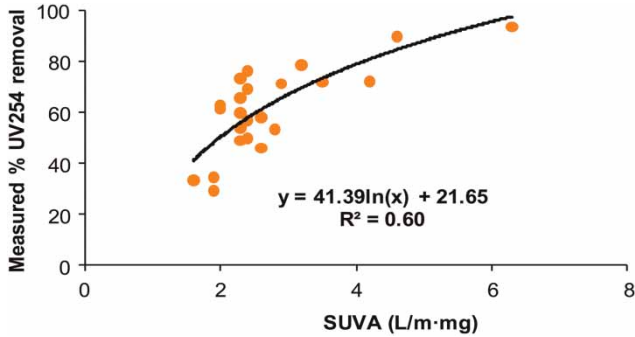


Figure 3 | Raw water SUVA and measure UV₂₅₄ percentage removal.

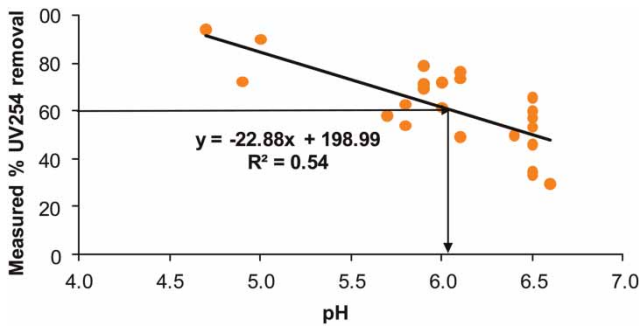


Figure 4 | pH and measured UV₂₅₄ removal percentage.

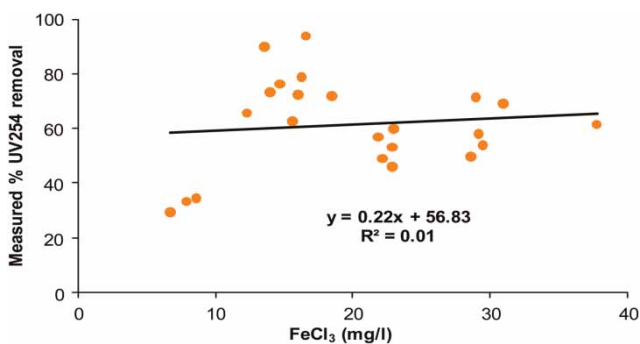


Figure 5 | Enhanced coagulation dosage and measured UV₂₅₄ removal percentage.

correlation between coagulant dosage and UV₂₅₄ percent removal was 0.01, clearly showing that coagulant dosage could not be the controlling factor for enhanced coagulation.

Several researchers have found the coagulation pH to be the parameter having a greater effect on achieving optimal NOM removal by coagulation (Bell-Ajy *et al.* 2000; Qin *et al.* 2006; Yan *et al.* 2008; Xie *et al.* 2011). This could

be linked to the varying distribution of the metal coagulants' species, and their hydrolyzing behaviour in raw water with low temperature (Sharp *et al.* 2006; Yu *et al.* 2007; Sulaymon *et al.* 2009). USEPA recognized the coagulation pH to be determined by the raw-water alkalinity. Results from this study are in agreement with these findings, indicating that the coagulation pH was another controlling factor for UV₂₅₄ removal rather than the coagulant dose for the varying-alkalinity samples analyzed.

Figure 6 shows a relationship between the predicted pH values from jar tests' curves and the actual or measured pH values from batch tests. Under perfect conditions, a straight line fit of the distribution passing through the origin would have a slope of 1.00 and pH results from this study a slope of 0.96 and a correlation constant of 0.83 which is relatively high. The same analysis was done for UV₂₅₄ as in Figure 7. The UV₂₅₄ results had an even better slope and correlation constant with values of 0.99 and 0.93 respectively. This approach or 'model' showed good predictive ability when applied to predicting optimum coagulation pH and the resultant UV₂₅₄ percent removal.

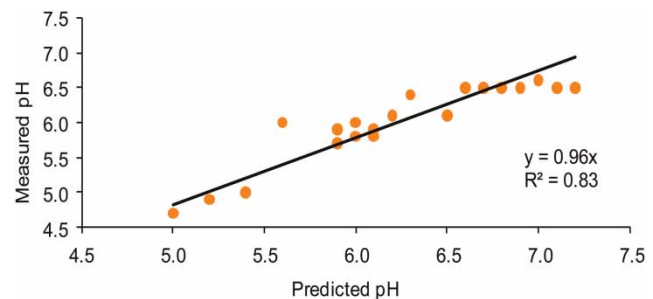


Figure 6 | Predicted and measured pH values of all waters.

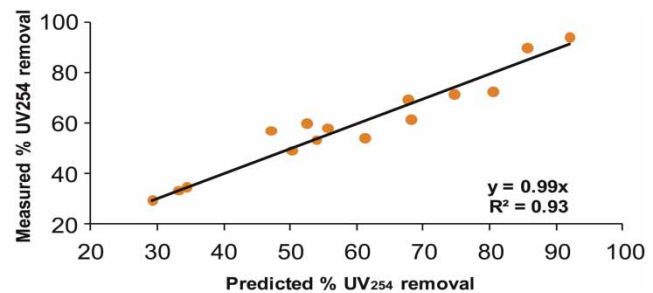


Figure 7 | Predicted and measured UV₂₅₄ percentage removals.

CONCLUSIONS

- Turbidity was reduced to levels low enough for the filtration treatment process at the dosages adopted for enhanced coagulation, hence it is not compromised by enhanced coagulation.
- Raw water SUVA and coagulation pH were the noticeable factors greatly affecting enhanced coagulation.
- A titration curve can be used with success to target specific pH values for jar testing purposes. The pH values and UV₂₅₄ removal percentage interpolated from the jar tests closely matched those from independent batch tests.
- Optimum dosage for enhanced coagulation, for all samples irrespective of water source and season, corresponded to pH values between about 4.5 and 7.0. Waters of low alkalinity require the addition of lime to maintain the pH in this range.

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