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Enhanced Flocculation Using Drinking Water Treatment Plant Sedimentation Residual Solids

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Abstract: Inefficient removal of total organic carbon (TOC) leads to the formation of carcinogenic disinfection by-products (DBPs) when a disinfectant is added. This study is performed in an effort to develop a simple, non-invasive, and cost-effective technology that will effectively lower organic precursors by having water utilities reuse their treatment residual solids. Jar tests are used to simulate drinking water treatment processes with coagulants—aluminum sulfate (alum), poly-aluminum chloride (PACl), and ferric chloride and their residual solids. Ten coagulant-to-residual (C/R) ratios are tested with water from the Missouri River at Coopers Landing in Columbia, MO versus alluvial ground waters. This treatment results in heavier floc formation and leads to improved sedimentation of organics and additional removal of aluminum and iron. An average of 21%, 28%, and 33% additional TOC removal can be achieved with C/R ratios <1 with alum, PACl, and ferric chloride, respectively.

Keywords: treatment residual solids; coagulation; flocculation; TOC removal; turbidity; disinfection by-products

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1. Introduction

Percentage removal of natural organic matter (NOM) affects the efficiency of drinking water treatment process [1]. More specifically, NOM reduction may influence how effective disinfection using chlorine is in terms of amount of disinfectant needed and the concentration of disinfection by-products (DBPs) formed as a result [2–4]. It is known that biologically refractory humic and fulvic acid fractions of NOM, generally of allochthonous origin, are most reactive with chlorine [5].

A typical drinking water treatment plant uses a sequence of coagulation, flocculation, sedimentation, filtration, and disinfection unit processes to treat incoming raw water with an aim to decrease the total organic carbon (TOC) and turbidity to a required limit [6,7] and produce finished water with little to no taste and odor issues. In addition to these parameters the finished water is also expected to comply with the rules such as total coliform rule, maximum residual disinfectant level (MRDL), and stage-II D/DBP rule. Many feasible distribution system models have been investigated in recent years to help utilities with operational decision-making [8], but advanced technologies to increase the TOC removal efficiency have mostly had a high price and complexity associated with them. Achieving a balance between TOC reduction and chlorination has been a great concern to both environmental officials and the utility managers since the late 1970s when research began to show a direct link between trihalomethane (THMs) formation and reaction between TOC and free chlorine [5].

Coagulants such as aluminum sulfate and ferric chloride are used for coagulation and the floc that is formed as a result is allowed to settle down in a sedimentation basin before the water enters the filtration stage where the disinfectant is added. As observed in Missouri, in many small-scale utilities the floc formed is usually too small or light in weight for it to settle down and travels beyond the final clarification step into the filtration stage resulting in more frequent back wash cycles [9]. This also allows for higher concentrations of TOC to react with chlorine, forming higher concentrations of DBPs even before the finished water leaves the system [10,11]. Factors such as detention time in first and second stage basins in the 2-stage water treatment process, floc thickness (robust floc is denser and will settle out easily), raw water quality and coagulant type, and concentration used can dictate how efficient the treatment process is in removing TOC.

There are many advanced treatment technologies such as granulated activated carbon (GAC), enhanced coagulation, membrane filtration, and MIEX that provide efficient TOC removal [12], but are economically infeasible for small-scale utilities. This study is performed as an effort to develop a novel, simple, non-invasive, and cost-effective technology that will effectively lower the organic precursors/TOC while helping the water utilities to reuse their treatment residual solids that would otherwise need to be disposed of. These residual solids are composed of mostly un-used coagulant, settled organic materials (suspended and dissolved), and impurities.

There are many aspects that need to be taken into consideration when recycling residuals including, but not limited to the feasibility of this approach for a continuous process, the cost benefits of this technology and its impact on microbial quality of the water. This lab-scale study is conducted using a well-known method among utility community (jar tests) as a proof of concept to provide basis for future pilot scale continuous process studies as at a treatment plant. Though many operators use jar tests as a basis for their coagulant dosing decisions, it would be beneficial to conduct a pilot study when introducing a new technology such as residuals recycling. Some, but very limited information is currently available about impact of residuals recycling on microbial water quality. D'Adamo et al. conducted a full-scale evaluation of multiple residuals management options and recycling was investigated as part of it. The dissolved oxygen and coliforms results from the study determined recycling to be a potential safe option for residuals management [13]. Further investigation in this area during pilot scale study is needed to evaluate the full spectrum of microbial parameters such as bacterial number and biodegradable dissolved organic carbon (BDOC). Another aspect that needs future examination is cost-benefit; this proposed treatment can potentially decrease the amount of coagulant used by finding an optimal coagulant to residual dosing ratio thereby providing cost savings to a utility. This optimal ratio will differ for each utility depending on their raw water quality, unit process treatment efficiency, residual composition, and residuals recycling mechanism and so will their extent of cost savings.

It is known that even an increase as little as 0.5 mg/L in TOC concentration with right precursors can lead to DBP compliance issues for the treatment plant, therefore any additional TOC removal beyond this is used as the criterion for effectiveness of the proposed treatment.

2. Materials and Methods

This research was conducted using jar testing to simulate drinking water treatment unit processes in the laboratory. Jar tests are a widely accepted and used method for understanding the effect of treatment process changes at lab scale [14] and water treatment utilities are often able to replicate the results at plant scale. Standard jar test allows for simulation of rapid mix during addition of chemicals, flocculation, and sedimentation under constant temperature and pressure conditions.

Raw water from the Missouri River and from an alluvial ground water located in the McBaine Bottoms near the City of Columbia, Missouri (USA) was collected over a period of 1 year to capture water quality changes during fall, spring, and summer seasons. Characteristics of incoming water and residuals are presented in Table 1.

Table 1. Characteristics of incoming water.

Source	pH	TOC (mg/L)	Turbidity (NTU)	Aluminum (mg/L)	Iron (mg/L)
River Water	(6.9–8.45)	(5.8–10.4)	(20.9–445)	*	*
Alluvial GW	(7.1–7.52)	(4.0–5.1)	(20–28)	0.07	5.34
Sludge Type	**pH**	**% Solids**	**Alkalinity (mg/L)**	**Al Ion (mg/L)**	**Fe Ion (mg/L)**
Alum	7.06	11.0	56	0.565	0.13
PACl	7.15	15.1	60	0.175	0.18
Ferric	7.3	43.9	58	0	0.9

* Testing not conducted.

Aluminum sulfate (alum), poly-aluminum chloride (PACl), and ferric chloride ($FeCl_3$) from Hawkins Inc., Columbia, MO are the three commonly used coagulants in the water industry, and their respective residual solids (sludges) are used for jar testing. Three coagulants and their respective residual solids were collected from three treatment plants on the same day, making them relatively similar in age. Nine coagulant-to-residual (C/R) ratios (4, 2, 1, 0.66, 0.5, 0.4, 0.33, 0.28, and 0.25) in addition to a control (coagulant with no residual solids) were tested with all three coagulants and both of the raw waters collected during every season. Coagulant doses of 100–150 mg/L were used for alum and ferric chloride and 20–30 mg/L was used for PACl. These are the typical doses that plant operators use in Missouri. Four times the amount of coagulant to one part of residual results C/R equal to 4. Metal ion concentrations in residuals are usually used for comparison and results analysis. Different treatment plants can produce residuals with different characteristics such as metal ion concentration, pH, alkalinity, percent solids etc., depending on the site-specific conditions such as detention time, presence of algae, use of pre-oxidant, source water quality etc. Seasonal variations in source water can also affect the residual characteristics at the same treatment plant. Therefore, a simple ratio of constant coagulant concentration to varying residuals (dry weight of precipitate) was used in this work to make the practical application of the proposed technology more feasible for utilities of all sizes. Utilities will need to perform jar tests with their own coagulant (at same concentration/weight they usually use) and varying residuals weights to come up with a ratio that works best for their system.

The jar test method used involves addition of coagulant and residual solids together to all jars simultaneously, which is followed by mixing for 30 s at 100 rpm (revolutions per minute) to simulate rapid mix, 30 min at 35 rpm to simulate flocculation, and finally 30 min at 0 rpm for settling/sedimentation. Samples were collected for water quality testing before and after the jar test from each jar. Noteworthy, the coagulant concentration was maintained constant throughout the process with residual solids concentrations being the only fluctuating variable making it easier to interpret the effect of residual solids on the treatment efficiency of the process.

UV_{254}, pH, TOC, turbidity, aluminum, and iron tests were conducted on the source and effluent water from each jar test. Every jar test was repeated twice for redundancy and all samples were collected in triplicate for each of the above water quality tests. UV_{254} was measured using a Varian Cary 50 Conc. UV-visible spectrophotometer from Varian, Inc., Milpitas, CA, USA following standard method 5910 B [15]. TOC was measured using the combustion infrared method following standard method 5130 B [15]. A Hach DR5000 from Hach Company, Loveland, CO, USA was used to measure aluminum and iron concentrations in the samples following Hach method 8012 and 8008, respectively. Turbidity was measured using Hach method 8195 and Hach method 8196 was used to measure the pH [16].

Percentage additional TOC removal for each C/R ratio tested was calculated as the percentage difference between the TOC concentration of effluent water using just the coagulant (no residual solids) and that of water treated with C/R ratios as shown in Equation (1).

$$\text{\% TOC removal (additional)} = \text{\% (Effluent TOC (coagulant only)} - \text{Effluent TOC (C/R))} \quad (1)$$

3. Results and Discussion

3.1. C/R Ratios and TOC Removal

This analysis is intended to illustrate the effect of each C/R ratio tested on water quality in terms of TOC removal. Data from jar tests using nine C/R ratios, alum, PACl, and $FeCl_3$ as coagulants and their respective residual solids with Missouri River and alluvial ground water as incoming water is presented below. Though water from different seasons has been tested, similar results were obtained. The overall average of the results is presented in this section. Efficiency analysis is done in terms of percent additional TOC removal with respect to the control (coagulant with no residual solids).

Data from the jar tests with nine C/R ratios on Missouri River show that an additional reduction of TOC can be achieved with alum, PACl, and $FeCl_3$ when the ratio is <1.0. This pattern can be clearly seen when the TOC concentration in effluent jars with C/R < 1.0 are compared against their respective controls as shown in Figure 1. This reduction can be attributed to the weight the residual solids added to the system allowing for heavier floc formation. Heavier floc leads to reduced carry-over, thus improving the sedimentation efficiency of the basins. When C/R > 1.0 (that is, when the concentration of residual solids is lower than that of the coagulant), the efficiency of the treatment process decreases. The TOC concentrations of the effluents with all three coagulants compared to their respective controls is higher by at least 26%. This implies that when residual solids concentration is less than the coagulant concentration, the amount of additional weight is not enough for this approach to make a difference. Therefore, instead of increased TOC removal, the system is under-performing, resulting in worse water quality with respect to their controls by at least 26%.

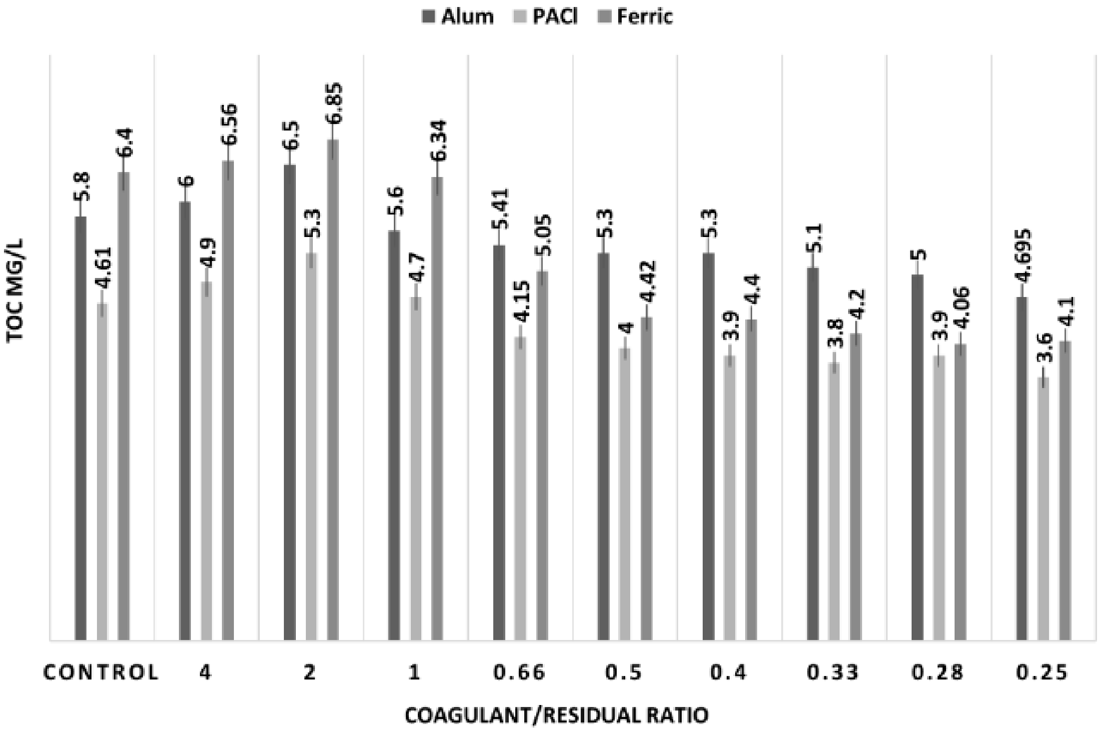

Figure 1. Surface water total organic carbon (TOC) removal using nine coagulant-to-residual (C/R) ratios.

When C/R = 1.0, the system is either working similar to the control or better, but not significantly better. The differences in quality of effluent from the jars in this case depend more on the type of coagulant used rather than the ratio with which it is treated.

Analysis of the data shows that a 3.5–23.1% additional TOC removal can be achieved with C/R < 1 for alum, a 10.5–24.6% for PACl, and 1.0–44.7% for $FeCl_3$ relative to the control. Average percent additional TOC removal for alum, PACl, and $FeCl_3$ are 17%, 20%, and 33%, respectively, for all C/R ratios < 1. It can be concluded that when enough residual solids are added into the system with constant coagulant concentration ([R] > [C]), a considerable additional TOC decrease can be achieved resulting in better finished water quality. The optimum C/R ratio differs with each coagulant used and the raw water quality coming in; hence extensive jar tests need to be conducted before choosing a ratio that works best for a specific system.

From the initial nine C/R ratios tested, five best ratios for each coagulant were chosen and the jar tests were repeated with water collected from a different season. The best ratios for alum are 1, 0.5, 0.33, 0.28, and 0.25, for PACl are 1, 0.5, 0.33, 0.28, and 0.25, and for $FeCl_3$ are 0.66, 0.5, 0.33, 0.28, and 0.25. Figure 2 shows that the results from the jar tests with the six optimum ratios confirm the results from the earlier analysis. Even with change in incoming water quality the efficiency of treatment with residual solids did not change over 5% of originally tested water, though the optimum ratio for each coagulant can be different.

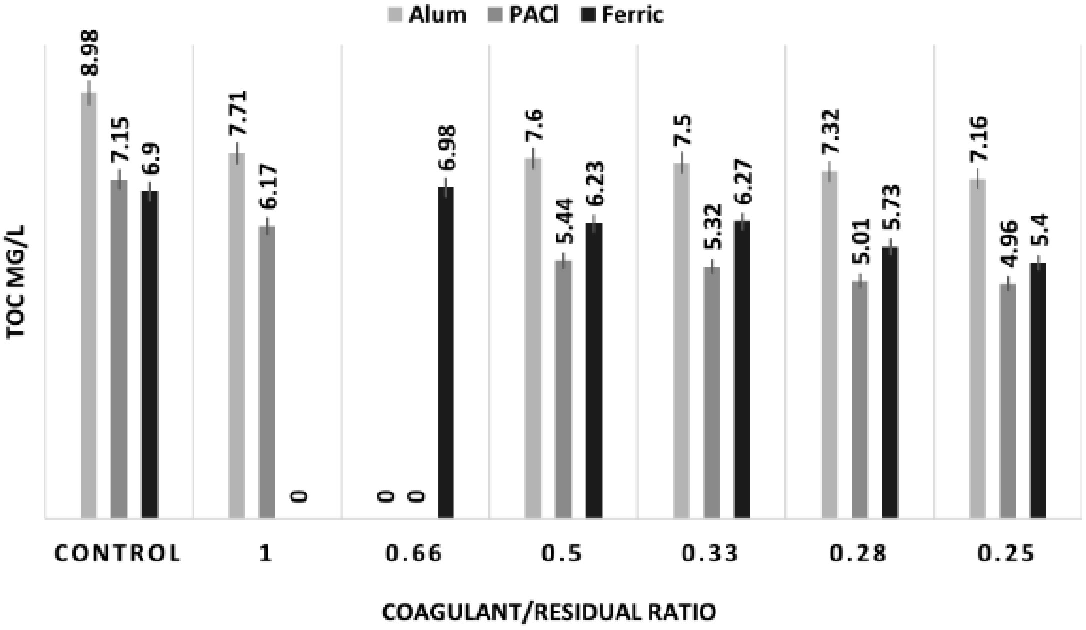

Figure 2. Surface water treatment using selected five C/R ratios based on net positive reduction in TOC (C/R > 1 didn't result in additional TOC removal).

Analysis of the data shows that a 16.6–22.5% additional TOC removal can be achieved with the selected ratios for alum, a 27.2–36.2% for PACl, and 9.5–24.4% for $FeCl_3$ relative to the control. Average percent additional TOC removal for alum, PACl, and $FeCl_3$ are 19.4%, 32%, and 15.4%, respectively for all selected ratios.

The quality of incoming ground water in terms of NOM species is considered to be different than that of a river [17]. Typically, TOC and turbidity concentrations of ground water are lower than the surface water [17]. In order to determine whether or not, treatment with residual solids will work with ground water, water from alluvial wells was collected and tested with the selected five C/R ratios using all three coagulants.

The data from the jar test shows that treatment with residual solids still was able to achieve additional TOC removal with respect to controls as shown in Figure 3. Although the effect of each coagulant by itself on the water seems different than that on surface water and so are the ratios, the percentage difference in TOC removal is on average over 17%. This can be clearly seen in the graph when the effluent TOC concentrations are compared to controls. Analysis of the data shows that a 14.1–38.2% additional TOC removal can be achieved with the selected ratios for alum, a 21.2–32.1% for PACl, and 6.3–42.6% for $FeCl_3$ relative to the control. Average percent additional TOC removal for alum, PACl, and $FeCl_3$ are 25.4%, 28.5%, and 24.6%, respectively, for all selected ratios.

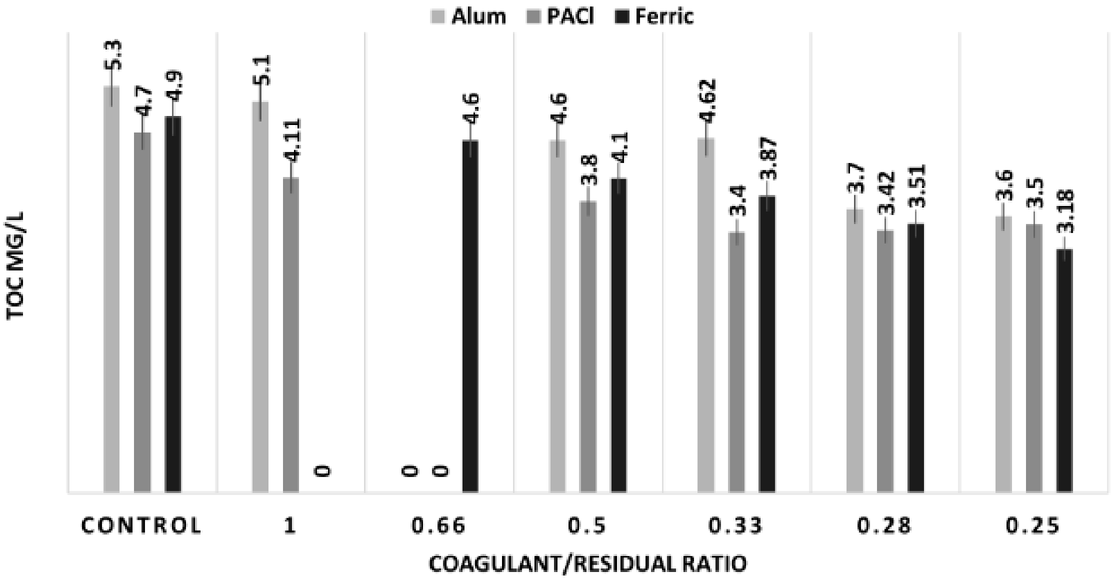

Figure 3. Ground water TOC removal using selected five C/R ratios.

For a ground water system, the best choice of coagulant can be different from that of surface water and therefore, so is the optimum C/R ratio of each coagulant. In determining the optimal ratio for a ground water system, additional jar testing with the water needs to be conducted. The result of this treatment on both types of water source is enhanced TOC reduction, but percentage reduction for each source differs. This difference needs to be taken into consideration before making a treatment change decision. Enhanced TOC removal even if by a small percentage can have an impact on a utility's ability to be compliant with DBP stage 2 regulations. Additional TOC removal allows for reduced use of disinfectant to maintain required minimum residual and can potentially lead to decreased formation of regulated and emerging DBPs.

3.2. C/R Ratios and Aluminum Removal

Aluminum concentrations of raw water differ depending on the source and so are the concentrations in effluent water depending on the treatment used. Using aluminum-based coagulants such as alum and PACl can also increase the concentrations of aluminum in the effluent water, so this was an area of concern during this study [18]. New treatment technologies that are aimed at improving water quality with respect to one contaminant or water quality parameter may sometimes degrade it with respect to another. Therefore, aluminum concentrations were tested before and after treating water with residual solids for all three coagulants used.

The concentration of aluminum in the raw ground water was measured to be 0.04 mg/L. As shown in the Figure 4, the residual aluminum in the water after being treated with alum, PACl, and $FeCl_3$ is 1.45 mg/L, 1.26 mg/L, and 0.07 mg/L, respectively. Noteworthy, the residual aluminum in water treated with alum and PACl is higher than that treated by $FeCl_3$ as well or than in the raw water. This could be due to a technical error, or an outlying circumstance.

Analysis of the data from jar tests shows that contrary to our assumption that residuals might increase the aluminum concentrations in the effluent water, the treatment with residual is shown to actually decrease the aluminum residuals notably compared to the controls. A 13.2–65.6% additional aluminum removal can be achieved with the selected ratios for alum and 13.6–83.1% for PACl relative to the control. Average percent additional aluminum removal for alum and PACl are 40.3%, and 37.7%, respectively for all selected ratios. Aluminum residual in the control and in the treated water as in the case of $FeCl_3$ is not significant as the coagulant itself is not aluminum based. It can be concluded that addition of residuals not only improves the finished water quality with respect to TOC concentration, but also with respect to aluminum concentrations.

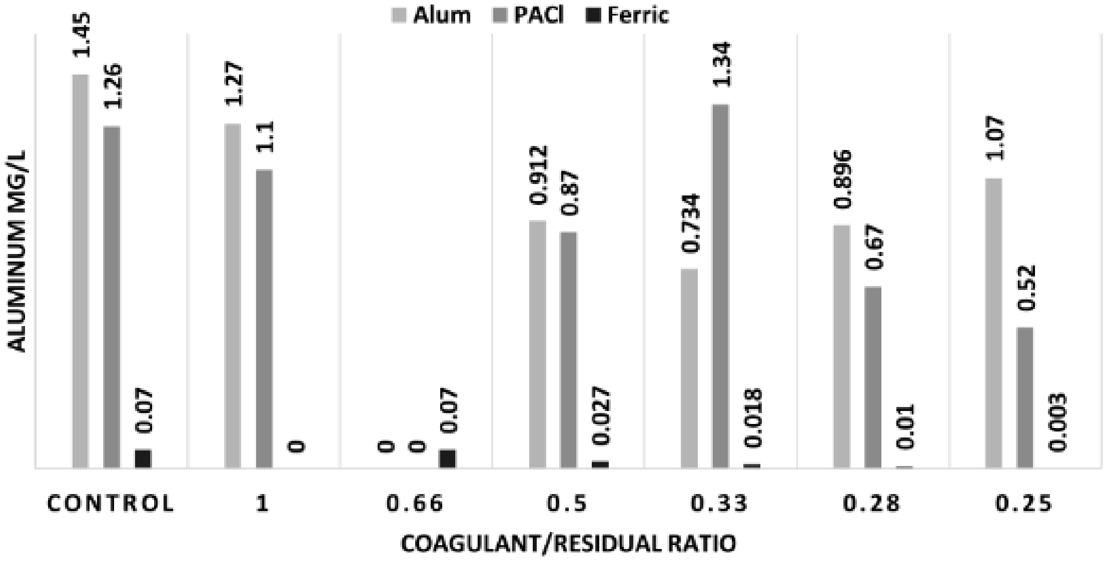

Figure 4. Aluminum residual removal using selected five C/R ratios.

3.3. C/R Ratios and Iron Removal

Most drinking water utilities have to deal with iron concentrations in their source water on a day-to-day basis. Presence of iron ions in natural and treated waters is known to have adverse effects on disinfection leading to increased formation of DBPs [2,19,20]. With increasing drought conditions, changes in the land use, and decreasing dissolved oxygen concentrations in the water sources, iron concentrations in raw water is becoming a bigger problem [21]. Aeration is typically used to precipitate iron from the raw water entering a treatment plant, but usage of iron-based coagulants to decrease TOC concentrations can sometimes have an adverse effect with respect to iron residual concentrations in finished water. The proposed treatment using residual solids raises concerns of increased iron concentrations in the effluent water as one of the coagulants used is $FeCl_3$. Therefore, iron concentrations were tested before and after treating water with residual solids for all three coagulants used.

The concentration of iron in the raw water was measured to be 5.34 mg/L. The residual iron in the water after being treated with alum, PACl, and $FeCl_3$ was 3.08 mg/L, 1.98 mg/L, and 2.76 mg/L, respectively.

Analysis of the data from jar tests shows that contrary to our assumption that residual solids might increase the iron concentrations in the effluent water, the treatment with residual solids is proven to decrease the iron residuals considerably compared to the controls as shown in Figure 5. A 67.2–98% additional iron removal can be achieved with the selected ratios for alum, a 12.91–64.9% for PACl, and 35.9–80.2% for $FeCl_3$ relative to the control. Average percent additional iron removal for alum, PACl, and $FeCl_3$ are 85%, 36.35%, and 59.7%, respectively for all selected ratios. It can be concluded that addition of residual solids not only improves the finished water quality with respect to TOC concentration, but also with respect to iron concentrations.

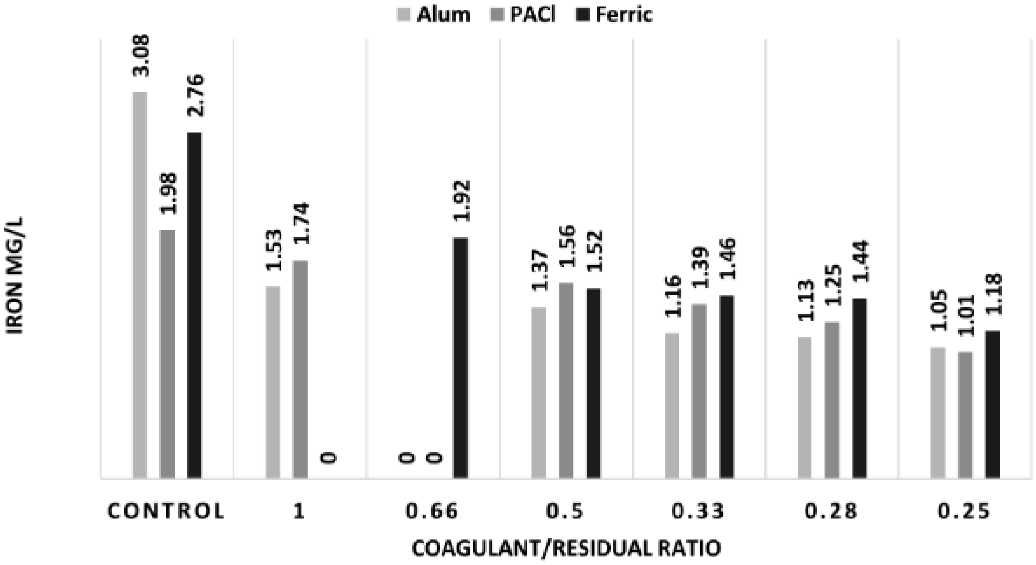

Figure 5. Iron residual removal using selected five C/R ratios.

3.4. pH Stability

The drinking water treatment process is a pH-based system. pH differences can be the result of coagulants added as part of the treatment and they are usually adjusted by addition of a base or an acid before disinfection [22]. Different disinfectants work effectively at different pH ranges. For example, chlorine works well at lower pH (pH 6–7) when compared to chloramines (pH 7–8.5) [19]. Therefore, water utilities adjust their pH throughout the process to allow for different unit processes to work more efficiently. However, pH adjustment can sometimes be expensive or hard to achieve, and the expenses of which could negate the benefit from this process. Different coagulant usage results in different pH in the effluent water and this raises the concern about dramatic pH changes as a result of proposed treatment with residual solids [23]. Therefore, pH is measured before and after treating water with residuals for all three coagulants used for both surface and alluvial ground water and are compared against their respective controls as shown in Figure 6.

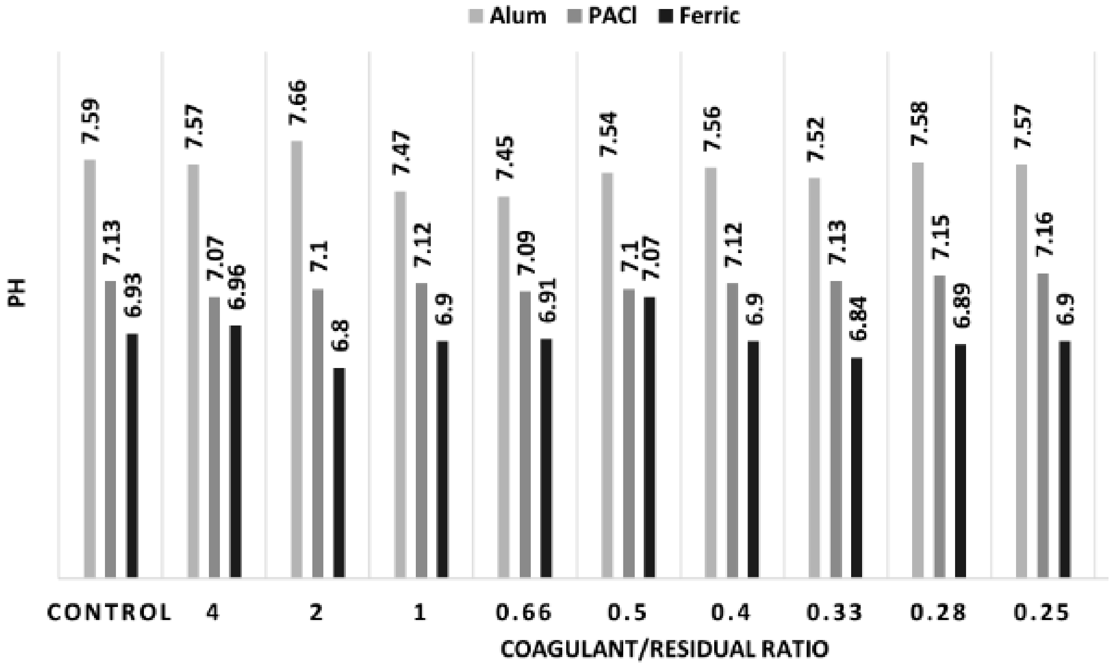

Figure 6. pH change because of treatment with residuals.

Analysis of the data from jar tests shows that contrary to our assumption that residuals might change the pH in the effluent water; the proposed treatment with residual is shown to cause no significant (+/− 0.2) changes in pH when compared to the controls. Therefore, no additional efforts are required for pH adjustment as a result of addition of residual solids.

4. Conclusions

This study was performed as an effort to develop a cost-effective technology that will provide additional TOC removal while helping the water utilities reuse their treatment solids. Enhanced TOC removal can be achieved when $C/R < 1$. The percent additional TOC removal for alum, PACl, and $FeCl_3$ use in surface (river) water is 16.6–22.5%, 27.2–36.2%, and 0.5–24.4%, respectively, and use in alluvial ground water is 14.1–38.2%, 21.2–32.1%, and 6.3–42.6%, respectively. A slight increase in TOC is observed when $C/R > 1$. Enhanced aluminum and iron removal can also be achieved with this proposed treatment. The percent additional removal of aluminum residual for alum and PACl addition is 13.2–65.6% and 13.6–83.14%, respectively, whereas percent of additional iron residual removal for alum, PACl, and $FeCl_3$ is 67.2–98%, 12.91–64.9%, and 35.9–80.2%, respectively. As seen with the metals tested, it is believed that the proposed treatment will increase the overall removal of pollutants and pathogens in the water, but monitoring of indicator organisms is recommended to determine the impact on microbial populations. Further investigation in this area during pilot scale study is needed to evaluate the full spectrum of microbial parameters such as bacterial number and BDOC. No significant changes in pH were observed as a result of the proposed treatment.

The percent additional TOC removal depends on the raw water quality and its changes, C/R ratios used, and type of coagulant needed. Residuals produced at a plant are generally one-third the volume of coagulants used. Additional studies on continuous application are needed, while reuse of recycled residuals can be an optimal solution for residuals availability issues. Finding a right match of coagulant and C/R ratio for a certain kind of raw water requires additional jar testing and data analysis. Treatment using residual solids is shown to be a simple, non-invasive, and cost-effective technology that will effectively lower the TOC concentrations by having the water utilities reuse their treatment residual solids. Another aspect that needs future examination is cost-benefits; this proposed treatment can potentially decrease the amount of coagulant used by finding an optimal coagulant to residual dosing ratio thereby providing cost savings to a utility. This optimal ratio will differ for each utility depending on their raw water quality, unit process treatment efficiency, residual composition, and residuals recycling mechanism and so will their extent of cost savings. This study used constant coagulant concentration and varying residuals to evaluate the effect of residuals on the water quality in terms of TOC, pH, aluminum and iron concentrations. Utilities need to perform further analysis with varying coagulant and residuals concentrations on their water to obtain cost benefits from decreased need of coagulants. The cost of a utility's existing residuals management process can be eliminated to most part as the volume of residuals used in this proposed treatment is higher than what's typically produced at a utility leading to further cost benefits. Landfills are a common disposal method and the proposed treatment will allow utilities to save on the associated cost while reducing environmental loading.

This lab-scale study was conducted using a well-known method among utility community (jar tests) as a proof of concept to provide basis for future pilot scale continuous process studies as at a treatment plant.

Author Contributions: Conceptualization, S.R.P. and E.I.; methodology, S.R.P.; validation, E.I.; formal analysis, S.R.P.; investigation, S.R.P. and J.C.; resources, E.I.; data curation, S.R.P. and J.C.; writing—original draft preparation, S.R.P.; writing—review and editing, E.I.; visualization, S.R.P.; supervision, E.I.; project administration, E.I.; funding acquisition, E.I., H.S., J.Y., and B.H.

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