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Effect of Al speciation on residual turbidity and Al minimization by coagulation with single and dual dosing

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ABSTRACT

It is challenging for a drinking water treatment plant to carry out effective coagulant dosing with polyaluminum chloride (PACl) for turbid-varied water. The goal of this study is to investigate effects of Al speciation on the minimization of residual turbidity and Al in coagulation-sedimentation for low and high turbid water. Two coagulant dosing approaches, including single and dual dosing with different commercial PACl and $FeCl_3$ coagulants, were used to evaluate coagulation performance in terms of turbidity and residual Al. The results showed that single dosing by PACl-1 with high monomeric Al (48%) is effective in reducing turbidity at low turbidity, but its residual Al is higher than that by PACl-2 with high monomeric Al (54%) and polymeric Al (35%), while PACl-2 coagulation brings less turbidity reduction than PACl-1 coagulation regardless of Al minimization. Dual dosing with $FeCl_3$ and PACl improve Al reduction but turbidity reduction improvement depends on Al/Fe dosing ratio. The most effective reduction in residual turbidity and Al is achieved at the Al/Fe ratio of 1:2 for low-turbidity water, whereas dosing ratio is insensitive to residual turbidity and Al at high turbidity. Dual dosing can achieve more effective residual turbidity and Al minimization for low turbidity water.
Key words | coagulation, $FeCl_3$, PACl, residual Al, water treatment

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INTRODUCTION

Coagulation is an essential treatment process used to aggregate small particles in drinking water treatment. Studies showed that water properties greatly affect the effectiveness of coagulant performance (Yu *et al.* 2006; Ng *et al.* 2013). In low turbidity water, existing particles could be too small in size, thus, limiting the aggregation with coagulants as well affecting the structure of flocs formed, reducing overall effectiveness of coagulation (Shen 2005; Huang *et al.* 2015; Zhang *et al.* 2018). Single dosing with Al-based coagulant was not effective in reducing residual Al after coagulation even though at low dosage, especially for low turbid water (10–15 NTU) with high pH value ($pH > 8$) based on Al hydrolyzed chemistry (Duan & Gregory 2003). In Taiwan, specifically during the typhoon seasons, water turbidity always increases due to excessive upstream erosions by heavy rainfall to as high as 40,000 NTU (Lin *et al.* 2008b). Similarly, the Yellow River as one of the most important local urban water resources in China also contains high concentrations of solids (Song *et al.* 2015). In China, it is feasible to treat the high turbid water using a pre-sedimentation tank, but, in Taiwan, due to land scarcity, most existing drinking water treatment plants (DWTP) rely on the use of hydrolyzed coagulants, such as polyaluminum chloride (PACl) to lower high turbid water, which normally results in excess Al in finished water when overdosing occurs. It is known that high levels of aluminum may have some adverse effects on the human central nervous system, specifically the brain, together in the presence of other substances, such as citrate, or other external conditions, especially in diseases such as Alzheimer's disease and dementia (Taylor *et al.*

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1992; Kawahara & Kato-Negishi 2011; Killin *et al.* 2016). Aluminum as a potent neurotoxin also can accumulate with additional immunotoxicity effects in the human body (Yasui *et al.* 1997; Zhu *et al.* 2014). To comply with drinking water standard limitations, in conventional DWTP either single or dual dosing with Al- and Fe-based coagulants are alternatively dosed to lower the residual Al of finished water to an acceptable level (<0.20 mg/L as Al) for drinking water safety limit in the United States (USEPA 2018) and (<0.16 mg/L as Al) for drinking water safety limit in Taiwan.

Letterman & Driscoll (1988) reported that controlling the final pH of PACl coagulation in the range 5.5–6 is an effective and feasible way for reducing residual Al in water. However, it is troublesome for operators to manipulate coagulation at a constant pH to improve turbidity reduction and achieve lower residual Al. Thus, ferric chloride ($FeCl_3$) coagulant has been widely used as an alternative coagulant to replace either some or all parts of PACl coagulant dose to achieve residual Al minimization for drinking water treatment (Qureshi & Malmberg 1985; Dong *et al.* 2014; Park *et al.* 2016). In theory, the chemistry of using dual coagulants is more complex than that of single coagulants. Studies have reported that there is no discernible improvement in coagulating ability by dual coagulation over single coagulant (Johnson & Amirtharajah 1983).

Although it has been proven that dual dosing with PACl and $FeCl_3$ coagulants would improve the reduction of residual Al in finished water for DWTP, it is difficult to constantly control the dosing ratio between Al- and Fe-based coagulants by use of dual coagulant dosing. Many studies have proven that Al speciation of PACl coagulants has a profound effect on the residual Al concentration and turbidity reduction (Lin *et al.* 2008a; Yang *et al.* 2011; Duan *et al.* 2014). A tailored PACl with low content of monomeric Al or high content of polymeric Al is effective in lowering final residual Al in the coagulation process (Kimura *et al.* 2013). An effective single or dual dosing approach is crucial to improve coagulation performance in achieving simultaneous reduction for turbidity and residual Al in drinking water treatment. However, limited researches have studied the effect of Al speciation on the particle removal and residual Al, such as particulate and dissolved Al, in coagulation by single or dual dosing with different Al/Fe dosing ratios for drinking water treatment.

This study aims to investigate coagulation performance for residual turbidity and Al minimization with the rational use of single (PACl) or dual coagulants (PACl + $FeCl_3$) in drinking water treatment for low and high turbidity water. The effects of Al speciation on coagulation with either single or dual dosing were evaluated to show under what conditions each or both coagulants are effective in removing particles and lowering residual Al.

MATERIALS AND METHODS

Raw water quality

Raw water was collected from Hsinchu Second Water Treatment Plant, Taiwan and tested for its characteristics. pH was measured using a pH meter (InoLab Multi level, WTW, Germany) at pH 9.0 ± 0.1 and 8.0 ± 0.1 for low and high turbidity water, while turbidity was measured using a turbid meter (2100P, Hach, USA) at 11 NTU and 735 NTU for low and high turbidity water, respectively. The rise of turbidity in river water up to 735 NTU occurred because of heavy rainfall. This condition is highly favorable especially in the typhoon season around summer. The average diameter of particles in raw water was determined at 37.27 μm by FlowCAM (FlowCAM, USA). The imaging of FlowCAM was conducted based on the method of a previous study (Lin *et al.* 2016).

Characterization of coagulants

Two commercial PACl coagulants, designated as PACl-1 and PACl-2, were used in this study. Al speciation of PACl-1 and PACl-2 was determined by Ferron method (Lin *et al.* 2008a). As shown in Table 1, the PACl-1 contains about 48% monomeric Al (Al_a), 8% polymeric Al (Al_b), and

Table 1 | Al species distribution of commercial PACl coagulants

Type of coagulants	pH	Basicity	Al_a (%)	Al_b (%)	Al_c (%)
PACl-1	4.14	1.4	48	8	44
PACl-2	4.2	1.5	54	35	11

44% colloidal Al (Al_c), while the PACl-2 has 54% monomeric Al (Al_a), 35% polymeric Al (Al_b), and 11% colloidal Al (Al_c) of total Al concentration. Aluminum concentration was analyzed by an inductively coupled plasma atomic emission spectrometry (ICP-OES 7000 series, Agilent, USA). In addition, a reagent-grade $FeCl_3$ coagulant was used for this study. The working solutions containing 2,000 mg/L Al or Fe were freshly prepared before each test.

Coagulation protocol

Standard jar trials (Phipps and Bird, USA) were conducted to evaluate coagulation performances. Two kinds of coagulant dosing approaches, including single dosing (PACl-1 or PACl-2) and dual dosing (PACl-1 + $FeCl_3$ or PACl-2+ $FeCl_3$), were carried out for this study. The initial pre-oxidation with NaOCl of 2 mg/L as Cl was conducted at 240 rpm for 30 sec. After that, rapid mixing was conducted at 200 rpm ($G = 350\ s^{-1}$) for one min followed by a slow mixing at 30 rpm ($G = 25\ s^{-1}$) for 22 min. The suspension was left undisturbed for 10 min. After settling, the turbidity of the supernatant was measured by a turbid meter (2100 P, Hach, USA) and the residual dissolved Al was quantified by ICP-OES (ICP-OES 7000 series, Agilent, USA). The zeta potentials of the suspension were measured via a laser zeta analyzer (Zetasizer nano ZS, Malvern Inc., UK) immediately after the rapid mixing without dilution. All tests were conducted in triplicate.

RESULTS AND DISCUSSION

Effect of Al speciation on turbidity and Al minimization for single dosing

Effects of coagulation by single dosing with PACl-1 and PACl-2 at various dosages on corresponding pH, residual turbidity, and Al for both low (11 NTU) and high (735 NTU) turbid water are shown in Figures 1 and 2, respectively. Dosage ranges were 0, 9.4, 18.8, 23.5, and 28.2 mg/L for dosing in low turbidity water, whereas for dosing in high turbid water they were 0, 18.8, 37.6, 56.4, 75.2, and 94 mg/L as 10% of Al_2O_3. As shown in Figure 1(a), for low turbidity water, residual turbidity decreased significantly with increasing dosages from 9.4 to 28.2 mg/L prior to dosing of PACl-1 coagulant to as low as 0.71 NTU at 28.2 mg/L. Residual Al increased gradually with increasing dosages of coagulant, with residual dissolved Al hitting the minimum safety level for residual Al of 0.16 mg/L Al (indicated by dotted line) at 18.8 mg/L and going over the minimum limit with a further increase of dosage to as high as 0.27 mg/L Al at 28.2 mg/L dosage. pH changes were insignificant throughout the dosing ranging between 8.73 and 8.84. Figure 1(b) shows that in high turbidity water, residual turbidity decreased significantly with increasing dosages from 18.8 to 94 mg/L to as low as 6.2 NTU at 75.2 mg/L with a slight increase to 6.4 NTU at 94 mg/L. Dissolved residual Al remained low and under the minimum safety level for residual Al at all dosages for high turbid water. pH changes were visible with further increase of dosage with pH prior to dosing at 8.04 to as low as 7.22 for dosage at 94 mg/L.

On the other hand, as shown in Figure 2(a), in low turbidity water, residual turbidity decreased significantly with increasing dosages from 9.4 to 28.2 mg/L prior to dosing of PACl-2 coagulant to as low as 1.32 NTU at 18.8 mg/L with slight increases in turbidity parallel with increasing dosages. Unlike PAC-1 dosing, PACl-2 dosing lowers the residual dissolved Al to 0.18 mg/L Al at 28.2 mg/L. pH changes were minimum throughout the dosing, ranging between 8.73 and 8.28. Figure 2(b) shows that in high turbidity water, residual turbidity decreased significantly with initial dosing at 18.8 mg/L to 36.6 NTU and continued to decrease with increasing dosages to as low as 15.17 NTU at 56.4 mg/L but gradually increased with dosage >56.4 mg/L. Dissolved residual Al remained low and under the minimum safety level for residual Al at all dosages in high turbidity water. Residual dissolved Al at the coagulant dosage of 18.8 mg/L was comparable at 0.043 mg/L Al for high turbid water compared with 0.097 mg/L Al for low turbid water. pH changes were minimum with further increase of dosage with pH prior to dosing at 8.04 to 7.41 for dosage at 94 mg/L. In the case of turbidity removal at optimum dosage, at low turbidity, removal rate was slightly better for PACl-1 coagulation at 89.27% than 87.89% for PACl-2 coagulation, whereas PACl-1 coagulation showed a similar trend at 99.16% than 97.94% for PACl-2

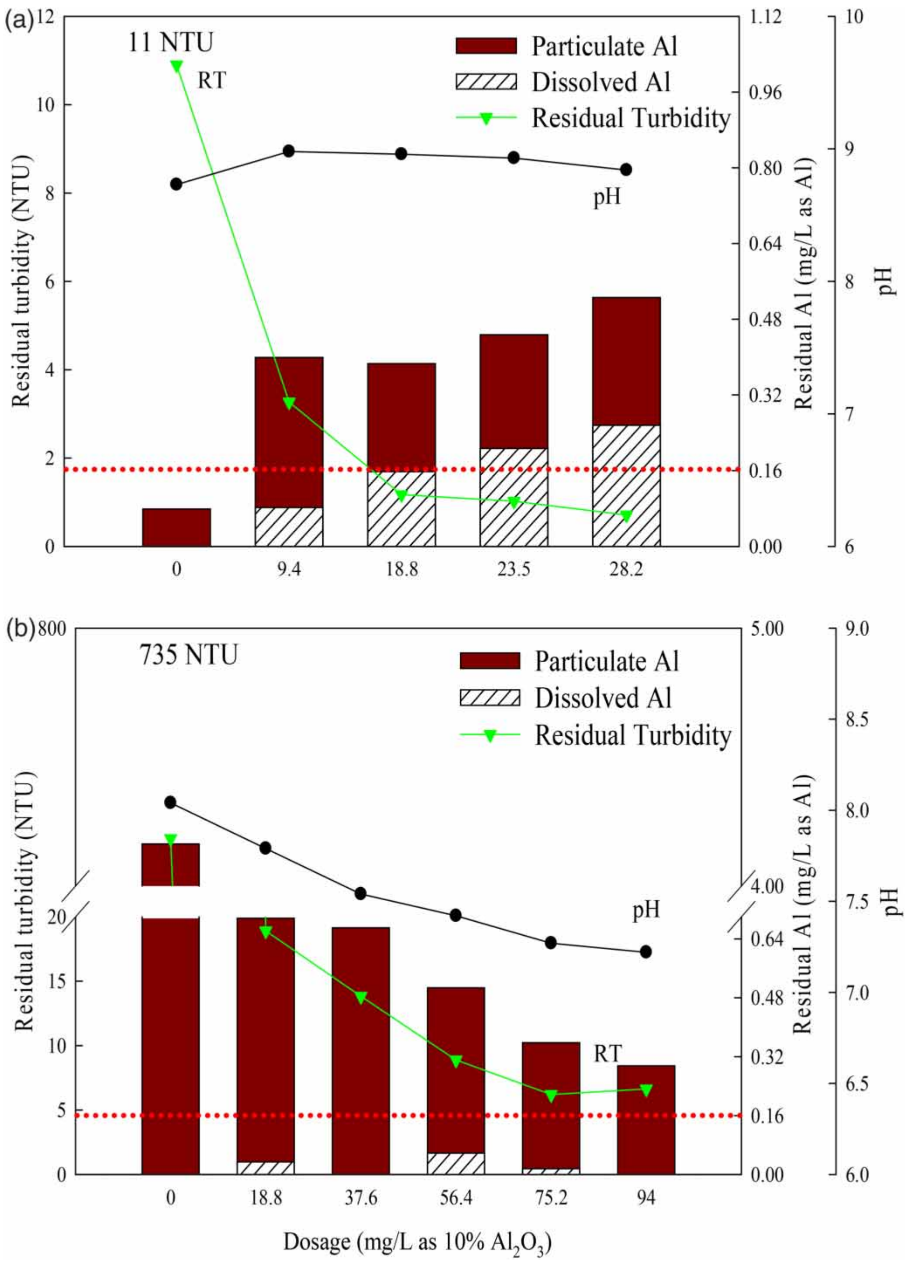

Figure 1 | Variations of residual turbidity and Al of supernatant and corresponding pH by coagulation with PACl-1 at various dosages for low and high turbid water: (a) 11 NTU and (b) 735 NTU.

coagulation at high turbidity. In the case of residual Al removal at optimum dosage, at low turbid water, both PACl-1 and PACl-2 showed insignificant difference in increase of particulate Al, whereas PACl-2 resulted in 38.6% less dissolved Al compared to PACl-1 for high turbid water. Overall, residual Al for low turbid water was increasing due to the addition of coagulants, especially for dissolved Al (even passing the regulation limit), whereas the removal of residual Al at high turbidity was significant with dissolved Al remaining lower than the regulation limit. On the other hand, PACl-1 resulted in 18% more total residual Al than PACl-2 in low turbid water, while in high

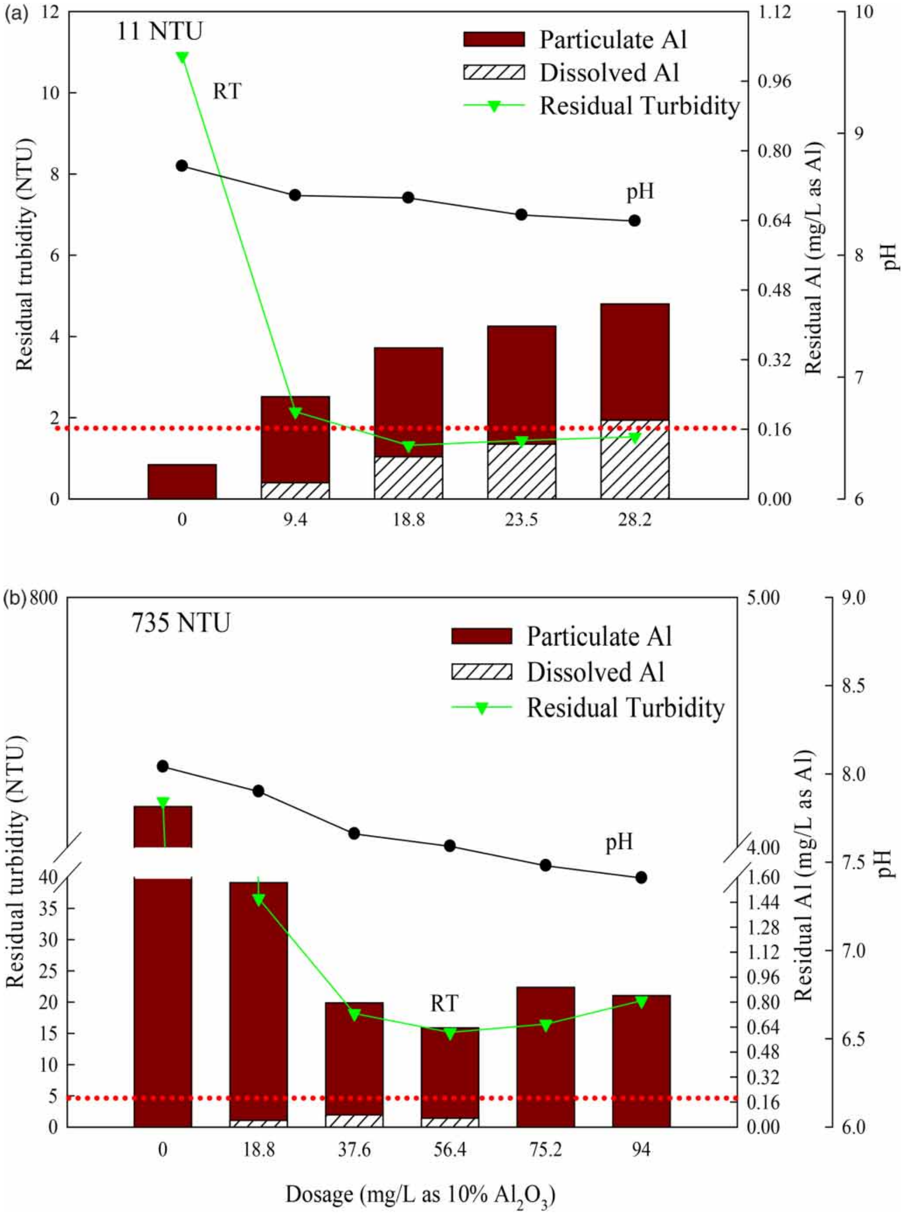

Figure 2 | Variations of residual turbidity and Al of supernatant and corresponding pH by coagulation with PACl-2 at various dosages for low and high turbid water: (a) 11 NTU and (b) 735 NTU.

turbid water, PACl-1 showed a better result with 45.98% less total residual Al.

As the above results show, there are marked differences in residual turbidity and Al between PACl-1 and PACl-2 coagulation, especially for low turbid water treatment. A study by Yang *et al.* (2010) indicated that the majority of residual Al will be in dissolved inorganic Al with monomeric Al as its main component and lower residual Al can be achieved with initial pH between 6 and 8. Other studies have also proven that lowering monomeric Al in PACl coagulant can lower the residual dissolved Al in solution based on water chemistry principle (Kimura *et al.* 2013). As discussed by Duan *et al.* (2014), monomeric Al may form small and weak monomeric Al complex which is hard to remove, in contrast with polymeric and colloidal Al species that can form bigger and more compact flocs that readily absorb particles. It is demonstrated that PACl-1 contains equally high monomeric ($Al_a = 48\%$) and colloidal Al ($Al_c = 44\%$) and low polymeric Al ($Al_b = 8\%$) level. Based on previous studies, colloidal Al favors enmeshment that is responsible for particle removal by sweep flocculation in the neutral or alkaline pH region (Lin *et al.* 2008c; Liu *et al.* 2009). Regarding the zeta potential variations with changing of pH, as shown in Figure 3, the zeta potentials became more positively charged with decreases in pH for PACl-1 and PACl-2 coagulation; similar

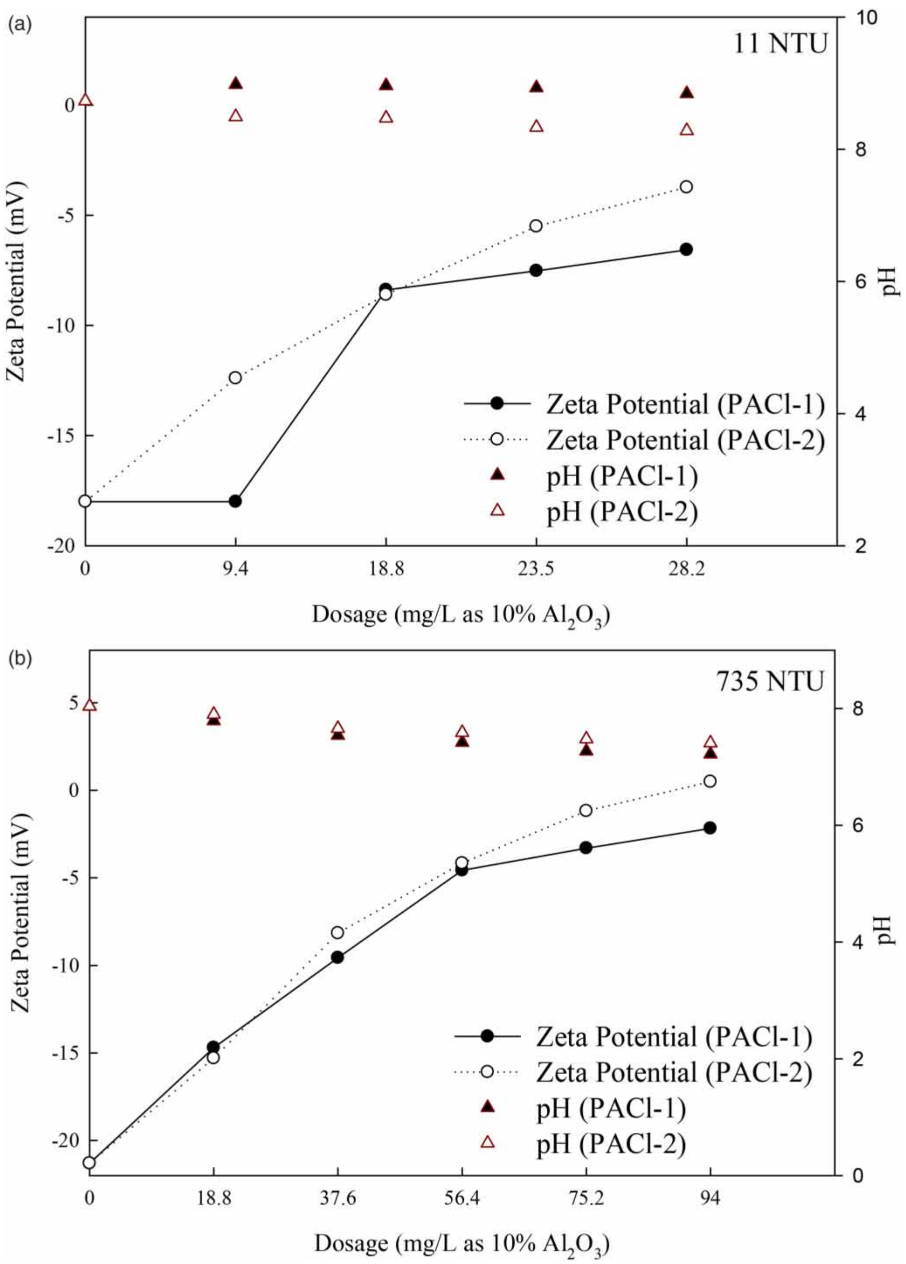

Figure 3 | Comparison in zeta potential changes with corresponding pH by coagulation with either PACl-1 or PACl-2 at various dosages for low and high turbid water: (a) 11 NTU and (b) 735 NTU.

results were found in other studies (Lin *et al.* 2008a; Priya *et al.* 2018). In addition, the pH after rapid mixing for PACl-2 coagulation is near to 8, and it is slightly lower than that for PACl-1 coagulation under various dosages at low turbidity, but the pH under various dosages at high turbidity for PACl-1 and PACl-2 coagulation is similar with a range from 7.6 to 8. Theoretically, the dissolved Al decreases as the pH of solution decreases at alkaline pH (Kimura *et al.* 2013). Thus, at alkaline pH, PACl-1 coagulation produces an effective particle destabilization by enmeshment to lower residual turbidity at either low or high turbidity. However, it would result in higher residual dissolved Al in supernatant after the coagulation-sedimentation process, especially at low turbidity. There are fewer particles remaining in low turbid water before coagulation which lowers the possibility of Al ion or hydrolyzed Al species adsorbing to the surface of suspended particles during coagulation (Yao *et al.* 2014). In contrast, PACl-2 contains high monomeric Al ($Al_a = 54\%$), low polymeric Al ($Al_b = 35\%$), and low colloidal Al ($Al_c = 11\%$) level. Owing to its polymeric Al, PACl-2 has higher charge neutralization ability, as shown in Figure 3, to destabilize particles, specifically native natural organic matter (NOM), repulsion force by neutralizing negative charges of particles to simultaneously lower residual turbidity and dissolved Al, accompanying sweep flocculation under alkaline pH at low turbidity (Duan & Gregory 2003; Zhang *et al.* 2018). At such a condition, PACl-2 coagulation is more effective to improve residual turbidity and Al reduction, in the meantime, after the coagulation-sedimentation process compared to PACl-1 coagulation. However, as turbidity increased to as high as 735 NTU, PACl-1 coagulation at optimum dosage (75.2 mg/L) is able to form bigger flocs to produce a more effective sweep flocculation along with strong adsorption of hydrolyzed Al, which results in greater reduction of residual turbidity and Al, while PACl-2 coagulation results in particle aggregation by both charge neutralization and sweep flocculation that are responsible for higher residual turbidity at lesser optimum dosage of 56.4 mg/L (Lin *et al.* 2008b). These results indicated that Al speciation has a profound effect on PACl coagulation with single dosing for residual turbidity and Al reduction. The content of monomeric, polymeric Al and colloidal Al are equally crucial to achieve both lower turbidity and dissolved Al level.

Effect of Al speciation on turbidity and Al minimization for dual dosing

Dual dosing effects of PACl-1 or PACl-2 with $FeCl_3$ coagulant at various dosing ratios towards corresponding pH, residual turbidity, and Al are shown in Figures 4 and 5. Dosing ratios were 0, 1:0, 1:1, 1:2, and 2:1 as Al:Fe for both coagulations on low and high turbidity water. The optimum PACl dosage for turbidity and Al reduction was chosen at 18.8 mg/L as Al with various $FeCl_3$ dosages for this study. As shown in Figure 4(a), in low turbidity water, residual turbidity decreased significantly to 2.19 NTU after initial dosing with only PACl-1 coagulation. Initial dual dosing with 1:1 and 1:2 ratios double the residual turbidity to 4.29 NTU and decreased it to 2.87 NTU, respectively. At dosing ratio of 2:1, residual turbidity can be further decreased to around 2 NTU. Compared to single dosing by PACl-1, dual dosing with $FeCl_3$ can further decrease the dissolved residual Al to 0.054, 0.012, and 0.081 mg/L for 1:1, 1:2, and 2:1 dosing ratio, respectively, accompanying little pH variation between 8.55 and 8.68. Figure 4(b) shows that in high turbidity water, residual turbidity decreased significantly after initial dosing to the lowest of 6.2 NTU with only PACl-1 coagulation. Dual dosing with 1:1 and 1:2 ratios almost tripled the residual turbidity to 17.73 NTU and later decreased to 13 NTU, respectively. Dosing ratio of 2:1 showed an increase in residual turbidity to 14.60 NTU. Residual dissolved Al remained low and under the minimum safety level for residual Al of 0.16 mg/L Al throughout all dosing ratios. Single dosing with only PACl-1 showed the lowest residual Al presence at 0.015 mg/L compared with 0.017 and 0.036 mg/L for 1:1 and 1:2 dosing ratio, respectively. pH changes were minimum throughout various dosing ratios between 7.27 and 7.37.

As shown in Figure 5(a), in low turbidity water, residual turbidity decreased to 4.2 NTU after initial dosing with only PACl-2 coagulation. Initial dual dosing with 1:1 followed by 1:2 and 2:1 ratios increased the residual turbidity to 5.29 NTU and later decreased to 3.09 NTU and the lowest at 1.97 NTU, respectively. Compared to single dosing by PACl-2, it is shown that dual dosing with $FeCl_3$ can further decrease the dissolved residual Al to 0.015 and 0.054 mg/L for 1:1 and 2:1 dosing ratio, respectively, accompanying little pH variations between 8.58 and 8.69. Figure 5(b) shows that in

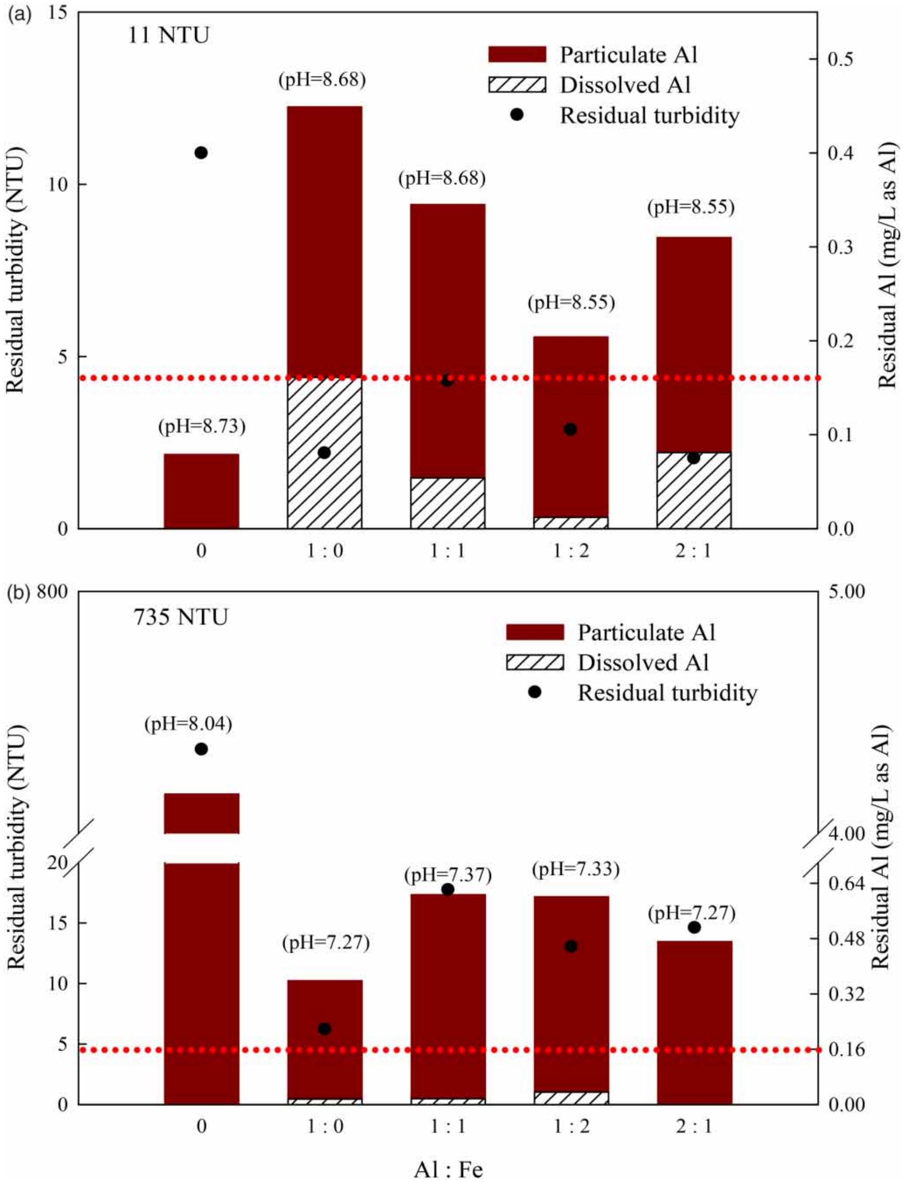

Figure 4 | The relationship between pH, residual turbidity, and Al of supernatant at various ratios of mixed PACl-1 and $FeCl_3$ coagulant for low and high turbidity water: (a) 11 NTU and (b) 735 NTU.

high turbidity water, residual turbidity decreased significantly after initial dosing with only PACl-2 to 15.17 NTU. Initial dual dosing with 1:1 followed by 1:2 and 2:1 ratios slightly increased the residual turbidity to 16.73 NTU and later decreased to 15.7 NTU and the lowest at 14.9 NTU, respectively. Residual dissolved Al remained low and under the minimum safety level for residual Al of 0.16 mg/L Al throughout all dosing ratios. Single dosing with only PACl-2 showed the highest residual Al presence at 0.057 mg/L compared with 0.022 and 0.037 mg/L for 1:1 and 1:2 dosing ratio, respectively. pH changes were minimum throughout various dosing ratios between 7.59 and 7.31.

For turbidity removal by coagulation with dual dosing, at low turbidity, both PACl-1 and PACl-2 dosing with $FeCl_3$ at 2:1 ratio showed the best removal with 81.28% and 81.92%, respectively. At high turbidity, both single dosing

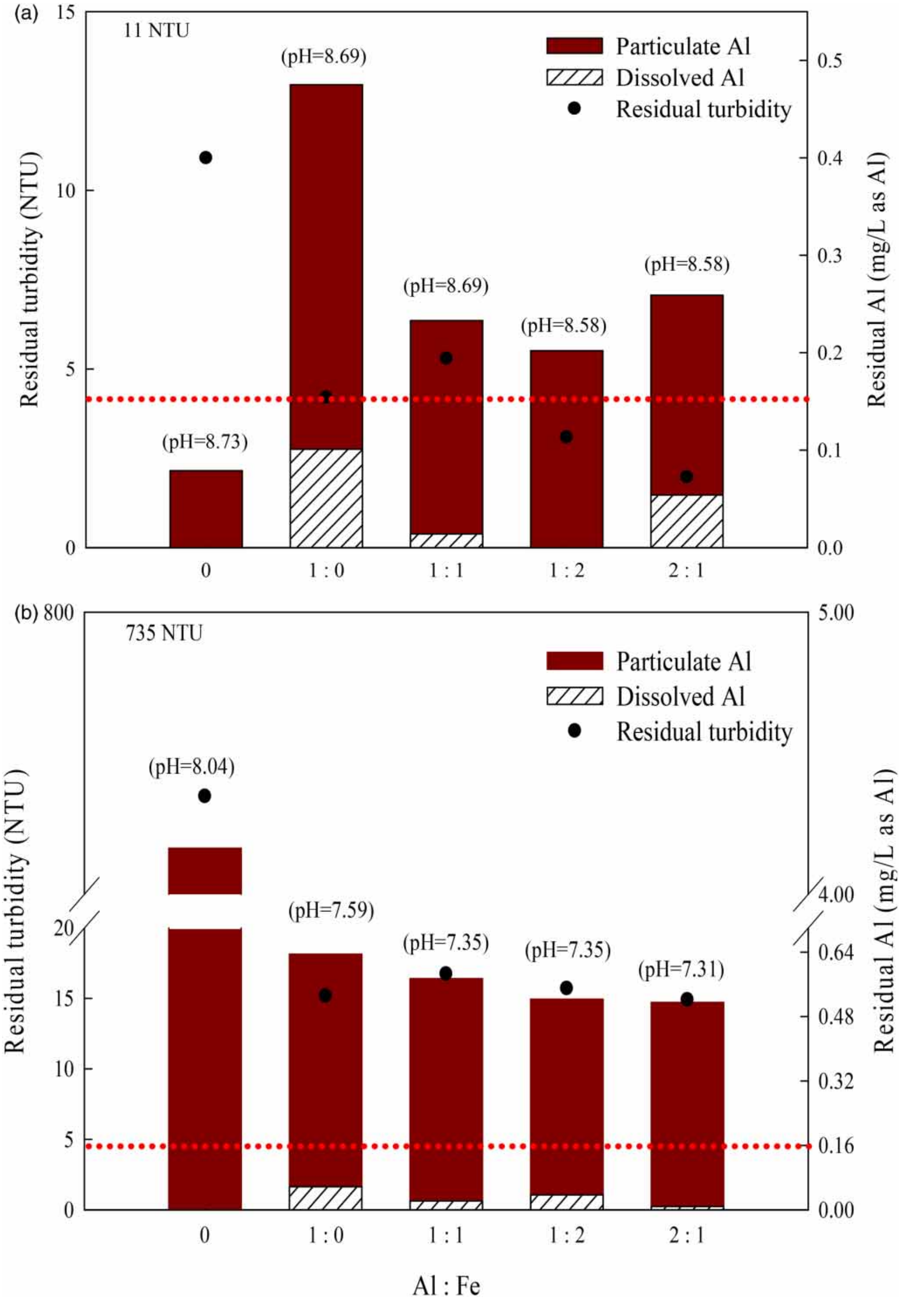

Figure 5 | The relationship between pH, residual turbidity, and Al of supernatant at various ratios of mixed PACl-2 and $FeCl_3$ coagulant: (a) 11 NTU and (b) 735 NTU.

and dual dosing at similar ratio showed a significant removal with 98.01% and 97.97%, respectively. For coagulation with dual dosing at 2:0 ratio, a similar trend for low turbid water was found after coagulation, but, dissolved Al was able to be minimized under the regulation limit. Similar trends were also found for high turbidity water. Dual dosing with PACl-2 and $FeCl_3$ resulted in 20% lesser total residual Al than PACl-1 at low turbidity, while PACl-1 showed a better result with 10% lesser total residual Al at high turbidity.

As mentioned above, although a previous study has reported that there is no discernible improvement in coagulating ability by dual coagulant over the single coagulant

(Johnson & Amirtharajah 1983), our study showed that the combination of PACl-1 or PACl-2 and $FeCl_3$ can improve the minimization of turbidity and Al for low turbid water, compared to single dosing. At low turbidity, as low as 11 NTU, shifting Al/Fe dosing ratio from 1:1 to 1:2 would improve in minimizing residual turbidity and Al reduction, while it would increase total residual Al level (particulate and dissolved Al) but decrease the residual turbidity when the Al/Fe dosing ratio changes to 2:1. The particle aggregation rate is quite low as coagulant dosing is conducted at low turbidity because few particles can contact each other to form flocs during aggregation. As reported by Duan & Gregory (2003), $FeCl_3$ possesses lower solubility at neutral pH than Al salts that it would be substantially hydrolyzed into large amounts of $Fe(OH)_3$ to frequently attach suspended particles, which offers opportunities for particle aggregation and floc formation. This results in trace ferric ions being retaining in treated water. However, coagulation with dual dosing by either PACl-1 or PACl-2 with $FeCl_3$ behaves differently in residual turbidity and Al reduction for high turbid water, even though low dissolved Al below the minimum safety level was achieved. At high turbidity, as high as 735 NTU, dual dosing with PACl-1 and $FeCl_3$ would produce higher residual turbidity and Al at various Al/Fe dosing ratios compared to single dosing with PACl-1, while dual dosing with PACl-2 and $FeCl_3$ would slightly improve turbidity and Al reduction as the Al/Fe dosing ratio is at 1:2 or 2:1. The particle aggregation rate is improved for coagulation of high turbid water because the majority of particles in suspension are easily destabilized and aggregated to floc due to frequent contacts between particles during aggregation. For this reason, the partial addition of PACl-2 containing higher polymeric Al level would improve particle destabilization by charge neutralization at dual dosing (Lin *et al.* 2008b) compared to the partial addition of PACl-1. The results showed that Al speciation has little effect on turbidity and Al reduction for coagulation with dual dosing. In addition, Al/Fe dosing ratio for coagulation with low turbid water exhibits stronger impact than that with high turbid water for residual turbidity and Al minimization.

The aluminum in the raw water was natively higher in high turbid water compared with low turbid water in the absence of dissolved aluminum. However, the residual dissolved aluminum only occurred after PACl coagulation. In both cases of PACl-1 and PACl-2 coagulation, the content of particulate Al as the major source of residual Al was effectively reduced with further sand filtration processes in the DWTP which can easily remove the residual particulate Al. In practice, high residual dissolved Al is more of a concern in treated water as the dissolved aluminum passes through a filter into the distribution system. In every case of coagulation test for this study, the level of dissolved Al after coagulation was achieved to within the safety range regulated by USEPA and Taiwan EPA. On the other hand, it is known that the solubility of ferric is lower than alumina at neutral pH where ferric content would easily transform into $Fe(OH)_3$ precipitates along with trace ferric ions during coagulation even though the dosage increases. These $Fe(OH)_3$ precipitates are also easily removed by filtration process in the DWTP. Therefore, trace of ferric ions in treated water would be within the permissible limit.

CONCLUSIONS

For low and high turbid water, Al speciation predominantly rules residual turbidity and Al minimization for single dosing with PACl, but it has little effect on turbidity and Al reduction for coagulation with dual dosing. A commercial PACl-2 containing 54% monomeric Al and polymeric Al as high as 35% exhibits a better ability to minimize residual turbidity and dissolved Al compared to PACl-1 containing polymeric Al as low as 8%. The PACl coagulation for residual turbidity and dissolved Al minimization in low turbid water treatment exists as Al speciation dependency. Al/Fe dosing ratio for coagulation with low turbid water exhibits stronger impact than that with high turbid water for residual turbidity and Al minimization. Enhanced coagulation for residual turbidity and Al minimization by dual dosing with commercial PACl and $FeCl_3$ can be achieved at Al/Fe ratio of 1:2 in low turbid water treatment, but the Al/Fe dosing ratio would be insensitive to the improvement in reduction of residual turbidity and Al. It is feasible to achieve simultaneous residual turbidity and both particulate or dissolved Al minimization using dual dosing with PACl and $FeCl_3$ at proper Al/Fe dosing ratio.

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REFERENCES

Dong, H. Y., Gao, B. Y., Yue, Q. Y., Sun, S. L., Wang, Y. & Li, Q. 2014 Flocs properties and membrane fouling of different monomer and polymer Fe coagulants in coagulation-ultrafiltration process: the role of Fe (III) species. *Chemical Engineering Journal* **258**, 442–449.

Duan, J. & Gregory, J. 2003 Coagulation by hydrolysing metal salts. *Advances in Colloid and Interface Science* **100–102**, 475–502.

Duan, S. X., Xu, H., Xiao, F., Wang, D. S., Ye, C. Q., Jiao, R. Y. & Liu, Y. J. 2014 Effects of Al species on coagulation efficiency, residual Al and floc properties in surface water treatment. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **459**, 14–21.

Huang, X., Sun, S., Gao, B., Yue, Q., Wang, Y. & Li, Q. 2015 Coagulation behavior and floc properties of compound bioflocculant–polyaluminum chloride dual-coagulants and polymeric aluminum in low temperature surface water treatment. *Journal of Environmental Sciences* **30**, 215–222.

Johnson, P. N. & Amirtharajah, A. 1983 Ferric chloride and alum as single and dual coagulants. *Journal of American Water Works Association* **75** (5), 232–239.

Kawahara, M. & Kato-Negishi, M. 2011 Link between aluminum and the pathogenesis of Alzheimer's disease: the integration of the aluminum and amyloid cascade hypotheses. *International Journal of Alzheimer's Disease* **2011**, 1–17.

Killin, L., Starr, J., Shiue, I. & Russ, T. 2016 Environmental risk factors for dementia: a systematic review. *BMC Geriatrics* **16** (1), 175.

Kimura, M., Matsui, Y., Kondo, K., Ishikawa, T. B., Matsushita, T. & Shirasaki, N. 2013 Minimizing residual aluminum concentration in treated water by tailoring properties of polyaluminum coagulants. *Water Research* **47** (6), 2075–2084.

Letterman, R. & Driscoll, C. 1988 Survey of residual aluminum in filtered water. *Journal of American Water Works Association* **80**, 154–158.

Lin, J. L., Chin, C. J. M., Huang, C. P., Pan, J. R. & Wang, D. S. 2008a Coagulation behavior of Al_{13} aggregates. *Water Research* **42**, 4281–4290.

Lin, J. L., Huang, C. P., Pan, J. R. & Wang, D. S. 2008b Effect of Al(III) speciation on coagulation of highly turbid water. *Chemosphere* **72**, 189–196.

Lin, J. L., Huang, C. P., Chin, C. J. M. & Pan, J. R. 2008c Coagulation dynamics of fractal flocs induced by enmeshment and electrostatic patch mechanisms. *Water Research* **42** (17), 4457–4466.

Lin, J. L., Hua, L. C., Wu, Y. T. & Huang, C. P. 2016 Pretreatment of algae-laden and manganese-containing waters by oxidation-assisted coagulation: effects of oxidation on algal cell viability and manganese precipitation. *Water Research* **89**, 261–269.

Liu, H. J., Hu, C. Z., Zhao, H. & Qu, J. H. 2009 Coagulation of humic acid by PACl with high content of Al_{13}: the role of aluminum speciation. *Separation and Purification Technology* **70**, 225–230.

Ng, M., Liu, S., Chow, C., Drikas, M., Amal, R. & Lim, M. 2013 Understanding effects of water characteristics on natural organic matter treatability by PACl and a novel PACl-chitosan coagulant. *Journal of Hazardous Materials* **263**, 718–725.

Park, H., Lim, S. I., Lee, H. & Woo, D. S. 2016 Water blending effects on coagulation-flocculation using aluminum sulfate (alum), polyaluminum chloride (PAC), and ferric chloride ($fecl_3$) using multiple water sources. *Desalination and Water Treatment* **57** (16), 7511–7521.

Priya, T., Prakash, P. & Mishra, B. 2018 Understanding the coagulant activity of zirconium oxychloride to control THMs formation using response surface methodology. *Ecotoxicology and Environmental Safety* **159**, 28–37.

Qureshi, N. & Malmberg, R. H. 1985 Reducing aluminum residuals in finished water. *Journal of American Water Works Association* **77** (10), 101–108.

Shen, Y. 2005 Treatment of low turbid water by sweep coagulation using bentonite. *Journal of Chemical Technology & Biotechnology* **80** (5), 581–586.

Song, W., Li, X., Sun, S., Yang, Y. & Jia, R. 2015 Test study of enhanced coagulation for conventional treatment with low temperature and low turbidity. *Journal of Residuals Science & Technology* **12**, S39–S46.

Taylor, G., Ferrier, I., McLoughlin, I., Fairbairn, A., McKeith, I., Lett, D. & Edwardson, J. 1992 Gastrointestinal absorption of aluminium in Alzheimer's disease: response to aluminium citrate. *Age and Ageing* **21** (2), 81–90.

US Environmental Protection Agency 2018 *2018 Edition of the Drinking Water Standards and Health Advisories Tables*. USEPA, Washington, DC, USA.

Yang, Z. L., Gao, B. Y. & Yue, Q. Y. 2010 Coagulation performance and residual aluminum speciation of $Al_2(SO_3)_4$ and polyaluminum chloride (PAC) in Yellow River water treatment. *Chemical Engineering Journal* **165**, 122–132.

Yang, Z., Gao, B., Cao, B., Xu, W. & Yue, Q. 2011 Effect of OH^-/Al^{3+} ratio on the coagulation behavior and residual aluminum speciation of polyaluminum chloride (PAC) in surface water treatment. *Separation and Purification Technology* **80** (1), 59–66.

Yao, M., Nan, J. & Chen, T. 2014 Effect of particle size distribution on turbidity under various water quality levels during flocculation processes. *Desalination* **354**, 116–124.

Yasui, M., Strong, M. J., Ota, K. & Verity, M. A. 1997 *Mineral and Metal Neurotoxicology*. CRC Press, Boca Raton, FL, USA.

Yu, J., Wang, D., Yan, M., Ye, C., Yang, M. & Ge, X. 2006 Optimized coagulation of high alkalinity, low temperature and particle water: pH adjustment and polyelectrolytes as coagulant aids. *Environmental Monitoring and Assessment* **131** (1–3), 377–386.

Zhang, Z., Jing, R., He, S., Qian, J., Zhang, K., Ma, G., Chang, X., Zhang, M. & Li, Y. 2018 Coagulation of low temperature and low turbidity water: adjusting basicity of polyaluminum chloride (PAC) and using chitosan as coagulant aid. *Separation and Purification Technology* **206**, 131–139.

Zhu, Y., Li, Y. F., Miao, L. A., Wang, Y. P., Liu, Y. H., Yan, X. J., Cui, X. Z. & Li, H. T. 2014 Immunotoxicity of aluminum. *Chemosphere* **104**, 1–6.

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