Fate and fractionation of aluminum in a full-scale Al-based drinking water treatment plant

Hongyuan Liu, Haoran Liu and Yawei Xie

ABSTRACT

Residual aluminum in drinking water is widely concerning due to its potentially harmful effect on human health and drinking water distribution systems. The fate and fractionation of aluminum and the factors influencing residual aluminum in a full-scale Al-based drinking water treatment plant (DWTP) was presented in Jiaxing, China. The results showed that treated water residual aluminum concentration was less than 0.1 mg/L regardless of the seasonal change of raw water aluminum concentration. The addition of secondary flocculation had a negligible influence on treated water residual aluminum concentration due to the efficient removal of particulate aluminum by sand filter. Residual aluminum concentration of treated water was lower (mean 0.037 mg/L) in summer (average water temperature was 29 °C) than that (mean 0.067 mg/L) in winter (average water temperature was 16 °C). Significant positive relationships between particulate aluminum concentration and particle counts, as well as the total aluminum concentration of treated water and turbidity, were found. Those relationships provided the possibility to estimate residual aluminum concentration by monitoring particle counts and turbidity.

Key words | aluminum fractionation, full-scale treatment plant, residual aluminum, seasonal change

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INTRODUCTION

Poly-aluminum chloride (PACl) is commonly used as a pre-hydrolyzed coagulant in drinking water treatment plants (DWTP) due to its effective performance for the removal of suspended particles and dissolved natural organic matter (Krupińska 2018). However, although pre-hydrolyzed coagulants are less sensitive to changes in pH and temperature of purified water, they still increase the residual aluminum in water (Krupińska *et al.* 2019). Excess aluminum intake has been linked to human health problems, such as bone disorders and dialysis encephalopathy (Zatta *et al.* 2009). Exposure to aluminum through drinking water may be a contributing factor in Alzheimer's disease and related disease progression (Krupińska 2020). In addition to health effects, residual aluminum could deposit as aluminum hydroxide on the pipe wall, resulting in the decrease of carrying capacity. Therefore, total aluminum concentration in treated water has been limited in many countries, such as America (0.05–0.2 mg/L), Japan (0.1 mg/L) and China (0.2 mg/L) (Wang & Cui 2004; Kimura *et al.* 2013).

Although some studies on predicting or controlling the treated water total aluminum concentration were conducted (Tomperi *et al.* 2013), few studies focused on the changes of aluminum fractionation in each unit of the full-scale plant, which is particularly important for understanding the characteristic of aluminum removal along the water treatment process. There are two kinds of aluminum fractionation in water, namely dissolved aluminum, particulate aluminum, and dissolved monomeric aluminum in the main form of dissolved aluminum (Yang *et al.* 2010). The high level of different forms of aluminum usually implies unreasonable operation of the treatment process. For example, the high concentration of dissolved aluminum in treated water may result from unsuitable Al-based coagulant dosage or coagulation operation (Edzwald & Kaminski

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2007), while the high concentration of particulate aluminum indicates a poor efficiency of the solid–liquid separation process (John & Edzwald 1990). Therefore, we could optimize the water treatment process and operation conditions to reduce the aluminum concentration in treated water. For example, Viraraghavan & Srinivasan (2002) reported that clarifier and filtration units effectively removed particulate aluminum. Aluminum fractionation showed influences on human health and the distribution network. For example, dissolved monomeric aluminum is high in toxicity to human health (Yang *et al.* 2010), while particulate aluminum in the distribution system could cause increased turbidity, and a loss in hydraulic capacity (Bérubé 2004).

In addition to the treatment process and operation condition, the physicochemical characteristics of water, typically pH and temperature, affect residual aluminum concentration by influencing the solubility of aluminum in water. Aluminum is soluble under acidic and alkaline conditions but is insoluble at near-neutral pH (Rubinos *et al.* 2007). Hence, under acidic and alkaline conditions, more particulate aluminum could dissolve into water increasing the dissolved aluminum concentration in treated water. Moreover, the literature indicates that the residual aluminum concentration increased with an increase of water temperature between 17 and 27 °C (Ma *et al.* 2017). There are two explanations for the increase. On the one hand, the solubility of particulate aluminum increases with increasing water temperature, leading to an increase in dissolved aluminum concentration. Aluminum hydrolysis, on the other hand, is an endothermic process. Dissolved aluminum levels increase due to the changing of hydrolysis equilibrium of $Al(OH)_3$.

However, current studies mainly focused on the aluminum concentration in treated water and a drinking water distribution network based on the bench scale experiment (Akbari *et al.* 2018). Little attention has been given to reduce residual aluminum concentration on a full-scale treatment plant. For DWTPs, the operation of the treatment process is complex and there are many uncontrollable factors. Therefore, it is more meaningful to study aluminum fate and fractionation in drinking water plants.

In the present work, aluminum fate and fractionation were studied at a DWTP, which was located in Jiaxing, China (Jiaxing DWTP). The objectives of this study were to: (1) evaluate aluminum fractionation variation and removal efficiency of each unit of the full-scale plant, (2) identify factors influencing the total aluminum concentration during water treatment, (3) investigate the relationship between turbidity, particle counts, and aluminum concentration.

MATERIALS AND METHODS

Treatment plant

Jiaxing DWTP is located in the south of Jiaxing. Raw water from Changshui river (surface water) went through a spectrum of water treatment processes, including coagulation, flocculation, sedimentation, biological aerated filter (BAF), biofilter, ozone, granular activated carbon (GAC), secondary flocculation, sand filter and clearwell. The characteristics of raw water and treated water in Jiaxing DWTP are shown in Table 1. The designed capacity of the Jiaxing DWTP was 1.5×10^5 m^3/d. PACl (Haixia Jingshuiling Chemical Co. Ltd, Jiashan, China) was used as a coagulant. In the routine operation, PACl (basicity of 73% and containing approximately 10% Al_2O_3) was added twice. For the first time, a high PAC concentration (2.5 mg/L, as Al_2O_3) was added for coagulation. For the second time, a lower PAC concentration (0.5 mg/L, as

Table 1 | Characteristics of raw water and treated water in Jiaxing DWTP during the study

	Summer (2018/5/19–2018/7/20)		**Winter (2018/11/09–2018/12/11)**	
Parameter	**Raw water (average)**	**Treated water (average)**	**Raw water (average)**	**Treated water (average)**
Color (Pt-Co units)	31	<5	31	<5
Turbidity (NTU)	26.02	0.03	34.94	0.05
Alkalinity (mg/L)	105.6	99.7	110.3	103.6
Hardness (mg/L)	154.2	148.5	162.1	165.2
COD_{Mn} (mg/L)	5.2	1.3	4.5	1.5
pH	7.5	7.1	7.2	7.1
DOC (mg/L)	4.1	1.4	3.9	1.4
Water temperature (°C)	29	29	16	16

Al_2O_3) was added to remove biological leakage from the GAC process. The air-to-liquid ratio of BAF was 0.5:1. The ozone dosage of the ozone contact tank was 2 mg/L.

Sample collection

Water samples were collected from 10 sites along the water treatment process during 2018/5/19–2018/7/20 and 2018/11/09–2018/12/11 (see Supplementary material, Figure SI-1). The samples were collected in acid-treated semi-rigid polyethylene bottles and then stored at 4 °C (Viraraghavan & Srinivasan 2002). To prevent the problem of aluminum leaching, water samples did not have contact with glass throughout the experiment.

Coagulation experiments

The bench-scale coagulation experiment was conducted to study the effect of initial pH on total aluminum concentration. Three kinds of samples, namely flocculation water samples, sedimentation water samples, and filtration water samples, were collected. Sampling details are provided in the Supplementary material.

Analysis methods and chemicals

Aluminum fraction analysis

Aluminum concentration was measured by the Chrome-azurol S Spectrophotometric Method (GB/T 5750.6-2006).

The concentration of total aluminum and three aluminum fractionations were divided according to the pretreatment method of Driscoll & Letterman (1995):

1. For total aluminum concentration (Al_T): the sample was acidified with nitric acid to pH = 2 for at least 1 h.
2. For dissolved monomeric aluminum concentration (Al_{DM}): the sample was filtered using a membrane filter (Millipore 0.45 μm cellulose acetate filters).
3. For dissolved aluminum concentration (Al_D): the sample was filtered using membrane filter (Millipore 0.45 μm cellulose acetate filters) and acidified with nitric acid to pH = 2 for at least 1 h.
4. Particulate aluminum concentration (Al_P) equals Al_T minus Al_D.

Water quality analysis methods and chemicals

Details are provided in the Supplementary material.

RESULTS AND DISCUSSION

Aluminum in the full-scale plant treatment process

Variations of Al_T along the treatment process of Jiaxing DWTP

As shown in Figure 1, Al_T in raw water was significantly ($P < 0.01$) lower (0.038–0.041 mg/L) in summer than in winter (0.274–0.308 mg/L). This variability may result from seasonal changes in soil and water properties. It was reported that Al_T was positively correlated with turbidity in raw water (Li *et al.* 2018). As shown in Table 1, the turbidity of raw water in winter (35 NTU) was higher than that in summer (26 NTU). Hence, the high Al_T may be related to high turbidity. In addition, we found rainfall was frequent between November and December 2018 and the pH of raw water in winter (7.2) was lower than that in summer (7.5). Heavy rainfall and lower pH caused dissolution of Al into soil water, increasing Al_T in water (Kim *et al.* 2006). Among all water treatment units, sedimentation and BAF showed high efficiencies, 80 and 70% respectively, in removing Al_T, and their performances were not affected by the season. However, ozonation and GAC had almost no effect on removing Al_T. There were two reasons for this observation: one was the lower Al_T in the influent; the other was related to the operation of GAC. The GAC process in Jiaxing DWTP was upward flow, which weakened its filtration effect for Al_T. In other words, biological and chemical treatments have a weaker removal effect on Al_T, while physical treatments have a stronger removal effect. Viraraghavan & Srinivasan (2002) reported that sedimentation and filtration units effectively removed Al_P and GAC was capable of removing part of the organic dissolved aluminum. These phenomena indicate that the removal of Al_T is likely related to the forms of aluminum.

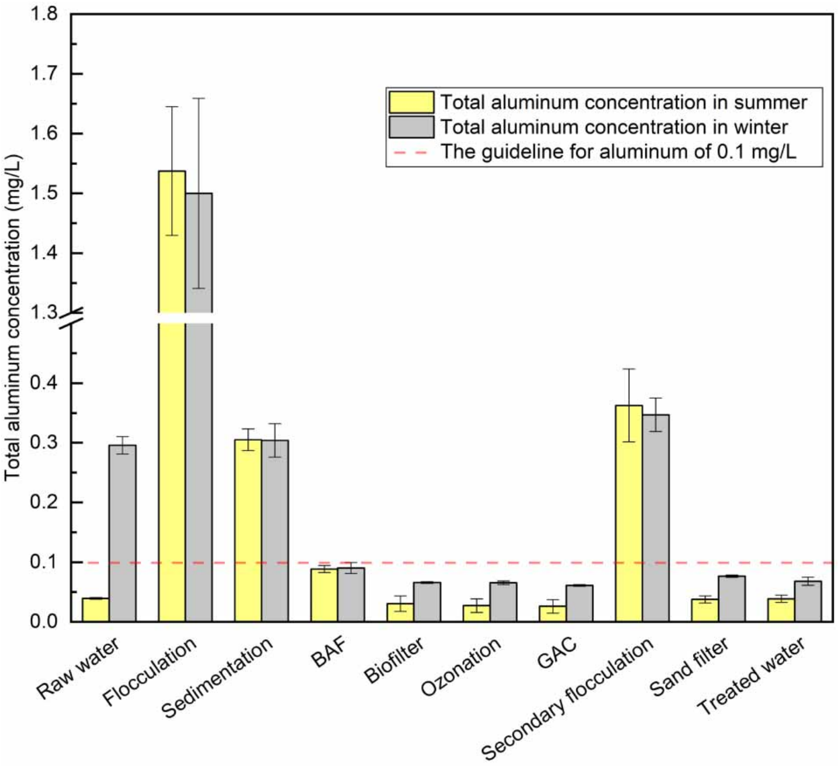

Figure 1 | The concentration of total aluminum along the treatment process of Jiaxing DWTP in summer (2018/5–2018/7) and winter (2018/11–2018/12).

In addition, although Al_T could be reduced to a level lower than 0.1 mg/L, which is the standard of Zhejiang province, China (Zhejiang Urban Water Industry Association 2018), it is worth noting that Al_T of treated water in summer was lower than that in winter. We found the removal performances of biofilters and sand filters were affected by the season. The change of water temperature may change aluminum fractionation and influence the removal capacity of the treatment process unit.

Variations of aluminum fractionation along the treatment process of Jiaxing DWTP

The increases of Al_T in flocculation water and secondary flocculation water were caused by PACl additions and the proportions of Al_P decreased along the treatment process, except for the two flocculation steps at which PACl was added (Figures 1 and 2). Al_P in flocculation water and secondary flocculation water accounted for more than 70% of the total. It was observed that the increase in the proportions of Al_P after the second PACl addition was much larger than that after the first PACl addition. However, this observation does not indicate more Al_P in secondary flocculation water than that in flocculation water because the increased Al_P is almost the same for the first PACl addition (0.04 mg L^{-1}/mg L^{-1} PACl) and the second (0.05 mg L^{-1}/mg L^{-1} PACl). On the other hand, PACl mainly contributed to Al_P, which was very low in GAC water. Therefore, the proportion of Al_P increased greatly in secondary flocculation water. Fortunately, although the second PACl addition could greatly increase Al_P, Al_P was largely removed after the sand filter treatment process (Figure 3(a) and 3(b)). At the same time, Al_D was also low. Comparing with Figure 1, we found that Al_D was the most important part of Al_T in treated water. In addition, Al_D and Al_{DM} in treated water fluctuated between 0.02 and 0.05 mg/L and Al_{DM} was the main constituent of Al_D in treated water (Supplementary material, Figure SI-2).

As shown in Figure 3(a) and 3(b), the performance of sedimentation was efficient for removing both Al_P and Al_D. Biofilters and sand filters had a removal efficiency of more than 80% for Al_P, while the removal efficiency of Al_D is poor, especially in winter. This suggested that

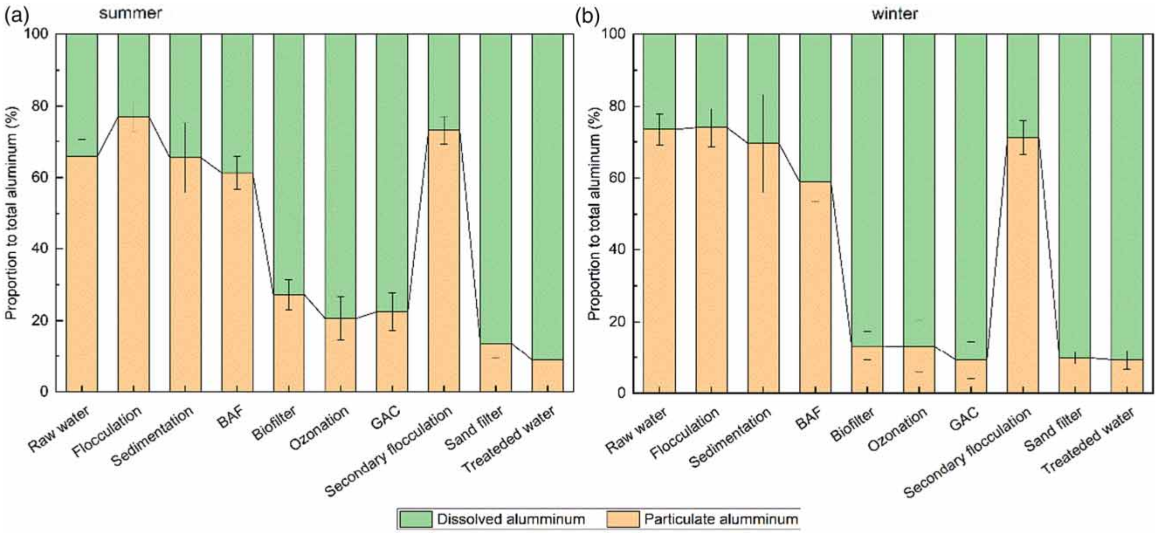

Figure 2 | The proportion of particulate aluminum and dissolved aluminum along the treatment process of Jiaxing DWTP.

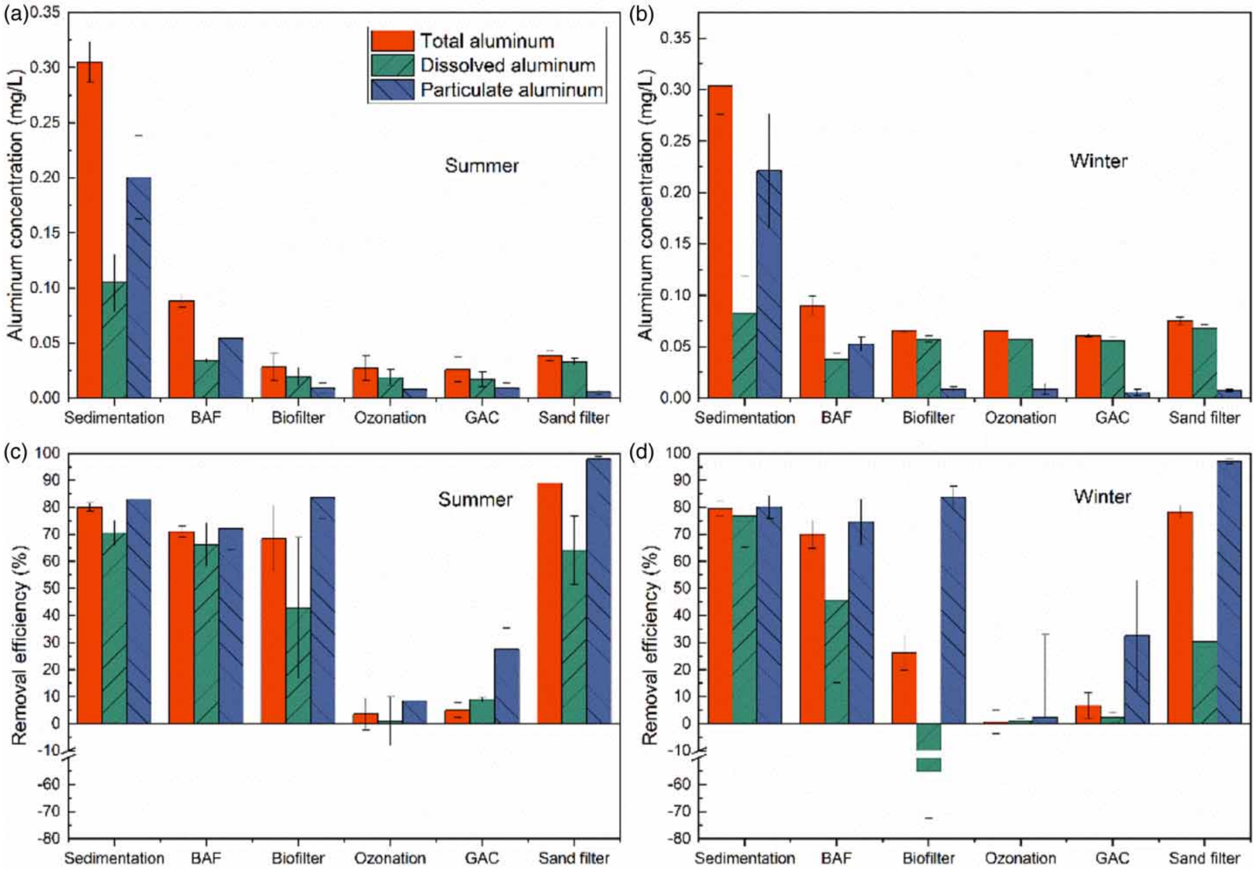

Figure 3 | Removal of aluminum fractionation along the treatment process of Jiaxing DWTP. The variations of aluminum fractionation in summer (2018/5–2018/7) (a) and winter (2018/11–2018/12) (b); the removal efficiency of different forms of aluminum of treatment process in summer (c) and winter (d).

sedimentation could simultaneously remove Al_P and Al_D efficiently while filtration mainly removed Al_P. In the coagulation process, PACl could form Al_P and Al_D by hydrolyzation. Since the flocs (formed newly in coagulation water) have a larger active site density and specific surface area than the solids (presented in raw water), aluminum hydrolysates tend to adhere to the flocs and destabilize them by surface enmeshment or charge neutralization. Then, Al_P and Al_D could be removed by sedimentation. For filtration, Al_P is trapped in the gap of the filter material, while Al_D could pass through the filter material, resulting in the presence of Al_D in sand filter water.

In addition, it should be noted although both Al_T and Al_P decreased, Al_D increased after the biofilter process in winter (Figure 3(d)). The dissolution of Al_P and re-dissolution of aluminum from biofilm to water may be potential explanations for the observation. Moreover, the low removal efficiency of the O_3/GAC process indicated that the adsorption effect of activated carbons on residual aluminum removal was poor.

Factors influencing Al_T

Effect of PACl dosage

The managers of Jiaxing DWTP increased PACl dosage from 2.5 to 3.0 mg/L as Al_2O_3 2018/6/7 due to the increased turbidity of the raw water. As shown in Supplementary material, Figure SI-3, the change of the operation condition resulted in an increase in residual aluminum in treated water. The mean Al_T in treated water was 0.0321 mg/L with a small fluctuation (95% confidence interval, CI: 0.0319–0.0323) during 2018/5/25–2018/6/16, while it was 0.0504 mg/L with a more significant fluctuation (95% confidence interval, CI: 0.0498–0.0511) during 2018/6/7–2018/6/17. This indicated that although Al_T could be reduced to a level that meets the limitation in treated water, the health risk for users would increase if the PACl dosage increased during operation.

Effect of water temperature

To study the effects of water temperature on Al_T in treated water, we selected two temperature ranges (27–29 and 15–17 °C, respectively) for the experiment. As shown in Figure 4(a), the mean Al_T in treated water at 27–29 and 15–17 °C were 0.037 and 0.067 mg/L, respectively. Different results were obtained for the influence of temperature on Al_T in treated water. Van Benschoten *et al.* (1994) reported that Al_T in treated water in summer was higher than that in winter. However, John & Edzwald (1990) reported that the highest Al_T occurred during the coldest periods. In theory, the influence of water temperature on Al_T could be related to the reaction rate and equilibrium of aluminum hydroxide hydrolysis. The solubility of Al_P increased with the increase of water temperature so that Al_D increased. However, for a complex system like the full-scale treatment processes, it is difficult to give an exact explanation for the observed phenomenon based on the influence of a single

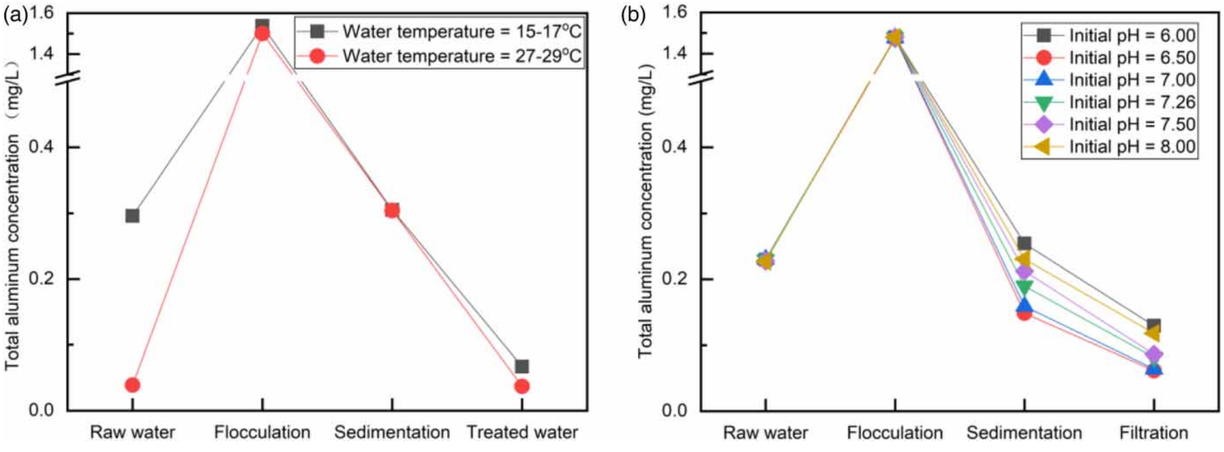

Figure 4 | Effect of water temperature on total aluminum concentration in water treatment processes (PACl dosage is 2.5 mg/L as Al_2O_3 for coagulation and 0.5 mg/L as Al_2O_3 for secondary flocculation. pH: around 7.3) (a). Effect of initial pH on total aluminum concentration in water treatment processes (coagulation experiments: bench-scale) (b).

factor. For Jiaxing DWTP, we found that bio-related processes, such as BAF, biofiltration, GAC and sand filtration (microorganisms often adhere on sand), performed higher removal rates of Al_D in summer than in winter (Figure 3(c) and 3(d)). Although we cannot distinguish the specific organisms, this seems to indicate that biological removal can control Al_D (Hydes 1989). Due to the seasonal variation of the bacterial community, the activity of microorganisms to remove aluminum may be lower in winter (Liu *et al.* 2017). Comparing with Figure 3(a) and 3(b), the removal efficiency of sand filters on Al_D was poor in winter, resulting in higher Al_T in treated water.

Effect of initial pH

To study the effect of initial pH on Al_T, a bench-scale coagulation experiment was conducted. The details are shown in the Supplementary material.

Correlation of residual aluminum concentration with particle counts and turbidity

Correlation of Al_P with particle counts

There is a positive correlation between Al_P and particle counts (Supplementary material, Figure SI-4). The addition of PACl increases Al_P and particle counts, and after the subsequent process, both reduced. This correlation was good only for the process before ozonation. Ozonation could convert higher molecular weight organic matters to lower ones (Yu *et al.* 2018), which increases particle counts. Therefore, the correlation is weak. It is worth noting that more than 80% of DWTPs do not have advanced treatment processes (Bei *et al.* 2019). Therefore, even though the correlation has such a defect, it may be useful for DWTPs without advanced treatments to predict Al_P in treated water with particle counts.

Here, we found a good correlation between Al_P and particle counts (>10 μm) ($R^2 = 0.866$) (Figure 5(a)). The particle counts and Al_P were analyzed, and the following equation was obtained:

$$C_{Pa} = 0.0001 \times C_p + 0.0173 \tag{1}$$

where C_{Pa} is the concentration of particulate aluminum (mg/L) and C_p is the concentration of particles (mL^{-1}).

The regression model indicated that if Al_P was <0.050 mg/L, particle counts were <327 mL^{-1}. For the conventional DWTPs, the managers could calculate Al_P by measuring the particle counts.

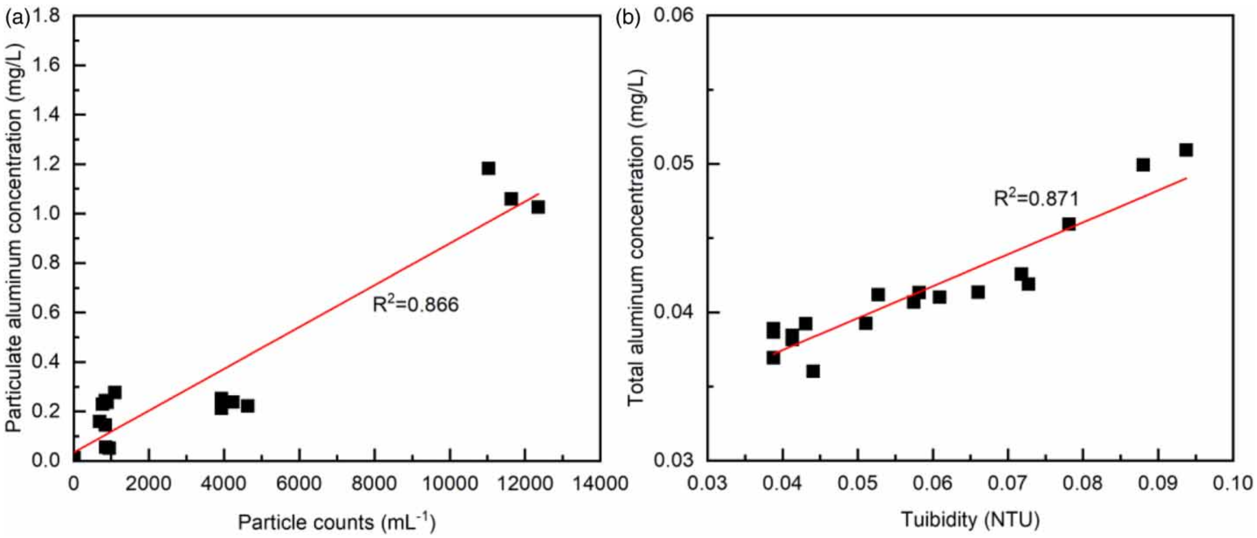

Figure 5 | Relationship between particle counts (>10 μm) and particulate aluminum concentration in water (a) and relationship between turbidity and total aluminum concentration in treated water (b).

Correlation of Al_T with turbidity

In addition, there is a positive correlation between Al_T and turbidity within 0.1 NTU in treated water. This was useful in Zhejiang province, where turbidity is required to be less than 0.1 NTU. Figure 5(b) shows a significant correlation between turbidity and Al_T ($R^2 = 0.871$) in treated water.

Here, using the data of treated water, the turbidity and Al_T were analyzed, and the following equation was obtained:

$$\text{Turbidity (NTU)} = 4.649 * C_{Ta} - 0.134 \tag{2}$$

where C_{Ta} is the concentration of total aluminum (mg/L).

For the general DWTPs, the managers could calculate Al_T by measuring the turbidity of treated water.

CONCLUSIONS

The aluminum fate and fractionation study was conducted in a full-scale Al-based DWTP. Here, the main conclusions drawn from this work are as follows:

1. Although Al_T could match corresponding standards after treatment, PACl dosage increases Al_T in treated water and the potential risk of exceeding the limit.
2. Al_P was the paramount fraction in raw water while Al_D was the dominant fractionation in treated water.
3. Sedimentation and BAF could effectively remove Al_P and Al_D. Filtration could effectively remove Al_P. The ability of O3/GAC to remove Al_P and Al_D is low.
4. Al_T in treated water in summer (27–29 °C) was lower than that in winter (15–17 °C).
5. For the processed water (before ozonation), the correlations of particulate aluminum with particle counts (>10 μm) were positive. For treated water, the correlations of total aluminum with turbidity were positive.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this paper is available online at https://dx.doi.org/10.2166/aqua.2020.005.

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