

## Variation of assimilable organic carbon during coagulation by aluminum and iron in drinking water treatment

Tao Liang and Jun Ma

### ABSTRACT

The variation of Assimilable Organic Carbon (AOC) during the coagulation of raw surface water and synthetic water was investigated. It was found that the removal rate of AOC concentration during alum coagulation of raw surface water was only about 24.5% at high coagulant dosages. A higher removal rate of AOC in the case of  $\text{FeCl}_3$  coagulation (about 65.9%) was observed under the same dosages, but AOC-NOX increased from  $143 \mu\text{g L}^{-1}$  to  $473 \mu\text{g L}^{-1}$  during  $\text{FeCl}_3$  coagulation. The results showed a relationship between the solution pH in the coagulated water and the reduction of AOC. The AOC of water decreased efficiently during coagulation in the case of synthetic water at lower coagulant dosages, and the case of coagulation with  $\text{FeCl}_3$  also resulted in a better removal of AOC. The lowest value of AOC achieved in the case of synthetic water ( $54 \mu\text{g L}^{-1}$ ), was close to the biostability criteria of drinking water ( $50 \mu\text{g L}^{-1}$ ). It can be concluded that not only large molecules but also low-molecular weight fractions of organics can be removed during coagulation. The performance of coagulation effect was evaluated based on the following parameters: pH, turbidity, UV absorbance at 254 nm ( $\text{UV}_{254}$ ) and Total Organic Carbon (TOC). The results showed that the coagulation effectiveness has a substantial impact on AOC reduction.

**Key words** |  $\text{Al}_2(\text{SO}_4)_3$ , assimilable organic carbon, coagulation,  $\text{FeCl}_3$ , pH

**Tao Liang** (corresponding author)  
**Jun Ma** (corresponding author)  
State Key Laboratory of Urban Water Resources and Environment,  
National Engineering Research Center of Urban Water Resources,  
School of Municipal and Environmental Engineering,  
Harbin Institute of Technology,  
2627 Mail Box,  
202 Haihe Road,  
Harbin 150090,  
China  
Tel.: +86 451 8628 3210  
E-mail: tao.liang2009@163.com

### INTRODUCTION

Not all organic carbons in water can be used by bacteria. Only low-molecular-weight (LMW) fractions can be directly utilized for metabolism (Van der Kooij & Hijnen 1984). Assimilable organic carbon (AOC) is the fraction of dissolved organic carbon (DOC) that is most easily consumed by microorganisms, resulting in a proliferation of microbial cells (Van der Kooij 1992). According to Hem & Efraimsson (2001), 50–70% of the AOC fraction consists of organic molecules smaller than 1,000 Da. AOC usually constitutes only a small fraction (0.1–9%) of total organic carbon (TOC) in drinking water, and it has been regarded as one of the most important quality parameters for microbiological stability in drinking water treatment and distribution systems (Van der Kooij 1992; Escobar &

Randall 2001). Removal of AOC during the production of drinking water not only deprives heterotrophic bacteria of a substrate indispensable for their survival and multiplication in the water phase, but also limits bacterial colonization in the drinking water distribution system (Monika *et al.* 2005).

Several biological tests have been developed to assess the level of biodegradable organic matter in water (Huck 1990). While the removal of AOC has been extensively studied and optimized during biological processes (Hu *et al.* 1999; Escobar & Randall 2001; Lehtola *et al.* 2001), limited studies have been focused on the variation of AOC in conventional water treatment, especially during the coagulation stage. Coagulation may be an essential step in reducing the concentration of AOC in drinking water.

doi: 10.2166/aqua.2009.091

However, the studies on the variation of AOC during coagulation reported in the literature are somewhat contradictory. Kaplan *et al.* (1994) reported that the conventional treatments had little impact on AOC concentration. Other investigators reported that the AOC during coagulation varied widely and was not stable, ranging from 0 to 85% by clarification (Huck *et al.* 1991). Additionally, Zhou *et al.* (1999) and Li *et al.* (2003) reported that the value of AOC increased after coagulation. Since coagulation is ubiquitously practiced and is a very important process in water treatment, the study of AOC variation during the coagulation process is needed.

The aim of this investigation is to evaluate the effects of coagulation conditions on AOC concentration. AOC is assumed as a parameter which partly represents the LMW fraction in water.

## MATERIALS AND METHODS

### Coagulation

The coagulation was conducted using either the River Songhua raw water or synthetic surface water as the target of study to assess the impact of coagulants on the AOC concentration without pretreatment. The first case is the process of coagulation and sedimentation with River Songhua raw water (known as Process I). Table 1 shows the characteristics of River Songhua raw water. The other is the process of coagulation and sedimentation with synthetic water (known as Process II). The synthetic water is prepared by spiking kaolin and fulvic acid (Jufeng Company, Shanghai, China) in Milli-Q ultra-pure water.

**Table 1** | Water quality of River Songhua (May 2007 to July 2007)

Parameter	Range	Average
pH	7.78–7.91	7.85
Turbidity (NTU)	20.8–83.6	30.1
Temperature (°C)	20–27	23
TOC (mg L <sup>-1</sup> )	5.08–5.97	5.35
UV <sub>254</sub> (cm <sup>-1</sup> )	0.106–0.601	0.188
AOC (μg L <sup>-1</sup> )	1,624–1,829	1,726
AOC-P17 (μg L <sup>-1</sup> )	1,497–1,669	1,583
AOC-NOX (μg L <sup>-1</sup> )	127–160	143

**Table 2** | Water quality of synthetic water

Parameter	Range	Average
pH	7.18–7.45	7.35
Turbidity (NTU)	19.60–20.10	20.00
Temperature (°C)	23–25	24
TOC (mg L <sup>-1</sup> )	9.88–10.42	10.37
UV <sub>254</sub> (cm <sup>-1</sup> )	1.004–1.013	1.010

The quality of the source water (process II) is summarized in Table 2. In this study, Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> and FeCl<sub>3</sub> are the coagulants used in both process I and process II. The tests were conducted by rapid mixing for 1 min at 120 rpm, which corresponds to a mean velocity gradient of 75 s<sup>-1</sup>, followed by a 20 min flocculation period at 30 rpm, which corresponds to a mean velocity gradient of 23 s<sup>-1</sup>, and a 30 min settling period.

### AOC analysis

The 400 mL water samples were collected in clean, carbon-free Erlenmeyer flasks in duplicate and were pasteurized for 30 min at 60°C in a water bath. To make sure that organic carbon is the only limiting nutrient, phosphate salt was added to the samples to achieve a final concentration of 370 mg/l of K<sub>2</sub>HPO<sub>4</sub>·3H<sub>2</sub>O. In case chlorine is present in the sample, sodium thiosulphate was used (30 mg L<sup>-1</sup> in the 600 ml flask). The strains used in the AOC test are: *Pseudomonas fluorescens* strain P17, which is capable of utilizing a wide range of low-molecular-weight compounds at very low concentrations, and a *Spirillum* sp. strain NOX, which only utilized carboxylic acids. Finally, samples were inoculated with strain P17 and strain NOX simultaneously, to reach an initial concentration of about 300 cfu ml<sup>-1</sup>. The prepared samples were incubated for 3 or 4 days at 20°C until maximum growth is reached. The maximal number of colonies and the yield values determined for each strain were used in the calculation of the AOC concentration. The yield factors determined in our laboratory are similar to the values determined by Van der Kooij (1990), i.e. 4.1 × 10<sup>6</sup> and 1.2 × 10<sup>7</sup> cfu/μg of C-acetate for P17 and NOX, respectively. The total AOC concentration is calculated as the sum of AOC levels measured with two bacterial strains (total AOC = AOC-P17 + AOC-NOX)

since the two organisms do not have a wide range of overlap in compounds that they can metabolize (LeChevallier *et al.* 1993).

## Experimental methods

For each raw water sample, pH, turbidity,  $UV_{254}$  (indicative of the humic and aromatic characteristics), TOC (indicating the mass of organic material) and total AOC concentrations are assessed after water treatment.

UV absorbance is measured at 254 nm with a Shimadzu UV-2550 spectrometer. TOC analysis is performed with a Multi N/C 3100 Total Organic Carbon Analyzer.

## RESULTS AND DISCUSSION

### Variation of AOC fraction in the coagulation of raw water

In this process, the coagulants  $Al_2(SO_4)_3$  and  $FeCl_3$  are dosed into the raw water directly without pre-oxidation, both of their dosages being 0.12, 0.18, 0.24, 0.30 and 0.36  $m\text{ mol L}^{-1}$ , as Al and Fe, respectively.

As shown in Figure 1, after the raw water from the River Songhua was treated by coagulation with  $Al_2(SO_4)_3$ , the total AOC concentration decreased very slightly. In the first case (Process I), the raw water had high levels of AOC, ranging from 1,726 to 1,303  $\mu\text{g L}^{-1}$ , of which, all of the AOC-NOX values were less than 310  $\mu\text{g L}^{-1}$ , while the AOC-P17 values ranged from 996–1,583  $\mu\text{g L}^{-1}$ , which

were much higher than the AOC-NOX values. Under the same dosage of coagulant, a higher removal rate for AOC in the case of  $FeCl_3$  coagulation was observed. The lowest value of AOC, at 597  $\mu\text{g L}^{-1}$ , was obtained at a dosage of 0.30  $m\text{ mol L}^{-1}$  of Fe.

Comparing the coagulation with  $Al_2(SO_4)_3$  and  $FeCl_3$  (Figure 2), the case of coagulation with  $FeCl_3$  also results in better removal of turbidity,  $UV_{254}$  and TOC after sedimentation, and the value of pH in the coagulated water is less than in the case of coagulation with  $Al_2(SO_4)_3$  under the same dosage.

Such variation and differences can be explained by the following reasons. First, the amount of AOC removal in the coagulation process is strongly related to the solution pH in the coagulated water. The variation of AOC removal rates in this study showed that the removal of AOC was favored for the low pH range in the coagulated water. Semmens & Ayers (1985) showed that the solution pH was an important factor in the coagulation process. At lower pH, more positively charged humic acid (HA) is enhanced and the coagulation is mainly due to charge neutralization. Typically, the best precipitation for humic substances has been obtained at pH 4–5 with ferric and at pH 5–6 with alum (Lefebvre & Legube 1990). After the coagulants were dosed into the raw water, the reduction of the solution pH in the case of coagulation with  $FeCl_3$  was more than in the case of coagulation with  $Al_2(SO_4)_3$ . With a more favorable pH value, iron salts typically resulted in better coagulation effectiveness and greater removal of AOC. In contrast, the fact that the pH value in the coagulated water with  $Al_2(SO_4)_3$  reduced less indicated that the AOC, which

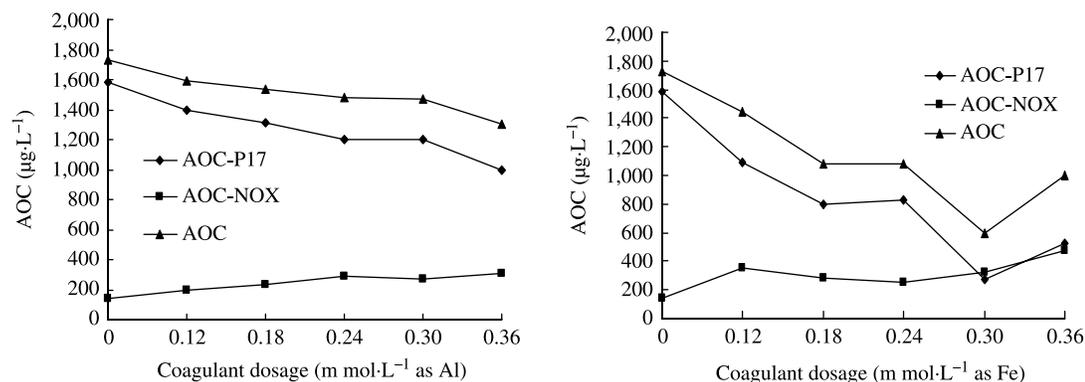


Figure 1 | Variation of AOC fraction in the coagulation of actual surface raw water.

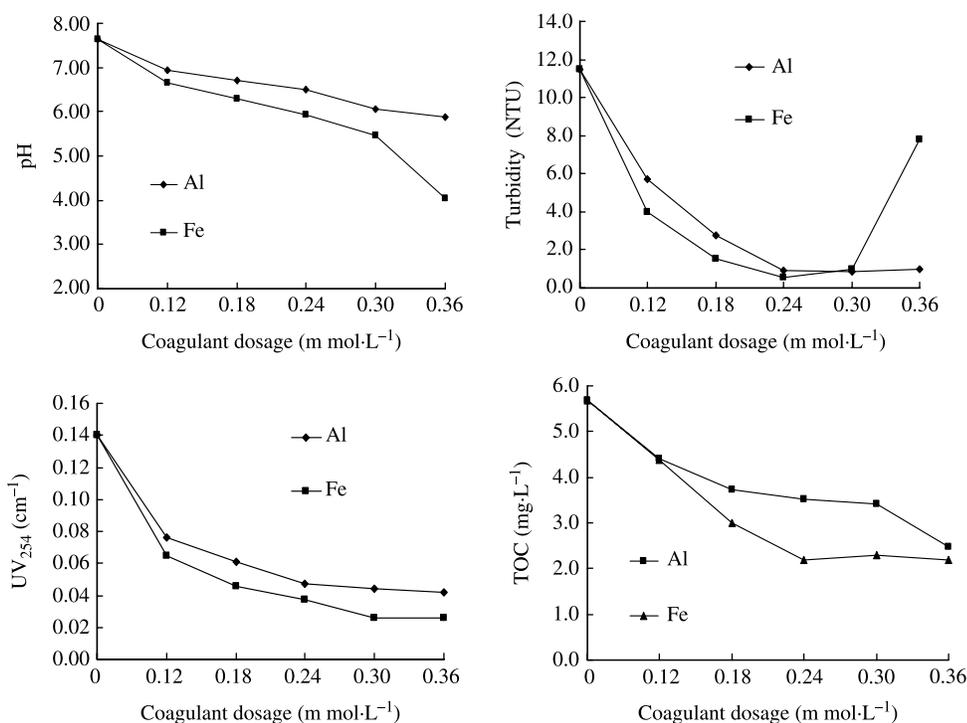


Figure 2 | Effect of water quality in the coagulation of actual surface raw water.

represents hydrophilic organics, are only slightly affected in the process of coagulation by  $\text{Al}_2(\text{SO}_4)_3$ .

Second, there is an equilibrium between adsorption and desorption. Flocs can be described as highly porous and loosely connected aggregates made of many primary particles (Randtke 1988). A floc has a very large specific surface area and polar functional groups, and can adsorb a large amount of organics in water (Schnitzer & Khan 1979). When the coagulant is dosed into water, the metal ions will combine with HA to form complexes. The formed metal hydroxide flocs can removed nonpolar LMW organics in water and the metal hydroxide flocs formed in the process will release the LMW organics which are adsorbed and complexed by HA (Zhang & Wang 1998). The increasing LMW organics play a very important role in enhancing AOC in water (Zhang & Wang 1999). The reason that the reduction of AOC in  $\text{FeCl}_3$  coagulation is higher may be related to more LMW organics being removed by the metal hydroxide flocs in the  $\text{FeCl}_3$  coagulation process. The adsorption of the LMW organics on the pre-formed metal hydroxide flocs is accompanied by the release of LMW from HA until equilibrium is reached (Zhang & Wang 1999).

The increase of AOC-NOX in the solution during the alum coagulation process might result from the release of LMW organics. Within the pH range investigated, the adsorption and desorption are two competitive reactions happening between the hydroxide in solution and the functional groups of HA.

Raw water is so complex that the effect of coagulation in raw water is influenced by many factors such as the coagulation conditions, the characteristics of natural organic matter (NOM) and the nature and concentrations of inorganic compounds. It is clear that the chemical characteristics of the TOC, as well as other physicochemical characteristics of raw water, will determine the degree of AOC removal by coagulation.

### The mechanism of AOC reduction by coagulation with $\text{FeCl}_3$

The data in Figure 1 also show that  $\text{FeCl}_3$  has a quite different effect on the AOC concentration from the case of coagulation by  $\text{Al}_2(\text{SO}_4)_3$ . The lowest value of AOC after coagulation is obtained at a dosage of  $0.30 \text{ m mol L}^{-1}$  of Fe,

since AOC begins to increase at a dose of  $0.36 \text{ m mol L}^{-1}$  of Fe. Starting from the dosage of  $0.30 \text{ m mol L}^{-1}$  of Fe, an abrupt AOC-NOX increase occurred, reaching a peak value of  $473 \mu\text{g L}^{-1}$ . Moreover, when  $\text{FeCl}_3$  is used at a dosage of  $0.30 \text{ m mol L}^{-1}$  of Fe, AOC-NOX begins to exceed AOC-P17. A water type is characterized by both the AOC concentration and AOC composition. Starting from a dosage of  $0.30 \text{ m mol L}^{-1}$  of Fe, compounds available for the strain NOX (most likely carboxylic acids) predominate in the AOC concentration.

A similar trend in AOC variation was observed in the catalytic oxidation process (Liang *et al.* 2008). Catalytic oxidation by-products include a great deal of carboxyl acids, which are mainly represented by AOC-NOX, and compounds available for strain P17 can be partly catalytically oxidized (Liang *et al.* 2008). Hence, AOC-NOX usually increases rapidly and AOC-P17 decreases in the catalytic oxidation process. This AOC variation cannot be observed in the coagulation process. It is assumed that a certain catalytic oxidation existed in the case of coagulation with  $\text{FeCl}_3$  but did not in the case of coagulation with  $\text{Al}_2(\text{SO}_4)_3$ .

Stumm & Morgan (1987) demonstrated that  $\text{Fe}^{3+}$  has a certain oxidation ability, and that some organics in water can be easily oxidized by  $\text{Fe}^{3+}$ . When  $\text{FeCl}_3$  is added to water,  $\text{Fe}^{3+}$  oxidizes the phenol hydroxyl group in fulvic acid to produce a semiquinone radical at once and  $\text{Fe}^{3+}$  is reduced to  $\text{Fe}^{2+}$  (Stumm & Morgan 1987). Again  $\text{Fe}^{2+}$  can reduce the quinone group to produce semiquinone radicals (Stumm & Morgan 1987). Hence,  $\text{Fe}^{2+}$ - $\text{Fe}^{3+}$  is the oxidation and reduction catalyst for fulvic acid to generate

free radicals (Stumm & Morgan 1987). When  $\text{FeCl}_3$  is used as the coagulant,  $\text{Fe}^{3+}$  also has a certain catalytic effect on the free radicals' formation reaction of fulvic acid (Zhang & Wang 1999). It is indicative that  $\text{Al}^{3+}$  has no catalytic effect, which might be the main reason that the variation of AOC in the case of coagulation with  $\text{FeCl}_3$  is different from the case of coagulation with  $\text{Al}_2(\text{SO}_4)_3$  when the coagulation dosage exceeds  $0.30 \text{ m mol L}^{-1}$  of Fe.

### Variation of AOC fraction in the coagulation of synthetic water

Most raw waters have quite different characteristics from synthetic waters. Using synthetic water may simplify the system in the coagulation process.  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{FeCl}_3$  are used as coagulants in both process I and process II to study their effect on AOC, and the dosages are 0.015, 0.030, 0.045, 0.060 and  $0.075 \text{ m mol L}^{-1}$  of Al and Fe, respectively.

It can be seen from Figure 3 that the AOC is reduced greatly with the increase of coagulant dosages. The data from Figure 3 show a decrease in AOC concentrations of 64% and 72% with an alum dosage of  $0.045 \text{ m mol L}^{-1}$  of Al (pH 5.1) and  $0.060 \text{ m mol L}^{-1}$  of Al at pH 4.9, respectively. The case of coagulation with  $\text{FeCl}_3$  also results in a better removal of AOC, and the removal rate of AOC is about 90% starting from the dosage of  $0.045 \text{ m mol L}^{-1}$  of Fe. The lowest value of AOC after coagulation ( $54 \mu\text{g L}^{-1}$ ) is close to the biostability criteria of  $50 \mu\text{g L}^{-1}$  (LeChevallier *et al.* 1993). AOC-NOX changes a little in process II except for one data point (dosage of  $0.045 \text{ m mol L}^{-1}$  of Al), and the value of AOC-P17 is always the largest proportion of

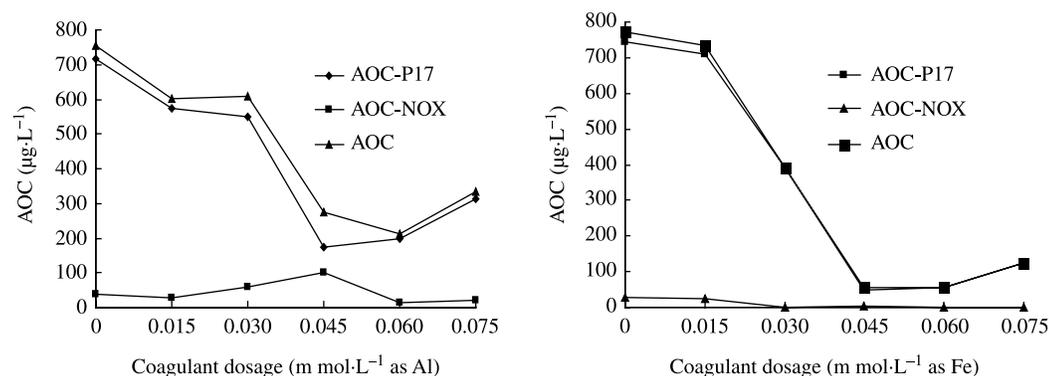


Figure 3 | Variation of AOC fraction in the coagulation of synthetic water.

the AOC. The AOC-NOX values for three data point in the case of coagulation with  $\text{FeCl}_3$  is lowest and decreased to  $0 \mu\text{g L}^{-1}$ . It is not observed that  $\text{Fe}^{3+}$  has a catalytic effect during coagulation in the case of synthetic water, which might be due to the low coagulant dosages in process II.

The coagulation with an alum dosage of  $0.045 \text{ m mol L}^{-1}$  of Al achieved similar turbidity,  $\text{UV}_{254}$  and TOC removals (81%, 98% and 84%, respectively). The optimum coagulant dose was identified as the lowest dosage at which there was maximum turbidity,  $\text{UV}_{254}$  and TOC removal. It would be expected that  $0.045 \text{ m mol L}^{-1}$  of Al is the optimum  $\text{Al}_2(\text{SO}_4)_3$  coagulant dose in the case of synthetic water. The data from Figure 4 show that  $\text{FeCl}_3$  also results in a better coagulation effect than  $\text{Al}_2(\text{SO}_4)_3$  in the case of synthetic water and  $0.060 \text{ m mol L}^{-1}$  of Fe is the optimum  $\text{FeCl}_3$  coagulant dose. Starting from a dosage of  $0.060 \text{ m mol L}^{-1}$  of Fe, TOC decreased to less than  $1 \text{ mg L}^{-1}$ .

Easton & Jago (1993) reported that some of the reduction in AOC levels might have been due to biological

activity because no oxidation was applied prior to coagulation, and there were long residence times within the sedimentation basins. In this study, the residence times for all the samples are the same, but there are large differences in AOC reduction in the cases of different coagulants. Therefore, biological activity is not the only reason for diminishing AOC content. Christlan *et al.* (2000) thought that AOC fractions include both low- and high-molecular-weight molecules. Most of the AOC removal is related to large molecules, whilst little or no AOC LMW fraction can be reduced by coagulation (Christlan *et al.* 2000). According to Christlan *et al.* (2000), low removal efficiency of AOC during coagulation would be expected if the AOC was composed of low-molecular-weight molecules. According to Hem & Efraimsson (2001), 50–70% of the AOC fraction consists of organic molecules smaller than 1,000 Da. However, starting from a dosage of  $0.045 \text{ m mol L}^{-1}$  of Al and Fe, the reduction of AOC exceeds 60%, which means that LMW fractions can also be removed in the coagulation process.

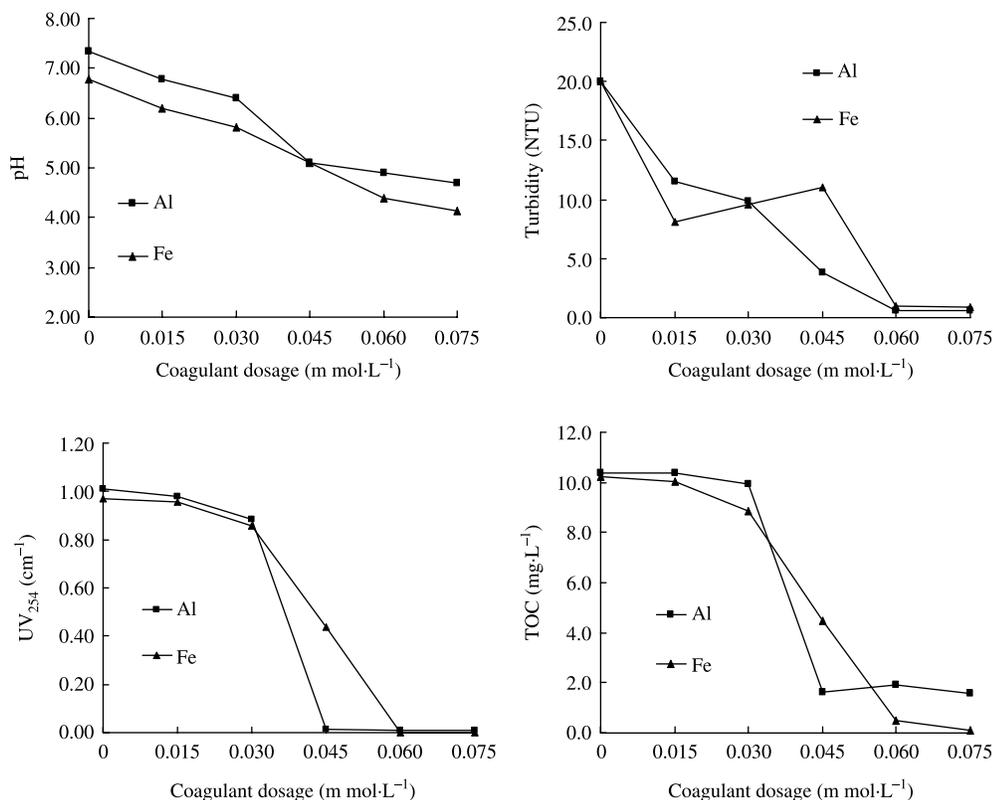


Figure 4 | Effect of water quality in the coagulation of synthetic water.

Coagulation pH is dependent on the coagulant type and the treated water sample. In synthetic water, the solution pH decreases greatly with low coagulant dosage. The high AOC removal rate is observed for the water sample with a high pH value reduction. The experimental data indicate that the pH of coagulation is a very influential parameter.

In comparison to the effect of AOC removal in both process I and II, the reduction of all the water quality parameters in the case of synthetic water at low dosages is much larger than the case of raw water at high dosages. It can be concluded that the AOC removal during the coagulation process would be determined by the composition of organic matter in the water. It can also be concluded that the coagulation effectiveness has a strong impact on AOC removal during coagulation. The better the coagulation effectiveness of coagulation, the more the LMW fraction can be adsorbed or precipitated through charge neutralization. It is assumed that enhanced coagulation could reduce AOC efficiently because of the high effectiveness of coagulation.

Increase of AOC in coagulation is not observed in this study, and we can expect that the existence of algae in the influent of coagulation may be a reason for the AOC increase in the reports of Zhou *et al.* (1999) and Li *et al.* (2003). If the algal cells are indeed leaking or breaking, one can expect a significant DOC and AOC increase in water (Frederik *et al.* 2007).

## CONCLUSIONS

This paper investigated the impact of coagulants on the variation of AOC during coagulation of actual surface raw water and synthetic water. Based on the results obtained, the following conclusions can be reached:

- (1) The case of coagulation with  $\text{FeCl}_3$  results in a better removal of AOC than the case of coagulation with  $\text{Al}_2(\text{SO}_4)_3$  under the same dosages in both actual surface raw water and synthetic water.
- (2) Removal of AOC by coagulation of actual surface raw water or synthetic water is correlated to the solution pH in the coagulated water and the coagulation

effectiveness. Enhanced coagulation could be an effective way of improving AOC removal.

- (3) AOC-P17 always constitutes the largest proportion of AOC in the coagulation process, unless the water is oxidized by excessive coagulant dosage of  $\text{FeCl}_3$ .
- (4) High removal efficiency of AOC during coagulation of synthetic water would be expected if the LMW fractions can also be removed in the coagulation process.

## ACKNOWLEDGEMENTS

The support from the Natural Science Foundation of China under the scheme of the Innovation Group Fund is greatly appreciated.

## REFERENCES

- Christlan, V., Kimberly, B. & Eva, I. 2000 Impact of enhanced and optimized coagulation of removal of organic matter and its biodegradable fraction in drinking water. *Water Res.* **34**, 3247–3257.
- Easton, J. & Jago, P. 1993 The impact of water treatment on AOC removal-experiences at UK water works. In: *Proc. AWWA WQTC Conf., Miami, FL, Nov. 7–11*. AWWA, Denver, CO.
- Escobar, I. C. & Randall, A. A. 2001 Case study: ozonation and distribution system biostability. *J. AWWA* **93**(10), 77–89.
- Frederik, H., Sebastien, M., Elisabeth, S., Oliver, K., Thomas, E. & von Gunton, V. 2007 Formation of assimilable organic carbon (AOC) and specific natural organic matter (NOM) fractions during ozonation of phytoplankton. *Water Res.* **41**, 1447–1454.
- Hem, L. J. & Efraim, H. 2001 Assimilable organic carbon in molecular weight fraction of natural organic matter. *Water Res.* **35**(4), 1106–1110.
- Hu, J. Y., Wang, Z. S., Ng, W. J. & Ong, S. L. 1999 The effects of water treatment processes on the biological stability of potable water. *Water Res.* **33**, 2587–2592.
- Huck, P. M. 1990 Measurement of biodegradable organic matter and bacterial growth in drinking water. *J. AWWA* **82**, 78–86.
- Huck, P. M., Fedorack, P. M. & Anderson, W. B. 1991 Formation and removal of assimilable organic carbon during biological treatment. *J. AWWA* **83**, 69–90.
- Kaplan, L. A., Reasoner, D. J. & Rice, E. W. 1994 Survey of BOM in US drinking waters. *J. AWWA* **86**, 121–133.
- LeChevallier, M. W., Shaw, N. E., Kaplan, L. A. & Bott, T. L. 1993 Development of a rapid AOC method for water. *Appl. Environ. Microbiol.* **59**, 1526–1531.

- Lefebvre, E. & Legube, B. 1990 Coagulation par le Fe (III) de substances humiques extradites d'eaux de surface: effets du pH et de la concentration en substances humiques. *Water Res.* **24**, 591–606.
- Lehtola, M. J., Miettinen, I. T., Vartiainen, T., Myllykangas, T. & Martikainen, P. J. 2001 Microbially available organic carbon, phosphorus, and microbial growth in ozonated drinking water. *Water Res.* **35**, 1635–1640.
- Li, L. Z., Yu, G. Z. & Wang, Z. S. 2003 Removal of assimilable organic carbon from surface water by water treatment processes. *Tech. Equip. Environ. Pollut. Control* **4**, 58–60.
- Liang, T., Ma, J. & Wang, S. J. 2008 Impacts of AOC by O<sub>3</sub>/TiO<sub>2</sub> catalytic oxidation in drinking water. *Chin. J. Environ. Sci.* **20**, 41–44.
- Monika, P., Koen, H. & van Keer, C. 2005 Investigation of assimilable organic carbon (AOC) in Flemish drinking water. *Water Res.* **39**, 2259–2266.
- Randtke, S. J. 1988 Organic contaminant removal by coagulation and related process combinations. *J. AWWA* **80**(5), 40–56.
- Schnitzer, M. & Khan, S. U. 1979 *Humic Substances in the Environment*. Chemical Industry Press, Beijing, China.
- Semmens, M. J. & Ayers, K. 1985 Removal of trace organics by coagulation from Mississippi water. *J. AWWA* **77**, 79–84.
- Stumm, W. & Morgan, J. J. 1987 *Water Chemistry*. Science Press, China.
- Van der Kooij, D. 1990 Assimilable organic carbon (AOC) in drinking water. In: McFeters, G. A. (ed.) *Drinking Water Microbiology*. Springer, New York, pp. 57–87.
- Van der Kooij, D. 1992 Assimilable organic carbon as indicator of bacterial regrowth. *J. AWWA* **84**(2), 57–65.
- Van der Kooij, D. & Hijnen, W. A. M. 1984 Substrate utilization by an oxalate-consuming *Spirillum* species in relation to its growth in ozonated water. *Appl. Environ. Microbiol.* **47**(3), 551–559.
- Zhang, G. & Wang, Z. 1998 Mechanism study on organics removal with the conventional water treatment process in fulvic acid contaminated surface water. *Environ. Eng. Ser.* **3**, 179–188.
- Zhang, G. C. & Wang, Z. S. 1999 A study on the removal of mutagens from water by various united water purification process. *Water Res.* **34**, 1781–1790.
- Zhou, L. H., Zhang, S. Q. & Han, F. 1999 Mechanism study of the coagulant impact on mutagenic activity in water. *Chin. J. Environ. Sci.* **20**, 41–44.

First received 1 December 2008; accepted in revised form 30 July 2009