



Differences in behaviour of three biopolymer constituents in coagulation with polyaluminium chloride: Implications for the optimisation of a coagulation–membrane filtration process

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ABSTRACT

Coagulation is often applied as a pre-treatment for membrane processes to reduce dissolved organic matter and to prevent membrane fouling. Biopolymers (BPs) have repeatedly been reported as major organic foulants, and coagulation conditions such as pH or dose have been optimised to minimise the remaining BPs. Optimisation however remains problematic because of the complex and heterogenous nature of BP. In this study, the behaviour of several BP fractions in a coagulation process was investigated by excitation–emission matrix-parallel factor analysis (EEM-PARAFAC) following liquid chromatography (LC)-fractionation. Using a series of jar tests, we found that BP removal depends on the type of source water, reflecting differences in charge neutralisation conditions in three samples of natural water despite nearly identical processes for removing humic substances. This result demonstrates the complexity of optimisation for BP coagulation. Fractionation of EEM-PARAFAC to BP by LC showed that at least three organic component groups (C1, C2 and C3) constitute BP. C1 is tryptophan-like organic matter that is often found in wastewater effluent, C2 is tyrosine-like organic matter that has a phenolic chemical structure, and C3 is a humic-like substance. C1 was removed thoroughly at acidic pH but not at neutral pH, while the removal of C2 was inefficient even with a significant change in pH or dose, indicating similar difficulties in a coagulation process. The difference in components C1 and C2 may partly explain the difference in efficiencies of removal of BP in water from different sources. Our investigation suggests that the optimisation or selection of appropriate pre-treatment processes for membrane systems should be substantially based on the composition of BPs (e.g., C1 and C2 components).

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1. Introduction

Low-pressure membrane filtration processes in water treatment, such as microfiltration (MF) and ultrafiltration (UF), often use coagulation as a pre-treatment to remove dissolved natural organic

matter (DOM) and to prevent membrane fouling (Huang et al., 2009; Matilainen et al., 2010). Many studies on membrane fouling have found that hydrophilic biopolymers (BPs) such as polysaccharides and proteins, as well as organic colloids that are hypothesised to be peptidoglycans enriched in amino sugars, are major organic foulants (Amy, 2008; Kimura et al., 2014; Tian et al., 2013). Because BPs are the most important foulants, coagulation conditions for best control of coagulation–membrane performance have been proposed at which most of the BPs can be removed.

However, conventional optimal coagulation conditions based on the removal of turbidity or colour may not be identical to optimal

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conditions for BP removal. A number of efforts to determine the optimal coagulation conditions for BP removal have been reported. Most of these studies focused on selecting optimal coagulant dose at near-neutral pH. Kimura and Ando (2016) concluded that the optimal coagulant dose (mg Al/L) is 20 times the BP concentration (mg/L) at pH 7, which indicates that the necessary coagulant dose varies with the BP concentration in the source water. In contrast, Wray and Andrews (2014) showed that a low polyaluminium chloride (PAC) dose (0.05 mg Al/L) is enough to achieve maximum BP removal at pH 7 for water from three kinds of sources, which contained 0.22, 0.38 and 0.45 mg BP/L; however, the optimal coagulant dose for each water sample was different. These studies, which were conducted under limited coagulation conditions, addressed the behaviour of BPs under changeable coagulation conditions, especially pH. There are as yet no studies investigating the dependence of BP coagulation characteristics on DOM characteristics in source water.

A BP is defined as a colloidal fraction with a molecular weight (MW) larger than approximately 100 kDa. It may be quantified by size-exclusion methods such as size-exclusion high-performance liquid chromatography (HPLC-SEC) (Huber et al., 2011) and flow field-flow fractionation (Baalousha et al., 2011). A larger nitrogen/carbon (N/C) ratio in BP may indicate more protein-like substances in comparison with polysaccharide-like substances. Three-dimensional fluorescence excitation–emission matrix (EEM) spectroscopy, an effective and highly sensitive method for detecting fluorescent substances, has increasingly been used in the analyses of protein-like and humic-like substances in aquatic DOM in recent years. The use of EEM as a detector for liquid chromatography (LC) has revealed that protein-like substances exist in the BP fraction (Her et al., 2003). However, identifying the behaviour of protein-like substances under different coagulation conditions using only EEM is difficult because the spectral ranges of different substances overlap. The recent approach of combining a multi-way statistical method (parallel factor analysis; PARAFAC) with EEM allows an individual EEM fluorescent spectrum to be decomposed into several fraction components (Ishii and Boyer, 2012). This combination has led to new methodological avenues in the investigation of DOM quality and quantity, and has been applied to a rapidly growing number of studies on drinking water and wastewater treatments (Markechova et al., 2013; Yang et al., 2015). By comparing the spectral characteristics with traditional fluorescence peaks, the fluorescent DOM constituents in natural water can be broadly decomposed into protein-like and humic-like components, both of which can be further separated (Yang et al., 2015).

Although BPs often consist of multiple components that have complex and heterogeneous properties, most studies on the treatability of BP in water treatment process have treated their behaviour as a single uniform organic component. As such multi-component characteristics of BP result in inconsistencies in the treatability of BP, no universal guidelines for the optimisation of BP elimination in coagulation processes exist.

In this study, we therefore investigated the behaviour of individual BP components in the coagulation process by fractionating and characterising BP samples through combined HPLC-SEC and EEM-PARAFAC analyses. BPs present in natural water were categorised into several components, and their coagulation properties such as optimal pH and dosage were examined individually.

As noted above, the controlled elimination of BP in the coagulation process remains challenging due to a lack of knowledge of BP components and their behaviour in coagulation processes. The results obtained in this study may finally aid the development of a novel efficient coagulant for BP removal and may provide insights into the optimisation of coagulation conditions in coagulation–membrane filtration processes.

2. Materials and methods

2.1. Raw water samples

Three types of surface river water in Japan were used as raw water samples (Fig. 1). Water from Kikuchi River (K-RW) was taken upstream of several large cities in the vicinity and very clean as drinking water source. Water from Edo River (E-RW), a river flowing past Tokyo, contained many more impurities. Yodo River water (Y-RW) was highly contaminated and had the worst quality among the three samples, as the river traverses several metropolitan areas and for approximately the river water is contaminated by wastewater (Yamamura et al., 2014).

Before analysis, particles larger than 10 μm were removed using a 10 μm stainless-steel cartridge filter. The characteristics of the three types of water are listed in Table 1. The total organic carbon (TOC) in K-RW was approximately half that in Y-RW (Table 1), indicating less organic matter. K-RW also had the lowest specific ultraviolet absorbance (SUVA), an indicator of hydrophobicity of organic matter (Table 1), suggesting that K-RW was rich in hydrophilic organic matter. The BP concentration for each water sample filtered through 0.45 μm membrane filters was determined by the method described in section 2.3 (Analytical methods). K-RW had the lowest BP concentration and Y-RW the highest, implying high and low water quality, respectively (Table 1, Fig. 2). This is consistent with the finding that water contaminated by wastewater always contains large amounts of BPs (Tian et al., 2013). However, the difference in BP concentration between K-RW and E-RW was insubstantial. The EEM contours of each raw water sample filtered through 0.45 μm membrane filters normalised against their TOC data are shown in Fig. 3, and the original EEM contours of each water sample can be found in Fig. S1.

2.2. Jar tests

Jar tests were performed at room temperature ($23 \pm 3^\circ\text{C}$) using six replicates of standard 1 L beakers and a six-paddle jar test apparatus. Commercial PAC (10% as Al_2O_3 , basicity = 50–55%; Taiki

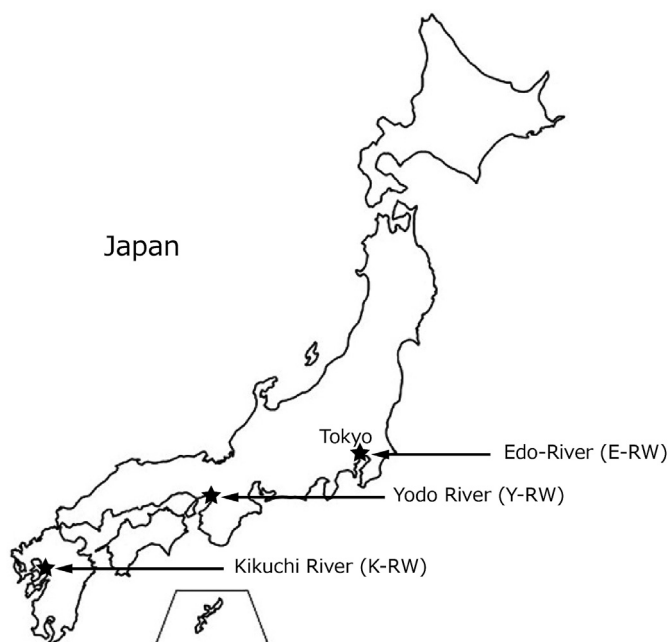


Fig. 1. Sampling sites for surface river water in Japan.

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