



# Influence of chlorination on metal binding by dissolved organic matter: A study using Log-transformed differential spectra



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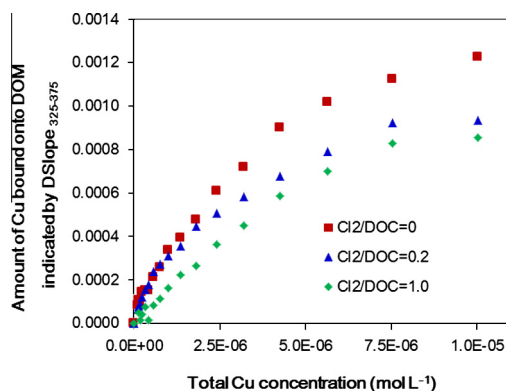
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## HIGHLIGHTS

- Little change of Fe(III) binding were affected by chlorine.
- The bound Al(III) decreased at high total Al(III) concentrations after chlorination.
- The increase of carboxylic group after chlorination lead to increase the bound Cu.
- DSlope<sub>325–375</sub> correlate strongly with concentration of SRFA-bound Fe, Al and Cu.

## GRAPHICAL ABSTRACT



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## ABSTRACT

This study employed the method of Log-transformed absorbance spectroscopy to probe interactions of Suwannee River fulvic acid (SRFA) and Aldrich humic acid (AHA) affected by chlorination with Fe(III), Al(III), and Cu(II) ions. Cl<sub>2</sub>/DOC ratios of 0.2 and 1.0 (mg/mg) were selected to represent chlorine doses that are commonly used in pre-chlorination and disinfection in water treatment processes. Spectroscopic data were interpreted based on non-ideal competitive adsorption coupled with a Donnan electrostatic sub-model (NICA–Donnan model) that was employed to quantify the constants describing relevant complexation equilibria. The data demonstrated that effects of chlorination on dissolved organic matter (DOM) and metal interactions depend on the metal, chlorine dose and intrinsic DOM properties. Fe(III) was observed to form strong complexes with DOM and relatively little change of Fe(III) binding were observed both SRFA and AHA affected by chlorine. The amount of bound Al(III) decreased significantly at high total Al(III) concentrations (>5 μM) due to the reducing of phenolic groups after chlorination. The amount of bound Al(III) decreased only slightly for chlorinated SRFA but for AHA Al(III) binding increased at low total Al concentrations (<1 μM) due to the generation of additional carboxylic groups after chlorination. It is different from Fe(III) and Al(III), the dominant involvement of Cu–DOM binding is carboxylic groups in both fulvic and humic acid, especially for humic acid, the increase of carboxylic group after chlorination for SRFA at low Cl<sub>2</sub> dose and for AHA at both low and high dose lead to increase the total amount of bound Cu(II).

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## 1. Introduction

Reactions between dissolved organic matter (DOM) and chlorine are extremely important for drinking water and wastewater treatment processes. Due to the oxidation of and chlorine

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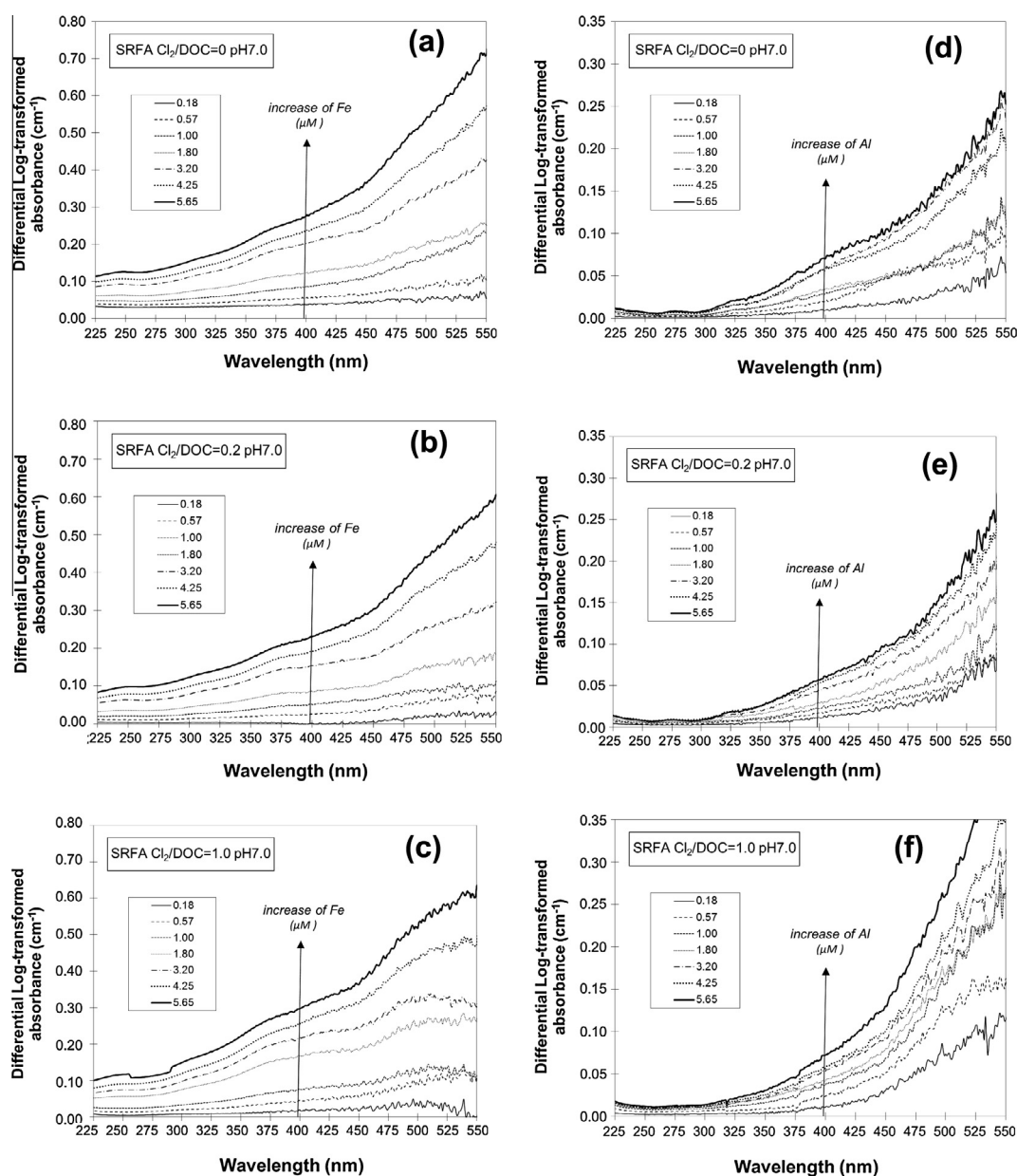
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incorporation in primarily aromatic halogen attack sites in DOM, these reactions also affect the behavior of DOM in its interactions with other system components, for instance with Fe and Al based coagulants and with other metal cations typical for water treatment and distribution systems (Aeschbacher et al., 2009, 2012; Lin and Valentine, 2009; Liu et al., 2009; Liu and Qi, 2010; Maurer et al., 2010; Macalady and Walton-Day, 2011).

Due to their diverse functional groups and site-specificity, the DOMs behave as heterogeneous ligands (Ritchie and Perdue, 2003; Leenheer, 2009), and this chemical heterogeneity will be reflected in their ion binding properties. The polyelectrolytic behavior of DOM will also affect ion binding due to electrostatic interactions (de Wit et al., 1993; Milne et al., 1995a; Cabaniss, 2011). The complexity of such a system is great and has led to the development of various models to be able to describe ion binding at different pH's and ionic strengths and over a wide range of concentrations for the different ions (Benedetti et al., 1995; Dudal

and Gérard, 2004; van Riemsdijk et al., 2006). The non-ideal competitive adsorption coupled with a Donnan electrostatic sub-model (NICA–Donnan model) is a well-attested metal complexation model in which most of the metal ions bound to NOM are bound to operationally defined carboxylic and phenolic functional groups and form non-discrete Donnan gel electrostatic interactions. To account for this non-uniformity, the dimensionless parameters  $m_{\text{carbox}}$  and  $m_{\text{phenolic}}$  that define the width of the distributions of  $pK$  values assigned to the prevalent carboxylic and phenolic groups, respectively, were introduced (Kinniburgh et al., 1996; Milne et al., 2001).

DOM–metal interactions have been typically examined with the methods of potentiometry that employs applicable ion selective electrodes (ISE) and voltammetric measurements. However, these methods are not inherently structure-sensitive, cannot be used for many cations and typically require unrealistically high concentrations of the system components (Kinniburgh et al.,



**Fig. 1.** Differential Log-transformed absorbance spectra of SRFA before and after chlorination recorded at varying concentrations of iron (a–c), aluminum (d–f) and copper (g–i) at pH7.0. DOC concentration  $5.0 \text{ mg L}^{-1}$ . Metal ions concentrations shown in the legend are in  $\mu\text{M}$ .

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