



Effective removal of trace thallium from surface water by nanosized manganese dioxide enhanced quartz sand filtration



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HIGHLIGHTS

- Enhanced quartz filtration could effectively remove trace Tl from surface using nMnO₂.
- Competitive Ca²⁺ reduced the enhancement efficiency due to competitive effects.
- Complex HA also hindered the enhancement efficiency of trace Tl filtration.
- A slight increase of Tl residual could be observed after backwashing.

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ABSTRACT

Thallium (Tl) has drawn wide concern due to its high toxicity even at extremely low concentrations, as well as its tendency for significant accumulation in the human body and other organisms. The need to develop effective strategies for trace Tl removal from drinking water is urgent. In this study, the removal of trace Tl (0.5 μg L⁻¹) by conventional quartz sand filtration enhanced by nanosized manganese dioxide (nMnO₂) has been investigated using typical surface water obtained from northeast China. The results indicate that nMnO₂ enhanced quartz sand filtration could remove trace Tl(I) and Tl(III) efficiently through the adsorption of Tl onto nMnO₂ added to a water matrix and onto nMnO₂ attached on quartz sand surfaces. Tl(III)-HA complexes might be responsible for higher residual Tl(III) in the effluent compared to residual Tl(I). Competitive Ca²⁺ cations inhibit Tl removal to a certain extent because the Ca²⁺ ions will occupy the Tl adsorption site on nMnO₂. Moreover, high concentrations of HA (10 mgTOC L⁻¹), which notably complexes with and dissolves nMnO₂ (more than 78%), resulted in higher residual Tl(I) and Tl(III). Tl(III)-HA complexes might also enhance Tl(III) penetration to a certain extent. Additionally, a higher pH level could enhance the removal of trace Tl from surface water. Finally, a slight increase of residual Tl was observed after backwash, followed by the reduction of the Tl concentration in the effluent to a “steady” state again. The knowledge obtained here may provide a potential strategy for drinking water treatment plants threatened by trace Tl.

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1. Introduction

Thallium (Tl) is one of the most toxic metals to humans and other aquatic organisms (Puddu et al., 1988; Hassler et al., 2007;

Tatsi et al., 2015). These studies reported that Tl is much more toxic than many metals, such as mercury, cadmium, and lead (Zitko, 1975; Repetto et al., 1998; Kazantzis, 2000; Lennartson, 2015; Campanella et al., 2016). Moreover, significant amounts of Tl can accumulate in the human body. Thus, Tl is commonly added to the list of priority metallic pollutants to be treated in drinking water sources (Zitko, 1975; Campanella et al., 2016; Pavoni et al., 2017).

In aquatic environments, Tl occurs in two common oxidation states, Tl(I) and Tl(III). Tl(I) resembles alkali metals and is highly

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soluble in aqueous solutions. Tl(III) is more stable because it forms complexes with other constituents in water, such as sulfates, chlorides, carbonates, nitrates, and humic acid (HA) (Kaplan and Mattigod, 1998; Lin and Nriagu, 1998b, 1999; Lis et al., 2003; Twining et al., 2003; Zhou and Cooke, 2006; Turner et al., 2010; Xiao et al., 2010; Nielsen et al., 2013). Water can become contaminated with Tl from different sources, including mining operations and some alloy production manufacturing processes (Ewers, 1988; Zhang et al., 1998; Lis et al., 2003; Sangvanich et al., 2010; Vanek et al., 2010, 2013; Xiao et al., 2012; He et al., 2015). Thus, either intentional or accidental discharge of wastewater from such sources might lead to surface water contamination by trace Tl. Water contaminated by trace Tl may threaten public health and the global environment. Thus, the drinking water criteria for Tl established by the USEPA and the Chinese Government are $0.2 \mu\text{g L}^{-1}$ and $0.1 \mu\text{g L}^{-1}$, respectively (Memon et al., 2008).

Until now, different treatment technologies have been utilized for Tl removal from water, including chemical precipitation, ion exchange, adsorption, and biological methods (Kikuchi et al., 1990; Naghizadeh and Afzali, 2007; Memon et al., 2008; Peter and Viraraghavan, 2008; Zolgharnein et al., 2011; Khavidaki et al., 2013), which rely on the mechanisms of exchange, complexation, and precipitation (Vink, 1998; Memon et al., 2008; Saljooghi and Fatemi, 2011; Zolgharnein et al., 2011; Khavidaki et al., 2013). However, there is only a small amount of data related to the removal of Tl in engineering applications. Although some researchers have indicated that adsorption is an effective way to remove Tl from water, few data could be found for the industrial application of adsorption. As such, experimentation is critical to develop appropriate treatment strategies for the removal of trace Tl from drinking water (Memon et al., 2008; Senol and Ulusoy, 2010; Pu et al., 2013).

Water treatment technologies that have simple structures are most desired because they typically are lower in cost and are more convenient (Sung and Turian, 1994; Ridal et al., 2001; Tang et al., 2009; McTaggart et al., 2015; Pan et al., 2017). Sand filtration is a widely adopted method in drinking water and wastewater treatment plants, among other methods. Enhanced filtration is an optimal way to improve the removal of various pollutants, including As, geosmin, Cr, tastes, odors, phosphates, etc. (Sung and Turian, 1994; Ridal et al., 2001; Newcombe and Cook, 2002; Wickramasinghe et al., 2004; Sun et al., 2009; Erickson et al., 2012; Wu et al., 2012; Liang and Maceiko, 2015; Choi et al., 2016). Generally, many methods have been proposed to accomplish enhanced filtration, including reduction, oxidation, and adsorption (Wickramasinghe et al., 2004; Sun et al., 2009; Erickson et al., 2012; Wu et al., 2012; Liang and Maceiko, 2015). Our previous research on the adsorption of Tl by nanosized manganese dioxide (nMnO_2) indicated that the method is highly efficient under neutral pH conditions, even when there is a high concentration of common constituents such as monovalent, divalent, or HA (Huangfu et al., 2015c). Thus, enhanced filtration that employs this novel nanosized particle, nMnO_2 , may offer an effective and economical option for water resources that are threatened by trace Tl.

Data obtained from previous work that evaluated the effectiveness of *in situ* MnO_2 indicate that the reducing products of MnO_2 that occur from permanganate oxidation can enhance flocculation and improve filtration for both high and low turbidity water (Zhang et al., 2007, 2009). Based on analyses that used an atomic force microscope (AFM), the authors previously proposed that the higher adsorption capacity for arsenic than for aged manganese dioxide mainly resulted from the smaller particle size of *in situ* formed MnO_2 (Zhang et al., 2008). These studies indicated that an *in situ* formed MnO_2 enhanced filtration process can effectively remove micro-pollutants, but they did not provide any

insight for the efficient removal of trace Tl.

Our recent work reported the enhancement efficiency of nMnO_2 for conventional coagulation, and our results revealed that nMnO_2 could significantly enhance the removal of trace Tl from representative surface waters (Huangfu et al., 2017). In this study, the removal of the trace element Tl (i.e., Tl(I) and Tl(III)) from representative surface water by conventional filtration processes enhanced with nMnO_2 was evaluated in a lab-scale sand filtration system. The effects of Tl valance, nMnO_2 dosage, Ca^{2+} concentration, HA concentration, pH, and backwash on Tl removal are considered. The residual Mn concentration of the effluent was measured, and an analysis of data obtained from a scanning electron microscopy-energy dispersive spectrometer (SEM-EDS) was performed to understand the mechanisms of trace Tl removal by nMnO_2 enhanced filtration. The results obtained here may be useful for the engineering application of nMnO_2 and related technologies in the reduction of trace Tl from surface water based on quartz sand filtration in related drinking water plants.

2. Materials and methods

2.1. Chemicals

Chemicals including TlNO_3 (99%), $\text{Tl}(\text{NO}_3)_3$ (99%), HA, and acetic acid 3-(N-morpholino)-propanesulfonic acid (MOPS, $\text{pK}_a = 7.2$) were obtained from Sigma-Aldrich Company. KMnO_4 , $\text{Na}_2\text{S}_2\text{O}_3$, NaOH, HNO_3 , NaNO_3 , and $\text{Ca}(\text{NO}_3)_2$ were obtained from Sinopharm Chemical Reagent Co., Ltd. All of the reagents except for HA were used as received from the manufacturer. All stock solutions used in the present research were prepared with double distilled water ($>18.2 \text{ M}\Omega \text{ cm}$).

HA stock preparation followed the procedure described by our previous study (Huangfu et al., 2017). Briefly, the HA stock was purified by means of repeated centrifugation (4000 r min^{-1}), pH adjustment and precipitation to remove ash, humin, and fats. A separatory funnel was then used to separate HA from fulvic acid by precipitating the solution for 48 h at a pH of approximately 3. The concentration of HA stock was characterized by its total organic carbon (TOC) content and was determined by employing a high-temperature oxidation method (Model Multi3100, Jana, Germany) after dilution (the measured sample values were $5\text{--}20 \text{ mgTOC L}^{-1}$). The stocks were stored in the dark at 4°C and were used within a month.

2.2. Surface water

Our preliminary work that employed adsorption experiments indicated that water conditions, such as competitive cations, HA, and other impurities in water, played an important role in Tl(I) adsorption and oxidation (Huangfu et al., 2015c). To confirm whether nMnO_2 could be an effective means to remediate natural source water, a river water sample (collected from Northeast China) was selected as a representative surface water sample. Before filtration, the river water sample was treated with 4 mg L^{-1} polymeric aluminum chloride (PAC), and the supernatant was collected for the filtration experiments. The important parameters of the supernatant are shown in Table 1. The TOC concentration was measured using a high-temperature oxidation method (Model Multi3100, Jana, Germany). Cl^- concentration was determined by ion chromatography (ICS-3000, DIONEX, USA). Turbidity was measured using Hach YSI 6136 turbidimeters. Other water quality parameters were measured using standard methods. Alkalinity was determined according to Standard Method 2320 B. The water pH was measured using an electronic pH meter (Cole Parmer pH 100 series) calibrated daily using pH 4, 7, and 10 buffers. Water hardness

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