



Manganese deposition in drinking water distribution systems



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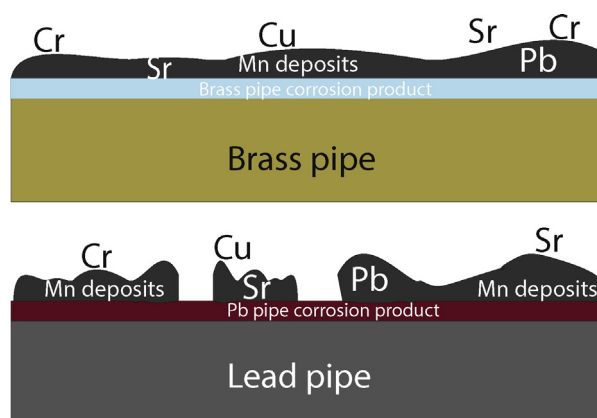
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HIGHLIGHTS

- Oxidation and deposition of Mn deposits in drinking water distribution pipes
- In-situ synchrotron-based μ -XANES and μ -XRF mapping
- Toxic metal sorption in Mn deposits

GRAPHICAL ABSTRACT



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ABSTRACT

This study provides a physicochemical assessment of manganese deposits on brass and lead components from two fully operational drinking water distributions systems. One of the systems was maintained with chlorine; the other, with secondary chloramine disinfection. Synchrotron-based in-situ micro X-ray adsorption near edge structure was used to assess the mineralogy. In-situ micro X-ray fluorescence mapping was used to demonstrate the spatial relationships between manganese and potentially toxic adsorbed metal ions. The Mn deposits ranged in thickness from 0.01 to 400 μm . They were composed primarily of Mn oxides/oxhydroxides, birnessite (Mn^{3+} and Mn^{4+}) and hollandite (Mn^{2+} and Mn^{4+}), and a Mn silicate, braunite (Mn^{2+} and Mn^{4+}), in varying proportions. Iron, chromium, and strontium, in addition to the alloying elements lead and copper, were co-located within manganese deposits. With the exception of iron, all are related to specific health issues and are of concern to the U.S. Environmental Protection Agency (U.S. EPA). The specific properties of Mn deposits, i.e., adsorption of metals ions, oxidation of metal ions and resuspension are discussed with respect to their influence on drinking water quality.

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1. Introduction

Manganese (Mn), ubiquitous in the environment, comprises about 0.1% of the earth's crust (WHO, 1999). Divalent manganese (Mn^{2+}) can be solubilized from soils and bedrock (Ljung and Vahter, 2007) into surface and ground waters, sources of drinking water. Manganese concentrations from natural processes are typically low, but can range up to 1.5 mg L^{-1} or higher. Manganese can also be introduced into surface and ground waters from anthropogenic activities. Heal (2001) investigated the relationship between land use and Mn concentrations in surface waters in Scotland. She reported that conifer afforestation, liming, fertilization addition, drainage ditch construction and plowing increased Mn concentrations in runoff (Heal, 2001). In addition flooding and waters associated with mining (surface and deep; NAWQA, 2000) can contribute to dramatic increases in Mn concentrations in ground water sources used for drinking water.

Soluble Mn^{2+} in drinking water can be oxidized to insoluble Mn^{3+} and Mn^{4+} (Cerrato et al., 2006; Kohl and Mediar, 2006; Manceau and Charlet, 1992; Negra et al., 2005) by oxidizing disinfectants and by metal oxidizing bacteria. The result is precipitation, and in the case of drinking water distribution systems (DWDS), Mn deposition on internal pipe surfaces or on established corrosion products. Mn oxides/oxyhydroxides are highly effective at adsorbing metal ions such as chromium (Cr), copper (Cu), iron (Fe), lead (Pb), and strontium (Sr) ions (Courtin-Nomade et al., 2009; Manceau and Charlet, 1992; Manceau et al., 1992; Negra et al., 2005; Nelson et al., 1999; Takahashi et al.,

2007; Tanaka et al., 2010; Villalobos et al., 2005). In addition Mn oxides can oxidize adsorbed metal ions. For example Mn oxides can oxidize Cr^{3+} to Cr^{6+} resulting in the metal ion being converted from a non-toxic to toxic state (Courtin-Nomade, 2009; Manceau and Charlet, 1992; Manceau et al., 1992).

Mn deposits can be readily resuspended (Cerrato et al., 2006; Schlenker et al., 2008; Sly et al., 1990) in DWDS by numerous factors including changes in water disinfection chemistry, especially transitioning from chlorine to chloramine (Torrey, 2013). Manganese in tap water at concentrations greater than 0.05 milligrams per liter of water (mg L^{-1}) can be detected as changes in color, odor, or taste to the water. Because of this Mn is regulated by the U.S. Environmental Protection Agency (U.S. EPA; U.S. EPA, 2004) for esthetic issues (e.g., black water).

Health effects from Mn ingestion are not a concern until concentrations are approximately 10 times 0.05 mg L^{-1} . Ingestion of elevated Mn concentrations from drinking water may cause neurological disorders (Alvarez-Bastida et al., 2013; Bourchard et al., 2011; Chai et al., 2010; Ljung and Vahter, 2007; Schlenker et al., 2008, U.S. EPA, 2004, U.S. EPA, 2012). An additional health concern, that has not been addressed by regulators, is the potential impact of adsorbed toxic metal ions released at elevated concentrations when Mn deposits disassociate. Therefore the action level (AL) established by several states for Mn concentrations is 0.5 mg L^{-1} to ensure protection against manganese toxicity and esthetic issues. This AL is consistent with the World Health Organization guidance level for manganese in drinking water (WHO, 2004). Individual states may abide by federal regulations or adopt more stringent standards (Torrey, 2013).

The purpose of this paper is to provide the first known physicochemical characterization of Mn deposits from two fully operational DWDS, one using chlorine; the other, secondary chloramine disinfection. A combination of traditional physicochemical characterization techniques and synchrotron-based in-situ micro X-ray adsorption near edge structure (μ -XANES) spectroscopy and X-ray fluorescence mapping (μ -XRF) were used to assess potentially toxic metal ions associated with the deposits. In this study, Mn deposition was evaluated on lead and brass surfaces to avoid any contamination from a corroding iron substratum.

Table 1
Selected water quality parameters of finished drinking waters for the two drinking water distribution systems that samples for this study were obtained.

Parameter	Utility	
	A	B
pH	7.26 to 7.69	8.6
Hardness (mg L^{-1} as CaCO_3)	91 to 196	91 to 177
alkalinity (mg L^{-1} as CaCO_3)	37 to 111	68
Ca (average)	NR	36
Chlorine – free (mg L^{-1})	NA	0.97
Chlorine – total (mg L^{-1})	0.0 to 4.3	NR
Orthophosphate ($\text{mg-PO}_4 \text{ L}^{-1}$)	2.1 to 3.0	NA
Phosphate ($\text{mg-PO}_4 \text{ L}^{-1}$)	NA	0.083
Fe (mg L^{-1})	ND	0.05
Mn (mg L^{-1})	0.0005	<0.002

NA – not applicable; NR – not reported; ND – not detected.

2. Materials and methods

2.1. Sampling

2.1.1. Water chemistry

Surface waters are used as source waters by both utilities. Utility A (UA) uses secondary chloramine and Utility B (UB), free chlorine. UA

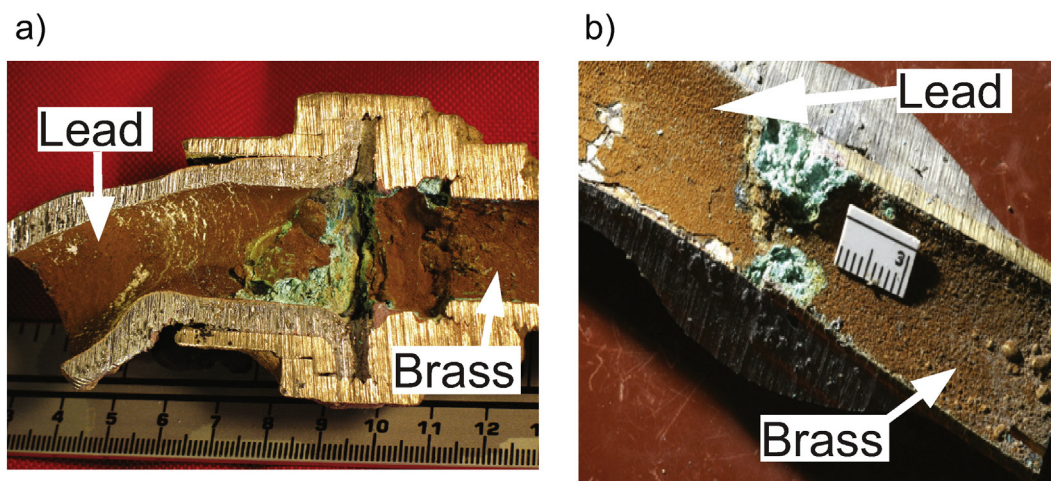


Fig. 1. Images of lead service line with brass connector from a) Utility A and b) Utility B. The scale in both images is in cm.

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