



Effectiveness of hypolimnetic oxygenation for preventing accumulation of Fe and Mn in a drinking water reservoir



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ABSTRACT

The accumulation of Fe and Mn in seasonally stratified drinking water reservoirs adversely impacts water quality. To control issues with Fe and Mn at the source, some drinking water utilities have deployed hypolimnetic oxygenation systems to create well-oxygenated conditions in the water column that are favorable for the oxidation, and thus removal, of Fe and Mn. However, in addition to being controlled by dissolved oxygen (DO), Fe and Mn concentrations are also influenced by pH and metal-oxidizing microorganisms. We studied the response of Fe and Mn concentrations to hypolimnetic oxygenation in a shallow drinking water reservoir in Vinton, Virginia, USA by sequentially activating and deactivating an oxygenation system over two summers. We found that maintaining well-oxygenated conditions effectively prevented the accumulation of soluble Fe in the hypolimnion. However, while the rate of Mn oxidation increased under well-oxygenated conditions, soluble Mn still accumulated in the slightly acidic to neutral (pH 5.6 to 7.5) hypolimnion. In parallel, we conducted laboratory incubation experiments, which showed that the presence of Mn-oxidizing microorganisms increased the rate of Mn oxidation in comparison with rates under oxic, abiotic conditions. Combined, our field and laboratory results demonstrate that increasing DO concentrations in the water column is important for stimulating the oxidation of Fe and Mn, but that the successful management of Mn is also tied to the activity of Mn-oxidizing organisms in the water column and favorable (neutral to alkaline) pH.

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1. Introduction

Controlling iron (Fe) and manganese (Mn) in drinking water is a complex challenge for water utilities because of the adverse effects of these metals on drinking water quality and the difficulties in preventing their accumulation in source reservoirs. Issues with water staining, odor, and taste can be attributed to elevated Fe and Mn concentrations (Sommerfeld, 1999; World Health Organization, 2004), and adverse human health effects have also been associated with chronic exposure from drinking water (Wasserman et al., 2006). In response to the aesthetic water quality effects, the World Health Organization (WHO) has established guidelines for Fe and Mn concentrations in drinking water at 0.3 and 0.1 mg/L, respectively (World Health Organization, 2004). In the United States, the United States Environmental Protection Agency (EPA)

has established secondary maximum contaminant limits for Fe and Mn concentrations in drinking water at 0.3 and 0.05 mg/L, respectively (United States Environmental Protection Agency, 2016). Treatment of Fe and Mn is most commonly accomplished by oxidation and filtration techniques (Kohl and Medlar, 2006; Sommerfeld, 1999); however, the accumulation of these metals in raw water increases the cost and difficulty of the water treatment process.

The development of reducing conditions in lake and reservoir sediments during thermal stratification leads to the reductive dissolution of Fe and Mn from the sediments and subsequent diffusion of these metals into the water column (Davison, 1993; Hem, 1972). Under oxidizing conditions (dissolved oxygen (DO) concentrations >2 mg/L), Fe and Mn occur naturally as oxidized, insoluble forms (Fe³⁺, Mn³⁺, and Mn⁴⁺) in watershed sediments (Jones and Bowser, 1978). However, anoxic conditions (DO concentrations < 0.5 mg/L) commonly develop within the sediments of lakes and reservoirs during thermal stratification that are

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favorable for the reduction of Fe and Mn to soluble forms (Fe^{2+} and Mn^{2+} ; Davison, 1993; Lerman et al., 1995). The reduction of Fe and Mn is known to occur abiotically via redox reactions with sulfides, organic acids, and Fe^{2+} (Mn reduction only); however, microbial activity also plays an important role in metal reduction through the production of sulfide, organic acids, and anaerobic respiration, which couples the mineralization of organic carbon to Fe and Mn reduction (Burdige, 1993; Lovley, 1987; Nealson and Saffarini, 1994). Reductive dissolution of Fe and Mn causes accumulation of soluble Fe and Mn in anoxic sediment pore waters, where concentration gradients drive their diffusion into the overlying water column (Davison, 1993; Hongve, 1997; Zaw and Chiswell, 1999). Transport of reduced Fe and Mn back into oxidizing zones in the sediments or water column can lead to these metals becoming oxidized, often precipitating as insoluble Fe- and Mn-oxhydroxides that are subject to sedimentation out of the water column (Davison, 1993; Hem, 1972).

Hypolimnetic oxygenation is increasingly used as the first stage in treating Fe and Mn *in situ* by maintaining well-oxygenated conditions in the hypolimnion of seasonally stratified lakes and reservoirs (Beutel and Horne, 1999; Bryant et al., 2011; Burns, 1998; Debroux et al., 2012; Dent et al., 2014; Gantzer et al., 2009; Gerling et al., 2014). Hypolimnetic oxygenation (HOx) systems increase DO concentrations by injecting air, pure oxygen, or oxygenated water into the hypolimnion (Beutel and Horne, 1999; Gerling et al., 2014; Singleton and Little, 2006). Physical mixing induced by the HOx system operation often accompanies the injection of oxygen, aiding in distributing DO throughout hypolimnion (Fig. 1; Beutel and Horne, 1999; Gantzer et al., 2009; Gerling et al., 2014). Successful management Fe and Mn using hypolimnetic oxygenation depends on maintaining a sufficiently oxidizing environment to quickly precipitate oxidized metals in the water column, where they will settle back to the bottom sediments, or ideally, to cause metals to precipitate prior to being released from the sediment pore waters into the hypolimnion water column.

The rate of metal oxidation in the water column is primarily influenced by DO concentrations, pH, and microbial activity (Diem and Stumm, 1984; Hem, 1981, 1972). The abiotic oxidation of Fe is faster than Mn under similar DO and pH conditions. For example, in air-saturated water with $\text{pH} > 6$, abiotic Fe oxidation is rapid, with soluble Fe having a half-time, defined as the time required for half of the initial mass of soluble Fe to become oxidized, of < 24 h (Davison, 1993). However, under similar conditions the half-time for soluble Mn can be > 1 year (Morgan, 1967). Soluble Mn oxidation occurs much faster in oxic lake water than observed under abiotic conditions, with several

studies observing soluble Mn oxidation half-times between 1 and 30 days at pH between 6.5 and 8.4 (Chapnick et al., 1982; Diem and Stumm, 1984; Kawashima et al., 1988; Tipping et al., 1984). Chapnick et al. (1982) and Diem and Stumm (1984) identified Mn-oxidizing organisms in the reservoir water and showed that when these organisms were removed by filtration, Mn oxidation was negligible within the duration of their incubation experiments (10–30 days). The mechanisms of biotic Mn oxidation are not fully understood, but there is evidence that strains of actinobacteria, cyanobacteria, firmicutes, proteobacteria, and fungi have some ability to cause Mn oxidation and are ubiquitous in aquatic environments (Aguilar and Nealson, 1998; Richardson et al., 1988; Tebo et al., 2005), suggesting that the rates of Mn oxidation in lakes and reservoirs are strongly influenced by the activity of Mn-oxidizing organisms.

Although decreased concentrations of Fe and Mn have been observed in oxygenated reservoirs, sediments can continue to release metals, even in oxic hypolimnia (Aguilar and Nealson, 1998; Bryant et al., 2011; Gantzer et al., 2009). For example, Gantzer et al. (2009) reported a 97% decrease in Mn concentrations in the hypolimnion of Carvins Cove, Virginia, USA following the installation and continuous operation of a HOx diffuser system. However, elevated Mn concentrations were still observed in the sediment pore waters while the overlying hypolimnion was well-oxygenated, indicating that upward diffusion of Mn mass continued during HOx operation (Bryant et al., 2011; Gantzer et al., 2009). Similarly, Dent et al. (2014) reported decreases of the mean hypolimnetic Fe and Mn concentrations in North Twin Lake, Washington, USA, by 71% and 73%, respectively, within 8 h of activating the oxygenation system; however, the mean concentrations reported in that study did not account for volume difference along the profile, which makes it difficult to determine whether the decrease in hypolimnetic metal concentrations can be attributed to rapidly enhanced oxidation or whether redistribution of mass in response to HOx-induced physical mixing also influenced concentrations along the depth profile.

Managing Fe and Mn treatment in drinking water systems is challenging and expensive. Thus, accurately characterizing the processes controlling metal removal in drinking water reservoirs at the whole-ecosystem scale is needed to optimize management strategies. As discussed above, metal concentrations are affected by a host of biogeochemical and physical processes in waterbodies (Aguilar and Nealson, 1998; Bryant et al., 2011; Diem and Stumm, 1984; Gantzer et al., 2009). While previous studies have examined the impacts of specific processes of metal concentrations in laboratory incubations or examined larger scale patterns at the lake or reservoir scale (Beutel et al., 2008; Diem and Stumm, 1984; Gantzer et al., 2009; Gerling et al., 2014), it remains unknown how redox and pH-dependent processes in the water column and sediments, physical mixing across concentration gradients, and the activity of metal-oxidizing bacteria together affect metal concentrations at the reservoir scale.

The goal of this study was to investigate the effects of hypolimnetic oxygenation on Fe and Mn oxidation at the whole-ecosystem scale in a drinking water reservoir. We manipulated the HOx system to experimentally create intermittent periods of hypolimnetic anoxia and well-oxygenated conditions over two years. Throughout the two-year experiment, we measured the effects of HOx system activation on soluble and total Fe and Mn accumulation in the hypolimnion and the rate of particulate metal sedimentation. In parallel, we conducted laboratory incubation experiments to evaluate the influence of microbial activity on Mn oxidation in the reservoir. We used the field and laboratory data to evaluate the importance of biogeochemical processes in Fe and Mn oxidation in oxygenated reservoirs.

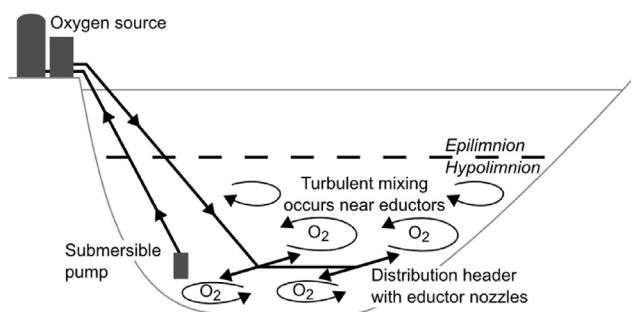


Fig. 1. Hypolimnetic oxygenation systems cause turbulence in the water column that may lead to physical mixing of the hypolimnion. This figure depicts the mixing induced by the side-stream supersaturation system installed in FCR. This figure is adapted from Gerling et al. (2014).

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