

## Research article

## Do constructed wetlands remove metals or increase metal bioavailability?

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## ABSTRACT

The H-02 wetland was constructed to treat building process water and storm runoff water from the Tritium Processing Facility on the Department of Energy's Savannah River Site (Aiken, SC). Monthly monitoring of copper (Cu) and zinc (Zn) concentrations and water quality parameters in surface waters continued from 2014 to 2016. Metal speciation was modeled at each sampling occasion. Total Cu and Zn concentrations released to the effluent stream were below the NPDES limit, and the average removal efficiency was 65.9% for Cu and 71.1% for Zn. The metal-removal processes were found out to be seasonally regulated by sulfur cycling indicated by laboratory and model results. High temperature, adequate labile organic matter, and anaerobic conditions during the warm months (February to August) favored sulfate reduction that produced sulfide minerals to significantly remove metals. However, the dominant reaction in sulfur cycling shifted to sulfide oxidation during the cool months (September to next March). High concentrations of metal-organic complexes were observed, especially colloidal complexes of metal and fulvic acid (FA), demonstrating adsorption to organic matter became the primary process for metal removal. Meanwhile, the accumulation of metal-FA complexes in the wetland system will cause negative effects to the surrounding environment as they are biologically reactive, highly bioavailable, and can be easily taken up and transferred to ecosystems by trophic exchange.

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## 1. Introduction

Wetlands influence global biogeochemical cycles due to their unique role in sequestering carbon, recharging groundwater, providing habitats for plants and animals, filtering pollutants from terrestrial runoff and atmospheric deposition, improving water quality in adjacent rivers/lakes, and regulating bioavailability of contaminants (Reddy and Delaune, 2008). Wetlands and their uplands are usually considered as sink, source, and transformers for contaminants (Reddy and Delaune, 2008).

Constructed wetlands are engineered systems that utilize natural processes associated with natural functions of vegetation, sediments, organisms, and microbial communities to remove contaminants from wastewaters (Kadlec and Wallace, 2009). During the past fifty years, cost-effective, technically feasible, and easily maintained constructed wetlands have successfully treated municipal, agricultural, and industrial effluents (Reddy and

D'Angelo, 1997; Mays and Edwards, 2001). Water-tolerant plant and saturated soil conditions are applied to mimic the optimal treatment conditions in the natural wetlands (IWA Specialit-Group, 2001). Treatment wetlands can be constructed in various hydrologic modes and the two basic categories are surface flow wetland and subsurface flow wetland (IWA Specialit-Group, 2001; Kadlec and Wallace, 2009). With the current technology, free water surface wetland, horizontal subsurface flow wetland, and vertical flow wetland are the three most widely used types that employ different flow patterns, layout, media, and plants (Kadlec and Wallace, 2009).

Constructed wetlands are designed to improve water quality by removing nutrients, metals, toxic organic compounds, and biochemical oxygen demand (Machemer and Wildeman, 1992; Reddy and D'Angelo, 1997; Mays and Edwards, 2001; Kohler et al., 2004; Hallberg and Johnson, 2005; Mainea et al., 2006). A range of physical, chemical, and biological processes are involved in regulating the fate of contaminants, including (1) settling of suspended particulate matters; (2) chemical filtration, precipitation, and adsorption through contact with water, sediment, litter, and plants; (3) breakdown and transformation of contaminants by

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micro-organisms; (4) predation and natural die-off of pathogens; and (5) uptake of contaminants and nutrients by plants and other organisms (Reddy and Delaune, 2008; Kadlec and Wallace, 2009). The engineering and design also contribute to the wetland function and duration (Reddy and D'Angelo, 1997). Meanwhile, constructed wetlands can adversely affect the associated trophic structure and surrounding environment as a result of the continuous accumulation and trophic flux of contaminants (Mays and Edwards, 2001).

Due to the complicated biogeochemical mechanisms and various wetland types, the removal processes and the treatment effectiveness of different constructed systems continues to be variable (Machemer and Wildeman, 1992; Reddy and D'Angelo, 1997; Kohler et al., 2004; Hallberg and Johnson, 2005; Mainea et al., 2006). Machemer and Wildeman (1992) studied the removal processes in a constructed wetland designed to treat mine drainage containing copper (Cu), zinc (Zn), manganese (Mn), and iron (Fe). They concluded the primary removal process of metals was organic adsorption for a young wetland and sulfide precipitation for a mature wetland (Machemer and Wildeman, 1992). Hallberg and Johnson (2005) explored the roles of microorganisms in remediating acid mine drainage in the treatment wetlands and found the dominant microbial isolate was an iron-oxidising and sulfur-oxidising moderate acidophile. Stormwater, as a variable that may influence the wetland function, was proved to be an insignificant source of contamination for a golf course wetland, which was efficient in improving quality of water from storm runoff and golf course tile drainage (Kohler et al., 2004). Mainea et al. (2006) assessed the removal efficiency of a large-scale constructed wetland designed to process industrial effluents, and compared the results to a previously constructed small-scale system. Chromium (Cr), nickel (Ni), Zn (Zn), nitrite, and nitrate were both retained and efficiently removed from water, but metals were retained by the macrophytes in the large wetland and in the sediment in the small wetland (Mainea et al., 2006).

The H-02 wetland system on the Savannah River Site (Aiken, SC, Fig. 1) was constructed to treat the process water and some storm runoff water released from the Tritium Processing Facility (Mills et al., 2011). It is a free water surface wetland, designed to

remove heavy metals, specifically Cu and Zn, from the water before it entering the Upper Three Runs Creek, a regulated stream that empties into the Savannah River. The free water surface wetland is similar to the hydrological regime of natural wetlands, that has an area of open water, floating vegetation, and emergent plants (Kadlec and Wallace, 2009). It functions as a land-intensive biological treatment system: the influent containing metals flow through a large area of shallow water, where metals and organic matter settle to the surface sediments and/or enter the biogeochemical cycles in the aquatic system (Bach et al., 2008).

This study examined Cu and Zn removal processes in the H-02 constructed wetland, as well as the biogeochemical processes and water chemistries that influence metal removal reactions. Monthly monitoring of metal concentrations and water quality parameters, including water temperature, pH, oxidation-reduction potential (ORP), alkalinity, dissolved organic carbon (DOC), and two major anions chloride and sulfate, were measured from 2014 to 2016. Metal speciation was modelled in the wetland waters and concentrations of each metal species was calculated. This study summarizes the metal removal reactions in the H-02 wetland and compares it to previous studies. The wetland function and duration, possible adverse effects to the surrounding environment, and recommendations for future studies were also discussed.

## 2. Material and method

### 2.1. Wetland description

As depicted in Fig. 1, wastewater enters the H-02 wetland system from several source pipes (PIPs) at the south end of the retention basin, providing hydrologic control of the wetland. The influent (INF) water exits the retention basin (REB) through a culvert in the northeast corner and flows to a splitter box that partitions the flow equally into two separate wetland treatment cells (WC1 and WC2). The half-acre, rectangular treatment cells consisted of a geo-synthetic liner and impermeable clay layer covered with 46–61 cm of wetland soil. The soil was amended with organic matter, fertilizer, and the gypsum (calcium sulfate) to

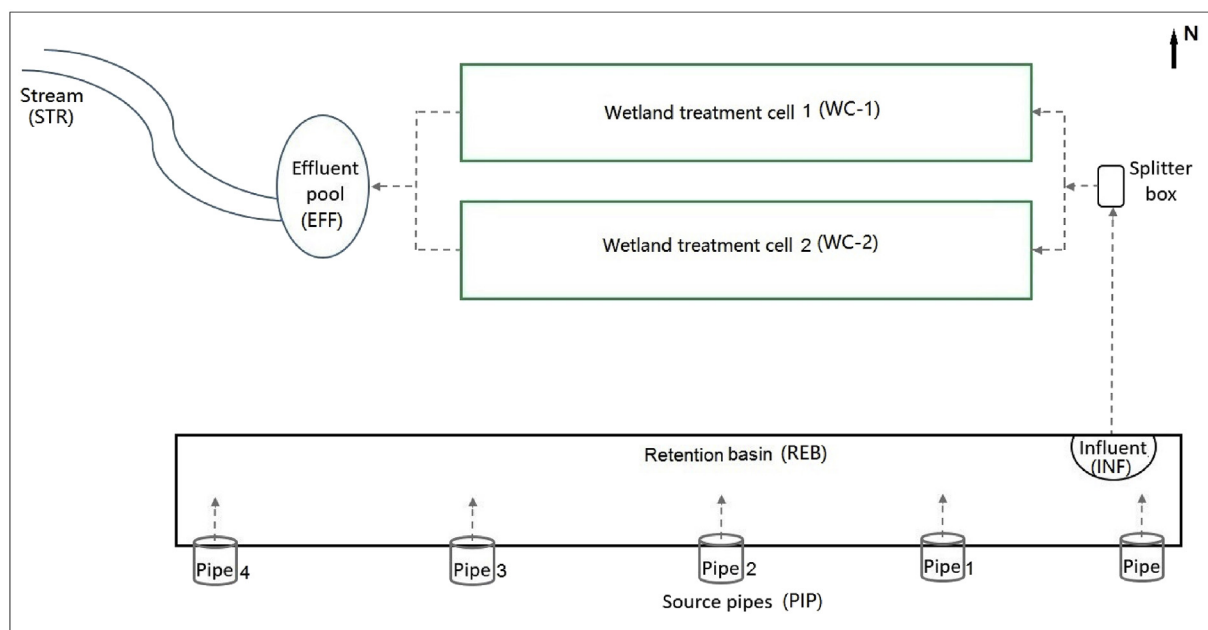


Fig. 1. H-02 wetland system with overlay of water flow. Broken lines indicate the water flow.

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