



Reactive transport modeling of early diagenesis in a reservoir lake affected by acid mine drainage: Trace metals, lake overturn, benthic fluxes and remediation

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ARTICLE INFO

Article history:

Received 18 June 2015

Received in revised form 13 October 2015

Accepted 15 October 2015

Available online 24 October 2015

Keywords:

Acid mine drainage

Reactive transport modeling

Trace metals

Benthic fluxes

Lake overturn

Organic matter sulfurization

ABSTRACT

The Sancho Reservoir in SW Spain has been impacted by acid mine drainage (AMD) since the Tharsis mine stopped activity in 1998. As a result, the reservoir exhibits low pH (~3.5) and high aqueous concentrations of sulfate, aluminum, iron and trace metals. Thus far, removal of contaminants by sediment burial has not been as effective as expected in improving water quality within the reservoir. To inform potential remediation strategies, a 1-D, non-steady-state reactive transport model with a comprehensive set of equilibrium and kinetic biogeochemical reactions is used to simulate the fate of trace metals and acidity in sediments affected by AMD. Two realizations of the model account for the spatial heterogeneity of bottom water oxygenation. A “permanently oxic” model represents shallow sediments above the thermocline, while a “holomictic” model represents the deeper sediments where bottom water oxygen levels oscillate between completely anoxic and oxic as a result of water-column overturn. The model is calibrated against an extensive dataset on the depth distributions of pore water and solid phase species. Model results imply that, under permanently oxic conditions, the sediments act as a sink for acidity (H^+) and aqueous Al, Zn, Cu, Co and Ni, but act as a source of aqueous Mn, Fe and As. The latter are released to the overlying water as a result of Mn and Fe (oxy)hydroxide reductive dissolution in the sediments. Below the thermocline, when bottom waters become anoxic, metal sulfides precipitate in the sediment. When the bottom waters subsequently become oxic, the metal sulfides are oxidized along the downward-penetrating oxygen front and the associated metals are released to the overlying water. On the order of 35% of the sediment pools of sulfide-bound Zn, Cu, Co and Ni, and ~25% of FeS are thus reoxidized. However, overall the sediments act as a net sink for the pollutants considered in the model. On an annual basis, about 10% of the total elemental masses of S, Al, Zn and Cu present in the water column of the reservoir are removed by burial in the sediments, but only ~2% for Co and Ni. For Fe, Mn and As, the corresponding values are 80, 70 and 98% respectively. The model predicts that, if AMD input to the reservoir were to completely cease, the sediments would reach a new steady state with negligible release of aqueous contaminants to the overlying water column within a few years.

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1. Introduction

Elevated concentrations of bioavailable metals in sediments may negatively impact biota and water quality (Luoma and Rainbow, 2008). The consequences can be especially detrimental in lakes and reservoirs, which are important ecosystems and key resource waters for

drinking, agriculture, industry and recreation. Therefore, the sustainable management of lakes and reservoirs stands to benefit from computational tools that simulate the fate and distribution of metals, and predict their behavior under variable conditions, for example, variable pollutant discharges, eutrophication, remediation actions, and climate change. However, existing water quality models mainly focus on the hydrodynamic functioning of lakes and reservoirs and ecological indicators in the water column, such as the chlorophyll, nitrogen or phosphorous concentrations (see Mooij et al., 2010a, 2010b for a review), but lack sophisticated representations of early diagenetic processes in the sediments (Paraska et al., 2014).

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Acid mine drainage (AMD), caused by the oxidative weathering of sulfide mine wastes, contains high concentrations of sulfate, iron, arsenic and metals and, therefore, represents a major environmental concern (Nordstrom, 2011). The effects of AMD discharges to lakes and reservoirs have been extensively documented (Herlihy and Mills, 1985; Peine and Peiffer, 1996; Nordstrom et al., 1999; Munk and Faure, 2004; Frommichen et al., 2004; Sarmiento et al., 2009; among others), with some studies focusing on the mobility and bioavailability of metals released from bottom sediments (Lopez et al., 2010; Lee et al., 2008). However, to our knowledge, little work has been carried out on the quantitative modeling of water–sediment exchanges of metals in AMD affected water bodies.

As a result of seasonal temperature changes, many lakes and reservoirs undergo alternating periods of mixing and stratification of the water column. During stratification, the hypolimnion may become anoxic due to oxygen consumption coupled to the oxidation of organic matter and other reduced chemical species (Müller et al., 2012). During mixing events, the water column becomes vertically homogenous and oxygen may then reach the bottom water. This redox oscillation causes drastic changes in the cycling of metals in bottom sediments and the associated benthic fluxes. Under anoxic conditions, Fe, As and Mn are released from the sediments, whereas many other metals are immobilized as mineral sulfide precipitates (Riedel et al., 1999; Banks et al., 2012a, 2012b). During oxic conditions, Fe, Mn and As are precipitated as oxide phases, while sulfides can be oxidized and the associated metals remobilized (Carbonaro et al., 2005). Torres et al. (2014) conducted core incubations of AMD affected sediments under oscillating bottom water oxygenation. By means of reactive transport modeling of the pore water profiles, they estimated rates of redox and dissolution/precipitation reactions, as well as exchange fluxes across the water–sediment interface (SWI).

When scaling results of laboratory experiment up to field conditions, variations in depositional fluxes and bottom water chemistry must be taken into account. This is especially true for AMD impacted lakes and reservoirs in a Mediterranean climate, where heavy storms and floods are common. During high flow events, large quantities of Fe and Mn oxides are eroded from the source mining areas and streambeds and transported as suspended load to the receiving water body (Cánovas et al., 2007, 2012). These oxides may contain variable amounts of adsorbed or co-precipitated As, Pb, Cu and Cr (Alpers et al., 1994; Smith, 1999; Casiot et al., 2003). In lakes and reservoirs, these oxides together with inert particulate material (silicates) and minerals formed in the water column (e.g., Fe(III)-oxyhydroxides) are deposited at the SWI and should be considered in models simulating water–sediment interactions.

More than 80 biogeochemical reactive transport models (RTM) for sediments have been recently reviewed by (Paraska et al., 2014). Among those some have been developed to reproduce metal cycling in lake and river sediments (Katsev et al., 2006; Massoudieh et al., 2007; Dittrich et al., 2009; Bessinger et al., 2012). Şengör et al. (2007) calibrated a 1-D reactive diffusive model to investigate local-scale processes on metal transport (Zn, Cu and Pb) through reductive dissolution of Fe(III) hydroxides in the contaminated Lake Coeur d'Alene sediment. However, relatively few RTM couple sediment transport processes (advection, bioirrigation and bioturbation) to comprehensive biogeochemical reaction networks in order to describe early diagenesis in lake, estuarine, and marine sediments. For instance, Van Cappellen and Wang (1996) presented a model that accounts for the coupling of the major redox element cycles to the degradation of organic matter. Canavan et al. (2006, 2007) expanded the previous model to include the partitioning of Ni and Zn between pore water and solid sediment in a coastal fresh-water lake, while Couture et al. (2010) developed a non-steady-state reactive transport model to account for historical changes in As deposition, cycling and accumulation in lake sediments. More recently, Arora et al. (2014) described a RTM that includes a full set of biochemical reactions together with

diffusion and sedimentation, and applied the model to Lake Coeur d'Alene sediments.

Here, we build on the work of Couture et al. (2010) and present a RTM for metal behavior in sediments affected by AMD under a seasonally variable bottom water oxygen regime. We use the model to simulate the annual cycling of major elements and trace metals in sediments of a reservoir lake receiving AMD. We compare the fate and benthic exchanges of the metals in shallow sediments that are under permanently oxic bottom waters with those in deeper sediments that experience oscillating O₂ concentrations. The model incorporates the reaction network and rates derived from previous core incubation experiments (Torres et al., 2014) from the same site, and couples the reaction rates to solid mass transport fluxes. The model is calibrated with sedimentation rates and pore water and solid sediment concentration profiles collected from different locations in the reservoir, and at different times of the year. We then assess how trace metal speciation changes under the two different bottom water redox regimes, and calculate metal fluxes across the SWI as well as net metal burial rates. We further use the model to predict the response of the benthic exchange fluxes in a remediation scenario in which AMD contaminated water is no longer supplied to the reservoir.

2. Study site

The Sancho Reservoir (58 km³) is located in the Odiel River Basin in SW Spain (Fig. S1, supplementary information); it has a surface area of 427 ha and a maximum depth of 40 m. The River Meca, the main tributary of the Sancho Reservoir, is heavily contaminated by AMD with high concentrations of trace metals, iron and sulfate, and a mean pH of 2.6 (Galván et al., 2009, 2012). The average stream flow is 61 km³ year⁻¹, most of which occurs during a few winter floods. During these floods, large amounts of particulate material are eroded from the riverbed and transported into the reservoir. The reservoir was built in 1962 to supply water for a paper mill refrigeration system. The water initially had a pH close to 5, but has become acidic and the metal content increased with time since the Tharsis mine closed in 1998 and water treatment ceased. The Sancho Reservoir currently exhibits a pH of around 3.5 (Ayora et al., 2013).

The reservoir is holomictic with an overturn in winter which generates an isothermal water column that lasts two months. During overturn, the entire water column is mixed, and dissolved oxygen reaches the sediment. During the rest of the year, the water column is stratified and anoxic conditions develop at water depths below 15 m. As a result, the reservoir can be divided in two different sections: one under permanently oxic conditions in the water column, the other experiencing stratification and oxygen concentrations fluctuating between fully oxic and anoxic (Fig. 1). This causes marked differences in the metal fluxes across the water–sediment interface (SWI) between the two sections of the reservoir. Moreover, the shallower and permanently oxic part is close to the river entrance and thus more strongly affected by flood events. Dating with ¹³⁷Cs and ²¹⁰Pb indicate net sediment accumulation rates of 1.3 and 1.6 cm year⁻¹ in the deep and shallow sections, respectively (Bolívar et al., 2012).

3. Experimental data

The experimental data used to build and calibrate the reaction network included in the early diagenetic model are described in detail in Torres et al. (2014). Briefly, three sediment cores were collected: two in the deep section of the reservoir during oxic (January 2010) and anoxic (November 2010) conditions, and one close to the river mouth (October 2010) under permanently oxic conditions (Fig. 1). To estimate the annual particulate fluxes, sediment traps were installed in the two sections of the reservoir, from September 2009 to September 2010. Water samples were also collected at the same locations at three water depths: in the epilimnion (2 m depth), metalimnion (18 m

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