Chemical Geology 269 (2010) 100–112

Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

Geochemistry and stable isotope investigation of acid mine drainage associated with abandoned coal mines in central Montana, USA

Christopher H. Gammons [a,*], Terence E. Duaime [b], Stephen R. Parker [c], Simon R. Poulson [d], Patrick Kennelly [e]

[a] Dept. of Geological Engineering, Montana Tech, Butte, MT 59701, United States
[b] Montana Bureau of Mines and Geology, Butte, MT 59701, United States
[c] Dept. of Chemistry and Geochemistry, Montana Tech, Butte, MT 59701, United States
[d] Dept. of Geological Sciences and Engineering, University of Nevada-Reno, Reno, NV 89557, United States
[e] Dept. of Earth and Environmental Science, Long Island University, Brookville, NY 11548, United States

ARTICLE INFO

Article history:
Received 14 November 2008
Received in revised 14 April 2009
Accepted 25 May 2009

Keywords:
Acid rock drainage
Geochemistry
Stable isotopes
Coal
Iron oxidation rate
Montana
Diurnal
Diel

ABSTRACT

The Great Falls-Lewistown Coal Field (GFLCF) in central Montana contains over 400 abandoned underground coal mines, many of which are discharging acidic water with serious environmental consequences. Areas of the mines that are completely submerged by groundwater have circum-neutral pH and relatively low concentrations of metals, whereas areas that are only partially flooded or freely draining have acidic pH (<3) and high concentrations of metals. The pH of the mine drains either decreases or increases after discharging to the surface, depending on the initial ratio of acidity (mainly Al and Fe^{2+}) to alkalinity (mainly HCO_3^-). In acidic, Fe-rich waters, oxidation of Fe^{2+} after exposure to air is microbially catalyzed and follows zero-order kinetics, with computed rate constants falling in the range of 0.97 to 1.25 mmol L^{-1} h^{-1}. In contrast, Fe^{2+} oxidation in near-neutral pH waters appears to be first-order with respect to Fe^{2+} concentration, although insufficient data were collected to constrain the rate law expression. Rates of Fe^{2+} oxidation in the field are dependent on temperature such that lower Fe^{2+} concentrations were measured in down-gradient waters during the day, and higher concentrations at night. Diel cycles in dissolved concentrations of Zn and other trace metals (Mn, Ni) were also noted for down-gradient waters that were net alkaline, but not in the acidic drains.

The coal seams of the GFLCF and overlying Cretaceous sandstones form a perched aquifer that lies ~50 m above the regional water table situated in the underlying Madison Limestone. The δD and $\delta^{18}O$ values of flooded mine waters suggest local derivation from meteoric water that has been partially evaporated in agricultural soils overlying the coal mines. The S and O isotopic composition of dissolved sulfate in the low pH mine drains is consistent with oxidation of biogenic pyrite in coal under aerated conditions. A clear distinction exists between the isotopic composition of sulfate in the acid mine waters and sulfate in the adjacent sedimentary aquifers, making it theoretically possible to determine if acid drainage from the coal mines has leaked into the underlying Madison aquifer.

1. Introduction

Acid mine drainage (AMD) is a well-documented environmental problem at many active and abandoned mine sites world-wide. The basic problem is simple: exposure of pyrite and other metal-sulfides to weathering under atmospheric conditions produces sulfuric acid, with subsequent mobilization of other toxic substances (metals, metalloids) into groundwater and surface water. The details of the process are complex, however, and involve a large number of gas-exchange, mineral precipitation, surface chemistry, and redox reactions, many of which are catalyzed by microbes (e.g., Nordstrom and Alpers, 1999; Nordstrom, 2003; Blowes et al., 2003; Blodau, 2006; Cravotta, 2008a,b). Although most active mines have a zero-discharge policy with respect to release of AMD to the environment, this is often not the case for abandoned mines, especially when there is no legally responsible owner. Hence, it is often the case that AMD from abandoned mines is released to down-gradient watersheds with little or no treatment.

An extensive literature exists on characterization and reclamation of AMD from coal deposits in the eastern U.S. (Growitz et al., 1985; Herlihy et al., 1990; Hedin et al., 1994; Cravotta, 2008a,b). In contrast, most studies of AMD in the western U.S. have focused on hard rock metal mining. Sites such as the Berkeley Pit copper mine in Butte, Montana, (Davis and Ashenberg, 1989; Pellicori et al., 2005), the Iron Mountain Cu–Zn mine in California (Edwards et al., 2000; Nordstrom et al., 2000) and the Summitville gold mine in Colorado (Gray et al., 1994) are highly publicized examples of AMD in the western U.S. that continue to pose significant environmental challenges. Comparatively little scientific

* Corresponding author.
E-mail address: cgammons@mtech.edu (C.H. Gammons).

0009-2541/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.chemgeo.2009.05.026

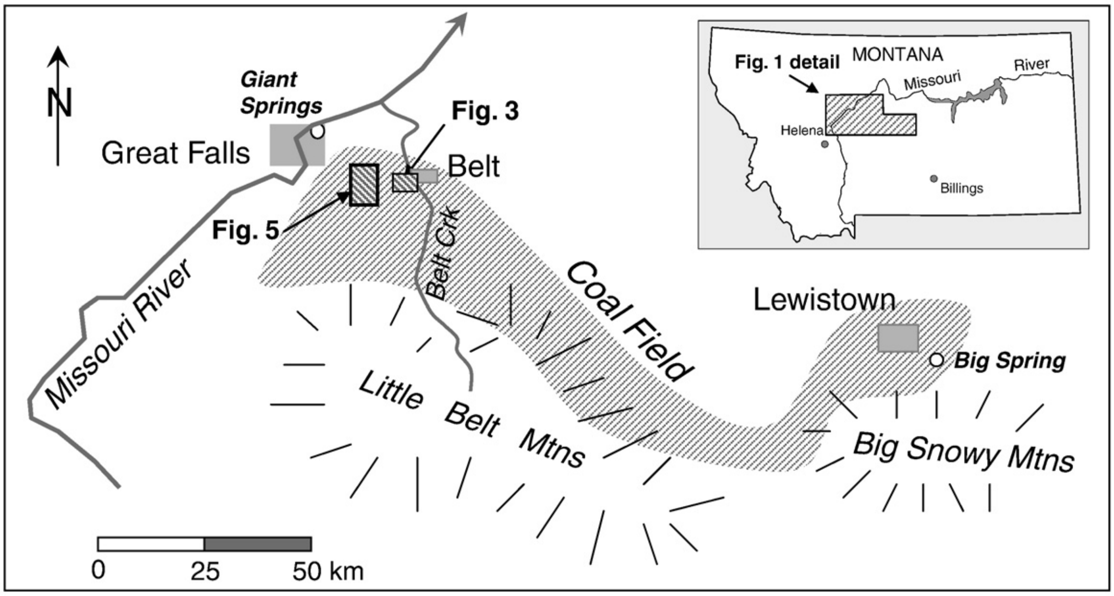

Fig. 1. Generalized location map of the study area.

study has been made on AMD associated with coal in the Rocky Mountain region, despite the fact that many western U.S. states are important producers of coal. This is largely because most of the coal fields actively being mined (e.g., the Powder River Basin of Wyoming–Montana) are low-S coals, and/or are being mined in such a way that major AMD problems have, to date, been avoided (e.g., Davis, 1984; Ferreira et al., 1989). The Great Falls-Lewiston Coal Field (GFLCF) of central Montana, U.S., is an exception to this rule. The purpose of this study is to summarize previous literature on the occurrence of AMD in the GFLCF, to examine spatial and temporal changes in the chemistry of the mine waters after they discharge to the surface and become aerated, and to use stable isotopes to help determine sources of water and dissolved sulfate in the abandoned coal mines, as well as the surrounding sedimentary aquifers.

1.1. Site geology and hydro-stratigraphy

The Great Falls-Lewistown Coal Field (Fig. 1) produced sub-bituminous to bituminous coal from large underground mines in the late 19th and early 20th centuries (Fisher, 1909). The largest mines were located near the western edge of the district near the towns of Belt and Stockett (Figs. 1 and 2), and it is in this region that the most serious environmental problems have occurred. Many of the abandoned mines are producing AMD that contaminates streams and alluvial groundwater (Osborne et al., 1983a, 1987; Karper, 1998). Reclamation of these sites has focused on removal of mine buildings and associated waste, as well as landscaping and revegetation of disturbed soils. Attempts to treat some of the acid mine drainages passively (e.g., open limestone channels, constructed wetlands) have failed due to the high acidity of the waters coupled with extreme cold temperatures in winter (McCurley and Koerth, 1994).

Fig. 3 is a schematic cross-section showing the main hydro-stratigraphic units in the study area. The coal seams of the GFLCF are located at the top of the Jurassic–Cretaceous Morrison Formation – mainly shale and siltstone – and are conformably overlain by clastic sediments of the Cretaceous Kootenai Formation (Vuke et al., 2002; Duaime et al., 2004). Erosionally-resistant sandstone units within the Kootenai Fm., including the Cutbank and Sunburst members, form the backbone of the broad, grassy uplands in this portion of the Rocky Mountain foothills. The coal seams crop out in deeply incised valleys formed by ephemeral streams that drain northward towards the Missouri River.

In the vicinity of Belt, the Jurassic Swift and lower Morrison Formations lie unconformably above the Mississippian Madison Group, a ~500 m thick marine limestone consisting of the Mission Canyon and underlying Lodgepole Formations. The Madison aquifer is an important local and regional source of potable groundwater. The Madison feeds two very large natural springs at either end of the GFLCF named Giant Springs and Big Spring (Fig. 1). Giant Springs, one of the largest fresh water springs in the U.S., discharges ~8400 L s^{-1} of

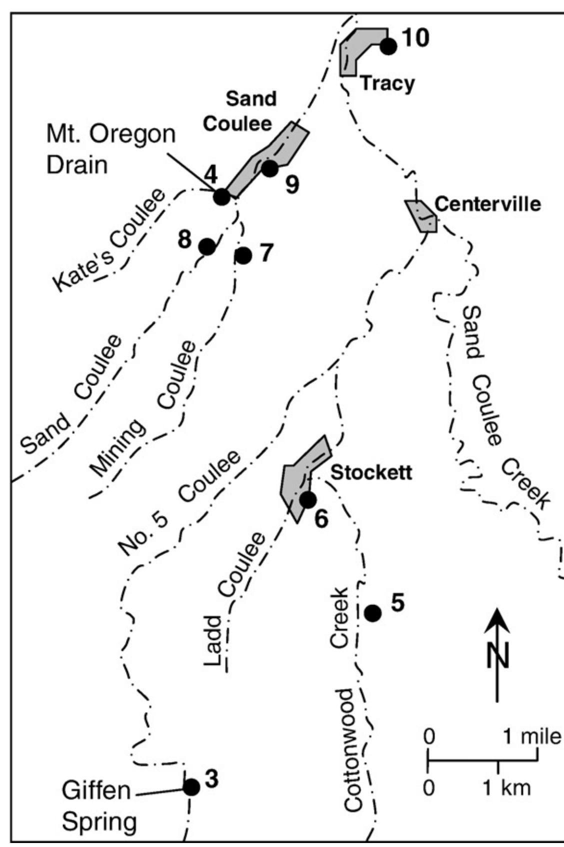

Fig. 2. Location map of the Stockett area. Solid circles show locations of mine drains discussed in this paper (see Table 1 footnotes for location names).