



Geochemical and mineralogical characterization of sulfur and iron in coal waste rock, Elk Valley, British Columbia, Canada



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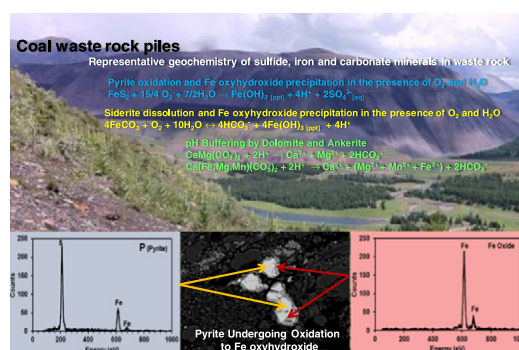
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HIGHLIGHTS

- Geochemical and mineralogical characterization of Fe and S in coal waste rock is discussed.
- Fe occur as pyrite, siderite, goethite, lepidocrocite in the waste rock dumps.
- S occur as sulfide minerals, organic sulfur and sulfate mineral (barite) in the waste rock.
- Iron oxyhydroxides are formed from oxidation of siderite and pyrite in the waste rock dumps.
- Oxidized S is released through the waste rock drains to the receiving surface water as sulfate.

GRAPHICAL ABSTRACT



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ABSTRACT

Exposure of coal waste rock to atmospheric oxygen can result in the oxidation of sulfide minerals and the release of sulfate (SO_4^{2-}) and associated trace elements (e.g., Se, As, Cd, and Zn) to groundwaters and surface waters. Similarly, reduced iron minerals such as siderite, ankerite, and the sulfide, pyrite, present in the waste rock can also undergo oxidation, resulting in the formation of iron oxyhydroxides that can adsorb trace elements released from the oxidation of the sulfide minerals. Characterization and quantification of the distribution of sulfide and iron minerals, their oxidation products, as well as leaching rates are critical to assessing present-day and future impacts of SO_4^{2-} and associated trace elements on receiving waters. Synchrotron-based X-ray absorption near edge spectroscopic analysis of coal waste rock samples from the Elk Valley, British Columbia showed Fe present as pyrite (mean 6.0%), siderite (mean 44.3%), goethite (mean 35.4%), and lepidocrocite (mean 14.3%) with S present as sulfide (mean 26.9%), organic S (mean 58.7%), and SO_4^{2-} (mean 14.4%). Squeezed porewater samples from dump solids yielded mean concentrations of 0.28 mg/L Fe and 1246 mg/L SO_4^{2-} . Geochemical modeling showed the porewaters in the dumps to be supersaturated with respect to Fe oxyhydroxides and undersaturated with respect to gypsum, consistent with solids analyses. Coupling Fe and S mineralogical data with long-term water quality and quantity measurements from the base of one dump suggest about 10% of the sulfides (which represent 2% of total S) in the dump were oxidized over the past 30 years. The S from these oxidized sulfides was

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released to the receiving surface water as SO_4^{2-} and the majority of the Fe precipitated as secondary Fe oxyhydroxides (only $3.0 \times 10^{-5}\%$ of the Fe was released to the receiving waters over the past 30 years). Although the data suggest that the leaching of SO_4^{2-} from the waste rock dump could continue for about 300 years, assuming no change in the rate of oxidation of sulfides, SO_4^{2-} is currently not a concern in receiving surface waters as the concentration levels are below regulatory limits.

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1. Introduction

Coal is an abundant and widely distributed fossil fuel, providing about 30% of global primary energy needs. It is also used in the production of 70% of the world's steel (World Coal Association, 2014). Surface coal mining in mountainous areas generally requires the removal of hundreds of meters of overlying rock to access coal seams, a process that generates large volumes of waste rock. Coal waste rock dumps often contain sulfur (S)- and iron (Fe)-rich minerals. The S is predominantly present as sulfide minerals (e.g., pyrite, marcasite, chalcopyrite, sphalerite, and galena) and organosulfur compounds whereas the Fe is present with S in pyrite, jarosite, troilite, and pyrrhotite and non-sulfur containing minerals such as siderite, ankerite, and illite (Chou, 2012; Waanders et al., 2003; Younger, 2004). Trace elements (e.g., Se, As, Cu, Cd, Zn, V, Pb, Ni, Mn, and Cr; all potential contaminants) are often associated with the primary sulfide minerals present in coal waste rock dumps (Huggins et al., 2009; Jamieson, 2011; Riley et al., 2012; Silva et al., 2011; Szczepanska and Twardowska, 1999).

Exposure of sulfide minerals present in waste rock dumps to oxidizing conditions (i.e., atmospheric oxygen and recharging water) can result in the mobilization of sulfate (SO_4^{2-}) and associated trace elements to receiving groundwaters and surface waters (Dang et al., 2002; Jamieson, 2011; Silva et al., 2011; Szczepanska and Twardowska, 1999; Younger, 2004). In the absence of carbonate minerals in the waste rock (to neutralize acids produced by the oxidation and subsequent hydrolysis reactions), the oxidation of sulfide minerals can also result in the discharge of acidic waters (Jamieson, 2011; Silva et al., 2011). In addition to sulfide oxidation, ferrous iron (Fe^{2+}) present in minerals such as pyrite, siderite, and ankerite can oxidize to ferric iron (Fe^{3+}), undergo hydrolysis, and precipitate as iron oxides and oxyhydroxides such as ferrihydrite, goethite, hematite, magnetite, and lepidocrocite (Silva et al., 2011; Zheng et al., 2007). These iron oxyhydroxides can adsorb trace elements and thus act as an internal geochemical control on the porewater concentrations of potential contaminants in waste rock dumps (Biswas et al., 2017a; Hendry et al., 2015; Jamieson, 2011; Zheng et al., 2007).

With coal reserves of 8.7 Bt (Stone, 2008), Canada is a major producer of steelmaking coal; production in 2013 was 34 Mt (Noble, 2014; World Coal Association, 2014). The Elk Valley, located in the Eastern Kootenay coal district of southeastern British Columbia (Fig. 1), is a major steelmaking coal production region (Goodarzi et al., 2009) in Canada. Although coal mining in the Elk Valley began in late 1897, large-scale (open cast) coal mining only began in the late 1960s. Currently, five active open-pit steelmaking coal mines are operating, producing 26.7 Mt of coal in 2014 (Teck Resources, 2015). The production of coal has resulted in the generation of about 4.7 billion bank cubic meters (BCM) of waste rock in the Elk Valley (Golder Associates, 2014).

The Elk River flows through the Elk Valley and into Lake Kootenay, crossing into the United States about 110 km downstream of the town of Sparwood (Fig. 1). Hauer and Sexton (2013) compared water quality of the Elk River Basin to the neighbouring Flathead River Basin and observed elevated concentrations of contaminants of interest (CIs). They observed SO_4^{2-} and Se concentrations downstream of the coal mines in the Elk Basin that were 40–50 and 7–10 times greater than background, respectively, and attributed the increased loading to the

regional surface coal mining. The presence of elevated concentrations of CIs in the transboundary Elk River system resulted in the establishment of a multi-disciplinary research program in 2012 with the goal of improving upon our limited understanding of the origin, fate, and transport of potential CIs (Se, Cd, Zn, SO_4^{2-} , and NO_3^-) in porewaters of the coal waste rock dumps in the Elk Valley. Such knowledge is critical to calculate present-day and future CI releases from the dumps and into the Elk River, define possible internal geochemical controls on the CIs, and propose potential measures to control the loading of CIs to groundwaters and surface waters. Understanding the present-day and future impacts on the receiving waters requires a detailed study of the distribution of primary S and Fe minerals in the waste rock dumps, their oxidation products, as well as their oxidation and leaching rates. The objectives of the current study were to quantify the distribution and mineralogical reservoirs of S and Fe in coal waste rock dumps in the Elk Valley and the rates of leaching of S (as SO_4^{2-}) and Fe from the dumps to the Elk River. The occurrence of S and Fe minerals have previously been determined in the coal waste rock piles as part of general geochemical and mineralogical characterization of coal waste rock to determine the occurrence and distribution of potential trace element contaminants and define the potential for acid mine drainage (Dang et al., 2002; Nganje et al., 2011; Ribeiro et al., 2013, 2010; Sahoo et al., 2014; Shahhoseiny et al., 2013; Szczepanska and Twardowska, 1999). However, the focus of the current study is to quantify the differences in the bulk mineralogical occurrence of sulfide and iron phases in freshly blasted coal waste rock as well as in weathered (aged) coal waste rock from dumps. These differences are used with the 10-year record of coal waste rock drain water chemistry to estimate present-day as well as future dump-scale oxidation and leaching rates of S (as SO_4^{2-}) and Fe to the environment. The objectives of the current study were attained by analyzing solid and porewater samples from freshly blasted waste rock as well as weathered waste rock from dumps in conjunction with a 10-year record of water chemistry and discharge rates from a rock drain located at the base of the dump. Analytical methods, including synchrotron-based X-ray absorption spectroscopy (XAS), inductively coupled plasma mass spectrometry (ICP-MS), LECO combustion analysis, ion chromatography (IC), and electron microprobe analysis (EMPA), were required to characterize dump samples in the detail required to meet the objectives. In addition, humidity cell experiments were used to simulate the geochemical weathering in waste rock samples in order to provide insights into geochemical processes governing the oxidation and leaching of CIs from the waste rock dumps. Further, analysis of rock drain data provided unique dump-scale estimates of sulfide oxidation and leaching rates not attainable using classical laboratory methods (e.g., humidity cells, columns, plug flow reactors, leach tests) or small-scale field trials (e.g., barrels, test piles, leach pads) that are subject to scaling factors (Malmström et al., 2000). The fact that the waste rock present at all the dumps in the Elk Valley comes from the same source rock suggests that findings of this study are applicable to the other regional dumps. Further, the unique dump-scale oxidation rates determined here may be applicable to other waste rock dumps with similar chemical/mineralogical composition. The findings of this study provide critical data that will allow us to define the long-term fate and transport of Se and As associated with the primary sulfide minerals in the waste

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