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Weathering of Zinc-(Zn)-bearing Mine Wastes in a Neutral Mine Drainage Setting, Gunnerside Gill, Yorkshire

C Tame^{a,1}, K.A. Hudson-Edwards^a, and H.A.B. Potter^b

^aDepartment of Earth and Planetary Sciences, Birkbeck, University of London, Malet St., London WC1E 7HX, UK

^bEnvironment Agency England & Wales, c/o Institute for Research on Environment and Sustainability, Newcastle University, Newcastle upon Tyne, NE1 7RU, UK

Abstract

Numerous areas throughout the world are affected by circum-neutral pH, low iron (Fe) drainage with high concentrations of zinc (Zn) arising from discharges from, and weathering of, mine wastes. Gunnerside Gill, a small upland tributary in the headwaters of the River Swale in Yorkshire, is such a site affected by historic lead and zinc mining. The aim of the study is to assess the controls on Zn mobilisation from the mine tailings and floodplain sediments to the river water through a column leaching experiment. Sphalerite has been identified as the primary Zn mineral in the bedrock within Gunnerside Gill. However, there is more evidence of secondary phases of Zn were including Fe oxides and phosphates present within the samples and the BCR data suggests it is these phases that appear to be undergoing the majority of the Zn dissolution.

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1. Introduction

Numerous areas throughout the world are affected by circum-neutral pH, low iron (Fe) drainage with high concentrations of zinc (Zn) arising from discharges from, and weathering of, mine wastes. Several historical mining areas in the UK are characterised by such circum-neutral, low Fe drainage, with high levels of dissolved Zn¹. Gunnerside Gill, a small upland tributary in the headwaters of the River Swale in Yorkshire, is such a site affected by historic lead and zinc mining in the Northern Pennines. The aim of the study is to determine the controls on Zn mobility from Gunnerside Gill mine tailings and river bank sediments using column leaching experiments and

E-mail address: rockology@hotmail.com

aqueous field data.

2. Site setting

Gunnerside Gill (54.3994° N, 2.0922° W) is a small upland tributary in the headwaters of the River Swale. Gunnerside Gill is underlain by a series of Carboniferous strata comprising silica cemented sandstones, limestones and cherts². Hydrothermal Pb, Zn, barium and fluorine-rich fluids were introduced into the Carboniferous strata by the intrusion of the Wensleydale Granite at depth, during the late Carboniferous or early Permian³. The volume of Zn produced from Gunnerside Gill is not well documented; although sphalerite was present with galena in Swaledale it was not commercially worked⁴. However, it has been estimated that 17796 tonnes of Pb were produced from mining in Gunnerside Gill⁵. The upper reaches of the River Swale, including Gunnerside Gill, were often mined by ‘hushing’. Hushing is a hydraulic form of mining in which dams were constructed upslope of the shallow mineral veins and sluices were opened to erode the overburden and expose the mineral veins. This practice resulted in the mobilisation of significant amounts of fine-grained metal contaminated sediments to the catchment⁶. In a previous study conducted by⁷ following flooding in 2000, samples of overbank and channel-edge flood sediment were recovered from along the River Swale. In the silt and sand sized fraction of the channel edge flood sediment recorded concentrations of Zn were found to be 4500 mg/kg as a result of the sediment input from Gunnerside Gill. Zinc concentrations in the clay and silt sized fraction of the overbank flood sediment were found to be 14000 mg/kg.

3. Materials and methods

Four samples of mine waste and floodplain sediment (mine waste: Gun 1, 2, 3; floodplain sediment Gun 9) from Gunnerside Gill were characterized by X-ray diffraction, electron microprobe analysis, total metal extraction and geochemical analysis and chemical sequential extraction. These samples were then used to model 10 years of weathering by undertaking a column leaching experiment. The columns comprised clear plastic tubes with acid washed quartz sand at the base and 1400 g of sample added in uniform layers of 1 to 2 cm. High-purity water was added on a weekly basis to mimic seasonal rainfall and the resulting leachate was sampled and subsequently analysed by ICP-AES.

4. Results and discussion

Gun 1, Gun 2 and Gun 3 are mixtures of waste rock and tailings, and Gun 9 is floodplain sediment from the lower reaches of Gunnerside Gill near its confluence with the River Swale. The general mineralogy of the samples is shown in Table 1. The Fe oxides contain variable proportions of Mn, Pb and Zn.

Table 1. Mineralogy of pre column experiment samples (X = major component, x = minor component, x_g = individual grains identified by EMP)

Sample	Quartz	Barite	Calcite	Fluoride	Clays	Cerussite	Galena	Fe Oxides	Sphalerite	Pb/Zn Phosphate	Pyrite
Gun 1	X	X	X		x	x		x _g			x _g
Gun 2	X	X	X	x	x		x _g	x _g			
Gun 3	X	X		x	x		x		x _g		x _g
Gun 9	X	X		x	x			x _g		x _g	

Based on the results of the BCR sequential extraction, as shown in Figure 1, the proportion of Zn in all four samples was found to be present mostly in the acid extractable or soluble/carbonate/cation exchange fraction, followed by the reducible or Fe/Mn oxihydroxide fraction in the case of Gun 1, Gun 2 and Gun 9 and the oxisible or organic matter/sulphide fraction in Gun 3. This is reflected in the original mineralogy of the samples, Fe oxides were

found to be present in Gun 1, Gun 2 and Gun 9, and sphalerite was identified in Gun 3. With the exception of Gun 1 the post column experiment solids showed a change in the proportion of Zn in each fraction. In Gun 2 the proportion of Zn in the organic matter/sulphide fraction increased and the Fe/Mn oxihydroxide fraction decreased, indicating the Zn has most likely been leached from the Fe oxides. In Gun 3 the Zn in the organic matter/sulphide fraction increased and reduced in the soluble/carbonate/cation exchange fraction, indicating the Zn release is likely to be from weakly bound mineral surface sites, Gun 9 showed a similar if less intense trend.

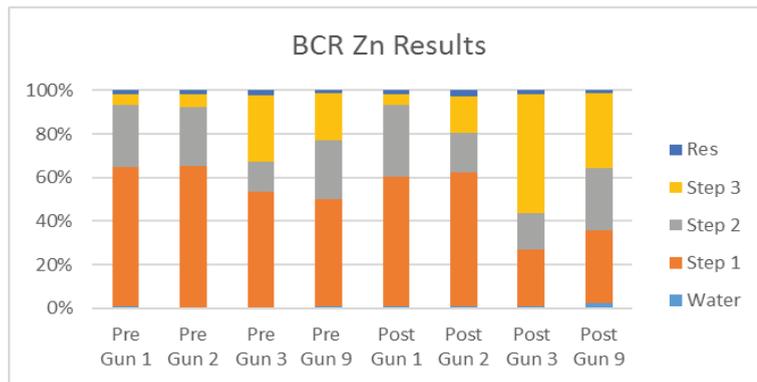


Fig. 1 BCR graph for the pre and post solid column material.

The difference between the pre and post column Zn concentrations within the solids is shown in Table 2. The percentage change in Zn between the pre and post column experiment for Gun 1 are minimal. The percentage changes are more noticeable in Gun 2, Gun 3 and Gun 9, which is reflected in the changes between the fractions in the BCR data as depicted in Fig. 1.

Table 2. Difference of Zn concentration between the pre and post column solids

Zn concentration in solid (mg/kg)	Pre column	Post column	% difference
Gun 1	5.8	5.6	3
Gun 2	17.5	10.6	40
Gun 3	11.2	5.8	48
Gun 9	6.9	4.2	38

The Environment Agency⁸ conducted monthly water monitoring of Gunnerside Gill over a one year period. The results show that 92% of the Zn concentrations exceeded the respective Environmental Quality Standards (EQS), calculated for Gunnerside Gill based on a hardness of 70 mg/L sampled in February 2010 as 50 µg/L for Zn. Figure 2 is a conceptual model that relates the EA monitoring data to the experimental column leaching data, GG1 to GG6 represent EA monitoring locations. The results indicate that Gun 1 is already weathered compared to Gun 2, Gun 3 and Gun 9 which are still leachable. With the exception of Gun 3 the column leaching Zn concentrations are generally consistent with those reported from the EA monitoring. This could suggest that Gun 3 in particular, when leached in the field, does not consistently or directly supply waters to the channel of Gunnerside Gill or that there is a change in the environmental conditions between the sampling site and the EA monitoring location. In addition several authors⁹ demonstrated that column leaching studies can over-estimate surface release rates and reaction rates in general by up to 3-4 orders of magnitude, respectively.

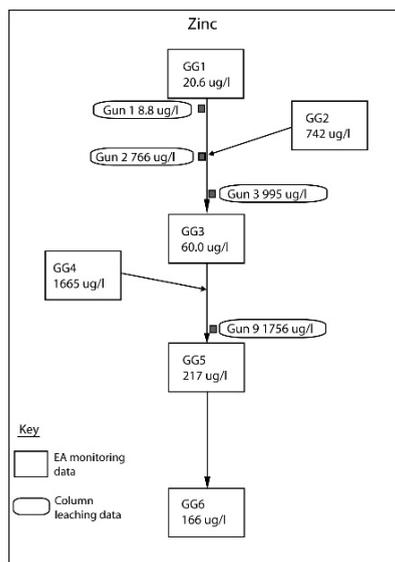


Fig. 2 Conceptual model of EA monitoring data (averaged over 1 year) and column leaching data (averaged from week 15 to week 40).

Although sphalerite has been identified as the primary Zn mineral in the ore, evidence of secondary phases of Zn were noted including Fe oxides and phosphate phases. Based on the BCR data it seems that the more soluble Zn and Fe oxides appear to be undergoing the majority of dissolution, following the results of the column leaching experiment. However, it is also likely that there will be a certain amount of sphalerite dissolution within Gunnerside Gill as a whole. Sphalerite has been identified in one of the samples (Gun 3), is present in the underlying bedrock and surrounding mine wastes, and is known to be relatively unstable in atmospheric conditions. In addition Gunnerside Gill is likely to represent a complex redox environment that will constantly influence the dissolution and precipitation of Zn minerals as the conditions dictate.

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