Numerical Modelling of Mine Pollution to Inform Remediation Decision-making in Watersheds

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Abstract

Prioritisation of mine pollution sources for remediation is a key challenge facing environmental managers. This paper presents a numerical modelling methodology to evaluate potential improvements in stream water quality from remediation of important mine pollution sources. High spatial resolution synoptic sampling data from a Welsh watershed were used to calibrate the OTIS solute transport model. Simulation of mine pollution remediation scenarios using OTIS revealed decreases in stream Zn concentrations between 9% and 62% under mean streamflow conditions. Remediation scenarios under low streamflow conditions were less effective (<1% to 17% decrease in Zn concentrations), due to diffuse and metal-rich groundwater inflows.

Keywords: OTIS, Mine Pollution, Remediation, Water Quality, Synoptic Sampling

Introduction

In the United Kingdom (UK), millennia of metal mining have produced substantial quantities of mine wastes that contaminate approximately 2800 km of watercourses (Johnston *et al*. 2008). Pollution from abandoned metal mines is recognised as a major cause of failure to achieve environmental objectives set out in statutory River Basin Management Plans (RBMPs) (Jarvis and Mayes 2012). As such, identification and prioritisation of mine pollution sources for remediation is one of the major challenges facing environmental managers.

In UK watersheds, potential improvements in stream water quality from remediation or treatment of mine pollution sources are evaluated using a mass balance approach (Jarvis and Mayes 2012). Under the approach, metal loads from point sources of pollution (e.g. drainage adits or tributary inflows) are subtracted from a total watershed metal load (calculated downstream from all known mine workings) to simulate the remediation of the point sources. However, there are two key limitations to this approach that may affect the efficacy of remediation and compliance with the mine pollution reduction targets set out in RBMPs. First, metal loading data from diffuse pollution sources are generally not available, therefore mass balance modelling of water quality improvements are limited to evaluation of point source remediation only. Second, the mass balance approach does not consider natural attenuation of metal pollutants by instream chemical reactions (e.g. first order decay processes such as sorption and precipitation). As mine pollutants in UK watersheds (Zn, Pb, Cd) generally behave in a non-conservative manner (Jarvis *et al*. 2019, Byrne *et al*. 2020), neglecting chemical reactions may over- or underestimate the effects of remediation on stream water quality.

The primary aim of this research was to demonstrate a modelling methodology that overcomes these limitations and allows environmental managers to evaluate potential water quality improvements from different remediation strategies in mined watersheds. Further details on the methodology and results are described in Byrne *et al.* (2020).

Methods

Synoptic Sampling

Working in the heavily mined (primarily Pb and Zn) Nant Cwmnewyddion watershed in central Wales, we conducted tracer dilution and synoptic sampling (multiple water quality samples collected across a watershed under steady-state flow conditions) experiments under mean (Q45) and extremely low (Q99) streamflow conditions in July 2016 and July 2018, respectively. A concentrated sodium bromide (NaBr) solution was injected into the stream during both experiments and allowed to reach plateau concentrations over the 2.5 km study reach. Synoptic sampling was then carried out at 25 stream sites and 18 inflow sites in 2016, and at 31 stream sites and 6 inflow sites in 2018. Bromide concentrations (filtered at 0.45 μm) were determined by ion chromatography, and Zn concentrations (filtered at 0.45 μm, acidified) were determined by inductively coupled plasma mass spectroscopy. As Br is considered to behave in a conservative manner in circum-neutral streams like the Nant Cwmnewyddion (Dzombak and Morel 1990), the observed dilution of Br was used to estimate streamflow at all stream sample sites using the tracer-dilution method (Runkel *et al*. 2013). Further details on the methodological approach can be found in Byrne *et al*. (2020).

Numerical Modelling

Zinc concentration and streamflow data from the synoptic sampling campaigns were used to calibrate the OTIS (One-dimensional Transport with Inflow and Storage) solute transport model (Runkel 1998). The OTIS model has been widely used to simulate hydrologic and geochemical processes in streams (Runkle 2007); however, application of the steady-state model to simulate watershed processes is rare.

There are four steps involved in the OTIS modelling process (Walton-Day *et al*. 2007). Physical transport parameters (dispersion coefficient (D); stream cross-sectional area (A); storage zone cross-sectional area (AS); storage zone exchange coefficient (α)) were determined from Br breakthrough curves at three 'transport sites' and from field measurements at a fourth transport site (**STEP 1**). OTIS models for filtered Zn in 2016 (Q45) and 2018 (Q99) were then calibrated assuming conservative transport. In this study, conservative transport assumes no removal of metal species from solution by sorption or precipitation processes. The study reach was first subdivided into twenty-four (2016) and thirty (2018) sub-reaches to bracket the location of synoptic samples. Then, lateral inflow concentrations (the difference in streamflow between the downstream ends of adjacent model reaches divided by the distance between the downstream ends of the model reaches) were estimated for each model sub-reach by using either sampled inflow concentrations or effective inflow concentrations. Effective inflow concentrations represent the mean dissolved Zn concentration entering a stream segment under the assumption of conservative transport, and may be developed using simple mass balance calculations on individual stream segments (Kimball *et al*. 2002). If the simulated Zn concentration profiles (assuming conservative transport) plotted above the observed Zn concentration profiles, reactive (non-conservative) transport was assumed (**STEP 2**). Simulation of non-conservative transport was then conducted by estimating first-order removal coefficients (λ) using non-linear least squares regression in OTIS-P (Runkel 1998) (**STEP 3**). Once calibrated, simulation of remedial alternatives was achieved by changing the lateral inflow concentrations of different mine pollution source areas to mimic potential remediation scenarios (**STEP 4**).

Remediation scenarios assumed a 94% reduction in inflow or source area filtered Zn concentrations, which is comparable with other mine pollution treatment schemes in the UK (Stanley 2020). The remediation scenarios simulated in this study, and based on consultation with the environmental regulator (Natural Resources Wales), included reductions in Zn from Wemyss Mine and Frongoch Adit and an aggregate of both sources.

Results and Discussion

Stream Chemistry and Streamflow

Zinc exceeded regulatory standards (15 µg L^{-1}) along the entire study reach and under both streamflow conditions (fig. 1). Stream Zn concentrations varied between 40 µg L^{-1} and 2901 µg L^{-1} under Q45 flow conditions (fig. 1a). Inflow concentrations from Mill Race Stream (4997 µg L^{-1}), which drains Wemyss Mine, and Frongoch Adit (3907 µg L^{-1}) appeared to be the main drivers of increased stream Zn concentrations. However, high Zn concentrations (maximum = 8514 µg L^{-1}) were also observed in several narrow diameter pipes draining the left bank of the stream opposite Graig Goch Mine. Stream Zn concentrations were substantially higher under Q99 flow conditions, varying between 18 µg L^{-1} and 8146 µg L^{-1} (fig. 1b). Increases in stream concentrations again appeared to be related to surface inflows from Mill Race Stream (5825 µg L^{-1}) and Frongoch Adit (9000 µg L^{-1}). A substantial increase in stream Zn concentrations was observed opposite Graig Goch Mine. However, only one of the drainage pipes (7358 µg L^{-1}) adjacent to Graig Goch Mine was flowing under Q99 flow conditions, and the negligible loading from this pipe cannot account for the observed increase in stream concentrations.

Streamflow at the end of the study reach was 27.5^{-1} s^{-1} under Q99 flow conditions compared to 203 L s^{-1} under Q45 flow conditions (see Byrne *et al*. 2020). Frongoch Adit (mine water) was the largest contributor to overall streamflow under Q45 (12%) and Q99 (35%) flow conditions. However, streamflow increases in the reach next to Graig Goch Mine accounted for 12% and 29% of total streamflow under Q45 and Q99 flow conditions, respectively (Byrne *et al*. 2020). In the absence of surface tributary inflows in this reach (flow from the narrow diameter pipes was negligible), these data indicate

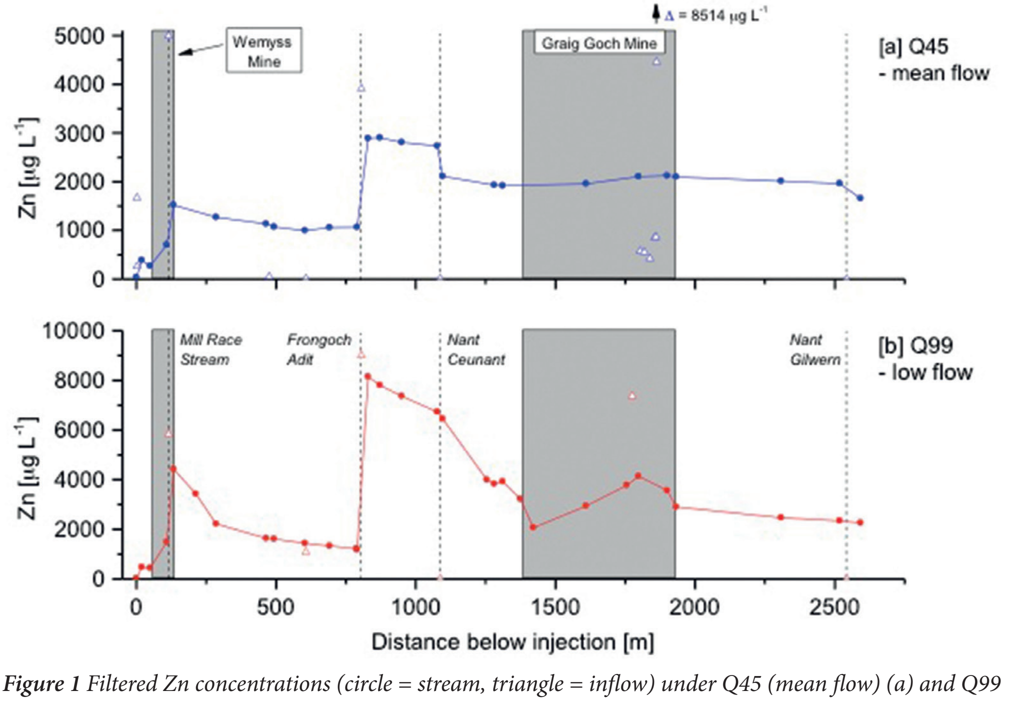

Figure 1 *Filtered Zn concentrations (circle = stream, triangle = inflow) under Q45 (mean flow) (a) and Q99 (low flow) (b) streamflow conditions in the Nant Cwmnewyddion. The locations of streamside mine wastes are indicated by grey boxes and labelled in (a). Major inflows (mine water and tributary) are indicated as dashed vertical lines and labelled in (b).*

diffuse groundwater was contributing to the increased streamflow in this reach. Zinc loads from this reach accounted for 20% and 31% of the total stream Zn load under Q45 and Q99 flow conditions, respectively.

OTIS Modelling

Initial conservative model simulations under Q45 streamflow conditions showed close correspondence between the simulated and observed Zn concentration data (fig. 2a). This indicates chemical (non-conservative) attenuation processes (e.g. sorption and precipitation) were relatively unimportant, and Zn was transported largely as a conservative solute. However, conservative model simulations under Q99 streamflow conditions showed the simulated data plotted above the observed Zn concentration data (fig. 2b), indicating removal of Zn by chemical reactions was important under extreme low flow conditions. Subsequent reactive (first-order decay) modelling demonstrated good fits with the observed Zn concentration data under both streamflow conditions.

Simulation of three mine site remediation scenarios (1. Wemyss Mine; 2. Frongoch Adit; 3. Wemyss Mine and Frongoch Adit) indicated variable improvements in stream water quality under different streamflows, and under different remediation scenarios (fig. 3). Under both streamflow conditions, the optimal scenario was remediation of both Wemyss Mine and Frongoch Adit, which achieved a 62% and 17% reduction in filtered Zn concentrations under Q45 and Q99 streamflows, respectively. However, it must be noted that remediation of Frongoch Adit alone could achieve similar improvements in stream water quality (52% and 16% reduction in filtered Zn under Q45 and Q99 conditions, respectively), primarily due to the large Zn loading from Frongoch Adit and that much of the filtered Zn originating from Wemyss Mine is removed by instream chemical reactions upstream from Frongoch

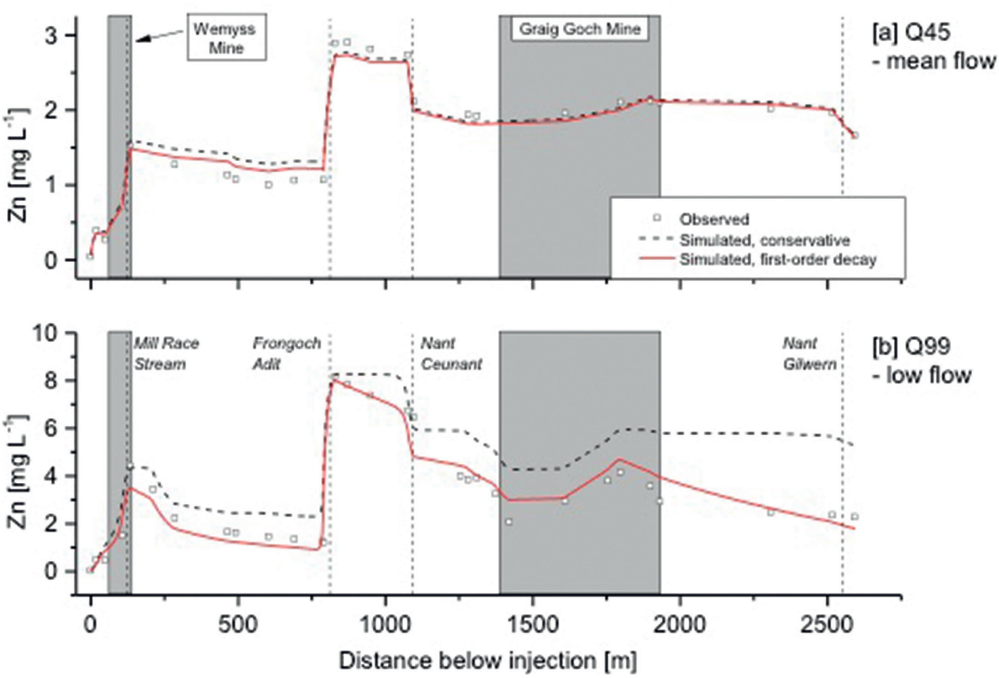

Figure 2 *Observed filtered Zn concentrations (squares), and simulated conservative (dashed line) and reactive (first-order decay)(red line) Zn concentrations under Q45 (mean flow) (a) and Q99 (low flow) (b) streamflow conditions in the Nant Cwmnewyddion. The locations of streamside mine wastes are indicated by grey boxes and labelled in (a). Major inflows (mine water and tributary) are indicated as dashed vertical lines and labelled in (b).*

Adit. It is noticeable also that the simulated reductions in filtered Zn concentrations were substantially lower under Q99 flow conditions. This is most likely due to high Zn concentration water entering the stream along an approximately 600 m length of the channel adjacent to Graig Goch Mine. Under Q99 flow conditions, this Zn-rich water is not diluted to the same extent as under Q45 flow conditions; and therefore, the effect on stream water quality and remediation efficacy is more severe.

Conclusions

This study demonstrates how numerical modelling of solute transport in mined watersheds can be used to simulate potential stream water quality improvements from different hypothetical mine site remediation alternatives. The OTIS model, calibrated with hydrological and Zn concentration data from synoptic sampling campaigns, simulated stream Zn reductions ranging from <1% to 62%, depending on the streamflow and the remediation scenario. Once calibrated, the OTIS model can easily simulate remediation of multiple sources of mine contamination across a watershed.

This study also demonstrates the importance of considering diffuse sources of mine contamination and reactive transport of solutes when simulating potential water quality improvements from different remediation approaches. OTIS simulations revealed how remediation of a single mine contamination source (Frongoch Adit) could yield similar water quality improvements to remediation of more than one source (Frongoch Adit and Wemyss Mine), due to in-stream attenuation of much of the Zn loading from one of the sources (Wemyss Mine). Furthermore, simulations of Zn concentrations under Q99 low flow conditions revealed remediation could be less effective than under Q45 mean flow conditions, largely due to untreated Zn-rich

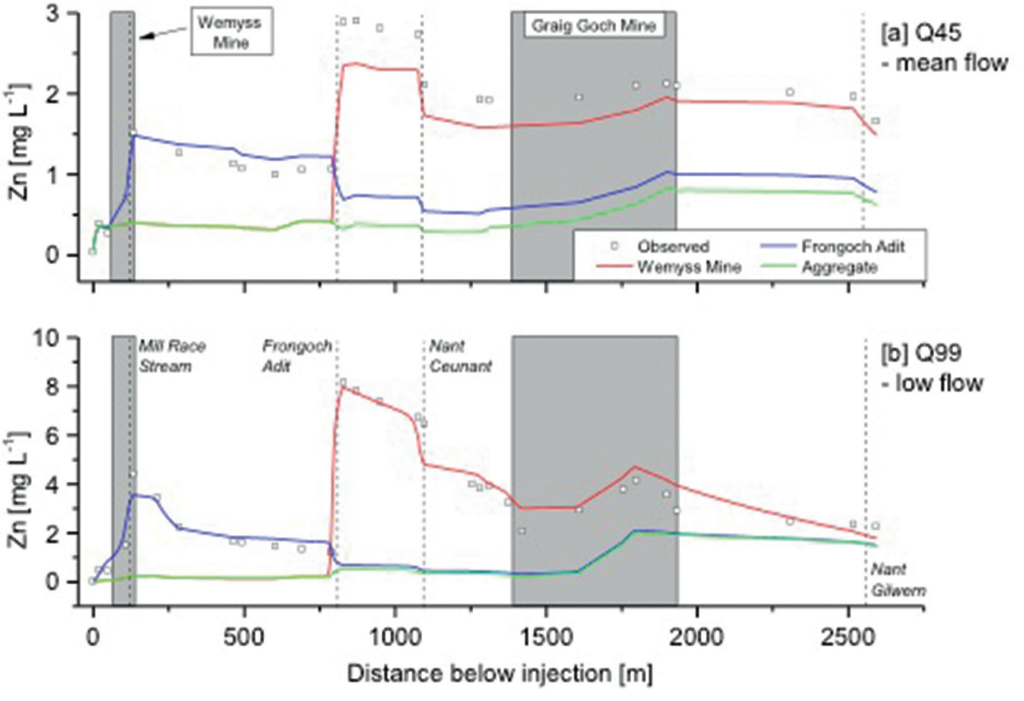

Figure 3 *Observed (squares) and simulated filtered Zn concentrations under Q45 (mean flow) (a) and Q99 (low flow) (b) streamflow conditions in the Nant Cwmnewyddion. Three different remediation scenarios are considered: Wemyss Mine (red line), Frongoch Adit (blue line), and an aggregate of Wemyss Mine and Frongoch Adit (green line). The locations of streamside mine wastes are indicated by grey boxes and labelled in (a). Major inflow (mine water and tributary) are indicated as dashed vertical lines and labelled in (b).*

water entering the system via diffuse subsurface pathways adjacent to Graig Goch Mine.

In many global regions, climate change is expected to increase the frequency and magnitude of flow extremes (low and high) in watersheds (Arnell *et al*. 2013). In temperate regions like the UK, this shift in hydrological regime is likely to increase the importance of diffuse contamination sources (Arnell *et al*. 2015), which may include runoff and erosion of surface mine wastes during high flows and contaminated groundwater efflux during low flows. Therefore, monitoring and modelling methodologies that consider diffuse sources of mine contamination will be needed by environmental managers to mitigate mine contamination in the future. Ongoing research in UK watersheds is focussed on simulation of solute transport and remediation effectiveness over a greater range of streamflows, and in particular during Q10 (high flow) events.

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