



Limitations on the role of the hyporheic zone in chromium natural attenuation in a contaminated urban stream



Barbara Palumbo-Roe^{a, *}, Vanessa J. Banks^a, Helen C. Bonsor^b, Elliott M. Hamilton^a, Michael J. Watts^a

^a British Geological Survey, Environmental Science Centre, Nicker Hill, Keyworth, Nottingham NG12 5GG, United Kingdom

^b British Geological Survey, The Lyell Centre, Research Avenue South, Edinburgh EH14 4AP, United Kingdom

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ABSTRACT

The urban hyporheic zone may offer natural attenuation potential for contaminants. This potential is contaminant-specific and may be spatially and temporally variable. The aim of this study was the assessment of the natural attenuation potential of the hyporheic zone of an urban stream receiving hexavalent chromium (Cr)-rich effluents from the historical land disposal of chromite ore processing residue (COPR) in Glasgow, Scotland. The evidence based approach involved the use of a network of multilevel piezometers for best capturing potential anoxic field conditions and fine-scale spatial gradients in solute concentrations of surface water and porewater. In-situ porewater sampling was integrated with sediment sampling and reach-scale monitoring of stream water quality. The results show a sharp decrease of total dissolved (filtered <0.45 μm) Cr concentrations at the surface water – sediment boundary in all profiles, from Cr mean values of 1100 $\mu\text{g l}^{-1}$ in surface water to 5 $\mu\text{g l}^{-1}$ in porewater. Chromium speciation analysis indicates that no Cr(VI) was detectable in the neutral pH, moderately reducing porewater, while it was the dominant species in surface water. Evidence of historical COPR detrital grains contributed to the total Cr concentrations (size fraction <150 μm) up to 8800 mg kg^{-1} in the streambed sediment. The abundance in the porewater of Fe (mean value = 1700 $\mu\text{g l}^{-1}$), mainly as Fe(II), a natural electron donor for Cr(VI) reduction to Cr(III), indicates a high natural attenuation potential of the hyporheic zone for downwelling dissolved Cr, through Cr(VI) reduction to Cr(III) and the formation of Cr(III) solids of low solubility. Authigenic Cr-rich rims on particles also documented active Cr precipitation from solution in the fine sediments. Large short-term changes of stream stage and stream water composition were not reflected in the hyporheic conservative (chloride) and reactive solute composition. This result indicates only limited surface water infiltration and suggests that small advective exchange might limit the effectiveness of the hyporheic zone for enhancing Cr surface water quality at the reach-scale. This is supported by further evidence from preliminary surface water quality synoptic sampling which shows only moderate to low downstream decrease in surface water Cr concentrations. The surface water investigation needs to be supported by combined water quality-flow monitoring and to be extended to a wider range of temporal and spatial scales to corroborate the reach-scale findings.

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1. Introduction

Awareness of the opportunity offered by the incorporation of the hyporheic zone into stream management for improving water quality and supporting biodiversity in urban areas is increasing

(Gessner et al., 2014; Lawrence et al., 2013; Ward et al., 2011). In order to improve the confidence in incorporating the hyporheic zone into urban water management, a better understanding of the natural contaminant attenuation capacity of the hyporheic zone in urban environments is needed (Smith et al., 2008). The hyporheic zone, a zone of exchange between the stream and the shallow subsurface, has the potential to mitigate diffuse pollution in urban streams. Yet, for some of the lowland streams with very fine and organic rich bed sediments, unfavourable hydraulic conditions,

* Corresponding author.

E-mail address: bpal@bgs.ac.uk (B. Palumbo-Roe).

limiting flow exchange between streambeds and surface water, are considered to constrain the efficiency of contaminant removal in the hyporheic zone (Herzog et al., 2015). Also largely unexplored, is the overall significance of the hyporheic zone attenuation potential in light of the large variability in stream flow regime and solute composition that urban watercourses may experience (e.g. as a result of storm runoff from impervious surfaces and variable wastewater effluent discharges). These variables can significantly affect the size and composition of the hyporheic zone, and the biogeochemical processes associated with it (Boulton et al., 1998; Mueller Price et al., 2015; Siergieiev et al., 2014; Zimmer and Lautz, 2013), and therefore, the contaminant attenuation capacity of the hyporheic zone.

Chromium (Cr) is a common historical contaminant in the urban environment due to its widespread use in industrial applications such as tanning, metallurgy and plating. Tons of chromite ore processing residue (COPR) have in the past been landfilled in urban areas (e.g. Hudson County, New Jersey, USA (Burke et al., 1991) and Glasgow, United Kingdom (Farmer et al., 1999)). Natural attenuation may be a viable option for remediation of Cr contaminated sites under certain settings (Palmer and Puls, 1996). Chromium attenuation in water is due to the reduction of Cr(VI), present as mobile and toxic chromate (CrO_4^{2-}) and dichromate (HCrO_4^-) anions, to Cr(III), whose solubility is limited by the formation of $\text{Cr}(\text{OH})_3$, Cr(III) co-precipitated with iron oxy-hydroxides or adsorbed to mineral phases (Ball and Nordstrom, 1998).

Several studies (Hellerich and Nikolaidis, 2005; Henderson, 1994; Higgins et al., 1998; Wadhawan et al., 2013; Whittleston et al., 2011) have reported Cr attenuation in groundwater, wetlands, estuarine sediments and soils. However, investigations within urban settings are lacking. Given their peculiarities, as described above, additional studies in urban streams are needed. Because of the strong dependence of Cr solubility and toxicity on Cr oxidation state, the hyporheic zone of some urban lowland streams with organic-rich silty-clay sediments, might have a high attenuation potential for Cr. In this study we investigated the occurrence and behaviour of Cr in the hyporheic zone of a Cr contaminated urban stream, the Polmadie Burn, a tributary of the river Clyde, Glasgow, UK, where Cr(VI) enriched waters have discharged into the stream channel for many decades. The catchment of the Polmadie Burn includes areas impacted by the historical disposal of chromite ore processing residue (COPR). Our goal was i) to provide a field-based evaluation of the hypothesis that Cr(VI) contaminated waters will not persist in the hyporheic zone of a stream characterised by fine sediments. Included in our evaluation was ii) a preliminary assessment of the impact of short-term stream stage fluctuations and compositional changes on hyporheic Cr water quality and iii) the importance at the reach-scale of the hyporheic zone as a Cr natural attenuation zone. By understanding the processes in the shallow hyporheic zone of the study site we aim to provide lines of evidence to complement the assessment of the linkage between urban land, groundwater and river contamination and the potential role of the hyporheic zone for enhancing water quality and improving aquatic habitats in an urban setting, to justify consideration in restoration work.

2. Study area

The Polmadie Burn (National Grid Reference [NGR] NS 60160 63255) (Fig. 1), is located within the Oatlands area of Glasgow, UK, approximately 3 km to the south-east of the city centre. It drains into the River Clyde in the study area and its catchment includes areas impacted by the historical disposal of COPR that originated from White's Chemical Works (operational between 1810 and 1967) in Shawfield in the immediate vicinity of the study site. The

chrome, derived from the processing of chromite ore, was supplied as chromium oxide from 1919 to 1945 and then as dichromate salts. Historical land uses of the catchment area ranged from farmland and residential to a colliery, railway sidings, industrial works and clay pits. Although the burn is situated upstream of the Clyde Tidal Weir at Glasgow, which is designed to stabilise the upstream banking of the River Clyde by maintaining a fixed water level, a tidal response in the groundwater levels was considered plausible. The stream levels respond rapidly to culverted drainage upstream of the study area.

Chromium concentrations in surface water up to 6.3 mg l^{-1} have been measured by Farmer et al. (2002). Existing UK water Environmental Quality Standards (EQS) for the protection of freshwater organisms range from 5 to $50 \text{ } \mu\text{g l}^{-1}$ total dissolved Cr. In a regional geochemical survey of the Clyde catchment, Fordyce et al. (2004) reported 3550 mg kg^{-1} Cr in bed sediments (<150 μm fraction) sampled from Polmadie Burn, largely exceeding the Canadian sediment quality guideline for Cr of 111 mg kg^{-1} (Macdonald et al., 2000). The Canadian standards rather than UK standards are considered in this study, as established sediment quality guidelines for metals in river and marine sediments are not available for the UK.

The bedrock geology beneath the Polmadie Burn comprises the Scottish Middle Coal Measures Formation, overlain in the south-east by the Scottish Upper Coal Measures Formation (British Geological Survey, 1993). These formations are Carboniferous in age and comprise cyclic alternations of mudstone, siltstone and sandstone with seat clay, seat earth and coal seams at some levels. The bedrock geology is overlain by superficial sequences of Quaternary River Terrace Deposits. These are covered by extensive sandy, silty, alluvial deposits laid down where local depressions coincide with the course of the modern river system, including the Polmadie Burn. The superficial geology of low permeability clays and silts (alluvium) and/or cohesive glacial till were considered to represent a barrier to significant deep groundwater flow gains or losses along the stream. The focus of the hyporheic zone investigation was, therefore, on the relationship between stream water and the upper ~50 cm of the streambed.

There are extensive areas of infilled and made ground to the south and south-east of the open section of the Polmadie Burn. Borehole evidence (records held by the BGS National Geoscience Data Centre) indicates that the alluvium extends to at least 20 m depth. More specifically, a borehole at National Grid Reference NGR NS 60036 63051 was taken to 15.0 m depth, but did not fully penetrate the alluvium. It encountered 4.50 m of made ground underlain by interbedded silty sand and sandy clay with silt on its parting.

The depth of incision of the Polmadie Burn is in the order of 3 m. This is elevated relative to that of the River Clyde. The channel has been canalised. In the lower, northerly flowing reach (site A, Fig. 1) the valley side slopes are about 17° and a small terrace of 1.0–1.5 m width has formed approximately 0.9 m above the valley floor on both sides of the valley. In the upper, north-westerly flowing reach (site B, Fig. 1), the valley sides are steeper, such that the profile takes more of a "U"-shape form and terrace development is minimal. The bed of the Polmadie Burn comprises a dark brown to dark grey organic-rich silty-clay. The presence of large debris (shopping trollies, waste, rugs, bricks) was also noticeable.

3. Methods

The multiple lines of evidence approach combined in-situ monitoring, hydraulic testing, sampling and geochemical/mineralogical characterisation of hyporheic water, surface water and streambed sediments, to analyse stream-subsurface interactions

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