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Erosion of the Alberta badlands produces highly variable and elevated heavy metal concentrations in the Red Deer River, Alberta

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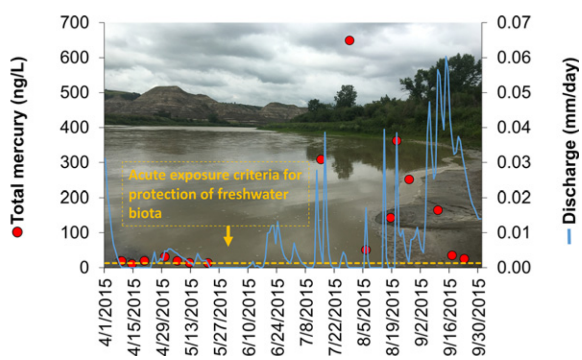
HIGHLIGHTS

- Erosion of contaminated soil is a major cause of riverine heavy metal contamination.
- Examined riverine heavy metal dynamics in a highly erosive watershed.
- No evidence of widespread enrichment of suspended sediments with heavy metals.
- Elevated riverine heavy metal concentrations due to high instream sediment mass.
- Erosion of 'background' soils produced levels typical of heavily impacted rivers.

GRAPHICAL ABSTRACT



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ABSTRACT

Erosion is important in the transport of heavy metals from terrestrial to fluvial environments. In this study, we investigated riverine heavy metal (Cd, Cu, Hg and Pb) dynamics in the Red Deer River (RDR) watershed at sites upstream ($n = 2$) and downstream ($n = 7$) of the Alberta badlands, an area of naturally high erosion. At sites draining the badlands, total water column Cd, Cu, Hg and Pb concentrations frequently exceeded guidelines for the protection of freshwater biota. Furthermore, peak concentrations of total Cd (9.8 μg L^{-1}), Cu (212 μg L^{-1}), Hg (649 ng L^{-1}) and Pb (361 μg L^{-1}) were higher than, or comparable to, values reported for rivers and streams heavily impacted by anthropogenic activities. Total suspended solids (TSS) explained a large proportion ($r^2 = 0.34$–0.83) of the variation in total metal concentrations in the RDR and tributaries and metal fluxes were dominated by the particulate fraction (60–98%). Suspended sediment concentrations (C_{sed}) and metal to aluminum ratios were generally not indicative of substantial sediment enrichment. Rather, the highly variable and elevated metal concentrations in the RDR watershed were a function of the high and variable suspended sediment fluxes which characterize the river system. While the impact of this on aquatic biota requires further investigation, we suggest erosion in the Alberta badlands may be contributing to Hg-based fish consumption advisories in the RDR. Importantly, this highlights a broader need for information on contaminant dynamics in watersheds subject to elevated rates of erosion.

1. Introduction

The contamination of surface waters with heavy metals (elements with a specific density > 5 g cm^{-3}; Callender, 2003) is a major

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environmental concern (Driscoll et al., 2013; Sutherland, 2000). Heavy metals exhibit a range of toxic effects to aquatic and terrestrial biota (Driscoll et al., 2013; Flemming and Trevors, 1989; Mebane, 2010) in addition to impacts on human health (Lanphear et al., 2005; Tchounwou et al., 2012). Although natural sources exist (e.g., weathering and wildfires), anthropogenic activities (e.g., mining and smelting, urbanization and industrial processes) substantially increase heavy metal fluxes to rivers (Horowitz and Stephens, 2008; Horowitz et al., 2012; Macklin et al., 2006; Sutherland, 2000). Metals in aquatic environments are conventionally classified as particulate or dissolved with dissolved forms operationally defined as material passing through a 0.45 μm filter (Nystrand et al., 2012; Owens et al., 2005). While dissolved forms are considered immediately bioavailable, the extent to which particulate forms are bioavailable will depend on the lability of metal species in particulate phase and the physicochemical properties of the environment (Eggleton and Thomas, 2004). A large proportion of heavy metal transport to the world's lakes and oceans occurs via rivers (Callender, 2003) and due to the strong affinity of metals for soil/sediment surfaces (Horowitz, 1991) most of this is associated with the particulate phase (Horowitz et al., 2012; Martin and Meybeck, 1979; Viers et al., 2009). There is now strong evidence for anthropogenic contamination of suspended sediments in many of the world's rivers (Horowitz et al., 2012; Owens et al., 2005; Viers et al., 2009), and erosion events are a major vector for the transport of suspended sediments and associated contaminants from terrestrial to aquatic environments (Horowitz et al., 2012; Rickson, 2014; Sutherland, 2000; Walling, 2005). As such, understanding the role of erosion on heavy metal dynamics in river systems is an active and important area of research.

Two factors govern the flux of metals to fluvial systems during erosion events: (i) the concentration of sediment bound metals; and (ii) the mass of sediment transported during erosion events. Both of these factors can be increased by anthropogenic activities (Burton and Johnston, 2010). Atmospheric deposition, mining activities, solid and liquid wastes, urban road networks, phosphate fertilizers and legacy pesticides are potential sources of soil contamination (Cadwalader et al., 2011; Nicholson et al., 2003; Roberts, 2014; Taylor and Owens, 2009; Wuana and Okieimen, 2011), while land clearing activities, including road construction, agricultural practices, and forestry activities, are major drivers of accelerated erosion (Swank et al., 2001; Burton and Johnston, 2010; Culp et al., 2013; Rickson, 2014). In addition to soil contamination, suspended sediments may also be enriched due to the selective erosion and transport of fine particles during erosion and/or resuspension events. This preferential erosion of fines (i.e., clays and silts, <63 μm) is higher during low energy erosion/resuspension events. Selective erosion of fines also occurs during high energy events; however, a greater proportion of the sediment load is in the coarse fraction (i.e., fine and coarse sands, >63 μm) (Quinton et al., 2001). Importantly, due to the relatively high surface area of fine vs. coarse sediment particles, metals are often enriched in silts and clays (Horowitz, 1991; Owens et al., 2005; Taylor and Owens, 2009). Because silts and clays generally make up the bulk of the suspended sediment load (Owens et al., 2005), enrichment of metals in suspended sediments can be substantial. For example, Quinton and Catt (2007) reported sediment bound metal concentrations in agricultural runoff that were above thresholds for the protection of aquatic biota and on average ≈4 times higher than the soils from which they were derived. This suggests that erosion driven contamination of rivers may not be limited solely to systems that drain heavily contaminated soils.

Badlands are highly erosive environments formed in arid and semiarid regions. They are typically characterized by sparse vegetation cover, lithologies which are dominated by active smectite clays, and the presence of steep slopes associated with well-defined rill systems (Kasanin-Grubin and Bryan, 2007; Liberti et al., 2009). Importantly, due to the high rates of erosion in these regions, rivers and streams that drain badlands are characterized by high concentrations of suspended sediment (Gallart et al., 2002); however, despite the potential for significant fluxes of sediment-bound metals in runoff from badlands, there is little information on riverine metal concentrations or fluxes in these systems. The Red Deer River (RDR) in western Canada drains the Alberta badlands and is characterized by high suspended sediment levels (Campbell, 1977). Importantly, the river is currently subject to fish consumption advisories due to elevated Hg concentrations in fish tissue (Alberta Health, 2009) and elevated metal concentrations in the water column have been reported downstream of the badlands (Anderson, 1996). However, the specific cause(s) of elevated Hg in fish tissue, or the role of sediments as a driver of elevated heavy metals in the RDR, have not been investigated. In this study, we examine the link between erosion in the Alberta badlands and the concentrations and fluxes of Cd, Cu, Hg and Pb in the RDR watershed. In addition to providing insights into heavy metal dynamics within the RDR itself, this study provides broader insights into the role of suspended sediment as a source of contaminants in highly erosional systems.

2. Methodology

2.1. Study area

The Red Deer River Watershed is located in southern Alberta and covers approximately 49,650 km^2. The headwaters of the RDR are in the Canadian Rockies where ≈70% of river discharge originates as snow melt (Tanzeeba and Gan, 2012). Peak flows in the RDR occur in the late spring/early summer months. The watershed transitions from alpine and foothills landscapes in the headwaters to prairie grasslands in the mid to lower reaches (Downing and Pettapiece, 2006). Average annual temperature is approximately 4 °C (taken at the city of Red Deer; http://climate.weather.gc.ca). Median annual precipitation across the watershed is 393 mm but varies from more 900 mm in the Rockies to approximately 270–400 mm in the grasslands (AMEC, 2009). Within the grasslands, the majority of annual precipitation occurs as rain during the May to September period (Downing and Pettapiece, 2006) with a smaller proportion (≈30% of annual precipitation) occurring as snow during colder months (Kasanin-Grubin and Bryan, 2007).

The underlying bedrock of the RDR watershed is formed predominately of Upper Cretaceous and Tertiary deposits (Campbell, 1977). Upper Cretaceous bedrock is dominated by shales, sandstones, thin ironstone bands, and coal seams while surficial Tertiary deposits consist of till, lacustrine deposits, and glacial outwash associated with Wisconsin glaciation (Campbell, 1977). In the mid to lower reaches of the watershed, the RDR is flanked by the Alberta badlands for approximately 300 km from the town of Nevis to Atlee near the Alberta-Saskatchewan border (Campbell, 1970; Bryan and Campbell, 1980) (Fig. 1). The Alberta badlands cover an area of approximately 800 km^2 (Campbell, 1977) and is formed from bedrock belonging predominately to the Horseshoe Canyon, Bearspaw and Dinosaur Park Formations (Price et al., 2013). Development of the Alberta badlands began approximately 15,000 years ago when meltwater incision exposed highly erodible bedrock during the retreat of Wisconsinan ice (Campbell, 1987).

Due to its highly erosive nature, the Alberta badlands contribute >70% of the sediment load to the RDR despite making up only a small proportion (≈2%) of the overall watershed area (Campbell, 1977). Average annual erosion rates within the Alberta badlands have been estimated to be approximately 4 mm yr^{-1} (Campbell, 1987). Within the badlands, sediment fluxes to the RDR are initiated primarily by intense but short lived convective rainstorms (Bryan and Campbell, 1980). Surface crusts develop rapidly on shale slopes upon wetting. Surface sealing contributes to low infiltration rates which generate substantial overland runoff over the sparsely vegetated surface (Campbell, 1970). This in turn leads to significant erosion primarily via sheet-flow and rilling (Campbell, 1987). In addition, surface runoff may enter fissures or desiccation cracks to transport sediment via piping (Campbell, 1987; De Boer and Campbell, 1990).