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Heavy metal mining and flood plain response in the upper Clyde basin, Scotland

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Abstract The Lowther Hills in the Southern Uplands of Scotland have a long history of metalliferous mining spanning over 700 years. Mining and ore processing generated significant quantities of metalliferous waste, but in the Clyde basin the geomorphological impact was largely restricted to headwater systems. In the mining district channel metamorphosis and flood plain aggradation resulted in active transformation of valley floors. With the cessation of mining, these headwater systems returned to laterally unstable single-thread gravel bed rivers. The pollution legacy remains acute with sediment-associated Pb values in excess of 150 000 mg kg^{-1}, surpassing UK contaminated land *action trigger values* by two orders of magnitude. Channel bank erosion rates of 30–50 mm year^{-1} are responsible for remobilizing metal-rich sediment from flood plain storage and now represents the dominant source of metals to downstream reaches. The historical and on-going significance of this process was evaluated 40 km downstream on the main flood plain of the River Clyde. At this site mining-derived sediments were incorporated by passive dispersal. A fingerprinting analysis indicated that the mining epoch contributed only a minor flux of sediment to the flood plain, nevertheless peak Pb values exceed 1 500 mg kg^{-1} illustrating more pernicious geochemical impacts.

Key words mining; geomorphological response; metalliferous sediments; flood plains; fingerprinting; temporal discontinuity; Scotland

INTRODUCTION

The Leadhills–Wanlockhead mining district in the Southern Uplands of southern Scotland has experienced a long, but intermittent, history of metalliferous mining extending back at least to Roman times. The ore field is small (*c.* 20 km^2), but at its peak contributed more than 90% of the total Scottish Pb and Zn production and as much as 10% of the national UK output of Pb (Smout, 1967). The eastern half of the mining district is located within the western headwaters of the River Clyde, most notably within the Glengonnar Water (Fig. 1). This tributary has a documented history of mining spanning over 700 years, initially for Au, but subsequently for Pb, Ag and Zn (Gillanders, 1981).

Early mining activity (notably from the thirteenth century onwards), was localized, intermittent and largely focused on alluvial gold deposits. Large-scale lead mining did not commence until the late eighteenth century, and thereafter production patterns reflected technological advances and international commodity prices. As processing techniques advanced, initially coarse-grained and ore-rich wastes became progressively finer-grained with a more varied mineralogy. Moffat (1991) estimated that *c.* 0.5 Mt of spoil remains in the immediate mining area.

The importance of physical transport processes to disperse this waste was confirmed by Hillier *et al.* (2001), who showed that the Pb in alluvial sediment was mostly present as cerussite ($PbCO_3$) and concentrated in the silt fraction. In the <6 μm fraction Pb was adsorbed onto organic matter, Fe-Mn coatings and clay minerals. The main aim of this study was to consider the geomorphological response and geochemical impact of metal mining in the upper Clyde basin and thus make a contribution to the growing body of literature considering the response of drainage basins to large-scale perturbations such as mining (cf. Miller, 1997).

BACKGROUND AND APPROACH

The morphology of the 650 km^2 upper Clyde study basin is controlled by lithology and structure, and can readily be divided into northern and southern components separated by the Southern Uplands Fault (Fig. 1(a)–(b)). To the south, Lower Palaeozoic sequences are dominated by folded and faulted Ordovician and Silurian mudstones, shales, greywackes and conglomerates. To the north, Upper Palaeozoic sequences, particularly Devonian–Permian sandstones, limestones and breccias occur along with Devonian aged andesetic and basaltic lavas and local outcrops of intrusive volcanics. Vein systems were emplaced during the Caledonian and Hercynian Orogenies (430–390 Ma).

The research focused on two reach scales. Firstly, within the mining district, on the flood plain of the Glengonnar Water, and secondly, 40 km downstream on the flood plain of the River Clyde at the Clyde–Medwin meanders (Fig 1(c)). The Glengonnar has a drainage area of *c.* 24 km^2, with a 12-km-long flood plain between 80 and 200 m wide. Rowan *et al.* (1995) documented phases of significant flood plain accretion in the Glengonnar flood plain linked to the historical mining activity. This involved channel metamorphosis and extensive channel–flood plain aggradation resulting from debris supply exceeding the transport capacity, thus conforming to the model of "active transformation" (Lewin & Macklin, 1987).

Forty kilometres downstream the River Clyde is, by British standards, a large lowland river actively migrating and reworking its flood plain (Brazier *et al.*, 1993). The valley floor attains widths in excess of 1 km and is bounded by glaciofluvial terraces. The main channel is highly sinuous and set within an active channel corridor *c.* 400 m wide producing a relatively flat, but highly segmented flood plain containing a range of relict landforms including palaeochannels, scroll bars and ox-bow lakes.

Flood plain histories were reconstructed using aerial photographs and maps dating back to 1846, complemented by field mapping and radiometric dating. Flood plain and channel sediments were collected from surface transects and from numerous trenches, sections and cores. A range of physico-chemical parameters was assessed including grain-size and heavy metal concentrations. Metal analysis was undertaken using atomic absorption spectrophotometry (AAS) and X-ray fluorescence (XRF) following standard procedures (Rowan *et al.*, 1995, 1999). Selected profiles were radiometrically dated using ^{137}Cs and ^{210}Pb techniques (Oldfield & Appleby, 1985).

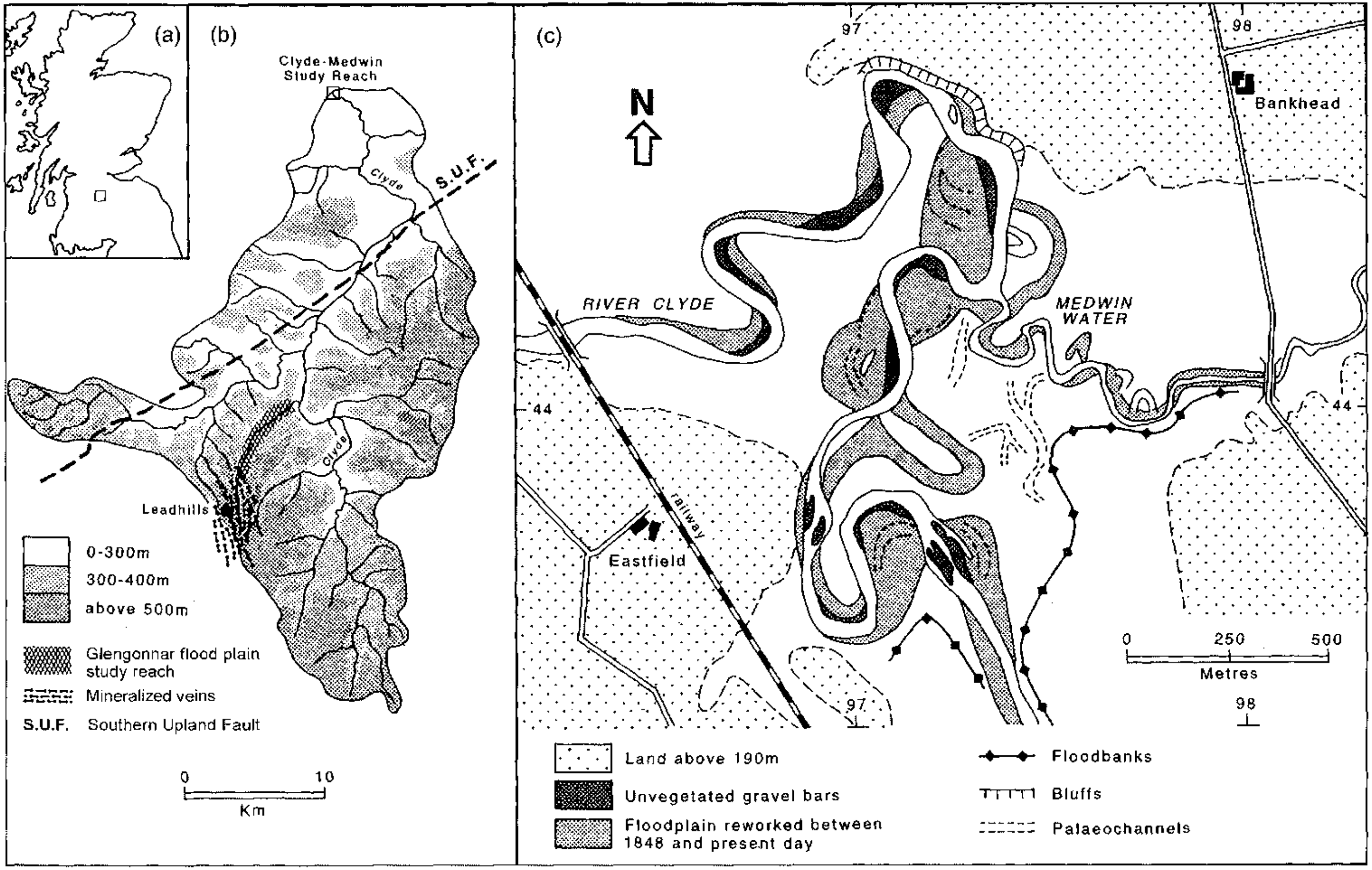

Fig. 1 (a) Location of study basin; (b) topography of the upper Clyde basin; (c) Clyde–Medwin meander study site.

FLOOD PLAIN RESPONSE—HEADWATER *vs* DOWNSTREAM REACHES

The Glengonnar Water

Metal levels in flood plain sediments show a complex spatial pattern, which resolves into three components: cross-valley variations relating to terrace remnants of different ages and metal content, the systematic decline of metal concentrations in the down-valley direction, and vertical variations corresponding to local aggradation histories. Typically, flood plain cross-sections show a series of terraces (0.4–2 m) stepping down to the present channel position. The most polluted soils (0–15 cm) are usually associated with the highest and oldest terrace remnants corresponding to the peak mining period in the early twentieth century. Within equivalent terrace surfaces Pb, Cu, and Zn levels declined logarithmically ($p > 0.5$) with distance downstream. This effect has been widely reported and results from processes including hydraulic sorting, loss to storage, and dilution effects through addition of tributary sediments (cf. Miller, 1997).

Variations of metal levels within vertical sequences have been correlated within mining output histories with peaks within the profile corresponding to periods of greater mining activity and associated flood plain aggradation (Rowan *et al.*, 1995). In the upper reaches of the Glengonnar system sediment-associated Pb levels >150 000 mg kg^{-1} surpass UK contaminated land *action trigger values* by two orders of magnitude. Action trigger values are defined as the level above which prolonged exposure is likely to cause injury to grazing animals or crop yields (ICRCL, 1990).

Temporal discontinuities in metal delivery

The potential for remobilization of metals from the flood plain was assessed from a survey of fine-grained sediment within the bed of the Glengonnar channel using the bed sampling technique developed by Lambert & Walling (1988). This method is designed to both quantify and sample a representative fraction of the readily mobilized fine-grained matrix of gravel-bed rivers. By sampling systemically down the length of

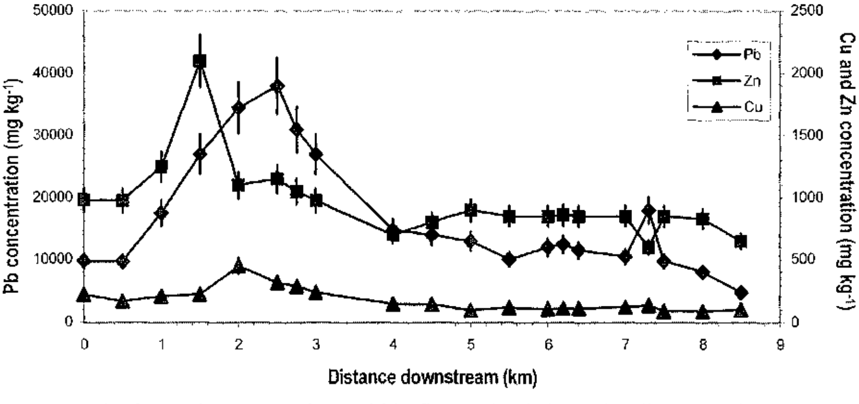

Fig. 2 Metal concentrations within fine-grained channel sediments, Glengonnar (error bars 95% CI).

the Glengonnar it was estimated that 30–60 t of fine-grained (<2 mm) sediment was stored in the transient channel sink. Concentrations of Pb, Zn and Cu within these sediments all peak within 2–3 km of the Glengonnar system (Fig. 2), correlating closely to the flood plain reaches with the highest metal levels.

For the Glengonnar flood plain, decadal-scale channel migration rates of 30–50 mm year^{-1} were established from sequential topographic maps and aerial photographs. Integrating flood plain metal levels and rates of channel activity the flood-plain-derived metals flux was estimated as *c.* 9 kg Pb m^{-1} year^{-1} (Rowan *et al.*, 1995). The resultant channel storage mass was thus 500–1000, 30–60, and 5–11 kg of Pb, Zn and Cu respectively (95% confidence interval—CI). Assuming that these matrix sediments are mobilized every 1–2 years, then this represents a potentially significant on-going supply of metals into the main channel of the River Clyde.

The Clyde meanders

Three cores to a maximum depth of 3 m were extracted from well-defined palaeochannels. The stratigraphy comprised a fine-grained unit (0.8–1.3 m thick) overlying gravels of undetermined depth (>3 m). The upper units were massive sandy-silts representing infill and over-bank deposits. Occasional lenses of gravelly-sand equated to discrete flood events. These palaeochannel fill sequences were easily distinguished from the underlying channel gravels by a sharp contact.

Down-profile variations in core geochemistry are exemplified using Pb and Zr (Fig. 3(a)). Significant quantities of Zr were found towards the base of all three cores, most notably in core 1 (not shown). Core 3 contains four distinct Pb peaks evidencing

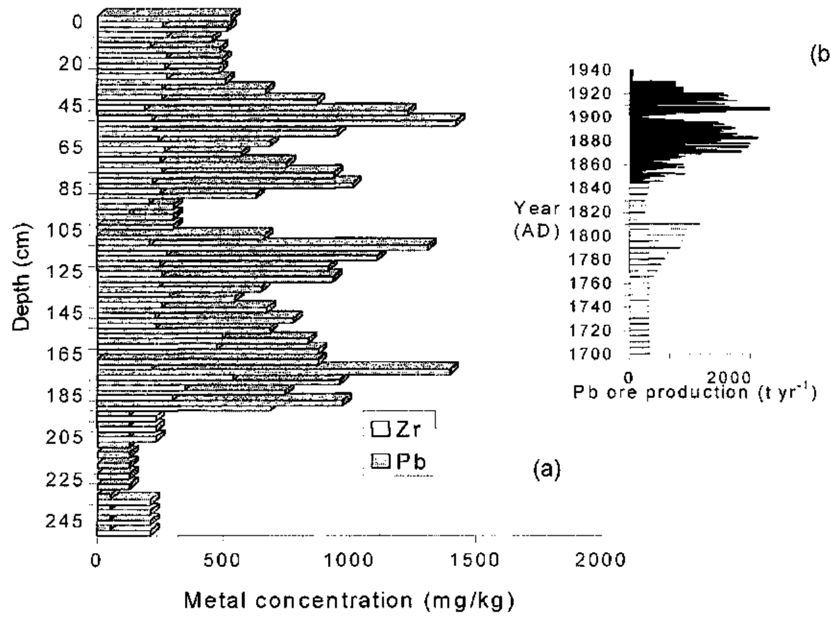

Fig. 3 (a) Pb and Zr variations with depth from Clyde-Medwin palaeochannel; (b) lead production from Leadhills mining field.

concentrations approaching 1500 mg kg^{-1}. The accretion history of the core was established using ^{210}Pb dating and matching sedimentary Pb peaks to peaks in mining activity (Fig. 3(b)). The upper Pb peaks were radiometrically corroborated as 1908 and 1880 respectively, the third peak was assigned to the Napoleonic period (*c.* 1810), while the fourth peak was associated with a significant increase in Zr. Both these lower peaks occur within the gravel sequences, and the Zr peak has been tentatively linked to alluvial gold mining during the Medieval period (*c.* 1600 AD) when hydraulic mining of Zr-rich placer deposits was widespread in the mining district (Rowan *et al.*, 1999). These metalliferous gravels were likely deposited as extensive point- and mid-channel bars within a channel system frequently switching position i.e. under conditions similar to the present day.

Variations in metal concentration result from both the intrinsic evolution of the meander system and external variations in metal supply. Channel change mechanisms have involved lateral migration, avulsions and neck cut-offs producing a highly segmented flood plain containing a relatively narrow (*c.* 400 m) corridor of metal-contaminated sediments reaching depths up to 3 m adjacent to the main channel (shaded zone in Fig. 1(c)).

Quantifying the mining flux at the Clyde meanders

An attempt was made to quantify the contribution of mining-derived sediments made to the Clyde meanders flood plain using a quantitative "fingerprinting" approach. The basis of this technique is to match selected physico-chemical properties of suspended sediment, or flood plain deposits, to the equivalent values of potential sources. A multivariate "unmixing model" was then used to apportion the flux back to its constituent sources (cf. Collins *et al.*, 1997). A novel element in this work was the use of a Bayesian framework to determine contributory coefficients along with a measure of model uncertainty i.e. the 95% confidence interval (Franks & Rowan, 2000).

Four source groups based on the major lithologies, Ordovician/Silurian, Devonian, Volcanics and Mining District, were sampled ($n = 49$). Both drainage basin samples and flood plain sediments (<125 μm fraction) were analysed by XRF yielding an optimized composite set of tracers (Pb, Rb, V, W, Ni and Cr). These tracers were then used in the unmixing model to determine the mean contribution from each source along with an estimate of the predictive uncertainty (cf. Small *et al.*, 2002).

Despite the areal dominance of Ordovician/Silurian lithologies across the basin, this source group typically produced less than half of the total sediment flux. Such upland areas are characterized by low intensity farming practices, and sediment supply is primarily from channel bank erosion. Most sediment is derived from the northern lowland areas of the catchment due to more intensive arable farming activity. The relative contribution (%) of mining-derived sediment is shown in Fig. 4. The discontinuous nature of the mining operations is clearly shown by the existence of four discrete peaks within the profile. During the Medieval period (*c.* 1500 AD) sediment was delivered from across the basin, and the contribution from the mining district was minimal. As the scale and intensity of mining operations grew the proportion of mining-derived sediments increased to maximum contributions of 1.8 ± 2.1% (95% CI). From map evidence, the enhanced supply of metalliferous sediments was

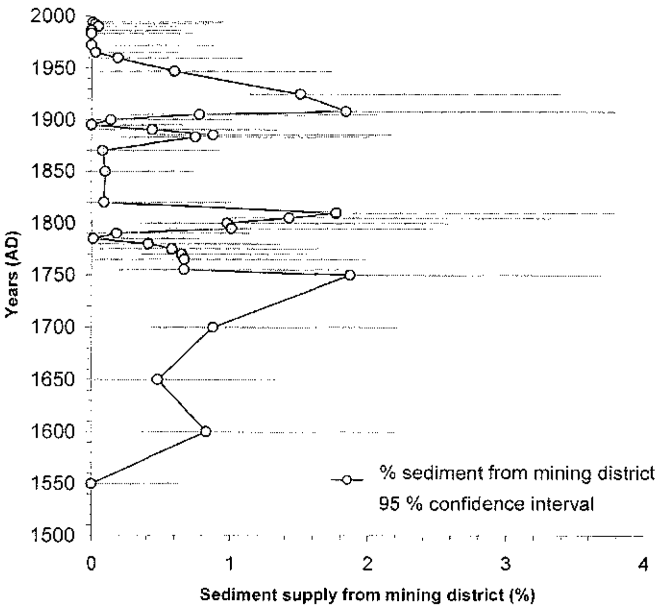

Fig. 4 Results of sediment fingerprinting for core 3 indicating the contribution of mining-belt-derived sediments since 1500 AD.

insufficient to affect the natural dynamics of the Clyde meanders. However, the geochemical impact is still clearly evident from the elevated soil surface metal levels i.e. Pb > 500 mg kg^{-1}, which while below UK Government "Action Trigger" values (ICRCL, 1990), remains an order of magnitude higher than the natural background levels. Metals are thus incorporated into the flood plain in a manner analogous to the "passive transport" mechanism described by Lewin & Macklin (1987) and testify to the on-going environmental legacy of the historical mining operations upstream.

CONCLUSION

Historical mining operations within the upper Clyde caused environmental impacts over a range of scales. In headwater systems, such as the Glengonnar, flood plains experienced active transformation through channel metamorphosis from single thread to braided systems and flood plain aggradation resulting from debris supply exceeding the transport capacity. These headwater systems have buffered the geomorphological response downstream as well as attenuating the geochemical impact.

At the Clyde meanders the geomorphological response was minimal, but metalliferous sediments were found to penetrate several metres within the near-channel zone. Variations in Zr and Pb levels were correlated with episodes of gold and lead extraction to give a chronological framework spanning *c.* 500 years. Variations in the provenance of fine-grained sediment were determined using a quantitative multivariate fingerprinting approach confirming that mining-derived sediments were incorporated by passive dispersal rather than by active transformation of the flood plain system. In quantitative terms the mining period contributed only a small fraction of the total sediment load, though the pollution implications are still acute. This combined stratigraphic and geochemical approach offers scope to explore the channel–flood plain response to drainage basin adjustments (climate, land use, flood frequency) within one of the most important water resource basins in Scotland.

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