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TRANSFER OF SEDIMENTS AND CONTAMINANTS IN CATCHMENTS AND RIVERS

Contaminated sediment dynamics in peatland headwater catchments

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Abstract

Purpose: Despite substantial research into Pb storage in peatlands, formal description of the mechanisms of contaminated sediment mobilisation is limited. This study explores the controlling factors of contaminated sediment dynamics in an eroding peatland in the Peak District National Park, UK.

Materials and methods: This study uses the Pb contamination stored near the peat's surface as a fingerprint to trace contaminated sediment dynamics in three severely degraded headwater catchments. A field portable XRF analyser was used across a range of catchment surfaces to examine patterns of contaminant storage and release.

Results and discussion: Lead concentrations varied greatly over a small spatial scale. Erosion is exposing high concentrations of Pb on interfluvial surfaces (up to $1660 \mu\text{g g}^{-1}$), and substantial amounts of reworked contaminated material (up to $1010 \mu\text{g g}^{-1}$) are stored on other catchment surfaces (gully walls and floors). A variety of factors have been shown to significantly control Pb release and storage in this environment, including wind action, aspect, and gully depth. Vegetation also plays an important role in retaining sediment-bound heavy metals within contaminated peat catchments.

Conclusions: This study provides the first comprehensive overview of the mechanisms controlling Pb release and storage in degraded peatlands. Previous assessments of Pb fluxes may have underestimated contaminant export from severely degraded systems. Wind has also been identified as an as yet unaccounted for vector for heavy metal transport in peatland environments.

Key words Blanket peat • Deposition • Erosion • Heavy metals • Vegetation • Wind

1 Introduction

The near-surface layer of peatlands in close proximity to urban and industrial areas can be contaminated with atmospherically deposited heavy metals (Vile et al. 1999; Rothwell et al. 2010a). As such, peatlands can represent significant sinks of anthropogenically derived heavy metals such as lead (Pb) (Shotyk et al. 2000; Farmer et al. 2005; Rothwell et al. 2007a). Many blanket peats in the UK are substantially degraded as a result of climate change, pollution, and mismanagement (Ferguson et al. 1978; Holden et al. 2007; Bonn et al. 2009). Consequently, there is concern that erosion is releasing substantial quantities of Pb contaminated sediment into the fluvial system (Yang et al. 2002; Shotbolt et al. 2006; Rothwell et al. 2007a, 2008b, 2010b), and that the removal of contaminated surface material is reducing the function of peatlands as a long-term sink for atmospherically deposited contaminants (Rothwell et al. 2008b).

Recent peatland Pb research has focussed on understanding the processes involved in the release of contaminated sediment. Rothwell et al. (2005, 2007a, 2008b) have shown that peat erosion is releasing substantial amounts of Pb into the fluvial system in both particulate and dissolved phases. Heavy metals can then be captured in impoundments on rivers such as reservoirs (Shotbolt et al. 2006) and floodplains (Chudaničová et al. 2016), leading to concerns over ecological and human health. Other studies have modelled Pb release under acidification and drought scenarios (e.g. Lucassen et al. 2002; Tipping et al. 2003). Further modelling (Rothwell et al. 2010b; Shuttleworth et al. 2015) and technical developments (e.g. portable XRF (pXRF), Shuttleworth et al. 2014) have led to an increased understanding of

the spatial and temporal patterns of Pb across upland peatlands in the UK.

Near-surface Pb storage has been shown to be highly heterogeneous due to spatial variability in atmospheric Pb deposition (Bindler et al. 2004; Farmer et al. 2005; Rothwell et al. 2007a). This can be further complicated by the erosion of surface material from exposed surfaces in degraded areas (Shuttleworth et al. 2015). This complexity presents a significant challenge when modelling catchment Pb fluxes. Rothwell et al. (2010b) derived a strong relationship between gully depth and sediment-associated lead concentrations, but Shuttleworth et al. (2015) found that this relationship broke down in areas of severely degraded peat where a higher than expected proportion of material derived from the contaminated surface was entering the fluvial system. Shuttleworth (unpublished data) also found that in some headwater catchments suspended sediment Pb concentrations exceed those stored on interfluvial surfaces, indicating that there may be substantial storage of Pb contaminated sediment elsewhere in the catchment. A deeper understanding of the mechanisms of Pb release and storage is therefore required to better quantify contaminant export from eroding peat systems.

Rothwell et al. (2007b) proposed that variability in the Pb content of fluvial sediments was likely to be due to differences in catchment erosion processes in degraded upland peatlands. A variety of controls on peatland erosion and sediment dynamics have previously been proposed (Table 1). Peat erosion is negligible below an established plant cover; bare peat, by contrast, is readily erodible (Bragg and Tallis 2001). An exposed peat surface is therefore a prerequisite for erosion to take place. Sediment 'preparation' is often cited as

an important control on sediment production and mobilisation on bare peat surfaces (e.g. Tallis 1973; Francis 1990; Labadz et al. 1991). Freshly exposed peat is fibrous, cohesive, and resistant to water erosion, while weathering produces a superficial friable layer on bare peat surfaces, which is readily mobilised and rapidly depleted (Evans and Warburton 2007). This loose sediment has a very low density which can be removed from bare peat surfaces by one of three key mechanisms: (i) through the action of running water (e.g. Labadz et al. 1991; Evans et al. 2006); (ii) wind (Tallis 1997; Warburton 2003; Foulds and Warburton 2007a, b); (iii) chemical oxidation (Francis 1990; Waddington and McNeil 2002; Evans and Warburton 2005). Evans and Warburton (2005) and Evans et al. (2006) provide a robust assessment of the full range of peat erosion processes from a single peatland site. However, these studies focussed on constructing sediment budgets and quantifying the catchment-scale export of organic sediment, and do not consider contaminants. To date, there has been no attempt to provide a comprehensive overview of the mechanisms which control Pb movement including its release and storage in degraded peatlands.

This paper employs the Pb contamination stored near the peat's surface as a fingerprint to trace contaminated sediment movement and storage in three severely degraded peat headwater catchments. Lead concentrations across different catchment surfaces (interfluves, gully walls, and gully floors) are quantified, and patterns in Pb concentration are investigated to identify the key controls on Pb-contaminated sediment dynamics.

2 Methods

2.1 Field area

The Bleaklow Plateau is an area of upland blanket peat that makes up part of the Peak District National Park (PDNP) in the southern Pennines, UK (Fig. 1a). The plateau lies between 500 and 633 m asl, and is situated between the cities of Manchester and Sheffield which were the heartlands of the English Industrial Revolution. Peat depths of up to 3 m (Evans and Lindsay 2010a) overlie a sandstone bedrock from the Millstone Grit Series (MGS) (Wolverson-Cope 1976) and fine grained head deposits of weathered MGS shales (Rothwell et al. 2005). Mean monthly temperatures vary between 13.2 °C (July) and 1.6 °C (February) (2004-2013), mean annual rainfall is 1313 mm (2004-2013), and the prevailing wind direction is WSW (254°) (Clay and Evans 2016). The PDNP supports a range of ecosystem services including tourism, food production, grouse moorland, drinking water and carbon sequestration (Bonn et al. 2009).

The peatlands of the PDNP are amongst the most degraded and contaminated in the world. Anthropogenic and climatic pressures have caused widespread erosion (Bower 1960a, 1960b, 1961; Tallis 1985; Bonn et al. 2009). Peak Pb concentrations of up to 1650 $\mu\text{g g}^{-1}$ can typically be found between 5 and 10 cm below the peat's surface, surface Pb concentrations in excess of 300 $\mu\text{g g}^{-1}$ are common, and Pb contamination is minimal below depths of 30 cm (Rothwell et al. 2007a, b). Bleaklow has been the focus of a multi-million pound restoration initiative (Shuttleworth et al. 2015), but this study concentrates on three headwater catchments in an actively eroding area of the plateau to the north of Bleaklow Head (Fig. 1b). This area has been purposefully left in its degraded state to act as a baseline for

comparison with restored areas. Consequently, the field site has been the focus of recent research into carbon release, pollutant dynamics, and peatland restoration (e.g. Clay et al. 2012; Dixon et al. 2014; Cole et al. 2014; Shuttleworth et al. 2014, 2015).

The three headwater catchments are typical of the area, with steep walled gullies with depths varying from around 1 m at the gully heads to 3-4 m at the gully mouths. Vegetation cover is sparse and bare peat is prevalent. Any vegetation present on interfluvial surfaces is composed of a mixture of low lying shrubs (*Calluna vulgaris*, *Erica tetralix*, *Vaccinium myrtillus*) and cotton grass (*Eriophorum spp.*) reflecting a mixture of the original pre-gully vegetation and some newly-established vegetation. Vegetation on gully walls and floors is dominated by cotton grass, which is thought to have established on these surfaces post-disturbance (Crowe et al. 2008). A small number of ericaceous shrubs are present on gully walls and appear to have originated at the peat's surface, but have been transported downslope during localised slope failure.

2.2 Field survey

Surface Pb concentrations were measured at 1 m intervals along four parallel transects (spaced approximately 10 m) running perpendicular to the three gullies (Fig. 1b). A total of 188 readings were taken using a handheld Niton XL3t 900 XRF analyser following the method outlined in Shuttleworth et al. (2014). The limit of detection (LOD) for Pb is 7 $\mu\text{g g}^{-1}$ (Shuttleworth et al. 2014). Where necessary, vegetation was gently removed and the peat's surface was lightly compacted by hand in order to present a smooth flat surface to the XRF

1 sensor (c.f. Ridings et al. 2000). There is no commercially available XRF Certified Reference
2 Material (CRM) for heavily contaminated peat so NCS DC73308 (Chinese stream sediment,
3 manufactured by NIST) was used as this has the most appropriate Pb concentration of the
4 CRMs available to the study. The relative percent difference (RPD) between the
5 concentration in the reference material and the concentration measured by the pXRF was
6 within 10 % for Pb. Samples from the top 1 – 1.5 cm of each sampling point were collected
7 using a stainless steel palette knife in order to determine the water content to correct for
8 the dilution effect of the high moisture content of the peat (c.f. Shuttleworth et al. 2014).
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24 **2.3 Data analysis**

25 ***2.3.1 Surface, catchment and vegetation effects***

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27 A general linear model (GLM) approach based on an analysis of variance (ANOVA) was
28 employed to determine the statistical significance of the influence of three factors
29 (catchment, surface type, and vegetation) and their interactions on Pb storage. Firstly, the
30 amount of contamination within each catchment may be influenced by heterogeneous
31 aerial deposition and storage (Bindler et al. 2004; Rothwell et al. 2007a) which would
32 restrict the amount of contaminated sediment available for redistribution on the different
33 surfaces. Secondly, Pb concentrations on the different catchment surfaces were compared
34 to investigate the relative amounts of contaminated sediment stored on each surface type.
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36 Finally, the presence or absence of vegetation was also included, as vegetation and
37 sediment dynamics are closely linked in blanket peats (Evans and Warburton 2005; Evans et
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al. 2006; Shuttleworth et al. 2015).

Initial investigation of the data showed that Pb storage on northwest (NW) facing gully walls was significantly different to Pb storage on southeast (SE) facing walls (2-tailed t-test, $p < 0.0001$) so these were included in the model as two separate surface types. Lead data within each factor were tested for normality (Anderson-Darling) and equality of variance (Levene); any factors that failed were square root transformed. Tukey's pairwise comparison was applied post hoc to assess where the significant differences lie. All relationships were tested at the 95 % level ($p \leq 0.05$). The magnitude of the effects of each significant factor and interaction were calculated using a generalised ω^2 (Olejnik and Algina 2003).

2.3.2 Directional trends

The nonparametric Spearman's rank correlation coefficient was employed to test for directional trends in the un-transformed data (Hollander and Wolfe 1973). Pb concentrations were tested against their corresponding easting value as a proxy for wind direction. The effect of gully depth on the pattern of Pb storage was assessed for both gully walls and gully floors. The pattern of Pb storage on gully walls was considered by investigating the relationship between Pb concentration and the depth of the sampling point below the interfluvial surface, similar to the method of Rothwell et al. (2010b). This analysis was carried out on individual sections of transect where there was a sampling frequency on gully walls of greater than five; in total five sections could be analysed. The pattern of Pb storage on gully floors was also considered. The gully depth map developed by

1 Evans and Lindsay (2010a) was not of a suitable resolution to derive accurate gully depth
2 values for the gully floor sampling points, so the relationship between Pb concentrations
3 and distance from the gully head was tested. This is based on the assumption that in
4 headwaters gully depth rapidly increases with distance from gully head (Evans and Lindsay
5 2010).

16 **3 Results**

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18 Figure 2 shows Pb storage on the different catchment surfaces, Table 2 summarises the Pb
19 concentrations which characterise each of the levels that make up the factors tested by the
20 GLM, and Table 3 summarises the results of the ANOVA. All surfaces store substantial
21 amounts of Pb contaminated sediment, but this storage is highly variable. Concentrations
22 across the field site range from below the limit of detection to 1660 $\mu\text{g g}^{-1}$, and Pb
23 contaminated and clean sediment can be found in all catchments and on all surface types.
24 Variation in Pb concentration is evident both between and within the catchments and
25 surface types; all bar one relative standard deviation (RSD) is in excess of 80 % (Table 2).
26 Interfluvial surfaces contain the highest Pb concentrations (mean 244 $\mu\text{g g}^{-1}$, max 1660 $\mu\text{g g}^{-1}$)
27 but there is also considerable Pb storage on gully floors and walls.

28
29 The total explanatory power of the ANOVA model is 27.6 % (Table 3). This reflects the
30 complex pattern of Pb storage displayed in Fig. 2, and is likely to be in part a product of the
31 interference of directional relationships. Despite this, surface type was found to be a
32 significant factor in the ANOVA, explaining 17.2 % of the variation in the data. Post-hoc

1 testing shows Pb storage on NW-facing walls is significantly lower than on SE-facing walls
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3 ($p=0.002$) and interfluve surfaces ($p < 0.001$), but no other significant differences were
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5 identified between surface types (Fig. 3b). Catchment is also significant, explaining 3 % of
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7 the variation in the data (Table 3); Pb storage in Catchment 1 is significantly lower than that
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9 of Catchment 3 ($p = 0.011$; Fig. 3a). The interaction between surface type and vegetation
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11 cover was also found to be significant, but vegetation cover alone was not (Table 3). Bare
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13 NW-facing walls contain significantly lower Pb concentrations than all other bare surfaces (p
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15 < 0.001), and vegetated interfluve surfaces ($p < 0.001$) and vegetated SE-facing walls ($p =$
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17 0.005). Vegetated gully floors also contain significantly lower Pb concentration than
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19 vegetated interfluve surfaces ($p = 0.008$).
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26 Although there is no statistically significant difference in Pb storage between bare and
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28 vegetated areas on individual surfaces, there are some interesting relationships which
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30 should be noted. Lead concentrations tend to be higher under vegetation on interfluve
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32 surfaces and on NW-facing gully walls; mean Pb storage on bare interfluve surfaces is 67 %
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34 of the mean vegetated value, while mean Pb storage on bare NW-facing walls is only 34 % of
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36 the mean concentration found under vegetation (Table 4; Fig. 3c). In contrast, Pb
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38 concentrations are substantially lower under vegetation on gully floors than on bare areas.
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40 Mean Pb storage on bare gully floors is almost 3 times higher than the mean vegetated
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42 concentration (Table 4; Fig. 3c). This indicates that vegetated and bare areas affect
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44 contaminant storage in different ways on the different surface types, and may explain why
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46 the GLM did not identify vegetation as a significant factor.
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When considering the relationship between Pb storage and gully depth, there is a significant negative relationship between Pb storage and distance from gully head on gully floors ($\rho = -0.359$, $p = 0.022$, 2 tailed) i.e. higher Pb concentrations are found near gully heads than gully mouths. Out of the five sections of gully wall transect, four did not show any relationship between Pb concentration and depth of sampling (all $p > 0.05$). The fifth section did produce a statistically significant result ($\rho = 1$); however, this was in the opposite direction than expected i.e. higher Pb concentrations were found at the base of the gully wall than near the top.

There was no correlation between easting (as a proxy for wind direction) and Pb concentration on interfluvial surfaces ($\rho = -0.116$, $p = 0.313$, 2 tailed).

4 Discussion

4.1 Patterns of contaminant storage

High concentrations of Pb were found in the study catchments. The highest Pb concentrations measured in this study ($1660 \mu\text{g g}^{-1}$) are comparable to Pb concentrations found in other studies in the southern Pennines (e.g. $1647 \mu\text{g g}^{-1}$, Rothwell et al. 2007a), and are similar to some of the highest values recorded elsewhere in the world (e.g. $1527 \mu\text{g g}^{-1}$ in Gola di Lago, Switzerland: Shotyk et al. 2000; $1650 \mu\text{g g}^{-1}$ in Fennoscandia tundra in Russia: Zhulidov et al. 1997). Lead storage varied between the three catchments with Catchment 1 containing significantly lower Pb concentrations than the other two catchments (Fig. 3a). This is to be expected, given that the near-surface record of Pb deposition in the southern

Pennines can vary by several orders of magnitude both horizontally and vertically over relatively short distances (Rothwell et al. 2007a). This variability could be due to a combination of factors including: differences in peat accumulation rates (e.g. Mighall et al. 2002), spatial heterogeneity in atmospheric deposition (e.g. Norton et al. 1997), spatial and temporal variation in plant community (e.g. Bindler et al. 2004), varying rates of decomposition (Biester et al. 2003) and removal of material by erosion (Shuttleworth et al. 2015).

There are some surprising similarities in the magnitude of Pb storage on the different catchment surfaces. Freshly exposed material on gully walls and floors should be relatively 'clean' compared to the contaminated near-surface material, but Pb concentrations on SE-facing gully walls and gully floors are similar to the levels found on the surface of interfluvies (Table 2) indicating that these are sites of substantial deposition and storage of reworked contaminated material. The maximum Pb concentrations found on these surfaces (555 and 1010 $\mu\text{g g}^{-1}$) exceed many previously reported maximum near-surface Pb concentrations found in peatlands around the world (e.g. 479 $\mu\text{g g}^{-1}$ in Bozi Dar, Czech Republic: Vile et al. 2000; 400 $\mu\text{g g}^{-1}$ in Lochnagar, Scotland: Yang et al. 2001). Rothwell et al. (2007b) found that re-deposited fluvial sediment (on floodplains, fans, and trash-lines) elsewhere in the Peak District contained 40 – 66 $\mu\text{g g}^{-1}$ Pb, but concentrations of these re-worked sediments were one or two orders of magnitude lower than those stored near the peat's surface ($\sim 1200 \mu\text{g g}^{-1}$). The high Pb concentrations recorded on gully floors and SE-facing gully walls in this study greatly exceed those recorded in reworked sediment by Rothwell et al. (2007b). NW-facing gully walls also store some Pb, but they are the 'cleanest' of the four surface types,

1 containing significantly lower Pb concentrations than the other three surfaces (Table 2). The
2 differences in Pb storage between NW- and SE-facing gully walls suggests that these
3 surfaces may be subject to different processes of deposition and removal.
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10 11 **4.2 Controls on contaminant storage**

12 The distribution of Pb within the catchments is clearly complex, but the statistical analyses
13 have highlighted several factors that influence contaminant storage including vegetation
14 cover, aspect, and depth of gully.
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25 **4.2.1 Vegetation cover**

26 The presence of vegetation has been shown to greatly influence sediment storage in
27 peatlands (Tallis and Yalden 1983; Evans and Warburton 2005; Evans et al. 2006;
28 Shuttleworth et al. 2015), and although vegetation cover alone was not found to control Pb
29 storage in this study (mean Pb storage on all bare and vegetated surfaces only differs by 9
30 $\mu\text{g g}^{-1}$ – Table 4), the interaction between surface type and vegetation cover is significant
31 (Table 3). The lowest Pb concentrations overall are found on bare NW-facing gully walls and
32 vegetated gully floors, while the highest concentrations are found on vegetated interfluvial
33 surfaces and bare gully floors (Fig. 3c; Table 4). Vegetation cover does not contribute to any
34 statistically significant differences in Pb storage on individual surface types. However, this is
35 likely due to the high variability in Pb values recorded on each surface (Table 2), and there
36 are some noticeable differences in mean Pb storage on bare and vegetated surfaces on
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three of the four surface types (Table 4; Fig. 3c). Higher mean Pb concentrations are found under vegetation than on bare areas on NW-facing gully walls and interfluvial surfaces, while the opposite relationship is evident on gully floors, where mean Pb storage is highest on bare areas (Table 4; Fig. 3c). This indicates that the presence or absence of vegetation does not have a consistent effect on Pb storage on the different surface types.

4.2.1.1 Interfluvial surfaces

The observed differences in Pb storage on vegetated and bare interfluvial surfaces in the study catchments are likely a product of differing rates of surface lowering. Shuttleworth et al. (2015) found that there is relatively little variation in Pb concentrations across the surface of intact areas of peatland ($290 - 400 \mu\text{g g}^{-1}$), while Pb concentrations on interfluvial surfaces in degraded areas range from 10's to 1000's $\mu\text{g g}^{-1}$. Exposed peat is highly susceptible to erosion (Tallis 1997; Evans and Warburton 2007) and Shuttleworth et al. (2015) propose that varying rates of surface erosion in degraded areas expose different stages of the Pb deposition profile at the peat's surface. Therefore, bare areas on interfluvial surfaces, being vulnerable to erosive processes, will have been stripped of some (and at some sampling sites, all) of the contaminated layer, resulting in surface Pb concentrations which range from below the pXRF limit of detection to $1050 \mu\text{g g}^{-1}$ (Table 2). Vegetation inhibits surface recession so any vegetation present on interfluvial surfaces will have protected the underlying peat from erosion, preserving the contaminated layer below.

4.2.1.2 Gully walls

In the Bleaklow area, the record of atmospheric Pb deposition is limited to the upper 30 cm of the peat profile (Rothwell et al. 2007a, b). When gully walls are cleared of superficial friable material, Pb concentrations are negligible (Shuttleworth et al. 2015), so any Pb-enriched material found on gully walls is interpreted to be reworked sediment derived from the near-surface contaminated layer. Lead storage is considerably higher under vegetation on NW-facing walls than on bare areas. This is similar to the observations of Pb storage on interfluvies, but the vegetation on these gully walls affects Pb storage differently as there is no in-situ contaminated surface to protect. Rather, vegetation intercepts contaminated peat particles which have been mobilised from the peat's surface as they move down the gully walls (Evans and Warburton 2005; Rothwell et al. 2010b) and then protect this redeposited sediment from subsequent erosion. Any contaminated sediment deposited on bare walls is vulnerable to weathering and is easily remobilised, leading to a relative enrichment of Pb contamination under gully wall vegetation.

No such relationship is evident on SE-facing gully walls where Pb concentrations are similar on bare and vegetated surfaces, providing further evidence that NW- and SE-facing gully walls may be subject to different processes of deposition and sediment removal.

4.2.1.3 Gully floors

Vegetated gully floors and floodplains have been shown to be important areas of sediment deposition in peatlands (Evans et al. 2005; Rothwell et al. 2007b), and gully floor vegetation

1 is often cited as pivotal in reducing connectivity between eroding surfaces and the fluvial
2 system (e.g. Evans and Warburton 2005; Crowe et al. 2008; Molina et al. 2009). It is
3 therefore surprising that mean Pb concentrations are three times higher on bare peat than
4 on vegetated surfaces, indicating greater storage of contaminated sediment on bare
5 surfaces. However, Evans and Warburton (2005) note that sediment is not efficiently
6 transported across vegetated alluvial fans, indicating sediment deposition may be limited to
7 the upstream extremity of vegetated surfaces. Thus, contaminated sediment may simply
8 not have reached the vegetated sampling locations on gully floors. Figure 4 shows freshly
9 deposited peat building up behind tussocks of *Eriophorum spp.* (cotton grass) on the floor of
10 Catchment 2, suggesting that vegetation on gully floors may be encouraging upstream
11 deposition in a similar manner to gully blocks which are used to restrict the flow of water
12 and sediment in peatland restoration initiatives (Evans et al. 2005). Bare areas of peat on
13 gully floors therefore represent significant deposition of reworked material derived from the
14 contaminated surface layer.

4.2.2 Aspect

15 Lead storage is significantly higher on SE-facing walls than on NW-facing walls (Fig. 2; Table
16 2), indicating that aspect plays an important role in determining the mechanisms behind
17 sediment dynamics on gully walls in eroding peatlands. The NW-facing gully walls will be
18 more prone to disturbance by frost heave and needle ice formation which would destroy
19 the structure of the surficial peat (Luoto and Seppälä 2000) producing a fluffy loose texture

1 that is easily dislodged and transported. Francis (1990) notes that frost heave preferentially
2 affects previously loosened peat, so any redeposited contaminated sediment on NW-facing
3 gully walls would have been particularly prone to frost action and easily removed, leading to
4 lower Pb storage. Birnie (1993) also reported maximum erosion on northerly aspects
5 suggesting that greater frost frequency was an important factor.
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13 Evans and Warburton (2007) cite running water as the dominant mechanism in mobilising
14 sediment on peatland gully walls. However, NW-facing gully walls in this study will also be
15 relatively more exposed to the prevailing wind than the SE-facing walls, which may also
16 contribute to the difference in Pb storage on these two surfaces. Aeolian processes have
17 received relatively little attention in the study of peatland erosion, but wind has been shown
18 to be a significant geomorphic process in peatlands in Finland (Seppälä 2001) and Canada
19 (Cummings and Pollard 1990), and Evans and Warburton (2007) note that wind-driven
20 volumetric losses from UK upland peatlands are comparable to agricultural soil losses. The
21 fact that maximum wind-driven UK peatland erosion has been shown to occur on SSW to
22 WNW aspects (Evans and Warburton 2007) would suggest that the differences in Pb storage
23 on the downwind facing gully walls could be driven by exposure to wind.
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45 **4.2.3 Gully depth**

46 Lead concentrations in gully floor sediments decrease with distance from the gully head,
47 supporting the concept that clean peat makes up a greater proportion of sediment with
48 distance downstream. As gullies deepen, relatively uncontaminated 'clean' peat represents
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1 a larger proportion of the exposed gully wall. The surface-derived Pb signal becomes
2 progressively diluted as contaminated and clean particulates mix (Rothwell et al. 2010b).
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5 Despite the evidence from gully floors that the proportion of clean to contaminated
6 sediment increases with gully depth, there is no relationship between gully depth and Pb
7 storage on gully walls. This is similar to the findings of Shuttleworth et al. (2015) which also
8 did not show any relationship between gully depth and Pb contaminated suspended
9 sediment. Rothwell et al. (2010b) derived the gully depth-sediment associated Pb
10 relationship in catchments where interfluvies were well vegetated, so any Pb would only
11 have been sourced from the contaminated layer exposed at the top of gully walls. In our
12 study catchments, no such relationship is evident due to the prevalence of bare interfluvie
13 surfaces, releasing large volumes of contaminated material (Shuttleworth et al. 2015),
14 masking any effect of an increasing pool of 'clean' peat as gullies deepen.
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35 **4.3 Implications for management and future research**

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37 Patterns of contaminant storage are consistent with sediment redistribution by both wind
38 and water erosion processes mediated by the presence of vegetation. This adds to the
39 growing body of evidence that re-vegetation is a key component of peatland restoration
40 schemes (e.g. Anderson et al. 2009; Worrall et al. 2011; Shuttleworth et al. 2015).
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49 The observation that wind erosion is a potentially significant vector for contaminated
50 sediment transport highlights a Pb fraction that is as yet unaccounted for in estimates of
51 peatland Pb budgets. Fine metal-laden airborne particulates have been shown to affect
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human respiratory health in urban environments (e.g. Fernandez Espinosa et al. 2002; Voutsas and Samara 2002), where sediment associated Pb contamination is not as severe as that found in the near-surface peats of the Peak District. Consequently, the airborne component of the Pb budget in eroded peatlands could also be important in toxicological terms, given the large number of people that visit the Peak District (in excess of 10 million tourist-days per year; Global Tourism Solutions 2009).

5 Conclusions

The legacy of atmospheric Pb deposition stored near the peat's surface has been successfully employed as a fingerprint to trace contaminated sediment movement and storage in degraded peat headwater catchments. High Pb concentrations are, as expected, associated with un-eroded peat surfaces which preserve the record of industrial pollution at the sites. High Pb concentrations are also observed in areas of sediment deposition indicating that legacy Pb pollution is mobile and reworked in this eroding peatland system. Previous assessments of Pb fluxes may have underestimated contaminant export from severely degraded systems such as these, and wind has also been identified as an unaccounted for vector for heavy metal transport in peatland environments. Vegetation plays a key role in protecting and trapping contaminated sediment, reinforcing the importance of re-vegetation in the restoration and management of these degraded systems.

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Figure captions

Fig. 1 Location of the study site. (a) The Bleaklow Plateau in relation to the cities of Manchester and Sheffield. The red star denotes the gullied field area, just north of the Bleaklow summit. The blue star denotes the location of the automatic weather station. (b) View down Catchment 2 (looking NE), showing transect markers running right to left across the photo. Transect markers are spaced at 2 m intervals

Fig. 2 (a) Schematic depicting mean lead concentrations measured along the four transects on the different catchment surface types (figure not to scale); (b) Interval plot (mean $\pm$ 95 % C.I.) of lead concentrations grouped by surface type

Fig. 3 Interval plots for factors and interactions which produced significant differences when comparing lead storage based on ANOVA depicting 95 % confidence intervals for the means. Note: the y-axis represents square-root transformed Pb values

Fig. 4 Freshly deposited peat accumulating behind tussocks of *Eriophorum* on the floor of Catchment 2

Figure 2
(a)

(b)

(a) Figure 3

(b)

(c)

Figure 4

[Click here to download Figure Fig4.jpg](#)

Table 1 Summary of selected controls on peatland sediment dynamics

Control	Mechanism	Effect	Study
Vegetation	Protects surface;	Reduces sediment	Evans and Warburton (2005);
	Traps mobilised	production;	Shuttleworth et al. (2015)
	sediment	Reduces sediment movement	
Weathering	Frost	Prepares' surface;	Tallis (1973); Francis (1990);
	heave/needle ice;	Produces readily	Labadz et al. (1991); Luoto
	Desiccation	mobilised sediment	and Sepälä (2000)
Erosion	Fluvial; Aeolian;	Mobilises sediment;	Holden and Burt (2002b);
	Mass movement	Deposits sediment	Foulds and Warburton (2007a, b); Warburton et al. (2004)
Degree of degradation	Surface removal;	Controls sediment	Shuttleworth et al. (2015);
	Gullyng	associated Pb concentrations	Rothwell et al. (2010b)

Table 2 Descriptive statistics for Pb each factor tested by the GLM. LOD = limit of detection

Factor	Level	<i>n</i>	Pb concentration ($\mu\text{g g}^{-1}$)			
			Mean	R.S.D.	max	min
Catchment	1	48	124	95.7	502	< LOD
	2	69	222	116	1660	< LOD
	3	71	213	91.5	1010	< LOD
Surface type	Interfluves	75	245	81.3	1660	< LOD
	NW-facing walls	43	80.5	121	382	< LOD
	SE-facing walls	37	207	60.4	555	< LOD
	Floors	33	209	119	1010	< LOD
Vegetation cover	Bare	137	191	103	1010	< LOD
	Vegetated	51	200	120	1660	< LOD

Table 3 ANOVA results for square-root transformed data. p = probability of factor being zero

and ω^2 = generalized proportion of variance explained. Significant results in bold

Source	p	ω^2
Catchment	0.015	0.033
Surface	0.000	0.172
Vegetation cover	0.642	0.000
Catchment*Surface	0.062	0.026
Catchment*Vegetation cover	0.227	0.002
Surface*Vegetation cover	0.003	0.043
Total		0.276

Table 4 Mean lead storage on bare and vegetated surfaces ($\mu\text{g g}^{-1}$)

	Bare	Vegetated
All Surfaces	191	200
Interfluves	223	338
NW-facing walls	51.2	156
SE-facing walls	215	192
Gully floors	281	99.4