



A 3300-year atmospheric metal contamination record from Raeburn Flow raised bog, south west Scotland



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ABSTRACT

There is now a plethora of records of atmospheric metal deposition across Europe based on total concentrations and calculated enrichment factors. However, to place such records into an archaeological context and to identify anthropogenic contamination signals more accurately, it is important to separate the signals derived from anthropogenic activities from those of a natural origin. This study presents a new 3300-year record from a bog in the vicinity of Hadrian's Wall and the Northern Pennine orefield in order to generate a new atmospheric metal deposition record for this archaeologically important part of the British Isles. For this purpose multi element geochemistry was undertaken to apportion the contribution of trace metals (lead, zinc, copper, arsenic and mercury) and sulphur as a result of mining/metallurgy and/or geogenic processes. To extract the different contributions through time we used total concentrations and enrichment factors (EF), and applied principal component analysis (PCA) to the dataset. The PCA extracted 7 components: at least two components are necessary to elucidate the trace metal distribution. Zinc, arsenic and lead are mostly related to atmospheric pollution, while mercury and copper appear to be more closely associated with organic matter. Based on these results four phases of lead contamination have been identified that date to: I, c. 2350–1500 cal BP; II, c. 1050–700 cal BP; III, c. 500–350 cal BP and IV, 250 cal BP–present. Copper enrichment also occurs during the Bronze Age (c. 3150–2800 cal BP). Peaks in other metals do not always correspond with lead and they may have been caused by other land use changes or processes that operate internally within the bog. Although the lead can be attributed to both anthropogenic and geogenic sources, its down profile pattern is in accordance with contamination records elsewhere in Britain and Europe, and the lead enrichment recorded at Raeburn Flow suggests that the Northern Pennine orefield was exploited for metals during the late Iron Age and Roman period.

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1. Introduction

Hadrian's Wall is one of the most iconic Roman archaeological features in Britain. The wall and its associated frontier has attracted a wealth of archaeological studies that have reconstructed all aspects of the social and economic livelihoods of people living along this frontier (e.g. Breeze and Dobson, 1987; Jones and Mattingly, 1993), as

well as environmental changes caused by activities such as agriculture (e.g. Dumayne and Barber, 1994; Manning et al., 1997). Lead mining is known to have taken place across Europe and in the British Isles throughout the Roman period, but hitherto archaeological evidence from the Northern Pennine orefield, which is closest to Hadrian's Wall and its hinterlands has been less forthcoming (e.g. Jones and Mattingly, 1993). In a previous study Mighall et al. (2004) found no evidence of lead enrichment during the Roman period in three peat records in Weardale and Teesdale and they suggested that any pre-medieval activity may only have been on a scale that was insufficient to generate a detectable contamination signal. This is

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surprising, given the relatively close proximity of the Northern Pennine orefield to Hadrian's Wall, which seems to be an obvious source of metals for the Romans to exploit along with other lead rich deposits found, for example, in SW Britain at Charterhouse in the Mendips, Alderley Edge in Cheshire and central-north Wales (Timberlake et al., in press). Such issues have wider significance in understanding the Roman's role in commanding and controlling the native economy (cf Manning et al., 1997) and therefore are worthy of further investigation.

The Romans are renowned for their exploitation of metals; especially lead which was used for a variety of purposes including aqueduct piping, plumbing, pewter, coffins and gutters for buildings, and for its association with silver, e.g. in coinage. Such was the demand for lead, Nriagu (1998) estimated that the peak in annual production during the Roman Empire was about 32,000 metric tons. This demand is also reflected in signals preserved in terrestrial archives such as peatlands and lake sediments. These archives have shown that the start of atmospheric metal contamination can be traced back to the Bronze Age, to as early as c. 2530 BC in Spain and c. 2400 BC in Britain (Martínez Cortizas et al., 2013). However, the most widespread early contamination phase appears to have commenced in the late Iron Age and peaked during the Roman period (Martínez Cortizas et al., 2013). Martínez Cortizas et al. (2013) describe this common narrative of historical contamination in more detail with higher concentrations of lead commencing around 1000–500 BC, peaking by the 1st–2nd centuries AD and collapsing by c. AD 400; a pattern that has been reconstructed by numerous researchers from peat bogs in Sweden, Britain, Germany, France and Switzerland (De Vleeschouwer et al., 2010). Indeed, Renberg et al. (2001) suggest that the chronology of this pattern is so consistent it can be used as a chronological marker.

The mineralized deposits of the Northern Pennine orefield are located primarily in the Weardale and Teesdale regions of County Durham, extending westward into the Alston Moor and Escarpment regions of Cumbria, northward into Northumberland and southward into North Yorkshire (<http://www.ukminingventures.com>). Despite the wealth of Roman archaeology to the north of this mineralised region, the first documented evidence of mining in the Northern Pennines dates to the 12th century AD, but there is some circumstantial evidence for earlier activity. A Roman date is preferred by Wooler (1924), Raistrick and Jennings (1965) and Lord and Morgan (2003) and it is suggested by others albeit tentatively (Jones and Mattingly, 1993), whilst Coggins (1986) speculates that pre-Roman activity may have taken place. Firm archaeological evidence for Roman exploitation is, however, still lacking.

The aim of this study is to reconstruct the atmospheric metal deposition history using a peat core from Raeburn Flow (SW Scotland), which lies 12 km from Hadrian's Wall and approximately 55 km from the Northern Pennine orefield in order to generate a new and more accurate record of atmospheric contamination history for this region. To achieve this aim we have analysed the geochemistry of the Raeburn Flow peat for the last c. 3300 years, with supporting radiocarbon dates. We applied multivariate statistical techniques (PCA) in order to separate potential signals derived from human activities, which could include both enhanced soil dust deposition and metal pollution, from those derived from natural processes, which could include naturally enhanced soil dust deposition and internal processes within the peat related potentially to diagenesis.

2. Materials and methods

2.1. Location, sampling and sub-sampling strategy

Raeburn Flow is a raised bog located in a special area of conservation (SAC) and a dedicated 'Site of Special Scientific Interest'

(SSSI) north of the Solway Firth in the Scottish Borders, Scotland, UK (Fig. 1). The site was previously studied with respect to ecological and climatic reconstructions using plant macrofossil and testate amoebae analyses (Mauquoy and Barber, 2002). Past peat cutting along the southern and eastern edges has lowered the water table and *Pinus sylvestris* and *Betula pubescens* have recently begun to establish across the bog surface. The raised bog phase spans at least the last 5000 years across parts of the bog (Mauquoy and Barber, 2002). A 360-cm long core (55° 02' 03.83" N and 03° 06' 27.41" W), consisting of *Sphagnum* peat, was retrieved in February 2011 using a Russian corer with a length and diameter of 50 × 7.5 cm respectively. Cores were wrapped in plastic cling film and protected in PVC tubing for transport. Additionally, rainwater was collected during the coring to determine the calcium/magnesium ratio at the site (cf Shoty, 1996). The cores were cut into 1 cm-thick slices in the laboratory and stored at 4 °C prior to analyses. All analyses were undertaken on single slices. Subsamples for total geochemical analysis were then freeze-dried and subsequently ground and homogenised.

2.2. Geochemical analyses

Fifty samples at 8-cm intervals were digested at Ecolab, Toulouse (France), following the protocol for total HF–HNO₃ digestion detailed in Le Roux and De Vleeschouwer (2011). Due to high organic content in a number of samples, an additional cleaning step using *aqua regia* was employed. Aliquots were measured by quadrupole ICP-MS at the *Géosciences Environnement Toulouse* laboratory (GET Observatoire Midi-Pyrénées, Toulouse) for a suite of 35 elements including lead, zinc, arsenic, copper, titanium, zirconium and sulphur. Five different certified reference materials (NJV 94-1 *Carex* energy peat, NJV 94-2 *Sphagnum* energy peat, NIST-1515 apple leaves, NIST-1547 peach leaves and NIMT/UOE/FM/001 ombrotrophic peat) were used to monitor the analytical quality of the measurements. All measured elements were above limits of detection. Accuracy was above 90% for all the measured elements excepted for titanium (75%). The measurements of two blank samples revealed values below limits of detection.

Mercury concentrations were determined using a direct mercury analyser at GET (Milestone[®]DMA-80). All samples were measured in duplicates. Certified Reference Material NIST-2685b was repeatedly measured throughout the whole measurement session. Results showed a high recovery rate of approximately 98% with an RSD of approximately 5%. Because the mercury analyser burns the sample at high temperature (850 °C), the ash content of each sample was also measured by weighing each sample before and after the mercury analyses.

2.3. Enrichment factors

We use enrichment factors (EF), calculated using the equation proposed by Shoty (1996), to identify periods of increased trace elements believed to have derived from human activities. As no clear background values for the pre-anthropogenic period can be derived with confidence, we use upper continental crust (UCC) values (Rudnick and Gao, 2003). Titanium (Ti) was used as the reference lithogenic element (as supported by the PCA results).

2.4. Chronology

An age-depth model was generated using ¹⁴C and ²¹⁰Pb radiometric dating techniques. Radiocarbon dates were determined at Chronolab, Belfast, using Accelerator Mass Spectrometry (AMS). Three samples of fresh peat were carefully cleaned to minimise the risk of contamination and *Sphagnum* macrofossils were handpicked

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