- **Physicochemical composition of wastes and co-located landscape designations at legacy**
- **mine sites in south west England and Wales: Implications on resource potential**
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Highlights (85 characters max)

Abstract

 This work examines the potential for resource recovery and/or remediation of metalliferous mine wastes in south west England and Wales. It does this through an assessment of the physicochemical composition of several key metalliferous legacy mine waste piles and an analysis of their co-location with cultural, geological and ecological designations. Solid samples were taken from 14 different sites and analysed for metal content, mineralogy, paste pH, particle size distribution, total organic carbon and total inorganic carbon. The majority of sites contain relatively high concentrations (in some cases up to several % by mass) of metals and metalloids, including Cu, Zn, As, Pb, Ag and Sn, many of which exceed ecological and/or human health risk guideline concentrations. However, the economic value of metals in the waste could be used to offset rehabilitation costs. Spatial analysis of all metalliferous mine sites in south west England and Wales found that around 70% are co-located with at least one cultural, geological and ecological designation. All 14 sites investigated are co-located with designations related to their mining activities, either due to their historical significance, rare species assemblages or geological characteristics. This demonstrates the need to consider the cultural and environmental impacts of rehabilitation and/or resource recovery on such sites. Further work is required to identify appropriate non-invasive methodologies to allow sites to be rehabilitated at minimal cost and disturbance.

1 Introduction

 There are few locations world-wide where historic metal mining is more evident than in mainland Britain. Extensive mining of major ores for metals such as copper, lead, tin and zinc at locations such as the Devon Great Consols in south west Devon and Parys Mountain in north west Wales fuelled profound global societal and industrial change (particularly during the Industrial Revolution) but as a consequence created a significant legacy of waste. Most mine sites in the UK were in peak operation in the 18th and 19th centuries and, as a result, mine sites were not subject to restoration practices which have been required in more recent years. In England and Wales alone, it has been estimated that there are over 8,000 disused metal mines located predominately in 12 ore producing regions (A. F. Jarvis 2007) (Palumbo-Roe 2010). Rather than simply rehabilitating such sites one option is to also recover any economically valuable metals that are present. Mine wastes and tailings are an obvious target for metals recovery as there are often significant quantities of such material in relatively easily accessible locations (i.e. above ground). To date, however, there is a paucity of studies that have characterised mine waste sites in terms of their metal content and extractability. This study presents the first effort to present these data for prominent legacy mine sites in England and Wales.

 Legacy mines also provide environmental or landscape 'resources'. This study also examines the resource potential of these legacy mine wastes in the context of site rehabilitation. Further to the potential recovery of economically valuable metals, there are often other drivers. For example, site remediation may: enable the land to be developed; enhance the conservation of industrial heritage and the related tourism features; and/or decrease the release of pollutants from the site into the surrounding environment. Similarly, there are also often a range of existing services that the mine sites provide which must be considered when implementing site remediation, including: cultural, scientific and educational features (such as historic industrial ruins); and rare fauna and flora. Thus it is important to appreciate the multifaceted value, both positive and negative, depending on perspective, that these sites currently have and would have if remediated. Within this a cost benefit approach must be applied to accurately assess to what extent the economic gain (that can be made through metal extraction) can offset the economic cost of such an intervention. This study thus considers multifaceted characterisation of value and resource through various lenses and the authors use the word "resource" in a wide sense (e.g. (Freeman 2014)) to cover both tangible resource of, for example, the metal/ore as well as functional and intangible resource stemming from the ecological, sociocultural and landscape value of the mine sites.

 In this work key geological, ecological and cultural designations (herein grouped under the umbrella of "environmental designations") co-located with the mines of the south west of England and Wales and, in particular, the case study legacy mine sites are presented as a means of assessing the potential consequences of the remediation of these sites. The specific aims of this paper are therefore to: (i) present data from the physicochemical characterisation of mine wastes from 9 major sites in the south west of England and 5 major sites from Wales; (ii) delineate the co-located environmental designations of the case study sites; (iii) appraise broader considerations of value and resource relevant to metal mine sites; and (iv) consider potential decision making tools to determine appropriate methodologies for optimising resource value. Very few studies currently exist which have applied this holistic approach to mine waste characterisation and to our knowledge this is the first time that the co-location of UK mine waste with geological, ecological and cultural designations has been examined.

2. Key drivers/deterrents for the reclamation of legacy mine waste

2.1. Environmental pollution

 A large number of historic metal mine sites world-wide are responsible for the release of metals and metalloids into surface and groundwater (Hudson-Edwards 2011) (Plumlee 2011). For example, a preliminary national assessment in 2009 revealed that as much as 6% of surface water bodies in England and Wales are currently adversely affected by pollution from historic metalliferous mines (Mayes, et al. 2009). In the UK ore extraction ceased at the majority of mine sites by the first half of the twentieth century, and as such ownership and/or legal liabilities for clean-up are often either unclear or orphaned (Palumbo-Roe 2010). This is also the case in many of the ore fields of North America (e.g. the USA and Canada have approximately 35,000 and 10,000 legacy metal mine sites respectively), Asia (e.g. Japan has approximately 5,500 legacy metal mines) and Europe (e.g. Sweden has approximately 1,000 legacy metal mines) (Mayes, et al. 2009). The financial cost of remediating and rehabilitating these mine wastes is significant. For example, in 2012 a series of joint reports commissioned by the Department for Environment, Food and Rural Affairs (DEFRA) and the Welsh Government in collaboration with the Environment Agency estimated that the total cost to remediate all of the water-related environmental problems associated with abandoned non-coal mines in the UK would be approximately £370 million, excluding operating costs, and take upwards of ten years (A. &. Jarvis, Prioritisation of Abandoned Non-Coal Mine Impacts on the Environment. The National Picture, Report SC030136/R2. 2012a) (A. &. Jarvis, Prioritisation of Abandoned NonCoal Mine Impacts on the Environment. Future Management of Abandoned Non-Coal Mine Water Discharges, Report SC030136/R12. 2012b). Moreover, the pollutant discharge from such sites often continues for many decades or even centuries, before water quality recovers to the pre-mining baseline. For example, despite ceasing major operations in 123 the late 18th century Parys Mountain in north Wales remains a major contributor of Cu and Zn to the Irish Sea, discharging an estimated 24 and 10 tonnes of each element respectively each year (Mullinger 2003).

2.2 Ecological resource

 The unique (and often extreme) physicochemical conditions and lack of disturbance has resulted in the development of a rich ecological resource on many different metalliferous mine wastes world-wide (Bradshaw 2000). For example, legacy mine sites often contain numerous species of rare metal-tolerant plants and lichens (Rodwell, et al. 2007), grasslands, wildflowers, orchids and important invertebrates, birds and mammals (e.g. the lesser horseshoe bat) (Barnatt and Penny 2004). In the UK this has resulted in specific recognition and protection for some mine waste sites. Examples include: the designation of Sites of Special Scientific Interest (SSSI) status for rare metal-tolerant plants, and lichens, and two priority habitats: Calaminarian grasslands (BRIG 2008) and Open Mosaic Habitats on Previously Developed Land (OMH) (BRIG 2010).

2.3. Geological and mineralogical resource

 The amount of metal produced at major UK mine sites has generally been relatively well recorded over the peak production years (i.e. during the Industrial Revolution), however, definitive figures for the quantity and type of waste produced are often lacking, with estimates typically calculated from predictions on the mineral to waste ratios, which are often highly variable, even for the same commodity (Palumbo-Roe 2010). To date a number of studies have 143 attempted to quantify the mass, distribution and composition of mine waste located at specific sites across the UK, however, a conclusive inventory is yet to be created due to the large number of mine waste sites and the inherent complexity of differentiating between the mine waste and the natural ground surface. As such a first estimate (e.g. to within an order of magnitude) for the mass and composition of mining waste present at many major legacy metal 148 mine sites in the UK has not yet been conducted with their associated economic value therefore unknown.

 Globally, historic ore beneficiation processes were typically less efficient than today and as such it is likely that appreciable concentrations of economically valuable metals were discarded as waste and are currently stored at legacy metal mine sites. Furthermore, the material has often already undergone size reduction during historic ore beneficiation and is often stored as unconsolidated material in relatively accessible locations (in piles above ground). Mine waste (in particular mine tailings waste) is also often of a relatively homogenous physical and chemical composition compared to other waste streams such as municipal solid waste. These extraction and processing activities have often resulted in the occurrence of rare and unusual geological, mineralogical or physiographical features deemed worthy of protection. Many mine wastes in the UK are therefore designated, for example, as Sites of Special Scientific Interest (SSSIs) because of these characteristics. Similarly, where relics demonstrate technological advancement of the mining industry they may also be designated as, for example, Scheduled Monuments.

2.4. Sociocultural resource

 The cultural heritage of many mine sites is considerable and the waste piles themselves are an intrinsically valuable component of this heritage landscape, i.e. in addition to remnant buildings and processing equipment (Howard, Kincey and Carey 2015). As such many landscape-scale historic mining districts have been granted official conservation status, for example the Cornwall and West Devon Mining Landscape World Heritage Sites as well as the numerous individual Scheduled Monuments and Listed Buildings that are associated with a rich legacy of mining. Physical features such as hushing scars; prospection pits and mine shafts; roads, tramways and leats linking the mines and settlements as well as the spoil tips themselves are regarded as valuable heritage (e.g. (Schlee 2007)). The ecological and cultural significance of mine wastes, coupled with their setting in the mine site and the wider landscape, provide a range of benefits to local people and visitors, with the former mine sites often being economically important for industrial heritage tourism (e.g. (Jones 2001)). These benefits can be framed as ecosystem or, perhaps more helpfully in this context, landscape services (Swanwick 2009). For example, prior to its World Heritage Site status being granted it was estimated that the mining attractions in Devon and Cornwall benefitted from nearly 1 million visitors each year, with around 2.5 million visitors to the region citing the mining heritage as an important consideration in their visit. This generates significant revenue to the local economy at an estimated £120 million per year (Atlantic Consultants 2003). Economic growth associated with mining heritage tourism has also been highlighted as a realistic development option in many economically marginal areas of Wales and there is active promotion led by the European Union for the maintenance of mining heritage e.g. the commercial Mining Heritage Network (Jones 2001) (Edwards 1996)).

 It is much more difficult to assign a monetary value to many of the other services provided by such sites which include recreation for local populations, cultural and spiritual enrichment, education and research (Bloodworth, Scott and McEvoy 2009) (Barnatt and Penny 2004) (Swanwick 2009). For example, local communities also often place an emotional value on mining landscapes (Ballesteros 2007). Many legacy mine sites also have educational and academic value and are often the subject of a diverse range of education and research in subjects from earth sciences, archaeology and engineering to social sciences and economic history. The cultural value of the sites is reflected by the wide number and type of stakeholders including archaeological and local history groups. However, the rural location of many mine wastes means that in addition to ecological and cultural resources arising from past mining activity there is likely to also be additional designations that may be adversely impacted on by pollution from the waste. Therefore it is crucial that the multifaceted nature of such sites and the landscapes in which they are located is understood.

3. Methodology

3.1 Site selection

 In England and Wales there are estimated to be over 3,000 legacy non-ferrous metal mines (A. F. Jarvis 2007) concentrated in three main ore producing regions: Cornwall and west Devon; Northumbria and north Humber; and Wales. The focus of this study is on the districts of Cornwall and west Devon and Wales because they both contain significant quantities of metalliferous legacy waste (Figure 1), representative of Cu/Sn and Pb/Zn mining areas, a high density of UK Mine Waste Directive sites (Palumbo-Roe 2010) and a range of cultural and environmental designations.

208 Cassiterite $(SnO₂)$, chalcopyrite $(Cu, FeS₂)$ and later arsenopyrite (Fe,AsS) bearing ore were principally extracted in Cornwall and west Devon and processed for Sn, Cu and As respectively. Chalcopyrite and galena (PbS) bearing ore was principally extracted in Wales and processed for Cu and Pb respectively. In many cases galena also contained relatively high Ag content, especially at Cwm Ystwyth, which was also extracted. Large quantities of sphalerite (Zn,FeS) was also mined, however, Zn was only occasionally removed and much remains in mine waste. The sites investigated which are located in south west England were: Alfred Consols (ALF), Caradon (CAR), Consols (CON), Devon Great Consols (DGC), Levant (LEV), Nangiles (NAN), Prince of Wales (PWM), Poldice (POL) and Wheal Maid (WHM). The sites investigated which are located in Wales were: Esgair Mwyn (EGM), Frongoch (FRG), Grogwynion (GRG), Parys Mountain (PYM) and Wemyss (WEM). Sites were selected which contain considerable mine waste volumes and are also located across different mining districts (as determined by different geographical and mineralogical constraints) of the region. Within

- south west England this was the Tamar valley and Tavistock (PWM, DGC), Caradon (CAR), Gwennap Kennal Vale and Perran Foundry (CON, NAN, POL, WHM), Port of Hayle (ALF) and St Just (LEV) mining districts. Within Wales this was the Central Wales (EGM, FRG,
- GRG, WEM) and Anglesey (PYM) mining districts.

 Figure 1. Location of metalliferous mines in south west England and Wales. Produced using BRITPITS database; Licence No. 2014/098BP ED British Geological Survey NERC. Boundary data from UK Data Service. URL: [http://census.edina.ac.uk.](http://census.edina.ac.uk/)

3.2 Sample collection procedure

 Mine waste samples were collected from each site following the methodology of ASTM D6009-12 (ASTM, Standard Guide for Sampling Waste Piles, D6009-12. 2012) which provides an appropriate method for the sampling of unconsolidated, aggregated waste piles. Many sites contained notable waste pile(s) of which the largest was typically targeted for characterisation (see Supplementary Data for sampling locations). Samples were collected using a stainless steel trowel at equal distances around the base of each mine waste pile at a depth of 0.2m. The sample depth of 0.2m was selected because it was determined as likely to represent a suitable compromise between sampling beneath the surface weathered zone whilst also exerting minimal aesthetic damage on the waste piles. Moreover, visual inspections in the field revealed the material, in almost all occasions, to be relatively homogenous with depth, i.e. no surface weathered zone could be identified.

 At most sites the mine waste is considered to be mine tailings (based on literature records and the relatively fine particle size observed). Each sample had a volume of approximately 5 L with a mass typically between 6 and 8 kg, depending on bulk density. Once collected the samples 245 were dried at 105° C for 24 hrs. The number of samples taken from each site was dictated by the pile volume, ranging from 3 samples taken for LEV to 21 samples taken from WHM (see Supplementary Data).

3.3 Sample and site characterisation procedures

 Composite samples were created for each site by riffling each sample 6 times and then mixing (using a mixing pad) each final aliquot together thoroughly. Each composite sample was then riffled to yield an appropriate mass for each analysis technique. Particle size distribution (PSD) measurements were performed via dry sieving and sedimentation (BS, Soil quality — Determination of particle size distribution in mineral soil material — Method by sieving and sedimentation. 2009) using 400 g from each composite. Uncompacted aggregate bulk density measurements were performed following BS 812: 1995 (BS, Testing aggregates of density. Part 2. Methods of determination. 1995). A cylinder of 1876 mL in volume was used and a tamping rod of 16 mm in diameter. Paste pH measurements were performed via ASTM D4972 – 13 (ASTM, Standard Test Method for pH of Soils 2013), using a 1:1 solid liquid ratio, i.e. 40 260 g from each composite and 40 mL of Milli-Q water (resistivity > 18.2 MQ cm). Samples were prepared for X-ray diffraction (XRD), inductively coupled plasma optical emission spectroscopy (ICP-OES), total organic carbon (TOC) analysis and total inorganic carbon (TIC) 263 analysis by crushing (to particle size <75 µm), using a Labtech Essa LM1-P puck mill crusher at 935 RPM for 120 seconds, a 200 g subsample of each composite sample. Each crushed sample was then prepared for XRD analysis by packing approximately 2 g of the material into an aluminium XRD stub. Analysis was performed using a Phillips Xpert Pro diffractometer 267 with a CuK α radiation source ($\lambda = 1.5406$ A; generator voltage of 40 keV; tube current of 30 268 mA). Spectra were acquired between 2 θ angles of 5–90°, with a step size of 0.02° and a 2 s dwell time. Each crushed composite sample was prepared for ICP-OES analysis via a 4 acid digest (EPA 1996). Firstly, 0.01 g was placed in a PTFE lined microwave digest cell and 3 mL of analytical grade 45.71% hydrofluoric acid (HF) was then added and left for 12 hrs. 6 mL of aqua regia solution (1:1 ratio of analytical grade 32% hydrochloric acid (HCl) and 70% nitric 273 acid (HNO₃)) was then added and the container was then placed in a microwave digest oven (Anton Paar Multiwave 3000) and heated at 200°C (1400 watts) for 30 minutes (after a 10 minute up ramp time period) and then allowed to cool for 15 minutes. The resultant solution 276 was then neutralised using 18 mL of analytical grade 4% Boric acid (H_3BO_3) at 150°C (900 watts) for 20 minutes (after a 5 minute up ramp time period) and then allowed to cool for 15 minutes. ICP-OES analysis was performed using a Perkin Elmer Optima 2100 DV ICP-OES. Total carbon (TC) measurements were performed using a Leco SC-144DR sulphur/carbon analyser. Samples of 0.35 g mass were loaded into the instrument and heated at 1350°C in a 281 pure O_2 (>99.9%) atmosphere. The concentration of CO_2 released by each sample was then measured using an infrared detection cell at a constant flow rate. Total inorganic carbon (TIC) measurements were performed using a Shimadzu SSM-5000A using 99.9% O2 at 500 mL/min and catalytically aided combustion oxidation performed at 900°C. Total organic carbon (TOC) was calculated by subtracting each TIC measurement from each samples corresponding TC measurement.

3.4 Hydrometallurgical extraction experiments

 Hydrometallurgical extraction experiments were conducted using a 1:10 solid-liquid ratio; 40 g of each composite sample and 400 mL of a 1M H2SO⁴ solution. Samples were sealed in 500 mL glass jars and constantly agitated at 200 RPM using a Stuart SSL1 orbital shaker table. Liquid samples for ICP-OES analysis were extracted from each batch system after 24 hrs and filtered using a 0.45 μm PTFE filter.

293 **3.5 Spatial analysis of mine locations, ecological and cultural designations**

 In addition to the analytical characterisation of the waste materials spatial analysis was undertaken to: i) understand the scale of past mining activity in the south west of England and Wales; and ii) examine the co-location of mine sites with areas protected for their geological, ecological or cultural benefits, particularly at the case study locations. The British Geological Survey BRITPITS database was used along with spatial data for the main geological, ecological and cultural designations [\(Table 1\)](#page-13-0) held by Natural England, Historic England and Natural Resources Wales. These designations were selected as they meet at least one of the following criteria: they are 'specified' ecological receptors under Part 2A of the Environmental Protection Act (1990) (DEFRA 2012), they are known or suspected to be co-located with past mining activity and there are spatial data available for them.

304 **Table 1 Ecological and cultural designations included in the study**

 The BRITPITS database details all known mine locations in Great Britain as point data categorised by the commodity (e.g. coal, Cu, Pb, gravel), type of mine (e.g. underground, open pit), status (e.g. active, ceased) geological age (e.g Carboniferous, Permian), lithostrat (e.g. Alluvium, West Maria Lode) as well as address and operator information. Co-ordinates are for the working or entrance to the mine (tolerance of 5 m) (Cameron 2012), not the location of the waste, but the assumption was made that all non-active mine sites have waste materials in their immediate vicinity. There are around 170,000 entries in the complete database, of which 4670 are non-active metalliferous mines in England, with 717 in the south west region and 3350 in Wales which are the focus of this study.

 The analysis was carried out in ArcMap 10.1. First, the BRITPITS data were limited to those mine sites with the metalliferous commodities (Sb, As, Cu, Ba, Au, Fe, Pb, Mn, Ag, Sn, Sr and Zn), those that were mine locations (as opposed to associated infrastructure such as rail depots and wharfs) and those that were non-active (ceased, inactive, dormant, historic; there were only two active metalliferous mines in the areas of interest). Where multiple commodities were mined BRITPITS contains duplicate records, one per commodity, so these records were merged.

 Next, the spatial joining function in ArcMap was used to identify which mine sites are co- located with the geological, ecological and cultural designations [\(Table 1\)](#page-13-0). Additional designations were also considered but no mine sites were co-located with these in SW England or Wales; these were Parks and Gardens, Battlefields and Nature Improvement Areas so these will not be discussed further. The split between geological and ecological, and cultural designations is arbitrary in some cases. Some designations have a clear basis in nature conservation (e.g. LNRs, SACs) or heritage (e.g. SMs, WHSs) whereas others are more nuanced. The decision was taken for cultural designations to include those where landscape and/or recreation as opposed to wildlife conservation is a primary objective (e.g. AONBs, National Parks) (Gaston, et al. 2006).

 Finally, this analysis was refined using the case study mine sites. The estimates of the spatial extent of the sampled spoil tips, drawn from aerial imagery, were used to gain further insight into the co-location with the designations. Polygons were drawn around an aerial view of the waste pile which has been sampled (see Supplementary Data for individual sampling locations) using the contrasting colour between the waste pile and the surrounding vegetation along with field observations as a guide. The specific designations at the site level were then examined more closely to identify which are dependent or independent on the mine waste as a way of exploring the opportunities and constraints for resource recovery. In addition, the case study sites were compared spatially to those on the inventory of Mine Waste Directive sites (Environment Agency 2014). These are known or are suspected to be causing a risk to water quality and/or human health and therefore likely to require remediation.

 To estimate the volume of waste in the case study locations polygons were used in conjunction with digital surface models produced using Light Detection and Radar (LiDAR). The data were at 1 m resolution with the exception of DGC and NYM where only a 2 m resolution was available. ArcMap was used to estimate the elevation of the land surface surrounding the waste material. This was estimated using at least ten points around the boundary of the polygon and the average elevation calculated. The polygon volume tool was then used to calculate the volume of waste above this elevation. This is a conservative estimate as the topography of sites was variable with many of the wastes being located on a slope. In addition, the presence of vegetation at some waste piles likely led to inaccurate readings due to it both shrouding the edge of the waste pile and also enabling greater elevations than the land surface to be recorded

353 in the LiDAR data. Figures 2 and 3 display location of case study mine sites in south west

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356 **Figure 2. Location of case study mine sites in south west England and their co-location** 357 **with statutory designations. Produced using BRITPITS database; Licence No.** 358 **2014/098BP ED British Geological Survey NERC. All rights reserved. AONB, SAC, SSSI** 359 **© Natural England copyright. Contains Ordnance Survey data © Crown copyright and** 360 **database right [2016]. SM, WHS © Historic England 2016. Contains Ordnance Survey**

361 **data © Crown copyright and database right 2016. Boundary data from UK Data Service**

362 **http://census.edina.ac.uk.**

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365 **Figure 3. Location of case study mine sites Wales and their co-location with statutory** 366 **designations and Landscapes of Historic Interest. Produced using BRITPITS database;** 367 **Licence No. 2014/098BP ED British Geological Survey NERC. All rights reserved. SAC,**

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4 Results and Discussion

4.1 Physicochemical characterisation of mine wastes

 Table 2 displays location, estimated volume, bulk density, total mass, paste pH and TOC data for mine waste taken from SW England and Wales respectively. The paste pH for all sites is recorded to be <7 and so cationic metal species are expected to be relatively mobile in the environment. TIC was recorded as 0.00% for all composite samples (except the EGM site where it was recorded as 0.04 wt.%) which indicates that the mine waste all have significantly 379 low carbonate alkalinity. The XRD patterns of the composite samples all indicate quartz (α - SiO₂) as the major crystalline component present with minor muscovite $(H_2KAI_3(SiO_4)_3)$ and 381 potassium feldspar $(K_5Na_5A1Si_3O_8)$ recorded for some samples (Appendix A). The original ore minerals arsenopyrite and chalcopyrite were not detected for any of the mine waste samples from SW England, and no Pb-bearing minerals, e.g. galena (PbS), were detected for the mine tailing samples from Wales. Particle size as a function of cumulative mass passing for all sites is shown in Appendix B. It can be noted that the PSD is relatively variable between sites, with a variation in 0.57-24.81 wt.% recorded for the silt and clay size fractions (particle size range <0.063 mm), upper and lower values for CAR and FRN respectively; a variation in 19.9-78.8 wt.% recorded for the sand size fractions (particle size range 0.063-2 mm), upper and lower values for LEV and DGC respectively; and a variation of 3.6-79.1 wt.% recorded for the gravel size fraction (particle size range 2-64 mm), upper and lower values for PWM and LEV respectively.

 As noted above, when estimating the volume of waste in each pile the average elevation from the area immediately surrounding the waste was used as a baseline, which has resulted in these estimates being conservative because much of the surrounding material is unlikely to be at the original elevation and the typography of some sites was extremely variable. For example, the volume of mine waste at DGC and GRG have been determined in other studies to be 274,250 (Mighanetara 2008) and 50,311 (Excal 1999) respectively compared to 198,923 and 9510 $m³$ here.

399 **Table 2. Location, volume, bulk density, total mass, paste pH, TOC and TIC data for mine waste. The location data refers to the location**

400 **where the first sample was taken from. * No LiDAR data; † mine waste is located in a valley floor so not possible to estimate volume using**

401 **LiDAR.**

402

 Table 3 displays metal concentration data for composite samples from each site. An indication is also provided of where values exceed various guideline concentrations developed to trigger risk assessments to protect human and ecological health. In general relatively high concentrations of As, Cu, Pb and Zn were determined for the sites located in SW England. For example, As concentrations were recorded as being greater than 0.1% for all sites (with the exception of ALF and NAN) with a maximum of 1.92% recorded for DGC. Relatively high concentrations of Cu and Sn were also recorded, with a maximum of 1.76 and 0.078% for CON and PWM respectively. Relatively high concentrations of Cu, Pb and Zn were recorded for samples taken from sites located in Wales. Particularly high concentrations of Pb were recorded for all sites, with a maximum of 4.67 wt.% recorded for FRG. Relatively high concentrations of Zn were also recorded with a maximum of 0.62 wt.% recorded for FRN. Moreover, a number of these metals and metalloids are determined to be exceeding guideline concentrations (some significantly) used to trigger risk assessments to protect human and ecological health. As was recorded to exceed human health guidelines for all sites sampled in SW England and PYM in Wales, whereas Pb was recorded to exceed both human and ecological health guidelines for all Welsh mine sites, and also CON, NAN and WHM for ecological risk. Cr, Cu, Zn and Cd were recorded as exceeding ecological guidelines for almost all sites, and Ni for a number of sites. As such it can be concluded that all sites comprise significant human health and ecological risks associated with toxic metal and metalloid concentrations.

 Although cut-off values are highly specific to the ore and mine setting, a survey of typical cut- off grades (percentage w/w) for a range of heavy metals indicates that Cu is economic at grades approximately >0.5%, Zn and Pb at >1% (Environment Agency 2012) and Ag at >0.02% (Douglas M. Smith 1982). A number of sites have yielded metal concentrations above this 426 threshold, namely: CON (Cu = 1.76%), LEV (Cu = 0.52%), PYM (Cu = 0.92%), EGM (Pb = 2.36%), FRN (4.67%) and GRG (1.30%). Metal concentrations (wt.%) for individual samples (which were used to create the composites) are displayed in the Supplementary Data. It can be noted that in general a relatively high variance was recorded between each sample, with a relative standard deviation (RSD) greater than 100% commonly recorded. This indicates that each mine waste pile is relatively heterogeneous. It can also be noted that there is a relatively close fit between the average of these data and the results for the composite sample, with a variance of <10% typically recorded for each metal. This demonstrates that the composite samples are a relatively good representation of the individual samples.

435 **Table 3. Notable metal and metalloid concentration data for composite samples from all sites where green cells indicate concentrations above screening**

levels for ecological risk¹ ; orange indicate those above guideline levels for human health risk2,3 436 **and red indicate those above both.**

437¹ Proposed Soil Screening Values under the framework for Ecological Risk Assessment (Environment Agency, Guidance on the use of soil screening values in ecological risk assessment. Science Report

438 SC070009/SR2b. 2008) NB these are not available for As; ² Category 4 Screening Values for public open space where there is considered to be a 'negligible tracking back of soil' (Defra 2014); ³ Soil Guideline

439 Value for Commercial land use (Environment Agency, Soil Guideline Values for Arsenic in soil. Science Report SC050021/Arsenic SGV. 2009a) (Environment Agency, Soil Guideline Values for cadmium in soil.

440 Science Report SC050021/Cadmium SGV. 2009b) (Environment Agency, Soil Guideline Values for Mercury in soil. Science Report SC050021/Mercury SGV. 2009c) (Environment Agency, Soil Guideline Values

441 for Nickel in soil. Science Report SC050021/Nickel SGV 2009d) (Environment Agency, Soil Guideline Values for Selenium in soil. Science Report SC050021/Selenium SGV. 2009e).

4.2 Mine waste resource value and hydrometallurgical extraction efficacy

 Key elements of economic value at each site (Cu, Zn, Ag, Sn and Pb) are shown in Table 4. This allows a first estimate of the total economic value for each key element at each site. It should be acknowledged, however, that this value could not be recovered in practice because of the limitations of mineral processing and the constraints imposed by the physicochemical properties of the material. Conversely when estimating the volume of waste in each pile the average elevation from the area immediately surrounding the waste was used as a baseline, which has resulted in these estimates being conservative because much of the surrounding material is unlikely to be at the original elevation and the typography of some sites was extremely variable. In addition as explained in Section 3.5 the single largest waste pile was sampled at each site. In many cases additional (but often minor) waste piles were observed at each site. These piles have not been accounted for both in terms of sample collection (see Supplementary Data for details) and total waste volume estimation using LiDAR. Moreover, the accurate sampling of large mine wastes piles is an intrinsically difficult exercise because the number of samples collected are limited by the resources and time available for any characterisation programme and the amount collected is never enough to fully characterise the waste pile (unless the entire waste pile is sampled and characterised). Also due to operational constraints relatively few samples were taken from each site (see Supplementary Data for details) and it is therefore almost certain that such samples do not entirely represent the overall mine waste pile. The results displayed in Table 4 should therefore be considered not as definitive but rather likely only to be accurate to the nearest order of magnitude.

 As an indicator of the ease of extraction using conventional hydrometallurgical processes the 464 recovery of metals in 1 M H_2SO_4 is also included (Table 5). The greatest Cu value is calculated 465 for the DGC mine $(\text{\textsterling}1,657,600)$ where reasonably high value of Sn $(\text{\textsterling}887,00)$ is also recorded. Relatively high value per tonne of Cu is also calculated for PYM (£32.15/tonne). Zn is not recorded in appreciable value (>£50,000) for any of the mine sites in the south west of England; however, relatively high value is estimated for a number of sites in Wales, including FRG (£1,989,000) and WEM (£104,700). Relatively low Ag value is recorded for all sites in the south west of England and a number of sites in Wales; however, relatively highly value is estimated for FRG (£552,400) and PYM (£11.99/tonne). Relatively low Pb value is estimated for all sites in the south west of England, whereas relatively high value is estimated for all sites in Wales, with maximum of £1,521,300 calculated for FRG. It can therefore be stated that the 474 deportment of value resides with Cu $>$ Sn $>$ Zn $>$ Pb $>$ Ag for the English study sites, whereas it is Pb>Ag>Zn>Cu>Sn for the Welsh study sites. When comparing these data to ore deposits (where economically valuable metals are typically present in much greater concentrations and total mass) it is unlikely that the mine wastes studied would be considered as suitable targets for resource recovery of the metals alone. However, the study has shown that the metal resource is present in quantities which are potentially sufficient to offset the costs of site remediation and rehabilitation. Furthermore the hydrometallurgical extraction data (Table 5) demonstrates 481 1M H₂SO₄ as able to solubilise Cu, Zn, As and Pb with reasonably high efficacy (often $>20\%$). In contrast Ag and Sn were determined as poorly soluble, with <5% dissolution recorded for 483 all mine wastes. Results therefore demonstrate that strong acids (such as $H₂SO₄$) could be successfully utilised (even at relatively low concentrations) for the significant removal of acid soluble metals such as Cu, Zn, As and Pb from UK mine wastes. Following subsequent recovery (e.g. via electrowinning) the value of such metals could then be utilised to offset a proportion of the remediation costs.

496

497 **Table 5. Percentage recovery of key elements in 1 M H2SO⁴ (200 RPM agitation speed,**

498 **1:10 solid-liquid ratio and 24 hrs reaction time).**

499

501 **4.3 Extent of mine sites in the south west of England and Wales and their association with**

502 **geological, ecological and cultural designations**

 This section focusses on the key considerations which are likely to impact the feasibility of implementing mine waste remediation and/or resource recovery processes. This considers the geological, ecological or cultural designations that are co-located in areas of mining and how they may act as constraints and opportunities for such interventions. This begins with an overview of the scale of this co-location in the south west of England and Wales followed by a more in-depth examination of the specific reasons for designation in the case studies areas.

509 There are 717 non-active metalliferous mines in the south west of England (Appendix C) and 510 3350 non-active metalliferous mines in Wales [\(Appendix D. Number of non-active mines by](#page-66-0) 511 [commodity in each type of geological, ecological and cultural designation in Wales](#page-66-0)

Commodit	Total number of mines in each designation															
y	To	L	${\bf N}$	S	S	S	\mathbf{A}	\mathbf{P}	\mathbf{O}	AO	\mathcal{C}	$\mathbf N$	S	W	L	More
	tal	N	N	S	\mathbf{A}	\mathbf{P}	W	H	M	NB	\mathbf{P}	\mathbf{P}	M	H	H	than $\overline{1}$
		$\mathbf R$	$\mathbf R$	SI	C	\mathbf{A}		1	H					S	$\bf I$	designa
																tion $(\%)$
Barytes	3	$\overline{0}$	$\overline{2}$	$\overline{2}$	$\overline{0}$	$\overline{0}$	$\overline{0}$	3	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\overline{2}$	2(67%)
Barytes	$\overline{4}$	$\overline{0}$	$\overline{0}$	θ	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\overline{4}$	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\overline{0}$	$\boldsymbol{0}$	$\overline{0}$	$\mathbf{1}$	1(25%)
Lead																
Copper	21	$\overline{2}$	$\mathbf{1}$	5	$\overline{4}$	$\overline{3}$	$\mathbf{1}$	$\overline{2}$	$\overline{0}$	6	$\overline{4}$	$\mathbf{1}$	$\mathbf{1}$	$\overline{0}$	$\mathbf{1}$	195
	3		3	8	$\mathbf{1}$		6	1				$\overline{2}$	$\overline{0}$		$\mathbf{1}$	(92%)
								2				$\overline{0}$			$\overline{7}$	
Gold	74	$\overline{0}$	$\overline{0}$	$\mathbf{1}$	9	3	$\overline{7}$	$\overline{7}$	$\overline{0}$	12	$\mathbf{1}$	6	$\overline{2}$	$\overline{0}$	5	74
				7				$\overline{0}$				$\overline{2}$			\overline{c}	(100%)
Gold	19	$\overline{0}$	Ω	θ	6	θ	$\overline{7}$	$\mathbf{1}$	θ	θ	θ	$\mathbf{1}$	θ	θ	$\mathbf{1}$	19
Copper								8				9			8	(100%)
Iron Ore	60	$\overline{0}$	$\overline{0}$	5	3	θ	9	5	θ	$\mathbf{1}$	$\overline{0}$	$\overline{0}$	$\mathbf{1}$	θ	$\overline{2}$	31
								9							$\overline{0}$	(52%)
Ironstone	17	5	$\overline{0}$	1	8	5	$\mathbf{1}$	$\mathbf{1}$	22	$\overline{2}$	$\overline{4}$	$\mathbf{1}$	θ	9	$\overline{7}$	100
	8			θ			$\overline{0}$	$\overline{7}$				9			5	(56%)
								7								
Lead	18	$\overline{0}$	5	$\overline{4}$	$\overline{2}$	$\overline{2}$	$\mathbf{1}$	$\mathbf{1}$	23	375	7	$\overline{2}$	$\overline{2}$	Ω	$\overline{4}$	1199
	47			$\overline{2}$	6	9	6	7				6	7		7	(65%)
				5	9		θ	9				θ			$\overline{4}$	
								1								

[Priority habitat data in Wales is presented as the area within a 1.6 km \(1 mile\)](#page-66-0) grid square so the location of the mine cannot be said to be [accurately co-located with the habitats. SSSI=Site of Special Scientific Interest, AW=Ancient Woodland, PH=Priority Habitat PH,](#page-66-0) [OMH=Open Mosaic Habitat on Previously Developed Land, SAC=Special Area of Conservation \(European\), SPA=Special Protection Area,](#page-66-0) [AONB=Area of Outstanding Natural Beauty, NP=National Park, CP=Country Park, SM=Scheduled Monument, WHS=World Heritage Site,](#page-66-0) [Landscape of Heritage Interest; Sources: mine data from BRITPITS Licence No. 2014/098BP ED British Geological Survey © NERC. All](#page-66-0) 517 rights reserved. All other data © Natural Resources Wales copyright. Contains Ordnance Survey data © Crown copyright and database right [2016.](#page-66-0)D). These are predominantly located in Sn and Cu mining areas of Cornwall (n=456), the As and Cu areas of Devon (n=49), the Pb and Zn areas of North East and Central Wales (n=2470), the Cu, Zn, Pb and Fe areas of North West Wales (n=819), and the ironstone regions of South Wales (n=45) and Gloucestershire (n=102). The majority of mine sites in SW England (68%) and Wales (72%) are co-located with at least one designation.

 There are mines located on many of the designated sites in both SW England and Wales. However, numbers are generally small for ecological or geological designations compared with the total number of designated sites in the region (Table 6). Despite this, in some cases the proportion of the area of such designations that are co-located with mines is much greater due to a few very large sites such as, for example, Exmoor Heath (10,000 ha), Plymouth Sounds (6,000 ha) and Dorset Heath (5,000 ha) SACs and Dorset Heathlands SPA (8,000 ha). Similarly, in Wales, despite only 8% of SSSIs being co-located with mines they account for 71% of the area of SSSIs, this is due to four very large sites of over 19,000 ha: Berwyn, Elenydd, Eryri and Migneint-Arenig-Dduallt (which are also SACs and/or SPAs). In Wales, there is a disparity between this effect for the SSSIs and European sites where a relatively modest proportion of area are co-located with mine sites due to the inclusion of far larger areas of coastal sites (e.g. Liverpool Bay, Cardigan Bay). It is not possible to discern from this overview whether the SSSI sites are designated for their geology or ecology or whether the LNRs, NNRs, SSSIs, SACs and SPAs are designated due to species and habitats that are intrinsically linked to the mining activities or whether they are coincidental to it. In the examples highlighted here the designations are not specifically linked to the presence of mine wastes. This is important as resource recovery could have a positive or a negative impact depending on the reasons for designation and this will be discussed in Section 4.4.

 Regarding priority habitats co-located with the mine waste it is possible in some circumstances to discern whether these are intrinsically linked to the presence of the mine waste. In SW England the largest number of mines are co-located with priority habitats other than OMH which are unlikely to be dependent on the characteristics of the mine waste and may even be negatively impacted by it. Although these habitats do not receive statutory protection local authorities are expected to consider their protection and enhancement in local planning policies. Resource recovery might therefore offer an opportunity for these habitats to be restored or enhanced if combined with remediation. Overall, 13 priority habitats are co-located with mine sites in SW England, but in terms of area this only accounts for 0.6% of priority habitats. The greatest number of mines were located on deciduous woodland (n=215), with less than 15 on the other 12 types. In Wales around 7% of priority habitats are co-located with mines. However, in Wales, with the exception of OMH, the priority habitat data is not represented as polygons but as 1.6 km grid squares indicating the presence of the habitat, each grid square then details the area covered by the habitat within this but the exact boundaries are not available. This means that mine sites appear to be co-located with several habitats and this has inflated the proportion of habitats co-located with mines. There were 3741 priority habitats co-located with almost all of the metalliferous mines in Wales (n=3258). As with SW England, the greatest number in Wales were on Broadleaved Woodland (n=2517). There were also substantial numbers co-located with Lowland Dry Acid Grassland (n=1608), Lowland Dry Heathland (n=1294) and Purple Moorgrass and Rush (n=1127). In both SW England and Wales a greater proportion of mine sites are co-located with OMH at 4% and 7%, respectively. This is not surprising given that this priority habitat is explicitly focussed on brownfield and previously developed sites, including mine wastes, and was in part based on an analysis of the BRITPITS data (Lush, Kirby and Shephard 2013). These sites are much more likely to be adversely affected by any resource recovery as they have developed over time due to the edaphic conditions on site so an alteration of these may change the species assemblages present.

 A far greater number of mines are co-located with areas of cultural significance representing both the rural landscapes together with the mining history of SW England and Wales. It is often impossible to disentangle the role of mining in some of the cultural designations. For example although AONBs and National Parks are not necessarily recognised for their mining activity *per se*, they are representative of the landscape character and cultural history of an area (e.g. mining is specifically mentioned in Cornwall and Tamar Valley AONB; Cornwall and Tamar AONB, 2015). The cultural designations generally operate at the landscape scale hence the large proportion of area co-located with mines for AONBs, National Parks and the World Heritage Sites (Table 6) demonstrating the ubiquity of mining in the heritage in these areas. There are two World Heritage Sites associated specifically with the mining heritage: the Blaenavon Industrial Landscape, which is recognised for the coal and ironstone mining activity and associated industries in south Wales; this makes up the vast majority of area of WHS in Wales (other two are castles and an aqueduct) and the Cornwall and West Devon Mining Landscape in SW England. Similarly, many of the Welsh mines are in the landscapes of historic interest designated by Natural Resources Wales.

 This spatial analysis demonstrates the significance of the mining legacy in SW England and Wales and its complex interaction with geological, ecological and cultural designations. It also illustrates that the decision as to whether to recover resources from former mine sites is likely to be dependent on a range of factors outside of the economic viability of such an endeavour and that these can only be determined at the site level.

587 **Table 6. Total number and area of designations in the south west of England and Wales,**

588 **those co-located with mine sites and the number of metalliferous mine sites in each**

589 **designation**

590 ¹ Caution should be used when using these figures as not all mines are represented in BRITPITS and the point locations are not necessary in 591 the same location as mine wastes; ² Refers to broad habitats as opposed to individual sites; ^a Includes a small portion of New Forest; ⁴Priority 592 habitat data in Wales is presented as the area within a 1.6 km (1 mile) grid square so the location of the mine cannot be said to be accurately 593 co-located with the habitats. Sources: mine data from BRITPITS Licence No. 2014/098BP ED British Geological Survey © NERC. All rights 594 reserved. Local Nature Reserve (LNR), National Nature Reserve (NNR), Site of Special Scientific Interest (SSSI), Special Area of 595 Conservation (SAC), Special Protection Area (SPA), Ancient Woodland (AW), Priority Habitat (PH), Open Mosaic Habitat on Previously 596 Developed Land (OMH), Area of Outstanding Natural Beauty (AONB) and National Park data for England © Natural England copyright. 597 Contains Ordnance Survey data © Crown copyright and database right 2016. Scheduled Monument and World Heritage Site data for England 598 © Historic England 2016. Contains Ordnance Survey data © Crown copyright and database right 2016. The Historic England GIS Data 599 contained in this material was obtained on 29th June 2015. The most publicly available up to date Historic England GIS Data can be obtained 600 from HistoricEngland.org.uk. All other data © Natural Resources Wales copyright. Contains Ordnance Survey data © Crown copyright and 601 database right 2016.

602 **4.4 Geological, ecological and cultural considerations at the case study sites: opportunities** 603 **and constraints to resource recovery and reclamation**

 All of the case study sites have some form of recognition either for their potential or known geological, ecological or cultural resources (Table 7). These can provide an opportunity for resource recovery or a constraint against it. For example, if mine waste is negatively impacting on ecological or cultural receptors that are not dependent on the characteristics of the mine waste then this could provide a powerful argument for resource recovery, decontamination and/or recovery of land value resource. However, some mine wastes have rare geological or ecological features or are valued for their cultural heritage and these could act as a constraint to resource recovery if the existence of these features were to be adversely affected by such activities.

 Taking potential constraints first, several of the sites are co-located with ecological designations that are directly related to the presence of mine wastes. In SW England CAR and POL are protected as SSSIs for their metallophytic bryophytes (liverworts and mosses) (Natural England 1999a). Bryophytes are adapted to Cu-rich substrates and include a number of internationally and nationally rare species, including one, *Cephaloziella integerrima,* which has only been recorded at two other sites since 1950 (Natural England 1999b). CAR is also designated as a SAC for its Calaminarian grasslands of the *Violetalia calaminariae* (JNCC 2015), recognised as one of the best in the UK and, globally, is one of only two known sites of the Cornish path-moss *Ditrichum cornubicum*, which is protected under the Wildlife and Countryside Act (1981) (Natural England 1999a).

 In Wales too, GRG and PYM are co-located with SSSIs, also designated for their bryophyte communities (Natural Resources Wales 2004) (Carmarthenshire County Council n.d.a) (Carmarthenshire County Council n.d.b) (Countryside Council for Wales 1995) (Countryside Council for Wales 1999). In addition, GRG is co-located with a SAC for its unique assemblage of metallophyte lichens (Calaminarian grasslands of the *Violetalia calaminariae*), one of which, *Epigloea filifera* has not been reported anywhere else in Britain (Natural Resources Wales 2004). The SSSI at PYM has over 125 lichen species and includes a *Lecidea* which is unique in Britain and possibly a new species (Countryside Council for Wales 1995).

 The designation at GRG is also both a SAC and SSSI for its alluvial shingle deposits which are associated with a mosaic of habitats including heathland communities not usually found in England or Wales. These support nationally scare species of beetle and otters the latter of which are protected under the Wildlife and Countryside Act (1981) and European Council Directive 92/43/EEC on the conservation of Natural Habitats and of Wild Fauna and Flora (Countryside Council for Wales 1999) (Natural Resources Wales 2004).

 The SSSIs at GRG and PYM, together with that at FRG are also designated for their geological characteristics. This includes mineralisations of the waste at FRG and PYM which are unique to Britain (Countryside Council for Wales 1995) (Countryside Council for Wales 1999). At GRG the fluvial geomorphology is characterised by an actively braiding river system which may be linked to the mining activity (Countryside Council for Wales 1999).

 In addition, six of the case study sites have been identified as potential OMH sites (ALF, CON, DGC, NAN, POL and WHM) although three of these, CON, POL and WHM, fall on the same OMH. The inclusion of OMH requires some caution as the initial inventory for these sites has been predominantly based on an analysis of previous land uses (e.g. BRITPITS and the National Land Use Database for Previously Developed Land) and aerial imagery. Therefore an ecological survey would need to be carried out to ascertain the presence of an OMH (Lush, Kirby and Shephard 2013).

 These designations have the potential to act as a significant constraint to resource recovery, specifically the management plan for one SSSI highlights that "care must be taken during preservation or derelict land operations to safeguard the specialised conditions the plants require" (Natural England 1999b). This means that any activities that changed either the physical or chemical characteristics of the waste are likely to be met with opposition. Many of the species are dependent directly on the elevated metal concentrations in the spoils (Batty 2005) or tolerant of them. The removal of the metals would reduce the toxicity of the spoils to

 other vegetation types which could then colonise the spoils potentially to the detriment of these rare species.

 Turning to the historic environment designations, all of the case studies in SW England, except ALF, fall within the Cornwall and West Devon Mining Landscape World Heritage Site. This World Heritage Site was designated in 2006 in recognition of the "contribution the area made to the industrial revolution and formative changes in mining practices around the world" (UNESCO 2006, 155). The designation also specifically recognises the significant ecological resources linked to this mining activity in the "distinctive plant communities of waste and spoil heaps and estuarine areas" (UNESCO 2006, 155). In addition there are numerous Listed Buildings (not discussed here) and Scheduled Monuments that are individually protected for their contribution to the mining landscape. Two sites, DGC and PWM are co-located with Scheduled Monuments whilst CAR is adjacent to one. These are protected for various built features including transport infrastructure, mine shafts, pumping engine houses and processing infrastructure (Historic England 2002) (Historic England 2006) (Historic England 2002). Interestingly the Prince of Wales Mine at Harrowbarrow Scheduled Monument specifically recognises the importance of the mine wastes as a record of the technologies in use at the time and as landmarks (Historic England 2006).

 None of the case study sites in Wales are in the Blaenavon Industrial Landscape World Heritage Site. However, all of them fall in one of three landscapes of historic interest (Table 7). All are recognised for their land management activities including agriculture and forestry but have a strong association with past mining (Dyfed Archaeology n.d.a) (Dyfed Archaeology n.d.b) (Cadw, Welsh Assembly Government, Countryside Council for Wales 2007). Although not receiving of a legal protection these landscapes are protected under planning policy from development that might have an adverse impact on their character (Welsh Government 2016) para.6.5.25. In addition there are several Scheduled Monuments associated with mining activity on the FRG and PYM sites (RCAHMW 2008) (RCAHMW 2000) (RCAHMW 2004) as well as many individual aspects of the mining infrastructure including the sublimation chambers and kilns at PYM (RCAHMW 2007).

 As already mentioned the mining landscapes have the potential to provide substantial economic benefits. Prior to its WHS designation the Devon and Cornwall mining landscape generated significant tourism industry and associated revenue to the local economy (Atlantic Consultants 2003), given that designations can play an important role in tourists choice to visit an area (Reinus and Fredman 2007) (Selman 2009) and the increase in heritage tourism in recent decades (Williams and Shaw 2009) this is likely to have increased since the designation.

 In terms of cultural designations not dependent on the mining activity none of the case study mines fell in the National Parks of SW England and Wales or AONBs in Wales despite the large land areas occupied by these designations. However, in SW England two case study sites are in AONBs: LEV and DGC. AONBs are designated in recognition of the area's landscape character, historic and natural environments. So although they are not specifically dependent on the mining legacy both the Tamar Valley and Cornwall AONBs recognise the significance of the mining heritage within their wider landscape (Cornwall AONB 2011, Tamar Valley AONB 2014) but would also be protective of contamination impacting on the natural environment.

 The value placed on heritage features is not straightforward. Whilst cultural aspects are valued by the public (Swanwick 2009) (Howley 2011), landscapes perceived as 'natural' or 'unspoilt' are often preferred (Swanwick 2009). The value of heritage features is subject to temporal changes, with features becoming increasingly important over time (English Heritage 2008). Landscape quality is inherently subjective and different groups have different preferences (Swanwick 2009). Although designations such as AONBs and National Parks in SW England and Wales explicitly recognise the contribution of the mining heritage to the overall landscape the individual features including wastes can also be perceived to have a detrimental impact on the quality of landscape (English Heritage 2008). Conversely, inappropriate restoration can also do more harm than good from both a nature conservation and landscape perspective. The Cornwall AONB has been estimated to generate bring in 4.5 million visitors to Cornwall estimated to spend £1.5 billion (Cornwall AONB 2011). Therefore any activities on mine sites need to balance the potential negative impacts on these designations. Resource recovery may fall under mineral planning, permission for which takes into account whether planned activities will have adverse effects on ecological systems, historic environments and human health (DCLG 2012) (Welsh Government 2016). Therefore the co-location of many waste sites with designated areas that may be detrimentally affected by resource recovery is a significant constraint.

 Turning to the potential opportunities for resource recovery to enhance or restore the ecological or cultural resources none of the case study mines in are co-located with sites protected for their geological or ecological characteristics not related to their mining legacy. However, DGC is adjacent to an ancient woodland; Clitters Wood. Several of the sites are co-located with or adjacent to priority habitats: ALF, WHM and NAN with lowland heathland, and WHM and PWM with deciduous woodland. In Wales all sites are co-located with at least three priority habitats (Table 6), but as already discussed these habitats overlap in the data so a more detailed assessment would be required. Ecological surveying and risk assessment would be necessary to determine whether priority habitats are affected by the mine sites. These habitats do not receive statutory protection *per se* but they are protected under planning policy (DCLG 2012) (Welsh Government 2016). As Table 3 demonstrates all of the case study sites have wastes with concentrations, particularly Cd, Cu and Zn, that may pose a risk to specified ecological receptors (e.g. SSSIs, SPAs, SACs, AONBs, National Parks), and this is likely to be the case across many of the abandoned mine wastes in the UK. They may also be impacting on aquatic ecology through mine water discharges (Mayes, et al. 2009), several appear on the Mine Waste Directive inventory (Table 7), or other designated terrestrial ecological receptors not co-located with the mine waste through the mobilisation of pollutants in water or food-chain transfer. The potential risk to ecological receptors is likely to add weight to the case for remediation and therefore act as an opportunity for resource recovery as a means of remediating the waste.

 It is clear from this study that there is substantial variation between mine wastes in terms of their characteristics and the context in which they are situated. A multitude of different perspectives will need to be sought when considering their long term management and whether resource recovery is appropriate. This will need to balance the requirements of a range of stakeholders and disciplines including environmental scientists, heritage professionals, ecologists and representatives from the different management bodies and regulators associated with these designations (Selman 2009). It should also be recognised that land managers, experts and the general public may have very different preferences in terms of the future of such sites and these views will also need to be considered (Bloodworth, Scott and McEvoy 2009) (English Heritage 2008) (Howard, Kincey and Carey 2015) (Selman 2009) (Swanwick 2009). Human Ecology Mapping (HEM) approaches offer promising spatial data gathering and analytical tools that may enable the views of multiple stakeholders to be considered (McLain 2013). These methods, particularly "sense of place" (see (D. R. Williams 1998)) might be useful in examining the resources and values of metalliferous mine sites integrating a spatial dimension with the human-landscape connection. Ultimately, the decision to recover resources from mine wastes needs to balance the potential negative impacts on geological, ecological and cultural designations with any positive impacts on those not explicitly dependent on the mining heritage.

778 **Table 7 Ecological and cultural designations co-located with the case study mine wastes in the south west of England and Wales**

^a Estimated total value of Cu, Zn, Ag, Sn and Pb at each site; Sources: mine data from BRITPITS Licence No. 2014/098BP ED British Geological Survey © NERC. All rights reserved. Mine Waste Directive (MWD) inventory data © Environment Agency copyright. Contains Ordnance Survey data © Crown copyright and database right 2016; Local Nature Reserve (LNR), National Nature Reserve (NNR), Site of Special Scientific Interest (SSSI), Special Area of Conservation (SAC), Special Protection Area (SPA), Ancient Woodland (AW), Priority Habitat (PH), Open Mosaic Habitat on Previously Developed Land (OMH), Area of Outstanding Natural Beauty (AONB) and National Park data for England © Natural England copyright. Contains Ordnance Survey data © Crown copyright and database right 2016. Scheduled Monument and World Heritage Site data for England © Historic England 2016. Contains Ordnance Survey data © Crown copyright and database right 2016. The Historic England GIS Data 786 contained in this material was obtained on 29th June 2015. The most publicly available up to date Historic England GIS Data can be obtained from HistoricEngland.org.uk; All other data © Natural Resources Wales copyright. Contains Ordnance Survey data © Crown copyright and database right 2016.

5. Decision making tools and technology options for intervention

5.1. Decision making tools for optimising resource value

 As discussed above legacy metalliferous mining waste sites have multifaceted value and resource associated with them. This results in the selection of the strategy for optimising resource value being a non-trivial problem and requires the consideration of a number of competing criteria to allow identification of appropriate approaches. In similar multi-criteria problems various decision support frameworks have been developed, many being based on Multi Criteria Decision Analysis (Q. Wang 2014), it is proposed that such an approached can be adopted here.

 In many environmental problems the criteria considered are classified within a sustainability assessment framework under three areas or pillars, namely: economic, environmental and social issue (Pettit 2011). However, for the problem considered here it is necessary to also consider the technical aspects of resource recovery from wastes. In the proposed approach three MCDA methods are adopted: simple ranking method, Analytic Hierarchy Process (AHP) and Compromise Programming, this allows either the individual use of one or the sequential use of all to allow sensitivity analysis to be undertaken (Pettit 2011). Typical criteria that can be used are listed in Table 8. The particular criteria considered and their method of assessment will depend on the nature of the particular site or inventory of sites considered. However, it can be seen that many of the environmental and social criteria can be directly related to the various ecological and cultural designations listed in Table 1, for example cultural receptor criteria can be linked to, for example, AONB, NP and LHI data and ecological receptor criteria to, for example, SAC, PH and SSSI data.

812 **Table 8. Examples of decision criteria**

813

 It is suggested that this methodology will be applied for two main purposes. This first of these is site specific and will aid comparison between different options and scenarios. For example, the choice between various ex-situ and in-situ remediation technologies can be made and compared against a 'do-nothing' scenario. The second purpose is to allow inventory appraisal where a number of sites at a regional or national inventory scale can be ranked for potential resource recovery and also enable classification of an anthropogenic deposit as a reserve or resource.

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823 **5.2. Technology options for resource recovery from metal mine wastes**

824 It has been demonstrated that many historic UK metal mine sites comprise 825 environmental/landscape resources in their existing state. However, in light of stricter future legislation associated with the European Union Water Framework Directive it is likely that intervention (namely for pollution control) will need to be implemented in the future at many sites. Given the multifaceted resource value of metal mine sites, these interventions need to be sensitive to the existing resource (as indicated by the site designations presented) and/or enhance the resource value of the sites, for example by protecting or enhancing industrial heritage. Thus the cost-benefit analyses might include the reduced cost of remediation when including metal resource recovery and the additional benefits might include preservation, protection and enhancement of industrial heritage with the possible tourist revenue generation that may arise. The methodology proposed by (Conesa 2008), which strives to protect the cultural heritage components of metal mine sites whilst rehabilitating the site from an environmental perspective, is suggested as a useful approach, and it could be extended to include metal resource recovery.

 If the resource comprises the mine site in its current form then remediation for pollution mitigation would have to be done either through established *in situ* techniques for preventing or reducing infiltration, reducing leachability and waste stabilisation or containment, *ex situ* techniques could only be applied where the impact was minimal and the site could be rehabilitated to a condition satisfying the appropriate stakeholders.

 Where the metal present are one of the resources to be recovered from the site then an important processing decision is whether the mine wastes can be excavated. If this is an option for the site then a wide range of standard processing routes are available for separation, comminution, concentration and/or recovery metals from excavated materials. For example, gravity separation methods might in some cases be applied to separate metal-bearing minerals from gangue minerals which can be returned to site. Metals can then be recovered from the metal bearing concentrate using established hydrometallurgical, biohydrometallurgical or pyrometallurgical approaches.

 If physical separation is not an option e.g. for cost reasons or because of mineralogy and metal deportment is not favourable, then the hydrometallurgical and/or biohydrometallurgical techniques of heap (or dump) leaching may be of particular utility for the removal of metals from mine wastes and tailings. These techniques are routinely used in the mining industry for recovery of metals (e.g. Cu, Au) from low grade ores. Material is placed on to an impermeable liner system and a lixiviant is recirculated though the pile, metals are recovered from the metal- rich "pregnant" liquor. Where material is fine (e.g. tailings) then the material can be agglomerated prior to heap leaching to improve subsequent recovery. Similar methods such as soil flushing have been adopted for decontamination of soils and sediments (Leštan 2008) (Seidel 1998) - these parallel methods are essentially only different in their aim: metals recovery or decontamination and thus are applicable within the context discussed here.

 In situ approaches for metal recovery could be attractive given the constraints for mine site reclamation discussed above, and in this context could under certain conditions be considered as a more "passive" remediation option (see (Cundy 2013). Phytoremediation (or phytomining depending on context) is an established *in situ* technology, however the process is very low intensity and intervention is still required for periodic harvesting, processing of the biomass for metal recovery also requires significant further processing. *In situ* heap/dump leaching and metals recovery is a promising option but requires that the material to be flushed overlies an impermeable stratum or engineered barrier. A pump and treat system can then be applied to ensure capture of the effluent downstream in collection boreholes/trenches without resulting in secondary pollution. A compromise may be to capture and recover metals already being released from sites in mine drainage. Low intensity metal capture are being developed for the

 "passive" treatment of metalliferous mine waters. Such systems use a variety of (bio)geochemical engineering approaches to achieve immobilisation of metals, including: precipitation, adsorption, microbiological reduction or oxidation and pH manipulation. Thus these technologies potentially offer low intensity harvesting of metals from legacy mine waste and would simultaneously achieve: (i) eventual decontamination of the mine waste, (ii) 878 protection of the environment from metal pollution and (iii) recovery of the metals. However, further research is required to design systems that capture metals in forms that are directly amenable to recycling.

6. Conclusions and implications

There are numerous drivers for the reclamation of legacy mine sites. Many wastes are likely to be causing significant breaches of water and soil quality guidelines in the UK and the mobilisation of these pollutant metals may be negatively impacting surrounding ecosystems. When considering site reclamation strategies a balance needs to be achieved, however, between protecting human, water and ecological receptors that may be at risk from metal pollution from mine wastes and designations that are dependent upon it. In addition, the simultaneous recovery of economically valuable metals from the mine wastes during site remediation may provide a useful mechanism to offset the cost of such activity.

This study has determined the physical and chemical composition of several prominent legacy metalliferous mine tailing waste piles in England and Wales across a range of parameters, including metal content, mineralogy, paste pH, particle size distribution, total organic carbon and total inorganic carbon. The colocation of cultural and ecological designations with the mine wastes have also been determined. The following can be concluded:

- 1) Several mine wastes investigated contain a number of different economically valuable metals (namely Cu, Pb and Sn) at concentrations close to or greater than typical minimum ore grade;
- 2) Several mine wastes investigated contain a number of different pollutant metals (namely Cr, Ni, Zn, As, Cd, Pb) at concentrations exceeding Soil Guideline Values; and
- 3) Most of the case study sites receive some form of protection either due to their historical significance, rare species assemblages or geological characteristics which may limit the potential for resources recovery and rehabilitation.

Results demonstrate that it is unlikely that the potential economic gain of extracting valuable metals from the mine waste will constitute a sole driver for intervention. Instead it is suggested that this value could be considered as a useful mechanism to offset site rehabilitation costs. A substantial number of mine sites in south west England and Wales are co-located with cultural or ecological designations, many of them due to the mining activities. These unique geological, ecological and cultural resources will act as a significant constraint to mine waste remediation and site reclamation if the existence of these features were to be adversely affected by such activities. This paper has demonstrated that an integrated assessment methodology for assigning and evaluating resource value is necessary to allow appropriate evaluation of resource potential. It is clear that further work is urgently required to apply similar holistic resource value determination approaches at other legacy mine sites in the UK and world-wide. This will enable the establishment of a reliable methodology for the quantitative assignment of resource value (economic, cultural, environmental, etc.) at such sites. In turn this will enable prioritisation of mine sites in most urgent need of rehabilitation, but also enable such rehabilitation and remediation processes to be conducted via methodology that is both at appropriate cost and disturbance to existing environmental and cultural designations.

Acknowledgments

We would like to thank Jeff Rowlands and Marco Santonastaso from the School of Engineering, Cardiff University for performing ICP-OES and TOC/TIC analysis respectively. We would also like to thank Geoff Smart, Mark Roberts and Theresa Mercer from the School of Engineering, Cardiff University for organising the mine waste sample collection. The work was financially supported by the Natural Environmental Research Council [grant number: NE/L013908/1]. Additionally we would like to thank the following people and organisations for facilitating access to the mine sites: Ainsley Cocks from Cornish Mining World Heritage Organisation and Alan Blamey from the Gwennap Parish Council (Consols, Nantgiles, Poldice and Wheal Maid); Mark Snellgrove of Bakes Down Farm and James Squier from Bidwells LLP (Devon Great Consols); Andy Crabb from the Dartmoor National Park Authority (Devon Great Consols); Caroline Vuliamy, Nick Russell and Daniel Ratcliffe of Historic England (Caradon, Prince of Wales); Mr Matt Glencross (Caradon); Chris Matthews of Duchy of Cornwall (Prince of Wales); John Bray (Frongoch); Peter Stanley and Tom Williams from National Resources Wales (Esgair Mwyn, Frongoch, Grogynion and Wemys); Philip Goodman from National Resources Wales, Ian Cuthbertson from Anglesey Mining and David Jenkins from the Amlch Industrial Heritage Trust (Parys Mountain); Robert Power of the National Trust (Levant); Mark and Robert Body of Nanpusker Farm (Alfred Consols). Finally we would also like to thank Samantha Davenport from Natural England for early access to the spatial OMH data, Jonathan Rothwell from Natural Resources Wales for spatial OMH and PH data and Paul Satchell at UWE for assistance with waste volume calculations.

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Appendix A. XRD spectra for the mine tailing composite samples. LHS (from bottom): ALF, CAR, CON, DGC, LEV; middle (from bottom): NAN, POL, PWM, WM; RHS (from bottom): EGM, FRN, GROG, PYM, WEM.

Appendix C. Number of non-active mines by commodity in each type of geological, ecological and cultural designation in the South West of England

Sources: mine data from BRITPITS Licence No. 2014/098BP ED British Geological Survey © NERC. All rights reserved. Local Nature Reserve (LNR), National Nature Reserve (NNR), Site of Special Scientific Interest (SSSI), Speci of Conservation (SAC), Special Protection Area (SPA), Ancient Woodland (AW), Priority Habitat (PH), Open Mosaic Habitat on Previously Developed Land (OMH), Area of Outstanding Natural Beauty (AONB) and National Park data © Natural England copyright. Contains Ordnance Survey data © Crown copyright and database right 2016. Scheduled Monument and World Heritage Site © Historic England 2016. Contains Ordnance Survey data © Crown copyright and database right 2016. The Historic England GIS Data contained in this material was obtained on 29th June 2015. The most publicly available up to date Historic England GIS Data can be obtained from HistoricEngland.org.uk.

Appendix D. Number of non-active mines by commodity in each type of geological, ecological and cultural designation in Wales

¹Priority habitat data in Wales is presented as the area within a 1.6 km (1 mile) grid square so the location of the mine cannot be said to be accurately co-located with the habitats. SSSI=Site of Special Scientific Inte PH=Priority Habitat PH, OMH=Open Mosaic Habitat on Previously Developed Land, SAC=Special Area of Conservation (European), SPA=Special Protection Area, AONB=Area of Outstanding Natural Beauty, NP=National Park, CP=Country Park, SM=Scheduled Monument, WHS=World Heritage Site, Landscape of Heritage Interest; Sources: mine data from BRITPITS Licence No. 2014/098BP ED British Geological Survey © NERC. All rights reserved. All other data © Natural Resources Wales copyright. Contains Ordnance Survey data © Crown copyright and database right 2016.