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## 2 **Phytoremediation: metal decontamination of soils after the sequential forestation of** 3 **former opencast coal land**

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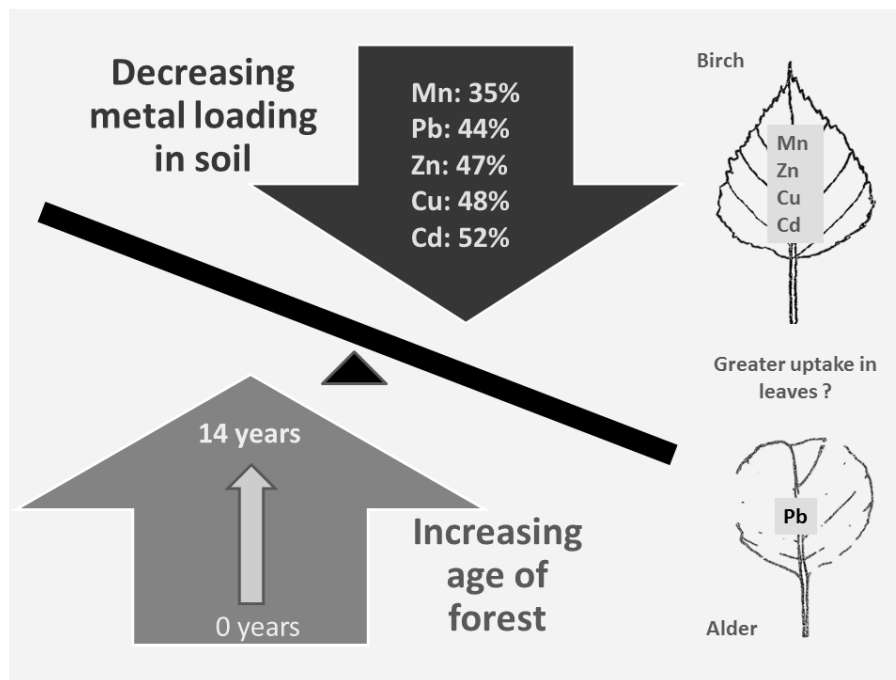
### 10 **Abstract.**

11 Forest phytoremediation through forestry may be an effective means for reducing the metal loading  
12 in lands reclaimed after surface-coal-mining in the UK. Planted with mixed woodland, soil loadings  
13 of 5 key metals (Zn, Cd, Mn, Pb and Cu) decreased, significantly and progressively, compared to soils  
14 left as grassland, through a 14 year forestation chronosequence on land reclaimed from the former  
15 Varteg opencast coalmine, South Wales, UK. Fourteen years after initial tree planting, soil metal  
16 loadings decreased by 52% for Cd (4.3 mg.kg<sup>-1</sup> per year), 48% for Cu (2.1 mg.kg<sup>-1</sup> per year), 47% for  
17 Zn (7.3 mg.kg<sup>-1</sup> per year), 44% for Pb. (7.1 mg.kg<sup>-1</sup> per year) and 35% for Mn (45 mg.kg<sup>-1</sup> per year).  
18 Analysis of metal loadings in the leaves of *Alnus glutinosa* (L. Gaertn) (Common Alder) and *Betula*  
19 *pendula* (Roth) (Silver Birch) found both species involved in metal uptake with birch taking up more  
20 Cd, Cu, Zn and Mn and Alder taking up more Pb. Concentrations of Zn, Mn and Cd (Birch only)  
21 increased significantly in leaves from, but not soils under, older plantings. Since different tree  
22 species take up metals at different rates, mixed plantings may be more effective in forest  
23 phytoremediation.

24 **Keywords:** forest phytoremediation; chronosequence; technosols; coal-land reclamation; South  
25 Wales; metal loadings in soils; metal loadings in leaves.

#### **Highlights**

- Isolates the effect of forestation in reducing soil metal loadings of five key metals.
- 14 year forestation chronosequence.
- Foliar analysis provides a direct link between tree growth and reduced metal loadings in soils.
- Mixed species plantings may be more effective in forest phytoremediation.
- Decontamination of former surface-coal-mine land in Wales



27

28

### Graphical Abstract

29

## 30 1. Introduction

31 This project aims to determine whether forestation is an effective means of reducing metal  
 32 contamination in the soils of former surface coal-mine (opencast) sites. It employs a series of  
 33 forestry test plots planted at different times across a 14-year period (1993 – 2007) on the same  
 34 terrace-bench of a surface-coal-mine land that was created and reclaimed in 1963. This planting  
 35 created an experimental framework where the only difference between test plots is the length of  
 36 time that they have been planted to trees. This study shows how this forestation is associated with  
 37 changes in metal concentrations in the developing soils on this site.

38 This study explores the effectiveness of two tree species, Common Alder (*Alnus glutinosa* L. Gaertn.)  
 39 and Silver Birch (*Betula pendula* Roth) in mitigating the levels of five key metals: Cadmium (Cd),

40 Copper (Cu), Manganese (Mn), Lead (Pb) and Zinc (Zn) in the marginally-contaminated soils of a  
41 former opencast coal mine in Torfaen, South Wales, UK. The Varteg Hill Extension Opencast mine  
42 was restored as grassland, albeit without great success. In recognition, in 1970, the UK's National  
43 Coal Board established a series of grass seeding trials just a kilometre north of Varteg in an attempt  
44 to remedy the problem. These National Coal Board test plots remain visible but their outcomes have  
45 never been published.

46 This study forms part of a larger research project that aims to determine best practices for reversing  
47 land degradation on formerly reclaimed opencast coal-lands in South Wales. This employs  
48 forestation as a means for the self-sustainable reconstruction of soils as well for its wider benefits to  
49 environmental regeneration and carbon sequestration (Haigh, 2018). Here, in order to reverse on-  
50 going land degradation, experimental tree plantings in trial plots were planted semi-annually  
51 between 1991 and 2007 (Haigh et al., 2018; Plamping et al., 2017).

52 Previous studies of mine-soil chronosequences often involved sites created from the same parent  
53 materials but at different times (Schafer et al, 1980; Walker et al., 2010). However, this describes a  
54 'forestry chronosequence', which is a series of forestry plantings where the trees are planted in  
55 different years but the soils beneath created simultaneously by a single land reclamation event (Karu  
56 et al., 2009). Many previous studies have explored the negative impacts of metal polluted soils on  
57 tree growth (Menon et al., 2007, Liu et al, 2013, Pulford and Watson, 2003). However, there are few  
58 comparable longitudinal studies of the long-term impacts of forestation on soil metal loadings.  
59 Insam and Domsch (1988) compared microbial biomass and soil carbon levels on a 30-year  
60 chronosequence of reclaimed opencast brown-coal mine spoils near Cologne, Germany, while  
61 Klaminder et al. (2006) used a 20-220 year forestry chronosequence to calculate a 250-year  
62 residence time for Pb in the mor organic layer of coniferous forest in northern Sweden. This also

63 found that the response could be much faster (Mean Recovery Time <50 years) in the early stages of  
64 forest succession when broadleaved litter fall dominates over conifer. In India's Jharia Coalfield,  
65 Bandyopadhyay, Rana and Maiti (2018) compared metal concentrations beneath 3 and 25 year old  
66 Eucalyptus trees. They found that Zn, Pb and Cu soil concentrations in the older reclaimed mine soils  
67 were, respectively, 3.46, 1.55 and 1.44 times lower.

68 The UK's surface- coal-mining industry is rapidly disappearing. However, it has created a legacy of  
69 disturbed land, some contaminated, but most 'reclaimed', if not always to a high standard (Haigh,  
70 2000). Metal contamination is not, usually, a major problem for former surface-coal-mines in Wales.  
71 While deep-mine coal-spoils may contain high levels of toxic metals, the mineral ores associated  
72 with such contamination are a relatively small part of surface-coal-mine spoils. These are dominated  
73 (90-95%) by 'overburden', the shale and sandstone rocks that overlay the coal-seams.  
74 Consequently, generally, these spoils have lower levels of contamination. This said, many surface-  
75 coal spoils contain 'hot spots' of severe contamination, which are often associated with coal and  
76 metal minespoils that have become mixed into the fill of the opencast pit.

77 Traditionally, in the UK, most former surface-coal-mine sites have been restored to grassland,  
78 especially in the uplands. This may not be the best policy for restoration where there is present or  
79 potential contamination by metals. In general, because of their greater productivity and biomass,  
80 trees have far greater potential value than grass for the amelioration of metal contaminated soils  
81 (Raskin, Smith and Salt, 1997; Pulford and Dickinson, 2006; Pajević et al., 2016). They need only  
82 accumulate small amounts of metal to be effective (Pulford and Dickinson, 2006). In addition, their  
83 roots penetrate much more deeply into the soil and involve more of the substrate in soil forming  
84 processes.

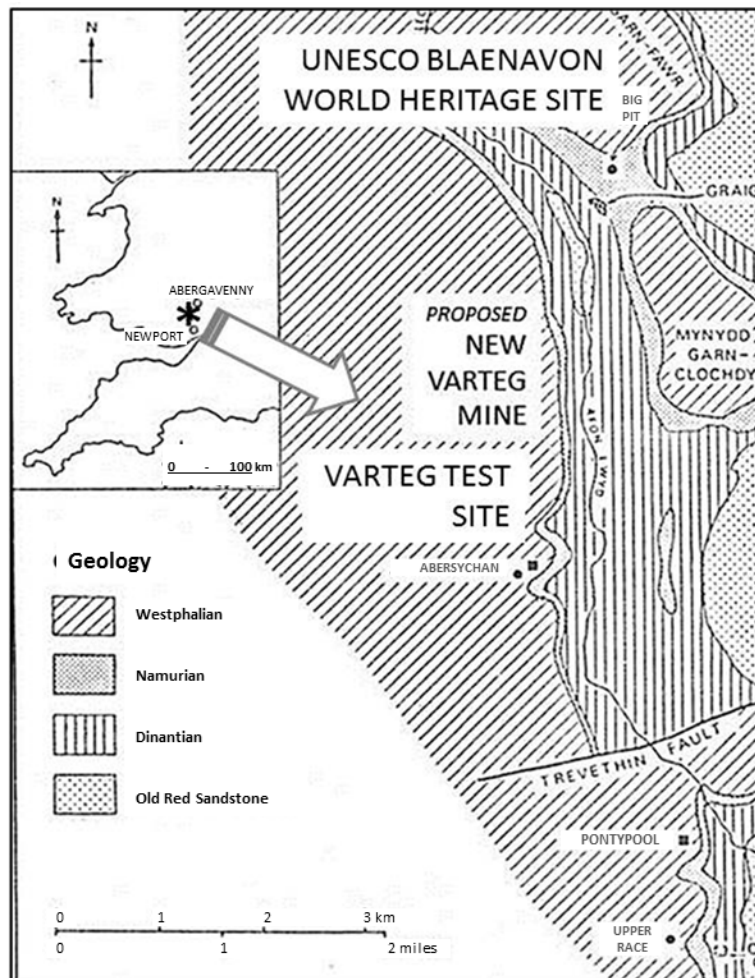
85 Currently, the main alternatives to the phytoremediation of metal-contaminated soils are physical  
86 approaches such as “dig-and-dump” and/or quarantining by containment, physico-chemical  
87 immobilisation or extraction (Wuana and Okiyeimen, 2011). However, while phytoremediation is  
88 relatively low-cost, these alternatives are expensive and better suited to the treatment of small  
89 problem sites than the large areas of marginal contamination typical of the UK’s former surface-  
90 coal-mines (Pulford and Watson, 2003, p.529).

91 However, forest phytoremediation requires many years to be effective and this is dependent on  
92 both the species planted and local site conditions that affect metal bioavailability (Pulford and  
93 Watson, 2003). Until recently, it was considered suitable only for very slightly contaminated soils  
94 (Ernst, 1996). Different species take up different metals in different proportions at different rates,  
95 which may mean that mixed plantings are more effective than monospecific (Samecka-Cymerman  
96 and Kempers, 2003). However, there remain questions about the phytoremediation effectiveness of  
97 tree species other than *Salix* and *Populus* spp (Raskin and Ensley, 2000). Further, forest  
98 phytoremediation has not always proved effective for sites where pollution is on-going. For  
99 example, a 23-year study of acid-soluble heavy metals (Cu, Pb, Cd, Zn) in the forest litter and humus  
100 horizons of soils affected by atmospheric deposition from the Middle Ural Copper Smelter, Russia,  
101 found that metal levels in soils remained largely unaffected by afforestation with spruce  
102 (Vorobeichik and Kaigorodova, 2017). This may be important because local research suggests that,  
103 even in the absence of the atmospheric deposition of pollutants, natural weathering of the mine-  
104 stones may lead to increased soil contamination through residual enrichment. Dang et al. (2002,  
105 1996) used SEM (Scanning Electron Microscope) and EPMA (Electron Probe Micro-Analysis) to  
106 compare ashed samples of weathered and unweathered mine-stones from two former South Wales  
107 surface-coal-mine sites just 8 km north of Varteg: Pwll Du (restored 1948) and Walters Group  
108 (restored 1994). They found that while some metals exist as sulphides, which can be oxidised to

109 sulphates through microbial action (Haigh, 2000), much is complexed with organic carbon, which is  
110 not mobilised by weathering and remains to enrich the weathered residual. This process carried Cu  
111 (145-675 mg/kg) and Zn (96-485 mg/kg) (cf. Table 1) to join Pb (23.4-74.9 mg/kg) above the  
112 thresholds for soil contamination. The present study asks if forest phytoremediation is an effective  
113 means for reducing soil decontamination in these circumstances.

## 114 **2. Site Description**

115 The test site lies 3 km south of the 'Blaenavon Industrial Landscape World Heritage Site' *"one of the*  
116 *best preserved, relict industrial landscapes in Wales, ... and [amidst] the only sizeable, abandoned,*  
117 *multiple period, opencast mineral workings in South Wales"* (Torfaen County Borough Council, 2013,  
118 p. 183). The forestry plots are located on a terrace bench created in 1963 during the reclamation of  
119 the Varteg Hill (Waun Hoscyn Extension) Opencast Coal Mine (51°44'50-53"N 03°07'72-98"W). This  
120 bench lies between 360- 370 metres above mean sea level on the western flank of Cwm Afon Llywd  
121 in Torfaen and on the western outcrop of the South Wales Coalfield (Figure 1). Pre-mining site  
122 geology was Carboniferous Coal Measure (Westphalian and Namurian) strata, mainly shales, often  
123 poorly consolidated mudstones, with harder sandstone and some thin coal seams (Neame, 2015).  
124 Spoil tips from coal and earlier iron mines were common in the pre-mining land surface and these  
125 wastes are mixed into the fill of the former surface-mine creating pockets of highly acid and metal-  
126 loaded spoil (Haigh and Kilmartin, 2017).



127

128

**Figure 1. Location of test site, Varteg, S. Wales**

129

Immediately north of the test site is the land of the proposed ‘Varteg Hill Reclamation and Coal

130

Recovery Scheme’, which includes large parts of the former Varteg Opencast mine area and which

131

has been subject to intensive investigation for an Environmental Impact Assessment (Smart, 2014).

132

This reports high levels of Fe and Mn in many groundwater samples, especially, in waters draining

133

Coal Measures strata and former coal-mine workings (Smart, 2014). Deep-mine coal spoils are

134

judged slightly contaminated, with high levels of As (Ove Arup and Partners Ltd, 2008). Twenty test

135

pit samples found contamination ‘hot spots’ for Cu, Zn, B and high ambient levels of iron and

136

sulphate (Smart, 2014). The report concluded, controversially, that ground contamination posed no

137 “significant risk either to human health or controlled waters” (Smart, 2014, p68). However, as Table  
 138 1 shows, several of the levels reported exceed guideline values recognised by the Environment  
 139 Agency (UK), CLEA (UK) or MAFF/EU, and the ‘Dutch List’ and suggest contamination issues for Cu,  
 140 Zn, Pb, and (possibly), As, Ni and Se.

141 **Table 1: Metals (mg/kg<sup>-1</sup>) recorded in 20 samples from test pits on adjacent ‘Varteg Hill**  
 142 **Reclamation and Coal Recovery Scheme’ lands (Smart, 2014).**

Metal (Emboldened metals are those examined by this study)	Range	Environment Agency 'Prompt Value' for soil of pH 6.5 (Nicholson et al., 2008)	CLEA Guideline Value (UK) (Residential or allotment land use).	'Dutch list' (target and) intervention values for soil remediation (VROM, 2000)	Maximum metals allowable in UK soils (MAFF, 1993) (nb. same as: EC Directive 86/278/EEC for soil of pH5.5).
As	6.7 – 62.5	n/a	500	(29) 55	50
<b>Cadmium (Cd)</b>	<b>&lt;0.1 – 0.76</b>	<b>1.0-1.1</b>	<b>1.0-1.8</b>	<b>(0.8) 12</b>	<b>3</b>
Cr	3.8 – 28.9	n/a	130	(100) 380	400
<b>Copper (Cu)</b>	<b>11.7 – 4630</b>	<b>58</b>	<b>n/a</b>	<b>(36) 190</b>	<b>100</b>
<b>Lead (Pb)</b>	<b>18.1 – 1075</b>	<b>260.7</b>	<b>450</b>	<b>(85) 530</b>	<b>300</b>
Hg	<0.1 – 0.43	n/a	10-26	(0.3) 10	1
Ni	5.1 – 85.6	21	130-230	(35) 210	60
Se	<0.5 – 3.1	n/a	120	(0.7) 100	3
<b>Zinc (Zn)</b>	<b>18.5 – 742.5</b>	<b>n/a</b>	<b>n/a</b>	<b>(140) 720</b>	<b>250-300</b>

143  
 144 Currently, soil profiles on the reclaimed land consist of a thin organic layer, typically 2-3 cm  
 145 thickness, above weathered clays with clasts of sandstone, coal shale and some coal. Many soil  
 146 profiles display a clay-enriched, high density, essentially water-impermeable, layer at 18-30 cm.  
 147 Deeper, the proportion of large cobbles of sandstone and coal-shale increases. Below 50 - 70cm are  
 148 unweathered, often clay-veneered, mine-spoil cobbles with bridged voids (Haigh et al., 2015; Haigh  
 149 and Sansom, 1999).  
 150 Autocompaction, caused by the breakdown of water-unstable minestones near the soil surface, is  
 151 ubiquitous on local former surface coal-mine sites (Haigh and Sansom, 1999). At Varteg, soil bulk  
 152 density is typically high: 1.63 g/cm<sup>3</sup> at 35-40cm and 1.81 g/cm<sup>3</sup> at 50 cm depth. These high bulk  
 153 densities negatively affect soil organic carbon levels, which fall below 1% when bulk density >1.5  
 154 g/cm<sup>3</sup> (Merrington, 2006). Moffat and Bending (1992) suggest that, ideally, the soil bulk density for



155 forestry on disturbed land should be  $<1.5 \text{ g/cm}^3$  ( $<50 \text{ cm}$  depth) and not  $>1.7 \text{ g/cm}^3$  at  $100 \text{ cm}$   
156 depth. Here, the higher bulk densities may reduce root penetration, except through fissures;  
157 indeed,  $1.8 \text{ g/cm}^3$  is often considered the limiting maximum bulk density for root penetration (Haigh,  
158 2000).

159 Soil pH is a key influence on metal partitioning between the soil and soil solution (Bolan et. al.,  
160 2003). Soil pH recorded in 63 field tests under 3m high trees averaged pH 5.8 (sd. 0.3), while under  
161 dense forest in test plot PA93, Humphrey (1997) recorded pH 6.6 (sd. 0.2) in 24 tests. Moffat and  
162 Bending (1992) advise that soils for reclamation forestry should have a pH in the range of 3.5 – 8.5.  
163 Merrington (2006) proposes trigger values of  $\text{pH}>6$  for metal retention and  $\text{pH}>5$  for effective  
164 microbial functioning.

165 Organic matter increases a soil's moisture holding capacity and soils used for tree planting should  
166 contain  $>0.8 \%$  organic matter. However, the Varteg soils have only  $0.3 \%$  to  $0.5 \%$  organic matter,  
167 perhaps even less because Loss on Ignition (LOI) tests can be affected by the combustion of mineral  
168 coal fragments. Soil fertility levels (N, P, K and Mg) are also very low (Haigh et al., 2018).

169 Monthly mean air temperatures range from  $2.5^\circ\text{C}$  to  $15^\circ\text{C}$ . Rainfall (1971-2000) was  $1543 \text{ mm/year}$   
170 at Cwmavon Reservoir,  $0.8 \text{ km}$  to the north on the Afon Lwyd valley floor. Locally, natural soils  
171 remain at field capacity for 285-325 days/year; evaporation is estimated as  $472 \text{ mm/year}$  on rough  
172 grazing land (Robinson, 1989).

### 173 **3. Methodology**

#### 174 **3.1 Tree Species Selection.**

175 The tree species recommended for the reclamation of former coal-lands in the UK include:

176 *“generally undemanding or ‘pioneer’ species...rowan, birch, alder, poplar, larch and pine”* (Forestry

177 Commission (Scotland), 2001, p. 7). Willow is also planted, typically at riparian or boggy sites or as  
178 part of a mixed planting. However, *Alnus* (Alder) and *Betula* (Birch) are widely used and two key  
179 species are selected for this study (Grobelak, 2016; Pulford and Watson, 2003; Haigh et al., 2018).

180 Alder species are often used in the phytoremediation of metals, where bacterial endophytes are  
181 central to the process (Ma et al., 2016). *Alnus glutinosa* (L.) Gaertn., the UK's only native alder, is  
182 known to thrive in metal contaminated and waterlogged soils and on coal-land reclamation sites for,  
183 at least the first 5 years, >10 years at Varteg (Moffat, 2014; Haigh et al. 2018). Alder also  
184 accumulates metals such as Pb, Zn and, especially, Cu in its roots and root nodules, of which a small  
185 proportion is transferred to leaves and shoots (Lorenc-Plucińska et al., 2013). Nevertheless, the  
186 leaves and leaf litter from *Alnus glutinosa* grown on polluted soil contain greater amounts of Cu, Zn  
187 and Cd (but similar amounts of Pb) than trees grown on non-polluted soils. Since *Alnus* is a nitrogen  
188 fixer, it tends to increase soil acidity, which may also increase metal mobility, although rates of litter  
189 decomposition do not seem to be affected (Scheid et al., 2009).

190 Silver birch, *Betula pendula* (Roth), like other *Betula* species, is a pioneer coloniser of lighter infertile  
191 soils. It has a strong rooting system that enables it to penetrate into the deeper layers of soil, which  
192 also helps it access and accumulate larger amounts of metal. Silver birch has been suggested for use  
193 in the biomonitoring of soil contamination because of the close correlation between the levels of Cu  
194 and Pb in its leaves and in the soil (Klink et al., 2006). Mycorrhizal associates (VAM) facilitate metal  
195 uptake by the plant and ameliorate Zn toxicity (Baker and Walker, 1990). Birch, a hyperaccumulator  
196 of Zn, has a high tolerance of polymetallic soil contamination, although shoot and root growth in  
197 young seedlings can be inhibited, especially by Cd (Dmuchowski et al., 2014, Hermle et al., 2006;  
198 Osteras et al. 2000). Hence, within limits, both *A. glutinosa* and *B. pendula* are tolerant of soil metal  
199 contamination and, while total metabolic activity can be impaired, the density of associated

200 microorganisms (bacteria and fungi) inhabiting root and rhizosphere, is not (Złoch et al., 2014;  
201 Scheid et al., 2009).

### 202 **3.2 Metal Selection**

203 Coal-mine spoils are a common source of heavy metal contamination (Siegel, 2002). The metals  
204 found in surface-coal-mine spoils originate, variously, from the geochemistry of the native rocks,  
205 past industrial activities including mining, air pollution, and from the addition of soil amending  
206 substances during reclamation (Alloway, 2013; Haigh, 1995; Siegel, 2002). An inventory of normal  
207 agricultural soils in the UK found that atmospheric deposition was the major source (25-85%)  
208 followed by manure and sewage sludge (Nicholson et al., 2003). However, in coal-mine spoils, many  
209 metals are mobilised by weathering and microbiological processes in the relatively oxidizing and  
210 sometimes acidic environments of the newly displaced and fractured minespoils (Dang, et al., 2002;  
211 Haigh, 2000). Acids caused by pyrite dissolution are aggressive; they can degrade aluminosilicates  
212 and other minerals releasing previously stabilised metal components (Johnson, 2003). For example,  
213 Cadmium (Cd) is mobilised at pH 6.0 - 5.5, and its mobility increases by 60% -90% at pH 4.0, while  
214 Zn, Ni and Cu become mobile at 5.0-5.5 (Blake and Goulding, 2002).

215 Different researchers have selected different clusters of metals for analysis in similar contexts. For  
216 example: Alvarez et al (2003) selected: Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn; Moreno-Jiménez et al.  
217 (2009): Cd, Cu, Fe, Mn, and Zn; Maiti et al. (2016): Cd, Cu, Fe, Mn, Ni, and Zn; Niu et al (2015): Cd, Cr,  
218 Ni, Pb and Zn, and Pietrzykowski et al. (2014): Cd, Cu, Pb and Zn. This study selects Cd, Cu, Mn, Pb,  
219 and Zn, all five are among the 11 key metals listed by Alloway (2013, p.3). Four, Cd, Cu, Pb, and Zn  
220 are counted strongly hazardous, while Mn is only problematic at very high loadings (Vodyanitskii,  
221 2016). As for bioavailability, Cottenie et al. (1982) regard two, Cadmium (Cd) and Zinc (Zn), as

222 readily mobilised by plants, two, Lead (Pb) and Copper (Cu), much less so while Manganese (Mn) is  
223 'average'.

### 224 3.3 Experimental Design and Methods

225 This project evaluates the forest phytoremediation potential of five key metals (Cd, Cu, Mn, Pb and  
226 Zn) by two tree species commonly used in land reclamation: Common Alder (*Alnus glutinosa* L.  
227 Gaertn.) and Silver Birch (*Betula pendula* Roth). It explores soil and foliar samples from 5 forestation  
228 plots planted between 1993 and 2007 (Table 2) into the (marginally) contaminated soils of a former  
229 surface-coal-mine in South Wales, UK. (Haigh et al, 2018). These data are compared with data from  
230 the soil prior to planting plot MD07 (BL07: 'Background Level').

231 **Table 2. Varteg Test Plots and their date of plantation**

Site Code	Age of Plantation	Year of Planting
PA93	16-17	1993
TA94	15-16	1994
SH97	12-13	1997
CA03	6-7	2003
MD07	2-3	2007
BL07	0	<i>Not planted (Control)</i>

232  
233 Successive plantings of similar, adjacent, mixed species, forestation plots in 1993, 1994, 1997, 2003  
234 and 2007 has created a set of trees and soil that differ only in the length of time that they have been  
235 forested. This study aims to establish whether soil metal loadings of Zn, Cd, Mn, Cu and Pb are  
236 different beneath Alder and Birch trees of different age and through foliar analysis to determine  
237 whether these trees are directly involved in these changes. The null hypothesis is that there is no  
238 significant difference in metal loadings between test plots.

239 Soil samples were collected at a depth of 20-30cm, using random number tables to create co-  
240 ordinates for sampling, and replicated in 2009 and 2010. Soil compaction made it difficult to collect

241 samples from deeper layers. The 210 soil samples were tested for standard physical and chemical  
242 parameters including: soil pH, soil texture, soil moisture content, soil organic content - loss on  
243 ignition and carbonate content (following: Heiri, et al., 2001). Metals were assayed by an Atomic  
244 Absorption Spectrophotometer (AAS) using an Agilent Technologies SpectrAA 220FS and EDTA-Na<sub>2</sub>  
245 extraction (Quevauviller, 1996). Soil test results were calibrated against tests undertaken,  
246 independently, at the University of Wales Trinity St Davids and data published for the adjacent  
247 Varteg Hill Reclamation and Coal Recovery Scheme (Faber-Maunsell, 2006). Results were compared  
248 with the contamination action/treatment thresholds suggested by the widely used “Dutch List”  
249 (VROM, 2000).

250 Foliar samples (>10g per tree – dry weight, n=234) were collected in 2009 and 2010 from mature  
251 larger leaves in the upper three quarters of the tree, where metal content might be maximal. These  
252 samples were collected from the ‘control’ sections of each plot, where the only additives supplied to  
253 the trees were 750g of spent mushroom compost on planting (cf. Haigh et al., 2018). Foliar samples  
254 were digested using the MLS-1200 MEGA Microwave Digestion System. Soil and foliar sample  
255 collection was replicated in 2009 and 2010. During analysis, soil and foliar reagent blanks were  
256 analysed after every 15 tests and standard solutions after each 10 samples, to check against  
257 instrument drift.

#### 258 **4. Results.**

259 The results are reported in three stages. First, soil results prior to the 2007 planting (BL07) are  
260 contrasted with those measured on the same site after planting (MD07) in 2009 and 2010. Second,  
261 these results are compared with those from soil testing on four older plantings (CA03—2003; SH97 -  
262 1997; TA94 - 1994 and PA93 – 1993). Finally, results from the foliar tests are considered.

263

264 **Table 3. Soil metal loadings before planting (BL07) and 2 and 3 years after planting (mg kg<sup>-1</sup>)**

Mean (sd.)	Cd	Cu	Mn	Pb	Zn	Sample n=
<b>BL07 (2007)</b>	1.16 (2.63)	63.92 (1.52)	1879 (385)	224 (34)	192.8 (29.2)	213
<b>MD07 (2009)</b>	1.12 (2.33)	58.00 (2.00).	1598 (275)	224 (32)	177 (24.0)	210
<b>MD07 (2010)</b>	1.07 (1.98)	51.21 (1.98)	1532 (242)	200 (28)	169.7 (30.51)	234
<b>Significance of difference: 2007 to 2010 (p=)</b>	0.004	0.003	0.002	0.025	0.005	-

265

266 Table 3 shows that, in all cases, recorded metal levels are highest in the pre-plantation tests and  
 267 lowest in the third, 2010, data collection: 2007>2009>2010. A Wilcoxon Signed Ranks test suggests  
 268 that the sequence 2007>2009>2010 is significant (p<0.01). Overall, levels of Cu, Mn, Pb and Zn in  
 269 the soil decreased by 14-18% but Cd just 6%. In the case of Cu, and possibly Mn and Zn, the total  
 270 reduction is greater than (or close to) one standard deviation (sd.) of the mean. T-testing confirms  
 271 that the decrease in all metal levels, from before planting (2007) until 2-years after planting (2010),  
 272 is significant.

273 Table 4 displays data from tests conducted in 2009 and 2010 on all five plots. Again, the metal  
 274 loadings recorded in 2009 are, as a whole, significantly greater than those of 2010 (p<0.0005), even  
 275 if relative differences are small. The decrease in metal concentrations across progressively older  
 276 plantations continues; in 46 of 50 cases the next older plot displays a lower level of each metal. This  
 277 is significant (Wilcoxon Signed Ranks: p<0.05). The four exceptions cases all come from the relatively  
 278 unsuccessfully forested CA03 plot, where concentrations of Cu (slightly) and Mn (considerably) are  
 279 higher on CA03 (planted 2003) than the younger MD07 (planted 2007).

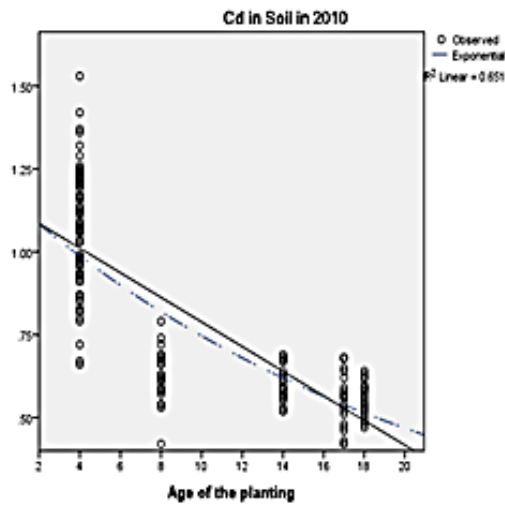
280 **Table 4. Metal loadings (mg kg<sup>-1</sup>) in soils across 14-year forestation chronosequence (n=423)**

MD07	Cd - 2009	Cd - 2010	Cu -2009	Cu - 2010	Mn - 2009	Mn - 2010	Pb - 2009	Pb - 2010	Zn - 2009	Zn - 2010
MEAN	1.12	1.07	58.23	51.21	1598.3	1533.8	223.62	199.49	176.75	169.68
Min	0.43	0.28	32.39	30.42	1233.5	1003.0	134.11	131.16	83.64	75.43
Max	2.85	2.45	78.61	69.66	2353	2171.5	285.38	265.32	232.03	221.82

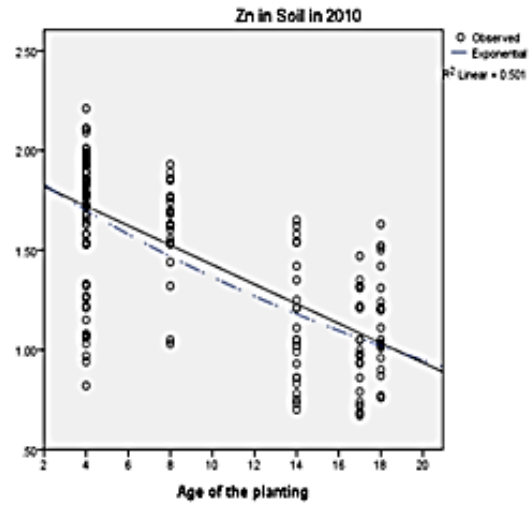
sd	2.33	1.98	5.66	7.13	275.3	241.7	32.36	27.60	24.43	30.46
<b>CA03</b>										
MEAN	0.70	0.63	60.46	52.31	2058.46	2012.69	197.86	189.25	155.45	146.94
Min	0.30	0.42	32.53	31.58	1332.13	1292.06	122.74	128.74	73.53	75.52
Max	1.27	1.25	68.68	63.85	2686.42	2568.73	278.62	268.17	203.33	193.17
sd	0.87	0.77	7.22	7.70	299.33	277.23	34.56	37.46	43.35	35.30
<b>SH97</b>										
MEAN	0.64	0.60	49.16	43.00	1366.02	1340.04	142.8	131.95	124.56	111.74
Min	0.58	0.52	32.32	31.16	1034.75	1048.92	104.96	117.03	66.08	70.8
Max	0.73	0.69	67.28	59.83	1557.69	1498.63	176.44	168.89	176.05	165
sd	0.34	0.49	4.68	5.84	687.54	928.43	8.68	9.78	28.43	27.79
<b>TA94</b>										
MEAN	0.61	0.59	41.00	37.58	1374.56	1337.48	130.83	123.52	100.03	97.3
Min	0.32	0.37	22.05	25.93	1033.96	1048.82	100.25	104.96	63.86	67.08
Max	0.82	0.78	65.73	59.32	1532.03	1498.04	157.09	143.04	155.75	147.55
Sd	0.87	0.99	6.99	7.43	790.44	845.72	10.26	8.05	24.55	22.45
<b>PA93</b>										
MEAN	0.57	0.56	36.53	33.27	1276.02	1227.10	127.23	120.4	94.32	89.43
Min	0.11	0.14	22.56	27.37	1132.32	1116.48	99.4	101.24	78.2	67.32
Max	0.68	0.64	43.52	40.71	1353.84	1322.83	137	131.57	174.11	163.79
sd	0.48	0.47	3.50	3.56	432.92	493.92	5.50	6.11	24.53	25.32

281

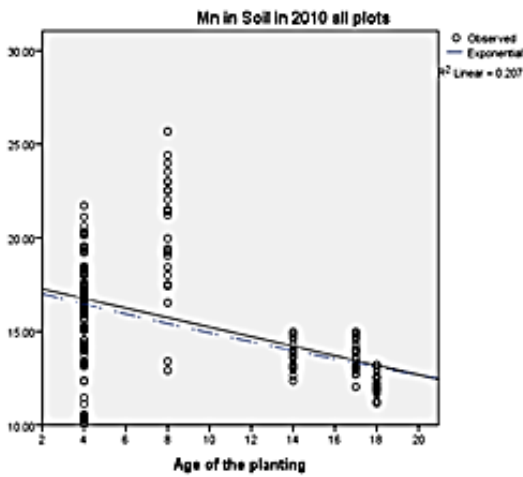
282 Regression analysis demonstrates the negative trend linking the amount of time trees have been  
283 planted and the amount of metals present in soil. Figure 2, which combines the data collected in  
284 2009 and 2010, shows the significant ( $p=0.002$ ), negative, best-fit trend line linking increasing age of  
285 trees with decreasing soil metal concentration. It depicts both linear and exponential best-fit lines,  
286 and, while the differences are small, they suggest that the rate of decrease in soil metal  
287 concentrations could be declining with time after forestation. In sum, 14 years after forestation with  
288 Alder and Birch, soil metal concentrations have decreased relative to background levels (BL07) by  
289 52% for Cd ( $0.043 \text{ mg.kg}^{-1}$  per year), 48% for Cu ( $2.07 \text{ mg.kg}^{-1}$  per year), 47% for Zn ( $7.21 \text{ mg.kg}^{-1}$  per  
290 year), 44% for Pb. ( $7.16 \text{ mg.kg}^{-1}$  per year) and 35% for Mn ( $44.82 \text{ mg.kg}^{-1}$  per year).



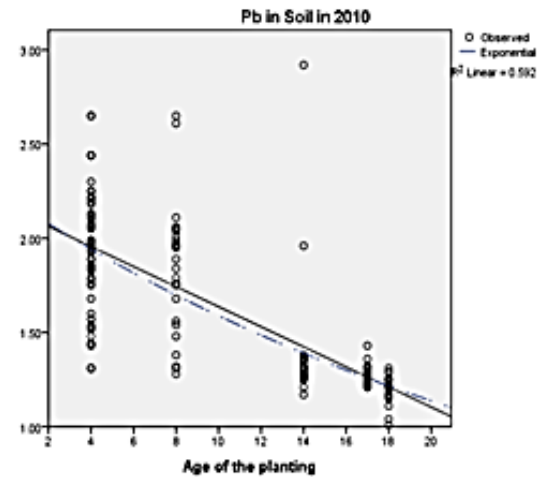
a) Cd in soil



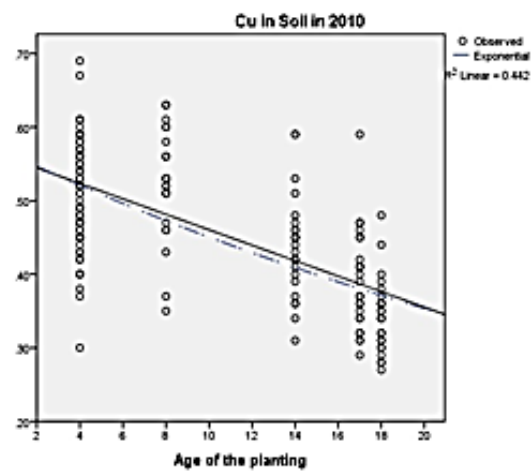
b) Zn in soil



c) Mn in soil



d) Pb in soil



e) Cu in soil

**Figure 2: Regression between metal loadings in soil (mg/kg) and age of tree plantation (years) (with linear  $R^2$  values: all cases:  $p < 0.005$ ).**



292 The pre-planting data (BL07) indicate baseline metal loading in grassed soils. Of course, these are  
 293 not the initial soil concentrations on site; they are what remain after 44 years. Similarly, the data  
 294 from the forestation plots show the soil metal loadings that remain after 46-47 years, including  
 295 between 2-3 years under forest (MD07) and up to 16-17 years under forest (PA93). These results,  
 296 however, do show the impact, through time, of changing the land use from grass to forest.

297 The significant and progressive decrease in metal loadings following forestation suggests that trees  
 298 play a role but it does not prove the connection. Hence, foliar analysis was undertaken to prove that  
 299 uptake by trees is directly involved.

300 Table 5 describes the average metal loading ( $\text{mg kg}^{-1}$ ) of Alder and Birch leaves across the whole  
 301 series of forest plantings from 1993 to 2007 as recorded in 2009 and 2010. The Alder leaf data  
 302 shows that metal uptake remains fairly consistent across the whole chronosequence; there is a  
 303 moderate uptake of Mn, Zn and, to some extent, Cd, although Pb may show a slight negative trend.  
 304 Findings for Birch are similar, although only four of the six test plots include this species. For both  
 305 species, the sequence of foliar volumetric uptake is  $\text{Mn} > \text{Zn} > \text{Cu} > \text{Pb} > \text{Cd}$ . However, Birch leaves  
 306 contain higher levels of Cd, Cu, Mn and Zn, while Alder leaves contain more Pb. Unlike Alder, Birch  
 307 shows a stronger uptake in the leaves of trees planted 2-3 years previously (MD07) than those  
 308 planted 15-16 years ago (TA94). The exception is Mn, where the trend is reversed. In theory, of  
 309 course, the total uptake of trees increases with age because of their greater biomass (Haigh et al.,  
 310 2018; Plamping et al., 2017).

311 **Table 5. Comparison of foliar metal uptake ( $\text{mg kg}^{-1}$ ) by Alder and Birch (2009-2010) (Alder n= 111;**  
 312 **Birch n=123).**

Plot	Alder	Birch	Alder	Birch	Alder	Birch	Alder	Birch	Alder	Birch
<b>MD07</b>	<b>Cd</b>	<b>Cd</b>	<b>Cu</b>	<b>Cu</b>	<b>Mn</b>	<b>Mn</b>	<b>Pb</b>	<b>Pb</b>	<b>Zn</b>	<b>Zn</b>
MEAN	0.20	<b>0.62</b>	4.51	<b>14.00</b>	239.65	<b>448.78</b>	<b>4.25</b>	2.38	67.72	<b>88.86</b>
MIN	0.00	0	2.01	9.22	167.01	305.13	1.99	1.02	38.54	39.02

MAX	0.75	1.13	6.88	20.01	405.00	572.30	6.47	4.75	107.53	145.23
Std Dev	0.41	0.58	0.91	2.01	50.31	70.66	1.31	0.60	13.49	23.67
<b>CA03</b>	<b>Cd</b>		<b>Cu</b>		<b>Mn</b>		<b>Pb</b>		<b>Zn</b>	
MEAN	0.32		4.91		307.74		4.65		77.13	
MIN	0.06		3.02		189.00		2.15		52.12	
MAX	0.52		7.64		433.21		6.92		98.56	
Std Dev	0.36		1.08		58.51		1.33		11.92	
<b>SH97</b>	<b>Cd</b>	<b>Cd</b>	<b>Cu</b>	<b>Cu</b>	<b>Mn</b>	<b>Mn</b>	<b>Pb</b>	<b>Pb</b>	<b>Zn</b>	<b>Zn</b>
MEAN	0.22	<b>0.58</b>	4.72	<b>14.70</b>	243.68	<b>465.20</b>	<b>3.82</b>	2.78	61.76	<b>81.60</b>
MIN	0.04	0.01	2.01	10.21	201.19	322.01	2.53	1.95	31.24	31.21
MAX	0.42	0.84	7.94	18.45	354.00	599.00	6.49	4.52	89.43	125.63
Std Dev	0.48	0.54	1.32	2.91	39.72	63.31	1.39	0.70	17.86	23.01
<b>TA94</b>	<b>Cd</b>	<b>Cd</b>	<b>Cu</b>	<b>Cu</b>	<b>Mn</b>	<b>Mn</b>	<b>Pb</b>	<b>Pb</b>	<b>Zn</b>	<b>Zn</b>
MEAN	0.23	<b>0.52</b>	4.80	<b>10.95</b>	266.74	<b>498.12</b>	<b>3.59</b>	2.41	60.05	<b>71.86</b>
MIN	0.05	0	2.04	6.8	195.02	211.03	1.53	1.01	32.19	22.06
MAX	0.31	0.6	8.43	13.56	371.42	699.53	6.78	3.21	78.42	120.40
Std Dev	0.48	0.54	1.42	1.39	30.06	98.71	1.26	0.65	9.54	25.01
<b>PA93</b>	<b>Cd</b>		<b>Cu</b>		<b>Mn</b>		<b>Pb</b>		<b>Zn</b>	
MEAN	0.25		5.33		291.02		3.75		58.49	
MIN	0.00		0.00		136.22		1.94		32.45	
MAX	0.30		10.30		398.05		5.77		70.41	
Std Dev	0.41		2.02		64.36		1.22		15.57	

313

314 Table 6 displays correlations between the metal loadings in soils and leaves across the full range of  
315 plantings. The first column displays, after applying a Bonferroni correction, the significant negative  
316 correlations between metal loading in the soil and age of the plantation (cf. Figure 2). The top row  
317 explores correlations between plantation age and foliar metal levels by tree species. It shows  
318 positive correlations between the age of plantation and the loading of Mn and Zn in both Alder and  
319 Birch leaves and Cd in Alder alone. Table 7 also displays several positive significant correlations  
320 between metal levels in soils and leaves for all metals except Cd. This means that as the plantation  
321 ages, there are higher levels of metals engaged with the biological system.

322 The loading of Mn and Zn in soils also correlates with that in leaves for both species, which suggests

323 a relationship with the level of contamination, while the positive correlation ( $p=0.002$ ) between Zn  
 324 in soil and Mn in leaves implies a mutuality of some kind. These data, however, confirm that the  
 325 trees are engaged actively with the mobilization of, at least some, of the metals in the soils.

326 **Table 6: Correlations between soil metal loadings and foliar metal concentrations for Alder and**  
 327 **Birch trees across all Varteg plots.** (Values show Pearson's 'r' and significance where  $p<0.005$  is  
 328 rated 'significant').  
 329

	Age of Trees	Mn Alder	Mn Birch	Cd Alder	Cd Birch	Zn Alder	Zn Birch
Age of Trees		0.573 $p=0.002$	<b>0.385</b> $p=0.001$	<b>0.468</b> $p=0.001$		<b>0.302</b> $p=0.001$	0.205 $p=0.004$
Cd Soil	<b>-0.830</b> $p=0.001$						
Cu Soil	<b>-0.651</b> $p<0.0005$						
Mn Soil	<b>-0.388</b> $p<0.0005$	0.453 $p=0.003$	0.348 $p=0.005$				
Pb Soil							
Zn Soil	<b>-0.672</b> $p=0.001$	0.366 $p=0.004$	0.393 $p=0.002$			0.489 $p=0.005$	0.589 $p=0.002$

330  
 331 **5. Discussion**

332 This investigation explored a 14 year forest chronosequence on land of the same age and character  
 333 where parts were forested with similar species at different dates and other parts left as grassland.  
 334 This allowed the assessment of the long term impact of forest upon metal loadings in the soil.  
 335 Common Alder and Silver Birch were studied, both are native species widely used in UK land  
 336 reclamation. Metal selection was guided by their plant availability; Cd and Zn are readily mobilised,  
 337 Pb and Cu much less easily, while Mn is 'average' (Cottenie et al., 1982). Four of the five metals are  
 338 rated strongly hazardous, while Mn is hazardous only at very high loadings (Vodyanitskii, 2016).  
 339 The plantation of mixed woodland on marginally metal contaminated lands, which were reclaimed  
 340 in 1963 from the Varteg Hill Extension Opencast Coal-mine, was associated with a significant long  
 341 term reduction in the loadings of all five metals compared to land left under grass. The reduction

342 ranged between 35% (Mn) and 52% (Cd). By contrast, Baltrėnaitė et al., (2016), whose focus was  
343 uptake by plant tissues only, found that the effectiveness of Birch for the purification of a sewage-  
344 sludge polluted soil was 0.2-0.3% per year (<4.2% across 14 years) for Mn and Zn but only 0.04-  
345 0.07% per year for Cu and Pb (<0.98% across 14 years) while that for Alder reached only 0.01-0.04%  
346 per year for Cu, Mn and Pb (<0.6% across 14 years). Clearly, there is more involved in metal  
347 decontamination than simply the planted trees. Elsewhere in the UK, *Populus* sp. has been  
348 estimated to reduce soil Cd and Zn contamination by 5.6 mg Cd kg<sup>-1</sup> and 96 mg Zn kg<sup>-1</sup> over 20-years  
349 (French, 2004; French et al. 2006). The non-linear regression graphs of Figure 2 and negative  
350 correlations in Table 6 confirm that the rate of metal depletion is decreasing as soil loadings  
351 decrease. However, if present depletion rates continued: Cd and Zn would be depleted in about 15  
352 years, Cu and Pb within 20 and Mn within 30 years. If Placek et al. (2016) are correct in suggesting  
353 that the 'self-cleaning of soils' can take several hundred years, then it is clear that forestation with a  
354 mixed woodland is an effective catalyst of these processes.

355 Table 7 compares the uptake of metals by tree leaves on the Varteg with data collected for the same  
356 species in other contexts. The table shows that, typically, Birch leaves contain higher loadings of all  
357 the metals tested except Pb. The Varteg data, in general, fits within this spectrum of other results  
358 and with other data from its local context (Smart, 2014). Several studies report low uptake of Pb and  
359 Cu in Alder leaves (Huang et. al., 1997; Schmidt, 2003; French, 2004). Butkus and Baltrėnaitė (2007)  
360 also found Alder leaves to contain more Pb but also more Cu and Mn. Samecka-Cymerman et al  
361 (2003) also found low uptake Pb and Cu into leaves of Birch (*Betula pendula*) grown on  
362 contaminated black coal spoils in Poland. However, on the Varteg, the Cu loading of Birch leaves  
363 and amount of Mn in the soil is high, while the amount of Zn is relatively low (Table7).

364

365 **Table 7. Comparisons of metal loadings (mg.kg<sup>-1</sup>) in contaminated soils (italics) and in the leaves of**  
 366 **Alder and Birch (means and +/- standard deviations)**

	<i>Varteg Soil Range of Means (Data from Smart (2014))</i>	Varteg Birch (and Alder) leaves: Range of Means	<i>Silesia, Black Coal Dumps (Samecka-Cymerman et al., 2003)</i>	Silesia, Birch leaves (Samecka-Cymerman et al., 2003)	<i>Lithuania, sewage sludge amended soils (Butkus and Baltrėnaitė, 2007).</i>	Lithuania Birch (and Alder) leaves (Butkus and Baltrėnaitė, 2007).	<i>Belgium Dredged river sediment (Depth: 30-45cm) (Mertens et al., 2006a)</i>	Leaves (Alder) (Mertens et al., 2006b)
Cd	1.16 +/- 0.32 (<0.1 - 0.8)	0.5-0.6 (0.2 - 0.3)	0.6-1.2	0.3-0.6	n/a	n/a	5.9 +/- 0.7	(<0.23)
Cu	64 +/- 1.5 (11.7 - 4630)	11-14 (5.3 - 4.5)	41-68	4.6-5.3	25.3 +/- 5.9	2.3 (2.8)	53.9 +/- 6.1	(5.8 +/- 0.9)
Mn	1879 +/- 385 (n/a)	465-498 (308-244)	265-715	170-650;	108 +/- 28	108 (113)	n/a	n/a
Pb	216.0 +/- 34.2 (18.1 - 1075.0)	2.4-2.8 (4.7-3.6)	35-189	10-15	4.4 +/- 1.3	0.7 (1.0)	74.3 +/- 8.1	(5.0 +/- 0.5)
Zn	200 +/- 29 (19-743)	72-78 (77-59)	85-181	160-409	114 +/- 47	n/a (214)	359 +/- 46	(65 +/- 12)

367  
 368 Leaves are important but not the only parts of plants where metals accumulate and different metals  
 369 accumulate in different proportions in different plant tissues (Rana and Maiti, 2018). For Birch, the  
 370 Zn accumulation sequence is branches<fine roots<large roots <stem< leaves and, for Alder, fine  
 371 roots<large roots<stem <leaves<branches. Mn accumulates more in the fine roots than leaves of  
 372 Birch while for Alder the Mn sequence is large roots<fine roots<stem<leaves<branches (Butkus and  
 373 Baltrėnaitė, 2007). Pb accumulates most in the branches of Alder but in the leaves of Birch while, in  
 374 both species, Cu accumulates more in leaves. Rosselli, Keller and Boschi (2003) report that while  
 375 Betula and Salix transfer Zn and Cd into leaves and twigs, Alnus tends to exclude them from above-

376 ground tissues. They conclude that it is not possible to convert metal uptake in leaves to uptake of  
377 the tree from the soil. So, the foliar data from the Varteg does no more than demonstrate that trees  
378 are directly involved in the uptake of metals, and that there are differences in the uptake of different  
379 metals by different tree species. They do, however, support the argument for using mixed species  
380 plantations in forest phytoremediation.

381 More importantly, trees are not the only active biological agent involved. Forestation provides the  
382 soil with a sheltering canopy, a biologically active forest floor and litter layer, a rhizosphere of roots,  
383 fungal mycorrhiza and associated organisms, collectively a biochemical reactor that controls the  
384 mobilisation of metals and, separately, uptake by trees (Richter, 1987). Successful reclamation  
385 depends on the formation of a living soil system. At Varteg, results show that, while humus  
386 formation remains at an initial stage, there is a large and active microflora, especially in those plots  
387 where the trees have received fertiliser. Here, Noustorova et al (1999) recorded a total microflora of  
388  $196-286 \times 10^3$ g, of which >80% were ammonifying bacteria. Earthworm biomass is also greater under  
389 Varteg's trees than outside the plantations (Plamping et al., 2017). The reduction of soil metal  
390 loadings is a system process; many organisms may be involved.

391 Birch, generally, has a higher loading of metals in its leaves than Alder, which has, occasionally, been  
392 called a metal excluder, despite many studies, including this one, showing it capable of significant  
393 metal uptake (cf. Gamalero et al., 2009; Lorenc-Plucińska et al., 2013). The key observation is that  
394 these species take up different amounts of different metals, which means that mixed plantings may  
395 be most effective for phytoremediation.

396 Metal loadings in the leaves collected at Varteg are not unusually high apart from Mn (Millaleo, et.  
397 al. 2010; Migeon, et. al. 2010) and Cd and Zn in Birch (Boregard and Rydin, 1989; Butkus and  
398 Baltrėnaitė, 2007). In northern France, Mignon et al. (2009), investigated 25 woody species grown

399 on Cd, Zn and Pb contaminated land and found the highest accumulations of Zn and Cd in Poplar  
400 leaves (*Populus tremula* × *Populus tremuloides*), Pb concentration low across all species, and that  
401 Birch (and Oak – also planted at Varteg) accumulated more Mn than other woody species. It is  
402 possible that the characteristics of mine spoils encourage relatively high metal uptake. Borgegard  
403 and Rydin (1989) report on heavy metal uptake in Birch in a soil cover over Cu tailings and found that  
404 the leaves contained more Zn, Pb and Cd than comparable uncontaminated soils; the BL07 test show  
405 that the same is true for Mn, Cd and Zn on the Varteg plots. However, in this study, foliar metal  
406 loadings increase over time whereas soil metal loadings decrease.

407 There is a well-known, albeit complex, positive association between the uptake of Cd and Zn which  
408 may be species dependent (Liu et al., 2013). Mn has been commended used as a soil amendment  
409 for the remediation of metal contaminated soils and, here, there is a significant positive correlation  
410 between Zn in soil and Mn uptake, while (McCann et al., 2015). Rana and Maiti (2018) also find a  
411 correlation between Cu and Pb in Eucalyptus. High levels of Mn are found in soils with low organic  
412 matter and soils that are easily waterlogged, which is certainly the case at Varteg, and also in soils  
413 with pH <5.5 (Schulte and Kelling, 1999). Soil pH is thought to affect the availability of Zn more than  
414 any other factor, with maximum Zn availability at pH 5.4 (Schulte, 2004). Similarly, low pH facilitates  
415 the availability of Cd in soil (Kirkham, 2006). On the Varteg pH is generally in the range of 5.2– 5.7,  
416 which may help explain the uptake of these metals.

417 In sum, these decreases in soil metal loadings may be attributed to two processes. First, the  
418 mobilization effected by organic activity may transform the metals into forms that are easily carried  
419 away by leaching, subsurface and surface runoff. Dispersed into the wider environment, these  
420 contaminants are, here, rendered undetectable by dilution (Haigh and Kilmartin, 2017). Second is  
421 the uptake or absorption of metals by leaves, other plant tissues, and/or by the trees fungal and

422 rhizospheric associates (Liu et al., 2013). Some of this mobilised metal may be recycled into the soil  
423 by the decay of organic tissues, while some may be bio-transformed into inert forms and/or held  
424 outside the soil within the standing biomass (Sinha et. al., 2007). Indeed, the input of organic matter  
425 by trees could counter the mobilisation of some metals including Pb, which can form insoluble  
426 complexes with organic matter; this is why the residence time of Pb in rich organic soils can be very  
427 long (Manceau et al., 1996). However, if this were the case here, of course, the result would be  
428 enrichment of the top-soils by inputs of metals taken up from the buried minespoils. In fact, metal  
429 loadings decline and the likely reason is leaching into the soil solution and surface or near –surface  
430 runoff (Ross, 1994). However, given high rainfall, runoff and ambient metal pollution levels offsite, it  
431 is probable that any contribution to off-site pollution loadings is too small to be detected (Haigh and  
432 Kilmartin, 2017).

## 433 **6. Conclusion.**

434 In coal mine-spoil derived soils of the same character and age, the plantation of mixed woodland  
435 results in large, significant and progressive decreases in the soil loadings of five key metal  
436 contaminants. As the age of the tree plantation increases, the level of soil metal contaminant  
437 loading decreases progressively, so confirming that forestation ameliorates soil metal loadings.  
438 Here, soil loadings of five key metals (Zn, Cd, Mn, Pb and Cu) decrease, significantly, under *Alnus*  
439 *glutinosa* (L. Gaertn) (Common Alder) and *Betula pendula* (Roth) (Silver Birch). Fourteen years after  
440 tree planting, soil metal loadings, relative to land left under grass, decreased by 52% for Cd (0.043  
441 mg.kg<sup>-1</sup> per year), 48% for Cu (2.1 mg.kg<sup>-1</sup> per year), 47% for Zn (7.3 mg.kg<sup>-1</sup> per year), 44% for Pb.  
442 (7.1 mg.kg<sup>-1</sup> per year) and 35% for Mn (45 mg.kg<sup>-1</sup> per year). Foliar analysis indicated that trees were  
443 directly involved in for drawing down soil metal loadings for all five of the metals. There were  
444 significant positive correlations between the age of the trees and the amount of Mn, Zn and Cd



445 (Alder only) in leaves. However, there was little other evidence for metal accumulation on site and it  
446 is probable that most of the mobilised metals were dispersed into the larger environment by runoff  
447 and leaching (cf. Haigh and Kilmartin, 2017). More generally, differences in the metal uptake of  
448 Birch and Alder suggest that mixed plantings may be more effective than monospecific for forest  
449 phytoremediation.

450 Here, in South Wales, the forestation of marginally contaminated soils on reclaimed former surface-  
451 coal-mine soils has resulted in a significant decline in soil metal loadings on even on the most  
452 recently planted test sites. Measurement from soils across the full 14 year forestation  
453 chronosequence found that metal levels declined significantly ( $p= 0.003$  to  $p=0.0002$ ) through time.  
454 This paper has demonstrated that forestation with *Alnus glutinosa* (L. Gaertn) (Common Alder) and  
455 *Betula pendula* (Roth) (Silver Birch) is an effective means of (Cd, Cu, Zn, Pb and Mn) metal  
456 remediation on the moderately contaminated lands produced by opencast coal-mining in South  
457 Wales.

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