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Ecological Effects of *In Situ* Sediment Contaminants

Consequences of environmental contamination by lead mining in Wales

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Abstract

Lead mining in Wales originated before the Roman Occupation. The main active period was from 1750–1900 when zinc and copper were also mined and during this period only simple and inefficient ore processing methods were available. Consequently large amounts of copper, lead and zinc compounds were lost to the environment and have since become incorporated in sediments and soils. Locally, pollution may still occur from drainage from abandoned mines and by mobilization of mine tailings. This paper describes the present state of Welsh rivers and reviews the distribution of contaminants in sediments and soils. The uptake of heavy metals by plants and the consequences for human health are also discussed.

Introduction

The British Isles were once among the major producers of non-ferrous metal ores but after 1880 the industry declined in the face of foreign competition and, except for tin, British ore output is now negligible (Richardson, 1974). Evidence of this once prosperous industry can be seen in many parts of Great Britain especially Wales. In every county in Wales there are abandoned workings with their attendant waste heaps most of which are still bare of vegetation. In addition to the visible reminders of past industrialization there is also an invisible inheritance in the form of widespread contamination of soils, sediments and biota by toxic metals which were released to the environment when the mines were working. This paper presents an overview of research on heavy metal pollution in Wales.

Heavy metals may be defined as those having a metallic density $>6\ g \cdot cm^{-3}$. Chief concern focuses on Cu and Zn, which are essential micronutrients but may be harmful when present in large concentrations and on Cd, Hg and Pb, which have no known beneficial metabolic role but are known toxins. These metals are ordinarily present in rocks, sediments and soils but locally may become concentrated in rocks as ore bodies and generally in the environment through pollution as a consequence of mining the ores.

Lead and zinc ores in Wales normally occur as veins or lodes associated with faulting in the host rocks (Imm, 1959). The main carriers of the veins are Ordovician and Silurian sediments and Carboniferous limestone. Associated with these ores are the gangue minerals quartz, calcite and barite. Galena (PbS) and sphalerite (ZnS) were mined extensively in Wales, whereas chalcopyrite ($CuFeS_2$) was less common and the most notable mine was at Parys Mountain, Anglesey. Gold was mined at a number of localities which form a broad belt north of Dolgellau. Further south an isolated mine at Pumpsaint, Dyfed was an important producer during the Roman Occupation.

The major elements in the ore minerals are invariably accompanied by minor or trace amounts of other metals. It has been known since prehistoric times that silver can be extracted from lead after galena has been smelted and many of the mines in Wales were opened primarily to obtain silver. Other important constituents are antimony, arsenic, bismuth, cadmium, gallium and germanium. Davies (1983) has provided fuller accounts of the accessory metals in ore minerals. The important point is that these 'guest' metals are also potential contaminants when ores are milled and smelted.

Mining in Wales predates the Roman invasion in AD 43. It virtually ceased during the Dark Ages (5th to 11th centuries) and became economically important again after the 15th century when there was a growing demand for silver for coinage and lead for armaments (Richardson, 1974). The industry peaked in the middle decades of the nineteenth century and only a few mines were still worked during this century. Some 313 base metal mines are recorded in Wales and adjacent areas of England. Figure 1 illustrates the main mining areas of England and Wales. Figure 2 illustrates the distribution of lead mines in the Principality.

The techniques for separating ore from rock remained broadly the same throughout the whole mining period and entailed total reliance on the use of water. Water wheels drove the ore crushing machinery in most Welsh mines (Trevithick's Cornish steam engine never became popular in Wales) and the lighter rock was separated from the denser ore by hydraulic action in jigs and buddles. The waste water from the processing machinery carried both dissolved metal compounds and suspended ore particles and was released into rivers without pretreatment.

At first, smelting took place near the mines but diminishing supplies of wood for charcoal in the eighteenth century led to the development of specialised smelting centres near the coalfields of north and south Wales. There were 20 major smelters just outside Swansea in the Tawe valley and 11 smelters near Holywell along the Dee estuary. Thus, Wales was one of the major metallurgical areas of the world a hundred years ago. Its smelters handled ore not only from other British mining fields but also from abroad; e.g., the mines around Fredericktown in south eastern Missouri sent their copper concentrates to Swansea (Richardson, 1974; Beynon & Betteridge, 1979; Chadwick, 1981).

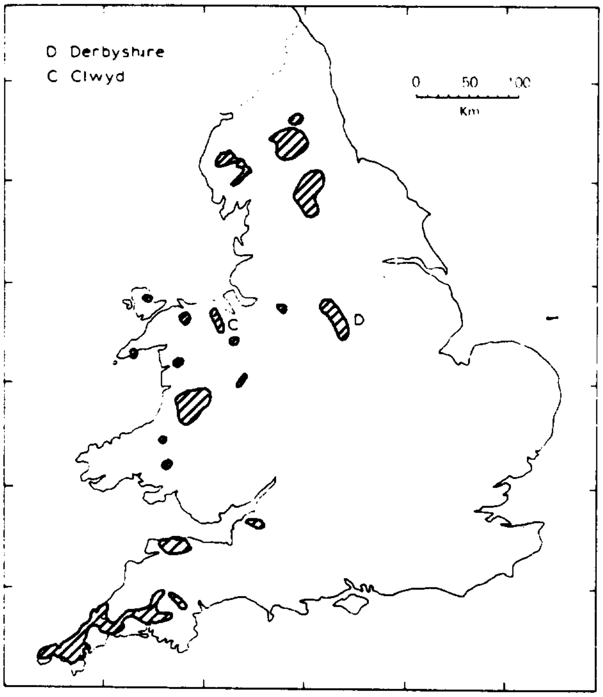

Fig. 1. Generalized locations of lead-zinc-copper mining fields in England and Wales.

Although the ore dressing and smelting process improved with time they were always inefficient. In the middle of the nineteenth century, farmers living in the Aberystwyth area complained that mine waste was deposited on fields in valley bottoms when local rivers flooded and was ruining their land. The consequence was an enquiry by the Rivers Pollution Commissioners. In their report (Frankland & Morton, 1873) they remarked that 'Lead mining is, in many instances carried on in an extremely slovenly and wasteful manner... we cannot doubt that reckless waste of valuable materials is permitted in many mines'. They analysed waste from jigs and concluded that 6.8 tons of ore were lost in every 100 tons of waste and calculations from their data lead to the conclusion that approximately 35% of the ore raised to the surface was lost to the environment during ore dressing. The subsequent smelting process was also inefficient. Overall, it can be estimated that for every ton of silver-free lead which was produced as much as 2 tons may have been lost to the environment (Davies, 1980).

The toxic nature of lead compounds was well understood and there was a wide variety of local names in Great Britain for lead poisoning: potter's rot, Devonshire colic, painters' colic, lead palsy, belland (Derbyshire) and *y ballan* or *y belen* (Wales). Yet the mining industry was entirely unregulated until 1876 when the Rivers Pollution Prevention act (39 & 40 Vict.) passed into law. But it came too late to be effective in Wales since the industry was then in a state of rapid decline. The environmental damage had been done.

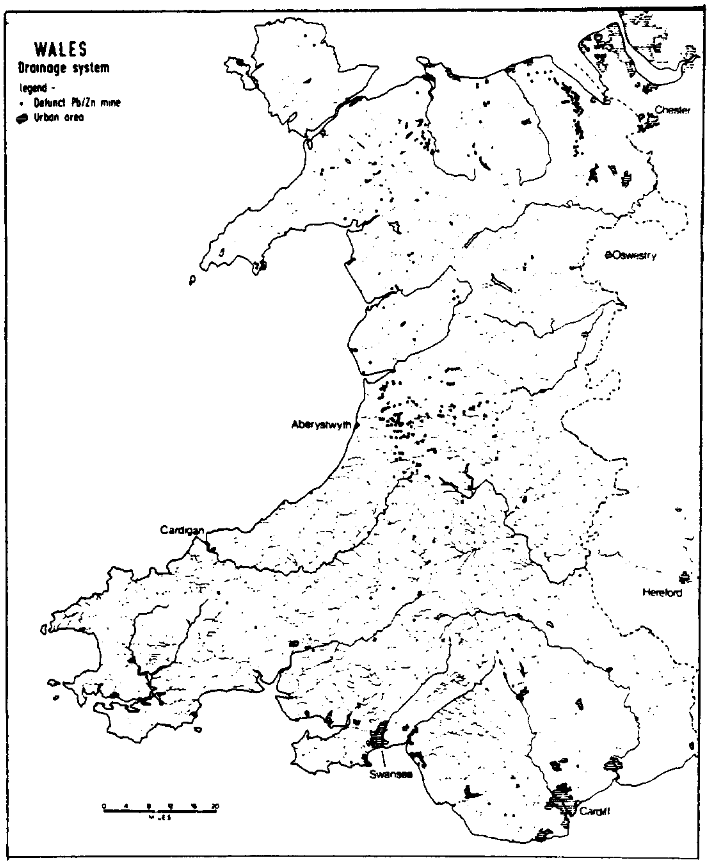

Fig. 2. Distribution of old lead-zinc mines in Wales superimposed on the drainage system of the country. Selected urban areas are also shown.

Pollution of rivers and sediments

The rivers of Ceredigion, especially the Ystwyth, were seriously polluted prior to the Rivers Pollution Act of 1876 and their reputation as first class salmon and trout rivers had been destroyed. Scientific investigations of river pollution and its consequences did not start until after the Great War of 1914–1918. In one of the earliest papers, Carpenter (1927) demonstrated that dissolved lead caused the formation of mucus on the gills of fish which then suffocated them. Newton (1944) reported that stickleback (*Gasterosteus aculeatus* L.) was the species most sensitive to lead pollution locally.

There was a general improvement in water quality after 1876 throughout Wales. The Ystwyth was the worst affected river and the slowest to recover. A survey of the lowest reaches made in 1919 (Carpenter, 1924) revealed that the fauna was restricted to 9 species, mostly insects, and the water lead concentration was 0.4 mg/L. By 1922 the number of species had increased to 26, again mostly insects, and the lead concentration had fallen below 0.1 mg/L. By 1939 the number of species had increased to 63 (but no fish) and the lead concentration was below 0.05 mg/l (Lauri & Jones, 1938; Jones, 1940). In 1975 fish had returned to most of the river but were still absent from a 3 km stretch below the Cwm Ystwyth mine. The river Rheidol recovered much faster and an account of this has been given by Jones & Howells (1973). They reported that dissolved lead has decreased from a range of 0.2–0.5 mg/L in 1919–1921 to <0.04 mg/L in 1971–1972. Zinc concentrations appear not to have decreased. Trout were first caught again in 1952 but salmon were not reported until 1970. More recent work has been concerned with the dynamics of the rivers and their dissolved constituents. In particular, the relationships of solute concentrations and river stage have been investigated. Grimshaw *et al.* (1976) have emphasized the importance of recording discharge at the time of sampling. In the case of zinc they concluded that concentrations are highest at low flows and during the initial stages of storm run-off. The annual yield of zinc for the Ystwyth in both 1973 and 1974 was 72 tonnes. Other Welsh rivers are still polluted to a greater or lesser extent. Abdullah & Royle (1972) reported that many rivers draining the old mining districts are still mildly polluted because of mine drainage and the release of solutes from waste heaps. Tributaries of the Conwy drain a group of lead mines near Llanrwst; Thornton *et al.* (1975) have reported up to 30 μg Pb/L and 1230 μg Zn/L. In an oyster hatchery in the estuary, 18 km below the mines, these dissolved metals caused intermittent failure of the larvae to develop and settle (Elderfield *et al.*, 1971).

There have been periodic setbacks in the recoveries of Welsh rivers especially when old workings are accidentally breached (Newton, 1944). The adit drainage from mines can represent a continuing significant point pollution source and may necessitate special treatment. In the Rheidol valley the drainage from the Cwm Rheidol mine is passed through a static limestone cleansing system which was designed both to raise the pH of the drainage water and to precipitate lead on to the limestone chippings (Treharne, 1962). However, the efficiency of this unit has deteriorated in recent years particularly since an incident in 1969. In October of that year the entrance plug to Number 9 level was accidentally breached during exploratory prospecting and caused a major fish kill in the river Rheidol. This incident illustrates the point that although a satisfactory effluent quality can be ensured during the working life of a mine, control is much more difficult once a property has been abandoned.

Although there is a general consensus that water quality continues to improve, the situation with regard to river sediments is much more complex. A floodplain is constructed partly by overbank sedimentation during flood inundation and partly by the deposition of material in point or channel bars within the river channel. In the Ystwyth, the floodplain is constantly renewed through meander development when outer banks are eroded and inner banks build up. Davies and Lewin (1974) investigated a meander loop in the Rheidol valley which had grown over 130 years by both lateral expansion across the valley floor and by down-valley enlargement. Using earlier survey material, it was possible to allocate areas of the floodplain to one of four age zones all formed by marginal channel accretion rather than over-bank deposition. Lead contamination was worst in the oldest section, contemporaneous with maximum lead mining activity, declined with increasing juvenility but was appreciable even in most recent sections. But the decline in zinc contents of the sediments was much less marked and was thought to indicate continual entrappement of Zn^{+2} in the sediments. Lewin *et al.* (1977) have established that channel sediments account for the highest metal values and very high levels away from the present river channel are often identified with old abandoned channels.

Much of the published work is concerned with lead and zinc but some papers have shown that the other metals found in ores may also contaminate water or sediment. Davies (1976) demonstrated that where the lead contents of sediments were raised as a consequence of pollution so were the mercury levels. Floodplain soils in an uncontaminated river valley contained up to 0.09 μg Hg/g soil whereas in the Ystwyth valley the mean concentration was 0.247 μg Hg/g and the maximum was 1.78 μg Hg/g. In the case of silver, Jones *et al.* (1983) calculated the background concentrations of silver in soils in west Wales to be 0.01 – 0.166 μg/g whereas in soils developed in Ystwyth valley floodplain sediments the median silver content was 1.2 μg/g with a range of 0.11 – 6.8 μg/g. In contaminated stream waters in the river Ystwyth catchment, silver in the filtered (<0.45 μm) water contained 3 – 230 ng Ag/L compared with <1 ng/L in uncontaminated waters (Jones *et al.*, 1984). The authors also proposed that *Scapania undulata* was useful as a biological monitor of dissolved silver since it concentrated the metal from water and there was a very good linear relationship between plant and water contents.

Soil contamination

Aquatic ecosystems are still polluted by drainage from old mines and erosion of mine waste or tailings still contributes to river sediments. But the main impact of mobilisation of metal-rich materials from mine wastes is on the terrestrial ecosystem (Davies *et al.,* 1983).

The waste heaps on old mine properties are often bare of vegetation. They have poor moisture retention properties and their steep sides make them unstable. The major plant nutrients are lacking and they contain excessive contents of heavy metals, especially zinc. The plants which do colonize these heaps tend to germinate in rabbit or sheep droppings and they are usually metal tolerant ecotypes. Bradshaw (1970) and Antonovics *et al.* (1971) have discussed the emergence of metal tolerance in certain species and have listed the plants where this property has been identified. Bradshaw & Chadwick (1980) have described how tolerant ecotypes may be used to reclaim and revegetate metal contaminated soils.

A lack of vegetative cover renders mine tailings very prone to mobilization. Wales is generally hilly or mountainous and spoil heaps are often located on slopes. Although visible toxic effects rarely extend beyond a few meters from the waste material, analysis of soils taken from fields below the tips shows that pollutants may be transported for hundreds of meters downslope (Davies, 1977). Windblow is also a mechanism whereby toxic tailings can be transported to neighbouring agricultural land. Davies & White (1981) reported an investigation at a mine near Aberystwyth. Most of the <2 mm fraction of the spoil was of sufficiently small diameter to move by saltation and the material <2 μm approximated to the composition of PbS. Deflation was caused primarily by dry northerly winds and movement of spoil could be detected as far as 1800 m downvalley. Similar findings have been reported by Jones *et al.* (1985) with particular reference to silver.

In contrast with the continuous interest in the ecology of polluted Welsh rivers after 1920, the only report of soil contamination prior to 1968 was a paper by Griffith (1919) in which he concluded that many cases of soil infertility could be ascribed to lead pollution. Davies (1968) drew attention to the persistance of lead residues in soil and to the presence of excessive amounts of other metals, especially cadmium, copper and zinc. Alloway & Davies (1971) found that all the soils collected from riverside fields throughout the Ystwyth valley were contaminated by lead which ranged from 90 to 2900 μg/g compared with 12–72 μg Pb/g soil in the nearby, uncontaminated Aeron valley. Analysis of soils collected from cross-sections across contaminated valleys has confirmed that water-borne contaminants are the principal source of lead and other metals (Alloway & Davies, 1971; Davies, 1976; Jones *et al.,* 1983) in alluvial soils. Lewin *et al.* (1977) concluded that the general order of severity of lead pollution depends on sediment type, namely, channel active since 1800 > channel active before 1800 > non-channel flood sediments > slope deposits (scree or head).

The nature of the ores which were mined exerts a strong influence on the nature of soil contamination. In west Wales, the principal ores were those of lead and zinc and most zinc production was restricted to late in the nineteenth century. Consequently, lead is the major soil and sediment contaminant together with silver and mercury which occur as guest elements in galena. Zinc is also important and levels of cadmium are elevated (Alloway & Davies, 1971; Davies, 1976; Jones *et al.,* 1983). In Anglesey, near the old Parys Mountain copper mine, Alloway & Davies (1971) reported up to 1405 μg Cu/g soil compared with a background concentration of <30 μg Cu/g soil. In north-east Wales, the zinc ores are especially rich in cadmium and Davies & Roberts (1975, 1978) found the following maximum metal concentrations in soil: Pb = 48000 μg/g, Zn = 50000 μg/g and Cd = 540 μg/g.

Three general processes have contributed to metal contamination of soils in Wales: 1. flood inundation has affected alluvial soils in old mining areas;

2. atmospheric transport brought fallout from smelter smoke to agricultural land and continues to mobilize the fine material in tailings; and, 3. gravitational transport downslope from spoil heaps has affected many fields on hill slopes. The consequence is a widespread and complex pattern of soil contamination in each mining region. Davies & Roberts (1978) published computer derived isoline maps for metals in soils of north-east Wales. Comparison of the mapped patterns with the distribution of old mines and smelters allowed the explanation of these patterns in terms of the three transportation mechanisms listed above. A simplified version of these maps is given in Fig. 3.

Biological implications of soil contamination

There is a general although not simple relationship between the heavy metal content of soils and of plants growing on those soils. Uptake through the roots is influenced by soil parameters such as acidity or redox potential and different species absorb metals to different extents. But in general, the higher the heavy metal concentration of the soil the higher will be the concentration in plants. Cadmium and lead are toxic to animals and people and the accumulation of these metals in foodstuffs raises the question whether human health might be impaired by ingesting small amounts of toxic metals. Davies & Roberts (1975) grew radish (*Raphanus sativus* L.) in gardens in the Halkyn Mountain area of north Wales. They demonstrated that the plants from some gardens contained more than the then British legal limit for lead in food (2 mg/kg fresh weight) and several of the plant cadmium concentrations were high enough to cause concern. Subsequently, Davies & White (1981) confirmed these

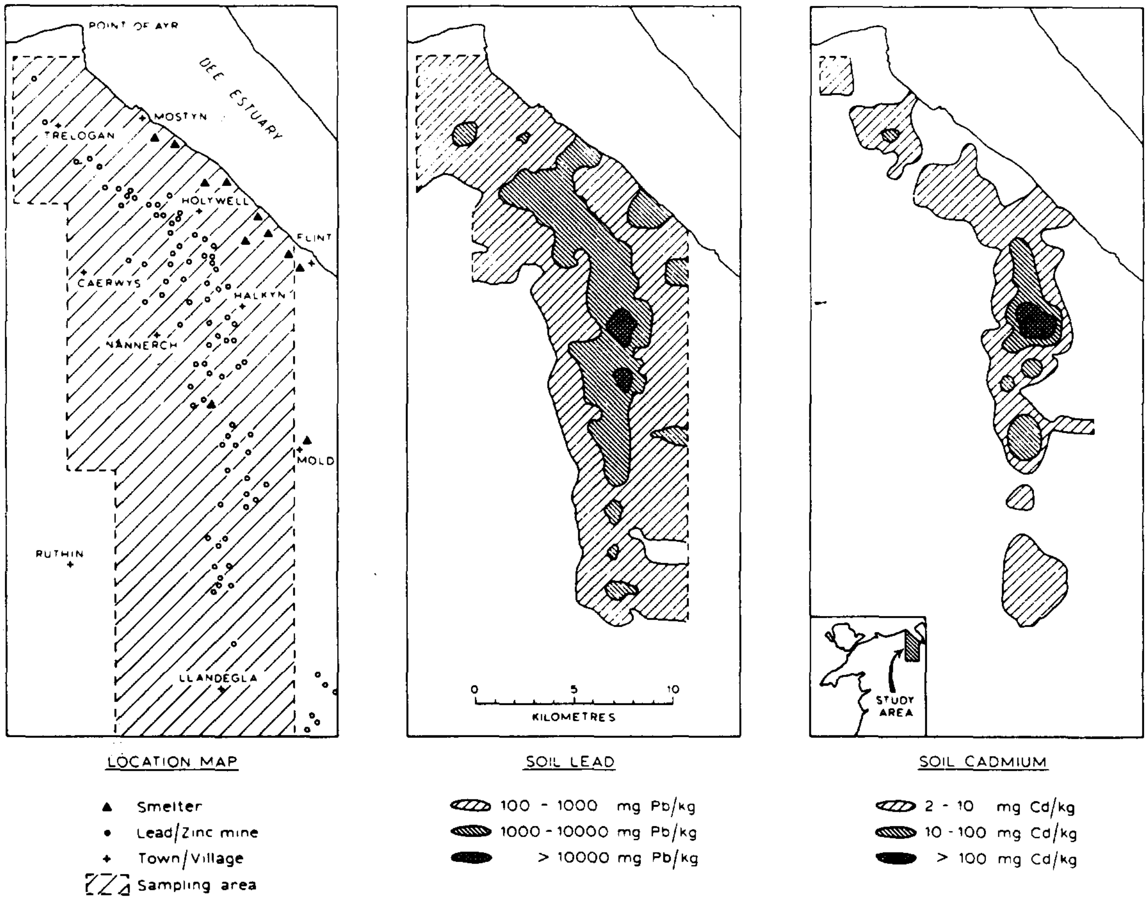

Fig. 3. Distribution of cadmium and lead-contaminated soils in the Halkyn Mountain area of Clwd, north-east Wales.

results for other vegetables. They noted that lead tended to be retained in plant roots but enough could still be translocated to edible leaves to exceed the legal limit for food lead. Cadmium was much more mobile and leaf concentrations were especially high in lettuce. The medical implications of these data were investigated by Gallacher *et al.* (1984). Women residents in the area had blood lead concentrations that were about 50% higher than those of women residing in a control area 30 km to the west. Blood lead concentrations were related to the consumption of home grown produce. Those with the highest consumptions had blood lead concentrations that were 28% higher than those of women who consumed no locally grown vegetables. The data suggested that an increase in soil lead of 1000 μg/g was associated with an incease of blood lead of about 42 μg/L.

Conclusion

This paper has presented only an overview of research in Wales. Heavy metals in the Welsh environment have been the subject of ecological, geographical, geological and medical research for over 60 years. As a consequence, the lead mining areas of the Principality are now widely regarded as classic sites for the study of environmental pollution. Many of the contaminated areas are very well documented and provide some of the best field areas in the world for the study of environmental pathways of toxic metals. The techniques which have evolved for the study of sediments, soils and biota and the data derived from the application of these techniques have an international applicability. The lessons learned from the Welsh experience can help safeguard those environments which are now undergoing industrialization or where new metals mines are being opened.

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