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# The Environmental Impact of Industrialisation in South Wales in the Nineteenth Century: ‘Copper Smoke’ and the Llanelli Copper Company

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## SUMMARY

The development of industrial towns on the south-western side of the south Wales coalfield, such as Llanelli, Swansea, Neath and Port Talbot, was based to a large extent upon the growth of metal industries. This paper focuses on a major environmental problem associated with this development: that of ‘copper smoke’ produced by the region’s important copper smelting industry. The first part of the paper discusses the social, economic and legal history of the copper smoke problem, and the technology employed to control emissions. The second part deals with the chemical composition of copper smoke and estimates the output and dispersion of copper smoke and other metal fumes from the Llanelli Copper Co.’s works in the 1860s using a Gaussian plume model. It is argued that the extent of long and short range deposition of sulphur and heavy metals from these works was significantly high and that the industry was therefore a major polluter, particularly when copper smelting was at its peak in the region in the mid- to late- nineteenth century.

## 1. INTRODUCTION

The development of industrial towns on the south-western side of the south Wales coalfield provides environmental historians with an interesting case study of the impact of early industrialisation on the environment. A feature of towns such as Llanelli, Swansea, Neath and Port Talbot is that their development was based to a large extent upon the growth of metallurgical industries. Indeed, by the mid-nineteenth century the scale of production and the variety of metals

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manufactured in these places led south Wales to be regarded as the world's leading centre for industrial metallurgy.

The economic transformation of this region inevitably posed serious environmental problems. A geochemical survey of stream sediments conducted in the 1970s indicated high levels of arsenic, copper, lead, zinc and other elements given off by smelting in south Wales (Wolfson, 1978). Worst affected was the Lower Swansea Valley, an area so badly scarred by nearly 300 years of non-ferrous metal manufacturing that it became the subject of a major study and reclamation project in the 1960s and 1970s (Hilton, 1967; Bromley and Humphreys, 1979; Lavender, 1981; Bridges, 1988; Bridges and Morgan, 1990). While the Lower Swansea Valley Project provides the most dramatic and well-documented example of the cumulative effects of metal working on the local environment, it is less easy to assess the pollution problems faced by our forebears in the nineteenth century, or to estimate the relative contribution of early metal industries to long term and long distance environmental degradation (cf. Quinn, 1989). Few attempts were made to measure pollution levels until relatively recently, and the data required for accurately estimating emissions a century or more ago are rarely available. Furthermore, the variety of industrial activity in south Wales, the complexity of the processes employed in the metallurgical industries, and the lack of surviving records for the majority of the metal works, makes it particularly difficult to assess pollution levels in this region in the nineteenth century. The purpose of this paper is not therefore to attempt to measure aggregate pollution levels in the metal-working towns, but to focus on one aspect of industrial pollution – 'copper smoke' – for which reasonable data are available. The lack of surviving records of the majority of the copper works also precludes a full survey of the copper industry, and so after a general discussion of the problem, the second part of this paper attempts to measure the output of copper smoke and fumes from associated lead and silver production from one of the largest and better documented non-ferrous metal smelters, the Llanelli Copper Co.

## 2. COPPER SMOKE

Copper smoke was recognised by experts such as Angus Smith, the first Chief Alkali Inspector, as one of the worst forms of industrial pollution in nineteenth century Britain (PP, 1872). The output of copper smoke was considerable at times in Lancashire, Tyneside and Anglesey, but most especially in south Wales, which dominated world copper production in the first half of the nineteenth century. While visitors to Swansea commented that copper smoke ascended 'not in "volumes" but in "encyclopaedias"' (Hall and Hall, 1861), the French social scientist Frédéric Le Play, in an earlier career as Professor of Metallurgy at the Ecole des Mines, Paris, calculated in the mid-1840s that in a single year about

92,000 tons of sulphurous acid were projected into the atmosphere above Swansea by the copper smelters alone, ignoring the output of other industries and the domestic burning of coal (Le Play, 1848).

The environmental problem of copper smoke manifested itself in three ways. Firstly, the smoke was frequently inhaled by furnace workers and local residents, causing respiratory problems and concern over public health. The health issue was confused, however, by a lack of understanding of the chronic effects of the smoke on humans and by the absence of reliable or easily interpreted morbidity and mortality statistics; some experts even regarded copper smoke as beneficial to health (eg. Williams, 1854). A second, and more apparent, problem was the dry deposition of particles of copper, sulphur, arsenic and other pollutants which contaminated crops and pasture and which in turn poisoned grazing animals. The so-called *efrydd-dod* (smoke disease) became a contentious issue between farmers and industrialists. Thirdly, the sulphur dioxide contained in copper smoke reacted with the moisture in the air to form sulphurous ($H_2SO_3$) and sulphuric ($H_2SO_4$) acids to produce a potent form of acid rain which could have devastating effects. As one farmer complained, ‘it shrivels up the grass and the straw almost as if a dash of lightning had gone over it’ (*Cambrian*, 6 August 1858).

It was the damage to herbage which caused most concern. Not only did the pollution contribute to the destruction of the natural beauty of the land surrounding copper works to create ‘a volcanic appearance’ (Webb, 1812), but it also affected the productivity of nearby agricultural land. Copper smelters faced numerous complaints from landowners and tenant farmers over damage to land, and land values were affected (Rees, 1980-1). For instance, an assessment of land values in Glamorgan in the 1860s showed they had fallen by half since an earlier valuation in smoke affected districts (*Cambrian*, 15 September 1865); while in the 1890s the owner of the large Margam estate, Miss Emily Talbot, was forced to reduce farm rents by between 35 to 75 per cent because of damage caused by the nearby Rio Tinto Co. works at Cwmavon (PP, 1894).

During the eighteenth and nineteenth centuries, various attempts were made to combat the copper smoke problem in south Wales. Local government intervention in the early eighteenth century established the principle of ‘zoning’ the industry on the outskirts of Swansea, so that the prevailing westerly winds took the smoke over wasteland to the east. This was the most effective form of control imposed upon the industry given that national legislation on smoke and noxious vapours introduced in the nineteenth century by-passed copper smelting. With regard to smoke legislation, an important distinction was made between copper ‘smoke’ and smoke produced by combustible fuels. In an 1846 Parliamentary report Sir Henry De La Beche and Lyon Playfair stated that ‘The emission of that which is improperly termed smoke from certain manufactories, such as the copperworks of Swansea, is another question, and involves very different considerations from the prevention of smoke from fuel’ (PP, 1846). The reason for this distinction was that copper smoke was not only the product of combustion but it contained furnace fumes for which no satisfactory technology had been developed to reduce emissions. This argument was also used by the House of Lords Committee on Noxious Vapours in 1862 and the Royal Commission on Noxious Vapours in 1878, whose reports formed the basis for the Alkali Acts of the latter part of the nineteenth century. Both bodies considered pollution from copper works and the methods used to deal with the problem, but judged that no economical remedy was available. This was in contrast to their findings on the alkali industry which could employ the relatively inexpensive technology of the Gossage tower to reduce hydrochloric acid emissions considerably. However, the Royal Commission did recommend that copper works should be monitored by the inspectorate in the hope that a technological solution would eventually be found. This recommendation was not taken up, but the inspection of copper and other metal smelting works was introduced in the Alkali Act of 1906 (which made no provision to enforce reductions in emissions) (Carter, 1964).

In the absence of legislative control over copper smoke, the motivation for copper smelters to deal with pollution came primarily from the threat of legal action or the economic incentive to recover by-products from the fumes. On several occasions smelting companies faced indictment for public nuisance, but the only successful prosecution occurred in Lancashire in 1770, when the Macclesfield Copper Co. was taken to court by the Liverpool Corporation and forced to relocate its works outside the town. Numerous private nuisance actions were taken against smelters over damage to private property, but the plaintiff rarely succeeded. The biggest success against a smelting company again occurred in Lancashire, when landowner William Tipping was awarded damages against the St. Helens Smelting Co. which was also forced to relocate its works. There are no records of similar achievements in south Wales (Newell, forthcoming).

Aside from any arguments that the courts became increasingly supportive of industry in nuisance cases (McLaren, 1983), problems occurred in identifying the source of pollution when there were so many potential offenders within close proximity, and in determining the extent of damage caused by pollution. Legal costs also discouraged potential plaintiffs from taking action. However, various references exist to out of court compensation payments, which is how the dispute between Miss Talbot and the Rio Tinto Co. was resolved, indicating that some redress was made for the damage caused in certain cases.

## 3. TECHNOLOGICAL RESPONSES

A number of firms did invest in methods to restrict the output of copper smoke. Most of these were associated with attempts to recover by-products such as sulphur and arsenic, or to restrict the loss of copper. The high sulphur content of certain copper ores had long attracted the attention of chemical manufacturers: a patent for making sulphuric acid from copper ores using hydro-metallurgical techniques was taken out as early as 1637 by a Capt. Thomas Whitmore, for instance (Woodcroft, 1969). In the 1840s William Gossage patented a method for collecting sulphur given off by copper ores using his condensing tower, although it was not taken up commercially (Allen, 1907). The most widely used technology involved connecting furnaces to chimneys by long flues and chambers where some of the arsenic, sulphur and copper would condense and could be recovered. During the eighteenth century it is probable that this method was used to recover sulphur for the manufacture of sulphuric acid at copper works in the Bristol area, and such apparatus was certainly in use at the copper mines on Anglesey by the 1780s (Day, 1973).

The search for a technical solution to the copper smoke problem was given a fillip in 1821 when a fund was established in Swansea 'for obviating the inconvenience arising from the smoke produced by smelting copper ores' (Anon., 1863). The fund provided prize money for the best invention to deal with the problem and a competition was held. The prize was not awarded, however, because it was judged that none of the designs submitted could fully alleviate the problem. But a system of flues and chambers with water showers for condensing gases designed by the smelter John Henry Vivian with the assistance of Michael Faraday and Richard Phillips was highly commended and received much publicity. Other companies did adopt this method (with or without the addition of showers), usually in conjunction with tall chimney stacks to disperse the smoke. Some of the constructions were extremely large and expensive, notably those built at the Llanelli, Pembrey, Spitty and Cwmavon works. At the latter a system was constructed whereby 2 furnaces were connected to a flue nearly a mile long which ran up the side of a large hill to a stack, the top of which was at a height of over 1,000 feet. The Pembrey stack was 270 feet high, the first of the Llanelli Copper Co.'s two large stacks, completed in January 1831, was 230 feet high, and the second Llanelli stack, which came into operation in 1861 and known as *Stac Fawr* (Big Stack), was 320 feet high (Percy, 1861). The system of culverts connected to *Stac Fawr* were erected at a cost of £7-8,000. The manager of the works claimed that 'the copper dust and very nearly all the arsenious fumes are condensed' by this system, which was described in some detail by Dr. Edward Ballard of the Medical Department of the Local Government Board:

> Most of the flues from the calciners and furnaces are conducted by long and capacious underground flues to the chimneys. But in what I may call the roasting-house, where all the roasters in use are ranged on one side and a number of ore furnaces on the other side, the flues rising from all these furnaces enter a capacious brick flue running through the length of the house on each side a few feet above the furnaces. Each of

> these flues terminates in a separate chamber, 20 feet long by 20 feet high, and 6 feet wide, which is partly underground and which is provided with a baulk or partition of brick rising up several feet from the bottom, against which the current of gas, &c. is made to beat, and by which deposition is favoured. The flues from these two chambers unite then in a flue 6 feet wide and 8 feet high, and this leads to a very long wide chamber, divided by partitions so as to constitute a zig-zag. From this flue conducts to another deposit chamber close to the foot of the taller stack. Deposit occurs in all these chambers and flues ... The deposit in these large flues contains on analysis 23.44 per cent. of copper, of which 19.59 are soluble in water, and 5.60 per cent. of arsenic. (PP, 1878-9).

The efficiency of such systems, which were used in a variety of metal works (Tucker, 1972), was nevertheless limited. Greater success was achieved by the use of high tension electricity in the flues to increase the rate of precipitation, but by the First World War Vivian and Sons was the only non-ferrous metal smelting company to have utilised this method (Carter, 1964).

John Henry Vivian and others recognised in the 1820s that the copper smoke problem might best be tackled by improved furnace technology (Anon., 1863). Most copper smoke was produced in the first process of smelting, known as calcination (see below). It was argued that it was theoretically possible to conduct this process and at the same time oxidise the sulphur driven off the ores to produce sulphuric acid in the furnace, thus eliminating sulphurous gas emissions at source. The chemistry was understood, but Vivian and others believed at the time that the technical problems in designing appropriate furnaces for commercial use were insurmountable. The basic problem was that the oxidation necessary to produce sulphuric acid was prohibited in existing furnaces by the presence of coal smoke, which caused a different chemical reaction to take place. It was thought that it was not possible to develop a commercially viable furnace that could keep the fumes emitted from the ores separate from the smoke given off by furnace fuel. Various attempts were made at overcoming this problem. Pascoe Grenfell and Sons went as far as installing furnaces designed by Herr Schafheutl of Munich in 1848 (Percy, 1861) and employing a process devised by Dr. Gurlt of Birmingham in the 1860s (Grant-Francis, 1881), but both failed to work satisfactorily and were abandoned.

Despite the technical difficulties, two types of furnace were devised which could overcome the problem, and both were used commercially to a limited extent from the 1860s. The first to be developed was the muffle furnace, which separated the copper ore fumes from the coal smoke, thus allowing the necessary reaction to take place. This furnace was brought to the attention of the 1862 Select Committee on Noxious Vapours by Peter Spence who went on to use his own patented design in the Goole Alum and Smelting Works (*Chemical News*, 20 September 1867). Versions of the muffle furnace were also used by Charles Lambert at the Port Tennant Works in Swansea and by Newton Keates and Co. in St. Helens (PP, 1878). The second type of furnace produced very little coal smoke, as it required fuel only to begin calcination. Once ignited, this furnace then used the sulphur in the ore as fuel, thus eliminating the presence of coal smoke and indeed significantly reducing the amount of coal required in calcination. This furnace was developed by Moritz Gerstenhöfer in Germany, where smelter smoke was also becoming a major problem (Schramm, 1990). Vivian and Sons obtained the British patent for the furnace, which received highly favourable reports in *The Times* (4 December 1865) as well as in the local press, being hailed as the solution to the copper smoke problem. Such claims were premature, however, as in practice both types of furnace had major limitations. At best, the furnaces could remove only 33 to 40% of the sulphur, and neither furnace was suited for smelting ores of less than 20% sulphur content, indeed the Gerstenhöfer furnace could not operate with a lower proportion of sulphur (PP, 1878). This gave these furnaces limited applicability to a smelting industry which at the time was handling an increasing quantity of imported ores with a low sulphur content. Furthermore, the furnaces were particularly expensive to install along with the other equipment required for sulphuric acid manufacture, such as lead chambers, steam boilers, concentrating pans and stills; and the Gerstenhîfer furnace could calcine only finely ground ore, which meant further expense on costly crushing equipment (PP, 1878).

On the commercial side, sulphuric acid was not a particularly valuable by-product. As a bulky, low value good, it could be sold profitably only to local buyers as it was obtainable cheaply elsewhere by the chemical industry, which was its main consumer. In south Wales the scope for selling acid was limited, the principal users being tinplate manufacturers who required the acid for 'pickling', or removing scale and impurities from sheets of iron prior to plating (Trubshaw, 1883). Vivian and Sons also opened their own superphosphate fertiliser works and alkali works to utilise the acid. At Lambert's Port Tennant works, the acid was used in the manufacture of copper sulphate. Lambert regarded this enterprise as unprofitable and Pascoe Grenfell decided not to introduce similar apparatus as he felt it was not worth the expense. Similarly, the Llanelli Copper Co. did not make use of the sulphur collected in the flues, which it considered would be a 'loss-making venture' (PP, 1878). Thus only three south Wales companies and one Lancashire company utilised the new furnaces. The technology portrayed as solving the copper smoke problem therefore had very little impact on the industry overall.

## 4. LLANELLI AND THE LLANELLI COPPER WORKS

While most growth of the south Wales copper smelting industry occurred around Swansea and Neath, four works were opened in the Llanelli district in the first half of the nineteenth century. The first, and most important, were the Llanelli Copper Co. works which began production in 1804. These were established by a partnership headed by a Birmingham industrialist, Charles Nevill, whose family was to have a long association with the works. The Spitty works were erected in 1809 on the western side of Llanelli; the Cambrian works were opened in 1830 but closed in 1841 and later converted in to lead-silver works; and the Pembrey works were erected in 1846, also to the west of the town (Evans, 1939). Llanelli was well located for copper smelting and other heat intensive industries since coal could be obtained cheaply from the south Wales coalfield. The town was also well served by good port facilities which was important given that all supplies of ore had to be imported. Much of the ore was shipped direct from Cornwall and Ireland or was trans-shipped from Swansea, where imported ore was sold at sales known as 'ticketings' (Newell, 1990).

During the first half of the nineteenth century, the industrial development of Llanelli was dominated by the growth of non-ferrous metal production. As well as producing copper, the copper works also smelted zinc for use in the manufacture of brass and Yellow Metal (another copper-zinc alloy used to sheath the wooden hulls of ships), and several lead-silver works were in operation, including those of the Llanelli Copper Co. The output of non-ferrous metals continued to grow until the 1870s, but then a slow decline set in. The copper industry was affected initially by the collapse of the Cornish mining industry in the 1860s and 1870s, although increased imports of ore and part smelted ore ensured that the industry survived into the twentieth century. The lead-silver industry also declined in the last quarter of the nineteenth century, but the fall-off in non-ferrous metal production was more than offset by the rapid growth of the tinplate and iron and steel industries in the district, which ensured that Llanelli continued to be an important industrial town (Evans, 1939). Overall, copper smelting in south Wales declined rapidly in the early twentieth century and had almost ceased by 1920 (Newell, 1990).

The diversity of industrial activity, and in particular the presence of lead and copper works and widespread use of coal, ensured that Llanelli possessed major sources of atmospheric pollution. Unfortunately, the absence of production data for many of the industrial works makes it impossible to assess aggregate pollution emissions for the town. However, the surviving records of the Llanelli Copper Co. for the period c.1850 to c.1870 provide sufficient information to make an assessment of the extent and nature of copper smoke emissions and of the output of fumes from the company's lead and silver works. The Llanelli Copper Co. also provides an interesting case study in that it employed high chimney stacks. Indeed, *Stac Fawr*, to which all of the calcining furnaces were connected, was for a time the highest chimney in England and Wales (see Figure 1).

It is noticeable that three of the four copper smelting works in the Llanelli area constructed extremely tall chimneys to disperse the copper smoke, whereas few companies did so in the Swansea district. This was possibly because the copper works in Llanelli were less favourably situated and smoke was more often blown

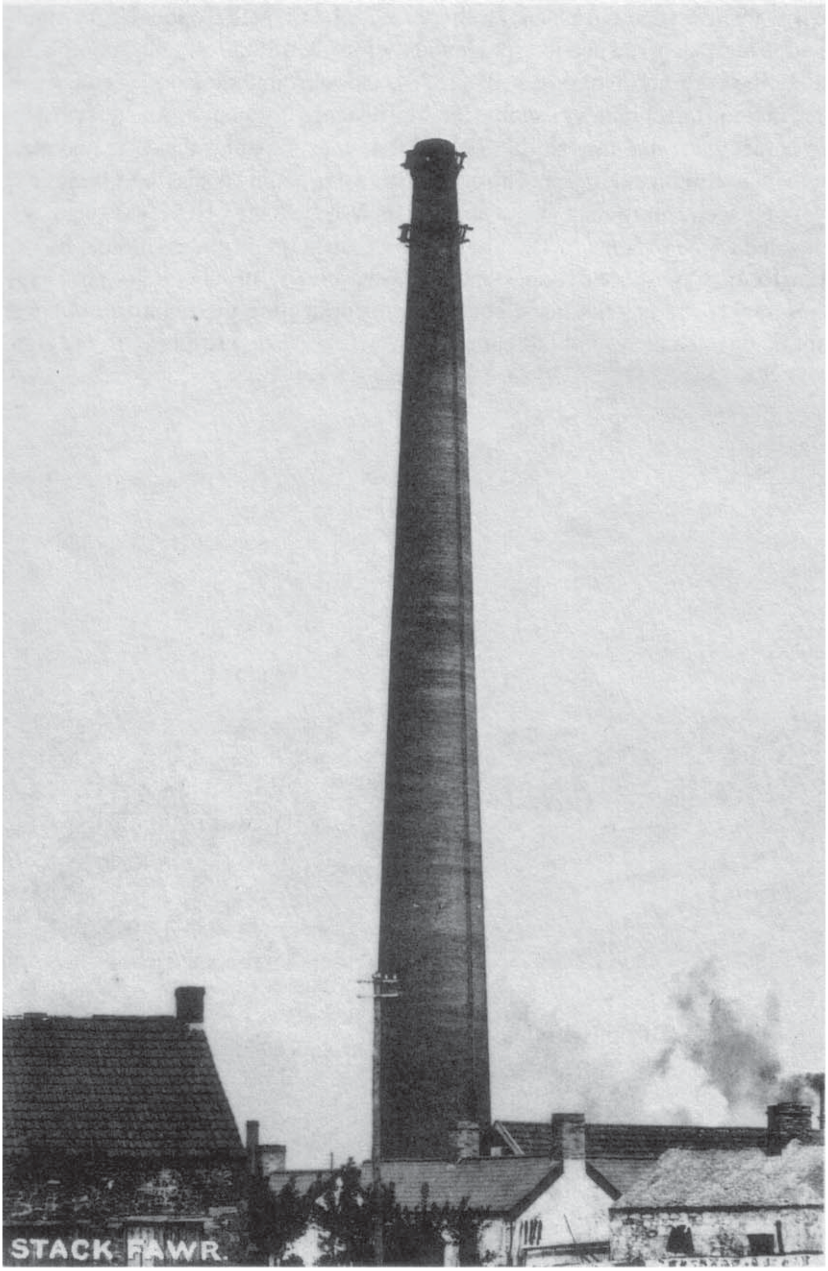


FIGURE 1. ‘Stack Fawr’, c.1894 (© Bodleian Libary, Oxford)

across the town and farm land (Evans, 1939). The Llanelli Copper Co. certainly made a number of out of court settlements before the 1860s following complaints about the burning-up of crops (PP, 1878). The surrounding land was relatively flat, making tall chimneys suitable for long distance dispersion. And it is perhaps no coincidence that the families behind the two companies that invested most heavily in equipment to deal with copper smoke in south Wales (the Vivians and Nevills) were prominent in local politics. When Henry Hussey Vivian was elevated to the peerage in 1893 he certainly made use of the claim that he had transformed 'poisonous copper smoke into a useful by-product' (Vivian, 1989). Presumably the Nevills made equal play of protecting the town from copper smoke through the use of tall chimneys whilst bringing prosperity to the area.

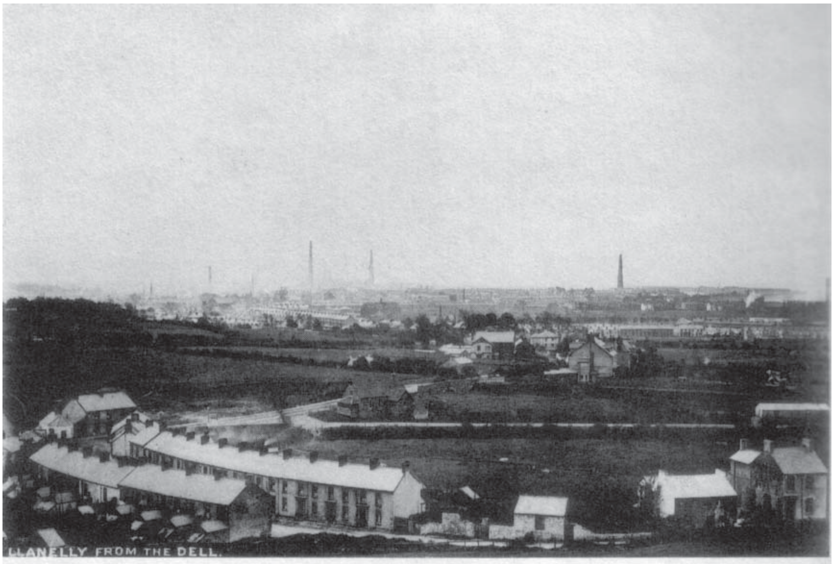


FIGURE 2. 'Llanelly from the Dell', c.1894. ©Bodleian Library, Oxford

## 5. CHEMISTRY OF COPPER EXTRACTION

During the eighteenth and nineteenth centuries what became known as the 'Welsh process' of copper smelting developed in the copper industry of south Wales. The chief characteristics of the Welsh process, which of course varied between different smelting works and was modified over time, were that it utilised coal-fired reverberatory furnaces and a wide range of ores of varying chemical composition. Details of the process can be found elsewhere (Napier, 1852/3; Percy, 1861; Vivian, 1823), but what is of relevance here is the chemistry of the Welsh process. The process was based upon four main stages, although in practice these may have been conducted in six or more separate operations in different furnaces. The stages were as follows:

### i. *Partial Roasting (or Calcination)*

In this process the prepared ore was heated to red heat to reduce its sulphur and arsenic content as well as to convert available copper to copper sulphide ($Cu_2S$). The large amounts of sulphur and arsenic driven off at this stage made calcination the principal cause of copper smoke.

From a consideration of the thermodynamics involved it is also possible that some metal sulphides were converted into oxides at this stage. For example, at 400°C zinc sulphide can be converted to its oxide, whilst the iron, copper, lead and silver sulphides remain unaffected. This is important because such oxides can be removed into the slag in the next stage.

$$2ZnS + 3O_2 \xrightarrow{400°C} 2ZnO + 2SO_2$$

### ii. *Smelting*

The product of this partial roasting (a calcine) was melted with a silica based (acid) flux. This separated any oxides present into the slag. The first smelt usually removed the amphoteric metals such as zinc, arsenic and antimony leaving a copper enriched mixture of ores, known as 'matte' or 'regulus', together with a slag. The matte comprised mainly $Cu_2S$ and FeS along with any other metals in the sulphide form which had not been converted into oxides in the partial roast. It was also important that the total proportion of copper present at this stage was not too high otherwise copper would have been lost into the slag, as predicted by equilibrium thermodynamics. In practice, a copper content of 13% or less was usual. By controlling temperature, any lead present could either be retained in the matte or transferred to the slag for reclamation. If a liquid slag was produced, tin could also be reclaimed. Careful control of temperature in the process was necessary to prevent the loss of lead, silver, and tin to the atmosphere.

### iii. *Converting*

The matte was again heated with a silica based flux, but this time with copious quantities of air and at a temperature (dictated by thermodynamics) of about 1200°C (Moore and Boyce, 1990). The aim of this process was to remove the iron from the copper and convert the $Cu_2S$ to Cu.

$$Cu_2S + O_2 \xrightarrow{1,200°C} 2Cu + SO_2$$

Metallic copper rather than $Cu_2O$ was formed by this process because of the mutual reduction reaction:

$$Cu_2S + 2Cu_2O \longrightarrow 6Cu + SO_2$$

This produced 'blister copper', known as such because the bubbling of $SO_2$ through the melt produced a blister effect. Silver and other metals were present in the blister copper.

### iv. *Fire Refining*

The final stage, used to produce copper of approximately 99.5% purity, was again a carefully controlled melt using oxidising fluxes or air, and organic matter as reducing substances. The process known as 'poling', whereby a stout pole of green wood was thrust under the surface, is still employed in the industry. This oxidation/reduction continued until all the impurities were removed to the slag. At this stage the slag was very valuable as it contained any precious metals which had been present in auriferous or argentiferous copper ores.

By the late nineteenth century electro-refining processes were being developed to refine copper to purities exceeding 99.5%, which was increasingly demanded by the electrical industry. In Britain, however, fire refining remained the predominant technology into the twentieth century (Newell, 1990). Although the latter three stages of the Welsh process were fairly fixed, varying only for the recovery of different secondary metal values, the partial roasting stage differed according to the character of the ores used. The copper ores exploited in the nineteenth century fell into two broad classes: the copper sulphide based ores ($Cu_2S$-FeS-$Sn_2S$) and the carbonate/oxide based ores ($Cu_2CO_3$-CuO-$Cu_2O$). For the sulphide based ores the processes were as above, but for the carbonate/oxide based ores the partial roasting stage was sometimes excluded, the ores being smelted immediately, but with the addition of sulphur into the furnace charge. The reason for this is that the copper in the oxide form can react with the silicate minerals of the gangue and be lost into the slag.

There were, of course, gangue components together with large amounts of iron, sulphur, and silicate present in the ores purchased by smelters. The presence of these materials also affected the chemistry of the smelting processes and the composition of copper smoke. For instance, the 'whiteness' of the smoke noted by Le Play (1848) was caused by sulphuric acid, a little hydrochloric acid and water vapour, whereas the 'poisonous' nature of copper smoke mentioned by Vivian (1989) was due mainly to the oxides of sulphur from the ore as well as 'arsenic trioxide' ($As_4O_6$) from the ore and gangue. In addition lead, cadmium, antimony, and traces of sulphur and hydrofluoric acid were present in copper smoke depending on the ores being used and smelting conditions.

In summary, the main species emitted from copper smelting works were: $CO_2$, CO, $SO_2$, $SO_3$, $H_2SO_3$, $H_2SO_4$, $H_2O$, $As_4O_6$, HCl, S, $Sb_4O_6$, Pb, and Cd.

Copper smoke is usually taken to include all except $CO_2$ and CO. The principal chemical reactions of the smelting process producing copper smoke were:

$$C_{15}H_8(\text{coal}) + 16O_2 \longrightarrow 2CO + 13CO_2 + 4H_2O$$

$$Cu_2S + O_2 \longrightarrow 2Cu + SO_2$$

$$2H_2O + 2SO_2 + O_2 \longrightarrow 2SO_3 + 2H_2O \longrightarrow 2H_2SO_4$$

$$SO_2 + H_2O \longrightarrow H_2SO_3$$

$$FeAsS \longrightarrow FeS + As(g)\ (700°C)$$

$$FeS_2 \longrightarrow Fe + S_2$$

$$2Sb_2S_3 + 9O_2 \longrightarrow 2Sb_2O_3 + 6SO_2$$

$$2As_2S_3 + 9O_2 \longrightarrow As_4O_6 + 6SO_2$$

$$H_2SO_4 + NaX \longrightarrow HX + Na_2SO_4 \text{ where X = Cl, F, minor.}$$

On the basis of this, it is possible to estimate the composition of a common form of copper smoke as it leaves the top of a chimney. In doing so it is assumed that the principal type of ore being smelted is Cornish (grey) copper sulphide ore. This assumption is valid for the 1860s, although in the latter part of the decade the output of the Cornish mining industry fell sharply and from then on south Wales smelters became increasingly dependent upon supplies of imported ores and regulus.

| | COAL or ORE | | | COPPER SMOKE | |
|---|---|---|---|---|---|
| 'Element' | % | mol/kg in ore | Volatiles | mols/kg ore out of stack | Composition (ppm, vol) |
| {Coal} | | | | | |
| | 1.0 | 0.02 | Bl Smoke (100%) | 0.04 | 0.37 |
| | 1.5 | 0.23 | $SO_2$(100%) | 0.44 | 4.4 |
| {Ore} | | | | | |
| S | 23.4 | 7.19 | $SO_2$ and $SO_3$ (91%) | 6.54 | 65 |
| | | | $H_2SO_3$ (9%) | 0.65 | 6.5 |
| | | 0.15 | $H_2SO_4$ (1.4%) | 0.15 | 1.5 |
| Cu | 40.6 | 6.39 | Cu (0.001%) | $6\times10^{-5}$ | $6\times10^{-4}$ |
| Fe | 5.0 | 0.09 | — | | |
| Si | 0.4 | 0.14 | — | | |
| As | 7.7 | 1.03 | $As_4O_6$ (10%) | 0.10 | 1.0 |
| Sb | 12.4 | 1.02 | $Sb_4O_6$ (1.0%) | 0.01 | 0.1 |
| Ag | 0.6 | 0.06 | Ag (0.01%) | $6\times10^{-6}$ | $6\times10^{-5}$ |
| Zn | 3.7 | 0.57 | — | | |
| Pb | 0.2 | Trace | PbO (0.01%) | $10^{-7}$ | $10^{-6}$ |
| Other | 2.5 | — | | | |
| Balance oxygen, nitrogen and carbon dioxide (90% volume) | | | | | |

TABLE 1. Estimations of composition of Copper Smoke[1]

An average composition of Cornish grey ore is included in Table 1, taken from Napier (1853) with a slight modification. The composition of the coal is assumed to be 1.5% sulphur and 1% smoke (Brimblecombe and Bowler, 1990).

The estimations given in Table 1 are founded on the properties of the elements and compounds concerned, as well as experimental and observational evidence given in the references cited. The mixture of ores assumed in Table 1 is consistent with Percy's measurement of 9% copper content at the outset of smelting, though less than Le Play's 13%. Napier states an optimal mixture was in the range of 9 to 13% (Napier, 1852). The large contribution of $H_2SO_4$ and $H_2SO_3$ is consistent with Le Play's observation that $H_2SO_3$ was given off from the calcining and roasting processes, and the small amount of $H_2SO_4$ with his further observation that $H_2SO_3$ alone could not be responsible for the whiteness of the smoke. It is also consistent with Percy's observation that rain made the smoke whiter.

## 6. THEORY OF PLUME MODELS USED

Gaussian-type plume models are of two basic types: the diffuse source model and point source model. The former is applicable to situations in which the source of the contaminants is not sharply identifiable but spread over a large area. This approach is demonstrated in a similar study of atmospheric pollution in York (Hipkins and Watts). The other model can be used for individual stacks, and is therefore appropriate in this case.

The Gaussian dispersion equation for a point source at an elevation is (Hanna, 1973; Smith, 1968):

$$\chi = (2/\pi)^{0.5}\left[\frac{fQ_p}{\sigma_z rU(2\pi/16)}\right] \exp\left[-\frac{H^2}{2\sigma_z^2}\right]$$

where:

$H$ = Effective stack height (m) (see below)
$f$ = Normalised percentage frequency with which wind blows toward sector.
$Q_p$ = Source strength of pollutant ($\mu gs^{-1}$)
$\sigma_z$ = vertical dispersion parameter (m) (see below)
$\chi$ = surface concentration of pollutant ($\mu gm^{-3}$)
$U$ = windspeed at effective stack height ($ms^{-1}$)
$r$ = distance downstream from point source (m)

The effective stack height (H) is given by:

$$H = 2.9\left[\frac{[wR^2(T_p - T_e)]}{[U(A + 3)]}\right]^{0.33}$$

where:

$w$ = Initial vertical speed of plume ($ms^{-1}$)
$R$ = Stack radius (m)

$T_p$ = Initial absolute temperature of plume (K)
$T_e$ = Absolute temperature of environment (K)
$A = dT_e/dz$, the lapse rate of the atmosphere ($Km^{-1}$)

and finally:

$$\sigma_z = ax^b$$

where:

x = effective boundary layer height, (m) for conversion of $\phi$ (flux) to concentration

a and b are dimensionless empirical constants as follows:
a = 0.40, b = 0.91 [sunny day, turbulent conditions]
a = 0.15, b = 0.75 [cloudy day, average conditions]
a = 0.06, b = 0.71 [night-time, unmixed conditions].

The model assumes that the expanding plume has a Gaussian distribution in both the x and z dimensions, i.e. that the plume entrains surrounding air and expands outwards in a regular way. This is often not the case (Brimblecombe, 1986), but the model is acceptable for average conditions. The model gives a (maximum) spot ground level concentration of the pollutant over a grid square on the scale appropriate to the resolution of the calculations. Here the calculations were performed on a scale of 100m, hence the results refer to grid squares of 100m. The pollutant is assumed to be normally distributed across each square, and the spot concentration produced is the maximum (i.e. peak of the Guassian curve) concentration in the square. Clearly this will give unrealistically elevated concentrations. This situation can be rectified by integrating the concentration of pollutant across the area of model square (1 $km^2$), and then dividing by the area of the actual square of which it is the centre. The second area will vary with distance from the source. Table 2 gives correction factors calculated for these areas. For the regional model a correction factor of 0.16 was chosen as a suitable average. The long range model was corrected continuously with distance from source.

An alternative method of correcting for the peak values is to use a model which calculates average concentrations. A model which estimates long term average ground level concentrations (Scorer and Barrett, 1962; Brimblecombe,

| Distance from source (m) | Actual grid area (m2) | model area (m2) | Correction factor |
|---|---|---|---|
| 5,000 | 300,000 | 10,000 | 0.33 |
| 10,000 | 600,000 | 10,000 | 0.17 |
| 15,000 | 850,000 | 10,000 | 0.12 |
| 50,000 | 50,000,000 | 10,000 | 0.04 |

TABLE 2. Correction factors for conversion of square to dodecahedral grid

1992) was used to estimate concentrations closer to the stack. The model is designed for distances up to 60 stack heights away from the stack. It uses a radial as opposed to grid system, and is based on an upper and lower cone in which pollutants are dispersed. Only the pollutants from the lower cone are assumed to come into contact with the ground. The model can be described as:

$$\chi = Q_p f/[2\pi r^2(\beta + \alpha)0.125]$$

where:

$r$ = distance from stack (m) – up to a maximum of 60 stack heights
$\beta$ = angle from horizontal of top cone
$\alpha$ = angle from horizontal of bottom cone.

The results of the Gaussian and Scorer and Barrett models were combined, and the average results used. More weighting was placed on the Scorer and Barret model nearer the stack, and more on the Gaussian model away from the stack.

The parameters of the point source model are appropriate for gases but not for the much faster deposition rates of particulates (Sturges and Harrison, 1986b). The faster deposition rates of particulates mean that the assumptions of normal dispersion in both axes across the face of the plume are violated, as the particles settle toward the ground much faster than they move across the plume. A model specifically designed to cope with this problem (Brimblecombe, 1992) was used to check the results from the point source model.

Heavy metal particulates are removed from the atmosphere by rather different mechanisms than gases, and will fall under the influence of gravity. Particle size is important because there are two forces acting on the particles: gravity and the effect of wind (floating). The sum of these effects will be determined by particle size. For the heavy metals, a specialist model (Brimblecombe, 1992) was used which comprehended these effects and calculated fluxes of particles to the ground. Mean particle sizes were assumed for the heavy metals, which were treated as one class. The mean particle size assumed for these metals was $1\mu$ (Harrison, 1986). Heavy metals in this work were defined as being composed of 0.05% Cu, 0.34% Pb, 0.02% Ag, 27% Sb, and 72% As. By adapting data on the effect of particle size on particle residence times (Slinn, 1983), and assuming a boundary layer height of 800m, ground level concentrations were obtained. As expected, the specialist model predicted much higher concentrations closer to the stack, where the resolution of the main model is very poor. For the heavy metals the two datasets were combined to produce the final map.

It is not possible to include the effects of landscape on atmospheric turbulence in the simple models used here. While south Wales has a hilly terrain, it is necessary to assume a flat landscape for these models, which therefore do not allow for the funnelling of pollutants along the valleys and atmospheric mixing that would have occurred. Because of this, it should be noted that the long range results are likely to be seriously optimistic for trajectories to the north of Llanelli.

## 7. DATA

The models deal with the main chimneys of the company’s copper, lead and silver works. The chimneys were about 500m apart, and were modelled separately (resolution 100m). The results for each chimney were combined to give overall concentration levels.

Table 3 contains the final coal and ore datasets adopted for the models. Data are available for the amount and type of ores bought by the company for its various works in this period (Nevill MSS). Some of the descriptions of the ore are precise, such as ‘x tons of Cornish copper ore, (Wheal Maria)’. Frequently distinctions are made between various types of sulphide, oxide and carbonate ores. In other cases only the basic type of ore is recorded, eg. ‘copper ore’.

**Compositions (ex stack, tonnes)**

| Year | Black smoke | Cu | As | Sb | Ag | Pb | $H_2SO_4$ | $H_2SO_3$ | $SO_2 + SO_3$ |
|---|---|---|---|---|---|---|---|---|---|
| 1861 | 553 | 0.062 | 82 | 31 | 0.026 | 0.41 | 410 | 521 | 6100 |
| 1862 | 653 | 0.071 | 95 | 35 | 0.03 | 0.47 | 473 | 602 | 7100 |
| 1863 | 701 | 0.077 | 102 | 38 | 0.033 | 0.51 | 511 | 650 | 7600 |
| 1864 | 599 | 0.065 | 86 | 32 | 0.028 | 0.43 | 429 | 546 | 6400 |
| 1865 | 639 | 0.067 | 89 | 33 | 0.029 | 0.44 | 443 | 565 | 6700 |
| 1866 | 522 | 0.053 | 70 | 26 | 0.023 | 0.35 | 352 | 448 | 5300 |
| 1867 | 610 | 0.062 | 83 | 31 | 0.027 | 0.41 | 414 | 527 | 6200 |
| 1868 | 468 | 0.046 | 61 | 23 | 0.02 | 0.3 | 306 | 389 | 4600 |
| 1869 | 460 | 0.046 | 62 | 23 | 0.02 | 0.31 | 308 | 392 | 4652 |
| 1870 | 460 | 0.046 | 61 | 23 | 0.02 | 0.3 | 306 | 389 | 4600 |
| 1871 | 460 | | | | | | | | |

TABLE 3. Coal and elemental budget data for model

An elemental composition of ore purchases was made based on a sample of entries, and this was applied to data for annual ore purchases (see Table 4). In order to generate the figures that appear in Tables 3 and 4, certain assumptions were necessary. For those cases when the descriptions were not detailed enough to assign a mine or area, the compositions of average copper, lead and silver ore had to be adopted. Ore compositions from specific mines are known (Napier, 1852), unspecific copper ore was assumed to be the most common ore for the metal, eg. for copper it was assumed to be Cornish grey copper ore.

| | Cu | Fe | S | Si | Sb | As | Se | Zn | Pb | Ag | Other |
|---|---|---|---|---|---|---|---|---|---|---|---|
| ta$^{-1}$ | 1093 | 740 | 952 | 687 | 544 | 147 | 7 | 20 | 720 | 48 | 76 |
| % | 21.1 | 14.3 | 19.8 | 13.3 | 10.5 | 2.8 | 0.1 | 0.4 | 13.9 | 0.9 | 4.3 |

TABLE 4. Mean elemental composition of ore for the lead, silver and copper works used in the model

The Nevill accounts record only the cost of coal. To convert monetary values into quantities it was necessary to estimate the local price of coal. Elsewhere, measurements of the ratio of ore to coal used in copper smelting at about this time are available (ranging from 1:1.6 to 1:1.86), (Vivian MSS) and these provide a check for consistency given that the amounts of ore smelted are known.

Meteorological data were supplied courtesy of the Meteorological Office. The data are recent, and it assumed here that regional windfields have not changed appreciably since the 1860s. The data consist of raw wind direction and speed distributions (10 year means) for both Mumbles Head and Milford Haven. The Milford Haven data are continuous, whereas those for Mumbles Head are 10 minute spot averages. Both distributions were similar in terms of windspeed and direction correlations, and also percentage direction frequencies. As might

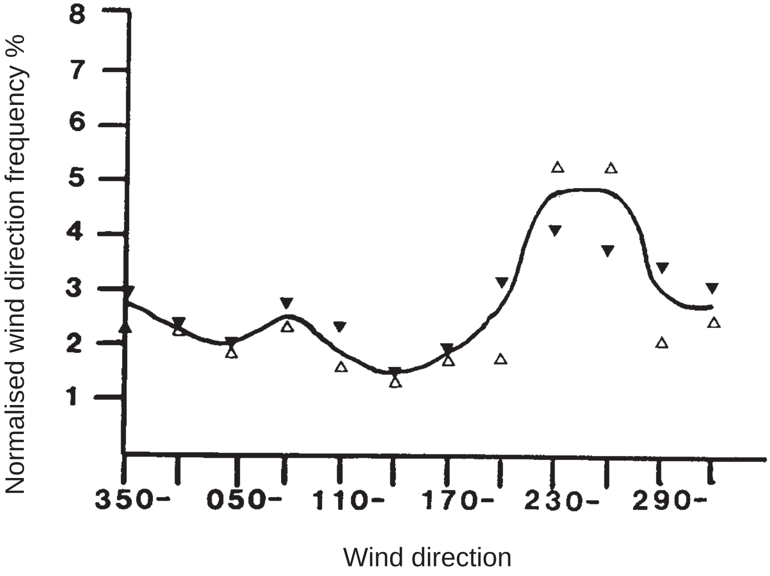


FIGURE 3. Normalised wind direction frequency data for the two meteorological sites. Mumbles Head; Milford Haven

be expected, the Milford Haven data have higher mean windspeeds in the maximal direction. The normalised wind direction frequency data were calculated for each of the 12 directions supplied (see Figure 3), and the mean of the two sites used in the model. In both cases the overall average windspeed across all directions was about 11 knots or 5.66 ms-1 (see Figure 4). These 12 directional normalised wind frequency data were used to run the models.

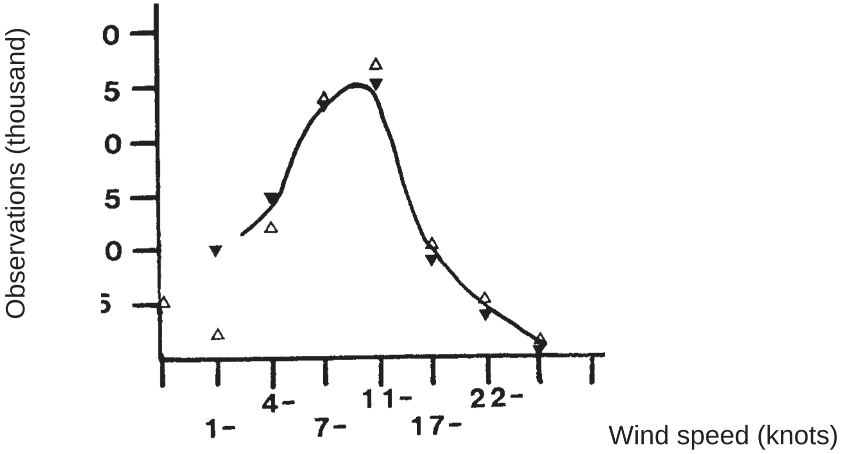


FIGURE 4. Wind speed frequency data for the two meteorological sites.
Mumbles Head; Milford Haven

## 8. METHOD

The point source model was run for each of the 12 directions for which meteorological data were available, and the results corrected as above and plotted on a dodecahedral grid. The data for 1861 were used to run the model as the assumptions employed mean that any change from year to year is in amount not speciation. The correction factors for subsequent years are shown in Table 5.

| | 1861 | 1862 | 1863 | 1864 | 1865 | 1866 | 1867 | 1868 | 1869 | 1870 |
|---|---|---|---|---|---|---|---|---|---|---|
| Ore smelted ta$^{-1}$ | 28774 | 33267 | 35929 | 30181 | 31214 | 24723 | 29123 | 21505 | 21643 | 21505 |
| Normalised to 1861 | 1.00 | 1.16 | 1.24 | 1.04 | 1.08 | 0.86 | 1.01 | 0.74 | 0.75 | 0.74 |

TABLE 5. Ore smelted, 1860-1870[2]

The worst year was probably 1863, with about 27% more pollution than 1861. By 1870 output had dropped such that pollutant fluxes were about 17% less than 1861 levels, although the quality of data from 1869 onwards is very poor. The other models were also run for 1861 and the results treated in the manner discussed above.

Given the patchy nature of the historical records, it has been necessary to make assumptions about the data used in the models, the most important of which concern the composition of ore. These assumptions will inevitably affect the results produced by the models, but because it is not possible to assess the reliability of the historical sources on which the assumptions are based, neither is it possible to test statistically the accuracy of the results obtained.

## 9. RESULTS

Local maps of estimated ground level pollution for $SO_2$ and $SO_3$, $H_2SO_4$, $H_2SO_3$, heavy metals, and black smoke are given in Figure 5. The scale is such that the maps show about 560 km2 around Llanelli with a range of about 20 km in every direction.

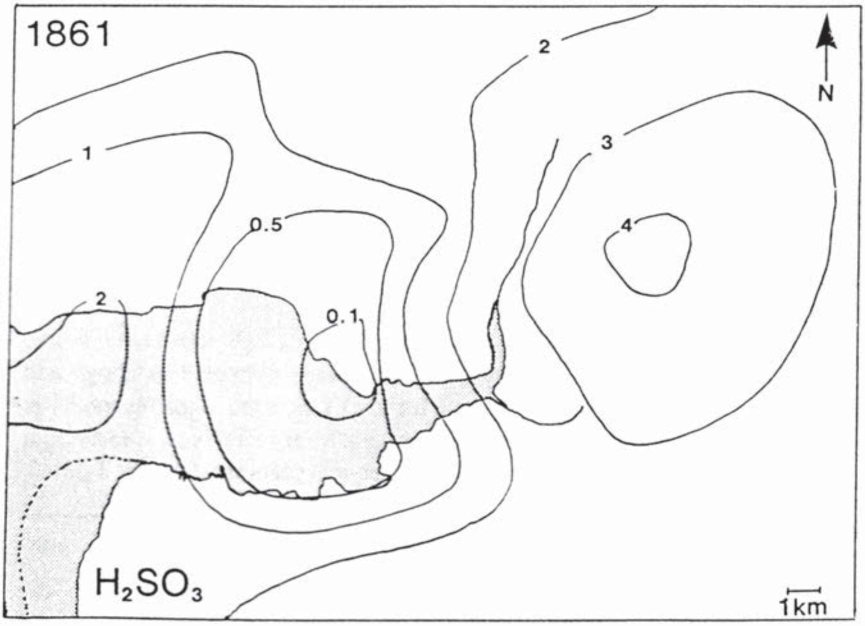
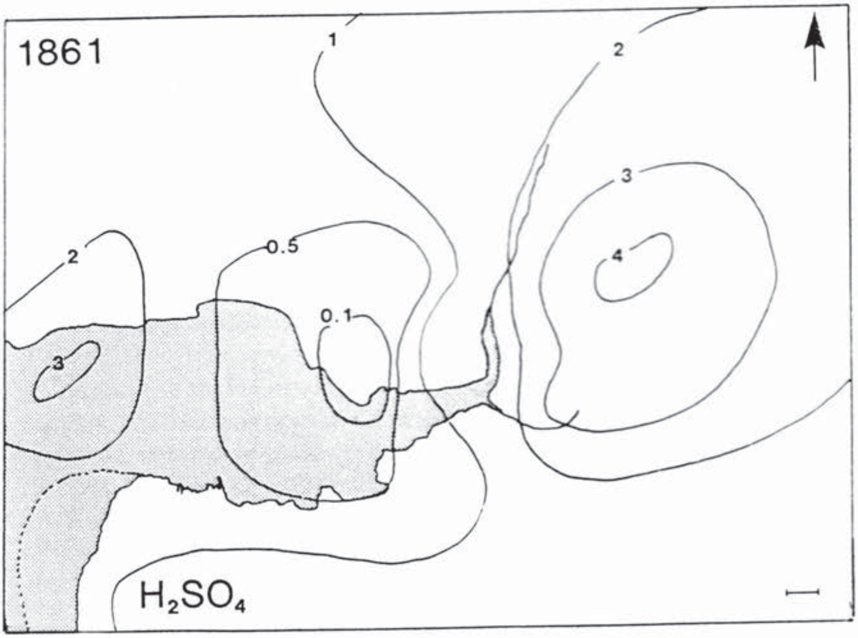
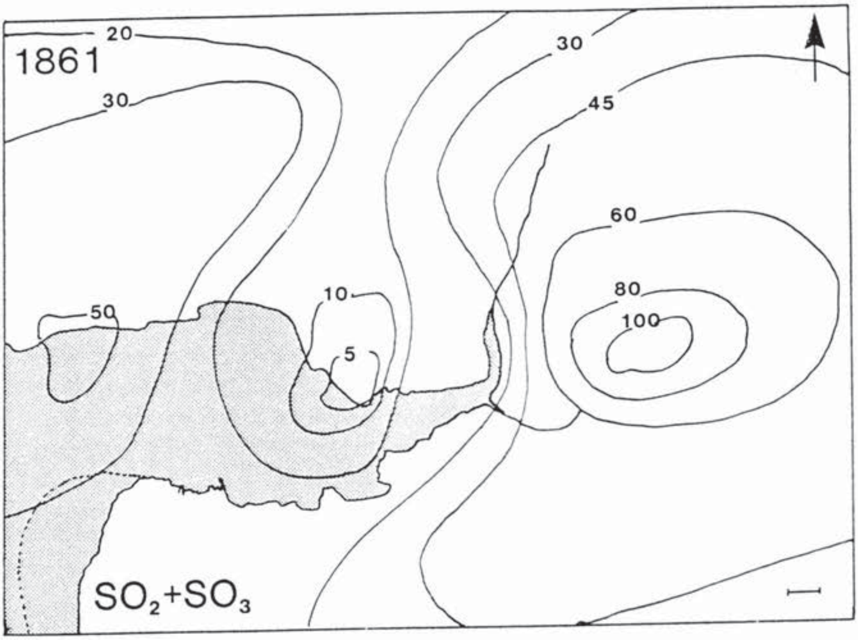
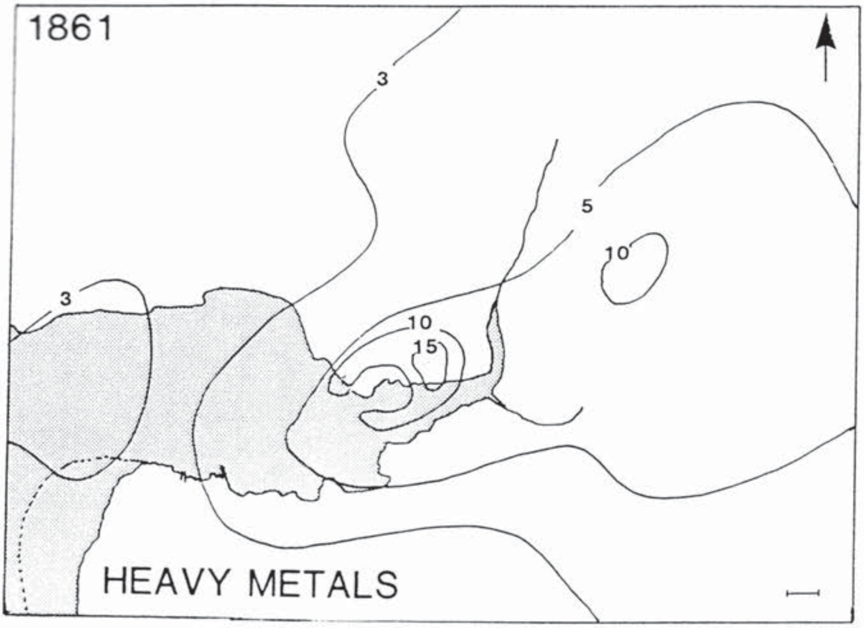
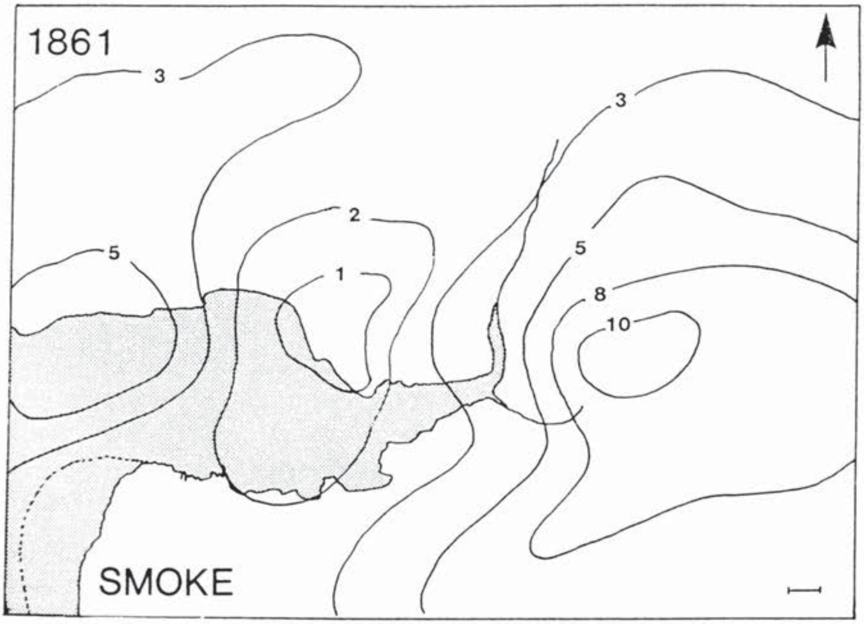


FIGURE 5. Pollution maps (local scale) for the various components modelled. Scale bar is 1km. All concentrations in $\mu gm^{-3}$. Heavy metals composed of As,Sb,Pb,Ag,Cu (0.72:0.27:01(rest)).


1861
1
2
3
4
2
0.5
0.1
3
$H_2SO_4$


20
1861
30
30
45
60
80
100
50
10
5
$SO_2+SO_3$


1861
3
5
10
10
15
3
HEAVY METALS


1861
3
3
2
5
1
5
8
10
SMOKE

The effects of the heavy metal emissions are especially bad (10-15$\mu gm^{-3}$) within about 12km of the smelting works. The heavy metal values given include lead, arsenic, antimony, silver and copper. Due to the different atmospheric chemistry of these components, it is likely that the heavy metals deposited closer to the plant (< c.8km) were mostly lead, copper and silver, whilst those deposited further away were probably arsenic and antimony. For comparison, in modern cities with dense traffic levels of 5$\mu gm^{-3}$ can be reached. Modern smelters can contribute to local lead levels up to around 5$\mu gm^{-3}$ (Sturges and Harrison, 1986ab; Elsom, 1992). A recent EC regulation has set a limit value of 2$\mu gm^{-3}$ annual average, and anything much above 1.5$\mu gm^{-3}$ is seen as a health hazard (Elsom, 1992). However, it is more likely that the overall intake of heavy metals would have been dominated by intake from food and water rather than air (Waller, 1991). In this respect, the dispersion of heavy metals over a large area is significant in that it would have contributed to the widespread contamination of crops, grazing land for livestock and water supplies.

The other pollutants tell a similar story, with high levels over a very wide area. The most disturbing are the $SO_2$ and $SO_3$ concentrations of 100$\mu gm^{-3}$ over 10 km away. These concentrations from a single smelting works are comparable to total concentrations found in York at about the same time, for instance, and in the latter case the pollutant was spread over a much smaller area (Brimblecombe and Bowler, 1990; Hipkins and Watts, 1992). Because it proved difficult to predict independent equilibrium concentrations of $SO_2$ and $SO_3$ thermodynamically, and because they are not separated in the contemporary literature, they are treated together here as well.

Constant exposure to the levels of $SO_2$ and $SO_3$ indicated here would probably have caused significant chronic respiratory problems within the local population. However, this form of pollution is most notorious for its acute effects, such as the high death rates associated with the synergistic effects of coal smoke and sulphur oxides in London ‘smogs’ (Elsom, 1992). Although the results presented here are annual averages, they do suggest that under the right meteorological conditions short-term peaks in copper smoke could have had severe acute effects on the local population in south Wales as well.

Also of concern are the likely concentrations of other metals and non metals that were probably present in the fumes but not included in this analysis. These might include beryllium, cadmium, bismuth, mercury, nickel, selenium, etc. Neither does this study include possible contributions from halogens, (notably HCl), principally because of uncertainty over ore compositions. The combined health effects of the particles, the chemical effects of the mixture of unmeasured pollutants and radionuclide emissions from flue deposits was potentially serious.

As discussed above, the longer range model is likely to be compromised for Northern trajectories by landscape effects. Figure 6 shows the distribution map for $SO_2$ and $SO_3$ over a wider area. Again it is clear that the effluent from the one plant is very important both locally and further afield. For comparison, in modern

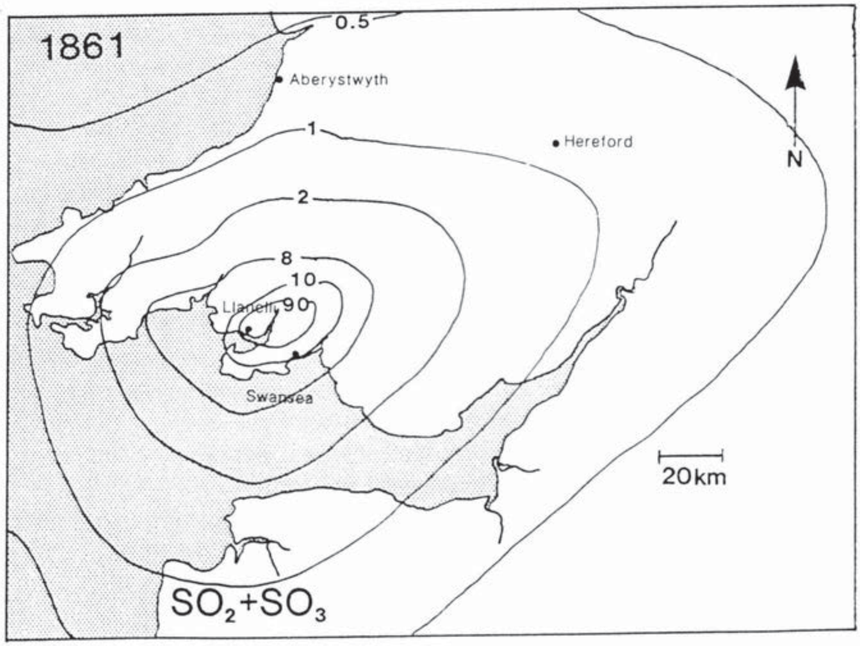


FIGURE 6. Pollution maps (regional scale) for $SO_2+SO_3$. Scale bar is 20km. All concentrations in $\mu gm^{-3}$.

industrial cities peak $SO_2$ concentrations can reach 20$\mu gm^{-3}$. The concentrations in this work are annual averages. Again when the pollution from the rest of Llanelli and Swansea are added to that modelled, the effects would have been felt over much of south Wales as well as the north Devon and Somerset coasts.

## 10. CONCLUSIONS

Although one of the largest firms in the industry, it is estimated that the Llanelli Copper Co. accounted for about only 15% of copper production in south Wales at this time. Most production took place in and around Swansea at works which had lower stacks than those used by the Llanelli Copper Co. This suggests that other works may have been more serious local polluters than those studied here.

This paper indicates that pollution levels from one producer alone were disturbingly high. When emissions from other metal works, other industries and domestic coal users are taken into account it is apparent that actual concentrations downwind from Llanelli and Swansea would have been exceptionally high.

The maps also show clearly the effect of the very high chimney in transporting pollutants over long distances, and the effect of the prevailing windfields. These are consistent with the contemporary observations of damage to land in the locality, and they also substantiate claims made by landowners and others that pollution from the metal works was damaging land and poisoning grazing animals much further away but which were impossible to prove at the time.

Overall, the findings of this paper corroborate the views of Angus Smith as to the seriousness of the pollution problems of the copper industry and its ancillary activities. They also point to further studies which could be undertaken to shed additional light onto the problems of atmospheric pollution from smelting works. For instance, the alarming concentration levels indicate a serious health hazard. Whilst these were not picked up at the time, it should be possible in retrospect to re-examine local mortality data to see whether high-risk areas differ from those less or unaffected by smelter smoke. In addition, the problems of nineteenth century south Wales bear similarities to recent pollution problems in Eastern and Central Europe. It may be possible to utilise information from studies on these contemporary problems to gain further insights into the probable effects of copper smoke and other smelter fumes on the British population in the last century. In doing so it will contribute to a small but growing literature which is exposing the previously neglected social costs associated with the British Industrial Revolution.

## NOTES

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[1] Estimates for percentages of components emitted into the atmosphere are as follows: 1.4% of sulphur from the ore as sulphuric acid (Percy, 1861); total acids in smoke ($H_2SO_3 + H_2SO_4$) 16 $gm^{-3}$, calculated from PP (1911); total sulphur in smoke (acids + $SO_2$ + $SO_3$) = 130 $gm^{-3}$, calculated from Percy (1868) assuming his figures are w/w percentages; hence split ores sulphur as 90% $SO_2$ + $SO_3$ and 9% $H_2SO_3$ and 1.4% $H_2SO_4$ from calculations/estimates (Le Play, 1848; Percy, 1861; PP, 1911); lead emissions and speciation taken from modern smelter data (Sturges and Harrison, 1986a,b,c; Harrison, 1986); coal sulphur and smoke data (Brimblecombe and Bowler, 1990); estimates of antimony relative to arsenic (Napier, 1852); absolute numbers estimate. The ranking of metals, eg. copper and silver to antimony, by comparison of boiling points (Moore and Boyce, 1990), the published analyses of culvert deposits and copper smoke, (Napier, 1853; Percy, 1868; Le Play, 1848) and estimates by the authors. General data on chemistry of elements from Greenwood and Earnshaw (1984).

[2] The data from 1868 is very patchy and assumptions have been made. In some cases the records for these years were not available at all so data from previous years was used and extrapolated.

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