

INTERNATIONAL SYMPOSIUM

Environmental health aspects of lead

**Die gesundheitlichen Aspekte
der Umweltverschmutzung durch Blei**

**Les problèmes sanitaires posés par le plomb
présent dans l'environnement**

OCCURRENCE AND DISTRIBUTION OF LEAD AND
OTHER METALS IN TWO AREAS OF UNUSUAL
DISEASE INCIDENCE IN BRITAIN

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SUMMARY

The occurrence of lead, zinc, copper and cadmium has been investigated in two historic metal mining areas of Britain, namely, Cardiganshire and the Tamar Valley. Cancer and other diseases appear unusually prevalent in both areas. Total and plant-available lead and other metals are enriched in soils of the mineralized zones and lead is also unusually high in unpolluted soils. Enhanced amounts of heavy metals are entering the food chain through vegetables and drinking water but the significance of this is uncertain.

INTRODUCTION

Environmental factors may be relevant in the aetiologies of several diseases and, in Britain, maps of some of these diseases have revealed greater incidences in the north and west which reflects a recurrent geographical distinction between "upland" and "lowland" Britain. For example, stomach cancer and lesions of the central nervous system (10), anencephaly (11) and pernicious anaemia (13) have their greatest prevalence in parts of upland Britain.

Upland Britain, broadly that part lying north and west of a line drawn from mouth to mouth of the rivers Tees and Exe, is characterised by old rocks, organic-rich acid soils, higher rainfall and lower temperature, soft drinking water, extensive historic lead mining with local pollution and a lower population density. Of these factors much attention has been paid to a possible causative role of toxic trace metals. Soils rich in lead have been associated with multiple sclerosis (5, 15) and lead leached from spoil heaps with high stomach cancer in Cardiganshire (9). Cancers, diseases of the central nervous system and congenital malformations are more prevalent in the defunct metal mining district (Tamar Valley) of Cornwall/Devon (12) and in one village three parts with contrasting cancer experience had three different water supplies, the worst part being served by well water derived from mineralised strata (1).

The occurrence and distribution of lead and other metals in Cardiganshire and the Tamar Valley have been investigated. The following review is partly based on previously published work (2, 3, 4, 6, 7).

THE STUDY AREAS

North Cardiganshire and the Tamar Valley are probably representative of other old mining areas in upland Britain. Both are underlain by hard Lower Palaeozoic sediments. In the Tamar Valley there are many igneous intrusions and the area is flanked by the granite bosses which form the high ground Dartmoor and Bodmin Moor. Altitudes in the Tamar Valley range from sea level to about 300 m, but in Cardiganshire to above 800 m. Soils are generally acid brown earths and gleys, which become peaty above 300 m, and land use is chiefly grassland and forestry, with some horticulture in the Tamar Valley. Metal mining began before the Roman Occupation but was most active between 1700 and 1900 A.D. Output in Cardiganshire was primarily lead, silver and, latterly, zinc and in the Tamar Valley, arsenic, copper and lead. Now, the only evidence of past industry is scattered, abandoned mine workings and spoil: both areas are popular for holidays and tourism.

TOTAL TRACE METALS IN SOILS

In both areas the major pollutants were lead and zinc, and copper was prominent in the Tamar Valley. Cadmium was also studied since it occurs as a 'guest' element in the zinc ore sphalerite.

To establish the normal soil range for each metal, samples (0-15 cm) were taken for analysis from soils similar to those of the mineralised zone but definitely free from pollution. Trace metal contents in soil are best described in terms of a log-normal distribution and 99.9% of a population is contained within

the range mean $\pm 3 \times$ standard deviation. Results based on this calculation are given in Table 1 and the data allow polluted soils to be identified; e.g. a lead content > 106 p.p.m. suggests unusual enrichment, most probably by contamination. The lead values are interesting, for the abundance in the lithosphere is 16 p.p.m. and between 10 and 20 p.p.m. is commonly regarded as an average for soil. In a recent survey in the U.S.A. the (geometric) mean lead content was reported as 16 p.p.m. whence the normal range was 4-120 p.p.m. (14). Yet values for Welsh soils formed on Ordovician and Silurian sediments are consistently higher and 40 p.p.m. Pb is typical. In contrast, the mean lead level for soils on Triassic sandstone near Birmingham was approximately 20 p.p.m. (8).

There are several sources of pollution in mineralised regions. Undisturbed soils overlying ore bodies become metal enriched (exemplified in Table 2) by natural processes. Fall-out from smelters allows enrichment downwind. The environs of spoil heaps are often phytotoxic because of contamination by wind blown fines or by drainage water from the spoil and only tolerant species, such as strains of *Agrostis tenuis*, grow there. Pollutants are also dispersed over a wide area with no apparent deleterious effects. In the mineralised part of the Tamar Valley 34 apparently uncontaminated, fertile pastures were sampled: 14 contained anomalous levels of at least one of the four metals analysed (Table 3) and the mean metal values for these 14 fields were all higher than the normal means of Table 1. The River Ystwyth, in Cardiganshire, contains within its floodplain some of the best local land. Before 1876 the uncontrolled injection of mine effluent into the river resulted in the deposition of finely ground ore particles on to these fields during floods with consequent poisoning of crops and stock. But these fields are now apparently fertile and productive. Summary data for samples from 17 of these fields are given in Table 3 and the very high levels of lead should be noted.

The data in Table 3 also illustrate how the nature of the regional mineralization controls pollutant levels. Cardiganshire was primarily a producer of lead and zinc whereas the Tamar Valley produced much more copper: the mean values for lead, zinc and copper reflect these differing emphases.

For rural people a significant contact with the physical environment is through home-grown vegetables. Accordingly, 12 gardens in the Tamar Valley were sampled, including some outside the mineralized part. Summary data are presented in Table 3. Every sample was anomalous with respect to at least one metal and it is suspected that traditional gardening practices, such as heavy use of animal manure or application of coal ash or soot may contribute to this enhancement.

PLANT AVAILABLE METALS IN SOIL

Only a small fraction of the total metal in soil is available for absorption by plants. The content of available metals is customarily estimated by extracting soil with one of a variety of solutions. Often, these solutions are diluted acids and acetic acid (2.5%, v:v) is commonly used. Available metals in all samples were estimated in this way and, in general, there was a direct, linear relationship between the total metal content and its availability to plants. This is exemplified by the data in Table 4 which are the results of computing the correlation coefficient between total and available metal values for the soil samples

including those of Table 3.

In practice, uptake of an element by a plant depends not only on the availability of that element but also on factors such as soil reaction, antagonism and synergism by other elements, plant species and differential uptake during the growing cycle. Radish (*Raphanus sativus*) has been used as a convenient test crop to measure actual uptake and nine experimental plots were harvested from Tamar Valley gardens. The results (Table 5) generally confirm the evidence from acetic acid extraction of soil that availability is proportional to total lead content. They do not support the contention that lead is rendered largely unavailable in neutral and alkaline soils. Radish plots were also grown in north Cardiganshire gardens (2) and broadly similar results were obtained, the main difference being that the lead values were generally higher: the Cardiganshire mean was 64 p.p.m. Pb with range 28-161 p.p.m., compared with the Tamar Valley (Table 5) mean of 38 p.p.m. Pb and range 14-74 p.p.m.

Within a plant, although lead tends to accumulate mostly in the root the amount in the leaf also rises as uptake increases. Radish was grown (2) on a control soil and a high lead soil (44 and 11,200 p.p.m. Pb, respectively): the control radish contained 10 p.p.m. Pb (dry matter) in both the root and leaves whereas the other contained 499 p.p.m. Pb in the root and 136 p.p.m. Pb in the leaves. Moreover, the very high lead content of the second soil emphasises the point that lead is not very phytotoxic and "Toxicity symptoms are no sure guide to the plant's metal content and maximum acceptable lead concentration are exceeded in apparently healthy crops" (4).

TRACE METALS IN WATER

Allen-Price (1) indicted the quality of Tamar Valley water supplies in his study of cancer distribution but he published no analyses. Accordingly, a very limited survey was made of rural domestic water quality. One litre samples were collected during the middle part of the day after rejecting the first three litres from the tap: it was assumed this would obviate the possibility of lead contamination by acidic water standing in lead pipes. Table 6 records the results. The data are too few to justify close argument but they do point to the need for further investigations. Assuming plumbosolvency may be a problem in soft waters below pH 7.8 then about half the samples are suspect. The World Health Organisation has advisory standards for cadmium and lead in water and some national organisations advise even lower lead limits. Therefore, samples have been identified in Table 6 where 50% of W.H.O. European standard was reached: 5 samples are suspect. It is worth noting that the two suspect lead samples (506 and 518) correspond with samples 16 and 53 in Table 5, where high radish leads are recorded, and both water supplies were derived from wells. No standards are set for the other three metals but, when compared with reports in the literature for natural waters, some of these values are also high.

CONCLUSIONS

It is not claimed in this paper that lead and other heavy metals are involved in the cause of diseases such as cancer. There appears to be a geographical association between high rates of several diseases and some westerly parts of Britain but correlation is not necessarily indicative of cause and effect.

There is an emerging appreciation that some rural environments in 'upland Britain' are richer in lead and other metals than similar parts of 'lowland Britain'. Apart from the question of mining pollution, uncontaminated soils on Ordovician and Silurian sediments consistently yield lead values two to three times higher than soils on younger rocks. The situation is compounded by the extent of historic lead mining and its associated pollution: e.g., there is at least one defunct lead mine in every Welsh county. Lead and other pollutants have been dispersed throughout the mineralized regions without necessarily causing visible environmental damage. The inference from the work on lead in soil, in vegetables and in tap water, is that relatively large amounts are entering the food chain and that rural inhabitants of these areas are exposed to a greater 'lead insult' than similar populations elsewhere in Britain. But the significance of this is very uncertain; e.g. the dietary importance of locally produced food in rural areas is not known and populations are so small that the medical data need careful interpretation.

Current work includes more detailed investigation of the extent and nature of heavy metal pollution and how metal uptake by plants may be influenced.

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TOTAL METAL CONTENT : p.p.m. DRY SOIL

<u>METAL</u>	<u>Geometric Mean</u>	<u>Range</u>	<u>Sample Size</u>
Pb	40	15 - 106	29
Zn a	131	69 - 250	17
b	61	22 - 172	12
Cu	18	4 - 61	29
Cd	1.5	0 - 3.3	29

a = Cardiganshire

b = Tamar Valley

Table 1. Normal levels of metals in soils of the study areas. Zinc values differed significantly in the two areas and are quoted separately.

SAMPLE NO.	1	2	3	4	5	6
TOTAL LEAD (p.p.m. dry soil)	45	68	142	64	58	15

Table 2. Lead contents of soil along a 100 m transect across a lead ore vein near Mold, Flintshire.

TOTAL METAL CONTENT (p.p.m. DRY SOIL)

	<u>Pb</u>		<u>Zn</u>		<u>Cu</u>		<u>Cd</u>	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
TAMAR VALLEY (14 pasture samples)	122	35 - 310	136	20 - 258	82	9 - 193	2.0	1.1 - 4.5
(12 garden samples)	260	41 - 522	396	73 - 793	131	25 - 684	2.7	1.3 - 4.3
YSWTYTH VALLEY (17 pasture samples on alluvium)	1419	90 - 2900	455	95 - 810	30	17 - 42	2.5	1.2 - 4.0

Table 3. Metal contents of contaminated soils.

METAL PAIR (Total v. Available in soil)	LINEAR CORRELATION COEFFICIENT (r)		
	TAMAR VALLEY		YSTWYTH VALLEY
	(46 pastures)	(12 gardens)	(17 alluvial soils)
Pb	0.717***	0.885***	0.867***
Zn	0.542***	0.727**	0.903***
Cu	0.774***	0.810**	-
Cd	NS	0.746**	-

*** 0.1%)

** 1.0%) Statistical

* 5.0%) significance

NS < 5.0%)

Table 4. Relationships between total and available metal contents of soil.

SAMPLE NO.	SOIL pH _s	LEAD CONTENT (p.p.m.)	
		Plant Ash	Available in Soil
8	7.0	40	14.8
10	5.5	32	2.3
14	6.6	32	18.3
15	5.3	19	6.0
16	7.1	74	8.7
17	6.6	29	5.0
21	5.3	14	1.0
53	5.7	51	14.1
63	6.6	55	22.3

Table 5. Lead content of radish grown in Tamar Valley gardens. Soil reaction (pH_s) was determined in 0.01 M CaCl₂.

SAMPLE NO.	pH	METAL CONTENT AS $\mu\text{g. l}^{-1}$				
		Pb	Zn	Cu	Cd	Mo
501	9.3	<	232	23	2	1.5
502	8.9	<	20	8	12*	0.5
503	7.0	<	8	4	9*	0.4
504	7.8	<	6	4	2	0.8
505	9.0	25	5	5	2	0.8
506	5.9	2960*	53	190	2	0.2
507	8.4	<	5	7	2	3.0
508	8.9	<	3	7	2	2.2
509	9.0	<	5	5	2	4.2
511	8.7	<	5	10	2	1.4
512	7.7	<	11	125	2	0.2
516	7.5	<	300	5	9*	0.6
518	7.0	55*	16	265	3	<
522	7.6	<	<	3	3	0.4
523	6.2	<	16	<	2	0.2
524	8.8	<	2	<	3	10.2
DETECTION LIMIT	-	20	2	2	2	0.2

< below detection limit.

* $\geq$ 50% of WHO European limit.