Sources and pathways of environmental lead to children in a Derbyshire mining village

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Abstract

Garden soil and housedust samples, from households in a Derbyshire village closely associated with historic lead mining, have highly elevated lead levels. Handwipe samples from children also have relatively high lead concentrations suggesting that elevated levels of lead are transferred to the child by the soil- dust-hand-mouth pathway. However, this is not reflected in their blood lead concentrations which are within normal UK ranges and less than predicted by some lead exposure models. SEM analysis of soil grains has revealed that many are composed of pyromorphite [$Pb_5(PO_4)_3Cl$], a stable soil-lead mineral. This mineral is formed from the weathering of galena [PbS] but it is not clear to what extent weathering has occurred in the soil. Pyromorphite has an extremely low solubility which may contribute to a low human bioavailability of lead in these soils, resulting in the lower than expected blood lead concentrations.

Introduction

There has been much concern recently, that intellectual development in young children may be impaired by long term environmental exposure to lead (MRC, 1985; MRC, 1988). Publications concerning this exposure consider garden soil and housedust as important sources of lead: for example, Yankel *et al.* (1977), Duggan (1983), Duggan and Inskip (1985). Young children inadvertently ingest dust by hand to mouth activity and sucking objects (Harvey *et al.*, 1985) and may even eat soil or dust, the habit known as pica. Two British studies have considered the relative importance of different pathways of lead to young children; one by the Royal Commission (1983) in their review of lead in the environment and the other by Davies *et al.* (1990) based on surveys undertaken in Birmingham. Both conclude that housedust accounts for approximately half of a young child's total daily lead exposure.

Elevated levels of lead have been found in Derbyshire stream sediments and soils (Colbourn and Thornton, 1978; Webb *et al.*, 1978) where there is a long history of mining. Village communities in Derbyshire closely associated with past mining activities contain raised lead concentrations in soil and housedust samples (Barltrop *et al.*, 1975; Culbard *et al.*, 1988). An investigation of lead exposure in the local population was conducted by Barltrop *et al.* (1974, 1975). Mean blood lead concentrations of 25.0 $\mu g\ dL^{-1}$ were reported for children living in these villages, although no adverse health effects were apparent.

Blood lead levels have been falling at approximately 4% per annum since the early 1970s (DoE 1990), coinciding with the reduction of lead content in petrol, paint and food. During the same period, analytical instrumentation has greatly improved the accuracy of blood lead determinations. This paper examines the sources and pathways of environmental lead to children in Winster, one of the Derbyshire mining villages.

Winster Village

Winster village (GR SK241604) is situated in Derbyshire, Central England. It is underlain by mineralised limestone and three lead veins run directly through the village at shallow depth (Figure 1). Lead mining in the Winster area dates back to the Roman occupation of Britain and culminated in an intensive lead mining phase involving mechanisation during the eighteenth century. The lead mining industry collapsed at the end of the eighteenth century and no mining has taken place in the immediate vicinity of Winster since this time.

Present day Winster village can be divided into two parts (Figure 1). The central part of the village comprises eighteenth century housing built during the heyday of mining whilst newer housing, mostly post-Second World War, has been built on the eastern periphery of the village. The older houses form an irregular array almost completely enclosing a central hillock of mine spoil which forms the gardens of many of the surrounding houses. Very little evidence of past mining activities is present in the orderly rows of the newer houses, although two houses have mine shafts in their gardens.

Method

Sampling

Soil and dust samples were collected in January 1988, handwipe samples in May 1988 and venous blood samples in September 1989. Forty-five households were sampled at random within Winster. Samples of housedust, garden soil and if applicable, vegetable garden soil were collected

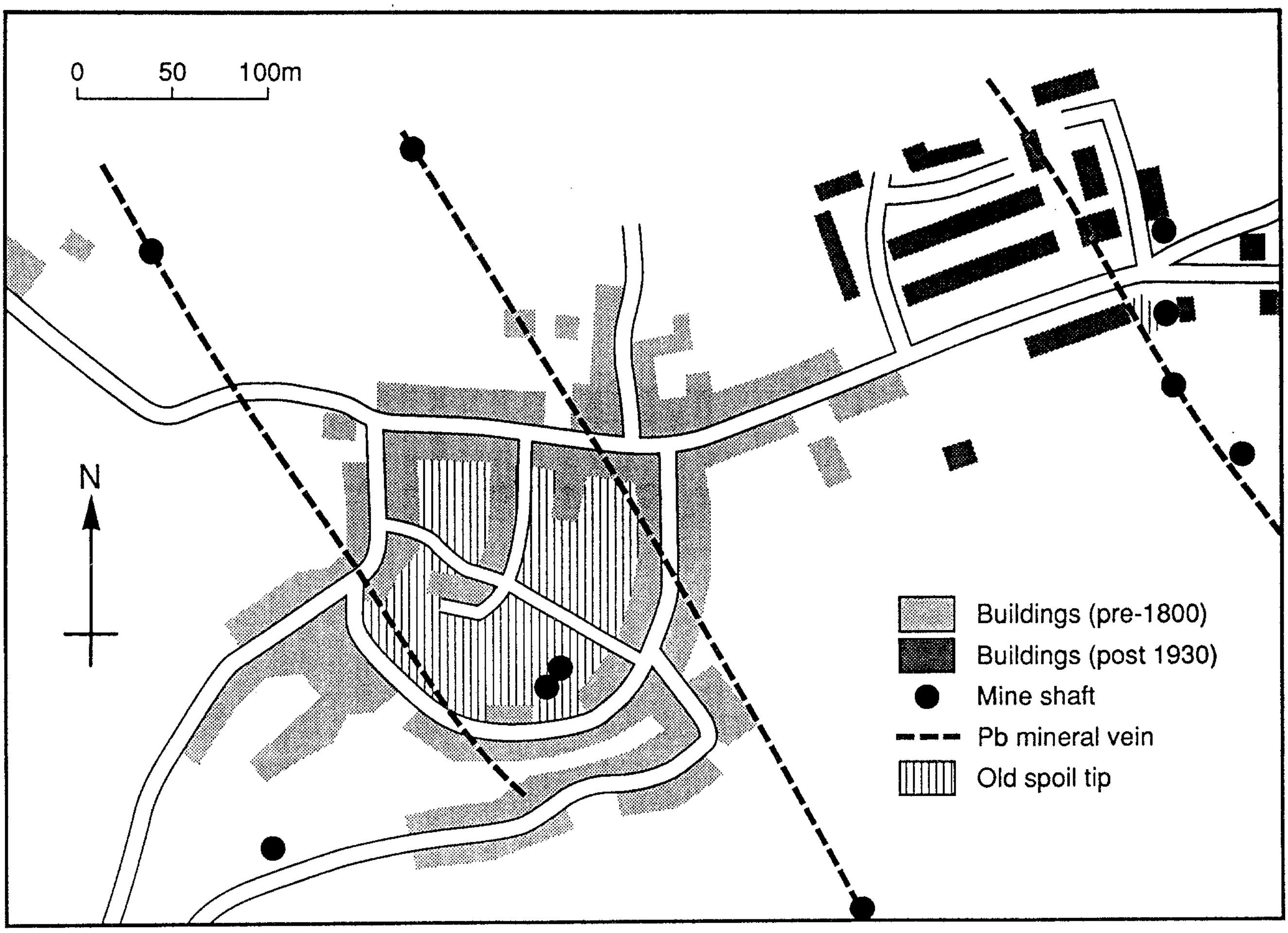


Figure 1 *Sketch of Winster village showing housing and mining features.*

Table 1 *Lead in environmental samples, handwipes and children's blood in Winster (this study), Birmingham and a UK national survey.*

	Winster (present study)			Birmingham study (Davies et al., 1990)			UK national survey (Culbard et al., 1988)[1]		
	n	*GM*	*Range*	*n*	*GM*	*Range*[2]	*n*	*GM*	*Range*
Garden soil (Pb μg g^{-1})	42	7,140	2,400–22,800	86	313	92–1,160	4,126	266	13–14,100
Vegetable soil (Pb μg g^{-1})	11	9,580	2,235–22,160		N/A		193	270	24–2,560
Housedust (Pb μg g^{-1})	45	1,560	462–6,240	94	424	138–2,093	4,638	561	5–36,900
Lead loading (Pb μg m^{-2})	44	275	14.2–3,560	93	60	4–486		N/A	
Handwipes (μg Pb per pair of hands)	10	13.1	3.8–39.5	704	5.7	1.9–15.1		N/A	
Children's blood (Pb μg dL^{-1})									
1–3 year-olds	3	6.9	6.2–8.3	97	11.7	6–24		N/A	
1–8 year-olds	10	9.4	6.2–20.7		N/A			N/A	

[1] including London but excluding geochemical hotspots.
[2] 5th and 95th percentiles.
N/A – not analysed.
GM – geometric mean.

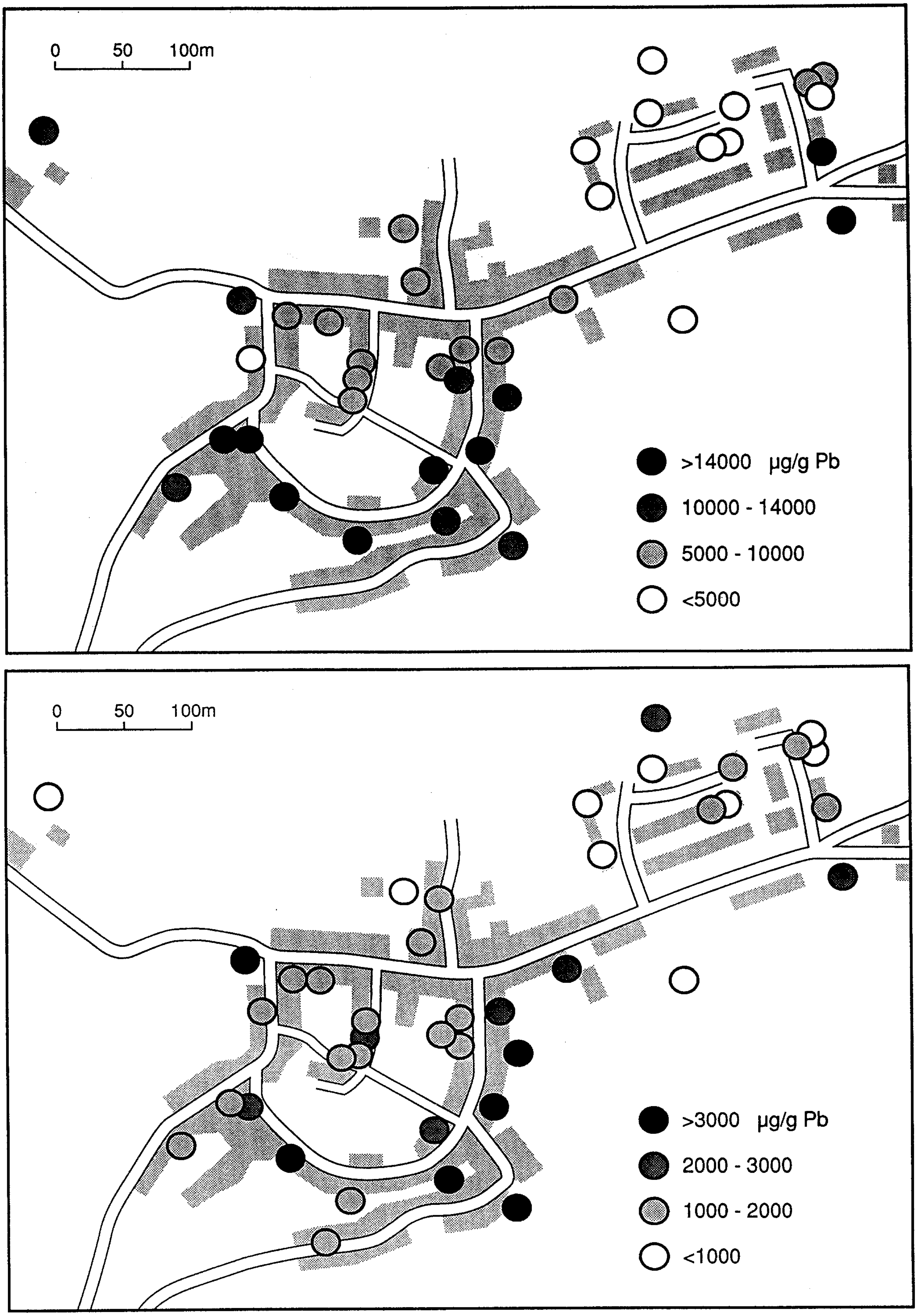
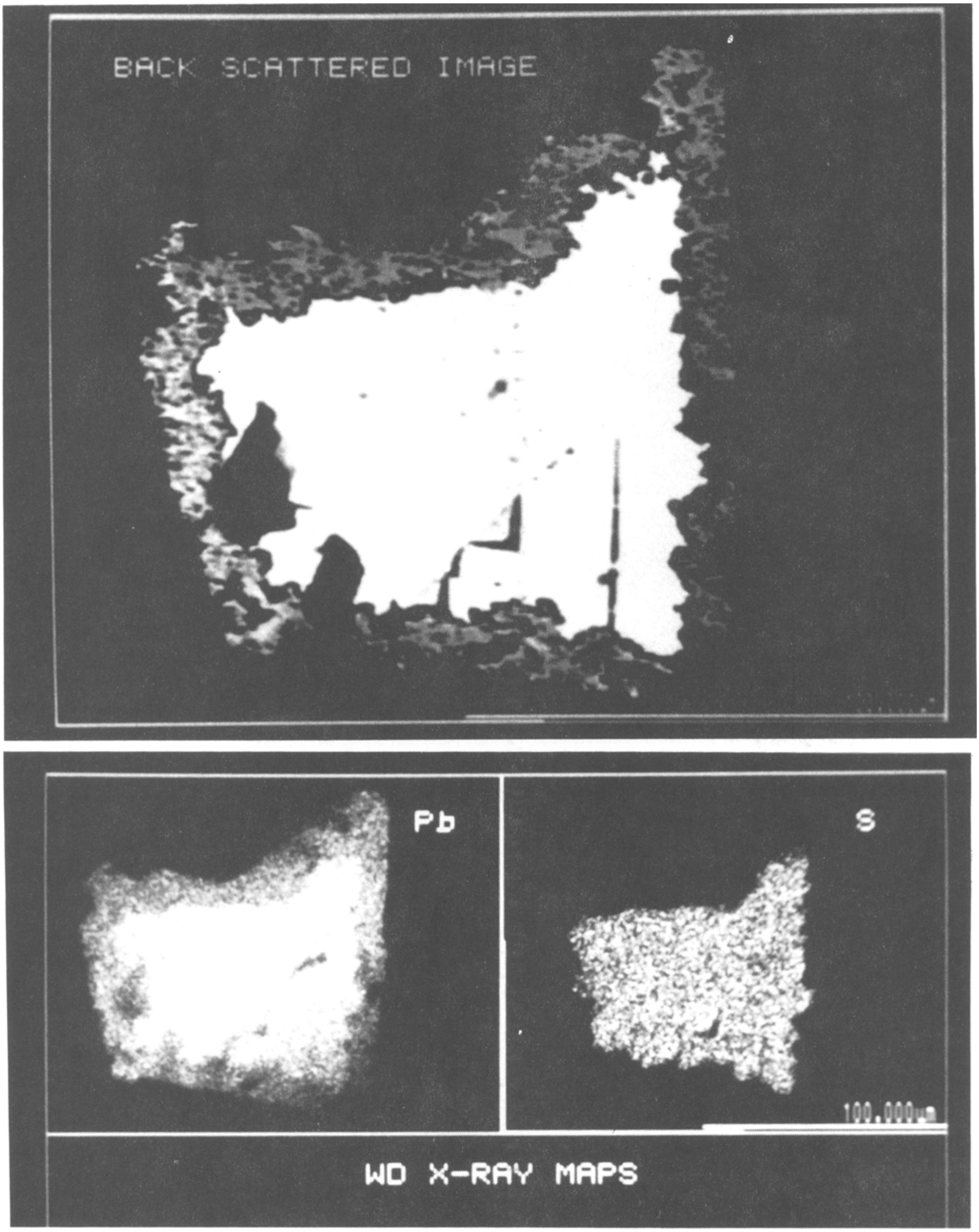


Figure 2(a) *Distribution of lead in garden soil, Winster.* ***(b)*** *Distribution of lead in housedust, Winster.*



Figures 3(*a*) and (*b*) *SEM analysis of grain of galena showing outer weathered zone. Both lead and sulphur are present in the inner core but sulphur is absent in the weathered zone. Quantitative analysis indicates that this weathered zone is composed of cerussite (lead carbonate).*

from each household. Four square metres of each householder's living room carpet was vacuumed and the dust collected in a filter thimble inserted into the vacuum nozzle (Watt *et al.*, 1983). This method allowed dust loading and hence dust lead loading (the *amount* of lead per square metre) to be calculated. Soil samples comprised 25 (0–5 cm) depth subsamples of exposed earth. To evaluate sample variability, four duplicate housedust samples were taken during the same week and four duplicate soil samples 5 months later.

Table 2 *House age and lead concentration in Winster.*

	n	*GM*	*Range*
		(Pb μg g⁻¹)	
Houses <60 years old			
Housedust	16	925	462–2,290
Garden soil	16	4,300	2,400–17,600
Houses >60 years old			
Housedust	29	2,080	1,000–6,240
Garden soil	26	9,720	3,920–22,800

GM – geometric mean.

The quantity of lead present on the hands of young (1–2.5 yr old) Winster children attending playgroup was determined using the handwipe technique of Davies and Watt (1986). The sampler's hands were first washed and wiped, then each child's pair of hands were wiped with a total of three wet-wipes which were placed in a self-sealing plastic bag and retained for analysis. Repeat samples were taken from two children on different days to evaluate variability.

Venous blood samples were taken from children aged 1–8 yr by a local practitioner and analysed at the Trace Element Unit, Southampton General Hospital.

Analysis

Dusts were passed through a mm sieve to remove carpet fibres whilst soils were disaggregated, sieved to <2 mm and ground to a powder in a "Tema" mill. Soils and dusts were digested in 4:1 concentrated nitric-perchloric acid and taken to dryness over 24 hours (1 hour 50°C, 3 hours 150°C, 18 hours 180°C). Samples were then leached in 5M HCl and made up to final volume with de- ionized water (DIW). (Thompson and Walsh, 1989.)

Handwipes were digested in warm concentrated HNO_3 and taken to dryness by boiling at 80°C. The residues were leached in HCl and made up to volume with DIW. (Davies and Watt, 1986.)

Solutions from soil and dusts were analysed using an ARL 34000 inductively coupled plasma atomic emission spectrometer; lead in handwipe samples was determined by a Perkin–Elmer 5000 flame atomic absorption spectrophotometer.

Throughout the analysis a rigorous data quality control procedure was adhered to. Each batch contained 10% duplicate pairs, 5% house reference materials and 5% reagent blanks. Analytical error was within ± 10% for soils and dusts, and ± 20% for handwipes.

SEM analysis

X-ray maps were compiled for individual Pb-rich grains in polished block samples of soil from the Winster area using a JEOL 733 scanning electron microscope (SEM). X-ray mapping involves the collection of X-rays as the electron beam scans across a field of view. These X-rays are analysed by energy dispersive (EDX) or wavelength dispersive (WDX) systems. Multi-element analysis is achieved by EDX but this system cannot differentiate between lead and sulphur, essential for the analysis of galena (PbS). Galena grains were therefore analysed using single element analysis by WDX which can differentiate between lead and sulphur. X-ray mapping has two major limitations: (i) light elements *e.g.* carbon, oxygen and hydrogen cannot be analysed and (ii) the maps yield qualitative information only.

Results

A summary of lead levels found in Winster soil and dust samples and on the hands of young children is presented in Table 1 together with comparative data from two similar UK studies. Geometric means are reported as the data shows a typical positively skewed distribution.

Soils and Dusts

The mean concentration of lead in housedust is 1,560 $\mu g\ g^{-1}$ and that in garden soil is 4.5 times greater at 7,140 $\mu g\ g^{-1}$. Mean lead loading in dusts from the village was found to be 275 $\mu g\ m^{-2}$. A soil-trodden carpet at one farm contained 19,400 μg Pb m^{-2} but this is an exceptional case and not included in the mean. All the garden soils exceeded 2,000 $\mu g\ g^{-1}$ Pb and 50% of the housedusts contained more than 1,000 $\mu g\ g^{-1}$ Pb with 33% of the housedusts exceeding 2,000 $\mu g\ g^{-1}$ Pb.

Duplicate samples indicate a sample variability maximum of 35% in garden soil, 60% in dust lead concentration and 50% in dust lead loading. These figures are similar to those reported by Laxen *et al.* (1988) and reflect the considerable temporal and spatial variation in household dust lead concentration and lead loading. Soil variability is thought to be a product of the sampling strategy coupled with heterogeneity associated with extremely high lead levels. However, the mean household lead levels found in this study are in broad agreement with those from two other studies on the Derbyshire mining villages (Barltrop *et al.*, 1975 and Culbard *et al.*, 1988), demonstrating that a representative sample can be taken in a study of this kind.

Figures 2(a) and (b) show the spatial distribution of household lead concentrations in Winster. Houses in the centre of the village, which are all pre 1800, generally contain greater quantities of lead in both housedust and garden soil than the newer houses to the east of the village centre. This is clearly shown in Table 2 where the geometric mean of lead concentrations in older households is double that of newer households.

Hand and blood Lead

The geometric mean of lead on the hands of Winster children aged 1–2.5 yr is 13.1 μg Pb per pair of hands, significantly greater at the 95% confidence level than the mean of 5.7 μg Pb previously reported in Birmingham (Davies, 1989). Two duplicate samples gave variabilities of 21% and 66%. This is in good agreement with the average geometric coefficient of variation (Kirkwood, 1979) of 34% from hand lead values (with the exception of one outlier of 406.1 μg Pb) reported by Davies and Watt (1986).

Blood sampling in Winster was limited because of the

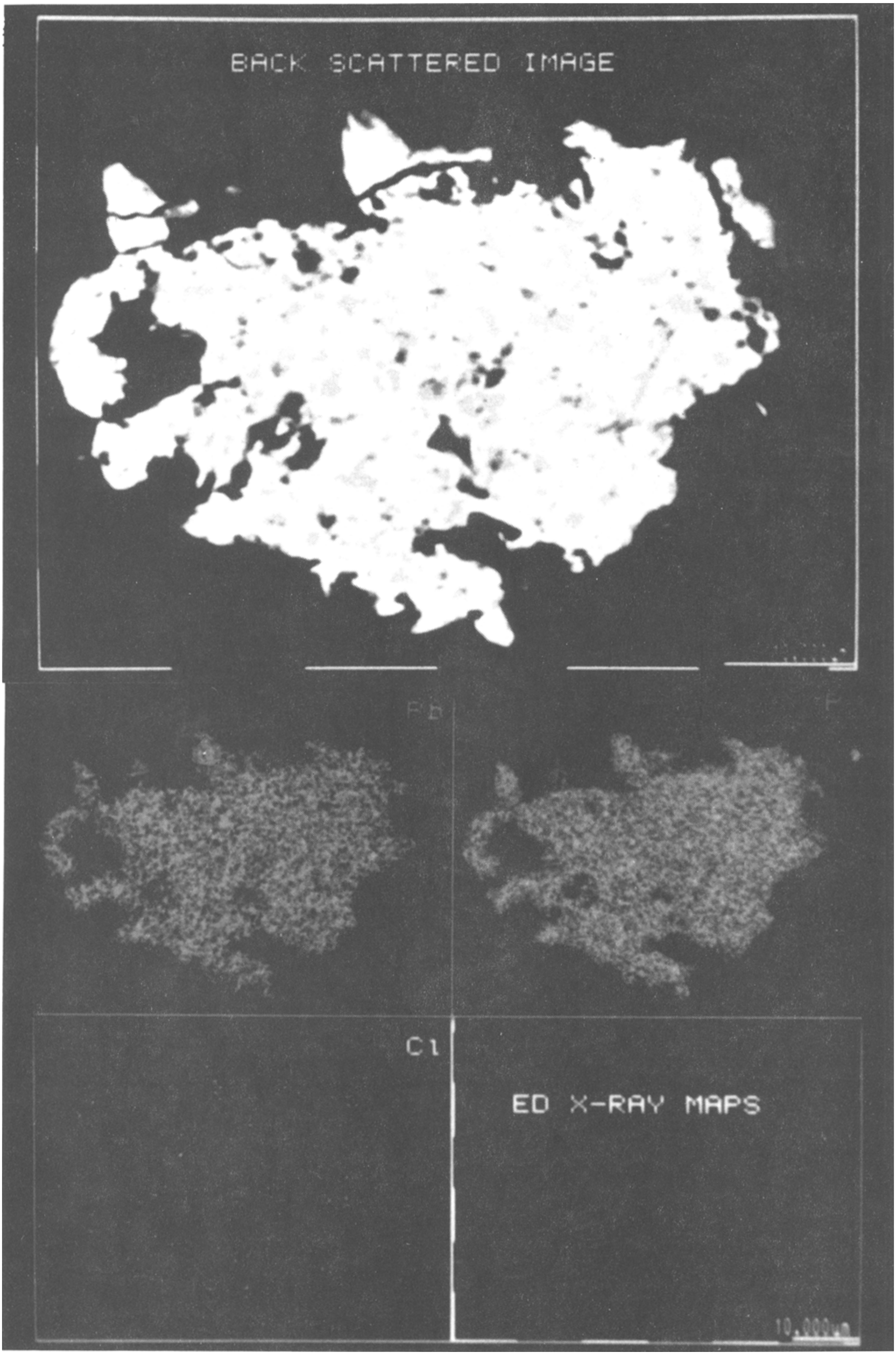
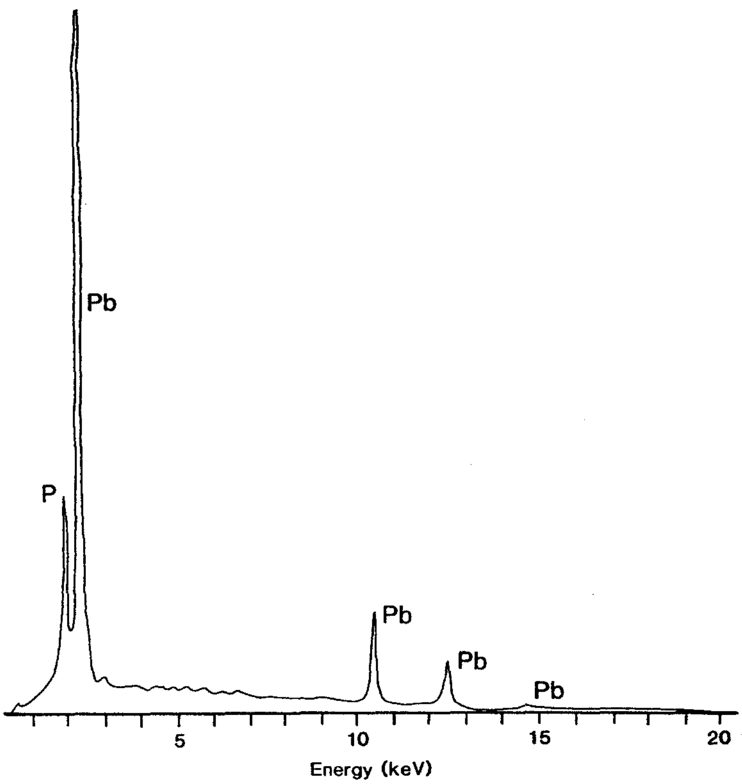
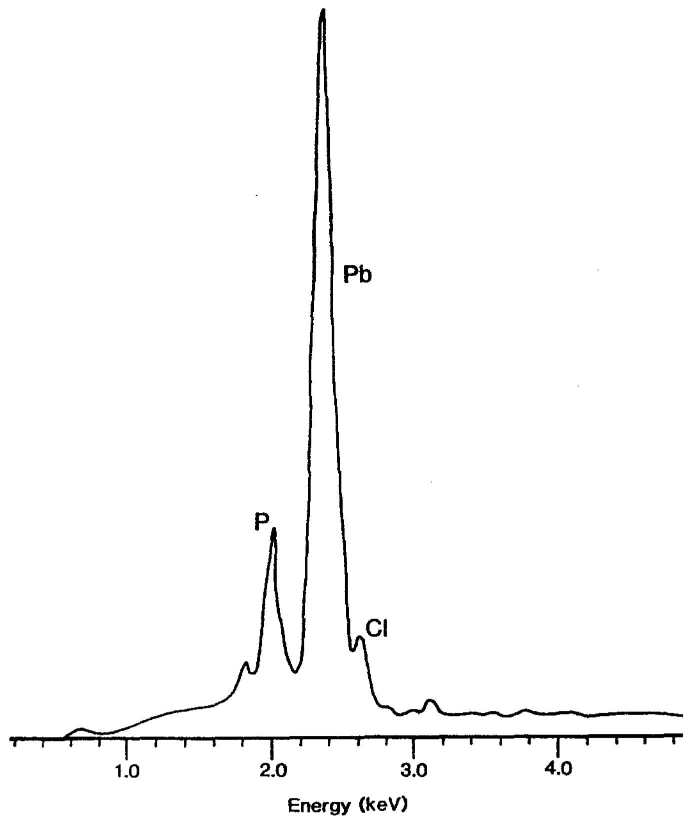


Figure 4(a) and (b) *SEM analysis of grain of pyromorphite [$Pb_5(PO_4)_3Cl$] showing the presence of lead and phosphorus with minor amounts of chlorine.*





Figures 5(*a*) and (*b*) *SEM qualitative energy dispersive spectrum shows a chlorine peak on the expanded scale.*

small population size, prohibiting any rigorous statistical analysis. The overall geometric mean of lead in children's blood (1–8 yr) is 9.4 μg dL^{-1} and 6.9 μg dL^{-1} for those aged 1–3 yr. Typically, children's blood lead reaches a maximum at 2 years (Duggan, 1983; RCEP, 1983). Our apparently anomalous result is probably caused by the extremely small sample size (n = 3).

Discussion

From Table 1, it is evident that mean lead levels in all Winster environmental samples are greatly elevated above averages reported in the UK National Survey (Culbard *et al.*, 1988) and the Birmingham Study (Davies *et al.*, 1990). The mean concentration in garden soil is greater than the national average by a factor of 27. This extreme elevation of soil lead, to the point at which it is very much greater than that of housedust has been found in other historic mining areas (Davies *et al.*, 1985; Thornton, 1988; and Moffat, 1989) and implies a strong external source of contamination entering the home. In Winster village, high lead concentrations (>10,000 μg g^{-1}) in garden soils are generally coincident with mineral veins and old spoil tips, mostly occurring in the older part of the village (Figures 1 and 2(a), Table 2). Household lead levels are known to increase with house age (Thornton *et al.*, 1985; Davies and Thornton, 1987) but this effect could not solely account for the gross elevation of soil lead content. Thus, the primary source of elevated environmental lead levels in Winster is geological mineralisation that has been brought to the surface by past mining activities.

The elevated amount of lead present on Winster children's hands implies that the high environmental lead levels are transferred to the child by the soil-dust-hand (or soil-hand) route. The dust may then be inadvertently ingested by hand-to-mouth activity (Harvey *et al.*, 1985). However, the relatively large amounts of lead on children's hands are not reflected in the childrens blood lead levels (GM = 9.4 μg dL^{-1}), which are within the normal UK range. Recent analyses of children's blood lead have produced geometric means of 11.7 μg dL^{-1} (age 1–3 yr) for Birmingham (Davies *et al.*, 1990), 10.7 μg dL^{-1} (age 6–9 yr) for Edinburgh (Laxen *et al.*, 1987), and 7.3 μg dL^{-1} (age 6–7 yr) for children attending schools situated on heavily trafficked roads (DoE, 1990). Strehlow and Barltrop (1988) examined children's blood lead levels in urban and rural areas over a period of three years. Geometric means of 7.4 to 8.3 μg dL^{-1} were reported for London and 6.4 to 6.8 μg dL^{-1} for Suffolk (age 5–7 yr) and were found to be significantly different for each year. Thus, Winster childen's blood lead concentrations may be considered elevated for a rural UK area but are similar or only slightly elevated compared to urban areas and all values fall below the recommended action level of 25 μg dL^{-1} (DoE and Welsh Office, 1982).

Several authors have predicted a rise in blood lead levels associated with an increase in dust and soil lead concentrations: for example, a literature review by Duggan and Inskip (1985) suggests that, on average, blood lead levels increase by 5 μg dL^{-1} for a 1,000 μg g^{-1} rise in dust or soil lead concentration. If a national dust lead concentration average of 500 μg g^{-1} and a blood lead level average of 10.0 μg dL^{-1} in children are assumed, then the expected blood lead level in Winster children would be 15.0 μg dL^{-1} based on lead concentrations in housedust (GM 1,500 μg g^{-1}). A response-curve of blood lead versus total exposure to dust and water lead for Edinburgh children (Laxen *et al.*, 1987) gives a predicted blood lead of 13.3 μg dL^{-1} for a lead concentration in dust of 1,500 μg g^{-1}. The Birmingham regression model (Davies *et al.*, 1990) produces an increase in blood lead from 11.7 μg dL^{-1} (the geometric mean value for Birmingham) to 13.6 μg dL^{-1} if Winster mean dust lead loading is inserted in the model.

The blood lead levels reported in this study are appreciably lower than predicted values. The above models are based on the urban/inner-city environment where the major sources of lead are fossil fuel residues and paint, whilst the primary source of lead in Winster is of geological origin. Thus, chemical forms of lead present in mine-waste soils will be different to those present in urban soils and dusts, reflecting these contrasting sources. The bioavailability of lead has been shown to be influenced by chemical speciation (Barltrop and Meek, 1975; Chamberlain *et al.*, 1978) and may contribute to the discrepancy between predicted and actual blood lead levels in this study.

SEM X-ray maps show galena weathering directly to cerussite ($PbCO_3$) in the soil environment (Figure 3), consistent with the pH (6.6–8.2) of these soils (Garrels and Christ, 1965). Many lead-rich grains were composed of pyromorphite ($Pb_5(PO_4)_3Cl$) (Figures 4 and 5), which is a stable soil-lead mineral (Nriagu, 1974). The mode of formation of this mineral is not clear. It may have formed in the mineral vein and then been incorporated into the soil or formed within the soil as a result of further weathering of galena.

Pyromorphite is only very slightly soluble ($K_{sp} = 10^{-84.4}$) (Nriagu, 1973), less so than galena ($K_{sp} = 10^{-27.5}$) (Smith and Martell, 1976). It could contribute to a low bioavailability of lead in these soils, below that of urban soils where more soluble forms of lead are present (*e.g.* lead chloride and bromide). A low soil-lead bioavailability would reduce the uptake of lead into the child so that despite increased lead intake via the soil- dust-hand-mouth route, blood lead concentrations could remain within normal UK ranges.

Conclusion

Past lead mining activities have caused the gross elevation of lead concentrations in garden soils and the contamination of housedusts in the Derbyshire village of Winster. There is evidence to suggest that the high levels of lead are transferred in part to the child via the soil-dust-hand-mouth pathway. However, blood lead concentrations of children living in the village are within normal UK ranges. The solid phases of Winster soils contain appreciable amounts of pyromorphite, a stable soil-lead mineral. The extremely low solubility of this mineral may contribute to a low human bioavailability of soil-lead, resulting in the 'normal' blood lead concentrations found in young Winster children.

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