LEAD CONTAMINATION OF ROADSIDE PASTURE

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S U M M A R Y

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Lead released in the exhaust gases of petrol-engined vehicles contaminates the air, soil, and vegetation of the roadside environment. It may, therefore, present a threat to the health of farm livestock grazing near major roads, and might also contaminate animal products which are to be consumed by man.

This thesis investigates the pathways through which farm animals may be exposed to vehicular lead. The study involves the measurement of lead levels in air, pasture, soil and animal tissues, together with a number of field and laboratory experiments. In addition, moss bags were used to investigate rates of deposition of airborne lead to plant surfaces.

The deposition of airborne lead was found to be greatest near the motorway, and this produced elevated levels of lead in pasture grass and soil, particularly within fifty metres of the road. Lead uptake from the soil via plant roots was shown generally to be only a minor source of this metal to herbage, the exception being at background sites during the summer months.

The deposition rate of lead showed a marked seasonal variation which produced corresponding changes in the lead concentration of the grass. Thus, the lead concentration of pasture was ten fold greater during summer than winter months at all sites. For a given rate of deposition, plant growth processes result in there being a ceiling level of contamination of pasture. This growth and decay of leaves also accounts for the low lead levels in the roadside grass during summer compared with winter months, even though traffic flow is greatest at this time.

Results from this study suggest that levels of lead in pasture adjacent to rural motorways are not a threat to the health of farm livestock. In addition, it is recommended that winter grazing near major roads should be avoided so as to reduce the possibility of contamination of offals by lead.

Key words: Lead, Food-chain, Motorways, Pasture, Moss-Bags

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CHAPTER 1

INTRODUCTION TO THE RESEARCH STUDY

1.1 SOURCES OF LEAD IN THE ENVIRONMENT

Lead is a metallic element which is a member of Group VIA of the Periodic Table. It is ubiquitous in nature, being a trace constituent of rocks, soil, water, air and plant and animal organisms. The element has an average crustal abundance of 16 µg/g, but occurs in much greater concentrations as lead ores, the principal one being galena (lead sulphide). It would seem that man has mined lead since prehistoric times, although its use and dispersal in the environment has increased greatly since the industrial revolution. On a global scale, man's present rate of usage is of the order of four million tonnes annually, which is very high compared with the natural rates of mobilisation of lead by weathering and volcanic activity, estimated as only 210,000 tonnes each year (HMSO, 1974).

As a result of man's long association with the metal, it has become clear that, along with the technological benefits, there is also the danger of poisoning of both human and other forms of life. Therefore, in response to this hazard, a vast amount of scientific literature now exists concerning the health effects of lead for man, and the distribution of lead in the ecosystem. These two aspects are reviewed in detail by Waldron and Stöfen (1974) and Nriagu (1978).

The present study is concerned with one aspect of lead in the environment, that is the release of lead particulates from motor vehicles as a result of fuel combustion. Table 1-1 shows that lead used in the manufacture of petrol accounted for only some seventeen per cent of the total amount of lead used in industry in the UK in 1974. However, unlike some industrial uses, none of the lead added

TABLE 1-1

Utilisation of Lead in the UK in 1974

(Butler 1979)

Use	Amount (kg.10^6)	% of Total
Batteries	80.1	24.6
Lead in petrol	56.1	17.3
Ammunition	7.3	2.3
Cable covering	44.4	13.7
Sheet, foil and pipe	47.6	14.6
Collapsible tubes	1.4	0.4
Pigments	36.7	11.3
Alloys	32.2	9.9
Miscellaneous	19.3	5.9
TOTAL	325.2	100

to petrol can be recycled, and thus, all of this lead is released into the environment.

On a worldwide basis, the use of lead in petrol accounts for over 60 per cent of the total emission of this element to the atmosphere (Butler, 1979). In addition, in regions with high vehicle density, vehicular emissions may account for over 90 per cent of the lead in the air (HMSO, 1974). Therefore, the use of lead alkyls in petrol results in this source being a major contributor of environmental lead.

1.2 THE USE OF LEAD IN PETROL

Lead additives have been used commercially in petroleum fuel since 1923. Their function is to prevent combustion of the fuel without detonation, resulting in the efficient use of fuel and minimising engine wear. If the fuel does detonate it creates a shock wave which produces a metallic or pinking sound. In consequence, lead additives are known as anti-knock agents as they prevent the occurrence of pinking.

The anti-knock agents are organo-lead derivatives known as lead alkyls. Two compounds are presently in use, of which tetraethyl lead (TEL) is the most important, while tetramethyl lead (TML) has only been used in the last decade. When fuel is combusted the lead alkyls are converted to lead oxides, which would reduce performance if allowed to deposit in the engine. Therefore, compounds known as scavengers (dibromoethane and dichloroethane) are also blended into the petrol to react with the lead oxides formed during combustion to form halides, which are volatile at engine operating temperature, and are carried off with the exhaust gases. In addition to this inorganic lead, a comparatively small amount of the lead additive will be lost from the motor vehicle by evaporation. This organic lead represents only a very small percentage of the total air lead concentration (Harrison and Perry, 1977) and is, therefore, of minor significance.

The lead content of petrol in the UK is subject to statutory control, (Motor Fuel - Lead Content of Petrol - Regulations 1976 - SI 1976 No 1989), and the current maximum permitted level is 0.45 g/l. Government policy has been to restrict the total amount of lead emitted from motor vehicles in the UK to a level below that reached in 1971. This has resulted in a phased reduction in petrol lead content during the 1970s as shown in Table 1-2. A further reduction to a maximum of 0.4 g/l will come into effect in January 1981 as a result of the EEC Directive adopted in 1978 (78/611/EEC).

Figure 1-1 shows the estimated total lead emitted by motor vehicles in the UK; this assumes that 70 per cent of the lead added to petrol is released with exhaust emissions. The histogram also includes the predicted emission for 1980 and 1981 as reported by the Department of Transport (1979). The data shows that the total lead emission has varied by only about 10 per cent over the past decade and is likely to remain at a level of 7,000 - 8,000 tonnes in the early 1980s.

1.3 VEHICULAR LEAD AS A POLLUTANT

The release of vehicular lead into the atmosphere is not a new phenomenon in the UK. Yet more recently there has been a growing awareness of the possible health effects that may result from lead levels below those producing any clinical symptoms in humans. Countries such as the United States of America and Japan have responded by making lead-free petrol available. This action was partially

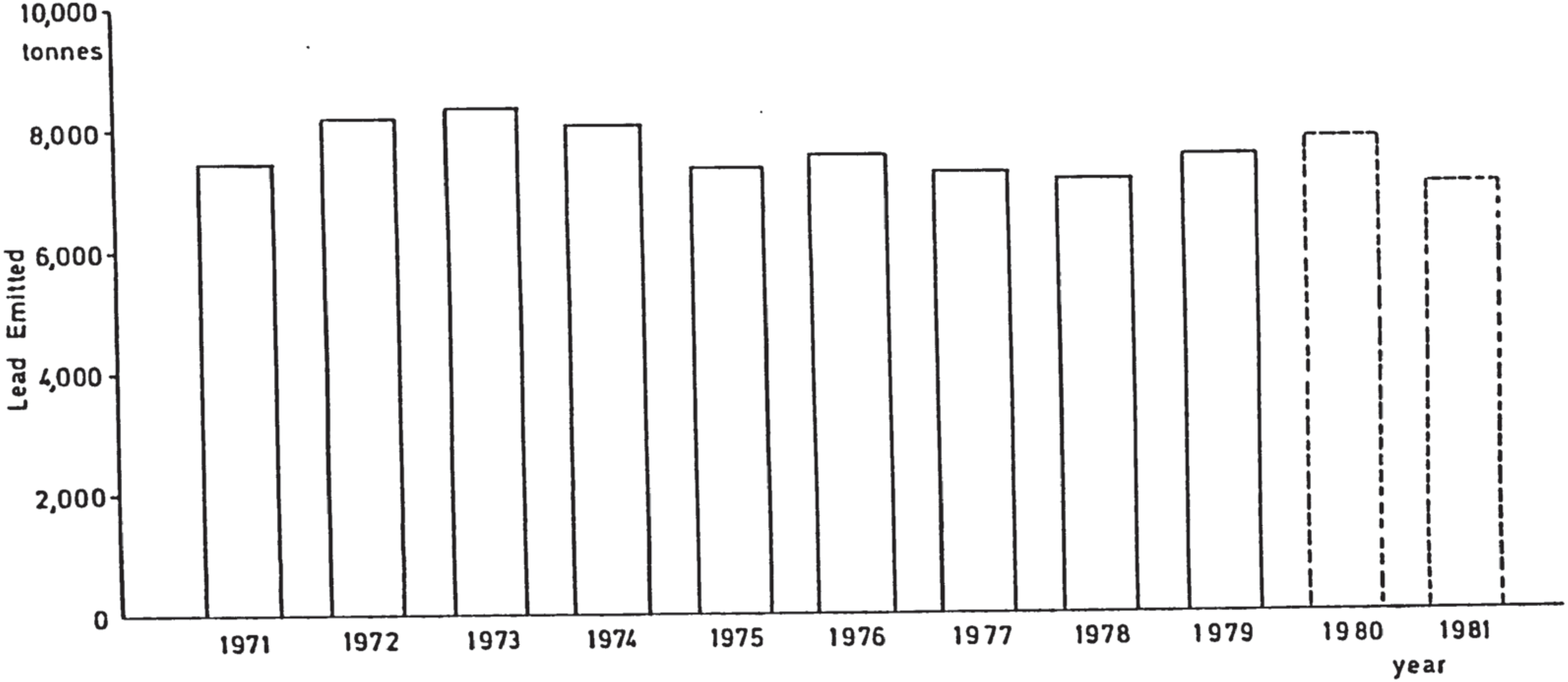
TABLE 1-2

Maximum Permitted Lead Content of Petrol in the UK

Year	Maximum level (g/l)
1971 - 1972	0.84
January 1973 - November 1974	0.64
November 1974 - November 1975	0.55
November 1975 - January 1978	0.5
1978 - 1980	0.45
January 1981	0.4

FIGURE 1-1 Total Amounts of Lead Emitted from Petrol Engined Vehicles in the UK

(Dept. of Transport 1979)



motivated by the need to control gaseous emissions from vehicles by use of catalytic convertors, which are adversely affected by lead.

The exposure of the population in the UK to lead has been recently considered by Lawther et al. (1980) who conclude that lead in the air is a minor source of lead to humans compared with that ingested from food. However, their report does state that the continued fall-out of lead year by year from the air to the ground will increase the concentration in the soil, and may increase the uptake of lead by plants. These workers were unable to assess the contribution that this pathway of lead from the air to man via food makes to the total human body burden of lead. Yet, they believe that the short growing season of many crops, the removal of lead during preparation, washing and cooking, and the fact that crops grown near roads generally form only a small proportion of the total diet, results in lead in air making only a small contribution to the body intake of lead. This conclusion is not in agreement with the findings of all other workers. Tjell et al. (1979) and Rabinowitz (1972) suggest that the majority of lead associated with plants is the result of airborne deposition even at sites quite remote from major roads. This means that atmospheric lead may be an important source of contamination of food ingested by man.

In addition to the direct intake of lead from plants, man may receive lead from animal products as a result of the transfer of lead through the food chain. Animals ingesting crops contaminated by air-borne lead are likely to receive greater amounts of lead than man from this plant source, because forage is not washed or treated to reduce its lead content prior to consumption. Therefore, the possibility exists of the absorption of quite high levels of lead by grazing animals and the contamination of meat and milk products by the deposition of this lead in animal tissues. In more exceptional circumstances, it is conceivable that the ingestion of forage contaminated by lead presents a threat to the health of farm livestock. It would seem, therefore, that the contamination of pasture grass and other forage crops by vehicular lead may be an important aspect of the broader problem of the pollution of the environment by lead. This may be particularly so in areas adjacent to the 2,000 miles of motorway in the UK which, in addition to carrying a high traffic flow, often pass through agricultural regions. It is this aspect of roadside lead pollution that the present work sets out to investigate.

1.4 BACKGROUND TO THE RESEARCH STUDY

During the period 1974-1977 a small team of research workers within the Interdisciplinary Higher Degrees Scheme of the University of Aston in Birmingham were investigating the social and economic effects of motorway development upon the farming community. Financial support came from SRC/SSRC Joint Committee Studentships and a Wolfson Foundation Grant to Dr D J van Rest. Many of the findings of this study, which was carried out in collaboration with the National Farmers Union, are reported by Hearne (1977) and Bell (1978). During the course of the group's work, a number of farmers posed the question as to whether land, next to a motorway, was suitable for agricultural purposes in view of the much-publicised occurrence of elevated lead levels in soil and vegetation near major roads. With this question in mind, Dr C Vick, a member of the IHD research team, undertook a pilot study to investigate the concentration of lead in pasture grass bordering the M6 motorway in parts of the English Midlands.

Vick (1976) found that elevated levels of lead did occur in pasture grass at four sites adjacent to the motorway. It was decided that a more detailed investigation was warranted to assess the significance of this roadside contamination. Thus, the three year study reported in this thesis was initiated in conjunction with the Department of Construction and Environmental Health of the University of Aston, a department which has extensive experience in trace element analysis. Financial support again came from a SRC/SSRC Joint Committee Studentship and the Wolfson Grant. In May 1978 further financial support was provided by The Associated Octel Company Limited who also liaised with the supervisory team. (Despite the company's commercial interest, the relationship was at all times scrupulously correct).

1.5 AIMS AND PURPOSES OF THE STUDY

Preliminary objectives during the first months of the project were to verify the earlier findings of Vick, and to investigate the situation in 1977. Some research was also undertaken to evaluate the suitability of the analytical technique which was to be used to measure the lead content of various biological materials. Once these initial objectives were achieved, the aims and purposes of the research programme were established as follows:-

1. To understand the physical processes governing particulate deposition and the atmospheric lead concentration in the vicinity of a motorway.
2. To investigate the movement of lead in the agricultural ecosystem and to identify the factors determining the level of roadside contamination by vehicular lead.
3. To assess the possible toxicological hazard of lead emitted by motor vehicles, both for livestock grazing

near major roads, and for man as a consumer of milk and meat products from such animals.

4. To suggest practical measures that may be taken to minimise the exposure of farm animals to this roadside lead.

1.6 RESEARCH APPROACH

It was considered that the best approach, to achieve the objectives outlined above, lay in obtaining an understanding of the mechanisms which determine the behaviour of lead once emitted by motor vehicles into the agricultural environment. Thus, it was decided to undertake a detailed case study of a particular farm, bisected by the M6 motorway and situated in Cheshire. Some smaller scale investigations, at other sites, would also be undertaken for comparison purposes. This would allow a number of environmental factors, farming practices, and traffic flows to be closely monitored and investigated for possible correlations with changes in pasture lead concentration. The results obtained could then be compared with published data concerning trace metal pollution at other sites and the possible toxicological effects evaluated. In addition, it was decided to monitor the levels of zinc, a biologically essential trace element, to compare its occurrence in the agricultural ecosystem with that of lead.

Whilst the use of case studies has the disadvantage of perhaps basing generalised conclusions on data taken from atypical or variable sites, it does allow an in-depth study to be carried out with limited resources. It is believed that the site chosen for detailed investigations is typical of much pasture land adjacent to motorways in the UK, and that the study farm is probably more exposed to traffic pollution than most which border a motorway.

1.7 THE CASE STUDY

A. The Region

The site selected for the detailed study of roadside lead pollution is located approximately 1.5 km north of junction 17 of the M6 motorway near Sandbach in Cheshire (OS map reference SJ 762636). This section of the road was constructed in 1962 and its six lanes carried an average traffic flow of over 46,000 vehicles per day in 1978. The site may be described as rural, being situated in the most important dairy farming region in England, and is relatively free of industrial pollution (Cheshire CC. 1977). The location of the study farm and other fields investigated is shown on the enclosed maps (Figures 1-2, 1-3).

The topography of the region may be described as undulating and open, with the study area having an elevation of 60 to 70 metres above sea level. This part of 'The Cheshire Plain' receives an average annual rainfall of 805 mm which varies seasonally, with greatest levels in late summer and winter months. The bedrock of the area is a Keuper Marl, and no mineralisation is known in the region which could enhance the trace metal content of local soils. As a result of the importance of dairying, as well as beef cattle and sheep to a lesser degree, most of the land in the Sandbach district is under permanent pasture.

B. The Study Farm

The study farm is a long established dairy unit with a total land area of 52 hectares. Some 37 hectares remain as a permanent or semi-permanent perennial ryegrass (*Lolium perenne*) pasture. Most of the grassland is situated to the west of the motorway, although three

FIGURE 1-2 Location Map of Study Area in Cheshire

FIGURE 1-3 Map of Motorway Fields at the Study Site

Aston University

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PLATE 1

View of Study Site North of Motorway Bridge

PLATE 2

View of Study Site South of Motorway Bridge

fields are eastward of the road and connected to the central part of the farm by a motorway bridge. The map of the site (Figure 1-3) shows that seven of the fifteen fields are adjacent to the motorway. From Plates 1 and 2 it can be seen that fields A, B and G are level with the motorway, although the land rises relative to the road from south to north. Thus, field C is nearly four metres below the level of the road while field F is above the motorway, which is set in a steep embankment. South of field C, the land again rises relative to the road so that field E is level with the motorway. The site pictures also show that there is little gradient on the motorway itself, and that the road is bordered by a four-bar wooden fence which is situated ten metres from the edge of the hard shoulder of the motorway.

Sixty to seventy adult Friesian cattle graze the pasture from March until November together with up to thirty young animals. The animals are allowed to graze over all the pasture in fields with access to the farm-yard, but strip grazing is undertaken in other areas. Sixteen hectares of grassland are kept for silage which is used as a winter feed for the cattle. All fields have water troughs which receive a piped mains supply and there is little access to surface water on the farm. During winter months, the dairy cattle are stall-fed and are kept in farm buildings situated 200 metres west of the motorway.

The soil of the area is a loamy fine sand with a pH of 6.5-7.4. A soil analysis report was commissioned from the Ministry of Agriculture Fisheries and Food (MAFF) in February 1979 and this found that all fields were satisfactory in lime, potassium, magnesium and phosphorus (Appendix 1). The farmer places a dressing of phosphate and potash (375 kg/ha) in the autumn and nitrate (250 kg/ha) after each cut of grass during the summer. Lime is also added every three or four years to maintain a near-neutral soil pH.

C. Other Study Fields

The study also involved the investigation of a number of other individual fields both near the motorway and distant from it which were not part of the dairy farm. In particular, field 1, located in Figure 1-3, was closely investigated. This pasture was grazed by sheep for the whole year as well as horses, and occasionally cattle in summer. From Plate 2 it can be seen that this field is level with the motorway and has an undulating topography similar to field B west of the road.

Other pastures examined are quite distant from the motorway and are located at points C1, C2 and C3 in Figure 1-3. All fields were perennial ryegrass pastures which were grazed by cattle during the summer months. The soil type was similar to the study farm in terms of texture, pH, moisture and organic carbon content.

D. Other Motorway Sites

As a comparison with the M6 site, a dairy farm adjacent to the M5 motorway in Gloucestershire was also investigated (OS map reference SO 915323). This is comparable to the M6 site in a number of ways including the following:-

a) It is a six lane, rural motorway.

b) There is no known mineralisation in the area which could enhance the trace metal content of soils.

c) The motorway is orientated in a north-south direction.

d) The pasture consists of perennial ryegrass west of the motorway, although it is Italian ryegrass (Lolium multiflorum) to the east.

e) The topography of the area is undulating and the motorway bordered by a four bar wooden fence.

The difference between the M6 and M5 sites include soil types, traffic flow and possibly a small climatic variation.

A further minor investigation of roadside pasture was carried out adjacent to the M1 motorway in Hertfordshire (OS map reference TQ 962144). This differed from the other two sites in a number of ways being only a four lane motorway and situated in a deep cutting. The ryegrass pasture itself was screened from the road by a belt of coniferous trees and a solid wooden fence. Only horses grazed this field, which had an undulating topography, and lay to the east of the motorway.

1.8 THE COLLECTION OF SAMPLES

A. The Sampling Programme

The two year sampling programme was set up with the aim of monitoring the movement of lead from the exhaust pipes of motor vehicles to the various sinks in the environment. In designing the procedure for sample collection, the possible pathways by which lead moves through the ecosystem had to be considered. Figure 1-4 is a simplified flow diagram of the possible pathways by which emitted lead may reach farm livestock and other compartments of the ecosystem.

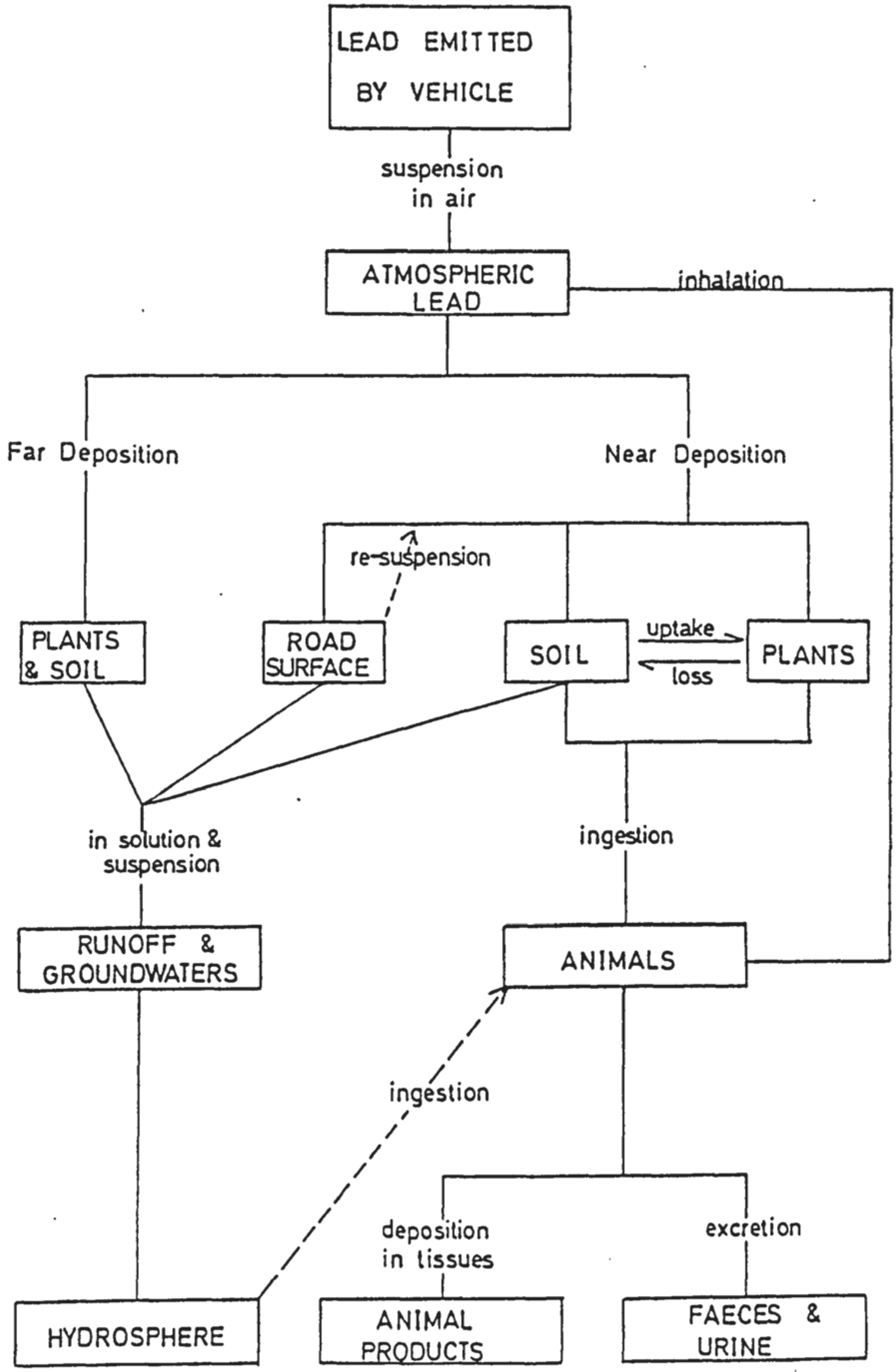
The critical factors determining the initial contamination of the roadside environment by lead are:-

i) the level of emission,

ii) the degree of dispersion,

iii) the rate of deposition of the lead.

FIGURE 1-4 Environmental Pathways of Vehicular Lead at the Motorway Study Sites



These primary factors may be distinguished from secondary processes influencing the movement of lead within the ecosystem. These might include lead uptake from the soil by plants, the growth and decay of leaves, and the effect of rainfall by leaching lead from vegetation surfaces and soils. Thus, these secondary factors may adjust the levels of contamination which result from the primary processes. A sampling programme and a number of experiments were, therefore, undertaken to investigate the influence that these various factors may have upon the level of contamination of the roadside environment, and the exposure of farm livestock to vehicular lead.

B. The Samples Obtained

The following range of samples was collected:-

i) Millipore filters

Air was drawn through a 0.8 μm pore size Millipore filter in the manner described fully in Chapter 2. The suspended particulates collected on the filter were then analysed for their lead content.

ii) Moss Bags

The deposition rate of airborne lead to vegetation was measured by the use of moss bags. A monitoring programme was set up for a period of one year and is discussed fully in Chapter 3.

iii) Pasture Grass

The above-ground portion of grass was sampled from sites both adjacent to and distant from the motorways to determine the level of lead contamination.

iv) Soil

The surface soil was also sampled at the same time as grass and its lead content determined.

v) Other Samples

A number of other materials, including rain water, milk and blood were analysed to study the movement of lead in the agricultural ecosystem.

The research programme, therefore, involved a large number of chemical analyses of a wide range of materials. Details of sample collection, preparation and chemical analysis are referred to in later chapters, but the general principles of sample preparation and analysis for trace metal content will now be considered.

1.9 ANALYSIS OF SAMPLES

A. Sample Preparation

Samples of grass, soil and moss formed the greater part of the materials to be analysed for trace metal content. After collection, all such samples were kept in frozen storage at a temperature of minus 18°C.

In preparation for chemical analysis, the grass and moss was placed in paper bags and dried in an oven for a period of 24 hours at 80°C. Samples were then cut up using stainless steel scissors. A ventilated dust box was constructed for this process as the dry moss produced a large amount of dust which could have contaminated both laboratory surfaces and other samples. Approximately one gram of the dry material was then accurately weighed out into a 100 ml conical flask ready for an acid digestion procedure described below.

Soil samples, on the other hand, were placed on filter papers in a large oven and left for a period of seven days at a temperature of 35°C. The dried samples were then lightly ground with a pestle and mortar, and the material passing a 2 mm sieve placed in a storage bottle.

Approximately 1 g of this soil was accurately weighed out into a Kjeldahl flask in preparation for acid digestion.

All samples of soil, grass and moss were digested with 10 ml of hot, concentrated nitric acid (Analar), which was allowed to simmer on a hot plate or Kjeldahl apparatus for one hour. After this time, the acid was left to cool, diluted with deionised water and filtered using a Whatman No 1 paper. The resulting filtrate was then made to 25 ml with deionised water and used directly for determination of the metal content.

B. Determination of Metal Content

The lead and zinc content of the samples was determined by atomic absorption spectrophotometry using the 283.3 nm and 213 nm spectral lines respectively. A Perkin Elmer (Model 360) instrument was used in the flame mode for zinc and either in the flame or flameless mode for lead. A deuterium arc background corrector was also employed during all measurements. The flameless mode consisted of a Perkin Elmer HGA-74 graphite furnace. Instrumental conditions for both the flame and flameless operations are shown in Table 1-3. All results were recorded on a Perkin Elmer 056 chart recorder.

Appropriate standards were prepared fresh on the day of analysis from 1,000 µg/ml stock solutions. These were made to contain a 5 per cent concentration of nitric acid to resemble the test solutions. Standards in the range 0.5 - 10 µg/ml were used for lead determination by the flame technique, and in the range of 0.1 - 0.5 µg/ml for the flameless mode. For zinc, standards between 0.5 - 4 µg/ml were used.

1.10 STUDY OF THE ANALYTICAL TECHNIQUE

Numerous methods have been suggested for the destruction of

TABLE 1-3

Instrumental Conditions for Atomic Absorption Spectrophotometer

1. Flame Mode (for lead and zinc)		
	Lead	Zinc
Wavelength (nm)	283.3	213
Spectral slit width (nm)	0.7	0.7
Chart recorder response (mV)	10	10
Light source	Electrodeless discharge lamp (Perkin Elmer)	
Background corrector	Deuterium arc lamp	
Flame gas	Air/Acetylene	
Air pressure	3.0 kg/sq cm	
Air flow rate	21 litre/min	
Acetylene pressure	0.85 kg/sq cm	
Acetylene flow rate	3.8 litre/min	
2. Flameless Mode (for lead)		
Wavelength (nm)	283.3	
Spectral slit width (nm)	0.7	
Gas flow (argon)	miniflow/gas stop	
Sample size (µl)	20	
Chart recorder response (mV)	10	
Light source	Electrodeless discharge lamp (Perkin Elmer)	
Background corrector	Deuterium arc lamp	
Drying temp (°C)	154	
Drying time (secs)	30	
Charring temp (°C)	330	
Charring time (secs)	30	
Atomising temp (°C)	730	
Atomising time (secs)	10	

organic matter prior to dissolution for metal determination by atomic absorption spectroscopy. These have been reviewed and discussed by Gorsuch (1970) and Christian and Feldman (1970). Basically the methods involve two types of approach:-

1) Wet oxidation
2) Dry ashing

Each technique has its own particular drawbacks, but with regard to lead, Gorsuch (1970) concluded that wet oxidation techniques were advantageous, giving better recoveries of added lead. He found that mixtures of nitric and perchloric acids, or hydrogen peroxide were most satisfactory.

A number of workers, such as Lagerwerff (1970), have used a four to one mixture of nitric and perchloric acid because of its strong oxidising action. This reagent was used in preliminary studies in the present work, but no advantage was found over concentrated nitric acid. In fact a number of disadvantages for its use exist including the following:-

i) Perchloric acid mixtures are potentially explosive
ii) When using the flameless mode for the determination of lead in perchloric acid digests, pronounced matrix effects were observed, a finding also reported by Julshamn (1977). This resulted in a standard curve which was linear over only a small range of concentration. In contrast, when nitric acid only was used the curve was much improved.

With regard to the nitric acid digests, the following points would suggest its preferred use:-

i) The lead and zinc content of the National Bureau of Standards orchard leaves (reference material 1571) was determined. The results found that this technique gave a recovery of 90 per cent of that obtained by the laboratories of the Office of Standard Reference Materials which used a perchloric and nitric acid digestion. This figure is confirmed by Little (1978, personal comm.) who obtained a recovery of 92 per cent.

ii) The usefulness of this technique is also reported by Chamberlain et al. (1978) of the Atomic Energy Research Establishment (Harwell) for a major study of lead pollution near the M4 motorway (UK).

It was also decided to use concentrated nitric acid to measure the lead content of soil as there is no reliable and universal soil extractant which reflects lead uptake by plants (Burridge, 1979 pers. comm.). A number of other investigations were undertaken to check the reliability of the analytical technique. These include:-

1) The analysis of a number of replicate samples in each batch of thirty.

2) A frequent check of the reproducibility of samples. An example of such a study for grass, soil and moss is shown in Table 1-4. It is possible that a considerable part of the variance in these results may be attributed to the non-uniformity of the materials.

3) The effect of storage time on the lead and zinc content of samples was also examined. It was found that when the acid solutions were stored in glass storage bottles no change in metal concentration was apparent over a period of four weeks. On the other hand, non-acidified solutions did show a loss of lead and zinc on storage.

TABLE 1-4

Reproducibility of Analysis for Moss, Grass and Soil for Lead and Zinc Content

(µg/g dry wt)

Index	Soil		Moss		Grass	
	Lead	Zinc	Lead	Zinc	Lead	Zinc
Mean	23.8	39.7	19.5	40	3.3	20.8
Range	22.5-25.2	37.5-42.5	18-22	34-44	3.0-3.6	18-23.5
Standard Deviation	0.95	2.2	1.1	3.5	0.26	2.0
Number of analyses	6		10		5	

4) An examination of the effect of the length of heating during sample digestion was also undertaken. It was found that a variation in the time for which a sample was allowed to simmer, from thirty to sixty minutes, did not have a significant effect upon the measured value of metal content.

5) Elemental recovery experiments were also undertaken. Excellent recoveries were obtained, these being 90 per cent for zinc and 100 per cent for added lead.

Thus, a high degree of confidence may be assumed for the results obtained in this study. However, because of the effects of sample preparation and the analytical method used, caution is necessary when comparing the results of this work with that of other workers who have used different techniques for trace metal determination.

CHAPTER 2

THE VEHICULAR LEAD SOURCE

2.1 INTRODUCTION

This part of the thesis considers, in some detail, the emission of lead from motor vehicles and the factors which influence its dispersion in the atmosphere in the vicinity of a motorway. The resultant airborne lead concentration has important implications for the rate of deposition of lead to plant and soil surfaces, and also for the exposure of livestock to lead through the process of inhalation.

A review of published papers is presented to describe the characteristics of airborne lead and to show typical concentrations in the atmosphere. These data are then related to both measurements of air lead levels at the M6 study site and to predicted concentrations given by a mathematical model. The findings are used in later parts of this work to explain the mechanisms which determine the levels of lead contamination of roadside pasture, and the significance of inhaled lead for farm livestock.

2.2 LEAD EMISSION

The amount of lead arising from a given length of motorway will be a function of both the total number of vehicles and the concentration of lead in the petrol which is undergoing combustion. Something between 60 and 80 per cent of the lead in the fuel is emitted, whilst the remainder is retained in the lubricating oil, the oil filter, and as exhaust system deposits (Habibi 1970, 1973; Larsen and Konopinski 1962, Hirschler and Gilbert 1964). The level of emission is also influenced by such factors as mode of driving, vehicle age and road gradient. In general, lead emission increases with the age of the vehicle, on inclined slopes and is greatest during acceleration, particularly after periods of stop-start driving conditions. At the motorway study area, petrol driven vehicles are normally travelling at a constant mean speed of approximately 95 km/hour on a level stretch of road.

2.3 PHYSICAL PROPERTIES OF THE EMITTED LEAD

The physical characteristics of lead emitted by motor vehicles has been determined both by the collection of exhaust gases and the examination of suspended particulates collected near major roads. Hirschler (1957) found the emitted lead in exhaust gases leaving the tail-pipe of saloon cars to be from 0.01 µm to several millimetres in diameter. Habibi (1973) established that the average particle size in the exhaust emissions increased with accumulated mileage and during "start-stop" driving conditions.

A number of techniques are available to investigate the particle size distribution of atmospheric aerosols (Butler 1979, Chamberlain et al. 1978). Particle diameters may be expressed in terms of aerodynamically equivalent spheres of unit density, a method utilising the Anderson cascade impactor; equivalent projected area diameters independent of density, by means of electrostatic precipitators; diffusional mean equivalent diameter, using a diffusion battery; or the diameter determined by observational measurements with the electron microscope. The scientific literature contains a wide range of measured particle size distributions for exhaust lead (Little and Wiffen 1977). However, Little and Wiffen (1978) undertook a detailed study during the summer months of the particle size distribution of airborne lead beside the M4 and M40 motorways (UK). They used a number of techniques and found that most of the lead was associated with particles of less than 0.1 µm diameter. It was concluded, by these workers, that the mass median equivalent diameter (derived from the Anderson impactor data) was less than 0.3 µm for lead-containing particles near the motorway, and that the particles did not coagulate to any significant degree. A maximum of 10 per cent of the airborne lead was associated with particles exceeding a diameter of about 2 µm.

However, Butler (1979) has suggested that coagulation of the lead aerosol becomes significant with time and that emitted particulates may become attached to ambient aerosols. Also, Chamberlain et al. (1978), have measured a broad peak in particle diameter between 0.3 and 1 µm for aged lead aerosols in rural Oxfordshire, compared with a mass median equivalent diameter of less than 0.3 µm near the M4 motorway (UK).

Little information is available concerning possible seasonal variations in the particle size distribution of lead aerosols. Chamberlain et al. (1978) report a slightly higher mass median equivalent diameter of particles during autumn than summer in London, but this was not significant for the length of the sampling period. They also found an increase in particle size under foggy conditions which may have resulted from hygroscopic growth of particles, or by a greater aggregation in poorly dispersive conditions. On the other hand, in the United States of America, Gilette and Winchester (1972) found little difference in the aerosol size distributions under different weather conditions at urban and remote sites.

2.4 CHEMISTRY OF EMITTED LEAD

A number of studies have shown that inorganic lead is by far the most important form of lead in the atmosphere (Harrison and Laxen 1977;

Day et al. 1977; Harrison and Perry, 1977). In general, organic lead accounts for no more than 2 per cent of total atmospheric lead, except under certain conditions such as on garage forecourts.

Particulate lead leaves the engine along with other exhaust gases, largely in the form of lead chlorobromide. In addition, oxides, sulphates and phosphates of lead may be present in particles which are temporarily deposited in the exhaust system prior to emission (Habibi 1973). The actual form of lead in the atmosphere presents problems of investigation, mainly because of the small particle size and the chemical reactions which occur in ambient air. Changes may occur, possibly with the aid of photochemical reactions, to produce a complex mixture of oxides, sulphates, carbonates and ammonium salts of lead (Habibi 1973; Olsen and Skogerboe 1975; Ter Haar 1971; Butler 1979). Biggins and Harrison (1978) suggest the compound $(NH_4)_2\ SO_4PbSO_4$, formed by reaction with atmospheric ammonia and acid sulphates, as an intermediate form in the conversion of the lead halide to the sulphate. The chemistry of particulates, and the complex changes which occur in the atmosphere as well as on plant and soil surfaces, would appear to be areas worthy of further study because of their importance in determining solubility and chemical behaviour of the various compounds. Chamberlain et al. (1978) report that lead associated with motor vehicle exhaust, whether fresh or aged, is extremely soluble in distilled water at $20^{o}C$. In contrast, the lead associated with lead oxide aerosols dissolves much more slowly.

2.5 CONCENTRATION OF LEAD IN AIR

The lead concentration of ambient air in the form of suspended particulates is typically of the order 0.1 - 10 ng/m^3 in remote areas of the world, increasing to 0.1 - 1.0 $\mu g/m^3$ in rural areas and

0.5 - 10 $\mu g/m^3$ in urban regions (Nriagu 1978; Lawther et al. 1980). Airborne lead levels at the roadside have been extensively studied (e.g. Butler et al. 1975; Daines et al. 1970; Little and Wiffen 1977; Bevan et al. 1974; Page et al. 1971; Cholak et al. 1968). These, and other workers, have shown the importance of a number of factors determining the air lead concentration in the vicinity of roads, such as traffic density, wind speed and direction, atmospheric mixing height, mode of driving and local topography of the highway.

Airborne lead levels decrease rapidly with distance from a road and this is most apparent upwind of a highway (Daines et al. 1970; Page et al. 1971). Studies in the UK have recently been reviewed by Lawther et al. (1980) who suggested that, in general, airborne lead levels at 50 metres from a road are about 20 per cent of the kerbside value. This rapid decline in air lead concentration with distance from the roadside is a result of vertical and longitudinal dispersion of the pollutant plume, and the deposition of particulates to surrounding soil and vegetation. Calculations by Chamberlain et al. (1979) have shown that upward diffusion of exhaust gases is the most important factor, and that deposition within 100 metres of the road accounts for only about 10 per cent of the lead emitted on the M4 motorway (UK). The possibility of deposition being a more important factor under certain meteorological conditions is discussed in Chapter 3.

Peak airborne lead levels are generally found during periods of high traffic flow, low wind speed and greatest atmospheric stability (Day et al. 1977; Hickman et al. 1973; Bevan et al 1974; Daines et al. 1970). It is, therefore, the combination of source strength and dispersion processes which greatly influence air lead concentration. Chamberlain et al. (1979) and Nishida et al. (1977) show that relatively high lead levels may occur at roads in deep cuttings where dispersion of exhaust gases is poor.

2.6 EVIDENCE OF TEMPORAL VARIATIONS IN ATMOSPHERIC LEAD CONCENTRATION

A. Diurnal

A diurnal variation in airborne levels in the vicinity of some roads is well documented. Butler et al. (1975) and Hogbin and Bevan (1976), for example, show the occurrence of morning and evening peaks in concentration which correlate with periods of greatest traffic flow.

B. Seasonal

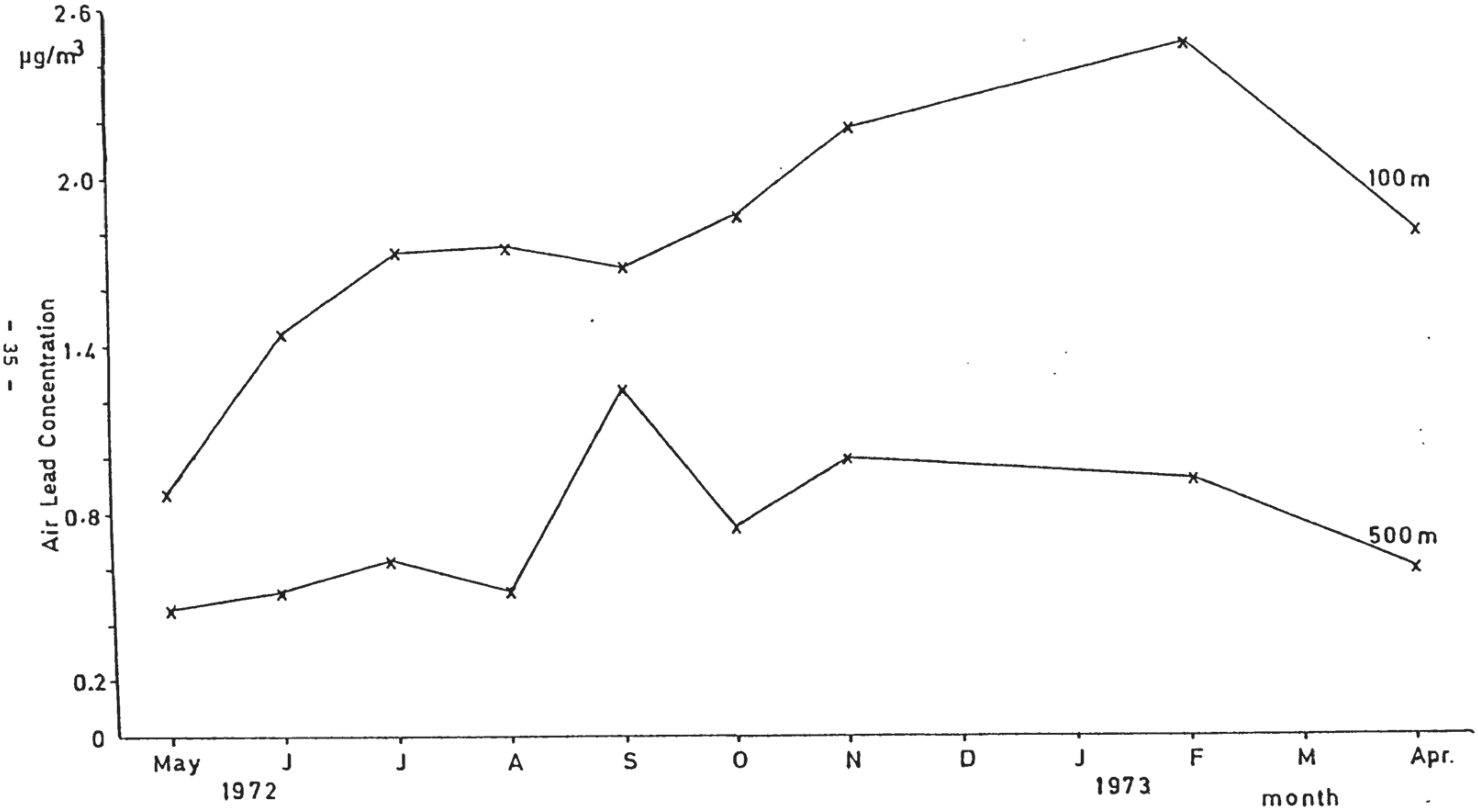
In spite of the numerous studies which have investigated atmospheric lead, few consider a possible seasonal variation in levels. However, four main studies in Britain show the occurrence of higher levels of airborne lead in the vicinity of major roads in winter, although peak traffic flow occurs during the summer months.

i) Butler et al. (1975) investigated the Gravelly Hill interchange on the M6 motorway in Birmingham. Figure 2-1. shows the average monthly air lead concentrations during 1972 and 1973. Peak levels occur during the period November to February and are a factor of 2 to 3 greater than concentrations in May. This seasonal variation is more pronounced at 45 metres from the motorway than at 500 metres due to the diminishing effect of the vehicular lead with distance.

ii) Lawther et al. (1972) report the air lead concentration measured over periods of three months during 1966-70 at two sites in London. The mean lead concentrations were generally found to be 50 to 200 per cent greater in winter than summer months.

iii) Bullock and Lewis (1968) detail a study of atmospheric pollution in Warwick High Street during 1965/66. The air lead concentration

FIGURE 2-1 Atmospheric Lead Levels near the M6 Motorway in Birmingham (Butler et al. 1975)



was found to be directly related to levels of traffic flow, yet to the rear of buildings whose frontage borders the High Street, air lead levels in winter were approximately 50 to 100 per cent greater than summer values, and this variation was not related to changes in traffic volume.

iv) Day et al. (1977) report the results of air monitoring for lead beside an urban motorway in Bristol during three 9 week study periods in 1975/6. Higher lead levels were found in winter and were thought to be the result of the more frequent occurrence of certain meteorological conditions, producing high lead levels during these months. These weather factors were low wind speeds, high relative humidity, variable wind direction and infrequent precipitation.

In addition, Pollitt (1976) found that the highest peaks in air lead concentration occurred during winter in the urban area of Salford. He suggested that this may be the result of a greater lead emission from vehicles in this period.

Chamberlain et al. (1979) make use of the ratio (C/Q), where C = lead concentration near the ground ($\mu g/m^3$) and Q = source strength in a defined area ($\mu g/m^2/S$), and predict a higher value in winter than in summer. This is a result of poor dispersion of the pollutant plume during winter because of the greater atmospheric stability. Based on the data obtained by other workers, Chamberlain et al. found a winter to summer ratio for C/Q of 1.6 for rural areas in the UK.

Data for the average weekly lead concentration of air at the central reservation of the M4 motorway during 1973-75, showed a seasonal change, with peak levels in autumn and lowest levels in winter (Hogbin and Bevan 1976). When the airborne lead values were adjusted for changes in traffic speed and flow during the year (to normalise the data), it was concluded that the seasonal fluctuation was almost entirely explained by the variation in average wind speed. However, the sample site was subject to turbulence produced by traffic, and this may have overwhelmed any possible effects of light winds, temperature inversion and changes of barometric pressure which may be significant a short distance from the road.

Therefore, in summary, the available information indicates the occurrence of seasonal variation in airborne lead levels in the vicinity of roads in the UK. Concentrations some 1.5 to 3 times greater in winter than summer occur, except immediately adjacent to the road where traffic density and turbulence produced by vehicles are the important variables. The higher winter values appear to be largely the result of poor dispersion due to the greater atmospheric stability in this period compared with summer months.

Several workers in countries other than the UK report highest atmospheric lead levels in cities and suburbs during winter months (Australia : O'Connor et al. 1978; Germany : Rentschler and Schreiber 1977; Spain : Marino and Romero 1977; USA : Ewing and Pearson 1974; Colucci et al. 1969; Edwards and Wheat 1978; Japan : Kezaburo et al 1975). In general, these studies show a 2 to 6 fold annual variation in airborne lead levels. Peak concentrations were found to correlate with the occurrence of calm weather, cool air temperature and low mixing depth of the atmosphere. Daines et al. (1970) monitored air lead levels at approximately 10 and 150 metres distance from a highway in the USA and found highest monthly average values during the autumn months when wind velocity and mixing depths are at a minimum. They were also able to show that the total area influenced by lead from vehicles on the road was greatest in this period. Cholak et al. (1968)

did not find any significant seasonal change at a roadside in Cincinnati, USA, but peak lead levels occurred during the autumn months at a sampling site approximately 400 metres distant from the road.

The studies of airborne lead in other countries, therefore, are further evidence of the importance of meteorological factors, particularly atmospheric stability, in determining the atmospheric lead concentration resulting from a pollution source.

2.7 CONCLUSION

Lead is emitted from motor vehicles largely in the form of lead chlorobromide and is predominantly associated with submicron particulates in the engine exhausts. Dispersion in the atmosphere results in a rapid decline in airborne lead levels with distance from a road. The air lead concentration shows a diurnal variation which can be related to traffic flow and a seasonal change which is associated with certain meteorological conditions.

2.8 AIRBORNE LEAD CONCENTRATIONS DETERMINED IN THE PRESENT STUDY

METHOD

Mean airborne lead concentrations during a period of twenty-four or forty-eight hours were taken over periods of two to three weeks in February, June and August of 1979. The sampling site was situated some 200 metres west of the M6 motorway and at a height of 1.5 metres. The findings were then related to the concurrent meteorological conditions as recorded by the Keele weather station (Staffordshire).

An air pump sampler was used to draw approximately $10m^3$/hour of air through a millipore filter of pore size 0.8 µm. The filter was protected by a plastic cover from rain and turbulence and situated at a distance of one metre from the side of a farm building. Ideally, the sampling should be carried out away from all obstructions such as buildings, which both create turbulence and may restrict air movements. However, the availability of an electrical power supply, and protection from rainfall and animals, are essential factors influencing the choice of site. It was decided that the relatively low height of the building (2 metres) and the placing of the filter some distance from the face of the structure would minimise the influence of sampling position on measured levels of airborne lead.

The filters were changed every twenty-four or forty-eight hours. The June and August filters were weighed before and after exposure to measure the total weight of suspended particulate matter collected during the sampling period. Prior to analysis the filters were air dried, cut up with stainless steel scissors and placed in a 50 ml conical flask.

Digestion was carried out by means of 2ml of concentrated nitric acid. The solution was warmed on a hotplate for five minutes and then allowed to cool, filtered and made to 10ml with deionised water. The lead (and zinc) content was then determined using flame atomic absorption spectrophotometry.

RESULTS AND DISCUSSION

Table 2-1 shows the air lead concentrations for each of the sampling periods. Highest average lead concentrations occurred in February 1979 and minimum in August. These values are within the range of 0.1-1 $\mu g/m^3$ given by Nriagu (1978) as typical of rural areas. Peak values in each sampling period can be related to the occurrence of strong easterly winds which carry vehicle emissions from the motorway toward the sampling site. This is in contrast to the airborne zinc levels which have a mean daily concentration of 0.1-0.13 $\mu g/m^3$ in the three sample periods and do not vary with wind direction. This would suggest, therefore, that the airborne zinc 200 metres west of the motorway is the result of regional sources, and that zinc arising from the road has little influence at this distance. Table 2-1 also shows the total weight of particulate material collected on the filters during each 24-hour period. A greater weight of particulate was collected in June and this also had a higher lead content than during August. Values are comparable with those of Cawse (1974) who found 0.51 per cent lead in suspended particulates at seven urban sites in the UK. It is difficult to determine which factors are responsible for the highest air lead concentrations in February and minimum in August, but the following factors may be significant.

(i) Changes in source strength

The amount of lead emitted at the motorway will be proportional to the number of petrol-driven vehicles on the road. Table 2-2 shows the mean 24 hour traffic flows for each of the three sampling periods.

TABLE 2-1

Mean Daily Concentration of Lead and Total Weight of Suspended Particulates 200 metres West of the M6 Motorway

Sampling period (during 1979)	Air lead concentrations (µg/m³)		Total weight of particulate (g)		% lead in particulates
	Mean	Range	Mean	Range	
February (15 days)	0.42	0.09-1.4	-	-	-
June (11 days)	0.36	0.14-0.65	0.0018	0.009-0.0026	0.54
August (13 days)	0.11	0.02-0.65	0.0008	0.0002-0.0015	0.46

TABLE 2-2

A Summary of the Environmental Data Influencing Airborne Lead Levels 200 metres West of the M6 Motorway

Month (during 1979)	Traffic flow* (Mean 24 hr)	Wind direction as % time westerly and easterly		% time** calm weather	Mean daily wind speed (m/s)
		East	West		
February	20,000	21	55	1.5	4.5
June	29,000	38	43	19	1.5
August	33,000	62	3	17	1.75

* For Junctions 14-15 on M6 motorway (pers. comm. 1979, Department of Transport) "- cars only"

** Wind speed less than 0.5 m/s

Mean traffic flows in August are about 65 per cent greater than in February. The density of traffic will also vary greatly from day to day depending on weather conditions and commuter patterns.

(ii) Wind Direction

The motorway is unlikely to have much influence on airborne lead levels 200 metres west of the road if winds are strong prevailing westerlies. During such periods, air lead concentrations at the sample site will be determined by regional sources eastward of this point. Table 2-2 describes the wind direction as determined by the Keele weather station for each of the three sampling periods. The situation is complexed by a wind occurring which is not from the prevailing direction for at least a short period of time on most days.

(iii) Atmospheric Stability

This may be described as the degree of turbulence in the atmosphere which results from convection, eddy diffusion and movement of air masses. The greater the stability the poorer is the dispersion of atmospheric lead aerosols, and this will result in an increase in the airborne lead concentration. In general, conditions of dispersion are poorer in winter than in summer. Atmospheric stability also has a diurnal cycle with greatest stability during the hours of darkness when traffic flow is at the lowest level.

(iv) Rainfall

Chow and Earl (1970) report lower lead concentrations in the atmosphere after rainy periods in California (USA). However, rainfall occurs for only about 10 per cent of the time in the UK and thus it is unlikely to have a significant effect upon the mean 24 hour lead concentration. No correlation was apparent between the occurrence of rain and the airborne lead level at the study site.

(v) Wind Speed

Although wind speed is one factor determining the degree of atmospheric stability, the horizontal movement of air may differ for similar conditions of vertical diffusion. As the air pump sampler does not operate isokinetically, a doubling of the wind speed results in a halving of measured airborne lead levels. Wind speed may vary greatly during a single day and even more so over the 11 to 15 day sampling periods. Table 2-2 shows the mean daily wind speed for each of the three sampling periods, which is greatest in February and least in June.

CONCLUSION

It appears, therefore, that the main factor tending to produce high airborne lead levels at the sampling site is a stable atmosphere when associated with light easterly winds. Variations in traffic flow do not correlate with the measured airborne lead concentrations which are lowest during the month of August when the traffic flow is greatest.

The daily mean lead concentrations are similar for the June and February sampling periods. This is probably the result of the occurrence of easterly winds of similar frequency during these two months. However, the calmer conditions during June would tend to produce a higher airborne lead level than the strong winds of February. The data suggests, therefore, that the poor dispersion conditions in winter, due to the stability of the atmosphere, are sufficient to offset both the influences of higher wind speed and lower traffic flow, both of which tend to reduce the airborne lead levels relative to the summer months.

The lower mean daily lead concentration in August, as compared with June, is probably the result of the prevailing westerly winds. In consequence, the sampling site was upwind of the motorway for the greater part of this period and the air lead concentration was largely dependent upon regional sources westward of the study area.

The effect of environmental factors on air lead levels at the study area was investigated further by use of a mathematical model of dispersion which has been developed by Butler (1979).

2.9 THE BUTLER ATMOSPHERIC DISPERSION MODEL

A. Background to the Model

Meteorological factors are important in determining the degree of turbulence in the atmosphere which has important consequences for the rate of vertical, lateral and horizontal spread of a pollutant plume. A number of workers have attempted to quantify the degree of dispersion of an atmospheric pollutant under varying meteorological conditions and have produced mathematical models to predict airborne concentrations downwind of a source. The background and development of such models is described by Green and Lane (1964), Smith (1975) and Pasquill (1974).

In all applications of dispersion models it must be realised that, at best, they apply to idealised situations of air flow and topography. Pasquill (1974) has stated that "there is a basic inaccuracy and unrepresentativeness of the calculations of dispersion, even in circumstances for which the dispersive action of the atmosphere is most clearly understood". Scorer (1978) points out that theories designed to account for meteorological factors cannot be better than very crude.

Butler (1979) has adapted existing models to predict the air lead concentration resulting from motor vehicles travelling on a level motorway with an undulating surrounding topography. It is, therefore, applicable to the M6 study area. This form of model assumes that turbulent diffusion is far more important than molecular diffusion as a mechanism of dispersion. At a given distance downward of the source the pollutant concentration is regarded as having a Gaussian distribution in the vertical and crosswind directions when averaged over a wide front and with time. The motorway is regarded as a series of mutually independent point sources each producing a pollutant plume, the effect of which is summed to obtain the lead concentration at a point downwind of the road.

The spread of the pollutant plume may be described by the standard deviation of its dimensions in the vertical and crosswind directions. Pasquill (1974) calculated standard deviations from a knowledge of the atmospheric stability which is related to conditions of wind speed and solar radiation. Nine stability categories were developed ranging from unstable (A) to very stable (F) as a practical substitute for direct measurements of turbulence, which are complex to undertake and are not recorded on a routine basis. The original categories have been modified by the Meteorological Office to include the stable category G, producing a total of ten classes. These are shown in Table 2-3.

TABLE 2-3

Characteristics of Pasquill Stability Categories

Wind Speed (m/s)	Daytime* Incoming solar radiation (mW/cm^2)				Within 1 hr of sunset or sunrise	Night time Cloud amount oktas		
	Strong	Moderate	Slight	Overcast		0-3	4-7	8
<2	A	A-B	B	C	D	F/G	F	D
2-3	A-B	B	C	C	D	F	E	D
3-5	B	B-C	C	C	D	E	D	D
5-6	C	C-D	D	D	D	D	D	D
6	C	D	D	D	D	D	D	D

* excluding 1 hour after sunrise and 1 hour before sunset

B. Limitations and Assumptions of the Butler Model

The following limitations and assumptions are made when applying the Butler (1979) model:

i) it applies strictly to open level country;

ii) the material in the pollutant plume should not react chemically or coagulate to form larger particles that may be deposited;

iii) a plume which contacts the ground is completely reflected without loss by deposition;

iv) the position of the plume centre is determined by the wind direction and the mean wind speed employed in the model is representative of the stability category;

v) uncertainties in the value for standard deviation of the plume spread will increase with distance from the source;

vi) the influence of the vehicle wake on the dispersion of the lead aerosol may be accounted for by adding a value of 27 metres to the downwind distance, a correction factor suggested by Calder (1973).

C. Criticisms of the Model

i) An estimate must be made of the percentage of lead in the fuel which is emitted as an aerosol, and this effectively behaves as a gas in the atmosphere for the purpose of the model. This involves two distinct stages:

a) the amount of the combusted lead which is retained in the vehicle. This has been discussed previously and suggested as being about 20 per cent of the lead in the fuel.

b) the amount of lead which is associated with particles which are deposited on soil and vegetation at the roadside. Few workers have measured this factor in the field and the data which is available shows considerable variation.

For the purposes of this model, a value of 12.5 per cent of the emitted lead is presumed to be deposited on the soil and vegatation of the roadside environment. This value represents the lower range of that calculated by Little and Wiffen (1978) for the percentage of the emitted lead that is deposited within 100 metres of the M4 motorway (UK). Accordingly, it is suggested that 70 per cent of the lead in the fuel is in the form of a lead aerosol which is not subject to deposition. Further, the greater part of this deposition occurs within 10 metres of the motorway and the pollutant plume does not undergo any significant losses by deposition after this point.

ii) The effect of vehicle wake on dispersion of the pollutant plume will depend upon both traffic density and the orientation of the wind relative to the road (Sistla et al. 1979). Thus, the factor suggested by Calder (1973), as used in this model, may be inadequate and result in some error of the predicted values of airborne lead.

iii) Sistla et al. (1979) suggest that the Pasquill type of atmospheric stability categories may not properly represent atmospheric conditions near roads, whilst Hickman et al. (1979) believe that the basic Gaussian model for estimating pollutant dispersion does not truly reflect the complex atmospheric motions around roads.

2.10 APPLICATION OF THE BUTLER MODEL TO THE STUDY SITE

The following data were applied to the Butler Model to predict the airborne lead concentration during the period November 1977 to November 1978 :

A) Traffic flow and lead emission rate

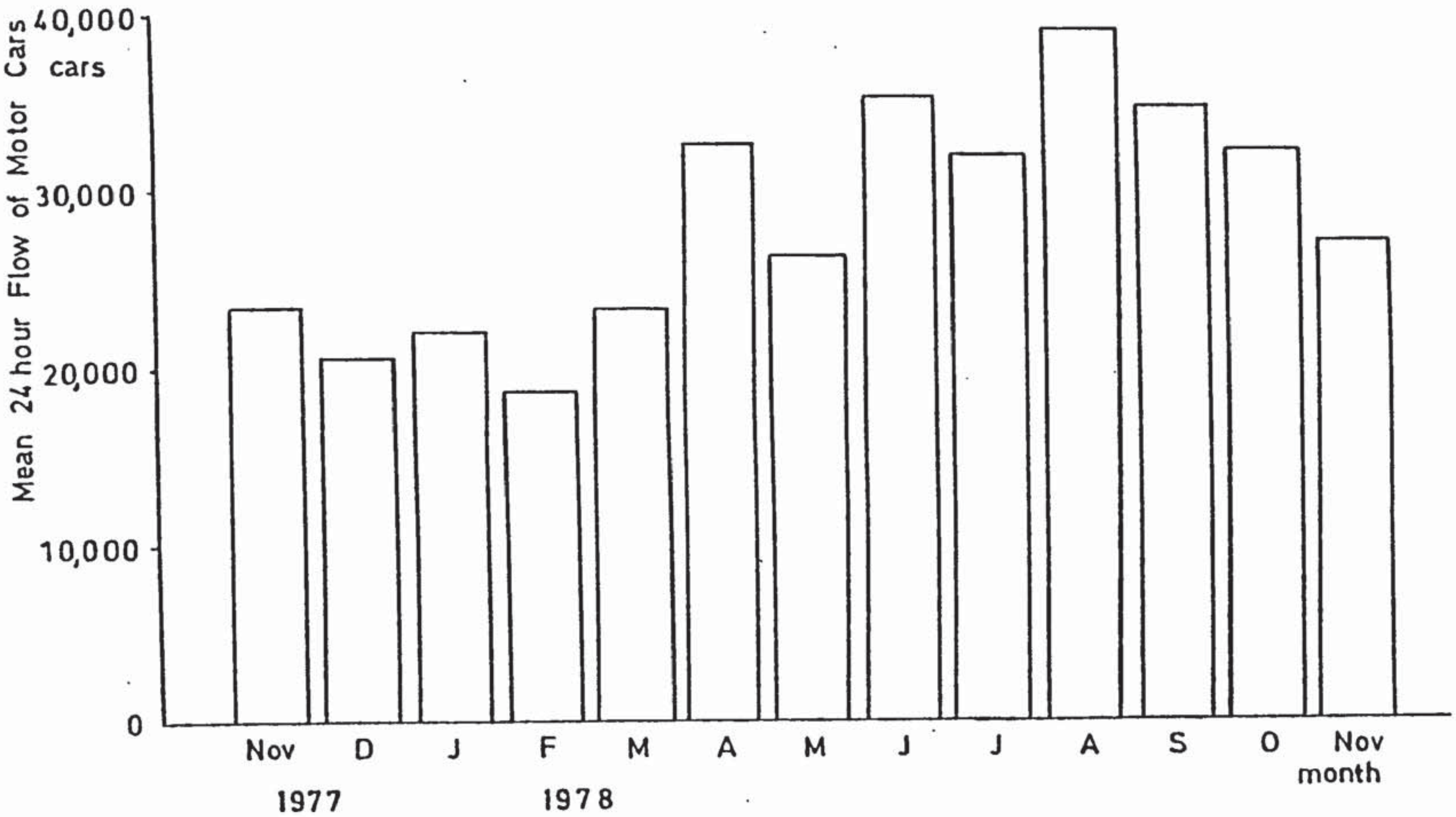
Counts were obtained for the 16-hour Friday, Saturday and Sunday flow of motor cars between Junctions 14 and 15 of the M6 motorway. These monthly statistics are recorded by the Department of Transport, Southwark Street, London. The census point is situated two junctions south of the study area, but traffic density shows little variation between these two stretches of motorway (Department of Environment, 1974). A conversion factor was then applied to calculate a mean 24-hour flow of motor cars. It must be recognised that, in addition to the error from vehicle counting systems, the mean flow may not be representative of the month because of the wide variations in traffic density which results from climatic and human factors (e.g. Bank holidays).

Figure 2-2 shows the seasonal variation in the mean 24-hour flow of cars for each month during the period November 1977 to November 1978. Peak traffic flows occurred in August and are approximately two-fold greater than the minimum in February. Traffic flow may be used to give an estimate of the average rate of lead emission from the motorway by assuming a petrol lead content of 0.4 g/l. Vehicles are assumed to be travelling at an average speed of 95 km/hr (65 mph) and consuming 4.5 litres (1 gall.) of petrol for every 40 km (25 miles) travelled. Also, 70 per cent of the lead in the petrol is estimated as having been released as a conservative aerosol into the atmosphere.

B) Atmospheric Stability

Data for the percentage frequency of occurrence of each of the ten modified Pasquill Stability categories was obtained for Manchester's Ringway Airport, which is situated about 20 km NNE of the study area. It is probable that the absolute frequencies of occurrence of each

FIGURE 2-2 Mean Daily Flow of Motor Cars Each Month between Junctions 14 and 15 of the M6 Motorway in 1977/8



category will differ from the Sandbach site, but relative frequencies in each month should be similar due to both sites being subject to similar synoptic conditions (Farmer 1979 pers. comm.).

Table 2-4 shows the accumulated percentage frequency of occurrence for categories A to C/D and for classes D to G for the 13-month period. The frequency of occurrence of the more stable categories is greatest during winter months. Unstable conditions are most common in May and least frequent in the months November to March.

C) Wind Speed

Although wind speed is a factor used to derive the atmospheric stability categories, each stability class may occur in association with certain ranges of wind velocity (Table 2-3). Thus, a doubling of wind speed during a period of the same stability category will result in a halving of airborne lead levels downwind of the motorway. A clear, seasonal variation in the mean daily wind speed does occur and is discussed in detail in Chapter 3. Minimum velocities occur in May and speeds are generally higher in winter months. Caution must be applied when incorporating wind speed into the model because of its relationship to stability category.

2.11 PREDICTED AIRBORNE LEAD CONCENTRATIONS AT THE STUDY SITE

A) Variation with distance from the motorway

Figure 2-3 shows the predicted decline in airborne lead levels with distance downwind of the M6 motorway in October 1977. The model predicts a 50 per cent decline in lead levels from ten to sixty metres from the motorway. Better dispersal conditions during stability category C result in a mean air lead concentration of 0.75 $\mu g/m^3$ over a 90 metre distance from the motorway fence, compared with a mean of 1 $\mu g/m^3$

TABLE 2-4

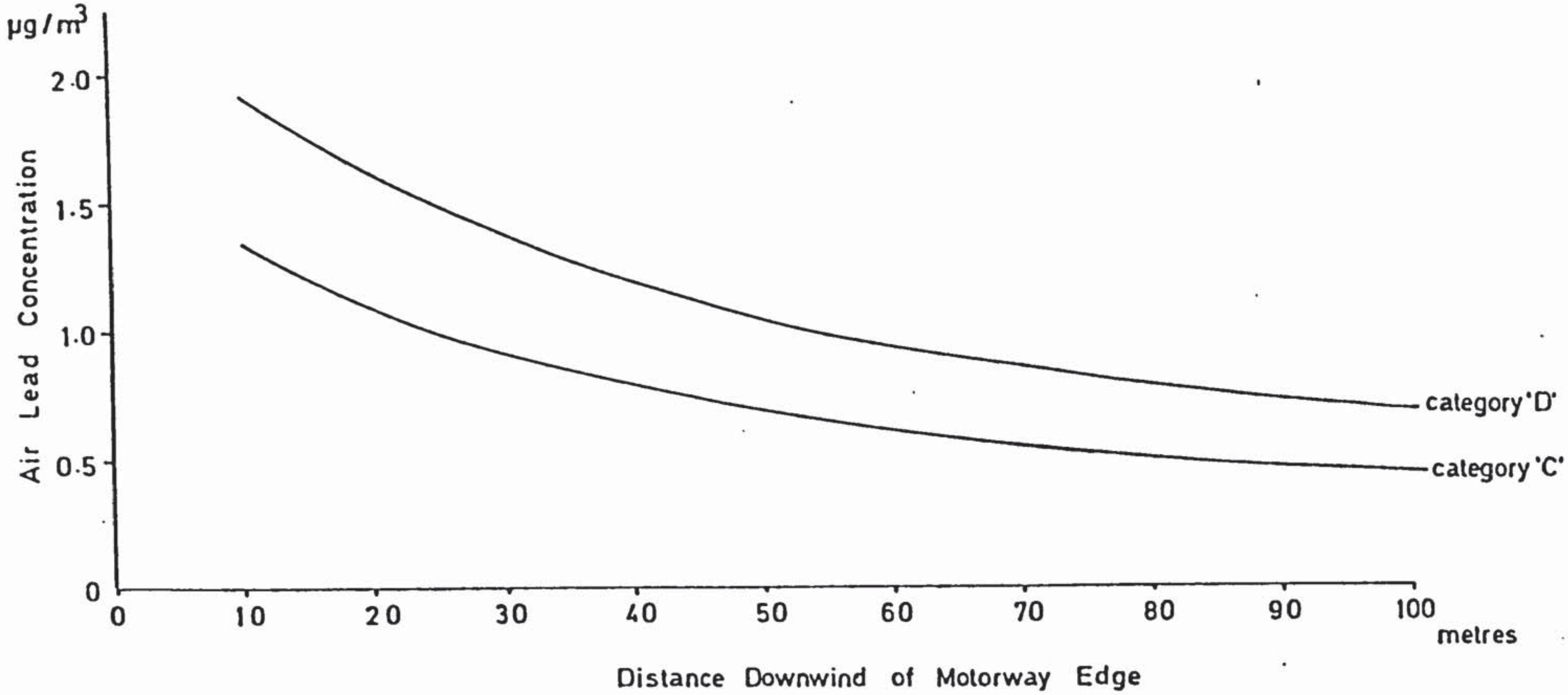
Percentage Frequency of Occurrence of Pasquill Stability Categories

A - C/D and D - G for Manchester during the period

November 1977 to November 1978

Month	% time A - C/D	% time D - G
Nov. 1977	8.1	91.9
Dec.	10.1	89.9
Jan. 1978	9.4	90.6
Feb.	15.4	84.7
Mar.	11.5	89.7
Apr.	27.4	72.6
May	45.6	54.4
June	33.6	66.4
July	37.6	62.4
Aug.	35.3	64.7
Sep.	17.8	81.2
Oct.	19.5	80.5
Nov.	11.2	88.8

FIGURE 2-3 Predicted Airborne Lead Levels Downwind of the M6 Motorway in October 1977 for a Wind Speed of 2m/s and two Different Atmospheric Stability Categories



under category D, with a wind speed of 2 m/s. These values refer to the average airborne lead level downwind of the motorway, whereas concentrations at any particular site will vary continually depending upon traffic flow, atmospheric stability, and wind speed and direction relative to the road.

B) Seasonal variations in airborne lead concentration

The model has been used to evaluate the relative effects of the seasonal variations in traffic flow and atmospheric stability on the airborne lead levels in the study area. Table 2-5 shows the predicted air lead concentration 10 metres downwind of the M6 motorway during November 1977 to November 1978 for each of four stability categories and a wind speed of 2 m/s. Stability categories outside the range of the B to E classes generally occur for less than 10 per cent of the time in any month and are thus of less significance to airborne lead concentrations. A wind speed of 2 m/s is used because it may occur under each of the four stability categories considered. However, mean daily wind speeds are generally greater than 2 m/s except during the calmer summer months.

Categories D - G are most frequent in all months of the year because they represent all night-time conditions as well as an overcast day. The dispersion during daylight hours may be of particular significance as a result of the traffic flow being greatest at this time, especially during summer months. On this basis, Table 2-5 has been divided according to the stability categories which will have the greatest influence on dispersion at the motorway during each month. It presumes, therefore, that stability categories B and C are important during summer months, especially during May, but are of little significance in winter. The airborne lead concentration most representative of each month is a

TABLE 2-5

Predicted Air Lead Concentration 10 metres Downwind of the M6 Motorway under varying Atmospheric Stability Conditions and a Wind Speed of 2 m/s

Month	Lead emission rate from M6 μg /m/s	Air Lead Concentration for different Atmospheric Stability Categories (μg/m^3)			
		B	C	D	E
1977					
November	8.5	0.8	1.1	1.6	2.0
December	8.7	0.82	1.15	1.65	2.07
1978					
January	8.0	0.75	1.05	1.5	1.9
February	6.7	0.64	0.9	1.2	1.6
March	8.4	0.8	1.1	1.5	2.0
April	11.8	1.1	1.5	2.2	2.8
May	9.5	0.9	1.25	1.8	2.26
June	12.8	1.2	1.7	2.4	3.0
July	11.6	1.1	1.5	2.2	2.8
August	14.1	1.34	1.8	2.6	3.4
September	12.6	1.2	1.65	2.4	3.0
October	11.6	1.14	1.5	2.2	2.8
November	9.75	0.92	1.3	1.85	2.3

Key to Table

- (solid box) 80% time D - G
- (dashed box) 60% time D - G
- (partly dashed box) 70-80% D - G
- (partly dashed box) 60-70% D - G

value within the range of numbers enclosed by the boxes in the Table. It is apparent from the Table that the seasonal variation in the atmospheric stability category offsets the differences in source strength (i.e. traffic flow) between winter and summer. The results suggest, therefore, that airborne lead levels in February for a 2 m/s wind speed may exceed those in May despite the considerable difference in traffic flow (Figure 2-2).

It is difficult to quantify the effect of the seasonal variation in mean daily wind speeds upon the air lead concentration. The resolution of the model is probably not sufficient to warrant a detailed study of wind speed under each stability category for every month. In addition, the monthly variations in wind speed are partially accounted for by the incorporation of wind data into the stability category. Thus, the predicted values of airborne lead in Table 2-5 are probably higher than actual levels for all months except May and August when mean wind speeds were approximately 2 m/s. During November, December and March mean daily wind speeds are twice the 2 m/s value and about 50 per cent greater during the remaining months (see Chapter 3, Figure 3-9). Thus, although the values in Table 2-5 tend to overestimate concentrations during most months, the model is able to show that changes in atmospheric stability counteract the effect of the seasonal variation in traffic flow upon airborne lead levels in the study area. Yet it would seem that, on average, differences in mean daily wind speeds result in lower predicted air lead concentrations in winter than in summer months. This is in contrast to the published work reviewed previously and to the few air lead measurements taken at the M6 study site. Therefore, it appears that the atmospheric dispersion model only partially explains the measured seasonal variation in airborne lead near major roads in terms of traffic density, atmospheric stability and mean daily wind speed.

2.12 COMPARISON OF PREDICTED AND MEASURED AIRBORNE LEAD LEVELS 200 METRES WEST OF THE MOTORWAY

A number of difficulties restrict the usefulness of a comparison between the predicted airborne lead levels and those actually measured 200 metres west of the M6 motorway. These are as follows :

i) traffic density varies considerably from day to day and information specific to the periods of air sampling is not available;

ii) the sampling site was rarely downwind of the motorway for a full 24-hour period, thus making most measured values incomparable to the predicted levels;

iii) the model is unable to account for winds which are not perpendicular to the road and for the effect of calm weather conditions upon air lead levels. These factors may not be of great significance at the study site because winds almost parallel to the M6 (i.e. within 15^{o}) generally occur for less than 15 per cent of the time in any one month and calm weather rarely occurs for more than 10 per cent of the total time. In addition, Calder (1973) shows that the pollutant concentration downwind will be quite insensitive to small changes in the orientation of the wind with respect to the motorway;

iv) the situation of the sampling site may be criticised because of the effect of buildings upon air flow;

v) a number of stability categories may occur during a 24-hour sampling period.

Thus, while recognising these problems, Table 2-6 compares the predicted air lead concentration 200 metres downwind of the motorway with measured values during days of prevailing easterly winds. The predicted values are based on the mean daily wind speed for each of the sampling periods, and the prevalence of category D in February, and category C in June and August 1979. Predicted values do, therefore, fall within the range of measured values obtained at the study site, although the model does not explain the higher values which occurred in February 1979. These peak winter values are not associated with periods of low wind speed and it seems unlikely that traffic flow varied sufficiently to account for the variation. Thus, the peak values may be a result of the siting of the air sampler, or represent air pollution episodes which the model is unable to predict because of the limits upon its resolution. This inability of the model to account for peak winter lead concentrations may explain why predicted lead levels do not show the same seasonal variations that were found by the field studies of other workers reviewed earlier in this chapter.

The measurements obtained 200 metres west of the motorway may also be used to obtain a value for the importance of regional sources of airborne lead at the study area. During the days of strong westerly winds that occurred in the three sampling periods, airborne lead concentrations ranged from 0.02 to 0.24 $\mu g/m^3$. These values, therefore, are less than 10 per cent of the predicted concentrations 10 metres downwind of the M6 motorway. Insufficient data was obtained to investigate any possibility of seasonal variations in the importance of regional sources, although Cawse (1974) found that air lead and zinc concentrations were about 1.7 times higher in winter than in summer at non-urban sites in the UK. Measurements at the M6 site have not identified any seasonal variation in airborne zinc levels although peak daily values occurred in February.

TABLE 2-6

Comparison of Measured and Predicted Airborne Lead Concentrations 200 metres Downwind of the M6 Motorway

Month (During 1979)	Mean daily wind speed (m/s)	Measured air lead concentration ($\mu g/m^3$)	Predicted air lead concentration ($\mu g/m^3$)
February	4.5	1.07 1.41 0.83 0.23 0.11	0.12
June	1.5	0.36 0.65 0.51 0.21	0.49
August	1.75	No data	0.45

2.13 CONCLUSION

An atmospheric dispersion model has shown that airborne lead levels decline rapidly with distance from the road. The model demonstrates that changes in the stability of the atmosphere tend to offset the seasonal variation in traffic flow. However, this method is unable to account for the peak airborne lead concentrations which occur in winter months. These were found both, by measurement at the M6 study site, and are confirmed by other studies in the UK. In general, however, predicted levels do agree favourably with on-site measurements.

Regional sources result in an ambient air lead concentration which is probably no greater than 10 per cent of the concentration 10 metres downwind of the M6 motorway. The model predicts that lead levels at the motorway fence (10 metres from the road edge) are unlikely to exceed 3 $\mu g/m^3$ for any considerable period of time.

CHAPTER 3

THE DEPOSITION OF ATMOSPHERIC LEAD

3.1 INTRODUCTION

The release of lead from motor vehicles and its dispersal in the atmosphere have been discussed in the preceding Chapter. In time, much of this lead will be removed from the atmosphere and deposited on the earth's surface, whilst the remainder enters plant or animal systems. Deposition of airborne lead close to major roads may result in elevated levels of this metal in soil and herbage which could, in turn, present a risk of toxicity for grazing animals. In addition, deposition far from the source may result in a subtle increase in the lead concentration of soils and the food crops grown there.

Initially, this Chapter considers the mechanisms of deposition of atmospheric lead with reference to the published findings of other workers. Data concerning measured rates of deposition in the M6 (Sandbach) study area are then reported and discussed in relation to environmental conditions prevalent at this site.

3.2 THE PROCESSES OF DEPOSITION

The physical characteristics of suspended particulates are important determinants of the means by which lead is removed from the atmosphere. Particle size will be a major factor determining the transport of aerosols in the atmosphere by influencing the rate of sedimentation and adhesion to surfaces. Chapter 2 has demonstrated that lead emitted from motor vehicles is largely associated with submicron-sized particles.

The rate of deposition of lead from the atmosphere to the earth's surface has been estimated as 0.8 mg/m^2 in the northern hemisphere and

0.4 mg/m^2 in the southern (Nriagu, 1978). Cawse (1974) calculated average lead deposition rates in non-urban areas of the UK as 40 mg/m^2 for the year 1972 and 27 mg/m^2 in 1973. Although there appears to be a considerable difference between these figures it should be noted that they were calculated from only seven sampling sites. An average value for the UK is given by Chamberlain (1974) as 36 mg/m^2/year.

There are two principal mechanisms by which lead-containing particulates are removed from the atmosphere; these are wet and dry deposition. The mechanisms are discussed below in relation to the range of particle size associated with lead aerosols released from motor vehicles.

A. Wet Deposition

Wet deposition includes all matter brought down to the earth's surface by precipitation. Lead in rainfall results from two distinct processes;

a) Washout - whereby particulate matter is collected by precipitation from the atmosphere below the cloud base.

b) Rainout - which results from the incorporation of particulates into water droplets during cloud formation.

Rainout mechanisms depend upon the capture, by cloud droplets, of Aitken nuclei (i.e., particles of radius < 0.1 μm) as a result of Brownian diffusion, whilst washout removes particles of radius 5 - 10 μm by inertial impaction. There is, therefore, a range of particle size (approximately 0.05 - 5 μm) for which aerosols are not efficiently removed by rain, and this is termed the 'Greenfield window'.

Thus, on the basis of the results of particle size distribution obtained by Little and Wiffen (1978), referred to earlier in Chapter 2, it would seem that most of the lead emitted by motor vehicles occurs as a particulate which is efficiently removed by rainout processes, even though a considerable proportion may fall within the 'Greenfield window'.

The contribution that wet deposition makes to the total removal of atmospheric lead is discussed by Coello et al. (1974) and Nriagu (1978). Coello et al. (1974) state that lead removal by precipitation, is a major mechanism. However, Nriagu (1978) suggests that the relative removal efficiency of wet and dry processes is difficult to estimate, and will depend upon distance from the source, weather conditions, initial release height and pollution type. Calculations of flux rates due to wet and dry processes for particles of 0.1 - 1 µm diameter show the two mechanisms to be of comparable importance (Gatz, 1975). Further support for this finding comes from Cawse (1974), who found that dry deposition of lead, and zinc, to a filter paper was 52 and 44 per cent respectively of total deposition of these elements at rural sites in the UK. These findings may be criticised however, as deposition of particles to a filter paper may not be representative of that to the ground surface. At the roadside, Chamberlain (1975) has shown that rainfall has a negligible effect upon primary deposition.

B. Dry Deposition

Dry deposition consists of the processes of sedimentation, diffusion and impaction of particles on to surfaces. Gravitational settling is only important for particles greater than 0.3 mm in diameter and is, therefore, of little significance for lead from motor vehicles (Butler, 1979). Particles between 5 and 50 μm in diameter may be precipitated by turbulent deposition at the kerbside, but this process will also be of minor importance for vehicular lead emissions.

For submicron particles, the diffusion and impaction on to surfaces will be the predominant form of dry deposition (Chamberlain, 1975). The rate of deposition by these two processes depends not only upon particle characteristics and meteorological factors, but upon the nature of the surface itself (Chamberlain and Chadwick, 1972; Little and Wiffen, 1977). Thus, any measure of dry deposition of submicron particles is only applicable to the collecting surfaces used in the study or, at best, can only be related to other materials which have a similar surface structure.

A number of workers have investigated the factors which determine dry deposition rates of submicron particles to soil and vegetation surfaces. Wedding et al. (1975) used a wind tunnel fumigation system to measure deposition of 0.3 μm diameter particles to leaf surfaces. Total deposition on rough, pubescent leaves was ten times greater than to smooth, waxy leaves. Carlson et al. (1976) used soya bean leaves (Glycine max) fumigated with a lead chloride aerosol (1 - 3 μm diameter). Negligible amounts of lead were removed by wind re-entrainment, even at speeds of 6.7 m/s, even though this produced a rapid fluttering motion of the leaves. Chamberlain (1975) reports that particles smaller than 5 μm in diameter are unlikely to bounce off a surface on impact unless the incident wind speed is very high and the surface smooth and hard. He also states that, for these small particles, the collection efficiency greatly depends upon the micro-roughness of the surface rather than stickiness or wetness, which are more important for the larger particles.

The efficiency of deposition to a surface may be described by the term deposition velocity (Vg). This is defined by the following expression:

$$Vg = \frac{\text{particle deposition flux}}{\text{particle concentration above surface}} = \frac{\mu g/cm^2/s}{\mu g/cm^3} = cm/s$$

For very low wind speeds, the deposition velocity will be almost equal to the terminal velocity of the particle, but at higher speeds, impaction on surface roughness elements becomes progressively more important (Chamberlain, 1967).

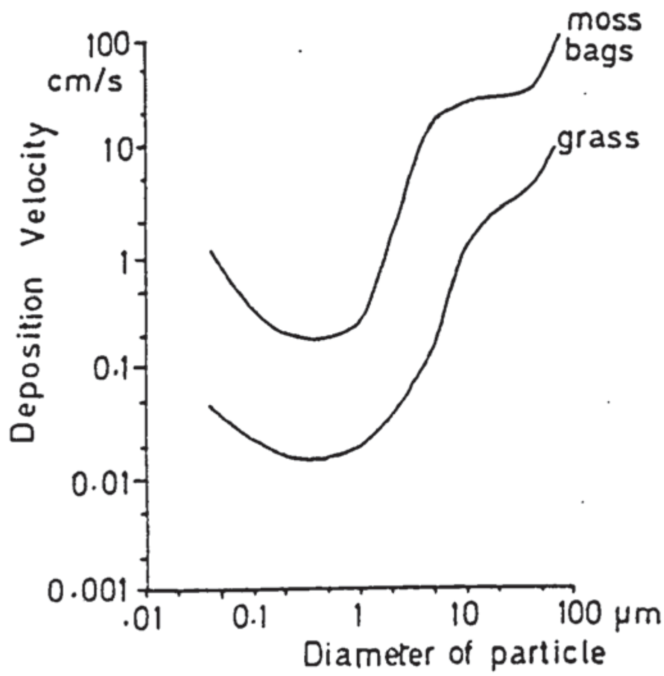
Little and Wiffen (1977) used radio-actively labelled petrol engine exhausts to investigate dry deposition to leaves from eleven tree species. Their wind tunnel investigations showed that rough, or hairy surfaces, collected up to eight times more lead than smooth leaf surfaces. Deposition velocities to all leaves increased with wind speed and were greater for fresh aerosols than aggregated, aged exhaust emissions. These workers also compared deposition velocities to senescent and living foliage, and found that the effect of leaf condition on deposition was species dependent.

Clough (1975) discusses the importance of particle size on deposition velocity to grass. Figure 3-1 shows that deposition velocity, for a given wind speed, increases with particle size for diameters of 1 to 100 µm. A minimum value for Vg occurs for particles of 0.5 µm in diameter, with an increase for smaller particles, due to the greater importance of molecular diffusion. This explains the higher deposition velocity for fresh exhaust aerosols (0.01 - 0.02 µm diameter) compared to that for aged aerosols (0.2 μm diameter) found by Little and Wiffen (1977). It also suggests that deposition is more efficient for lead aerosols at the roadside than for aggregated particulates found some distance from the source.

Therefore, in conclusion, the amount of lead which will be deposited on a plant or soil surface relative to the concentration of particulates in the air, will increase with wind speed and surface roughness. It will also vary with particle size, being least for particles of approximately 0.5 μm in diameter but will increase progressively for smaller and larger diameters.

FIGURE 3-1

The Effect of Particle Size on Deposition to Moss Bags and Grass (Clough 1975)



3.3 LEAD DEPOSITION NEAR ROADS

Few studies have attempted to estimate the fraction of the total lead emitted by motor vehicles which is deposited on soil and vegetation near to major roads. Two distinct approaches have been used by Little and Wiffen (1978) to study lead deposition near the M4 motorway (UK). This road has a traffic flow of approximately 90,000 vehicles per day.

(i) Long Term Study

This involves the calculation of the total amount of lead which has been released by motor vehicles since the opening of the road, and also that which is present in soil and vegetation near the road. The difference in values is assumed to be the amount of emitted lead removed from the roadside environment by atmospheric transport.

(ii) Short Term Study

This is based on the direct measurement of lead deposition rate to grass surfaces at the roadside over a relatively short period of time. This information is used, together with values of deposition velocity to soil and road surfaces (obtained from wind tunnel experiments), to calculate the percentage of the lead that is currently being released and which is deposited near the road.

Using the long term approach, these authors report a value of 40 per cent for the proportion of emitted lead which was deposited within 100 metres of the motorway. This includes lead deposited on the road surface and the central reservation, as well as grass and soil bordering the road. The short term approach gave a mean value of only 10 per cent during dry weather conditions in the month of August 1976, although it was recognised that values would vary widely depending on meteorological factors such as wind speed. These workers suggest that the discrepancy between the two values obtained by the different methods may be partially explained by the process of re-suspension of lead from the road surface. This is considered to be of particular importance in wet weather when the lead on the road surface is carried by the spray to adjoining areas. However, because the lead will be present in large water droplets, this will be deposited mostly within 5 metres of the carriageway. Subtraction of the quantity of lead deposited on the road surface from the value of deposition obtained by the long term approach, gave a value of 22 per cent for the proportion of emitted lead which is accounted for by soil and vegetation within 100 metres of the motorway. Thus, it was concluded that between 10 and 30 per cent of the emitted lead is deposited within 100 metres of the motorway.

This study of lead deposition near the M4 motorway may be criticised for not investigating possible seasonal variation in the deposition rate of airborne lead to the grass surface. A greater deposition in winter months could partially explain the differences found in the results of the long and short term studies. In addition, Wesley et al. (1977) suggest that diurnal variation in atmospheric stability should be taken into account in order to evaluate the deposition processes over natural surfaces. Thus, periods of several hours, as used to measure lead deposition to grass at the M4 study site, may not be representative of deposition for a full 24 hour period. An investigation of seasonal variation in the deposition rate of lead to roadside vegetation is presented later in this Chapter.

A number of other estimates of the proportion of the emitted lead which is deposited near roadways comes from studies undertaken in countries other than the UK. In New Zealand, Ward et al. (1975) used the long term approach to study lead deposition near a road with relatively low traffic density (only 1,200 vehicles/day). These workers calculated that approximately 58 per cent of the emitted lead had been deposited on soil and vegetation within 250 metres of the road. It is unlikely that this figure would differ for a distance of only 100 metres because, as discussed in Chapter 2, most of the lead deposited near roads settles out within 50 metres. Other estimates are given by Huntzicker et al. (1975) and Reiter and Katen (1971) for roads in the USA. These workers calculated values of 6, and 10 to 30 per cent respectively for the proportion of emitted lead which is deposited within 150 metres of a road, based on the mechanisms of atmospheric transport and deposition of lead.

In conclusion, therefore, the available evidence suggests that a relatively small proportion of lead emitted by motor vehicles is deposited within 100 metres of the road. Most of the lead is transported away from the roadside environment as a result of dispersion in the atmosphere, and is eventually deposited over a much wider area some considerable distance from the source.

One method for examining the deposition rate of metals to vegetation is by the use of moss bags. A discussion on their usefulness and suitability for this purpose is now presented.

3.4 MOSS BAGS AS A MEASURE OF TRACE METAL DEPOSITION

Ruhling and Tyler (1968) measured the metal content of indigenous mosses in Sweden as an indicator of the level of non-ferrous metal contamination of an environment. This idea was extended by Goodman and Roberts (1971) who transplanted mosses from an uncontaminated to a polluted site in South Wales (UK). They continued this experiment further by suspending the contaminated moss in nylon mesh bags and concluded that moss was a useful indicator and integrator of aerial metallic burdens. The moss bag was, therefore, an extension of the idea of using vegetation as an indicator of atmospheric pollution, such as had been undertaken by Everett et al. (1967) in the UK with privet leaves.

Perhaps one of the best-known studies using moss bags was that of Little and Martin (1974) who monitored heavy metal pollution around a zinc and lead smelting complex at Avonmouth, UK. These workers wished to investigate the quantities of metals actually impinging on vegetation, and thus, a sampling technique which resembled the plant cover itself was considered most applicable. Their paper discusses the physical nature of moss shoots, which readily entrain and entangle particles and provide a large surface area for metal ion binding. They point out that it is important that the exposure period of the moss should not be sufficiently long for saturation of all ion exchange sites to occur, or for replacement of one ion by another, higher in the electro-chemical series. In addition, if exposed for too long, it is suggested that the capacity of the moss to retain increasingly large burdens of insoluble particulate may be exceeded. Deposition rates of airborne lead to moss bags were calculated for their study area and found to be up to 339 μg lead/g moss/day near the smelter, and 0 - 32.6 μg/g/day at greater distances. Variation between months was thought to be largely due to fluctuations in production at the smelting complex. During the Avonmouth study, Little (1974) demonstrated the importance of the height of the suspended moss bags on metal deposition rate. Raising the height of the bag from 0.1 to 0.6 metres above the ground surface resulted in an increase in the deposition rate. This was believed to be associated with the change in wind speed with height which results from the decreasing effect of ground surface friction.

A further study involving moss bags is that of Ratcliffe (1975). He used indigenous moss and grass samples, and suspended moss bags in the vicinity of a lead battery factory to monitor atmospheric lead deposition. The moss bag data were analysed for a correlation with lead levels in indigenous moss and grass, and with meteorological factors. The correlation of the lead content of moss bags, after exposure for one month, with lead levels in indigenous moss and grass was poor. He suggests that this was the result of differences in the exposure of moss bags and indigenous vegetation, changes in the rate of grass growth, and a seasonal fluctuation in the lead content of the grass. No studies were undertaken during the winter months when little grass growth occurs. A good correlation with the lead content of moss bags, collected after one month, and the wind speed and direction for that month, was found, particularly at greater distances (2.4 Km) from the factory. Short term studies showed that meteorological parameters accounted for only 40 per cent of the weekly variation in lead accumulation by moss bags, but this was thought to be due to the fact that a one week exposure of the bag is insufficient to measure slow deposition rates accurately.

These earlier studies, therefore, concluded that moss bags are a useful indicator of the aerial input of metals on to vegetation surfaces. However, to provide further information, the moss bag must be studied to investigate how it reacts to factors such as air metal concentration, particle size, leaching by rainfall, and wind speed. It also needs to be compared with other measures of particulate deposition (e.g., deposition gauge) and the actual deposition to the indigenous surface vegetation. Such factors have been studied by a few workers using both field and wind tunnel experiments. Clough (1975) used radio-actively tagged particles of a wide size range (0.03 - 32 μm) to investigate deposition to moss bags and trays of Italian ryegrass under varying wind speeds in a wind tunnel. Results showed that large particles were collected more efficiently than those of smaller size, although, as discussed previously, the deposition velocity (Vg) increases for very small particles (Figure 3-1). For all particle sizes, Vg increased with higher wind speeds. Thus, Clough concluded that the collection characteristics of moss bags vary with particle size and wind speed in the same way as grass surfaces, which is the vegetation above which moss bags are usually suspended. In addition, he found that the deposition velocity to moss bags was considerably greater than to grass, due to differences in the plant surface characteristics (Figure 3-1). These findings were confirmed by Little and Wiffen (1977) who used radio-actively labelled petrol engine exhausts in a wind tunnel study of deposition velocity to grass and moss surfaces.

Further findings of the wind tunnel investigations undertaken by Clough (1975) include:

a) A higher deposition velocity for > 20 μm diameter particles to wet than dry moss surfaces as a result of reduced bounce-off. This was, however, not significant for smaller particles.

b) The prolonged spraying of moss bags with water was ineffective at removing particles.

c) Once deposited, particles were not re-entrained from moss bags by the wind.

Clough suggests that moss bags are not suitable as monitors of air particulate content, but they are a satisfactory measure of dry deposition if sheltered from the rain. He calculates that the deposition velocity of 0.5 μm particles to a moss bag, suspended at a height of 1.5 metres, is approximately 15 times greater than the rate to grass surfaces on the ground. However, the collection of rain droplets is probably of quite similar efficiency for both moss and grass. Therefore, the use of moss bags for the assessment of total deposition to grass is not satisfactory as the results obtained are critically dependent upon the fraction of the deposition which is the result of rainfall. This paper does not, however, consider the relative importance of wet and dry deposition as a source of airborne pollutants to vegetation in the field.

Field studies by Roberts (1972), in South Wales, UK, showed that moss bags exposed to wind, but protected from rain, collected 95 per cent of the metals retained by a fully exposed bag. Also, when partially protected from wind by a polythene screen, but open to rainfall, the moss collected only 20 - 40 per cent of the metal retained by a fully exposed bag. Thus, wet deposition is believed to be a minor source of metal addition to moss bags, particularly in an area of high airborne concentration. Further, this work found that the accumulation of metals by moss bags proceeded in a linear manner for up to ten weeks after exposure, even in heavily polluted areas.

Goodman, Smith and Inskip (1975) also undertook a field study of airborne metals using moss bags in South Wales, UK. At each of nine sites, results obtained were compared with the amount of particulates collected by a total deposit gauge, a dry deposit gauge (horizontal filter pad covered by perspex but open at the sides), an air concentration gauge (air sucked through a filter) and trays of grass (*Festuca rubra*). Hairiness and wetness of the vegetation contributed to the following difference in the percentage of airborne particulates retained:

moss bags > grass > total deposit gauge > dry deposit gauge

The trays of *Festuca rubra* placed at the same height above ground as moss bags, and sampled weekly for a month, were found to accumulate approximately one half of the quantity of metals collected by moss bags per unit surface area. A good correlation was found between metal levels retained by moss bags, total and dry deposition gauges, but not with the air concentration gauge. The authors suggest that this reflects the importance of wind speed, and direction relative to the pollution source on deposition rates, rather than just air metal concentrations.

It may be concluded, therefore, that moss bags are a useful measure of dry deposition of trace metals to grassland because of the similarity in collection characteristics of grass and moss surfaces. The method is less suitable if wet deposition processes are the major source of atmospheric contamination of vegetation, although this has been shown not to be the case in fairly polluted sites in the UK.

3.5 CONCLUSION

Airborne lead is removed from the atmosphere by processes of wet and dry deposition. Dry deposition appears to be the most important process near a source of pollution, such as a motorway, whereas wet deposition becomes progressively more important with distance from the point of emission.

Few workers have determined the proportion of the lead emitted by motor vehicles that is deposited near a major road. The available evidence suggests that the greater part of this vehicular lead is transported away from the roadside environment by processes of atmospheric dispersion.

Difficulties are encountered when measuring the rate of lead deposition to vegetation because of the influences of wind speed, particle size and surface features on the efficiency of capture of particles. Moss bags are, however, useful as integrators of the aerial lead burden, and for estimating the long term (usually 2 to 6 weeks) loading rates into an ecosystem.

3.6 MOSS BAGS AS A MEASURE OF LEAD DEPOSITION AT THE M6 STUDY AREA

A. The Aims of the Study

i) To obtain a measure of the rate of lead input from the atmosphere to the pasture grass bordering the M6 motorway.

ii) To compare rates of lead deposition near the motorway with sites distant from the road, and to investigate the influence of wind direction on deposition in the study area.

iii) To investigate any possible seasonal variation in lead deposition rate, and the environmental factors influencing the scale of deposition.

B. Method

The moss bags were prepared using Sphagnum moss (*Sphagnum papillosum*) collected from the Bramble Hill region of the New Forest, Hampshire. This is a site remote from heavily used roads and industrial premises.

At the laboratory, the moss was washed in running tap water and sorted so as to discard other plant material. The resulting moss was then rinsed a number of times with deionised water, hand squeezed, and used directly for the moss bags.

Initially, the suitability of three types of moss bag were investigated. All were prepared by placing the moss in a pocket of fine nylon mesh (2 mm square holes), although the form of the bags varied in the following manner:

1) 15 g of damp moss were placed in an 8 x 8 cm square flat bag.

2) 30 g of damp moss were placed in a flat, rectangular bag with dimensions of 13 x 11 cm.

3) 15 g of damp moss were made into a ball and placed in a circular bag.

Examples of the three types of bag were suspended from exposed, peripheral branches of trees for a period of four weeks, and at a height of approximately 1.5 metres. Five sites were used within the study area at distances ranging from 10 to 1,500 metres from the motorway. This trial experiment showed that rectangular bags were most efficient at collecting lead (and zinc) with little difference between the two types. Thus, to minimise the amount of moss used, it was decided to use the square bag containing 15 g of moss. After oven drying, this yielded about 1.5 g of dry material which was sufficient for metal determination.

A monitoring programme was set up in November 1977 which involved the exposure of five moss bags at each of eight sites located in Figure 3-2. After a period of between four and five weeks, the moss was replaced by a fresh set of bags. Thus, data was obtained for the lead and zinc deposition rate during approximately monthly intervals until the study was discontinued in November 1978.

C. Lead Concentration of Moss Bags in Relation to Distance from the Motorway

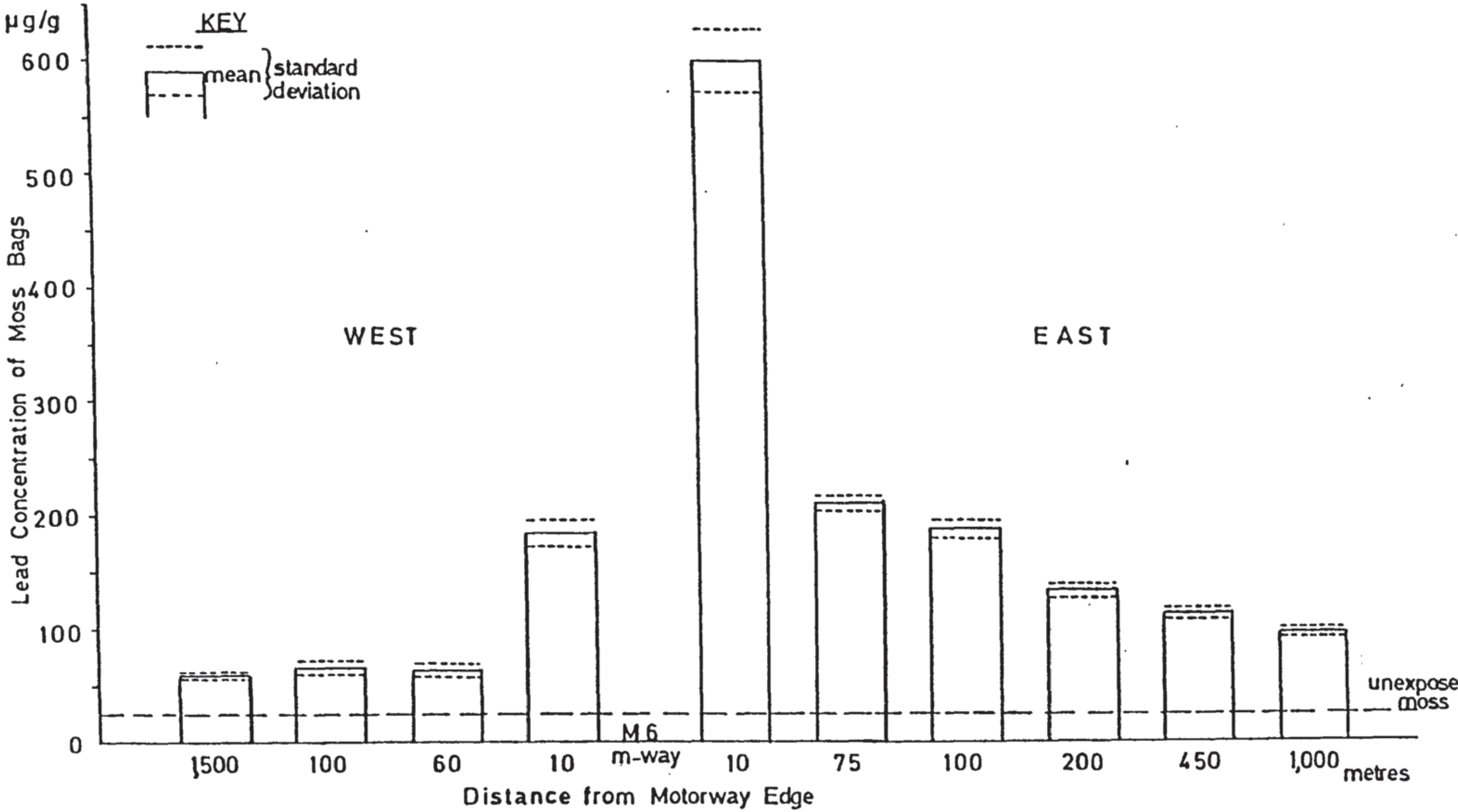
During late February and early March 1978 a detailed study of the deposition of airborne lead at varying distances from the motorway was undertaken. Figure 3-3 shows the mean lead content and standard deviation of five bags placed at each of ten sites in the study area.

FIGURE 3-2 Location of Moss Bags

Aston University

Illustration removed for copyright restrictions

FIGURE 3-3 Lead Concentration of Moss Bags Exposed During February/March 1978



It can be seen that lead deposition is greatest 10 metres east of the edge of the motorway and decreases with distance from the road. The lead content of the moss suspended 10 metres from the western edge of the motorway is almost equal to that 100 metres east of the road. This reflects the occurrence of a strong, westerly wind during this period which carried the pollutant cloud to the east of the motorway.

Statistical analysis shows that the lead content of moss bags 1,000 metres east of the M6 is significantly greater than those 1,500 metres upwind of the motorway. Therefore, during the February/March exposure period, moss bags were able to detect lead emissions from the motorway at a distance of at least 1,000 metres downwind. In contrast, there was no significant difference between the lead content of the moss bags 60 metres upwind of the motorway and those at the 1,500 metres west site.

The influence of the prevailing wind direction upon the deposition of lead each side of the motorway may be clearly demonstrated by consideration of the levels found in the moss bags placed either side of the motorway at a distance of 10 metres from the edge of the hard shoulder. Table 3-1 compares the mean lead content of the bags with the prevailing wind direction, for the period November 1977 to November 1978. Prevailing wind direction has been determined from data concerning the percentage frequency of occurrence of winds greater than 0.5 m/s at the Keele weather station (Staffordshire). Results show that the lead concentration of moss bags was highest during each month at the site which is downwind of a strong prevailing wind. This finding confirms the results relating to atmospheric lead levels referred to earlier in Chapter 2.

TABLE 3-1

Lead Levels in Moss Bags as Influenced

by Prevailing Wind Direction

Date of Bag Exposure	Lead concentration of Moss Bags 10 metres from motorway edge (μg/g)		Direction toward which prevailing wind occurs
	West of M6	East of M6	
17.11.77	226 (40)	214 (29)	{E}
15.12.77	372 (44)	773 *	{E}
16. 1.78	658 (43)	680 (78)	{E}
23. 2.78	184.5 (24)	601 (60)	E
31. 3.78	514 (52)	186 (7)	W
5. 5.78	144 (17)	187 (34)	{W}
1. 6.78	183 (6)	641 (102)	E
11. 7.78	134 (3)	601 (11)	E
8. 8.78	94 (14)	507 (46)	E
6. 9.78	54.5 (5)	513 (46)	E
5.10.78	140 (16)	305 (11)	E

( ) = standard deviation of five bags.

* = < 3 bags analysed.

E/W = prevailing wind blows towards east/west of motorway and is dominant over other directions by > 15% of the total time.

{E}/{W} = prevailing wind direction, but dominant by only < 15% of the time.

Table 3-2 compares the mean lead content and standard deviation of the moss bags placed at each of the main survey points during the twelve month sampling period. Highest lead levels were found in moss suspended near the motorway in all months of the year. In general, the lead concentration of moss bags placed 10 metres east and 10 metres west were respectively 2 to 10 and 3 to 15 times greater than levels 1,500 metres west of the motorway.

It would appear that the 1,500 metre west site was not significantly influenced by vehicular emissions from the motorway as moss bags at this distance had a consistently lower lead content than moss suspended at other sites. This was the case even when the prevailing wind was a strong easterly during April 1978. In addition, a study during March 1978 found no significant difference between the lead concentration of moss bags placed 1,500 and 2,500 metres west of the motorway. The evidence, therefore, suggests that this distance may be regarded as a background site at which the levels of lead contamination reflect regional sources.

D. Lead Deposition Rates and Seasonal Variation

The lead deposition rate to moss bags was calculated by subtracting the mean lead concentration of the moss before exposure from the mean lead content of the bags after four weeks in the field. This value was then divided by the number of days for which the moss bag was exposed to give lead deposition rate as µg of lead deposited/gram of dry moss/day. This then allows the comparison of data concerning different exposure periods.

Figure 3-4 shows the lead deposition rate at each of seven monitoring sites. All sites show a higher rate of lead deposition in

TABLE 3-2

Mean Lead Content of Moss Bags and Standard Deviation Before and After

Exposure Near M6 Cheshire 1977/78 (µg/g dry wt)

Date of Exposure 1977/78	Distance West of M6			Distance East of M6					Unexposed moss
	1,500 m	100 m	10 m	10 m	100 m	200 m	450 m	1,000 m	
17.11.77	23 (3.9)	50 (12.9)	226 (40.2)	214 (28.7)	-	48 (8.8)	46 (7.4)	-	10.2 (1.8)
15.12.77	75 *	91 (10.5)	372 (44.2)	773 *	-	168 (17.4)	136 (27.6)	-	22 (3.3)
16. 1.78	118 (5.8)	171 (4.4)	658 (43)	680 (78)	-	178 (12.4)	151 (8.8)	-	24.5 (1.5)
23. 2.78	62 (8.2)	66 (8)	184.4 (24)	601 (60)	187 (15)	112 (12.7)	112.8 (11.5)	95 (8.6)	27.6 (1.5)
31. 3.78	58 (6.1)	122 (7.5)	514 (51.6)	186 (6.9)	81 (12.4)	57 (4.6)	78 (5.8)	62 (4.6)	35 (1.0)

* < 3 bags exposed and recovered

( ) = standard deviation of five bags.

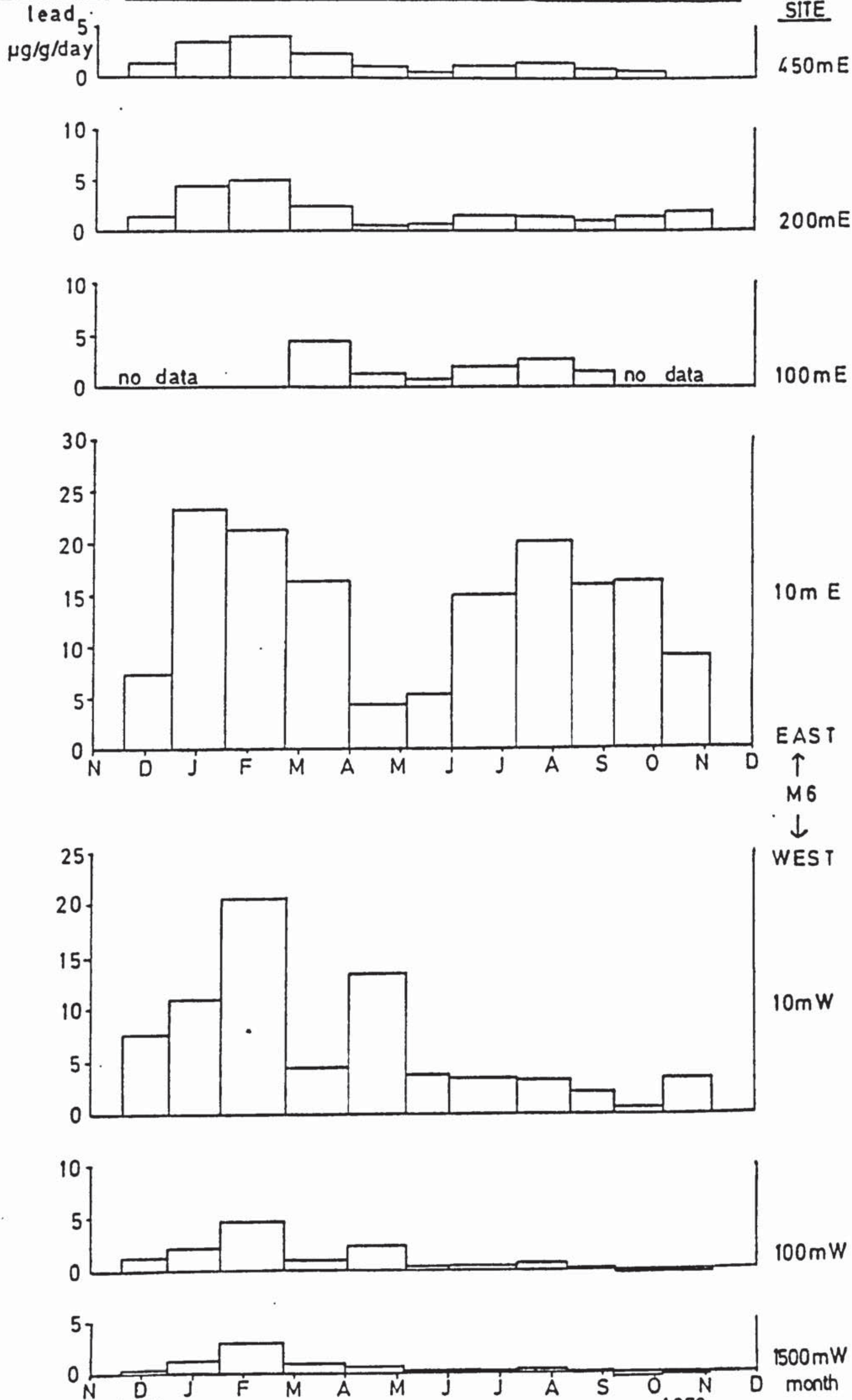
TABLE 3-2 (Continued)

Date of Exposure 1977/78	Distance West of M6			Distance East of M6					Unexposed moss
	1,500 m	100 m	10 m	10 m	100 m	200 m	450 m	1,000 m	
5. 5.78	45.6 (2.4)	52 (3.5)	144 (17.5)	187 (34)	65.5 (4.2)	59.6 (10.9)	54.4 (3.2)	-	39.8 (1.5)
1. 6.78	46.5 (2.2)	59.4 (2.3)	183 (6.5)	641 (102)	124 (21.8)	111.8 (10)	89 (4.6)	64 (3.5)	39.8 (1.5)
11. 7.78	45.9 (1.3)	63.5 (4.4)	133.7 (2.9)	601 (11)	117 *	83.3 (23)	86.2 (3.0)	63.3 (3.4)	39.8 (1.5)
8. 8.78	42.1 (2.7)	46.8 (1.6)	94 (13.9)	507 (46)	89 (9.2)	69.6 (2.9)	66.5 (3.6)	58.4 (3.9)	39.8 (1.5)
6. 9.78	29.5 (3.0)	34.5 (6.7)	54.4 (5.2)	513 (46)	-	85.3 (5.9)	65.3 (4.4)	45.7 (1.6)	39.8 (1.5)
5.10.78	33.4 (1.2)	34.9 (2.1)	140.3 (15.9)	304.7 (11.2)	-	97.8 *	40 *	52.6 *	39.8 (1.5)

* < 3 bags exposed and recovered

( ) = standard deviation of five bags.

FIGURE 3-4 Deposition Rate of Lead to Moss Bags 1977/8 (µg/g/day)



winter months, with peak values during January and February 1978. Lowest deposition rates occurred during the spring and summer months. A negative value appears on the graph for two sites during late summer. This apparent loss of lead from the bags during exposure was only statistically significant at the 1,500 metres west site in September and October 1978 and is discussed further later in this Chapter. Other notable features of the graph are:

i) The secondary peak in lead deposition rate during summer months at the 10 metres east site. This corresponds to the period of peak traffic flow described in Chapter 2. Its effect upon deposition is just detectable at the other three sites leeward of the motorway.

ii) The relatively high deposition rate during April 1978 at the 10 metre west site which corresponds with the occurrence of a strong easterly wind. The effect of this wind upon deposition is also detectable 100 metres west of the motorway.

iii) The higher lead deposition rate during December/January than the January/February period at the 10 metres east site, when peak deposition occurs in this latter period at all other sites. The difference between these two periods is not, however, of statistical significance. Only three moss bags were recovered at this site during the December/January period and these show considerable variability in lead content.

In conclusion, therefore, a considerable seasonal variation occurred in the rate of lead deposition to moss bags at all survey points during 1977/78. Highest rates of deposition were found during winter months and minimum in summer. This finding is in agreement with Day et al. (1977) who report that moss bags suspended near a motorway in Bristol (UK) had peak lead concentrations during the winter months.

Although the moss bag survey was continuous for a period of only one year, some smaller scale investigations were carried out in April, August and November of 1979. Lead and zinc deposition rates were similar to those found during the corresponding months of 1977/78 (Table 3-3). Thus, the results suggest that the seasonal change in deposition rate of atmospheric lead and zinc to moss bags is a phenomenon which occurs repeatedly every year.

E. A Comparison of Lead and Zinc Deposition Rates to Moss Bags

The deposition rate of airborne zinc to moss bags at each survey point is shown graphically in Figure 3-5 for the period November 1977 to November 1978. The results indicate that the motorway is a source of zinc, but only the moss bags situated 10 metres from the M6 were contaminated significantly. All other sites were found to have a similar rate of zinc deposition which was less than the values measured adjacent to the motorway. These results are, therefore, in agreement with the findings of airborne zinc measurements, taken 200 metres west of the M6, which were found to be independent of wind direction relative to the motorway.

A seasonal variation in zinc deposition occurred which is very similar to that found for lead. Peak zinc deposition rates occurred in January/February at all sites and minimum during summer months. As the zinc must originate dominantly from regional sources, it further supports the findings for lead at the 1,500 metres west site, which suggests that deposition from ambient atmospheres is greatest during the winter months. The findings would suggest that similar factors are determining the lead and zinc deposition rate to moss bags. These

TABLE 3-3

A Comparison of Lead and Zinc Deposition

Rates to Moss Bags in 1978 and 1979

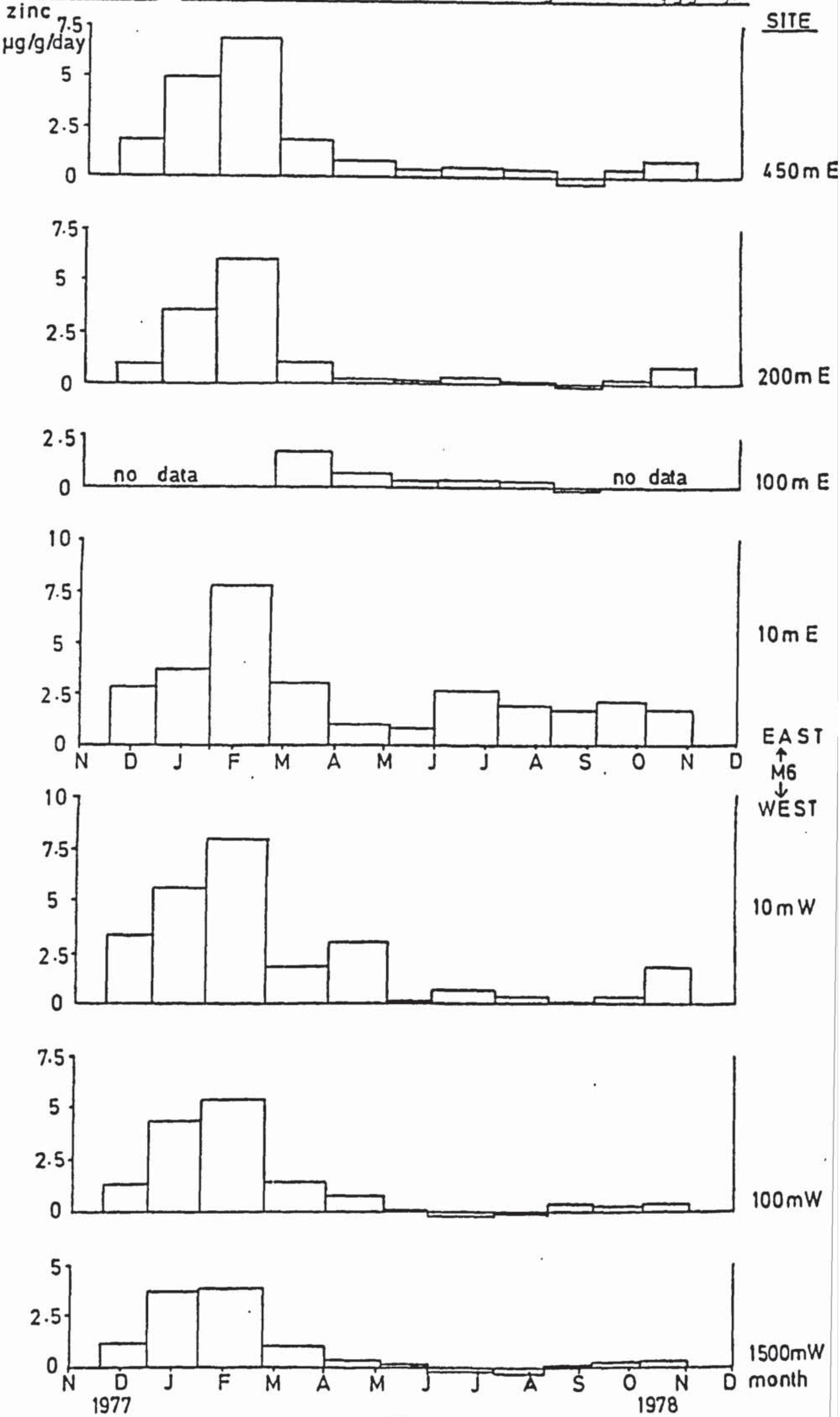
1. Lead

Month and Year	Lead Deposition Rate µg/g/day			
	1,500 m west	10 m Downwind	10 m Upwind	450 m east
April 1978	0.66	13.7	4.5	2.4
April 1979	0.56	12.5	5.3	1.6
August 1978	-	-	1.9	-
August 1979	-	-	1.2	-
November 1977	0.5	7.7	7.2	-
November 1979	1.0	18.0	2.9	-

2. Zinc

Month and Year	Zinc Deposition Rate µg/g/day			
	1,500 m west	10 m Downwind	10 m Upwind	450 m east
April 1978	0.43	2.97	1.0	0.69
April 1979	0.56	2.4	1.1	0.77
August 1978	-	-	0.01	-
August 1979	-	-	0.19	-
November 1977	1.2	2.9	3.4	-
November 1979	0.45	1.7	0.9	-

FIGURE 3-5 Deposition Rate of Zinc to Moss Bags 1977/78 (µg/g/day)



must either be source factors, which determine the level of airborne lead and zinc, or environmental influences which result in a seasonal variation in the efficiency of collection and retention of airborne metals by the moss bag. These factors are now discussed.

3.7 FACTORS INFLUENCING THE RATE OF LEAD DEPOSITION TO MOSS BAGS

The calculation of the lead and zinc deposition rate to moss bags has been based upon the metal concentration of the moss after an exposure period of four weeks. This metal content will be the resultant of the metal input to the bag by deposition, minus possible losses from the bag by wind re-entrainment and leaching by rainfall.

A. Lead Input to the Moss Bag

(i) Source Strength

The amount of lead emitted from motor vehicles travelling on the motorway will be proportional to the traffic flow. This shows a considerable seasonal variation as discussed in Chapter 2. Peak traffic flow occurs during the summer and this is in contrast to the lead deposition rate to moss bags, which is greatest during winter. Therefore, although the influence of an increase in source strength during mid-summer is demonstrated by a rise in the lead content of moss bags, traffic flow rate is not a major factor determining the variation in lead deposition rate.

To demonstrate the importance of factors other than variation in traffic flow, the deposition rate to moss bags 10 metres either side of the motorway has been added together for each of the eleven exposure periods. This gives a measure of lead deposition near the motorway which is independent of wind direction. The summed deposition was then normalised for the mean hourly flow of petrol-engined motor vehicles, expressed as hundreds of vehicles per hour. Therefore, a value is obtained which will be termed the traffic normalised deposition efficiency (TNDE) with the units of μg of lead/g of moss/day/100 vehicles/hour.

Figure 3-6 shows the TNDE for moss bags at a distance of 10 metres from the M6 motorway during November 1977 to November 1978. Results show that this index of deposition was greatest during winter and least during the summer months. Lead deposition to moss bags in February/March was five times more efficient than in May with respect to the source strength. Therefore, although traffic factors accounted for some of the variations in lead deposition rate found during 1977/78, it is presumably the effects of meteorological factors which produce the seasonal variation in TNDE.

(ii) <u>Meteorological Factors</u>

Meteorological factors will influence both the processes of wet and dry deposition to the surfaces of moss bags.

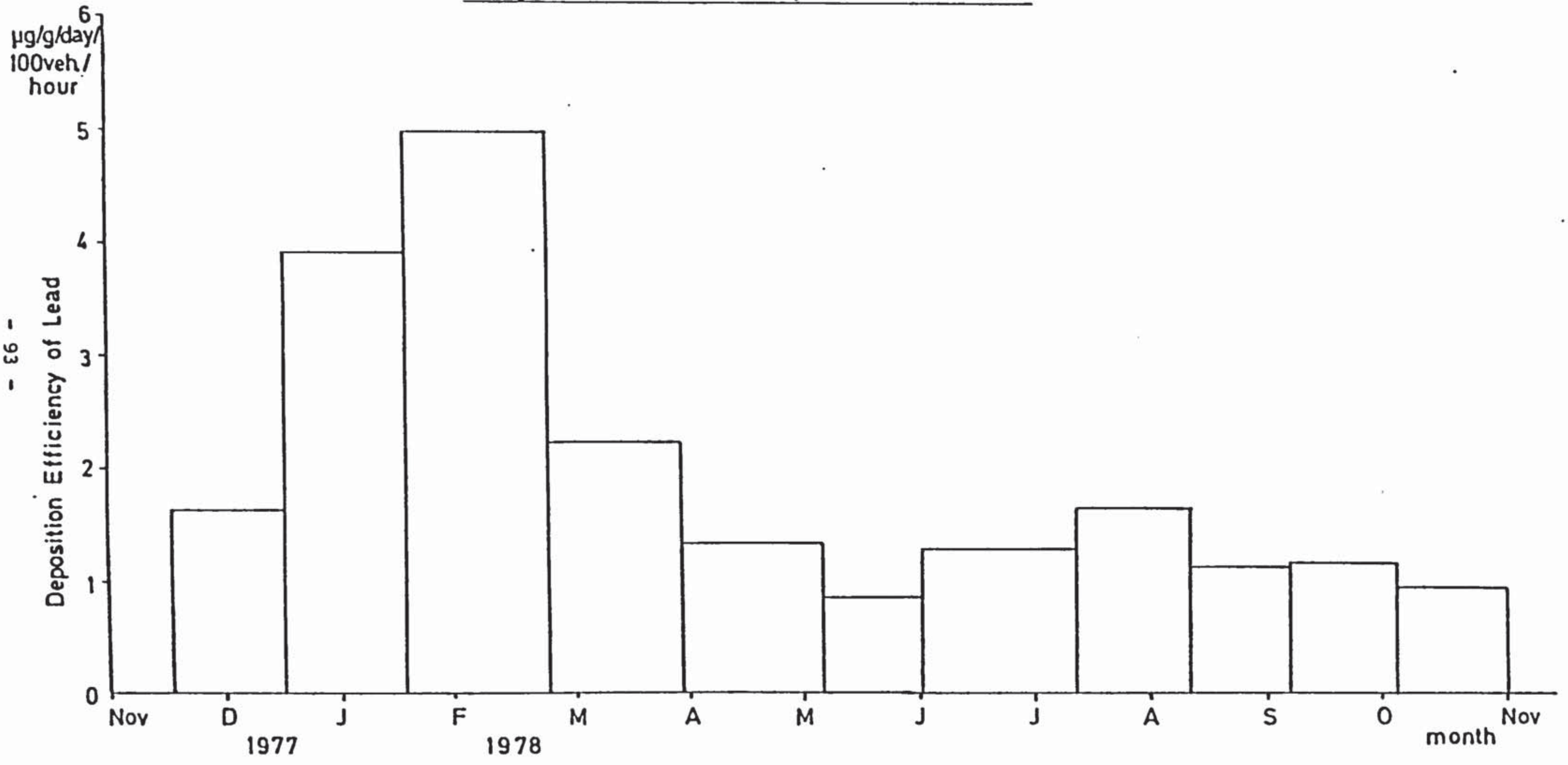
(a) <u>Wet Deposition</u>

The lead input to the moss bag from precipitation will depend on both the lead concentration in the rain and the amount of rainfall which occurred during the exposure period.

1) Lead Concentration;

An investigation of the lead concentration of the rainfall was undertaken during March until September 1978 at the 1,500 metres west site. Rain was collected by the use of a glass funnel and a plastic collecting vessel. A few drops of concentrated nitric acid were placed

FIGURE 3-6 Traffic Normalised Deposition Efficiency of Lead to Moss Bags 10 metres from the Edge of the M6 Motorway in Cheshire



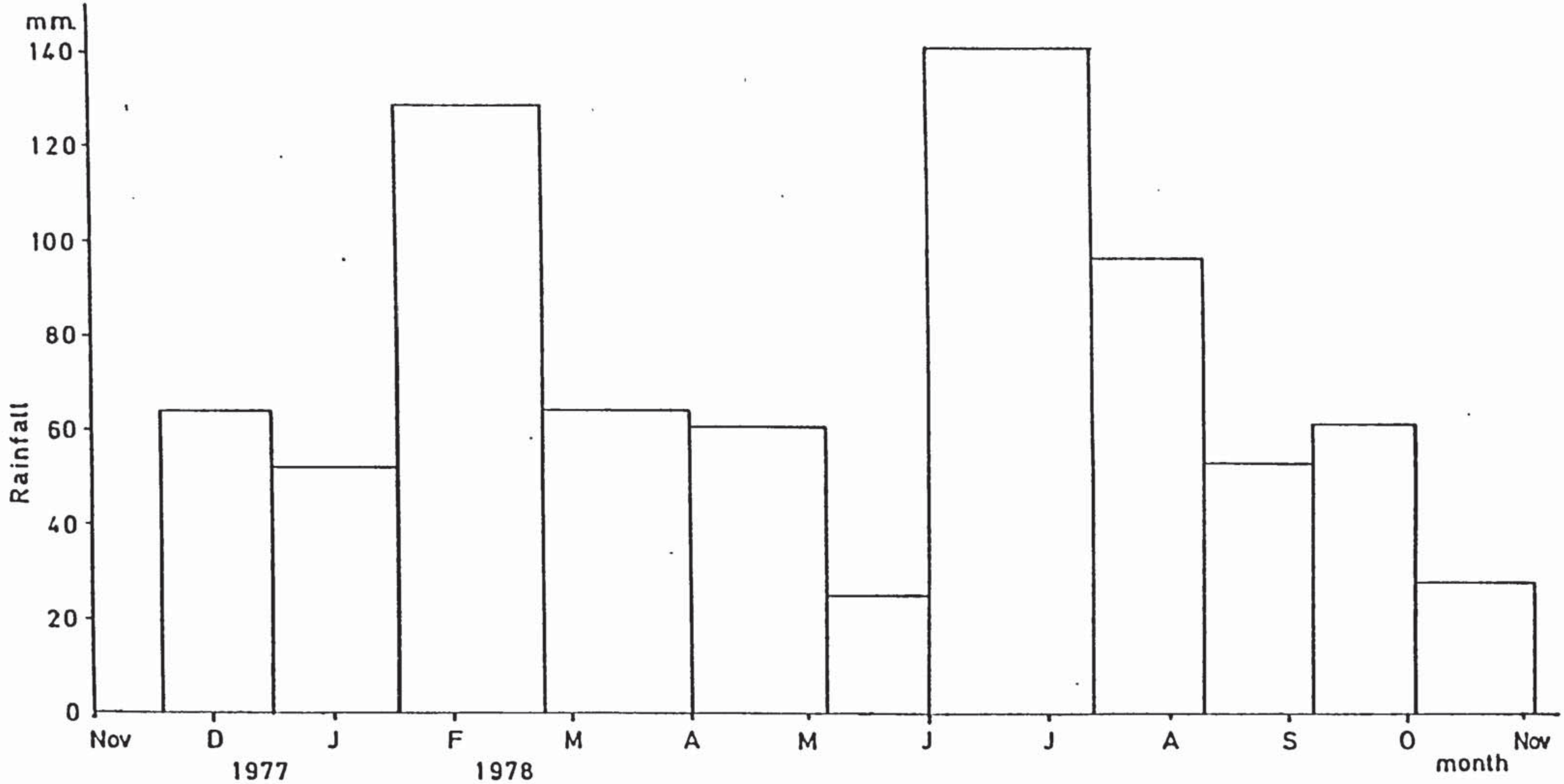
in the vessel prior to exposure to minimise metal loss by adsorption to surfaces. At the end of the moss bag exposure period, the rainwater was filtered (Whatman No. 1 Paper) and the filtrate analysed for lead content using flameless atomic absorption spectrophotometry. The lead concentration of the rainwater was found to range from < 10 to 15 μg/l. These measurements are not considered very satisfactory because dry deposition of soluble lead could add to the actual concentration of the rainfall. In addition, evaporation and some algal growth may have influenced the concentration before laboratory analysis. However, the data are quite comparable to that obtained by Harrison et al. (1975) who report lead levels ranging from 15 - 40 μg in weekly rainfall samples collected through a filter paper at a rural site in Cambridgeshire. Cawse (1974) found a two-year average lead concentration of 41 μg/l in rainfall to which soluble dry deposits may have contributed. This was at seven non-urban sites in the UK.

There is little published data on which to establish whether a seasonal variation occurs in the lead concentration of rainfall in the UK. Cawse (1974) reports that levels of wet deposition were generally higher in winter, but this was not significant at all sites. Samples taken at the Sandbach study site did not depict any trend in the lead concentration of rainfall during March to September 1978.

2) Rainfall Amount;

Figure 3-7 shows the total amount of rainfall, as measured at the Keele weather station, for the moss bag exposure periods. A double peak in the rainfall distribution occurs with maximum levels in February and June, and minimum in May and October. Therefore, the seasonal variation in the amount of rainfall does not correlate with the variation in lead deposition rate to moss bags. Even if the

FIGURE 3-7 Total Rainfall at Keele During Moss Bag Exposure Periods in 1977/8



February rainfall contained 40 μg/l of lead, a simple calculation suggests that wet deposition must be of secondary importance to dry deposition as a source of lead to moss bags.

If it is assumed that the suspended moss bag presents an area of catchment for rainfall equal to the area of one surface of the bag, and that 100 per cent of the lead in the rain is absorbed by the moss and retained, only 20 μg/g of lead is accounted for by rainfall during January/February. This value may be compared with the lead concentration of the moss bags, after exposure in this period, in Table 3-2, which are at least 100 μg/g at all sites. Therefore, rainfall would appear to contribute, at most, 20 per cent of the total lead content of the moss bags. It is probable that this calculation greatly overestimates the amount of rainfall that is incident on a freely suspended flat bag of moss and the retention capacity of the moss. This calculation is in agreement with the findings of Roberts (1972) who found that dry deposition accounts for perhaps 90 per cent of the lead deposition to moss bags in relatively polluted sites.

A further possible influence of rainfall upon deposition to moss bags comes from the resuspension of lead that has settled on the road surface by incorporation into spray water generated by the passage of vehicles. Little and Wiffen (1977) suggest that road spray may act as an important source of lead to soil and vegetation, but only within five metres of either side of a motorway. There is no published data concerning any possible seasonal variation in lead deposition rates from road spray and the importance of this source might be expected to be a complex function of traffic flow, rainfall amount, frequency and intensity. It is, however, unlikely that road spray was a significant source of lead to moss bags at the M6 study area because they were placed no nearer than 10 metres from the motorway. In addition, if road spray was a major source of lead to moss bags at 10 metres, it might be expected that the lead content of the moss would be similar after the June and February exposure periods during which similar volumes of rainfall occurred.

Further evidence suggesting that contaminants in the road spray are not a major source of lead to moss bags comes from the findings of Pope et al. (1979). These workers collected spray adjacent to the M1 motorway (Hertfordshire) and found that zinc concentrations exceeded those of lead, although dry deposition may have had some influence on their data. Yet at the M6 study site, lead deposition rate to moss bags 10 metres from the motorway are by far in excess of those for zinc.

(b) <u>Dry Deposition</u>

The rate of deposition of lead-containing particulates to the surface of a moss bag will depend upon a number of factors. The influence of meteorological conditions, moss surface and particle characteristics will now be discussed in view of the seasonal variation in lead deposition to moss bags found at the M6 study site.

1) Atmospheric Concentration of Lead;

The use of an atmospheric model of pollution dispersion, together with measurements of airborne lead at the study site, have been discussed in Chapter 2. It was concluded that the higher summer traffic flow did not result in a correspondingly greater airborne lead concentration when compared with winter months. This could, at least in part, be explained by the greater stability of the atmosphere during winter.

It would appear, therefore, that airborne lead levels do not show a large seasonal variation within the M6 study area. Even if average airborne lead levels were greater over the winter period, this would seem unlikely to explain the magnitude of the seasonal variation in deposition rates to moss bags found during 1977/78 in the vicinity of the motorway.

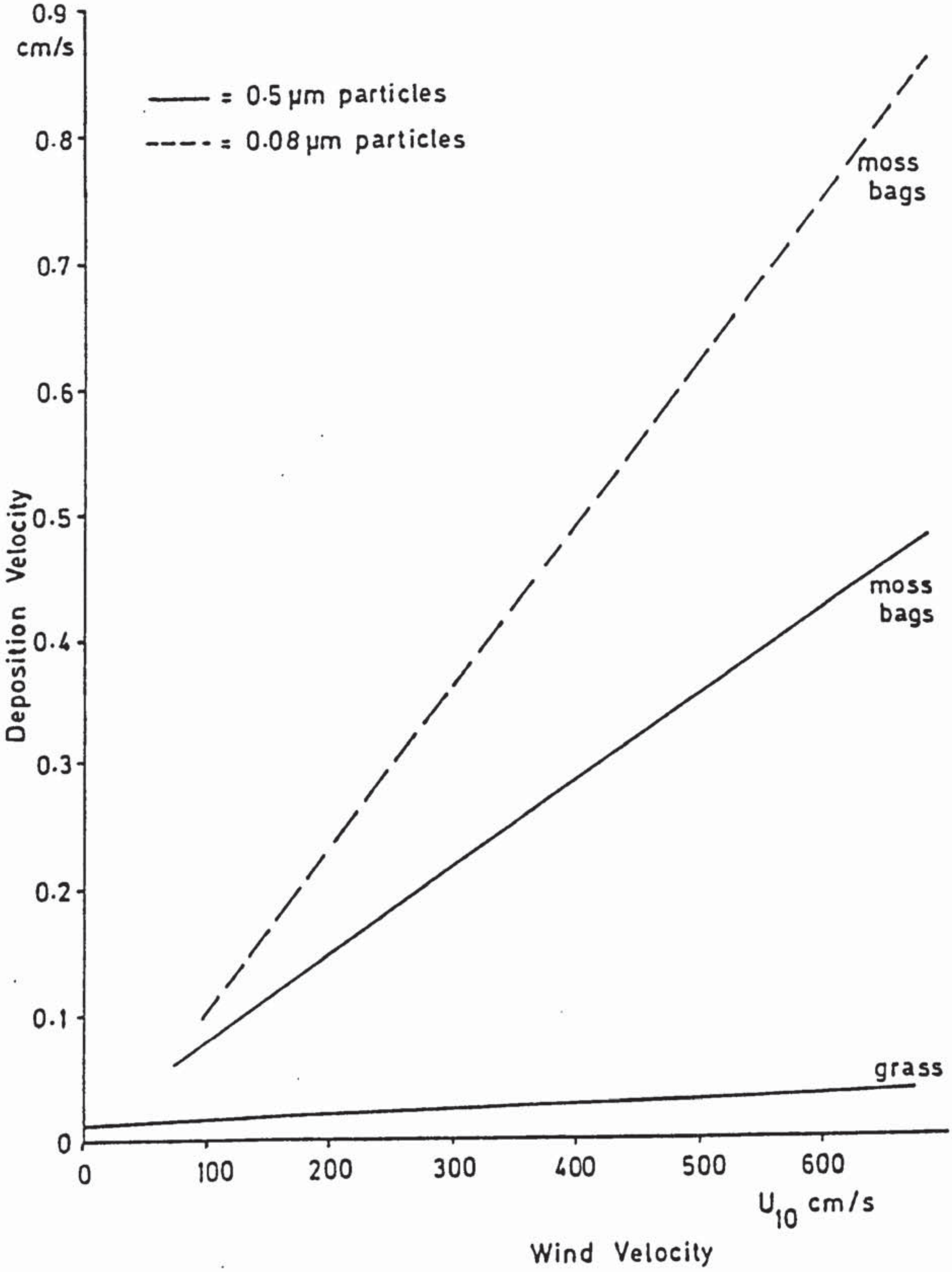
Regional sources, such as the combustion of fossil fuel for the generation of heat and power, combined with the more stable atmospheric conditions were suggested by Cawse (1974) to be responsible for a 1.7 times higher airborne lead and zinc concentration during winter than summer months at seven non-urban sites in the UK. A seasonal variation of this order could only partially explain the monthly differences in deposition to moss bags at the background site (1,500 metres west).

2) Wind Speed;

Wind speed has been demonstrated to have an important influence upon the dry deposition velocity of particulates of a size range characteristic of petrol engine exhaust emissions (Little and Wiffen, 1977; Clough, 1975). For a given airborne concentration of lead particulates, the deposition to moss bags will be greater for a higher wind speed.

Figure 3-8 shows the effect of wind speed upon the deposition velocity of 0.5 and 0.8 μm diameter particles to moss bags (and grass) reported by Clough (1975). Particle concentration in the air was measured by use of air filters which were operated isokinetically, and the moss bags were suspended at a height of 10 cm above the floor of the wind tunnel. The graph shows that deposition velocity (Vg) increased linearly with wind speed and that a doubling of wind

FIGURE 3-8 The Effect of Wind Speed on Deposition of 0.5 and 0.8 µm Particles on Grass and Moss Bags (Clough 1975)



velocity produced an approximately two-fold increase in Vg for both sizes of particle.

Figure 3-9 shows the mean daily wind speed for each month during the period November 1977 to November 1978 at the Keele weather station. Highest wind speeds occurred during the winter months with minimum in May. However, wind speed does not correlate with the measured variation in lead deposition rates which peaked during January/February and declined during March and April 1978. In addition, the mean daily wind speed varied by less than a factor of two over the yearly period, which is considerably smaller than differences in lead deposition rate to moss bags during winter and summer. The wind characteristics during 1977/78 were further investigated by the calculation of the wind run (i.e., speed x hours) for each of the moss bag exposure periods (Figure 3-10). This was also found not to correlate closely with measured deposition rates, as the peak wind run occurred during March.

It appears, therefore, that the higher wind speed in winter months may be a contributory factor to the seasonal variation in lead deposition rate to moss bags. However, it does not fully explain the measured changes in lead and zinc deposition to moss bags in the study area.

3) Particle Size;

Meteorological conditions may influence the rate of coagulation of particulates in the atmosphere (Chamberlain et al., 1978). These workers show that there is a tendency for a greater aggregation of lead aerosols during winter weather conditions, as a result of the stability of the atmosphere, and possibly due to hygroscopic growth

FIGURE 3-9 Mean Daily Wind Speed at Keele for Each Month during November 1977 to November 1978

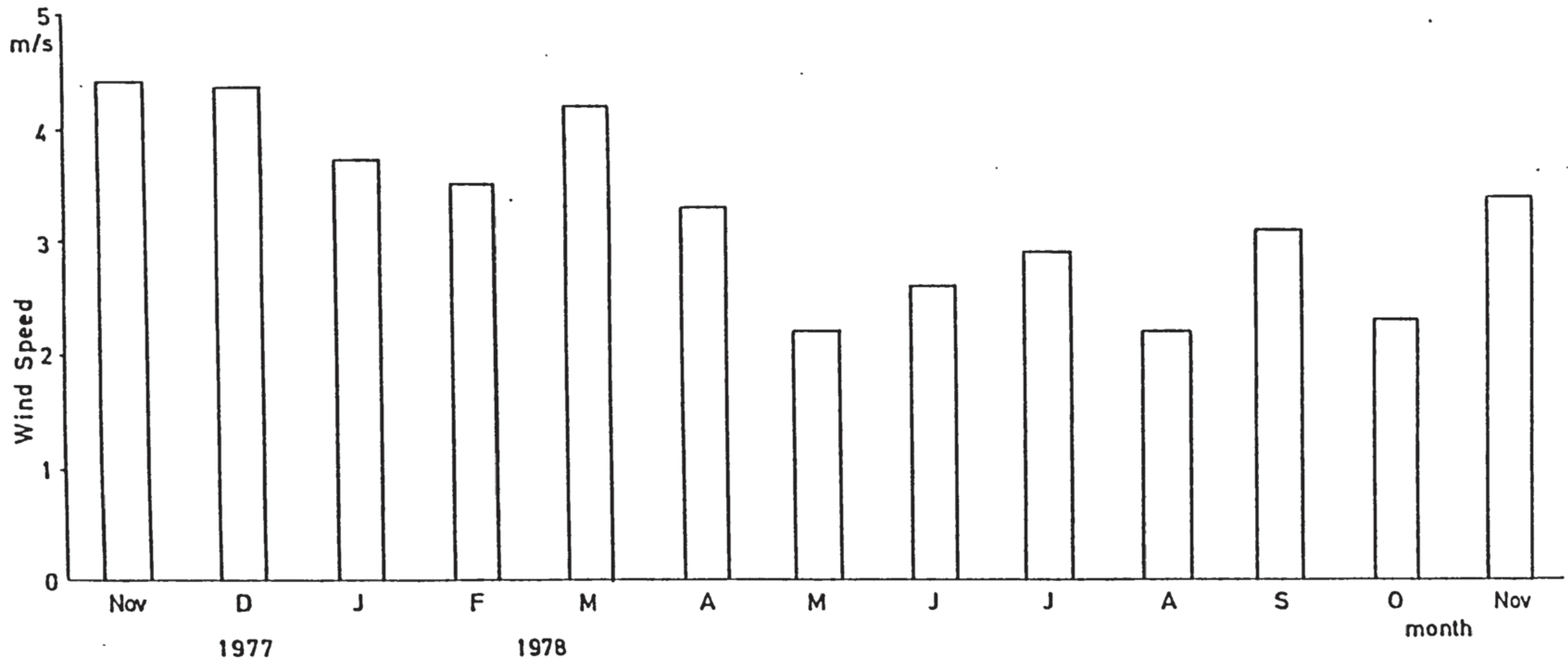
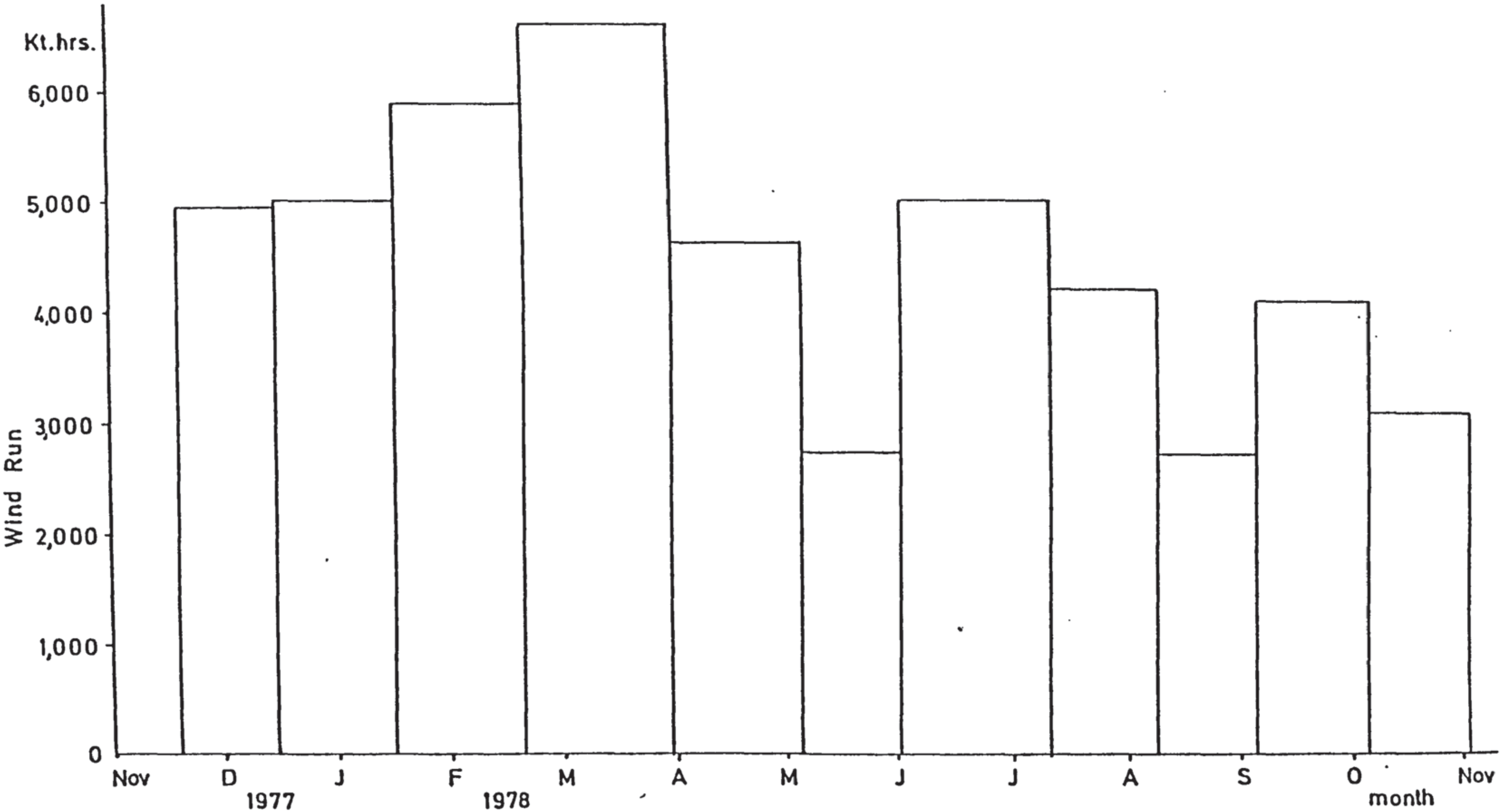


FIGURE 3-10 Wind Run (All Directions) at Keele during Moss Bag Exposure Periods



of particles. However, as deposition velocity is minimal for 0.5 μm particles at all wind speeds (Clough, 1975), it would seem unlikely that sufficient aggregation of lead aerosols from motor vehicles could occur to influence the deposition velocity to moss bags significantly.

4) Moss Surface;

Little and Wiffen (1977) found that deposition of lead aerosols was greater to dry than wet moss, due to the more open structure of the dry moss which had a greater effective surface area for deposition. This factor would appear not to be significant when compared with deposition rates measured at the M6 study site, presuming that the amount of rainfall is an indicator of the wetness of the bag.

B. Lead Loss from the Moss Bag

It is conceivable that lead deposited on a moss bag is more readily lost by rain leaching and wind re-entrainment during summer than winter months. Clough (1975) has, however, demonstrated that particulate losses by wind re-entrainment and from spraying moss bags with water are insignificant. To investigate the situation in the field at the M6 study site, moss bags were placed near the motorway to collect a high concentration of lead and then placed at the background site for four weeks to assess if any lead was lost from the moss. It was felt that acidic rainfall could be a more efficient leaching agent than the water spray used in laboratory studies, and thus a field study was thought to be most appropriate.

Ten moss bags were suspended 10 metres west of the M6 motorway for a period of four weeks during July 1979. Five of the bags were then transferred to the 1,500 metres west site and the remainder returned to the laboratory for analysis. After a period of four weeks, the moss bags were collected from the background site and their metal content compared with those exposed only at the motorway. No significant difference was found between the lead and zinc content of the two sets of moss bags (Table 3-4). Therefore, despite a rainfall of 30 mm, little of the lead and zinc collected adjacent to the motorway was lost during the subsequent four week exposure period at the background site.

This field experiment was repeated in greater detail during the winter of 1979/80. Twenty moss bags were exposed at a distance of 10 metres from the motorway during November 1979. Ten bags were then transferred to the 1,500 metres west site for a period of four weeks until early January 1980. During this time of exposure at the background site, 160 mm of rainfall was recorded at the Keele weather station. However, Table 3-4 shows that similar findings occurred in that no significant changes were found in the lead and zinc content of the moss bags.

It may be concluded, therefore, that very little of the lead and zinc deposited on moss bags at the roadside is removed by rainfall leaching or wind re-entrainment during summer and winter months. There was some evidence, however, for a decrease in lead concentration of the moss when exposed at the background site in September/October 1978. The amount of lead involved was small and appears to reflect a loss of the lead present in the moss before exposure rather than the leaching of newly deposited material.

It has not been possible to fully identify the mechanisms which are responsible for the higher lead and zinc deposition rates during

TABLE 3-4

An Investigation of the Loss of Lead and Zinc from Moss Bags in the Field

Date and Site of Moss Bag Exposure	Lead concentration of Moss Bags µg/g		Zinc concentration of Moss Bags µg/g	
	After exposure near motorway	After a further 4 weeks at 1,500 m west site	After exposure near motorway	After a further 4 weeks at 1,500 m west site
August 1979 10 m west	67 (14)	65.5 (10)	47.5 (7)	48 (4)
November 1979 10 m east	507 (64)*	568 (24)*	92 (4)*	87.5 (8)*

( ) = standard deviation of five bags.

( )* = standard deviation of ten bags.

winter than summer months. Dry deposition appears to be the most important process of deposition to the moss bag, and wind speed and airborne lead concentration result in this being a more efficient mechanism during winter months. However, variations in the rate of lead and zinc deposition do not correlate closely with the available data concerning these two factors.

It is conceivable that the high winter deposition rates are the result of air pollution episodes which, as suggested in Chapter 2, may account for the peak air lead concentrations measured at the 200 metres west sampling station in February 1979. Therefore, further research is recommended to determine the daily or weekly variability in the deposition rate of lead to vegetation during a period of moss bag exposure.

3.8 CONCLUSION

Lead deposition rate, as measured by moss bags, decreases with distance from the M6 motorway and is least upwind of the road. A pronounced seasonal variation in the amount of lead deposition occurs, with peak values in winter and minimum during summer months. This variation is not the result of changes in traffic flow, but is probably due to the influence of meteorological factors upon the mechanisms of dry deposition. Loss of lead from the bags during an exposure period is not a significant factor.

The seasonal variation in the rate of lead deposition to the moss bag surface, both near to and distant from the motorway, will have important implications for the level of contamination of pasture grass in the study area. This is considered in detail in Chapter 5.

CHAPTER 4

LEAD CONTAMINATION OF ROADSIDE SOIL AND VEGETATION

4.1 INTRODUCTION

Lead particulates are released by motor vehicles into the atmosphere and are subject to processes of removal from the air to the ground surface. These removal mechanisms have been discussed previously in connection with the measurement of the rate of lead deposition to moss bags. The processes of wet and dry deposition will also contaminate soil and vegetation in the vicinity of a motorway, and could conceivably create a potential health hazard for grazing animals due to the toxic nature of lead compounds.

This Chapter reports measurements of the lead concentration of grass and soil at the M6 study site. The results will then be compared with the findings from other sites and the data reported in the scientific literature. This information is then used in the final Chapters to assess the significance of roadside lead, both for the health of farm livestock and as a contaminant of animal products which are used for food by man.

4.2 THE LEAD CONTENT OF SOIL AND PASTURE GRASS AT THE STUDY SITE IN CHESHIRE

A. Sample Collection

A soil and grass sampling programme was initiated in October 1977 with the following aims:

i) To determine the level of lead contamination of the pasture grass and soil in fields bordering the motorway.

ii) To assess the area of pasture that is clearly influenced by lead emissions from the motorway source.

iii) To identify any seasonal changes in the lead content of soil and vegetation.

Sampling was concentrated along a transect perpendicular to the motorway and extending to the full width of fields A and E shown in Figure 1-3. In addition, grassland sites C1, C2 and C3 were established for random sampling within a 10 x 10 metre area which was situated beneath the corresponding moss bag survey positions. Occasional grass and soil sampling was undertaken in other fields on the dairy farm as a comparison with the transect field. All sites were permanent or semi-permanent perennial ryegrass (Lolium perenne) pasture and were grazed at variable intervals by farm livestock.

Grass samples were obtained by placing a one-metre square quadrat at a given point along the transect. The above-ground (> 1 cm) portion of the plant was then removed by cutting with stainless steel hand shears and placed in a polythene bag for return to the laboratory. Care was taken not to include any soil material, and the occasional weed species was discarded. This quadrat sampling procedure was carried out at five or ten metre intervals near the motorway and with less frequency toward the more distant edges of the fields. At sites C1 - 3, five one-metre square quadrates were placed randomly within the sampling area and the grass harvested in the manner described. The sampling procedure was repeated every month until the summer of 1979, with a period of more frequent investigation in the first four months of that same year.

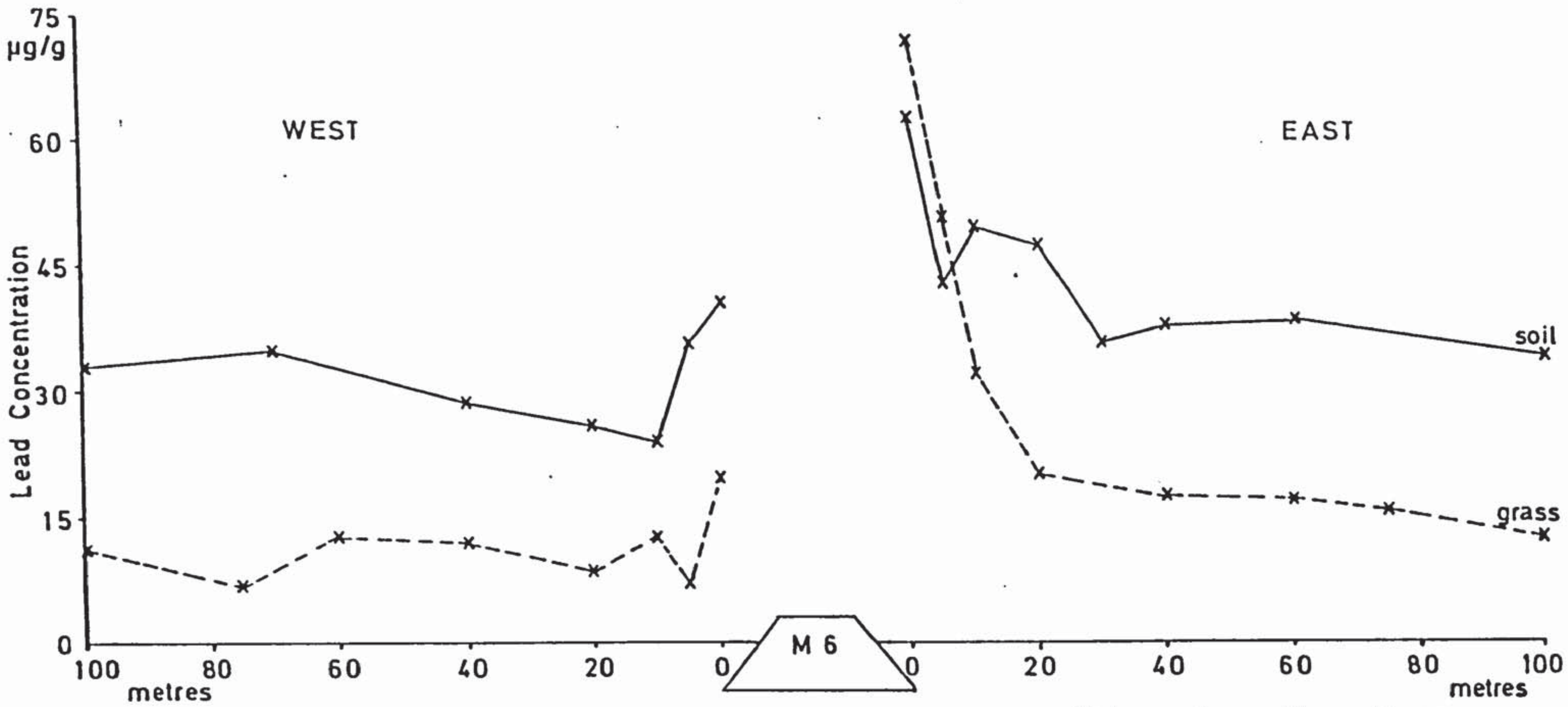
Soil sampling was carried out at the same time as the collection of the pasture grass. At the centre of each one-metre square quadrat a stainless steel trowel was used to take the top 5 cm of soil, and this was placed in a labelled polythene bag for return to the laboratory. Sampling of soil was undertaken for the period October 1977 to November 1978. All soil and grass samples were prepared and analysed in the manner previously described in Chapter 1.

B. Lead Concentration of Soil and Grass with Distance from the Motorway

Figure 4-1 shows that peak levels of lead in the top soil and vegetation occurred near the M6 motorway in November 1977. At this time, the soil had a higher lead concentration than the grass at all sample quadrats except immediately east of the motorway. In agreement with other workers, it was found that lead levels decline rapidly with distance from the motorway and little change in concentration occurs beyond a distance of about 50 metres from the road edge. A slight elevation in pasture lead content is detectable at a distance of 100 metres east of the motorway, but at 200 metres levels are not significantly different from the background site. Similar findings were obtained by the investigation of other pastures on the dairy farm.

The lead concentration of soil and pasture grass is greater to the east than to the west of the motorway. This is a result of the influence of the prevailing wind direction upon the deposition of atmospheric lead, a finding in agreement with that of the moss bag survey described in Chapter 3. Clearly, wind direction has an important influence upon the level of lead contamination in pasture adjacent to a motorway. Therefore, it may be concluded that, in general, the effect of exhaust emissions from a major road on lead content of soils and plants is not detectable beyond a distance of 50 to 100 metres, and that soil has an elevated lead concentration over a shorter distance than vegetation.

FIGURE 4-1 The Lead Concentration of Grass and Top Soil Near the M6 Motorway in November 1977



4.3 THE RELATIONSHIP BETWEEN TRAFFIC FLOW AND THE LEVEL OF LEAD CONTAMINATION

A. Soil

Table 4-1 compares the lead concentration of the top 5 cm of soil found in this study with the data of other workers who have investigated lead contamination near roads in the UK. The highest lead levels occur near the roadway and values adjacent to the more major routes exceed the 2 - 200 µg/g range given by Nriagu (1978) as normal for an uncontaminated soil. Although comparison between the data of different workers requires caution, due to variation in sample preparation and chemical analysis techniques, sample collection and site characteristics, the results in Table 4-1 do show a broad correlation between soil lead content and traffic density. Similar findings of a relationship between traffic flow and the lead concentration of roadside soil have been reported by a number of workers such as Chow (1970) and Cannon and Bowles (1962).

Chow (1970) states that the total traffic flow since the opening of the road, rather than the current traffic density, determines the lead concentration of the top 5 cm of an undisturbed soil. This has been shown to occur because the lead compounds in the top soil are not readily leached out into deeper layers due to their insoluble nature (Smith, 1976). In consequence, levels of lead at a depth of greater than 15 cm are not elevated above the background concentration for top soil, even at the central reservation of the M4 motorway (Little and Wiffen, 1977). The distribution of lead within the profile of the loamy, fine sand soil at the M6 study site was investigated by sampling at five depths, both near to, and 1,500 metres distant from the motorway. Table 4-2 shows that highest lead levels are associated with the top 5 cm of soil at both sites influenced by the motorway and

TABLE 4-1

Total Lead Concentration of the Top 5 cm of Roadside Soil in the UK

Author	Davies and Holmes (1972)				This Study					Bevan et al. (1974)	Little and Wiffen (1978)	
Date	July/August 1970				December 1978		July 1978		February 1979	March 1972	August 1976	
Location	4 roads near Birmingham				M5 motorway (Tewkesbury)		M6 motorway (Sandbach)		M1 motorway (Aldenham)	M4 motorway (Harlington)	M4 motorway (Heston)	
Traffic ** vehicles/day	503	7,500	17,500	19,000	26,000		46,000		74,000	90,000	90,000	
Sampling position relative to road	*	*	*	*	E	W	E	W	East	*	N	S
Distance from road edge												
C.R.	-	-	-	-	-	-	-	-	-	3,000	5,500	
0-2 metres	27	65	78	87	-	-	-	-	-	1,800	1,621	3,740
5	-	-	-	-	-	-	300	129	-	-	304	594
10	-	-	-	-	60	42	60	47	200	760	-	-
15	-	-	-	-	52	-	50	46	93	-	210	241
20	-	-	-	-	43	45	50	34	-	84	-	-
25	22	45	51	47	45	-	38	-	-	-	-	-
30	-	-	-	-	48	51	29	43	-	-	169	313
50	-	-	37	22	46	46	33	40	-	-	167	118
100	-	-	-	-	47	48	32	38	-	-	202	-
Background	-	-	25	-	37		30		59	-	200	

* Mean of samples taken each side of road.
** Mean annual 24 hour flow of all vehicles.

C.R. = Central Reservation.

TABLE 4-2

Variation in the Total Lead Concentration of

Soil with Depth at the M6 Study Area

Depth in Soil (cm)	Lead Concentration of Soil (µg/g)		
	10 metres East	10 metres West	1,500 metres West
0 - 2	76	50	31
2 - 5	71	23	32
5 - 10	51	16	24
10 - 15	24	11.5	32
15 - 20	16.5	11.5	17

at the background site. At depths of greater than 10 - 15 cm, there is little difference between the soil lead concentration at the three sites. This finding is, therefore, further evidence that lead is not removed from the surface soil by leaching processes.

B. Vegetation

In contrast to soil, the level of surface contamination of plant surfaces at the roadside will be the result of quite recent lead emissions rather than the total traffic flow since the opening of the road. This is because of the seasonal growth cycle of plants, which results in the loss of deposited lead from the plant canopy to the soil when leaves senesce and decay (Lagerwerff and Specht, 1970). Therefore, because of the seasonal changes in plant growth, the time of sampling must be expected to be an important factor determining the lead content of vegetation.

Table 4-3 compares the lead concentration of roadside pasture found by other workers in the UK with the results of this study. All data shown are for samples taken during the summer months so that any possible influence of a variation in lead concentrations due to plant growth will be minimised. All studies found an elevated level of lead adjacent to the road, and a rapid decrease with distance to a near background concentration at approximately 50 metres from the motorway edge. There is a general relationship between the traffic density and grass lead concentration, although, as discussed for soil, caution is required in the comparison of different sites.

TABLE 4-3

The Lead Concentration of Roadside Pasture in the UK

Author	Davies and Holmes (1972)				This Study		Vick (1976)	
Date	July/August 1970				August 1979		August 1975	
Location	South West Birmingham				M5 Tewkesbury		M6 near Birmingham	
Traffic ** vehicles/day	503	7,500	17,500	19,000	26,000		36,000	
Sample position relative to road	*	*	*	*	E	W	S	N
Distance from road edge (metres)								
0 - 2	14	38	44	50	-	-	-	-
5	-	-	-	-	-	-	-	-
10	-	-	-	-	31	7	75	30
15	-	-	-	-	23	3	25	37
20	12	21	35	22	-	6	32	35
30	-	-	-	-	-	4	-	15
50	-	-	20	10	-	4	-	-
100	-	-	-	-	-	2	-	-
Background	-	-	-	10	2	2	-	-

* Mean of samples taken both sides of road during dry weather conditions.

** Mean annual 24 hour flow of all vehicles.

Continued overleaf

TABLE 4-3 (Continued)

Author	Associated Octel Co Limited (1978 pers. comm.)		This Study		This Study	Little and Wiffen (1978)	
Date	August 1975		August 1978		July 1979	August 1976	
Location	M6 Cheshire (Peover)		M6 Sandbach		M1 Aldenham	M4 Heston	
Traffic ** vehicles/day	44,000		46,000		74,000	90,000	
Sample position relative to road	E	W	E	W	E	S	N
Distance from road edge (metres)							
0 - 2	-	-	-	-	-	1,180	572
5	-	-	73	41	-	770	390
10	55	33	41	33	21	-	-
15	-	-	42	26	10	628	228
20	-	-	33	10	5	-	-
30	-	-	30	6	-	228	226
50	27	17	12	11	-	162	83
100	22	9	8	4	6	-	75
Background	-	-	3	3	5	75	75

** Mean annual 24 hour flow of all vehicles.

A. Soil

Table 4-4 compares the lead content of the top 5 cm of the soil adjacent to the M6 motorway (Sandbach) in January and July 1978. Clearly, there is no significant difference between the soil lead concentration in winter and summer months. The lead which has been deposited between these two sampling dates is not sufficient to have had any significant effect on the lead content of the top soil. This is probably the result of the great mass of soil relative to the quantity of newly deposited lead.

TABLE 4-4

The Lead Concentration of the Top 5 cm of Soil at the M6 Study Site in January and July 1978

Sample Site (metres from motorway fence)	Lead Concentration of Soil (μg/g)			
	East of M6		West of M6	
	January	July	January	July
0	66	60	41	47
5	52	50	31	46
10	42	50	38	34
15	39	38	31	33
30	32	29	40	41
50	23	33	41	40
100	36	32	40	38

B. Vegetation

i) Review of Published Work

No published work involving the long term investigation of lead contamination of roadside pasture appears to be available. Everett et al. (1967) did, however, find a higher lead concentration of privet leaves during winter than summer months, both near roads and at remote sites in the UK. There are also a number of workers who have reported a considerable seasonal variation in the lead concentration of grassland in environments other than beside major roads. These studies may be divided into three types on the basis of the major lead source to the plant:

a) Sites which are not contaminated by local industrial emissions, major roads, or mineral ore deposits.

b) Sites contaminated by mineral ore deposits which have been mined by man, and contain tailings of high lead content near the ground surface.

c) Sites contaminated by the deposition of airborne lead which has resulted from industrial processes.

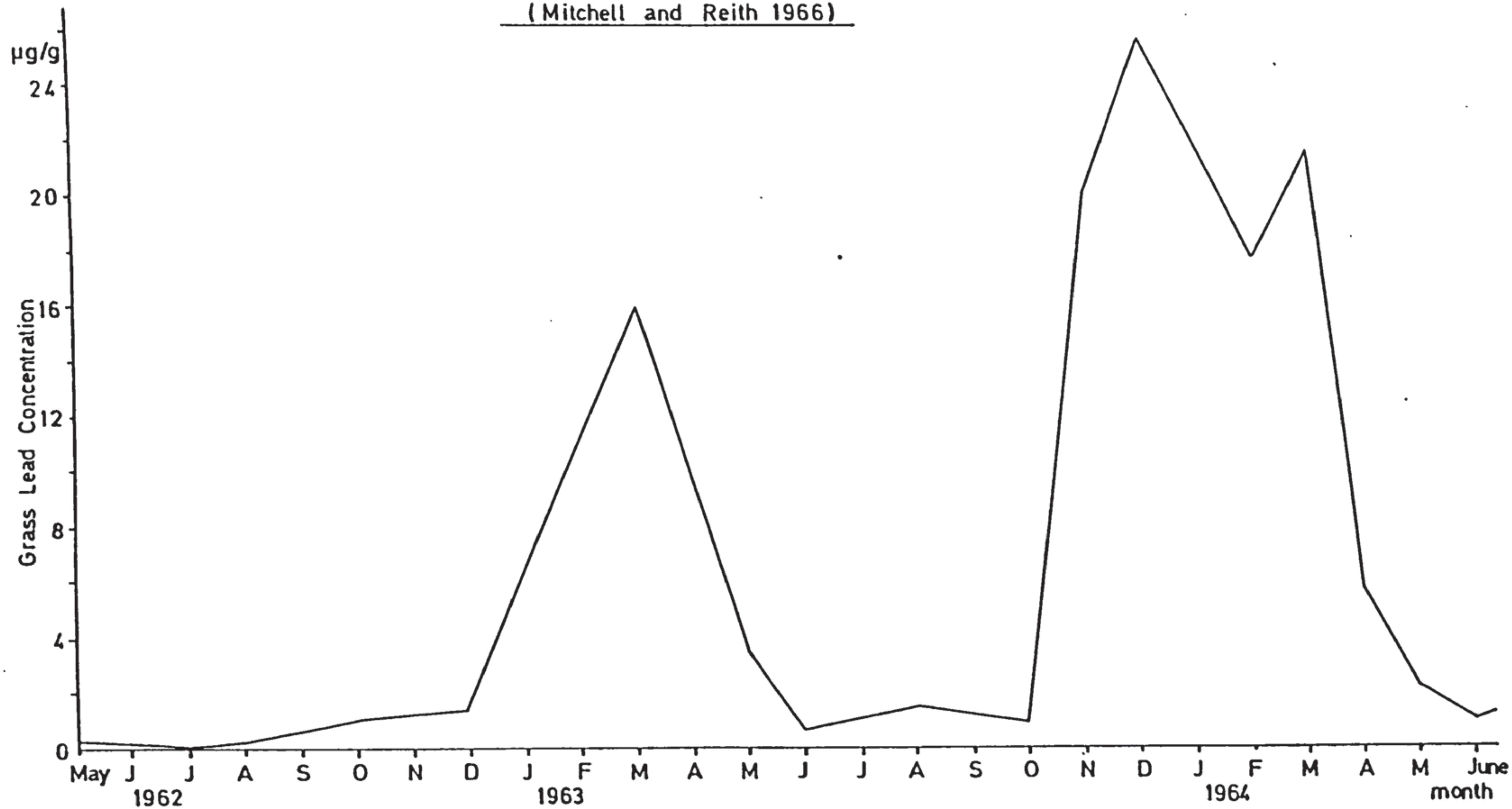
The findings of these studies will now be considered:

a) Uncontaminated Sites

Mitchell and Reith (1966) report a large seasonal variation in the lead content of pasture grass growing on a number of different soil types in North East Scotland. Figure 4-2 shows that levels of 0.3 - 1.5 μg/g (dry weight) occurred during periods of active grass growth, but during autumn the lead concentration increased to reach a peak of 30 - 40 μg/g by late winter. In spring, the lead levels declined rapidly with the onset of new growth. This paper also reports

FIGURE 4-2 Seasonal Change in the Lead Content of Mixed Pasture during 1963/4 in N.E. Scotland

(Mitchell and Reith 1966)



the results of Davey (1957) who found an increase in lead levels from summer to late autumn in mixed herbage which had been cut every three to four weeks to simulate grazing conditions.

Further evidence for a seasonal change in pasture lead concentration in the UK is provided by Wilkins (1978). This worker measured the lead concentration of grass at over 500 sites in Pembrokeshire (Wales) and found that levels increased from approximately 1 μg/g in June to over 4 μg/g in March.

b) Contamination by Mining Activities

A number of studies have investigated the lead content of pasture in lead-rich soils and disused mine-workings in Derbyshire. Shearer et al. (1940) reported that the lead content of grass collected from four fields in January showed a large increase over samples taken in November. Allcroft and Blaxter (1950) found higher lead levels in pasture during January than May. More recent studies in Derbyshire by Thornton and Kinniburgh (1978) found a ten-fold decline in grass lead concentration from February to May at both lead-rich and uncontaminated sites.

c) Contamination by Lead Smelters

In the UK, Little (1974) and Goodman and Roberts (1973) report a seasonal variation in the lead content of pasture grass in the vicinity of smelting works. However, perhaps the most detailed investigation of this seasonal variation is that of Rains (1971, 1975), who studied the lead content of the annual wild oat (*Avena fatua*) in the vicinity of a lead smelter in California, USA. He found that lead levels were greatest during the non-growing season, and declined rapidly with the appearance of new growth in spring.

Further information concerning vegetation contaminated by smelter emissions in the USA is reported by Dorn et al. (1975) and Knight and Burau (1973), and in Canada by Schmitt et al. (1971). These workers also found highest levels of lead in pasture during the winter months. Similarly, the findings of Mueller and Stanley (1970) identified higher lead levels in dry, over-wintered grass compared to fresh green growth in a contaminated region of California, USA.

In conclusion, therefore, it would appear that the pasture lead concentration is higher during winter than summer months at both lead-rich and relatively uncontaminated sites.

ii) <u>The Results of This Study</u>

a) <u>Lead Content of Herbage at the M6 Study Area</u>

Figure 4-3 shows the lead concentration of pasture grass along the study transect adjacent to the M6 motorway for the months of March and July 1978. Lead levels in March are approximately ten times those measured in July at all sample points. The influence of the vehicular lead emission upon the pasture is detectable over a greater distance during March, but there is little variation in lead levels beyond a distance of 60 metres from the motorway fence.

To demonstrate this seasonal variation more clearly, Figure 4-4 shows the mean lead concentration of grass samples taken at 0, 5 and 10 metres from the motorway fence and the mean of the five random samples taken at the 1,500 metres west site. The graph shows the large seasonal variation which occurred at all sites during the period October 1977 to July 1979. Although not included in the graph, samples taken as part of an investigation of the lead distribution in the plant canopy (Table 5-11) have shown that a similar seasonal rise and fall in

FIGURE 4-3 The Lead Concentration of Grass near the M6 Motorway in March and July 1978

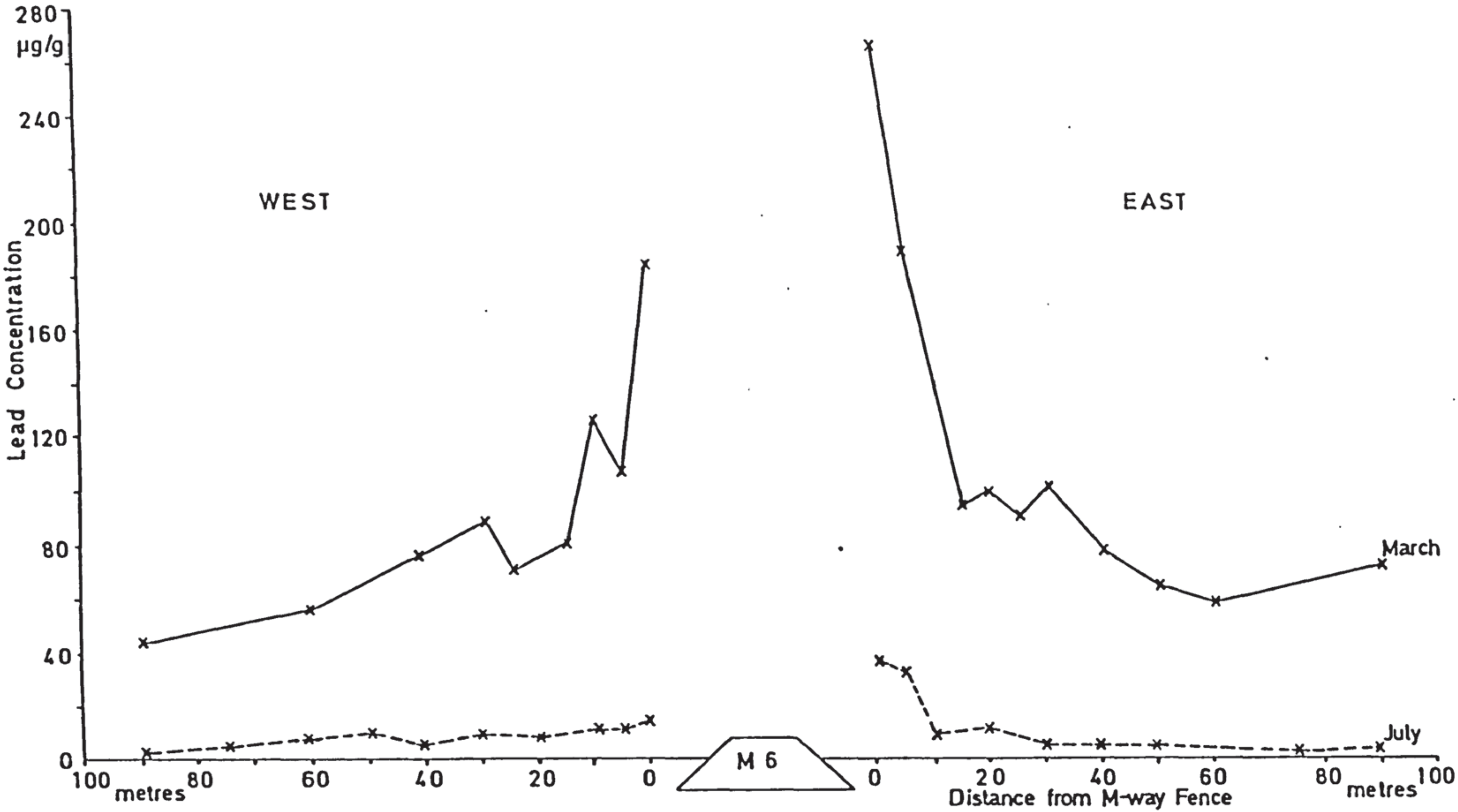
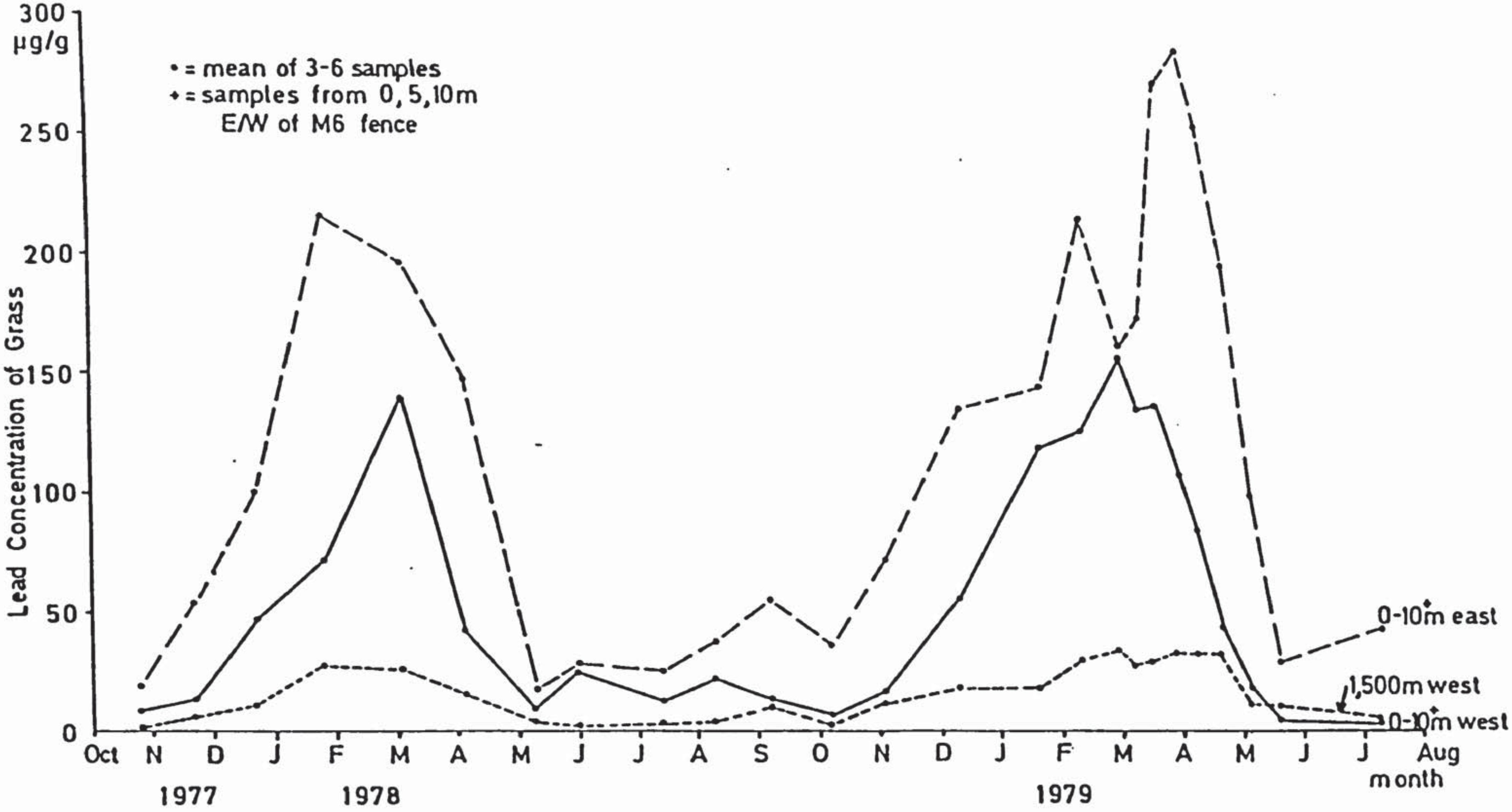


FIGURE 4-4 Seasonal Variation in the Lead Concentration of Pasture Grass at the Study Area in Cheshire



lead levels occurred between the autumn of 1979 and the spring of 1980.

Therefore, it may be concluded that a rapid rise and decline in lead concentration occurred during each autumn and spring respectively of the thirty month sampling period. This resulted in a ten-fold change in pasture lead content over periods of only two months to produce maximum lead levels in winter and a minimum in summer.

Highest lead levels were found in grass east of the motorway fence at all sampling occasions. This is a result of the prevailing westerly wind which greatly influences the rate of lead deposition to plant surfaces by its effect upon airborne lead concentration (Chapter 2). Crump et al. (1980) demonstrate how the strength of the prevailing wind between sampling occasions can be related to the relative rate of change in grass lead concentration immediately east and west of the motorway during the winter of 1977/78. This is further demonstrated by the pasture lead concentration in late February 1979 when levels east and west of the M6 motorway are almost equal. Prior to this date, the prevailing wind was easterly, resulting in a greater lead deposition west of the motorway and a convergence of the values for the lead content of pasture. However in March, the wind direction changed and the prevailing westerlies during the first two weeks of that month resulted in a rapid rise in levels east of the motorway, accompanied by a small decline to the west.

The lead concentration of pasture grass at the 1,500 metres west site showed a similar seasonal change to the pasture near the motorway. Levels at this background site were approximately one tenth of the lead content of the roadside grass in all months of the year. The lead content of pasture at the 1,000 metres east site was not significantly different from the 1,500 metres west site, suggesting that grass at these locations was not influenced by lead emissions from the motorway.

The possible mechanisms producing the seasonal change in lead concentration, both near to and distant from the M6 motorway, are discussed in detail in Chapter 5. It is clear, however, from a comparison of Figures 3-4 and 4-4, that the pasture lead concentration varies seasonally in a manner similar to that found for deposition rate to moss bags. In addition, the relationship between the lead content of pasture at the background and motorway sites during 1977/78 is of the same order as that for deposition rate to moss bags placed at these sites. This suggests, therefore, that the rate of deposition of atmospheric lead is an important factor determining the lead concentration of pasture grass in the M6 study area.

b) The Lead Content of Pasture at Other Sites

The investigation of the grass lead concentration in fields adjacent to the M5 motorway (Gloucestershire) and the M1 motorway (Hertfordshire) identified a similar seasonal variation to that found at the M6 (Sandbach) site (Table 4-5). This shows, therefore, that the seasonal variation in lead concentration is of a general occurrence and that the M6 study farm was not exceptional.

Further evidence that seasonal change in the lead content of roadside pasture is a widespread phenomenon is provided by the work of Vick (1976). Her study identified peak levels during winter months at three sites adjacent to the M6 motorway in the English Midlands. In addition, data from the Associated Octel Company Limited (unpublished personal communication, 1978) shows that winter pasture consistently contained a much higher lead concentration than summer growth during the years 1970 - 1976 in fields adjacent to the M6 motorway (see Appendix 2). Their sampling site was situated approximately 10 km north of the Sandbach study area and was similar in a number of respects,

TABLE 4-5

The Lead Content of Pasture at the M5 and M1 Motorway Study Sites

1. M5 (Gloucestershire)

Date of Sampling	Lead Concentration of Grass (μg/g)			
	100 m West	0 - 10 m * West	0 - 10 m * East	100 m East
April 1978	17	51	143	34
August 1978	4	10	72	10
December 1978	17	48	155	49
March 1979	30	117	160	54
August 1979	2	6	-	-

* Mean concentration of samples at 0, 5 and 10 metres from motorway fence.

2. M1 (Hertfordshire)

Date of Sampling (1979)	Lead Concentration of Grass to the East of the Motorway Fence (μg/g)		
	0 - 10 m	100 m	250 m
February	103	84	56
July	11	7	2

including topography, type of fencing, grass species and grazing patterns.

The evidence, therefore, indicates that the seasonal variation in the lead content of pasture is repeated annually and is not the result of exceptional factors during one or two years.

4.5 ZINC CONTENT OF SOIL AND PASTURE GRASS AT THE M6 STUDY AREA (SANDBACH, CHESHIRE)

The moss bag survey had shown that elevated levels of airborne zinc occur at the motorway fence, although this effect was not detectable at a distance of 60 metres from the road. This finding, therefore, corresponds to the small increase in soil and pasture zinc concentration found adjacent to the M6 motorway during 1977/79 which is demonstrated by the results for November 1977, shown in Table 4-6. In general, zinc levels at the motorway fence were no greater than about 50 per cent higher than background values for grass and soil, and this effect was restricted to an area within 10 metres of the fence.

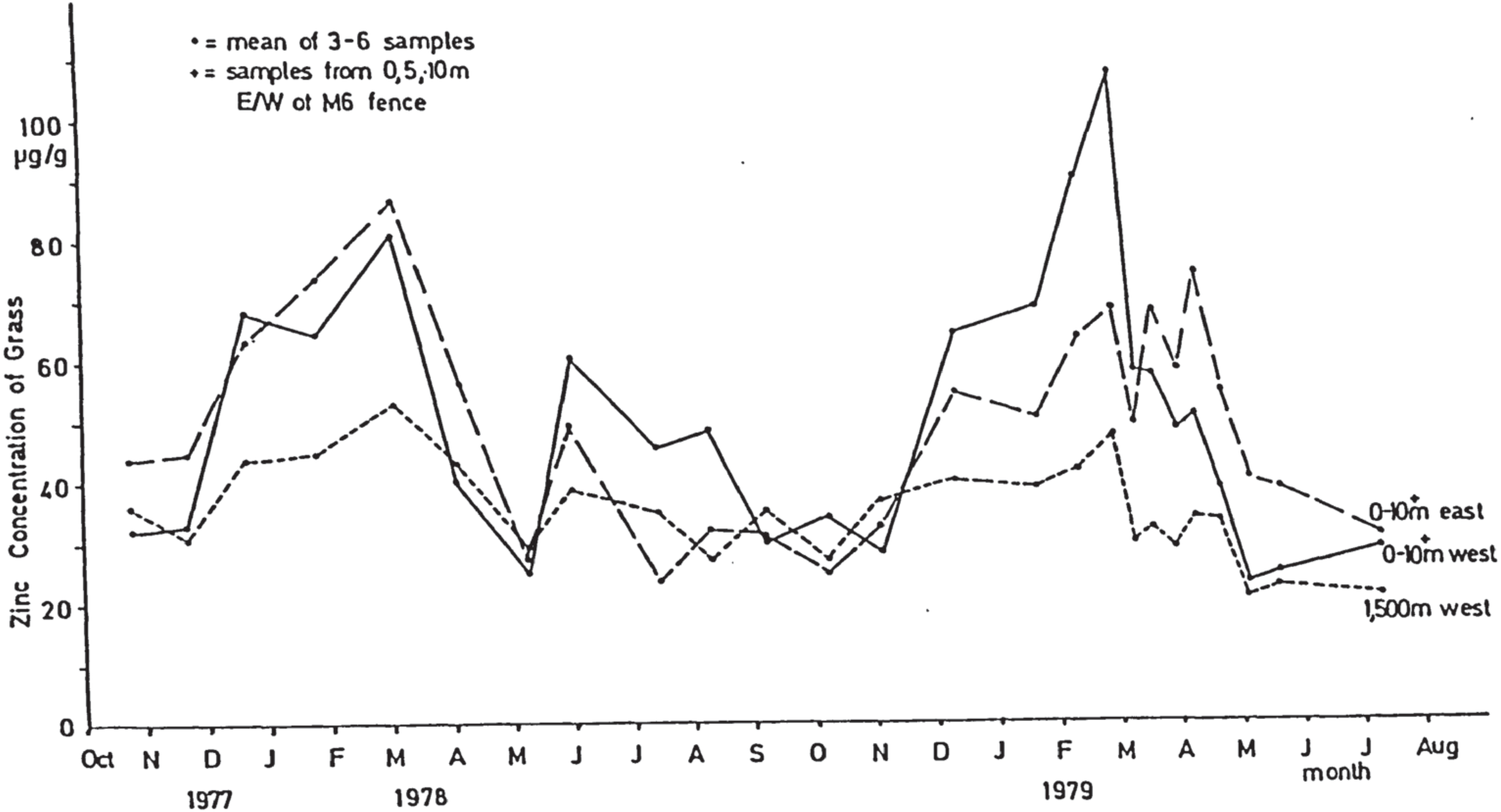
Figure 4-5 shows that pasture zinc content did vary seasonally with winter values being approximately 50 per cent greater than summer concentrations at most sites. This seasonal change was more pronounced adjacent to the motorway fence where the peak winter zinc levels were nearly three times greater than the lowest summer values. It is clear, however, that the pasture zinc concentration did not show as great a seasonal variation as did the lead concentration. This is despite the fact that the zinc deposition rate to moss bags showed a similar order of seasonal change as lead. The results suggest, therefore, that deposition of airborne lead and zinc have a more significant effect upon the lead concentration of herbage than upon the zinc content. Further evidence for such a conclusion is discussed in Chapter 5.

TABLE 4-6

Zinc Content of Surface Soil and Pasture Grass at the M6 Study Site During November 1977

Distance from motorway fence (metres)	Zinc Concentration of Pasture Grass (μg/g)		Zinc Concentration of Surface Soil (μg/g)	
	East of motorway	West of motorway	East of motorway	West of motorway
0	55	35	77	63
5	40	32	61	56
10	38	34	61	43
20	33	36	52	39
40	35	46	51	55
100	29	31	43	49
450	30	-	38	-
1,500	-	31	-	36

FIGURE 4-5 Seasonal Variation in the Zinc Concentration of Pasture Grass at the Study Area in Cheshire



4.6 CONCLUSION

Elevated concentrations of lead occur in the soil and pasture grass in the vicinity of a motorway, although the effect is largely restricted to an area within 60 metres of the road adge. Lead levels in pasture grass show a large seasonal variation both adjacent to a motorway and at background sites. Peak lead concentrations occur during winter months and are approximately ten times greater than the minimum values found in summer.

The seasonal variation in the rate of deposition of atmospheric lead corresponds to the measured changes in pasture lead concentration at the M6 study site. This suggests that airborne lead is the major source of lead to the pasture grass whereas the zinc content of herbage is not influenced by atmospheric deposition to the same degree.

CHAPTER 5

MECHANISMS DETERMINING THE GRASS LEAD CONCENTRATION IN THE VICINITY OF A MOTORWAY

5.1 INTRODUCTION

An understanding of the processes which determine the lead concentration of a plant is necessary to predict the circumstances which might result in high levels of contamination. Such an understanding may allow recommendations to be put forward concerning the type of farming practice which would minimise the exposure of farm livestock to roadside lead.

The study of the factors which influence the lead concentration of herbage is presented in this part of the thesis under two main sections:-

i) Part 1 considers the occurrence of lead in the pasture grass and the relative importance of airborne and soil sources of the metal.

ii) Part 2 discusses the factors which produce the seasonal change in the lead content of pasture.

PART 1 AN INVESTIGATION OF THE SOURCES OF LEAD TO PASTURE GRASS

5.2 INTERNAL v EXTERNAL LEAD

The major factors which influence the level of lead contamination near roads have been reviewed by a number of workers (Hunter, 1976; Coello et al, 1974; Smith, 1976; Lepp, 1975; Peterson, 1978; Page et al. 1971; Ter Haar, 1971; Holl and Hampp, 1975). These factors may be considered as conditions affecting:-

i) the level of contamination that results from the presence of lead-containing deposits on the surface of the plant;

ii) the plant tissue lead concentration which has resulted from lead uptake from the soil via the root system or possibly by absorption through the foliar surfaces.

Before any assessment of the importance that environmental factors may have upon the total plant lead concentration, the relative significance of the external deposits and the in-tissue lead must be distinguished. Four main types of approach have been used both in this study and by other workers to try and measure the contribution that the two methods of plant contamination make to the total lead concentration. These are:

A. Leaf washing studies.

B. Study of plant cuticles.

C. Greenhouse studies of lead uptake from the soil by plants.

D. Field studies of plants in both contaminated and relatively uncontaminated environments.

These experiments are now discussed.

5.3 WASHING OF LEAF SURFACES

Various workers have investigated the effect that the washing of leaf surfaces has upon the lead content of foliage collected from the roadside environment (Haney et al. 1974; Daines et al. 1970; Kloke and Riebartsch, 1964; Lerche and Breckle, 1974). These studies, together with that of leaves collected from vegetation contaminated by smelter emissions (Little, 1973; Rains, 1971) show the effectiveness of washing to be determined by a number of factors such as plant species, washing agent, condition of the leaf, and the physical and chemical form of the lead present. In addition, it is uncertain whether a particular washing agent leaches any of the in-tissue lead from the leaf.

Therefore, although a leaf washing experiment may indicate that a certain proportion of the total plant lead is readily removed, and thus is likely to be present as a surface deposit on the leaf, this technique does not quantify the ratio of in-tissue lead to external contamination. As a preliminary investigation of the occurrence of lead in or on grass leaves growing in the M6 study area, the following experiment was undertaken.

Method

The above ground portion of perennial ryegrass was collected from five one-metre square quadrats located adjacent to the motorway fence in fields B and I and at the C1 site shown in Figure 1-3. Sample collection was undertaken in early October 1979 when the grass was still actively growing, and during January 1980 when growth was negligible. Once returned to the laboratory, the grass from five quadrats at each site was thoroughly mixed by hand. From this mixture, 50 g (fresh weight) random samples were taken and placed in 100 ml round-bottomed flasks. To each weighed sample 100 ml of one of five washing agents (distilled water, rainwater, detergent, chloroform, dilute acid) was added and the resulting mixture shaken for one hour. The solution was then decanted and the grass rinsed by shaking with a further 50 ml of the washing agent for 15 minutes. This was then decanted and the metal content of the grass determined in the manner described previously in Chapter 1. The results were then compared with the lead and zinc concentration of unwashed grass which had also been taken randomly from the bulked harvest of five quadrats.

Results and Discussion

Table 5-1a shows the effect of the five washing agents on the lead concentration of the grass. The dilute acid wash and detergent wash were equally effective, and removed between 70 and 90 per cent of the

TABLE 5-1

The Effect of Various Washing Agents on the Lead and Zinc Concentration of Grass

a) Lead

Date of Sampling	Field Location	Lead Concentration before Washing (μg/g)	Percentage of Lead Removed by Washing				
			Distilled Water	Rain Water	10% 'Decon 90' Detergent	10% Nitric Acid	Chloroform
October 1979	C1	3.5	35	9	72	-	25
	E1	75	11	40	85	-	0
January 1980	C1	14	4	-	90	90	0
	E1	174	9	-	92	88	5
	B	30	-	-	92	-	-

C1 = Control 1

E1 = East 1

TABLE 5-1 (Continued)

b) Zinc

Date of Sampling	Field Location	Lead Concentration before Washing (μg/g)	Percentage of Lead Removed by Washing				
			Distilled Water	Rain Water	10% 'Decon 90' Detergent	10% Nitric Acid	Chloroform
October 1979	C1	21	0	0	11	-	0
	E1	23	0	0	34	-	0
January 1980	C1	36.5	13	-	40	67	0
	E1	50	2	-	52	65	0
	B	36.5	-	-	29	-	-

C1 = Control 1

E1 = East 1

lead from the grass. Distilled water, rainwater and chloroform removed no more than 40 per cent of the lead and less than 10 per cent for some samples. Chloroform was the least effective washing agent, which may suggest that dissolution was the most important mechanism of lead removal rather than displacement of particles by mechanical action.

Arvik and Zimdahl (1974a) have used chloroform to remove wax deposits associated with the cuticles of plants. In consequence, it was expected that the chloroform wash would remove lead particulates attached to surface wax deposits on the ryegrass leaves. However, the ineffectiveness of the chloroform wash suggests that either the wax layer was not removed, or else lead particulates remained attached to non-waxy regions of the leaf, as observed for other plant species by Rentschler (1977).

Table 5-1b shows that zinc is less readily removed from the grass by the various washing agents than is the case for lead. This is in agreement with the findings of Little and Martin (1972) who found a detergent solution removed 80 per cent of the lead from elm leaves but only 25 to 33 per cent of the zinc. It is most likely that the greater zinc retention is a result of a much higher percentage of the zinc occurring within plant tissues compared with lead which may be largely a surface deposit.

Conclusion

The results show that most of the lead associated with the grass leaves is strongly bound and the greater proportion can only be removed by strong washing agents such as detergents or acids. Provided that the detergent or acid did not leach lead from within the plant tissue, the results show that the majority of lead occurs as a surface deposit. Also, a comparison between the January and October samples suggests that the relative amounts of lead that can be removed by washing does not differ greatly from autumn to midwinter.

5.4 STUDIES OF PLANT CUTICLES

One possible source of lead into plant tissues is the entry of airborne lead into leaves through the foliar surface. This could conceivably occur by diffusion of lead in solution across leaf epithelia or by lead aerosols entering stomatal apertures. Studies concerning foliar lead intake are reviewed by Zimdahl (1976) who concludes that uptake through leaf epithelia can occur, but the significance of the process remains to be resolved. Arvik and Zimdahl (1974) showed that the presence of wax deposits reduced the penetration of lead solutions across isolated plant cuticles.

Peterson (1978) also reviews studies concerning foliar uptake and concludes that the intact cuticle excludes significant absorption. However, he suggests that the possible entry of lead through cracks in the cuticle and damaged regions of the leaf remains to be investigated. This paper also considers the findings of Roberts (1976) who found evidence for lead aerosols entering the stomatal cavity in an area of high airborne concentrations, but suggested that surface tension over stomatal pores prevents the entry of lead in solution.

The surface characteristics of three grass species at the study site were investigated by electron microscopy. Pieces of leaf blade and sheath were dried in carbon dioxide and prepared for stereoscan examination by a gold spluttering technique. Plates 3 and 4 show the occurrence of crystalline wax deposits in the vicinity of stomata of <u>Festuca rubra</u> and <u>Holcus lanatus</u> respectively which are occasional grass species in the pasture near the motorway fence. The surface wax occurs over most regions of the leaf and thus may be expected to

PLATE 3

Electron Microscope Picture of the Surface of a leaf of Festuca rubra

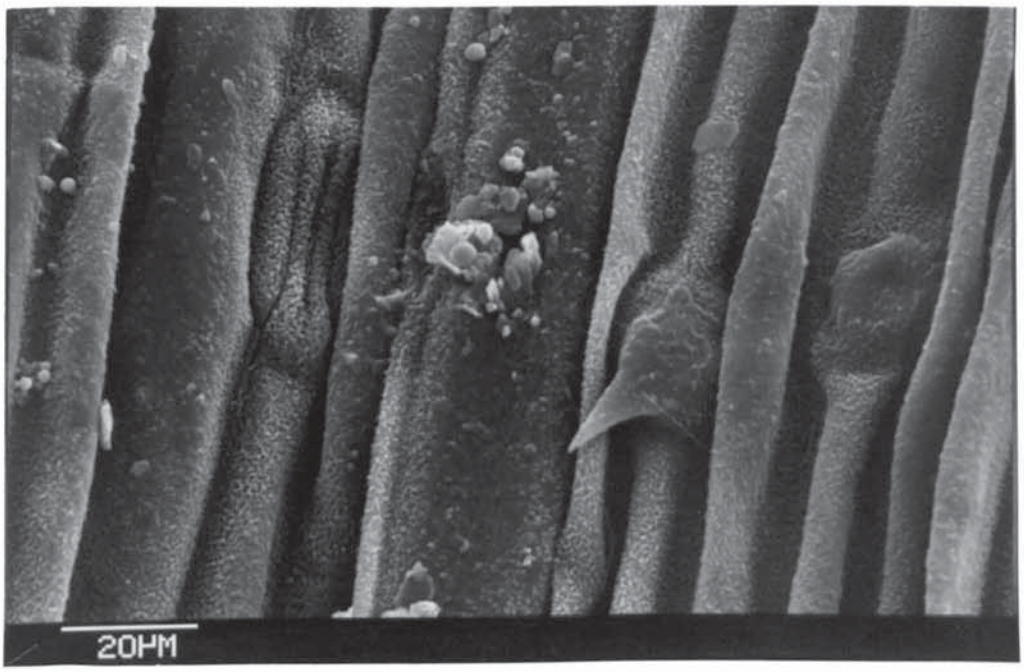
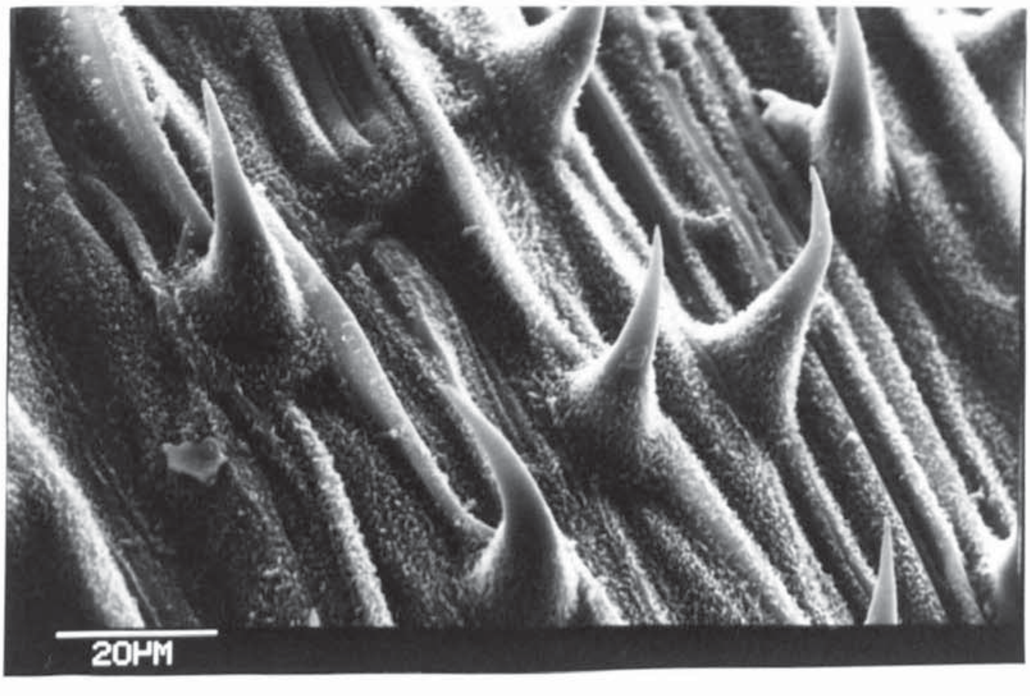


PLATE 4

Electron Microscope Picture of the Surface of a leaf of Holcus lanatus



minimise foliar absorption of lead in solution. Plate 5 shows in detail the heavy deposition of wax over the stomata of Festuca rubra which may prevent the entry of lead aerosols into stomata as suggested for pine needles by Godzik et al. (1979).

Stomata are not as clearly visible on the surfaces of leaves of Lolium perenne. It would appear that stomata are situated in the troughs of the furrowed surface texture shown by Plate 6. A pore structure which is probably a stomatal opening is present in the top left-hand corner of the photograph. It would also appear from Plate 6 that there is a lack of a rough textured crystalline wax deposit over large areas of the leaf of Lolium perenne. However, Leafe (1980, pers. comm.), by using electron microscopy, has identified wax deposits on leaves of perennial ryegrass which are aggregated around the stomata. Harris et al. (1974) report that the waxy cuticular deposits of ryegrass leaves may be removed by steam containing petroleum ether. Further work on studying both the effectiveness of this wash in removing wax deposits and the influence of this wax removal upon plant lead concentrations is recommended.

In conclusion, it would appear that studies of plant cuticles have shown that foliar absorption of airborne lead is unlikely to be of great significance for roadside vegetation. Possibly it does occur more readily in plant species without a waxy cuticle, although this appears not to be the case for the grasses investigated in this work. Factors controlling the environmental cycling of in-tissue lead are likely to differ quite considerably from those determining the movement of surface lead deposits. However, the process of foliar absorption is still dependent upon the factors which initially determine the level of surface contamination of the plant, and the airborne lead concentration in the air boundary layer of the leaf. In contrast, lead uptake via

PLATE 5

Electron Microscope Picture of Stomata of Festuca rubra

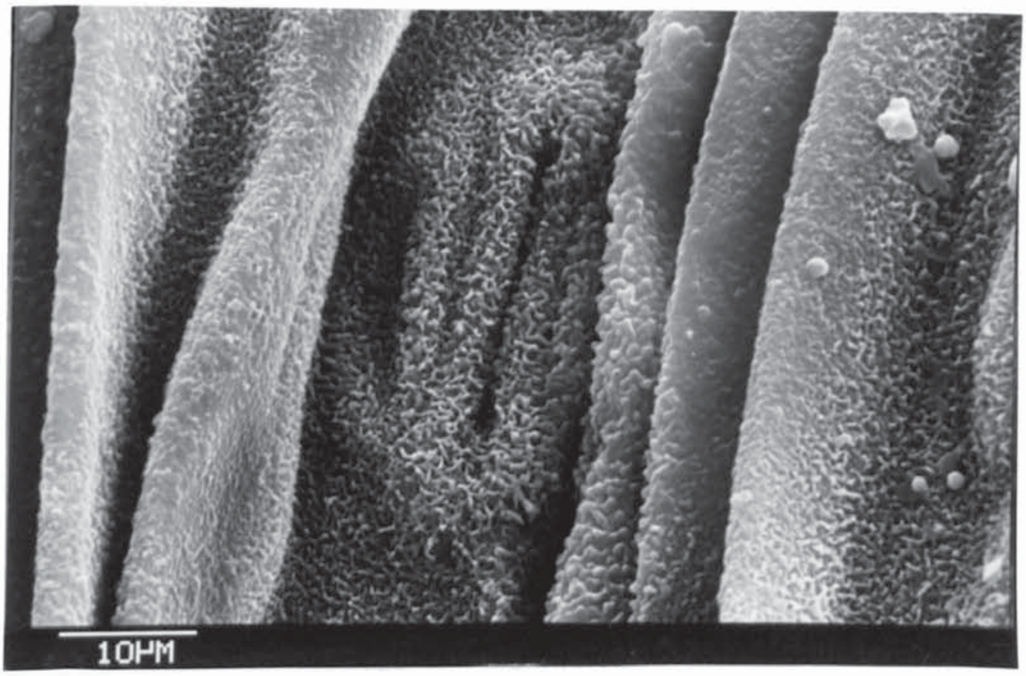
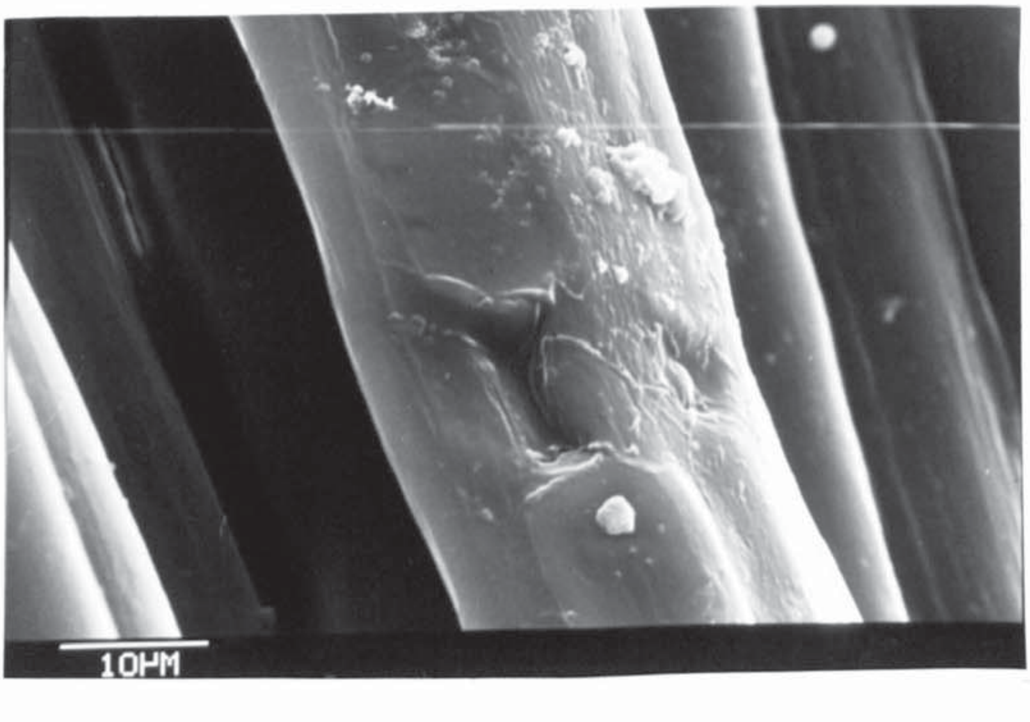


PLATE 6

Electron Microscope Picture of the Surface of a leaf of Lolium perenne



the roots will occur independently of current concentrations of air-borne lead.

5.5 GREENHOUSE STUDIES OF LEAD UPTAKE BY PLANTS

A. Published Work

The general findings of many plant growth studies (e.g., Baumhardt and Welch, 1972; Laitinen, 1974; Marten and Hammond, 1966; Rains, 1975; and Hassett and Miller, 1977) indicate that soil lead is largely unavailable to plants, although an increase in soil lead concentration is reflected by a relatively small rise in plant tissue levels. Lead availability to plants has been shown to increase with lower soil pH (Macleans, 1969) and to be influenced by many other factors such as soil texture, organic matter content, calcium concentration and presence of other metals (Zimdahl, 1976). The binding or exchange capacity of the soil, which is often directly correlated with soil organic matter content, is a particularly important determinant of lead availability to plants. Lead may become bound to organic material and then is not taken up by plant roots (Zimdahl and Koeppe, 1979).

The majority of lead entering plant roots is as a result of non-metabolic processes, although some active uptake may occur (Arvik and Zimdahl, 1974b). Tanton and Crowdy (1971) show that lead may move from the "free space" of the root into xylem vessels. Transport may then occur in xylem vessels of the stem to the leaves (Glater and Hernandez, 1972).

The lead concentration of roots is generally greater than shoots. Thus, although movement of lead can occur from roots to aerial parts of the plant, the root is considered as a barrier to transport of this metal under most soil conditions (Wallace and Romney, 1975). Malone et al. (1974) and Jones et al. (1973a) have demonstrated the translocation of lead from root to shoot in ryegrass, but less than 30 per cent of total uptake was involved. An increase in the lead concentration of shoots of perennial ryegrass has been observed when sulphur was deficient (Jones et al. 1973b) and for tree seedlings when phosphorus was limiting (Rolfe, 1973). Ter Haar (1975) also found an increase in the lead concentration of grass leaves when grown in a culture solution in which water was withheld. It is not clear if this is the result of an increase in translocation of lead to shoots or a normal rate of transport to the aerial plant, which has poor growth resulting in a build-up of lead concentration.

B. An Investigation of Lead Uptake from the Soil by Grass Grown at the Study Site

The uptake of lead by ryegrass from soil taken from the study farm was investigated by growing grass from seed in samples of top soil in the laboratory. This was undertaken as follows:-

Method

A sample of a "Gro-Plan" mixture of Italian ryegrass (*Lolium multiflorum* : variety Motherwitzer) and clover, which is occasionally used on the study farm, was obtained for the growth experiments. In July 1978, soil and dust was collected from the motorway verge and topsoil from the site 1,500 metres west of the M6. The soils were then dried before use in this experiment.

Three 0.5 m^2 experimental trays were set up as follows:

a) 5 g of seed mixture was sown at a depth of 2 cm in the control soil (pH 6.8) and watered with deionised water.

b) 5 g of seed mixture was sown at a depth of 2 cm in the dust and soil (pH 6.7) collected from the motorway verge and watered with deionised water with the same frequency as Tray 1.

c) 5 g of seed were spread over cotton wool soaked in a nutrient solution made up in deionised water. The tray was covered with a perforated black polythene sheet to minimise contamination of the growth medium by airborne lead.

After a period of twenty days, one half of the tray was harvested. It was found necessary to harvest all of the hydroponically grown grass to obtain a sufficient yield for analysis. After a total of ninety days, the remaining grass in each tray was harvested, together with that produced by a new hydroponic tray set up after harvest 1. All grass material and growth media were analysed for metal content as described previously (Chapter 1).

Results and Discussion

Table 5-2a shows the metal content of the growth media and of the seeds (after washing in deionised water) used in the experiment. Levels of lead, copper, cadmium and zinc are elevated in the motorway verge soil compared with the control soil. The lead levels in the verge soil are about 150-fold greater than at the 1,500 metres west site.

Table 5-2b shows the concentration of each of the four metals in the grass after a period of twenty days. If total soil metal concentration is compared to the concentration in the grass, it is clear that lead is less readily taken up into the plant shoots compared with the other three metals. Also of interest is the higher concentration of lead in the roots than shoots of grass in the hydroponic tray. This is in agreement with the findings of other workers

TABLE 5-2

The Uptake of Trace Metals by the Root System and Their Translocation to the Shoot in Ryegrass

a) Metal Content of Grass Seed and Growth Media

Sample	Lead (µg/g)	Copper (µg/g)	Cadmium (µg/g)	Zinc (µg/g)
Seed	1.0	4	0.08	34
C1 soil	24	4	0.3	40
Motorway verge soil	3,800	200	2.0	300
Hydroponic solution*	< 0.005	0.08	< 0.001	< 0.05

* in µg/ml

b) Metal Content of Grass at Harvest 1 (20 days)

Sample	Lead (µg/g)	Copper (µg/g)	Cadmium (µg/g)	Zinc (µg/g)
C1 shoot	< 0.5	11	0.15	70
Motorway verge shoot	4	17	0.4	85
Hydroponic; 1) root	3	8	0.25	65
2) shoot	< 0.5	11	0.5	73

c) Metal Content of Grass at Harvest 2 (90 days)

Sample	Lead (µg/g)	Copper (µg/g)	Cadmium (µg/g)	Zinc (µg/g)
C1 shoot	5	14	-	185
Motorway verge shoot	40	54	-	207
Hydroponic shoot	1.5	13	-	112

who have suggested that the root is a barrier to transport of lead in plants. This finding also suggests that, despite all precautions, the culture solution was not lead-free and indeed, the seed itself contributed some metal to the solution. However, the low levels in the grass shoot show that airborne deposition did not add significantly to the lead concentration of the foliage in the three experimental trays.

Table 5-2c shows the metal concentration of the grass shoots after ninety days. The lead content of the grass in all three trays has increased approximately ten-fold. Zinc levels have more than doubled, and an increase was apparent for copper only from the contaminated motorway soil. Lead concentrations in the shoots are still, however, low compared with the very high soil levels at the motorway verge. It is not possible to distinguish if the increase in shoot lead concentration between the two harvests is a result of changes with plant age or due to nutrient stress, as was found by Jones et al. (1973) and Rolfe (1973). A more complex experiment is required to investigate this further. This would involve the use of suitable controls to separate the effects of plant ageing and stress due to nutrient deficiency upon the lead content of grass leaves.

Conclusion

The laboratory growth studies have shown that lead is taken up from the soil by ryegrass, but only small amounts are translocated to the above-ground portion of the plant compared with the concentration in the soil. Copper, zinc, and cadmium are more readily transported to the grass foliage than is the case for lead. The metal concentrations of the grass leaves increased with time and this was most significant for lead.

5.6 FIELD STUDIES OF LEAD IN PLANTS

A. Published Work

Studies of the total lead content of vegetation, its soil support, and the distribution of the element in the plant give an indication as to the source of lead. Normally, the lead concentration of plants is less than that of the soil in which they are growing at both uncontaminated (Warren and Delavault, 1960) and lead enriched sites (Badham et al. 1979; Alloway and Davies, 1971; Kirkham, 1976). Yet in areas of high atmospheric deposition of lead, such as near smelting works and major roads, the plant foliage may have a greater lead concentration than surface soil. This suggests, therefore, that surface contamination of foliage is a major source of lead to plants in the roadside environment.

A number of studies of roadside crops have shown that foliage contains much higher levels of lead than plant organs not exposed to airborne contamination, such as cereal grain, bulbs and tubers, and inner leaves of cabbages and lettuces (Schuck and Locke, 1970; Sommer et al. 1971; Ter Haar, 1971; Egan, 1972). For example, Ter Haar (1971) grew a number of food crops near major roads and also in greenhouses with filtered and unfiltered air. Near roads and in unfiltered air, lead concentrations of leaves, husks and chaff were two to three times higher than in filtered air. However, no increase was found in the edible portion of the plants, which were not in contact with the aerial lead.

Some quantitative assessments of the contribution of deposited lead to total concentration of plants have been made. Lagerwerff (1971) grew radishes in soil contaminated by automobile lead, both exposed to aerial contamination and protected in a greenhouse with filtered air. It was concluded that aerial lead accounted for 40 per cent of the total lead content of radish tops. In a further greenhouse experiment, Ter Haar et al. (1969) grew radishes and perennial ryegrass in filtered air (containing 0.03 μg Pb/m^3) and in an ambient atmosphere (containing 0.96 μg Pb/m^3). They concluded that 50 per cent of the lead content of grass was from aerial contamination, but most associated with radishes was from the soil. Two further studies using isotopically labelled lead have concluded that airborne lead accounts for over 80 per cent of the lead in vegetation. Rabinowitz (1972) used stable lead isotopes to distinguish uptake from the soil by oats and lettuce from atmospheric contamination. Lead was added to soil in experimental trays to produce concentrations ranging from 97 to 1,300 μg/g. The crops were planted in this soil in a greenhouse and after two weeks the trays were placed near a major road or at a remote site. It was found that aerosols were the major source of lead to the plants even at the remote site when the soil contained 97 μg/g of lead. Rabinowitz concluded that 80 per cent of the lead content of crops grown in the Salinas Valley of California (USA) resulted from the deposition of atmospheric lead which had been emitted by motor vehicles. Tjell et al. (1979) grew ryegrass (Lolium multiflorum) in a soil to which radio-actively labelled lead had been added. They concluded that over 90 per cent of the total grass lead content originated from airborne deposition during the period June 1977 to March 1978 at a relatively unpolluted site in Denmark.

The results of field investigations would suggest, therefore, that in general, airborne lead is the major source of lead to vegetation, particularly to species with a large leaf area relative to the dry weight of the plant. This situation may vary depending upon factors such as the availability of soil lead to plants, the plant species, and the rate of lead deposition from the atmosphere. There is little quantitative information concerning the relative importance of soil and atmospheric sources of lead, and the investigations which have been carried out may be criticised because experimental conditions may differ from the field.

B. Investigations at the Sandbach Study Site

a) The Use of Moss Bags to Quantify the Effect of Airborne Deposition Upon the Lead Concentration of Pasture

The collection characteristics of moss and grass surfaces have been shown to vary in a similar manner for a wide range of particle sizes and wind speed conditions (Chapter 3). Therefore, moss bags are a useful measure of the dry deposition rate of particles to the vegetation above which they are suspended (Clough, 1975). However, the moss bag is contaminated by lead only as a result of deposition from the atmosphere, whereas rooted plants may additionally pick up lead by uptake from the soil. In addition, the grassland may lose lead from its plant canopy by the processes of death and decay, abrasion, rainfall, leaching and possibly in association with the peeling of cuticular wax. Thus, although the collection characteristics of grass and moss have been shown to be similar, the retention ability of the grass surface and that of the moss bag suspended above the pasture has not been fully investigated. This was considered to be worthy of study.

i) The Retention and Collection of Airborne Lead by Grass and Moss

Method

In the winter of 1979 a field study showed that lead collected by moss bags adjacent to the motorway was strongly retained over a period of four weeks when placed at a background site (Chapter 3). This experiment was expanded to include grass bags. These were constructed by weighing out 10 grams (fresh weight) of perennial ryegrass leaves harvested from the 1,500 metres west site during October 1979. The weighed out sample of grass was then placed in a nylon mesh bag (8 x 8 cm) of the same dimensions as the moss bags. Ten grass bags were then suspended alongside the moss bags during November 1979 at the 10 metres east site. After a period of one month, five of the grass bags were removed to the 1,500 metres west survey point and exposed for a further four weeks. The remaining five grass bags at the 10 metres east site were returned to the laboratory for analysis. The grass was analysed in the same manner as the moss, which has been described previously in Chapter 3.

A further group of five grass bags was suspended adjacent to those containing moss at both the 1,500 metres west and 10 metres west sites during November 1979 in order to compare the collection characteristics of the grass and moss under conditions of different atmospheric contamination.

Results

Table 5-3 compares the lead concentration of the grass and moss bags. Clearly, both types of bag depicted a maximum deposition of lead 10 metres east of the motorway and a minimum 1,500 metres west of the road. The moss collected a greater amount of lead. This is to be expected as the surface features of moss make it a more efficient collector of particulates (Clough, 1975; Little and Wiffen, 1977). Even though the moss bags are more efficient at collecting lead the use of grass bags, particularly in highly contaminated areas, has advantages in that grass of a low lead content is more readily available.

TABLE 5-3

A Comparison of the Lead Concentration of Moss Bags and Grass Bags after One Month Exposure during November/December 1979

Location Bags Exposed	Lead Concentration (µg/g)	
	Moss Bags	Grass Bags
1,500 m west	49 (4)	23 (1.6)
10 m west	100 (12)	83 (8)
10 m east	507 (64)	345 (52)
10 m east then placed at 1,500 m west for four weeks	568 (24)	454 (122)
Unexposed moss	20.6 (2.0)	3.3 (0.2)

The results of the transfer of grass bags to the background west site show that no significant losses of deposited lead occur over a four week period, a finding in agreement with the results for moss bags. Indeed it would seem that a small but not significant gain in the lead content of the bags has resulted from the continued exposure.

Conclusion

The results show that non-growing grass surfaces do collect lead in proportion to the level of airborne contamination in a similar manner as moss surfaces. The moss is, however, a more efficient collector, but neither grass nor moss bags appear to lose any significant amount of the lead once it is deposited.

ii) The Relationship between Deposition Rate to Moss Bags and Pasture Lead Concentration

Method

At each of six survey points, the mean lead deposition rate to moss bags was calculated as was the mean lead concentration of the grass below the bags. The grass sampling was carried out at approximately the same time as the collection and replacement of moss bags and thus the pasture lead content was compared with the deposition rate during the previous four to five weeks. Although not all of the moss bag survey points were immediately above the grass sampling transect (see Figures 1-3 and 3-2), it is unlikely that deposition rates would vary greatly over lateral distances of up to 50 metres.

Data concerning the winter and summer months was treated separately, as the rapid growth of grass in summer may result in changes in pasture lead concentration which are independent of the monthly changes in deposition rate. During winter, however, the pasture grass will have very limited growth, and thus may be regarded as resembling the grass bag as a collection surface for airborne particulates. This is important as the similarity of grass and moss bags for collecting and retaining atmospheric lead has already been demonstrated.

Results and Discussion : Winter Months

Figure 5-1 shows the relationship found between grass lead concentration and deposition rate to moss bags during the winter months of 1977/78 at the survey point 200 metres east of the M6 motorway. This clearly shows a strong linear relationship even though deposition rate and grass lead concentration vary by a factor of three over the winter period. A preliminary report of the relationship has been presented by Crump and Barlow (1980), but a more detailed discussion will now be given.

If all of the lead associated with the pasture resulted from the deposition of airborne particulates, it would be expected that the grass lead content would be zero if no deposition had occurred. However, extrapolation of the straight line graph depicting the relationship between grass lead concentration and deposition rate in Figure 5-1 to a point of zero deposition, predicts that the pasture grass will have a lead concentration of approximately 10 μg/g (dry weight) which has not resulted from atmospheric sources.

Table 5-4 shows the correlation coefficients for deposition and grass lead concentration at each of the six survey points. Also shown in this Table are the predicted values for the lead concent of pasture grass, which has not resulted directly from atmospheric sources. This was found by calculating "a" in the equation $y = a + bx$. The method of determining the importance of airborne lead for plant lead

FIGURE 5-1 The Relationship between Grass Lead Concentration and the Deposition Rate of Lead 200 m East of the M6 Motorway during the Winter of 1977/8

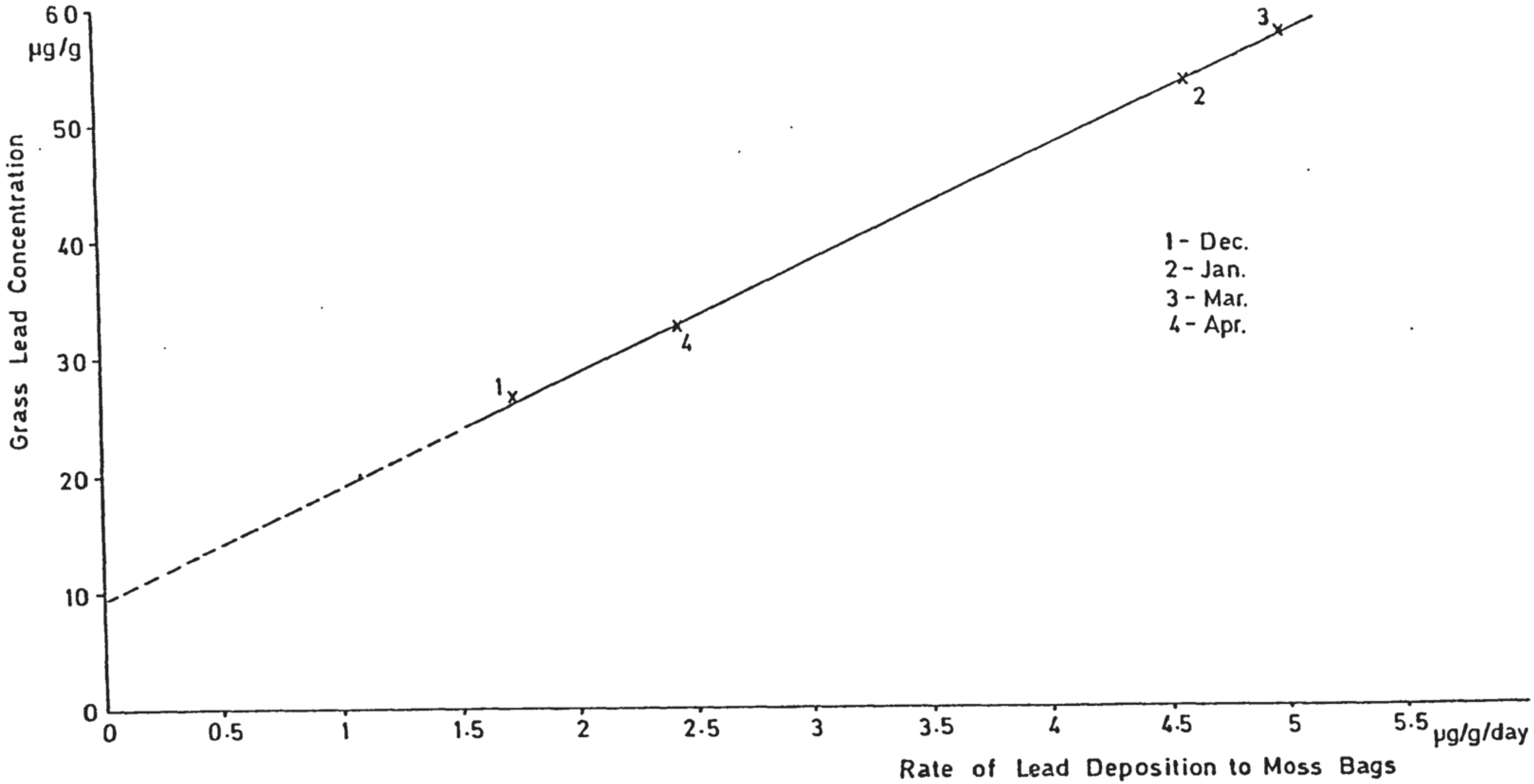


TABLE 5-4

Predicted Lead Concentration in Plant Tissues based upon the Correlation Coefficient for Deposition Rate and Total Grass Lead Levels in Winter

Site (distance) from M6 in metres)	Number of Samples	Correlation Coefficient (Significance)	Predicted Lead Concentration in Tissue (μg/g)
450 m east	4	0.997 (0.001)	6.5
200 m east	4	0.999 (0.001)	9.5
10 - 20 m east	4	0.986 (0.01)	43.5
10 - 20 m west	4	0.99 (0.01)	5.0
100 m west	3	0.99 (0.01)	8.0
1,500 m west	3	0.98 (0.02)	3.5

concentration presumes that the relationship between deposition rate and grass lead concentration remains linear for low levels of contamination. Although this cannot be shown with the available data, it is encouraging that a linear relationship has held for deposition rates and grass lead levels ranging from 0.46 to 23.5 μg/g/day and 10.7 to 28.7 μg/g respectively. Despite the small number of samples, the correlation coefficient was found to be statistically significant at the 0.02 or better probability level.

This linear relationship between grass lead concentration and deposition rate during the winter months provides a means of quantifying the relative importance of atmospheric lead and other sources as a contaminant of grassland. Unlike field and laboratory studies described previously, this investigation has involved minimum disturbance of actual environmental conditions. Lead, which has not resulted from airborne deposition, must have originated either from uptake from the soil via the roots or the presence of lead-containing soil particles on the surface of the plant. It appears unlikely that soil particles on the plant surface contribute to the total grass lead concentration in winter, at the M6 study site, as grass lead levels are higher than the lead content of the topsoil. In addition, it is improbable that this source of lead would remain constant for the winter period, which must be the case for the linear relationship between moss and grass to occur. Therefore, the most plausible explanation is that the non-atmospheric lead is the result of uptake from the soil by grass roots and is present in the plant tissues rather than as a surface deposit. Further evidence for this hypothesis comes from Mitchell and Reith (1966) who suggested that an increase in lead translocation to grass shoots during the autumn may account for a seasonal change in pasture lead concentration. These workers found that soil contamination on the plant surface did not explain the autumn rise in lead levels. In addition, the seasonal change in the lead content of grass growing on a disused mining site was not due to external soil contamination (Thornton, 1980, personal communication).

Table 5-5 compares the predicted concentration of non-atmospheric lead in the plant with the total soil lead content at each of the survey points. The results suggest that the tissue lead concentration in winter is greater in soils with a higher lead content. The predicted non-atmospheric grass lead does, however, seem high in relation to the soil lead content at the 10 - 20 metres east site. Possibly the mean value of soil lead concentration is not a good reflection of the rooting soil over the 10 metres distance, during which soil lead declines rapidly from a value of about 80 μg/g at the motorway fence to perhaps 30 μg/g at a distance of 20 metres from the roadside.

Also in Table 5-5, the predicted non-atmospheric lead content of the ryegrass is compared with the minimum total lead concentration of the grass found during the following summer. Clearly, the predicted winter values are 1.5 to 3 times greater than the minimum total grass lead content in summer. This suggests, therefore, that the internal lead concentration increased during autumn, possibly as a result of a greater translocation of lead to the shoots, or due to continued transport from the roots in a period of slow winter growth. Support for both of these mechanisms comes from the work of Rolfe (1973), Jones et al. (1973), and Ter Haar (1975), referred to earlier in this Chapter.

At the M6 study site it would seem that the internal plant lead content remains quite constant during winter, or else a linear relationship between deposition rate and lead concentration would not

TABLE 5-5

Comparison of Predicted In-Plant Lead Concentration during Winter with Total Concentration of Lead in Soil and Summer Grass

Site (distance from M6 in metres)	Predicted Lead Concentration in Plant Tissue (µg/g)	Lead Content of Surface Soil (µg/g) (soluble in conc. nitric acid)	Minimum Total Lead Content of Grass in Summer (µg/g)
450 m east	6.5	27	2.5
200 m east	9.5	27.5	3.0
* 10-20 m east	43.5	54	17
* 10-20 m west	5.0	36	9.0
100 m west	8.0	36.5	3.0
1,500 m west	3.5	24	2.0

* Mean lead concentration in soil and grass at 10, 15, and 20 m from motorway edge.

have occurred. In addition, new spring growth must have a very low in-plant concentration to account for the total summer lead level in pasture being less than the predicted internal concentration during winter.

Table 5-6 shows the predicted internal plant lead concentration as a percentage of the total grass lead content during winter. The results show that the deposition of airborne lead was a more important source of this metal to the pasture than uptake from the soil. In general, lead uptake from the soil accounted for between 20 and 40 per cent of the total lead content of the grass leaves during the winter period. At the 10 - 20 metres west site, uptake from the soil accounts for less than 12 per cent of the total. This is probably the result of the relatively low lead content of the surface soil compared with the deposition rate to moss bags in this period. The results also show that atmospheric contamination is the most important source of lead to pasture at the 1,500 metres west site, which has been shown not to be directly influenced by lead emissions from the M6 motorway.

The Summer Months

In contrast to the winter period, a poor correlation was found between the deposition of lead to moss bags and pasture lead concentration during the summer months. Figure 5-2 shows the scatter of points found for the 10 - 20 metres east site. Similar findings were obtained by Ratcliffe (1975) in an investigation of lead levels around a battery factory during June to August 1973.

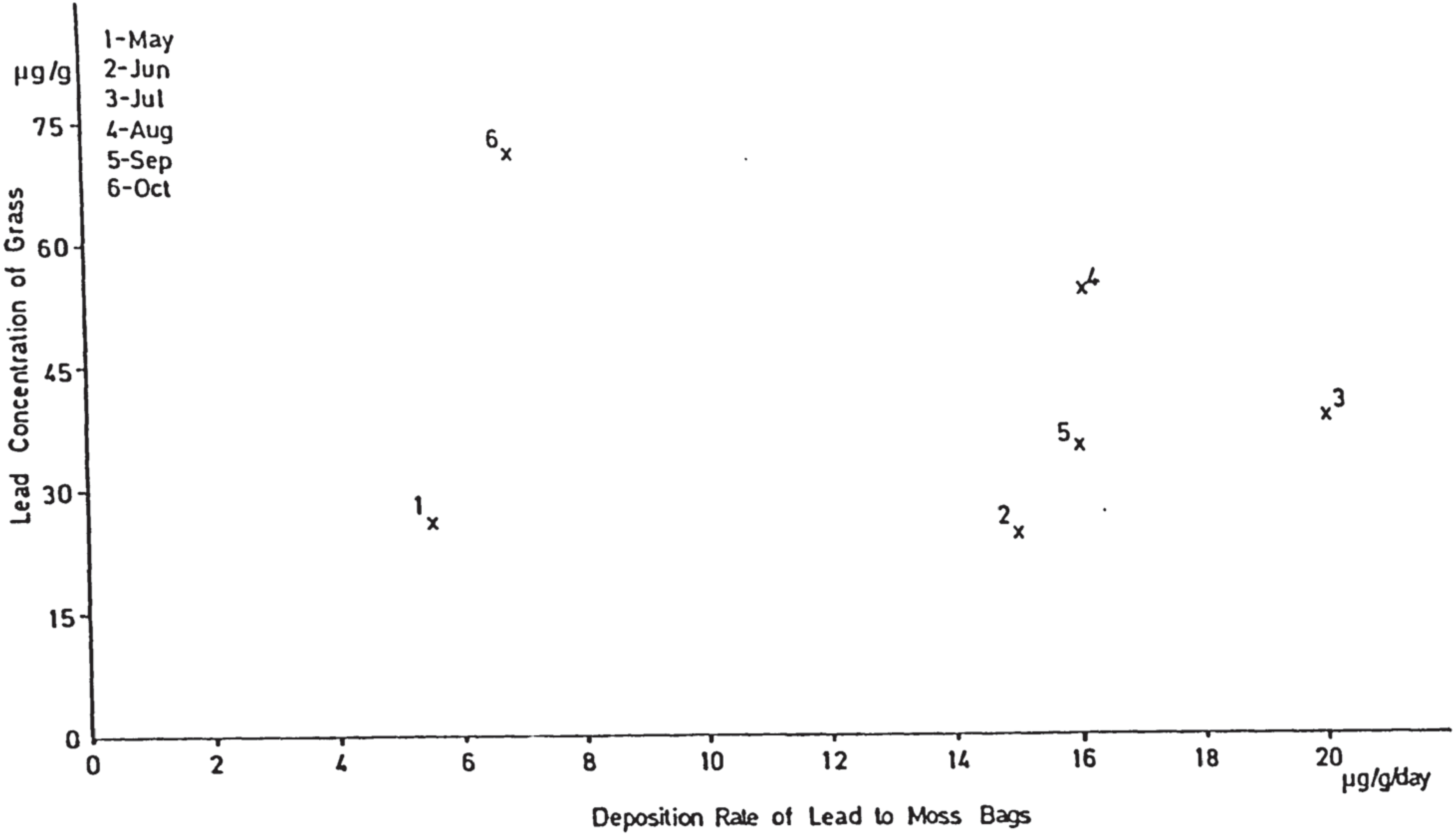
The poor correlation of data in summer is probably the result of the rapid and variable growth of the grass which, at the study site, is also subject to grazing and cutting. This means that the efficiency of deposition of airborne particulates to pasture may vary

TABLE 5-6

Predicted Internal Lead Concentration of Grass Shoots in Winter As a Percentage of Total Grass Lead

Site : Distance from motorway edge (metres)	Range of Grass Lead Concentration found in winter (µg/g)	Predicted Internal Lead Content of Shoots as Percentage of Total Grass Lead
450 m east	16 - 28	41 - 24
200 m east	26 - 58	37 - 17
10-20 m east	100 - 216	44 - 20
10-20 m west	42 - 138	12 - 4
100 m west	14 - 45	55 - 17
1,500 m west	11 - 27	31 - 13

FIGURE 5-2 The Relationship between Grass Lead Concentration and Deposition Rate of Lead to Moss Bags at the 10-20m East Site during the Summer of 1978



during the summer, and thus it will not remain in proportion to the deposition rate of lead to the unchanging moss surface. In addition, the rapid growth and senescence of grass foliage, and the removal of herbage by animals, do result in changes in the lead content of pasture by processes which do not act upon the moss bag. Therefore, no linear correlation exists between deposition to moss bags and lead content of grass which can be used to predict the amount of lead in summer pasture. However, the following evidence would suggest that, except within fifty metres of the road edge, most of the lead associated with the grass may result from non-atmospheric sources during the summer:

i) Laboratory growth experiments described earlier in this thesis (Section 5.4) and by other workers, have shown that some translocation of lead from roots to shoots can occur in actively growing grass. The experiment using soil from the background site, in which ryegrass was grown, found that translocated lead may result in a pasture lead concentration of up to 3.5 μg/g. This may be compared with grass lead levels at the background site during the summer of 1978 which ranged from 2 to 4 μg/g.

ii) Levels of lead deposition to moss bags are low during the summer. In addition, for several months, no significant increase in the lead content of moss bags was measured at the background site following exposure for periods of four to five weeks.

A further factor which suggests that aerial deposition does not have a major effect on the grass lead content at background sites is that of surface contamination by soil particles. As the grass lead concentration during summer is less than that of the topsoil, any soil particles which occur on the leaf surface as a result of rain splash or contact between the grass and the ground may well make a significant contribution to the lead content of the pasture. In contrast, the elevated lead levels occurring in summer pasture and within fifty metres of the motorway almost certainly result from airborne deposition of vehicular lead. This is because soil lead levels in this area are not greatly enhanced and the growth experiments described previously have shown that any increase in the lead content of the soil has relatively little effect upon the amount of lead translocated to grass shoots. In addition, moss bags have found lead deposition rates near the motorway to be considerably greater than at the background site.

Conclusion

During the winter months, a linear relationship exists between the deposition rate of lead to moss bags and the lead content of the rye-grass pasture beneath the bags. This is a result of the similar collection and retention characteristics of the moss and grass for airborne lead. The correlation can be used to distinguish between the contribution that atmospheric deposition and the uptake of soil lead via plant roots makes to the total lead content of pasture grass. In general, 60 to 80 per cent of the lead associated with the ryegrass foliage in winter months results from the deposition of airborne particulates.

During the summer, no clear relationship between the lead deposition rate and grass lead concentration was apparent. In this period, uptake of lead from the soil via plant roots, and the presence of soil particles on leaf surfaces, accounts for the majority of the lead associated with the pasture. However, within fifty metres of the motorway, deposition of airborne lead remains the major source of lead to pasture grass in all months of the year.

b) A Comparison of the Zinc Concentration of Pasture and Deposition Rate to Moss Bags

Both deposition rate to moss bags and grass zinc concentration were highest during the winter months at the M6 study site. This suggests that airborne material may contribute significantly to the grass zinc content. The relationship between grass zinc concentration and deposition rate is shown graphically for winter (Figure 5-3) and summer months (Figure 5-4) at the site 200 metres east of the motorway. Clearly, there is a poor linear relationship between the data in winter compared with that for lead, and the data points for summer months are very scattered.

Table 5-7 shows the correlation coefficients obtained for the relationship between deposition rates and zinc concentration of pasture grass during December to April. The low values of "r", which range from 0.5 to 0.8, are probably the result of a large fraction of the total grass zinc content occurring in plant tissues, rather than as external contamination. In addition, it is possible that this internal zinc concentration changes during the winter months. Zinc is, therefore, an interesting comparison with lead because the two elements differ in the relative importance of aerial deposition as a source of the trace metal to the pasture.

A better linear relationship was found for the zinc data in winter than summer, a finding suggesting that surface contamination by zinc is of greater importance during the non-growing season. The available data does not allow an accurate assessment of possible variation in zinc uptake from the soil during the year. A predicted value of tissue zinc concentration was obtained using the data with the

FIGURE 5-3 The Relationship between Grass Zinc Concentration and Deposition Rate to Moss Bags 200m East of the M6 Motorway during the Winter of 1977/8

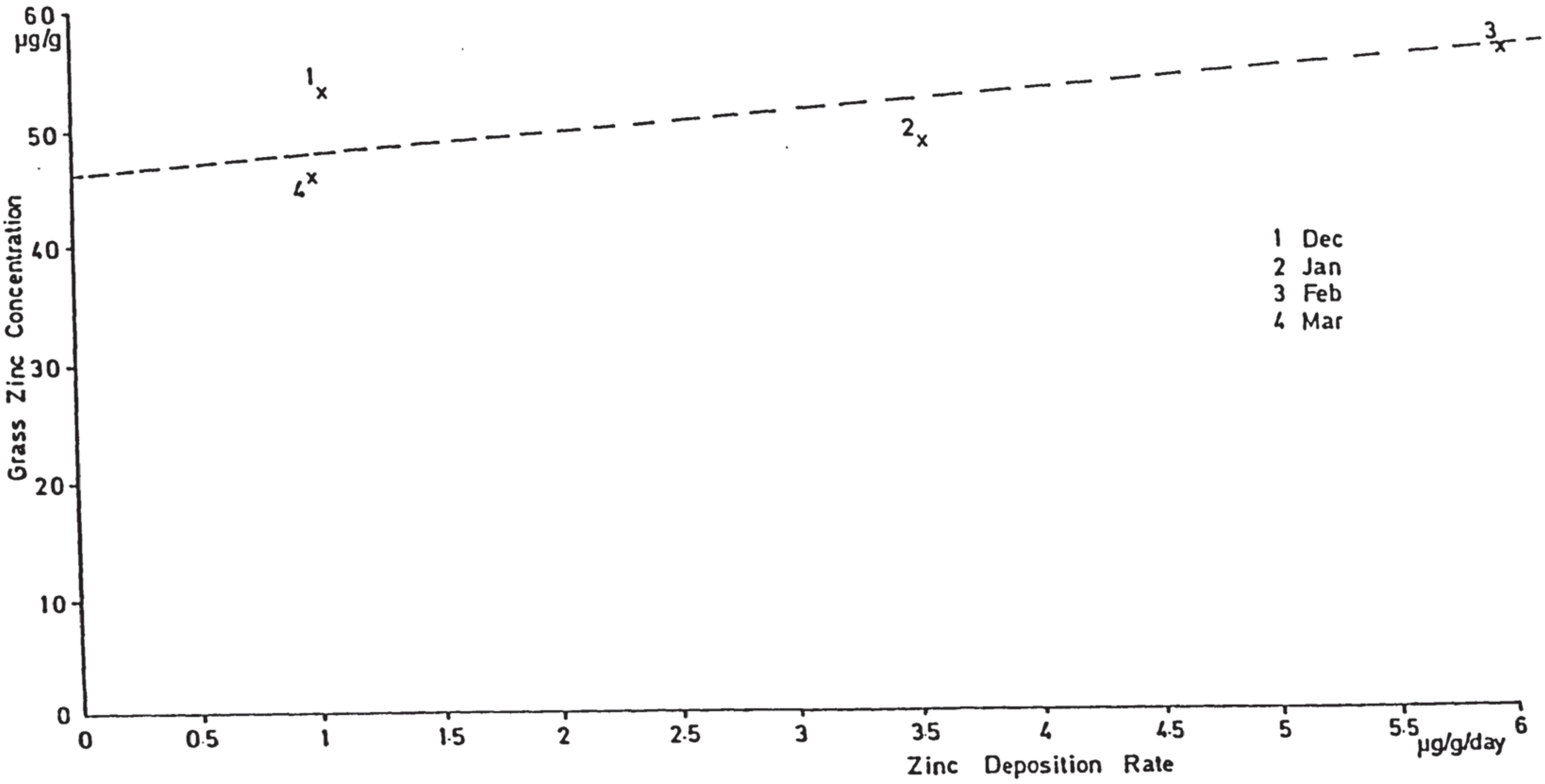


FIGURE 5-4 The Relationship between the Zinc Concentration of Grass and the Deposition Rate of Zinc 200m East of the M6 in Summer 1978

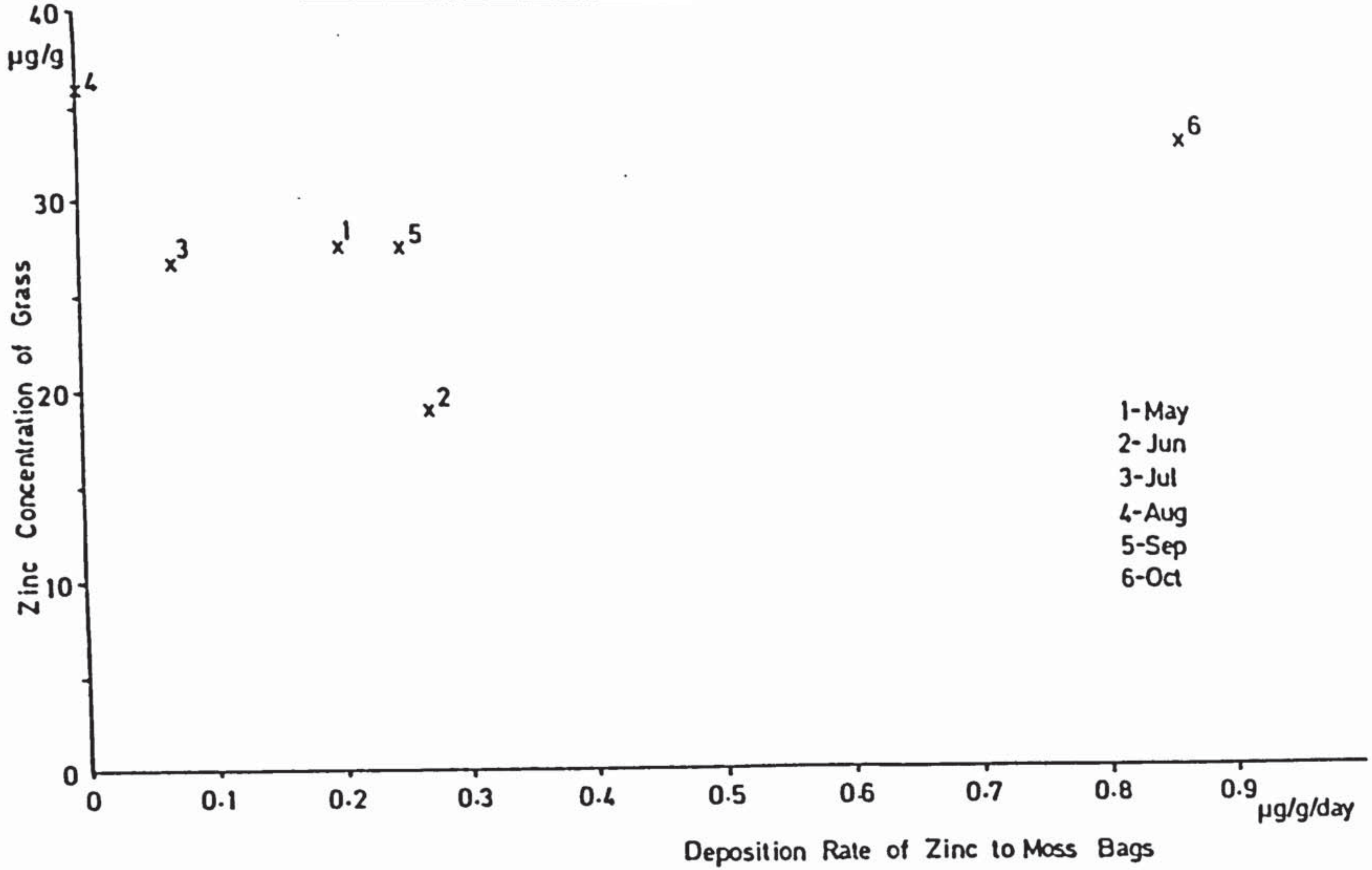


TABLE 5-7

The Correlation Coefficient of the Relationship Between Grass Zinc Concentration and Deposition Rate During Winter

Site : distance from M6 in metres	n (number of samples)	Correlation Coefficient	Significance
450 m east	4	0.81	0.1
200 m east	4	0.64	NS
10 - 20 m east	4	0.67	NS
10 - 20 m west	4	0.51	NS
100 m west	4	0.68	NS
1,500 m west	4	0.73	NS

NS = Not Significant

best linear relationship (450 metres east site), and this gave a value of 27 μg/g during winter months which may be compared with summer pasture levels of 20 - 35 μg/g. This, therefore, suggests that higher winter deposition probably accounts for the seasonal change in grass zinc content found in the study area, and that there is little seasonal change in the amount of zinc uptake per unit dry weight of above-ground biomass.

c) An Investigation of Soil Factors Influencing Lead Uptake by Pasture Grass

The humus content and pH of the soil have been shown to influence the availability of lead to plants and hence uptake via the root system (Zimdahl, 1976). Therefore, to investigate the possibility that these factors may be responsible for a seasonal variation in lead uptake, the organic carbon content and pH, together with the soil moisture content, were determined for samples of topsoil taken monthly at the study site from October 1977 to October 1978. The method and detailed results of this study are given in Appendix 3, but show no detectable change in these physical soil factors that can be related to the variation in the lead concentration of pasture grass. It may be concluded, therefore, that a seasonal change in the amount of lead uptake by the plant is not the result of the influence of soil pH and organic carbon content. In addition, a slight elevation in soil pH occurred near the motorway, as shown by Table 5-8, which may tend to reduce the availability of lead to plants in this most contaminated area. Such a finding is, however, not borne out by the predicted uptake of lead via plant roots at the 10 metres east site during the winter months.

TABLE 5-8

Variation in pH of Surface Soil with Distance

from the M6 Motorway in December 1977

Distance from motorway fence (metres)	pH of Surface Soil	
	West of motorway	East of motorway
0	7.6	7.4
5	8.5	7.4
10	7.2	7.4
20	7.0	7.1
50	6.6	6.9
100	5.5	6.3
450	-	7.4
1,500	6.9	-

5.7 CONCLUSION

Most of the lead associated with pasture grass which is within fifty metres of the motorway edge occurs as a surface deposit. During the winter months, aerial deposition also accounts for the majority of lead associated with pasture, even at sites not directly influenced by vehicular lead emissions from the motorway. However, at these background sites, uptake from the soil via the root system, and plant surface contamination by soil particles, are probably more important sources of lead for summer pasture than airborne particulates.

It appears that the lead content of the above-ground portion of the grass sward which results from lead uptake from the soil, increases in the autumn months. This internal plant lead remains at this higher level during winter and declines with the appearance of new growth in spring. In contrast to lead, the majority of the zinc associated with the pasture is the result of uptake from the soil during all months of the year.

PART 2 FACTORS PRODUCING A SEASONAL CHANGE IN THE LEAD CONTENT OF PASTURE GRASS

5.8 THE FACTORS TO BE CONSIDERED

Investigations at the study site have shown that the following two factors do act to produce a ten-fold seasonal change in pasture lead content:

i) A variation in the rate of lead deposition to vegetation. This is a result of meteorological conditions which produce a greater rate of deposition of atmospheric lead to plant surfaces during winter.

ii) An apparent increased uptake of lead from the soil during the winter months which may be due to a slower rate of biomass production. This is, however, of minor significance as the majority of lead associated with the pasture during winter is present as a surface deposit.

There are also a number of other possible mechanisms producing a seasonal change in lead concentration. Rains (1971) suggested that the growth cycle of the plant resulted in seasonal variation in the lead content of wild oats (Avena fatua) in the vicinity of a lead smelter. He proposed that lead built up on leaf surfaces during winter months and this was not removed by rainfall. In spring and midsummer, new growth resulted in a dilution effect of the deposited lead and thus lead levels decline in spring, remain low during summer, and increase again as growth slows in the autumn. A short term study of lead contamination of three crops adjacent to a road in Scotland found a change in lead levels during June to August (Maclean and Shields, 1977). These authors suggest that the seasonal changes in lead levels are related to differences in plant growth rate, but appear not to have assessed the possible influence of changes in wind speed and direction, traffic flow and rainfall.

A further factor which may act to produce higher concentrations of lead in winter is that of a decrease in the dry weight per unit area, due to the senescence of leaves in winter. This possibility was investigated by Mitchell and Reith (1966), and Rains (1975), but found not to be significant.

After spraying solutions of ^{85}Sr, ^{51}Cr and ^{210}Pb on to grassland, Chadwick and Chamberlain (1970) found a greater retention of deposited radionuclides by grass during winter than summer months. In a subsequent review paper, Chamberlain (1970) proposed that the production and loss of crystalline wax deposits by the growing plant may account for the more rapid decline in the level of surface contamination during the summer months. In winter, wax production ceases and thus any particles present on the cuticle of the leaf will not be lost as readily as when wax crystals are shed during summer. Chamberlain's study found that the difference in retention of radionuclides between winter and summer could not be accounted for by variation in the amount of rainfall.

A number of short term studies have suggested that rainfall may influence the level of lead contamination of roadside vegetation. Chamberlain et al. (1979) and Davies and Holmes (1972) report higher lead levels in roadside grass during dry periods in the UK, compared with samples taken during wet weather. However, these studies have not considered the influence of other meteorological and growth factors upon grass lead concentration in any detail, and rainfall was found not to account for the seasonal change in lead levels found by Rains (1971).

In summary, therefore, in addition to changes in the rate of deposition of atmospheric lead and uptake from the soil, the following factors must be considered as possible mechanisms of seasonal change in pasture lead concentration:

A. The grazing and cutting of pasture;

B. The removal of surface contamination by rainfall;

C. Changes in the plant surface which may influence the retention of lead particulates;

D. Growth factors.

These factors will now be considered in more detail.

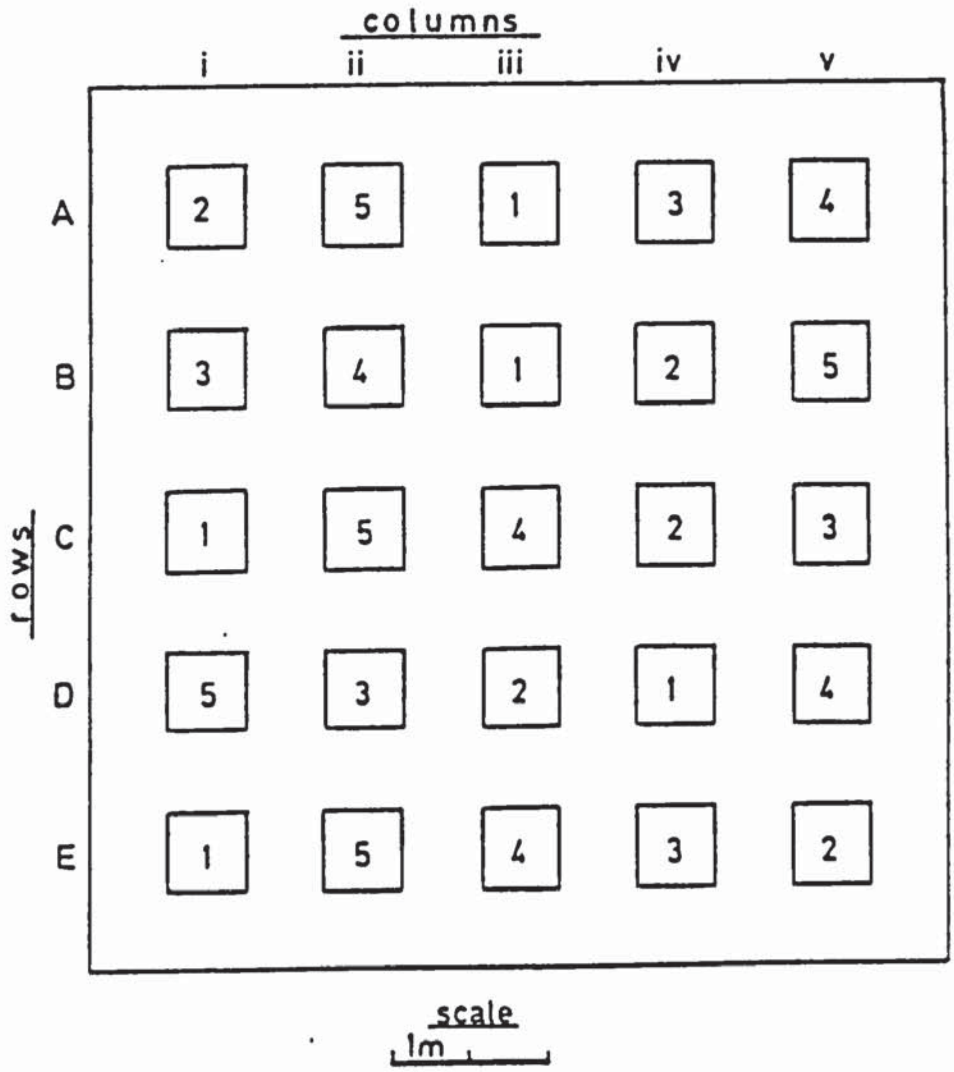
5.9 GRAZING AND CUTTING

The removal of herbage by grazing and cutting appears not to be an important mechanism of seasonal change in the lead concentration of pasture. This is because a similar change in lead levels occurred in both fields which were grazed in all months of the year and those utilised only during the summer. However, as grazing pressure may have differed significantly between fields and with time, a field study was undertaken to investigate the seasonal variation in the lead content of pasture which was protected from grazing. In addition, the lead content of the regrowth of grass which had been defoliated was compared with uncut pasture in the summer months.

Method

In the autumn of 1978 a 15 x 15 metre area of perennial ryegrass pasture was enclosed in a barbed-wire fence in a field 1,000 metres west of the M6 motorway (see Figure 1-2). The experimental area was then divided as shown by Figure 5-5 in order to provide 25 plots of one metre square area with spaces for access between. The five plots

FIGURE 5-5 Diagram of Field Plot Investigation of the Effect of Growth and Grazing upon Pasture Lead Content



Date of Harvest of plots (1979/80)

1 March
2 June
3 July
4 November
5 February

in each row were then assigned at random to one of five dates for harvest of the grass. The first date of harvest was in March 1979 and the last in December 1979. At each harvest, the total dry weight of the plot was measured and the lead concentration of a sub-sample of the grass determined.

At the third harvest in July 1979, the plots which had been cut in two previous harvests were again investigated to determine the influence of the earlier defoliation on the lead concentration. Unfortunately, farm animals gained access to the study area in December and the relatively low yield at Harvest 4 suggests that the plots may have been interfered with prior to this date. However, the protective fence remained secure at least until after Harvest 3 had been taken.

Results

Table 5-9a shows the mean lead concentration and dry matter yield of the five plots cut at each harvest. Results show that a rapid decline in lead concentration occurs in the ungrazed pasture during the spring months, a finding repeated in the grazed field areas.

The lead content of the uncut grass remains low during the summer and Harvest 4 shows the autumn rise in lead concentration.

Table 5-9b compares the lead concentration of grass which remained uncut until Harvest 3 with that of the regrowth following defoliation in June and March. Results show no significant difference between the regrowth and the uncut pasture in the summer of 1979.

Conclusion

A seasonal variation in the lead content of grass occurs in pasture which is not grazed by livestock. Simulated grazing during the summer did not significantly affect the lead concentration of the grass.

TABLE 5-9

Results of the Field Plot Experiment at a Background Site

a) Change in Yield and Lead Content of Ungrazed Pasture During 1979

Factor	Harvest 1 (30/3/79)	Harvest 2 (5/6/79)	Harvest 3 (18/7/79)	Harvest 4 (3/11/79)	Harvest 5 (21/12/79)
Yield (g dry wt.)	28 (13)	273 (10)	1,050 (148)	540 (76)	Grazed -
Lead Concentration (μg/g)	37.5 (8.5)	0.9 (0.25)	1.4 (0.3)	11.5 (0.6)	16 (2.1)

( ) = Standard Deviation of 5 values.

b) Effect of Simulated Grazing on the Lead Content of Pasture

Factor	Uncut until 18/7/79	Regrowth since 30/3/79 until 18/7/79	Regrowth since 5/6/79 until 18/7/79
Yield (g)	1,050 (148)	854 (208)	648 (244)
Lead Concentration (μg/g)	1.4 (0.3)	1.6 (0.4)	1.4 (0.3)

( ) = Standard Deviation of 5 values.

5.10 REMOVAL OF DEPOSITED LEAD BY RAINFALL

An investigation of the effectiveness of distilled water and rainwater at removing lead from pasture grass has shown that only 4 - 40 per cent of the total lead was removed by a quite vigorous washing procedure (Table 5-1). However, this study may be criticised because the washing agent remains in contact with the grass and the procedure does not simulate rainfall. A further washing experiment was carried out using grass collected from the M6 study area during January and August 1978. This involved washing grass under a fast running tap for a period of two minutes followed by a rinse in distilled water. The results show that no significant losses of lead occurred (Table 5-10). It suggests, therefore, that lead is not readily removed by running water striking the grass surface. This finding is, perhaps, in contrast to that of Little and Wiffen (1977) who used radioactively labelled petrol engine exhaust (Pb^{203}) to contaminate grass in a wind tunnel. Subsequently, simulated rainfall was found to remove 50 per cent of the deposited lead. Possibly, in their study, the plant surface was saturated by lead particulates so that a large part of the removed lead was not in direct contact with the leaf, and was therefore less strongly retained.

The data concerning pasture lead concentration during 1977 to 1979 at the study site would also suggest that rainfall is not a significant factor producing seasonal change in the level of grass contamination. This is because:

a) the seasonal variation in lead concentration of pasture did not correlate with the quantity and pH of rainfall between any two sampling periods (Crump et al. 1980);

b) the smoothness of the change in grass lead concentration with

TABLE 5-10

The Effect of a Running Water Wash

Upon the Lead Content of Grass

Sample Site (Distance from road edge in metres)		Lead Concentration of Grass (μg/g)			
		January 1978		August 1978	
		Unwashed	Washed	Unwashed	Washed
East	5	-	-	73	73
	10	280	230	41	47
	20	170	160	-	-
	30	135	155	30	31
	100	43	42	8	8
	150	59	67	-	-
	450	-	-	9	8
West	5	-	-	38	39
	10	95	90	-	-
	20	55	30	-	-
	30	40	40	-	-
	100	27	27	4.5	3.5
	1,500	26	28	3.5	3.5

(-) indicates no samples analysed.

time shown in Figure 4-4 is itself indicative that factors other than the variable occurrence of rainfall determine the pasture lead content.

5.11 CHANGES IN THE PLANT SURFACE

Electron microscope investigations of the surface features of three grass species were undertaken and the results have been discussed previously in Section 5-3. This study also involved the examination of grasses both during the summer and winter months, and no difference in the surface features of the leaves in the two periods was apparent. This similarity of surface features during winter and summer suggests that the deposition velocity of particles to grasses does not vary seasonally as a result of changes in the micro-roughness of the grass leaf. It is not possible to assess from the electron microscope study whether the peeling of wax deposits on the leaf surface is an important mechanism influencing the presence of lead particulates on the plant surface. However, the presence of a waxy cuticle on the grass leaves suggests that this phenomenon could occur. Plates 7 and 8 compare the surface features of a green and senescent leaf of Holcus lanatus. Clearly, the contracted brown leaf presents a very different surface for deposition and may influence wetability of the leaf. Little and Wiffen (1977) found that the influence of leaf senescence upon the deposition velocity of particulates was species dependent for a number of tree leaves. Thus, it is possible that if the plant canopy has a greater proportion of senescent leaves during winter, the deposition efficiency of particulates to the grassland will differ from summer months. However, no data is available to quantify the effect of senescence upon the efficiency of particle catchment. In the field, this may not be of great significance as organic matter is quite rapidly broken down and thus, leaves can only remain as part of the

PLATE 7

Electron Microscope Picture of the Surface of a Green Leaf of Holcus lanatus

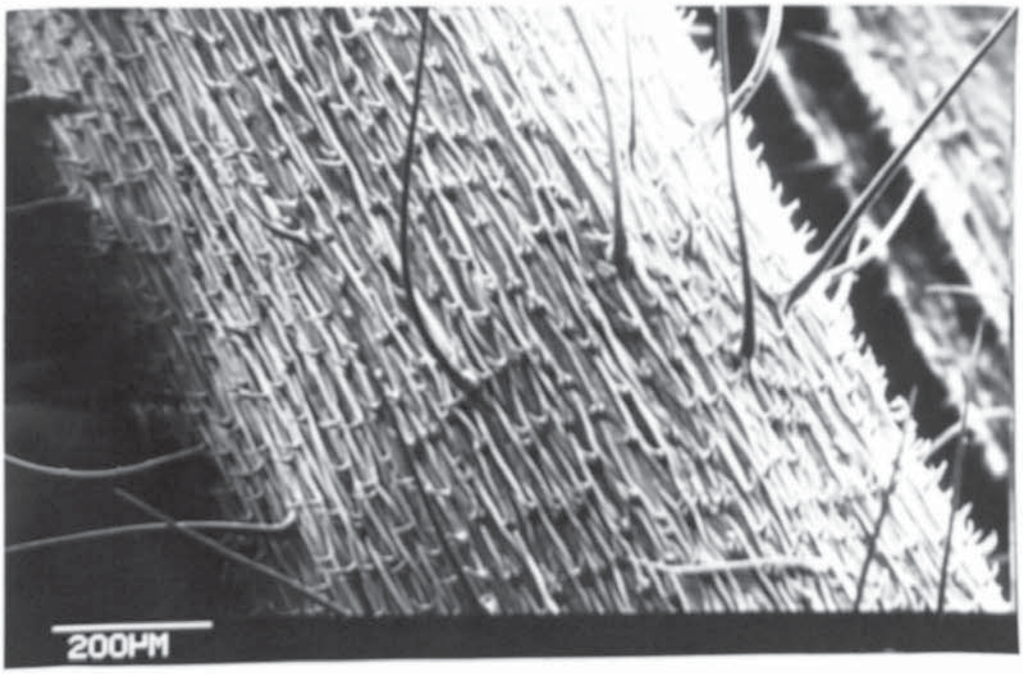
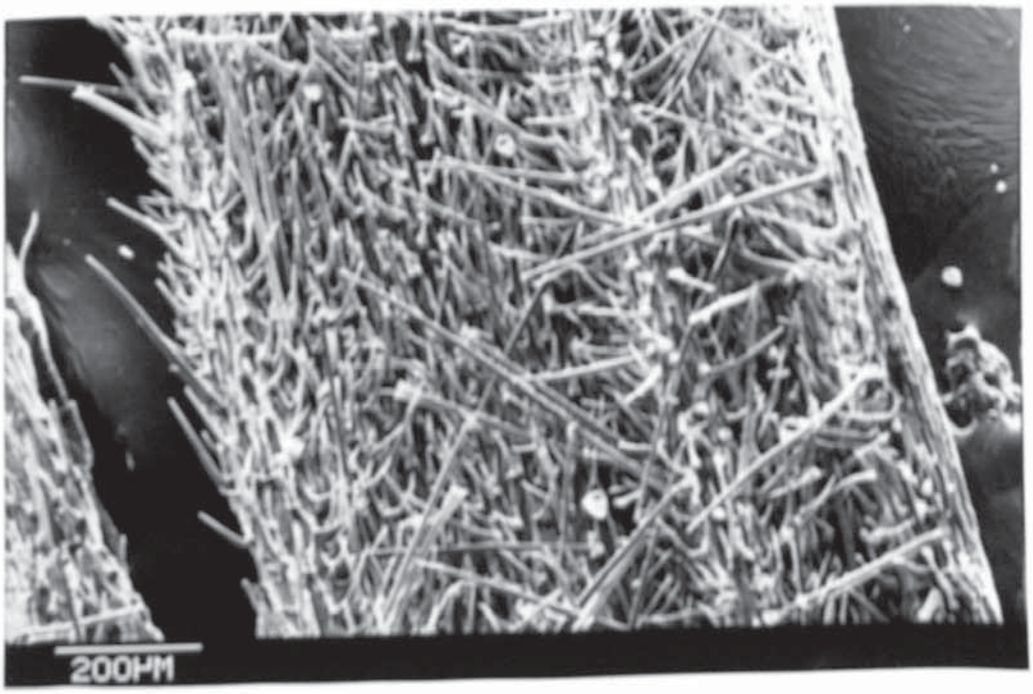


PLATE 8

Electron Microscope Picture of the Surface of a Senescent Leaf of Holcus lanatus



plant stand for a relatively short period of time (Hunt, 1965).

5.12 GROWTH FACTORS

A. The Influence of a Change in Biomass

Although growth occurs continually within the canopy of a perennial ryegrass pasture, due to the production and senescence of leaves even in midwinter, it is only during the warmer months that a significant change in the biomass per unit of time occurs (Thomas and Norris, 1977). Anslow and Green (1967) measured the annual dry matter yield from a number of cultivated grass species in the UK. Maximum production occurred usually within the first two weeks of May, and there was little species difference in the date of commencement of the spring growth. This period of rapid growth was associated with an increasing size of tillers and a greater erectness of the plants. These workers suggest that, during the summer, it is reasonable to assume that dry matter accumulates at a constant rate, if cut every month and well-fertilised, but at a rate perhaps 40 per cent lower than the spring peak. In mid-August, the rate of dry matter production begins to decline further, until there is no net yield from the pasture in late November or early December.

There are two distinct effects that a change in biomass may have upon the pasture lead concentration. Firstly, the change in height and density of the vegetation may influence the deposition per unit surface area of leaf and secondly, new growth containing a low level of lead will give rise to a lower average lead concentration. These effects would be greatest in spring when a rapid burst in production of dry matter occurs. The significance of these factors will now be discussed in relation to the M6 study site.

i) Change in height and density of vegetation

It is difficult to assess the effect that changes in vegetation structure have upon the deposition rate of airborne lead. A greater production of dry matter would dilute a constant fallout of lead to the ground surface. However, the taller and denser pasture also has a greater surface area for impaction and diffusion of particles. Therefore, if particulates are available for deposition, the effect of growth dilution will be offset by the deposition per unit surface area remaining constant.

A number of workers have shown that wind speed decreases from the top of the plant canopy to the soil (Monteith, 1966). This effect presumably increases with height and thickness of the canopy. As wind speed is an important factor determining dry deposition of small particulates, perhaps surfaces near the soil and in dense vegetation, have a lower deposition rate than more exposed leaves. This decrease in deposition velocity due to a reduced wind speed will be partially offset by a greater sedimentation of larger particles.

Investigations at the M6 study site found that taller and denser plots of grass did not have a significantly different lead concentration from swards with relatively poor growth in a field experiment during the summer of 1979. This experiment compared the yield and lead concentration of one-metre-square plots of ryegrass near the motorway which had been fertilised with nitrogen at different rates to produce a variation in growth rate. Three fertiliser treatments were used with twelve replicates of each and four cuts of the grass were taken over the growing season. The experiment and results of the statistical analysis of the data is described more fully in Appendix 4.

In conclusion, therefore, it would appear that the change in structure of the plant stand with season is not in itself an important factor producing a change in pasture lead concentration.

ii) Change in yield of pasture

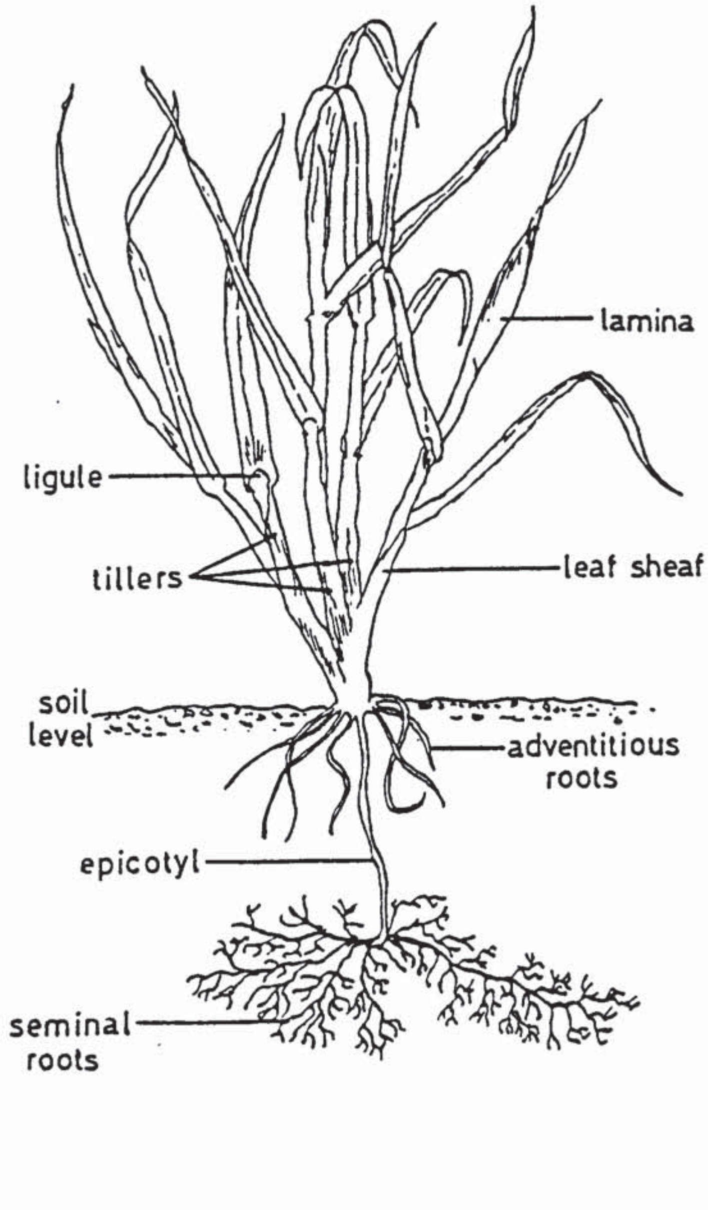
If the peak concentration of lead in pasture during winter at the M6 study site resulted from a gradual build-up of lead on the vegetation, it would be expected that the new growth in spring would produce a reduction in concentration proportional to the change in the dry matter yield. This presumes that the new growth has a relatively low lead concentration, a finding by Rains (1975) in greenhouse studies using wild oats. At the study site, this is also likely to be the case due to the low lead deposition rates found during the spring months. However, the field trial investigation of ungrazed pasture 1,000 metres west of the M6 motorway showed that lead concentration declined by a factor of forty, while yield increased by only ten-fold (Table 5-9).

This suggests, therefore, that the spring decline in grass lead concentration is only partially the result of growth dilution. Other mechanisms must also be acting which result in a loss of deposited lead from the plant canopy. One possible factor may be the death and decay of grass leaves and tillers.

B. The Influence of the Life Cycle of Individual Leaves and Tillers Upon Pasture Lead Concentration

The manner in which grass grows has been described by Jewiss (1966) and Langer (1972). Figure 5-6 shows the structure of the vegetative grass plant which consists of a collection of above ground tillers or shoots and the root system. Each tiller is composed of a number of foliar organs made up of leaf blades (laminae) and leaf sheaths.

FIGURE 5-6 Diagram of a Grass Plant



The stem of the plant is very contracted and only during flowering does it elongate to produce a flowering culm.

New leaves originate from a region of the stem apex where cell division and enlargement produce a lamina which moves up inside the sheaths of older leaves. The region of cell division is restricted to the leaf base and thus, the tip of the lamina represents the oldest portion of the leaf and is the first part to senesce. The rate at which the new leaves become visible externally varies with species and with season. This rate of appearance is greatest for periods of favourable growth in spring and summer, and least during cold winter months. Langer (1972) suggests that, in general, a new leaf appears every five to six days in summer, but may take twenty or more days in winter. In addition, for a given set of environmental conditions, the rate of leaf appearance remains fairly constant.

Alberda (1966) found that under ideal growth conditions tillers of Lolium perenne supported a maximum of four healthy leaves. When the fifth leaf emerged the first yellowed and died, so that the rate of leaf appearance was approximately equal to that of leaf death. In a field study of the growth of perennial ryegrass in Wales, Thomas and Norris (1977) found a maximum of 3 to 3.2 leaves per tiller. The rate of leaf appearance was quite closely related to temperature and was greatest during summer (approximately one new leaf/tiller/10 days) and least in winter (one new leaf/tiller/50 days). Even at upland sites in the coldest months of winter, these workers were able to measure a turnover of dry matter as the result of the appearance of new leaves and senescence of the oldest foliage. Hunt (1965) found that a new leaf appeared every 10 to 19 days on each tiller in an undefoliated stand of Italian ryegrass during winter in Wales. As there were approximately three living leaves per tiller, it is clear that about one-third of the plant canopy was replaced by this new growth every 16 to 19 days.

In addition to the turnover of organic material as a result of the growth and senescence of individual leaves, for perennial grasses, the tiller population itself may change markedly. An individual tiller has a restricted, but variable life span. Some tillers may appear and flower in one season, others flower in the year following their formation and a proportion remain vegetative, surviving for only a period of weeks or months. The production and subsequent development of the tillers will depend greatly upon environmental conditions. Thomas and Norris (1977) found that the rate of new tiller production was slow during winter, accelerated during April and May, and declined as the perennial ryegrass entered the heading phase. However, as individual tillers, in general, survive for several months, tillering will be of secondary importance to the process of leaf death and decay.

In conclusion, the individual grass plant and sward are in a dynamic condition. This results in a continual turnover of organic material in the plant canopy which occurs at a greater rate during summer than winter months. The average longevity of a grass leaf is, therefore, greater during winter than summer by a factor of perhaps five- to ten-fold. This value may vary considerably depending upon environmental conditions, but it is clear that winter leaves are exposed to airborne lead for a greater period than the summer growth. Thus, if lead builds up on the surface of the plant, and is not readily removed by leaching processes, the difference in the longevity of summer and winter leaves may be a significant mechanism producing the seasonal change in the lead content of the plant canopy. Evidence for a build-up of lead with leaf age in the pasture at the M6 study site comes from the following investigations:

i) The lead concentration of grass leaves of different age at the study site

Method

Grass samples were taken periodically from either side of the motorway (fields B and 1) and at the control site (C1) from autumn until early summer during 1979/80. In the laboratory, a sub-sample of the grass was separated into green and brown leaves. The brown, senescent leaves are older than the green leaves. Only the senescent leaves still attached to the tillers were investigated in order to allow the new growth to be compared with old leaves which were not yet influenced by decomposition. Leaves which had only partially senesced were discarded to ensure that the two classes of leaf were of a different age. The lead content of the whole pasture and that of the brown and green leaves was determined as described previously in Chapter 1.

Results and Discussion

Table 5-11 compares the lead content of green and brown leaves with that of the whole grass sample. Results show that the lead concentration of the brown leaves is very much higher than the newer green growth, and this is particularly so during the winter months. The lead content of the whole pasture sample has a value somewhere between that of green and brown leaves, reflecting the proportion of these two leaf classes or, in effect, the average age of the plant canopy.

To ensure that the differences in lead concentration were not due to the loss of dry material upon senescence, the length of ten brown and ten green leaves, taken from each site in February 1980, was measured. The individual leaves were then oven dried and the weight

TABLE 5-11

The Lead Content of Green and Brown Ryegrass Leaves

Site (Distance from road edge)	Date of Sampling	Lead Concentration (μg/g)		
		Whole Pasture	Green Leaves	Brown Leaves
1,500 m West	17 October 1979	3.5	1.5	4.0
	21 December 1979	16 (2)	6 (1.5)	18 (3)
	5 January 1980	14.5	1.5	22
	18 February 1980	20	2.0	38
	29 March 1980	13.5	5.0	39
	19 May 1980	7.0	2.0	11
10 m East	17 October 1979	75	19	124
	21 December 1979	164	30	184
	5 January 1980	174	17	281
	18 February 1980	254	16	266
	29 March 1980	132	29	330
	19 May 1980	21	4.0	39
10 m West	* 29 November 1979	48 (5)	12 (4)	70 (7)
	21 December 1979	27	11	70
	5 January 1980	30	3.5	93
	18 February 1980	34	6.0	107
	29 March 1980	27	13	109
	19 May 1980	15	5.0	23

( ) = standard deviation of 5 values.

* = sample from field C not B (Figure 1-3).

per unit length of lamina compared. In general, the dry weight of brown leaves was approximately 15 per cent lower than that of the green leaves. Thus, differences in dry weight between senescent leaves and new growth could not account for the three- to nearly twenty-fold difference in lead concentration. It would appear, therefore, that lead built up on the surface of grass leaves with increasing age. The possibility that the deposition rate of lead was greater to the senescent than the green foliage as a result of a change in the micro-roughness of the surface, may also be a minor contributing factor.

Clearly, the lead concentration of the pasture is dependent upon the mass of senescent leaves and the degree by which this is diluted by new green growth. The greater longevity of leaves, together with the higher lead deposition rate, results in the senescent foliage having a very much higher lead content in winter than in summer months. Green leaves show less variation from winter to summer as they have not been exposed to the atmosphere sufficiently to be severely contaminated by deposition. In addition, it is possible that the senescent leaves have a higher internal lead concentration because of the translocation of lead from the roots, as an increase in the lead concentration of foliage at senescence had been found by Guha and Mitchell (1966) for other plant species.

Conclusion

The lead content of older senescent leaves is considerably greater than that of green, newer growth. This suggests that lead builds up on the plant surface with time and is not removed by rainfall. In consequence, the seasonal variation in the average age of leaves in the plant canopy will tend to result in higher lead levels during winter than summer months.

ii) <u>The consequence of the linear relationship between deposition rate and grass lead concentration for the behaviour of lead in the plant canopy</u>

Moss bags, suspended above the pasture grass, were exchanged for fresh, uncontaminated moss every four to six weeks at the M6 study site. Therefore, if lead was building up on the surface of grass leaves over the winter period, a linear relationship between the deposition to moss bags and pasture lead concentration could not have occurred. This is because, during the winter, the grass lead content declined and increased simultaneously with changes in the lead deposition rate measured by the exposure of moss bags for periods of only one month. There must, therefore, be a mechanism by which lead is removed from the plant canopy so that the pasture lead concentration is determined by deposition over the exposure period and not by the lead content of the grass prior to this time.

Rainfall has already been demonstrated not to result in any significant loss of lead from the grass surface. Therefore, the most probable mechanism is the turnover of organic material in the plant canopy which results from the growth cycle of individual leaves and tillers. This process has been shown to occur even in the coldest winter periods in the UK, and the available information suggests that one to two months may represent the average longevity of a leaf in a perennial ryegrass pasture during winter. The occurrence of lead production and senescence will, however, be continuous rather than corresponding to the fixed periods of four to six weeks when moss bags were exchanged. Yet, because the deposited lead builds up on the surface of individual leaves, the greatest part of the lead content of pasture is associated with the oldest foliage.

The oldest canopy leaves sampled would have been undergoing development at the start of the moss bag exposure period. They have, therefore, been exposed to airborne lead for a similar period of time as the moss bags. Before the next set of moss bags are collected and the grass sampled, these senescent leaves, and the lead on their surfaces, will be lost from the plant canopy by decay processes. Hence the lead content of pasture is determined largely by senescent foliage which is of a similar age as the moss bag exposure period. In consequence, the study at the M6 site found a linear relationship between the lead deposition rate to moss bags and the pasture lead concentration over the winter months.

In summer months, the shorter longevity of grass leaves will result in the grass lead concentration being determined by the accumulated deposition over periods of only one or two weeks. Therefore, a linear relationship would not occur between deposition rate to moss bags and pasture lead concentration. In addition, growth dilution and grazing will further distort any relationship between lead content of moss bags and grass.

In conclusion, therefore, the occurrence of a linear relationship between lead deposition rate to moss bags and pasture lead concentration during the winter months suggests that a turnover of organic material, due to the growth cycle of individual leaves and tillers, does have an important influence upon pasture lead concentration. If individual leaves did not have such a very limited life span it would be expected that the lead content of the pasture grass would be greater for the same rate of deposition of airborne particulates. This is because lead appears to build upon the surface of the plant with time without significant losses due to rainfall or other weathering processes. Possibly the peeling of the waxy cuticle of the grass leaf has some influence upon the lead concentration of the summer pasture. However, this seems unlikely as this process must be of secondary importance to the effect of the rapid turnover of organic matter due to the death and decay of individual leaves and tillers.

5.13 THE RELATIVE IMPORTANCE OF PLANT GROWTH PROCESSES AND CHANGES IN DEPOSITION RATE AS A MECHANISM OF SEASONAL CHANGE IN PASTURE LEAD CONCENTRATION

Investigations at the study site have identified that variation in the rate of deposition of atmospheric lead and the life cycle of individual leaves are the two major factors producing a seasonal change in lead concentration. Uptake of lead from the soil via plant roots is a significant, but minor mechanism of seasonal change. Therefore, it remains to determine the relative importance of deposition rate and growth processes as factors determining the grass lead content.

If growth processes in the summer remained at the same rate as during winter months, it would be expected that a linear relationship would exist between the deposition rate to moss bags over a four week period, and the level of surface contamination of grass resulting from aerial deposition. Therefore, to investigate the effect of growth processes, the level of surface contamination resulting from airborne deposition during the summer months was predicted on the basis of the winter correlation between deposition rate and grass lead concentration. Figures 5-7 and 5-8 compare these predicted values for lead levels on the plant surface in all months of the year with the measured total lead content of the pasture at two sites in the study area. Near the motorway, the moss bag data predict that grass lead levels would increase during midsummer to a concentration approaching peak values in the previous winter. Presumably, the rapid growth and senescence of leaves

FIGURE 5-7 A Comparison of Measured Grass Lead Concentration with Predicted Levels of Plant Surface Contamination by Atmospheric Deposition 1,500m West of M6 during 1977/8

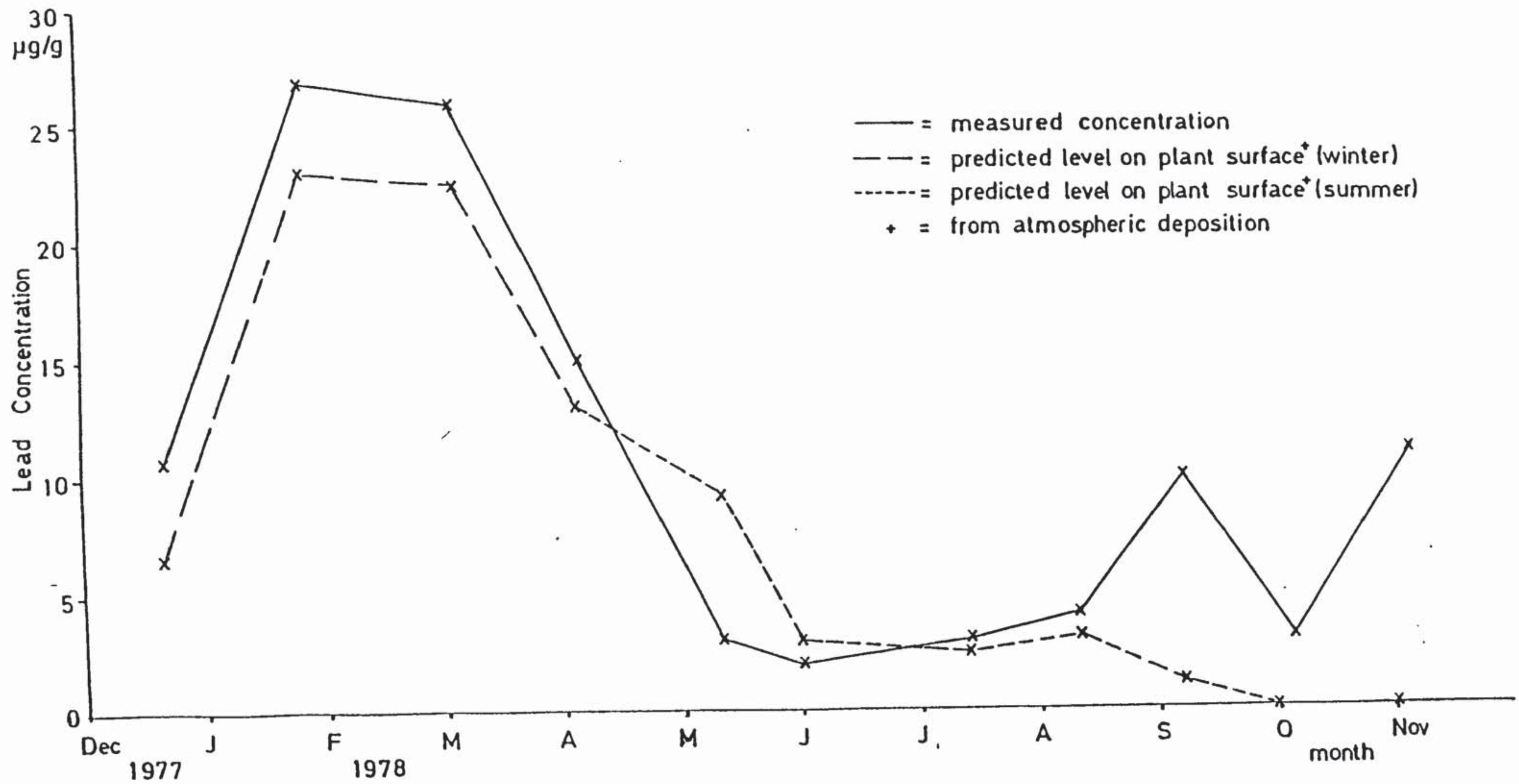
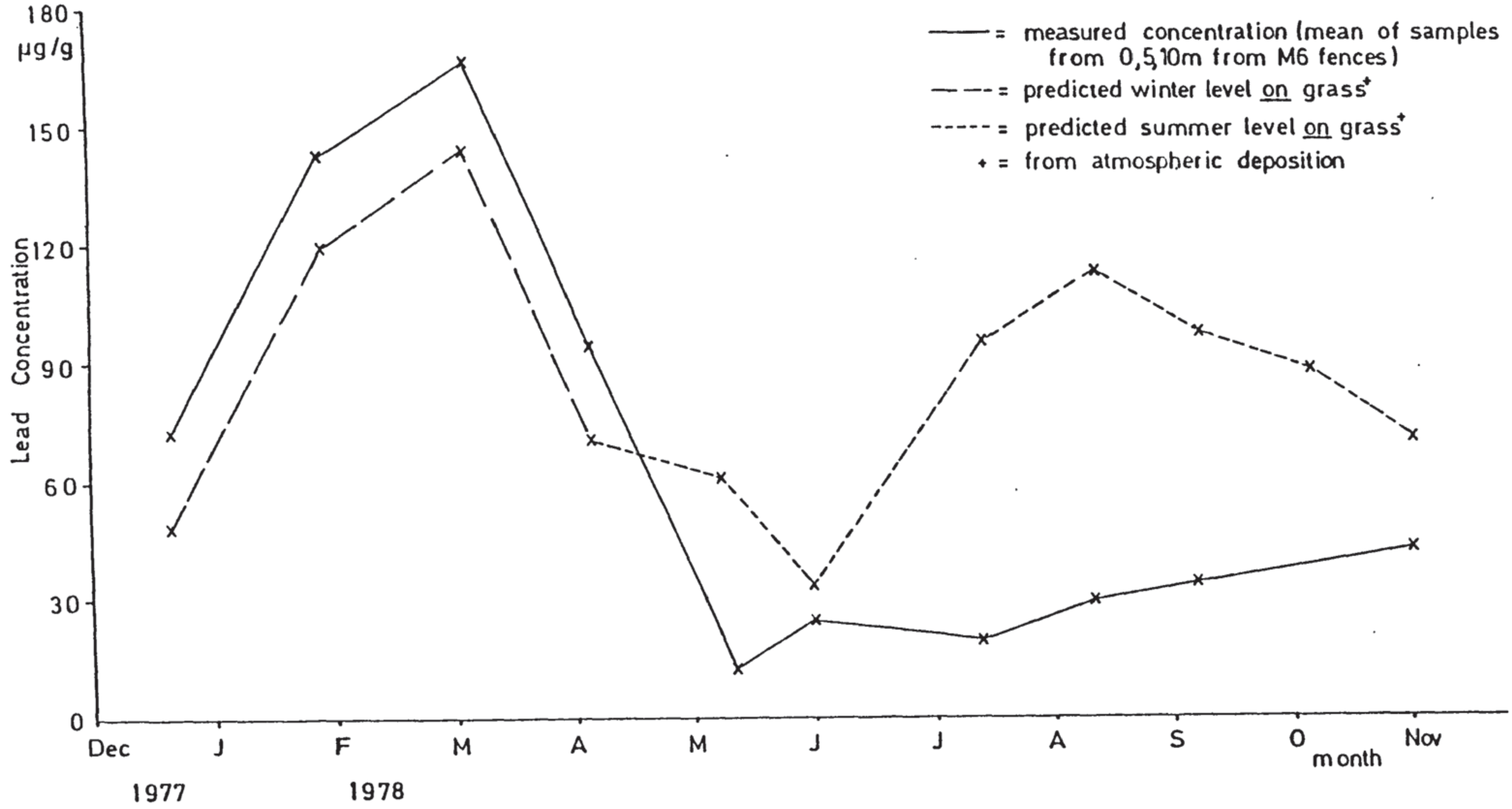


FIGURE 5-8 A Comparison of the Mean Lead Concentration 10-20m Either Side of the M6 with Predicted Levels of Plant Surface Contamination by Atmospheric Deposition



is the major factor resulting in summer grass lead levels remaining below 40 µg/g despite the increase in lead deposition rate produced by high traffic flow in July to September.

At the 1,500 metres west site, the predicted level of lead in pasture is less than the concentration measured during the summer months. Thus, presumably, the greater part of the lead associated with the grass resulted from uptake of available lead from the soil via plant roots, or is present as a surface deposit of soil particles which are on the grass as a result of rain-splash or contact of leaves with the ground. Therefore, growth processes would seem to have less effect on the lead concentration of the pasture where the deposition of airborne material is a minor source of lead for vegetation.

5.14 CONCLUSIONS

The seasonal change in grass lead concentration at sites in excess of 50 metres from the motorway is largely determined by the rate of deposition of atmospheric lead. Changes in the rate of growth of grass with season have little influence on lead levels at background sites.

Near the motorway, the rate of deposition of atmospheric lead shows less seasonal variation than at background sites. However, plant growth processes result in levels of lead in summer pasture remaining considerably less than during winter months. This effect would appear to be largely due to the rapid turnover of organic matter during the growing season which restricts the build-up of lead on individual leaves in the grass sward.

CHAPTER 6

THE SIGNIFICANCE OF ROADSIDE LEAD FOR FARM LIVESTOCK

6.1 INTRODUCTION

This chapter considers the possible effects that roadside lead may have upon the health of farm livestock and also that meat and milk products might become contaminated by lead from motor vehicles. Firstly, animal health effects are considered by reviewing the published data of other workers who have investigated the toxicity of lead for domestic animals. This information will then be used to assess the significance of estimated rates of lead intake for livestock which graze fields bordering the M6 motorway in Cheshire.

The second part of this chapter discusses the possible influence that roadside contamination may have on the lead content of meat and milk products. This section consists of a comparison of data obtained for the study area with the findings of other workers.

6.2 REVIEW OF PUBLISHED WORK ON HEALTH EFFECTS OF LEAD

A. The Occurrence of Lead Poisoning in Farm Livestock

Lead poisoning in farm livestock has been studied because of its economic importance. Osweiler et al. (1973) report that lead poisoning is the single, most common toxicosis in Iowa, USA. This acute toxicity is predominantly the result of cattle licking lead based paints, discarded batteries, used crank case oil and other materials containing very high concentrations of lead. Chronic lead poisoning of cattle, on the other hand, is comparatively rare.

Lead toxicity in horses is principally the result of cumulative poisoning over a number of months (Aronson 1971). This is because horses are more discriminating than cattle when foraging, but are more susceptible to elevated lead levels. Thus, it is not uncommon for the death of horses to occur after grazing the same fields as apparently unaffected cattle (Allen 1975; Egan and O'Cuill 1970).

Sheep would seem to be the more robust of the farm animals, having a greater tolerance to both single and cumulative doses of lead (Allcroft 1951). It would appear, therefore, that in a situation of environmental contamination by lead, such as in the vicinity of industry, mine workings and roadsides, the greatest hazards are the cumulative poisoning of horses, and the possible accumulation of lead in other animal tissues. Man being higher in the food chain than the domestic animals, may, therefore, consume such tissues which have an elevated lead content.

B. Lead Uptake by Animals

The farm animal may be exposed to lead from the air, soil, water and forage in its environment. Figures 6-1 demonstrates the modes of entry into an animal and the possible effects of the absorbed lead. Inorganic lead released from motor vehicles may enter the body by ingestion and inhalation. Absorption of inhaled lead is strongly dependent on the size of lead particulates, and it is the smallest fraction of particle sizes ($< 2\ \mu m$) which are both most readily deposited in the lungs and most prominent in the atmosphere near major roads (Chamberlain et al. 1978).

Lead may be ingested by livestock due to its presence in herbage, drinking water and other feeds. In addition, soil may be consumed along with forage and act as a significant source of lead (Harvey 1979; Colbourn 1976). However, of the total lead ingested, only 2 to 10 per cent is absorbed through the gut wall (Allcroft 1951; Dinius et al. 1973). The value probably varies depending upon factors such as age of the animal, nutrient balance and chemical form of the lead (Allcroft 1951).

C. Clinical Effects of Absorbed Lead

Studies concerning the health effects of absorbed lead fall into two main categories:

(i) the investigation of animals exposed to high levels of lead in the environment, and

(ii) laboratory experiments using feeding trials to observe animals which have ingested a known amount of lead.

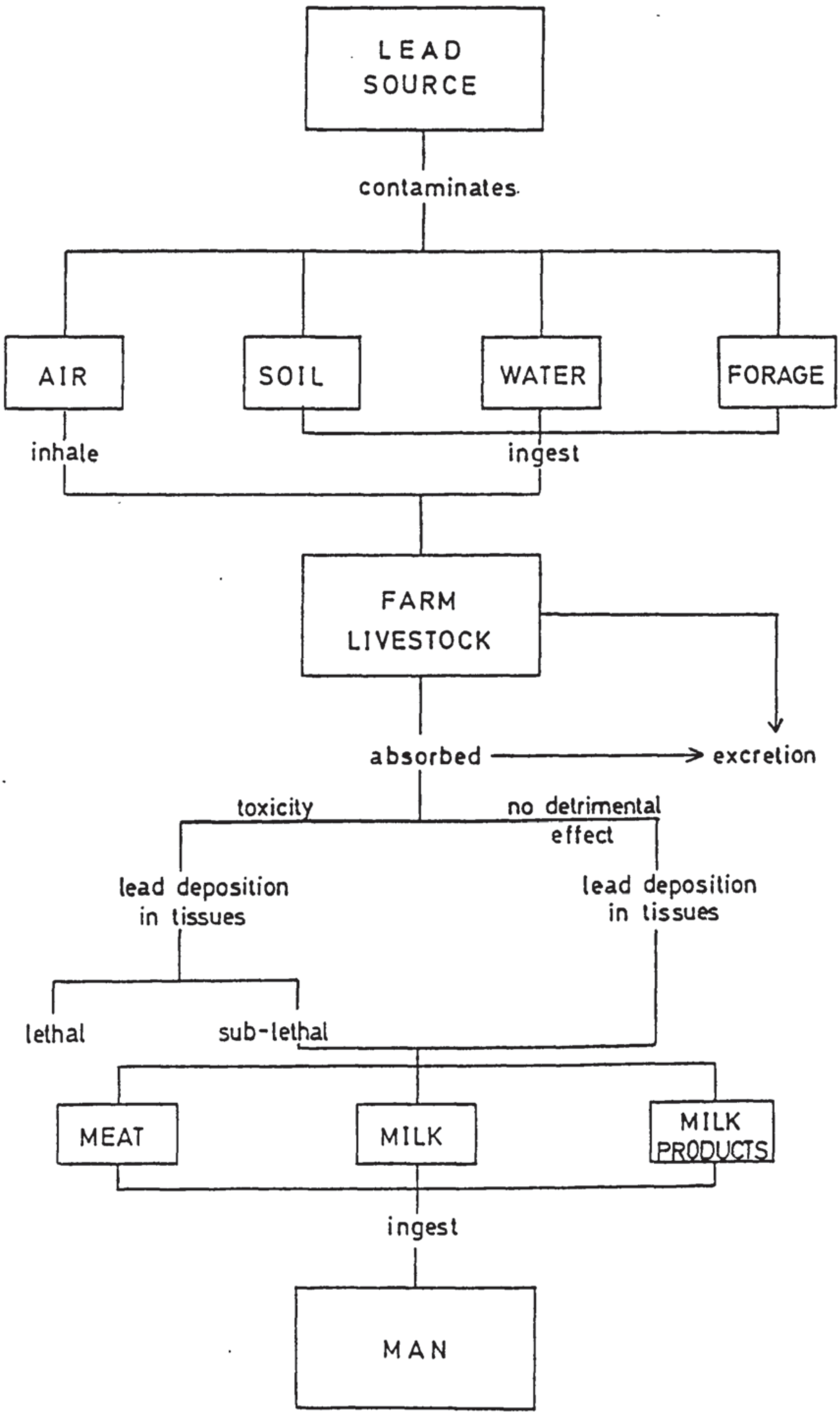
Figure 6-1 classifies the possible effects of the absorbed lead as either toxic or having no detrimental effect. Thus, toxicity is defined to include both clinical symptoms shown by individuals, and a chronic poisoning effect which is probably only identified by the study of populations. Such chronic effects include an alteration in growth and reproduction rate compared with groups not exposed to elevated lead levels. Sub-clinical effects of lead, such as the changes in the level of the enzyme δ-aminolevulinic acid dehydratase in the blood, for which no observable response has been found in terms of the well-being of the population, are not considered in this literature review.

D. Effects of High Level Lead Intake

Aronson (1975) reviews the literature on toxicity of lead to domestic animals and concludes that potentially lethal concentrations of lead in the forage are 80 μg/g dry weight for horses (1.7 mg lead/kg body weight) and over 200 μg/g for cattle (5.7 mg/kg) in the total diet when ingested over several months. A factor contributing to the susceptibility of horses to cumulative lead poisoning is their occasional ingestion of contaminated roots and soil when grazing (Aronson 1975).

Sheep, being more tolerant of lead than cattle, may withstand

FIGURE 6-1 Pathways of Lead to Man Through Animal Products



350 µg/g dry weight (8 mg/kg) without observable symptoms (Allcroft and Blaxter 1950). These figures of lethal concentrations in the forage are broad guidelines, and the actual value will depend upon a number of conditions such as animal health and age, possibly the type of forage, and the presence of other pollutants. Generally, it is the young animal which is most susceptible to lead poisoning (Schmitt et al. 1971; Stewart and Allcroft 1956).

The symptoms of clinical lead poisoning have been described by a number of workers. These are quite variable, both between individuals and species. Observable signs such as diarrhoea, colic and disturbances of the central nervous system (excitement, stupor, motor abnormalities and blindness) may occur in cattle.

There are few reports of substantiated clinical cases of lead poisoning of sheep in the literature (Forbes and Sanderson 1978). However, Stewart and Allcroft (1956) do report the occurrence of locomotor disability and generally sluggish behaviour of sheep grazing old lead mining areas in spring.

There are numerous reported observations of lead poisoning of horses, and clinical symptoms include difficulty in breathing (roaring), stiffness of joints, clumsiness, enlarged joints, facial paralysis, muscular weakness and poor appetite (Forbes and Sanderson 1978). When clinical symptoms appear gradually, measures may be taken to treat poisoning in horses (Allen 1975), sheep (Stewart and Allcroft 1956) and cattle (Hammond and Sorensen 1957).

E. Effects of Low Level Intake

The possible result of ingesting lead levels below those producing clinical symptoms in domestic animals is less well documented. Perhaps the most frequently observed effect is of an increase in abortion rate in pregnant ewes (James et al. 1966; Allcroft and Blaxter 1950; Egan and O'Cuill 1969; Buck 1970). Carlson et al. (1973) report no significant changes in blood haemoglobin, packed cell volume, and total red and white blood cells when ingesting 0.5 to 16 mg lead/kg body weight during a 23 week period. A diet containing up to 1,000 μg/g lead acetate had no effect on feed consumption or weight gain of twenty lambs over a 84 day period (Fick et al. 1976). However, Stewart and Allcroft (1956) found a lower weight gain in healthy lambs grazing on lead contaminated soil compared with a control group.

Aronson (1971) concludes that there is little evidence of sub-clinical levels of lead causing abortion in horses and cattle. Dinius et al. (1973) found no effect of 10 to 100 μg/g lead (as the chromate) in the diet of bull calves on their feed consumption, weight gain, electrocardiography and heart and respiration rates over 100 days. Hay containing 100 μg/g of lead harvested from a roadside did not result in any symptoms of toxicity when fed to dairy cattle for 34 days (Bovay 1971). Willoughby et al. (1972) found no change in feed intake until other clinical symptoms appeared when feeding three to four week old fillies 800 μg/g lead in their diet.

Therefore, few health effects due to a low level ingestion of lead have been identified, although this would seem an area worthy of further study.

F. Studies Concerning Roadside Lead

It would appear that there are no reported cases of clinical poisoning of domestic animals as a result of grazing roadside pastures. Few studies have investigated animals in the vicinity of major roads although tissue lead levels have been studied in cattle (Froslie et al.

1975 ; Rameau 1968) and sheep (Ward et al. 1978a,b) which have grazed pastures bordering roads in countries other than the UK. There appears to be no published data concerning the health of horses grazing roadside pasture. However, because of the small area of highly contaminated pasture involved, Aronson (1972) suggests that it is unlikely that cattle and horses would eat sufficient quantities of this herbage to result in clinical lead poisoning. This is a consequence of the rapid decline in grass lead concentration with distance from the road discussed earlier in Chapter 4. The possibility that roadside lead may have some detrimental health effect on livestock was considered in greater detail with reference to the M6 study site.

6.3 THE SIGNIFICANCE OF ROADSIDE LEAD FOR THE HEALTH OF LIVESTOCK NEAR THE M6 MOTORWAY IN CHESHIRE

No known cases of lead poisoning are reported for animals grazing pastures bordering the M6 study area. Grass lead levels reported in Chapter 4 show that the herbage is unlikely to be toxic to the grazing livestock when compared with the concentrations which are tolerated by cattle, sheep and horses. The highest lead levels in the study area occurred to the east, and within ten metres of the motorway fence. This peak value of 280 µg/g is probably not toxic to sheep grazing this area even if it were to form the total diet of these animals for many months.

One further possibility is that the presence of other pollutants in the forage makes the animals more susceptible to lead poisoning. Merry and Tiller (1978) briefly review the published work concerning the complex interactions when more than one toxic element is present in the diet of a ruminant. A high dietary intake of zinc may aggravate lead toxicity whilst cadmium and zinc, as well as cadmium and copper, may be mutually antagonistic. In view of reports of elevated heavy metals other than lead in roadside soils (e.g. Lagerwerff and Specht 1970) the pasture grass at the M6 study site was investigated to determine the copper, cadmium, mercury and manganese content, in addition to the lead and zinc levels reported in Chapter 4. Analysis for all these trace elements was by atomic absorption. For mercury a cold vapour technique was employed as described by Barlow et al. (1979). Table 6-1 compares the metal content of the pasture at the motorway fence with control sites and results suggest that these other metals are unlikely to be a threat to the health of farm livestock. One finding, perhaps of some importance, was that of a high level of cadmium (3-5 μg/g) in grass growing in a depression in Field B (see Figure 1-3) and situated approximately 50 metres from the motorway fence. This elevated concentration is unlikely to have resulted from motor vehicles, but is probably due to the application of phosphate and potash fertilizer which was subsequently found to have a high cadmium content (5.5 μg/g).

It would seem unlikely that health effects result from the ingestion of roadside herbage contaminated by lead at the study site. However, a more detailed assessment of the exposure of livestock to lead is necessary because other pathways of entry into the animal exist. These include inhalation of airborne particulates, and ingestion of drinking water and feeds which contain lead. An assessment of the importance of these other lead sources, together with that resulting directly from vehicular lead, involved the following two approaches:

(i) <u>An investigation of the lead content of animal faeces</u>

In order to obtain some indication of the amount of lead being ingested by cattle and sheep in the study area, a number of faecal samples were taken from animals at pasture. Table 6-2 shows the lead concentration of the faeces which was determined by the same method of chemical analysis as was used for soil and grass. The

TABLE 6-1

Copper, Mercury, Manganese and Cadmium Content of Grass in the M6 Study Area

Sample site (Distance from motorway border fence)	Metal Content of Grass µg/g (dry wt)			
	Copper*	Cadmium*	Manganese**	Mercury*
10 metres east	48	0.7	120	.063
10 metres west	16	0.6	82	-
1500 metres west	7.5	0.5	82	.043
Normal concentration in plants (Bowen 1966)	14	0.6	630	0.018

* Sampled December 1977

** Sampled April 1978

TABLE 6-2

The Lead Content of Animal Faeces at the

M6 Study Site

1. Cattle

Site	Date of sampling	Lead concentration (µg/g)
Control Sites	December 1978	18
	July 1978	29
	March 1979	9, 24
	May 1979	17.5, 19.5
	November 1979	22, 34
	December 1979	21.5
Within 50 metres west of motorway	July 1978	23
	May 1979	54, 56
	November 1979	29, 30, 29

2. Sheep

Site	Date of sampling	Lead concentration (µg/g)
Within 200 metres east of motorway	December 1978	48
	February 1978	36, 40, 43
	May 1978	30
	January 1979	32, 31, 48, 45
	February 1979	34, 31, 37
	March 1979	92, 24
	April 1979	47, 33
	May 1979	41, 94, 46, 50
	October 1979	129
	January 1980	64

lead content of faeces from cattle grazing near the motorway was, in general, similar to that for cattle at the background sites. These values may be compared with the findings of Riner et al. (1974) who report a lead content of cattle faeces of 7.5 to 20 µg/g when the animals were ingesting a feed containing 1.5 µg/g of lead. The results suggest, therefore, that quite low levels of lead are being ingested by cattle in the study area.

The data for the lead content of sheep faeces is more difficult to interpret because of the lack of samples from background sites. Concentrations are generally higher than for cattle, and this is probably the result of the greater lead levels in the pasture east of the motorway which is grazed by sheep. It is also possible that the gut absorption characteristics of sheep and cattle differ which may partially account for the phenomenon. The limited data obtained for the M6 site suggests that the lead content of sheep faeces is typically between 30 and 60 µg/g and exceptionally over 100 µg/g. This may be compared with the findings of Allcroft (1951) who reports a faecal lead concentration of 20 to 200 µg/g for sheep which had been grazing over old lead mining areas in Derbyshire (UK). He observed no abnormal symptoms in these sheep over a period of six years. It would seem unlikely, therefore, that the sheep at the study site are absorbing sufficient lead to produce any detrimental health effects.

(ii) Estimation of the amount of lead absorbed by the animals

The amount of lead taken up from the environment by the farm livestock may be estimated by consideration of the lead concentration of the animal's forage and the air that it breathes. This will be different for the cattle, sheep and horses in the study area because of the management system applied to each type of livestock.

Sheep were at pasture during all months of the year whilst dairy cattle grazed only during the period of April till November. In winter, the dairy cattle are stall fed with hay, silage, mineral feed, concentrate and barley which was found to contain between 0.5 and 5 µg/g of lead. The sheep also receive additional feed, and hay is made available to the animals in the field during the coldest winter months. Horses only grazed the field immediately east of the motorway during midsummer and were kept in fields and stables situated approximately 500 metres west of the motorway at other times. During the summer months, the mean lead concentration of the grass within 10 metres of the motorway fence was less than 50 µg/g and thus, was not a threat to the health of the horses.

If the dairy farm is considered, the rapid decline in levels of lead contamination with distance from the motorway means that lead poisoning of animals is most unlikely to occur. Pasture grass at distances of more than 50 metres from the motorway fence is not greatly affected by vehicular lead emissions. Therefore, only about 20 per cent of the total area of pasture on the study farm is significantly contaminated by roadside lead. This is despite the fact that, compared with other farms in the Sandbach region, this farm has a large proportion of its fields bordering the motorway. Thus, the highly contaminated pasture forms only a minor part of the total diet of the dairy cattle. In addition, the levels of lead in the topsoil were sufficiently low to discard the possibility of intoxication as a result of ingesting soil along with herbage.

A further source of lead to the farm livestock is that of airborne particulates which are inhaled by the animals. If the data concerning lead uptake from the human lung, reported by Chamberlain et al. (1978), is applied to the dairy cattle, it can be demonstrated that inhalation is a minor source of lead compared with ingestion. These workers found that for man, approximately 35 per cent of the inhaled vehicular lead was deposited in the lung, and that 50 per cent of this lead was then transferred to blood erythrocytes. On this basis, if the mean daily airborne lead concentration 10 metres downwind of the motorway for each month is between 1 and 3 $\mu g/m^3$ (Chapter 2) and the cow inhales 130 m^3 of the air per day (Comar, 1966), then between 0.23 and 0.69 mg of lead will be absorbed by the animal each day. This assumes that the animal remains downwind of the motorway and close to the motorway fence for a full 24 hour period. A similar calculation of the intake of lead by ingestion may be undertaken assuming that the animal consumes 13.6 kg of dry matter/day (Thornton and Kinniburg 1978). Then, if the pasture lead content were as low as 10 $\mu g/g$, and only 2 per cent of the ingested lead was absorbed through the gut wall, 2.7 mg of lead would enter the blood each day. Clearly, the inhaled lead accounts for less than 10 per cent of the total lead absorption. This is in agreement with the calculations of Chamberlain (1975) who suggests that inhaled lead is a minor source of lead to grazing animals compared with the ingestion of herbage.

Ward et al. (1978a) found that inhalation of airborne lead, by sheep kept within three metres of a road in New Zealand, resulted in a significant rise in the lead content of tissues over a period of only five days. Their study would, therefore, suggest that inhalation of roadside lead may be of greater significance than the above calculations have indicated. However, these workers did not make any measurements of air lead concentration and only small numbers of animals were used. Although it is most unlikely that the animals are exclusively within three metres of a major road in the UK, the findings of the New Zealand study do warrant further investigation to evaluate the relative importance of inhalation and ingestion as a source of lead for farm livestock.

A further source of lead to farm animals is that of drinking water. At the study site, animals have access only to drinking troughs to which piped mains water is supplied. Lead levels in the water were not above normal (<10 µg/l) and thus no danger of lead poisoning would come from this source. Investigations of sediment in drainage ditches which receive runoff from the M6 motorway showed very high lead levels (up to 1,200 µg/g). These high levels could possibly be ingested by animals drinking from the stream in which the sediment had been disturbed. The lead concentration of the sediment declined quite rapidly with distance downstream of the motorway to a level of only 40 µg/g at 100 metres. Therefore, if farm animals do have access to streams and ditches which receive runoff from the motorway, the sediment immediately downstream from the inflow could be a significant source of lead.

6.4 CONCLUSION

There would appear to be no danger of lead poisoning to farm livestock as a result of the inhalation or ingestion of the lead released by motor vehicles on the M6 motorway in Cheshire. The most important source of lead to animals in the study area would appear to come from ingestion of contaminated herbage within 50 metres of the motorway fence. However, at present, lead levels are not sufficient to produce toxicity to sheep and cattle even if the animals grazed continuously within this area. Horses are, perhaps, more at risk, although there is no danger of lead poisoning if grazing is limited to the midsummer months. It is suggested that the greatest risk to livestock may occur where background levels of lead are relatively high, such as found near the M4 motorway (London) by Little and Wiffen (1977). At such a site, roadside lead may add to the already elevated level and increase the risk of exceeding the lethal limit.

Although the lead concentration of roadside pasture is sufficiently low to prevent clinical symptoms of toxicity, it is possible that contaminated forage, and the inhalation of airborne particulates, produces an increase in the lead content of animal tissues which are to be comsumed by man. This possibility will now be considered.

6.5 CONTAMINATION OF MEAT AND MILK PRODUCTS BY ROADSIDE LEAD

A. Bovine Species

Specific studies of tissue lead concentrations in cattle grazing near highways are few. Blaxter (1950) fed road dust which was 2 per cent lead to a calf for 125 days so that a total of 105 grams of lead was ingested. Only 2 per cent of this lead was absorbed and the long term deposition in tissues was small. Froslie et al (1975) looked at bone concentrations of lead in cattle which had been grazing in the vicinity of a highway since birth, and found lead values no higher than a control group. Bovay (1971) reports a four-fold increase in the lead content of milk when cows are fed roadside hay containing 10 µg/g of lead. In contrast, Rameau (1968) found no significant difference in the lead content of milk from cows grazing near a highway with 30 to 110 µg/g of lead in forage when compared with animals grazing distant from the road. There is, therefore, little evidence at present of the occurrence of high lead levels in tissues of cattle as a result of grazing near roads, although research in this field is rather limited.

Unfortunately, it was not possible to obtain specimens of edible parts of the animals considered in this study. However, samples of blood and hair were procured and these, it is suggested, may give a good indication of the animals' exposure to lead. In addition, milk samples were taken monthly during 1978 from three cows. Samples of blood, milk and hair were analysed for lead content by the methods described by Barlow (1977a, b) and Barlow and Kapel (1980) respectively. These results are now compared with the findings of other workers.

a) Blood

Reference to Table 6.3 shows the range of blood lead levels found by various workers. It would appear that cattle not exposed to contaminated forage have blood lead levels ranging from 5 to 30 µg/dl.

TABLE 6-3

Blood Lead Levels in Cattle

Author & Country of Study	Level of Lead Exposure	Blood Lead Levels (µg/dl)
Allen (1975) UK	'Normal' pasture	<30
Allcroft (1951) UK	'Normal' pasture	18
Ward et al. (1977) New Zealand	'Normal' pasture	5-25
Hatch & Funnel (1969) Canada	1. 'Normal' pasture 2. Clinically poisoned animal	5-25 35-240
Buck (1970) USA	Clinically poisoned	>35
Hammond & Sorenson (1957) USA	Clinically poisoned	>150
Dorn et al. (1975) USA	1. Normal summer pasture 2. Normal winter pasture 3. Pasture near smelter (summer) 4. Pasture near smelter (spring)	6.6 19 28 58
Hammond & Aronson (1964) USA	1. Pasture near smelter (270 - 320 µg/g)	126
Harbourne et al. (1968) USA	1. Calves grazing old mine workings (0.2 to 1% lead in soil)	70-140
Osweiler et al. (1973) USA	Fatally poisoned	20-380
Vick (1976) UK	1. Normal pasture 2. Near M6 Motorway (Warwickshire)	28.5 29
Colbourn (1976) UK.	1. Normal pasture in Derbyshire (December) 2. Populations with above normal blood lead	0-24 16-36
Clegg (1974) UK	March herbage containing 1. 40-70 µg/g 2. 50-100 µg/g 3. 100- 720 µg/g	mean (range) 10 (1-40) 8 (0-26) 10 (1-26)
*Dinius et al (1973)	1. Calves fed 100 µg/g for 75 days 2. 100 µg/g for 100 days	30 20
*Aronson et al. (1968)	Calves fed 1.5-3 g of lead (killed 5-7 days later)	27
Kelliher et al. (1973)	1. Steers fed 15 mg/kg for 100 days 2. After 100-283 days	250 60-100

* = Feeding trial

Clinically poisoned animals have a concentration of at least 35 μg/dl. Caution is necessary when comparing values reported in the table because a number of different analytical techniques have been used and inter-laboratory comparisons have found the results of blood lead analysis very variable (Ter Haar 1975). In addition, few workers have considered a possible seasonal variation in blood lead levels as was found by Dorn et al. (1975).

The blood-lead investigation in the present work involved the taking of venous blood from ten dairy cows on the study farm. Samples were taken in March and November to correspond to periods of stall feeding and field grazing. The results showed that the lead levels in the twenty samples were all below 10 μg/dl, the detection limit for the method of analysis used. This would suggest that the cattle are not exposed to high levels of lead. These findings are in agreement with those of Vick (1976) who found no significant difference between the blood lead content of heifers grazing near the M6 motorway in Warwickshire and a control population. The blood results for the M6 study farm were further supported by the analysis of hair taken from three cows in March and November 1979 which showed lead levels below 2 μg/g.

b) Milk

One possible pathway of lead from domestic animals to man is through milk and milk products. Potter et al. (1971) found that only 0.02 per cent of ingested lead appeared in the milk during a period of six days. Feeding cows 11 mg lead/kg of body weight for two weeks resulted in 0.16 per cent of the lead appearing in milk solids (Lynch and Smith 1974).

Table 6-4 shows the lead content of milk obtained from control and polluted sites. It would appear that normal levels are less than 13 μg/dl, with generally higher concentrations in contaminated areas, exceptionally up to 300 μg/dl. These values may be compared with the

TABLE 6-4

The Lead Content of Cow's Milk

Author & Country of Study	Level of Lead Exposure	Lead Concentration of Milk (µg/dl)
Hammond & Aronson (1964) USA	Pasture containing 27 to 320 µg/g	300
Djuric et al (1971) Yugoslavia	1. Normal pasture (<10µg/g) 2. Pasture near smelter (100-430 µg/g)	2.0 160-190
Allen (1975) UK	Pasture near smelter 1. In spring 2. In winter	 1.6 1.1
Lagerwerff & Brower (1973) USA	Smelter area 1. spring 2. winter	 14 4.7
Dorn et al (1975) USA	Normal pasture 1. autumn 2. spring Near smelter 1. autumn 2. spring	 13 7 3.5 2.5
Bruhn & Franke (1976) USA	Various farms in California	9
HMSO (1975) UK	Smelter area	3.5
Egan & O'Cuill (1968) Ireland	1. Old mining area (580 µg/g)	28
Rameau (1968) Belgium	1. Normal pasture 2. Near road (30-110 µg/g)	2.6 3.3
Bovay (1971) Switzerland	1. Roadside hay (100 µg/g)	4 times level of control
Lewis (1972) USA	Near smelter	6.0
Schmitt et al (1971) Canada	1. Normal pasture 2. Near smelter	0-12 0-11
*Blanc et al (1977)	99 µg/g in diet	6.4
*Aronson (1975)	near lethal level	300
*Marshall (1963)	13 mg/45 kg body weight for 125 days	<5.0
*Lynch & Smitt (1974)	11 mg/day for 14 days	0.6

* = Feeding Trial

lead content of pasteurised milk for retail sale which is reported as being in the range of 1 to 8 µg/dl in the UK (HMSO 1975).

A few studies have looked at possible seasonal variations in the lead concentration of milk, and results suggest that a change can occur in some instances (Lagerwerff and Brower 1973, Dorn et al. 1975, Allen 1975). Highest levels were found by Lagerwerff and Brower (1973) when cattle were at pasture during summer, which they suggest is because of the use of less contaminated feed in winter. Dorn et al. (1975) found highest values in the winter when lead deposition to pasture was greatest, but no information relating to the composition of the diet throughout the year is reported. Allen (1975) found no significant difference in the lead content of milk taken from cows in March, after several months of stall feeding, and in July when the animals were at pasture.

A survey of metal contaminants in milk and milk products (Milk Marketing Board 1978) found that lead levels in milk from contaminated areas were higher than milk from other regions and that ingestion by the cow is possibly the greatest source of lead contamination of milk. However, results from the present study found a lead content of less than 10 µg/dl in all milk samples examined, suggesting that vehicular lead is not resulting in significant contamination of this animal product.

c) <u>Animal organs</u>

Probably the most important pathway of lead from livestock to man is through edible meat products. However for cattle, Dinius et al. (1973) found that only 0.03 per cent of the ingested lead is deposited in tissues. Most of the work concerning the lead concentration of animal tissues has been carried out during autopsy as a means of identifying the cause of death. Allcroft (1951) suggests a concentration of 40 µg/g (wet weight) in the kidney cortex and 10 µg/g in the liver as diagnostic of lead poisoning. However, liver tissue may contain less than 3 µg/g in a clinically poisoned cow (Hatch and Funnell 1969).

The major deposition of lead in the body occurs in the bones, yet this is of less importance to man than accumulation in liver, kidney and muscle which are important sources of food. Table 6-5 shows the lead concentration of a number of organs both from animals fed experimentally in the laboratory and those grazing at pasture. Normal levels in liver and kidney are 0.3-1.5 μg/g, whereas clinically poisoned cattle may have 10-72 μg/g in the liver and 25-137 μg/g in kidney tissue. Dinius et al. (1973) show no effect of 10 μg/g in the diet, but when fed 100 μg/g for 100 days the lead concentration of bovine kidneys and liver increased by four-fold. Fortunately, the lead concentration of muscle tissue appears not to exceed 1 μg/g, even in a poisoned animal.

It would appear, therefore, that the most likely pathway of roadside lead to man is through the consumption of offal rather than any other meat or milk product. However, as dairy cattle spend only a limited period of time grazing fresh pasture, and this is during the summer months when grass lead levels are lowest, it is unlikely that high concentrations of lead will occur in the offal. For beef cattle, which may spend a greater proportion of the time at fresh pasture, the situation may not be as satisfactory and this would seem an area worth of further study.

B. Sheep

Although there are fewer studies concerning the lead concentration of sheep tissues, a greater proportion of the information specifically refers to roadside contamination. Table 6-6 shows the lead concentration of blood and organs related to lead exposure. Normal blood-lead levels appear to be in the range of 5-25 μg/dl, but concentrations up to 240 μg/dl have been found in sheep from contaminated areas. Ward et al.

TABLE 6-5

The Lead Concentration of Bovine Tissues

Author & Country of Study	Level of Lead Exposure	Tissue Lead Concentration µg/g (wet wt) Liver	Kidney	Other
Allcroft (1951) UK	1. Normal diet 2. Poisoned calves	0.3 >10	1.5 >40 (C)	- -
Allcroft (1950) UK	poisoned calf	72	-	muscle 1.0
Hatch & Funnell (1969) Canada	Poisoned	43	137	-
Lewis (1972) USA	Smelter area	0.2	-	muscle 0.4 bone 20
Egan & O'Cuill (1970) Ireland	1. poisoned at mining area	-	25	-
Schmitt et al. (1971) Canada	Smelter area	1-5	1-5	muscle 0.06-0.3
Flanjak & Lee (1979) Australia	Eight locations in New South Wales	<0.85	<2.3	-
HMSO (1975) UK	Smelter area 1. general level 2. high samples	 <1.0 >2.0	 <1.0 -	
Frosher et al. (1975) Norway	1. Adult calves near highway & control 2. Young animals	- -	- -	bone 15+ 6+
*Dinius et al. (1973)	1. Control animals & those fed 10 µg/g for 100 days 2. 100 µg/g for 100 days	0.6 2.3	1.2C 4.7C 2.1M	
*Aronson et al (1968)	Calves fed 1.5 to 3 g lead orally and killed 5 to 7 days later	2.2-3.0	2.8-8.6 (C)	bone: 14.5-22.5 muscle: 0.02-0.1

NB The current UK maximum lead level in food for human consumption is 1 µg/g (fresh weight) with a value of 2 µg/g for liver (Lead in Food Regulations 1979, SI 1254 1979)

+ = ash weight　　M = medulla
* = feeding trial　　C = cortex

TABLE 6-6

The Lead Concentration of Sheep Tissues

Author & Country of Study	Level of Exposure to Lead	Tissue Concentration (µg/g wet weight) Blood µg/dl	Liver	Kidney	Other
Sharma (1971)	1. *16 mg/kg/day for 1 week	60	-	-	-
Carlson et al (1973)	1. *4.5 mg/kg 27 weeks 2. 2.3 mg/kg	30 17	- -	- -	- -
Allcroft (1951) (UK)	1. normal pasture 2. mining area	13 -	1.0 4.8	- -	- -
Ward et al (1977) (New Zealand)	survey 250 sheep	20	-	-	-
Allcroft & Blaxter (1950) (UK)	1. mining area 2. control	20 ± 1 13.9 ± 1	- -	- -	- -
Butler et al (1957)	Lambs from mining area (162-174 µg/g)	70	58	77	rib bone 275 (dry wt)
Clegg & Rylands (1966) (UK)	lambs from mining area	85-240	12-100	6-100	
Stewart & Allcroft (1956) (UK)	1. ewes (normal) 2. poisoned lambs from mining area (> 400 µg/g)	55 144	- 30	- 38	- -

continued........

TABLE 6-6 (continued)

Author & Country of Study	Level of Exposure to Lead	Tissue Concentration (µg/g wet weight) Blood µg/dl	Liver	Kidney	Other
Froslie et al (1975) (Norway)	roadside (4.6 µg/g) hay for 5 months and a control	no difference from control			-
Ward et al (1978 a,b) (New Zealand)	roadside (60 µg/g) for 6 months	<100	20	154c	bones 30
	1. control	20	0.8	0.7	0.12
	2. 55 µg/g in feed for 9 days	110	8.4	39c	1.9
	at roadside for 14 days (19 µg/g)	140	-	-	-
Ward et al (1978 a,b) (New Zealand)	1. 10 days after remove from highway	0.6	-	-	-
	2. and after 6 months	0.2	-	-	-
Ward & Brooks (1979) (New Zealand)	1. Within 50 metres of a road for 136 days	-	-	-	hair 4-15 µg/g
Colbourn (1976) (UK)	1. lambs in mining district	-	30-60 (dry wt)	<2-42	brain <2-6 rib bone 12-30

* = Feeding Trial

(1978b) found that blood lead concentrations responded rapidly to a change in the level of exposure. When the sheep were removed from the source of contamination (a major road) a large decrease occurred within ten days, but levels remained slightly above normal for over six months. Table 6-6 also shows that the lead concentration of the liver and kidneys may reach 100 µg/g in sheep from contaminated areas, compared with about 1 µg/g in background regions.

No blood samples from sheep were available in the present study, but in the spring of 1979, wool samples were taken from 10 sheep which had grazed the field immediately east of the motorway during the previous winter. These samples were analysed both in the unwashed state and after a washing procedure. The wash involved the shaking of a one-gram sample of wool for an hour in 100 ml of deionised water. The lead content of the wool was determined by the method of Barlow and Kapel (1980) and the results are shown in Table 6-7. These values relate to the distal 2 cm of wool taken from the back of the animal.

Clearly, the lead content of the wool is quite variable, as was the effectiveness of deionised water at removing lead. The results do, however, compare well with the 4 to 26 µg/g level found for water-washed wool by Ward and Brooks (1979). These workers sampled wool from sheep which were grazing within 50 metres of a road carrying 5,000 vehicles/day in New Zealand. They also reported a good correlation between lead in wool and in blood, so that a range of 4 to 26 µg/g in wool was related to a blood level of 16 to 170 µg/dl. Although caution is necessary in comparing the results of the present study with those of the New Zealand workers, it can be seen that if a similar relationship does occur for the sheep at this site, then the blood lead concentration of the ten sheep was approximately 40-45 µg/dl.

TABLE 6-7

Lead Content of Washed and Unwashed Wool taken from Sheep Grazing near the M6 Motorway in Spring 1979

Sample	Lead Concentration of Wool µg/g (dry wt)	
	Unwashed	Washed
1	9.5	2.0
2	15.5	9.5
3	12.0	2.0
4	3.0	6.5
5	21.0	21.5
6	13.0	16.0
7	9.0	5.5
8	17.5	9.5
9	15.0	9.5
10	8.0	10.5
Mean	12	9

The results of the wool analysis could not themselves be regarded as sufficient evidence to suggest that the blood lead levels of the sheep are elevated. However, the concentration of lead in the pasture grass measured at this site must certainly result in a significant increase in blood lead levels of the sheep during winter months. If the results of roadside trials reported by Ward et al. (1978a,b) are applicable to the M6 study site, then it would seem probable that the lead content of kidney and liver tissue exceeds the UK statutory limit of 2 µg/g for liver and 1 µg/g for kidney (Lead in Food Regulations, 1979 SI 1254).

C. Horses

The tissue lead concentration in horses is of less interest in the UK because the meat is rarely eaten by man. Normal blood lead levels appear to be about 20 µg/dl (Ward et al. 1977, Willoughby and Brown 1971, Allen 1975). Clinically poisoned animals have a blood lead level in excess of about 35 µg/dl (Allen 1975, Schmitt et al. 1971). Lewis (1972) suggests that 4 µg/g in liver tissue reflects lead poisoning, compared with normal levels of 0.04 µg/g. His study also investigated the lead content of mane hair after washing in acetone and ethyl ether, and found concentrations of 1.4 µg/g at control sites. In the vicinity of lead smelters, he found a two to five-fold increase in the lead content of the mane hair at 50 per cent of the sites investigated.

In July 1979, the lead content of unwashed mane hair of three horses which were grazing the field immediately east of the M6 motorway was between 10 and 19.5 µg/g. If comparable with hair from cattle and sheep, these results suggest that the mane hair may reflect a contaminated environment, though the pasture lead content does not approach a lethal level during the summer months. Further research is necessary to determine if the roadside contamination could affect the condition of horses at a sub-lethal level of exposure.

6.6 CONCLUSION

The results of this study, in confirmation of the work of other authors, suggests that the only tissues likely to contain high levels of lead as a result of roadside contamination are liver and kidney. Lead levels reached in dairy cattle are probably lower than those found in animals such as sheep, which graze the roadside herbage all through the year. It is suggested that further work should be undertaken to investigate whether offal from sheep which have grazed roadside pasture contains lead levels in excess of UK statutory limits. The results of this study suggest that this is likely to be the case.

CHAPTER 7

RECOMMENDATIONS TO FARMERS AND PLANNERS FOR MEASURES TO MINIMISE THE EXPOSURE OF LIVESTOCK TO ROADSIDE LEAD

7.1 INTRODUCTION

The level of lead contamination of roadside pasture does not, at present, constitute a known health hazard for farm animals. A possible exception to this is where vehicular lead from a motorway adds to the level of lead in pasture which is already contaminated by emissions from other sources. Perhaps the greatest problem is the possibility of high levels of lead building up in liver and kidney tissues which are to be eaten by man.

If the contamination of such tissues by roadside lead is confirmed as being a serious problem in the UK, there are two approaches which authorities may use to reduce the level of contamination. These are:

i) To reduce the lead content of petrol or prohibit it completely.

ii) To recommend measures to farmers and road planners which may be used to reduce the exposure of farm livestock to vehicular lead.

Any change in the lead content of petrol beyond levels already determined for January 1981 (EEC Directive 78/611/EEC) are likely to be made in response to the possible health hazard of lead to children in the urban environment. This factor, and the economic considerations of the removal of lead from petrol, are beyond the scope of this study. Therefore, this chapter will discuss the possible action that may be taken by the farmer and road planner to minimise the exposure of live-stock to vehicular lead, assuming that present discharge levels are maintained.

7.2 POSSIBLE ACTION BY THE FARMER

A proposed new road development must include an assessment of the impact of the scheme upon the environment. At present, the contamination of agricultural land is not a major consideration affecting planning decisions. It is, however, suggested that this should not be the case, and that planners ought to be aware of the effects that road design and traffic flow may have upon the level of roadside lead. The following considerations must be incorporated into the planning decision.

A. Land Area Contaminated by Lead

The area of land which may be contaminated by lead emissions from the proposed development includes a strip 60 metres either side of the road. A scheme which minimises the area of agricultural land in this strip will be more favourable on this assessment. In view of the rapid decline in the lead content of soil and vegetation with distance from the road, and the effect of prevailing wind direction upon lead levels, it may be useful to use a ranking system which recognises that land within 20 metres of the road, and that downwind of the road is the most highly contaminated. This more detailed analysis would be particularly appropriate where agricultural land occurs on only one side of the proposed road.

B. Land Use

Crops which have edible portions exposed to the atmosphere present the greatest risk for the transfer of roadside lead to man through the food chain. Therefore, crops such as pasture grass and leafy vegetables are less suitable for regions contaminated by airborne lead than root crops, such as potatoes, or cereals, where the edible grain is protected by the husk. The environmental impact of the road development should, therefore, have regard to the type of farming in the region affected by the scheme.

C. Soil Lead Content

The lead content of the soil prior to the new development should be investigated. This is because some areas of land along the proposed route may already be receiving quite high levels of lead due to the application of sewage sludge, local industrial emissions or other traffic sources. In addition, the soil may be "naturally" high in lead, and thus the contamination produced by vehicular lead emissions from the new road must be assessed with regard to existing levels. It is possible that the new source of lead may produce total concentrations in soil and vegetation which exceed toxic thresholds for grazing animals.

D. Surface Water Run-off from the Road

In Chapter 6 of this study, it was suggested that contaminated sediment, present in the run-off from the road surface, could, in some instances, be a significant source of lead for farm animals. The road planner should, therefore, take note of the fate of run-off water and if the drainage water does pass through agricultural land, one of the following measures may be considered:

a) Treatment of run-off water to remove suspended solids,

b) Provision of piped mains water for all animals and the prevention of access to drainage water by livestock,

c) Discharge of road run-off to an enclosed sewage system rather than as storm flow in open drainage channels.

E. The Conditions for Dispersal of Atmospheric Lead

The design of the road may have a significant influence upon the efficiency of dispersal of vehicular emissions in the atmosphere. Wind tunnel investigations by Nishida et al. (1977) found that when a road is in a deep cutting the dispersion of exhaust gases is restricted. Best dispersion of emissions occurred with elevated sections of roadway, although raised levels of pollutant may occur some distance from the road, as was found by Middleton et al. (1979) for the M6 motorway in Birmingham (UK). It may be expected that good dispersion of atmospheric lead reduces both the amount of lead inhaled by farm livestock and the deposition of airborne particulates at the roadside. However, the significance of this to the farm environment close to a major road has not been fully assessed.

A further consideration of dispersal is the use of shelter belts to influence the transport of motor vehicle lead emissions. Chamberlain et al. (1978) show that although hedges and trees may result in a doubling of the deposition rate to the ground surface by collecting particles on plant surfaces, this will have little effect on the total air lead concentration. This study does not, however, consider the effect that changes of wind speed due to the shelter belt may have upon desposition in the vicinity of this plant barrier. Little (1974) found that the collection of airborne metals by moss bags was reduced leeward of an elm hedge by 50 per cent compared with moss bags windward of this shelter belt. This could, therefore, have a significant effect upon the lead content of crops separated from a road by a tree line. Further research is necessary to investigate this possibility as a measure of minimising the contamination of pasture grass and other crops which are grown near major roads.

The planner should also be aware of the influence of driving conditions upon the amount of lead released by motor vehicles. A greater release of lead per vehicle will occur on graded sections of road (Chamberlain et al. 1978) and under stop-start driving conditions which may result from certain types of road junction (Habibi 1973).

F. Volume and Type of Traffic

The level of lead contamination of roadside soil and vegetation will be related to the number of petrol-engined vehicles using the road (Chapter 4). In addition, the seasonal variation in the volume of traffic will influence the lead content of roadside crops. This is because the deposition efficiency of lead at the roadside is greater during winter than summer months (Chapter 3). Therefore, a proposed route whose traffic is predominantly made up of holiday travellers during summer months may result in less deposition per vehicle flow than a road carrying commuter traffic, which is quite evenly distributed in volume during all months of the year.

Ideally, the road planner should have available a model which would enable him to predict the lead content of crops growing adjacent to the proposed road on the basis of the estimated traffic flow. Chamberlain et al. (1979) suggest that the lead content of grass one metre from a road carrying 1,000 vehicles per day is about 50 µg/g. However, this statement is based on only three sample transects, one of which was taken in Switzerland by Bovay (1970) and two near Birmingham (UK) by Davies and Holmes (1972). The data, therefore, is far from sufficient to conclude a meaningful relationship between traffic flow and pasture lead content which could be used as a guide for the prediction of levels of roadside contamination at any particular site.

Chamberlain (1975) suggests that the following relationship may be used to predict the level of lead contamination of roadside crops:

$$C = 12.5X$$

where C = Lead concentration in grass (μg/g)

and X = Lead concentration in the air (μg/m^3)

The equation is only applicable to actively growing grass. Caution is, however, necessary in the use of this method of prediction. Lead levels in herbage nine metres from the M4 motorway in 1977 were 108 μg/g when the grass was growing rapidly (Chamberlain et al. 1979). Yet air lead measurements at this site showed levels not exceeding 4 μg/m^3 which the above equation predicts would result in a grass lead concentration of only 50 μg/g. Applying the equation to the study site used in this work shows that the predicted grass lead content may indeed be of the right order of magnitude, but only in some situations. Thus, in the summer of 1978, when air lead concentration was estimated to be 2 μg/m^3 (as detailed in Section 2.10) the predicted grass lead would be 25 μg/g. This compares reasonably with the value found by analysis for a lead concentration in pasture of 30 to 60 μg/g at a distance of ten metres downwind of the motorway.

It may be concluded therefore, that there is no accurate method at present by which a planner may calculate the lead content of roadside vegetation based on the air lead level. Indeed, in Chapter 3 of this thesis, it has been shown that the mean air lead content is only one factor determining the rate of deposition of the emitted lead to plant surfaces. Meteorological factors also have a direct effect upon deposition rates and possibly fallout of airborne particulates is particularly pronounced during a certain combination of meteorological conditions and air lead concentrations. Thus, it is suggested that the development of a model to predict lead levels in roadside pasture is better based upon a direct measure of deposition rates of lead.

The present work has shown that the level of lead contamination of roadside pasture depends upon the rate of deposition of atmospheric lead over a period of four to six weeks in winter, and of perhaps one to two weeks in summer. This is the result of deposited lead building up on the surface of individual grass leaves whose life span varies seasonally. However, further work is necessary to relate the volume of traffic to the deposition rate of the emitted lead. The present work has used a Traffic-Normalised Deposition Efficiency (TNDE) described in Chapter 3, to compare deposition rates to moss bags at the same site in different seasons. Further study is needed to compare deposition rates during the same season for different traffic flows and other varying site conditions (e.g. road gradient and surrounding topography).

The object of this proposed work is to identify a relationship between traffic flow and the deposition rate of lead for a number of types of roadside environment (e.g. a road embankment, an elevated roadway with a shelter belt and exhibiting a steep gradient). Only when the effect of site conditions has been evaluated can progress be made towards developing a model which is of assistance to planners for predicting pasture lead levels. Until such time, any estimate of roadside lead levels is best based on the available evidence for a number of sites in the UK given in Table 4-3. It should be noted that if winter grazing of pasture is undertaken in the region under consideration, the levels of lead in winter pasture may be up to ten-fold greater than those given in the table.

7.3 POSSIBLE ACTION BY FARMERS

A number of measures recommended to planners to reduce the exposure of livestock to vehicular lead could only be carried out with the assistance of the farmer. Perhaps, through financial compensation for the road development, the farmer will restrict animal access to drainage waters and plant trees and hedges if these are shown to reduce the lead deposition to his land. Other measures must involve adaptation of the farm management system which again may be compensated for by the road builder. These changes might involve field use which could be altered on farms bordering existing roads.

The following recommendations are put forward as suggestions to minimise any health risk to farm livestock and reduce the contamination of farm products by roadside lead.

A. Field Use

i) Crop type

Where possible, fields which are adjacent to a major road and particularly those downwind, should be used for crops whose edible portions are not directly exposed to airborne lead. Therefore, crops such as lettuce, cabbage, brussels sprouts and pasture grass should be discouraged in favour of cereal grains, potatoes, carrots, sugar beet and other root crops. Investigations as part of this study have found low levels of lead in root vegetables and cereal grain, compared with grasses and leafy crops growing in the vicinity of major roads (Table 7-1). Such measures are possible and may be quite effective even on the M6 study farm, which has seven of its fifteen fields bordering the motorway. Two fields are used by the farmer to grow potatoes and barley, and at present these are not adjacent to the motorway. To minimise exposure of the dairy cattle to roadside lead it is

TABLE 7-1

A Comparison of the Lead Content of Foliage with that of some Other Plant Organs

Site	Lead concentration of grass	Other crops investigated	Lead concentration of the crops µg/g
15 metres east of M5 motorway (Glos.) (August 1979)	23	Wheat 1. stem 2. chaff 3. grain	 19 7 0.8
250 metres east of M1 motorway (Herts.) (February 1979)	56	A. Leeks (Edible part) B. Brussels sprouts	1.3 5.0
10 metres from A5 trunk road (Staffs.) (December 1978)	-	Carrots 1. leaf 2. tuber	 125 1.0
30 metres east of M6 motorway (Staffs.) (October 1977)	-	Sugar beet 1. leaf 2. tuber	 14 0.4

suggested that the two fields bordering the eastern edge of the motorway are used for barley and potatoes rather than as permanent pasture. This would have the added advantage of regular ploughing of the land preventing the accumulation of deposited lead in the surface layer of the soil.

The regular ploughing of the land may also be achieved by using the roadside fields for growing an annual grass (e.g. Lolium multiflorum). The possible benefit upon the lead content of the top 5 cm of soil is demonstrated by a comparison of ploughed and unploughed transects of soil sampled from an Italian ryegrass pasture east of the M5 motorway in December 1978. The grass had senesced at this stage and the farmer had ploughed only one half of the field allowing samples of soil to be taken from the two transects only two metres apart. There is a detectable difference in the lead concentration of the two soils adjacent to the motorway as shown in Table 7-2. It may be expected that a greater effect would be found for a field which had not been ploughed for many years.

A reduction in the lead content of surface soil by ploughing may reduce both lead uptake by the crop from the soil and the lead intake of farm animals as a result of ingestion of soil along with forage. Therefore, a change in field use together with the more frequent ploughing of the land may reduce the exposure of farm animals to roadside lead.

ii) Livestock

A further important factor to consider is the species of livestock grazing the most heavily contaminated fields.

At the study area, sheep, cattle and horses graze the fields east

TABLE 7-2

A Comparison of the Lead Content of a Ploughed and Unploughed Soil adjacent to the M5 Motorway (Glos.)

Distance from motorway fence (metres)	Lead Concentration of Surface Soil (μg/g)	
	Unploughed	Ploughed
0	61	42
6	52	35
10	43	46
20	48	40
30	48	45
50	54	49
100	47	39
200	38	37

of the motorway. As already discussed, horses are particularly susceptible to lead poisoning. Therefore, it would seem that these animals should be excluded from roadside fields. In addition, highest concentrations of lead in herbage occur during winter months. It is, therefore, recommended that livestock are not kept at pasture near major roads during the winter periods. Further, it is suggested that the grazing of roadside pasture is best restricted to the months of May to October. This is particularly important for young livestock as these are more susceptible to lead poisoning than mature animals (Allcroft 1951).

B. Field Management

Besides the type of use made of a roadside field, the management of the land itself provides opportunities to make changes which may minimise lead contamination of farm produce. Such changes include the following:

i) Soil characteristics

Soil properties have been shown to influence the uptake of lead by plants via the root system (Chapter 5). However, as most of the lead associated with roadside grass is as a surface deposit, a change in the rate of uptake of this metal from the soil will have only a minor influence upon the total lead content of pasture. Further, there is probably little scope for reducing plant uptake because acidic conditions which increase the availability of soil lead to grass roots are not normally found in well-maintained pastures where soil pH is controlled by liming.

ii) Grass species

Grass species may differ in the ability of their leaf surfaces to collect and retain airborne lead due to variation in surface micro-roughness (Chapter 5). However, no significant difference was found between the lead content of the pubescent leaf of Holcus lanatus and the non-hairy foliage of Lolium perenne. This minor investigation involved the sampling of individual tillers of Holcus which was an occasional species among the perennial ryegrass pasture. A further study is necessary to compare the lead content of pasture dominated by one of a number of grass species to determine if lead levels in forage may be reduced by the use of certain grasses. A further influence that grass species may have upon pasture lead content is through differences in production of dry matter. However, Anslow and Green (1967) found little difference in the date of commencement of spring growth and length of growing season of a number of cultivated species of grass in the UK. This suggests, therefore, that the timing of the spring decline in pasture lead content and the increase in lead levels during autumn until late winter, is unlikely to be altered by the use of grasses other than Lolium.

iii) Productivity of grassland

A further factor which may influence the lead content of forage is the productivity of the grassland. However, the field trial experiment described in Appendix 4 suggests that a high yielding, well-fertilized pasture may have a similar lead content to a grassland of relatively poor growth. This is because the major growth mechanism influencing the lead content of pasture is the life span of the individual leaves. Although the level of nitrogen fertilizer may have an important influence upon the growth rate of individual leaves, the rate of tillering and yield of pasture, it appears to have little influence upon the longevity of leaves (Ryle 1964, Langer 1972). Therefore, although the high rate of production of a well-fertilized pasture may contribute to the spring decline in grass lead levels, it would seem that the application of fertilizer has a relatively small effect on the lead content of pasture over a full growing season. A similar finding is reported by Tunney et al. (1972) who compared the lead content of fertilized and unfertilized plots of grass which were growing on mine tailings containing high levels of lead.

iv) Cutting and grazing of pasture

The system of grazing and cutting of pasture may also influence the amount of lead taken in by grazing animals. If the pasture is strip grazed it is suggested that this should be carried out by dividing the area into strips perpendicular to the road, rather than parallel to it. Thus, animals will not be restricted to an area of pasture which is most highly contaminated, and where air lead levels are greatest. A similar principle may be applied when cutting and collecting grass for hay or silage. If the crop is collected by machinery travelling perpendicular, rather than parallel to the motorway, it will ensure that the contaminated crop is diluted by forage of a lower lead content.

Investigations of the lead content of green and brown grass at the M6 study area found that most of the lead in pasture was associated with the oldest leaves. Therefore, any system of grazing or cutting which minimised the ingestion of senescent leaves would reduce the level of exposure for farm animals to roadside lead. The aim of limiting the accumulation of senescent leaves in the pasture is in agreement with the objectives of Hunt (1965) for maximising pasture production. He suggests, that to achieve the greatest yield of grass, grazing and cutting management must ensure that dry matter losses by decay are kept at a minimum while gains by photosynthesis are maximum. This may be achieved by a close cut of the grass once the canopy reaches the near-complete light interception stage as subsequent to this, dead matter accumulates in the sward.

Before any advice could be given to farmers, the relative effect of occasional close cutting or more frequent lenient harvesting on the lead content of a contaminated pasture should be investigated by field trials. If conditions result in a rapid loss of senescent leaves from the plant canopy by decay processes then it may not be necessary to manage the pasture in a manner that limits the production of dead plant material. In this context, the field trial experiment carried out 1,000 metres west of the M6 motorway found no difference in the lead content of swards of different ages. Possibly this may differ in a more contaminated environment such as at the roadside, where surface contamination by lead may be of greater importance. However, Leafe (pers. comm. 1979) suggests that the average leaf age of a grass sward is little affected by grazing and consequently there may be little that the farmer can do to minimise the pasture lead content by adjusting cutting and grazing regimes.

7.4 CONCLUSION

If further research into roadside lead pollution concludes that the exposure of farm livestock to lead must be reduced, then there are a number of effective and practical measures that may be taken by planners and farmers. Some of these actions are quite inexpensive and not in conflict with the farmers wishes to maximise production. Whether such measures are preferable to the removal of lead from petrol is a consideration beyond the scope of this study, as many economic and social issues are involved.

CHAPTER 8

CONCLUSIONS

It is apparent, from the work described in this thesis, that elevated levels of lead do occur in the air, pasture and soil adjacent to some major roads in the UK. This is as a result of the emission of particulates containing lead from the exhaust pipes of motor vehicles which use leaded petroleum fuel. The amount of lead released by an individual vehicle depends upon a number of factors, including the mode of driving, the road gradient, the age and capacity of the engine and the lead content of the petrol.

Detailed investigations of selected roadside sites, as part of this study, have provided an appreciable understanding of the factors controlling the pathways of vehicular lead in the environment. Once released into the atmosphere, the concentration of lead aerosols is determined by the atmospheric stability and diffusion processes. Upward dispersion of the exhaust gases rather than horizontal movement is probably the major mechanism that produces the rapid decline in the airborne lead concentration with distance from a road. Thus, the application of a Gaussian-type model of the dispersion of a pollutant plume in the atmosphere predicts that air lead concentrations 60 metres downwind of the M6 motorway at the study site are 50 per cent of those 10 metres from the road edge.

In contrast to dispersion processes, the deposition of atmospheric lead to the ground and other surfaces will have a minor influence upon atmospheric lead concentrations. Available evidence would suggest that only between 10 and 30 per cent of the emitted lead is deposited within 100 metres of a rural motorway. Although this constitutes a small part of the total lead released, the deposited material contaminates forage and other agricultural crops, and thus it is a pathway by which vehicular lead may reach farm products which are to be consumed by man.

The total amount of lead emitted by petrol-engined vehicles travelling on the M6 motorway shows a marked seasonal variation. This is because traffic flow during summer months is appreciably greater than in winter. However, measurements taken over 200 metres downwind of the motorway suggest that a corresponding seasonal variation in atmospheric lead levels does not occur, and indeed, peak concentrations were found during winter months. The Gaussian dispersion model partially accounts for this phenomenon by demonstrating the influence of atmospheric stability on the spread of a pollutant plume. The more stable atmospheric conditions in winter result in a less efficient vertical dispersion of exhaust emissions than during summer months. However, when horizontal air movements are incorporated into the model the results suggest that highest atmospheric lead levels occur in summer. This is, therefore, in contrast to the measured values of atmospheric lead undertaken in this study and by other workers. It is suggested that, perhaps, certain episodic conditions produce peak winter lead levels which cannot be predicted on the basis of traffic flow, wind speed and Pasquill stability categories as used in the model.

Lead particulates are removed from the atmosphere by a number of deposition processes. Moss bag studies of the deposition of airborne lead in the vicinity of the M6 motorway, have shown dry deposition to be the major process producing lead contamination of grassland. At a distance of 10 metres from the motorway edge, the rate of deposition of airborne lead to moss bags was between two and fifteen times greater than at a background site. The rate of lead deposition to the bags showed a marked seasonal variation at all survey points in the study area. Deposition rate was approximately ten fold greater during winter than summer months, both near and distant from the motorway. This seasonal variation could only partially be explained by the effect of changes in wind speed, mean daily atmospheric lead concentration and the diameter of the lead particulates; all factors which influence the rate of deposition to surfaces. Therefore, it is suggested that, in winter, short periods of relatively high lead deposition may occur within the four week exposure period of the moss bags. These periods might well correspond to the peaks in airborne lead concentration shown to occur at the 200 metres west air sampling site.

The rate of deposition of lead was found to decrease rapidly with distance from the motorway. In consequence, although elevated levels of lead occur in grass and soil near the motorway, the influence of vehicular emissions is slight beyond a distance of 50 metres from the road edge. Highest levels of soil and grass contamination were found downwind of the prevailing westerly winds, a finding to be expected.

In addition to surface contamination of the grass, the lead content of the herbage may be further enhanced by the uptake of this metal from the soil via plant roots, and its subsequent translocation to the leaves. It was found possible to distinguish between the airborne and non-atmospheric sources of lead by investigating the relationship between the deposition rates to moss bags and the grass lead concentration. The use of the correlation found between these two factors, in the manner described by Crump and Barlow (1980), showed that, in winter, airborne lead accounted for 60 to 80 per cent of the total lead content of the herbage. This was found at a number of sites situated at distances ranging from 10 to over 1,500 metres from the motorway. The remaining fraction of the lead in the grass shoots must have resulted from non-atmospheric sources, and is due to translocation of the metal from the plant roots.

The moss bag data concerning deposition rates of airborne lead also indicates that, during the summer months, surface contamination by atmospheric lead remains the major source of lead for herbage growing within approximately fifty metres of the motorway. However, at sites more distant from the road, non-atmospheric sources probably account for the majority of lead in pasture. These sources include the roots from which translocation may take place and the particles of soil on the surface of the plant, which have reached their position as a result of rain splash or contact of leaves with the ground. The presence of soil particles on leaf surfaces is of little significance for the lead content of winter pasture because, even at the background site, the grass lead level exceeds that of the top soil during this period.

Both near the motorway and at the background sites, the lead concentration of pasture grass showed a marked seasonal variation. Highest levels occurred during the winter months and were approximately ten fold greater than the minimum values found in summer. This seasonal variation corresponds quite closely to the changes in the rate of deposition of airborne lead to moss bags found at the study site.

The moss bag study has shown that at sites beyond a distance of fifty metres from the motorway, the seasonal change in the lead concentration of pasture grass occurs as a result of the variation in the rate of deposition of airborne lead. However, near the motorway, plant growth processes account for lead levels in summer pasture remaining at only a fraction of those found in winter. Thus, despite high traffic flow producing a deposition rate of airborne lead in summer comparable to peak winter values, the lead content of roadside grass showed a similar pronounced seasonal variation to that found at background sites.

The most important growth factor influencing the lead content of pasture was that of the life cycle of individual leaves and tillers. This determined the time for which a particle, deposited on the grass surface, remained associated with the plant canopy before being lost to the soil by the processes of leaf death and decay. In consequence of the greater longevity of leaves during winter than summer months, the same rate of lead deposition produces a higher level of lead contamination of winter pasture than of summer growth. This occurs as a result of the build up of deposited lead on the grass surface with time which has been shown not to be removed by rainfall or other weathering processes. Therefore, the continual process of turnover of organic matter in the grass sward determines a 'ceiling level' of lead contamination of pasture for a given long term deposition rate (i.e. several weeks). Other plant growth factors, such as the seasonal variation in biomass production, have been shown to have only a minor effect upon the lead content of herbage.

A further mechanism producing a seasonal change in grass lead concentration is that of the translocation of the metal from root to shoot in the plant itself. As the rate of grass growth slows in the autumn, the concentration of lead in the foliage, which has resulted from translocation of the element from the roots, increases at least three fold. This concentration of lead in plant tissues appears to remain constant during the winter months, until the leaves are replaced by new growth in spring which is of low lead content. However, the amount of lead from all non-atmospheric sources is of minor significance compared with airborne deposition except, perhaps, during the summer months at sites not influenced by emissions from the motorway. Thus, the uptake of lead from the soil plays only a minor role in the seasonal change in the lead content of pasture.

The occurrence of a large seasonal change in the lead content of herbage has important implications for the level of exposure of farm livestock to lead. This is as a result of the different management systems for each type of farm animal. Thus, sheep are more exposed to vehicular lead than dairy cattle and horses because they are kept at pasture during winter months when the grass is most highly contaminated. In contrast, dairy cattle and horses are stall fed with silage and other feeds of low lead content in winter, and only graze pastures during the summer months when grass lead levels are minimum.

Consideration of the amount of lead absorbed from the lungs and gastro-intestinal tract by an animal kept on a farm adjacent to the M6 motorway would suggest that no danger of clinical poisoning exists as a result of the vehicular exhaust emissions. In addition, there appears to be little likelihood of contamination of meat and milk products obtained from dairy cattle. Supporting evidence for this was provided by the presence of low levels of lead in bovine blood, milk, hair and faeces. It is, however, possible that the liver and kidney tissues of sheep grazing the roadside pasture may have elevated lead levels. The consumption of this offal by man is, therefore, a pathway by which lead additives in petrol may add to the total level of lead ingested. This, and other pathways by which lead in air may contribute to the total lead content of man's diet, is an area largely neglected by the recent review of lead and human health produced by Lawther et al. (1980). It would seem, therefore, to be an area worthy of further research to investigate fully the contribution that lead in the air makes to the total lead content of the human diet.

In areas where background lead levels in the environment are already high, such as near urban areas and some industrial premises, airborne lead arising from a major road may contaminate pasture grass to produce concentrations that could be detrimental to the health of grazing animals. Other workers have measured grass lead levels near the M4 motorway in a London suburb and these were found to have much higher lead concentrations than the samples examined in this study. Therefore, more general surveys of lead levels in pasture adjacent to major roads are necessary to fully assess the exposure of livestock grazing in such areas. Further research into the factors determining the rate of deposition of lead to plant surfaces may assist the development of a model to predict the lead content of roadside pasture based upon site characteristics, season and traffic flow.

From the results of this study, it is clear that, in rural agricultural regions, the release of lead from motor vehicles does not present a hazard to the health of farm livestock or to the quality of dairy products. However, the available evidence would suggest that liver and kidney tissues of sheep might have an elevated lead content as a result of grazing roadside pastures during winter months. This finding, however, remains to be confirmed by further research. If future studies should recommend that levels of roadside lead contamination are too high, there a number of practical and inexpensive measures that farmers and planners may implement to minimise exposure of livestock to lead. Alternatively, lead may be removed from petrol, although the economic and social considerations make this measure seem unlikely to be undertaken in the near future.

In summary, therefore, the aims and purposes of the study set out in Chapter 1 of this thesis have been achieved as follows:-

1. <u>Understanding the factors governing airborne lead levels and rates of deposition of lead near a motorway.</u>

   Meteorological factors have been shown to have an important influence upon air lead concentration and the rate of deposition of lead to plant and soil surfaces. In consequence, the highest rate of deposition of atmospheric lead to plant surfaces occurs in winter when the traffic flow, and hence the total amount of emitted lead, is least. Further research is required to assess the relative importance of wind speed, atmospheric stability and particle size upon airborne lead levels and the deposition rate of atmospheric particulates.

2. Factors determining the level of contamination of pasture by vehicular lead

During the winter months, the effect of vehicular emissions from the motorway upon the lead content of pasture is detectable up to a distance of 200 metres downwind of the road. However, in summer the lead concentration of pasture is not elevated beyond a distance of 50 metres. Within this roadside zone, the majority of the lead associated with the pasture has resulted from the deposition of airborne particulates.

The rate of deposition of atmospheric lead near the motorway did not show as great a seasonal variation as was found at background sites. However, plant growth processes resulted in levels of lead in summer pasture being considerably less than during winter months. This effect would appear to be largely due to the rapid turnover of organic matter during the growing season which restricts the build-up of lead on individual leaves in the grass sward. Thus, for a given rate of deposition of lead to vegetation, a ceiling level of lead contamination of herbage exists which depends upon the rate of leaf turnover. Rainfall and other weathering processes have little effect upon the lead content of the pasture.

In areas not directly influenced by lead emissions arising from the motorway, airborne deposition remains the major source of lead to pasture during the winter months. However, in summer, deposition of atmospheric lead is slight and the lead present in pasture must have resulted from one of the following two sources:

(i) soil particles on the leaf surfaces which are present as the result of rain splash or contact of leaves with the ground, or

(ii) the uptake of lead from the soil by plant roots and its translocation to the shoots.

3. Toxicological hazard of vehicular lead

Present levels of lead in roadside pasture in rural areas are not a threat to the health of farm livestock. In addition, it would seem unlikely that meat and milk products from dairy cattle are contaminated by lead as a result of grazing pasture in the vicinity of a motorway. However, liver and kidney tissues of sheep, which may be consumed by man, probably do have an elevated lead content as a result of the animals grazing roadside pastures during the winter months.

4. Measures to minimise exposure of farm livestock to vehicular lead

A number of measures could be implemented by farmers and planners to reduce the exposure of farm animals to roadside lead. The most necessary and effective action is to cease the grazing of roadside fields during the winter months. Further measures may be required where background levels of lead in soil and herbage are high due to contamination from other sources such as industrial premises and urban regions. Thus, it is recommended that further research should be undertaken to investigate roadside farms which may be influenced by additional sources of lead. Further it is suggested that a sampling programme should be set up to monitor the effect that future changes in the lead content of petrol and variations in traffic flow may have upon the level of lead contamination of roadside pasture.

Therefore, the general conclusion of this thesis is that present levels of contamination of roadside pasture by vehicular lead are not a major problem for farmers in the UK. However, local circumstances do exist which warrant further investigation because of the possible contamination of offal which is to be consumed by man.

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APPENDIX 1

SOIL ANALYSIS REPORT (M.A.F.F.)

Aston University

APPENDIX 2

THE LEAD CONTENT OF PASTURE GRASS

NEAR THE M6 MOTORWAY AT PEOVER, CHESHIRE

DURING 1970-1976

(THE ASSOCIATED OCTEL CO. LTD).

APPENDIX 3

PHYSICAL SOIL PROPERTIES AT THE M6

STUDY SITE NEAR SANDBACH, CHESHIRE

DETERMINATION OF PHYSICAL PROPERTIES

OF SOIL FOR THE M6 STUDY SITE

Method

Soil samples taken from the M6 study site in the manner described in Chapter 1 were investigated to determine the soil pH, organic carbon content and moisture content. The following techniques were employed:

A. Soil pH:

1) 20 g of fresh soil was weighed into a 250 ml beaker.
2) 100 ml of distilled water was then added and the mixture stirred vigorously.
3) The pH of the mixture was then determined by using a Pye Unicam (Model 293) pH meter.

B. Soil Moisture and Loss on Ignition:

1) A weighed crucible was filled with fresh soil and reweighed.
2) The damp soil was dried in an oven for 24 hours at a temperature of 100°C.
3) The crucible was reweighed and the weight of water lost by drying calculated.
4) The dry soil was then placed in a furnace at a temperature of 500°C for 12 hours.
5) The soil was then reweighed and the loss of organic matter due to ignition was calculated.
6) Both the weight of water and organic material lost from the soil was calculated as a percentage of the weight of the remaining inorganic matter.

TABLE A-1

pH of Surface Soil at the M6 Study Site

Date of Sampling	Soil pH				
	* 1,500 m West	** 0 - 100 m West	** 0 - 100 m East	* 450 m East	* 1,000 m East
1977					
October	6.8	6.7	6.9	-	-
November	6.5	7.1	7.2	7.5	-
December	6.5	6.9	6.9	7.6	-
1978					
January	6.6	7.2	6.9	7.5	-
March	6.8	7.0	7.1	7.4	7.2
April	6.4	7.2	6.7	7.5	7.2
May	6.3	7.1	6.7	7.4	7.2
June	6.3	7.0	6.7	7.5	7.1
July	6.6	6.9	7.0	7.6	7.5
August	6.9	7.3	7.2	7.5	7.4
September	6.6	6.8	6.5	7.3	7.0

* = Mean of 5 samples.

** = Mean of samples at 0, 5, 10, 20, 30, 40, 50, 75, 100 m from M6 fence.

TABLE A-2

Organic Carbon Content of Surface Soil

at the M6 Study Site

Date of Sampling	Organic Carbon Content as Percentage of Mineral Weight of Soil				
	1,500 m West	0 - 100 m West	0 - 100 m East	450 m East	1,000 m East
1977					
October	10	11	11	-	-
November	9	9.5	9	7	-
December	10	10.5	8.5	7	-
1978					
January	8	11	9	11	-
March	11	11	10.5	10	13
April	10	12	12.5	11	13
May	8.5	10	13	10	18
June	10	10	11.5	12	17
July	13	10	11	5	13
August	11	12.5	9	8	15
September	12	10.5	8.5	9.5	9

TABLE A-3

Moisture Content of Surface Soil at the M6

Study Site During 1977/8

Date of Sampling	Moisture Content as Percentage of Soil Mineral Weight				
	1,500 m West	0 - 100 m West	0 - 100 m East	450 m East	1,000 m East
1977					
October	41	48	49	-	-
November	43	43	46	40	-
December	46	44	45	35	-
1978					
January	46	52.5	47	54	-
March	61	58.5	53	55	67
April	53	63.5	63.5	58	68.5
May	41	44.5	60	52	73
June	25	8	23	16	35.5
July	55	46	38	26	68
August	58	74	50	48	68
September	59	49	42	43	46

APPENDIX 4

THE EFFECT OF THE GROWTH RATE OF GRASS UPON ITS LEAD CONTENT (FIELD PLOT EXPERIMENT)

A FIELD INVESTIGATION ON THE EFFECT OF THE GROWTH RATE OF GRASS UPON ITS LEAD CONTENT

Background

It was considered likely that the structure and yield of a grass sward might have a significant influence upon the catchment of air-borne particulates. In addition, a more productive area of grassland may result in a dilution of the fallout of airborne material. Thus, growth rate may influence the lead content of pasture near major roads where vehicular lead is a contaminant.

Method

In the autumn of 1978, a 15 metre square of grassland, situated in the corner of a permanent pasture and adjacent to the motorway fence (Figure 1-3), was enclosed by a barbed wire fence to prevent access of farm animals. The area was further protected by an electric fence which proved successful in deterring dairy cattle from entering the site.

The experimental pasture was divided on a grid basis into 36, one metre square plots with spaces for access between. Figure A-3 shows the layout of these plots, divided into six rows and six columns each containing six plots of grass. The columns were divided into two blocks of three in order to provide replicate plots for each of the six distances from the motorway (i.e. the rows). Within each row of a block the plots were assigned at random to one of three classes as shown in Figure A-3. These three classes, or treatments, involved the use of different application rates of nitrogen fertiliser and are also given in Figure A-3.

In April 1979, all 36 of the experimental plots were harvested. This was carried out by the use of hand shears to remove all the herbage present 1 cm above the ground surface. The grass from each plot was placed in a labelled polyethylene bag and returned to the laboratory. When harvesting was completed all plots were given a standard dressing of phosphate and potash (375 kg/ha). The plots were then treated with nitrogen fertiliser (105 Wiles 13 : 13 : 20) at the levels of application shown in Figure A-3.

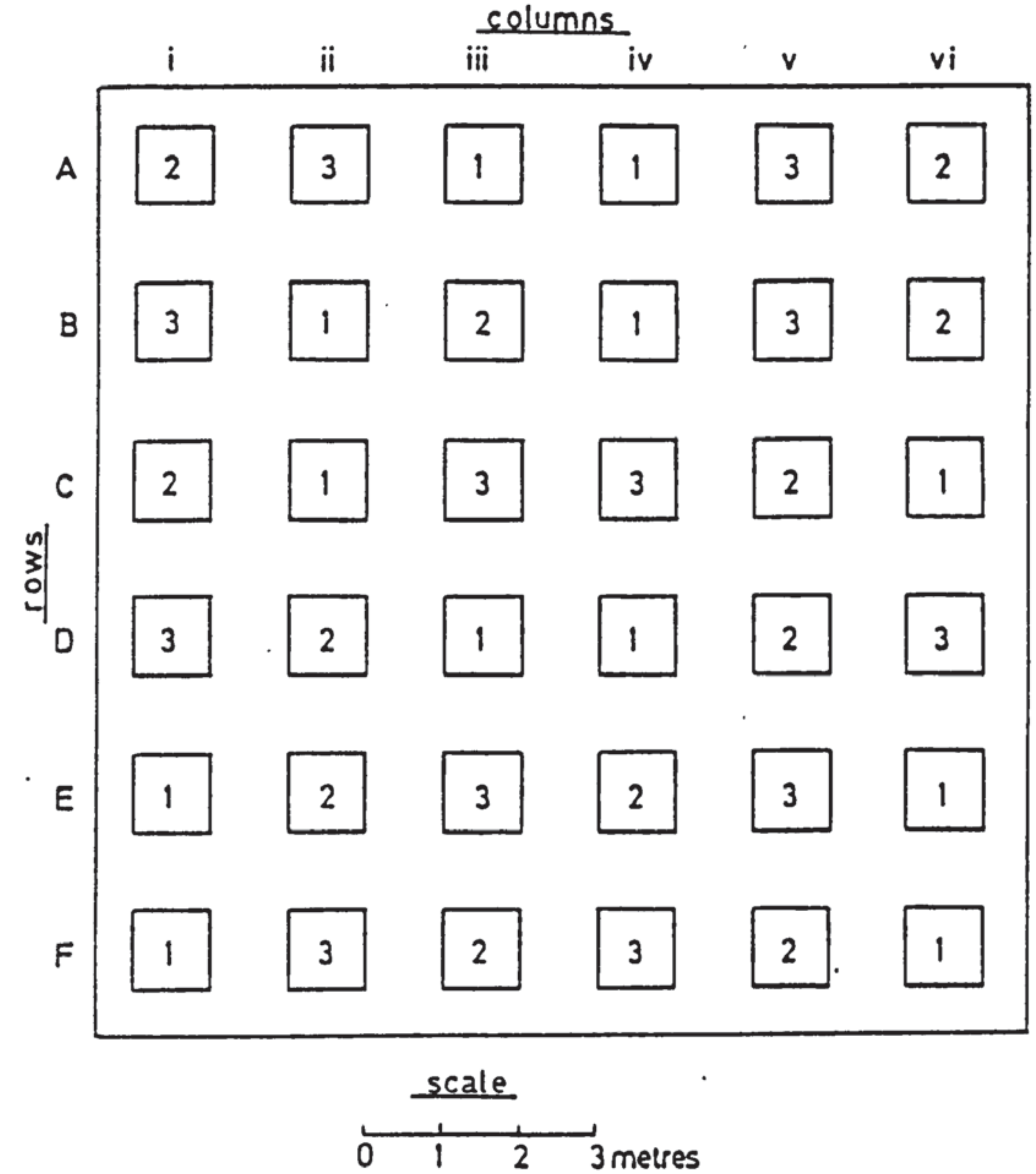
Subsequent harvesting of the plots was carried out in May, August, and November of 1979. Following each cut, the nitrogen fertiliser was again applied. An exception to this was after the August cut when the class '1' plots did not receive any additional fertiliser. This, it was hoped, would maximise the differences in yield between the three treatments at the November harvest. After each harvest was complete, the grass growing between the plots was also cut to ensure that it did not affect the rate of deposition of airborne lead to the experimental areas.

Once returned to the laboratory, the fresh weight of the grass harvested from each plot was recorded and sub-samples were taken to determine the dry weight and metal content. The dry matter yield was found by measuring the weight loss from fresh grass when dried in an oven at $80^{\circ}C$ for 24 hours. Lead and zinc content of the grass was determined in the manner described previously in Chapter 1 of this thesis.

Results and Discussion

The data obtained at each harvest for the dry matter yield and metal content of the grass was analysed statistically using a one-way analysis of variance. Table A-4 shows the effect of fertiliser treatment

FIGURE A-3 Diagram of Field Plot Experiment to Investigate the Effect of Growth Rate on Pasture Lead Content



Fertiliser (nitrogen) Application after each harvest:

1 = 50 Kg/ha

2 = 150 Kg/ha

3 = 300 Kg/ha

TABLE A-4

Results of a Statistical Analysis to Determine the Effect of Fertiliser Treatment on the Yield and the Lead and Zinc Contents of Pasture Grass

Harvest (1979)	Significance of 'F ratio' for differences between fertiliser treatments		
	Yield	Lead Content	Zinc Content
3 April	NS	NS	NS
30 May	0.001	NS	0.01
2 August	0.05	0.2	NS
29 November	0.01	NS	NS

NS = not significant at better than the 0.2 probability level.

on the yield and the lead and zinc concentrations of the pasture grass. It may be seen that, subsequent to the application of fertiliser after the April harvest, the three different nitrogen treatments produced a significant difference in the plot yields. This did not, however, result in a significant difference in the lead content of the pasture, even though there is a tendency for a greater lead concentration in the well fertilised plots at the August harvest. Possibly the taller fertilised grass was a more efficient collector of airborne lead in this period, but the results cannot be regarded as conclusive.

Fertiliser treatment had no effect on the zinc content of the grass except during May when levels were greater in the well fertilised grass. It is not known if this is the result of the deposition of airborne zinc or due to uptake of the trace element from the soil. The latter would seem to be the most likely source of the zinc.

With regard to the effect of growth rate upon pasture lead content, it should be noted that grass lead levels were low during the experimental period. The strong westerly winds during the summer of 1979 resulted in lead concentrations in the roadside plot of only about 5 μg/g in May and August, rising to between 40 and 50 μg/g in November. There was, however, a significant decline in the lead content of the grass with distance from the road at all harvests. Table A-5 shows the results of the statistical test applied to assess the significance of the change in lead concentration with distance. The Table also shows that there was no statistically significant variations in yield with distance from the motorway, and that a gradient in the zinc concentration of grass existed only during April.

Conclusion

The application of fertiliser to control the growth rate of

TABLE A-5

Results of a Statistical Analysis to Investigate Changes in Yield and Trace Metal Content of Pasture with Distance from a Motorway

Date of Harvest (1979)	Significance of 'F ratio' for variation with distance from the motorway		
	Yield	Lead Content	Zinc Content
3 April	NS	0.05	0.01
30 May	NS	0.01	NS
2 August	NS	0.2	NS
29 November	NS	0.2	NS

NS = not significant at better than the 0.2 probability level.

perennial ryegrass had little or no effect upon its lead content. It is possible that growth rate would have a more significant influence on the lead concentration in a more contaminated environment, where deposition of airborne lead is greater than in the present study. The reason for this is that uptake of lead from the soil may have been the major source of lead to the pasture in this experiment, at least during May and August. The results for November, when the grass was quite heavily contaminated by airborne lead suggest, however, that this is unlikely to have been the case. It would seem, therefore, that the pasture yield is not an important factor determining the lead content of herbage.

APPENDIX 5

PUBLISHED PAPERS