STUDIES OF ATMOSPHERIC POLLUTION DUE TO LEAD IN THE VICINITY OF URBAN MOTORWAYS

by

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SUMMARY.

Lead levels in the atmosphere and in topsoil or dust increase as one approaches cities, due to the greater use of petrol driven vehicles. The building of an urban motorway through the Gravelly Hill district of Birmingham increased the traffic density of that area. The effect of the opening and use of the interchange on atmospheric lead concentrations was studied. An equation to predict the pollution level at a reference site from two weather variables, wind speed and temperature, and traffic was derived. Also the equations relating seven remote sites to the reference site were calculated.

Three houses were studied to check the degree of penetration by the lead and particulate aerosols. One property had double glazing which enabled the effectiveness of this as a barrier to particulate aerosol penetration to be studied. The lead levels in the blood of some inhabitants of these houses were determined and found to lie in the normal range.

The particulate aerosol was collected on filter papers, some of which were observed under high magnification by an electron microscope. This, coupled with x-ray energy spectrometry, has enabled titanium, vanadium, zinc and lead rich particles to be identifiable by their size and shape.

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CHAPTER 1. INTRODUCTION

1.1 LEAD IN.

Elements comprising less than 0.01 per cent of an organism are commonly called trace elements. Of these, lead is ninth in order of magnitude of abundance in sea-water, and seventh in man (exceeded by Fe, Zn, Cu, Rb, Sr, and Al), I.C.R.P. (1960). Some are essential for optimal biological function, although large amounts of essential or non-essential trace elements may be toxic, ENGEL (1971). Lead has not been found to have an essential function and appears to have disease effects at all concentrations in human-beings. Thus it is necessary to identify and control the conditions that cause exposure. Man has evolved in the presence of a certain amount of inorganic lead from natural sources, PATTERSON (1965). Therefore, assessment of the threat from environmental pollution needs to be considered in relationship to background levels. The natural quantity of lead mobilized annually is c.180 kt, whereas c.2000 kt were mined in 1970, BRYCE-SMITH (1971b). Hence, lead is becoming available to all who handle and process the ore and to those who work with the metal mechanically and chemically. The number of people who work with lead, as the element, are few and their working conditions are carefully controlled. However, with the advent of the motor car, the distribution of lead throughout the biosphere has occurred.

Lead is put into motor fuel - petroleum, as a mixture of alkyllead compounds, mainly as tetraethyl- and tetramethyllead. These are added to prevent knock, the shockwaves caused by uncontrolled release of chemical energy in the engine. This it seems to do by an inhibitory mechanism, which affects the rate of oxidation of petrol in the combustion chamber, ROSS & RIFKIN (1956). The addition of alkylleads enable one to obtain maximum performance from an engine in relation to its size and weight, by having a high compression ratio. Removal of lead would reduce performance and increase fuel consumption, along with exhaust emissions. (Substitution of lead by aromatic compounds may enhance the discharge of cancer producing material).

Of the lead that is burnt in the engine, about 50 per cent is emitted from the tailpipe as an aerosol of inorganic lead particles. These are in the size range of stable atmospheric aerosols, ROBINSON & LUDWIG (1967) which leads to the widespread dispersion of lead compounds.

The penetration of man, along with motorized transport, to all parts of the world has ensured the global production of such aerosols. These are a source of lead to man directly, by inhalation, and also, after deposition, through the food chain.

The exposure of people to increasing levels of lead in air, food and water may cause poisoning. Even before the onset of toxic symptoms, it appears that lead may have many undesirable effects on human biochemistry. Therefore all routes of lead delivery to the body should be monitored and controlled where feasible.

The building of an urban motorway and interchange in residential areas of Birmingham gave rise to concern regarding its possible effects on the health of the local population. The Public Health Committee of Birmingham council commissioned a survey of atmospheric lead levels in the vicinity. The following is a result of that study conducted by the Department of Chemistry, Aston University.

1.2 NATURAL BACKGROUND LEAD LEVELS IN AIR

The natural level of lead in the atmosphere is difficult to assess. According to PATTERSON (1965) there was a calculable level of airborne lead, before man's industrial history of c. 0.0005 $\mu g\ m^{-3}$. This derived from volcanic dusts, soil erosion and aerosolic sea salts. Current values that may be used as background levels of lead in the atmosphere come from the mid-Pacific and from inland remote areas. The former has been determined by CHOW, EARL & BENNETT (1969) and a range of 0.0003 - 0.007 $\mu g\ m^{-3}$ found, the average being 0.003 $\mu g\ m^{-3}$. Levels measured in other remote places fall in the range 0.02 - 0.6 $\mu g\ m^{-3}$, with a mean of c. 0.05 $\mu g\ m^{-3}$, CHOW & EARL (1970), and JOST, MULLER & JENDRICKE (1972). These are the levels to be expected in the absence of local vehicular traffic. Such areas are usually remote from human habitation and bear little resemblance to the background levels experienced by most populations of industrialised countries. The air lead burden of rural environments is generally reported to lie between 0.1 - 0.5 $\mu g\ m^{-3}$, with a mean of 0.3 $\mu g\ m^{-3}$, TER HAAR (1972), LANDAU, SMITH & LYNN (1969) and TEPPER (1971). Our own measurements at a site in the Welsh countryside suggested an average level of 0.18 $\mu g\ m^{-3}$, Table 10, whilst in Aberystwyth a concentration of 0.1 $\mu g\ m^{-3}$ was found, BUTLER & MAC MURDO (1974a).

Hence, it seems that due to the production of lead aerosols by the ubiquitous motor car, the atmospheric load increases in proportion to the concentration of such sources, CHOLAK, SCHAFER & STIRLING (1961). Natural air and that remote from man's activity have lead burdens of 1 - 2 orders of magnitude smaller than those taken to represent minimum exposure to Western man. This latter occurs in rural areas, though it should be noted that in industrialised nations, such as Britain, the majority of people live in the residential areas around cities. The atmospheric lead levels of such suburban conurbations are higher again than those found in rural districts, commonly reported as being c. 1 $\mu g\ m^{-3}$. The range of literature values is small, varying from 0.4 $\mu g\ m^{-3}$, CHOW & EARL (1970), to 2 $\mu g\ m^{-3}$, DAINES, MOTTO & CHILKO (1970).

1.3 NATURAL LEAD LEVELS IN WATER

The natural level of lead in water is as indefinable as that in air. The concentration will vary from district to district depending upon the geological character of the water-shed area. PATTERSON (1965) has estimated the average natural level (that present before man's mining activity) to be 0.5 $\mu g\ l^{-1}$. This agrees well with the background concentration reported by ABDULLAH & ROYLE (1972) for Welsh river water of 0.7 $\mu g\ l^{-1}$. Where leaching had occurred, from known metallic veins such as in the Ystwyth and Twymyn valleys, levels of between 2 - 17 $\mu g\ l^{-1}$ may be found. Lead may also find its way into river water through industrial processes. Although the rivers themselves were not analysed, the Bay of Liverpool and the Bristol Channel have concentrations 40 - 60 times greater than average sea-water (0.03 $\mu g\ l^{-1}$), ABDULLAH, ROYLE & MORRIS (1972). The radioactive fingerprint technique used by TINKER (1971) to support the argument that lead pollution in cities is mainly from motor vehicles has not yet been applied to the lead found in these waters. However, it is known that oceanic sediment has a very different Pb^{206}/Pb^{204} ratio to that of snow, due most probably to the age of the sediment, TATSUMOTO & PATTERSON (1963).

Most water consumed by Western man is piped to him from reservoirs, thus the natural level does not represent the background concentrations to which he is currently exposed. One standard by which the lead content of tap water may be assessed is the WHO specification for drinking water. This was 50 $\mu g\ l^{-1}$ (1963) but has recently been raised to 100 $\mu g\ l^{-1}$, REED & TOLLEY (1973). Conflicting evidence about plumbosolvency and quality of tap water has been presented, REED & TOLLEY (1967) and CRAWFORD & MORRIS (1967). In studies of potable water, LEWIS (1965) and TOLLEY & REED (1970) found most levels fell below the recommended concentration.

1.4 NATURAL LEAD LEVELS IN FOOD

The natural level of lead in foodstuffs is low. The concentrations calculated by PATTERSON (1965) are about 0.03 ppm for animal tissue and 0.4 ppm for vegetation. This has changed since the motor car produces a widespread lead aerosol and the nutrient sources of potential foods may be contaminated over large areas. Plants may contain abnormal levels of lead, due to uptake from soil and water or by deposition from the atmosphere without displaying toxic symptoms, TOLAN & ELTON (1972). The former source is small since plants appear to have a natural selectivity against root uptake of lead, though this may be due to soil factors. Lead is bound in soil by an ion exchange mechanism, and absorption depends upon pH, carbon and phosphate content, crop type and date of harvest, TER HAAR (1972). The latter source, atmospheric deposition, may be significant as grass growing near roads has been found to contain elevated levels of lead, CANNON & BOWLES (1962) and CHOLAK, SCHAFER & YEAGER (1968). Deposited lead may be retained to the extent of 10 per cent by foliage, 1 per cent by fruit and 0.1 per cent by grains, PATTERSON (1965).

Concentrations reported are generally below 0.5 ppm wet weight for fresh foods, SCHROEDER & BALASSA (1961) and BRYCE-SMITH & WALDRON (1973), but preparation and canning could affect this. The British food regulations, H.M.S.O. (1961), permit a maximum lead concentration in most foods of 2 ppm wet weight. This has been questioned, BRYCE-SMITH & WALDRON (1973) not only on the basis of adult intake, but also in relation to children. Following a survey of lead in food, H.M.S.O. (1972), the permitted maximum for baby foods was lowered to 0.5 ppm of lead.

1.5 NATURAL LEVELS OF LEAD IN THE BODY

Natural levels of body lead may be found by correspondence between the levels of lead in the environment and man. It has been calculated to be about 2 mg for a 70 kg frame, PATTERSON (1965). The present average body burden is reckoned by the same author to be 200 mg.

Data acquired from 150 accidental deaths in the USA suggest a current burden of 120 mg/70 kg, SCHROEDER & TIPTON (1968). The vast majority of this lead is laid down in the bones but is not inactive there, MANTON (1973). The relatively heavy tissues in the body which clean the body fluids, also concentrate lead. The liver contains about 1.7 ppm wet weight and the kidneys c. 1.1 ppm. Hair also carries a heavy concentration of lead, c. 50 ppm, SCHROEDER & TIPTON (1968). These authors also found that the levels in many tissues of American subjects increased with age, whilst samples from other parts of the world did not. The natural level of lead in the blood has been estimated by PATTERSON (1965) to be 0.025 μg per 100 ml whole blood. Average levels found recently are c. 20 μg per 100 ml but are subject to dietary, occupational and even seasonal influences.

1.6 SOURCES OF LEAD IN THE ENVIRONMENT

Lead is brought into the biosphere by mining or industrial processing in quantities many times those mobilised by natural weathering, BRYCE-SMITH (1971b). Uses of lead solders and batteries release lead into different phases of the environment due to inefficiency in processing. The burning of coal and lead contaminated wastes also gives rise to the distribution of lead. Some emissions are unavoidable but they are generally localised. More widespread contamination of food and water can occur due to leaching from the lead glazing of containers, and from the solders used in sealing the joints of water pipes and tins. By radioactive fingerprinting, general pollution in all phases appears to be due to the use of tetraalkyl leads in motor vehicles, AULT, SENCHAL & ERLBACH (1970) and CHOW & EARL (1970).

United Kingdom petrol sales have increased exponentially to 14,000 kt in 1970, BRYCE-SMITH (1971a). Much of the lead added to petrol is emitted in the exhaust - estimated range from 25 to 80 per cent, ENGEL, HAMMER, HORTON, LANE & PUMBLEE (1971) and HALL (1972). The rate of emission depends upon the age of the engine and exhaust system, the grade of petrol, the running mode of and the load on the engine.

This lead in the exhaust, which is in the particulate form, is usually of such a size that it forms a stable lead aerosol, ROBINSON & LUDWIG (1967). The compounds emitted are the chlorobromide and two complexes of ammonium chloride with lead (II) chlorobromide. If phosphorous additives are present in the petrol up to 20 per cent of the emission can be present as the complex: $3\ Pb_3\ (PO_3)_2.\ Pb\ Cl.Br$. These compounds undergo atmospheric chemical reactions to form the oxide and oxy-carbonate of lead. It should be noted that the classic solubility, in distilled water, of these compounds, is low, but in biological media solubility can be markedly enhanced, SMITH (1971).

1.7 ABSORPTION OF LEAD BY THE BODY

Lead enters the body mainly via the gastro-intestinal and the respiratory tracts. The former is fed by the diet and by drinking water; it appears to pass about 10 per cent of the dietary intake of lead into the blood stream, ALEXANDER (1973) and THOMPSON (1971) though this may reach 50 per cent in children, BRYCE-SMITH & WALDRON (1973). The diet is the principal source of uptake in the general population. No foodstuff is known to concentrate lead selectively but all foods contain some trace. Absorption of the metal by plants through their roots depends upon soil conditions, and is retained from atmospheric deposition according to crop type. Storage and cooking of foods may also increase the dietary content of lead due to leaching and abrasion of lead alloys and glazes used in containers for these processes, SHUCK & LOCKE (1970) and PATTERSON (1965). In raw foods, levels reported by SCHROEDER & BALASSA (1961) and TOLAN & ELTON (1972) were found to lie between 0.1 - 1.0 ppm wet weight. Permitted levels are 10 ppm in the U.S.A., ENGEL (1971) and 2 ppm in the U.K., H.M.S.O. (1961). Thus most fresh precooked foods studied comply with these, suggesting that deposited lead is not yet at a critical level. However, such surveys miss two major sources of population exposure. The Ministry of Agriculture, Fisheries and Food, H.M.S.O. (1972) and ARCHER (1973) analysed some tinned foods and found elevated levels in these. Also since children eat more food in relation to their body weight and have a higher metabolic rate, KING (1971) these regulations may still leave them at risk.

Drinking water is recommended to contain less than 0.1 ppm of lead and in two US surveys, SCHROEDER & BALASSA (1961) and ENGEL (1971) the concentration generally did not exceed 0.05 ppm. Two British surveys, however, H.M.S.O. (1972) and REED & TOLLEY (1973) report an average level of 0.16 ppm in Glasgow and 53 per cent of samples analysed greater than 0.1 ppm respectively. Lead is more soluble and apparently a more effective toxin in soft water. Fish, whose T.C.$_{50}$ (the toxic concentration that kills 50 per cent of the fish) in hard water is 1 - 5 ppm of lead acetate, have a T.C.$_{50}$ in soft water of 0.1 - 0.5 ppm. The water for Birmingham consumption comes from Wales and at 100 ppm hardness is moderately soft, HAMILTON & MINSKI (1973). Of dietary lead in the U.K. c. 10 per cent is attributed to water and beverages.

The atmosphere may also be a significant source of lead. Estimates of the daily lead burden from this source vary from 6 per cent of the total, KEHOE (1959) and KING (1971) to 20 per cent, PATTERSON (1965) and MONIER-WILLIAMS (1949). Lead is emitted from motor vehicles in particles with an average mass median diameter of 0.3 μm, at a concentration of c.15000 μg m^{-3}, MUELLER, HELWIG, ALCOCER, GONG & JONES (1963) and LEE (1972). Thus most are in the range capable of lung penetration, and it has been found that about 40 per cent of particles less than 1 μm are deposited in the lung, NOZAKI (1966). The halides typically exhausted are quite soluble in serum and give rise to an absorption rate of inhaled lead of 30 - 80 per cent, FUGAS, WILDER,PAUKOVIC,HRSAK & STEINER-SKREB (1972) and SCHROEDER (1971). It should be noted in this context that inhaled lead absorbed by the lungs or by the gastro-intestinal tract after cilliary action, is often associated with other chemicals; the interactive, disease effect of which is unknown.

Other sources of lead to the body, besides occupational exposure, which in terms of numbers is limited, include smoking. Cigarettes have about 1 μg of lead each in their smoke, PATTERSON (1965) and ENGEL (1971), and may damage the lungs in such a way as to enhance the uptake of lead, since it has been reported that smokers tend to have higher blood levels than non-smokers, McLAUGHLIN & STOPPS (1973). Roadside dust may also prove a significant source of lead to children. Concentrations of lead in urban dusts vary from 700 - 3000 ppm and the licking of dirty fingers or eating dirty sweets could contribute a very substantial proportion of any recommended safe level of ingestion for children, McCALDIN (1965) and DAY, HART & ROBINSON (1975).

1.8 ASSESSMENT OF BODY DOSAGE OF LEAD

To assess the dosage of a pollutant as a biological agent, it is necessary to consider the amount absorbed against the time for which it is biologically active. For lead, this is about six months, so when evaluating low-dose response relationships long term averages are probably the most meaningful, FIRST (1969).

The lead content of the diet has been found to vary widely between 100 - 3000 μg day $^{-1}$, SCHNEIDER (1970) and THOMPSON (1971), averaging about 300 μg day $^{-1}$. Although this average lies below the WHO (1972) limit of 3000 μg per week, the possible range can leave many at risk. Ten per cent of this intake is absorbed, BERMAN (1966) and BARLTROP & KILLALA (1967), giving rise to a body burden from food of c.30 μg day^{-1}.

An adult is estimated to breathe an average 20 m^3 per day, which provides about 6 μg day^{-1} (0.4 - 10.0 μg day^{-1}), depending on the ambient atmospheric level. This is assuming an average rate of absorption after inhalation and deposition of 50 per cent. In addition to this, cigarettes can provide a further 5 - 15 μg day^{-1} depending on the number consumed.

From the literature surveyed the daily intake is about 400 μg day^{-1} of which the quantity absorbed is in the range 25 - 90 μg day^{-1}, averaging about 40 μg per day. From both the lungs and the gastro-intestinal tract the lead passes into the blood stream, by which means it circulates around the body and reaches equilibrium between the organs of the body that absorb it.

The lead intake is excreted largely in faeces, at a rate of c.200 μg day^{-1}, and urine, c.40 μg day^{-1}, MONIER-WILLIAMS (1949) and KEHOE (1961). Some more of the absorbed lead is lost into sweat and hair, but overall c.20 - 30 μg day^{-1} is retained largely in bones and heavy tissues, where it has a half life of between 2 - 3 years, BLACK (1962).

The total body burden of lead is probably 100 - 200 mg for a 70 kg framed adult, KEHOE (1961) and SCHROEDER & TIPTON (1968), and has been reported to increase with age, NUSBAUM (1965). At these levels no overt clinical symptoms have become apparent, which may be a reflection of the multiplicity of symptoms or the nature of clinical diagnosis.

1.9 INDICATORS OF BODY DOSAGE OF LEAD

The most widely used indicator for body burden is the blood lead level. All organs of the body contain some lead, but it is most readily stored in bone tissue by the displacement of calcium from its phosphate, GOLDSCHMIDT (1958). Ninety per cent of the body lead is therefore found in the skeleton and is relatively inert, though it is capable of mobilization - particularly in times of stress - and may affect Ca^{++} metabolism, SCHROEDER & TIPTON (1968) and BERMAN (1966). The level of lead intake that may affect health, by unbalancing body chemistry, needs to be assessed using a readily accessable tissue, i.e. blood, faeces or urine. Of these, urine and faeces levels are elevated and reduced more rapidly than those in blood. If raised, blood lead concentrations may remain high for a period of weeks, compared to one of days for urine and faeces, BARLTROP & KILLALA (1967). Numerous reports of blood lead measurements suggest an average level of 20 μg per 100 ml whole blood. Clinical lead poisoning has not been observed below 80 μg per 100 ml in adults and 50 μg per 100 ml in children, ENGEL (1971). However, it has been noted that the average level is nearer that at which clinical symptoms of poisoning are found than for other pollutants, BRYCE-SMITH (1971b). Acceptable ranges of blood lead level have been described for adults, LANE (1968), although children, who appear to be the portion of the population at greatest risk, are often judged by the same standards. The probability of sequelae to lead poisoning in children is high, c.80 per cent, but depends largely upon the symptoms by which the poisoning is recognised, BYERS & LORD (1943) and PERLSTEIN & ATTALA (1966). It should be noted that there is no evidence in the literature concerning a follow up study of adults with lead poisoning. One of the consequences of lead poisoning in childhood, reported by the aforementioned authors, is mental retardation. This is supported by MONCRIEFF, KOUMIDES, CLAYTON, PATRICK, RENWICK & ROBERTS (1964) who measured blood levels in "normal" and "retarded" children, but subsequent studies provide conflicting evidence, GORDON, KING & MACKAY (1967), MILLAR, BATTISTINI,CUMMING, CARSWELL & GOLDBERG (1970) and BRYCE-SMITH & WALDRON(1974).

The reported lead levels in blood have been "steady" for a number of years, without obvious effect, MILLS (1971), although they are about 100 times the natural concentration in human blood. Present concentrations appear to be correlated with potential for atmospheric exposure, GOLDSMITH & HEXTER (1967) and ENGEL (1971), but require careful interpretation, since they are affected by current disease or exposure to lead, time interval since disease or exposure, and season, BEATJER (1959), GORDON, KING & MACKAY (1967), and McLAUGHLIN, LINCH & SNEES (1973). The levels do appear to bear direct relation to enzymic activity in blood cells, due to the attraction of lead for sulphydryl groups, which associates 90 per cent of the lead in the blood with the erythrocyte fraction, BERMAN (1966). d-Aminolevulinic acid dehydratase (d-ALAD) has a linear response to blood lead over a range of 5 - 95 μg per 100 ml, with a correlation coefficient of -0.9, HERNBERG & NIKKANEN (1970). This inhibition may not have a physiological effect, SECCHI, ALLESSIO & CAMBIAGHI (1973), although the enzyme catalyses the production of porphryns, precursors in the biosynthesis of haeme and anemia is one of the symptoms of excess lead retention. HERNBERG & NIKKANEN (1970) suggest that d-ALAD activity may be a better indicator of lead exposure than blood lead. This has yet to be evaluated.

1.10 EFFECTS OF LEAD ON THE BODY

The effects of elevated levels of lead in the body are many and various in character. They appear as abdominal cramps, constipation, loss of appetite, aememia, fatigue, insomnia, motor-nerve paralysis and encephalopathy, ENGEL (1971). When diagnosed in childhood, it may lead to subsequent kidney disease, mental retardation, recurrent seizures, cerebral palsy, or optic nerve atrophy, most of which can be attributed to inhibition of growth of the central nervous system, BYERS & LORD (1943), PERLSTEIN & ATTALA (1966) and EMMERSON (1969). Investigation of the possibility of lead poisoning in children has been recommended in cases of pica, sudden changes of behaviour - such as fits of depression or irritability - or other inexplicable phenomena like hyperactivity or anemia, MONCRIEFF,KOUMIDES,CLAYTON, PATRICK, RENWICK & ROBERTS (1964) and BRYCE-SMITH & WALDRON (1974).

The body burden, which seems to increase with age, BARRY & MOSSMAN (1970), can be transmitted from mother to child. This may cause spontaneous miscarriage as evidenced by the occurrence of higher levels of lead in stillbirths than in children between 0 - 1 years of age. Generally, women appear more susceptible to lead poisoning than men, STUIK & ZIELHUIS (1974), and especially during pregnancy, which may be due to the higher rate of calcium metabolism at this time.

Clinical lead poisoning has always been associated with elevated exposure, usually due to occupational use of lead. This source is becoming increasingly rare since legislation requires ever more stringent control of industrial emissions, REYNOLDS (1969). Hence adult males are rarely considered to be at risk as the medical staff of a lead works are trained to look for any of the multiferous symptoms that may arise before the onset of clinical poisoning and also because adults are deemed to recover on removal from source.

For children, pica - the chewing and eating of non-food items - seems to be the major source of poisoning but inhalation has also been recognised as a cause. Increasing traffic in and around cities causes a resultant increase in atmospheric and road-dust burdens of lead in the suburban residential areas. Thus children playing out of doors are more likely to absorb large quantities of lead if this trend continues.

The action of lead in causing poisoning is through enzymic inhibition.

Lead has an affinity for sulphydryl groups and may affect the activity of any enzyme that has such groups in the template of an active site, GOLDBERG (1968). The ready availability of lead to all sections of the community has raised the questions: are there disease effects at non-toxic levels? and what is a safe level of intake?

This has resulted in the measurement of intake by means of amino-levulinic acid (ALA) or coproporhrin levels in blood or faeces/urine samples. The former is not always responsive enough to ensure detection, and the latter usually vary only after the blood lead has passed 40 µg per 100 ml, so are too insensitive.

More recently it has been suggested that the level of enzyme activity of d-ALAD may be a more accurate reflection of current burden. In the erythrocyte fraction of blood, lead is known to inhibit d-ALAD and sodium/potassium-activated adenosine triphosphatase over a wide range of concentrations, HERNBERG, NIKKANEN, MELLIN & LILIUS (1970) and SECCHI, ALESSIO & CAMBIAGHI (1973).

Other systems inhibited by lead include d-ALAD, glucose-6-phosphate dehydratase, and pyruvate decarboxylase in the brain, DOLOWITZ, FAZEKAS & HIMWICH (1937) and MONCRIEFF, KOUMIDES, CLAYTON, PATRICK, RENWICK & ROBERTS (1964). These may affect the availability of energy to the working brain, causing headaches etc. and to the growing brain, causing encephalopathy. Also lead competes with calcium for sites in its phosphate matrix and so is deposited in bones. This pool is mobile and may give rise to calcium deficiencies, which in their turn are known to cause anxiety states, PITTS & McCLURE (1967). These primary mechanisms are important themselves, as may be the consequent secondary effects.

Increasing lead level causes a build up of ALA, which has been found to induce hyperactivity,McGILLION, MOORE & GOLDBERG (1973), and defective nerve conduction, BECKER, VILJOEN & KRAMER (1971). Lead inhibits pyruvate decarboxylase, which leads to raised pyruvate levels, BROWN & McARTHUR (1972). This may lower the energy supply to the brain, PETERS (1953). If the pyruvate is metabolised to lacate by reduction, anxiety states can be induced due to the unavailability of calcium.

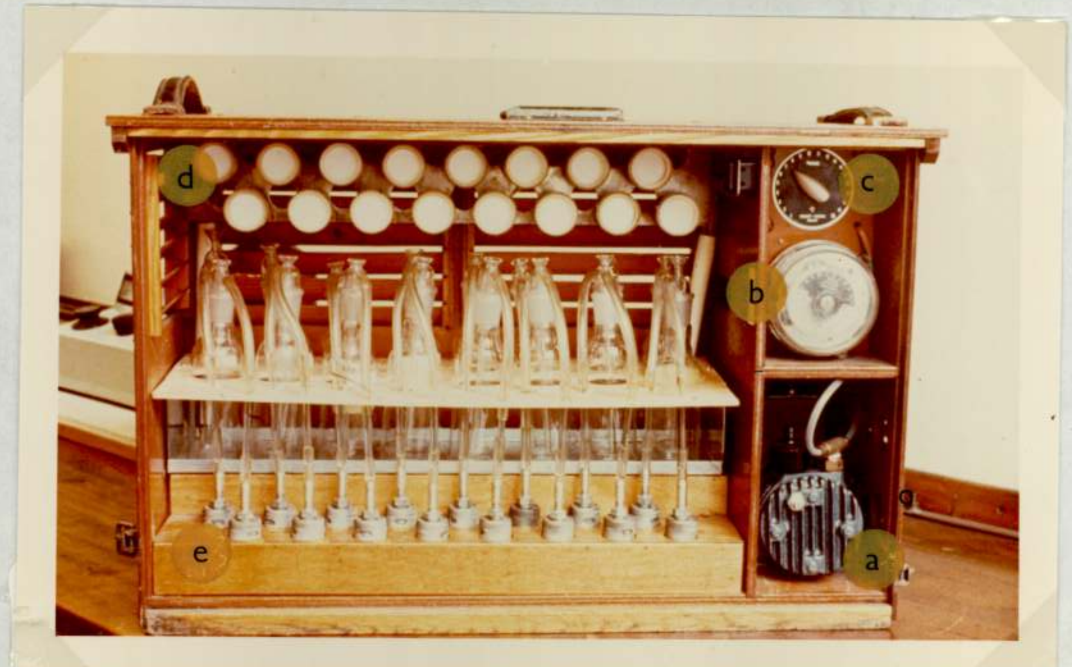


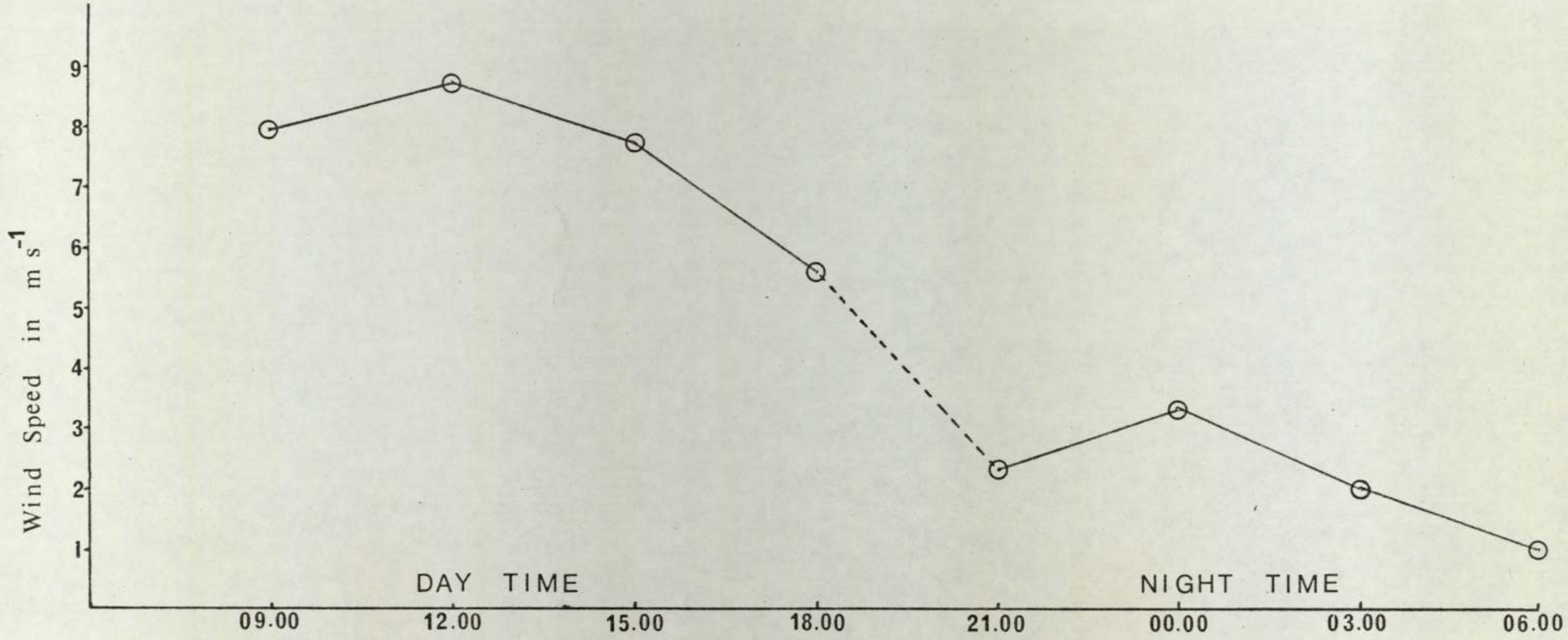
Figure 1: A photograph of an open monitor box, showing a) the vacuum pump, b) the time clock, c) the counter mechanism, d) the filter holders, and e) the schraeder valve sockets.

CHAPTER 2 - EXPERIMENTAL PROCEDURE.

2.1 EQUIPMENT

Essentially two different methods of observing the atmospheric lead aerosol concentration are available. These involve either monitoring air on a continuous basis, or collection of a time integrated sample by aggregation of that part of the atmosphere in which the pollutant is to be found. Lead is admitted to the atmosphere in both gaseous and particulate form, as tetra-alkyllead and inorganic lead salts respectively. THILLEZ (1967) proposed a method for the continuous measurement of total atmospheric lead. Trials with equivalent apparatus were not encouraging, so a method of particulate collection by atmospheric filtration was adopted. This has been applied by numerous workers, DIXON & METSON (1960), DAINES, MOTTO & CHILKO (1970) and TEPPER (1971), using cellulose-ester membrane filters, whose efficiency has been established by FARRAH (1967), MEGAW & WIFFEN (1963) and GROFFMAN & WOOD (1971).

The apparatus to which they were fitted is shown in Figure 1. The pump (a) - an Edwards Carbon Vane rotary vacuum type capable of drawing c.20 l min^{-1} through a filter paper - was controlled by a Sangamo time-clock (b). The latter switched the pump on twice per day for approximately an 11½ hour period in each case, once at about 07.30 and a second time at around 19.30. This time sequence was chosen for two reasons. Firstly, the "daytime" period (07.30 - 19.30) would collect an atmospheric sample during the time of maximum road use, whilst the complimentary interval is that of minimum traffic flow. Also, ambient atmospheric conditions vary more greatly between the "day" and "night" time periods selected, than during them, Figures 2a, b & c. When the pump is switched on, a counter mechanism (c, Figure 1) is activated. This unit can direct a current to any one of twenty outputs. Whenever the power is interrupted, the resumption triggers the counter, which moves to pass the current to the next output in sequence. In this monitor, the counter is made to operate a series of solenoid valves, each of which, when open, connects the pump to



<u>Figure 2a</u>: A graph of wind speed against time for one day.

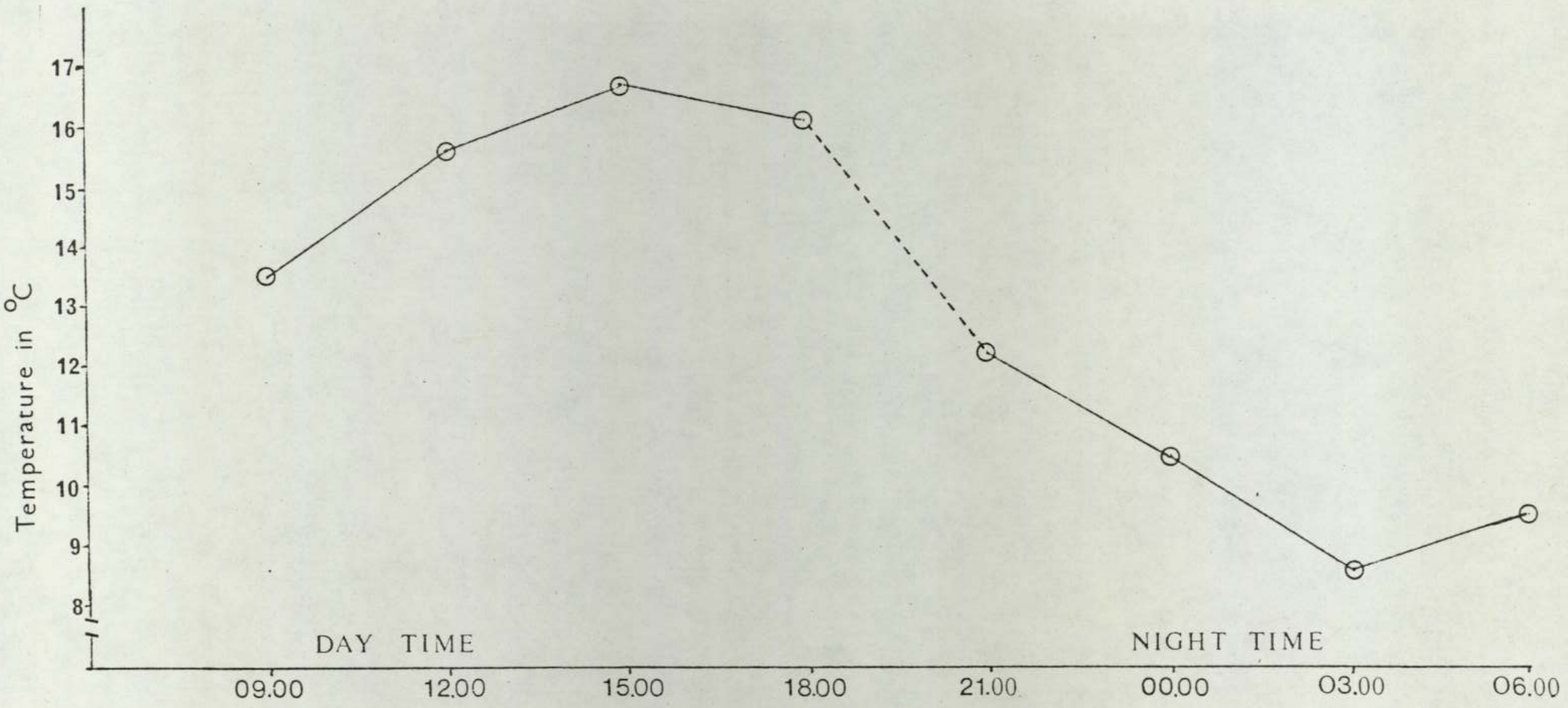


Figure 2b: A graph of temperature against time for one day.

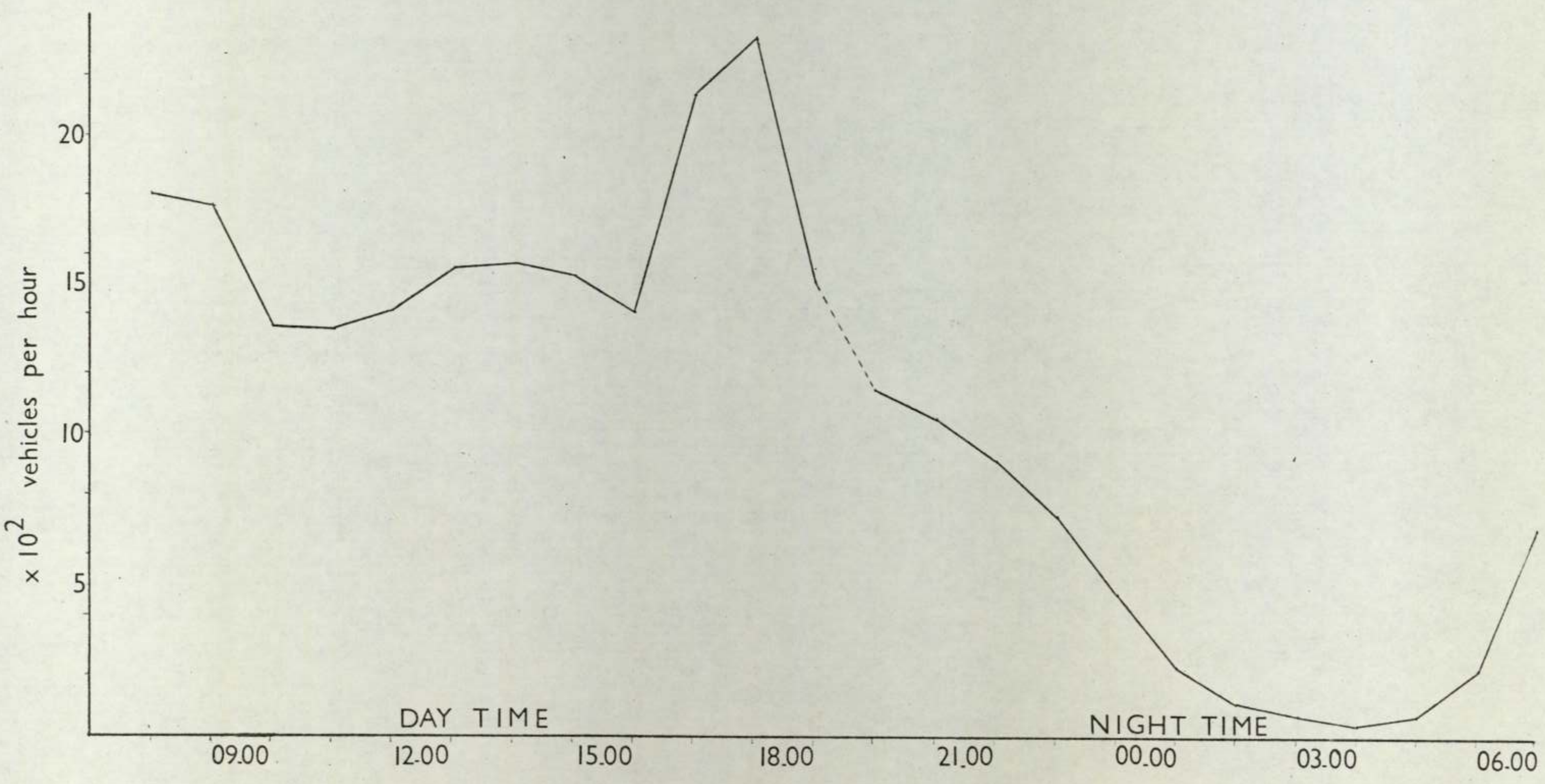


Figure 2c: A graph of traffic against time for one day.

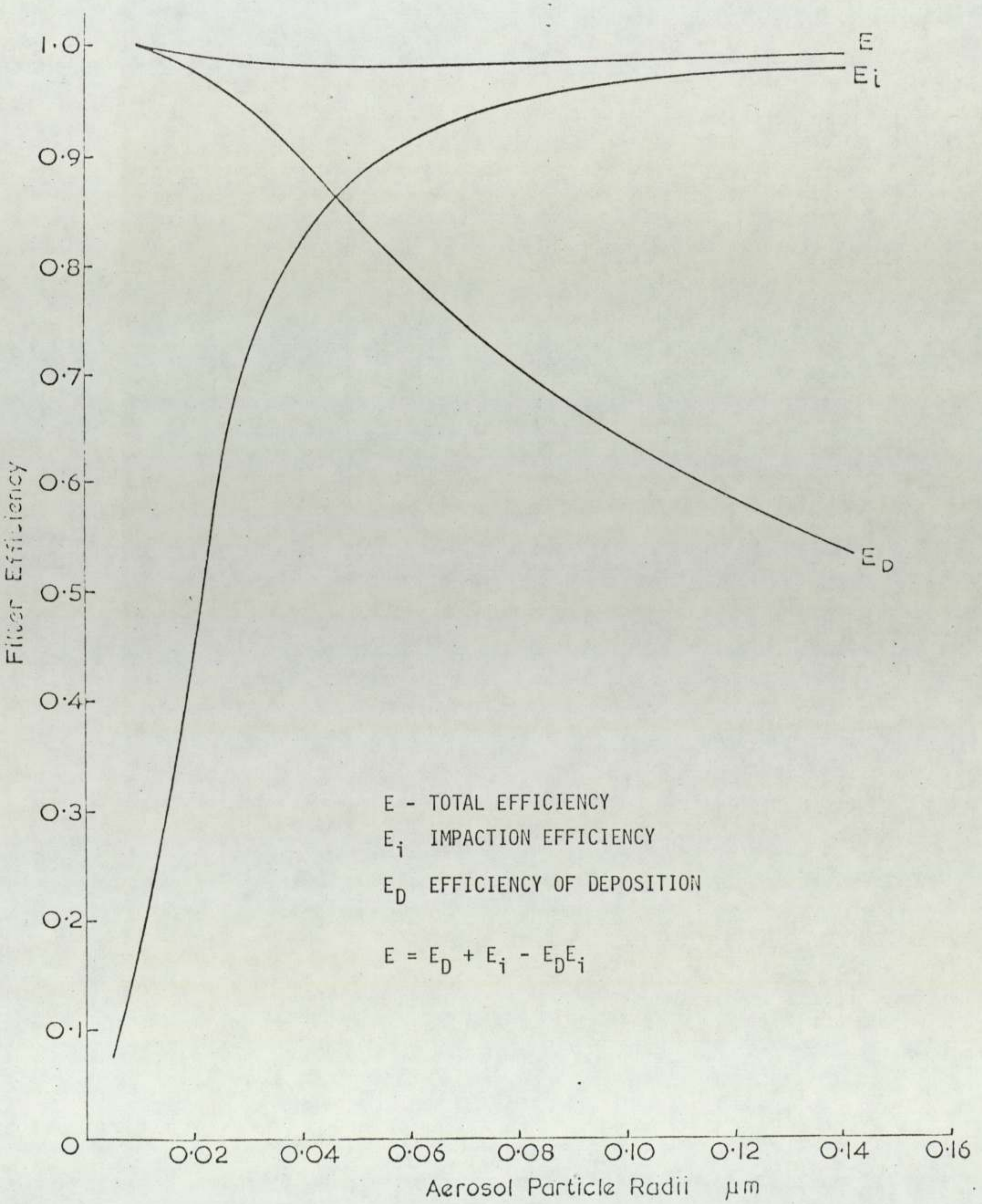
two inlets, where the incoming air may be sampled. One (d) is designed to take Millipore filter holders, and the other (e) is a Schrader valve socket. The whole monitor is run from a mains supply (240 volts, 13 amps) and is the unit that has been used throughout our investigations.

The filters used were 37 mm diameter Millipore type AA, with a pore size of 0.8 μm, which had been recommended by FARRAH (1967) and used by GROFFMAN & WOOD (1971). The efficiency of these filters has been questioned. At first inconsistent results were obtained, FITZGERALD & DETWILER (1957) and WALKENHORST (1959). Then practical studies by KUBIE, JECH & SPURNY (1961) and theoretical modelling by MEGAW & WIFFEN (1963) suggested that membrane filters were 99.9 per cent efficient to particles with as small a radius as 0.01 μm. Also the former found that neither the humidity of the sampled atmosphere, nor the lessening of the possibility of electrostatic deposition caused any reduction in efficiency. The theoretical model was further developed by PICH (1964) and employed by SPURNY & PICH (1965). The latter paper suggested that minima occurred in the curves of surface velocity and particle radius against efficiency. The most recent critique, from SEELY & SKOGERBOE (1974) and LAMOTHE, DICK, CORRIN & SKOGERBOE (1974) derives from results obtained by collecting samples on graphite and Millipore filters at the same site. As velocity at the Millipore filter surface increased, the efficiency apparently decreased. Calculations using the theory used by SPURNY & PICH, with parameters specific to our conditions, do indeed show a minimum in collection efficiency of 99 per cent at a particle radius of 0.09 μm, Figure 3. The question of absolute efficiency of Millipore filters is unresolved, but our results are presented as a valid self-consistent series of measurements.

2.2 SAMPLING SCHEDULE

For a week's sampling at one site, 16 filters were employed. They were first weighed and then placed in Millipore plastic holders. These were used to transport the filters to the sampling site and to support them

Figure 3: A graph of membrane filter efficiency against aerosol particle radiu



during sampling. After opening the monitor, the current was interrupted for 5 - 10 minutes while the filters were exchanged. The counter was then reset, the pump started to collect the rest of that day's sample and the monitor locked. In any one week therefore 7 "night-time", 6 complete and two half-"day-time" samples were taken. This used 15 of the 16 filters put out. The unused one acted as a blank to check the variability in particulate loading due to deposition whilst being transported or on site. It also acted as a check on the weighing procedure, since the filters were not dessicated before and after use. This was considered unnecessary because the filters reached a stable weight in the laboratory atmosphere within 1 - 2 days, and so could be reweighed after this time interval. The consistency of the blank, which generally returned between 0.0 - 0.2 mg heavier than its weight before exposure, again suggested that dessication was unnecessary. A picture of 3 sets of filters as they came back from the experiment described in Chapter 4 is shown in Figure 4.

2.3 ANALYTICAL PROCEDURE

The whole filter was analysed since PIERCE & MEYER (1971) had shown that analysis of anything less could lead to uncertainty in the determination of anywhere between 60 and 150 per cent. This being true whether 10 or 50 per cent of the filter was analysed. Methods of dissolving the lead from the filter involve the use of nitric acid alone or a nitric acid hydrogen peroxide mixture, BULLOCK & LEWIS (1968) and TONKELAAR & BIKKER (1971), but each left part of our sample undissolved. Another method, dry ashing before dissolution, has been shown to cause variable loss of lead compounds, GEORGII & JOST (1971) and BURNHAM, KANABROCKI & HATTORI (1969). So a method of digesting the paper and sample was adopted, for which the use of concentrated perchloric acid and concentrated nitric acid is established, DAINES, MOTTO & CHILKO (1970) and TEPPER (1971). The filter was placed in a 50 ml beaker and 3 ml of lead free concentrated (72%) nitric acid added. This was heated on an electric hot-plate until the

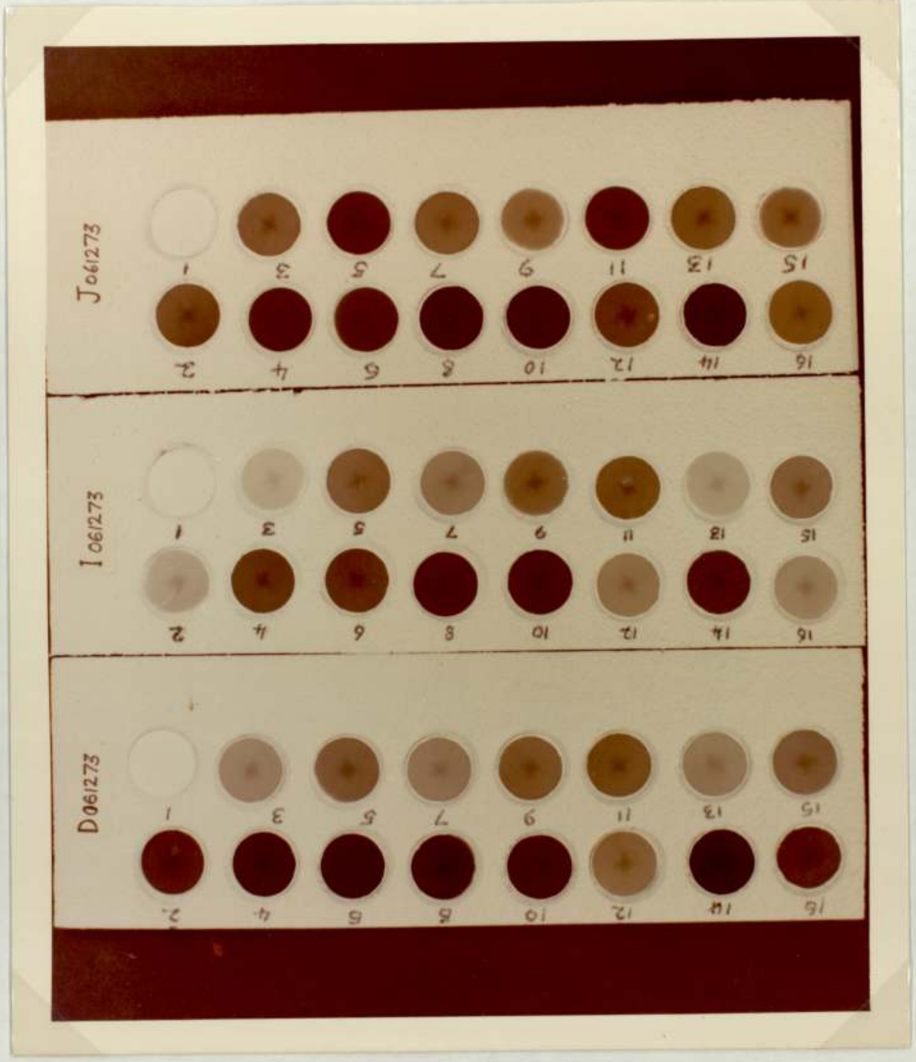


Figure 4: A picture of the filters returned from site D, inside (I) and outside (J) of house A, as a result of monitoring from 6th to 13th December, 1973.

filter paper had dissolved, then a 50/50 (v:v) mixture of nitric acid and lead free concentrated (60%) perchloric was added to keep the boiling solution to about 5 ml. When dense white fumes of perchloric acid appeared, 1 ml of nitric acid was added in such a manner as to wash the sides of the beaker. The solution was again made to fume, then removed from the heat and allowed to cool. The room temperature, clear solution was washed from the beaker with deionised water into a 10 ml volumetric flask, which was filled to the mark, mixed, transferred to a vial and stored for analysis. The resulting solutions were analysed, within one or two days of preparation, by atomic absorption spectrophotometry. This method for determination of lead in solution has been widely used, ELWELL & GIDLEY (1961), BULLOCK & LEWIS (1968) and GEORGII & JOST (1971). Few papers quote the actual emission line of the lead specific lamp, which is used to measure the concentration of lead atoms above the flame in the instrument. The choice is between the 217 nm line, PIERCE & CHOLAK (1966) and the 283.3 nm line, THILLEZ (1967). Since the latter was recommended by the makers Perkin Elmer, in their handbook (1968), it was used for all determinations from October 1971 to December 1973.

The photometer used was a Perkin Elmer 303, which is a split beam instrument, whose output is in per cent absorption. The flame is formed by burning an air-acetylene mixture into which solutions can be atomised. The output was produced on a chart recorder. To analyse solutions of unknown concentrations the following sequence was used:- Firstly a base line was established using double distilled water, then the absorption of each of four standards was found by atomising them into the flame for half a minute each, a time in which the signal became steady. Double distilled water was run for half a minute between standards and the eight unknowns that followed. The standard solutions were then re-run as a check on instrument stability. If an unknown had an absorption that lay outside the range covered by the standards, new ones were prepared using a Hook and Tucker auto-diluter, and the unknown run again between the new standards. All standard solutions were prepared from a stock solution of

1000 ppm lead nitrate in dilute nitric acid. Each week's monitoring of the atmosphere at a particular site, produced 16 samples (15 contaminated and one 'blank'), all of which were put through the same dissolution and ananysis procedure. The blank was considered a measure of any experimental contamination due to transport, electrostatic deposition on site, storage, reagents or containers, and also a measure of the matrix effect. This is an effect caused by atomising a mixture other than a dilute solution in water. It has been noted that this effect is small for lead in nitric and perchloric acids, ELWELL & GIDLEY (1961).

The measure of absorption was taken as the height of the peak above the base line signal - from distilled water. The resulting measure, of percentage absorption, was converted to absorbance units for each reading, using a table provided in the Perkin Elmer handbook. Then a graph of absorbance against concentration was drawn of the results from the standards, averaged between those run before and those after the unknowns being calculated. The concentration in the eight unknown solutions could be read directly from this graph. To find the concentration in the 15 unknown solutions due to atmospheric pollution, the level found in the blank filter solution - c.0.1 μg ml^{-1}- was subtracted from those found in the other solutions.

To calculate the average atmospheric lead concentration found by a filter, the following equation was used:

$$C_a = \frac{(Xa - b) \times 10 \times 10^3}{F.R. \times S.P.} \quad \mu g\ m^{-3}$$

C_a is the average lead concentration over a sampling period 'a'.

X_a is the concentration of lead in μg ml^{-1} found in the solution of the filter used during a period 'a'.

b is the concentration of lead in μg ml^{-1} in the solution of the appropriate blank filter.

10 is the factor used because each filter digest is made up to 10 ml.

FR is the flow rate of air through the filter during sampling as measured by a rotameter, this is in litres per minute so the 10^3 factor is required to convert litres to m^3.

SP is the length of the sampling period 'a' in minutes.

2.4 WEATHER DATA

The weather data used was selected from readings taken at Elmdon Airport, which is about 5 miles from Gravelly Hill. The records there cover numerous variables measured every third hour. The eight daily values are taken at: 02.50, 05.50, 08.50, 11.50, 14.50, 17.50, 20.50 and 23.50 h. For comparison with a "daytime" lead concentration, the mean of readings at 08.50, 11.50, 14.50 and 17.50 was used; whilst with a "night-time" level, the average of those at 20.50, 23.50, 02.50 and 05.50 was compared. Eleven variables were extracted:

a) wind speed	m sec^{-1}
b) wind direction	degrees from North
c) visibility	kilometres
d) temperature	degrees Centigrade
e) relative humidity	per cent
f) atmospheric pressure	millibars
g) dewpoint	degrees Centigrade
h) vapour pressure	mm of mercury
i) rainfall	mm
j) quantity of sunshine	hours
k) total cloud	eights of cover.

2.5 TRAFFIC MEASUREMENTS

After the ground level road system had been built, some traffic counts were made with pneumatic tubes by Warwickshire traffic engineers. Since a permanent record of traffic volumes was required for this investigation, arrangements were made with the Transport and Road Research

Laboratory to install Streeter-Amet equipment. This has counted traffic using the Salford Circus entrance-exit roundabout of the junction since April 1972. It received signals from sub-surface loops, which lay one under each lane of the four roads leading onto the roundabout. The signals from these seven loops were accumulated for an hour and the totals then printed onto a paper tape. "Day-time" traffic totals are summations of the 12 counts after and including the 08.00 figure. The "night-time" ones result from the summation of those after the 19.00 h. total.

Motorway traffic counts are also available from equipment especially designed and constructed by the Electrical Engineering Department of the University of Aston, on behalf of the Transport and Road Research Laboratory. Loops, whose positions are shown labelled A to L in Figure 5, were buried under each carriageway and created specific signals. Each signal was amplified and channeled to activate a unique Veeder-Root counter, as shown in Figure 6. Total coverage requires the use of 28 loops to account for all the traffic between junctions 5 and 7 of the M6. Figure 7 shows the counters (a), along with the digital clock (b) which displays the day, date and time. The whole unit is set in a cabinet below a camera (c). The flash-lights (d) and the camera are activated by the time-clock (e), the former has an interval determined by the timer (f). The time-clock may be set to obtain photographs at intervals varying from 1 - 12 h. This equipment was located at the Midlands Motorway Police Control Centre, Perry Barr, Birmingham.

2.6 MONITORING SITES

We measured atmospheric levels of lead at a number of sites around the junction. The selection of these depended on the availability of electricity and the safety that could be afforded the monitors during their operation. Thus schools, houses, gantries and a block of flats were selected as likely sites. The positions used are shown on the map Figure 8 by site letter and are described below.

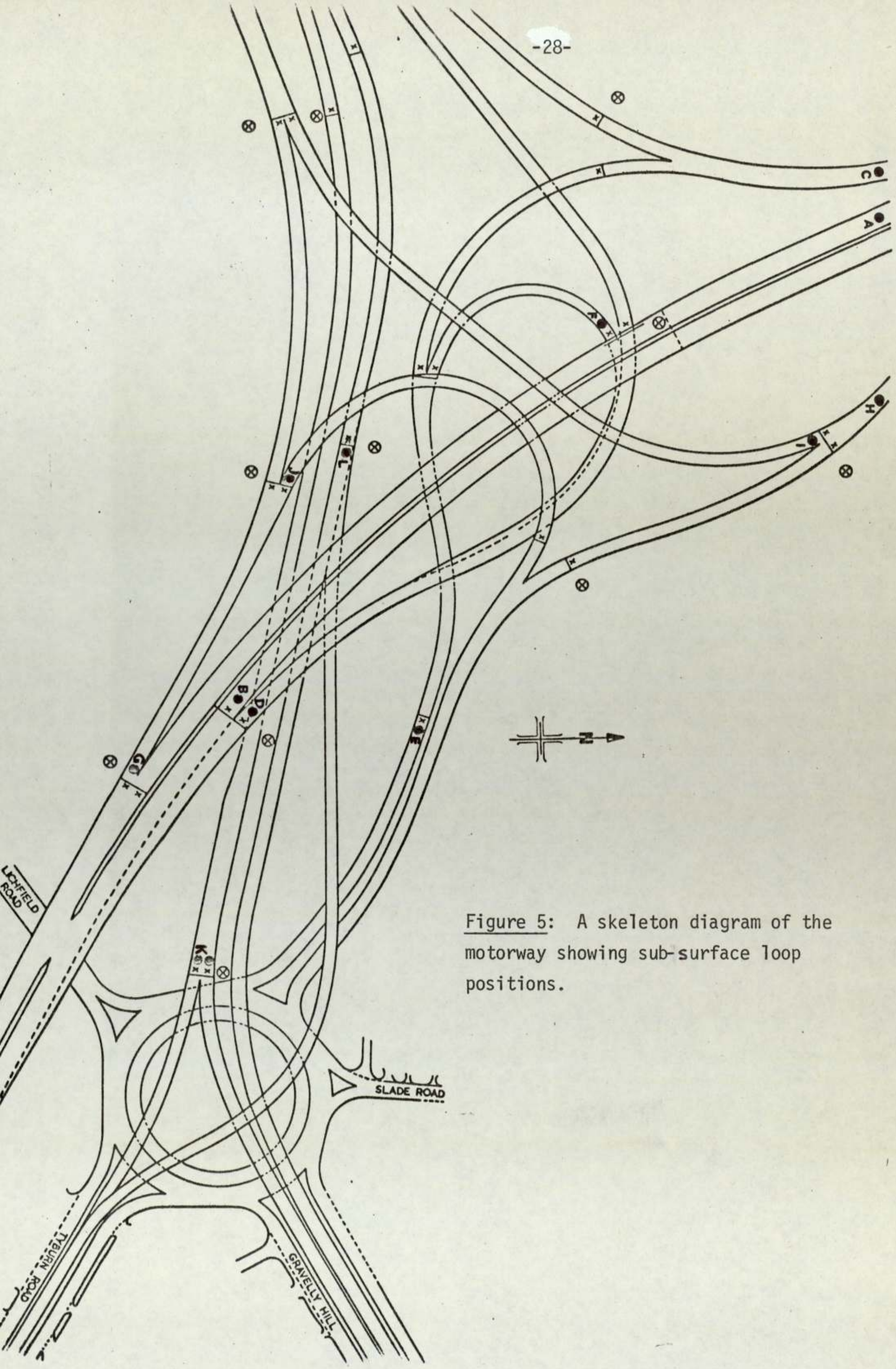


Figure 5: A skeleton diagram of the motorway showing sub-surface loop positions.

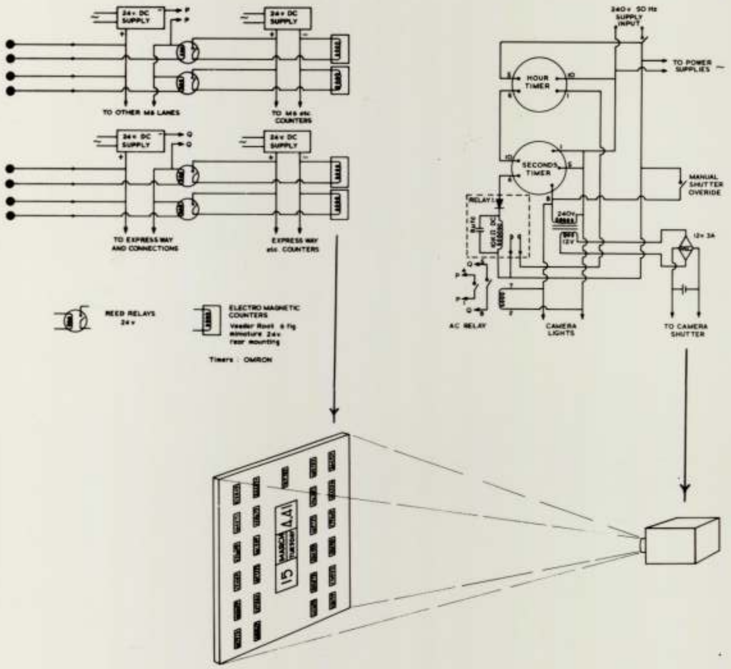


Figure 6: A circuit diagram showing the connexions between the sub-surface loops and veeder root counters. The wiring of the photographic circuit and the counter display are also shown.

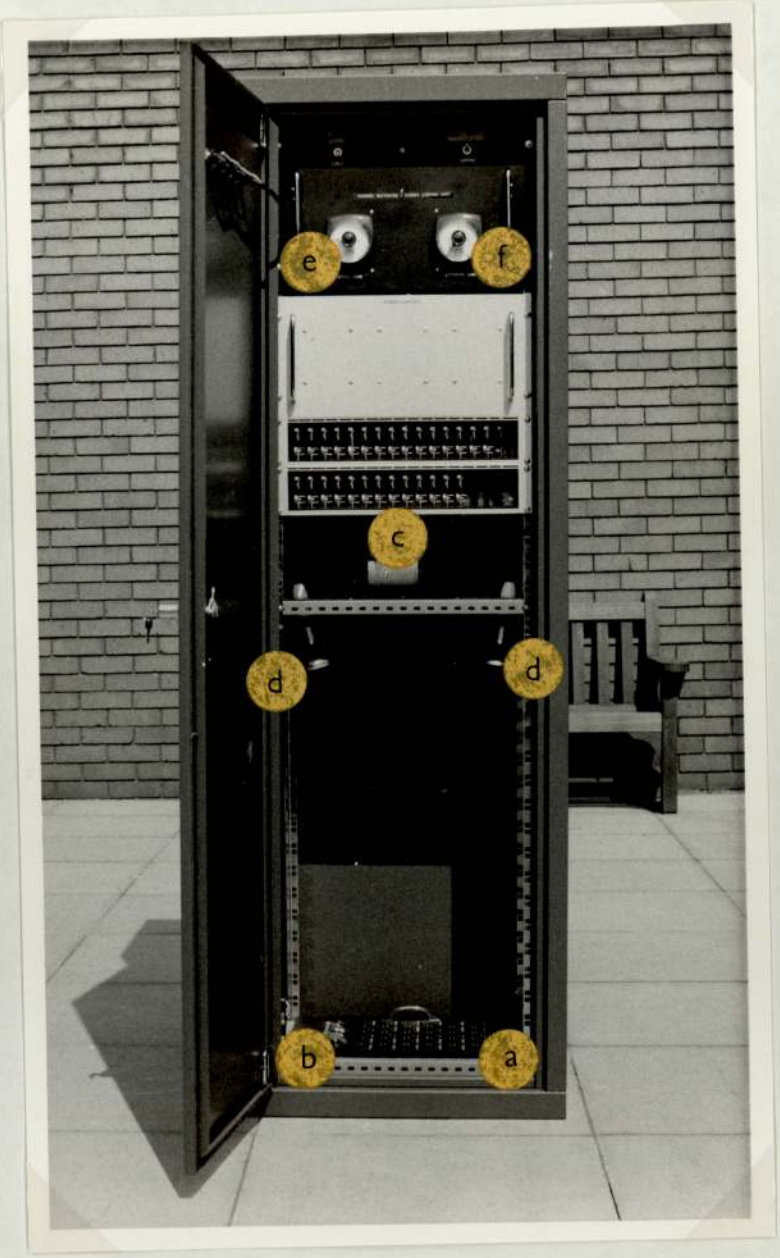


Figure 7: A picture of the motorway traffic monitor/recorder showing a) the counters, b) the digital clock, c) the camera, d) the flash lights, e) the time clock and f) the flash interval timer.

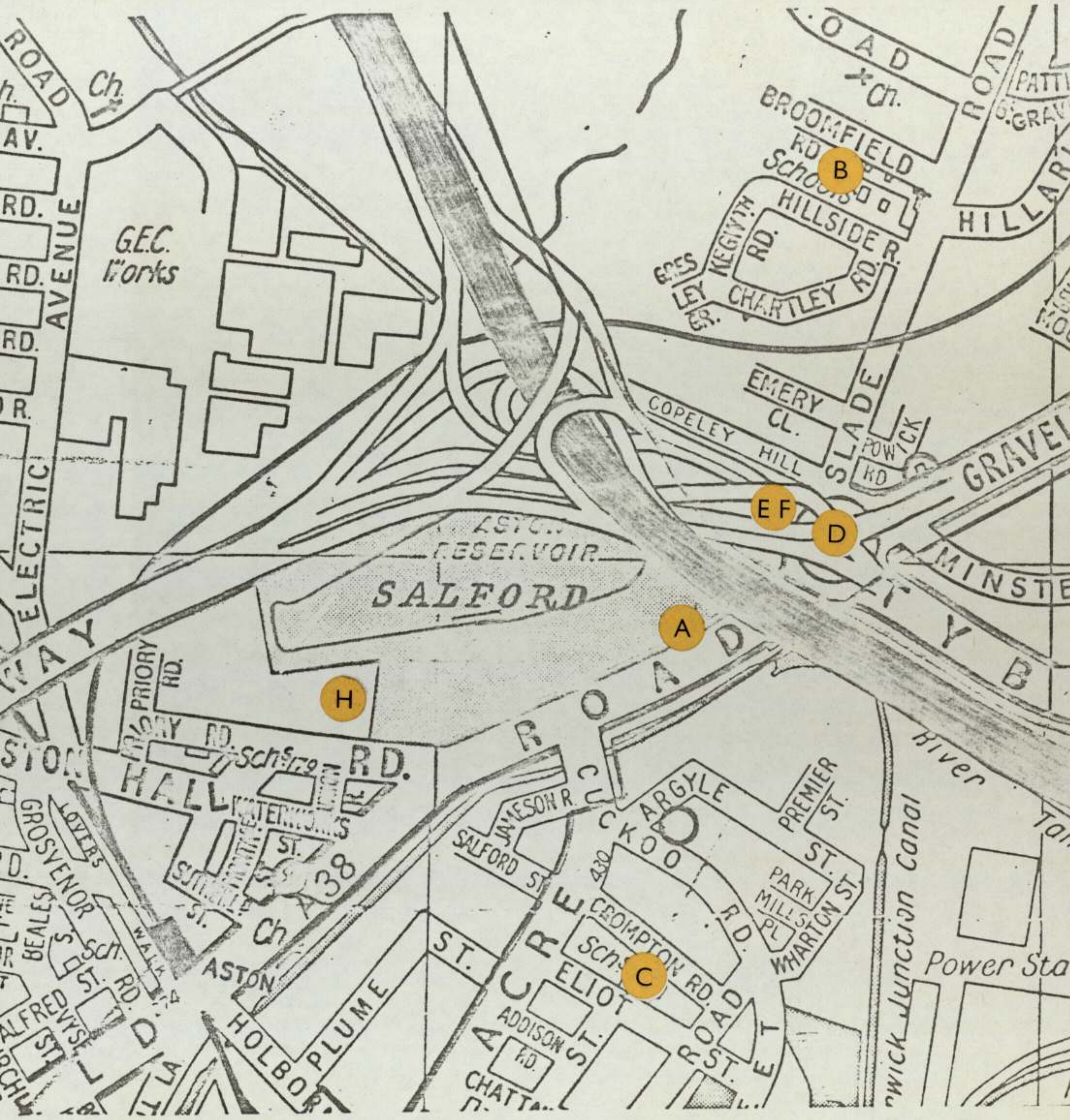


Figure 8: A picture of the Gravelly Hill area showing monitoring site positions.

SITE A: A position alongside the A38 road, 120 m south of the Salford Circus roundabout. A monitor was bracketed, 2½ m above the pavement, to a lamp-post from which an electricity supply was made available. This site was used continuously from October 1971 to May 1972, to gain some knowledge of lead levels prevalent before the motorway opened in May 1972.

SITE D: A permanent site situated on top of the conveniences at the centre of Salford Circus roundabout. The monitor was 3 m above the surrounding subway, but about level with the road surface which was 45 m distant. Monitoring at this site has been continuous since April 1972.

SITE B: Slade Road Schools. The equipment was mounted 2½ m above the playground of the school and about 510 m from the Salford Circus roundabout. The ground between the roundabout and the school is open parkland which rises c.30 m to the school. Measurements were taken here from January to November 1972 and February to June 1973.

SITE C: Eliot Street School. The monitor was mounted on a flat roof about 2½ m above the playground and 580 m from Salford Circus roundabout. The intervening area was partially residential and partly industrial but densly packed with buildings. Measurements were taken here from March 1972 to January 1973 and in June 1973.

SITES E & F: Gantry sites. F was mounted 5.5 m vertically above E on a gantry 56 m from the Salford Circus monitor. The monitor at E was placed beside an access road to the M6 and was controlled from the clock in the monitor at F so that the two were synchronous.

SITE H: Murdoch Point. This towerblock was 600 m from the Salford Circus roundabout and lay equidistant from the A38, Lichfield Road and the A38(M), Aston Expressway. The monitor was placed on the top of the tower about 36 m above the surrounding country, which was mainly flat parkland between the tower and the motorway.

CHAPTER 3. RESULTS AND DISCUSSION

3.1 LEAD LEVELS

The atmospheric concentration of lead has been monitored at numerous sites in many parts of the world. The levels found near urban motorways are reported from the United States as being c.10 μg m^{-3} for a traffic level of 2,500 vehicles per hour, DAINES, MOTTO & CHILKO (1970). ATKINS & KRUGER (1968) also report values of between 5 and 10 μg m^{-3} for a similar traffic flow. The monitoring of roadside levels that we undertook suggests an average level of 2 μg m^{-3}, at site A. The nearby traffic density on the A38, Lichfield Road was c.700 vehicles per hour. At site E, the base of a gantry, the level averaged 2.5 μg m^{-3} and the traffic level was of the same order as that at site A.

Fall-out from the atmosphere is fairly rapid near the road, LAMOTHE, DICK, CORRIN & SKOGERBOE (1974), with apparent exponential decay occurring. BESNER & ATKINS (1970) found a 45 per cent drop in 60 m with no subsequent dimunition up to 120 m. The background level is found when a stable aerosol has been formed, this quantitatively depends upon the neighbourbood, and would cause the "night-time" level to be little different from the "day-time" one. The results from the various sites are shown in Table 1. From this it appears that a background aerosol has been formed at 600 m from Salford Circus and that this is possibly the range of influence of the motorway. The average concentration of this aerosol was 0.8 μg m^{-3} after the motorway opened. A site (H) about equally distant from Salford Circus as the school sites, but 40 m high, had an average level of 0.5 μg m^{-3}.

3.2 VARIATION OF LEAD LEVELS

Two monitors were run side by side for two weeks and the results found are presented in Table 2. The average difference between readings taken for adaytime period was negligible, whilst at night-time, one monitor always gave a lower value by, on average, 0.25 μg m^{-3}. This is less than the

SITE AND DISTANCE FROM TRAFFIC	DURATION OF SAMPLING	AVERAGE DAY-TIME VALUES	AVERAGE NIGHT-TIME VALUES	PERCENTAGE $\frac{\text{NIGHT}}{\text{DAY}}$
0 m A (2 m above traffic)	1971/72 8 months	2.60	1.66	64
E	1972/73 2 months	2.50	1.25	50
F (5.5m above traffic)	1972/73 7 months	1.91	1.11	58
45 m D	1972/73 20 months	2.16	1.42	66
500 m B	1972/73 16 months	0.83	0.80	97
580 m C	1972/73 12 months	0.80	0.86	108
600 m H (36 m above traffic)	1973 2 months	0.48	0.52	108

Table 1: A comparison of mean day and night-time lead concentrations in $\mu g\ m^{-3}$ from sites A to H.

a) Day-time Values

DATE	BOX A	BOX B
26.04	1.60	1.88
27.04	0.99	1.35
28.04	1.08	0.91
29.04	3.27	3.35
30.04	3.15	3.12
01.05	1.36	1.31
02.05	1.50	1.25
03.05	1.13	1.29
04.05	1.09	-
05.05	1.20	0.91
06.05	1.42	1.61
07.05	2.14	2.18
08.05	1.87	1.52
AVERAGE ± 1 S.D.	1.68 ± 0.53	1.72 ± 0.58

b) Night-time Values

DATE	BOX A	BOX B
25.04	0.85	0.96
26.04	0.67	1.01
27.04	0.57	0.78
28.04	1.44	1.68
29.04	3.78	4.06
30.04	0.65	0.69
01.05	0.59	0.86
02.05	0.39	0.69
03.05	0.65	1.02
04.05	0.48	-
05.05	0.70	0.92
06.05	1.76	2.27
07.05	1.88	2.12
08.05	0.41	0.72
AVERAGE	1.06 ± 0.79	1.37 ± 0.86
DAY AND NIGHT AVERAGE	1.37 $\mu g\ m^{-3}$	1.54 $\mu g\ m^{-3}$

Table 2: Lead levels in $\mu g\ m^{-3}$ found by two monitors run synchronously at Salford Circus during the period 25th April to 08th May 1974.

variability in the readings (0.79 - 0.86 $\mu g\ m^{-3}$). Therefore despite possible faults in the filters these monitors and the analysis procedures give self-consistent results.

The 12 h average values of lead concentrations monitored at the various sites have been averaged into monthly figures and these are presented in Tables 3 - 9. The concentrations found at a remote site in Wales using the same equipment as at Gravelly Hill are given in Table 10. The maximum and minimum lead levels monitored in each month, with the date on which they were found, are given in Tables 11 - 17.

3.2.1 VARIATION WITH DAY AND NIGHT

The lead level is less during the night than in the day-time for sites within the influence of the motorway. Outside the boundary, the rate of influx of aerosolic lead is equalled by its rate of removal and the variation in levels found is due to changes in meteorological factors. From Table 1 it can be seen that sites B, C and H fall approximately on this boundary. The other four sites suggest that the decrease in level from day to night is 40 per cent on average, depending on site and time of year. This is least during March/April when the drop is c.17 per cent rising to c.55 per cent in the winter months, December and January. The decrease in traffic density from day to night is a steady 70 per cent throughout the year. If traffic is the major source of the lead aerosol, then there must be some retention of particulate from day to night. This is supported by TER HAAR, HOLTZMAN & LUCAS (1967) who estimated the half life of an urban lead aerosol to be between 1 and 4 weeks, and FRANCIS, CHESTERS & HASKIN (1970) who estimate a mean residence time of 9.5 days.

According to HABIBI (1970) 30 per cent of the lead burned during propulsion of a car is emitted in a stable aerosolic size range. GILLETTE (1972) suggests that coagulation of these particles and their subsequent removal by sedimentation is the major mechanism for aerosol ageing. This is in the absence of rainfall. During precipitation episodes, PUESCHEL,

1971	DAY TIME	NIGHT TIME	24 h AVERAGE
OCTOBER	3.51 (30)	2.10 (30)	2.80
NOVEMBER	3.22 (28)	2.14 (27)	2.68
DECEMBER	3.85 (30)	2.68 (27)	3.26
1972			
JANUARY	3.01 (24)	1.46 (23)	2.23
FEBRUARY	2.47 (11)	1.32 (11)	1.89
MARCH	2.01 (17)	1.78 (19)	1.89
APRIL	1.59 (27)	1.08 (28)	1.33
MAY	1.16 (21)	0.69 (21)	0.97

Table 3: Monthly average 12 h lead levels monitored at Site A in μg m^{-3}.
The number of contributions to each average is shown in parentheses.

1972	DAY TIME	NIGHT TIME	24 h AVERAGE
JANUARY	1.16 (8)	1.00 (8)	1.08
FEBRUARY	1.03 (11)	0.81 (11)	0.92
MARCH	0.90 (20)	0.83 (22)	0.86
APRIL	0.51 (24)	0.53 (26)	0.52
MAY	0.58 (22)	0.46 (23)	0.52
JUNE	0.54 (29)	0.55 (30)	0.54
JULY	0.60 (31)	0.68 (30)	0.64
AUGUST	0.51 (29)	0.56 (30)	0.53
SEPTEMBER	1.45 (30)	1.05 (30)	1.25
OCTOBER	0.81 (31)	0.72 (31)	0.76
NOVEMBER	1.25 (27)	0.78 (26)	1.01
DECEMBER			
1973			
JANUARY			
FEBRUARY	1.09 (26)	0.89 (27)	0.99
MARCH	0.91 (31)	1.30 (31)	1.10
APRIL	0.53 (20)	0.69 (22)	0.61
MAY	0.64 (17)	0.91 (18)	0.78
JUNE	0.69 (7)	1.09 (6)	0.89

Table 4: Monthly average, 12 h lead levels in $\mu g\ m^{-3}$ monitored at Site B. The number of contributions to each average is given in parentheses.

1972	DAY TIME	NIGHT TIME	24 h AVERAGE
MARCH	0.99 (13)	1.24 (16)	1.11
APRIL	0.93 (30)	0.83 (24)	0.88
MAY	0.78 (22)	0.50 (23)	0.64
JUNE	0.64 (17)	0.61 (17)	0.62
JULY	0.66 (28)	1.05 (30)	0.85
AUGUST	0.54 (24)	0.60 (27)	0.57
SEPTEMBER	1.02 (30)	1.07 (30)	1.04
OCTOBER	0.76 (31)	0.71 (31)	0.73
NOVEMBER	1.05 (29)	0.98 (28)	1.01
DECEMBER	0.92 (31)	0.74 (30)	0.83
1973			
JANUARY	1.49 (31)	0.75 (30)	1.12
FEBRUARY			
MARCH			
APRIL			
MAY			
JUNE	0.56 (21)	0.57 (21)	0.56

Table 5: Monthly average, 12 h lead levels in $\mu g\ m^{-3}$ monitored at Site C. The number of contributions to each average is shown in parentheses.

1972	DAY TIME	NIGHT TIME	24 h AVERAGE
MAY	1.04 (31)	0.69 (30)	0.87
JUNE	1.70 (29)	1.20 (30)	1.45
JULY	2.03 (30)	1.45 (31)	1.74
AUGUST	2.29 (23)	1.24 (24)	1.76
SEPTEMBER	2.27 (30)	1.12 (29)	1.70
OCTOBER	2.16 (30)	1.58 (29)	1.87
NOVEMBER	2.97 (28)	1.41 (29)	2.19
DECEMBER	2.29 (31)	1.28 (30)	1.78
1973			
JANUARY	2.59 (30)	1.19 (29)	1.89
FEBRUARY	3.36 (22)	1.63 (23)	2.50
MARCH	2.69 (26)	2.58 (27)	2.63
APRIL	2.15 (30)	1.92 (29)	2.04
MAY	1.65 (31)	1.09 (31)	1.37
JUNE	1.41 (30)	1.19 (29)	1.30
JULY	1.71 (29)	1.35 (30)	1.53
AUGUST	1.47 (31)	1.14 (30)	1.30
SEPTEMBER	1.95 (28)	1.36 (30)	1.66
OCTOBER	2.68 (24)	1.54 (24)	2.11
NOVEMBER	2.70 (29)	1.67 (29)	2.18
DECEMBER	2.06 (31)	1.78 (31)	1.92

Table 6: Monthly average, 12 h lead levels in $\mu g\ m^{-3}$ monitored at Site D. The number of contributions to each average is shown in parentheses.

1972	DAY TIME	NIGHT TIME	24 h AVERAGE
DECEMBER	2.24 (24)	1.08 (25)	1.66
1973			
JANUARY	2.76 (27)	1.41 (28)	2.08

Table 7: Monthly average, 12 h lead levels in $\mu g\ m^{-3}$ monitored at Site E. The number of contributions to each average is shown in parentheses.

1972	DAY TIME	NIGHT TIME	24 h AVERAGE
DECEMBER	2.39 (24)	0.91 (22)	1.65
1973			
JANUARY	2.21 (26)	0.74 (22)	1.47
FEBRUARY	1.99 (28)	1.29 (24)	1.64
MARCH	2.07 (23)	1.56 (19)	1.81
APRIL			
MAY	1.28 (7)	0.70 (7)	0.99
JUNE	1.51 (29)	1.00 (24)	1.25
JULY	1.89 (11)	1.58 (9)	1.73

Table 8: Monthly average, 12 h lead levels in $\mu g\ m^{-3}$ monitored at Site F. The number of contributions to each average is shown in parentheses.

1973	DAY TIME	NIGHT TIME	24 h AVERAGE
MAY	0.48 (9)	0.49 (11)	0.48
JUNE	0.48 (25)	0.54 (23)	0.52

Table 9: Monthly average, 12 h lead levels in $\mu g\ m^{-3}$ monitored at Site H. The number of contributions to each average is shown in parentheses.

DATE	DAY TIME	NIGHT TIME
10.7	-	0.07
11.7	0.11	0.14
12.7	0.08	0.08
13.7	0.39	0.50
14.7	0.06	-
AVERAGE	0.16	0.20
COMBINED AVERAGE	0.18	
PERCENTAGE OF LEAD IN TOTAL PARTICULATE	0.40	

Table 10: Lead concentrations in $\mu g\ m^{-3}$ found at a site near Newtown, Montgomeryshire, Wales, between July 10th - 14th, 1972.

	DAY TIME		NIGHT TIME	
1971	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
OCTOBER	8.24 (29)	1.11 (17)	4.48 (28)	0.18 (22)
NOVEMBER	6.35 (28)	0.66 (21)	12.22 (30)	0.37 (21)
DECEMBER	12.14 (01)	0.81 (26)	11.25 (04)	0.52 (28)
1972				
JANUARY	10.55 (21)	0.91 (27)	3.70 (13)	0.45 (01)
FEBRUARY	4.65 (07)	0.86 (02)	2.22 (03)	0.96 (08)
MARCH	4.26 (08)	0.45 (25)	4.37 (23)	0.60 (26)
APRIL	3.05 (20)	0.48 (30)	2.35 (13)	0.40 (30)
MAY	3.05 (16)	0.13 (23)	2.15 (16)	0.27 (21)

Table 11: Maximum and minimum values of the particulate lead aerosol concentration in $\mu g\ m^{-3}$, monitored at Site A. The numbers in parentheses are the dates when the levels shown were monitored.

	DAY TIME		NIGHT TIME	
1972	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
JANUARY	2.43 (31)	0.57 (27)	2.87 (30)	0.29 (29)
FEBRUARY	2.07 (07)	0.39 (08)	1.73 (03)	0.10 (11)
MARCH	2.10 (22)	0.03 (12)	2.26 (15)	0.09 (10)
APRIL	1.44 (13)	0.10 (03)	1.70 (12)	0.11 (23)
MAY	2.46 (18)	0.22 (08)	1.14 (06)	0.10 (07)
JUNE	1.31 (19)	0.06 (18)	1.56 (28)	0.13 (17)
JULY	1.93 (06)	0.16 (18)	2.62 (05)	0.12 (03)
AUGUST	1.15 (11)	0.24 (27)	1.70 (10)	0.05 (11)
SEPTEMBER	1.54 (12)	0.02 (02)	1.48 (07)	0.03 (03)
OCTOBER	1.31 (06)	0.24 (15)	1.57 (23)	0.11 (16)
NOVEMBER	2.45 (16)	0.22 (12)	2.10 (24)	0.10 (29)
DECEMBER				
1973				
JANUARY				
FEBRUARY	6.60 (25)	0.06 (03)	1.80 (28)	0.03 (07)
MARCH	2.02 (08)	0.29 (12)	5.38 (10)	0.21 (02)
APRIL	2.24 (26)	0.09 (22)	1.69 (26)	0.10 (08)
MAY	1.68 (24)	0.14 (20)	2.34 (24)	0.15 (29)
JUNE	1.36 (04)	0.10 (07)	2.33 (01)	0.51 (02)

Table 12: Maximum and minimum values of the particulate lead aerosol concentration in $\mu g\ m^{-3}$, monitored at Site B. The numbers in parentheses are the dates when the levels were monitored.

	DAY TIME		NIGHT TIME	
1972	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
MARCH	2.73 (20)	0.34 (26)	5.57 (19)	0.20 (26)
APRIL	4.64 (17)	0.16 (01)	4.24 (17)	0.09 (05)
MAY	2.48 (20)	0.17 (07)	1.47 (02)	0.12 (24)
JUNE	1.02 (12)	0.25 (03)	1.69 (01)	0.11 (30)
JULY	1.85 (06)	0.13 (17)	7.94 (25)	0.16 (03)
AUGUST	1.30 (23)	0.05 (27)	1.35 (28)	0.14 (08)
SEPTEMBER	4.27 (22)	0.26 (02)	3.29 (26)	0.21 (03)
OCTOBER	1.59 (23)	0.04 (19)	2.07 (31)	0.12 (29)
NOVEMBER	2.98 (08)	0.16 (12)	2.67 (04)	0.14 (27)
DECEMBER	4.02 (21)	0.23 (28)	1.90 (21)	0.19 (06)
1973				
JANUARY	4.71 (31)	0.23 (26)	2.06 (08)	0.14 (20)
FEBRUARY				
MARCH				
APRIL				
MAY				
JUNE	1.13 (21)	0.28 (24)	1.56 (08)	0.08 (11)

TABLE 13: Maximum and minimum values of the particulate lead aerosol concentration in $\mu g\ m^{-3}$, monitored at Site C. The numbers in parentheses are the dates when the levels shown were monitored.

	DAY TIME		NIGHT TIME	
1972	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
MAY	2.76 (18)	0.29 (04)	1.59 (31)	0.08 (08)
JUNE	2.73 (26)	0.67 (18)	2.95 (11)	0.13 (17)
JULY	4.49 (06)	0.66 (28)	4.72 (29)	0.03 (27)
AUGUST	9.32 (20)	0.63 (31)	2.72 (11)	0.40 (31)
SEPTEMBER	9.07 (01)	0.55 (09)	4.55 (26)	0.33 (02)
OCTOBER	7.91 (12)	0.49 (29)	3.25 (03)	0.36 (25)
NOVEMBER	8.37 (17)	0.57 (12)	5.31 (16)	0.35 (09)
DECEMBER	7.40 (18)	0.40 (08)	3.66 (23)	0.28 (08)
1973				
JANUARY	6.23 (09)	0.47 (14)	2.59 (04)	0.19 (14)
FEBRUARY	9.59 (25)	1.19 (04)	4.23 (02)	0.30 (07)
MARCH	7.11 (13)	0.62 (24)	7.41 (10)	0.31 (02)
APRIL	3.90 (14)	0.67 (27)	3.53 (14)	0.33 (04)
MAY	3.34 (31)	0.80 (13)	3.35 (25)	0.24 (04)
JUNE	2.63 (26)	0.26 (02)	2.75 (03)	0.32 (09)
JULY	2.70 (02)	0.39 (01)	2.53 (08)	0.45 (18)
AUGUST	2.77 (29)	0.49 (05)	2.75 (29)	0.22 (04)
SEPTEMBER	4.14 (24)	0.86 (01)	3.92 (26)	0.39 (27)
OCTOBER	5.55 (30)	1.01 (08)	7.69 (29)	0.40 (10)
NOVEMBER	6.70 (20)	1.03 (12)	5.85 (30)	0.39 (08)
DECEMBER	6.56 (31)	0.71 (16)	2.62 (31)	0.05 (15)

Table 14: Maximum and minimum values of the particulate lead aerosol concentration in µg m^{-3}, monitored at Site D. The numbers in parentheses are the dates when the levels shown were monitored.

	DAY TIME		NIGHT TIME	
1972	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
DECEMBER	5.88 (21)	0.37 (11)	3.62 (08)	0.20 (20)
1973				
JANUARY	5.58 (04)	0.37 (21)	3.67 (28)	0.17 (20)

Table 15: Maximum and minimum values of the particulate lead aerosol concentration in μg m^{-3}, monitored at Site E. The numbers in parentheses are the dates when the levels shown were monitored.

	DAY TIME		NIGHT TIME	
1972	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
DECEMBER	7.78 (21)	0.51 (27)	2.65 (21)	0.09 (08)
1973				
JANUARY	5.47 (16)	0.31 (21)	3.11 (16)	0.07 (20)
FEBRUARY	3.91 (17)	0.98 (22)	4.57 (16)	0.14 (12)
MARCH	5.01 (10)	0.62 (28)	6.06 (10)	0.36 (24)
APRIL				
MAY	3.14 (25)	0.30 (31)	0.91 (27)	0.37 (28)
JUNE	10.60 (17)	0.13 (03)	1.98 (07)	0.17 (19)
JULY	5.55 (05)	0.68 (01)	3.47 (06)	0.50 (01)

Table 16: Maximum and minimum values of the particulate lead aerosol concentration in μg m^{-3}, monitored at Site F. The numbers in parentheses are the dates when the levels were monitored.

	DAY TIME		NIGHT TIME	
1973	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
MAY	0.92 (18)	0.25 (28)	0.99 (25)	0.08 (31)
JUNE	1.06 (12)	0.01 (09)	1.61 (05)	0.07 (12)

Table 17: Maximum and minimum values of the particulate lead aerosol concentration in μg m^{-3}, monitored at Site H. The numbers in parentheses are the dates when the levels were monitored.

VAN VALIN & PARUNGO (1974) note that larger particles (radii 5 - 10 μm) are removed by inertial impaction with raindrops. Small particles (radii <0.01 μm) are removed by Brownian diffusion into cloud droplets. These studies leave a particle size range from 0.05 μm to 5 μm for which an atmosphere aerosol is stable. JUNGE (1963) found that when atmospheric aerosol particles are around 0.3 μm diameter the aerosol is very stable. This is supported in studies by ROBINSON & LUDWIG (1967) and LUNDGREN (1970).

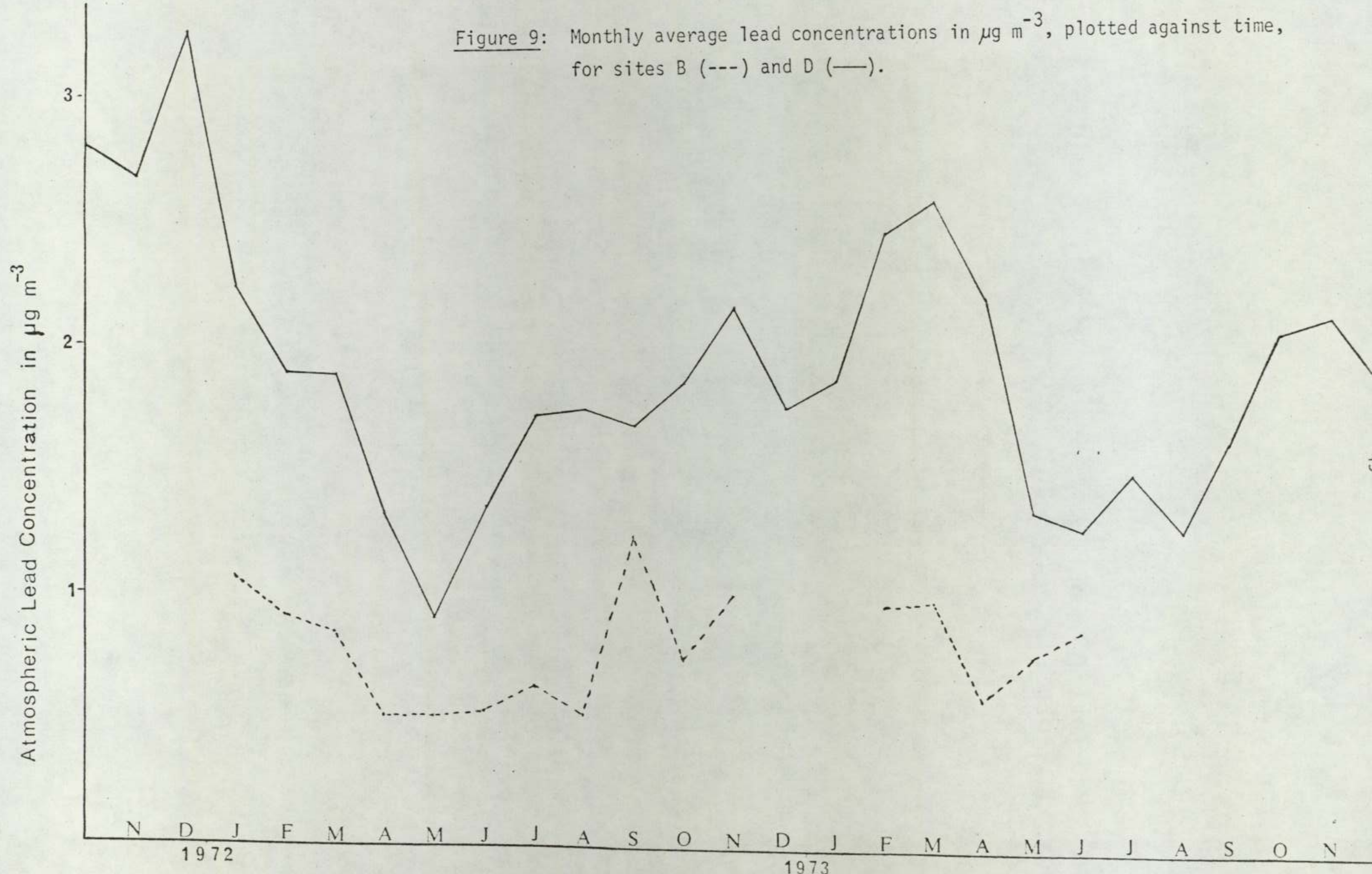
3.2.2 VARIATION WITH SEASON

The seasonal fluctuation of atmospheric lead concentration has been noted by CHOW & EARL (1970), LAWTHER, COMMINS, ELLISON & BILES (1972) and COLE & LYNHAM (1972) and is described as due to meteorological factors. This dependence on season can be seen in our results (Figure 9) where the higher levels can be seen to be found during the two winter quarters, October to December and January to March. The change in concentration in the atmosphere is caused by many factors including ordinary dilution, due to incoming fresh air, turbulent dilution, due to up currents, eddies and gustiness, turbulent coagulation, and wash-out by rain. To see if we could relate the change to the atmospheric conditions, we abstracted 11 meteorological variables and correlated the lead concentration to them.

3.2.3 VARIATION WITH WEATHER

LUNDGREN (1970) found the atmospheric level of lead to be correlated with relative humidity (correlation coefficient $r = 0.36$) and with wind speed ($r = -0.45$). To process our results the lead and particulate concentrations, the weather variables and the traffic total for each 12 h period were entered into the university ICL 1900 computer and analysed by its statistical programme XDS 3/22. The lead level was considered linearly dependent of each parameter and, for site D, of the eleven, three variables showed a fair correlation (Table 18). These were wind speed ($r = -0.28$), visibility ($r = -0.29$) and atmospheric pressure ($r = 0.28$).

Figure 9: Monthly average lead concentrations in $\mu g\ m^{-3}$, plotted against time, for sites B (---) and D (——).



	SITE D	SITE B
WIND SPEED	-0.28	-0.32
WIND DIRECTION	0.03	0.05
VISIBILITY	-0.29	-0.29
TEMPERATURE	-0.14	-0.21
RELATIVE HUMIDITY	-0.02	-0.08
ATMOSPHERIC PRESSURE	0.28	0.13
DEW POINT	-0.17	-0.19
VAPOUR PRESSURE	-0.14	-0.18
TOTAL RAINFALL	-0.07	-0.08
QUANTITY OF SUNSHINE	0.13	0.01
TOTAL CLOUD	-0.09	-0.21
TRAFFIC	0.32	0.07

Table 18: Correlation coefficients between lead levels measured at sites B & D and the eleven weather parameters.

The former has been found before so a negative correlation was expected as wind speed is a measure of ordinary and turbulent dilution. Visibility, dependent on wind speed and relative humidity, is a reciprocal measure of the moisture and dust load of the atmosphere, CHARLSON & PIERRARD (1969). Thus this negative correlation is not unexpected. No correlation was found with relative humidity (r = -0.02) although that with atmospheric pressure is appreciable. The former may be due to a reduction in variance of relative humidity around site D because of the presence of three cooling towers for apower station about 600 m East of Salford Circus. The dependence of lead level on atmospheric pressure is probably a mirror of the dependence on wind speed since these two are related (Table 19).

To develop a predictive equation, a better fit between predicted and actual levels can be obtained by incorporating more than one variable. Many of the weather parameters change in concert and to get an equation that allows for as much variance as possible, the variables used should be virtually linearly independent. Taking a coefficient of >0.4 to indicate dependence, a matrix of correlation coefficients (Table 19) shows wind direction, total rainfall and total cloud to be completely independent of other weather parameters. They also bear little relation to the lead levels monitored (Table 18). The wind speed is correlated to the visibility (r = 0.51) and the atmospheric pressure (r = -0.43), so in relating lead levels to more than wind speed, these latter must be excluded. Of the remaining five variables, the temperature has a correlation coefficient of greater than 0.4 with relative humidity, vapour pressure, quantity of sunshine, and dewpoint. Thus temperature may be used to represent the variability in meterological conditions not covered by wind speed. Monthly averages, maxima and minima of wind speed and temperature are given in Tables 20 - 23.

3.2.4 VARIATION WITH WIND DIRECTION

As mentioned above the wind direction does not show a linear correlation with lead levels. This is not unexpected for site D as it is at the centre

VARIABLE	1	2	3	4	5	6	7	8	9	10	11
1 WIND SPEED	1.00	0.29	0.51	0.09	-0.31	-0.43	-0.06	-0.09	0.08	0.15	0.15
2 WIND DIRECTION		1.00	0.21	-0.08	-0.16	-0.11	-0.16	-0.17	-0.06	0.03	-0.01
3 VISIBILITY			1.00	0.27	-0.57	-0.28	-0.01	-0.04	-0.09	0.35	-0.14
4 TEMPERATURE				1.00	-0.44	0.00	0.85	0.84	0.04	0.42	0.10
5 RELATIVE HUMIDITY					1.00	-0.01	0.07	0.07	0.17	-0.78	0.21
6 ATMOSPHERIC PRESSURE						1.00	-0.01	0.00	-0.31	0.06	-0.08
7 DEW POINT							1.00	0.98	0.14	0.01	0.25
8 VAPOUR PRESSURE								1.00	0.15	0.02	0.22
9 TOTAL RAINFALL									1.00	-0.10	0.21
10 QUANTITY OF SUNSHINE										1.00	-0.31
11 TOTAL CLOUD											1.00

Table 19: Matrix of correlation coefficients between weather parameters measured between October 1971 and July 1973.

1971	DAY TIME	NIGHT TIME	24h AVERAGE
OCTOBER	4.80	3.30	4.1
NOVEMBER	4.81	3.88	4.4
DECEMBER	4.22	4.12	4.2
1972			
JANUARY	4.89	4.17	4.5
FEBRUARY	4.51	3.48	4.0
MARCH	5.15	3.79	4.5
APRIL	6.65	5.02	5.8
MAY	6.29	4.07	5.2
JUNE	5.17	3.57	4.4
JULY	3.84	2.57	3.2
AUGUST	4.37	3.14	3.8
SEPTEMBER	3.34	2.24	2.8
OCTOBER	4.11	2.87	3.5
NOVEMBER	5.06	5.04	5.1
DECEMBER	4.91	4.59	4.8
1973			
JANUARY	4.11	3.57	3.8
FEBRUARY	5.35	4.66	5.0
MARCH	4.50	2.79	3.6
APRIL	5.89	4.35	5.1
MAY	5.87	3.93	4.9
JUNE	4.71	2.81	3.8
JULY	3.60	2.46	3.0
AUGUST	4.47	2.63	3.6
SEPTEMBER	4.53	2.94	3.7
OCTOBER	3.91	2.61	3.3
NOVEMBER	4.93	4.46	4.7
DECEMBER	5.33	5.07	5.2

Table 20: Monthly average wind speeds in m s^{-1}, as measured at Elmdon Airport, Birmingham.

1971	DAY TIME		NIGHT TIME	
	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
OCTOBER	9.56 (15)	0.77 (29)	9.82 (18)	0.38 (25)
NOVEMBER	10.07 (21)	0.38 (11)	10.33 (21)	0.00 (10/18)
DECEMBER	9.18 (11)	0.64 (27)	9.56 (20)	0.64 (05)
1972				
JANUARY	13.39 (27)	1.02 (06)	11.60 (26)	0.00 (05/30)
FEBRUARY	7.40 (02)	1.79 (07)	8.29 (02)	0.77 (03)
MARCH	11.60 (27)	1.79 (16)	10.33 (01)	0.00 (18)
APRIL	9.95 (02)	2.04 (13)	7.55 (28)	2.19 (16)
MAY	13.01 (27)	1.53 (18)	9.95 (25)	0.13 (16)
JUNE	8.29 (21/22)	2.30 (01/12)	9.18 (17)	0.38 (15)
JULY	7.01 (04)	0.64 (22)	7.14 (03)	0.00 (11/22)
AUGUST	7.91 (08)	1.02 (13)	7.01 (17)	0.26 (10)
SEPTEMBER	5.48 (11)	0.77 (26)	4.46 (08/19)	0.00 (26/27)
OCTOBER	7.78 (29)	2.30 (06/10)	6.76 (29)	0.00 (03/05)
NOVEMBER	9.69 (09)	0.64 (26)	12.75 (11)	1.02 (16)
DECEMBER	11.86 (05)	0.13 (18)	10.20 (11)	0.26 (17)
1973				
JANUARY	8.03 (27)	0.26 (09)	8.42 (14)	0.77 (05)
FEBRUARY	10.46 (12)	0.77 (26)	9.05 (22)	0.00 (25)
MARCH	8.03 (01)	0.51 (19)	8.16 (31)	0.00 (20)
APRIL	12.50 (02)	2.40 (27)	8.03 (01)	0.77 (25)
MAY	10.84 (10)	2.04 (24)	9.56 (07)	0.00 (24)
JUNE	11.09 (20)	1.79 (05/23)	7.65 (12)	0.13 (05)
JULY	6.76 (11)	0.51 (24)	6.63 (06/11)	0.13 (23)
AUGUST	10.20 (06)	1.02 (27)	10.46 (05)	0.00 (18)
SEPTEMBER	8.16 (28)	1.15 (20)	7.78 (29)	0.00 (08)

Table 21: Maximum and minimum values of the wind speed in m s^{-1}. The numbers in parentheses are the dates when the extremes occurred.

1971	DAY TIME	NIGHT TIME	24h AVERAGE
OCTOBER	12.44	8.55	10.5
NOVEMBER	6.69	4.56	5.6
DECEMBER	6.76	6.64	6.7
1972			
JANUARY	4.08	3.05	3.6
FEBRUARY	4.57	2.98	3.8
MARCH	8.36	4.60	6.5
APRIL	9.63	6.58	8.1
MAY	12.23	8.11	10.2
JUNE	13.47	9.89	12.2
JULY	16.15	13.98	15.1
AUGUST	16.90	12.47	14.7
SEPTEMBER	12.97	8.79	10.9
OCTOBER	11.51	8.29	9.9
NOVEMBER	6.88	5.42	6.2
DECEMBER	5.94	5.05	5.5
1973			
JANUARY	4.92	4.04	4.5
FEBRUARY	5.34	3.43	4.4
MARCH	7.75	3.30	5.5
APRIL	11.44	5.15	8.3
MAY	13.26	9.17	11.2
JUNE	16.51	12.49	14.5
JULY	17.09	12.93	15.0
AUGUST	18.76	13.47	16.1
SEPTEMBER	16.16	10.48	13.3
OCTOBER	10.34	7.26	8.8
NOVEMBER	6.50	4.91	5.7
DECEMBER	5.02	4.57	4.8

Table 22: Monthly average temperatures in ^{o}C as measured at Elmdon Airport, Birmingham.

	DAY TIME		NIGHT TIME	
1971	MAXIMUM	MINIMUM	MAXIMUM	MINIMUM
OCTOBER	19.00 (02)	5.50 (13)	14.25 (10)	0.00 (14)
NOVEMBER	12.75 (03)	1.50 (29)	11.18 (02)	-3.38 (10)
DECEMBER	13.15 (21)	1.15 (29)	11.98 (20)	1.85 (29)
1972				
JANUARY	8.40 (26)	-3.75 (31)	8.03 (23)	-8.25 (30)
FEBRUARY	6.70 (08)	-0.03 (01)	5.45 (02)	0.35 (09)
MARCH	13.85 (19)	2.20 (11)	10.35 (31)	0.12 (12)
APRIL	13.90 (01)	6.70 (23)	10.40 (01)	3.20 (24)
MAY	14.88 (22)	9.98 (15)	11.15 (23)	1.97 (16)
JUNE	16.10 (17)	9.50 (10)	13.60 (25/30)	5.80 (01)
JULY	21.80 (18)	10.45 (28)	19.98 (29)	6.73 (11)
AUGUST	20.80 (14)	1.50 (26)	15.50 (06)	7.40 (10)
SEPTEMBER	17.10 (01)	9.80 (09)	13.20 (03)	3.06 (25)
OCTOBER	14.50 (02/09)	7.10 (21)	12.79 (09/26)	0.94 (05)
NOVEMBER	14.20 (06)	1.70 (17)	13.17 (06)	-1.63 (24)
DECEMBER	11.70 (14)	0.20 (22)	12.13 (13)	-0.27 (22)
1973				
JANUARY	10.30 (23)	0.00 (17)	9.79 (23)	-2.30 (16)
FEBRUARY	10.00 (20)	-0.80 (15)	9.18 (06)	-4.81 (14)
MARCH	13.00 (23)	2.70 (11)	8.43 (03)	-2.68 (10)
APRIL	14.60 (26)	3.30 (02)	10.02 (04)	-1.49 (08)
MAY	19.70 (26/27)	8.50 (03)	14.87 (27)	2.82 (01)
JUNE	22.50 (16)	11.60 (20)	17.68 (18)	7.07 (04/13)
JULY	22.80 (05)	10.60 (04)	17.34 (05)	9.03 (07)
AUGUST	24.80 (16)	13.30 (21)	19.03 (16)	9.16 (17)
SEPTEMBER	23.20 (05)	10.00 (30)	17.27 (02)	5.04 (30)

Table 23: Maximum and minimum values of the temperature in degrees Centigrade. The numbers in parentheses are the dates when the extremes occurred.

of a circular source. A dependence on wind direction is expected for remote sites, and this was checked at sites B and C. The number of occasions that the wind comes from a particular direction and the wind speed are dependent on wind direction. Also averaging wind direction over twelve hours can mask gross fluctuations, so that conclusions should be carefully drawn.

At B, the motorway lies in a 70^0 arc from 170^0 to 240^0 from North. The results of considering 36, 10^0 sectors around the sampling site, averaging the lead values found in each sector and plotting these radially - the distance from the centre being proportional to the lead level, are shown in Figure 10. Lead levels were averaged over the year May 1972 to May 1973 and no enhancement was found due to the motorway. When the wind blew from Birmingham across the motorway to Slade Road schools, which it did 30 per cent of this time, the average lead concentration was 0.63 $\mu g\ m^{-3}$. When the wind was in any other direction, the level was 0.89 $\mu g\ m^{-3}$. The respective wind speeds being 4.9 $m\ s^{-1}$ within the arc of the motorway and 4.0 $m\ s^{-1}$ outside it.

A similar graph for site C is shown in Figure 11. This results from averaging levels found between March and August 1972. At Eliot Street schools the motorway subtends an angle of 60^0, from 310^0 to 10^0 from North. A mean lead value of 0.81 $\mu g\ m^{-3}$ was found when the wind was blowing from the direction of the motorway towards the sampling point, which it did 12 per cent of the time. When the wind came from outside this arc, a value of 0.72 $\mu g\ m^{-3}$ was obtained. The wind speeds for this period were respectively 4.3 and 5.0 $m\ s^{-1}$. Again no noticeable enrichment of the atmospheric lead aerosol at site C due to carry over from the motorway is found. This suggests that these sites are monitoring a background level of lead, which is c.0.8 $\mu g\ m^{-3}$.

The effect of wind direction was assessed in another way using two sites, 4 km North-East of Salford Circus. The sites, whose positions are shown in Figure 16, are beside the M6, which runs from South-East

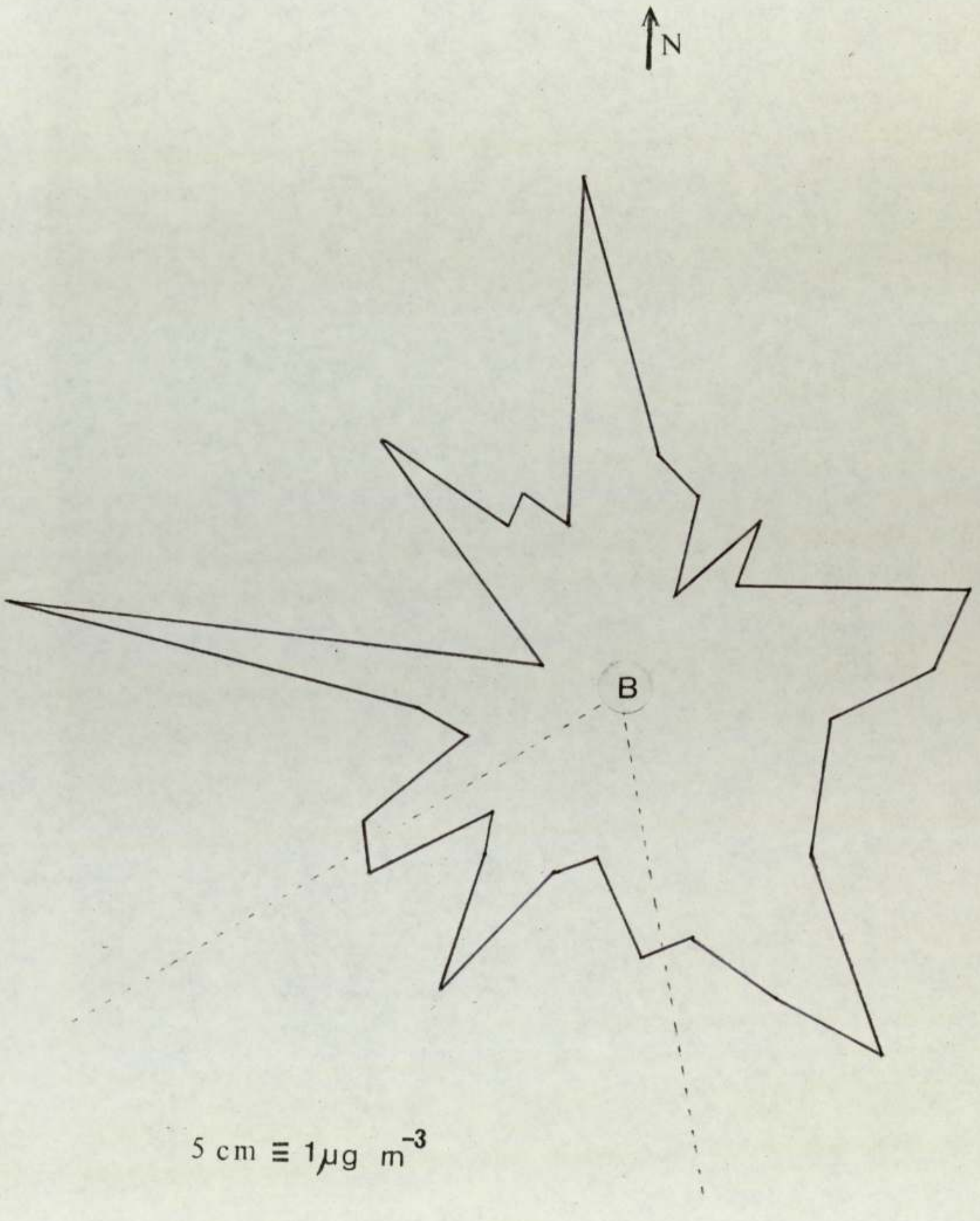


Figure 10: A radial plot of lead level against wind direction for site B.

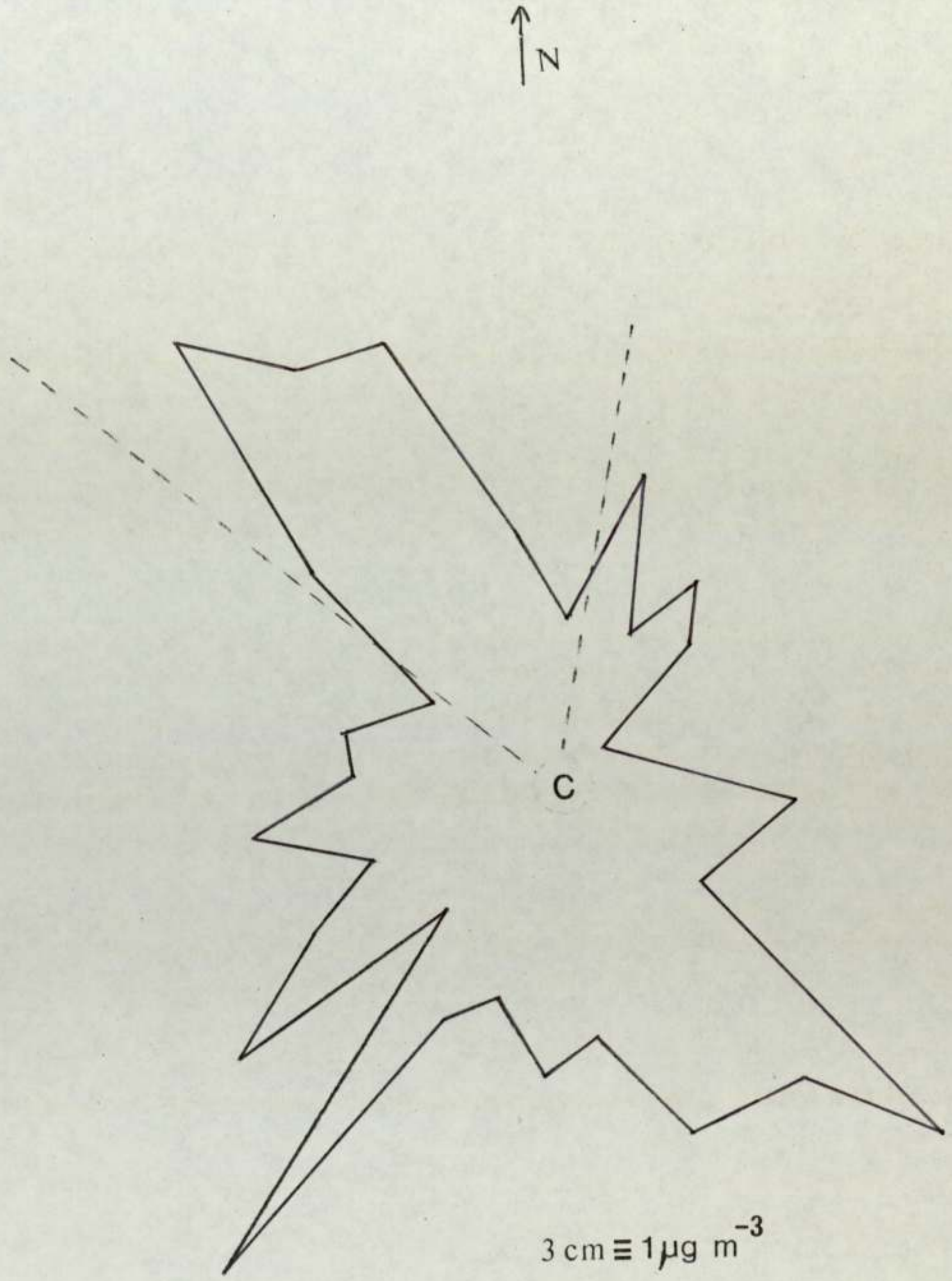


Figure 11: A radial plot of lead levels against wind direction for site C.

to North-West between two main roads. Altogether the lead concentration was monitored here for 37 days during April and May 1974. The lead levels found were compared with predicted values from a predictive equation for site D. The ratio of predicted to actual value was set against the appropriate wind direction, results for each 10° sector averaged and entered on Figure 12. This analysis is equivalent to multiplying the answer derived from a predictive equation by a factor which is dependent on wind direction.

$$(Pb)_{actual} = (Pb)_{predicted} \times WDR.$$

where WDR can vary between 0.0 and 1.0 depending upon the wind direction for that period. The 75 actual atmospheric lead determinations made during this period were compared with their predicted values from the equation including the wind direction factor and a Spearman rank correlation coefficient of 0.8 obtained. This shows the influence of wind direction in relation to the M6 road direction with respect to the sites monitored. The highest factors, 0.5 - 0.7, are associated with wind directions from 140 - 160° and 260 - 290° from North. These arcs are mainly aligned along the M6 for a distance of 750 - 1000 m on either side of the sites monitored. In contrast, winds blowing across the motorway in segments 0 - 120, 170 - 240 and 320 - 340° from North have low factors, 0.1 - 0.3.

3.2.5 VARIATION WITH TRAFFIC

It was mentioned in Section 3.2.1 that the decrease in lead level from day to night (25%) did not reflect the fall in traffic density (70%). This is either due to aerosol persistence or increased lead emission from other sources during the night time. The former seems more likely since the local industry works 24 h per day and the percentage lead in particulate varies little from day to night (from 1.37 to 1.29 per cent respectively). Better correlations between lead level and traffic are found at sites nearer the road, at site A $r = 0.40$, at site D $r = 0.32$, whilst at B $r = 0.07$.

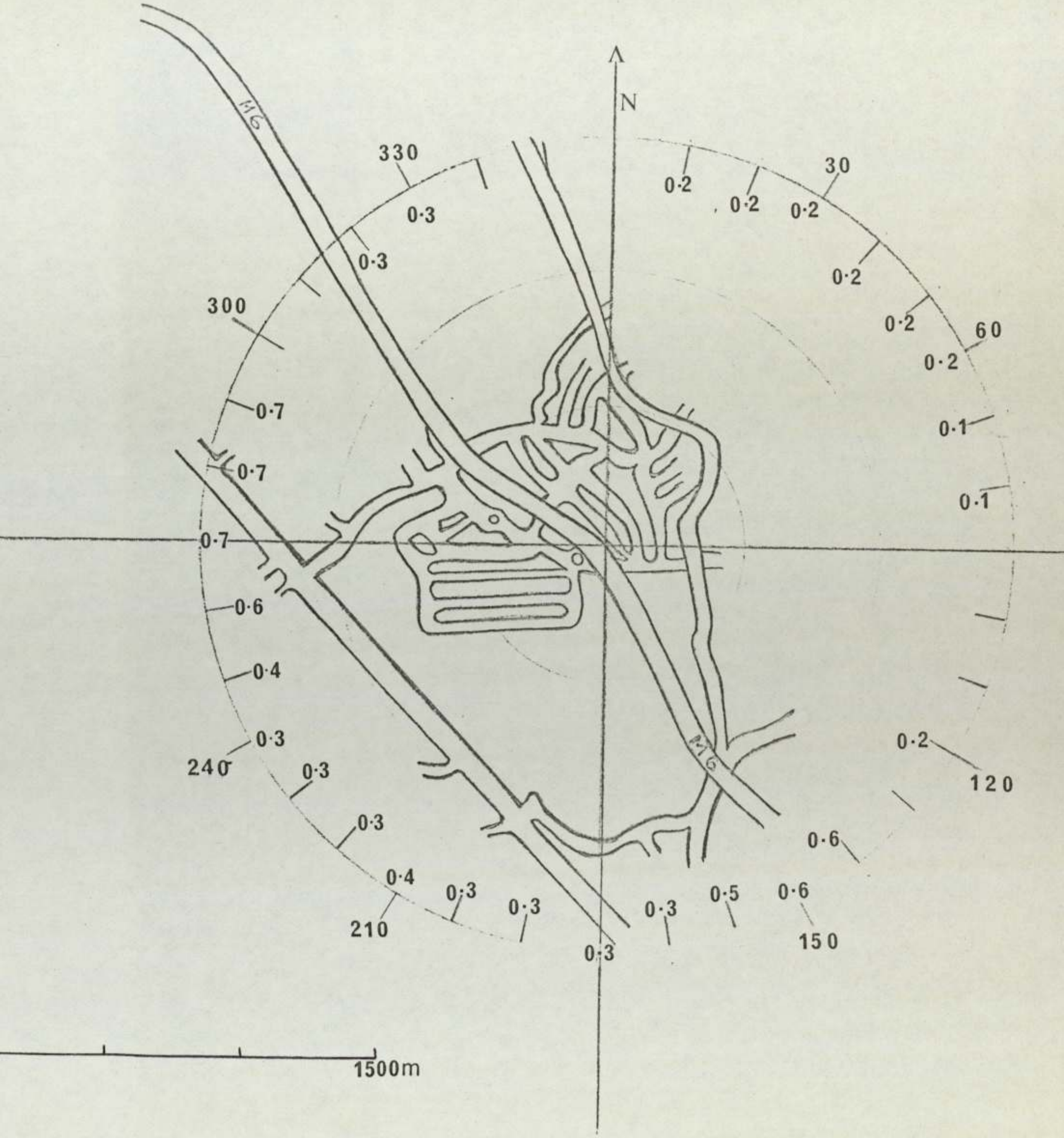


Figure 12: A diagram showing WDR factors placed around the compass for results from houses B and C.

The total traffic through this area has increased by a factor of 2.5 since the motorway opened in May 1972, Figure 13, Tables 24 & 25. The Salford Circus traffic has however decreased by 40 per cent, from 3.95×10^5 vehicles per week to 2.37×10^5 vehicles per week. The traffic load has thus been spread from the ground level road system to the multi-level interchange and elevated motorway. As a result the lead level at site D has decreased due to spatial dilution of sources. Meanwhile the levels monitored at site B have shown an increase from 0.66 $\mu g\ m^{-3}$ in February to June 1972 to 0.87 $\mu g\ m^{-3}$ in the same period during 1973, whilst the wind speed only dropped by 0.3 $m\ s^{-1}$. The stability of the aerosol at Slade Road schools is suggested by the fact that it shows little dependence on traffic ($r = 0.07$) otherwise having coefficients of correlation, with respect to the weather parameters measured, similar to those for site D.

3.3 PREDICTIVE EQUATIONS

Making certain assumptions, the amount of lead emitted into the atmosphere near the Gravelly Hill complex can be estimated. The proportion of petrol engine driven vehicles on the M6 and around Salford Circus averages 60 per cent whilst that along the A38(M) is about 80 per cent of the total, the others being mainly diesel propelled. Those with petrol engines are taken as consuming, at the rate of 11 km l^{-1}, fuel which contains on average 0.45 g of lead per litre. The amount of lead, originally in the petrol that is discharged, has been estimated at 75 per cent, HIRSCHLER, GILBERT, LAMB & NIEBYLSKI (1957) and Chamberlain (1974) or 50 per cent HABIBI (1970) and CRIDER, LEE, PATTERSON & WAGMAN (1971). Taking the latter figure, the quantity of lead emitted over 800 m sections of the M6 and A38(M), together with that from 400 m about Salford Circus, is 11.5 kg in one week or 10.2 $\mu g\ m^{-1}\ s^{-1}$. This figure is similar to ones quoted by BOVAY (1970) and CHAMBERLAIN (1974). The line source strength, in this case 10.2 $\mu g\ m^{-1}\ s^{-1}$, can

MONTH & YEAR	TOTAL NUMBER OF VEHICLES	
1972	DAY TIME	NIGHT TIME
APRIL	36,110	15,470
MAY	24,350	7,390
JUNE	25,250	7,650
JULY	23,490	8,140
AUGUST	23,240	8,630
SEPTEMBER	25,200	7,030
OCTOBER	26,740	7,480
NOVEMBER	26,510	7,220
DECEMBER	23,920	7,260
1973		
JANUARY	24,690	8,310
FEBRUARY	20,440	6,220
MARCH	18,150	5,580
APRIL	17,690	5,590
MAY	20,460	6,510
JUNE	26,480	8,490

Table 24: Monthly average 12 h traffic levels measured at Salford Circus April 1972 - June 1973.

Table 25:

(a) Total number of vehicles recorded at Salford Circus by the Streeter-Amet counters on a 24 h basis for 1 week in April 1972.

	Number of vehicles.
Monday	55,210
Tuesday	54,650
Wednesday	55,590
Thursday	57,180
Friday	59,170
Saturday	46,350
Sunday	32,890
WEEKLY TOTAL	361,040

(b) Total number of vehicles on the M6, A38(M) and Salford Circus Roundabout on a 24 h basis for one week in March 1973 and 1974 and January 1975.

	Total Number of vehicles.		
	March 1973	March 1974	January 1975
Monday	121,332	139,268	143,349
Tuesday	107,804	139,106	142,587
Wednesday	115,104	142,088	143,966
Thursday	116,742	143,977	145,699
Friday	116,703	152,548	148,153
Saturday	94,511	97,007	94,224
Sunday	76,443	86,457	76,954
WEEKLY TOTAL	748,639	900,451	894,932

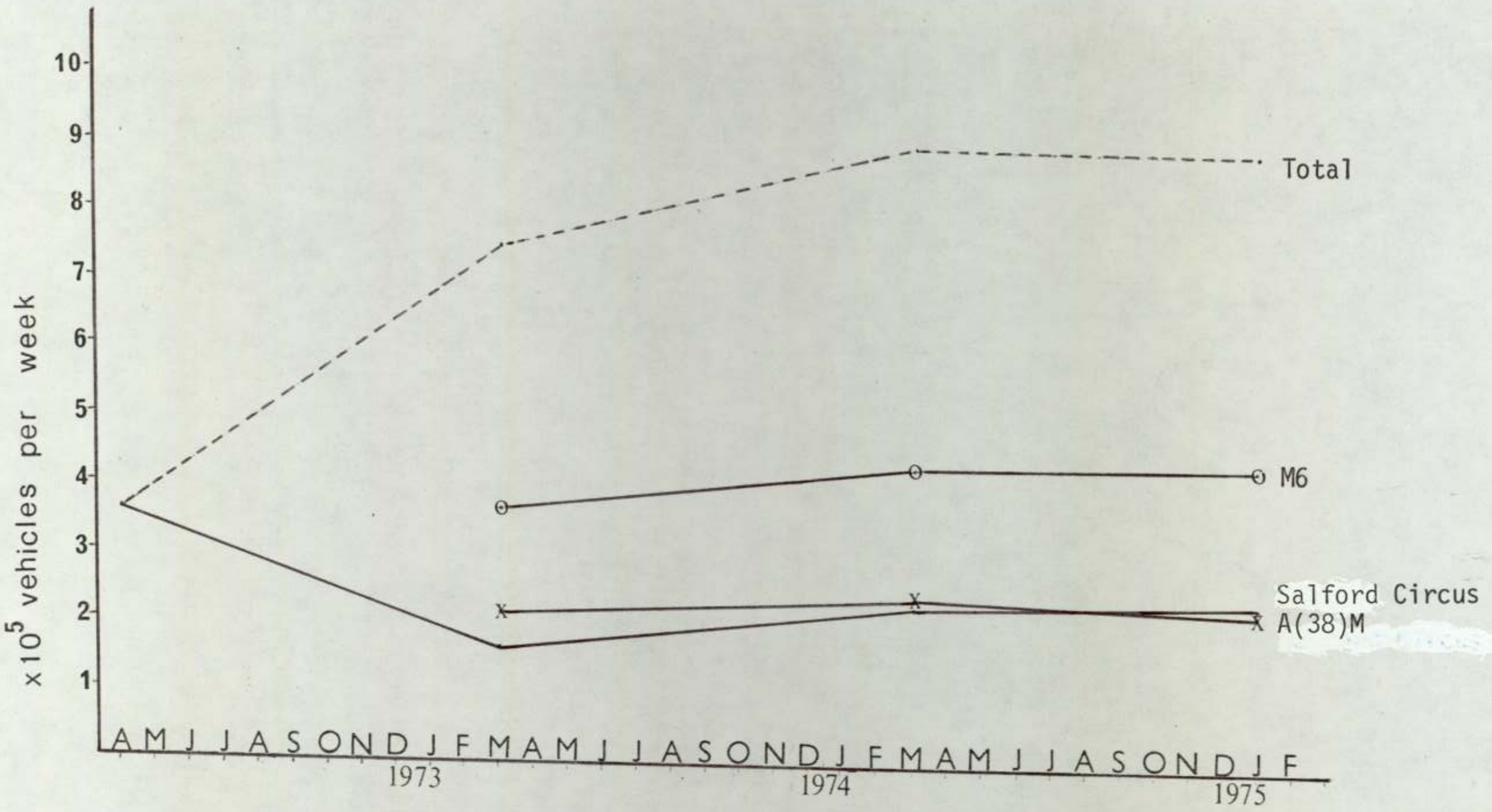


Figure 13: A graph showing the total, and component traffic densities around the junction from May 1972 - January 1975.

be used to predict the concentration, $\Theta(x,o)$ μg m^{-3}, at ground level x meters downwind of source with the equation

$$\Theta(x,o) = Q/uh$$

where $\Theta(x,o)$ is the concentration x m downwind of and o m above the source.

Q is the source strength in μg m^{-1} s^{-1}

u is the wind speed in m s^{-1} and,

h is the equivalent height of the pollutant cloud. This is predicted from the formulae $h = 1.26 \times \sigma_z$ PASQUILL (1961) and $\sigma_z = (0.5\ C_z^2)^{\frac{1}{2}} \times \exp(1 - n/2)$ SUTTON (1953) where σ_z is the standard deviation of the vertical distribution of pollutant in m, and in neutral conditions of atmospheric stability $C_z = 0.12$ and $n = 0.25$. σ_z may also be predicted by the equation $\sigma_z = a(x+c)^b$ where $a = 0.0856$ and $b = 0.8650$ when x is less than 500 m, CALDER (1973). Here a and b are the parameters determined with $c = 0$ so as to give a close fit to the PASQUILL-GIFFORD diffusion curves. Using the equation $\Theta = Q/u.h$ and the data from CHOLAK, SCHAFER & YEAGER (1968), DAINES, MOTTO & CHILKO (1970) and SCHUCK & LOCKE (1970), CHAMBERLAIN (1974) has produced the graph (figure 14). Our data, normalised to a traffic flow of 1000 vehicles per hour, is marked on the graph by the symbol D at (45 m, 1.23 μg m^{-3}). Thus estimating some parameters required for this formula shows our results are similar to those found elsewhere.

We have developed a less empirical equation to predict lead levels in the vicinity of Gravelly Hill using the weather variables mentioned earlier. Specifically the dependence of lead concentration on the two weather variables, wind speed and temperature and on traffic was found, for site D, to be:

$$(PBD) = 2.19 - 0.151\ (WSP) - 0.072\ (TMP) + 6.7 \times 10^{-5}\ (SCT)$$

where PBD is the average lead concentration for a twelve hour period at site D, in μg m^{-3}.

WSP is the average wind speed over twelve hours, in m s^{-1}.

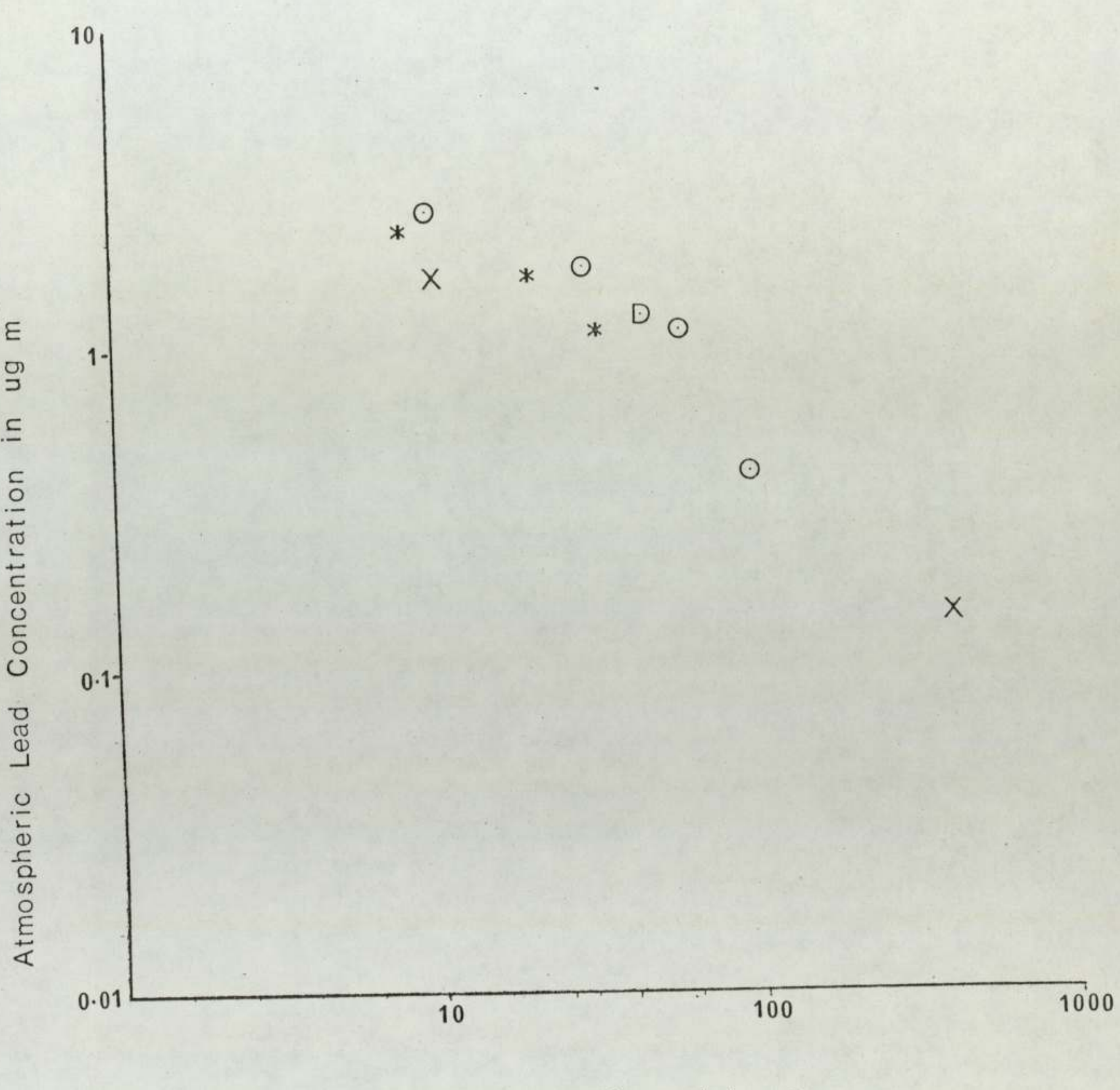


Figure 14: A graph showing the change of atmospheric lead levels with distance from a road carrying 1000 vehicles per hour. Results plotted are from DAINES, MOTTO & CHILKO (1970) - *, CHOLAK, SCHAFER & YEAGER (1968) - X, SCHUCK & LOCKE (1970) - 0, and this report - D.

TMP is the average temperature over twelve hours, in degrees centigrade.

SCT is the total traffic around Salford Circus in 12 hours. The predicted values of PBD correlate with the actual values giving a correlation coefficient of 0.53. This equation, modified to predict monthly averages, takes the form

$$(PBD) = 2.19 - 0.2\ (WSP) - 0.072\ (TMP) + 1.1 \times 10^{-6}\ (SCT)$$

These parameters, PBD, WSP, TMP and SCT, have the same units as before but the time scale is now one month instead of twelve hours. This equation has a correlation coefficient between actual and predicted values of 0.6 and a P value less than 0.01. The actual levels, at site D are shown plotted against the predicted levels, on a monthly basis, in Figure 15.

The twelve hour predictive relationship was calculated for site B:

$$(PBB) = 1.43 - 0.13\ (WSP) - 0.042\ (TMP) + 1.6 \times 10^{-5}\ (SCT).$$

where PBB is the average lead concentration for a 12 h period at site B. This has a correlation coefficient of 0.43 with the actual lead levels. Such equations were not calculated for the other sites but the relationship between the remote site and site D can give a prediction of the concentration at that position. It was found that lead levels monitored at sites A, B, C, E and H correlated well (average $r = 0.39$) with D. The equations linking sites are of the form

$$(PBB) = 0.45 + 0.168\ (PBD),$$

and the others like this one are found in Table 26. The constant is taken to represent the contribution to the lead level from local sources. The quotient of PBD is a measure of the dilution which occurs between the source, as measured at site D and the remote site. The correlations suggest either that the lead from traffic near Salford Circus is that collected at the other sites, or that the aerosol varies similarly at sites due to similarities of weather pattern within a circle of 500 m radius around site D. The fact that the correlations are better for sites close to site D than for those further removed (B and C) points to

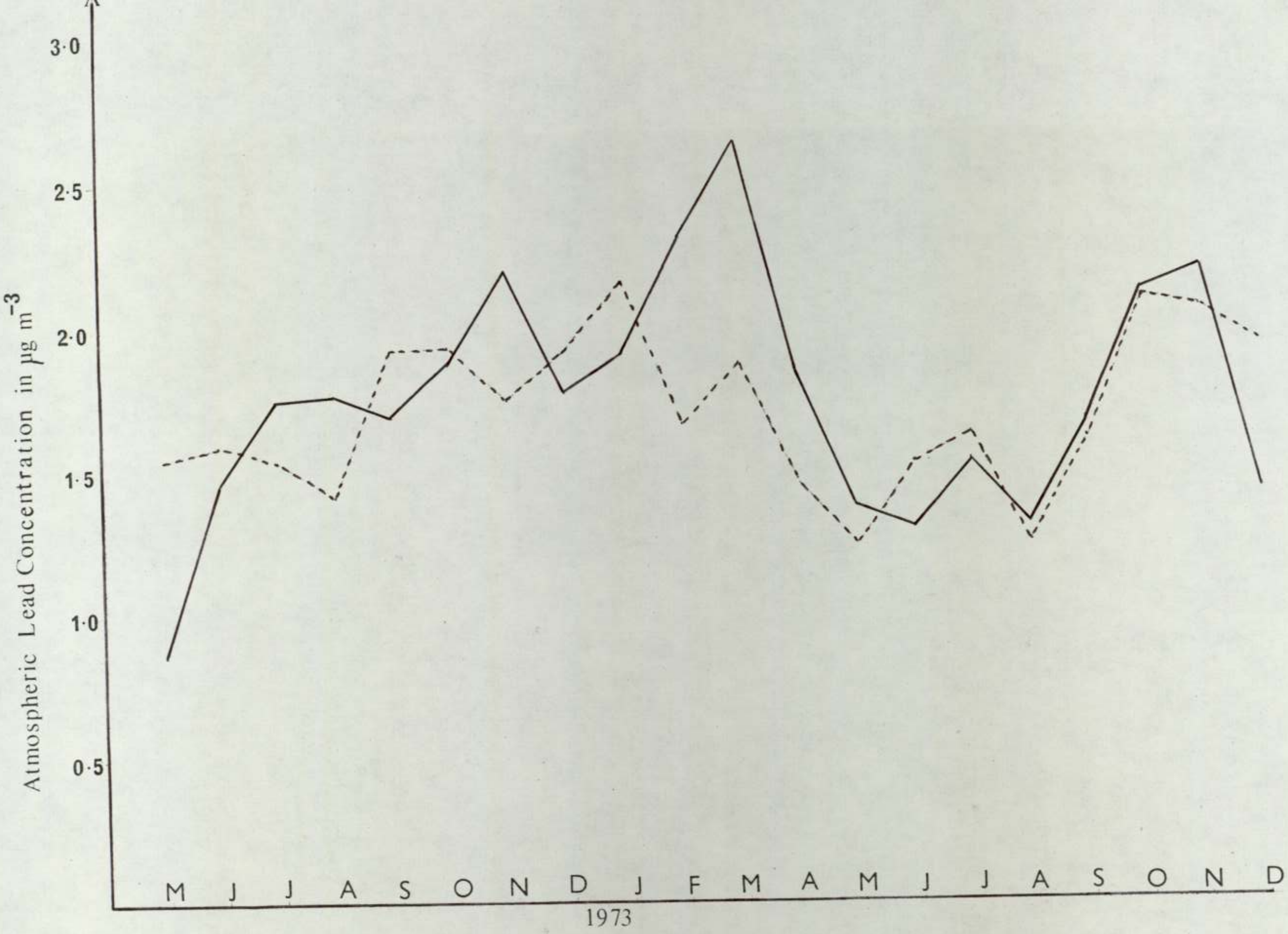


Figure 15: A graph showing the correspondence between the actual (—) and the predicted (---) monthly average lead levels monitored at site D.

$(PBA) = 0.29 + 0.597\ (PBD) \quad r = 0.51$

$(PBB) = 0.45 + 0.168\ (PBD) \quad r = 0.26$

$(PBC) = 0.45 + 0.171\ (PBD) \quad r = 0.34$

$(PBE) = 1.03 + 0.483\ (PBD) \quad r = 0.44$

$(PBF)^{*}= 0.68 + 0.481\ (PBE) \quad r = 0.49$

$(PBH) = 0.28 + 0.175\ (PBD) \quad 4 = 0.41$

* note PBF is predicted from PBE not PBD.

Table 26: The equations relating lead levels monitored at remote sites to those found at the reference site, D.

the former explanation for sites close to site D and the latter for remote sites. Thus while sites A, D, E and F are dependent on local traffic density and weather parameters, values found at sites B, C and H are more dependent on wind direction and other weather variables than on source strength. Hence weather factors which affect the transport of the lead aerosol are not picked out by differences in their correlation with lead between sites B and D.

3.3 THE EFFECT OF ATMOSPHERIC LEAD ON BLOOD LEAD LEVELS

GOLDSMITH & HEXTER (1967) used actual blood lead levels and estimated atmospheric concentrations to show that a logarithmic regression existed between these two. The relationship they found was:

$$\log_{10} \text{BLL} = 1.265 + 0.2433 \log \text{PBL}$$

where BLL is the blood lead level in μg per 100 ml whole blood and PBL is the ambient atmospheric lead level in $\mu g\ m^{-3}$. Putting a 24 h average level, from site D, of 1.75 $\mu g\ m^{-3}$ into this equation predicts the blood lead level of residents near Gravelly Hill to be 21.1 μg per 100 ml of blood. From a study by WALDRON (1975) of residents within 800 m of the Gravelly Hill interchange, the average blood lead level in October 1973 was 21.5 μg per 100 ml (Table 27) which agrees well with the prediction.

A relationship between increased atmospheric lead concentrations and increasing blood lead levels is supported by LUDWIG, DIGGS, HESSELBERG & MAGA (1965) and KNELSON, JOHNSON, COULSTON, GOLDBERG & GRIFFIN (1972). The former found a trend in atmospheric and blood lead levels from a rural to an urban environment. The latter paper contains a predictive equation to estimate the increase in blood lead level expected for a particular atmospheric concentration, which is

$$\text{IBL} = 0.327 \times L + 3.236 + (0.914L + 0.85)\times\log_{10}(L \times V \times R \times D \times 10^{-3})$$

where IBL is the increase in blood lead concentration in μg per 100 ml of blood.

DATE SAMPLES TAKEN	MALES (41 subjects)	FEMALES (58 subjects)	COMBINED AVERAGE
May 1972 (pre motorway opening)	14.41	10.93	12.67
October 1972 - February 1973	18.95	14.93	16.94
October 1973	23.73	19.21	21.47

Table 27: Data reproduced from the article by WALDRON (1975).

Lead levels in blood in µg per 100 ml.

L is the air lead concentration in $\mu g\ m^{-3}$.

V is the pulmonary ventilation in m^3 per day.

R is the fraction of inhaled lead that is retained.

D is the duration of exposure in days.

The average length of time between May 1972 and October 1972 to February 1973 is 240 days and in this time the blood lead level of 99 subjects increased by 4.27 μg per 100 ml on average. May 1972 to February 1973 is a period of about 550 days over which the increase averages 8.8 μg per 100 ml of blood. Setting $V = 20\ m^3$ per day, R = 35 per cent and $L = 1.75\ \mu g\ m^{-3}$ the predicted increases in blood lead level for 240 and 550 days are 4.88 and 5.74 μg per 100 ml respectively.

Normal blood lead levels range from 5 to 40 averaging 25 μg per 100 ml, KEHOE (1961), SCHROEDER & TIPTON (1968) and GOLDWATER & HOOVER (1967). Thus the values found near Gravelly Hill are within the range found elsewhere and in the normal range according to the categories of LANE et al. (1968).

3.5 PARTICULATE LEVELS

Particulate matter forms a stable atmospheric aerosol when the particulate size ranges from 0.05 to 5 μm. Such an aerosol has a maximum concentration as the removal mechanism, excluding wash-out by rain since it only rains 5 - 10 per cent of the time, is by coagulation precipitation. Atmospheric particulate concentrations were measured coincidentally with the lead levels. The 12 h average levels for the various sites are presented in Table 28. Levels monitored near the motorway - on average $160\ \mu g\ m^{-3}$ - are only slightly higher than those measured at a distance from it (on average $120\ \mu g\ m^{-3}$). This is presumably due to continuous renewal of the aerosol by traffic, which is known to emit particulate in the stable aerosol size range, HABIBI (1970). There appears to be a fall in the particulate levels monitored at sites B and C between before and after the motorway opened. This may be due to the difference in

SITE	DAY TIME	NIGHT TIME
A	178	127
B (pre motorway opening)	120	113
B (post motorway opening)	110	101
C (pre motorway opening)	156	118
C (post motorway opening)	121	114
D	156	106
E	245	159
F	154	120

Table 28: The averaged 12 h particulate concentrations in µg m^{-3} of the atmosphere at the various monitoring sites.

season during which the measurements were made.

The correspondence between lead and particulate is marked at site D. The predicted lead levels from the equation PBD = 0.81 + 0.009 PTD (where PTD is the 12 h average particulate level in $\mu g\ m^{-3}$ measured at site D) have a correlation coefficient with the actual levels of 0.65. Results from site B give the equation PBB = 0.43 + 0.004 PTB (where PTB is the 12 h average particulate level in $\mu g\ m^{-3}$ measured at site B) whose $r = 0.33$. The lack of correspondence at site B suggests somewhat different sources for the particulate and for the lead monitored there. Also the quotient of PTB is half that of PTD suggesting the particulate collected at site B contains less lead than that found at site D. The per cent content of lead in particulate figures for each site are given in Table 29. The lowest level is for a remote site in Wales, whilst the higher ones are found at sites A, D and F showing the dominance of a leaded particulate aerosol near traffic. The average of these, 1.2 per cent, is similar to levels found in Cincinnati by CHOLAK (1964), 1.42 - 1.85 per cent and in Cincinnati and Philadelphia by SMITH (1971), 1.7 and 1.5 per cent respectively. WALLER, COMMINS & LAWTHER (1965) found percentages of 0.8 in traffic and 0.6 as a background level in London. The percentage at site E is less than that at site F presumably due to the collection at site E of larger, short lived particles which contain a higher proportion of carbon and iron, HIRSCHLER,GILBERT, LAMB &NIEBYLSKI (1957).

Sites A, D, E and F can be expected to have monitored the traffic aerosol created by use of the junction. The particulate concentrations found at sites B and C have only a small correlation coefficient with those at site D (PTX = 85 + 0.2 PTD; $r = 0.25$ (where X = B or C)). This suggests that the aerosols monitored at sites B and C contained only a small proportion of the particulate generated by traffic on the junction. The ratio of particulate levels found at site B to that at site D is 0.83:1 whilst the ratio of lead levels is 0.47:1 showing the appearance of an external aerosol which dilutes the lead aerosol.

SITE	DAY TIME	NIGHT TIME
A	1.50	1.36
B (pre motorway opening)	0.87	0.82
B (post motorway opening)	0.83	0.72
C (pre motorway opening)	0.56	0.81
C (post motorway opening)	0.68	0.73
D	1.37	1.29
E	0.98	0.72
F	1.58	0.78
Remote site in Wales 24 h average	0.40	

Table 29: The average percentage of lead in particulate for the various monitoring sites.

Since the lead is only one per cent of the particulate collected what is the other 99 per cent? It is known that lead is emitted from car exhausts in conjunction with chlorine, bromine, carbon, iron and sulphur, HIRSCHLER & GILBERT (1957). Together these can make up 98 per cent of the exhaust particulate. However the atmosphere also contains an aerosol created by the wear of materials used by man, by soil erosion, by plants and by micro-organisms. These all exist in equilibrium; collisions causing agglomeration and sedimentation. The extent of particulate present due to re-entrainment of roadside dust depends on traffic density and speed, SEHMEL (1973). This was found to be between 10^{-3} and 1 per cent. The amount of asbestos in the atmosphere, possibly due to brake-lining wear, was determined by RICKARDS & BADAMI (1971) to be less than 0.1 μg m^{-3}. PIERSON, BRACHACZEK & KUMMER (1971) surveyed airborne particulate near a traffic tunnel for debris from tyres. They found that less than 40 per cent as much particulate matter came from tyre wear as from vehicle exhausts. Of the material we collected, all was oxidised or made soluble by the perchloric acid/nitric acid digestion, but we did not study the composition in more detail than to find the metallic content.

3.6 ALKYLLEAD LEVELS

A mixture of tetramethyl- and tetraethyllead together with methyl-ethyllead compounds is added to petrol as an antiknock agent. It can be absorbed through the skin, by mucous membranes and is preferentially distributed to the lipid phases of the body including the brain, ENGEL, HAMMER, MORTON, LANE & PLUMBLEE (1971). It is metabolised in the body to other complex ionic species of lead namely triethyl- and diethyllead, CREMER (1959). Triethyllead is formed in the liver, BRYCE-SMITH (1972) and this can cause significant decreases in the d-aminolevulinic acid activity of men working with leadalkyls, MILLAR, THOMPSON, GOLDBERG, BARRY & LOWE (1972). Triethyllead also causes exciteability, restlessness and loss of concentration. Also it inhibits tissue respiration in brain cells studied in-vitro. This form is relatively stable but is largely metabolised in 24 hours, to a dialkyllead. This is similar in physio-chemical properties to Pb^{2+} but has recently been shown to be genetically active, disrupting spindle fibre mechanism, AHLBERG, RAMEL & WACHTMEISTER (1972).

Most alkyllead added to fuel is destroyed during combustion, although LUTMER, BUSCH & MILLER (1967) found 3 per cent unburnt. Alkylleads may also reach the atmosphere from fuel-tank, carburettor and crank case evaporation. In Stockholm it was found that between 2 and 10 per cent of the total lead aerosol may be present as alkyllead vapour, LAVESKOG (1970). In Los Angeles a figure of 2 per cent was measured, STOPPS (1968) whilst for 6 cities in the U.S.A., PURDUE, ENRIONE, THOMPSON & BONFIELD (1973) found 4 - 14 per cent of total lead may be present as alkyllead. COLWILL & HICKMAN (1972) have monitored even higher levels, 7 - 21 per cent, on the central reservation of a motorway. The percentages represent concentrations between 0.10 and 3.57 $\mu g\ m^{-3}$ of organic lead.

We conducted a survey of alkyllead levels near site D, in Salford Park. The method used was largely that developed by MOSS & BROWETT (1968), although we undertook the final analysis spectrophotometrically. Initially samples took three weeks to collect as a very slow flow rate, $1.5\ l\ min^{-1}$, was used. The atmospheric levels of alkyllead, as measured during three periods between December 1972 and March 1973, were considered high since they represented, on average, 50 per cent of the total lead collected. A similar series of experiments was run in November and December 1973. These gave similar results to the previous series except when a filter of smaller pore size was used, STEWART (1974). The earlier trials were conducted with 0.8 μm filters acting as the particulate lead trap, latterly 0.45 then 0.22 μm filters were used. The percentage organic lead of the total decreased from 70 to 45 and then 15, suggesting the passage past the filter of some particulate material. Numerous difficulties were found with the method of determining organic lead and the results given in Table 30 must be regarded critically.

DATE in 1973	VOLUME SAMPLED m^3	FILTER PORE SIZE μm	ORGANIC LEAD LEVEL μg m^{-3}	PARTICULATE LEAD LEVEL μg m^{-3}
6/12 - 3/1	36.0	0.8	2.0	1.0
16/1 - 8/2	35.0	0.8	1.6	2.1
19/2 - 13/3	31.5	0.8	1.6	2.13
19/10-25/10	9.2	0.8	2.2	1.1
29/10 - 2/11	6.4	0.8	-	2.3
2/11 - 8/11	8.8	0.8	3.4	1.0
9/11 -15/11	9.3	0.8	0.29	0.08
19/11-22/11	4.4	0.8	-	2.2
23/11-29/11	8.3	0.45	1.8	2.0
29/11- 6/12	7.3	0.22	-	2.0
7/12-10/12	3.7	0.22	0.38	0.7

Table 30: Data from analyses of organic and particulate lead levels in Salford Park taken during 1973.

CHAPTER 4. RESULTS OF MONITORING AT OTHER SITES INSIDE AND OUTSIDE THREE HOUSES IN THE VICINITY OF GRAVELLY HILL.

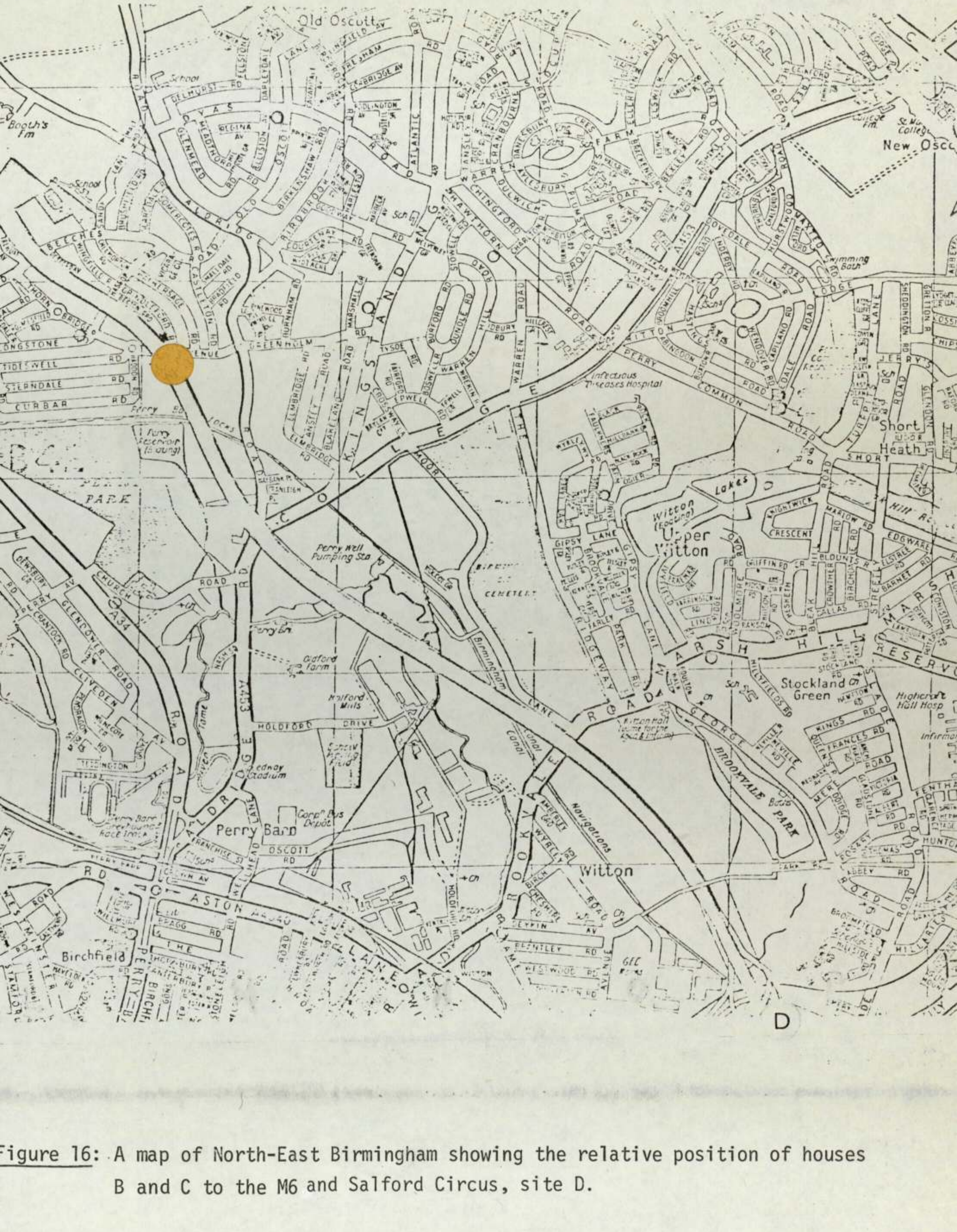
4.1 INTRODUCTION

It was noted by JACOBS, GOLDWATER & FERGANY (1962) that little work had been done on the characteristics of indoor suspended particulate, this still seems to be true. Their paper suggests that the particulate loading of an inside atmosphere is equal to or slightly higher than the corresponding outside one. FUGAS, WILDER, PAUKOVIC, HRSAK & STEINER-SKREB (1972) made a study to assess the total exposure of Zagreb dwellers to atmospheric particulate lead. This required the measurement of the lead contents of internal and external atmospheres. Their three sites showed that indoor levels were about 67 per cent of the outdoor values.
DAINES, SMITH, FELICIANO & TROUT (1972) looked at the lead concentrations of domestic atmospheres in homes at three distances from a main road. They found that at 4 m penetration was 50 per cent, at 38 m, 62 per cent and at 122 m, 70 per cent. LEE (1972) reports particle sizes and particulate concentrations inside and outside some public buildings in Hartford, Connecticut. These places have, on average, an indoor particulate level 75 per cent of the outdoor one. Also, whereas 40 per cent of the external particulates was less than 0.5 μm in diameter, the corresponding figure for the internal particulate was greater than 60 per cent.

We monitored the atmosphere inside and outside three houses near the Gravelly Hill interchange:

PROPERTY A: A semi-detached house in Powick Road about 160 m from Salford Circus and c. 25 m above the level of the roundabout.

PROPERTIES B & C: These were both 4 km distant from Salford Circus, at the Thornbridge Avenue-Grindleford Road junction, but as can be seen, in Figures 16, 17 and 18, were close to the motorway. Both properties were similar in design and age.



igure 16: A map of North-East Birmingham showing the relative position of houses B and C to the M6 and Salford Circus, site D.

Figure 17: A photograph of house B showing the proximity of the M6 and the monitor placed above the front porch.

Figure 18: A photograph of house C showing the proximity of the M6 to this double glazed property. (extreme left).

4.2 HOUSE A

4.2.1 EXPERIMENTAL

A series of synchronous samples were taken inside and outside this house during the period 29.11 to 19.12.1973. The time sequence was arranged to be as similar as possible to that used at site D, i.e. two samples were taken per day between the times 07.30h to 19.30h and the other from 19.30h to 07.30h. The resulting filters were all analysed in the same manner according to the procedure in Section 2.3.

As mentioned above, the house lay 160 m from the monitor at Salford Circus and about 25 m above it. At the house, efforts were made to disturb daily routine as little as possible. Only two points were specifically mentioned:

(a) the windows in the room containing the monitor should be kept closed, which was not unreasonable for the time of year.

(b) the door to the hallway should be kept open so that the air within the house could equilibrate freely with the atmosphere of the room in which we were monitoring.

4.2.2 RESULTS AND DISCUSSION

At the time of sampling, the traffic density around Salford Circus was about 240,000 vehicles per week of which c.60 per cent had petrol engines. On the motorway and access roads the volume of traffic was 690,000 vehicles per week, approximately 420,000 of which were petrol driven.

The averaged results are given in Table 31, and the 12 h levels are presented graphically in Figures 19 and 20. The first shows the daytime values from site D, plotted alongside those from inside and outside the house. The second graphs corresponding levels monitored at night. It is apparent from Table 31 that there is little difference between the concentrations inside and outside the house, both being about 0.9 μg m^{-3}.

From Figures 19 and 20, the close relationship between inside and

	INSIDE HOUSE	OUTSIDE HOUSE	SITE D
Daytime	1.07	1.14	2.16
Night-time	0.66	0.62	0.98
Combined Daytime/ Night-time Average	0.87	0.89	1.57

per cent penetration 98

Table 31: Atmospheric lead concentrations in $\mu g\ m^{-3}$ found inside and outisde a house, in the vicinity of an urban motorway, between November 29th and December 19th, 1973.

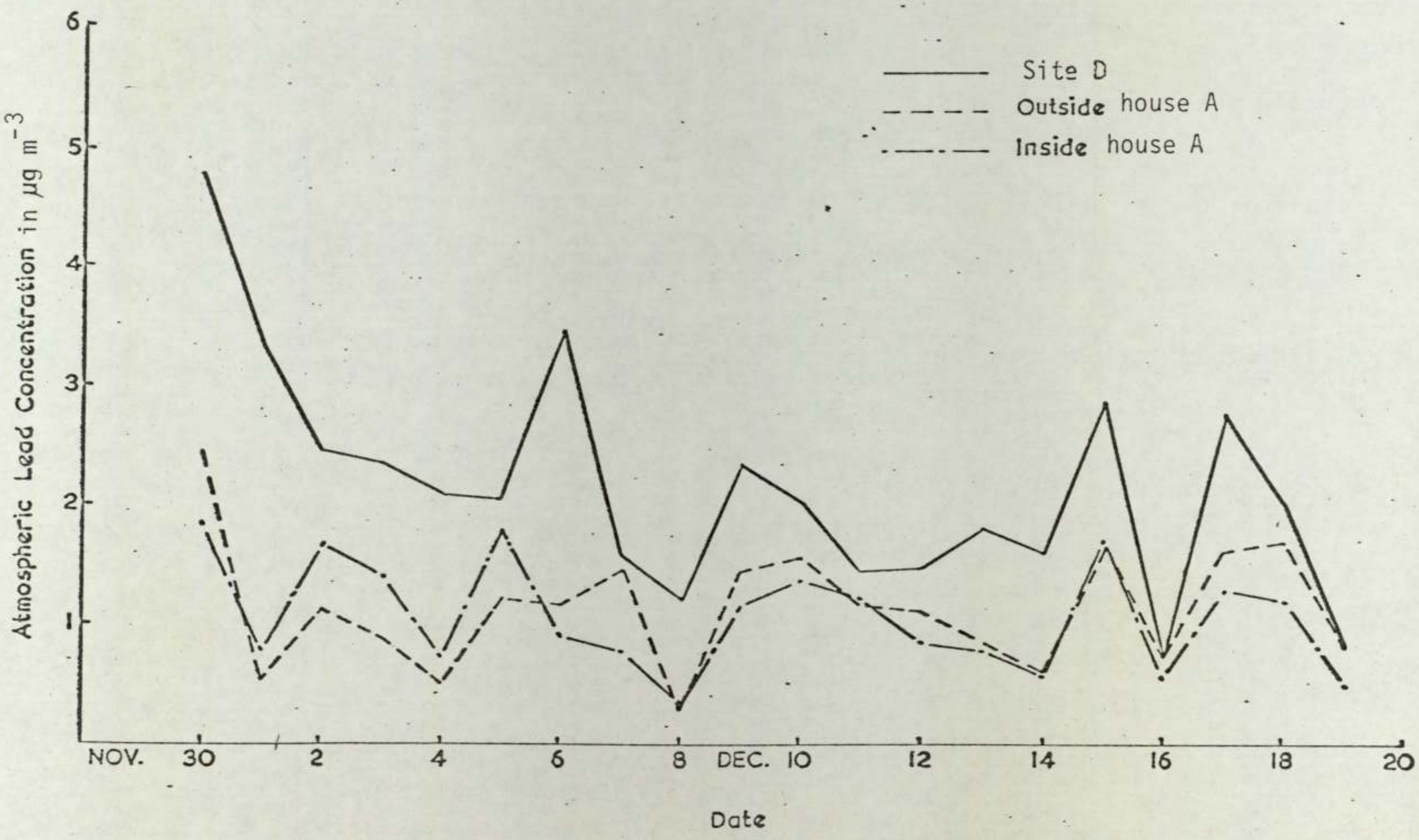


Figure 19: A graph of lead levels monitored at house A during the day time periods against the date on which they were found.

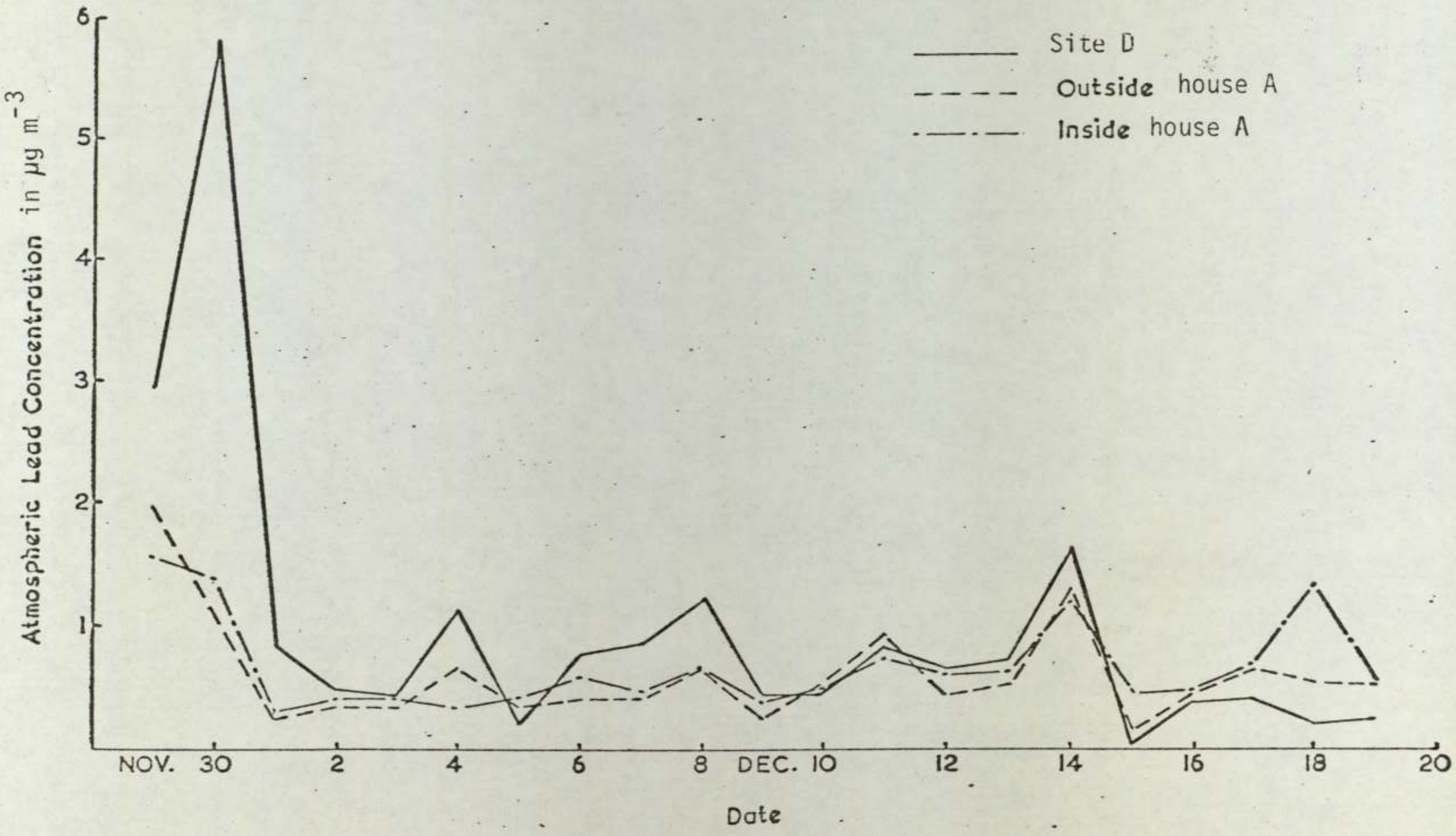


Figure 20: A graph of lead levels monitored at house A during the night-time periods against the date on which they were monitored.

outside levels can be seen. This suggests that the aerosol in the house is being continuously renewed from the external one, and that the house therefore affords little protection from the external lead aerosol. This is in keeping with the study by DAINES, SMITH, FELICIANO & TROUT (1972) who found increasing per cent penetration into houses with distance from a main road. The change in lead aerosol with distance is largely due to loss of larger particles, therefore per cent penetration is probably dependent on particle size, indeed LEE (1972) found a predominance of particles with smaller sizes inside buildings. This is significant with respect to the ability of the aerosol to penetrate and be retained in the lungs.

The lead level found at the house is 56 per cent of that monitored at the roundabout. This is slightly higher than is expected from literature values of decrease in concentration with distance. It may be due to the time of year when this study was conducted, which is condusive to the maintenance of atmospheric aerosols. The penetration of the house may also be seasonally dependent since in times of high humidity particulate is less likely to agglomerate CHAMBERLAIN, CLOUGH,HEARD, NEWTON, STOTT & WELLS (1975). However traffic figures suggest 70 per cent of the local, vehicle produced lead aerosol derives from free flowing motorway traffic which tends to produce particulate in the smaller size range.

The internal concentration of lead is more frequently higher than the corresponding external one during the night. Thus the fall in level from day to night outside the house is not reflected inside, which may be due to differences in removal mechanisms. However at night the correlation, between inside and outside levels (r = 0.94), is better than that during the day (r = 0.74). This suggests that an equilibrium condition, between the internal and external aerosols, is not immediate due to the interference of a permeable barrier, the walls and windows of the house. The stability of external conditions at night gives rise to an increased likelihood that the state of equilibrium may be reached.

The lead levels at night also vary over a smaller range than do those found during the daytime. These two factors combine to give a higher correlation between values found inside and outside at night compared to the more variable daytime levels.

Assuming the concentrations measured inside to be linearly dependent on those found outside, equations were developed relating the two, Table 32. If the internal aerosol was totally derived through equilibrium with the external one, then the constant in the equation, that proportion of the inside lead level which is independent of the outside lead concentration, should be zero. The fact that it is higher (0.33) during the day than at night (0.17) suggests that the constant may appear due to activity within the house stirring up particulate which has settled or been brought in on clothing. It may also be due to differing modes of penetration by the external atmosphere, perhaps dependent upon meteorological factors. The removal effect of precipitation and the dilution effect of high wind speeds are not reflected inside so the constant may be a compensator for low external values.

4.3 HOUSES B and C

4.3.1 EXPERIMENTAL

These are considered together as they are close spatially, both being situated at a point where the road on which they lie is traversed by an elevated section of the M6 (Figures 17 and 18). Both houses were semi-detached and three bedroomed, but one had roof insulation and double glazing in all rooms except the kitchen and bathroom.

The experimental procedure was as for house A, the sampling schedule being synchronised with the monitor at Salford Circus. The equipment was installed in house B in the downstairs front room and above the front porch, during the period 01.03 to 22.03.1974. At the other house, C, they were placed in the upstairs back bedroom and on the flat roof of the garage for the time 26.04 to 15.05.1974. The external monitor was,

<u>Table 32</u>: Regression equations for the dependence of lead concentrations in $\mu g\ m^{-3}$ found inside on those monitored outside House A.

Time of Day	Equation *	Validity Limits	Correlation Coefficients r.
DAYTIME	$(Pb)_I = 0.66\ (Pb)_0 + 0.33$	$0.27 < (Pb)_0 < 2.46$	0.74
NIGHT-TIME	$(Pb)_I = 0.82\ (Pb)_0 + 0.17$	$0.27 < (Pb)_0 < 1.96$	0.94

* $(Pb)_I$ denotes lead concentration inside House A

$(Pb)_0$ denotes lead concentration outside House A.

in both cases, about 10 m from the parapet wall of the motorway. The arrangements made with the occupants of these two houses were the same as at house A.

4.3.2 LEAD LEVELS: RESULTS AND DISCUSSION

The two periods over which samples were taken had similar average wind speeds (2.37 m s^{-1}, house B: 2.47 m s^{-1}, house C) and similar average traffic densities on the M6. The density during monitoring at house B was 3.81×10^5 vehicles per week, whilst that during monitoring at the second house was 4.09×10^5 vehicles per week. 80 per cent of these vehicles use the road during the daytime. Not all the vehicles were motor cars and from a number of manual counts of the M6 traffic it was found that on average 60 per cent were petrol propelled.

The averaged results are given in Tables 33 and 34, their 12 h levels are plotted consecutively for house B in Figure 21 and for house C in Figure 22. The results were analysed by the university computer in a similar fashion to the previous data. This gave regression equations for the dependence of the values monitored inside on those outside and those outside on ones from Salford Circus, Table 35.

Again the close relationship between lead levels found outside and inside the house can be seen, Figures 21 and 22. This is reflected in the correlation coefficients between these when a linear dependence is considered, Table 35. It is noteworthy that the lower correlation ($r = 0.92$, house B: $r = 0.76$, house C) is found for the house with double glazing. This suggests that the barrier between internal and external atmospheres has been made less permeable and equilibrium, between inside and outside aerosols, less easily attainable. The equations of dependence are:

$Pb_i = 0.11 + 0.59\ Pb_o$ for house B.

$Pb_i = 0.12 + 0.46\ Pb_o$ for house C.

These support the former assertion since the quotient of Pb_o is lower

Table 33: Atmospheric lead concentrations in $\mu g\ m^{-3}$ at House B monitored between March 1st and 22nd, 1974.

	Inside House	Outside House	Salford Circus	Percentage Lead Inside House/Outside House
Daytime	0.70	1.01	2.99	69
Night-time	0.68	0.96	1.53	71
Combined Daytime-Night-time average	0.69	0.98	2.26	70

Table 34: Atmospheric lead concentrations in $\mu g\ m^{-3}$ at House C monitored between April 25th and May 15th, 1974.

	Inside House	Outside House	Salford Circus	Percentage Lead Inside House/outside House
Daytime	0.49	0.75	1.56	65
Night-time	0.39	0.66	0.97	59
Combined Daytime-Night-time average	0.44	0.70	1.26	63

Figure 21: A graph comparing particulate lead concentrations in $\mu g\ m^{-3}$ inside (—) and outside (---) house B for consecutive day (⊙) and night (⊡) values.

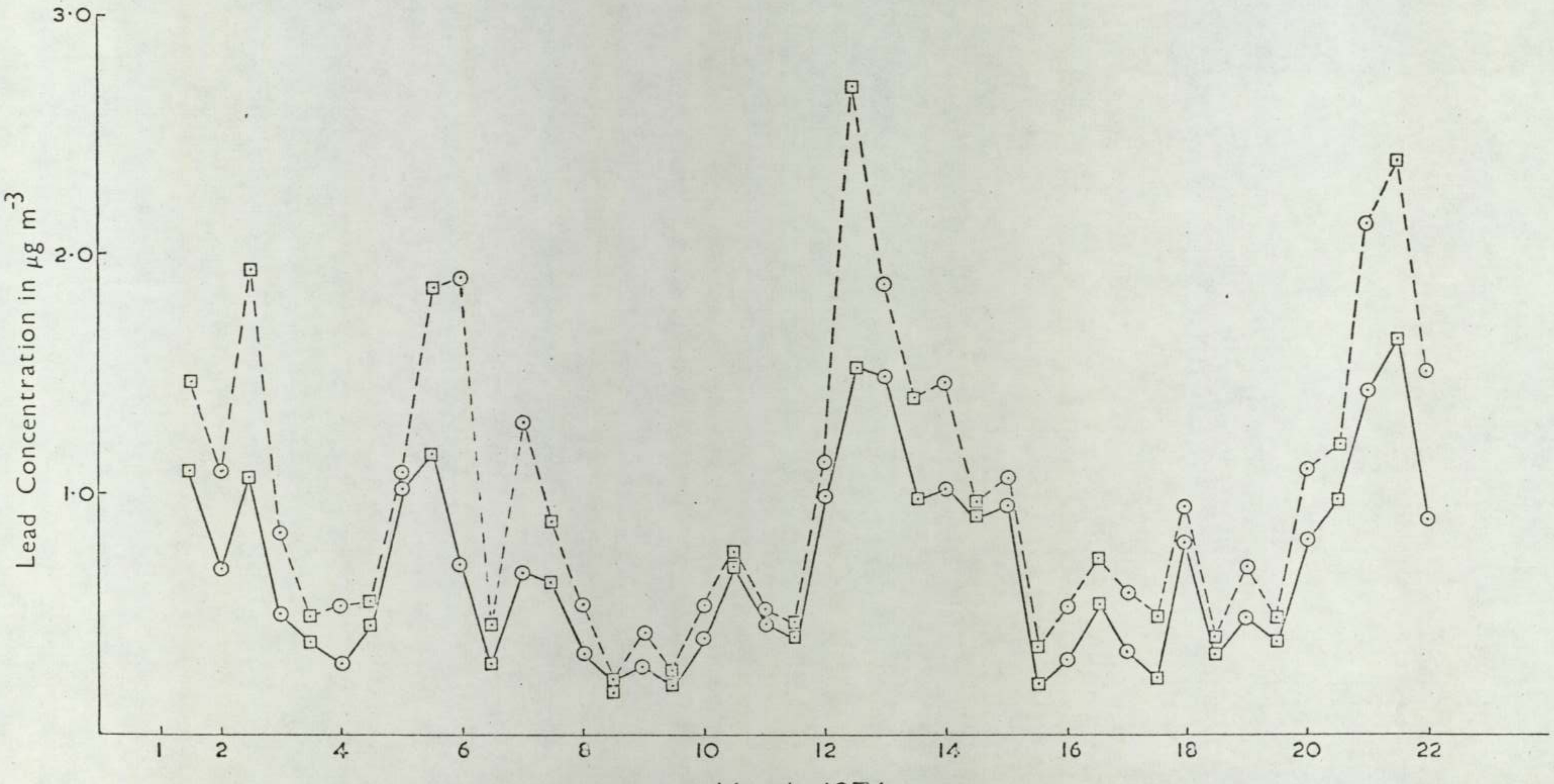


Figure 22: A graph comparing particulate lead concentrations in $\mu g\ m^{-3}$ inside(—) and outside (---) house C for consecutive day (⊙) and night (⊡) values.

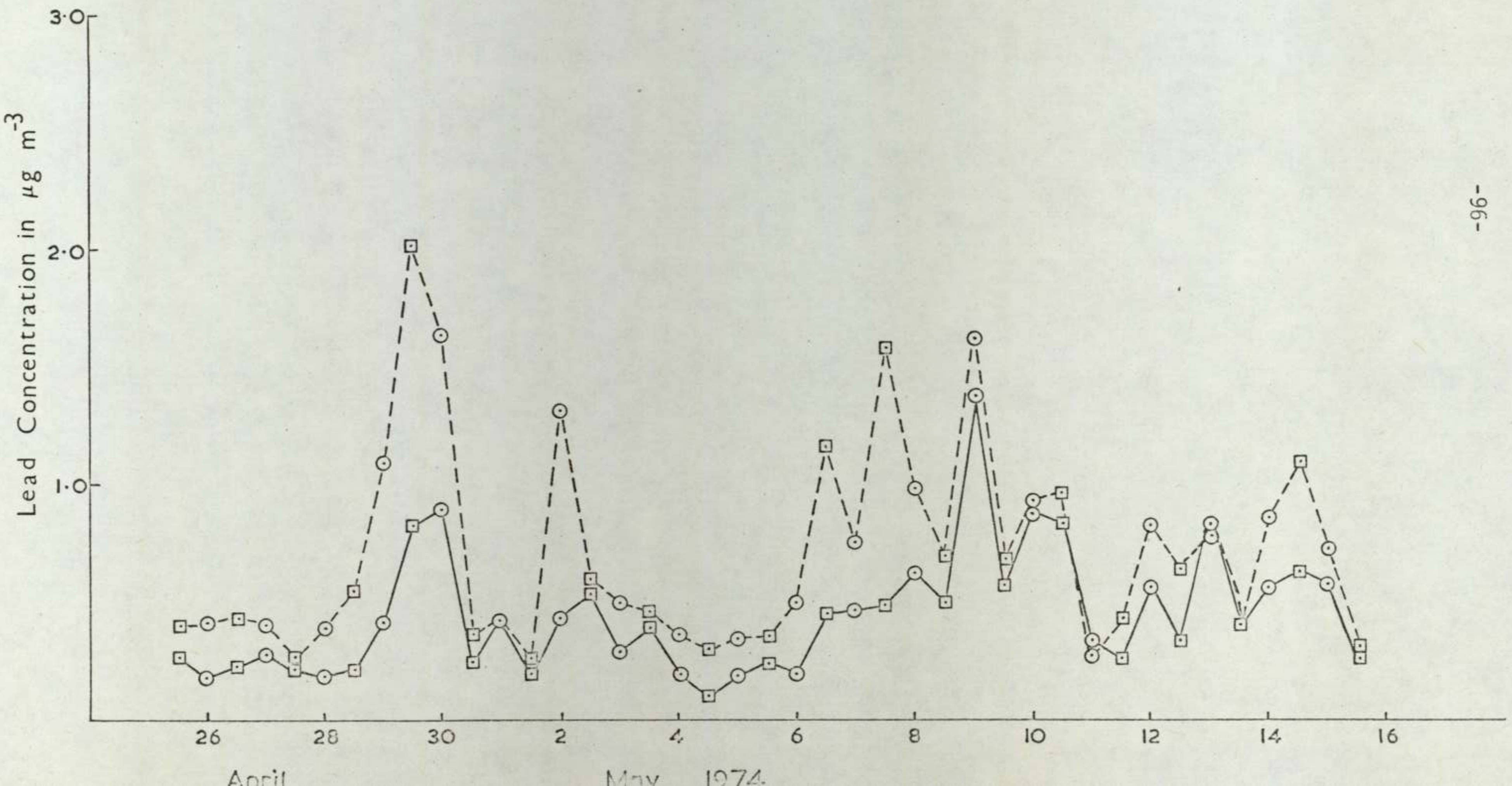


Table 35: Regression equations for the dependence of lead concentrations measured inside on those outside Houses B and C.

Locations	Day Equation *	r	Night Equation *	r	Day and Night Combined Equation *	r
House B Inside-Outside	$Pb_i = 0.61Pb_o + 0.09$	0.85	$Pb_i = 0.59Pb_o + 0.12$	0.96	$Pb_i = 0.59Pb_o + 0.11$	0.92
House C Inside-Outside	$Pb_i = 0.59Pb_o + 0.05$	0.80	$Pb_i = 0.33\ Pb_o + 0.17$	0.75	$Pb_i = 0.46Pb_o + 0.12$	0.76
House B Outside-Salford Circus	$Pb_o = 0.19Pb_d + 0.45$	0.64	$Pb_o = 0.49Pb_d + 0.21$	0.75	$Pb_o = 0.22Pb_d + 0.48$	0.59
House C Outside-Salford Circus	$Pb_o = 0.34Pb_d + 0.22$	0.60	$Pb_o = 0.51Pb_d + 0.17$	0.85	$Pb_o = 0.39Pb_d + 0.21$	0.72

* Pb_i denotes lead concentration inside the house
Pb_o denotes lead concentration outside the house
Pb_d denotes lead concentration at Salford Circus, site D.

for house C than for houses A and B. Also house C is the only one to show a lower correlation between the inside and outside night-time lead levels. This may be due to the effect of the roof insulation reducing upward heat loss and thus reducing convection currents, which would induce in-drafts. Such drafts would favour rapid equilibrium formation but these are presumably also inhibited by the decrease in permeability due to double glazing. The equations relating lead concentrations outside to those inside for the two houses B and C, both have constants which are less during the day (0.05 and 0.09) than at night (0.12 and 0.17). The constant is less likely to be due to activity within the house than to be a compensating factor for periods of low external lead levels. The per cent penetration for these two houses B and C is 70 and 63 respectively. Thus there does seem to be some protection afforded by double glazing. The penetration is lower here than at house A presumably due to the proximity of the source of the lead aerosol.

The degree of correlation between the levels measured outside the houses and those found at site D is good ($r = 0.59$, house B: $r = 0.72$, house C). Thus a concentration determined at Salford Circus can give a fair indication of that to be found 4 km distant - but close to the motorway. The data was also analysed to compare predicted values for site D, from the equation in Section 3.3, with the levels monitored outside the houses. Such comparison requires wind direction to be taken into account. The results of this are mentioned in Section 3.2.4 and show a correlation of 0.8 between concentrations, measured outside the two houses, and those predicted for site D. This extent of correspondence suggests that the aerosol at Thornbridge Avenue, around houses B and C has a meteorological dependence similar to the aerosol at site D. This is of interest since the proximity of the source, in the former case, would cause the mean aerosol size to be larger than at site D.

4.3.3 PARTICULATE LEVELS: RESULTS AND DISCUSSION

Figures 23 and 24 indicate that particulate concentrations inside follow the fluctuating values existing outside the houses. The average levels found for all three properties are given in Table 36. This, surprisingly, shows a larger penetration of particulate into the house with double glazing than into the one without. However the equations of dependence of particulate concentrations inside on that outside, Table 37, show that, as for the lead aerosol, the quotient of Pt_0 is lower for house C than for B and that the correlation is less. The extent of the penetration of the particulate aerosol is similar to that of the lead aerosol, varying between 60 and 70 per cent. Table 38 shows the percentage lead content of the particulate collected at the three houses. It is generally higher inside the property - except at house C, which suggests that a greater quantity of unleaded aerosol is infusing this house, since the level of particulate penetration is higher here than for house B.

4.4 BLOOD LEAD LEVELS

As mentioned above, Section 3.4, exposure to lead aerosols can lead to increasing blood lead levels. Occupants of these three houses were thus requested to give blood samples for analysis. From house A, two of the adult occupants gave venous samples on December 10th 1973 and these were analysed by Birmingham City Analyst. The values found averaged 24 μg per 100 ml of blood. One adult male from each house B and C gave blood samples and the levels found were 19 and 15 μg per 100 ml of blood respectively. These values are in the normal range, but should be checked periodically to investigate the possibility of change with time because WALDRON (1975) has reported finding an increase amongst residents near Gravelly Hill since the motorway opened.

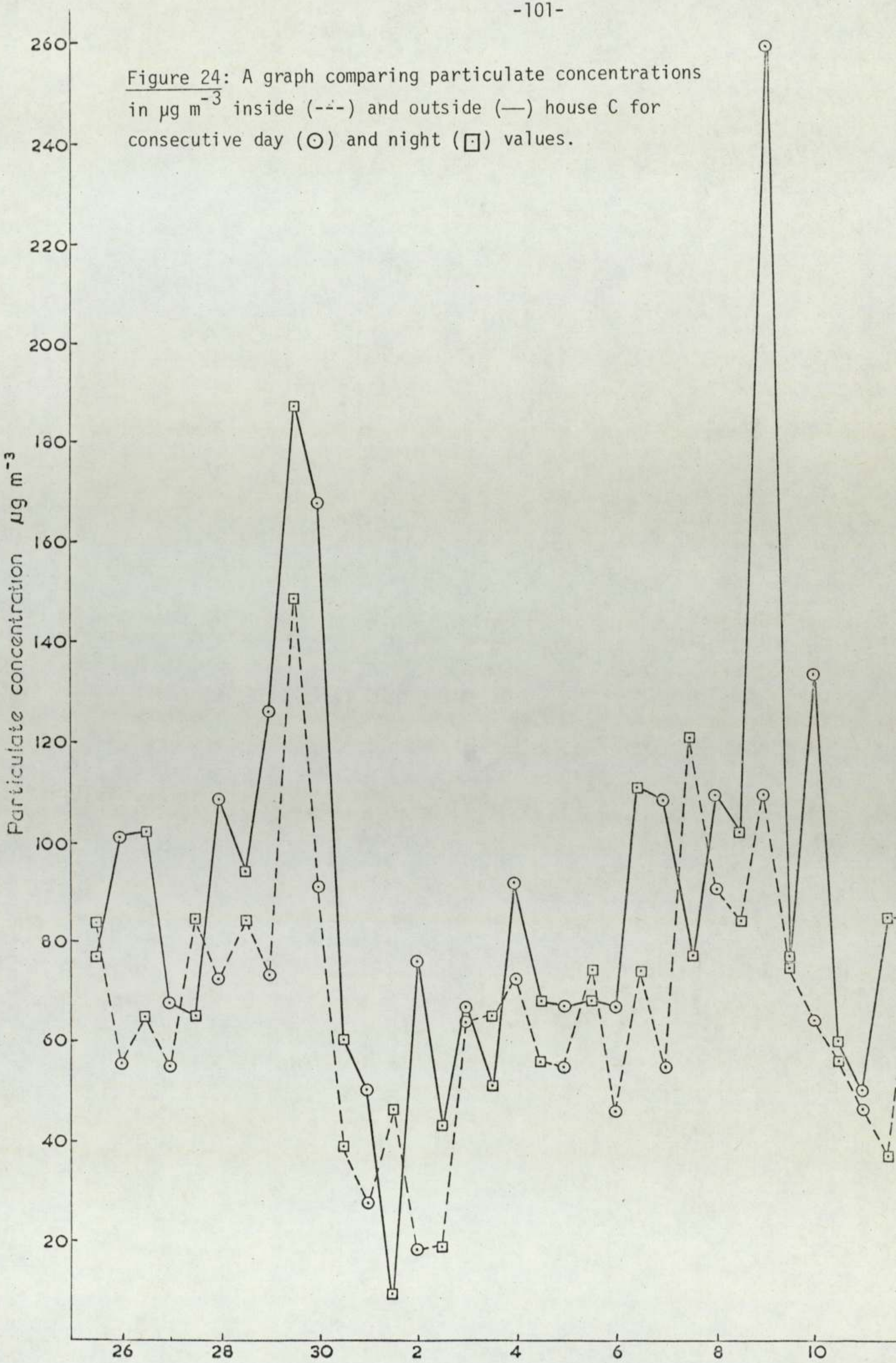


Figure 24: A graph comparing particulate concentrations in µg m^{-3} inside (---) and outside (—) house C for consecutive day (⊙) and night (⊡) values.

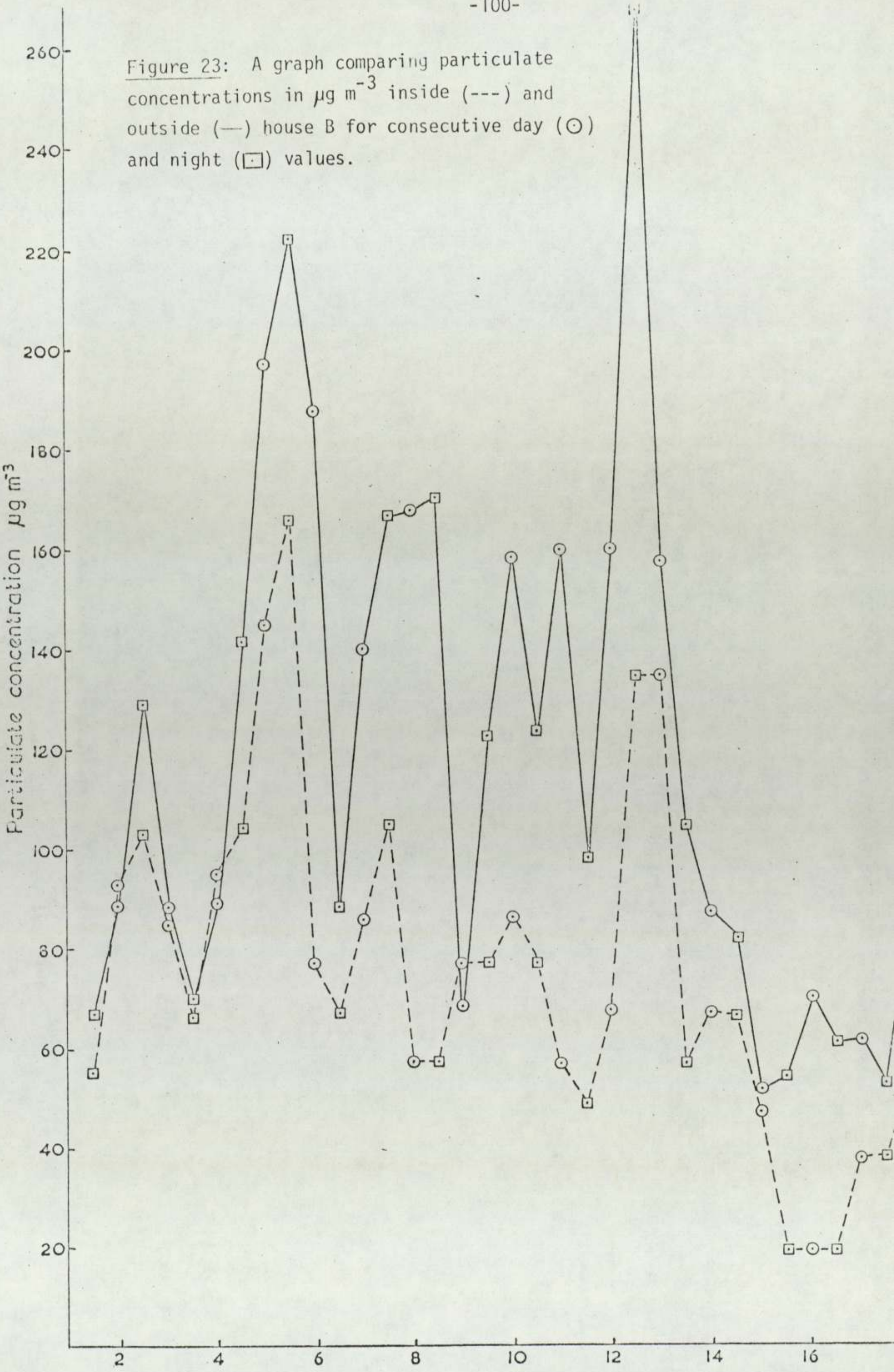


Figure 23: A graph comparing particulate concentrations in μg m^{-3} inside (---) and outside (—) house B for consecutive day (⊙) and night (⊡) values.

Table 36: Average particulate levels in $\mu g\ m^{-3}$ inside and outside the three houses.

HOUSE	A INSIDE	A OUTSIDE	B INSIDE	B OUTSIDE	C INSIDE	C OUTSIDE
DAY	126	175	76	114	63	103
NIGHT	79	119	74	114	70	81
COMBINED AVERAGE	104	147	75	114	66	92
PER CENT PENETRATION	71		66		72	

Table 37: Equations relating the particulate concentration measured inside houses B and C to the level outside.

HOUSE	EQUATION*	CORRELATION COEFFICIENT
B	$Pt_i = 18 + 0.50\ Pt_o$	0.71
C	$Pt_i = 30 + 0.41\ Pt_o$	0.67

* Pt_i represents the internal particulate concentration

Pt_o represents the external particulate concentration.

Table 38: Average percentage lead present in airborne particulate sampled inside and outside three houses.

Sample Time	HOUSE A		HOUSE B		HOUSE C		SITE D
	Inside	outside	Inside	outside	Inside	outside	
Day	0.85	0.65	0.99	0.98	0.85	0.82	1.25
Night	0.84	0.52	1.02	0.88	0.68	0.87	1.08
Combined Average	0.84	0.59	1.01	0.93	0.77	0.85	1.17

CHAPTER 5 - ELECTRON MICROSCOPE STUDY

5.1 INTRODUCTION

It has been noted earlier that an atmospheric aerosol is stable when the particulate matter that forms it has an average diameter of c.0.3 μm. Thirty per cent of lead particles emitted by motor vehicle exhausts, due to alkyllead combustion, have a size which falls within the stable range, HABIBI (1970). Many other trace elements are mobilised in such a way that they become associated with airborne particulate. These may derive from the burning of fossil fuels or metallic wastes. Notably, lead, chromium, zinc, copper vanadium and nickel are found at their highest concentrations in the smaller particles of an atmospheric aerosol, NATUSCH, WALLACE & EVANS (1974). To visualise such particles an electron microscope is required. Collection techniques for subsequent electron microscopy have been reviewed by BILLINGS & SILVERMAN (1962). Filters have been used and electron micrographs of particulate matter collected by filters have been published, LAWTHER, COMMINS, ELLISON & BILES (1972).

These authors found that the particulate collected consisted largely of aggregates of smaller particles whose diameter was less than 1 μm. Samples from Fleet Street and from a petrol engine showed small areas of material that was highly dense to the electron beam. The diameters of such areas was less than 0.015 μm. These, they said, it is reasonable to suggest contain part of the lead originally in petrol as alkyllead.

Some samples we collected at Gravelly Hill on millipore filters have been photographed under high magnification by an electron microscope. The elemental composition of areas under observation was analysed by x-ray fluorescence spectrometry.

5.2 EXPERIMENTAL.

Samples of the atmospheric particulate aerosol have been collected by millipore filters on a routine basis for some time. Some have been given over wholly for characterization by the electron microscope. Sections, 8 mm square, were cut from the filters and mounted on an aluminium stub. This was then coated with carbon using a carbon arc discharge for 6 s in an evacuated chamber at 10^{-5} torr. The coating allows impacting electrons in the microscope beam to be removed by making the surface conducting.

The prepared stub was placed in a Cambridge "stereo scan" scanning electron microscope, type 96113 mark 2A. This creates an incident electron beam which is accelerated through a potential difference of 20 keV. The photographs were of the resulting secondary, emitted electrons which enabled particles as small as 0.5 to 1.0 μm to be focussed. They were taken by a Zenith B SLR camera at f16, using a 40 s exposure time.

The microscope is coupled to a "Kevex" x-ray energy spectrometer 5000A, data processor 6000A and colour video display 5390. The x-ray detector in the instrument is a biased lithium doped silicon semiconductor, cooled in liquid nitrogen. It is housed in a compartment where it receives the radiation through a thin beryllium window. Beryllium absorbs x-rays with energies below 1 keV so this limits detection to elements having atomic numbers equal to or greater than that of sodium. The incident electron beam of the microscope generates characteristic x-rays which are intercepted by the detector positioned 2 cm above the surface of the sample. The signal produced by the detector is proportional to the energy of the incident x-rays. The spectrum that results from analysis of the signals is a series of approximately gaussian peaks projecting from the background. The positions and intensities of these peaks result from the elements in the 2 μm diameter beam of the microscope. The minimum detectable limit for the transitional elements is 0.9 per cent by weight of sample, WOLDSETH (1973).

5.3 RESULTS AND DISCUSSION

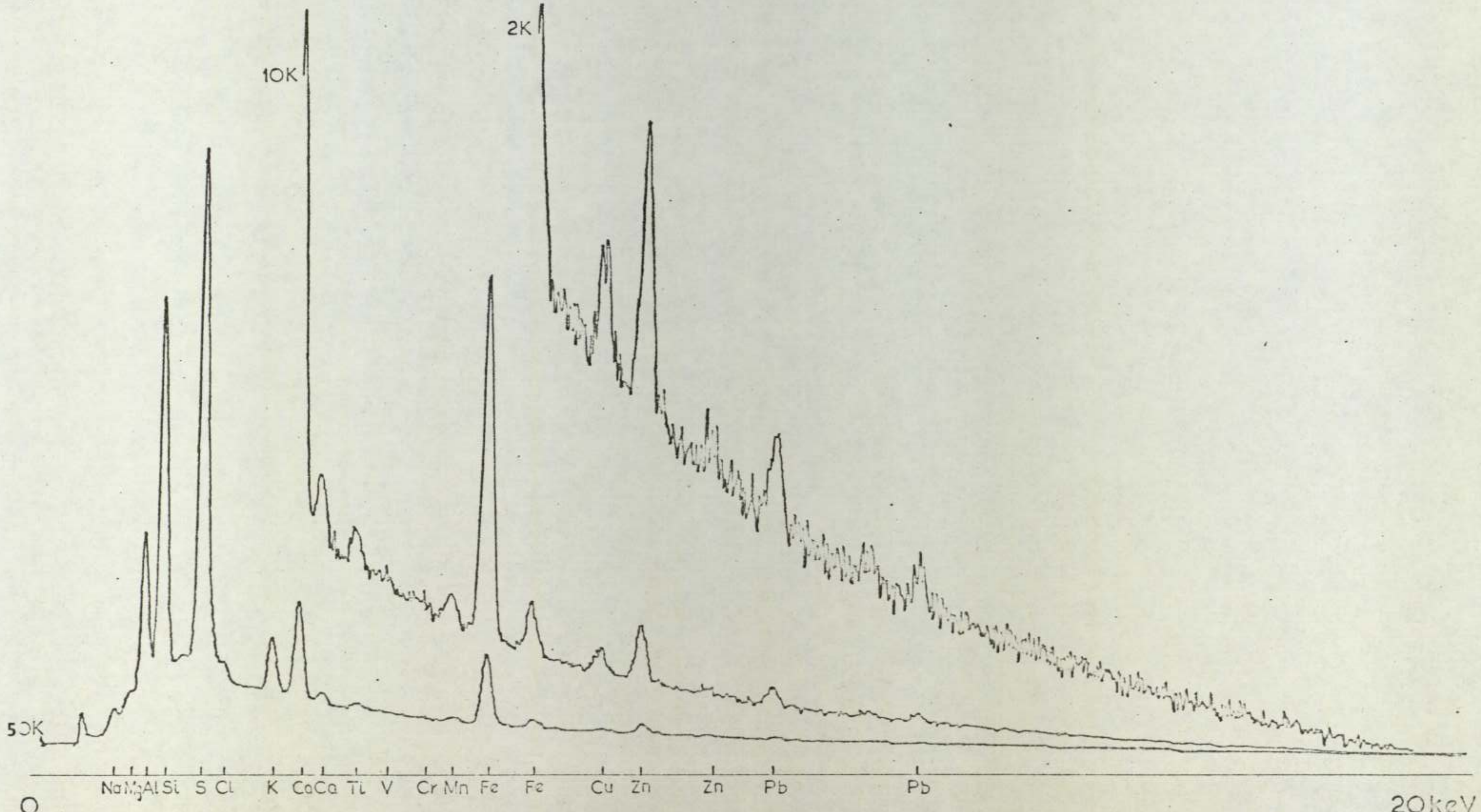
Qualitative analysis of a general area 50 μm square of a sample from Salford Circus is shown in Figure 25. The peaks have been allocated and the elements identified by comparison of their keV values as indicated on the Kevex display monitor with those given in the ASTM tables (1970). The spectrum shows the presence of elements typically found in surface soils such as sodium, magnesium, aluminium, silicon, potassium and calcium. The presence of titanium, vanadium, chromium, manganese, iron, copper, zinc and lead was also detected. These latter are known to be components of urban atmospheric aerosols in the U.K., PIERSON, CAWSE, SALMON & CAMBRAY (1973). The principle non-metallic element present appears to be sulphur although the lighter non-metals, except phosphorous, are undetectable.

Observing the general surface at higher magnifications shows the individual particles clearly. Some photographs and energy spectra are shown in Figures 26 - 44. Study of a number of particles of similar shape showed similarities in chemical composition, so the photographs that follow are generic types and are mentioned under their main constituent element:

(a) TITANIUM: Figure 26 shows an electron micrograph of a titanium bearing particle at a magnification of 5000. A photograph of the titanium K x-ray image at 4.51 keV for the same area as covered by Figure 26 is shown in Figure 27. The greater the density of the x-rays, the greater the degree of exposure of the film. Such an area - of increased exposure - occurs in the middle-centre of the Figure 27, which coincides with the size and shape of the particle. This confirms that the source of the titanium radiation, so apparent in the x-ray fluorescence spectrum (Figure 28), is the particle. The spectrum also indicates that chlorine and zinc are present.

(b) VANADIUM: Another particle is shown, under magnification of 5000, in Figure 29. The x-ray fluorescence spectrum of the same area is given in

Figure 25: Elemental composition of a typical aerosol as collected by a filter exposed at Salford Circus. The three traces derive from full scale expansions of 50,000. 10,000 and 2,000 counts respectively.



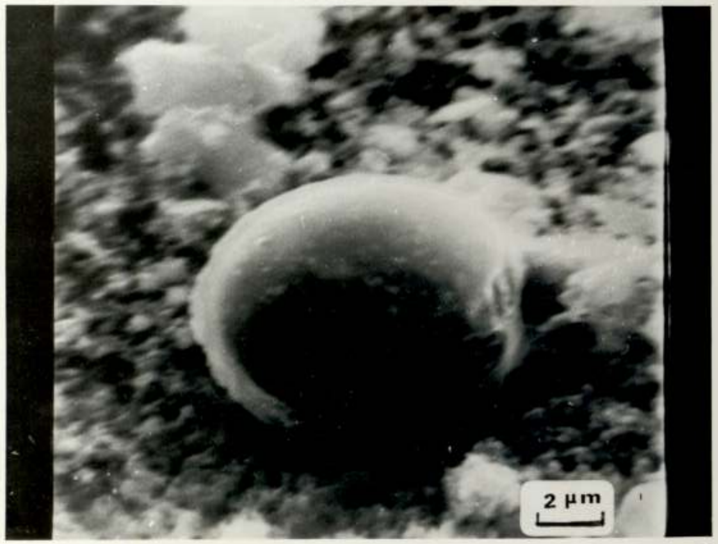


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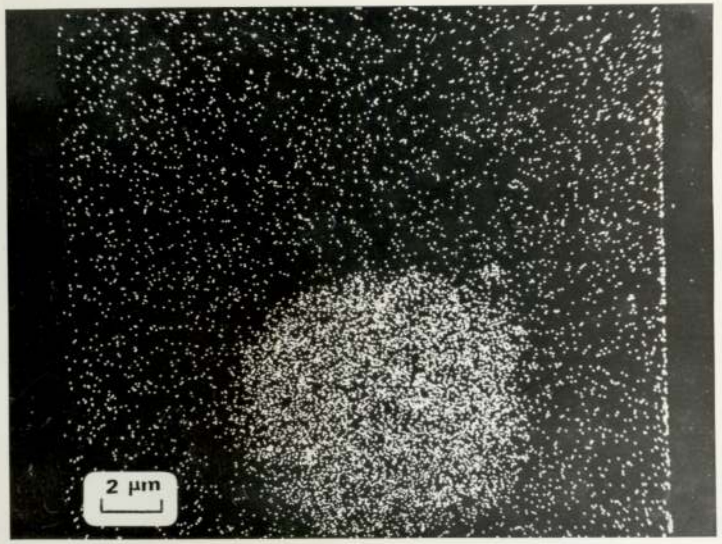
Figure 26: A photograph of a particle containing mostly titanium, at a magnification of 5000

Figure 27: The x-ray image due to titanium K_α radiation at 4.5 keV from the area shown in Figure 26.



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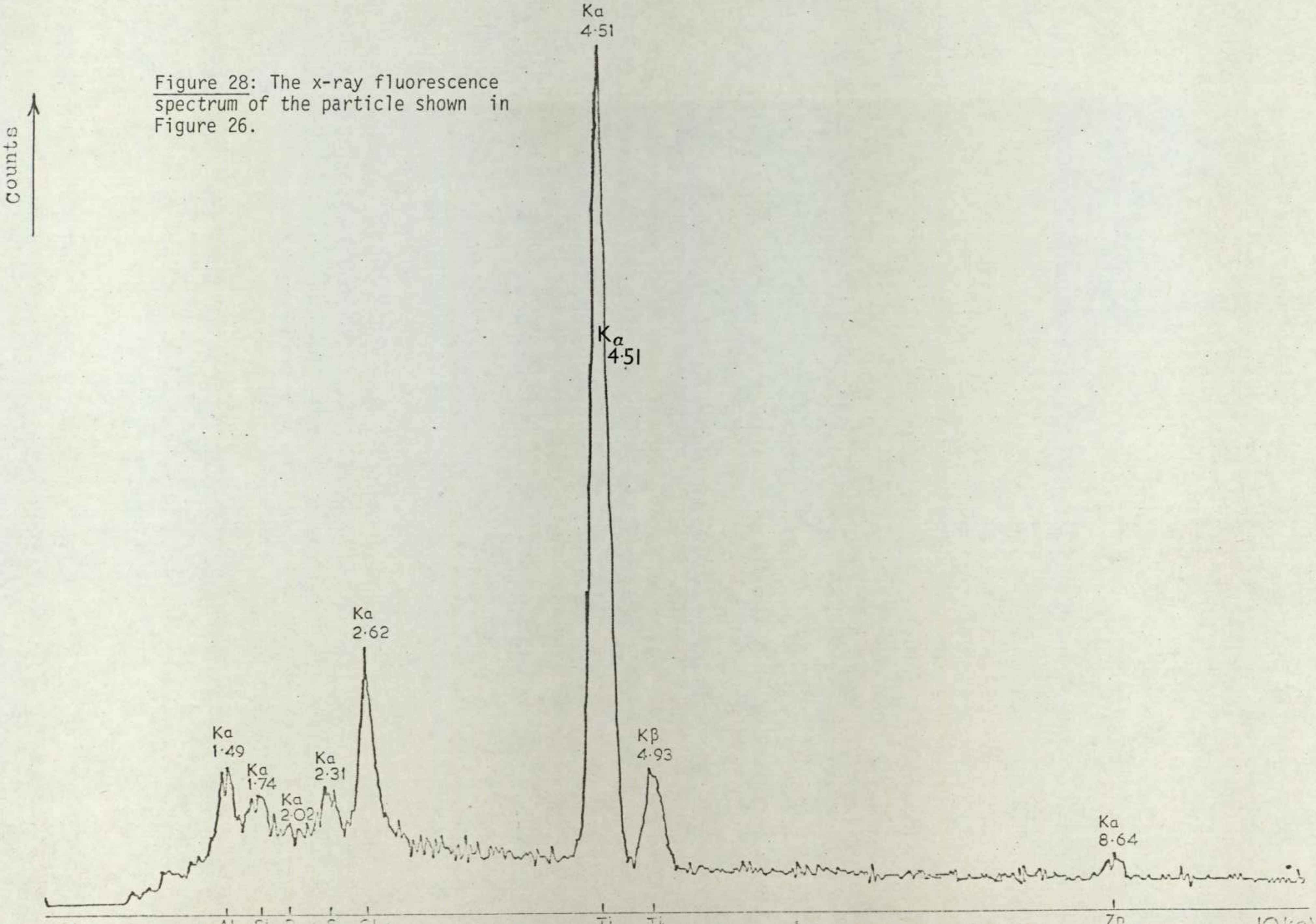

Figure 28: The x-ray fluorescence spectrum of the particle shown in Figure 26.

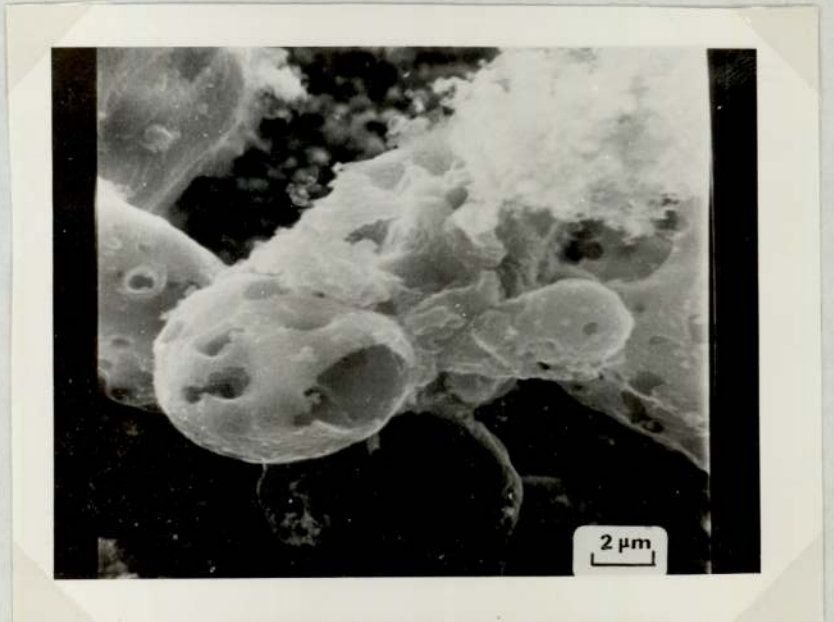

Figure 29: A picture of a particle consisting largely of vanadium, at a magnification of 5000 times.

Figure 30, and shows the presence of sulphur, vanadium and nickel. Discrete particles having a similar sponge-like appearance could be distinguished in other regions of the surface. These had similar spectra, vanadium,sulphur and nickel always being associated. Another typical example, at a magnification of 5000 times is shown in Figure 31. This particle has approximate dimensions of 12 μm by 15 μm, generally the length of long axis ranges from 9 μm to 20 μm. Since they were discovered on filters obtained from sampling 45 m from heavy traffic, their origin would appear to be diesel engine exhaust fumes.

(c) ZINC: Figure 32 is a photograph of an area at a magnification of 4000. The area is rich in aluminium, silicon, potassium, calcium, iron and zinc associated with sulphur, chlorine and presumably oxygen also. The x-ray fluorescence spectra of this area and of the particle in the centre right of the micrograph are shown in Figures 33 and 34. The latter reveals an enrichment of chlorine along with potassium and zinc compared to the general background.

The identification of a zinc rich particle was unusual. Generally, when the surface was scanned and analysed, zinc could only be located in an area through its association with chloride, but isolated zinc particles were difficult to find. It appears as though zinc is spread throughout "zinc rich areas" in the form of a film or in such a freely divided form (< 0.1 μm) that discrete zinc bearing particles could rarely be seen.

On a filter taken from the gantry, site F, another type of zinc rich particle was discovered in association with sulphur. An electron micrograph of a number of these zinc bearing particles is shown in Figure 35 at a magnification of 1,600. The corresponding energy spectrum, Figure 36, shows the presence of sodium, sulphur, potassium as well as zinc. The rectangular shaped particles range in size from 8 μm to 25 μm. Photographs of the K_α radiation from sulphur, at 2.31 keV, and from zinc, at 8.64 keV, at the same magnification as the electron micrograph (Figure 35) are given in Figures 37 and 38 respectively. These confirm that

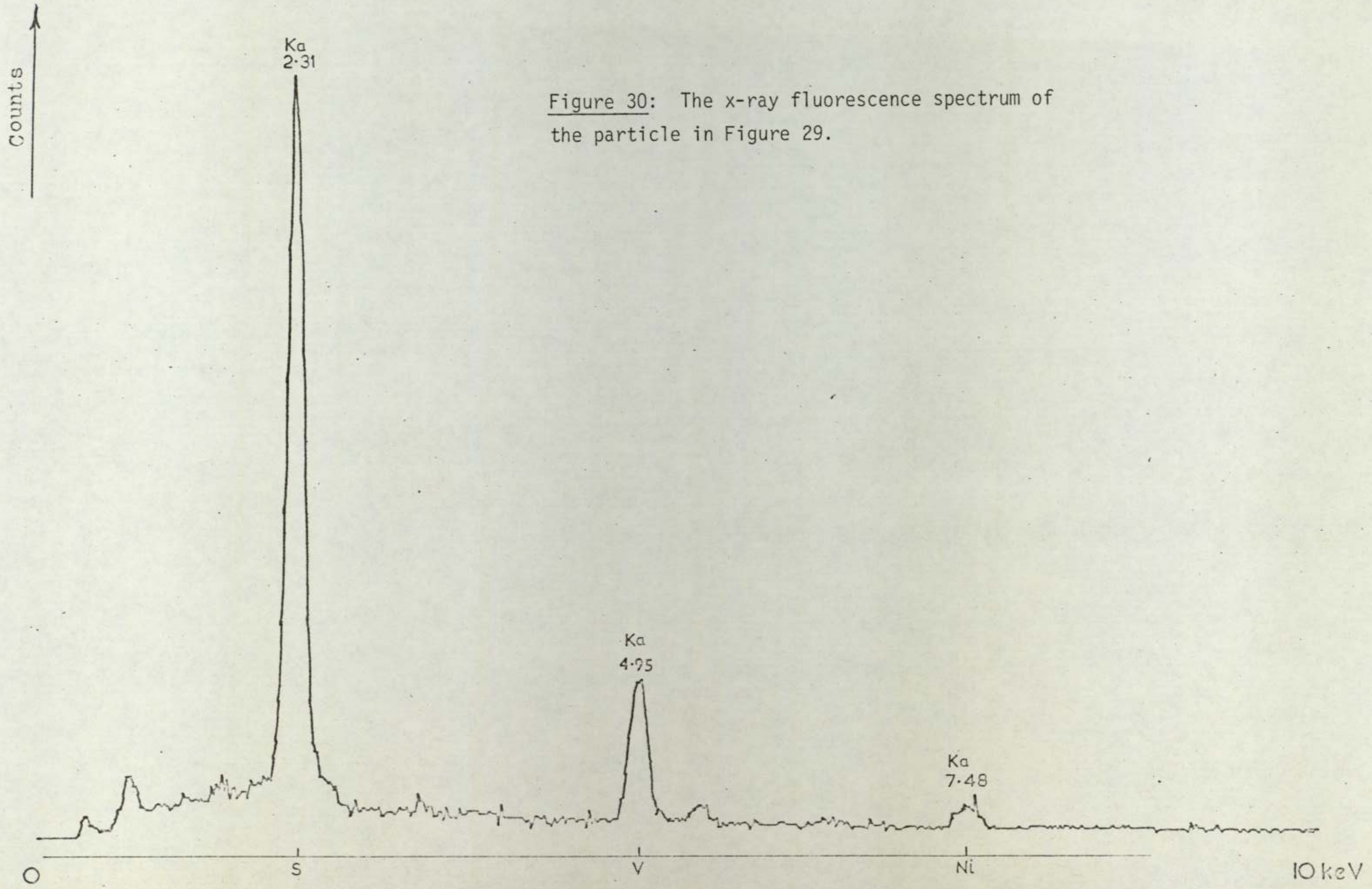

Figure 30: The x-ray fluorescence spectrum of the particle in Figure 29.

Figure 31: A vanadium bearing particle at a magnification of 5000 times showing the characteristic sponge-like appearance.

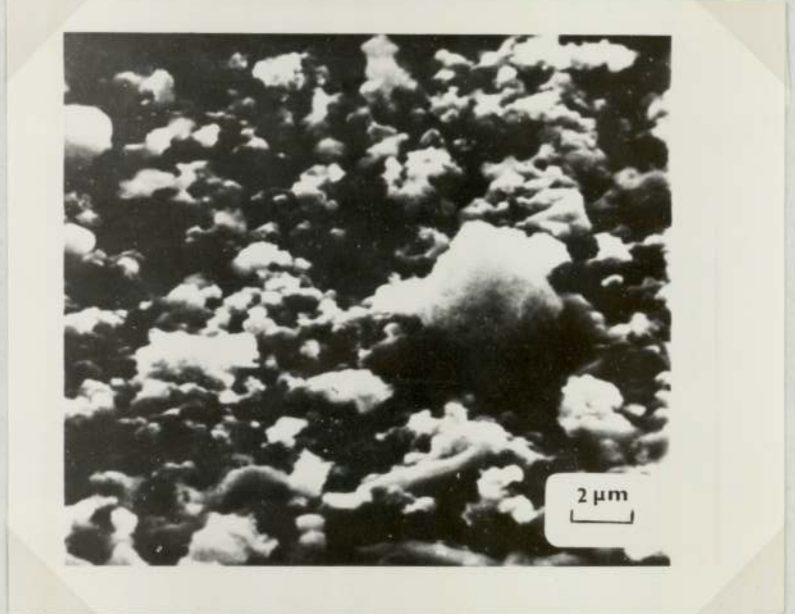

Figure 32: An electron micrograph of a general area of the surface at a magnification of 4000.

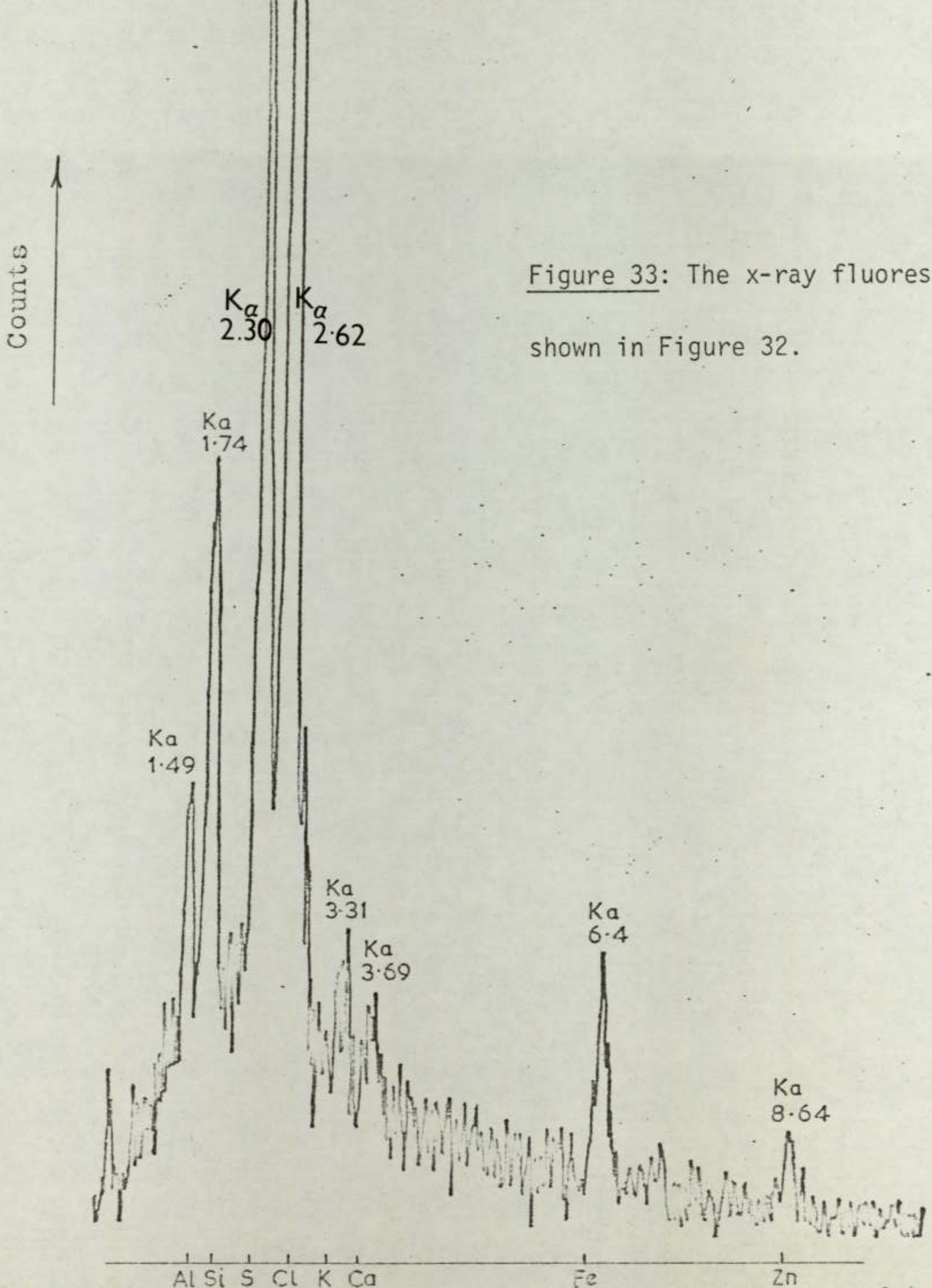

Figure 33: The x-ray fluorescence spectrum of the area shown in Figure 32.

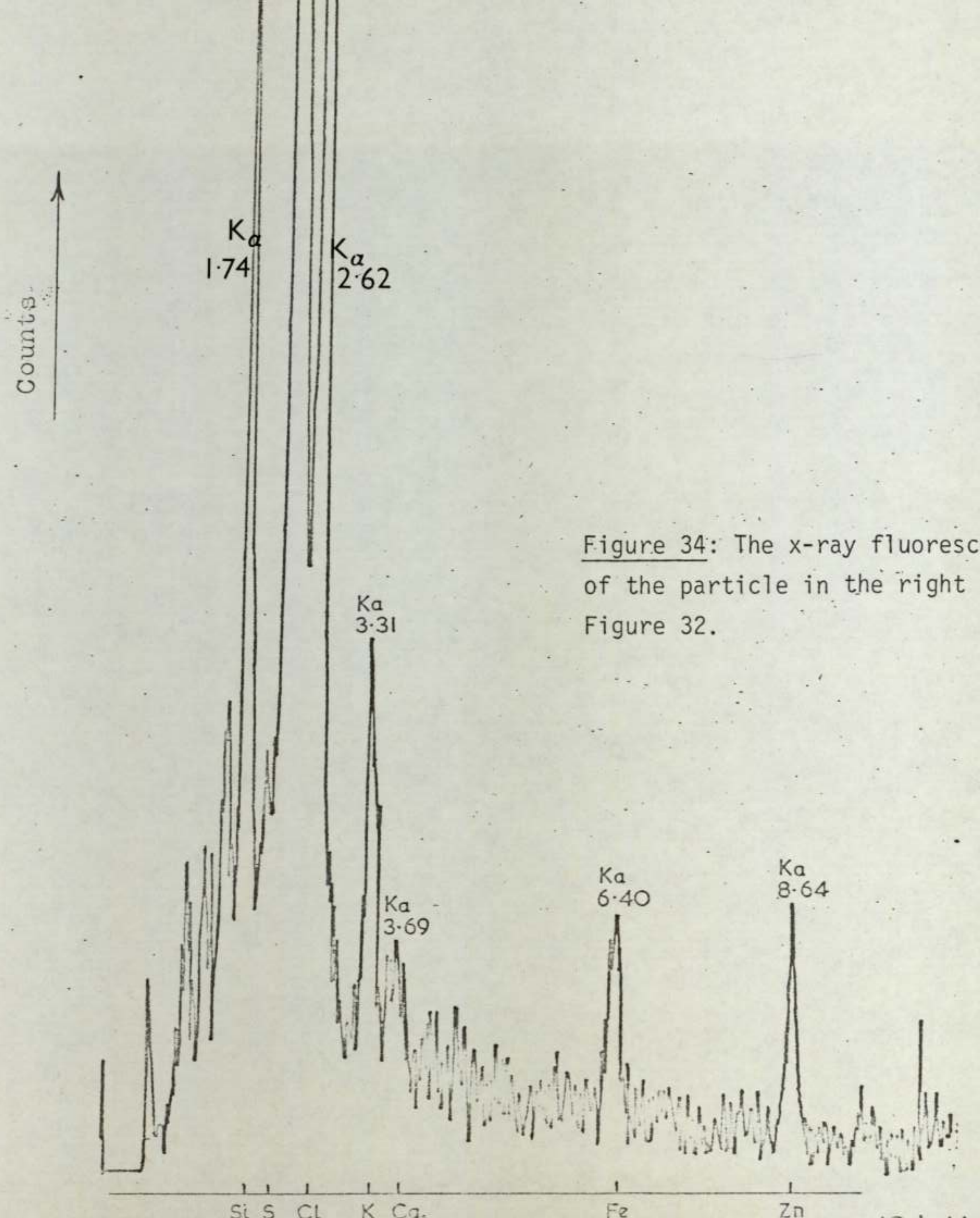

Figure 34: The x-ray fluorescence spectrum of the particle in the right centre of Figure 32.

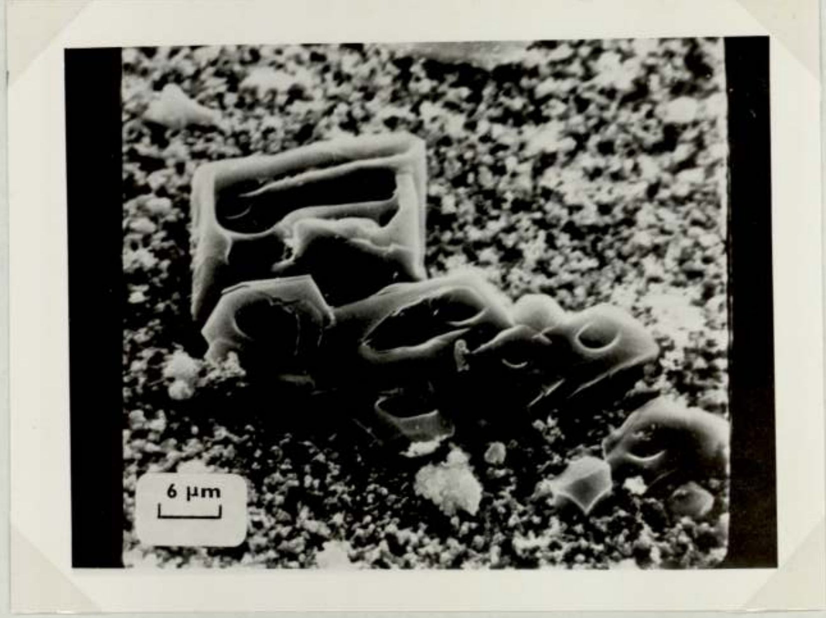

Figure 35: A zinc bearing particle found on a filter taken from site F, at a magnification of 1,600.

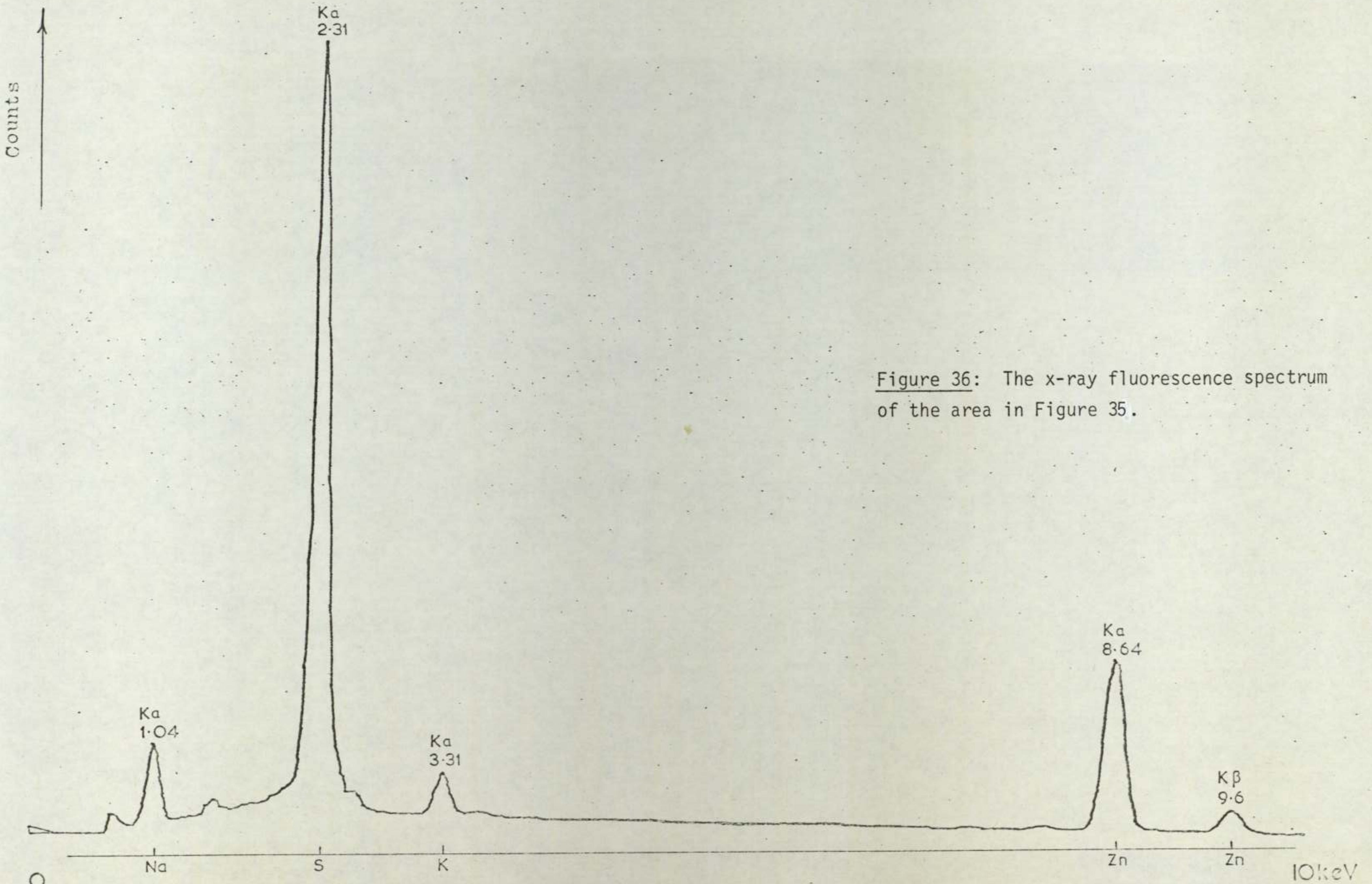

Figure 36: The x-ray fluorescence spectrum of the area in Figure 35.

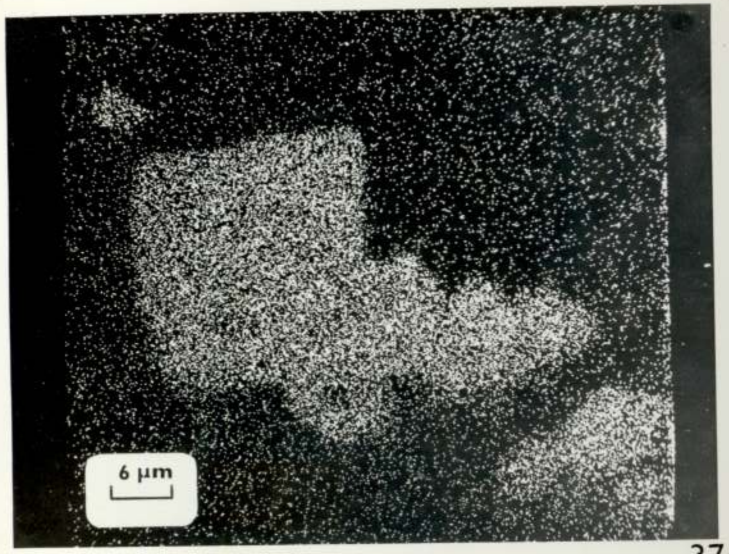

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Figure 37: The x-ray image due to sulphur K_{α} radiation at 2.31 keV from the area shown in Figure 35.

Figure 38: The x-ray image due to zinc K_{α} radiation at 8.64 keV from the area shown in Figure 35.

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the source of the radiation from these elements is located at the sites of the rectangular shaped particles.

(d) LEAD: Part of the surface of a filter from a monitoring site close to the kerb is shown in Figure 39 at a magnification of 5000. The lead $M_{\alpha\beta}$ x-ray image of the area is shown in Figure 40. This identifies the lead rich areas as being either side of the central "fluffy" region. The complete x-ray spectrum of the area is given in Figure 41. The approximate diameter of the lead rich particles is about 4 μm. Thus they belong to the size range associated with turbulent deposition.

In contrast to these lead particles, which were found close to the road surface, Figure 42 shows another particle at a magnification of 10000, identified on a filter from inside a house alongside the motorway. The particle is lead rich as shown by the x-ray image of the lead M radiation 2.39 keV, Figure 43. This particle is most probably derived from a petrol engine propelled vehicle since the x-ray fluorescence spectrum of the particle, Figure 44, indicates that chloride and bromide are still present. Also the particle is fairly young since chlorine and bromide are lost from the lead bromo-chloride emitted from motor car exhausts, PUESCHEL, VAN VALIN & PARUNGO (1974).

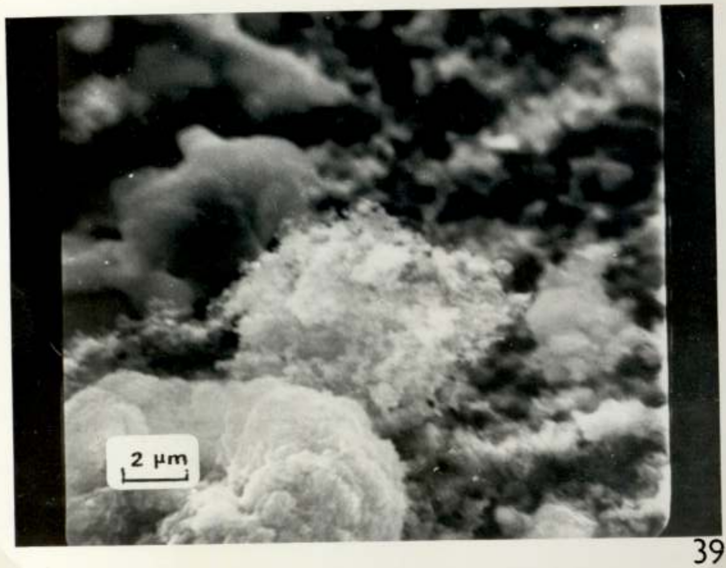

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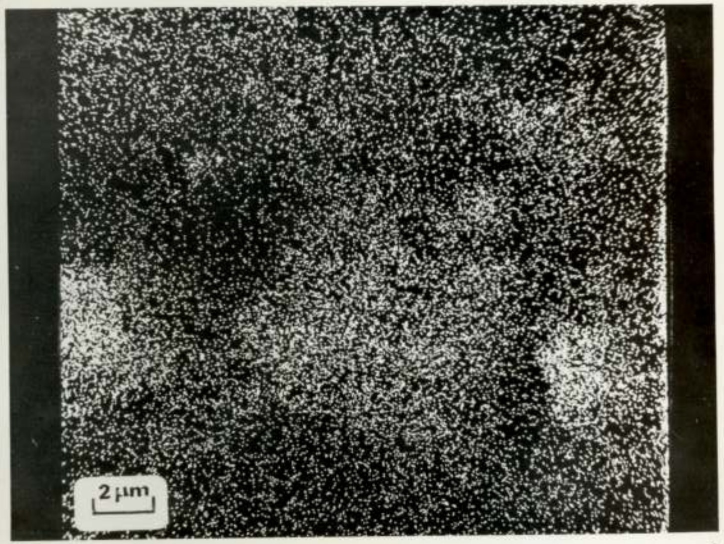
Figure 39: An electron micrograph of the surface of a filter taken from a kerbside monitoring site, at a magnification of 5000.

Figure 40: The $M_{\alpha\beta}$ x-ray image of the area shown in Figure 39.



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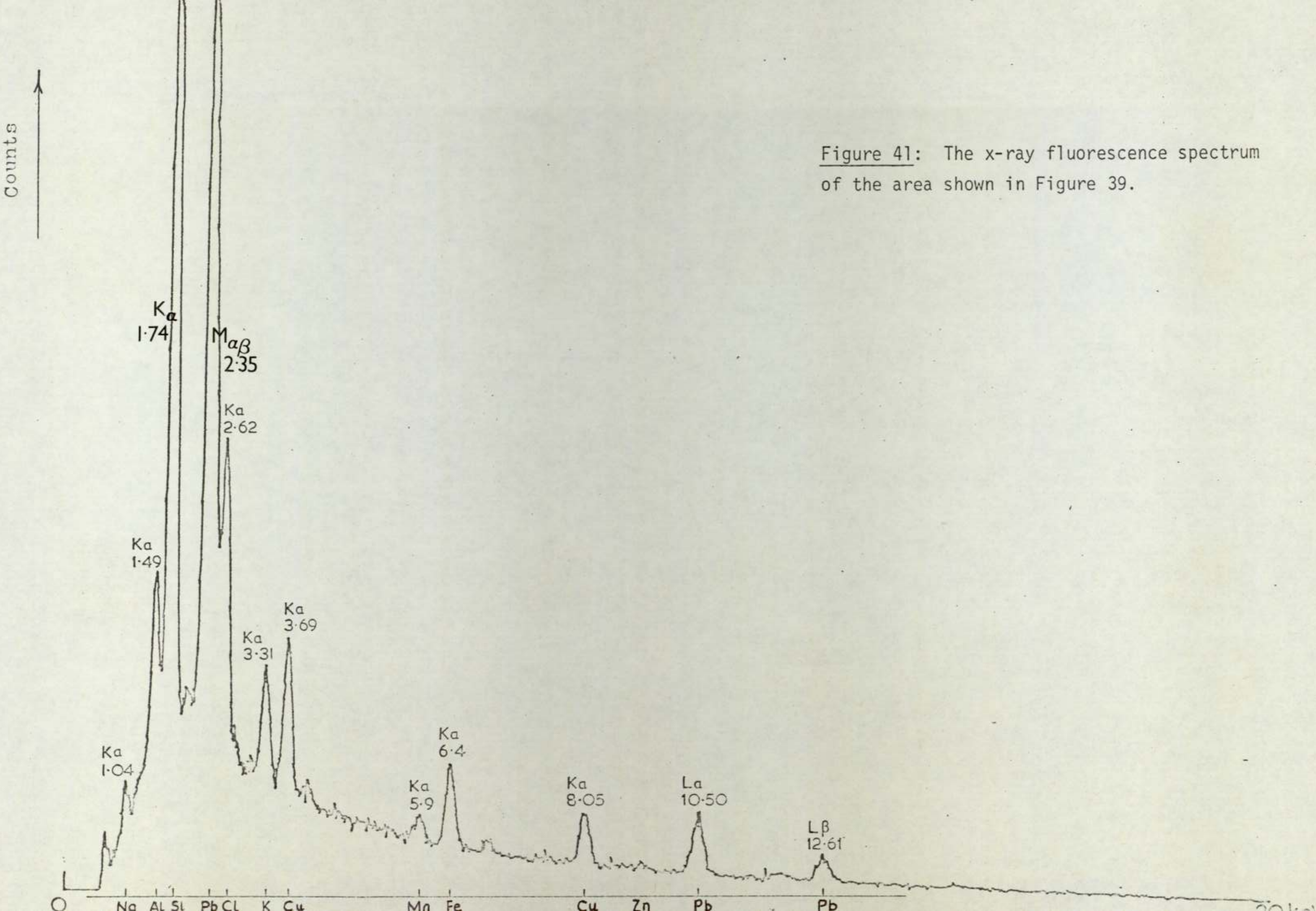

Figure 41: The x-ray fluorescence spectrum of the area shown in Figure 39.

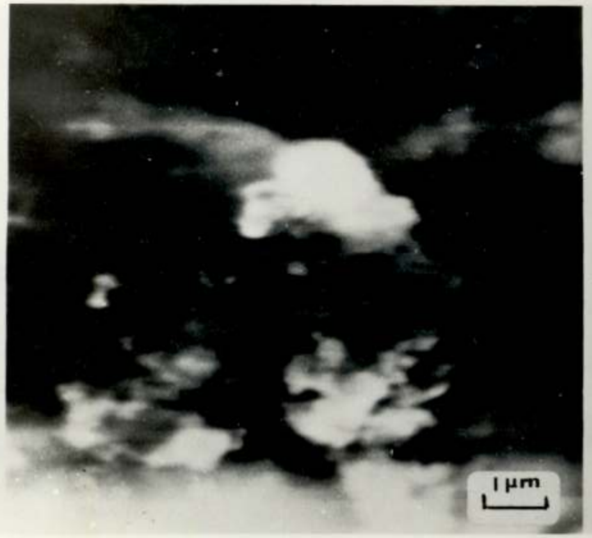

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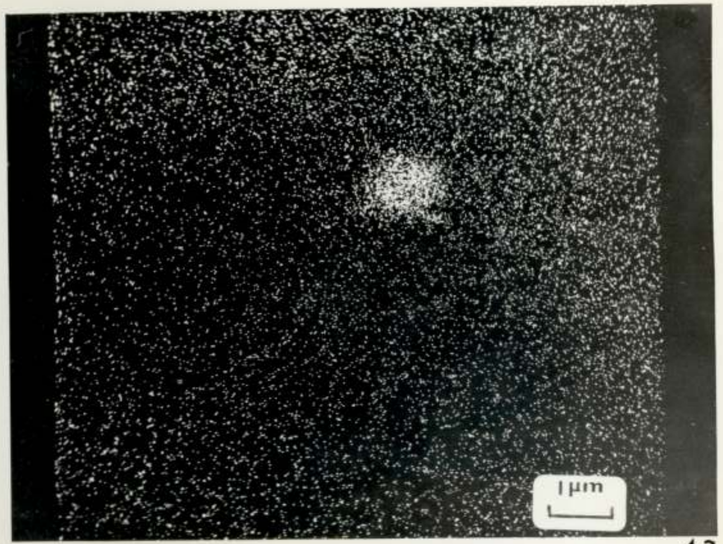
Figure 42: An electron micrograph of the surface of a filter taken from a house alongside the motorway at a magnification of 10,000.

Figure 43: The $M_{\alpha\beta}$ x-ray image of the area shown in Figure 42.



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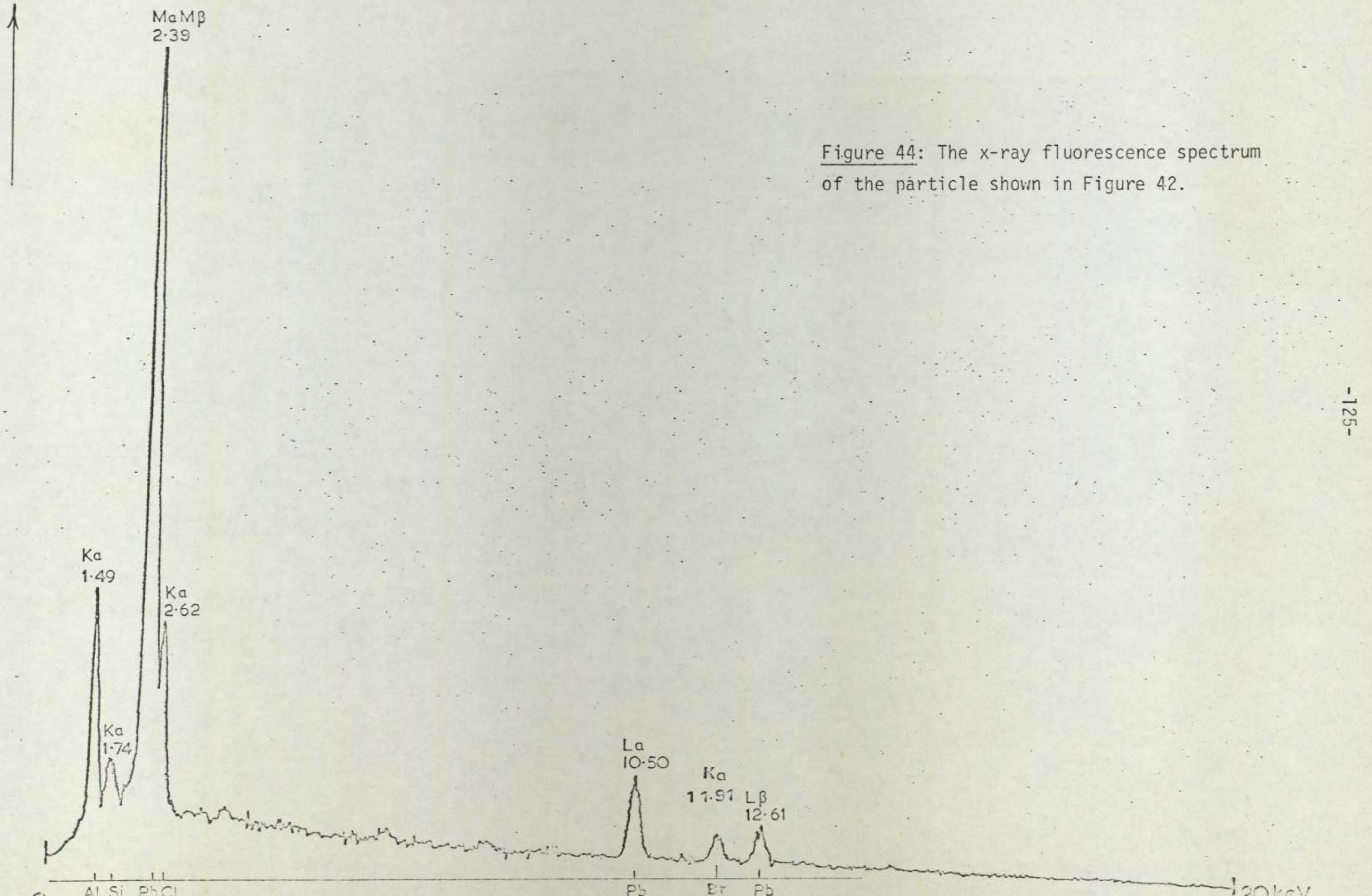

Figure 44: The x-ray fluorescence spectrum of the particle shown in Figure 42.

CHAPTER 6 - CONCLUSION

The monitoring of atmospheric particulate lead concentrations in the Gravelly Hill area of Birmingham has provided some useful information and layed the ground-work for further investigation. We have measured the lead levels for just over two years and found that the opening of a motorway complex in the vicinity resulted in a dilution of the local lead aerosol whilst causing an increase in the "background" lead concentration. It has been suggested that exposure to atmospheric lead levels greater than 2 μg m^{-3} for more than three months may be injurious. By this criterion the residents of the Gravelly Hill area are only likely to be at risk during the winter months, October to April, and then only if they live close to the motorway. The concentration exceeded 2 μg m^{-3} for three months at site D during one period, which was from February to April 1973. The blood lead levels of some residents was determined three times during the period of this study and, whilst most are still "normal", they do not yet appear to be in equilibrium. Thus the public health aspect requires continued monitoring of atmospheric as well as blood lead levels.

The sampling schedule on which we worked showed up differences between consecutive twelve hour average concentrations and, by accumulation,the differences from month to month. The weather and traffic were also monitored and the relationship between these and the lead level investigated. Statistical analysis suggests wind speed, temperature and traffic are the major influences causing the periodic fluctuations. Although the local traffic was directly measured, the weather data was obtained from instruments five miles distant. To refine the model these should be quantified as close to the monitors as possible. Relative humidity was expected to exert an influence on the fluctuation in lead level, but was found not to do so possibly due to the local environment. Also since this factor plays a part in the prevention of agglomeration, it affects particle size and thus the ability of particulate to penetrate houses and into the respiratory tract. However, a predictive equation, based on the three quantities mentioned above, was produced for the lead concentration at a reference site - the centre of a circular source, 90 m in diameter.

Five other sites, within 1 km of the reference site, were monitored and the dependence of the lead concentrations found at these on those at the reference site determined. Thus from a knowledge of three fairly accessible parameters the average atmospheric lead level for a particular period may be assessed at a number of points around the junction. Two more, remote sites were studied and, allowing for a fourth variable - wind direction - a relationship was found that had a good correlation between levels monitored there and those at reference site. The effect of wind direction deserves more study since the terrain varies with direction, from the reference site, and this can affect aerosol life time. Also the wind is more prevalent from certain directions than from others, so that such "down-wind" areas may receive more atmospheric lead and contain more deposited lead.

Lead levels were monitored both inside and outside three houses. The results found reaffirm the findings of other workers as to the degree of penetration and suggest the ability of double glazing to afford some protection from the external leaded aerosol. This protection may be valuable if the atmospheric lead concentration rises to a level considered unhealthy. Thus the degree of protection should be substantiated using properties further from the motorway.

The aerosol received closer scrutiny by electron microscopy linked with an x-ray energy spectrograph, which enabled the size, shape and composition of particles to be recorded. The general surface view shows the vast number of very small particles collected though individual particles could be picked out fairly readily. Thus this technique could be applied to very small samples collected for sizing by say an impactor, which would be useful in confirming data on the size distribution of aerosols inside buildings compared with the external one. Also, by taking more frequent samples, the degree of aerosol retention from day to night be assessed. The chemical composition of leaded particulate is known to vary with age of the aerosol and so the halide to lead ratio may be indicative of age and thus retention.

To meet future emission levels for exhaust gases and lead, it is probable that some system for the removal of lead from the exhaust gas or from petrol will be required. The former, effected by means of an alumina coated wire mesh, is probably an expensive method, which would require frequent scrutiny of exhausts. Removal of lead from petrol is possible if the octane rating is maintained by means of further refining or the addition of aromatic compounds. The latter method may increase the emission of hazardous polynuclear aromatic hydrocarbons, phenols and aldehyles, which also increase the photochemical reactivity of the exhaust gas. Increased refining of fuel would increase the cost of petrol and may cause increased fuel consumption. However, any increase in emissions of hydrocarbons, nitrogen oxides and carbon monoxide may be compensated for by recycling the exhaust gas, after burning and catalytic reduction.

Whilst tetraalkylleads are added to petrol it is advisable to monitor the resultant dispersion of lead. But it is also in the public interest to develop methods of reducing the other noxious products of combustion so that removal of lead may create cleaner exhausts.

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