'Children's Personal Exposure to Airborne Particulate Matter.'

A Thesis submitted to Middlesex University in partial fulfilment of the requirements for the degree of Doctor of Philosophy.

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To my family

Thank you

ABSTRACT

Personal exposure to particulate matter (PM_{10} and $PM_{2.5}$) for ten children aged 9 - 11 years was measured between January and September 1997 in the London Borough of Barnet. Personal, home, garden and classroom microenvironmental monitoring was successfully completed for all ten children. Each child was monitored for five consecutive days during winter, spring and summer, with the exception of one child who did not complete the summer session. All children completed daily time activity diaries to provide information on any activities that could potentially influence their exposure patterns. Each evening parents completed a household activity questionnaire providing information on all particle generating activities such as cooking and cleaning. Personal Environmental Monitors were used for the personal sampling and Harvard Impactors for the microenvironmental sampling.

The children's mean personal exposure concentrations for PM_{10} during winter, spring and summer were 69, 69 and 32 $\mu g/m^3$ respectively and for $PM_{2.5}$ 21, 24 and 15 $\mu g/m^3$ respectively. The strongest and most consistent associations were found between the personal and indoor exposure concentrations. The most significant correlations were observed between personal and home PM_{10} with a median $r_s = 0.66$. Classroom concentrations were the highest of all the sampled environments which could be attributed to the number of children present and the resuspension of particles. Ambient contributions of $PM_{2.5}$ to PM_{10} during the day were estimated to be 56%, which is comparable to other UK research. Indoor / outdoor concentrations were influenced by heating in the homes, however no significant specific particle generating activities in the home were found during the day. At night, home concentrations of $PM_{2.5}$ appeared to be influenced by the presence of smokers.

To determine potential sources of particulate matter, analysis of a sub sample of filters was undertaken using Scanning Electron Microscopy. Within the home particle composition was influenced by human activities predominantly; resuspended soil dust, skin flakes and fibres. The outdoor particles were predominantly biological in origin; pollen and insect debris. The composition of the personal exposure filters was a mixture of both environments and was dependant upon how much time each child spent in each of these environments. Smooth globular particles c.2μm in diameter were found on all filters and could be combustion related, possible vehicle derived or from cooking activities.

Measured data, along with the reviewed literature, provides some insight into the source apportionment of particulate matter. Analysis of questionnaire and time activity diaries also provides information on individual children's exposure patterns. Some estimation of potential health outcomes is discussed.

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1 Introduction

The Urban Pollution Research Centre (UPRC) at Middlesex University has investigated **'Children's Personal Exposure to Airborne Particulate Matter'.** Funded by the Engineering and Physical Science Research Council (EPSRC) CASE award in collaboration with Barnet Health Authority.

The thesis will report on the preliminary aims and objectives stated in the original registration document. It will then review published research to identify the sources of particulate matter along with associated health effects. The reviewed literature has been used to determine study design and methodology employed. Chapter 4 describes the methodology and all Standard Operating Procedures (SOP) have been included as appendices. There are two chapters that report the results of the data. Chapter 5 is a broad overview of the descriptive statistics. Correlations of all the data are reviewed along with potential sources of exposures and the results of this are used in Chapter 6 to investigate any significant relationships. Data from the questionnaires, time activity diaries and physical analysis of the collected particulates are investigated to explain the results seen in Chapter 5. Chapter 7 draws conclusions from all chapters and makes recommendations for further research.

1.1 Overview of Chapter 1

The aims and objectives of this research and, how the M.Phil and Ph.D. objectives have been met will be addressed in this chapter. Within the wider framework of the Ph.D., the following will also be reviewed:

- The programme of associated studies.
- Conferences and courses attended.
- Other research based work undertaken.

1.2 Registration Document Research Aims and Objectives

1) To establish a methodology for, and subsequently perform, a personal exposure monitoring programme of PM_{10} for a sample of 9-11 year old schoolchildren from within the area of Barnet Health Authority in North London.

2) To undertake an in-depth cross-sectoral study within the Barnet area to identify the prevalence of key respiratory diseases/symptoms in selected GP practices. This information will primarily be used for the selection of children.

3) Questionnaires and time activity diaries will be used to identify exposure patterns and the influence that confounding factors have upon the study.

4) To identify the source and the composition of the collected particulates by undertaking a physical and chemical analysis of the different size distributions.

5) To investigate children's exposure to PM_{10}, and then estimate the dose, uptake and likely effect upon children's health in order to assist in future health planning strategies.

6) To assess how the children's actual personal exposure compares to the national air quality standards, personal monitoring of children will be conducted throughout their daily activities both within the built environment and outside. From this data the impact of PM_{10} within the built environment will be assessed.

1.3 Discussion of Objectives

The preliminary objectives as stated in the registration document provided a starting point for this research. Not all objectives have been completed as different research interests have developed throughout the course of the Ph.D.

The initiative for the research originated from recommendations made by the Medical Research Council (MRC), (1994). This recognised the need to quantify the exposure of individuals to PM_{10} either through chamber studies or through personal exposure monitoring. This study has taken one aspect of this recommendation and has quantified the personal exposure of a sample of schoolchildren between the ages of 9 – 11 years to particulate matter.

The personal exposure of children to air pollution is of particular concern as they are considered to be a susceptible group of the general population with regard to air pollution related health problems (Scarlett *et al.*, 1996). Section 3.3.2. provides detail on the studies of children that have investigated air pollution health effects.

The advantages of monitoring children are that their exposure will generally be from within the London Borough of Barnet as they both live there and attend local schools. This

makes identification of sources of exposure to particulate matter easier to determine. Confounding issues of exposure to air pollution as a result of commuting through different areas of London, Environmental Tobacco Smoke (ETS) from smoking, and exposure in other social environments are also likely to be reduced when sampling children.

A proposed objective of the M.Phil. programme was to establish an acceptable methodology for the personal exposure monitoring of PM_{10}. Equipment available in the UK for assessing personal exposure is predominantly for use within the occupational exposure field of research. Particulates are usually collected over an 8-hour period within specific environments where the nature of the particulate matter is known. The size fractions that the occupational samplers collect reflect the inhalable or respirable fractions, Section 3.7 (Kenny, 1996a & 1996b). It was decided that such samplers would be unsuitable for using in this study due to the children's personal exposure resulting from a number of different environments with unknown particulate size fractions.

The Particle Total Exposure Assessment Methodology (PTEAM) study measured personal exposure to particulate matter over twelve hour periods both during the day and night (Spengler *et al.,* 1990, Thomas *et al.*, 1993). This PTEAM study and another study that was carried out in Boston (Rojas-Bracho, 1998) developed a monitoring protocol to determine personal exposure patterns of individuals. It was decided that the same equipment and methodology would be employed for this research. Total personal exposure to PM_{10} and $PM_{2.5}$ of schoolchildren throughout their daytime activities would be conducted. This would provide an indication of any seasonal variation in their exposure and the differences in each child's exposure with regard to their surroundings.

UK legislation now exists for particulate matter (PM_{10}) which will be discussed in Section 2.4. There is also legislation in the US for $PM_{2.5}$. This has been established as a result of the health concerns that have become evident from recently published community health studies (U.S.E.P.A., 1997). It was felt therefore that both size fractions should be included for measurement within this study of children's personal exposure.

There is a need to identify a relationship in the UK between children's personal exposure and their activities. From the results obtained it should be possible to show that the likely exposure of children to particulate matter is underestimated when using ambient measurements. Healthy children were included in the study, trying to identify a causal link between exposure to particulate matter and health impacts in this study would be unlikely to provide enough data to be epidemiologically relevant. Dr. Verne (Pers. Comm., 1996) an epidemiologist from Barnet Health Authority indicated that the number of confounding

factors and sample size would limit the viability of identifying health related problems resulting from exposure to particulate matter. It was then decided that healthy children would be included in the exposure analysis. The issues of confounding effects and sample selection have been discussed in Chapter 4.

Physical analysis of the collected particulates was undertaken to identify likely sources. As the particulate matter collection procedure was a cumulative process, the precise exposure patterns have not been identified. The physical analysis provides information about the different sources within the outdoor and indoor environments through the identification of specific particulates. Some chemical analyses were attempted, trace metal and Nicotine detection, they was unsuccessful and as such have not been included in the thesis.

1.4 Conferences, Seminars and Workshops

A variety of relevant conferences, seminars and workshops have been attended on the current research being carried out in this area. Conferences and Seminars were attended to promote the research and to liaise with individuals involved in the same field. The workshops attended were relevant to the understanding of the methodology, including training on a number of methods not eventually used in the final methodology.

- Paper presented at the **'Urban Air Quality - Measurement, Modelling and Management 2nd International Conference'**, Madrid, 3 - 5 March 1999.

- Paper presented at 'Tenth Conference of the International Society for Environmental Epidemiology & Eighth Conference of the International Society of Exposure Analysis', (ISEE & ISEA), Boston, USA, 15 – 18 August 1998.

- Paper presented at **'6th International Highway and Urban Pollution Symposium'**, ISPRA, Italy, 18 - 21 May, 1998.

- Poster presented at **'International Symposium on Health Effects of Particulate Matter in Ambient Air'**, Prague, 23 - 25 April 1997.

Paper presentation at the MUCORT '96 conference, Middlesex University. Also organised the conference as a Committee Member.

Attendance of the Faculty of Technology Inter faculty Research Seminar, June '97.

Attendance at the London Air Pollution Forum Meetings and Seminars.

University College London Workshop on Particulate Pollution, January 1996.

Aerosol Society Meetings, Birmingham 1996 & 1997.

Health and Safety Laboratory Workshop on Particulates, October '96.

Three week training sabbatical to Harvard University School of Public Health, August '96.

Worked in collaboration with London Borough of Barnet for Traffic data and central site monitoring.

Training on, and use of, equipment from Bartlett College of Architecture for Air Exchange Measurements and Building Design.

Attendance at the Standing Conference on Air Pollution, London, December '96.

Collaborative research with Atomic Energy Authority (AEA) Technology on a Chamber Study Sampler Comparison, July '97.

Attendance at School of Environmental Science Research Seminars.

Middlesex University postgraduate centre research workshops.

In contact with Imperial College, London, Mark Nieuwenhuijsen about EXPOLIS study in Oxford and data analysis procedures.

Attendance at the EPSRC Graduate School, Hartley Hall, Manchester, 4-9 September 1997.

Introduction to Urban Air Pollution, Short Course at UCL, January 1998

Attendance of the School of ES & E level 4 Research Methods Module

Member of the International Society of Exposure Assessment 1997 – 2000.

1.5 Publications

Copies of these have been included as Appendix 1.

Children's personal exposure to particulate matter in a UK urban environment. Preliminary results. A.J. Wheeler, R. Beaumont, I. Williams & R.S. Hamilton. **Epidemiology,** July 1998, Vol 9 No. 4 Pg S115.

Monitoring Children's Personal Exposure to Airborne Particulate Matter in London, UK - Method Development and Study Design. A.J. Wheeler, R. Beaumont, R.S. Hamilton & S. Farrow. **Science of the Total Environment,** 1 September 1999, Vol 235, Nos. 1-3, Pg 397-398.

Characterisation of Particulates Sampled During a Study of Children's Personal Exposure to Airborne Particulate Matter in a UK Urban Environment. A.J. Wheeler, I. Williams, R. Beaumont & R.S. Hamilton. **Environmental Monitoring and Assessment,** November 2000, Vol 65(1/2): 69-77.

2 Review of Particulate Matter

2.1 Introduction

This chapter reviews the literature relating to particulate matter, discussing the sources and size fractions of interest. The legislation for particulate matter in the UK and elsewhere is stated for the comparison of ambient outdoor concentrations with the personal exposure concentrations, as stated in Objective 6. An overview of the external sources of particulate matter found within the study area is provided to aid in the identification of potential exposure sources. Finally, as much of the research into personal exposure of particulate matter cites residential indoor environments as the primary exposure source, a review of indoor air quality has been undertaken.

2.2 Overview of Particulate Matter

Particulate matter consists of the solid and liquid droplets found in the atmosphere. Individually, these particles and droplets are invisible to the naked eye but collectively can appear as a cloud or haze (U.S.E.P.A., 1997). The size fractions investigated in this study were PM_{10} and $PM_{2.5}$, meaning particulate matter less than 10 microns (μm) or 2.5 μm in aerodynamic diameter (Brunekreef, 1994), these are considered to have the greatest impact upon human health. The historical legislation is reviewed to give context to the selection criteria.

Sources of particulate matter are varied, the larger size fraction under investigation in this study is generally referred to as PM_{10}. The sources of this size fraction tend to be natural, including wind blown soil, pollens and spores, and sea salt elements. The fine fraction of $PM_{2.5}$ is predominantly anthropogenic in origin. According to the Environmental Protection Agency Air Quality Criteria for Particulate Matter (U.S.E.P.A.,1996), PM_{10} is formed by crushing, grinding, and abrasion of surfaces, which breaks large pieces of material into smaller pieces. They are then suspended by the wind or by anthropogenic activity. Energy considerations limit the break-up of large particles and small particle aggregates generally to a minimum size of about 1 μm in diameter. Mining and agricultural activities are examples of anthropogenic sources of this size fraction. Fungal spores, pollen, and plant and insect fragments are examples of natural bioaerosols.

$PM_{2.5}$ is derived from combustion material that has volatilised and then condensed to form primary particulate matter or from precursor gases reacting in the atmosphere to form secondary particulate matter. This mode of particulate matter is formed by the nucleation of gas phase species, and grows by coagulation (existing particles combining) or

condensation (gases condensing on existing particles). They are composed of (a) freshly generated particles, in an ultra fine or nuclei mode, and (b) an accumulation mode, so called because particles grow into and remain in that mode (Wilson & Spengler, 1996). The graph in Figure 2.1 illustrates the formation and removal of the different particle size fractions in the ambient aerosol. The graph in Figure 2.2 shows the mass distribution of ambient particulate matter collected using size selective samplers. This diagram shows the mass of the different size distributions and it can be seen that the $PM_{2.5}$ mass is small compared to the PM_{10}. This is to be expected as the larger sized particles weigh more, however, if considering the health effects of these size fractions it may be more relevant to consider the number of particles that constitute each fraction. The number of particles in the $PM_{2.5}$ size fraction are greater than the PM_{10} fraction. See Section 2.3.4 for further details of particle number density.

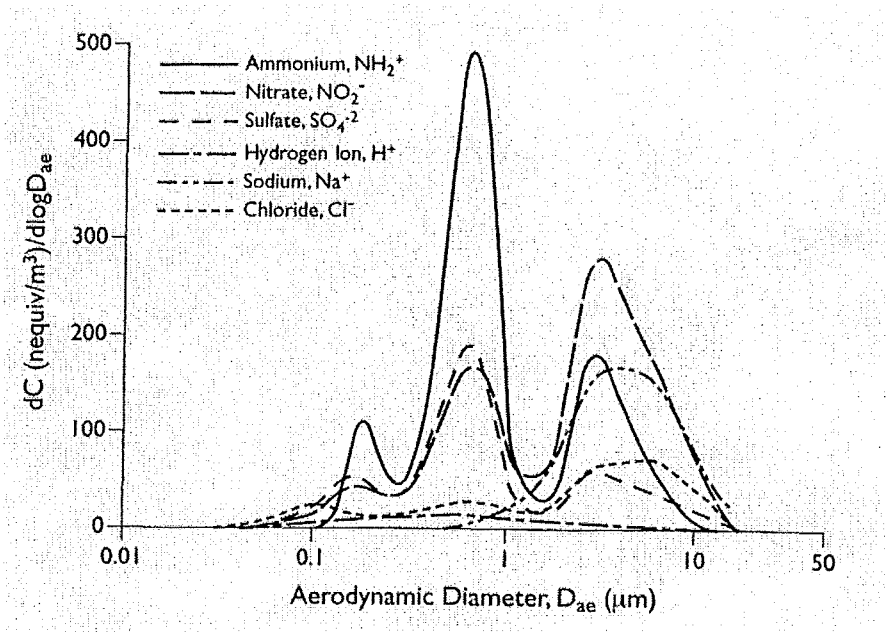


Figure 2.1 A schematic of an atmospheric aerosol size distribution showing the three modes, the main source of mass in each mode, and the principal processes involved in inserting mass into and removing mass from each mode (Koutrakis & Sioutas, 1996).

The Quality of Urban Air Review Group report (Harrison *et al.,* 1993) states an approximate breakdown of UK particulate matter composition as follows: ammonium ~5%, sulphate, nitrate and chloride ~30%, carbonaceous material ~40%, metals ~5% and insoluble material ~20%. Combining data from several studies derived this estimate. These studies were carried out in different places within the UK at different times using different techniques and cannot therefore be regarded as typical of specific locations within London. See Figure 2.3 for details of the estimated UK composition of both size fractions of particulate matter.

The composition of indoor particulates is not generally uniform. The general breakdown of particulate pollution within the home environment depends greatly upon the type of ventilation, heating, cooking fuel and numbers of smokers present (Wallace, 1996a). Section 2.6 discusses indoor air quality in greater depth. These factors have been included in the questionnaires developed for this research and are discussed further in Section 4.7.6.

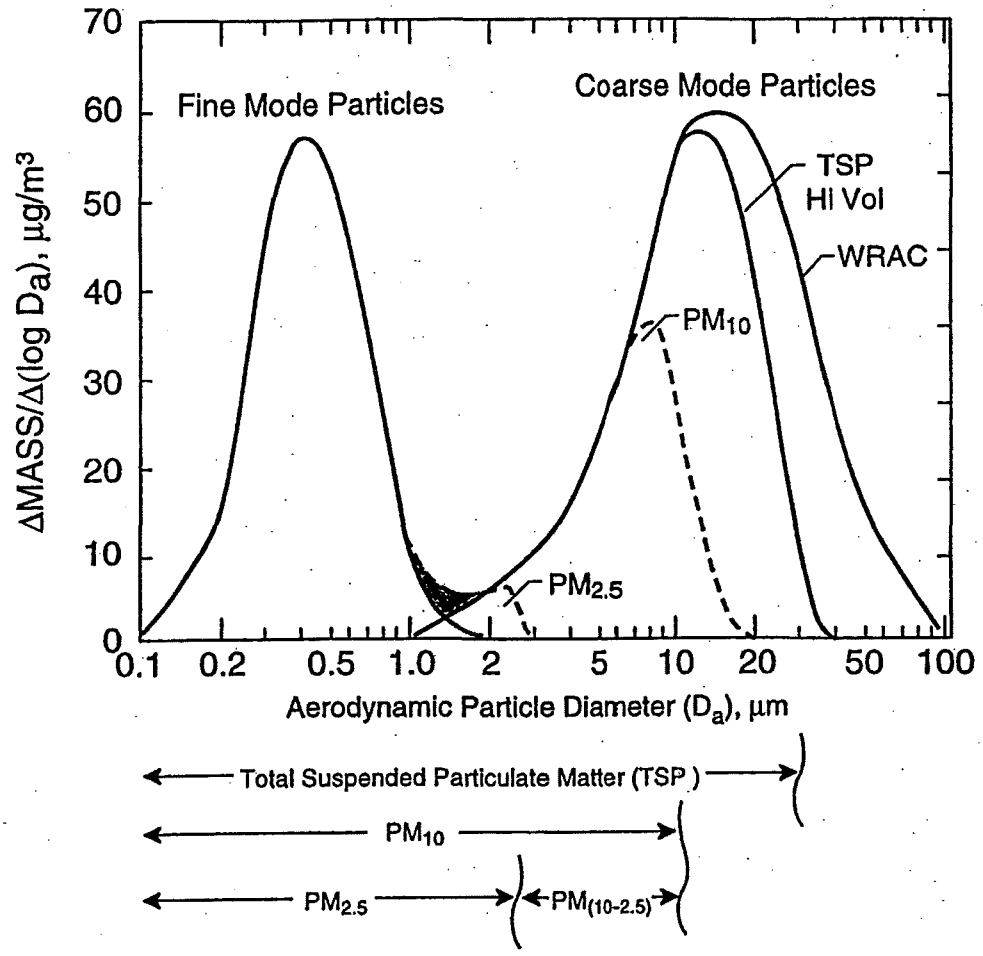


Figure 2.2 Mass distribution of ambient PM as a function of aerodynamic particle diameter (Lippmann & Maynard, 1999).

2.3 Characterisation of Particulate Matter

A number of different issues are relevant when discussing the nature of particulate matter. These include; their chemical composition, sources, number density, size distributions, physical morphology and size. These are discussed individually in Sections 2.3.2 to 2.3.5.

2.3.1 Particulate Matter Classification

The recommended classifications found in the International Standards Organisation document (ISO, 1983) details the nomenclature used by various authors and organisations.

The definitions of particulate matter are varied and for clarification purposes the different definitions are listed below, taken from the COMEAP report (Holgate, 1995).

Aerosol is technically defined as a suspension of fine solid or liquid particles in a gas, while common usage refers to the aerosol as the particulate component.

TSP Total Suspended Particulate a term describing the gravimetrically determined mass loading of airborne particles, most commonly associated with use in the US high volume sampler in which particles are collected on a filter for weighing.

PM_{10} particulate matter less than 10 μm in aerodynamic diameter (particles which pass through a size selective inlet with a 50% efficiency cut-off at 10 μm aerodynamic diameter). $PM_{2.5}$ is similarly defined, with a 2.5 μm aerodynamic diameter.

Smoke describes particulate matter <15 μm which predominantly originates from fossil fuel combustion.

Black Smoke non-reflective (dark) particulate matter, associated with the Smoke Stain technique developed in the 1960's.

Inhalable particles (also termed inspirable), particles which may be breathed in.

Respirable Particles can penetrate to the unciliated regions of the deep lung.

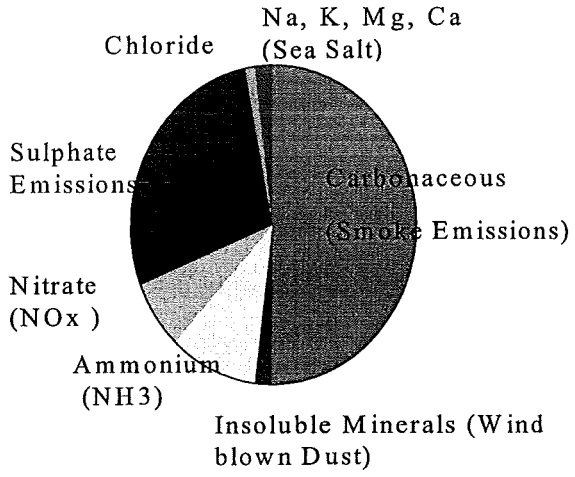
Thoracic Particle Mass describes that fraction of the particles which penetrate beyond the nasopharynx and larynx.

2.3.2 Composition of particulate matter

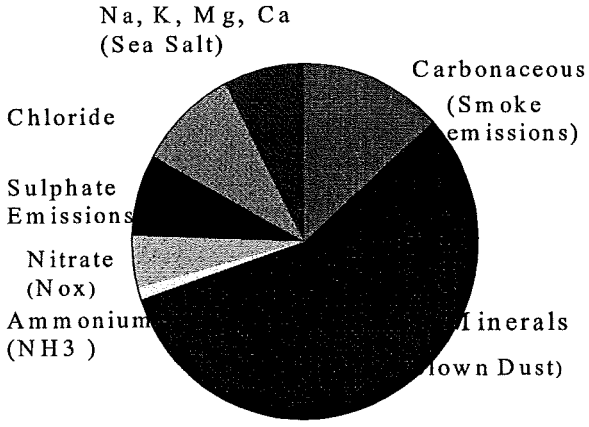
A number of studies have been carried out to determine the composition of particulate matter. However, particulate matter composition changes with regard to its source and its reaction with other aerosols whilst in transport. It is not known at present if UK particulate matter composition is similar to those of other countries where there are different sources and reaction mechanisms taking place. Hence, this study will use only UK research to provide an accurate description of UK particle composition. It has been stated that airborne particulate concentrations and composition are not expected to vary greatly from one location to another of the same type (e.g. urban) within the UK, (Harrison and Jones, 1995). As previously stated, sources of atmospheric particulate matter can be either natural or anthropogenic and the different sources result in different particle composition. Using the data obtained by Harrison and Jones (1995) which analysed particulate size ranges 2.5

and 2.5-15 μm, an approximate breakdown of UK particulate matter composition is given in Figure 2.3. This research has since been quoted in a number of reports including the Committee on the Medical Effects of Airborne Particles (COMEAP), Expert Panel on Air Quality Standards (EPAQS) and Airborne Particles Expert Group (APEG) reports on particles (Holgate, 1995, Seaton, 1995, Harrison *et al.* 1999). As previously stated the QUARG report recommends caution when assuming that this is representative of the UK particle composition. At this point however, there is insufficient data available so the approximate breakdown will be used in this research.

The smaller size fraction 2.5 μm differs from the coarser fraction as over 50% of the composition can be identified as carbonaceous matter from smoke emissions and 25% being sulphate emissions. The coarser fraction predominantly consists of insoluble minerals from wind blown dust with only 20% being carbonaceous in origin.



(a)



(b)

Figure 2.3 Typical Approximate Composition of Urban Airborne Particles: (a) Fine Fraction; (b) Coarse Fraction (Harrison and Jones, 1995)

2.3.3 Modal Distributions

The graph in Figure 2.1 demonstrates the different modes of particulate matter present within the atmosphere. The nucleation mode is the group with the smallest particle size. This mode consists of ions and nuclei (often of the dimensions of molecular clusters), and the particles into which they grow as a consequence of the condensation of vapours upon them. Particles arising from gas to particle conversion (e.g. sulphuric acid droplets from the oxidation of sulphur dioxide) are initially formed by condensation onto a nucleus. The size range of particles in this mode extends from that of molecular clusters 0.001μm in diameter to about 0.1μm. Condensation nuclei are usually present in very large number concentrations in urban atmospheres, but because of their small size they make a relatively small contribution to the total mass concentration. The lifespan of this size fraction is short as they rapidly grow into the accumulation mode. Removal processes of ultra fine particles include diffusion into rain droplets and accumulation into larger particles. The sources of these are vehicle exhausts, incinerators, and the chemical conversion of gases to particles in the atmosphere (Lee *et al.* 1986, U.S.E.P.A., 1996a).

The accumulation mode consists of particles that have grown from the nucleation mode by further condensation of vapours upon them or by coagulation. Their size range is usually about 0.1μm to about 3μm. These are relatively stable as the processes that remove particles from the atmosphere (e.g. diffusion, washout and sedimentation) are least efficient for particles in this size range (United Nations, 1979).

The coarse mode consists of particles greater than about 2.5 μm in diameter originating predominantly from mechanical processes such as erosion, resuspension and sea spray. Soil dust and most industrial dusts come within this category; as do pollens, mould spores and some bacterial cells. The lifespan of this size fraction is relatively short as the particles are easily removed from the atmosphere by deposition and rainfall, they tend to remain in suspension for only a few hours and therefore only travel a short distance (U.S.E.P.A., 1996a).

2.3.4 Particle Number Density

The majority of particulate mass is associated with the larger size fractions, as demonstrated in Figure 2.2. In contrast though the number of particles per unit volume increases with decreasing particle size. In terms of human health effects the smaller

particulates that are present in greater numbers penetrate deeper into the lung. The dose of these smaller particles into the lungs are therefore small in terms of mass, however the number is high and it may be that these are responsible for the observed health effects discussed in Section 3.3 (McAughey, 1997). The source apportionment of these smaller particulates is discussed in Section 2.3.2. The lack of available personal monitoring equipment for measuring particle number has resulted in the use of the integrated mass measurements described in Section 4.7.2.

A study of ambient particulate matter conducted in Germany by Peters *et al.,* (1997a) has shown that 73% of the particles counted were ultra fine (smaller than 0.1μm in diameter). However, these particles contributed only 1% to the mass concentration of fine particles. 82% of the overall mass was attributable to particles in the diameter range between 0.1 and 0.5 μm. Nevertheless, these particles contributed only 27% to the number concentration of fine particles. Particles with a diameter between 0.5 and 2.5 μm constituted less than 0.01% of the number concentration of fine particles, but 1% of the mass of fine particles. The study concluded that decreases in Peak Expiratory Flow (PEF), increased reporting of feeling ill during the day and cough were associated with the number and mass concentrations of the fine and ultra fine particles for a group of non-smoking adult asthmatics.

A UK study of ultra fine aerosol emissions from gas cooking in homes by Ross *et al.,* (1999) reported that gas combustion resulted in a very large number of ultra fine particles being produced. They also showed that this large number reflected a small amount of the total mass. The emitted particles also rapidly agglomerated which reduced the particle number and increased the mass. Within each home sampled for this study data was collected to identify the use of gas as a cooking fuel. This research has included homes with gas cooking and a questionnaire on their use was also conducted.

Watt and Kendall (1997) present a summary of particle number density. In a UK study carried out in Birmingham over a 24 hour period, it was found that between 12 midnight and 4 am, particle numbers drop suggesting that the emission sources are reduced, they then start to increase between 7 am until 7 pm. The coarse mode particles settle out leaving large numbers of small particles to agglomerate or be removed by wind action.

2.3.5 Physical Characteristics of Particulate Matter

The physical properties of particulates determine where they are deposited after being emitted and how long they remain in the atmosphere. The larger the size fraction of the particulates the more likelihood there is of them being washed out of the atmosphere by rainfall or sedimentation effects. The smaller ultra fine size ranges can remain in the atmosphere for up to several weeks before impaction or accumulation causes their removal (United Nations, 1979).

Particles in the atmosphere are varied in shape and those with similar settling velocities are assumed to have the same size. The aerodynamic diameter of a particle takes into account such variables as shape and density. A particle with a flat-plate like shape will have a smaller aerodynamic diameter and will remain in suspension for longer than a spherical particle with the same apparent physical diameter. A particle's atmospheric transportation needs to be considered in terms of the aerodynamic diameter rather than the mass (Seaton, 1995).

For the qualitative analysis of individual particles scanning electron microscopy can be employed. This uses accelerated electrons to strike the particle, generating a number of signals that can be used to interpret the particle's morphology and potential source. There are a number of limiting factors for the successful use of this method; these include the selection of filter media, optimal particle loadings, and sample handling (Casuccio *et al.*, 1983, U.S.E.P.A., 1996a). This technique is complex and expensive, hence the minimal number of filters that have been analysed, see Section 6.4 for the results of the analysis.

In a review of research conducted by Pooley and Mills, (1999) on particle morphology, several different types of particulate shapes were identified. Those particulates collected from roadside sites appear to be aggregates of random shapes. Diesel aggregates are spherical and appear as clusters in the shape of grapes or chains, these particles are generally very porous and can absorb gases easily, this alters their chemical composition. Some particulate morphology has been described in Section 6.4 in an attempt to determine the source apportionment of the collected particulates.

2.4 Particulate Matter Legislation

Particulate matter as an environmental pollutant has been defined differently over the years in UK legislation. In the past it was referred to as Black Smoke. It was primarily considered to be non-reflective particulate matter and was analysed by the darkness of stain obtained on a white filter paper through which air had been passed (Lippmann and

Maynard, 1999). Following the major smog episode of 1952 in London, legislation was set to reduce the number of smog events that occurred in UK urban areas. The 1952 episode led to 4000 additional premature deaths (Ostro, 1984, Schwartz and Marcus, 1990). The Clean Air Act was introduced in 1956 to reduce the emissions from domestic fires and to introduce smokeless fuels in smoke controlled areas. In 1976, The Royal Commission considered that black smoke was no longer a major air pollutant and as such the 1956 legislation remained unaltered (Fisher, 1996). In December 1991, meteorological conditions similar to those experienced in 1952 caused nitrogen dioxide levels to rise to record levels of 423 ppb, an exceedance of the World Health Organisation guideline level of 210 ppb. Black smoke concentrations also increased to a maximum daily average of 148$\mu g/m^3$ at a site in Westminster, well above the monthly mean standard of 43 $\mu g/m^3$. As a direct result of this pollution episode, it has since been estimated that in London during this period there was an increase of around 10% in overall death rates related to respiratory symptoms (Anderson *et al.* 1995). The Department of Health's Committee on the Medical Effects of Air Pollutants (COMEAP) and the Expert Panel on Air Quality Standards (EPAQS) have since made their recommendations to review this legislation. Both groups have proposed that there was a link between non-biological particles and adverse health effects (Holgate, 1995, Seaton, 1995). The EPAQS report concluded that, for any proposed standard, particulate matter should be measured using PM_{10} rather than Black Smoke. PM_{10} more closely represents the particles with the greatest health impacts due to their size and composition, replacing Black Smoke as the best method of measuring concentrations of airborne particles. Black Smoke is still appropriate for the measurement of building soiling. Health effects resulting from exposure to particulate matter will be discussed in Section 3.

A value of 50 $\mu g/m^3$ as a 99^{th} percentile measured over a 24 hour running period was suggested by the UK government as a PM_{10} standard to be achieved by 2005. For particles however, the existing objective is now known to be unachievable, so the Government proposes to replace the current objective with the less stringent European Union limit value, which is currently the only alternative nationally recognised target (UK DETR, 2000).

The EU has recently agreed limit values for PM_{10} of 50 $\mu g/m^3$ measured over fixed 24 hour periods, not to be exceeded more than 35 times a year, and an annual average limit value of 40 $\mu g/m^3$, both to be achieved by 2005. Indicative Stage II limit values have been set at 50 $\mu g/m^3$ not to be exceeded 7 times per year with an annual average of 20$\mu g/m^3$ (Harrison *et al.* 1999, EU Doc. 399L0030, 1999). The standard was decided upon as a concentration at

which individual's health effects are likely to be small and the large majority of individuals will be unaffected. The COMEAP report concluded that there was no evidence that healthy individuals are likely to experience acute effects to their health resulting from exposure to concentrations of particles found in UK ambient air. It was suggested however, that such effects would be found in individuals with pre-existing respiratory or cardiac disorders and by reducing the PM_{10} concentration such health effects would be reduced (Holgate *et al.*, 1995). Further discussion of the links between particulate matter and health is found in Section 3.3.

The World Health Organisation has declined to put forward a particulate matter standard as no threshold for health effects have been identified (ENDS, 1997). This implies, therefore, that there is no safe level for human exposure to particulate matter.

In 1997, the US set a new standard for particulate matter. This was based upon health implications that became evident from recent epidemiological studies. Recommendations state that the sources of the two size fractions, PM_{10} and $PM_{2.5}$, are different and should therefore be treated as such with separate legislation. A National Ambient Air Quality Standard (NAAQS) has been established for $PM_{2.5}$ with an annual mean value of 15 $\mu g/m^3$ and a 24-hour standard of 65 $\mu g/m^3$. The PM_{10} 24 hour NAAQS is 150 $\mu g/m^3$ with an annual value of 50 $\mu g/m^3$ (U.S.E.P.A., 1997). It is anticipated that future recommendations for particulate matter will cover the smaller size fractions of PM_1 and below.

In Japan, there is now legislation that covers indoor and non factory environments in an attempt to control the exposure of individuals to indoor sources of particulate matter (150 $\mu g/m^3$ averaging time for respirable particulate matter). It has been recognised that particulate concentrations measured at ambient monitors do not adequately reflect those found indoors (Peterson, 1992). In Norway, the Norwegian Health Authorities recommend upper limits for residential indoor PM_{10} levels of 90 $\mu g/m^3$ and 40 $\mu g/m^3$ for the $PM_{2.5}$ fraction (Ormstad *et al.* 1997).

2.5 Sources of Ambient Particulate Matter within the London Borough of Barnet

Research by Harrison and Jones (1995) cites the sources of primary airborne particulate matter within the UK urban environment. These include emissions from vehicles, stationary combustion processes such as coal burning, and industrial processes. Road transport is the major source of particulate emissions in London accounting for over 80% of the emissions whilst the stationary combustion processes account for only 5%.

The latest London Emissions Inventory (Buckingham, 1997) is a database that includes a number of sources regulated by the Environmental Protection Act of 1990. Within the boundaries of the London Borough of Barnet, this includes one Part A process which is a major combustion process, Brent Smelt works (an aluminium recycling plant). There are seventeen lower emission Part B processes. These include seven paint respraying garages, four crematoria, one adhesive coating process factory, one process that crushes and grades concrete, one cement works and two waste oil burners in the Barnet area. The locations of these processes in relation to the study's sampling sites are indicated in Figure 4.1.

The numbers of point sources within the London Borough of Barnet are small in comparison to the other boroughs. The contribution of industrial pollutants is less significant compared to other sources. The London Emissions' Inventory concludes that 78% of PM_{10} emissions in London are a result of road transport. Other sources include rail transport and area sources that include off-road vehicles and construction dust. There are major road networks within Barnet including the M1, A406 (North Circular Road), A1, A5, A41 and A1000 (Great North Road) all having traffic flows of over 25,000 vehicles per day (Brook, 1996). The high vehicle traffic flows make a major contribution to PM_{10} levels within Barnet when compared to other sources (Crabbe and Beaumont, 1998).

Recent research conducted in Harlem, New York City found that average $PM_{2.5}$ concentrations exhibited modest variations across four sites. The predominant factor influencing $PM_{2.5}$ was estimated to be from regional sulphate emissions. The study investigated the localized influence of diesel vehicle emissions of elemental carbon. It discovered that there was great spatial variation in diesel particulates related directly to transport type (Kinney *et al.*, 2000). This suggests that in cities that have high diesel use components of $PM_{2.5}$ resulting from these emissions could potentially be resulting in a stronger influence upon health on a very localised basis. It also suggests that distance from the road is unlikely to influence the spatial resolution of $PM_{2.5}$ in urban areas.

There will be some imported PM_{10} source contributions primarily from sources within neighbouring boroughs of Harrow, Brent and Camden as these are situated in the prevailing wind direction. Some sources found within Enfield, Hartsmere Borough Council and Haringey may also contribute (Crabbe and Beaumont, 1998).

These sources will influence personal exposure to particulate matter. Data has been collected using time activity diaries to determine the children's exposure to such sources in the outdoor environment. This is investigated further in Section 5.10.

2.6 Indoor Air Quality

The concentrations of air pollutants within the indoor environment are influenced by a number of factors summarised by Ashmore (1995) and include:

i) the number and location of indoor sources, and their rates of pollutant emission;

ii) the characteristics of the use of the specific sources;

iii) the size and structure of the building;

iv) the rate of air infiltration from outside;

v) the ventilation and mixing of air within the indoor environment;

vi) the removal rate of the pollutant, by physical deposition to surfaces or by chemical transformation;

vii) the outdoor concentration of the pollutant.

These factors have been cited in a number of the personal exposure studies reviewed in Section 3.5. Personal and indoor sources appear to be linked, therefore, altering any of these factors will also influence personal exposure concentrations.

The indoor sources of particulate matter within homes tend to be from ETS, cooking, heating fuel used, cleaning and resuspension of particles from people moving around. Abt *et al.*, (2000) used continuous methods to determine the sources of particulate matter within four homes. It was found that for particles between 0.7 –10 μm that sautéing, cleaning, presence of people and frying were the main contributors. The relationship between air exchange rates and indoor particle concentrations was also investigated in the paper. The findings suggested that at lower air exchange rates (<1 exchange / hour) there is a longer residence time allowing indoor sources of particulate matter to accumulate. When the air change rate increased to >~2 exchanges / hour there was a decreased variability in the indoor outdoor ratio, basically indicating that the indoor and outdoor concentration of particulate matter was the same. The follow up study by Long *et al.,* (2000) found similar results in nine households, they also saw a reduced correlation between indoor and outdoor particulate matter concentrations due to the indoor generated particles.

Wallace (1996a) reviewed three major indoor particle studies that showed indoor particulate matter concentrations exceeded those outdoors. A study carried out in two New York state counties assessed the influence of kerosene heaters, gas or wood stoves, or fireplaces, and cigarette smoking on indoor concentrations of $PM_{2.5}$ (Sheldon, 1988). The study was carried out over a winter period and results indicated that the mean indoor $PM_{2.5}$ concentrations were approximately double the outdoor concentrations in both counties. Smoking was again the most significant source within the home. It was also estimated from this study that in homes without any of the combustion sources 60% of the total $PM_{2.5}$ mass was from outdoor sources, and 40% from unidentified indoor sources.

The presence of animals within the home has been shown to increase the concentrations of particulate matter especially those associated with the size fraction 2.5 - 10 μm (Kamens *et al.*, 1991).

Santanam *et al.*, (1990) found that children living with smokers had a higher estimated exposure to particulate matter than those living in non-smoking households. Approximately 40% of the particulate matter was attributable to environmental tobacco smoke.

The PTEAM study (as described in Section 3.5) found that there was very little variation between rooms sampled, hence recommending that a single room within the homes could be used for monitoring. The mean indoor concentration for PM_{10} was 58.7 $\mu g/m^3$ compared to 62.6 $\mu g/m^3$ outdoors, $PM_{2.5}$ concentration were 36.3 $\mu g/m^3$ and 42.6 $\mu g/m^3$ respectively. The outdoor concentrations exceeded the indoor for this study. Possible reasons given for this include; the fact that the homes have no smokers present, and there are few indoor sources in the homes which also have air conditioning (Wallace, 1996a).

There have been very few assessments of particulate matter concentrations within schools, two being conducted in the Netherlands and one in the US. The study of children's personal exposure to particulate matter undertaken by Janssen (1998) included measurements within four schools. The classroom concentrations of $PM_{2.5}$ were highly correlated with the outdoor concentrations. During school hours, the outdoor values were about 5 $\mu g/m^3$ higher than the 24-hour averaged classroom concentrations. The summary of the data collected for PM_{10} concentrations for 3 of the schools estimated a 24-hour average of 40.4, 11.5 and 31.1 $\mu g/m^3$ higher than the ambient values. The high classroom concentrations and influence of physical activity found for PM_{10} (and not for $PM_{2.5}$) are probably a result of resuspension of coarse particles caused by the activity of the children.

This was proven when analysis of the particulate matter was undertaken and there was a strong correlation between the PM_{10} fraction and soil derived components. The most significant sources of the particles were from resuspension of settled dust and/or suspension of soil material brought in by the children's shoes. The two size fractions have apparent different origins. The $PM_{2.5}$ fraction reflected the ambient concentrations. The other Dutch study of particulate matter concentrations within classrooms was undertaken by Roorda-Knape *et al.,* (1998) and was designed to assess the respiratory health of children living near to motorways. PM_{10} measurements were made in a number of the classrooms during the weekdays whilst the children were at school. The results were highly variable and much higher (median = 73.1 $\mu g/m^3$) than those measured outdoors (26 $\mu g/m^3$). The classroom concentrations were not correlated with distance from motorways, traffic intensity or percentage of time downwind. A US study measured indoor and outdoor TSP concentrations in 6 schools and the indoor concentrations were lower than the outdoor concentrations, however these schools all had air filtration systems (Janssen, 1998). This could explain the differences in concentration between the Dutch and American schools.

Full air conditioning reduces the infiltration into the indoor environment of particulate matter from outdoor sources as the air is recirculated rather than being drawn in from outside. It has been shown that a reduction in air exchange can lead to an increase in the concentration of particulates from indoor sources (Dietz and Cote, 1982).

To understand the relationship between outdoor and indoor particulate matter concentrations a review of available literature was conducted. The ratio of indoor to outdoor particulate matter for PM_{10} during the day was estimated to be 0.3 for German homes by Müller (1991). A number of US studies of I/O during the day range from 0.5 to >1, these studies were undertaken in a number of different homes where the outdoor concentrations varied with the prevailing industrial sources that were present (Quackenboss *et al.,* 1989, Lioy *et al.,* 1990, Colome *et al.,* 1992, Özkaynak *et al.,* 1993). A study in Taiwan found ratios for both PM_{10} and $PM_{2.5}$ fractions of 0.61 and 0.68 respectively (Li, 1994). A study carried out in Oslo, Norway indicated that a median value of 1.43 for suspended particulate matter was found in homes that generally have higher indoor concentrations than outdoors. In an attempt to maintain the appropriate thermal conditions, these houses tend to be more airtight than homes in the US and other more temperate countries (Ormstad *et al.,* 1997). Studies of $PM_{2.5}$ have mostly been undertaken within the US and these tend to have higher ratios for indoors and outdoors ranging from 0.73 at night (Clayton *et al.,* 1993) to 1.4 during the winter (Wallace, 1996a). Other studies of daytime ratios were between 0.8 and 1.04 suggesting that there is less of a building

effect for the smaller sized particles (Clayton *et al.*, 1993, Özkaynak *et al.*, 1993, Wallace, 1996a).

2.7 Summary

The studies that have been reviewed show a number of factors that influence individuals' exposure to particulate matter. The outdoor concentrations are generally poorly correlated with the personal and indoor values in the majority of studies, except where the outdoor levels greatly exceed the indoor concentrations. The indoor concentrations are influenced predominantly by incidence of smoking, which particularly affects the $PM_{2.5}$ concentrations. Other factors that have been shown to influence personal exposure and indoor air quality are cooking, physical activities causing resuspension of particulate matter and heating fuel used within the home. There appeared to be a stronger correlation between the personal exposure concentrations and the indoor concentrations for the majority of the studies reviewed.

This research has assessed the potential sources of particulate matter reported in the literature and has incorporated measures to assess the impact that they have upon children's exposure.

Reviewing the characterisation of particulate matter has emphasised that the mass is not the only important component. Limitations of sampling equipment are primarily the reason for not investigating the number, mode and chemical composition. Some morphological analysis and mass concentrations have been investigated in Chapters 5 and 6.

The location of the microenvironmental sampling assumes no significant differences between rooms within a home, such that the main living room can be used to assess the quality of indoor air (Ju and Spengler, 1981). In the PTEAM study, the room-to-room variation using several indoor monitors is described. The results indicated that the integrated particle levels over 12 hours were less than 10% different between rooms. The indoor mean concentrations of all rooms were therefore used for the analysis of indoor and outdoor comparisons (Wallace, 1996a).

3 Human Exposure to Particulate Matter and Health Implications

3.1 Introduction

Understanding human exposure to particulate matter will provide information as to how such exposure, and the potential risk to health, can be reduced. The environments where people spend most of their time are at home and work, or school (U.S.E.P.A., 1996a). Several studies have now identified specific sources through the use of tracers and estimated the likely personal exposure of different population groups to particulate matter in domestic environments. These will be reviewed in Section 3.4.

The objectives as stated in Section 1.2 require the development of a suitable methodology for the personal exposure monitoring of children to particulate matter. With respect to this the literature review assesses previous research for methodologies, development of questionnaires, and the use of Time Activity Diaries (TAD). Much of the research reviewed on personal exposure monitoring has been carried out to determine the best methodology for undertaking Objectives 1, 3 and 4. Although Objective 5 was not fully achieved through this research the literature has been reviewed in an attempt to understand what the health implications are for exposure to particulate matter. The sampling equipment used for personal exposure sampling is also reviewed.

3.2 Particulate Matter Deposition within the Human Lung

The clearance mechanisms within the lungs are varied, depending upon the size and deposition site of the particulates. The diagram in Figure 3.1 shows the areas of deposition for the different sized particles. Particles that enter into the human respiratory system can be deposited in one of three regions, or be exhaled. These areas are referred to as nasopharyngeal, tracheobronchial and alveolar. The nasopharyngeal and tracheobronchial regions are cleared mostly to the stomach and components of the particles may be absorbed in the gut or excreted. Particles which are deposited within the alveoli are less efficiently cleared and can be absorbed into the bloodstream, or, if insoluble and inhaled in sufficiently large amounts may cause lung diseases such as pneumoconiosis or emphysema. Inhalation of some particles may lead to bronchial carcinoma induced by inhalation of such particles as asbestos fibres (Holgate, 1995). The human body reacts differently to particles identical in size but of different composition. Some examples of these reactions are:

a) the inhalation of biological aerosols which may cause a direct reaction of the lung tissue.

b) the number of inhaled particulates may result in the inflammation of the alveoli.

c) chemical components of particulate matter have been associated with cardiopulmonary effects.

Understanding the size and chemical composition of the particulates inhaled will aid in the identification of their potential effect on human health as discussed further in Section 3.3.

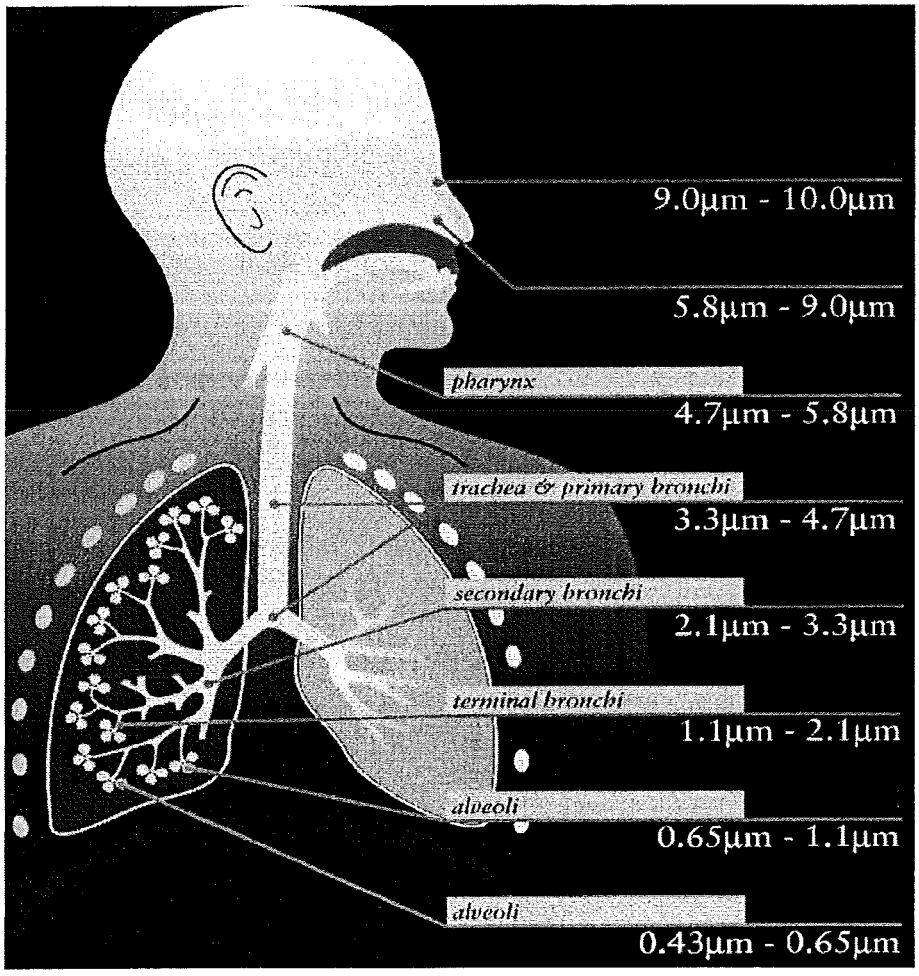


Figure 3.1 Sites of different sized particulates' deposition within the human lung. (Courtesy of Graseby Andersen).

3.3 Human Exposure and Health Effects

An individual's response to exposure to particulate matter depends upon a number of factors.

- The composition of the particulate matter i.e. size, number, shape and chemical characteristics (as discussed in section 2.3).

- The health of the individual inhaling the particulate, i.e. whether they are healthy or predisposed to allergic rhinitis, asthma or any other respiratory disorder.

- The concentration of the particulate matter within the individual's specific environment causing their exposure, dose and uptake to be unique.

There are two different definitions of exposure. One definition is that exposure is a function of 'contact at a boundary between a human and the environment with a contaminant of a specific concentration for an interval of time,' i.e. concentration in one environment. This does not account for the effects of an individuals' movement through different environments and time spent there (Ott, 1995). A more preferable definition of exposure is that at some instant of time there is a joint occurrence of two events:

1) a pollutant of known concentration is present at a particular location in space at a particular time, and

2) that the person is present at the same time and location in a space (Özkaynak, 1999).

By assessing these factors of activity and location in relation to the definition suggested by Özkaynak (1999), it is possible to model the likely dose and uptake of pollutants by individuals and assess the impact that specific multiple environments have on exposure to these pollutant concentrations.

It is important to identify groups of the population that are considered to be more sensitive to exposure to particulate matter so that policies can be implemented to reduce their exposure. Groups most clearly at risk from acute effects include those with pre-existing respiratory and/or cardiac disorders (Holgate, 1995). It has been suggested by Seaton, (1995) that the ultra fine particles are able to provoke alveolar inflammation, this causes a release of mediators capable, in susceptible individuals, of causing the exacerbation of lung disease and of increasing the blood's ability to coagulate. This may also explain the observed increases in cardiovascular deaths associated with urban pollution episodes. Other research by Peters *et al.*, (1997b) suggests that ultra fine particles are associated with decreases in peak expiratory flow (PEF) in asthmatics. Children may also receive an increased dose of particles to their lungs compared to adults, (see Section 3.3.2 for potential health effects). This research has selected children for the sample group as no research has been undertaken to assess their personal exposure to particulate matter in the UK.

3.3.1 Health Effects to Adults from Exposure to Particulate Matter

Epidemiology studies indicating mortality and hospitalisation as a result of respiratory illness show strong relationships with ambient particulate matter. In a review of studies

conducted in US urban areas, Dockery and Pope, (1994) found epidemiological evidence that a daily increase of 10 $\mu g/m^3$ exposure to PM_{10} resulted in an acute response of an increase of between 0.7% and 1.6% in daily mortality. The review by Ostro (1993) reached similar conclusions where a 10 $\mu g/m^3$ increase in exposure to particulate matter led to a mean increase of 0.96% in mortality. The primary cause of death was directly related to respiratory disease and cardiovascular disease being the secondary cause (Pope *et al.* 1995). Wordley *et al.,* (1997) conducted a study in the UK to determine the presence and magnitude of any relationships between short-term variations in ambient concentrations of PM_{10} and hospital admissions and mortality. The study found an association between all causes of mortality and all deaths from circulatory causes one day after high PM_{10} ambient concentrations. Mortality due to Chronic Obstructive Pulmonary Disease (COPD) was significantly associated with same day ambient PM_{10} concentrations. This has resulted in an estimate that an increase in 10 $\mu g/m^3$ exposure would cause a 1.1% increase in all causes of mortality for UK populations. Other studies have also indicated that an increase in particulates is associated with a short-term increase in mortality, especially in urban areas (Michelozzi *et al.* 1998). Consistent associations between the particulate matter concentrations and mortality have been demonstrated in a number of different countries, measured over different seasons (Dockery and Pope1994, Hoek *et al.* 1997, Katsouyanni *et al.* 1997, Lipfert, 1980, Schwartz and Marcus, 1990, Simpson *et al.* 1997). There does not appear to be a threshold value at which particulate matter stops having an effect upon human health and mortality. This was reflected in the WHO legislation for particulate matter as discussed in Section 2.4. In addition, as age increases it would appear that the particulate matter associations with mortality also increase.

Recent papers have suggested that the relationship between exposure to particulate matter and mortality may be closely linked to heart rate and heart rate variability (Gold *et al.,* 2000). There is a suggestion that on high pollution days heart rates are elevated and this could modify the autonomic control of the heart (Peters *et al.,* 1999, Pope *et al.,* 1999). Other suggestions have been forwarded that exposure to particulate matter increases plasma viscosity possibly leading to transient ischaemic events in people with existing coronary heart disease (Peters *et al.,* 2000)

The effect that an increase of 10 $\mu g/m^3$ of ambient particulate matter exposure has upon respiratory disease related hospital visits has also been widely studied (Dockery and Pope,1994, Gordian *et al.* 1996, Pope *et al.* 1995, Spix *et al.* 1998, and Wordley *et al.* 1997). All of the studies agree that an increase in ambient particulate matter is associated with increased hospital admissions for asthma, bronchitis, all respiratory diseases and

pneumonia. Estimates range between 1% and 2.4% increase for all respiratory symptom hospital admissions.

Asthma exacerbations, sometimes requiring medical attention, have also been associated with ambient PM_{10} exposure (Neas *et al.* 1994, Romieu *et al.* 1996, Dockery *et al.* 1989, Dockery & Pope 1994, Pope *et al.* 1995, Schwartz *et al.* 1993 and Wordley *et al.* 1997). Particulate matter may cause an increase in susceptibility to infectious disease by decreasing the lung's clearance, impairing macrophage function, or through other specific and non-specific effects on the immune system. Research conducted by Jarvis *et al.*, (1996) have shown an association in females between the use of gas for cooking with asthma symptoms and reduced lung function. In the review by Ghio & Samet, (1999) it is stated that airway reactivity after oxidant exposures can be associated with an influx of inflammatory cells. This inflammation is believed to increase reactivity of the airways through several putative mechanisms, including wall oedema, mediator release and epithelial damage. The relationships of inflammation with both ionisable metal concentrations and oxidant generation are consistent with the increase in bronchial reactivity observed with these particles. This suggests that the composition of the particulate matter is an important predictor of lung injury, with the primary source of these metal oxidants being of anthropogenic origin (Ghio & Samet, 1999).

A Norwegian study of hospital admissions for acute respiratory disease over a 3 year period suggests that PM_{10} is mainly an indicator of air pollution in general. The study sampled eight major air pollutants and found a stronger relationship with benzene and other VOC's. They suggest that vehicle emissions are more important for health effects of air pollution than just particulates alone. The study did not include $PM_{2.5}$ measurements which may be a more representative size fraction of vehicle emitted particulates. Again, multi-pollutant measurements are clearly needed to determine health effects (Hagen *et al.*, 2000).

Long term studies of individual's chronic health effects from exposure to air pollutants are limited. Abbey *et al.*, (1998) report that exposure to PM_{10} over a 20 year period is associated with a decrease in predicted Forced Expiratory Volume in one second (FEV_1) in non-smoking Californian males. Confounding effects within this study have been recognised. The most significant confounding effects were associated with short-term changes in air pollution. To identify the influence that this had the study was conducted during low pollution periods of the year.

The majority of these epidemiological studies are based upon ambient measures of particulate matter. Few personal exposure studies have been conducted that include health measurements, a result of the small numbers of participants that can be monitored at any time. Linking the health effects of these small numbers of samples to the epidemiological evidence is difficult as personal exposure studies are generally from selection biased groups within the population hence these results cannot then be applied to the wider population.

3.3.2 Health Effects of Children's Exposure to Particulate Matter

Children's response to air pollution episodes have been shown to be different from those experienced by adults. This may be due to (1) greater fractional deposition with each breath and/or (2) larger minute ventilation relative to lung size (Bennett *et al.* 1998). Allergic disease usually first develops and is common in children and adolescents. Sensitisation more commonly occurs with this age group as well. These are important issues when considering the effects of air pollution upon children's health (Carswell, 1995). An EPA report on the environmental health threats to children indicated that children's systems are still developing, including rapid changes in growth and development, immature body organs and tissues. They also breathe more air per pound of body weight, and because they play outside more they, they are more exposed to environmental threats (U.S.E.P.A., 1996b)

Studies indicate that children's exposure to particulate matter results in increased bronchitis symptoms and small decreases in lung function. A Harvard study investigating children's exposure to indoor air pollutants found an association between respiratory symptoms and the presence of some combustion sources such as kerosene heaters, wood stoves, gas cooking and cigarette smoking (Spengler *et al.* 1987). Other studies have also shown that domestic heating attributed emissions increase the risk of bronchitis (Herbarth *et al.* 1997). In the studies undertaken by Ware *et al.,* (1984) and Maier *et al.,* (1997), an assessment of childrens' respiratory health effects from exposure to indoor and outdoor air pollutants also assessed effects from passive smoking and gas cooking. After controlling for the confounding effect of parental education, it was found that there was no consistent pattern of increased risk for children from homes with gas stoves compared with children from homes with electric stoves. Associations were found between increased respiratory illness (around a 35% increase) and maternal smoking. Winter studies of Dutch asthmatic children with chronic respiratory symptoms found that there was an increase in the reported asthmatic attacks associated with particle exposure (Roemer *et al.* 1993). Other studies that

have measured lung function in schoolchildren have all identified decreases in FEV_1 associated with daily PM_{10} (Asgari *et al.* 1998, Dockery *et al.* 1989, Dockery & Pope 1994, Linn *et al.* 1996, Peters *et al.* 1997b, Pope & Dockery, 1992, Romieu *et al.* 1996, Scarlett *et al.* 1996 and Vedal *et al.* 1998).

Other studies have identified that an increase in indoor $PM_{2.5}$ leads to an increased cumulative incidence of lower respiratory symptoms, and is weakly associated with decreased pulmonary function levels in preadolescent children (Neas *et al.* 1994). It has been reported that respiratory illness in childhood may subsequently lead to the development of respiratory diseases in adulthood. There is also an associated risk of the development of COPD in smokers (Samet *et al.* 1983).

Research by Brunekreef *et al.,* (1997) identified a decrease in lung function associated with proximity to busy roads in the Netherlands. The study indicated that children living within 100m of motorways in the areas most exposed to truck traffic had poorer lung function than children living farther away. Wjst *et al.*, (1993) also found that high rates of road traffic in Germany diminished children's forced expiratory flow and increased respiratory symptoms. This was associated with a mixture of particulate matter and other common air pollutants resulting from vehicles.

The recently published report of the Scientific Committee on Tobacco and Health (Poswillo, 1998) acknowledges that passive smoking may be a cause of lung cancer and childhood respiratory disease. It also discusses the evidence that passive smoking is a cause of ischaemic heart disease and cot death, middle ear disease and asthmatic attacks in children. Neas *et al.,* (1994) studied indoor particulate matter and the effects of passive exposure to ETS in the home on respiratory symptoms and pulmonary function in children. The research indicated that indoor exposure to $PM_{2.5}$ is associated with an increase in the cumulative incidence of lower respiratory symptoms and is weakly associated with decreased pulmonary function level in preadolescent children. Research involving children living in Hong Kong also identified relationships between smoking and passive smoking with increased throat and nose problems, cough, phlegm and wheezing (Lam *et al.* 1998). Research in the Netherlands has also shown that exposure to ETS in the home is associated with an increase in the prevalence of cough and decreased lung function (Dijkstra *et al.* 1990), as did a study in New Zealand (Moyes *et al.* 1995). The review of children's health effects and exposure to ETS by Etzel *et al.,* (1997) summarised that children exposed to ETS had increased rates of lower respiratory illness and increased rates of middle ear

effusion, asthma, and sudden infant death syndrome. Exposure to ETS has also been associated with development of cancer during adulthood.

A study conducted in Southern California of asthmatic children showed a causal relationship between increased exposure to particulate matter and increased prevalence of chronic phlegm production and with bronchitis (McConnell *et al.,* 1999).

These studies, conducted in different countries assessed exposure and impact upon children's health to particulate matter both indoors and outside, raise a number of significant issues. The majority show an association between increased particulate matter concentrations and reduced lung function. Many also indicate that exposure to ETS is an important factor when determining lower respiratory disease, cough and middle ear effusion. This has also been acknowledged in the reanalysis of three studies undertaken by Schwartz & Neas (2000) where fine particles, $PM_{2.5}$, have been shown to have much stronger acute respiratory effects then coarse particles. There has been little research undertaken into the effect of childhood exposure on adult health later in life. Some links have been suggested, especially with cancer and COPD. These have not been quantified however, it is probable that by reducing children's exposure to particulate matter it may be possible to reduce the likelihood of these specific diseases occurring as an adult and it should also reduce the amount of time that the children are absent from school (Ransom and Pope, 1992).

A large European study, Pollution Effects on Asthmatic Children in Europe (PEACE) study included research from a number of cities to assess the acute effects of particles (PM_{10}), black smoke (BS), sulphur dioxide (SO_2) and nitrogen dioxide (NO_2) on the respiratory health of children with chronic respiratory symptoms. The study was conducted in the winter of 1993/1994 by 14 research centres in Europe. A total of 2,010 children, divided over 28 panels in urban and suburban locations, were followed for at least 2 months. The research showed that only previous day PM_{10} was negatively associated with evening PEF, but only in locations where black smoke was high compared to PM_{10} concentrations (Roemer *et al.,* 1998).

To summarise the health effects associated with adults' exposure, particulate matter does not appear to have a safe threshold of exposure. There are issues of confounding factors when considering the influence of air pollution effects upon health such as meteorology, occupational exposure, socio-economic status and pre-existing health effects. As stated a number of the studies which were reviewed accounted for these.

3.4 Rationale for Personal Exposure Studies

In the UK, the Department of the Environment, Transport and Regions (DETR), along with the Department of Health (DoH), identified the need for personal monitoring for three major purposes;

i) Epidemiology studies; personal monitoring will give increased support for the causality of statistical association and the specificity of action of the pollutant concerned.

ii) Toxicology; personal monitoring is an essential element in defining dose-response relationships on which assessments and risk management decisions can be based.

iii) Policy decisions in the area of air health effects and control actions, (Smith, 1994).

Prior to undertaking personal exposure studies several issues should be considered;

- purpose of the exposure assessment,
- most appropriate methodology for monitoring the specific pollutants,
- selection of an appropriate sample size,
- collection of the relevant survey data,
- modelling methods to be used (Loth & Ashmore, 1994).

Personal monitoring is essential to establish a known frequency distribution of the public's exposure to certain pollutants (Loth & Ashmore, 1994). The International Standards Organisation in 1981 stressed the importance of gathering information over different size ranges (expressed as Mass Median Aerodynamic Diameter, MMAD) to obtain databases for the evaluation of the health effects caused by inhalable particulate matter (Spagnolo & Paoletti, 1994). Personal exposure monitoring of particulate pollution is now recognised for inhalable, thoracic and also respirable fractions, with both European standards and International standards being set (Kenny, 1996a).

3.5 Review of Personal Exposure Research

Studies using personal exposure monitoring have shown that exposure to concentrations of a variety of air pollutants are substantially higher than estimates from fixed site monitors

(Wallace, 1993, and Mage & Buckley, 1995). Mage & Buckley, (1995) reviewed the literature from 14 studies available at the time to assess the relationship between personal exposure to particulate matter and fixed site monitoring. These studies included a number of different cut sizes, different seasons or were from annual sampling. The results indicated no significant relationships between the personal exposure monitoring and the ambient sampling. Seven of the studies also included indoor sampling and these showed a much stronger relationship with the personal exposure values. A reason suggested for the discrepancy between the indoor and outdoor correlations with the personal monitoring could be due to the different sources and composition of the particulate matter as discussed in Section 2.3.

In the UK there have been no studies of children's personal exposure to particulate pollution. The majority of personal exposure studies have been completed in the US (Wallace, 1996a). Some of the research has been carried out using a questionnaire based approach (Neas *et al.* 1994). Other US studies such as the Particle Total Exposure Assessment Methodology (PTEAM) included a small number of children in the cohort of individuals sampled (Clayton *et al.* 1993). It has been suggested that the findings from the US studies do have considerable implications for UK research (Loth & Ashmore, 1994).

3.5.1 US Personal Exposure Studies

The main goal of the PTEAM study was to estimate the frequency distribution of exposures to PM_{10} particles for all non-smoking Riverside, California residents aged ten and above. The findings showed that the population-weighted day time personal PM_{10} concentrations averaged about 159 $\mu g/m^3$, compared to the indoor or outdoor mean concentrations of 95 $\mu g/m^3$. The overnight personal PM_{10} mean was much lower (95 $\mu g/m^3$) and more similar to the overnight indoor (63 $\mu g/m^3$) and outdoor (86 $\mu g/m^3$) means. The major reason cited for this increased exposure was determined to be largely a result of the personal cloud effect, where individual's activities resulted in them being closer to sources or causing resuspension of particulate matter. Outdoor PM_{10} concentrations could explain about 25-30% of the variance observed in indoor concentrations, but only about 16% of the variance in personal exposures. Neither the indoor concentrations alone nor the outdoor concentrations alone, nor the time-weighted averages of indoor and outdoor concentrations, however, could explain more than about two-thirds of the observed variance in personal exposures. The major factors influencing indoor air quality were assessed to be the outdoor particle concentrations along with cooking and smoking. The correlations between the personal exposure concentrations and

the ambient measurements during the day were 0.37 and at night 0.54. The indoor and personal exposure values were more strongly correlated during the day and night with values of 0.63 and 0.88, respectively (Spengler *et al.* 1985, Özkaynak *et al.* 1993 and 1996). The 'extra' personal exposure concentrations could potentially be explained by resuspension during individual's activities.

The Harvard Six Cities Study reported by Spengler *et al.,* (1981) was one of the first studies to show that the indoor particulate matter concentrations can exceed those found outside. Cigarette smoke was identified as the major source of indoor particulate matter. Across all cities studied, except for Steubenville, the overall mean levels of particles were higher indoors than outdoors. An explanation for the higher ambient concentrations in Steubenville is that it lies in the heavily industrialised upper Ohio River Valley where there are large coke and steel plants, several coal-burning power plants, paper mills and smaller processing plants lining the river. It was concluded that for the majority of the cities, indoor levels were significantly higher than the outdoor concentrations. Even in homes without smokers, indoor particle concentrations were shown to equal or exceed outdoor levels. The personal exposure aspect of this study was conducted in Watertown, Massachusetts and Steubenville, Ohio and is reported in Dockery and Spengler (1981). They found that the personal Respirable Suspended Particulates (RSP) and outdoor correlations were strong (0.69). Approximately 48% of the variance in the personal measurements was explained by the outdoor values. An extension of this study was reported by Spengler *et al.* (1985) which attempted to quantify personal exposures to respirable particulate matter and gases for the population of Kingston and Harriman (Tennessee). In both towns, the averages of the personal and indoor concentrations of RSP were approximately 25 $\mu g/m^3$ higher than the outdoor RSP concentrations, suggesting the presence of significant indoor sources. Approximately 75% of the indoor samples and 95% of personal samples were above the mean outdoor average of 18 $\mu g/m^3$. All personal-ambient and indoor-ambient correlations were low and were not statistically significant. The personal-indoor correlations, however, were strong and statistically significant (0.7), most at the $p = 0.0001$ level. Only 1% of the personal exposures for the whole sample group could be explained by the outdoor RSP measure whilst 50% of the personal exposure variance could be explained by the indoor measures. In terms of the influence that this has upon epidemiological studies, it was suggested that the indoor concentrations should be considered to avoid misclassification of exposures.

The study reported by Lioy *et al.,* (1990), which was part of the Total Human Environmental Exposure Study, characterises the direct and indirect contribution of

outdoor PM_{10} to indoor air and personal exposures, and examines the factors that influence the actual magnitude of non-smoker personal exposures. The study was conducted in Phillipsburg (New Jersey) where the major point source was an iron pipe manufacturing company. Other sources include numerous residential, commercial and motor vehicle related area sources. Part of the study was to determine the influence that the point source had upon local PM_{10} and benzo(a)pyrene concentrations. It was discovered that on the days when the outdoor levels were >100 $\mu g/m^3$, as found on sampling days 4 and 8, the indoor/outdoor ratio was <1.0 for individual 11 and 10 of the 14 sampled participants, respectively. In general though, it was found that the personal concentrations exceeded the outdoor concentrations. The geometric mean values were 66, 42, and 48 $\mu g/m^3$ for the personal, indoor and outdoor environments, respectively.

An intensive personal monitoring study conducted in Waterbury, Vermont, involved 48 non-smokers, and used personal RSP samplers every other day for two weeks. The selection of volunteers was based upon willingness to participate and use of wood fuel as a primary or secondary heating source within their homes. The average personal exposure (36 $\mu g/m^3$) was always higher than outdoor values (17 $\mu g/m^3$) and also exceeded mean home levels (25 $\mu g/m^3$) for 43 of the 46 participants for which valid samples were available. Again, neither personal nor indoor RSP were strongly correlated with outdoor values. A relationship was observed, however, between personal exposure and in home concentrations ($r = 0.50$). Another factor that influenced personal exposure concentrations in this study was exposure to tobacco smoke. Those participants that were exposed to tobacco smoke for more than 2 hours a day had significantly greater RSP values, around 58% higher than non-tobacco smoke exposed participants. This study was conducted during the winter when it is assumed that the indoor concentrations are highest compared to summer when ventilation within homes is increased. This has been suggested as the reason why there is little correlation between the personal and outdoor values.

Research carried out at Harvard School of Public Health has attempted to identify the personal exposure of participants with COPD to PM_{10} and $PM_{2.5}$. The Harvard study sampled between 6 to 18 days during winter and summer. The personal exposure concentrations for PM_{10} and $PM_{2.5}$ for both seasons exceeded the ambient outdoor concentrations. The mean personal exposures during winter 1996 were 21 $\mu g/m^3$ for $PM_{2.5}$ and 43 $\mu g/m^3$ for PM_{10}. The summer values were 22 and 35 $\mu g/m^3$, respectively. Winter 1997 values were 22 and 38 $\mu g/m^3$ respectively. The outdoor concentrations for winter 96 were 12 and 18 $\mu g/m^3$ with summer concentrations of 18 and 26 $\mu g/m^3$, the final winter

concentrations were 11 and 19 $\mu g/m^3$. The correlation between the personal exposure concentrations and the outdoor ambient concentrations were low with a median r = 0.30. Simultaneous microenvironmental sampling of the home and garden using Harvard Impactors was undertaken (Rojas-Bracho *et al.* 1998).

A study of healthy senior citizens in Baltimore, MD. sampled over a 12 day period in both summer and winter showed higher correlations during the summer between personal $PM_{2.5}$ concentrations and ambient values Sarnat *et al.* (2000). Results indicated a median Spearman's r= 0.74 compared to winter where the value was r= 0.25, the major factor that influenced this relationship was ventilation of the participant's homes. There were no indoor measurements made in this study to determine the effects of indoor sources.

3.5.2 European Personal Exposure Studies

A recent study conducted in the Netherlands identified the relationship between personal and ambient PM_{10} for both adults and children (Janssen, 1998). The goals of the research were to:

i) evaluate the relationship between personal and ambient airborne particulate matter concentrations, within subjects, over time;

ii) evaluate potential differences between personal, indoor and ambient particulate matter concentrations.

The averaging time of the personal sampling was 24 hours with 4 to 8 measurements per participant conducted. The research compared the personal exposure of adults to PM_{10} with the ambient concentrations. The children's personal exposure to PM_{10} with the ambient concentrations was also completed. A small number of $PM_{2.5}$ measurements for the children were collected separately. The adults' personal exposure to PM_{10} exceeded both indoor and outdoor measurements with median values of 56, 35 and 42 $\mu g/m^3$, respectively. When the correlations between the personal and outdoor concentrations were adjusted to reject all measurements when exposure to ETS occurred, the median r increased from 0.5 to 0.81. The personal to indoor correlations also improved from 0.69 to 0.78. For the adult population living near a busy road, time spent in traffic and exposure to environmental tobacco smoke explained 75 % of the variance. Cleaning activities, cooking, time spent outdoors, ventilation and gender did not have a significant effect. For the non-ETS exposed adults, 50% of the daily variations in personal exposure could be explained by the ambient concentrations.

The children's personal exposure measurements were made during the weekdays only. Their personal exposures to PM_{10} were on average 67 $\mu g/m^3$ higher than the ambient concentrations. Measurements were also made within the schools and these had significantly higher concentrations than the ambient values. The school with the lowest concentrations also had the lowest personal exposure concentrations. When assessing the time weighted averages it was found that exposure to ETS and physical activity influenced their personal exposure concentrations. The correlation between the children's PM_{10} and ambient concentrations were 0.63 for non-ETS exposure and 0.59 for children with parental smoking. Parental smoking accounted for 35% of the children's exposure. The classroom exposure was the second most important cause of excess exposures and physical activity causing resuspension of particulate matter was the third. The children's exposure to $PM_{2.5}$ was closely related to the ambient concentrations, ETS being the most significant factor. The median correlation between the personal and ambient concentrations were 0.86 for all children and 0.92 for the children with non-smoking parents.

The largest European study currently being undertaken is the Air Pollution Exposure Distributions within Adult Urban Populations in Europe (EXPOLIS) as reported by Jantunen *et al.* (1998). The study was designed to assess the exposure distributions of target populations, determine the concentration distributions of the most important microenvironments, and assess the time activity distributions of target populations. The primary air pollutants under investigation are $PM_{2.5}$, carbon monoxide, and 30 Volatile Organic Compounds (Jantunen *et al.* 1998). Results of the selected populations sampled indicate that the selection procedure was unable to identify a random sample within all of the cities. The home and personal exposure data measurements will need to be corrected statistically to better represent the general populations of the cities or defined subgroups (Rotko *et al.*,2000).

Boudet *et al.*, (1997) measured total personal exposure of adults to fine particulate matter ($PM_{2.5}$). The personal exposure of urban adults to this size fraction is associated with ambient air and with traffic exhaust emissions. Preliminary results indicate that personal exposure to particulate matter over a 48 hour period is 104 $\mu g/m^3$ with 35 % of this mass being attributable to ambient air. The time spent outdoors amounts to only 11 % of the total time suggesting that traffic exhaust accounts for a large percentage of the total particulate mass.

3.5.3 Other Country's Personal Exposure Studies

Recently completed research undertaken in Toronto, Canada, had a primary goal of determining the distribution of air exposures to manganese in an urban population that uses the Methylcyclopentadienyl Manganese Tricarbonyl (MMT) additive in unleaded gasoline in automobiles (Pellizari *et al.* 1999). The study was based upon a similar design as the PTEAM study and was a large scale population-based exposure study. The design was to estimate three day average personal exposures to PM_{10} and $PM_{2.5}$. The personal exposures to particulate matter throughout the three days tended to be much higher than both the indoor and outdoor levels. The median for personal exposures to PM_{10} was 48.5 $\mu g/m^3$, indoor and outdoor medians being 23.1 and 23.6 $\mu g/m^3$, respectively. The median concentrations for the $PM_{2.5}$ fractions personal, indoor and outdoors were 18.7, 15.4 and 13.2 $\mu g/m^3$, respectively. The differences between the different environments were attributed to the presence of smokers. The correlations between the personal exposures and the outdoor fixed sites and roof sites were low (0.16 - 0.27). The highest correlation was found between the personal and indoor environment (0.56). This study also found that neither the roof nor fixed site concentrations can adequately predict personal particulate matter or manganese exposures.

Another Canadian study of COPD patients' exposure to particulate matter found that the mean personal and ambient exposure to $PM_{2.5}$ was 18 and 11 $\mu g/m^3$, respectively. The median correlation between the ambient and personal measurements was 0.48. When the tracer sulphate was used to identify the amount of personal exposure that can be predicted from ambient sources a median correlation of 0.96 was estimated, this suggests that using sulphate as a marker for outdoor combustion source particulate improves the models prediction of personal exposure (Ebelt *et al.,* 2000).

Research in South Africa to study the exposure and effects of indoor and outdoor air pollution on the health of children living in the Vaal Triangle is reported by Terblanche *et al.* (1992). The Vaal Triangle area is one of the most diverse regions for industrial development in Southern Africa. It has low level area source emissions (domestic coal burning) and is in close proximity to industries, which along with the topography and meteorology of the region make it a probable worst case scenario for South Africa. The median personal exposure concentrations of TSP for all children on schooldays and holidays were 310 and 298 $\mu g/m^3$, respectively. This exceeded the US 24-hour health standard of 150 $\mu g/m^3$ on 63% and 62% of the samples taken. The study went on to assess the health impacts of such exposures. Those children exposed to parental smoking had a

statistically significant higher prevalence of lower respiratory infections than those not exposed (25.7% v 20.8%).

A series of studies have been undertaken to assess the exposures of non-smokers to ETS and RSP. These have been undertaken in Stockholm, Barcelona, Turin, Paris, Bremen, Lisbon, Basel, Prague, Hong Kong and Kuala Lumpur. The studies have established RSP and ETS levels both at work and in all other locations, including the home. An attempt was made to randomly select the participants in each city for inclusion in a 24-hour exposure assessment. From the literature a variety of median concentrations were found and the majority of the highest exposures were evident in office workers who lived and worked with smokers. Housewives who lived with non-smokers had the lowest concentrations. The attempt to have a random selection of participants failed and tended to over-select predominant subgroups hence, trying to assume the wider population's exposure to RSP and ETS was not possible within acceptable levels of certainty (Phillips *et al.*1997, 1998a, 1998b, and 1999).

This study has attempted to incorporate aspects of all the reviewed study's methods to assess the major known sources of particulate matter for personal exposure. Further descriptions of the methodology are given in Section 4.2.

3.6 Personal Exposure Monitors

Personal monitors have to be worn within thirty centimetres of the breathing zone to ensure that the sampled air represents the air that the individual breathes (Health & Safety Executive, 1989). They also need to include a sizing mechanism and a media suitable for particulate collection. This enables both physical and chemical analysis of the particulates to be carried out. Rodes *et al.* (1991) indicated that the concentration levels of contaminants found within the breathing zone are affected by a number of factors. These factors have been found to include;

i) proximity to the source of the particulate matter,

ii) magnitude and direction of the convective air movements from the source and around the body,

iii) the character of the air turbulence within the breathing zone, and

iv) the presence of obstructions in the flow field.

To determine the concentrations of a specific pollutant a number of issues need to be addressed, including: why the sampling is being undertaken; when; where; how often and over what period of time samples are to be collected (Richtlinien, 1995).

3.6.1 Sampler Description

There are two forms of personal samplers available on the market, passive and active. The passive samplers rely upon molecular diffusion to deliver the analyte to the collection medium. These tend to be inexpensive, unobtrusive and, for large study groups, are easy to wear. However, such samplers usually require a long sampling period that does not give a suitable time resolution for the identification of peak pollution episodes.

Active samplers by comparison, tend to be more expensive, which results in smaller study groups. These samplers also tend to be more obtrusive, often restricting the study group's activities (Loth & Ashmore, 1994). The active samplers currently on the market collect the respirable, thoracic and inhalable fractions of particulate matter, as discussed below.

Respirable aerosol samplers collect particulate matter that can be inhaled into the human lung as far as the alveolar region (see Section 3.2). The review of personal samplers carried out by Kenny (1996a) discusses the different samplers available on the market and their efficiency. Small personal cyclone samplers are widely used for sampling respirable dust and give results that approximate to any of the occupational exposure conventions by operating them at appropriate flow rates. There is a drawback to using these samplers as the selection curves do not match all particle diameters, therefore causing significant sampling biases for some aerosol size distributions. Modifications to the cyclone geometry can correct this problem producing instruments with a much lower bias. New types of cyclone samplers that are a much better fit to the new convention being set by the European Standards Committee (CEN) can be constructed to operate at any desired flow rate. Cyclones with higher flow rates could be used for situations where analytical detection limits are a problem, such as the monitoring of respirable quartz (Kenny, 1996a). Disadvantages can be a reduction in the batteries life and an increase in the noise levels of the pumps.

There are few personal sampling instruments designed for the monitoring of particulates that penetrate into the human lung beyond the nasopharynx and pharynx regions (Holgate, 1995). This has been a neglected field in occupational hygiene and consequently there are no exposure limits yet for this fraction. The PM_{10} convention used for monitoring environmental air quality is similar, but is not an exact match to the CEN thoracic

convention. Commercially available personal samplers suited to this measurement include the CIP10-T which has been shown to match the CEN convention at a flow rate of 7 l m^{-1}. The problems with using this equipment in personal exposure studies, especially with children is the equipment is both expensive and cumbersome. The sampler selected for use in this study was the Personal Exposure Monitor (PEM, MSP Corporation, US) impactor which was originally designed to collect PM_{10} at a constant flow rate of 4 l min^{-1} on a 37 mm Teflon filter (Özkaynak *et al.* 1996). Recent modifications mean that a flow rate of 2 l min^{-1} is now used for the PM_{10} size fraction. The $PM_{2.5}$ size fraction requires a flow rate of 3 l min^{-1}. This means that both size fractions can now be collected simultaneously using a single pump. The weight is also reduced and hence, this is the sampler selected for use in this study.

The inhalable aerosol samplers require a suitable inlet to select the fraction of the aerosol that is capable of entering the nose and mouth during breathing. At present a large number of different personal samplers (and variations of these samplers) are used in different countries for sampling what is generally known as either 'total' or inhalable aerosol. The UK Health and Safety Executive developed the IOM inhalable sampler and when it was tested in laboratory conditions was found to slightly over sample compared with other European samplers. When their performance of a number of the samplers was tested it was found that they were within acceptable limits, particularly in the low external wind speeds thought to be most typical of indoor workplaces (Kenny, 1996b).

Given the small number of commercially available samplers for personal exposure sampling, it was decided that PEM would be the best option. The other samplers considered were primarily designed for use within the occupational environment where the particulate matter sources were well defined and external wind speeds were not an issue. The PEM samplers were robust and lightweight which is a significant factor when considering using children as participants. The pumps could also operate efficiently for the time period of 12 hours.

Ambient measurements were made using available comparable equipment. The Harvard Impactors have been shown to collect similar size fractions and concentrations to the PEM (Rojas Bracho *et al.*, 2000).

3.7 Sampling Methods

The use of personal monitoring to identify a specific population's exposure to particulate matter throughout their daily activities also requires information about their spatial and

temporal activity patterns. This information can be obtained through completion of questionnaires and time activity diaries. By monitoring background levels of pollutants within the home and workplace, it is possible to assess the sources of the pollutants that individuals are exposed to throughout their daily activities. By carrying out physical analysis of the particles it is possible to identify potential activities and sources that are responsible for an individual's exposure.

Time activity diary research has been used to assess personal exposure concentrations based upon time-weighted averages of individuals. These have been shown to underestimate personal exposure when compared to actual measurements as discussed in 3.5. Factors that decide the design of studies are primarily cost, accuracy and precision required. Questionnaires of activity data are useful for assessing a large number of individuals' exposure, this method is cheaper than direct personal exposure monitoring (Mage, 1991). Janssen (1998) found that the adults involved in the personal exposure sampling altered their behaviour when carrying the sampler. This was significant when compared to the days of non-sampling time activity diaries. The study did not find this to be the case for the children that were sampled in a similar way. This source of error has been identified in other research as the Hawthorne Effect and is most prominent in direct monitoring studies. By combining the direct and indirect methods a clearer indication of personal exposure can be made (Mage, 1991).

3.8 Summary

The research undertaken includes the collection of both PM_{10} and $PM_{2.5}$ for the children's personal exposure along with home, school and garden microenvironments to assess the potential variations in exposure patterns. Active sampling using PEM's and HI's was conducted, see Section 4.7

Other research has indicated that the exposure and health outcomes of individuals depend upon the environments that they frequent, the amount of time spent in each place, and their pre-existing health. This study incorporates the use of Time Activity Diaries for determining the children's location throughout the sampling period. No health measurements were made so no direct health outcomes are reported in this research.

Specific activities within the home affect the particulate matter concentrations, including cooking, cleaning, ventilation, heating, ETS and people moving around inside. To assess the impact that these activities have upon children's exposure patterns a daily household questionnaire was completed. A single air exchange measurement was also made to assess

how air tight each of the homes were. This will also influence the indoor/outdoor ratios for the transfer of particulate matter.

The health effects of exposure to particulate matter were reviewed for adults and children which indicated that there were significant potential health effects from exposure to particulate matter. The majority of these studies assessed exposure using outdoor ambient monitoring. Children's health effects have been shown to be largely related to decreases in lung function and lower respiratory diseases. There have not been enough studies to show that short term exposure to air pollution is directly responsible for causing ill-health among adults from exposure in childhood.

4 Materials and Methods

4.1 Introduction

This chapter describes the procedure for the selection of the children and a description of the sites. It also outlines the sampling methodology and techniques employed for the collection and analysis of the two particle size fractions for both the personal and microenvironmental sampling.

The sampling programme was designed to incorporate the objectives of the research as stated in Section 1.3. The objectives 1, 3 and 4 require the development of:

i) suitable methodologies for the sampling of children's personal exposure to particulate matter

ii) questionnaire and time activity diaries to determine the influence of confounding factors to children's exposure patterns

iii) analysis of the particulate matter to identify their source and composition.

4.2 Sampling Programme

Children were selected for inclusion in this personal exposure study to identify their exposure to particulate matter (PM_{10} and $PM_{2.5}$) during their normal daily activities. The sampling programme monitored each child's exposure over a five-day period, three school days and both weekend days, repeated on three separate occasions between January and September 1997 as recommended by Wallace (1996b, Pers. Comm.). Ten children were included in the study and sampled once per season with one child sampled per week. Research in South Africa of children's personal exposure to particulate matter, as described in Section 3.4, showed that the children's exposure patterns were not significantly different during the weekdays but were different at weekends (Terblanche *et al.* 1992). The children were sampled during the school week on Wednesdays to Fridays; any patterns in school day exposure should become evident. Exposure patterns over a weekend were assumed to be different for all children due to the variety of different activities that are carried out, hence both days were included in the sampling programme. One child was monitored during each 5-day sampling period each season.

The difference in children's seasonal exposure has been identified through a study in the Netherlands (Roemer *et al.,* 1993). By monitoring within different seasons it was anticipated that a representative sample of the children's exposure throughout the year

would be identified for a UK urban environment. The children were sampled during winter, spring and summer, equipment constraints prevented sampling being conducted throughout all four seasons. Many of the children also moved to different schools after September preventing their inclusion in further monitoring.

The date of the sampling was selected by the families so that it did not coincide with any inconvenient times, ensuring the families were not discouraged from completing three monitoring periods, see Table 4.1 for details of the sampling schedule. The inclusion of the same children for each of the sampling periods relates to the initial objectives for quantifying individual's personal exposure, (see Section 1.3). Previous research has highlighted the importance of undertaking repeated personal exposure assessments to particulate matter to provide sufficient data for correlations between personal and ambient particles within subjects over time. They also recognised the need to ensure compliance throughout the monitoring periods as personal measurements are labour intensive (Janssen, 1998).

Table 4.1 Sampling Schedule

Season	Winter										Spring										Summer									
Week	1	2	3	4	5	6	7	8	9	10	1	2	3	4	5	6	7	8	9	10	1	2	3	4	5	6	7	8	9	10
Participant																														
1	✓														✓	✓														
2	✓											✓															✓			
3		✓												✓							✓									
4			✓															✓											✓	
5				✓									✓												✓					
6[1]							✓													✓										
7[2]									✓																		✓			✓
8						✓					✓											✓								
9					✓												✓											✓		
10									✓										✓				✓	✓						

[1] Due to non compliance problems only two sampling sessions were completed, participant 10 sampled for an extra week.
[2] Scheduled spring appointment had to be postponed due to early arrival of family baby, participant 1 sampled instead.

Prior to the commencement of the sampling period all the children were issued with Time Activity Diaries to complete, (see Appendix 4 for a copy of the diary and also section 4.7.5 for further discussion of their design). This 'practice' diary ensured the children had no problems completing them when the sampling commenced. Mage (1991) and Janssen (1998) both recognised the fact that when carrying out personal exposure studies adults are likely to alter their behaviour patterns on measurement days, as this was not the case for children involved in personal monitoring this study did not investigate in depth compliance issues.

children involved in personal monitoring this study did not investigate in depth compliance issues.

4.3 Site Description

The children's homes were located in the London Borough of Barnet as illustrated in Figure 4.1. There are a number of point sources within Barnet and these are also shown on the map. Sources of air pollution (as discussed in Section 2.5) within Barnet include emissions from road transport, rail transport from diesel trains and prescribed processes (as defined under the Environmental Protection Act, 1990), such as crematoria, waste oil burners, vehicle resprayers, an adhesive coating process, one concrete crushing process and one process involving the blending and packing of concrete (Crabbe and Beaumont, 1998).

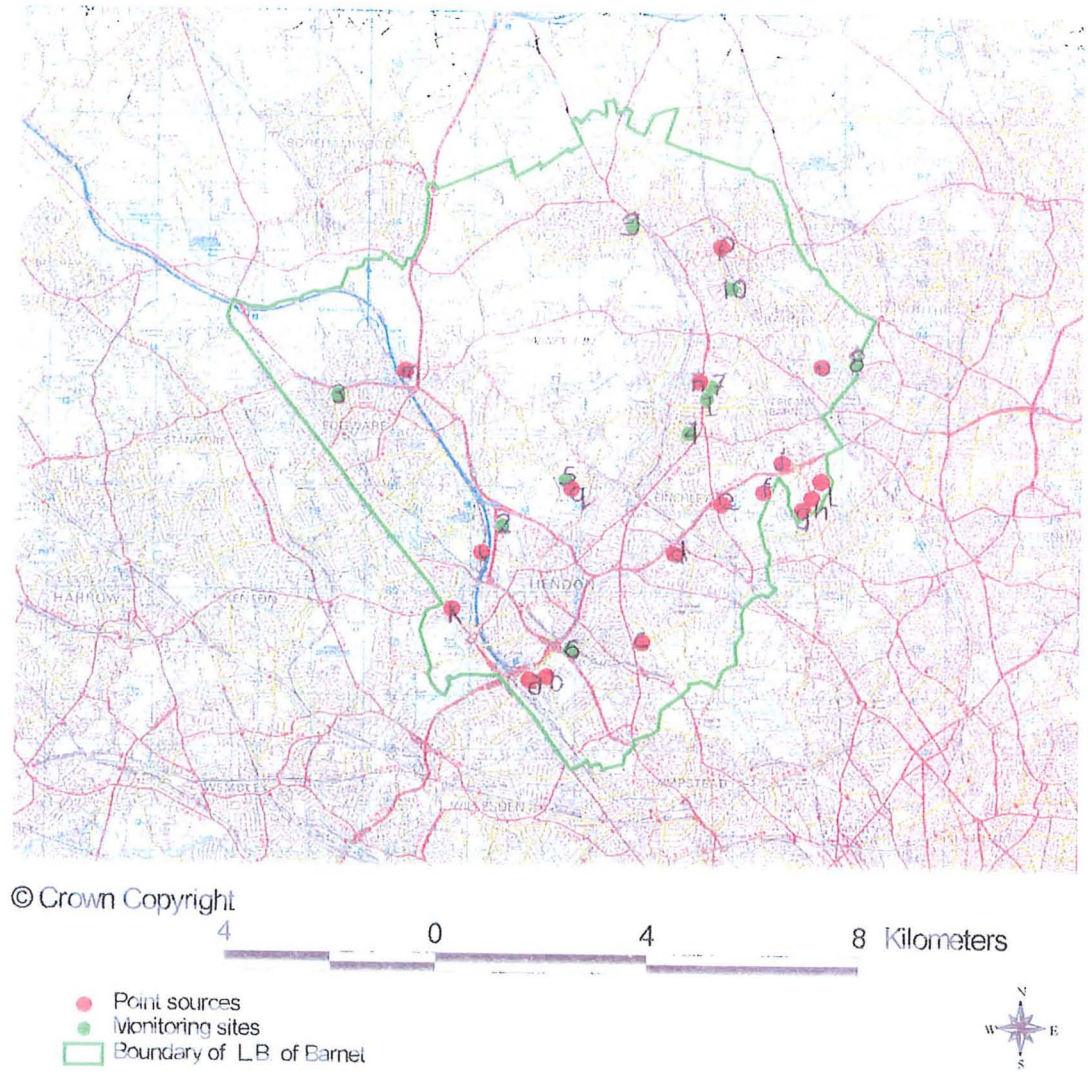


Figure 4.1 Locations of Participants in Relation to Point Sources within the London Borough of Barnet.

4.4 Selection Procedure of the Families

The selection of the ten children involved in this study was made using the questionnaire distributed in March 1996 by Barnet Health Authority (see Appendix 3 for a copy of the questionnaire). This questionnaire was issued to all schoolchildren aged 8-11 years old within the Barnet Health Authority region. The research aims were to identify any relationships between household characteristics and children's health. A number of questions relating to demographic issues, housing, child and family health were investigated. Based on the responses to this questionnaire and willingness to be included in further research, children were selected.

Research on indoor air quality in other countries have shown that cooking and ETS are major sources of indoor particulate matter (Özkaynak *et al.* 1996). These criteria were included in the selection of suitable families in an attempt to determine whether this would also be the case for homes in Barnet. Other selection criteria included the children being between 9 - 11 years old, as this age group are generally able to complete time activity diaries and are responsible enough to comply with the study requirements. The use of gas for cooking and heating was included as a requirement as research has indicated that this is an important source of particulate matter (Zartarian *et al.,* 1998). All the children attended primary schools where the majority of the lessons were conducted in one classroom. This was an important factor when considering the location of the microenvironmental monitors within the school, as described in Section 4.7.3. With all lessons being conducted in one classroom this ensured that the monitoring reflected the children's environment when analysing the potential source apportionment of the particulate matter.

Having requested families that fulfilled these criteria a search was undertaken using the database of collated information from the Barnet Health Authority questionnaire to identify suitable children. All data was coded so that only the child's name and school information was available. Head teachers were then approached for permission to contact the relevant families, meetings were set up to discuss the amount of involvement required from the schools and families. At this point a number of suitable children were rejected from the study due to the lack of co-operation from schools. Initially 37 children were identified as being suitable for inclusion, after contacting the schools and approaching the parents through telephone conversations 13 were considered to fall within the requested criteria.

Meetings with the families were then arranged. The equipment was demonstrated at these meetings to ensure a complete understanding of the involvement required from the families. At this stage it became evident that three families were unsuitable for inclusion, due to the lack of commitment or suitability of the houses for locating the equipment. The final ten families were then interviewed and suitable sampling dates arranged.

4.5 Details of the Families and Homes involved in the Monitoring Programme.

The final families that were selected included seven boys and three girls aged between nine and eleven years of age. A number of questions were raised during these initial meetings regarding suitability of the home and garden, availability of the family, and what sort of activities the child was involved in. Appendix 5 shows the content of the questionnaire. Any potential problems and questions were raised at this point. All the families were involved on a voluntary basis therefore it was necessary to ensure complete understanding

about the programme and willingness to participate throughout the three separate monitoring periods.

All head teachers were approached to determine the schools' willingness to be involved in the programme as there was also equipment that required daily access located in the child's classroom.

The different characteristics of the individual children and their homes are important as these may influence the sources of particulate matter. A number of studies have investigated what factors within homes influence particle concentrations; these have been reviewed in Chapter 2.

Of the ten families involved, three had residents that smoked cigarettes regularly and also permitted smoking within the home. Smoking in the home was not allowed in any of the other seven houses. All houses had gas hobs with three having electric ovens. The heating systems for all houses used gas as the fuel and had radiators in all rooms. Six of the houses had open gas fires that were used infrequently during cold weather spells, use of the gas fires was noted in the daily Household Activity Questionnaires, these are described in Section 4.7.6 and an example of one is in Appendix 6. It has been found that gas fires cause an increase in indoor particulate concentrations (Wallace, 1996a). A summary of the housing characteristics are displayed in Table 4.2.

The houses were different in design and had different types of windows. Three houses had double-glazing, four were single glazed and three had secondary glazing. Air exchange measurements were undertaken to identify the influence these may have upon the external to internal movement of air, and particulate matter, see Section 4.7.7 for the overview of this method and Appendix 10 for the methodology.

Three of the houses had pets that were permitted indoors. One house had a dog; another two cats and the other had a budgerigar. Five of the children walked to and from school every day, three travelled by car, one travelled by car to school then walked back home and one travelled into school by car and returned home by bus. Three of the houses were being decorated and having home improvement work carried out, although none had any work undertaken during the monitoring period.

Table 4.2 Characteristics of each participant and their home

Child Number	1	2	3	4	5	6	7	8	9	10
Sex (M/F)	M	F	M	M	M	M	M	F	F	M
Smoker in family	N	Y	N	Y	N	N	N	N	N	Y
Smoking allowed in home	N	Y	N	Y	N	N	N	N	N	Y
Gas Fire	Y	Y	N	N	N	Y	Y	Y	Y	Y
Pet at home	Dog	N	N	N	Cat	N	N	N	Bird	N
School travel mode	Walk	Walk	Car	Walk	Bus	Walk	Car	Car	Walk	Car
Window type[1]	D	S	Sec	S	S	S	D	D	S	S
Air Exchange Rates (ac/hr)	0.4	0.6	0.8	1.2	0.8	0.6	0.7	0.5	0.5	1.1

[1] D=Double glazing, S=Single glazing, Sec=Secondary glazing

In all the houses, the indoor monitors were located in the communal room used most frequently by the child under investigation. Generally, this was the living room although in two of the houses a second downstairs room was used. In the literature review in Section 3.5.1, the PTEAM study found that room to room variation within the selected houses when integrated over 12 hour collection periods was generally less than 10%. Resulting in the use of the main living room to collect the home sample (Wallace, 1996a).

The outdoor monitors were all located in the back garden of the houses; this was primarily a result of the availability of power sources and also to ensure the security of the equipment. In research conducted in the Netherlands by Roorda-Knape *et al.,* (1998) distance from motorways did change the concentration of both PM_{10} and $PM_{2.5}$ when the distance from the motorway increased from 15 metres to 115 metres. However, despite the concentration differences being significantly different they were small. Most of the homes that were sampled in Barnet were between 20 – 50 metres from major roads.

4.6 Study Design

During the day and night time monitoring periods a number of measurements were made. During the day personal PM_{10} and $PM_{2.5}$ concentrations were obtained for each child using the Personal Environmental Monitors (PEM) (see Figure 4.2 for the schematic design of the sampler). Harvard Impactors (HI, ADE, US) were used for the microenvironmental sampling, (see Figure 4.3 for the design of the sampler). The HI collected both size fractions for the indoor monitoring in the classroom, home and the ambient monitoring carried out in the garden. Sampling was carried out in the garden of each of the homes

throughout the day whilst the indoor samplers were set on a timer to be activated according to the child's presence. During the night time period no personal sampling was carried out, only the home and garden measurements were made, (Section 4.7.3 describes this method). It has been shown in other personal exposure studies that during the night personal exposure to particulate matter reflect home indoor concentrations (Özkaynak *et al.*, 1996).

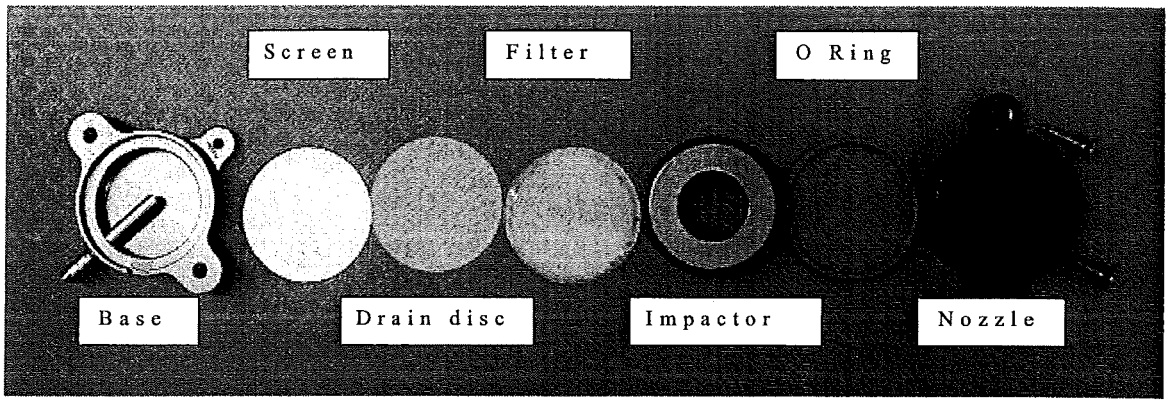


Figure 4.2 Schematic Diagram of Personal Exposure Monitors (MSP Corporation, US).

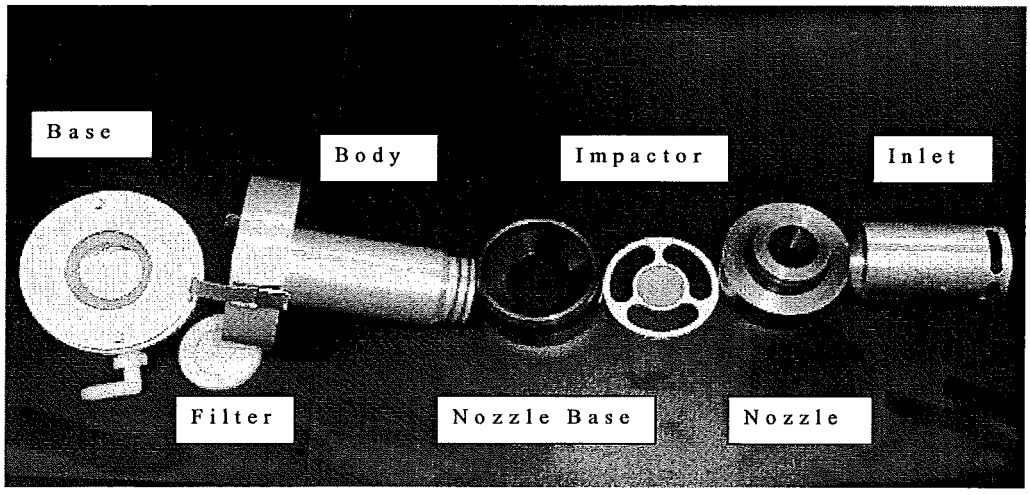


Figure 4.3 Schematic Diagram of Harvard Impactor (ADE, US)

4.7 Monitoring Methodology

This section describes the methodology used for the collection of particulate matter. The different equipment used and the data collection procedures are included. The objectives 1 and 3 as discussed in Section 1.3 are fulfilled with the development of the methods. The laboratory methods are found in Appendices 8 and 9.

4.7.1 Sampling Equipment

The equipment used for the personal monitoring was selected on the merits of a number of requirements as discussed in Section 3.6. As the study determined the exposure of children, it was felt that the primary requirement for the samplers should be that they were lightweight and robust. Other factors that had to be considered were the cost of the

samplers and pumps along with the running costs of filters and batteries. Many personal samplers are specifically designed for occupational exposure and this poses a problem for exposure assessment studies where the participants are exposed to a variety of microenvironments. The particulates can be a variety of sizes and external conditions of weather can influence the collection efficiency of some of the samplers, as discussed in Section 3.6.

It was decided that cyclone samplers would not be suitable for use within the study as they required a pump for each size fraction to be collected. This would be too heavy for the children to carry. Due to the weight and cost of the CIP10-T model of samplers it was felt that they would also be unsuitable. Inhalable samplers would not operate at the required collection efficiency as they depend upon low external wind speeds. The Personal Environmental Monitors (PEM) were selected as they had been used in other personal exposure studies, they are also lightweight and robust, further details in Section 4.7.2.

4.7.2 Personal Environmental Monitors (PEM)

The PEM collected the two cut sizes of PM_{10} and $PM_{2.5}$. Figure 4.2 shows the component parts of the samplers. These are small inertial impactors specifically designed for personal monitoring (Thomas *et al.,* 1993). The flow rate through each sampler was split to 2 litres per minute (l min^{-1}) and 3.2 l min^{-1} consecutively using a portable pump that ran at 5.2 l min^{-1} (Buck HF, Negretti Automation Ltd, Aylesbury, UK). At this flow rate the required cut point was obtained. The particles were collected onto 37 mm 2 μm pore size Teflon filters (Gelman R2PJ037, Gelman Sciences) which were placed downstream of a mineral oil-coated impactor plate. In addition, two 10 cm long elutriators were added to the inlets of both the PM_{10} and $PM_{2.5}$ PEM to minimise the particle collection from clothing.

The samplers were mounted on the left shoulder strap of a small rucksack weighing approximately 1.5 kg. The personal samplers were carried throughout the daytime monitoring period. Both parents and teachers supported and encouraged each child to ensure their compliance. Plate 4.1 shows a participant wearing the equipment.

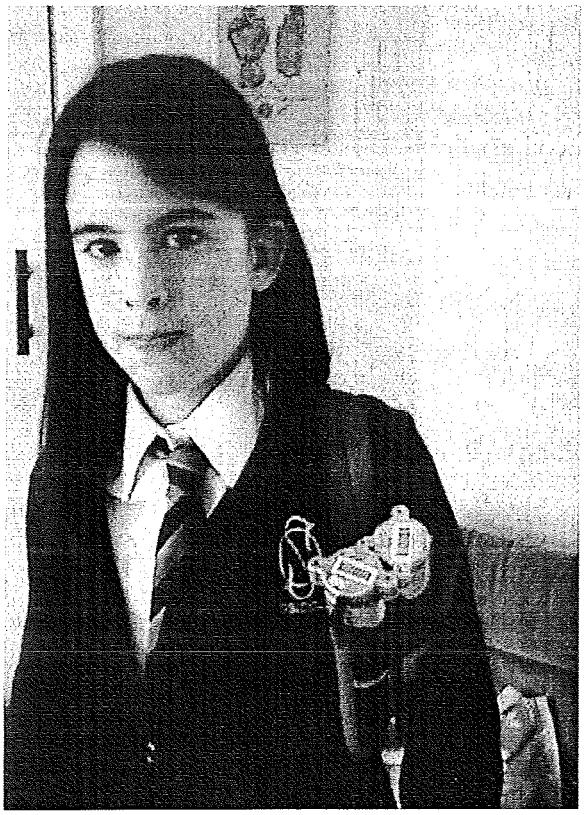


Plate 4.1 Example of participant wearing personal sampler

At the beginning and end of each 12-hour sampling period, all flow rates were measured using a calibrated rotameter. The total sampling volume and elapsed time were also measured using the digital readout from the pump. The pump was then switched off and the PEM's stored in resealable bags to avoid any further exposure and contamination when being transported to the clean room. The PEM's remained in the bags until they were disassembled and the filters removed for weighing.

4.7.3 Microenvironmental Monitoring

Harvard Impactors with cut sizes of PM_{10} and $PM_{2.5}$ were used for microenvironmental sampling (see Figure 4.3). These were located in the back garden of the houses, in a downstairs communal room most frequently used by the child, and in the classroom at school.

These operated at a flow rate of 10 l min^{-1} each. The garden HI's ran continuously throughout the daytime and night time monitoring periods. The indoor samplers were set using a timer to operate when the child was present. The HI's were changed in the morning and evening visits and the flow rates checked and noted at the beginning and end of each period with the calibrated rotameter. If the flow rate was out of the required range it was adjusted using a clamp system on the tubing.

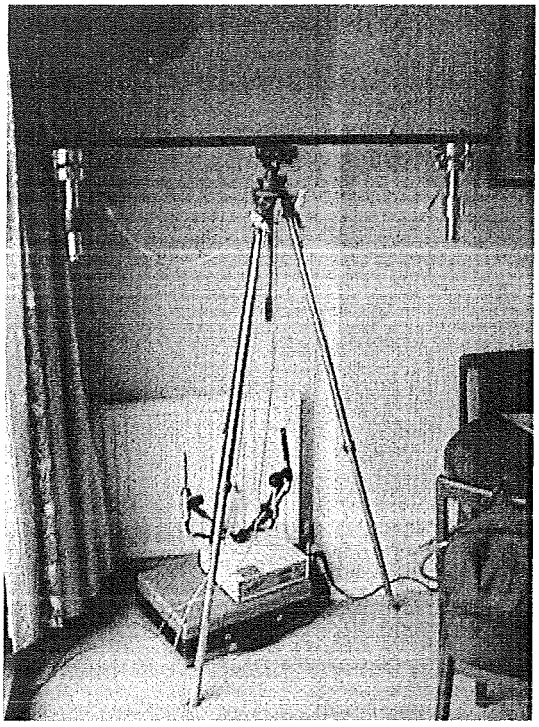


Plate 4.2 Example of Indoor HI setup at home during Summer Sampling.

Turner *et al.*, (2000) demonstrated the collection efficiency for both size fractions at 10 l min^{-1} with reproducible results. The $PM_{2.5}$ sampler showed a collection efficiency of 50.7% at 2.52 μm cut size. The PM_{10} had a 53.6% collection efficiency at 10.3μm cut size. This shows that the samplers are effective for the monitoring requirements of 50% collection efficiency at 2.5 and 10 μm as discussed in Section 3.6.1.

Rojas Bracho *et al.*, (2000) tested for any bias of these two methods and found that there was no significant bias in PEM measurements relative to the HI measurements.

Comparing the concentrations of the collected data with the AUN sites is slightly more problematic. The equipment used at the AUN sites are TEOM monitors which in a study by Smith *et al.*, (1996) found that during episode conditions an underestimation occurred which was ascribed to the standardised higher temperature sampling which leads to a loss of volatiles.

4.7.4 Ambient Conditions

The outdoor weather conditions of temperature, wind speed and direction were collected for every 5-minute interval using the Metlog Weather Station (R & D Electronics). Rainfall data from the Environment Agency site at the Mill Hill Golf Course was provided for 15-minute intervals. Indoor temperature, humidity and wind velocity was recorded at 5 minute

intervals sing a Casella 500 unit (Casella, UK). The maximum and minimum temperature and humidity values were recorded in the school using a hygrometer (Fischer, UK).

4.7.5 Time Activity Diaries

Each child completed a Time Activity Diary (TAD) every day that monitoring took place. (Appendix 4). The time intervals were 15 minutes each and had space for noting if any smoking had occurred close to them as recommended by Geyh, 1996 (Pers. Comm.). The diary was tested using the children of staff members at Middlesex University to ensure that it was simple enough to complete on the timescale required. Some revisions meant that in the relevant time interval the child's location and activity could be noted.

During the evening visits, the TAD was checked and any missing data discussed with the child and completed accordingly. This was carried out to ensure that the children had completed the diaries accurately. Discussing with the parents the times that the activities were carried out and then comparing with the school timetable ensured accuracy of the diaries. Interviewer bias was not considered to be an issue as the children completed the diaries themselves throughout the day and checking them in the evening was primarily to ensure that they were legible and accurate.

4.7.6 Household Activity Questionnaire

During the evening visits, a questionnaire was completed by the interviewer and the parents to identify any household activities that took place during the previous 24-hour period. Information about the type of housework that occurred was noted along with the duration of time that any cooking took place. Other information about the heating and ventilation, such as the duration of such events were also included. If any pets or smokers were in the house this was noted along with their location in the home. Section 2.6 reviews the relevant research and discusses the implications that these factors have upon air quality within the home. Any other activities were also recorded such as decorating and barbecues etc, (see Appendix 6 for a copy of the questionnaire). This data has been compiled into a database for analysis of the particulate matter concentrations and is analysed in Chapter 6.

4.7.7 Air Exchange Measurements

A method available for measuring air exchange rates is the pressurisation method also referred to as the 'blower door' technique (Infiltrator Series 900, Retrotec, UK). This is operated by placing an assembly in place of the conventional front door; as the blower input volume rate increases, pressure differential in a home is related to the cracks in the

building shell and ultimately, air infiltration rates. In addition, infrared scanners in the positive pressure mode can be used to locate major areas of leakage, such as the attic floor. Smoke sources near windows and electrical outlets will help to locate air leakage from within the home (Dietz & Cote, 1982). The fan is used to pressurise and depressurise the building. The flow of air through the fan is determined at a given pressure differential by comparing measurements to a calibration curve, a pressure versus flow curve can be determined by taking measurements at several fixed pressure differentials, e.g. from 10 Pascal's to 70 Pascal's, at 10 Pascal intervals. These data are then used to determine the effective leakage of the structure. Appendix 10 explains the methodology for this process. The air exchange rate of the individual houses will provide information for the analysis of the source apportionment of particulate matter. If the house has a high air exchange rate it is likely that the indoor particulate matter concentrations will relate closely to the ambient concentrations.

4.8 Analysis Methodology

This section describes the methodology used for the filter analysis. Objective 4, as discussed in Section 1.2, was developed to analyse the collected particulates. To determine the concentration of particulate matter collected on the filters, each one was weighed using a microbalance. A selection of the filters were also analysed using the Scanning Electron Microscope to identify the sources of the particulates using their physical characteristics.

4.8.1 Filter Weighing

The filters were all conditioned in clean room facilities with environmentally controlled temperature and humidity. All weighing was carried out using the Cahn-34 Microbalance (Avery Berkel, Birmingham, UK). The use of calibration weights, lab blanks and field blank filters ensured quality control and assurance.

The filters used were Teflon 2μm pore size, 37 and 41 mm diameter (Gelman R2PJ037 & R2PJ041, Gelman Sciences, UK). These were selected as the recommended filters for the PEM's and HI's.

All filters were conditioned for at least 24 hours at constant temperature and humidity in the clean room. Any variation in temperature greater than ±5°C around 20°C resulted in the filters being left for another 24 hours until constant conditions are reached. Humidity values of 40% ±5% were also required for 24 hours prior to weighing. This reduced the effects of static charge and moisture adsorption on the filters and collected particles. This

method is important when justifying the results as they ensure that the filters were not contaminated and that constant weighing took place. Appendix 7 shows the weighing protocol.

4.8.2 Scanning Electron Microscopy

A selection of 54 of the total filters sampled were analysed using the Scanning Electron Microscope (SEM, 240 Stereoscan, Cambridge Instruments, UK linked with a AN1085 EDX, Oxford Instruments, UK). This method is suitable for identifying the physical structure and sizing of the particles. The selection of these filters was determined by the availability of complete filter sets for all the environments sampled. This subset of filters had also been stored in the freezer to prevent any breakdown or loss of particulate matter after weighing.

A small section of the filter was cut and removed, then gold coated (SEM Coating Unit E5100, Polaron Equipment, UK) for 2 minutes onto a 12 mm stub (Agar Scientific, UK). The filters were magnified to identify the physical structure of the specific particulates. Analysis of the individual particles was conducted using the McCrone particle reference atlas (1973). The information obtained in this analysis was used to identify sources of particulate matter within the different sampled environments, as discussed in Section 6.4.

4.9 Summary

The methodology employed for the selection of suitable children for the personal exposure monitoring programme was successfully implemented according to the initial objectives as stated in Section 1.2.

The development of the time activity diaries and questionnaires were successful for identifying the likely confounding factors that could influence the concentration of particulate matter in the microenvironments that were sampled.

The final methodology did not incorporate any measurements of dose and health effects, as the final sample size was not representative of the general population, it was felt that this made any assumptions of health effects inconclusive.

The methodology developed has identified children's personal exposure during an integrated 12 hour daytime period. It has also successfully incorporated

microenvironmental monitoring to determine potential sources of the children's personal exposure.

A suitable sampler was used to conduct the personal monitoring that was small, lightweight, robust and proven in the field of personal exposure research.

5 Statistical Interpretation of Particulate Matter Data

5.1 Introduction

This chapter highlights all of the monitoring data statistics. The descriptive data for the annual, seasonal and individual microenvironments are presented with interpretation. The Time Activity Diaries (TAD) are also analysed and interpreted in terms of the amount of time that the children spent in specific environments throughout the different seasons. Estimations of personal exposure using the time activity data and microenvironmental data were attempted using time weighted averaging models.

Where the descriptive statistics indicate associations then correlation analyses and Wilcoxon Signed Rank Tests, were conducted. Where possible, investigations of associations between variables were completed and determination of which of these associations required further analyses. Where no evident associations occur then further analysis was not undertaken.

The ratios between $PM_{2.5}$ and PM_{10} have also been investigated and compared to the literature to assess the comparability of the results to other studies. All mean values quoted are geometric means as the data was not normally distributed as indicated in Section 5.3. Similarly, the geometric standard deviations have also been used and have the same units as the corresponding geometric means; these range between 1 – 4.

All data analysis was conducted using SPSS vers. 7.5 (Noruses, 1993) and SAS vers. 8.0.

5.2 Data Quality Assurance

5.2.1 Filter Blanks

The filters used for collecting particulate matter were quality assured using two types of blank filters. Laboratory blanks identified errors associated with the microbalance and weighing protocol. Field blanks identified errors associated with loading filters into the sampling equipment and consequent transportation. Any contamination of the blanks could also occur in field samples. Corrections were made to field sample data using these blanks.

Where the laboratory blank mean masses were significantly different from zero, as determined using a Student T-Test, then the corresponding field blank values were

corrected. This was only found in the case of the personal $PM_{2.5}$ filters. Negative values associated with lab blanks could be attributed to off gassing of the filters over time, this can occur when the filters have not been equilibrated in the constant temperature and humidity settings for at least a month prior to use. Where the field blanks were significantly different from zero, using the Student T-Test, these were used to correct the corresponding field sample weights, i.e. all field samples for the personal $PM_{2.5}$ monitoring were corrected for the field blanks by subtracting 5.37μg/m^3 from each field sample.

Damaged filters include those weighed using the London Borough of Greenwich Department of Environment microbalance. The location of their balance caused it to be unstable, the low masses collected in this study requires a balance that does not fluctuate by ±10μg. The on and off weights for the filters were consequently unstable and therefore considered unreliable. A result of this is the loss of many of the HI $PM_{2.5}$ and PM_{10} filters.

Where the standard deviation for the blanks is higher than the mean value this indicates that there is variability above and below the mean value. Large values indicate that there are some outlier or extreme values, where there does not appear to be a reasonable explanation to void the filter then they have been included.

The PEM's have previously been tested for accuracy and precision with results indicating a precision of ± 1.99μg/m^3 for both size fractions. The accuracy of the PEM's in comparison with collocated TEOM indicated an overestimation by the $PM_{2.5}$ PEM of 8% and 20% for PM_{10} PEM. Reasons for this difference point to the loss of semi volatile organics from the TEOM (Williams *et al.*, 2000). There is no significant bias in PEM measurements relative to HI measurements, using the mean relative difference between collocated PEM-HI pairs (Rojas-Bracho *et al.*, 2000).

Table 5.1 Blank Filter Data

Filter Type	*Laboratory Blank*			*Field Blank*			*Samples*	*Damaged*	*Flow Problems*
	N	*Mean (μg/m^3)*	*SD*	*n*	*Mean (μg/m^3)*	*SD*	*n*	*n*	*N*
PEM[1] $PM_{2.5}$	51	-1.03	±3.03	15	5.37	±3.48	150	4	17
PEM[2] PM_{10}	54	-0.82	±3.33	15	6.22	±4.36	146	8	0
HI[3,4] $PM_{2.5}$	127	-0.22	±7.40	85	1.81	±1.62	518	112	53
HI[5] PM_{10}	121	0.96	±8.01				526	90	70

[1] Personal Environmental Monitor Blank Filter Data for $PM_{2.5}$ Fraction
[2] Personal Environmental Monitor Blank Filter Data for PM_{10} Fraction
[3] Harvard Impactor Monitor Blank Filter Data for $PM_{2.5}$ Fraction
[4] Field Blank Data for both Size Fractions Totalled
[5] Harvard Impactor Monitor Blank Filter Data for PM_{10} Fraction

5.2.2 Levels of Detection (LOD)

Having determined the blank values for the different filter types, the level of detection for the samples were assessed. The normal analysis of the level of detection is 3 x Standard Deviation (SD) divided by the corresponding sampling volume for each sample type with all values that exceed this being acceptable for using in further analysis (Özkaynak *et al.* 1996 and Janssen, 1998).

The data in Table 5.1 show that the SD for the personal $PM_{2.5}$ blank field filters is 3.48 multiplied by 3, giving a LOD of 10.44 $\mu g/m^3$; twenty-four samples fell below this value. The PEM PM_{10} LOD being 13.08 $\mu g/m^3$; three samples did not exceed this LOD. The HI LOD for $PM_{2.5}$ and PM_{10} were 5.1 $\mu g/m^3$; thirty-two samples from the different microenvironments did not exceed this LOD. The LOD values are comparable to those found by Rojas-Bracho using the same methodology (2000). The samples that do not exceed the LOD were assigned half the LOD and all analysis was conducted using these values (Phillips *et al.* 1997 & 1999).

5.3 Overview of Descriptive Statistics

Of the 150 days personal monitoring measurements that were attempted 23% of the PM_{10} size fraction and 16% of the $PM_{2.5}$ size fraction were invalid. Around half of these invalid measurements were due to flow problems, 17% due to pump problems (such as power loss) and 32% due to Microbalance problems, primarily at Greenwich.

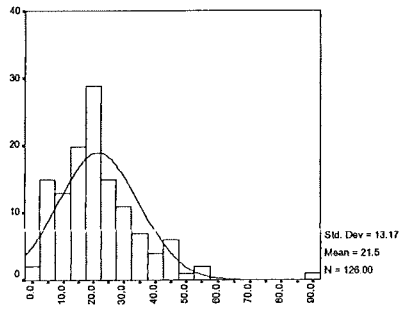
For the $PM_{2.5}$ microenvironmental sampling during the daytime within the home, classroom and garden 16%, 10% and 19% were invalid. The corresponding invalid PM_{10} measurements were 20%, 11% and 27%, respectively. Invalid night time measurements for $PM_{2.5}$ in the home and garden were 12% and 11% with the corresponding invalid PM_{10} measurements being 20% and 19%. Of these losses encountered the Microbalance problems accounted for about 34% and the flow rate of the pumps for about 66%.

Missing data resulted in 70 $PM_{2.5}$ observations where all data is valid and 69 for PM_{10} out of a possible 150. The majority of the missing data occurred during the winter sampling period with only 12 of the 50 observations being completed for $PM_{2.5}$ and 15 PM_{10}. The Greenwich microbalance problem was responsible for the majority of these losses, most of the missing data are the home and garden filters, only 5 personal $PM_{2.5}$ samples and 12 of the personal PM_{10} observations are missing for the winter sampling period.

To assess the appropriate analytical procedures for the data, a number of preliminary tests were undertaken. The normality of all the data, using all observations in all seasons, was assessed and the distribution and normality curves plotted. The y-axis represents the frequency distribution. Figure 5.1 a - j and Figure 5.2 a - f indicate that the data are skewed. According to McBean & Rovers (1998) for data that do not fulfil the necessary assumptions for the parametric analyses, the non-parametric methods are as powerful as or more powerful than the equivalent parametric tests. It is acknowledged that non-parametric tests may be wasteful of information and usually they have a smaller efficiency than the corresponding parametric methods, provided that the assumptions of the standard (parametric) methods can be met. Sarnat *et al.*, (2000) showed similar results for a personal exposure study in Baltimore, MD using the same sampling equipment where the data were not normally distributed and non-parametric analyses were consequently conducted.

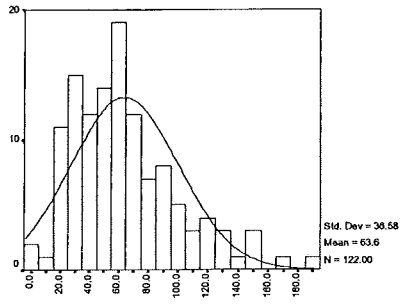
All of the PM_{10} concentrations exceed the $PM_{2.5}$ concentrations. This is as expected as the PM_{10} samplers are designed to also collect the $PM_{2.5}$ fraction.

Where outliers are apparent in the frequency distribution plots these values were further investigated using the time activity diaries and questionnaire data that applied to that specific value. Any significant activities where particulate matter was likely to have been generated has been analysed further in Section 6.3. Most of the outliers for the personal exposure concentrations had corresponding home or school values. Only a very few of these outliers could not be explained by corresponding microenvironmental concentrations and in these cases the household questionnaire and time activity diaries were analysed in an attempt to identify any activities or environments where the child may have been exposed but there was no particulate monitoring data available.



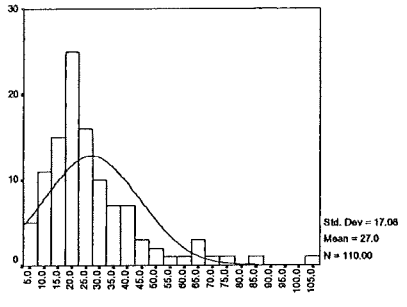
Child's PM2.5 Concentrations

a)



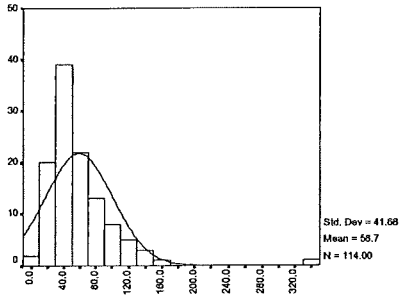
Child's PM10 Concentrations

b)



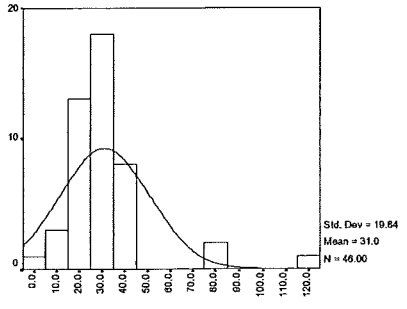
Home PM2.5 Day Concentrations

c)



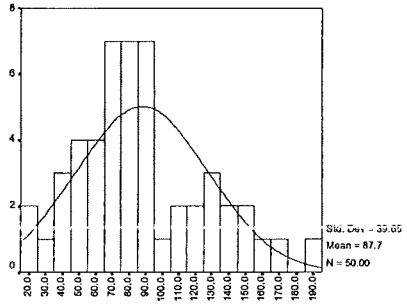
Home PM10 Day Concentrations

d)



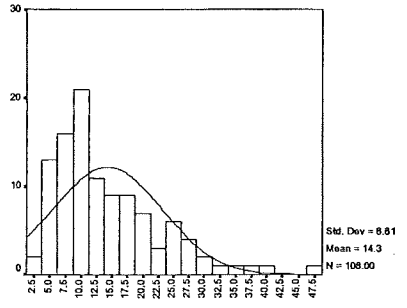
School PM2.5 Day Concentrations

e)



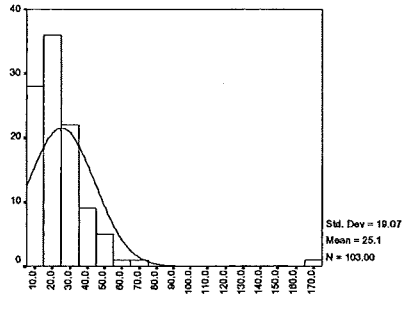
School PM10 Day Concentrations

f)



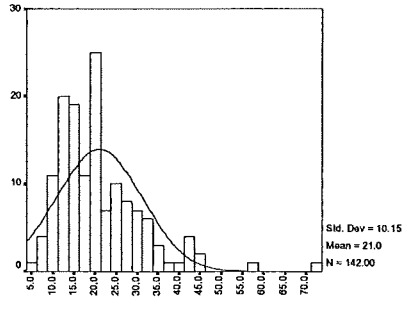
Garden PM2.5 Day Concentrations

g)



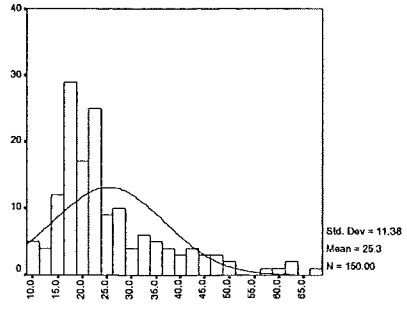
Garden PM10 Day Concentrations

h)



Brent AUN PM10 Day Concentrations

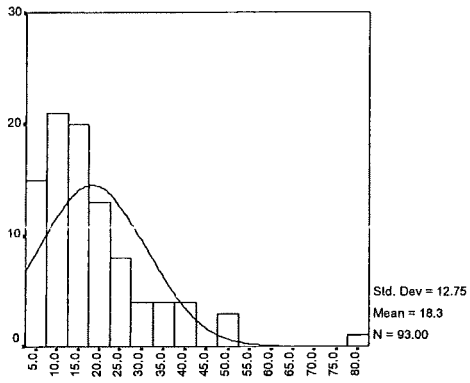
i)



Haringey AUN PM10 Day Concentrations

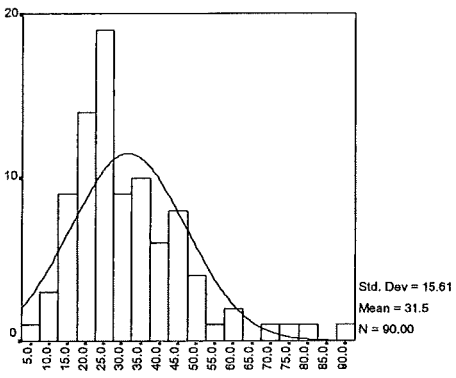
j)

Figure 5.1 a-j) Normality Plots of Day time Microenvironments & AUN sampled ($\mu g/m^3$). (Frequency is plotted on the y axis)



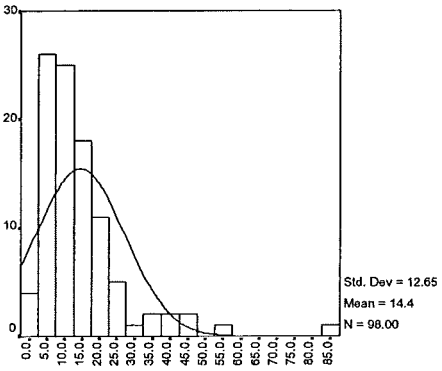
Home PM2.5 Night Concentrations

a)



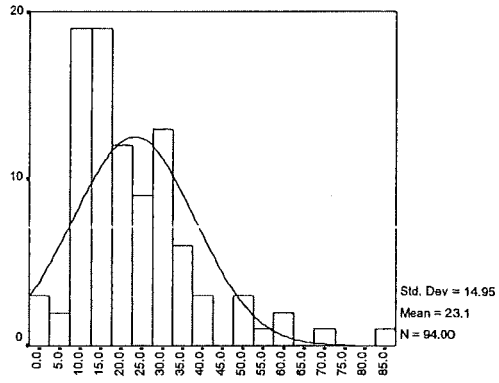
Home PM10 Night Concentrations

b)



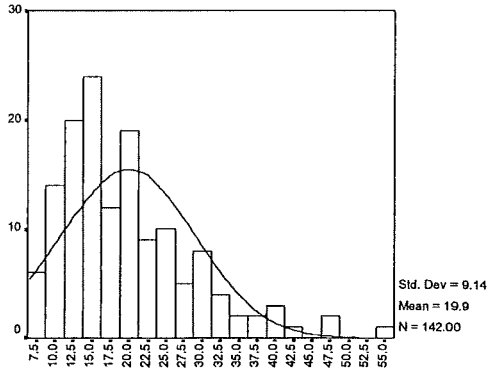
Garden PM2.5 Night Concentrations

c)



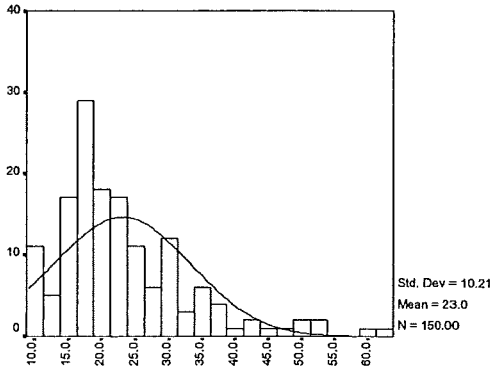
Garden PM10 Night Concentrations

d)



Brent AUN PM10 Night Concentrations

e)



Haringey AUN PM10 Night Concentrations

f)

Figure 5.2 a-f) Normality Plots of all Night time Microenvironments & AUN sampled ($\mu g/m^3$). (Frequency is plotted on the y axis)

5.4 Personal – Outdoor Relationships

A summary of the average personal exposure concentrations for all children in each season is presented in Table 5.2 indicating whether it was a school day or not. This shows that nearly all mean personal concentrations for the seasons were winter > spring > summer, suggesting that the children's exposure to both PM_{10} and $PM_{2.5}$ were seasonally influenced. The only difference was found for the school day $PM_{2.5}$ concentrations which were winter > summer > spring, although the actual values were all $20\mu g/m^3 \pm 3$ suggesting that there was little variability between the seasons.

The percentage of daily personal exposure concentrations that exceeded the equivalent garden outdoor concentrations in each season is also indicated in Table 5.2. Personal PM_{10} concentrations when the children were at school showed the highest percentage of days where ambient concentrations were exceeded. During non school days, both PM_{10} and $PM_{2.5}$ had the lowest number of days when personal concentrations exceeded ambient concentrations. This could be a reflection of the children's exposure from the classroom along with their activity patterns. The summer season non school days show the lowest percentage of personal concentrations exceeding ambient for both particle sizes, this could be a result of having open windows and doors during the summertime, this is investigated further in Section 6.3.3.

The data for each of the three seasons show that the geometric means for each child's personal PM10 concentrations were greater than both the outdoor AUN and garden sites, this is in agreement with the literature reviewed in Section 3.5. In response to objective 6, as stated in Chapter 1, analysis of the AUN site data was undertaken and if used to directly predict the personal exposure of children to PM_{10} over the corresponding 12-hour period, it would underestimate their exposure. Section 6.5 analyses this further.

The personal $PM_{2.5}$ concentrations also exceed the outside $PM_{2.5}$ concentrations, again suggesting that the outdoor ambient sites would under predict the personal exposure of children living in an urban environment. This disagrees with the study undertaken in the Netherlands by Janssen (1998) and hence further analysis of the data will be undertaken, see Section 5.8.

Table 5.2 Summary of Daytime Personal Concentrations in each Season.

	Winter				Spring				Summer			
	PM_{10}		$PM_{2.5}$		PM_{10}		$PM_{2.5}$		PM_{10}		$PM_{2.5}$	
Day[1]	S/D	NS/D	S/D	NS/D	S/D	NS/D	S/D	NS/D	S/D	NS/D	S/D	NS/D
N	18	28	21	28	22	24	19	24	8	38	7	38
geometric mean ($\mu g/m^3$)	89.7	54.6	23.8	18.3	79.7	50.1	17.5	17.1	65.9	28.9	20.5	13.3
geometric Std. Dev. ($\mu g/m^3$)	1.5	2	2.2	2.2	1.5	1.8	2.2	1.6	1.4	1.6	1.5	1.9
Max. value ($\mu g/m^3$)	171	139.9	91	53.6	193	116.7	50	36.1	92	63.4	38	40.9
Ambient geometric mean ($\mu g/m^3$)	27.9	20.7	9.4	14.2	19.6	19.5	11.1	10.5	21.9	20.4	10	11.8
Ambient geometric Std. Dev. ($\mu g/m^3$)	2.6	1.7	2.3	2.3	1.7	1.6	2.3	1.9	1.7	1.8	2.3	2.3
% > Ambient Value[2]	94	75	86	69	100	91	75	76	100	58	86	45

[1] S/D =School Day and NS/D = Non School Day
[2] Ambient value used is Garden unless data is void then AUN site is used

The data shown in Tables 5.3 - 5.5 indicate the daytime geometric means and ranges for each child's exposure concentrations for winter, spring and summer. The means for all 3 seasons are included in Table 5.5. The day concentrations represent all samples collected during the daytime, usually 7.30 until 19.30. The night time concentrations refer to the 19.30 until 7.30 samples collected overnight. These data are shown in Tables 5.6 – 5.8 with the 3 season's summary included in Table 5.8. The range of each child's data for the individual seasons and microenvironments sampled are also included to show the variability in the collected data, the values fall within the reviewed literature and are lower than those where a predominant point source is present.

The range of personal exposure data shows that there was great variability within and between each child. The 5 day means ranged from 5 – 49 $\mu g/m^3$ for $PM_{2.5}$ and 21 – 115 $\mu g/m^3$ for PM_{10}. The greatest range was found for PM_{10} which would suggest that personal activities could be responsible for this effect upon exposure. The breakdown into school and non school days in Table 5.2 suggests that much of this variation can be explained by

the higher exposures on school days compared to non school days. Section 5.10 investigates the seasonal effects upon children's activity patterns.

In Tables 5.3 – 5.8 NS indicates that no sampling occurred for the period indicated, either due to holidays so no sampling was carried out at the school or there were compliance problems. The problems indicated in Section 5.2 are included in the NS statement. When comparing the number of successfully collected data points with the studies in the Netherlands and Boston this study had a slightly lower success rate (Janssen, 1998 and Rojas-Bracho *et al.*, 2000). An example of low data collection is illustrated in the indoor / outdoor collection of PM_{10} and $PM_{2.5}$ in Bangkok, Thailand where only 54% of the original data set was viable for analysis (Tsai *et al.*, 2000). The majority of the problems resulted from the pump flow and microbalance issues previously mentioned in Section 5.2. When the children were not at school 12-hour daytime measurements were made in the home.

5.5 Indoor – Outdoor Relationships

The indoor concentrations of PM_{10} from the homes and classrooms both exceed the outdoor ambient measurements as can be seen in Tables 5.3 – 5.8. The classroom concentrations were consistently the highest of all the environments sampled throughout the three seasons, this agrees with the study undertaken by Janssen (1998).

The indoor concentrations of $PM_{2.5}$ from the homes and classrooms exceeded the personal and garden $PM_{2.5}$ concentrations in all seasons. The study by Janssen (1998) showed that the $PM_{2.5}$ concentrations within the classroom were correlated to ambient concentrations so further investigation of this data will be undertaken in section 5.8.1.

The seasonal 5 day mean home PM_{10} and $PM_{2.5}$ concentration values range from 28 - 79 $\mu g/m^3$ and 17 - 31 $\mu g/m^3$ respectively. On some of the days sampled the home environment values exceed the Norwegian legislation set for indoor air quality, however when averaged over the usual 5 days of sampling this lowers the mean, see Section 2.4. The summer values for indoor PM_{10} and $PM_{2.5}$ tend to be lower which again may be indicative of the ventilation rates being higher than in the winter or spring.

The seasonal 3 day mean classroom PM_{10} and $PM_{2.5}$ concentrations for the children range between 58 - 93 $\mu g/m^3$ and 19 - 43 $\mu g/m^3$ respectively; these were higher concentrations than those found within the homes and it may be possible that this was the major source of exposure for all of the children. It would have been useful to establish where the

particulates were derived from using further analysis of the elemental particulate composition such as x-ray fluorescence (XRF), however this was not within the scope of this research. A review of the published literature suggests that the majority of the particulates in classrooms are soil related, the other factor influencing the particulate concentrations are the number of active children present (Janssen, 1998, Ashmore, 1999, Pers. Comm.).

The 5-day mean PM_{10} home concentrations at night exhibited less seasonal variation than during the day, ranging from 20 - 45 $\mu g/m^3$. At night the home concentrations were lower suggesting that activities within the home were reduced. Analysis of the questionnaire data will provide information about when specific activities were conducted within the homes (see Section 6.3).

At night, the home $PM_{2.5}$ concentrations appear to reflect those of the garden except for children 2, 4 and 10 where a parent was a smoker. The significance of Environmental Tobacco Smoke (ETS) upon indoor air quality has been cited in a number of studies (Janssen, 1998, Santanam *et al.*, 1990, Sheldon, 1988, Spengler *et al.*, 1981).

The data for the night concentrations, shown in Tables 5.6 - 5.8, indicate that the home indoor concentrations for both $PM_{2.5}$ and PM_{10} were around 30% lower than during the day whilst the outdoor concentrations do not appear to differ greatly. Possible suggestions for this could be that particle generating activities and resuspension of particles indoors predominantly occurred during the day, as stated in the literature review of data from indoor air quality studies, Section 2.6. Further assessment of the source apportionment of the particulate matter will be undertaken in Chapter 6.

Table 5.3(a) Winter Mean Day Concentrations of all Microenvironments for all Children, $PM_{2.5}$ ($\mu g/m^3$)

Child ID No.	Home			Garden			Class			Personal		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	4	39	22-67	5	9	3-25	2	29	21-40	4	18	5-46
2	4	37	22-68	3	23	17-37	3	37	33-45	5	25	5-45
3	3	24	23-42	4	12	10-16	NS	NS	NS	4	30	19-54
4	2	51	43-60	NS	NS	NS	2	30	26-34	5	18	5-28
5	NS	NS	NS	NS	NS	NS	NS	NS	NS	4	22	7-38
6	3	14	8-30	3	14	8-21	2	20	15-25	5	11	5-37
7	NS	NS	NS	1	49	49	3	63	25-125	4	30	20-44
8	NS	NS	NS	1	3	3	3	11	3-24	5	15	5-24
9	3	26	17-52	3	13	10-19	2	52	36-77	4	49	27-91
10	3	24	18-42	3	10	7-14	NS	NS	NS	4	16	5-44
mean		*29*			*12*			*29*			*21*	
std.dev.		*1.8*			*2.1*			*2.5*			*1.9*	

Table 5.3(b) PM_{10}.

Child ID No.	Home			Garden			Class			Personal			Brent AUN			Haringey AUN		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	3	98	93-102	5	19	8-43	2	77	68-87	5	100	63-140	5	18	9-34	5	20	9-46
2	4	64	44-102	3	80	43-171	3	114	62-155	5	75	55-115	5	29	18-39	5	34	25-49
3	4	79	70-85	4	20	18-23	NS	NS	NS	4	68	55-120	5	18	15-21	5	22	19-27
4	2	105	76-145	1	30	30	1	93	93	4	115	59-171	5	20	14-28	5	23	16-32
5	NS	NS	NS	NS	NS	NS	NS	NS	NS	3	67	49-98	5	31	21-72	5	32	17-68
6	4	57	29-133	3	25	17-34	3	55	28-124	5	48	17-128	5	18	13-29	5	21	17-25
7	4	31	9-89	3	18	6-32	3	52	22-86	1	105	105	3	25	22-28	5	36	28-47
8	3	46	27-72	2	12	7-21	3	94	78-117	5	75	54-103	5	18	10-27	5	18	9-30
9	3	31	23-39	NS	NS	NS	3	94	69-131	2	113	98-129	5	26	21-46	5	26	18-59
10	3	28	21-34	4	28	20-57	NS	NS	NS	4	22	7-53	NS	NS	NS	5	25	17-42
mean		*52*			*24*			*79*			*69*			*21*			*24*	
std.dev.		*1.9*			*2.0*			*1.7*			*1.8*			*1.5*			*1.5*	

Table 5.4 (a) Spring Mean Day Concentrations of all Microenvironments for all Children, $PM_{2.5}$ ($\mu g/m^3$)

Child ID No.	Home			Garden			Class			Personal		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	10	28	17-86	10	10	5-16	2	27	27	8	20	13-29
2	3	30	12-56	4	13	7-21	1	26	26	5	17	5-28
3	4	34	32-36	4	28	22-34	2	32	32-33	5	14	5-50
4	5	19	6-33	5	9	3-20	2	29	20-41	5	20	17-24
5	5	13	11-18	5	7	5-10	3	23	20-28	4	5	5
6	4	20	15-27	5	13	7-27	2	33	32-34	1	19	19
7	5	19	15-21	5	15	8-34	NS	NS	NS	5	11	5-32
8	4	24	22-31	3	9	8-11	2	30	25-36	3	23	17-32
9	5	20	10-45	4	13	9-26	3	30	20-41	4	27	17-36
10	4	43	20-103	4	9	7-12	3	22	17-36	4	25	16-41
mean		*23*			*11*			*27*			*24*	
std.dev.		*1.8*			*1.7*			*1.3*			*1.7*	

Table 5.4(b) PM_{10}.

Child ID No.	Home			Garden			Class			Personal			Brent AUN			Haringey AUN		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	9	73	48-127	9	20	15-31	2	94	80-111	10	71	42-117	10	18	14-31	10	22	17-34
2	2	46	38-54	3	27	16-39	2	58	51-65	5	35	7-58	5	20	8-34	5	27	13-38
3	5	78	34-136	4	37	22-49	2	124	114-135	5	77	46-153	5	25	17-33	5	27	18-42
4	5	44	11-83	4	17	12-22	3	80	75-90	5	63	44-94	5	16	9-23	5	22	14-26
5	5	41	31-49	4	14	10-24	3	73	58-84	5	45	29-67	5	15	13-19	5	16	13-22
6	4	49	32-76	5	18	11-36	2	88	46-169	1	38	38	5	13	9-21	5	20	14-26
7	2	48	47-49	3	16	12-24	NS	NS	NS	3	21	16-31	5	18	12-30	5	19	17-27
8	3	56	48-65	2	21	17-26	3	69	24-135	4	75	48-193	5	15	12-21	5	19	12-27
9	5	27	19-54	5	21	14-49	3	51	35-62	5	83	64-96	5	22	12-44	5	26	18-42
10	5	70	38-157	4	12	6-27	3	83	40-159	5	75	37-149	5	13	9-20	5	19	17-27
mean		*56*			*20*			*76*			*69*			*17*			*22*	
std.dev.		*1.9*			*1.6*			*1.6*			*1.6*			*1.5*			*1.4*	

Table 5.5(a)Summer Mean Day Concentrations of all Microenvironments for all Children, $PM_{2.5}$ ($\mu g/m^3$).

Child ID No.	Home			Garden			Class			Personal		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
2	4	24	18-28	3	26	17-40	NS	NS	NS	5	14	11-18
3	5	26	14-43	5	9	3-23	2	11	9-15	3	11	5-19
4	4	11	3-26	4	3	3	NS	NS	NS	3	13	5-28
5	5	26	17-40	5	23	17-31	NS	NS	NS	5	24	18-41
6	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
7	3	20	12-41	3	7	3-19	2	20	15-27	4	12	5-20
8	4	18	11-24	5	16	10-28	2	28	23-34	5	17	11-38
9	5	13	3-41	3	12	7-25	NS	NS	NS	4	16	5-29
10	9	18	3-64	8	10	3-26	3	23	12-44	7	13	5-28
mean		*19*			*13*			*16*			*15*	
std. dev.		*1.8*			*1.9*			*1.7*			*1.9*	
3 season mean		***23***			***12***			***26***			***20***	
3 season std.dev.		***1.8***			***1.9***			***1.9***			***1.9***	

Table 5.5 (b) PM_{10}.

Child ID No.	Home			Garden			Class			Personal			Brent AUN			Haringey AUN		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
2	5	43	30-67	4	36	31-44	NS	NS	NS	3	21	16-25	5	25	17-33	5	31	20-45
3	3	82	49-110	3	19	14-35	1	50	50	4	49	28-81	5	13	5-27	5	21	16-31
4	4	34	17-59	4	11	6-16	NS	NS	NS	5	26	7-63	5	13	11-16	5	20	15-25
5	5	45	33-52	4	39	34-50	NS	NS	NS	4	29	22-35	5	34	24-43	5	40	30-50
6	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
7	3	43	38-48	3	25	13-48	2	74	71-77	3	35	23-55	5	19	10-43	5	23	11-57
8	5	41	25-70	5	27	18-43	3	127	80-195	5	54	27-92	5	22	13-35	5	29	19-61
9	5	27	16-66	4	16	7-32	NS	NS	NS	4	48	35-63	5	18	7-57	5	24	11-62
10	9	35	9-125	8	17	7-29	3	80	43-142	8	34	18-62	10	15	8-29	10	18	10-24
mean		*40*			*21*			*87*			*32*			*18*			*23*	
std.dev		*1.8*			*1.8*			*1.6*			*2.3*			*1.7*			*1.6*	
3 season mean		***49***			***21***			***79***			***54***			***19***			***23***	
3 season std.dev.		***1.9***			***1.8***			***1.6***			***2.1***			***1.6***			***1.5***	

Table 5.6 Winter Night Mean Concentrations of all Microenvironments for all Children, $PM_{2.5}$ and PM_{10} ($\mu g/m^3$).

Child ID No.	Home ($PM_{2.5}$)			Home (PM_{10})			Garden ($PM_{2.5}$)			Garden (PM_{10})			Brent AUN (PM_{10})			Haringey AUN (PM_{10})		
	N	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**
1	3	13	11-14	2	24	22-27	3	8	3-16	4	22	11-32	5	19	10-26	5	21	10-30
2	4	29	19-50	4	49	32-89	3	25	19-37	3	35	29-50	5	22	16-37	5	26	19-50
3	4	15	10-19	3	29	21-39	4	12	9-14	4	14	3-32	5	19	13-28	5	21	15-30
4	1	20	20	NS	NS	NS	NS	NS	NS	NS	NS	NS	5	17	11-24	5	19	12-27
5	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	5	26	8-41	5	31	12-48
6	3	9	5-16	4	26	14-51	3	10	3-21	2	29	21-39	5	15	11-22	5	17	9-25
7	2	19	15-24	3	24	6-24	3	19	3-86	3	29	16-59	3	30	29-32	5	35	24-60
8	1	6	6	NS	NS	NS	2	4	3-6	1	28	28	5	16	11-24	5	19	12-29
9	2	17	16-17	1	21	21	2	15	14-15	NS	NS	NS	5	26	22-30	5	26	22-31
10	1	82	82	1	73	73	3	9	5-14	3	14	11-15	NS	NS	NS	5	21	16-30
mean		*21*			*34*			*17*			*26*			*20*			*21*	
std.dev.		*1.9*			*2.0*			*2.2*			*2.2*			*1.6*			*1.5*	

Table 5.7 Spring Night Mean Concentrations of all Microenvironments for all Children, $PM_{2.5}$ and PM_{10} ($\mu g/m^3$).

Child ID No.	Home ($PM_{2.5}$)			Home (PM_{10})			Garden ($PM_{2.5}$)			Garden (PM_{10})			Brent AUN (PM_{10})			Haringey AUN (PM_{10})		
	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range	N	Mean	Range
1	8	10	6-26	9	25	14-32	9	8	3-21	9	17	10-28	10	16	12-21	10	19	15-25
2	3	24	20-30	2	40	34-46	4	24	16-38	4	34	25-48	5	24	13-36	5	26	15-36
3	4	22	14-48	4	42	34-69	4	26	24-45	4	41	32-59	5	24	20-38	5	29	22-45
4	3	12	10-14	4	25	24-25	4	12	10-14	4	14	9-18	5	15	9-26	5	17	10-29
5	4	4	3-6	3	19	16-25	4	4	3-7	4	10	8-15	5	11	7-20	5	14	9-23
6	3	12	10-13	3	22	19-24	4	10	5-22	3	11	8-18	5	14	9-26	5	17	10-29
7	3	12	9-16	3	32	26-44	4	8	5-10	4	16	16-22	5	15	13-21	5	19	17-23
8	4	10	7-13	2	15	13-18	4	9	6-13	2	17	17	5	15	13-19	5	20	18-26
9	4	13	10-16	4	17	15-19	4	11	6-23	4	12	3-33	5	14	11-25	5	20	17-27
10	4	27	21-40	4	42	34-60	3	9	6-15	4	13	8-17	5	14	9-16	5	17	15-18
mean		*13*			*26*			*11*			*17*			*16*			*19*	
Std.dev.		*1.8*			*1.5*			*1.9*			*1.9*			*1.5*			*1.4*	

Table 5.8 Summer Night Mean Concentrations of all Microenvironments for all Children, PM_{10} and $PM_{2.5}$ ($\mu g/m^3$).

Child ID No.	Home ($PM_{2.5}$)			Home (PM_{10})			Garden ($PM_{2.5}$)			Garden (PM_{10})			Brent AUN (PM_{10})			Haringey AUN (PM_{10})		
	N	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**	**N**	**Mean**	**Range**
1	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
2	4	30	25-38	4	43	35-51	3	20	11-37	4	40	27-84	5	29	19-42	5	32	21-49
3	4	13	6-36	3	27	22-31	2	5	3-10	4	9	3-31	5	16	7-26	5	19	11-26
4	4	10	6-22	4	24	12-40	4	4	3-7	4	13	7-18	5	14	10-21	5	18	12-30
5	3	19	13-34	4	28	18-47	3	25	18-41	4	36	28-52	5	35	23-47	5	41	31-52
6	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
7	4	10	6-22	4	22	12-44	4	8	3-23	3	20	8-42	5	18	11-40	5	19	11-43
8	2	20	14-30	4	30	17-57	4	15	9-44	3	24	14-56	5	21	16-41	5	21	15-44
9	4	10	3-38	4	25	17-49	4	11	3-57	3	24	12-70	5	14	7-56	5	17	9-62
10	7	29	20-52	8	43	24-80	7	10	3-19	7	17	9-31	10	16	7-24	10	18	9-27
mean		*17*			*31*			*10*			*20*			*18*			*21*	
Std.dev.		*2.0*			*1.6*			*2.3*			*2.3*			*1.7*			*1.6*	
3 season mean		***15***			***28***			***11***			***19***			***18***			***21***	
3 season std.dev.		***1.9***			***1.6***			***2.1***			***2.1***			***1.6***			***1.5***	

5.6 PM_{10} – $PM_{2.5}$ Relationships

The mean PM_{10} and $PM_{2.5}$ concentrations for each child are ranked in Tables 5.9 and 5.10. Ranked mean values are represented as 1 to 10, highest to lowest concentrations. Ranking the mean data determines a trend between the two size fractions. These tables also show the ratio of the concentrations representing the contribution that $PM_{2.5}$ makes to PM_{10}. Where the ratios are low then the contribution of $PM_{2.5}$ to PM_{10} is smaller. The smallest ratios are typically between the personal $PM_{2.5}$ and PM_{10} concentrations with a median value of 0.34, whilst the largest ratios are found at night in the garden. It has been suggested that personal activities are responsible for creating a 'personal PM_{10} cloud effect' (Spengler *et al.,* 1981). This could be responsible for these differences between the two size fractions. The reason why the indoor and classroom $PM_{2.5}$ and PM_{10} ratios are lower than the garden ratios are less clear, this could be a result of specific indoor activities resuspending the larger particles.

The ratios for the two size fractions fall within previously published values. In a study of six U.S. cities' fine and coarse particle mass, Spengler and Thurston,(1983) found that approximately 60 - 68% of the inhalable fraction of particles was contained in particles less than 2.5 μm diameter for five of the cities with only one city demonstrating a lower percentage of 50%. Janssen *et al.,* (1997) in their analysis of airborne particulate matter at street and background locations in the Netherlands found that the ratio of $PM_{2.5}$: PM_{10} was 0.56 and ranged from 0.21 - 0.79. Rojas-Bracho *et al.,* (2000) found that PM_{10} was approximately 60% $PM_{2.5}$. The values for the outdoor ratios in this study's evaluation of children's personal exposure fall within these limits. The range of the children's garden values during the day are 0.45 - 0.76 with a median value of 0.56. This suggests that during the day 56% of PM_{10} can be attributed to $PM_{2.5}$. Studies of other urban sites within the UK have been undertaken and data from Birmingham shows that the ratio between $PM_{2.5}$: PM_{10} is 0.56 (Harrison *et al.,* 1997). The contribution of $PM_{2.5}$ to the PM_{10} fraction represented about 60% in the study undertaken in Leeds by Clarke *et al.,* (1984).

At night, the range of garden values were 0.45 - 0.81 with a median value of 0.59. The contribution that $PM_{2.5}$ makes to PM_{10} is similar at night to that found during the day.

There appears to be little consistency between the ranks and the size fractions for each environment. Only when comparing the same size fraction for both the personal exposure and the home do the majority of the children's ranked means match.

Table 5.9 Day Ranks and Ratios of PM_{10} and $PM_{2.5}$ Concentrations.

Child ID[1]	Personal			Home			Garden			Class		
	Ranked			Ranked			Ranked			Ranked		
	$PM_{2.5}$	PM_{10}	Ratio	$PM_{2.5}$	PM_{10}	Ratio	$PM_{2.5}$	PM_{10}	Ratio	$PM_{2.5}$	PM_{10}	Ratio
1	2	1	0.26	1	2	0.40	9	6	0.51	5	4	0.33
2	3	8	0.46	2	5	0.58	1	1	0.45	3	3	0.39
3	6	4	0.27	3	1	0.35	2	2	0.59	9	2	0.21
4	7	6	0.37	7	6	0.40	10	10	0.49	4	5	0.35
5	10	7	0.31	9	7	0.43	6	3	0.53	7	7	0.31
6	9	5	0.29	10	4	0.32	4	5	0.65	6	9	0.38
7	8	10	0.45	6	9	0.51	3	7	0.76	1	10	0.67
8	5	3	0.26	5	3	0.38	7	4	0.49	10	1	0.19
9	1	2	0.40	8	10	0.67	5	8	0.70	2	8	0.53
10	4	9	0.46	4	8	0.59	8	9	0.58	8	6	0.27
Median			0.34			0.42			0.56			0.42

Table 5.10 Night Ranks and Ratios of Concentrations.

Child ID	Home			Garden		
	Ranked			Ranked		
	$PM_{2.5}$	PM_{10}	Ratio	$PM_{2.5}$	PM_{10}	Ratio
1	8	4	0.44	9	5	0.46
2	2	1	0.63	1	1	0.64
3	3	3	0.49	2	6	0.81
4	6	7	0.50	10	10	0.52
5	10	8	0.36	6	4	0.53
6	9	5	0.41	5	7	0.65
7	5	10	0.63	4	3	0.51
8	7	6	0.47	3	2	0.45
9	4	9	0.61	8	8	0.78
10	1	2	0.70	7	9	0.67
	Median		0.50	Median		0.59

[1] Child Identification Number

5.7 Personal PM_{10} – $PM_{2.5}$ Relationships

Tables 5.11 and 5.12 give each individual child's range of data for the microenvironments sampled. Table 4.2 shows the characteristics of each child and their home environment. The variations exhibited in Table 5.11 and 5.12 for the individual children may possibly be explained by these different characteristics, further investigation using the questionnaire data will provide insight into this in Chapter 6.

There were some variations between and within the individual children's personal PM_{10} concentrations ranging from 35 - 80 $\mu g/m^3$. The $PM_{2.5}$ personal concentrations did not exhibit such wide variations, having a range of 12 - 27 $\mu g/m^3$. There is little consistency in the variability of the two size fractions, for the personal $PM_{2.5}$ concentrations six of the ten children have higher standard deviations suggesting that there is a wider variability between and within these exposure measurements than the PM_{10} concentrations.

The classroom values show some of the highest concentrations collected in the study for both size fractions, the standard deviations for the majority of the children are lower for the PM_{10} concentrations. These high values would therefore appear to be consistent for within child variability.

A similar pattern is exhibited in the home values where many of the highest concentrations were also found. Again, the variability appears to be greater for each child's home $PM_{2.5}$ concentration, suggesting that on a daily basis concentrations fluctuate, further investigation of the activities conducted within the homes may provide some explanations for this variability.

The garden values show some of the lowest overall concentrations and much of the variability between sampling days occurs in the PM_{10} size fraction. Sporadic local sources could be responsible for this. The fact that $PM_{2.5}$ concentrations in a UK urban environment are generally traffic related along with the smaller size fraction leading to greater spatial homogeneity of these particles would suggest a reason for the reduced within child variability.

At night, the home concentrations typically fall and the variability between each child's measurements also reduces for both size fractions. The garden concentrations show little consistency in variability although there is a slight reduction in concentrations from the daytime although not by much suggesting consistent emissions during the day and night occurred.

Table 5.11 Day Descriptive Statistics for Individual Children in all Seasons ($\mu g/m^3$).

Child Identification Number		Home $PM_{2.5}$	Home PM_{10}	Garden $PM_{2.5}$	Garden PM_{10}	Class $PM_{2.5}$	Class PM_{10}	Personal $PM_{2.5}$	Personal PM_{10}	Brent AUN PM_{10}	Hari[1] AUN PM_{10}
Child 1	Min	17	49	3	8	21	68	5	42	9	9
	Max	87	127	25	43	40	111	46	140	34	46
	Geomean	*31*	*79*	*10*	*19*	*28*	*85*	*19*	*80*	*18*	*21*
	St. Dev.	1.7	1.4	1.8	1.5	1.3	1.2	1.5	1.4	1.4	1.4
	Number	14	12	15	14	4	4	12	15	15	15
Child 2	Min	12	30	7	16	26	51	5	7	8	13
	Max	68	103	40	171	45	155	45	115	39	49
	Geomean	*30*	*51*	*19*	*43*	*33*	*85*	*19*	*43*	*24*	*31*
	St. Dev.	1.7	1.4	1.7	1.8	1.3	1.7	1.8	2.5	1.5	1.4
	Number	11	11	10	10	4	5	15	13	15	15
Child 3	Min	14	34	3	14	9	50	5	28	5	16
	Max	43	136	34	52	33	136	54	153	33	42
	Geomean	*28*	*79*	*14*	*25*	*19*	*92*	*17*	*64*	*18*	*23*
	St. Dev.	1.4	1.6	1.8	1.6	1.9	1.7	2.2	1.6	1.6	1.3
	Number	12	12	13	11	4	3	12	13	15	15
Child 4	Min	3	11	3	6	20	75	5	7	9	14
	Max	60	145	20	30	41	93	28	171	28	32
	Geomean	*18*	*47*	*5*	*15*	*29*	*83*	*16*	*54*	*16*	*21*
	St. Dev.	2.3	2.1	1.7	1.6	1.4	1.1	2	4.2	1.4	1.3
	Number	11	11	9	9	4	4	14	14	15	15
Child 5	Min	11	32	5	10	21	58	5	22	13	13
	Max	40	52	31	50	28	84	11	98	72	68
	Geomean	*19*	*43*	*13*	*24*	*23*	*73*	*15*	*43*	*25*	*28*
	St. Dev.	1.6	1.2	2	1.9	1.2	1.2	2.8	1.5	1.7	1.7
	Number	10	10	10	8	3	3	13	12	15	15
Child 6	Min	8	29	7	11	15	28	5	17	9	14
	Max	30	133	27	36	34	169	37	128	29	26
	Geomean	*17*	*53*	*14*	*21*	*25*	*67*	*12*	*46*	*16*	*20*
	St. Dev.	1.6	1.6	1.7	1.5	1.4	2.1	1.8	1.9	1.5	1.2
	Number	7	6	8	8	4	5	6	6	10	10
Child 7	Min	13	9	3	6	15	22	5	16	10	11
	Max	21	89	49	48	125	77	44	106	43	57
	Geomean	*19*	*38*	*15*	*18*	*43*	*58*	*16*	*35*	*20*	*25*
	St. Dev.	1.4	1.8	2.1	1.9	2.4	1.8	2.4	2	1.6	1.7
	Number	8	9	9	9	5	5	13	7	13	15
Child 8	Min	12	25	3	7	3	24	5	27	10	9
	Max	31	72	28	43	36	195	38	193	27	61
	Geomean	*21*	*47*	*11*	*21*	*19*	*93*	*17*	*67*	*18*	*22*
	St. Dev.	1.4	2	2.2	1.7	2.8	1.8	1.7	1.6	1.4	1.6
	Number	8	10	9	9	7	11	13	14	15	15
Child 9	Min	3	16	7	7	20	35	5	35	7	11
	Max	52	66	26	49	77	131	91	129	57	62
	Geomean	*18*	*28*	*13*	*18*	*37*	*69*	*27*	*72*	*22*	*25*
	St. Dev.	2	1.6	1.6	1.8	1.6	1.6	1.8	1.5	1.8	1.7
	Number	13	13	10	9	5	6	12	11	15	15
Child 10	Min	3	9	3	6	12	40	5	7	8	10
	Max	103	157	26	58	44	159	44	149	29	42
	Geomean	*24*	*42*	*10*	*18*	*22*	*81*	*17*	*38*	*14*	*20*
	St. Dev.	2.3	2.2	1.5	1.8	1.6	1.8	1.8	2	1.4	1.4
	Number	16	17	15	16	6	6	15	17	15	20

[1] Haringey AUN Site

Table 5.12 Night Descriptive Statistics for Individual Children in all Seasons ($\mu g/m^3$).

Child Identification Number		Home $PM_{2.5}$	Home PM_{10}	Garden $PM_{2.5}$	Garden PM_{10}	Brent AUN PM_{10}	Haringey AUN PM_{10}
Child 1	Min	6	15	3	10	10	10
	Max	26	31	21	32	26	30
	Geomean	*11*	*25*	*8*	*18*	*17*	*20*
	St. Dev.	1.5	1.3	1.9	1.5	1.3	1.3
	Number	11	11	12	13	15	15
Child 2	Min	20	32	11	35	13	15
	Max	50	89	39	84	42	45
	Geomean	*28*	*44*	*23*	*36*	*25*	*28*
	St. Dev.	1.3	1.4	1.5	1.4	1.4	1.5
	Number	11	10	10	11	15	15
Child 3	Min	7	21	3	3	13	11
	Max	48	69	45	59	28	45
	Geomean	*16*	*33*	*14*	*17*	*19*	*22*
	St. Dev.	2	1.4	2.3	3.8	1.5	1.4
	Number	12	10	10	12	15	15
Child 4	Min	6	12	3	7	9	10
	Max	22	40	14	18	26	30
	Geomean	*12*	*24*	*7*	*13*	*15*	*18*
	St. Dev.	1.5	1.4	2.1	1.4	1.4	1.4
	Number	8	8	8	8	15	15
Child 5	Min	3	16	3	8	7	9
	Max	34	47	41	52	47	52
	Geomean	*8*	*23*	*9*	*19*	*22*	*26*
	St. Dev.	2.3	1.5	2.4	2.1	2	1.8
	Number	7	7	7	8	15	15
Child 6	Min	5	14	3	8	9	9
	Max	16	51	22	39	26	29
	Geomean	*10*	*24*	*10*	*16*	*15*	*17*
	St. Dev.	1.5	1.5	2	1.9	1.4	1.4
	Number	6	7	7	5	10	10
Child 7	Min	6	6	3	8	11	11
	Max	24	44	86	59	40	60
	Geomean	*13*	*20*	*10*	*21*	*19*	*24*
	St. Dev.	1.6	2	2.8	1.8	1.5	1.6
	Number	9	10	11	10	13	15
Child 8	Min	6	13	3	14	11	12
	Max	30	57	44	56	24	44
	Geomean	*11*	*24*	*10*	*22*	*17*	*20*
	St. Dev.	1.7	1.7	2	1.7	1.4	1.4
	Number	11	6	10	6	15	15
Child 9	Min	3	15	3	3	7	9
	Max	38	49	57	70	30	62
	Geomean	*12*	*20*	*12*	*16*	*17*	*21*
	St. Dev.	1.8	1.5	2.1	2.9	1.8	1.6
	Number	10	9	10	7	15	15
Child 10	Min	20	24	3	8	7	9
	Max	82	80	19	31	21	30
	Geomean	*31*	*45*	*10*	*15*	*15*	*18*
	St. Dev.	1.6	1.4	1.6	1.5	1.4	1.4
	Number	12	13	13	14	15	20

5.8 Spearman Rank Correlation Analysis

Spearman's correlation analysis of the data was carried out to identify any significant and non-significant associations between the different microenvironments. Scatter plots of the data were also drawn to simplify the analysis procedure. Significant correlations are considered to be p values of 0.05 and smaller.

Correlations were only considered on sampling sizes of 4 observations or more as p values could not be calculated for smaller sample sizes. The study design, as discussed previously, sampled one child for 5 consecutive days during winter, spring and summer. As such, it is only possible to analyse within child variability and not between child variability as there are many confounding effects that could be responsible for the differences found between children.

Where it is apparent there are outliers in the data these have been analysed further. If it was found that the removal of the observations did not improve the significance of the results then these were left in as there are no suitable reason for removing them.

5.8.1 All Day-time Data, Correlation Interpretation

The correlation analyses in Tables 5.13 and 5.14 provide evidence of the daytime correlations between the different microenvironments for all available observations throughout the sampling sessions. The association between personal PM_{10} and garden PM_{10} were non significant, however the associations between personal PM_{10} with the AUN sites were significant although the coefficients are small. Section 5.3 of the descriptive statistics indicated that the majority of the personal exposure concentrations exceeded the outdoor values. The fact that the association between the personal and garden is non significant but the personal and ambient are could be suggestive of the influence of local pollution sources.

The correlations between the home PM_{10} concentrations with the AUN sites were not significant. No further analysis of this relationship will be carried out. The classroom concentrations were not significantly associated with any of the outdoor microenvironments. The study by Janssen (1998) indicated a correlation between $PM_{2.5}$ in the classroom and the ambient sites. Further analysis of the correlations between the classroom and outdoor concentrations will be investigated in Sections 5.8.3 and 5.8.4. It is unclear why there is a significant association between the home and classroom PM_{10}

concentrations, this could just be a result of resuspended particulates in the indoor environments.

The strongest associations were found between the personal PM_{10} and $PM_{2.5}$ concentrations and the corresponding home concentrations as seen in Tables 5.13 and 5.14. Interpretation of the seasonal concentrations in Section 5.8.3 may indicate whether concentrations within the home were influenced by the different seasons. Whether the children's activities altered during different seasons will be investigated in Section 5.10 as a reason for any potential seasonal variation in exposure patterns.

Significant associations were found between the $PM_{2.5}$ and PM_{10} size fractions in the garden, home and personal measurements as seen in Table 5.15. This supports the analysis of the ratios indicated previously which found that >50% of the PM_{10} is attributable to $PM_{2.5}$ (see Table 5.9).

The association between the classroom $PM_{2.5}$ and PM_{10} size fractions were not significant, suggesting that the sources of PM_{10} were not related to the $PM_{2.5}$ fraction. This may be indicative of the effect of resuspension of the larger size fraction due to the number of children present in the classroom. There was a significant correlation between the personal $PM_{2.5}$ concentration and the class $PM_{2.5}$ which will be analysed further in Section 5.8.3.

Some suggestions for these high concentrations and lack of correlations with the outdoor concentrations have been made. Ashmore (Pers. Comm., 1999) suggested that due to the large number of children inhabiting one room that resuspension of the particles was a major source. In unpublished research of particle numbers within classrooms, he found that as soon as children entered into the classroom then particle numbers increased by a factor of 2-3. Janssen (1998) in the elemental analysis of PM_{10}, found that mass concentrations and most elemental concentrations were considerably higher than outdoor concentrations, especially during school hours. It was concluded that the causes for this are from the resuspension of the coarse particles and / or suspension of soil material caused by the activity of the children.

This study has highlighted the contribution that school PM_{10} and $PM_{2.5}$ have on personal exposure concentrations is not as significant as the home environment. This may be a problem with the small sample size and the missing data as the literature suggests that classroom exposure is usually a predominant source of personal exposure for children. It

is necessary to conduct further source apportionment studies on the particulate matter within schools to determine the sources of the particles and is discussed in Section 6.4.

Significant associations existed between the garden PM_{10} sites and both the AUN sites in the London Boroughs of Brent and Haringey. This suggests that the background concentrations of PM_{10} in the urban area of Barnet were fairly consistent.

The correlation between the two AUN sites exhibited the strongest association indicating that the background PM_{10} within the local area was uniform, suggesting that a single site may be suitable for monitoring ambient PM_{10} concentrations within Barnet. Further analysis of this will be undertaken in Section 5.9.

Table 5.13 Summary of 3 Season Means for Daytime PM_{10} Correlation Analyses for all Children.

		Home	Garden	Personal	School	Brent AUN	Haringey AUN
Home	r	1.00000	0.22888	0.59064	0.34637	0.07126	0.13513
	p		0.0330	<.0001	0.0357	0.4636	0.1536
	n	113	87	95	37	108	113
Garden			1.00000	0.07442	-0.14042	0.72611	0.75191
				0.4933	0.4848	<.0001	<.0001
			103	87	27	97	103
Personal				1.00000	0.34540	0.17905	0.17969
					0.0290	0.0545	0.0477
				122	40	116	122
School					1.00000	0.13508	-0.01498
						0.3653	0.9177
					50	47	50
Brent AUN						1.00000	0.84267
							<.0001
						142	142
Haringey AUN							1.00000
							150

Table 5.14 Summary of 3 Season Mean Daytime $PM_{2.5}$ Correlation Analyses for all Children.

		Home	Garden	Personal	School
Home	r	1.00000	0.41991	0.52485	0.23167
	p		<.0001	<.0001	0.2020
	n	110	93	91	32
Garden			1.00000	0.38844	0.28564
				0.0002	0.1130
			108	89	32
Personal				1.00000	0.50703
					0.0019
				126	35
School					1.00000
					46

Table 5.15 The 3 Season Mean Day time Correlations between PM_{10} and $PM_{2.5}$ for all Children in each Environment.

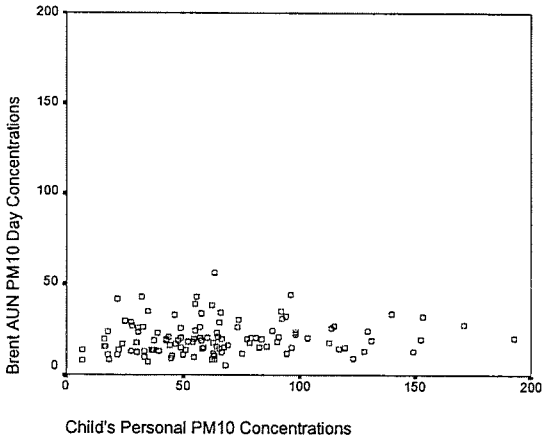
		Day	Night
Home	r	0.71167	0.76798
	p	<.0001	<.0001
	n	93	79
Garden		0.77906	0.78943
		<.0001	<.0001
		87	84
Personal		0.56508	
		<.0001	
		112	
School		0.24192	
		0.1181	
		43	

Table 5.16 Summary of 3 Season Mean Night time PM_{10} Correlation Analyses for all Children.

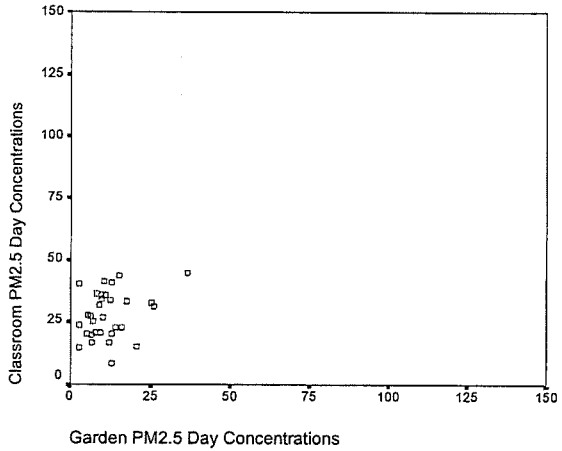
	Home	Garden	Brent AUN	Haringey AUN
Home	1.00000	0.47679	0.46705	0.38280
		<.0001	<.0001	0.0002
	90	80	87	90
Garden		1.00000	0.80489	0.80712
			<.0001	<.0001
		94	88	94
Brent AUN			1.00000	0.96161
				<.0001
			142	142
Haringey AUN				1.00000
				150

Table 5.17 Summary of 3 Season Mean Night time $PM_{2.5}$ Correlation Analyses for all Children.

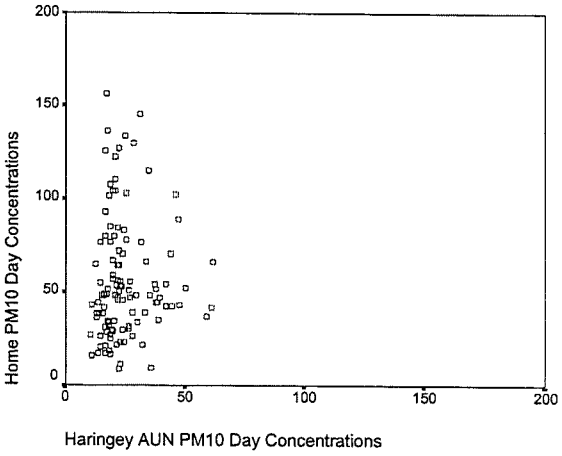
	Home	Garden
Home	1.00000	0.63075
		<.0001
	93	83
Garden		1.00000
		98



a) $r = 0.18$ $p = 0.055$ $n = 116$

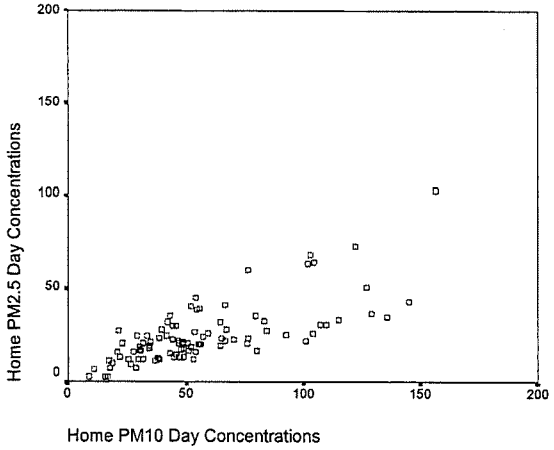


b) $r = 0.29$ $p = 0.11$ $n = 32$

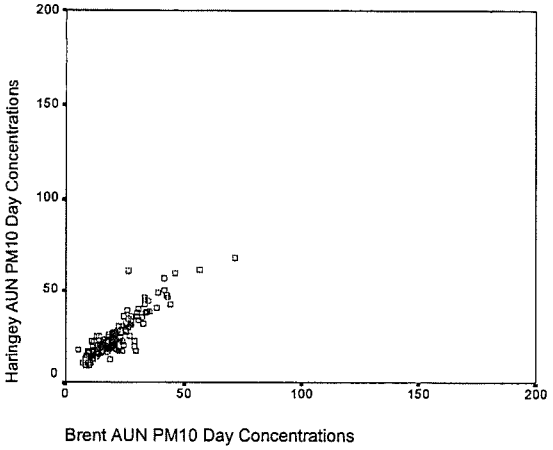


c) $r = 0.14$ $p = 0.15$ $n = 114$

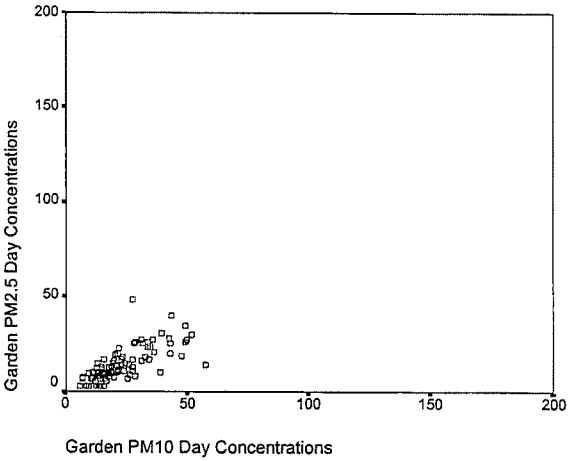
Figure 5.3(a-c) Examples of Weak or No Correlations, Day Concentrations ($\mu g/m^3$).



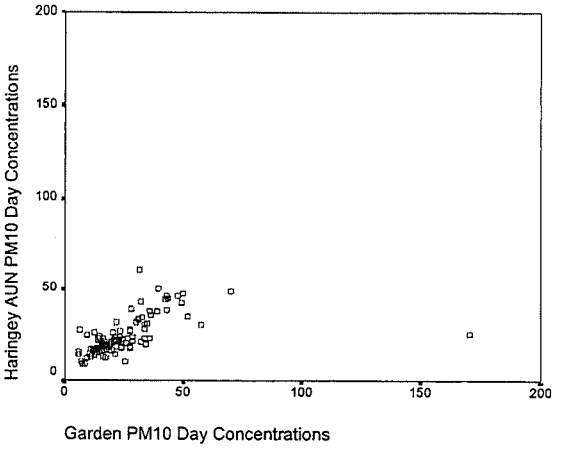
a) $r = 0.71$ $p = <0.0001$ $n = 93$



b) $r = 0.84$ $p = <0.0001$ $n = 142$



c) $r = 0.78$ $p = <0.0001$ $n = 87$



d) $r = 0.75$ $p = <0.0001$ $n = 103$

Figure 5.4(a-d) Examples of Significant Correlations, Day Concentrations ($\mu g/m^3$).

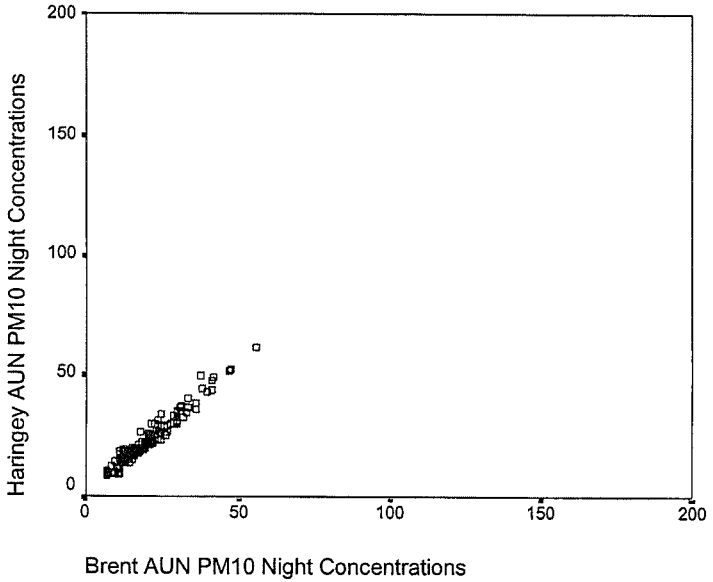
5.8.2 All 3 Seasons Night Data, Spearman Rank Correlation Interpretations

All associations between the different microenvironments during the night were significant as can be seen in both Tables 5.16 and 5.17. Possible reasons for this are that particle generating activities occurred more frequently during the day and this reduced the influence that the outdoor particulate concentrations had upon the indoor environments, see Section 2.6. This was also suggested by the differences in the ratios between particulate size fractions as seen in Section 5.3.

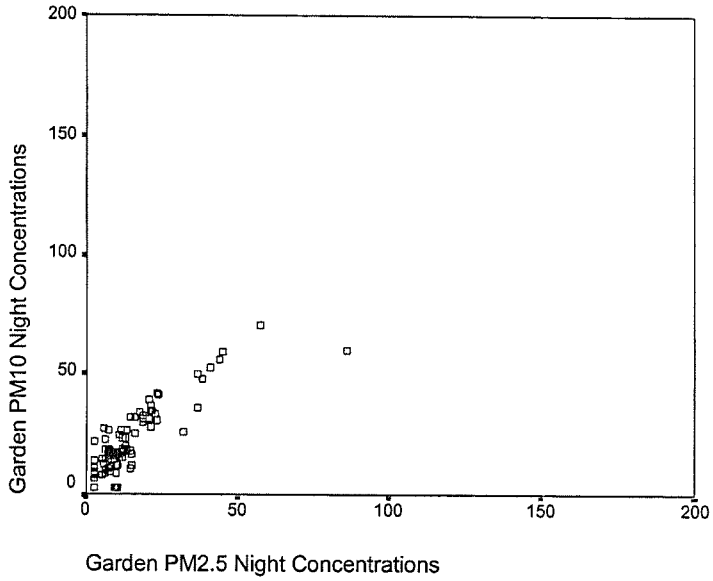
The associations between the home and the outdoor sites for PM_{10} exhibited smaller coefficients, which could be due to greater variability in the indoor concentrations. Possible reasons for this have been explained previously, see Section 5.6. The $PM_{2.5}$ associations were all significant and the coefficients were greater than 0.6 suggesting a more linear association.

The coefficients between the $PM_{2.5}$ and PM_{10} size fractions for both the garden and home sites were greater than 0.75 but were not greatly different during the night than when compared to the day. This was indicated in Table 5.10 where at night the contribution of $PM_{2.5}$ to PM_{10} in the outdoor samples were approximately 59% compared to 56% during the day.

A number of issues have been raised with the analysis of the total data collected over the three seasons. Where it is evident that there was clearly no association between the microenvironments, no further analysis will be undertaken.



a) $r = 0.96$ $p = <0.0001$ $n = 142$



b) $r = 0.78$ $p = <0.0001$ $n = 87$

Figure 5.5(a-b) Examples of Significant Correlations, Night Concentrations ($\mu g/m^3$).

5.8.3 Individual Seasons, Spearman's Rank Correlation Analysis

The observations for all of the children have been used in the correlation analysis of the seasonal data. The sampling programme was undertaken in winter, spring and summer as previously described in Section 4.2. Table 5.18 shows the summary for the temperature and rainfall in each of the seasons that were sampled. The spring sampling session had the highest rainfall. This would suggest that the indoor environments would have windows closed more frequently and hence, potentially, the ventilation would be reduced. This may also have an effect upon personal exposures which appear to be influenced more by indoor concentrations than outdoor during the winter and spring. The seasonal correlations that have been investigated are shown in Table 5.19.

The results from Table 5.14 show that the children's personal exposure concentrations have r = 0.51 for $PM_{2.5}$ when correlated with classroom concentrations, when the data is separated seasonally as shown in Table 5.19 and Figure 5.6 only during the winter sampling period is the personal and classroom association significant. The meteorological conditions were wet and cold so there was more likelihood that the windows were kept closed and the children remained indoors throughout their break periods, potentially leading to the resuspension of particulates and less dispersion. If weather conditions were dry and warm then the opposite is likely. As one child was sampled per 5 days it is not possible to determine the effects that meteorological conditions have upon such personal exposures. Other significant associations when the data were combined, and remained so when analysed separately for each season, included $PM_{2.5}$ associations between the home and garden, and the personal and home associations as seen clearly in Figure 5.8. The PM_{10} associations were less clear, personal and home associations were only significant during the winter and spring as can be seen in Figure 5.9, whilst the associations between the home and garden were only significant during the summer.

Table 5.18 Total Rainfall and Average Temperature in each Season.

Season	Time	Total Rainfall (cm)	Average Temperature (°C)
Winter	**Day**	18.0	11.0
	Night	5.6	7.0
Spring	**Day**	78.7	15.0
	Night	59.0	10.0
Summer	**Day**	34.3	21.3
	Night	27.0	16.0

The PM_{10} correlations between the children's personal exposure and classroom, shown in Table 5.19, were similar to the $PM_{2.5}$ correlations; again the same explanation of meteorological conditions could be used.

Associations between the children's personal exposure concentrations and the home were evident for $PM_{2.5}$ during the winter, spring and summer. When analysed on a seasonal basis, as seen in Table 5.19, there does appear to be some variation in the coefficients, with the winter having the strongest association. It is possible that sources of $PM_{2.5}$ within the home do not vary over seasons, although other factors such as ventilation and heating may. This is expected and supports the literature in Section 2.3.

The PM_{10} association between personal exposure concentrations and the home concentrations exhibited some seasonal variations. The associations were significant during the winter and spring but not the summer. Possible reasons for this could be attributed to the children's activity patterns within different seasons and also the different activities carried out in the houses. Investigation of the children's activity patterns is discussed in Section 5.10. Analysis of the seasons by school day or non school day did not improve the correlations significantly, if anything the reduction in the number of observations reduced the significance of the associations.

The associations between the AUN sites and the garden concentrations were all significant for all seasons for PM_{10}.

Table 5.19 Summary of Seasonal Spearman's Rank Correlations for all Children.

PM_{10} Winter	Home	Garden	Personal	School	Brent AUN	Haringey AUN
Home	1.00000	0.12456	0.63478	0.05495	-0.31806	0.12461
		0.6114	0.0009	0.8585	0.1133	0.5118
	30	19	24	13	26	30
Garden		1.00000	0.06883	-0.54286	0.46165	0.72643
			0.7669	0.2657	0.0405	<.0001
		25	21	6	20	25
Personal			1.00000	0.63516	0.19539	0.23101
				0.0147	0.2759	0.1629
			38	14	33	38
School				1.00000	0.35320	0.11455
					0.1796	0.6508
				18	16	18
Brent AUN					1.00000	0.76262
						<.0001
					43	43
Haringey AUN						1.00000
						50

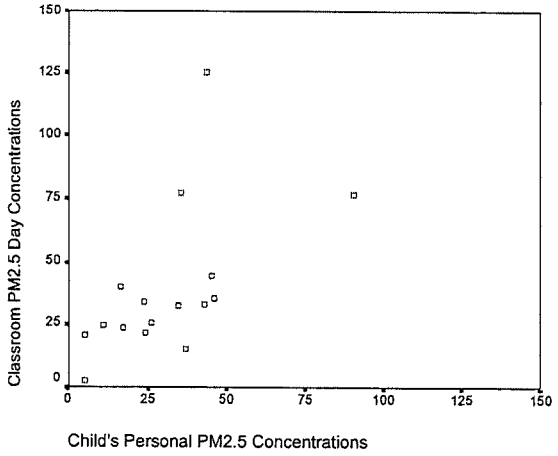
PM_{10} Spring	Home	Garden	Personal	School	Brent AUN	Haringey AUN
Home	1.00000	0.21467	0.50126	0.64461	0.04847	0.06727
		0.2086	0.0013	0.0052	0.7605	0.6721
	42	36	38	17	42	42
Garden		1.00000	0.12587	-0.35824	0.61296	0.64786
			0.4645	0.2085	<.0001	<.0001
		40	36	14	40	40
Personal			1.00000	0.08421	0.13968	0.21896
				0.7241	0.3601	0.1484
			45	20	45	45
School				1.00000	-0.24864	-0.33515
					0.2526	0.1180
				23	23	23
Brent AUN					1.00000	0.84827
						<.0001
					50	50
Haringey AUN						1.00000
						50

PM_{10} Summer	Home	Garden	Personal	School	Brent AUN	Haringey AUN
Home	1.00000	0.36987	0.26638	0.00000	0.32641	0.30366
		0.0372	0.1340	1.0000	0.0398	0.0536
	41	32	33	7	40	41
Garden		1.00000	0.02425	0.82143	0.85531	0.82405
			0.8988	0.0234	<.0001	<.0001
		38	30	7	37	38
Personal			1.00000	0.54286	0.12070	0.19960
				0.2657	0.4704	0.2231
			39	6	38	39
School				1.00000	0.66667	0.70000
					0.0710	0.0358
				9	8	9
Brent AUN					1.00000	0.86180
						<.0001
					49	49
Haringey AUN						1.00000
						50

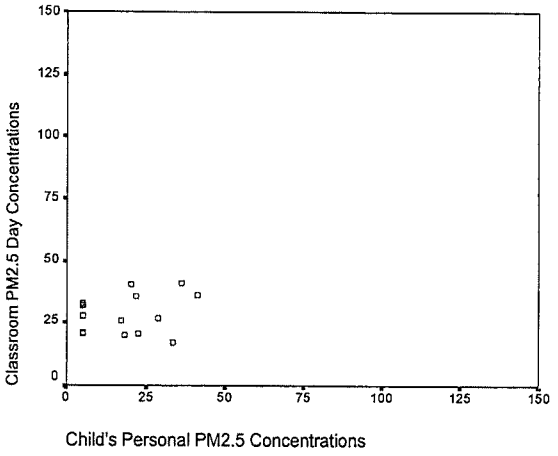
$PM_{2.5}$ Winter	Home	Garden	Personal	School
Home	1.00000	0.73929	0.57170	-0.14286
		0.0016	0.0105	0.7872
	22	15	19	6
Garden		1.00000	0.32919	0.09009
			0.1564	0.8477
		23	20	7
Personal			1.00000	0.63576
				0.0081
			44	16
School				1.00000
				17

$PM_{2.5}$ Spring	Home	Garden	Personal	School
Home	1.00000	0.45065	0.47522	0.20330
		0.0031	0.0045	0.4184
	44	41	34	18
Garden		1.00000	0.43689	0.37461
			0.0110	0.1256
		44	33	18
Personal			1.00000	0.19106
				0.5129
			39	14
School				1.00000
				20

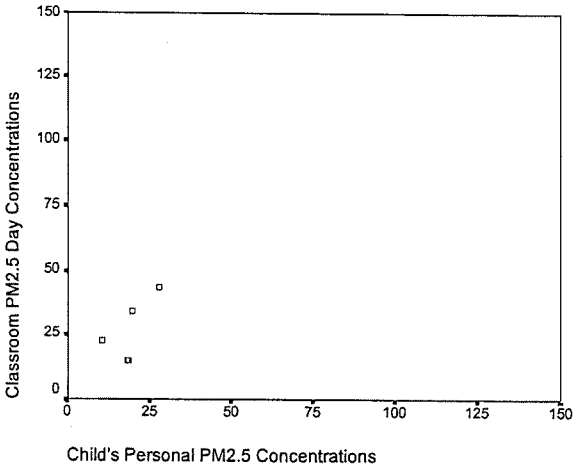
$PM_{2.5}$ Summer	Home	Garden	Personal	School
Home	1.00000	0.35181	0.41472	0.09524
		0.0327	0.0096	0.8225
	44	37	38	8
Garden		1.00000	0.40935	0.45047
			0.0132	0.3104
		41	36	7
Personal			1.00000	0.60000
				0.2848
			43	5
School				1.00000
				9



a) Winter $r = 0.64$ $p = 0.008$ $n = 16$

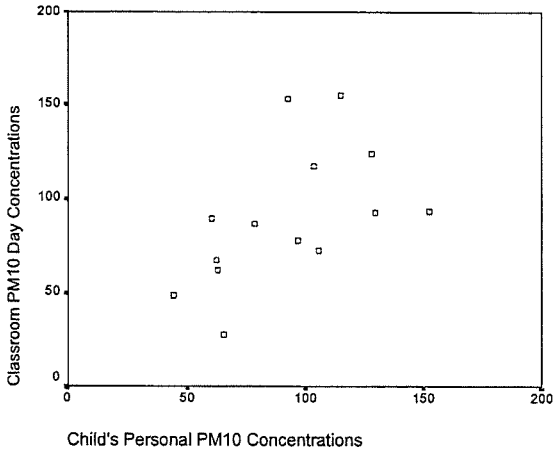


b) Spring $r = 0.19$ $p = 0.51$ $n = 14$

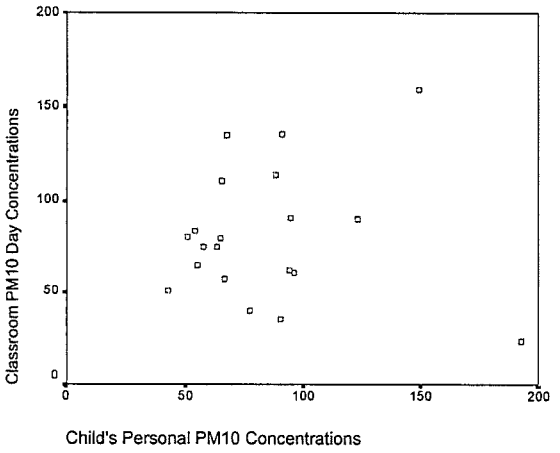


c) Summer $r = 0.6$ $p = 0.29$ $n = 5$

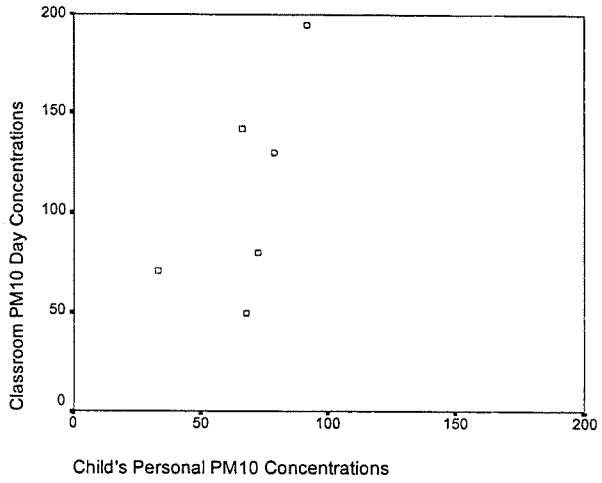
Figure 5.6a-c) Individual Seasons for all Children's Personal $PM_{2.5}$ Correlated with Class $PM_{2.5}$ ($\mu g/m^3$).



a) Winter $r = 0.64$ $p = 0.015$ $n = 14$

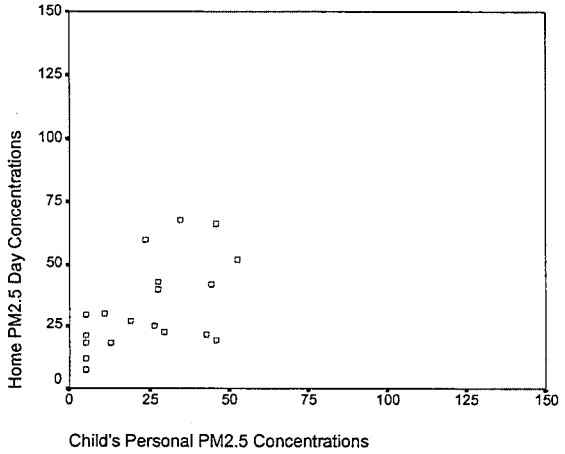


b) Spring $r = 0.08$ $p = 0.72$ $n = 20$

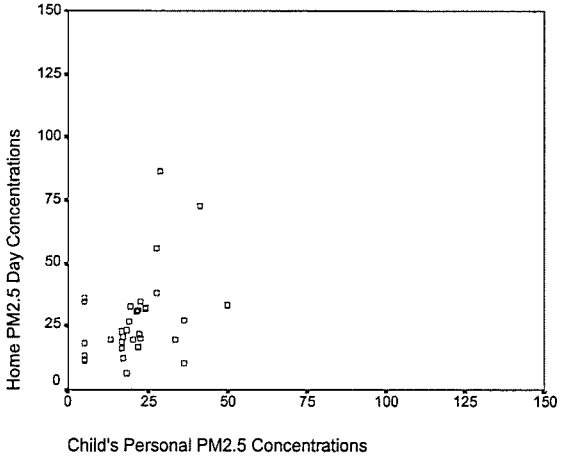


c) Summer $r = 0.54$ $p = 0.04$ $n = 6$

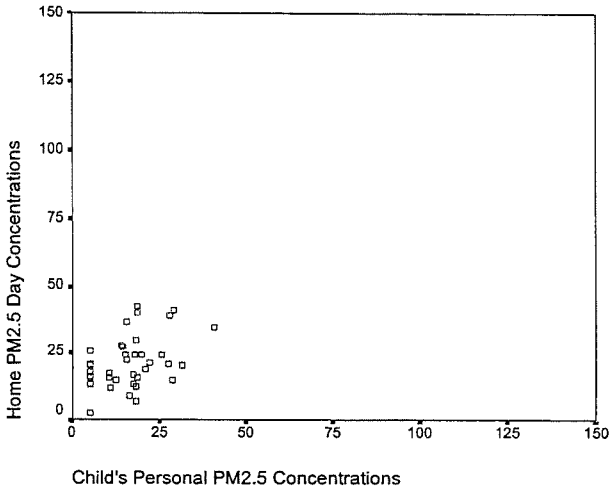
Figure 5.7 a-c) Individual Seasons for all Children's Personal PM_{10} Correlated with Class $PM_{10}(\mu g/m^3)$.



a) Winter $r = 0.57$ $p = 0.01$ $n = 19$

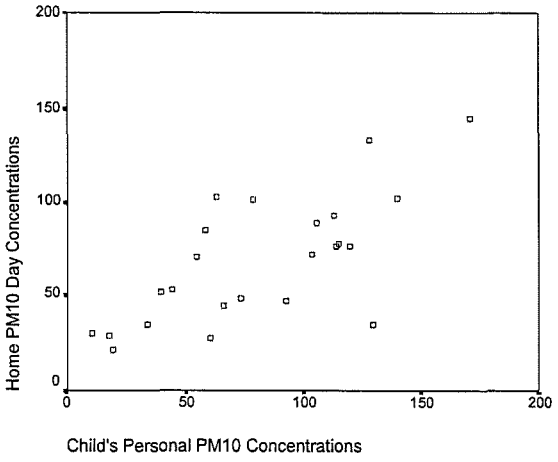


b) Spring $r = 0.48$ $p = 0.005$ $n = 34$

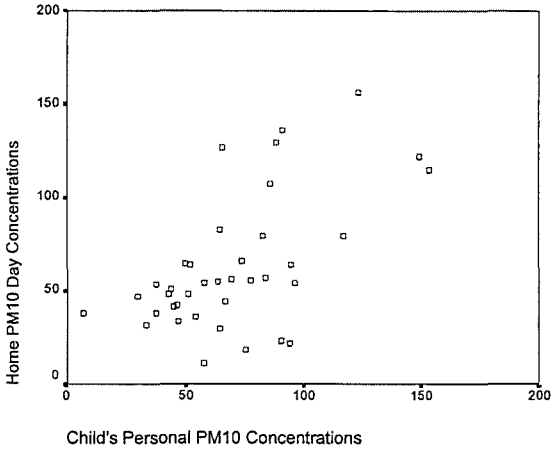


c) Summer $r = 0.42$ $p = 0.01$ $n = 38$

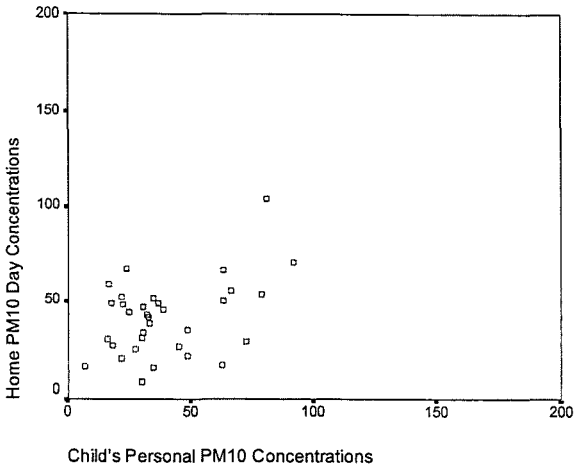
Figure 5.8 Individual Seasons for all Children's Personal $PM_{2.5}$ Correlated with Home $PM_{2.5}$ ($\mu g/m^3$).



a) Winter $r = 0.64$ $p = 0.001$ $n = 24$



b) Spring $r = 0.5$ $p = 0.001$ $n = 38$



c) Summer $r = 0.27$ $p = 0.13$ $n = 33$

Figure 5.9a-c) Individual Seasons for all Children's Personal PM_{10} Correlated with Home PM_{10} ($\mu g/m^3$).

5.8.4 Spearman's Rank Correlation Analysis of Individual Children's Data

The seasonal correlation analyses indicated that there were some seasonal variations between the personal exposure concentrations and the home and school. In order to identify whether these variations were due primarily to seasonal factors or, if other factors influenced the correlations, some further analyses of the individual children's data has been undertaken. Due to the small number of sampling points per child, all three seasons have been combined for each of the children. It is not possible to draw any significant conclusions for the personal classroom or classroom outdoor correlations as so few of the individuals have sufficient data points so none of this data has been included.

When the correlations were analysed for the individual children's personal $PM_{2.5}$ and home $PM_{2.5}$ (as shown in Table 5.20), six of the children had significant associations between the personal and home exposures with coefficients ranging from 0.61 – 0.94. Reasons for the lack of association in the other 4 children are less clear as there are no significant associations with any of the other microenvironments sampled, further analysis of the time activity diaries will be undertaken to try to identify activity patterns. It is possible that the classroom exposure is the most significant location responsible for the majority of these children's exposure but because of missing data or lack of power in the study design these relationships may have been missed.

When analysing the personal PM_{10} and home PM_{10} correlations there were only four of the children with significant correlations, children 3, 4, 8 and 10 with coefficients ranging from 0.74 – 0.77. The activities and children's activity patterns within the homes need to be evaluated further to identify any specific links to exposure, see Section 6.

When analysing the correlations by child for school and non school days there was insufficient power to determine if there were significant differences between the types of days.

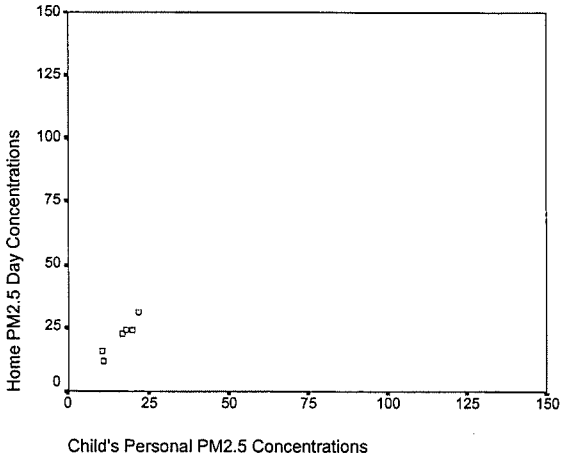
Table 5.20 Correlations of Individual Children's Personal Exposure Concentrations with Home Concentrations.

Child Identification Number Correlations	Child $PM_{2.5}$ & Home $PM_{2.5}$			Child PM_{10} & Home PM_{10}		
	n[I]	r	p	n	r	p
1	11	0.73	0.01	12	0.44	0.15
2	11	0.48	0.13	9	0.55	0.13
3	9	-0.12	0.76	10	0.77	0.01
4	11	0.64	0.03	11	0.74	0.01
5	9	0.81	0.01	9	-0.18	0.64
6	4	0.74	0.26	5	0.70	0.19
7	7	-0.26	0.57	5	0.00	1
8	6	0.94	0.01	9	0.75	0.02
9	10	0.62	0.05	10	0.27	0.45
10	13	0.61	0.03	15	0.74	0.01
Median		**0.63**			**0.66**	

There is variation between children's exposure patterns which was suggested in Section 5.4, this diversity between the associations of personal and home concentrations would also suggest that using this indoor environment to predict children's general exposure would be unreliable.

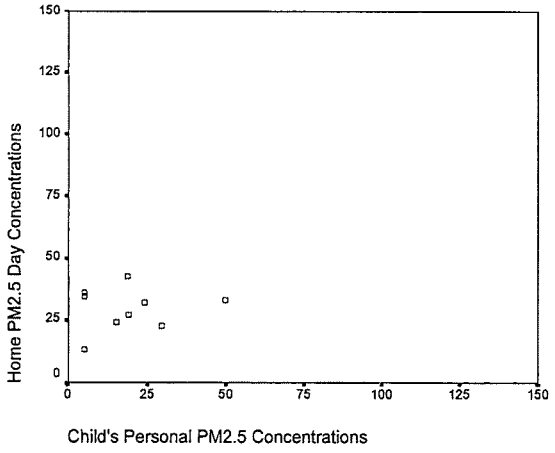
Analysis of the children's time activity diaries will also provide insight into their exposure patterns and how this could influence their personal exposure, Section 5.10. The household activity questionnaire data will be analysed in Chapter 6 along with the other source apportionment data analysis.

[I] Number of data points



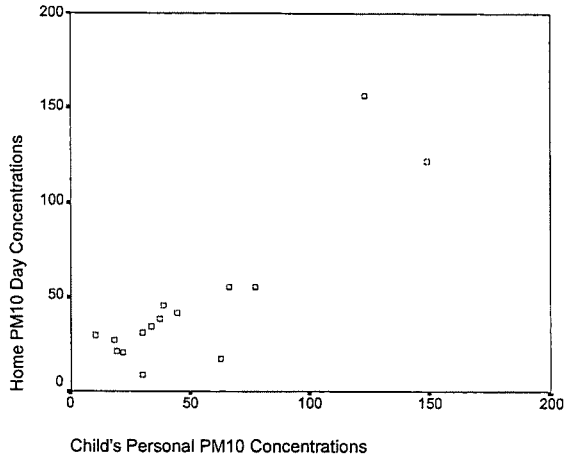
$r = 0.94$ $p = 0.01$ $n = 6$

Figure 5.10 Child 8 Personal $PM_{2.5}$ and Home $PM_{2.5}$ Strong Correlation ($\mu g/m^3$).



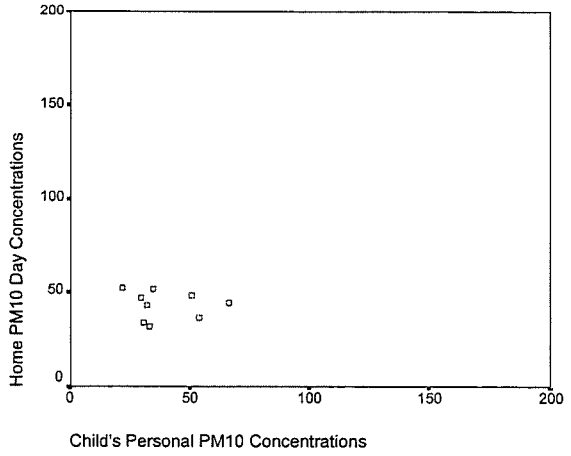
$r = -0.12$ $p = 0.76$ $n = 9$

Figure 5.11 Child 3 Personal $PM_{2.5}$ and Home $PM_{2.5}$ Weak Correlation ($\mu g/m^3$).



$r = 0.74$ $p = 0.01$ $n = 15$

Figure 5.12 Child 10 Personal PM_{10} and Home PM_{10} Strong Correlation ($\mu g/m^3$).



$r = -0.18$ $p = 0.64$ $n = 9$

Figure 5.13 Child 5 Personal PM_{10} and Home PM_{10} Weak Correlation ($\mu g/m^3$).

5.9 Wilcoxon Signed Rank Tests

The Wilcoxon Signed Rank Test is a non-parametric procedure used with two related variables to test the hypothesis that the two variables have the same distribution. It makes no assumptions about the shapes of the distributions of the two variables. This test takes into account information about the magnitude of differences within pairs and gives more weight to pairs that show small differences. The test statistic is based on the ranks of the absolute values of the differences between the two variables (Noruses, 1993).

A null hypothesis was assumed for the tests that there was no significant differences between the variables, hence if the hypothesis is rejected it can be assumed that the variables are different. The complete data sets for these tests are found in Appendix 11, only those accepted variables are included in Table 5.21.

The test provides further evidence for the association between the personal PM_{10} concentrations with the home and school PM_{10} data. The personal $PM_{2.5}$ does not indicate that there was a significant relationship with the indoor environments as was suggested by the correlation analyses in Section 5.8.4. The median r value for both of the $PM_{2.5}$ correlations at home and school were lower than the corresponding PM_{10} data. This suggests that there may be a greater variation due to specific activities that the child is exposed to in the home and classroom especially from the resuspension of PM_{10}.

Table 5.21 Summary of Wilcoxon Signed Ranks Tests, Accepted Variables

			n	Null Hypothesis (Accept/Reject)
Personal PM_{10}		**Home PM_{10} Day**	114	Accept
		School PM_{10} Day	50	Accept
Garden PM_{10}	**Day**	**Haringey PM_{10}**	150	Accept
	Night	**Haringey PM_{10}**	150	Accept
Home $PM_{2.5}$		**School $PM_{2.5}$**	46	Accept

The test indicates that there is no difference between the garden PM_{10} concentrations and the Haringey AUN site during the day or night. There is a significant difference however between the garden PM_{10} and Brent AUN site, the Brent and Haringey AUN sites also indicate that there is a significant difference between them. This suggests that the use of a single monitoring site in Barnet would be insufficient to assess the outdoor PM_{10}

concentrations as previously assumed in Section 5.8.1. Issues of point sources, as described in Section 2.9, may influence the background sites. Meteorological conditions may also have influenced the sources of particulate matter from outside of the sampling area especially if the prevailing winds were from areas with higher or lower background concentrations than those experienced in the immediate sampling area.

The Wilcoxon test showed that home PM_{10} concentrations were again not associated with any outdoor sources indicating that the sources of this size fraction were predominantly found within the home. The home $PM_{2.5}$ concentrations appeared to be associated with the school $PM_{2.5}$ concentrations, however this is likely to be a result of the fact that there is little spatial variability for this size fraction, see Table 5.11. The variation between these two environments may be small and hence the Wilcoxon test does not find them to be significantly different.

The Wilcoxon Signed Ranks Tests support the previous correlation analyses that the school concentrations for the PM_{10} size fraction cannot be attributed to outdoor sources. It does not support the correlation analysis for the $PM_{2.5}$ size fractions which may be due to the wide range in r values found for individual children.

The Wilcoxon Signed Rank Test has provided further evidence to support the Spearman's Rank Correlations that were undertaken in Section 5.8. Some of these associations have been disputed and suggest that the outdoor sampling sites are independent of each other, which could be a result of point sources. The unusual finding that the classroom $PM_{2.5}$ results are independent of the outdoor concentrations despite the strong correlations when using the Spearman's test are difficult to understand especially as there are unlikely to be any combustion sources for this size fraction within the classroom.

5.10 Time Activity Diary Analysis

The analysis of time activity diaries has been used to assess the activity patterns of children living within an urban area of London. The data in the diaries indicates how much time was spent indoors at home, school and other environments, all time spent outdoors and the time spent in enclosed transportation (includes cars, buses and trains, walking and cycling was classed as time spent outdoors). The data in Table 5.22 represents the children's personal exposure sampling time only, the rest of the time at night assumed that the children were at home.

All the children completed diaries prior to the sampling period so that they could become accustomed to completing them accurately. Janssen (1998) and Mage (1991) both found that adults tended to alter their activity patterns when involved in personal exposure monitoring, reducing the amount of time spent outside, changing their activities due to the weight of the bag and through being self-conscious. Janssen's study involving children did not find this to be the case. As a result of these findings it was assumed that the children in this study would not alter their activities either. By encouraging both parents and teachers to assist in this study the children's compliance was ensured. The diary data for all seasons and children is shown in Figure 5.14. The total sampling time spent indoors is 79% which compares to about 82% for the Janssen (1998) study. Other evaluations of activity patterns have been assessed and these indicate about 87.2% of individuals time is spent indoors (Wallace, 1996a). This refers to an adult population living in the U.S. so it is not representative for UK children.

The seasonal analysis of the data represented in Table 5.22 indicated that there were some differences in the children's activity patterns throughout the different seasons. Two sided Student T-Tests assuming equal variance of the data were conducted for environments and seasons. The only occasion where the assumption of equal variance was void occurred between spring and summer in the classroom. The complete analysis is in Appendix 12. The variations in activity patterns were only significantly different whilst the children were at school. Activity patterns were significantly different between winter and summer, and for spring and summer. The amount of time spent at school and the activities at school were unlikely to change significantly throughout the school year as illustrated by the lack of significant differences between the winter and spring. The summer sampling period encompassed the majority of the holiday period when most of the children were not attending school, which would account for these seasonal differences in activity patterns.

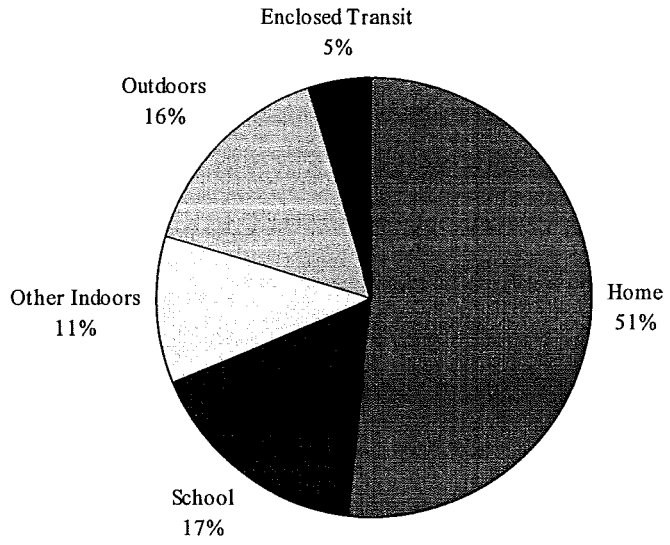


Figure 5.14 All 3 Seasons Time Activity Patterns for All Children.

The variability in personal exposure that was assumed to be a result of changes in the children's activity patterns, as discussed in Section 5.4, does not appear to be evident. There is no significant difference between the seasonal amounts of time spent outside.

The effects of rainfall during the spring, see Table 5.18, may therefore be the most influential factor on indoor concentrations, which in turn may be responsible for the differences in personal exposure as all of the analyses indicate a strong association between personal and indoor concentrations.

Table 5.22 Time Activity Diary Patterns for All Children during Sampling Period.

Season	n	Home (min.)	School (min.)	Other Indoors (min.)	Outdoors (min.)	Enclosed Transit (min.)
Winter	49	390	124	73	95	29
	%	56	17	10	13	4
Spring	55	350	144	68	107	34
	%	49	21	10	15	5
Summer	45	357	66	104	123	44
	%	52	9	15	18	6
Annual	149	362	121	76	109	34

The influence upon time spent inside, outside and time spent travelling by each child has been investigated in an attempt to identify if there are any relationships between these activity patterns and particulate concentrations. A summary of the day time values for the personal, home and classroom along with specific factors that may influence particulate generation and time spent outside and travelling are found in Table 5.23.

There does not appear to be a single factor that can explain why particle concentrations are either high or low. Child 1 has the highest PM_{10} concentrations for personal and home, a potential reason for this may be the presence of a dog and the lack of house cleaning that was evident. Resuspension of this size fraction from the carpets and furnishings as the child moved around the home is likely to impact upon personal exposure concentrations. Child 9 has some of the highest personal exposures, the $PM_{2.5}$ exposure could be explained by the high school $PM_{2.5}$ concentrations, however the PM_{10} concentrations are less easily explained. The child spent a quarter of their time indoors elsewhere which included music school and guides which have many other children present and could result in the resuspension of PM_{10} however these environments were not sampled so it is not possible to verify this fact.

The children that have a low personal exposure to particulate matter do not appear to behave differently to the other children. The concentrations in the home and classroom appear to have more of an influence upon their exposure than activity patterns.

There are only ten participants in this study and their activity patterns may not represent those activity patterns of other children living within Barnet.

Table 5.23 Summary of Particulate Concentrations and Potential Influencing Factors.

	Personal ($\mu g/m^3$)		Class ($\mu g/m^3$)		Home ($\mu g/m^3$)		Characteristics	Time Outside %	Travel Time %
Child	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}			
1	19	80	28	85	31	79	Dog / Dusty home	24	3
2	19	43	33	85	30	54	Parent smoked / Home next to M1	16	5
3	17	64	19	92	28	79		9	9
4	16	54	29	83	18	47	Parent smoked /No downstairs' carpets /AER 1.2	16	4
5	15	43	23	73	19	43	Cats	20	6
6	12	46	25	67	17	53	Poor compliance	10	1
7	16	35	43	58	19	38		14	6
8	17	67	19	93	21	47		19	7
9	27	72	37	69	18	28	25% time spent in other indoors	14	5
10	17	38	22	81	24	42	Parent smoked	13	1

5.11 Time Weighted Averages to predict Personal Exposure

Using the time activity data along with the microenvironmental concentrations it is possible to attempt some indirect personal exposure estimates. This method uses a time averaged approach to estimate personal exposures using average microenvironmental concentrations and the total time spent in each microenvironment. As stated previously, there are only 70 observations where all corresponding microenvironments were successfully captured, interpretation of the results for a wider population is not possible.

Using three different models based on the following model;

$Conc_{tw} = ((Conc_{class} * time_{class} + time_{home} * Conc_{home} + time_{garden} + (720 - time_{class} - time_{home} - time_{garden}) * Conc_{other}))/720$

The concentration other was multiplied by either home or garden or another concentration. Model 1 uses the home concentration to estimate any unknown exposure whilst Model 2 uses the garden and Model 3 uses a more conservative approach and divides by the time spent in each environment.

<u>$PM_{2.5}$ Model to estimate personal $PM_{2.5}$ exposure.</u>

Model 1 - Home concentration

Model_1=((school$PM_{2.5}$*class+home*home$PM_{2.5}$+garden*garden$PM_{2.5}$+(**720**-class-home-garden)* home$PM_{2.5}$))/**720**

Model 2 – Garden concentration

Model_2=((school$PM_{2.5}$*class+home* home$PM_{2.5}$+garden* garden$PM_{2.5}$+(**720**-class-home-garden)* garden$PM_{2.5}$))/**720**

Model 3 – Time spent in each environment

Model_3=((school$PM_{2.5}$*class+home* home$PM_{2.5}$+garden* garden$PM_{2.5}$)) / (class+home+garden)

Table 5.24 Time Weighted Averages for Personal Exposure Concentrations ($PM_{2.5}$)

Spearman Correlation Coefficients Prob > |r| under H0: Rho=0 Number of Observations

		pem2_5	Model 1	Model 2	Model 3
pem2_5	r	1.00000	0.49623	0.53219	0.51084
	p		<.0001	<.0001	<.0001
	n	126	70	70	70

Model 1 results show that the estimated personal exposure when compared to the actual personal exposure is underestimated. This is shown by the r = 49.6%. Model 2 uses the garden concentrations instead of home concentrations. This has the best estimate explaining 53.2% of the personal exposure. Model 3 looks just at the time spent in each environment. This improves upon Model 1 explaining 51.1%.

PM_{10} Model to estimate personal PM_{10} exposure.

Model 1 - Home concentration

Model_1=((schoolPM_{10}*class+home* homePM_{10}+garden* gardenPM_{10}+(**720**-class-home-garden)* homePM_{10}))/**720**

Model 2 – Garden concentration

Model_2=((schoolPM_{10}*class+home* homePM_{10}+garden* gardenPM_{10}+(**720**-class-home-garden)* gardenPM_{10}))/**720**

Model 3 – Time spent in each environment

Model_3=((schoolPM_{10}*class+home* homePM_{10}+garden* gardenPM_{10})) / (class+home+garden)

Models 4 – 6 use the imputed garden PM_{10} values.

Model_4=((schoolPM_{10}*class+home* homePM_{10}+garden* imputed gardenPM_{10}+(**720**-class-home-garden)* homePM_{10}))/**720**

Model_5=((schoolPM_{10}*class+home* homePM_{10}+garden* imputed gardenPM_{10}+(**720**-class-home-garden)*garden10))/**720**

Model_6=((schoolPM_{10}*class+home* homePM_{10}+garden* imputed gardenPM_{10})) / (class+home+garden)

The PM_{10} models 1,2 and 3 are all the same as the $PM_{2.5}$ models. Models 4,5 and 6 use an imputed value for the garden where any values are missing, this was created using the mean of the 2 AUN sites for that corresponding day. This increases the n from 69 to 87. Due to the variability of the concentrations in the home and at school it is not possible to derive an imputed value for these environments as this would increase the uncertainty of the data, however the correlations between the garden and AUN sites are all significant so there is less uncertainty in these estimates.

Table 5.25 Time Weighted Averages for Personal Exposure Concentrations (PM_{10})

Spearman Correlation Coefficients
Prob > |r| under H0: Rho=0
Number of Observations

			Model 1	Model 2	Model 3	Model 4
pem10						
pem10	r	1.00000	0.54396	0.50570	0.56003	0.61524
	p		<.0001	<.0001	<.0001	<.0001
	n	122	69	69	69	87

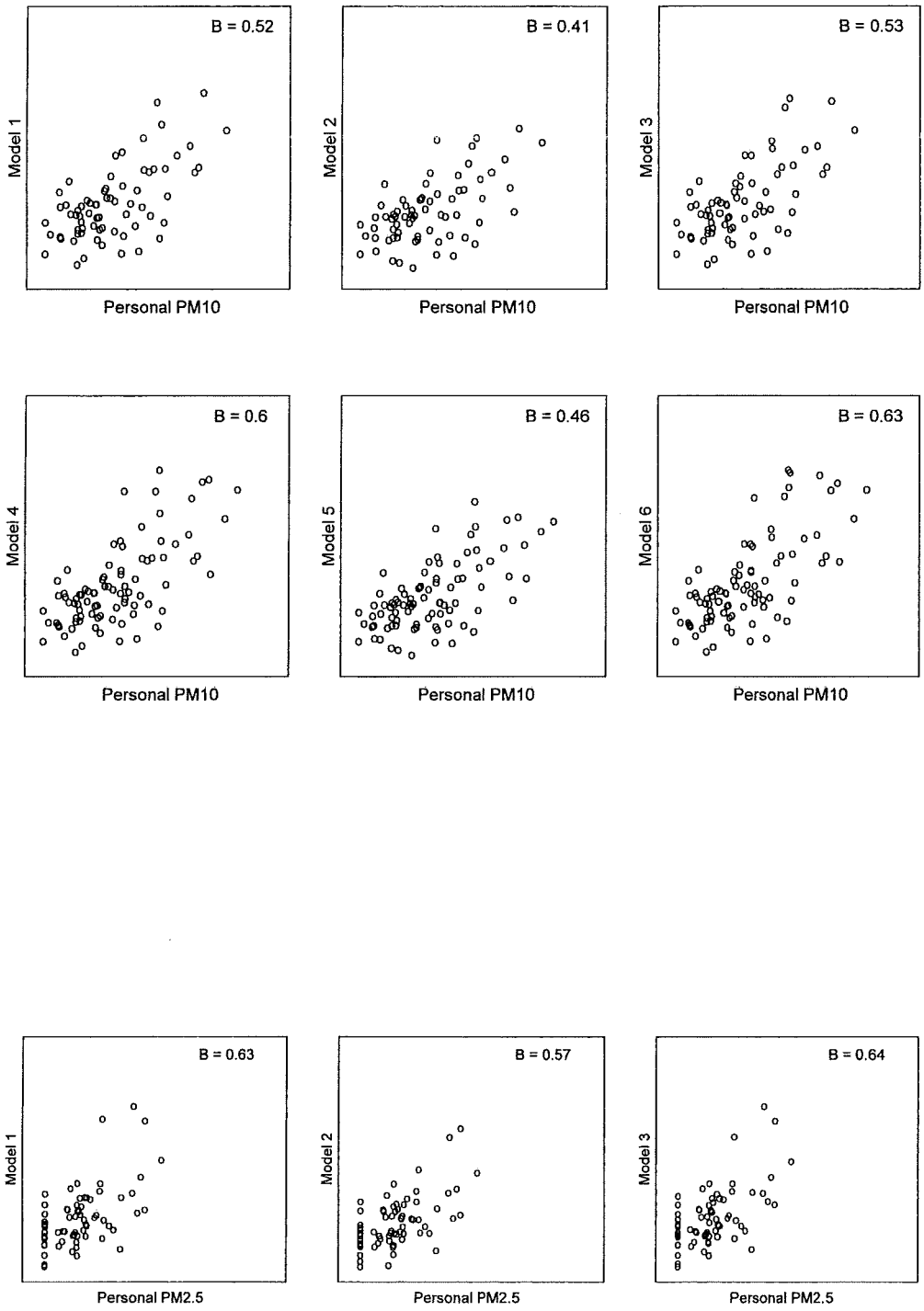
	Model 5	Model 6
pem10	0.57679	0.63327
	<.0001	<.0001
	87	87

Unlike the $PM_{2.5}$ time weighted averages the best model using the actual data is Model 3 which looks just at the time spent in each of the main environments rather than assuming that the home or garden concentrations can estimate the unknown concentrations. When the extra observations are imputed for the garden this model again provides the best estimate of personal exposure explaining 63.3% of the actual concentrations found.

Using scatter plots of the time-weighted outcomes with the actual personal exposures it is possible to see how the two compare using the different model structures. The PM_{10} scatter plot x and y axes are 0 to 200μg/m^3 whilst the $PM_{2.5}$ are 0 to 100μg/m^3. The B provides an estimate of the slope between the actual values and the calculated model responses.

The model results for both size fractions are not dissimilar to the correlation results discussed in section 5.8.1. When analysing the association between the personal and home environments for all children the r values were 0.59 and 0.52 for PM_{10} and $PM_{2.5}$ respectively. This would suggest that the home environment is responsible for much of the children's personal exposure. The model results range from 0.50 – 0.63 and 0.50 – 0.53 for PM_{10} and $PM_{2.5}$ respectively which would suggest that the time spent outdoors and in the classroom were less significant sources of exposure.

Figure 5.15 Scatter Plots of Time Weighted Averages ($\mu g/m^3$), Models 1 – 6 with Slope.



The scatter plots for all models are shown in Figure 5.15 it is evident from the slope (B) that the time-weighted averages consistently under predict the actual personal exposure concentrations. There is no evidence that there are any outliers driving this association.

Duan and Mage (1997) in their discussion of indirect estimates of personal exposure using microenvironmental averages suggest that this method is vulnerable to systematic measurement error. It does have advantages despite this as it allows cheaper measures of exposure and requires less burden on the subjects.

5.12 Discussion and Conclusions

The descriptive statistical analysis has highlighted a number of issues, some of which require further investigation in Chapter 6. The key issues in this chapter are summarised.

The methodology selected was successful in determining personal exposure with the majority of the data exceeding LOD, with comparable success rates to other similar research methods.

All of the PM_{10} concentrations exceeded the $PM_{2.5}$ concentrations. Personal exposure concentrations for the seasons were winter> spring> summer, suggesting that the children's exposure to both PM_{10} and $PM_{2.5}$ were influenced by season.

There was greater variability in personal exposure concentrations within and between children compared to all of the other sampled environments, this was especially evident when comparing school and non school day exposure patterns.

It is evident that it is not possible to directly assess children's 12-hour personal exposure to either PM_{10} or $PM_{2.5}$ using outdoor monitoring sites as these would underestimate the actual exposure concentrations. Other literature has reached the same conclusions and these have been discussed in greater detail in Chapter 2.

The home PM_{10} and $PM_{2.5}$ concentrations during the day and night were greater than the outdoor monitoring sites. Sources of particulate matter within the home require further investigation. It is not possible to use an outdoor monitoring site to assess indoor concentrations of PM_{10} and $PM_{2.5}$ as these would underestimate concentrations found within the home. At night there were significant correlations between the home and

outdoor concentrations for both size fractions. The classroom concentrations of PM_{10} and $PM_{2.5}$ during the day exceed the outdoor concentrations.

Ambient contributions of $PM_{2.5}$ to PM_{10} during the day were estimated at 56 % which is comparable to other UK research. The contribution of the classroom $PM_{2.5}$ fraction to the PM_{10} fraction is approximately 34% compared to the home where the value is 42% and outdoors is 55%. The lowest ratio of PM_{10} to $PM_{2.5}$ during the day was found for the personal exposure concentrations, this suggests that personal activities cause the resuspension of PM_{10}.

The outdoor sampling in the garden initially suggested with the correlation analysis that the relationship between the AUN sites and the garden were significant and hence a single monitoring site would be sufficient in Barnet to measure the outdoor concentrations. However, the Wilcoxon Signed Ranks Tests showed that there were significant differences between the two AUN sites and also between the garden and the Brent AUN site. Further investigation is required into whether local sources influence the AUN monitoring or whether meteorological conditions were responsible for this variation.

Personal PM_{10} exposure concentrations could be significantly associated with both the home and school. Personal $PM_{2.5}$ exposure associations were weaker than the PM_{10} when correlated with home and school. This suggests for the children included in this study that their personal exposure sources for PM_{10} were primarily from the indoor environment.

Children's activity patterns were not influenced by different seasons. The only activity patterns that were influenced by the seasons were time spent in the classroom and this could be a result of the summer sampling period falling during the majority of the children's holiday.

Time weighted averages of personal exposure estimated 53.2% of the actual $PM_{2.5}$ concentrations whilst the best estimate for the personal PM_{10} concentrations was 63.3%. All models under predicted the personal exposure concentrations as seen in the scatter plots.

6 Source Apportionment Analysis of the Particulate Matter

6.1 Introduction

This chapter analyses the source apportionment of the particulate matter collected during the study. Several techniques were employed for this with the intention to determine the potential sources of the children's personal exposure to particulate matter.

The indoor/outdoor (I/O) ratios, along with the air exchange measurements, were used to identify the potential influence that the outdoor concentrations of particulate matter had upon the indoor environment.

Data collected using the household questionnaire was analysed to assess the impact that specific activities may have had upon particle generation or resuspension of particulate matter within the home.

Physical analysis of a small sub sample of the filters has been performed using Scanning Electron Microscopy. This identified the potential differences in the particulate matter collected within different environments. A summary of the potential sources of particulate matter for children living within the Barnet area will be assessed.

A comparison of the personal exposure concentrations with the AUN legislation and potential health outcomes has also been conducted in accordance with the preliminary objectives as stated in Section 1.2.

6.2 Indoor / Outdoor Ratios

From the results presented in Chapter 5, it is evident that a number of sources of particulate matter influence the indoor concentrations. Weak correlations were found between both size fractions and the outdoor monitoring sites during the night, however no daytime correlations were found. The correlation between personal exposure and home concentrations for all children were shown to have some significance, although not highly significant the median $PM_{2.5}$ and PM_{10} values were 0.53 and 0.59 respectively. The personal and classroom correlations were also 0.51 and 0.35 respectively, hence the importance of understanding particulate sources within the indoor environment.

Table 6.1 shows the total ratios between the home and garden for both size fractions during the day and night. A number of studies have identified the ratio of particles found indoors and outdoors. These have been carried out in a number of different countries and for

different size ranges so some of them may not be directly applicable to the UK situation. Factors that are likely to influence this ratio are building design, including presence of air conditioning, outdoor concentrations and indoor particle generating activities, this was discussed in Section 2.6.

The ratios for both size fractions appear to be influenced by particle generating activities within the home, as the day ratios were higher than at night. Typically the PM_{10} ratios during the night were higher than the $PM_{2.5}$ ratios. This indicates that the generation and resuspension of PM_{10} within the home were influencing the relationship between the two particle size fraction concentrations.

The literature reviewed in Section 2.6 indicated that in homes with air exchange rates >2 that the indoor and outdoor ratios would show decreased variability. As the air exchange rates for the sampled homes does not exceed 2 air changes per hour some variability in the ratios shown in Table 6.1 are expected.

Table 6.1 Total Indoor/Outdoor Ratios and Air Exchange Rates for All Children.

Child ID	**Home: Garden $PM_{2.5}$**		**Home: Garden PM_{10}**		**Air Exchange Rate**
	Day	**Night**	**Day**	**Night**	**Air Changes/Hour**
1	3.38	1.25	4.02	1.45	0.4
2	1.97	1.32	1.35	1.32	0.6
3	3.16	1.19	3.07	4.3	0.8
4	3.5	1.89	2.83	1.85	1.2
5	1.92	0.87	2.13	1.33	0.8
6	1.53	1.55	2.63	1.68	0.55
7	2.02	2.47	3.02	1.5	0.7
8	1.98	1.09	2.22	1.22	0.45
9	1.56	1.17	1.41	1.95	0.5
10	3.94	4.68	3.67	2.94	1.05
Median	**2**	**1.285**	**2.73**	**1.59**	**0.65**

Table 6.2 Seasonal Analysis of the Indoor/Outdoor Ratios for All Children.

Child ID	Season	Home: Garden $PM_{2.5}$		Home: Garden PM_{10}	
		Day	Night	Day	Night
1	Winter	3.1	0.87	4.91	0.89
(ACH^{-1})	Spring	3.5	1.35	3.69	1.57
= 0.4	Summer	Not Sampled in Summer, Sampled for 10 Days in Spring			
2	Winter	1.37	1.19	0.82	1.77
(ACH^{-1})	Spring	2.79	1.26	1.89	0.98
= 0.6	Summer	1.34	1.50	1.36	1.15
3	Winter	1.94	1.27	3.97	5.86
(ACH^{-1})	Spring	1.16	0.86	2.11	1.04
= 0.8	Summer	5.10	1.66	3.34	7.10
4	Winter	*	*	2.54	*
(ACH^{-1})	Spring	2.66	1.03	2.62	1.81
= 1.2	Summer	4.89	2.54	3.21	1.88
5	Winter	Microbalance Problem			
(ACH^{-1})	Spring	2.72	0.94	3.14	2.09
= 0.8	Summer	1.12	0.79	1.12	0.77
6	Winter	0.95	1.44	1.70	0.99
(ACH^{-1})	Spring	1.68	1.62	3.32	2.14
= 0.55	Summer	Not Sampled in Summer			
7	Winter	*	4.92	3.73	0.93
(ACH^{-1})	Spring	*	*	*	*
= 0.7	Summer	2.02	1.77	2.66	1.69
8	Winter	*	1.09	3.48	*
(ACH^{-1})	Spring	2.76	1.09	3.79	1.08
= 0.45	Summer	1.39	1.10	1.65	1.26
9	Winter	2.07	1.14	*	*
(ACH^{-1})	Spring	1.75	1.45	1.36	2.48
= 0.5	Summer	0.78	0.90	1.47	0.88
10	Winter	2.37	5.91	1.26	4.93
(ACH^{-1})	Spring	7.10	3.28	6.54	3.37
= 1.05	Summer	2.77	3.51	3.06	2.40
Median	**Winter**	**2.01**	**1.19**	**2.54**	**1.77**
	Spring	**2.79**	**1.35**	**3.32**	**1.57**
	Summer	**2.06**	**1.50**	**3.06**	**2.4**
	Overall	**2.01**	**1.27**	**3.06**	**1.77**

When the ratios for the indoor / outdoor particle transfer were studied for any seasonal variations it appeared that the day ratios for both of the size fractions decreased during the summer, in several cases approaching 1 indicating that the indoor and outdoor concentrations were similar. A possible reason for this could be that the ventilation of homes during the summer sampling period was increased. The winter and spring ratios showed greater variation between day and night indicating that day time indoor particle generating activities and ventilation were potentially influencing indoor concentrations.

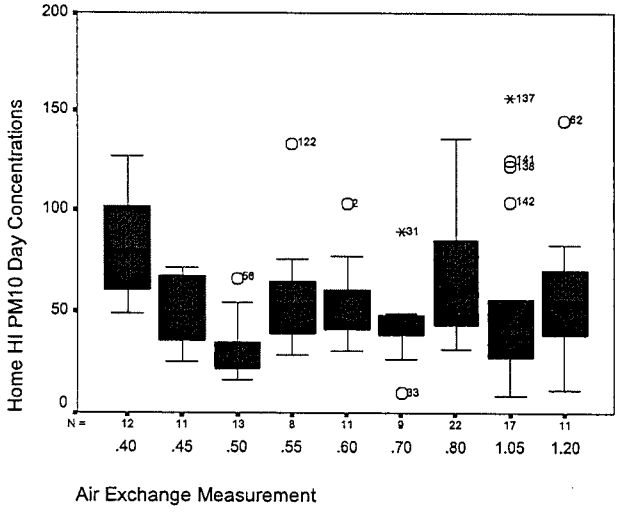
* Filter Problems, no samples

The results found in Table 5.18 show that the average ambient summer temperature was higher than the winter or spring. It could be assumed therefore, that ventilation of the homes was higher during the summer.

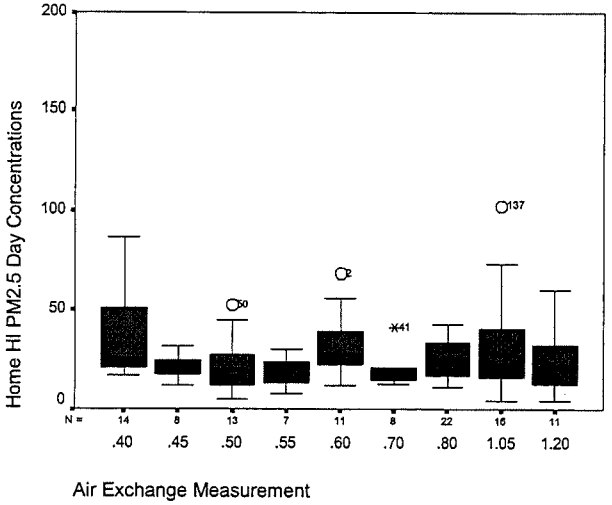
The air exchange measurements were also given for each of the houses. The air exchange measurements as described in Section 3.7.7 were a single measurement of the house. It was taken when all of the external doors and windows were closed and all internal doors were open, this represented the house at its most air tight. This situation is only likely to be reflected during the sampling period when the weather is cold and wet, i.e. most likely during the winter and spring periods. In the study by Kamens *et al.,* (1991) there was found to be no apparent relationship between indoor particle concentrations in the fine, coarse, or >10 μm size ranges and air exchange rates or outdoor wind speeds. It would appear that in Barnet there was also no relationship between the air exchange measurements and the two size fractions indoors at the different homes. Section 6.3.3 investigates the influence of daily ventilation changes on home concentrations of particulates.

The Figure 6.1 box plots show the home indoor mean concentrations and the air exchange measurements of the individual houses. The numbered points refer to data that were outliers. Figure 6.1 (a) represents the home PM_{10} concentrations and it can be seen that the range of the concentrations was greater than those found in Figures 6.1(b-d). This suggests that the majority of the particle generating activities occurred during the day and that air exchange rates did not influence the concentrations of PM_{10} within the home.

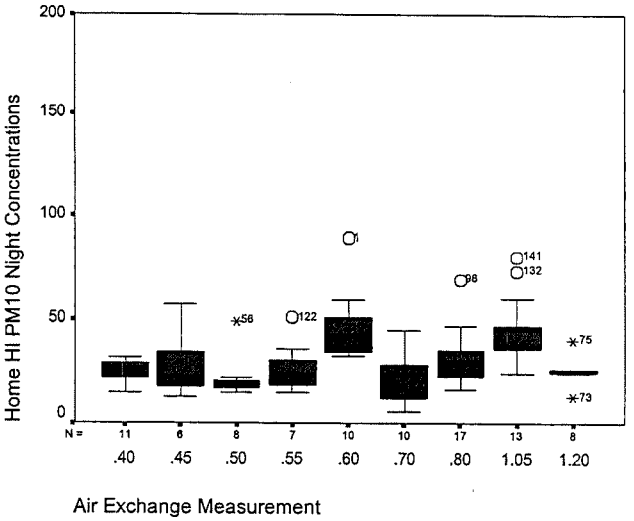
The $PM_{2.5}$ home day ratios were distributed evenly throughout the different air exchange rates with no significant variations being evident. This would suggest that there was no effect upon $PM_{2.5}$ concentrations within the home related to air exchange frequency. At night there appears to be no trends in the ratios that would suggest the air exchange rate has any influence upon indoor ratios for either size fractions. There does however, appear to be an effect from cigarette smoking. The two homes that have an adult who frequently smoked during the study were the homes with 0.6 and 1.05 ach^{-1}. The ratios at night for these two households were similar to the daytime ratios. The night time ratios of $PM_{2.5}$: PM_{10} have been discussed in Section 6.2 and Table 6.1 shows that the ratios within the home were lower during the night than the day except for the two homes where smoking occurred. The ratios indicate that about 50% of the PM_{10} is a result of the $PM_{2.5}$ which could explain the increase in the PM_{10} home concentrations seen in Figure 6.1c.



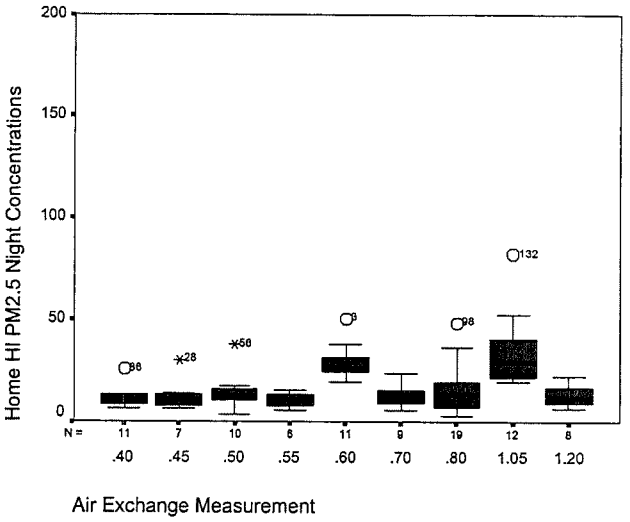
a)



b)



c)



d)

Figure 6.1 (a-d) Air Exchange Measurements (ach^{-1}) and Home Particulate Concentrations ($\mu g/m^3$).

6.3 Particle Generating Activities

Previous results, and the reviewed literature in Section 2.6, suggest that indoor activities potentially influence indoor/outdoor ratios of both size fractions. To investigate these potential sources data from the household questionnaire for cooking, including frying, grilling, baking and boiling, were aggregated into whether they had occurred during the sampling period. All cleaning, which included vacuuming, dusting, washing / drying of clothes and use of household cleaning sprays, were also aggregated to determine whether they had occurred during the sampling period. The other factors queried by the questionnaire were aggregated in a similar manner and the data for all of the children was analysed using box plots. This aggregation of the household data reduces the ability to recognise the peaks associated with specific particle generating activities, however, as the sampling period was integrated over a 12-hour period, it is unlikely that these activities will be significant in their effects.

6.3.1 Influence of ETS

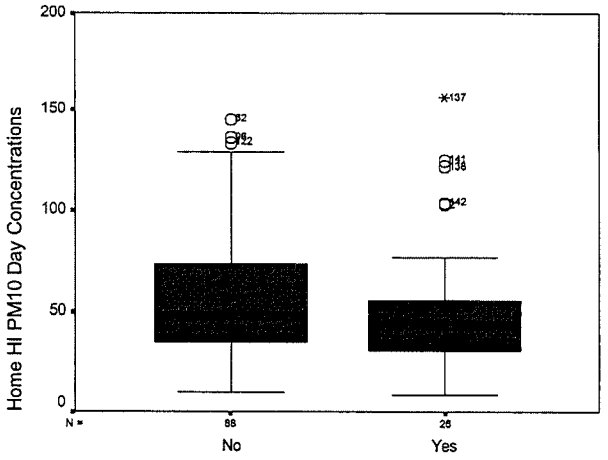
This study suggests that the homes with frequent smokers present have higher concentrations of particulate matter, especially $PM_{2.5}$, during the night than those homes without any smokers present. Another factor that may influence particulates associated to ETS is that smoking occurred more frequently during the evening when the smokers returned home from work. Some smoking occurred during the day however, the effect was more noticeable in the night sampling. All of this data was aggregated and the box plot of incidences when smoking occurred is shown in Figure 6.2 (a-d). Figures 6.2 (a) and (b) show the influence that the smoking incidences have upon day concentrations and these showed no significant effects upon indoor particulate matter concentrations. At night the influence of smoking upon home $PM_{2.5}$ and PM_{10} concentrations was more evident as seen in Figures 6.2 (c-d). The ratios of $PM_{2.5}$ to PM_{10} as discussed already in Section 6.2 suggest that the influence of $PM_{2.5}$ on the concentration of PM_{10} was greater at night. Also, there was less of an effect from other particle generating activities that occurred throughout the day.

When the box plots of smoking were plotted against the children's personal exposure as seen in Figure 6.7 (a) and (b), there would appear not to be a relationship. As the personal exposure concentrations were only sampled during the day it may be that the effects of other particle generating activities were overshadowing any smoking effects. This appears to be the case in the home and is also likely for the personal exposure concentrations.

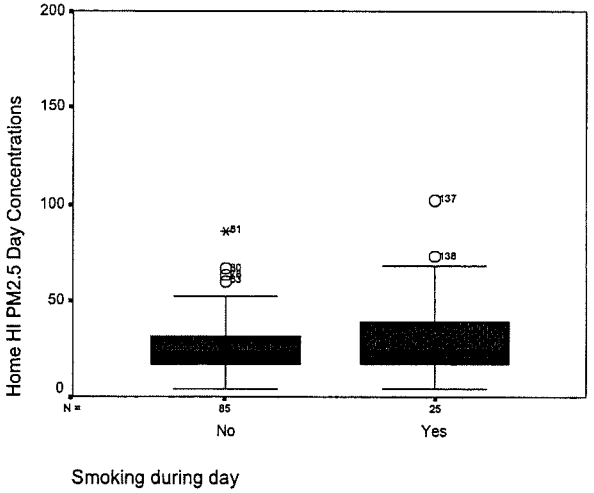
From the box plots, it would appear that there is some evidence that smoking influences the indoor concentrations, especially during the night time sampling. As the personal exposure of children assumed that personal exposure at night was the equivalent of the indoor concentrations then the children exposed to ETS could potentially have higher personal exposure to particulate matter. This is important when considering further research and analysis. When modelling the impact of specific activities upon exposure concentrations factors such as exposure to ETS and housing characteristics are essential components.

6.3.2 Influence of Cleaning and Cooking

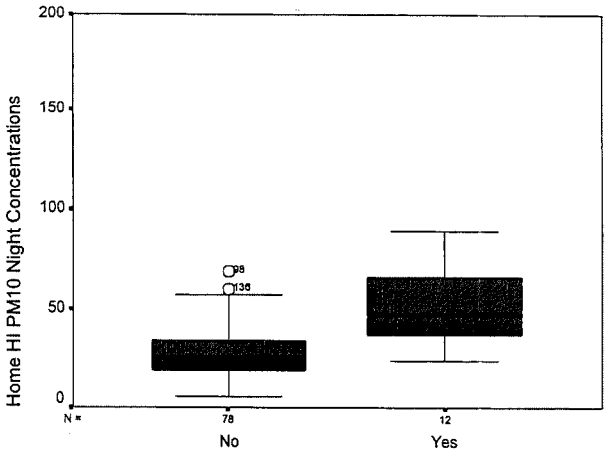
None of the other particle generating activities appeared to have any significant influence upon the concentration of either size fractions within the homes. Cleaning showed no significant effects upon particle generation as can be seen in Figure 6.3 (a-d). The effect of cooking upon PM_{10} concentrations during the day was quite varied and this is seen in Figure 6.4 (a) where there was a wide range of concentrations. A possible explanation is that the different types of cooking that took place were emitting different concentrations and sizes of particles, the equipment used integrates the sample over the 12 hours so any peaks related to cooking and cleaning were lost. This was discussed in Section 2.6.



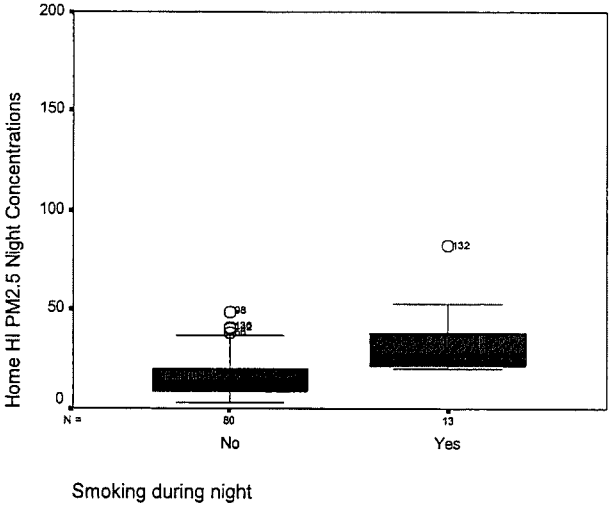
a)



b)

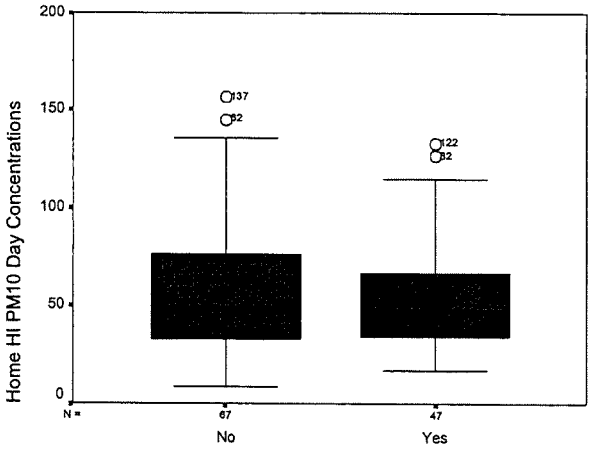


c)

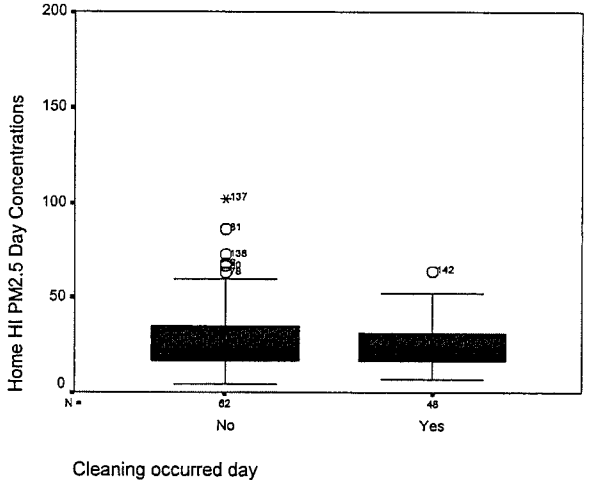


d)

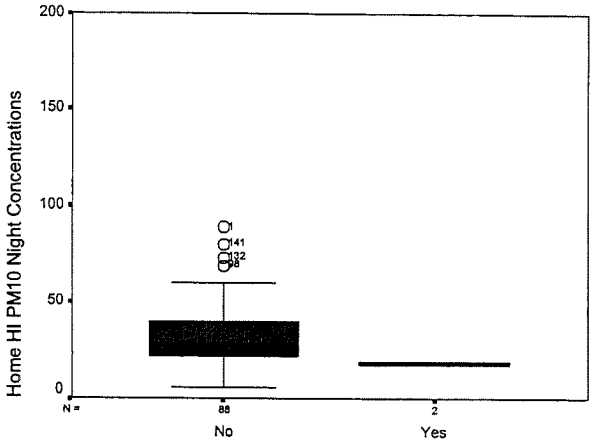
Figure 6.2 (a-d) Smoking Incidences and Home Particulate Concentrations ($\mu g/m^3$).



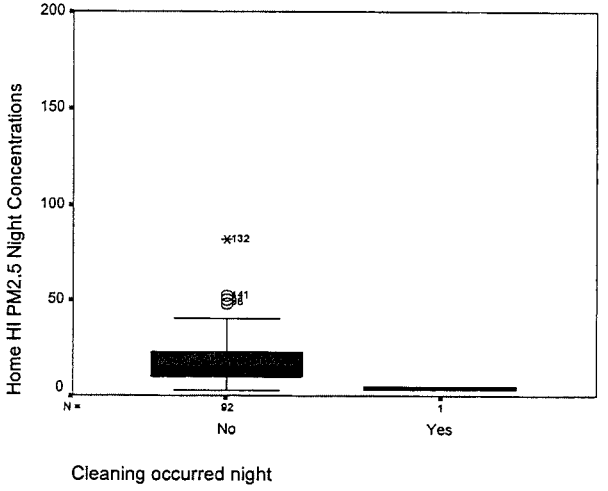
a)



b)

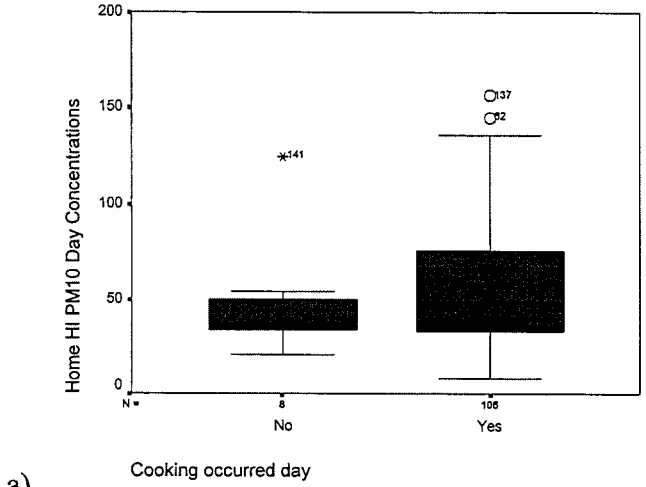


c)

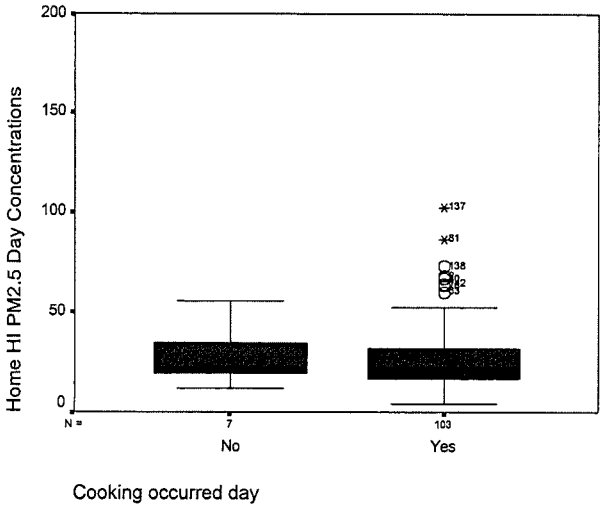


d)

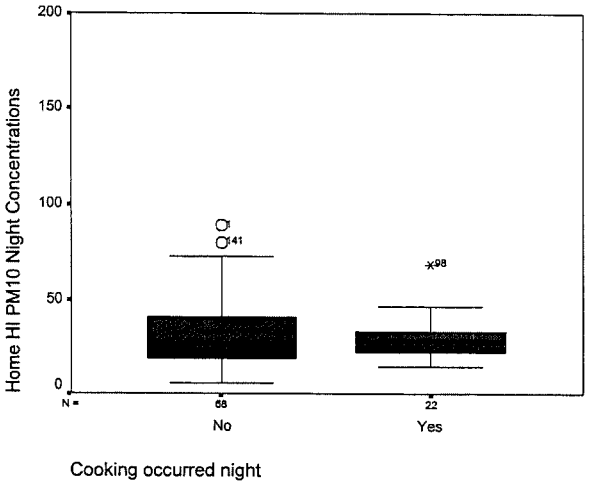
Figure 6.3 (a-d) Cleaning Activities and Home Particulate Concentrations ($\mu g/m^3$).



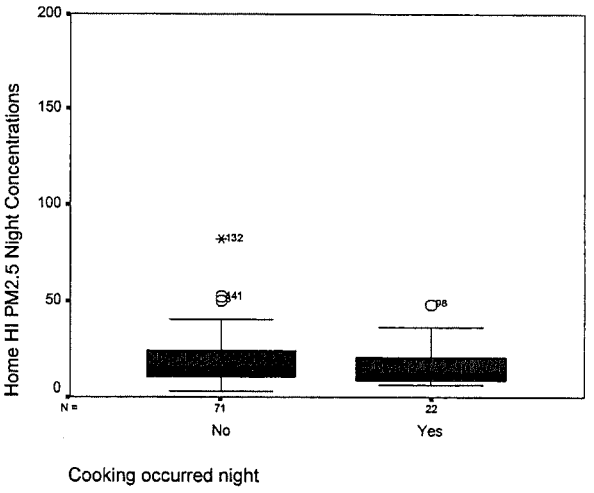
a)



b)

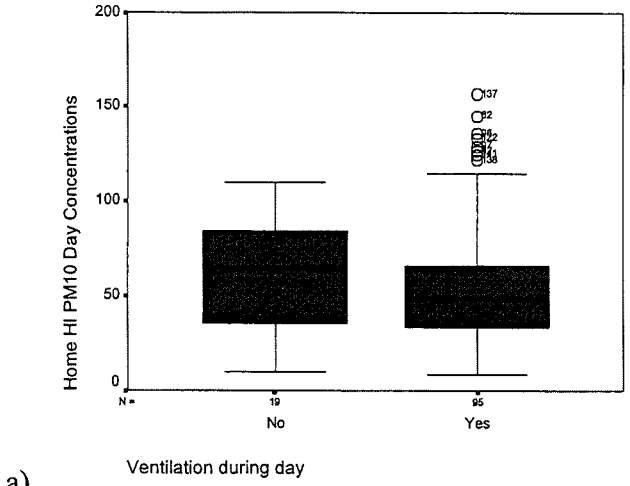


c)

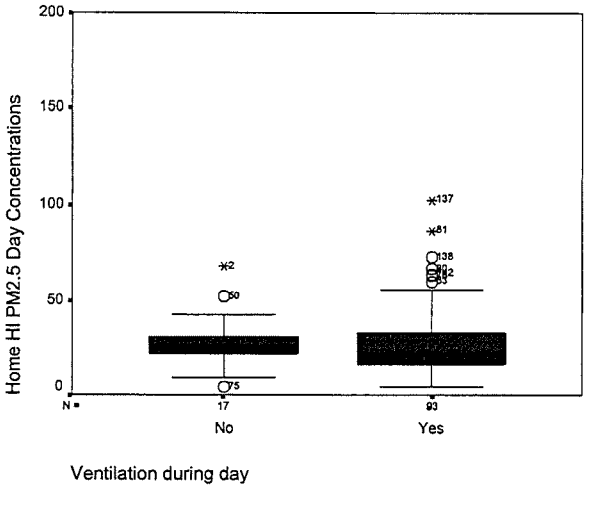


d)

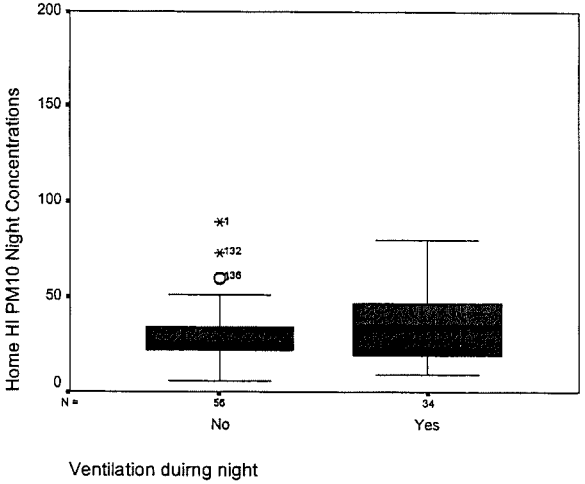
Figure 6.4 (a-d) Cooking Occurrence and Home Particulate Concentrations ($\mu g/m^3$).



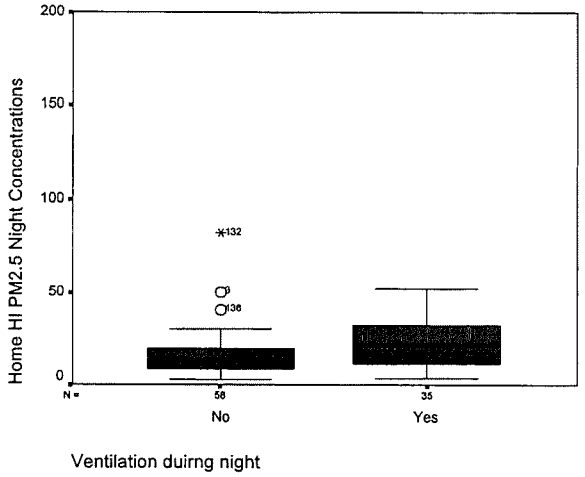
a)



b)

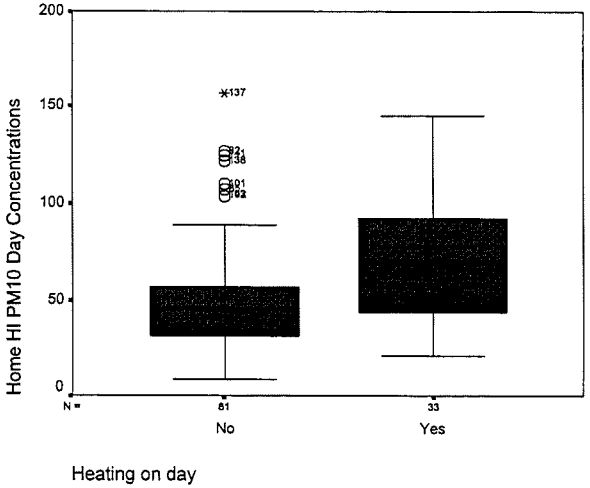


c)

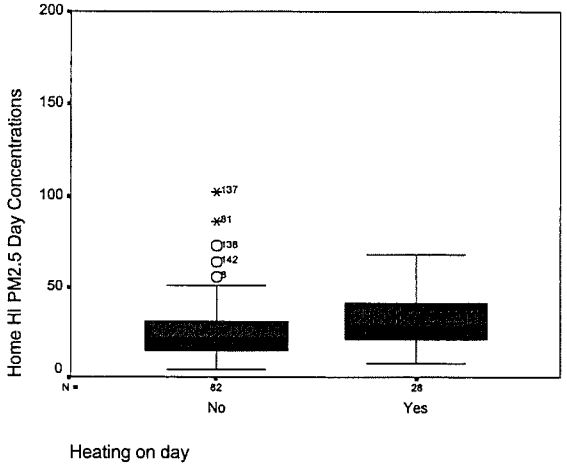


d)

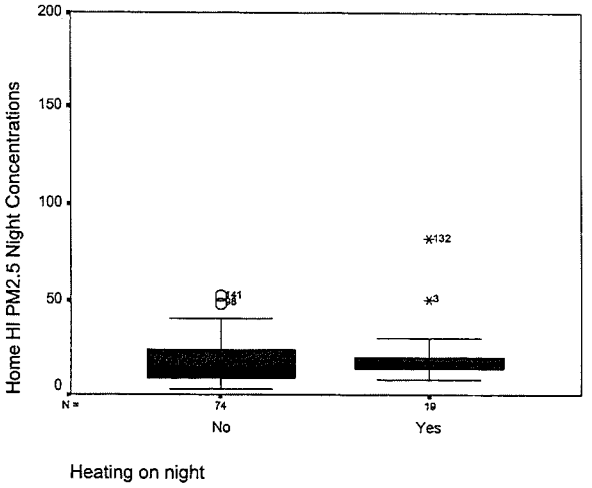
Figure 6.5 (a-d) Ventilation and Home Particulate Concentrations ($\mu g/m^3$).



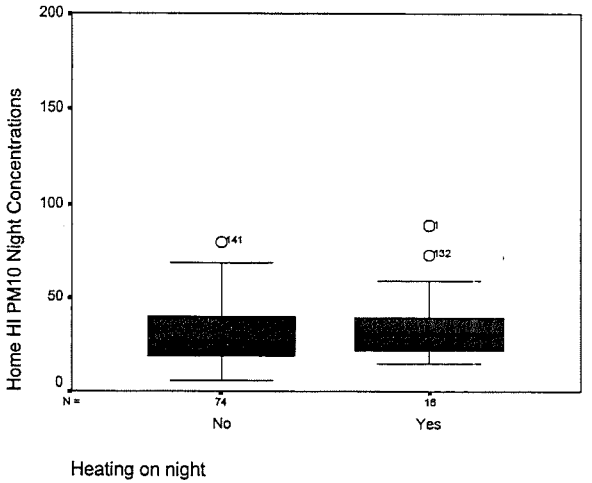
a)



b)



c)



d)

Figure 6.6 (a-d) Heating and Home Particulate Concentrations ($\mu g/m^3$).

6.3.3 Influence of Heating and Ventilation

There appears to be some relationship between the heating and concentrations of particulate matter in the different homes. This was shown when comparing the concentrations when the heating was on with when the heating was off, the results of the one way analysis of variance showed a marginally insignificant F test of 0.0575 for the $PM_{2.5}$ home concentrations whilst the home PM_{10} F test was significant at 0.002 during the day. This suggests that the PM_{10} concentrations during the day were influenced by whether there was any heating on. This could be expected as during the day most particle generating activities occur and if the heating was on it would not be expected for the windows to be open as well. This could lead to a build up of larger sized particles within the homes.

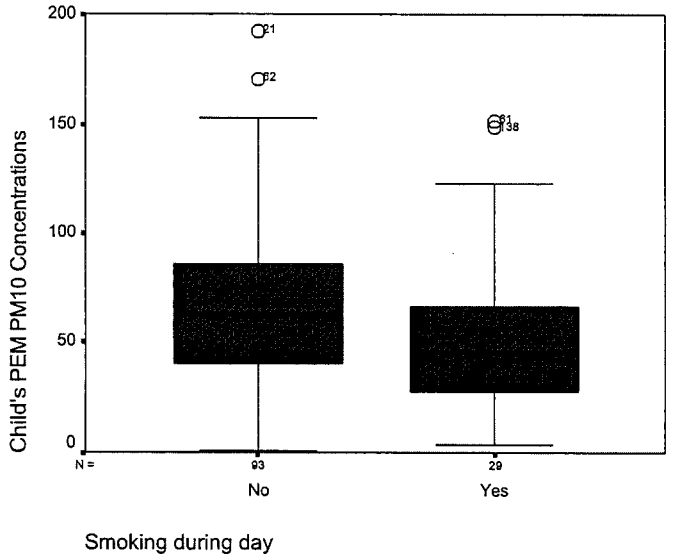
The $PM_{2.5}$ ratios of indoor to outdoor particles were lower during the night when little or no generation of PM_{10} was occurring. During the day, in the summer sampling period, the lowest ratios were found when the homes were well ventilated. The median ratio of $PM_{2.5}$ to PM_{10} was lower in the homes at night than during the day, as seen in Table 5.9 and Table 5.10. The influence of coarse particle generation within the home is the likely cause of this difference in these ratios, especially as the smaller particle size fractions are less likely to be influenced by building effects. The results of the one-way analysis of variance were both non significant.

The personal exposures and heating and ventilation appear to show a similar relationship, as seen Figure 6.8 and Figure 6.9. As these factors appear to have some influence upon the concentrations of particulate matter within the home there is also the likelihood that they will influence the children's personal exposure concentrations during the day, especially for children that spent a lot of time at home.

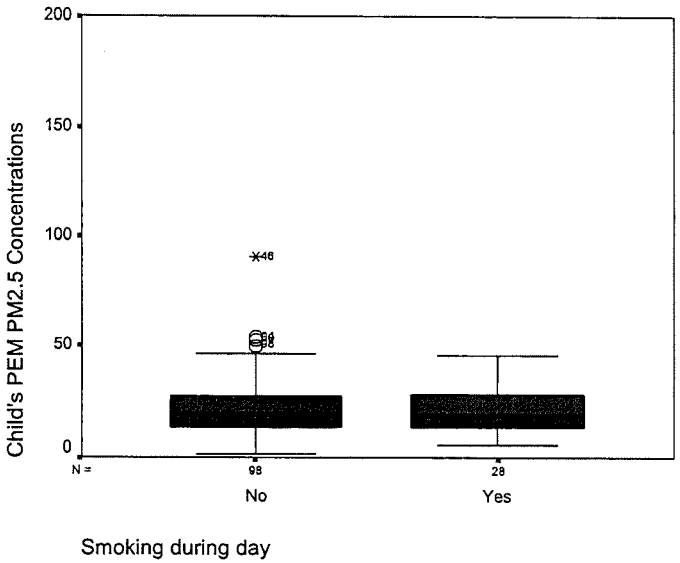
The influence of ventilation upon the indoor to outdoor ratios, particularly for PM_{10}, appears to be an important factor in source apportionment. To investigate this relationship further the ventilation times in all homes have been estimated from the questionnaire data and categorised into well, moderate and poor ventilation. This was achieved by aggregating the home ventilation for all children on all days into tertiles. The ventilation rates of the homes show some seasonal differences when plotting the indoor against the outdoor concentrations. When comparing the well ventilated to the moderate and poorly ventilated homes for the PM_{10} size fraction the data in the summer graph, see Figure 6.10 (c) shows a

closer relationship to the 1:1 line. This would suggest that during the summer the indoor and outdoor concentrations reflect each other more closely in homes that are well ventilated. This would support the facts stated in Sections 5.5 and 6.2 that during the winter and spring there is less ventilation and indoor sources of PM_{10} lead to a build up of particles.

The differences between the particle size fractions and ventilation categories during the night indicate that there is more of an effect upon indoor to outdoor ratios at night in poorly ventilated homes as compared with well-ventilated homes. This could be a result of the lack of particle generating activities that occur at night in the homes so that the particle concentrations reflect the outdoor concentrations for both size fractions. The greater variability may also be a result of the different air exchange rates in the individual homes. This could influence the pattern of deposition of the particles in each home Figure 6.1 illustrates the variability in particle concentrations and air exchange measurements. The home concentrations for both size fractions were lower at night compared to the day and the ratios shown in Table 6.1 also indicate there was more of a difference between the indoor and outdoor particle concentrations.

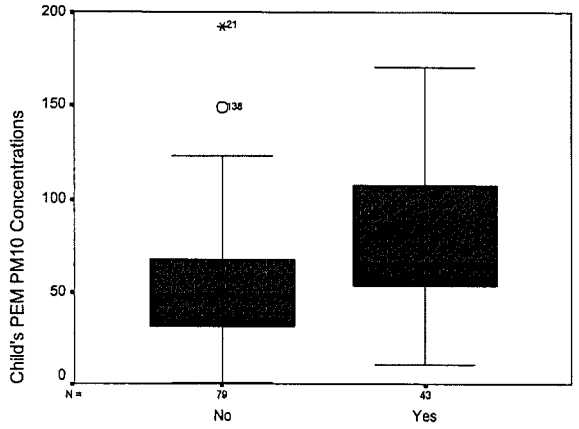


a)

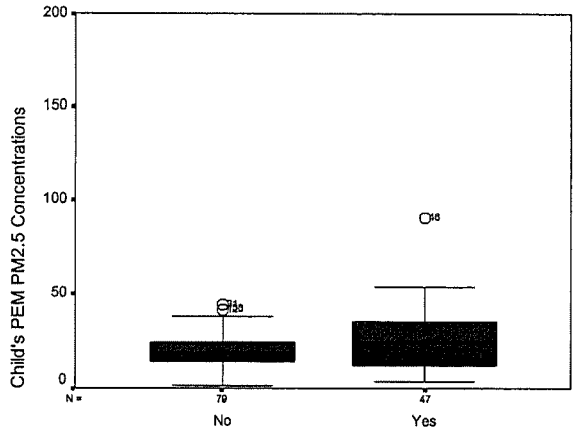


b)

Figure 6.7 (a-b) Smoking occurrence and Personal Exposure Concentrations ($\mu g/m^3$).

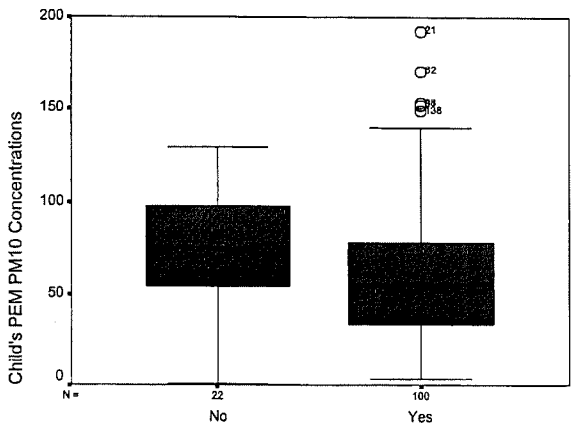


a)

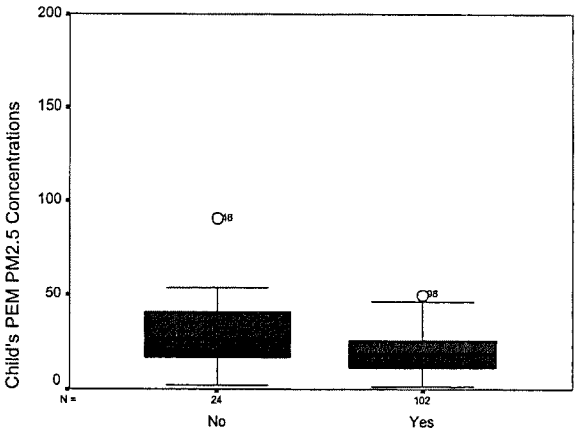


b)

Figure 6.8 (a-b) Heating and Personal Exposure Concentrations ($\mu g/m^3$).

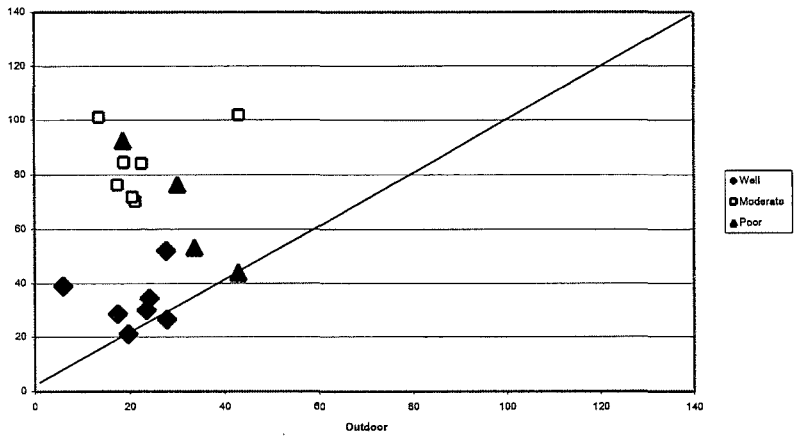


a)

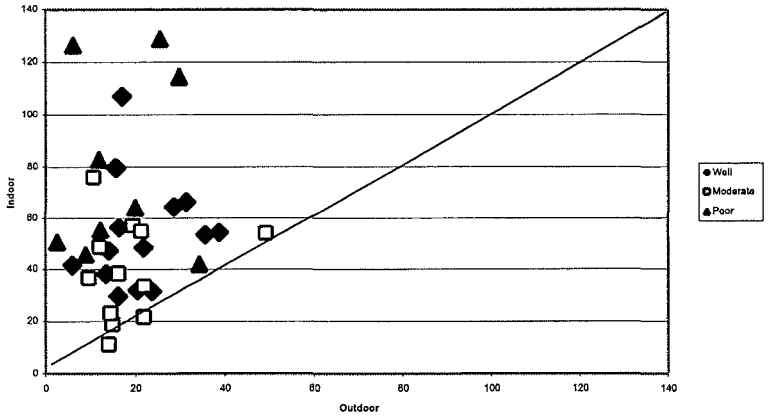


b)

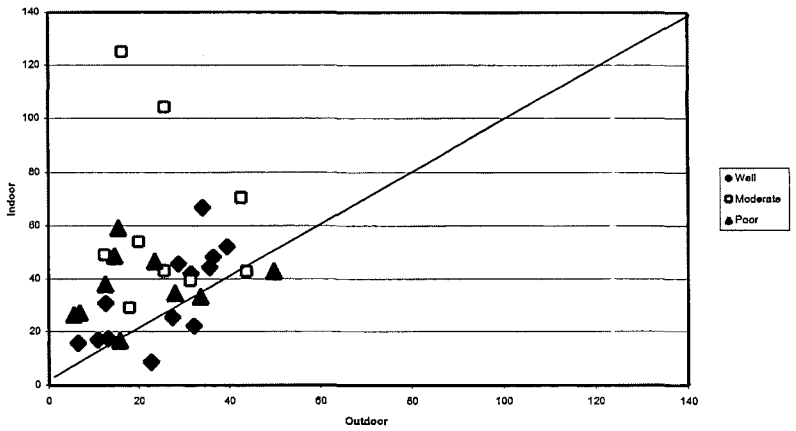
Figure 6.9 (a-b) Ventilation and Personal Exposure Concentrations ($\mu g/m^3$).



a)Winter



b) Spring



c) Summer

Figure 6.10 (a-c) Seasonal Daytime PM_{10} Indoor v Outdoor Effects of Ventilation

6.4 Scanning Electron Microscopy

Further analysis of the filters was undertaken in an attempt to ascertain the sources of particles collected in this study. The method of analysis is discussed in Section 4.8. A small sample of 54 filters were analysed using the SEM, these samples were selected as they were a complete, viable set of filters that had been refrigerated after gravimetric analysis to prevent any breakdown of the particles. The results will be discussed in terms of this small sample. It is intended only to give an insight into the possible sources of particulate matter found in personal exposure sampling in this area of London.

A summary of the particulate matter found on the filters of the different microenvironments is given in Table 6.3 using references from McCrone & Delly (1973). A selection of photographs illustrating the particle types are shown in Plates 6.1 - 6.8.

The SEM analysis has shown that the PEM's and HI's were collecting the intended size fractions and the loadings on the filters were evenly distributed. Plate 6.1 shows a blank filter with no particulate collection. It can be seen that there are potential difficulties in distinguishing between the Teflon threads of the filters and collected particles. The analysis of Teflon filters has to be made manually as computerised scanning methods cannot distinguish between the Teflon and the particles. This method of analysis is not generally undertaken with Teflon filters, however due to the small sample set it was felt that in this case such analysis was feasible.

It would appear evident that the sources of indoor and outdoor PM_{10} were different. In the home, the majority of the filters collecting PM_{10} had particles which were skin flakes, furnishing fibres and possibly soil derived coarse particles. The garden PM_{10} particles which were collected exhibited more insect and pollen debris.

The $PM_{2.5}$ filters for both indoor and outdoor environments do not appear to have any specific differences in the particulate matter characteristics, suggesting that this size fraction was not influenced by the particle generating activities within the home. This confirms the analysis of the ratios in Section 5.5, there appears to be more of an effect between the indoor and outdoor ratios during the day and night as compared to the PM_{10} size fraction.

The classroom filters for PM_{10} exhibited similar particulate collection as found in the home. There were a greater number of soil related particles, a possible result of resuspension as postulated previously in Section 5.8.4.

It is difficult to distinguish small ETS derived particles from the background globules of Teflon as seen in Plate 6.1, it was not possible to accurately identify ETS particles on any of the filters collected and analysed in this study.

The carbon based particles as seen in Plate 6.8 were collected from the home of the child where there was a faulty boiler. These particles can be assumed to be soot particles and are predominantly <0.7μm and would usually not be collected by the 2μm pore size filters. It is only because of the heavy loading of the filters that these have been trapped.

There were particles collected that were obviously much larger than the cut size of the samplers however, the sampler specifications as described in Section 3.6 were designed with a 50% collection efficiency hence some larger particulates were collected.

All of the filters appeared to have a loading of particles that were smooth and globular in shape and diameters of c. 2μm. It is possible that these particles were combustion derived, possibly from vehicles and due to their small size these were able to transfer easily from the outdoor to indoor environments. This again confirms the analysis of the ratios in Section 6.2.

The SEM analysis was successful for the physical characterisation of both size fractions. It provided evidence of the different sources of particulates for inside and outside, it also showed that personal exposure is a result of being exposed to both environments. This is an important concept to consider when setting legislation guidelines as discussed in Section 2.4.

Table 6.3 Summary of Particulate Matter Characteristics.

Environment	Particle Characteristics
Personal $PM_{2.5}$	Small angular particles c. 3.5μm diameter, smooth globular particles <2μm, some agglomerates, no skin flakes, fibres or insect debris. See Plate 5.3
Personal PM_{10}	Several large particles c. 30-40μm diameter which could be skin flakes, smooth globular particles <2μm, some granular, spherical particles c. 5μm. A small number of furnishing fibres. See Plate 6.3.
Home $PM_{2.5}$	Several agglomerates <4μm diameter, a small number of furnishing fibres, many smooth globular particles <2μm, no skin flakes. See Plate 6.4 and Plate 6.5.
Home PM_{10}	Many smooth globular particles <2μm, several skin flakes and furnishing fibres, granular angular particles <5μm, some agglomerates. See Plate 6.6.
Garden $PM_{2.5}$	Many smooth globular particles <2μm, some spherical sponge textured particles <5μm, no insect debris or fibres present.
Garden PM_{10}	Many smooth globular particles <2μm, some coarse irregular shaped particles c.5-6μm, some insect exoskeleton parts, some sponge like textured particles possibly pollen spores. See Plate 6.7.
Class $PM_{2.5}$	Granular angular particles c.3μm possibly soil derived, some skin flakes, some pollen granules, smooth globular particles <2μm.
Class PM_{10}	Same as Class $PM_{2.5}$ but more heavily loaded, some insect debris and fibres possibly from furniture or clothing.

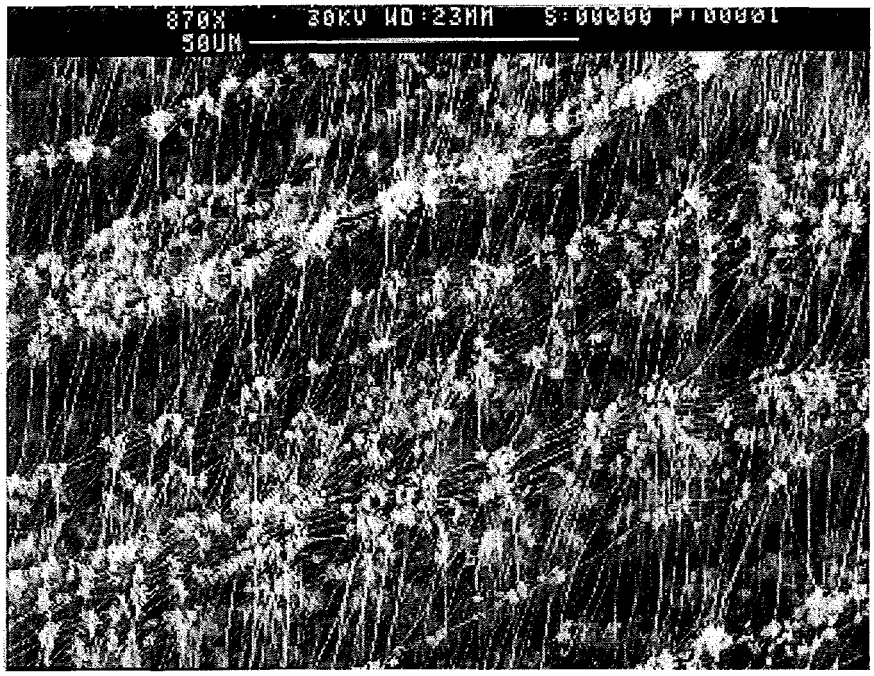


Plate 6.1 Blank Teflon Filter.

This figure clearly illustrates the problems of identifying specific particles as the threads and knots of the Teflon look like sampled particles. The SEM analysis can be computer controlled but only when using other types of filters, such as Nucleopore. The samplers and pumps that were used in this study were designed to operate with the Teflon filters which have a lower pressure drop across them compared to the Nucleopore filters.

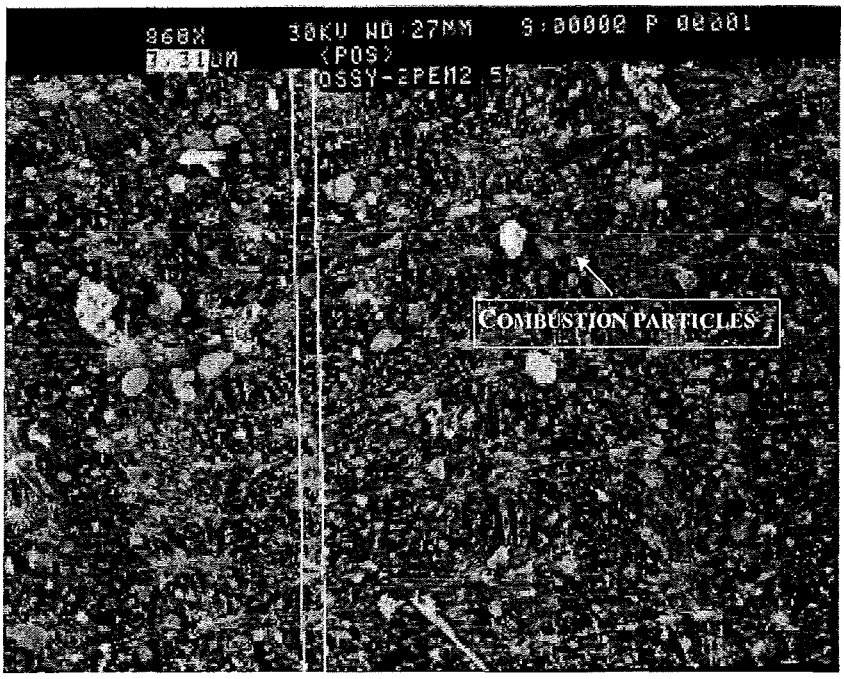


Plate 6.2 Example of Personal $PM_{2.5}$ Filter Particulates.

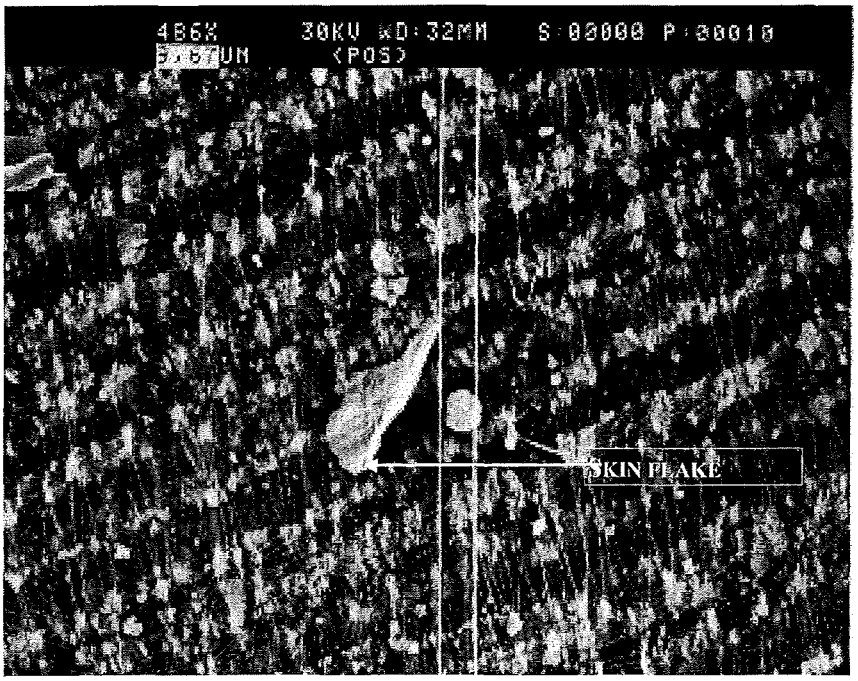


Plate 6.3 Example of Personal PM_{10} Filter Particulates.

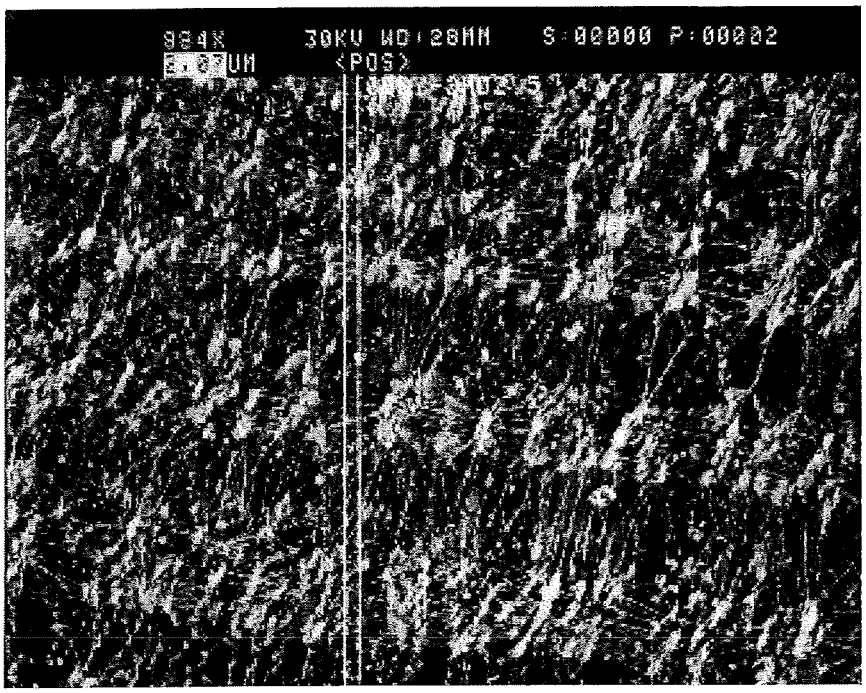


Plate 6.4 Example of Home $PM_{2.5}$ Filter Particulates (17 μg loading).

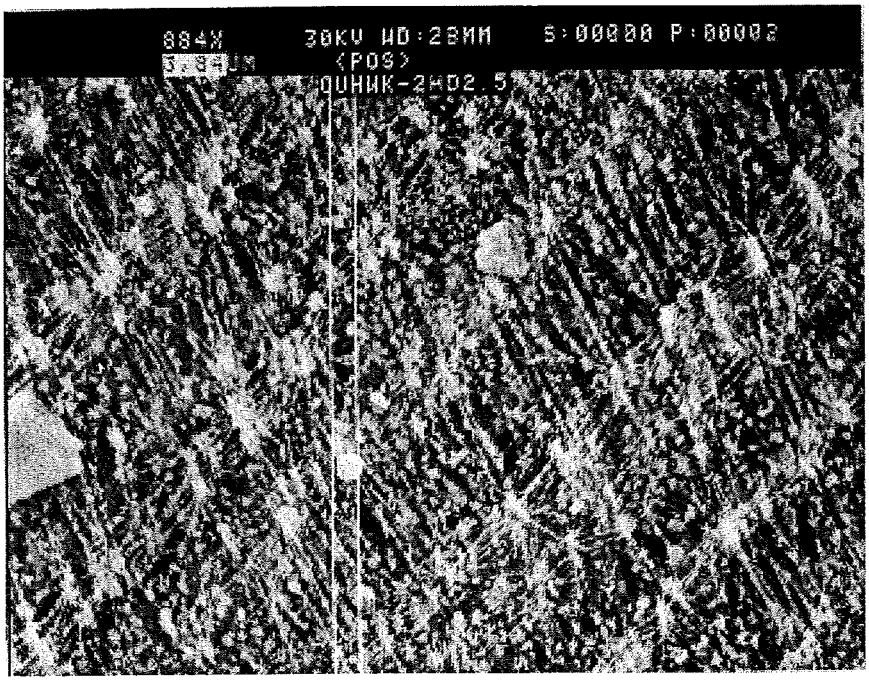


Plate 6.5 Example of Home $PM_{2.5}$ Particulates (27μg loading).

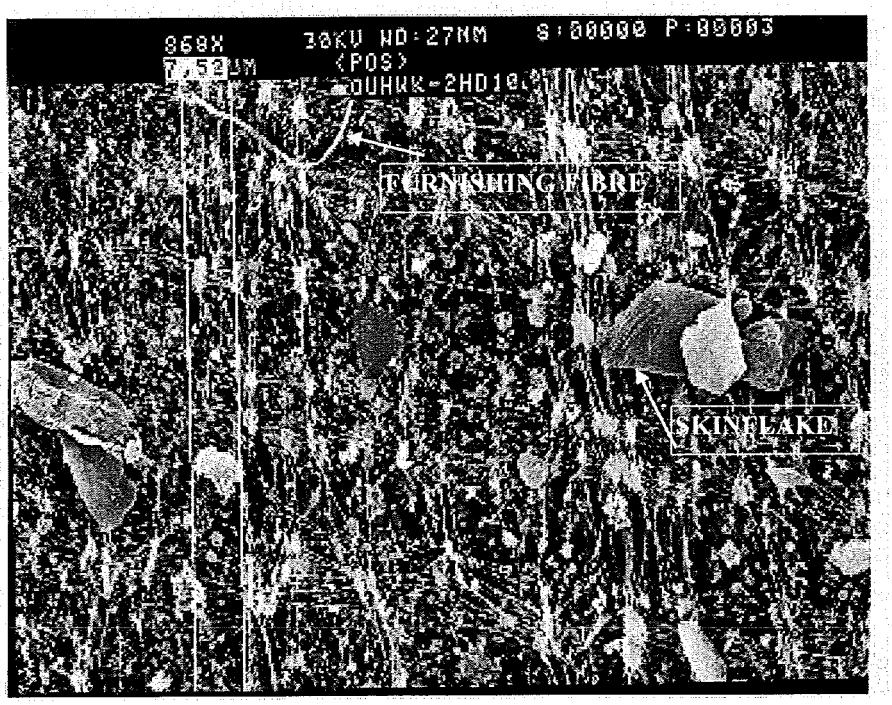


Plate 6.6 Example of Home PM_{10} Filter.

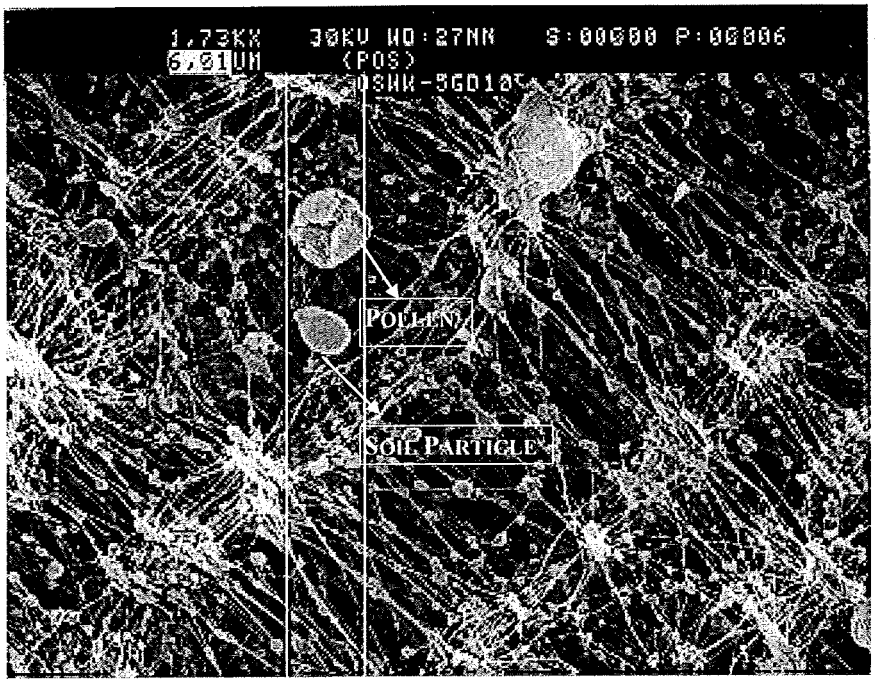


Plate 6.7 Example of Garden PM_{10} Filter.

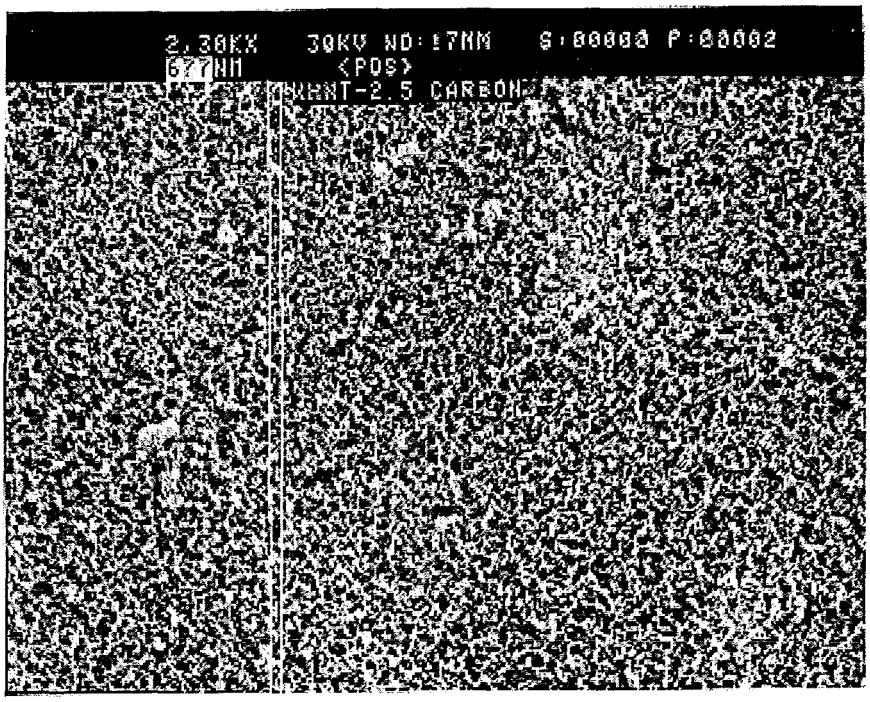


Plate 6.8 Example of Carbon Based Particles from a Faulty Boiler.

6.5 Personal Exposure and AUN data

To compare the daytime personal exposure with the AUN guidelines is problematic as discussed in Section 5.4. However, one of the objectives as stated in Section 1.2 is to assess how the children's actual exposure compares to the UK national air quality standards. One way of getting an indication as to whether the personal exposure concentrations for children living within Barnet are greater than the AUN recommended legislation is by assuming a rolling 24 hour average for 7am until 7am when the personal sampling occurred.

As only the daytime personal exposures were sampled the indoor night concentrations have been used for the assumed night time personal exposure patterns, this follows the recommendations made in the PTEAM paper (Spengler *et al.,* 1990, Thomas *et al.*, 1993). All available data is included in this analysis from all seasons. The personal data is the sum of the daytime personal concentrations and the night time home concentrations, divided by two to account for the two time periods. The Brent and Haringey AUN data for the daytime and night time concentrations is summed, again dividing the mean by two to account for the time periods. The average of the daily values is then used in a T-Test. The T-Test assumed a null hypothesis of there being no significant differences between the two variables, the results are in Table 6.4. The results reject the null hypothesis as there is a significant difference between the personal exposure concentrations and the AUN

concentrations, this was previously stated in Section 5.4. The mean 24-hour value for the personal PM_{10} is assumed to be 44 $\mu g/m^3$, this is just below the recommended 24 hour rolling average of 50 $\mu g/m^3$. This is an approximation but does provide some indication of the differences between personal exposure and AUN data for children living within the London Borough of Barnet. Another factor to consider is that the mean 12 hour personal exposure for these children over the three seasons was 53 $\mu g/m^3$ which could suggest that their short term exposures are higher and influenced by their activity patterns. These short-term exposures could be more important when considering potential health outcomes.

Table 6.4 T-Test of Personal Exposure and AUN.

t-Test: Two-Sample Assuming Equal Variances

	AUN	*Personal*
Mean	20.38298077	44.1145
Variance	116.6512693	579.0564
Observations	78	78
Pooled Variance	347.8538501	
Hypothesized Mean Difference	0	
df	154	
t Stat	-7.94620009	
P(T<=t) one-tail	1.89634E-13	
t Critical one-tail	1.65480742	
P(T<=t) two-tail	3.79268E-13	
t Critical two-tail	1.975486157	

The same procedures were undertaken for the personal $PM_{2.5}$, where the personal and indoor night time were added then divided by two to get a value of 20 $\mu g/m^3$ for the 24-hour average value of potential exposure. There is no AUN $PM_{2.5}$ standard for the UK at present, the US maximum standard for a recommended 24 hour exposure is 65 $\mu g/m^3$, the children in this research had lower personal exposures than the US standard, potential health outcomes will be discussed in Section 6.6.

6.6 Estimating Potential Health Effects for Children Living within Barnet

Having estimated the potential personal exposure of children living within the London Borough of Barnet some estimation of potential effects upon health can be assumed. The review of children's health effects, as discussed in Section 3.3.2, indicated a number of illnesses associated with exposure to both PM_{10} and $PM_{2.5}$. The UK legislation for PM_{10} as discussed in Section 2.4 was introduced because it was felt that at 50$\mu g/m^3$ 24-hour rolling average, healthy individuals would unlikely experience acute effects to their health. By

setting the legislation to this level it was felt that those individuals with pre-existing respiratory or cardiac disorders would be at less of a risk.

The potential personal exposure concentrations for the children in this study were below the recommended guidelines. Using the results of the only UK study of health outcomes for primary school children currently available it can be assumed that exposure to PM_{10} will have an adverse effect upon lung function (Scarlett *et al.,* 1996). Their research indicated that for exposure to particulate matter (PM_{10}) between 20-150 $\mu g/m^3$ there was a reduction in Forced Vital Capacity (FVC) of 1% (95%CI, 0.3 to 2%). The children's exposure in Barnet as discussed in Section 6.5 falls within this range of concentrations and as such it could be postulated that similar health effects could result for children living within Barnet. The summary of the study by Scarlett *et al.*, (1996) states that the findings for lung function are consistent with a number of studies from elsewhere, although the size of the effect is less. A recent review by Pope *et al.,* (1995) based mainly on American studies concluded that any increase of 10 $\mu g/m^3$ in PM_{10} levels is associated with a mean reduction of up to 0.35% in Forced Expiratory Volume (FEV). This is considerably more than the equivalent decrement of 0.09% in FEV that Scarlett *et al.*, (1996) found. For this research the findings from the UK study will more appropriate for assessing health outcomes as there is more likelihood that the particle composition is similar, as will be the activity patterns of the children.

The effects resulting from short-term exposure to $PM_{2.5}$ are also decreased lung function, lower respiratory disease symptoms and bronchitis, as discussed in the literature review in Section 3.3.2. The small numbers of long-term exposure studies estimate an increased risk for COPD and cancer in adults exposed as children. At present no legislation exists for $PM_{2.5}$ in the UK so no direct comparison is available. It has been shown however that children's 24-hour personal exposure concentrations for $PM_{2.5}$ exceed the outdoor concentrations and any future legislation should account for this difference.

6.7 Conclusions and Recommendations

The analysis of the ratios between indoor and outdoor concentrations has indicated that during the day, the particle generating activities that occur have a greater influence upon indoor concentrations.

Seasonal analysis of the ratios indicates that the summer day ratios for indoor and outdoor particles are lower, this has been shown to be a result of greater ventilation in the homes

during the summer. PM_{10} concentrations are more likely to be influenced by heating and ventilation. When the heating is on, windows are usually not open, leading to a possible build up of the larger sized particles in the homes. There is great variability in the data which could be a result of the differences in housing characteristics and activities that occur in each home. The small sample size of the dataset makes it difficult to identify which of the activities that are taking place during the day, are the cause of the high concentrations of particles within the home.

There appears to be no relationship between the one off air exchange rates taken when the homes were closed up and the concentrations of particles in both size fractions, this is not unexpected considering the reviewed literature in Section 2.6.

The homes where there was an increase in concentrations at night were found to be those that had a frequent smoker present. This indicates that smoking has an important influence upon indoor air quality especially at night when no other particle generating activities take place. The questionnaire data supports this, results indicate that smoking does have an influence upon home concentrations during the night when other particle generating activities were not occurring.

It would appear that cleaning and cooking activities do not specifically influence the concentration of either particle size fractions. Other studies, as already mentioned, have shown that instruments with greater time resolution do show that different types of cooking generate fine particulates. The 12-hour integrated samples collected in this study lose this definition.

The scanning electron microscopy analysis provides evidence of the different sources of particulate matter indoors and outdoors. Within the home it is clear that the particles are influenced by human activities as these are predominantly resuspended soil dust, skin flakes and fibres. The outdoor particles tend to be biological in origin e.g. pollen and insect debris. The personal exposure filters demonstrate a mixture of these two environments although they are child specific depending upon how much time was spent in the different environments.

There are smooth globular particles c.2μm in diameter that are found in all filters and are potentially combustion related, possible vehicle derived or a result of cooking. There is difficulty in distinguishing these particles from the filter matrix and further analysis of the particles using a different collection medium would provide greater insight into the source of these particles.

The $PM_{2.5}$ filters for both indoor and outdoor environments do not appear to have any specific differences in particulate matter characteristics, suggesting this size fraction was not influenced by particle generating activities in the homes.

There are particles collected that are obviously much larger than the cut size of the samplers however, the sampler specifications are designed with a 50% collection efficiency hence, some larger particulates have been collected. The SEM analysis showed that the samplers collected the intended size fractions and the filters were evenly loaded. Plate 6.1 shows the blank filter with nothing collected upon it. It can be seen that there are potential difficulties in distinguishing between all of the filters' Teflon threads and the particles. Very careful observation was required to identify the differences between the filter structure and particulates.

A greater understanding of particle type and source is necessary to determine the origins of the rest of the particles. It appears that ETS could be causing an increase in personal exposure concentrations within the home during the night however, it is not evident where the particles were originating from during the day. Greater source apportionment studies will need to be undertaken to assess this aspect. New developments of monitors that have a greater time resolution are becoming available and these will aide in the determination of specific sources from indoor activities. Some analysis has been undertaken using a TSI Scanning Mobility Particle Sizer which showed that ultra fine particles were produced by the gas oven, gas burners, and the toaster oven (Zartarian *et al.*, 1998).

7 Conclusions and Recommendations for Further Research

7.1 Discussion

This study has demonstrated that it is possible to identify children's personal exposure to particulate matter in the UK. Other research conducted in the Netherlands, as discussed in the literature review, used children as study subjects in a personal exposure to particulate matter study. Other personal exposure studies of particulate matter exposure have been conducted on either healthy or health compromised adults.

The children's exposure study design in the Netherlands was different to this one as multiple children were sampled at once, repeated sampling of up to eight measurements were obtained between January and May 1994 and 1995. They measured PM_{10} exposures for the children and outdoor concentrations, some classroom concentrations were also conducted during the second sampling session. Results indicated that the mean personal PM_{10} concentrations were 105$\mu g/m^3$ which were on average 67$\mu g/m^3$ higher than corresponding outdoor concentrations. These differences were attributed to exposure to ETS, classroom concentrations and indoor activity. There are several differences between the two studies, primarily the number of participants sampled per session, the sampling at different times of the year, the measurements of the homes as well as school and ambient locations. The Dutch study completed some XRF analysis whilst this study looked at the physical characteristics of the particles. Similar mass analysis results were obtained with the school concentrations exceeding all other sampled environments especially for PM_{10}.

The Boston study design methodology for sampling individuals with COPD was replicated in this study for use with children. The Boston study sampled two participants per every six days during winter and summer. Personal exposures exceeded indoor and outdoor measures for both size fractions and seasons. This has been attributed to the proximity of the participants to particle sources such as cooking and cleaning. The UK study of children did not find this to be the same as children are unlikely to be conducting these activities.

The Baltimore study utilised the same sampling equipment as the UK study. Healthy senior citizens were recruited for this study and were sampled during two seasons for 24 hour average time periods. Four to six individuals were included over a 12-day sampling period. Multiple pollutants were collected along with the particulate measures, personal and ambient locations were sampled. This study found that all personal exposure measurements were significantly lower than the corresponding outdoor measurements, this

was attributed to the limited exposure to indoor sources of particulate matter by these individuals.

Other studies that have been conducted included the PTEAM study where the findings showed that the population-weighted daytime personal PM_{10} concentrations averaged about 159 $\mu g/m^3$, compared to the indoor or outdoor mean concentrations of 95 $\mu g/m^3$. The overnight personal PM_{10} mean was much lower (95 $\mu g/m^3$) and more similar to the overnight indoor (63 $\mu g/m^3$) and outdoor (86 $\mu g/m^3$) means. The major reason cited for this increased exposure was determined to be largely a result of the personal cloud effect, where individual's activities resulted in them being closer to sources or causing resuspension of particulate matter. The study in Canada of personal exposure was a similar design as the PTEAM study and was a large-scale population-based exposure study. The design was to estimate three-day average personal exposures to PM_{10} and $PM_{2.5}$. The personal exposures to particulate matter throughout the three days tended to be much higher than both the indoor and outdoor levels. The median for personal exposures to PM_{10} was 48.5 $\mu g/m^3$, indoor and outdoor medians being 23.1 and 23.6 $\mu g/m^3$, respectively. The median concentrations for the $PM_{2.5}$ fractions personal, indoor and outdoors were 18.7, 15.4 and 13.2 $\mu g/m^3$, respectively. The differences between the different environments were attributed to the presence of smokers. The correlations between the personal exposures and the outdoor fixed sites and roof sites were low (0.16 - 0.27). The highest correlation was found between the personal and indoor environment (0.56). Similar correlations were found for the children's exposure, a smaller time resolution provided a clearer indication of potential exposure patterns, whereas a 3 day average results in much of this resolution being lost.

7.2 Conclusions

Conclusions are summarised reflecting the aims of the research established in Section 1.2.

Several studies, reviewed in the literature, have shown that the composition of particulate matter differs depending upon the source and size of the particulates. This study successfully collected both PM_{10} and $PM_{2.5}$ to assess children's personal exposure along with home, school and garden microenvironments. Physical analysis of the particles have indicated that there are differences in the composition of these particulates.

Other research has indicated that individuals' exposure depends upon the environments that they frequent and the amount of time spent in each place. This study successfully incorporated the use of Time Activity Diaries to determine the children's location

throughout the sampling time. Children's activity patterns were not influenced by the different seasons. The only activity patterns that were influenced by the seasons was time spent in the classroom, with significant differences being evident for the summer sampling period which was predominantly during the summer holidays.

The analysis of the ratios between indoor and outdoor concentrations has indicated that during the day the particle generating activities that occur indoors have a greater influence upon indoor concentrations. At night the ratios were lower especially for the $PM_{2.5}$ size fractions suggesting that the lack of activity and ventilation effects were influencing the build up of the larger particle sizes. Seasonal analysis of the ratios indicated that the summer day ratios for indoor and outdoor particles were lower, possibly due to greater ventilation of the homes during the summer. PM_{10} concentrations were influenced by heating and ventilation. When the heating was on the windows were not open, leading to a possible build up of the larger sized particles in the home. This is particularly evident for the daytime PM_{10} samples. There appeared to be no relationship between air exchange rates and the concentrations of particles in both size fractions.

The homes where there was an increase in indoor concentrations at night were found to be those that had a frequent smoker present. This indicates that smoking has an important influence upon indoor air quality especially at night when there are no other particle generating activities taking place. This is also supported by the questionnaire data which suggests that smoking influenced home concentrations during the night when other particle generating activities were not occurring.

The results from this study appear to indicate that cleaning and cooking activities do not specifically influence the concentration of either particle size fractions. Other studies, as already mentioned, have shown that instruments with greater time resolution do show that different types of cooking generate fine particulates. The 12-hour integrated samples collected in this study lose this definition.

The health effects of exposure to particulate matter were reviewed for adults and children, indicating that there were significant potential health effects from exposure to particulate matter. The majority of these studies assessed exposure using outdoor ambient monitoring. As no specific health outcomes were measured in this research it was only possible to assume similar results to those found by Scarlett *et al.*, (1996) from exposure to PM_{10}, these were estimated as a 1% decrease in FEV for school children in the UK.

It is evident that it was not possible to directly assess children's personal exposure to either PM_{10} or $PM_{2.5}$ using outdoor monitoring sites as these underestimated the actual exposure concentrations. Personal exposure was estimated as a mean value for PM_{10} of 44 $\mu g/m^3$ which does not exceed the recommended 24-hour rolling average of 50 $\mu g/m^3$.

The home PM_{10} and $PM_{2.5}$ concentrations during the day and night were greater than the outdoor monitoring sites. It was not possible to use an outdoor monitoring site to assess indoor concentrations of PM_{10} and $PM_{2.5}$ as these underestimated concentrations found within the home. The ambient measurements did not account for the differences in the particle compositions that were found using the SEM analysis either. At night there were weak correlations between the home and outdoor concentrations for both size fractions.

The classroom concentrations of PM_{10} and $PM_{2.5}$ during the day exceeded the outdoor concentrations. The contribution that the $PM_{2.5}$ fraction of the PM_{10} fraction is approximately 34% compared to the home where the value is 42% and outdoors is 55%. The lack of any data specifically associated with particle generating activities within the classroom has severely limited the analysis of the apportionment of the children's personal exposure sources. Only the SEM data indicated that there was a higher incidence of soil derived particles which could possibly be a result of the resuspension caused by many children in the room.

The outdoor sampling in the garden and the AUN sites initially suggested that there were significant relationships so that a single monitoring site would be sufficient in Barnet to measure the outdoor concentrations. However, the Wilcoxon Signed Ranks Tests showed that there were significant differences between the two AUN sites and also between the garden and the Brent AUN site. Further investigation is required into whether local sources influence the AUN monitoring or whether meteorological conditions are responsible for this variation.

The use of time weighted averaging models to estimate personal exposure to particulate matter using the time activity diaries and microenvironmental concentrations provided an underestimation of the actual exposure concentrations. The sample size of this population of children is too small to provide an accurate estimate of the exposure pattern of a wider population.

7.3 Recommendations for Further Research

A number of recommendations for future research related to this study can be made. These would not only improve the quality of the data collected but would also aid in the understanding of how particulate matter could potentially influence children's health.

The time period of the sampling was an integrated 12-hour exposure, the effects of specific particle generating activities were lost in this collection time. Having a method to determine the real time resolution would aid in the analysis of the exposure patterns, specifically within the home where cigarette smoking, cooking and cleaning have previously been cited as the dominant sources of particulate matter.

The effect of ventilation within the homes was shown to influence the concentrations of the different size fractions, only one air exchange measurement was made that reflected the likely ventilation frequency for each house during the winter. By having a continuous measurement of ventilation that reflected the sampling period a greater understanding of the ventilation effect could be determined.

Characterisation of particulate matter for specific source apportionment would greatly improve these studies. By collecting information on the activities that were undertaken in the school it would be possible to include this predominant source of the children's personal exposure. By being able to identify where the specific particles originate from it will be possible to legislate more effectively.

Further research into the personal exposure of a larger number of children sampled at the same time will reduce the number of confounding factors, such as meteorological conditions, that may be causing the differences between the individual children.

To determine the health effects of particulate matter for children further procedures would be necessary. These could include respiratory measures of lung function or prevalence of asthmatic symptoms. This was initially an objective but was not completed as part of the research due to financial constraints and the small number of children included, as a result of this the data cannot reflect the general population of children living in the UK.

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APPENDICES

List of Abbreviations and Units

ach^{-1}	Air Exchange Measurements (air changes per hour)
APEG	Airborne Particles Expert Group
AUN	Automatic Urban Network
CEN	European Standards Committee
CIP-10T	Model of Personal Exposure Monitor
COMEAP	Committee on the Medical Effects of Air Pollutants
COPD	Chronic Obstructive Pulmonary Disease
DETR	Department for the Environment, Transport and Regions
EPA	Environmental Protection Agency (USA)
EPAQS	Expert Panel on Air Quality Standards
EPSRC	Engineering and Physical Research Council
ETS	Environmental Tobacco Smoke
EXPOLIS	Air Pollution Exposure Distributions of Adult Urban Populations in Europe
FEV_1	Forced Expiratory Volume in One Second
HI	Harvard Impactor
HSE	Health and Safety Executive
ISO	International Standards Organisation
$l\ min^{-1}$	Litres per minute
MMAD	Mass Median Aerodynamic Diameter
MRC	Medical Research Council
NAAQS	National Ambient Air Quality Standards
PEM	Personal Exposure Monitor
PM_{10}	Particulate Matter less than 10 μm in aerodynamic diameter
$PM_{2.5}$	Particulate Matter less than 2.5 μm in aerodynamic diameter
ppb	Parts Per Billion
PTEAM	Personal Total Exposure Assessment Methodology Study
QUARG	Quality of Urban Air Review Group
RSP	Respirable Suspended Particulates
SPM	Suspended Particulate Matter
TAD	Time Activity Diary
TSP	Total Suspended Particulates
$\mu g/m^3$	micrograms per meter cubed concentration
μm	One millionth of a metre, micron
UPRC	Urban Pollution Research Centre
WHO	World Health Organisation

Appendix 3

Barnet Health Authority Questionnaire (March 1996)

Imperial College,
Centre for Environmental Technology,
48 Prince's Gardens,
South Kensington,
London.
SW7 2PE

Dear Parent/Guardian,

Respiratory diseases cause great distress to many people. They are of particular concern among children, and some chest diseases are becoming more common. In an attempt to establish the cause, parents/guardians of children attending schools within Barnet are being asked to complete this questionnaire which has been designed by Imperial College in conjunction with Barnet Health Authority. It would be very much appreciated if you would be willing to answer the questions in the attached questionnaire and return it, when completed, to your child's class teacher, in the envelope provided. Whether or not your child has symptoms, it is important that you should fill in the questionnaire.

All answers on this questionnaire will be kept entirely confidential. Should you have any queries concerning the questionnaire, please do not hesitate to contact me. Thank you for your co-operation.

Yours sincerely,

Heidi Cheung (PhD Researcher),
Telephone: 0171 594 9283 extn. 59281
Fax: 0171 581 0245

Dr. Stephen Farrow,
Director of Public Health.

ENVIRONMENT PROJECT QUESTIONNAIRE

PLEASE ANSWER ALL THE QUESTIONS BY TICKING THE APPROPRIATE BOX OR WRITING IN THE SPACE PROVIDED: ALL INFORMATION OBTAINED IN THE STUDY WILL BE KEPT CONFIDENTIAL. All questions mentioning "your child", refers to the particular child in this survey.

A. Your child's sex (please tick) Female ☐1 Male ☐2

B. The month and year your child was born .....month.....year

1. How many people live in your household? (including yourself)

   a. ..... adults (over 18 years)

   b. ..... young adults (16-18 years)

   c. ..... children (0-15 years)

2. Please give the full postcode of your PRESENT address
   e.g. SW13 9ZZ

   a. ☐☐☐☐☐☐☐

   How long has your child lived at the present address?
   (please tick one box)

   b. less than 1 year ☐1

   c. 1 - 4 years ☐2

   d. 5 - 9 years ☐3

   e. 10 years or more ☐4

3. Do your currently live in?

   a. a whole detached house (or bungalow) ☐1

   b. a whole semi-detached house/bungalow ☐2

   c. a whole terraced house ☐3

   d. a flat/maisonette (self contained) ☐4

   e. room in someone else's house ☐5

   f. other (please state) ☐6

   g. .....

4. Do you have a garage?

a. yes ☐1 no ☐2

if yes: (please tick one box)

b. the garage is not attached to your home ☐1

c. the garage is attached to the side of your home ☐2

d. the garage is built into the structure of your home ☐3

5. What is the lowest level of your living accommodation? (please tick one box)

a. basement ☐1

b. ground floor ☐2

c. 1st floor ☐3

d. 2nd floor or above, ☐4 If yes, please give the floor .....

6. How would you describe the level of traffic in your street? (Please tick one box)

None ☐1 Light ☐2 Moderate ☐3

Busy ☐4 Very Busy ☐5

7. Please tick if you have any of the cooking appliances below, and indicate approximately how many hours per week they are used:

		Yes	If yes, give hours
i.	Electric hob/rings	☐1	.....
ii.	Electric oven	☐2	.....
iii.	Microwave	☐3	.....
iv.	Gas hob	☐4	.....
v.	Gas oven	☐5	.....

vi. How old is the cooker in your home? (please indicate how many years) .....

8. Do you ventilate your kitchen when cooking?

Yes ☐ 1 No ☐ 2

If yes, how?

a. Cooker hood vent outside ☐ 1

b. Cooker hood ☐ 2

c. Wall/Window (extractor fan) ☐ 3

d. Wall vent or air brick ☐ 4

e. Open window ☐ 5

f. Other type of ventilation ☐ 6

(please describe) .....

9. **Inside your home**

What is the main method of heating in the home?

I. central heating by gas ☐ 1

II. heating by electricity ☐ 2

III. heating by oil ☐ 3

IV. storage heaters ☐ 4

V. wood stoves or wood fires ☐ 5

VI. paraffin heaters ☐ 6

VII. gas fires (mains gas) ☐ 7

VIII. gas fires (calor gas) ☐ 8

IX. coal fire/boiler ☐ 9

X. other type of heating ☐ 10

(please describe) .....

XI. What is your main method of heating in winter?

10. Does anyone in your home smoke cigarettes/cigars/pipe?

Yes ☐1 No ☐2

11. When was your home built?

pre 1900	☐1
1901-1940	☐2
1941-1960	☐3
1961-1980	☐4
post 1980	☐5

12. Is your home insulated with urea formaldehyde cavity insulation?

yes ☐1 no ☐2 don't know ☐3

13. In the past year have any of the following rooms been decorated or had any brand new furniture?

	your living room:	yes	no	don't know
a.	painted	☐1	☐2	☐3
b.	wall papered	☐1	☐2	☐3
c.	new carpet	☐1	☐2	☐3
d.	new floor covering	☐1	☐2	☐3
e.	new furniture	☐1	☐2	☐3
f.	new insulation material	☐1	☐2	☐3

	your kitchen:	yes	no	don't know
g.	painted	☐1	☐2	☐3
h.	wall papered	☐1	☐2	☐3
i.	new carpet	☐1	☐2	☐3
j.	new floor covering	☐1	☐2	☐3
k.	new furniture	☐1	☐2	☐3
l.	new insulation material	☐1	☐2	☐3

The room the study child sleeps in:		yes	no	don't know
m.	painted	☐1	☐2	☐3
n.	wall papered	☐1	☐2	☐3
o.	new carpet	☐1	☐2	☐3
p.	new floor covering	☐1	☐2	☐3
q.	new furniture	☐1	☐2	☐3
r.	new insulation material	☐1	☐2	☐3

any other rooms: (please describe) .....

		yes	no	don't know
s.	painted	☐1	☐2	☐3
t.	wall papered	☐1	☐2	☐3
u.	new carpet	☐1	☐2	☐3
v.	new floor covering	☐1	☐2	☐3
w.	new furniture	☐1	☐2	☐3
x.	new insulation material	☐1	☐2	☐3

4. How often do you use the following at home?

		Every day	Most days	About once a week	Less than once a month	Not at all
a.	disinfectant	☐1	☐2	☐3	☐4	☐5
b.	bleach	☐1	☐2	☐3	☐4	☐5
c.	window cleaner	☐1	☐2	☐3	☐4	☐5
d.	dry cleaning fluid	☐1	☐2	☐3	☐4	☐5
e.	fly spray	☐1	☐2	☐3	☐4	☐5
f.	incense	☐1	☐2	☐3	☐4	☐5
g.	household varnish	☐1	☐2	☐3	☐4	☐5
h.	aerosols or sprays (including hair spray)	☐1	☐2	☐3	☐4	☐5
i.	deodorants	☐1	☐2	☐3	☐4	☐5

		Every day	Most days	About once a week	Less than once a month	Not at all
j.	air fresheners	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
k.	moth repellent	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
l.	pesticides	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
m.	other chemicals	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
n.	please describe					

15. How often do you have any window open in your home?

	Windows almost always open	Windows open only when weather is good	Windows open occasionally	Windows almost never open
In summer:				
a. day	☐ 1	☐ 2	☐ 3	☐ 4
b. night	☐ 1	☐ 2	☐ 3	☐ 4
In winter:				
c. day	☐ 1	☐ 2	☐ 3	☐ 4
d. night	☐ 1	☐ 2	☐ 3	☐ 4

16. Are any of your windows double glazed?

yes all of them ☐ 1 yes some of them ☐ 2

no none of them ☐ 3 don't know ☐ 4

17. a. Is there any damp or condensation in your home?

yes ☐ 1 no ☐ 2

<u>If yes,</u> b. How much of a problem is it?

not serious ☐ 1

fairly serious ☐ 2

very serious ☐ 3

c. Is there any mould in your home?

yes ☐1 no ☐2

If yes, d. How much of a problem is it?

not serious ☐1

fairly serious ☐2

very serious ☐3

If you have damp/condensation/mould in your home please tick the boxes relating to the problems you get in each room.

	Condensation on windows/ walls/ ceilings	Damp patches on walls	Mould on walls	Damp on furniture, carpets or clothes	Mould on furniture, carpets or clothes	None
e. Kitchen (or Kitchen/diner)	☐1	☐2	☐3	☐4	☐5	☐6
f. Living room (or lounge/diner)	☐1	☐2	☐3	☐4	☐5	☐6
g. Hall/landing	☐1	☐2	☐3	☐4	☐5	☐6
h. Your child's bedroom	☐1	☐2	☐3	☐4	☐5	☐6
i. Other bedrooms	☐1	☐2	☐3	☐4	☐5	☐6
j. Other rooms	☐1	☐2	☐3	☐4	☐5	☐6

18. Please tick the types of floor covering present in the rooms of your home.

	wall-to-wall carpet	partly carpeted	lino	Tiles	floor boards
a. kitchen	☐1	☐2	☐3	☐4	☐5
b. living room	☐1	☐2	☐3	☐4	☐5
c. hall/landing	☐1	☐2	☐3	☐4	☐5
d. child's bedroom	☐1	☐2	☐3	☐4	☐5

e. How old is the floor covering in your living room? .....

f. How old is the floor covering in your child's bedroom? .....

19. **In the room where the child sleeps:**

		yes always	yes sometimes	no not at all
a.	Is there a window open at night?	☐1	☐2	☐3
b.	does your child have an electric blanket?	☐1	☐2	☐3
c.	is the heating on at night?	☐1	☐2	☐3

e. If yes, what type of heating? .....

f. How old is your child's pillow? .....

g. What type of pillow does your child use? (please tick one box)

foam ☐1 hollow fibre ☐2 polyester ☐3 feather ☐4

other ☐5 (please describe) .....

h. How old is your child's mattress? .....

20. Have you ever suffered from any of the following? Please tick any you have or have had.

Asthma	☐1	Eczema	☐2	Hay fever	☐3
Allergic rhinitis	☐4	Bronchitis	☐5		

Have you ever had any of the following problems?

		yes had it recently	yes in past, not now	no never	don't know
a.	attacks of wheezing with whistling on the chest	☐1	☐2	☐3	☐4
b.	cough/wheeze/ breathlessness often during the night	☐1	☐2	☐3	☐4
c.	asthma	☐1	☐2	☐3	☐4

21. Has **your partner** ever suffered from any of the following? Please tick any he/she has or has had.

Asthma ☐1 Eczema ☐2 Hay fever ☐3

Allergic rhinitis ☐4 Bronchitis ☐5

Has **your partner** ever had any of the following problems?

		yes had it recently	yes in past, not now	no never	don't know
a.	attacks of wheezing with whistling on the chest	☐1	☐2	☐3	☐4
b.	cough/wheeze/ breathlessness often during the night	☐1	☐2	☐3	☐4
c.	asthma	☐1	☐2	☐3	☐4

22. Has **your child** ever suffered from any of the following? Please tick any he/she has or has had.

Asthma ☐1 Eczema ☐2 Hay fever ☐3

Allergic rhinitis ☐4 Bronchitis ☐5

Has your child had any of the following problems?

		yes had it recently	yes in past, not now	no never	don't know
a.	attacks of wheezing with whistling on the chest	☐1	☐2	☐3	☐4
b.	cough/wheeze/ breathlessness often during the night or on getting up in the morning	☐1	☐2	☐3	☐4
c.	wheezing/ breathlessness when exercising	☐1	☐2	☐3	☐4
d.	asthma	☐1	☐2	☐3	☐4

e. Has your child ever been treated by a doctor or admitted to hospital because of asthma? yes ☐1 no ☐2

f. Has your child ever been diagnosed to have asthma by a doctor? yes ☐1 no ☐2

If the answer is **no never** to all parts of question 22, please go to question 26.

If the answer is **yes recently/yes in the past/don't know** to any part of question 22 please answer the following questions.

g. How many separate times has your child experienced cough/ wheeze/breathlessness during the night in the past 12 months?

once ☐1 twice ☐2 3-4 times ☐3 5 or more times ☐4 don't know ☐5 none ☐6

h. How many separate times has your child experienced cough/ wheeze/breathlessness during the day in the past 12 months?

once ☐1 twice ☐2 3-4 times ☐3 5 or more times ☐4 don't know ☐5 none ☐6

i. If your child has asthma, how many asthma attacks has your child had in the past 12 months?

once ☐1 twice ☐2 3-4 times ☐3 5 or more times ☐4 don't know ☐5 none ☐6

23. How many days altogether would you say your child has wheezed or experienced breathlessness in the past 12 months?

a. one day ☐1 2-3 days ☐2 4-9 days ☐3 10-19 days ☐4 20 or more ☐5 don't know ☐6 none ☐7

b. What do you think brings the wheeze or breathlessness on?

chest infection	☐1	being in a smoky room	☐2
cold weather	☐3	exercise	☐4
no idea	☐5	other (please tick and describe)	☐6

24. a. How old was your child when he/she **first** experienced an attack of wheeze/breathelessness/asthma?

year ..... month .....

b. How old was your child when he/she experienced the **most recent** episode of wheeze/breathlessness/asthma?

year ..... month .....

c. If your child no longer has attacks of wheeze/breathlessness/ asthma, how old was he/she when it last occured?

year ..... month .....

25. Has your child taken any medicine (pressurised inhaler, aerosol, nebuliser) for wheeze/breathlessness/asthma or for other chest problems?

a. <u>In the past</u>

yes ☐1 No ☐2 If <u>yes</u>, please describe .....

.....

b. <u>At present</u>

yes ☐1 No ☐2 If <u>yes</u>, please describe .....

.....

26. a. Have any of your other children had similar spells of wheezing/breathlessness/asthma?

yes ☐1 no ☐2 have no other children ☐3

if yes, please indicate for each child whether they are older/younger and whether they are male/female.

Please tick for each child

		Older	Younger	Male	Female
b.	Child 1	☐1	☐2	☐3	☐4
c.	Child 2	☐1	☐2	☐3	☐4
d.	Child 3	☐1	☐2	☐3	☐4
e.	Child 4	☐1	☐2	☐3	☐4

Please add additional children if necessary.

f. .....

.....

27. a. Do you have pet(s) in the house? yes ☐1 no ☐2

b. If yes, (please tick)

Cat ☐1

Dog ☐2

Bird ☐3

Other (please describe) ☐4

.....

.....

28. What educational qualifications do you and your partner have? Please tick all that apply.

		your self	your partner
a.	CSE or GCSE (D, E, F or G)	☐1	☐2
b.	O-level or GCSE (A, B or C)	☐1	☐2
c.	A-level	☐1	☐2

		your self	your partner
d.	Qualifications in shorthand/ typing/or other skills, e.g. haidressing	☐ 1	☐ 2
e.	Apprenticeship	☐ 1	☐ 2
f.	State enrolled nurse	☐ 1	☐ 2
g.	State registered nurse	☐ 1	☐ 2
h.	City & Guilds intermediate technical	☐ 1	☐ 2
i.	City & Guilds final technical	☐ 1	☐ 2
j.	City & Guilds full technical	☐ 1	☐ 2
k.	Teaching qualification	☐ 1	☐ 2
l.	University degree	☐ 1	☐ 2
m.	No qualifications	☐ 1	☐ 2
n.	Qualifications not known	☐ 1	☐ 2
o.	Not applicable, no such person		☐ 2
p.	Other (please describe)	☐ 1	☐ 2

.....

The question below ask about your <u>current</u> occupation.
As far as you can, please describe the actual job, occupation, trade or profession. (Use precise terms such as radio mechanic, woodworking machinist, toolroom foreman. If the occupation is known by a special name, please use that name. If in H.M. Forces, give the rank in addition to the actual job. Please also describe the type of industry or service given: i.e. give details of what is made, materials used or services given).

a. **your present job or last main job**

.....

.....

b. your partner's present job or last main job

.....

.....

30. a. Please state your child's country of birth

.....

b. How would you describe the race or ethnic group of your child?

Asian

- Bangladeshi ☐ 1
- Chinese ☐ 2
- Indian ☐ 3
- Pakistani ☐ 4
- Other ☐ 5 please describe .....

Black

- African ☐ 6
- Caribbean ☐ 7
- Other ☐ 8 please describe .....

White ☐ 9

Other ☐ 10 please describe .....

Not known ☐ 11

Person completing the questionnaire (please tick)

Child's mother	☐ 1	Child's father	☐ 2
Stepmother	☐ 3	Stepfather	☐ 4
Female guardian	☐ 5	Male guardian	☐ 6
Other	☐ 7		

(please specify relation) .....

PLEASE TURN TO BACK PAGE

WHEN COMPLETED, PLEASE RETURN THE QUESTIONNAIRE TO YOUR CHILD'S TEACHER, IN THE ENVELOPE PROVIDED BY WEDNESDAY OF THIS WEEK.
THANK YOU VERY MUCH FOR YOUR HELP

Research is continuing to enable us to understand the significance of indoor factors to health. In order that we can compare data provided by this questionnaire study with new research we are planning a second survey to carry out some measurements of pollutants in a sample of homes, if you would be willing to participate in the follow-up survey please would you indicate this by ticking the box below.

☐ 1

If you have any queries about this questionnaire, please contact

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Imperial College
Centre for Environmental Technology,
48 Prince's Gardens,
South Kensington,
London.
SW7 2PE

tel: 0171 589 5111 ext. 59281
fax: 0171 581 0245

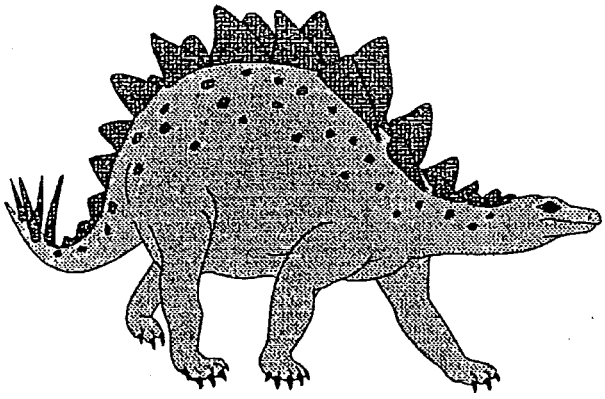
Appendix 4

Time Activity Diary

Try to remember to tick the circles in the diary when you change activities. Write the things that you do such as:

How you got to school,
What games you played outside,
If someone smoked a cigarette near to you and,
If you had to take the monitor off.

Thank you very much!

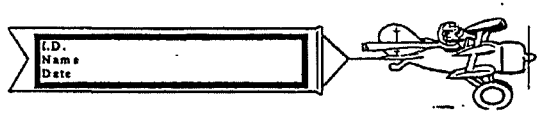


Name _____

School _____

Class _____

Day _____



	Time	Home Inside	Home Outside	Classroom	School Outside	Other Inside	Other Outside	Travelling	Near to smoker	Activity
	6.30	O	O	O	O	O	O			
☐	6.45	O	O	O	O	O	O			
	7	O	O	O	O	O	O			
M	7.15	O	O	O	O	O	O			
O	7.30	O	O	O	O	O	O			
R	7.45	O	O	O	O	O	O			
N	8	O	O	O	O	O	O			
I	8.15	O	O	O	O	O	O			
N	8.30	O	O	O	O	O	O			
G	8.45	O	O	O	O	O	O			
	9	O	O	O	O	O	O			
☐	9.15	O	O	O	O	O	O			
	9.30	O	O	O	O	O	O			
M	9.45	O	O	O	O	O	O			
O	10	O	O	O	O	O	O			
R	10.15	O	O	O	O	O	O			
N	10.30	O	O	O	O	O	O			
I	10.45	O	O	O	O	O	O			
N	11	O	O	O	O	O	O			
G	11.15	O	O	O	O	O	O			
	11.30	O	O	O	O	O	O			
☐	11.45	O	O	O	O	O	O			

	Time	Home Inside	Home Outside	Classroom	School Outside	Other Inside	Other Outside	Travelling	Near to smoker	Activity
	12	O	O	O	O	O	O			
	12.15	O	O	O	O	O	O			
A	12.30	O	O	O	O	O	O			
F	12.45	O	O	O	O	O	O			
T	1	O	O	O	O	O	O			
E	1.15	O	O	O	O	O	O			
R	1.30	O	O	O	O	O	O			
N	1.45	O	O	O	O	O	O			
O	2	O	O	O	O	O	O			
O	2.15	O	O	O	O	O	O			
N	2.30	O	O	O	O	O	O			
	2.45	O	O	O	O	O	O			
A	3	O	O	O	O	O	O			
F	3.15	O	O	O	O	O	O			
T	3.30	O	O	O	O	O	O			
E	3.45	O	O	O	O	O	O			
R	4	O	O	O	O	O	O			
N	4.15	O	O	O	O	O	O			
O	4.30	O	O	O	O	O	O			
O	4.45	O	O	O	O	O	O			
N	5	O	O	O	O	O	O			
	5.15	O	O	O	O	O	O			
	5.30	O	O	O	O	O	O			
	5.45	O	O	O	O	O	O			

	Time	Home Inside	Home Outside	Classroom	School Outside	Other Inside	Other Outside	Travelling	Near to smoker	Activity
	6	O	O	O	O	O	O			
	6.15	O	O	O	O	O	O			
E	6.30	O	O	O	O	O	O			
V	6.45	O	O	O	O	O	O			
E	7	O	O	O	O	O	O			
N	7.15	O	O	O	O	O	O			
I	7.30	O	O	O	O	O	O			
N	7.45	O	O	O	O	O	O			
G	8	O	O	O	O	O	O			
	8.15	O	O	O	O	O	O			
	8.30	O	O	O	O	O	O			
	8.45	O	O	O	O	O	O			
E	9	O	O	O	O	O	O			
V	9.15	O	O	O	O	O	O			
E	9.30	O	O	O	O	O	O			
N	9.45	O	O	O	O	O	O			
I	10	O	O	O	O	O	O			
N	10.15	O	O	O	O	O	O			
G	10.30	O	O	O	O	O	O			
	10.45	O	O	O	O	O	O			
	11	O	O	O	O	O	O			

Appendix 5

House Screening Questionnaire

House information

Address: _____
_____
_____
_____

Family name: _____
ID _____
Tel. No. _____

Sampling Dates
Period 1: _____
Period 2: _____
Period 2: _____

No. of Residents: _____ **Smokers?** _____

Smoking permitted in home? _____

Housing characteristics:
Cooking fuel, cooker _____ **Heating fuel** _____
grill _____ **Heating sources** _____
oven _____ **Humidifiers** _____

Ventilation:
Windows _____
Extractor fans _____

Pets: _____

Access to Garden: _____
Power source _____

Pump location: _____

Travel to School: _____

Regular activities: _____
_____

General times : _____
_____

Comments:

Appendix 6

Household Activity Questionnaire

Potential activities to influence PM concentrations during Monitoring Period. Date_____ I.D._____

1. **Was any housework carried out during the monitoring time?** Yes_____1 **GO TO QUESTION 2(i)**
No_____2 **GO TO QUESTION 3** **Child near?**

2. **Did the housework include;** (i) vacuuming Yes_____1 Time__:__ Room _____ _____
Time__:__ Room _____ _____
Time__:__ Room _____ _____
No_____2 **GO TO QUESTION 2(ii)**

(ii) dusting Yes_____1 Time__:__ Room _____ _____
Time__:__ Room _____ _____
Time__:__ Room _____ _____
No_____2 **GO TO QUESTION 2(iii)**

(iii) air fresheners Yes_____1 Time__:__ Room _____ _____
Time__:__ Room _____ _____
Time__:__ Room _____ _____
No_____2 **GO TO QUESTION 2(iv)**

(iv) clothes washing/tumble dryer Yes_____1 Time__:__ Room _____ _____
No_____2 **GO TO QUESTION 3**

				Child near?
3. **Did any cooking take place?**		Yes_____1	**GO TO QUESTION 3(i)**	
		No_____2	**GO TO QUESTION 4**	
	(i) oven	Yes_____1	Times__:__to__:__	_____
			Times__:__to__:__	_____
			Times__:__to__:__	_____
		No_____2		
	extractor fans/windows open	Yes_____1	Times__:__to__:__	
			Times__:__to__:__	
			Times__:__to__:__	
		No_____2	**GO TO QUESTION 3(ii)**	
	(ii) cooker....boiling	Yes_____1	Times__:__to__:__	_____
			Times__:__to__:__	_____
			Times__:__to__:__	_____
		No_____2		
	extractor fans/windows open	Yes_____1	Times__:__to__:__	
			Times__:__to__:__	
			Times__:__to__:__	
		No_____2	**GO TO QUESTION 3(iii)**	
	(iii) cooker.....frying	Yes_____1	Times__:__to__:__	_____
			Times__:__to__:__	_____
			Times__:__to__:__	_____
		No_____2		
	extractor fans/windows open	Yes_____1	Times__:__to__:__	
			Times__:__to__:__	
			Times__:__to__:__	
		No_____2	**GO TO QUESTION 3(iv)**	

			Child near?
(iv) grill	Yes_____1	Times__:__to__:__	_____
		Times__:__to__:__	_____
		Times__:__to__:__	_____
	No_____2		
extractor fans/windows open	Yes_____1	Times__:__to__:__	
		Times__:__to__:__	
		Times__:__to__:__	
	No_____2	**GO TO QUESTION 3(v)**	
(v) microwave	Yes_____1	Times__:__to__:__	_____
		Times__:__to__:__	_____
		Times__:__to__:__	_____
	No_____2		
extractor fans/windows open	Yes_____1	Times__:__to__:__	
		Times__:__to__:__	
		Times__:__to__:__	
	No_____2	**GO TO QUESTION 4**	
4. **Were any animals in the house?**	Yes_____1	Time__:__ Room _____	_____
	Type_____	Time__:__ Room _____	_____
		Time__:__ Room _____	_____
	No_____2	**GO TO QUESTION 5**	

				Child near?
5. **Did you have any heating on?**		Yes_____1	**GO TO QUESTION 5(i)**	
		No_____2	**GO TO QUESTION 6**	
	(i) central heating (inc. radiators)	Yes_____1	Time__:__ Room _____	_____
			Time__:__ Room _____	_____
			Time__:__ Room _____	_____
		No_____2	**GO TO QUESTION 5(ii)**	
	(ii) free standing heaters/fans	Yes_____1	Time__:__ Room _____	_____
			Time__:__ Room _____	_____
			Time__:__ Room _____	_____
		No_____2	**GO TO QUESTION 5(iii)**	
	(iii) open fires (coal, wood, gas)	Yes_____1	Time__:__ Room _____	_____
			Time__:__ Room _____	_____
			Time__:__ Room _____	_____
		No_____2	**GO TO QUESTION 5(iv)**	
6. **Was there any active ventilation?**		Yes_____1	**GO TO QUESTION 6(I)**	
		No_____2	**GO TO QUESTION 7**	
	(i) windows/doors open	Yes_____1	Time__:__to__:__Room__	_____
			Time__:__to__:__Room__	_____
			Time__:__to__:__Room__	_____
		No_____2	**GO TO QUESTION 6(ii)**	
	(ii) double/secondary glazing open	Yes_____1	Time__:__to__:__Room__	_____
			Time__:__to__:__Room__	_____
			Time__:__to__:__Room__	_____
		No_____2	**GO TO QUESTION 6(iii)**	
	(iii) air conditioning/extractor fans	Yes_____1	Time__:__to__:__Room__	_____
			Time__:__to__:__Room__	_____
			Time__:__to__:__Room__	_____
		No_____2	**GO TO QUESTION 7**	
7. **Were any smokers present in the house?**		Yes_____1	Time__:__Room No.____	_____
			Time__:__Room No.____	_____
			Time__:__Room No.____	_____
		No_____2	**END OF QUESTIONNAIRE**	

Appendix 7

Weighing Protocol

Weighing Methodology

Transfer the filters to individual plastic 50 mm diameter single vented petri dishes (Western Laboratory Services, UK). Uniquely mark each filter with a sticker. The Personal Environmental Monitors (PEM's) filters marked as PEM97-F or PEM97-C depending upon the size fraction being collected and a number. The Harvard Impactor filters marked HI97-F or C as in the PEM marking. Inspect all filters for any defects such as tears, if any are damaged return to the manufacturer. A rubber bulb was used to blow off any extraneous lint or particles from the filters prior to placing in the petri dishes.

Handle all filters using non-serrated forceps to prevent damage to the filter and contamination from using hands. Wipe the forceps with the lint-free tissues dampened with distilled water.

Prior to commencing weighing note the temperature and humidity readings, if they fall outside of the required ranges then do not commence weighing.

Commence weighing by calibrating the balance with a standard 200 mg weight. Place onto the pan using the forceps, then close the door. When the balance settles the ready light illuminates, once the readout has settled for ten seconds the weight can be noted. If it is 200 mg ± 3μg this is acceptable, if not then press the calibrate key. Remove the standard weight and allow to settle until the readout is zero ± 3μg. If this does not occur then recalibrate.

Weigh the control filter specific to the filter type being weighed, try to use one per box of filters. This should fall within ± 10 μg of its original weighing. If not then leave for a further 24 hours.

All filters are weighed twice at least 24 hours apart to ensure precision. The second weighing should fall within ± 10 μg of the first weighing, if not re-weigh again after a further 24 hours until two of the weighings fall within the ± 10 μg range.

Every tenth filter the control filter is reweighed. If it does not fall within the ± 10 μg of the original weighing then the previous ten filters are re-weighed.

After each filter is weighed the balance should re-zero to ± 3μg, if not, follow the same procedure to recalibrate again.

For OFF weighing the filters should be inspected for any problems such as tears or rips, any excessively large specks or bugs should be removed using the forceps.

At the end of the weighing the standard weight 200 mg should be re-weighed to within 200 mg ± 3μg. If this is out of range then re-weigh all filters again.

Appendix 8

PEM Assembly Method

Personal Environmental Monitor Assembly

Assembly of all PEM's took place in the clean room to reduce the likelihood of contamination. Bench tops were covered with Kimwipes to reduce contamination. PEM bases and tops were paired up. Each pair includes a PM_{10} and $PM_{2.5}$ PEM.

Backing screens are placed onto the bases using forceps, followed by backing pads which must be gently tapped to remove any excess lint present. Cover with lint-free tissues followed by Teflon filters. The impactor ring should be placed on top of the filter ensuring that the lip of the base is still showing.

Using forceps place a silicon spacer ring into the top. For the PM_{10} top ensure that five of the ten holes are covered with the sticky paper circles and for the $PM_{2.5}$ two of the ten holes are covered.

Clamp the tops and bases together using the screws and screwdriver. They should be hand tight but not tight enough to cause them to be curved.

Attach the label to the base which refers to the filter that is included in the PEM. Place both PEM's into two resealable bags for carriage into the field.

Cleaning of the PEM components requires one washing in liquid soap and distilled water and rinse twice in distilled water, then dry overnight in Kimwipes.

Appendix 9

Harvard Impactor Assembly

Clean the bench top with Kimwipes and distilled water, then cover surface with clean Kimwipes.

Remove the impactors from the resealable bags and place on top of the bench and tissues. Separate the HI's into the component parts. Remove the filter from the holder using the forceps and place into the relevant labelled petri dish. Remove the label from the HI base. Clean each component with a Kimwipe dampened with distilled water. Use a different Kimwipe for each type of component to prevent cross-contamination. Wipe the impactor plate, mark each time they are used. Add one drop of the mineral oil and allow to be absorbed into the sintered surface.

Assemble all components of the HI, leave the base separate to allow inclusion of the filter and holder. Attach two labels to the base that refer to the filter being inserted, this ensures that there is a label to remove in the field for identification on the log sheet whilst leaving one on the HI for identification in the lab when disassembling. The labels are coded with HI97-C and a unique number for the coarse fraction and HI97-F for the fine fraction. The impactors are identifiable for each size fraction through the colour of the nozzle, red being the coarse fraction and silver the fine fraction.

Insert the unused filter onto the filter holder using forceps, ensure that the backing pad and drain disc are centred. Place the filter holder onto the base part of the HI then clamp the rest of the assembled HI. Ensure that the unit is centred to allow the air to flow through the filter only.

Place each HI into a resealable bag then pair up the HI's so that there is a PM_{10} and $PM_{2.5}$ size in each one. Place these into a larger resealable bag for carriage into the field.

Appendix 10

Air Exchange Method

Depressurisation Mode
Select a suitable external door to assemble the equipment on. The door should be in a frame, i.e. not half of double doors as this is not a solid enough frame to mount the false door through.

Assemble the false door frame on the floor then fit into the door-frame of the house. Expand until it fits but is still loose enough to remove again. Place the canvas on the floor then lay the frame on top tucking the canvas around the frame. Attach the external tubing to the canvas and place the end of it away from the fan so that the pressure gauges are not influenced by the running fan. Pull the frame and canvas into the door-frame ensuring that the canvas is pulled tightly around the frame and all the edges are inside the house.

Tighten the frame using the fine adjustment clips. Place the fan into the opening in the canvas ensuring that the canvas fits around the fan rim.

For the depressurisation mode the fan should be on the inside of the frame.

Attach the pressure gauges to an upright object, i.e. a nearby door, ensure that the spirit level is balanced. Blow into the gauges to ensure that the needles are mobile. Use the screwdriver to alter the needles' position if they are not at zero.

Attach the tubing from the gauges to the fan and plug the fan into the mains.

Measure the volume of the house to ascertain the required range to use with the fan, close all windows and open all the interior doors. In general terms the larger the house the more air will be required to fill it, the ranges of the fan are altered by removing the discs from the front of the fan.

Switch on the fan to maximum, the upper pressure gauge reading should be greater than the flow gauge, if not alter the range on the fan. Take ten readings by altering the fan flow, readings should be at 2 Pascal intervals. Note both readings from the gauges.

Whilst the fan is in the depressurisation mode and at maximum flow take the smoke stick and identify any leaks around windows, doors and chimneys in the house.

Pressurisation Mode.
Remove the fan and reverse its position so that the air blows into the house. Change over the tubing so that the background pressure readings are taken inside the building.

Repeat the measurements at 2 Pascal intervals.

Enter the data into the specialised program to find the air changes per hour. The mean value for the pressurisation and depressurisation mode is used to give the value for the whole house. To estimate the value for a specific room it must be sealed from the rest of the house and the entrance door to the room used for assembling the false door.

Appendix 11

Wilcoxon Signed Rank Tests

Wilcoxon Signed Ranks Tests

Summary of all children's personal PM_{10} concentrations

H_0 = No significant difference between values, Confidence levels of 95% are assumed

	n	Z Value	Asymp[1]. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
Brent PM10 Day	142	-9.039	.000	Reject
Garden PM10 Day	103	-6.700	.000	Reject
Garden PM2.5 Day	108	-8.232	.000	Reject
Home PM10 Day	114	-1.348	.178	Accept
Home PM2.5 Day	110	-7.773	.000	Reject
Haringey PM10 Day	150	-8.806	.000	Reject
Personal PM2.5 Day	122	-9.107	.000	Reject
School PM10 Day	50	-1.022	.307	Accept
School PM2.5 Day	46	-5.258	.000	Reject

Summary of all children's personal $PM_{2.5}$ concentrations

	n	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
Brent PM10 Day	142	-.231	.818	Accept
Garden PM10 Day	103	-3.191	.001	Reject
Garden PM2.5 Day	108	-4.290	.000	Reject
Home PM10 Day	114	-7.908	.000	Reject
Home PM2.5 Day	110	-4.148	.000	Reject
Haringey PM10 Day	150	-3.395	.001	Reject
School PM10 Day	50	-5.430	.000	Reject
School PM2.5 Day	46	-2.375	.018	Reject

Summary of all children's garden PM_{10} concentrations

	n	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
Day				
Brent PM10	142	-4.697	.000	Reject
Garden PM2.5	108	-7.661	.000	Reject
Home PM10	114	-7.263	.000	Reject
Home PM2.5	110	-.747	.455	Accept
Haringey PM10	150	-1.273	.203	Accept
School PM10	50	-3.940	.000	Reject
School PM2.5	46	-2.408	.016	Reject
Night				
Brent PM10	142	-3.928	.000	Reject
Garden PM2.5	98	-6.575	.000	Reject
Home PM10	90	-5.098	.000	Reject
Home PM2.5	93	-3.765	.000	Reject
Haringey PM10	150	-.337	.736	Accept
School PM10	2	-1.342	.180	Accept
School PM2.5	3	0.000	1.00	Accept

[1] Asymptomatic Significance (2-tailed)

ummary of all children's garden $PM_{2.5}$ concentrations

	n	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
ay				
rent PM10	142	-6.674	.000	Reject
arden PM2.5	108	-8.148	.000	Reject
ome PM10	114	-8.062	.000	Reject
ome PM2.5	110	-7.412	.000	Reject
aringey PM10	150	-8.135	.000	Reject
hool PM10	50	-5.086	.000	Reject
hool PM2.5	46	-4.843	.000	Reject
ight				
rent PM10	142	-6.986	.000	Reject
ome PM10	90	-7.473	.000	Reject
ome PM2.5	93	-3.028	.002	Reject
aringey PM10	150	-7.822	.000	Reject
hool PM10	2	-1.342	.180	Accept
hool PM2.5	3	-1.069	.285	Accept

ummary of home PM_{10} concentrations

	n	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
ay				
rent PM10	142	-8.711	.000	Reject
ome PM2.5	110	-8.347	.000	Reject
aringey PM10	150	-8.351	.000	Reject
hool PM10	50	-2.748	.006	Reject
hool PM2.5	46	-3.931	.000	Reject
ight				
rent PM10	142	-6.911	.000	Reject
ome PM2.5	90	-7.277	.000	Reject
aringey PM10	150	-5.329	.000	Reject
hool PM10	2	-1.342	.180	Accept
hool PM2.5	3	-1.604	.109	Accept

ummary of home $PM_{2.5}$ concentrations

	n	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
ay				
rent PM10	142	-3.305	.001	Reject
aringey PM10	150	-.249	.803	Accept
hool PM10	50	-4.798	.000	Reject
hool PM2.5	46	-.935	.350	Accept
ight				
rent PM10	142	-2.473	.013	Reject
aringey PM10	150	-4.454	.000	Reject
hool PM10	2	-1.342	.180	Accept
hool PM2.5	3	-1.069	.285	Accept

ummary of school PM_{10} concentrations

	N	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
ay				
rent PM10	142	-5.958	.000	Reject
aringey PM10	150	-6.077	.000	Reject
chool PM2.5	46	-5.265	.000	Reject
ight				
rent PM10	142	-1.342	.180	Accept
aringey PM10	150	-1.342	.180	Accept
chool PM2.5	2	-1.342	.180	Accept

ummary of School $PM_{2.5}$ concentrations

	N	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
ay				
rent PM10	142	-3.876	.000	Reject
aringey PM10	150	-2.802	.005	Reject
ight				
rent PM10	142	-1.069	.285	Accept
aringey PM10	150	0.000	1.000	Accept

ummary of Brent AUN concentrations

	N	Z Value	Asymp. Significance (2-tailed)	Null Hypothesis (Accepted/Rejected)
ay				
aringey PM10	150	-8.253	.000	Reject
ight				
aringey PM10	150	-10.050	.000	Reject

Appendix 12

Time Activity Diary Student T-Test Results

T-Test:Two-Sample Assuming Equal variances

Home	Spring v Summer	Spring v Winter	Summer v Winter
Pooled Variance	42585.02783	42378.86827	40566
Hypothesized Mean Difference	0	0	0
df	98	102	93
t Stat	-0.249845367	-0.736195444	-0.405936072
P(T<=t) one-tail	0.401614954	0.231650751	0.342861029
t Critical one-tail	1.660550879	1.659930149	1.66140353
P(T<=t) two-tail	0.803229909	0.463301503	0.685722058
t Critical two-tail	1.984467417	1.983494258	1.985799827
Outside	Spring v Summer	Spring v Winter	Summer v Winter
Pooled Variance	10187.63451	7846.50662	7227.978495
Hypothesized Mean Difference	0	0	0
df	98	103	93
t Stat	-0.025391357	1.39918474	1.41580578
P(T<=t) one-tail	0.489897219	0.082380936	0.080086373
t Critical one-tail	1.660550879	1.659782356	1.66140353
P(T<=t) two-tail	0.979794437	0.164761872	0.160172745
t Critical two-tail	1.984467417	1.983262337	1.985799827
School	Spring v Summer	Spring v Winter	Summer v Winter
Pooled Variance	Unequal variance	22068.88967	17831.52688
Hypothesized Mean Difference	0	0	0
df	98	103	93
t Stat	2.616838848	0.415271661	-2.2255605
P(T<=t) one-tail	0.00514008	0.339403418	0.01423019
t Critical one-tail	1.660550879	1.659782356	1.66140353
P(T<=t) two-tail	0.010280159	0.678806836	0.02846038
t Critical two-tail	1.984467417	1.983262337	1.985799827
Other Inside	Spring v Summer	Spring v Winter	Summer v Winter
Pooled Variance	10260.74212	5987.021624	9762.478495
Hypothesized Mean Difference	0	0	0
df	98	103	93
t Stat	-1.832071119	-0.524913621	1.446452129
P(T<=t) one-tail	0.034988981	0.300385632	0.075706214
t Critical one-tail	1.660550879	1.659782356	1.66140353
P(T<=t) two-tail	0.069977962	0.600771264	0.151412429
t Critical two-tail	1.984467417	1.983262337	1.985799827
Enclosed Transit	Spring v Summer	Spring v Winter	Summer v Winter
Pooled Variance	1830.723562	1309.372904	1504.887097
Hypothesized Mean Difference	0	0	0
df	98	103	93
t Stat	-0.782195387	0.759862161	1.517968878
P(T<=t) one-tail	0.217992811	0.224536356	0.066207216
t Critical one-tail	1.660550879	1.659782356	1.66140353
P(T<=t) two-tail	0.435985621	0.449072713	0.132414432
t Critical two-tail	1.984467417	1.983262337	1.985799827