EXPOSURE TO FINE PARTICULATE AIR POLLUTION IN PRINCE GEORGE, BRITISH COLUMBIA

by

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Abstract

The relationship between ambient fine particulate pollution ($PM_{2.5}$) and children's personal exposure was investigated during the winter of 2001 in the city of Prince George, British Columbia. Personal exposures of 15 children and ambient concentrations on their respective elementary school roofs were collected for a 6-week period. $PM_{2.5}$ mass, sulphate (SO_4^{2-}) and absorption coefficient (ABS) were determined for all samples (ABS as a surrogate for elemental carbon (EC)). SO_4^{2-} and EC personal/ambient ratios were used as tracers of ambient $PM_{2.5}$ to estimate personal exposure to ambient and non-ambient sources. Both SO_4^{2-} and EC ratios were found to be reliable tracers for exposure to particles of ambient origin. A strong association was found between ambient generated exposure and ambient concentration suggesting ambient levels were an appropriate surrogate for exposure to ambient $PM_{2.5}$ sources. The almost equal contributions made by ambient and non-ambient sources to total $PM_{2.5}$ personal exposure demonstrate the importance of managing ambient air quality. These findings strongly support the use of ambient data for a longitudinal health study in this city.

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1 Literature Review and Study Rationale

1.1. Literature Review:

1.1.1. Particulate Matter Formation and Sources

Atmospheric particulate matter (PM) is a mixture of solid particles and/or liquid droplets that may vary in concentration, composition and size distribution. Aerosols are defined as suspensions of solid or liquid particles in a gas (Wilson & Spengler, 1996). Thus an aerosol includes both the particle and the gas in which they are suspended. Although aerosol and particle are different, they are often used interchangeably throughout the literature to refer to the particle only. The two main types of particles are primary particles, which are introduced into the air directly in solid or liquid form, and secondary particles that form in the air through gas to particle conversion. The study of atmospheric particulate matter is important because it can influence radiation, cloud properties, human health and vegetation (Kerminen, 1999). This influence depends on several properties of the aerosol including mass, number, size and chemical composition of the particles. The size distribution of particles within the aerosol is also an important characteristic as it can strongly affect particle behaviour and fate in atmospheric systems as well as their deposition in the human respiratory tract (Mitra *et al* 2002). The presence and location of modes in the size distribution has also been associated with formation mechanisms (Morawska *et al*, 1999).

There are generally three modes found in atmospheric aerosols: the nucleation or nuclei mode that represents particles less than 0.1 μm, an accumulation mode that accounts for particles in the size range between 0.1 µm and 2.0 µm in aerodynamic diameter and a coarse mode where particles are greater than 2.0 µm (Brasseur *et al,* 1999). According to Morawska *et al* (1999), most of the nuclei mode originates from the condensation and coagulation of hot, highly supersaturated vapours released during combustion and the accumulation mode generally results from processes of coagulation and heterogeneous nucleation. The origin of almost all particles in the coarse particle mode is from natural and anthropogenic mechanical processes. Generally, the nuclei and accumulation particle modes are considered to make up the fine particle component of an atmospheric aerosol, while the coarse range alone accounts for the coarse particle component. The terminology used to refer to the different size fractions is different throughout the literature but a recent review article suggests the most common definitions are a fine fraction with particles smaller than 1 µm and a coarse fraction that ranges from 1 to 10 µm (Mitra *et al,* 2002). This is a natural division in that particles below 1 µm are generated mainly from combustion sources and are smaller than particles which are generated from mechanical processes.

Coarse particles are the result of direct emissions or they are formed by the break up of bigger particles into smaller particles. As particles become smaller, more and more energy is required to break them up, resulting in a lower limit for a coarse particle of about 1 µm (Wilson and Suh, 1997). There are both natural and anthropogenic sources of coarse particles. Some natural sources include windblown soil, evaporation of sea-spray, volcanic ash, pollen, mold spores and parts of plants and insects. Anthropogenic sources of coarse particles include re-entrained dust generated by traffic on paved and unpaved roads, debris from the demolition of buildings, piles of material containing coarse dust and fly ash from industrial boilers and waste incinerators. The chemical components of coarse particles usually are dominated by crustal elements such as iron, calcium, silicon and aluminum as well as sea water species of sodium and chloride (Brasseur *et al,* 1999).

Fine liquid or solid particles can come directly from combustion sources such as an industrial plume or automobile but they are usually formed from gases through the processes of homogeneous and heterogeneous nucleation or gas-to-particle conversion. Substances with low saturation vapour pressure are generated in the gas phase by high temperature vaporization or through chemical reaction in the atmosphere (Wilson and Suh, 1997). Because of the low vapour pressure, gas preferentially partitions into the liquid or solid state resulting in the spontaneous growth of a particle (Brasseur *et al,* 1999). A considerable energy barrier must be overcome in order for homogeneous nucleation to occur, therefore gas species may alternately condense on existing particles resulting in growth of the particle. Heterogeneous nucleation is a similar process where vapour-to-liquid transition occurs in the presence of nuclei range particles or ions, resulting in particle formation and growth (Hinds, 1982). Particle formation can also occur when liquid droplets in the atmosphere such as cloud or fog provide an aqueous medium that is capable of gas uptake. As a result of this process, droplets that contain secondary components arise that can convert to dry particles with changes in atmospheric humidity (Harrison and van Grieken, 1998). Growth of particles in the nuclei mode can also result from the process of coagulation, where two small particles combine to form a larger particle (Wilson and Suh, 1997). The chemical composition of fine particles varies widely, but generally includes trace metals, semi-volatile hydrocarbons and soot. Ionic species including sulphate, nitrate and ammonium are also important fine fraction components that result from the oxidation of sulphur and nitrogen compounds (Brasseur *et al,* 1999).

Fine and coarse particles differ in sources, formation mechanisms, composition, atmospheric life-times, spatial distribution, indoor-outdoor ratios, and temporal variability, as well as size (Wilson and Suh, 1997). It is also suspected that they may differ in biological effects. Of the commonly measured size fractions, particulate matter less than 2.5 microns in aerodynamic diameter ($PM_{2.5}$) has been recognized as having the greatest effect on human health (CCME, 2000). This is due to their small size, ability to penetrate and deposit in the respiratory tract and their common origin from combustion processes suggesting a more reactive chemical composition (Brauer, 2002). For the purposes of this thesis fine particulate or fine particles will refer to the $PM_{2.5}$ size fraction. To help reduce and manage these effects the Canadian Council of Ministers of the Environment (CCME) has established a $PM_{2.5}$ Canada-Wide Standard of 30 $\mu g/m^3$ for a 24-hour period in the outside (ambient) air with the target of achievement for the year 2010 based on averaging of the 98th percentile over three consecutive years (CCME, 2000). A Reference Level was also set at 15 $\mu g/m^3$ for a 24-hour period (CEPA/FPAC, 1999). Research has demonstrated health effects above this Reference Level and it is an estimate of the lowest ambient PM level at which statistically significant increases in health responses can be detected (CEPA/FPAC, 1999). In the U.S., the 24-hour average $PM_{2.5}$ standard is 65μg/m^3 and is not to be exceeded more than once per year (EPA, 2003).

1.1.2. Health Effects of Particulate Matter

The two main ways of investigating health effects of air pollutants are through epidemiological (observational) and clinical (experimental) studies. In epidemiological studies, there are several approaches: time series, cohort, panel and cross-sectional. Time series, cohort and panel studies are all longitudinal in design and subjects are followed over time. In a time series epidemiological study, continuous temporal patterns of mortality, or other less severe health end-points, are compared with weather and air pollution patterns in search of consistent relationships, while other variables changing with time are also considered (Lipfert, 1995). This type of study is the most common observational method used for looking at the health effects of air pollution and is a population based analysis where individual differences are not taken into account. A cohort study is a specific type of longitudinal analysis where a sample population is identified and exposure is estimated for the members of the sample (Vedal, 1997). The sample group is then followed over a period of time looking for the occurrence of an adverse health outcome. This type of study does not generally conform to the traditional cohort study because it is often not possible to estimate exposure of each subject in the study although other important information is known about each individual. Panel studies involve a select group of individuals that are followed on a continuing basis while exposure, daily activity and health indicators are measured for each individual

(Bates and Vedal, 2002). Panel studies are often limited by sample size compared to other study types but can provide important insight into exposure effects for the study subjects as well as other individuals with similar activity patterns, health status etc. Cross-sectional studies attempt to reveal associations between air pollution and health by virtue of similarities in spatial patterns (Lipfert, 1995). A large sample population is assessed at one point in time (usually from different locations), for which data on current symptoms or illness can be obtained and exposure can be estimated from concurrent measurements of pollutant concentrations. All other spatial variables must be accounted for such as demography, socio-economic factors, climatic factors and other environmental factors. For all air pollution health studies the concept of an ecological study must be considered (Vedal, 1997). Most often the air pollutant concentration is only know for ambient conditions and health outcome data is only available at a population level; except for in a panel study it is not specifically known for each individual. This can result in measurement error for the individual and also result in problems with covariates that are not known at the population level.

Evidence of an association between respiratory health and high levels of particulate matter has existed since the 1970s (CEPA/FPAC, 1999). Analyses of several episodes of air pollution that contained high particle concentrations have shown a clear association with observed morbidity and mortality. Since that time a multitude of epidemiological studies have been undertaken to determine whether adverse effects also result from lower concentrations of air pollution and more specifically if they are due to particulate matter, another component of the air pollution mixture, or the combination of various pollutants. A detailed summary of epidemiological acute and chronic studies can be found in the National Ambient Air Quality Objectives for Particulate Matter Science Assessment Document (CEPA/FPAC, 1999). A comprehensive review is also provided in the current EPA Air Quality Criteria for Particulate Matter Document (EPA, 2003). Several other reviews of research regarding the health effects of fine particles specifically are also available (NRC, 1998; Pope, 2000; and Vedal, 1997). Most of the studies report a 3 to 9 percent increase in daily mortality for a 50 $\mu g/m^3$ increase in PM_{10} with both cardiovascular and respiratory disease mortality being affected and the latter having a greater increase (Bates and Vedal, 2002). The review by Pope (2000) reported typical changes in daily mortality of 0.5 to 1.5% for a 10 $\mu g/m^3$ increase in PM_{10} or a 5 or 6 $\mu g/m^3$ increase in $PM_{2.5}$. A few of the main generalizations reported by Bates and Vedal (2002) regarding the PM/mortality relationship are: one third of the 90 US regions analyzed did not demonstrate an association; associations were stronger with $PM_{2.5}$ compared to PM_{10}; there was no significant confounding by weather; and there was no evidence of a threshold below which there are no health impacts. An important study by Samet *et al* (2000) assessed the effects of five major pollutants on daily mortality rates in 20 U.S. cities (1987-1994) and found consistent evidence that PM_{10} was associated with the death rate from all causes as well as from cardiovascular and respiratory illness. For an increase in 10 $\mu g/m^3$ of particulate matter the study found a corresponding increase in the relative rate of death from cardiovascular and respiratory illness of 0.68 percent and 0.51 percent from all causes. Data from a long-running study of six cities in the U.S. showed a stronger association between $PM_{2.5}$ and mortality, with a 10 $\mu g/m^3$ increase in $PM_{2.5}$ concentration resulting in a 1.5% increase in mortality compared to an 0.8% increase for PM_{10} (Schwartz *et al*, 1996). Many studies have also looked at various indicators of morbidity such as hospital admissions, emergency room visits, school or work absences and several specific health indicators that can be measured directly from individuals and found strong associations with ambient levels of particulate matter (Bates and Vedal, 2002).

In more recent years, this field of research has rapidly expanded as government agencies in the United States, Canada and other countries begin to recognize the role that particulate matter plays in the health impacts of air pollution. A review was recently published by Lippmann *et al* (2003) summarizing the progress of research in the United States and identifying the achievement of a better understanding of PM health effects and scientific uncertainties. Epidemiological studies have continued to show that an association does exist between mass concentration of ambient particulate matter and adverse respiratory and cardiovascular health effects. Progress has been made in understanding the biological mechanism behind the health effects of fine particulate and the extent of both acute and chronic effects although more research is still needed. There are more than 150 published epidemiological studies and dozens of published reviews assessing the epidemiological evidence of the human health effect of particulate air pollution (Pope, 2000). A review by Pope (2000) concluded that a more complete understanding of the health effects of particulate air pollution will require additional contributions from toxicology, exposure assessment and other disciplines. There is still a significant amount of error that is unavoidable when performing a health study, and although technology and methods have improved, this error must be accepted in order to make important regulatory decisions. Numerous studies have suggested that an association exists and in each of these studies confounding factors are unlikely to have been the same. The relationship has been demonstrated in areas where several variables are different including pollutant mix, meteorology, time of year and with the use of different methodologies. The observed particle effects have persisted despite the many approaches used to control for different variables. Although it is still not completely clear whether it is concentration, number of particles, size or chemical composition that results in an adverse health effect, we do know that a health hazard is posed by the atmospheric aerosol and measures must be taken to protect public health.

1.1.3. Role of Exposure Assessment

One of the main criticisms of epidemiological studies has been misrepresentation of exposure. Data from ambient pollutant monitoring stations is most often used as a surrogate measure for personal exposure. Ambient monitoring at a single location in an urban area may be a good indicator of population exposure for some areas but in other locations where there are local sources and complex topography; there could be greater variability in outdoor concentrations. A number of studies have looked at the spatial variation of fine particles throughout an urban area and the nearby rural areas. Some have found that particles have concentrations at multiple outdoor sites that are well correlated with a central monitoring location especially in areas influenced mostly by regional air pollutants (Adgate *et* al, 2002; Burton *et al*, 1996; Clayton *et al*, 1993; Ozkaynak *et al*, 1996; Spengler *et al*, 1981; and Williams *et al*, 2003). However, this information is site specific and would only be accurate for areas with similar local sources, geographical features and meteorological conditions. Cyrys *et al* (1998) claimed that use of one monitoring site might indeed be inaccurate, especially in communities with substantial local sources of pollutants and during periods of low wind speed. Cyrys found significant differences in both PM_{10} and sulphate between a downtown site and two suburban sites of 30-40% and up to 17% respectively in Erfurt, Eastern Germany. Other recent research has shown that although fine particle mass concentration may be found in general to have a uniform distribution within a city, the chemical composition may be more variable and dependent on very local sources such as traffic (Kinney *et* al, 2000; Hoek *et al*, 2002; and Roosli *et al*, 2001). It has also been found that spatial variability of fine particles is often related to varying altitude rather than distance from a central monitor due to the stability of the atmosphere and the existence of surface inversions (Roosli, 2000).

Using a single measurement to represent the exposure of a population may lead to a biased interpretation of exposure-response relationships (Watt *et al,* 1995). Not only does pollutant concentration vary as you move away from a monitoring station, but it is also very different from indoor concentrations where the majority of an individual's time may be spent. Ventilation rates and air conditioning will drastically affect an individual's daily exposure to ambient particulate matter, as well as indoor sources of particles and other pollutants. A nationwide study in the United States showed that based on data from 9,386 respondents, 87.2% of a person's time is spent indoors, 7.2% in or near a vehicle and 5.6% outdoors (Wallace, 1996). A review by Wallace (1996) also shows that very few homes are free of important indoor sources of particles. Although the health impact of indoor sources is unknown they must still be considered in exposure estimates in addition to the impact of outdoor sources directly and via the penetration of outdoor particles into buildings. Other important factors that must be considered in an individual's exposure estimate are occupational exposure and personal habits such as smoking. Understanding personal exposure is essential to the understanding of pollutant health effects and research regarding the accurate assessment of exposure to fine particles is a vital research need (National Research Council, 1998).

1.1.4. Personal Exposure to Particulate Matter

Over the past 30 years research has shown that members of a population often have very different exposures to particulate matter (Brauer, 2002). An individual's exposure can be estimated indirectly using ambient measurements with micro-environmental models or by direct measurement via personal sampling or the use of biological markers (Monn, 2001). Often questionnaires assessing time-activity patterns of individuals are used to enhance these methods. Research involving personal monitoring, where subjects wear a small sampling device, has provided useful information regarding contact patterns between an individual and air pollution that has aided in the assessment of the relative importance of outdoor air pollutants (Brauer, 2002).

Many of the original studies that used personal sampling to directly measure exposure to particles showed little association between the personal measurements and simultaneous outdoor concentrations when cross-sectional correlations were calculated (Clayton *et* al, 1993; Dockery and Spengler, 1981; and Ozkaynak *et al*, 1996). These findings caused many to question the results of the majority of health effect studies that used ambient concentration as a surrogate for personal exposure. Several more recent longitudinal $PM_{2.5}$ exposure studies have shown stronger personal-outdoor correlations exist when data are analyzed by individual over time but the degree of association varies widely by individual (Ebelt *et* al, 2000; Janssen *et al*, 1999, 2000; Liu *et* al, 2003; Rojas-Bracho *et al*, 2000; Sarnat *et al*, 2000; and Williams *et al*, 2003). Personal-outdoor associations have been shown to be even stronger for the sulphate (SO_4^{2-}) component of fine particles indicating that it may be a more appropriate measure of exposure to ambient generated particles (Brauer *et al*, 1989; Ebelt *et al*, 2000; Sarnat *et al*, 2000; Stieb *et al*, 1998; and Suh *et al*, 1992). Use of the absorption coefficient of $PM_{2.5}$ filters, a surrogate measure of the elemental carbon of PM, has also lead to a stronger association between personal exposure and ambient concentrations (Janssen *et al*, 2000).

Understanding the relationship between ambient concentration and personal exposure is crucial for validating and interpreting the results of the many epidemiological studies that have found an association between ambient fine particles and various health indicators. Research has shown that many factors can influence this relationship including: location factors such as topography and local sources; meteorology; housing characteristics like the ventilation and infiltration of outdoor air indoors; and finally individual differences in activity (time spent outdoors, cooking, cleaning or during transportation) and personal habits (smoking). It is clear from the literature that the relationship between personal exposure and ambient particulate level is site specific and should be further investigated in areas where health impacts are suspected.

1.1.5. Review of Local Research

In 1996, a public opinion survey was conducted to determine public perceptions of outdoor air quality issues in the City of Prince George (Oster, 1997). Results from this survey suggested that Prince George residents were dissatisfied with the current airshed conditions and they believed that poor air quality affected their quality of life. Respondents expressed the need for more education on air quality issues and called for increased public awareness regarding the negative impacts and human health consequences of poor air quality in the city.

Monitoring of various air pollutant concentrations was initiated in the city in 1980. At that time particles were assessed by measuring total suspended particulate and dust-fall, the amount of particles that settle or are washed out of the air by rainfall (Lamble *et al*, 1998). Monitoring of PM_{10} started in 1990 and of $PM_{2.5}$ in 1994. Annual reports summarizing ambient pollutant concentrations and assessing annual trends have been produced by the BC Ministry of Water, Land and Air Protection, but there has been no attempt to validate the accuracy of this database in representing actual population exposure.

Health indicators for Prince George residents and the surrounding area reflect generally higher levels of illness and mortality compared to the provincial average (Prince George Airshed Technical Management Committee, 1996). The Northern Interior Health region (which includes Prince George) also has the second highest level of respiratory mortality in the province (Prince George Airshed Technical Management Committee, 1996). Research investigating the health effects of air pollution in the city has been undertaken in the past. These studies examined the links between hospital admissions and emergency room visits for specific categories of respiratory disease and the levels of total suspended particulate (TSP) and total reduced sulphur (TRS) (Knight *et al.*, 1988, 1989 and McNeney and Petkau, 1991). The study in 1989 showed a small but clear association between ER visits and TRS but the 1991 study actually showed fewer ER visits on high pollution days. It was suspected that the population size may have been too small to measure an effect and the availability of air pollution data only every six days was limiting. A follow-up study used sophisticated regression models to estimate the short-term increased risk of daily ER visits associated with PM_{10}, sulphur dioxide (SO_2) and ozone (O_3), while controlling for both temporal and meteorological effects (Ahkong *et al*, 2000). The findings for PM_{10} showed that a decrease in concentration from 20 to 10 $\mu g/m^3$ both one and three days previously was associated with a predicted decrease in ER visits of about 0.2%. Larger effects were observed for SO_2 and O_3 using different lag-times and levels. Interestingly, when the pollutants were considered simultaneously, PM_{10} had the least significant effect and O_3 had most significant effect.

In 1996 and 1998 respectively, the Prince George Airshed Technical Management Committee published a background report and airshed management plan assessing the air pollution status in the city and outlining a plan to improve air quality. The need for research in several areas was identified including a more detailed investigation of health impacts on city residents and source contributions to ambient levels for fine particles. Since that time, a limited amount of research has been undertaken. A source apportionment study of PM_{10} investigated the contributions of two sources, road dust and beehive burners, to ambient samples from both episodes of high PM_{10} concentrations and non-episodes (Breed, 1998 and 2002). Qualitative analyses of particle size, shape and chemical composition suggested that finer particles ($PM_{2.5}$) and relatively higher levels of sulphur, suggestive of combustion sources, dominated the non-episode samples while coarser particles (3-4 µm), suggestive of a road dust source, dominated the episodes. One of the research recommendations that came out of this study included further investigation into the health effects of PM on city residents. Preliminary modelling studies of sulphur dioxide have also been performed to assess the use of meteorological and air pollution dispersion models to estimate concentration levels throughout the area (Noullett, 1999 and McEwen, 2002). A study initiated by local industry used complex meteorological and pollution dispersion modelling to assess the impact of local pulp mill PM_{10} emissions on the airshed as well as other sources such as beehive burners and road dust (Jacques Whitford, 1999). Study findings indicated that the pulp mill contributions to ambient PM_{10} levels were more significant during non-episodic periods compared to episodic periods (levels of PM_{10} greater than 50 µg/m3). During episodes there was also a large contribution of PM_{10} that was not accounted for by industrial emissions. Another study has been initiated to assess the link between air quality episodes and synoptic climatology (Willis, 2004). A final study reviewing the evidence of different source contributions to PM_{10} episodes is also in progress (Sutherland and Fudge, 2002). Despite the valuable contribution that previous and current research has provided, the lack of information regarding actual population and individual exposure of city residents is apparent.

1.2. Rationale for the Present Study

The Prince George Airshed has many local sources of various air pollutants including several major industrial sources (pulp mills, sawmills and an oil refinery), vehicle emissions, locomotives, unpaved and paved road surfaces, vegetative burning, residential and commercial heating etc. (Prince George Airshed Technical Management Committee, 1996). Because the city and its local sources of air pollution are contained within a valley, there are often meteorological conditions that trap pollutants and result in episodes of poor air quality and unhealthy levels of air pollution exposure. The 2001 air quality report for the Prince George airshed shows annual average $PM_{2.5}$ levels to be higher than all other BC locations (Fudge *et al*, 2003). The 2001 report also showed a comparison of health risk between BC communities based on days where $PM_{2.5}$ levels were greater than 15 $\mu g/m^3$ and Prince George had the highest estimates based on both continuous and non-continuous data. The Canada-Wide Standard for $PM_{2.5}$ was almost exceeded (3-year average of 98th percentile greater than 30 $\mu g/m^3$) in the city for the periods from 1998 to 2000 and from 1999 to 2001 with average 98th percentiles of 28.2 and 29.6 $\mu g/m^3$ respectively (Lamble *et al*, 2002 and Fudge *et al*, 2003). In both 2000 and 2001, the 30 $\mu g/m^3$ level was exceeded with single-year annual 98th percentiles of 32.1 and 32.5 $\mu g/m^3$.

When high levels of a pollutant exist in an airshed a question regarding associated health effects must be posed. In order to investigate and begin to understand health effects of any air pollutant, an understanding of population exposure and the ability of ambient monitors to represent the airshed must first be achieved. The first step in this process is to develop a detailed knowledge of the spatial variation of the pollutant of interest in the airshed. Exposure to ambient air pollutants varies by location due to differences in local sources, regional contributions and meteorological conditions. Once spatial variation is understood it is possible to assess the ability of the local ambient monitoring network in representing ambient pollution levels throughout the airshed. Finally, to gain a complete understanding of population exposure, personal monitoring measurements can be taken to verify the accuracy of ambient area measurements in representing an individual's exposure to ambient generated sources. If a representative sample of a population is studied more closely and their exposure is determined with consideration of individual differences then this information would provide a more accurate estimate of exposure for the local population compared to the use of ambient monitoring data alone. Characterizing actual personal exposure is a fundamental component of understanding the relationship between air pollution and specific health outcomes and must be done before any causal relationship can be identified.

It is clear from the literature that a significant health effect exists from ambient particulate matter but it is still not known whether it is concentration, number of particles, size, chemical composition or some combination of these factors that results in the adverse health effect. An argument was made by Schwartz *et al* (1996) that the relationship between health outcomes and fine particles less than 2.5 μm is the most justified. They claim that $PM_{2.5}$ can readily infiltrate residential buildings with indoor levels similar to levels immediately outside the structure, thus population exposure to fine particle mass has a higher correlation with day-to-day ambient particle measures than with coarse particle mass or with reactive gaseous pollutant concentrations. Wilson and Suh (1997) also suggest that statistical associations found between daily PM indicators and health outcomes likely results from variations in the fine particle component and not the coarse component. Schwartz *et al* (1999) showed that episodes of high concentrations of coarse particles (PM_{10}) during dust storms in Spokane, Washington were not associated with increased mortality and concluded that crustal particles likely have little toxicity. Because actual exposure estimates for fine particles based on ambient levels may be more accurate than for other pollutants, there is a greater ability to accept a correlation to health effects. It is also the finer particles that are thought to contain the more toxic components due to their formation mechanisms, therefore, it is generally thought that these particles are responsible for the health effects apparent in epidemiological studies.

Airborne particulate matter is a top priority in the Prince George Airshed and data regarding spatial variation and human exposure will help managers to better understand the impact of particulate matter within different areas of the airshed and on the people living in those areas. The Prince George Air Quality Management Plan recommended that a health study be started no later than the year 2000 and also suggests that research be initiated regarding source contributions, spatial impacts of individual sources and meteorological effects (Prince George Airshed Technical Management Committee, 1998). This thesis begins to address these recommendations made in the 1998 Management Plan and will lead the way for more research in the Prince George Airshed regarding exposure to air pollutants and the associated health impacts.

1.3. Goals and Objectives

The primary goal of this thesis was to characterize the relationship between outdoor concentration and children's personal exposure to $PM_{2.5}$ in the city of Prince George and to evaluate whether or not the current ambient monitoring network was representative of both outdoor $PM_{2.5}$ levels at unmonitored locations throughout the airshed and actual personal exposure. This goal will be achieved through the following objectives:

Objective #1 **-** Characterize the spatial variability of both ambient and personal $PM_{2.5}$, sulphate and absorption coefficient throughout the airshed and assess the influence of meteorological conditions.

Objective #2 **-** Establish an understanding of the relationship between personal $PM_{2.5}$, sulphate and absorption coefficient exposure and the corresponding ambient concentrations.

Objective #3 **-** Assess the relationship between ambient generated exposures and ambient concentrations.

Objective #4 **–** Determine the relative contribution of ambient and non-ambient generated particles to total $PM_{2.5}$ personal exposure.

Objective #5 **-** Verify the use of absorption coefficient as a surrogate measure for elemental carbon.

1.4. Overview of Study and Structure of Thesis

This research project involved two components: a pilot study to test sampling procedures and establish the site specific relationship between absorption coefficient

(ABS) and elemental carbon (EC) and a field study to assess the relationship between outdoor concentrations and actual personal exposures to $PM_{2.5}$, sulphate (SO_4^{2-}) and EC (determined via reflectance and calculated absorption coefficients).

The pilot study was conducted from November 26, 2000 to January 19, 2001 at the Plaza air pollution and meteorological monitoring site of the BC Ministry of Water, Land and Air Protection (BC MWLAP) (a map of the study area is provided as Figure 3-1 in Chapter 3). Chapter 2 includes a description of the study design and methods used for the pilot study and a summary of the quality control that was undertaken to insure acceptable data quality. The relationship between elemental carbon and absorption coefficient is quantified and a comparison between the Harvard Personal Environment Monitor (HPEM) and the BC MWLAP continuously operated tapered element oscillating microbalance (TEOM) is provided. Problems with ambient sampling during cold winter conditions are also discussed.

The field study operated for a six-week period in 2001 on week-days only (February 5th to March 16th). Five area monitors and five personal monitors were operated simultaneously for 6 weeks during the winter enabling an assessment of the spatial variation of both ambient concentration and personal exposures throughout the city. Personal exposure samples were collected from children at five elementary schools located throughout the city and corresponding ambient samples were collected from their respective elementary school roofs. Total $PM_{2.5}$ mass concentration, sulphate and elemental carbon (via a determination of absorption coefficient) were determined for all of the ambient and personal samples collected. Chapter 3 compares the ambient and personal data. Analysis is performed by pooling across all individuals and schools, pooling by school and by assessing the longitudinal correlation between the personal and ambient measures for each individual. Chapter 3 also includes a general assessment of local meteorology during the study, particularly the impact of frequent temperature inversions. A general discussion of the impact of wind speed and direction is also included.

Chapter 4 takes the data analysis one step further using the personal to ambient ratio for sulphate and elemental carbon to estimate exposure to ambient and non-ambient generated sources. The relationship between the ambient exposure estimate and ambient concentrations is characterized and a regression model is provided to enable future estimation for children of Prince George during the winter. The contribution that ambient generated and non-ambient generated sources have to total personal exposures is also investigated. Air exchange rates and infiltration factors are also calculated for the residence of each subject in the study.

A summary of the discussion and conclusions made throughout the thesis is provided in Chapter 5. Further investigations planned for the study data and recommendations relevant to airshed management are discussed. Chapters 2, 3 and 4 are written as individual studies each including an abstract, introduction and conclusions.

2 Pilot Study

2.1. Abstract

A study sampling ambient fine particulate pollution ($PM_{2.5}$) was conducted from November 26, 2000 to January 19, 2001 in the city of Prince George, Canada. The purpose of this study was to test sampling procedures that would be used in a personal exposure study and to investigate the site specific relationship between elemental carbon levels and absorption coefficient. $PM_{2.5}$ concentrations obtained from a Harvard personal environment monitor (HPEM) were compared to data from the BC Ministry of Water, Land and Air Protection continuously operated tapered element oscillating microbalance (TEOM). The comparison showed that the HPEM concentrations were higher with a mean difference of 4.3 ± 3.2 µg/m^3. The medians and means of the TEOM and HPEM data were both significantly different but there was a high significant correlation with an R^2 of 0.93. Two methods were used for determining elemental carbon concentrations and the results from both methods demonstrated a strong and significant correlation with absorption coefficient with Pearson r values of 0.90 and 0.85 respectively. The site specific relationship between absorption coefficient and elemental carbon is described by the regression equation: Elemental Carbon (µg/m^3) = 0.34*absorption coefficient + 0.03.

A description of the study design and methods used for this pilot study and a summary of the quality control that was undertaken to insure acceptable data quality is also provided. Problems encountered due to cold winter weather are discussed and successful adaptations are described.

2.2. Introduction

In the city of Prince George, British Columbia, Canada fine particulate pollution ($PM_{2.5}$), or particles smaller than 2.5 μm in aerodynamic diameter, has been recognized as a serious health concern. Currently only one continuous monitor and one non-continuous monitor, on a 6-day cycle, monitor the ambient levels of this pollutant (Lamble *et al*, 2002). The annual averages in 2000 and 2001 were 9.51 and 9.46 $\mu g/m^3$, with corresponding 98th percentiles of 32.1 and 32.5 $\mu g/m^3$ that exceed the 30$\mu g/m^3$ level of the Canada-Wide standard (Fudge *et al*, 2003). There is a need for more detailed information about ambient levels of fine particulate throughout the city to determine if the current monitoring program is suitable for assessing community exposure (Prince George Airshed Technical Management Committee, 1998). Actual personal exposure measurements are necessary in order to make an assessment of health impacts on city residents (CEPA/FPAC, 1998).

A pilot study was run from November 26, 2000 to January 19, 2001 to test the sampling equipment and protocol and to train research staff. Fifty-nine 24-hour samples were collected on a daily basis excluding December 24th, 25th, 31st and January 1st. The first objective of the pilot study was to provide a comparison of ambient concentrations obtained from Harvard personal environment monitors (HPEMs) to those obtained from a tapered element oscillating microbalance (TEOM). Different sampling devices are used to measure $PM_{2.5}$ by various researchers so it is important to provide a comparison to a more common measuring device such as the TEOM. It would have been preferable to make a comparison to a federal reference method (FRM) for $PM_{2.5}$ but sufficient data were not available for such a comparison.

The second objective of the pilot study was to determine the relationship between an absorption coefficient (ABS) and actual elemental carbon (EC) concentrations for the Prince George Airshed. Elemental carbon determination involves lab analysis that destroys the filter. It also requires that a different filter type be used for sampling and the implementation of more stringent handling and storage procedures. During the main field study these requirements would not be possible so a different method was required to assess elemental carbon content of the fine particulate samples. Absorption coefficient has been suggested as a surrogate measure of elemental carbon concentration and a reliable indicator of traffic-related particulate matter (Cyrys *et* al, 2003; Fisher *et al*, 2000; Janssen *et al*, 2000, 2001; and Kingham *et al*, 2000). The relationship between absorption coefficient and elemental carbon may be different depending on local sources so it is necessary to provide a site specific comparison to support the use of absorption coefficient as a surrogate measure (Cyrs *et al*, 2003).

This chapter includes a description of the study design and methods used for this pilot study and a summary of the quality control that was undertaken to insure acceptable data quality. The relationship between elemental carbon and absorption coefficient is quantified and a comparison between the Harvard PEM and the continuously operated TEOM is provided. A discussion of problems with ambient sampling during cold winter conditions is also included.

2.3. Methods

2.3.1. Sampling Instrumentation and Procedure

Two monitoring stations were set up on the roof of the Plaza 400 building where the existing $PM_{2.5}$ monitors were located. Figure 2-1 shows the temporary monitoring site used for the pilot study.

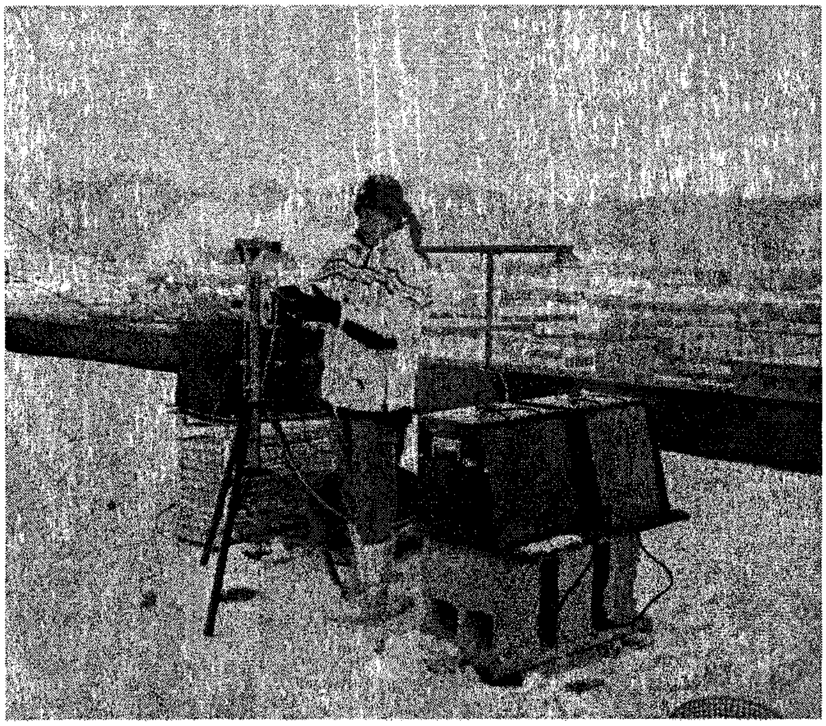


Figure 2-1 Pilot study monitoring site located on the roof of the Plaza building downtown Prince George where the Ministry of Water, Land and Air Protection operates $PM_{2.5}$ monitors.

The first station collected a 24-hour integrated sample on 37mm, 2 μm pore size Teflon filters (Pall Gelman #R2PJ037), suitable for determining total $PM_{2.5}$

concentration, absorption coefficient and sulphate level. The 2 μm pore size of the Teflon filters have 99.99% particle retention for a particle diameter of 0.3 μm, which is a standard test size. The second station collected a 24-hour integrated sample on a quartz fibre filter, 37mm (Pall 2500QAT-UP #7201), suitable for elemental carbon analysis. These two sampling stations were operated simultaneously in order to provide a means of comparing elemental carbon concentration to absorption coefficient calculated from a simple reflectance measurement taken from the Teflon filters.

The sampling device used for both filter types was a $PM_{2.5}$ Harvard personal environment monitor or HPEM designed by researchers at the Harvard School of Public Health and described by Demokritou *et al* (2001). The HPEM consists of a sampling inlet designed to direct particles smaller than 2.5 μm around a greased impactor plate and onto a 37 mm filter supported by a mesh screen. This sampler was designed for personal monitoring and is not generally used for collecting ambient $PM_{2.5}$ samples. It was selected for this study due to availability and cost. Using the personal monitor for ambient sampling also enabled a more straightforward comparison between ambient and personal measurements taken during the main field study. Other researchers have also used personal samplers for ambient measurements during a personal exposure study (Janssen *et al,* 1999 and Ozkaynak *et al*, 1996).

The samplers were suspended approximately 4 feet above the rooftop underneath a metal rain shield (stainless steel mixing bowl) using a powder-coated hanger (Figure 2-2). This protected the sampler from wind, rain and snow. By operating the personal samplers at the same location as the current continuous and non-continuous $PM_{2.5}$ monitors, a comparison could be made to assess the validity of using the HPEM as an ambient monitor in the Prince George airshed. A comparison to the Federal Reference Method for the $PM_{2.5}$ HPEM has been performed by Liu *et al* (2003). A linear regression coefficient (R^2), slope and intercept of 0.87, 0.88 and 1.64 were reported respectively. It was also found that a negligible bias existed when the $PM_{2.5}$ HPEM with greased impactor plates was compared with co-located results from a single-stage inertial Harvard Impactor (mean difference = 0.4 $\mu g/m^3$). Ward-Brown (2000) also compared the $PM_{2.5}$ HPEM to an ambient Harvard Impactor and reported an R^2, slope and intercept of 0.98, 1.08 and 1.25 respectively.

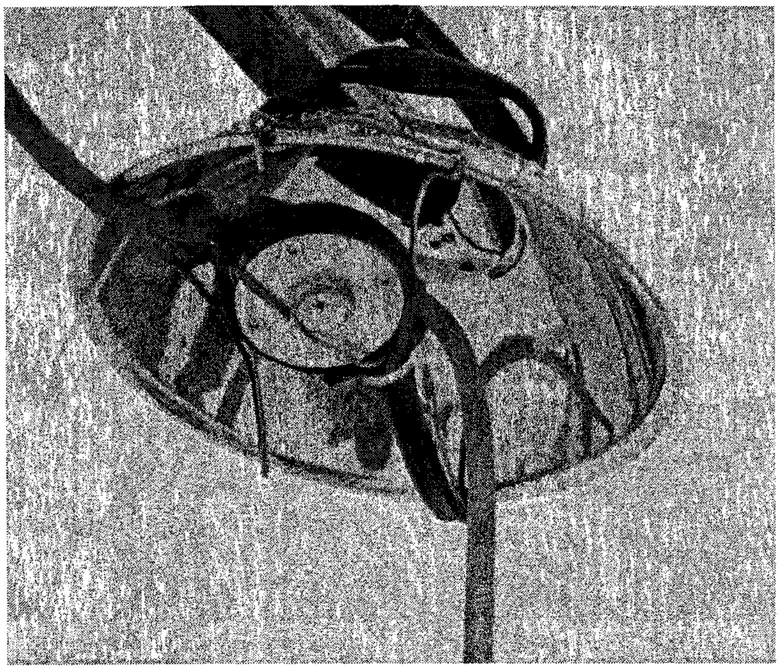


Figure 2-2 The Harvard $PM_{2.5}$ Personal Environment Monitor (HPEM) was suspended under a rain shield at the ambient monitoring sites. Heat tape is wrapped around the outside diameter to keep the sampler at 0°C. This prevented frost from forming across the sampler inlet.

Each HPEM was connected to a large flow controlled pump that produced a flow rate of approximately 4.0 litres per minute (LPM). The target flow range when starting samples was 3.8 to 4.2 LPM. The acceptable flow range when removing samples was 3.6 to 4.4 LPM or within 10%. A flow rate of 4.0 litres per minute and an acceptable range of ±10% was also reported by Ebelt (2000) and Janssen (1998b). Flow measurements were taken when the samplers were started and then again before the samplers were removed approximately 24 hours later. A BIOS frictionless piston meter (BIOS DryCal DC-1) was used to take all flow measurements. Initially, there were some problems with the BIOS in very cold weather but using a duffle bag and waterbed heater proved effective for keeping the instrument warm. Air was sampled through tubing from outside of the bag to ensure that there was no error introduced into the measurements due to heating. Precision rotameters (Matheson, model #603) were also used to take flow measurements for a few instances when there were operating problems with the BIOS during the pilot study. The rotameters were calibrated in the laboratory with the BIOS frictionless piston meter. Rotameters were needed in the main field study as there was only one BIOS and three field workers taking flow measurements simultaneously at the different monitoring sites. The rotameters also appeared to have some problems due to the cold weather, including irregular changes in flow and unstable readings, and were only used as a back up. The outdoor pumps were very consistent and flow adjustments were limited.

During the pilot study, 9 samples (17%) were lost due to problems with frost collecting on the sampling inlet. The frost plugged the sampler with no way of knowing the time frame that this occurred and consequently it was not possible to calculate sample volume. In order to prevent frost formation over the sampling inlet, the outer diameter of the sampler was encircled by plumbers heat tape (purchased at the local hardware store). This kept the sampler at 0° Celsius and proved effective at preventing frost formation for the remainder of the study.

The TEOM sampler (1400AB, Rupprecht & Patashnick) measures the mass collected on an exchangeable filter cartridge by monitoring the corresponding frequency changes of a tapered element. It is operated continuously by the BC Ministry of Water, Land and Air protection at a temperature of 40 degrees Celsius to remove water vapour. The sample flow is automatically adjusted by temperature and pressure to maintain a correct volumetric flow rate of 16.7 litres per minute. Direct audit and calibration procedures are performed for mass measurement and flow rate using NIST-traceable standards. No corrections are made to the data to account for the possible loss of semi-volatile particulate matter such as sulphate or nitrates due to the operation temperature.

2.3.2. Quality Control/Assurance

Throughout the pilot study both lab and field blanks were used as a means of ensuring that the samples were not contaminated. The lab blanks underwent the same steps that the actual samples did in the lab. They were loaded into samplers, tested for leakage by attaching to a pump and measuring any change in flow rate and then stored in a Ziploc bag at room temperature until after sampling was completed. They were then unloaded with the same batch of samples and the filters were stored for final analysis at room temperature. With each batch of samples a field blank was also loaded into a sampler, leak tested and than taken out into the field. Once it was in the field it was exposed to the air and then returned to the sealed Ziploc bag used for transport. The field blanks were then unloaded with the same batch of samples and stored. A second, extended field blank was also collected. The sampler was actually left in the field for the entire 24-hour sampling period but was not connected to the sampling pump. These additional field blanks were collected to ensure that there was not any additional contamination introduced in the field during the actual sampling period. The number of lab and field blanks collected was respectively 18 and 19% of the total samples collected. The number of extended field blanks collected was 7% of the total samples collected. On 5 days during the pilot study, successful co-located samples were collected. This was done to enable an assessment of the precision of the sampling method. At both of the sampling stations two samples were collected simultaneously on the same filter type in order to provide data to assess the accuracy of the sampling method. This was performed on 9% of the total sampling days.

2.3.3. Lab Preparation and Analysis

The HPEMs were cleaned and re-greased daily and underwent a deep-cleaning every 5 sampling days. All cleaning and loading of samplers followed the standard operating procedure (SOP) provided by the Harvard School of Public Health and is included in Appendix 5 (Ward-Brown, 2000).

Prior to sampling, the quartz fibre filters were pre-fired at 500°C for 3 hours to remove any possible contamination and then wrapped in tinfoil and stored in a hermetic glass jar kept in a refrigerator. After sampling the quartz filters were stored in an Analyslide holder (Pall 7231) and stored in a deep freeze. A mask was used with the quartz filters when sampling to concentrate the sample on a smaller area (~3/4 inch) of the filter. Before shipping to the Thompson Laboratory Building, Air Quality Research Branch of the Meteorological Service of Canada (Toronto, Ontario) for analysis, a stainless steel punch (McMaster-Carr 3427 A19) was used to cut and remove the concentrated area of the filter in preparation for the elemental carbon analysis. Elemental carbon analysis was performed via thermal optical transmittance (TOT) on the quartz fibre filters following the procedure described by Sharma *et al* (2002). This hybrid method was developed to enable comparison between two accepted methods of organic and elemental carbon determination that yield slightly different results. Comparison between the TOT method and both the NIOSH 5040 method and the Desert Research Institute (DRI) IMPROVE Thermal Optical Reflectance (TOR) approach is also shown by Sharma *et al* (2002). The two sets of elemental carbon data analyzed are identified as NIOSH and DRI/IMPROVE to indicate the method of analysis with which the results are comparable.

Before and after sampling, Teflon filters were stored at room temperature in sterile Petri dishes. The Teflon filters were weighed prior to sampling and then again after sampling. Before any gravimetric measurements were performed the filters were equilibrated for 48 hours in a temperature (21.9 ± 0.3°C) and humidity (41± 3%) controlled weighing room at the School of Occupational and Environmental Hygiene of the University of British Columbia in Vancouver. A microbalance (Sartorious M3P; 1 μg resolution) was used to make triplicate measurements of filter weight and agreement was required to be within 5 μg. An external calibration was performed daily using 5, 10 and 20 mg NIST-traceable weights. Frequent internal calibration and triplicate weighing of a test blank filter every 25 filters ensured that accuracy of the instrument was maintained during weighing sessions. Post weighing was performed on the pilot study filters prior to the start of the main field study in order to identify any possible problems. During the field study, filters were stored and then taken to the UBC laboratory in Vancouver for analysis at the end of the study.

After gravimetric analysis and a quality assurance check of the data, Teflon filters underwent reflectometry at the UBC lab. The “blackness” of the $PM_{2.5}$ filter was measured using a reflectometer (M43D Smoke Stain Reflectometer, Diffusion Systems Ltd., London, UK), which measures the reflection of the light incidence in percent. The reflectance analysis followed the standard operating procedure from the ULTRA study to determine absorption coefficient using a reflectometric method and was obtained from researchers at the University of Wageningen in the Netherlands (ULTRA, 1998). This procedure has been used by other researchers (Cyrys *et al,* 2003; Fischer *et al*, 2000; Gotshci *et* al, 2002; Janssen *et al,* 2000; and Kingham *et al*, 2000). Briefly, blank filters were used to set reflectance at 100 percent and then the reflectance was measured on five different spots on each sampled filter. An absorption coefficient (ABS) was then calculated from the average reflectance for each filter using the following formula (International Standard ISO 9835):

$$\text{(Equation 2.1)} \qquad \text{ABS (meters}^{-1}) = 0.5A \ln (R_F/R_S)/V$$

where A is the area of the stain on the filter (7.55×10^{-4} m^2); R_F is the average reflectance of the field blank filters in percent; R_S is the reflectance of the sample filter in percent; V is the sample volume in cubic meters (ISO, 1993). The absorption coefficient is multiplied by 10^{-5} for the purpose of reporting.

Sulphate analysis was also performed on the samples at the UBC lab after all gravimetric and reflectance measurements were completed. An extraction was done on the filters by wetting the filter with 100 µl of ethanol and sonicating in 5 ml of distilled/deionized water for 15 minutes in polyethylene containers (Ebelt *et al*, 2000 and Koutrakis *et al*, 1988). The extract was then analyzed using an ion chromatograph (Dionex, DX-300) with suppressed conductivity detection.

2.4. Results and Discussion

2.4.1. Data Quality

For the pilot study the mean increase of mass on the lab blanks, field blanks and extended field blanks were 4 µg, 5 µg and 6 µg respectively when one very high field blank was removed from the analysis. The amount of $PM_{2.5}$ collected on samples from the same batch of filters prepared with this one high field blank suggest that it was only the field blank that was contaminated and not all of the samples in that batch. Table 2-1 and Table 2-2 show statistical summaries for the pilot study blanks in concentration units obtained by dividing by the mean sample volume. Detection limits were also calculated for each component measured and are shown in Table 2-3. The calculations for limit of detection (LOD) are based on three times the standard deviation of the field blanks divided by the mean sample volume.

Table 2-1 Statistical summary of lab blanks, field blanks, and extended field blanks collected during the pilot study. For $PM_{2.5}$, values represent the mean difference between pre and post sampling filter weights (total average filter weight = 100.953mg). Sulphate and absorption coefficient (ABS) values are from post sampling analysis only. All values are converted to concentration units by dividing by the mean sample volume (5.748 m^3). Arithmetic means, standard deviation, minimum and maximum were calculated for each blank for total fine particulate ($PM_{2.5}$), sulphate (SO_4^{2-}) and absorption coefficient (ABS).

	$PM_{2.5}$ (µg/m^3)			SO_4^{2-} (µg/m^3)			ABS (m^{-1}x10^{-5})		
	Lab	Field	Ext. Field	Lab	Field	Ext. Field	Lab	Field	Ext. Field
N	12	13	4	12	13	4	12	13	4
mean	1	1	1	0.01	0.01	0.04	0.0	0.0	0.05
stdev	1	1	1	0.02	0.03	0.06	0.1	0.0	0.0
min	-1	-2	0	0.00	0.00	0.00	0.0	0.0	0.0
max	2	4	2	0.04	0.09	0.13	0.1	0.1	0.1

Table 2-2 Statistical summary of lab blanks, field blanks and extended field blanks collected during the pilot study for elemental carbon analyses. Arithmetic means, standard deviation, minimum and maximum were calculated for each blank for elemental carbon (EC) calculated using two different methods.

	EC (µg/m^3) NIOSH			EC (µg/m^3) DRI/IMPROVE		
	Lab	Field	Ext. Field	Lab	Field	Ext. Field
N	12	13	4	12	13	4
mean	0.00	0.00	0.00	0.17	0.18	0.28
stdev	0.00	0.02	0.00	0.31	0.21	0.24
min	0.00	-0.06	0.00	0.02	0.02	0.11
max	0.00	0.05	0.00	1.11	0.65	0.55

Table 2-3 Detection limits based on the field blanks collected during the pilot study. * One extreme blank was removed due to obvious contamination. LOD = 3*standard deviation of the field blanks/mean sample volume. For ABS LOD = 3* standard deviation.

	$PM_{2.5}$ (µg/m^3)	$PM_{2.5}$ * (µg/m^3)	SO_4^{2-} (µg/m^3)	ABS (m^{-1}x10^{-5})	EC NIOSH (µg/m^3)	EC DRI/IMPROVE (µg/m^3)
Limit of Detection (LOD)	4	3	0.08	0.1	0.07	0.64

Analysis of field and lab blanks provides information regarding possible error in the measurements and can be used to estimate accuracy and precision in the sampling method. It is important to note the negative mass values obtained for $PM_{2.5}$. There were 3 blank samples of the total 23 field and lab blanks collected during the pilot study that had a negative $PM_{2.5}$ mass. This was due to the weighing procedure and was a result of the post weights being less than the pre-weights. There are several factors that affect the resulting weight of a filter including changes in temperature, humidity, atmospheric pressure, static charge and vibrations (Jantunen *et* al, 2002). There are methods available to account for some of these effects but due to a relatively low effect this was not deemed necessary. The percent change in mass for the field blanks was very low (<0.01%) with all field blanks included and even lower for lab blanks that never left the weighing room and lab blanks that were taken to the Prince George laboratory for filter preparation. Standard weighing procedure was followed and an equilibrated weighing room was used. Although it is important to acknowledge this occurrence, it does not have a large impact on the study results considering the actual concentration levels observed during the study. When the actual precision of weighing the mass collected on a filter is a limiting factor, the measurement error will decrease with increasing ambient concentration (Lipfert & Wyzga, 1997). This error is accounted for by comparing field data to calculated limits of detection. None of the pilot study samples were below the detection limits for $PM_{2.5}$ mass, absorption coefficient or sulphate. For elemental carbon, 1 sample (2%) was below the detection limit for the NIOSH method and 2 samples (3.9%) were below for the DRI/IMPROVE method.

For comparison, Liu *et al* (2003) reported a detection limit of 4.5 $\mu g/m^3$ for total fine particulate using the $PM_{2.5}$ HPEM. Sarnat *et al* (2000) reported detection limits of

2.6 and 4.0 µg/m^3 for PM$_{2.5}$ and 2.6 µg/m^3 for SO$_4^2$ when using similar personal samplers. Janssen *et al* (2000) reported detection limits of 0.77 and 2.13 µg/m^3 for PM$_{2.5}$ and 0.08 and 0.15 m^{-1}x10^{-5} for absorption coefficients. Cyrys *et al* (2003) reported detection limits of 0.2 and 0.1 m^{-1}x10^{-5} for absorption coefficients and 0.25 µg/m^3 for elemental carbon. The detection limits shown in

Table 2-3 are consistent with or lower than those reported in similar studies.

It is also important to consider the precision or reproducibility of the measurements taken with regards to the sampling method used in the field and the analysis component undertaken in the lab. The co-located or duplicate samples collected provide information about precision as these samples were collected simultaneously and at the same location and underwent identical handling in the field and in the lab. During the pilot study 5 sets of co-located samples were successfully collected. Table 2-4 and Table 2-5 summarize the statistics on the difference between co-located samples. The true difference may be underestimated due to the low sample size with only five pairs of co-located measurements.

Table 2-4 Statistical summary for the difference between co-located samples collected during the pilot study. Mean difference, standard deviation, minimum and maximum were calculated for PM$_{2.5}$, sulphate and absorption coefficient measurements. Precision equals the standard deviation of the differences divided by the square root of 2.

N = 5	PM$_{2.5}$ (µg/m^3)	SO$_4$ (µg/m^3)	ABS (m^{-1}x10^{-5})
mean	1	.06	0
stdev	1	.06	0
min	0	.02	0
max	2	.16	.1
precision	0.7	0.04	0

Table 2-5 Statistical summary for difference between co-located samples. Mean difference, standard deviation, minimum and maximum were calculated for elemental carbon data provided by Environment Canada. Precision equals the standard deviation of the differences divided by the square root of 2.

N=4	EC (NIOSH) (μg/m^3)	EC (DRI/IMPROVE) (μg/m^3)
mean	.14	.53
stdev	.10	.39
min	.05	.31
max	.27	1.11
precision	0.07	0.27

A simple paired t-test for dependent samples was performed on the co-located data for each measured variable. These tests showed that the co-located samples were not statistically different from one another and provide assurance that the sampling and lab procedures were performed with adequate precision (p-values were all greater than 0.05). The measure of precision for $PM_{2.5}$, calculated by dividing the standard deviation of the duplicate differences by the square root of 2, was lower than the 2.2 μg/m^3 reported by Liu *et* al (2003) using the same HPEM samplers.

The flow measurements taken during each sampling period were crucial to determining the exact concentration of fine particles in the ambient air. The ambient sample pumps were very reliable and none of the samples had flows out of the acceptable range of 3.6 to 4.4 litres per minute (+/- 10%). Out of the 59 ambient samples collected during the pilot study only 7% had flows out of the target range of 3.8 to 4.2 litres per minute.

2.4.2. Instrument Comparison

The location of sampling during the pilot study was selected so that a comparison could be made between data collected by the current monitoring network and the data collected for this study using the $PM_{2.5}$ HPEM. Two different types of ambient samplers collect $PM_{2.5}$ samples at the Plaza site and are operated by the BC Ministry of Water, Land and Air Protection. The continuous $PM_{2.5}$ monitor is a TEOM, which provides hourly measurements. The non-continuous monitor is a Partisol sampler and collects a 24-hour integrated sample similar to that collected by the HPEMs. These samples are only collected every 6 days so there was not enough data available to enable a comparison of the Partisol to the HPEM. Figure 2-3 shows the comparison between the pilot study HPEM data and the Ministry TEOM. For the purposes of this comparison the hourly TEOM data was averaged over 24 hours using the corresponding time-frame in which the HPEM samples were collected.

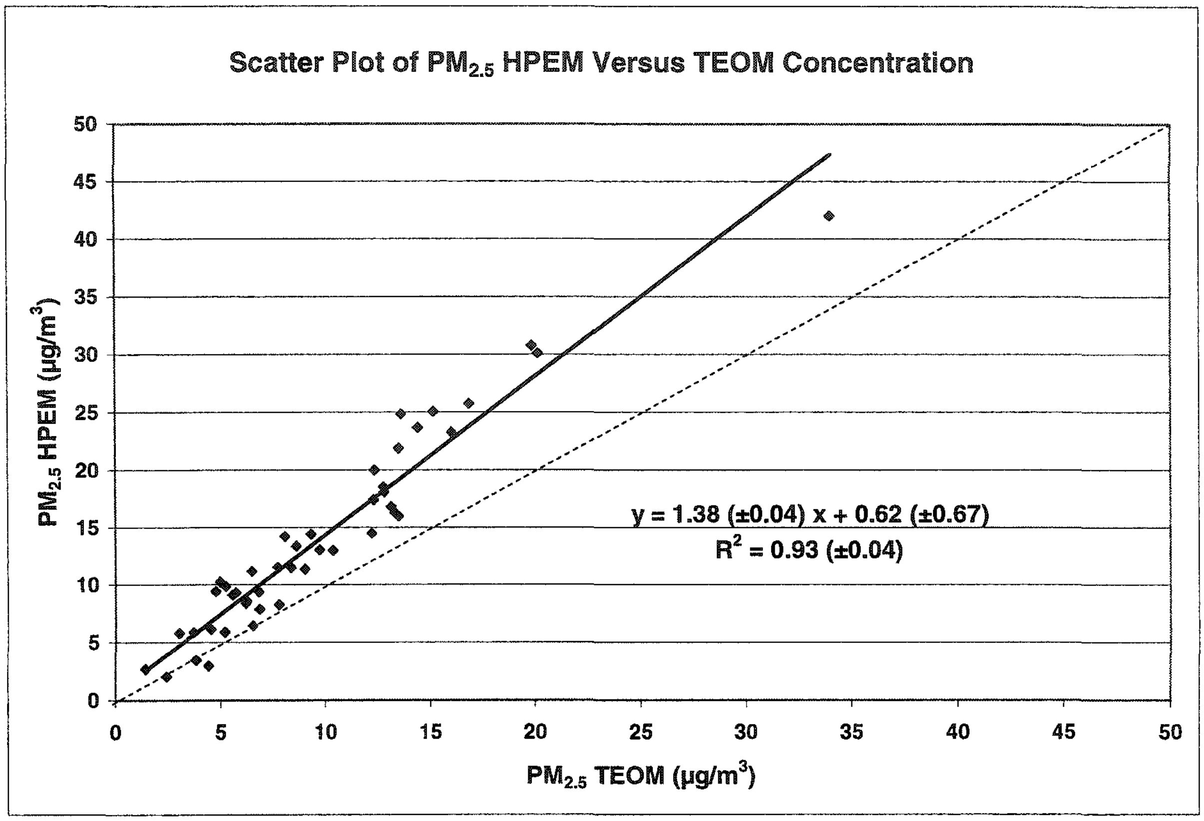


Figure 2-3 Comparison of $PM_{2.5}$ data collected during the pilot study using the HPEM and data from the Ministry operated TEOM. No scaling factor applied to TEOM data. (N=45)

The linear regression shows that the HPEM measurements were 38% more than those obtained with the TEOM. It is expected that the TEOM measurements would be less as it is operated with an internal temperature of 40°C and it is theorized that heating of the sample causes a loss of volatile particulate during the collection and measurement process (Chow, 1995). A TEOM monitor underreporting for the $PM_{2.5}$ size fraction has been reported by several others (Allen et al, 1997; Oh et al, 1997 and Williams et al, 2000). Williams (2000) compares a MSP PEM sampler (similar type of personal sampler to HPEM) to a TEOM and several other $PM_{2.5}$ samplers. This comparison showed that the MSP PEM consistently produced the highest mass concentration values and yielded a positive mass concentration bias of between 12 to 16% relative to the other monitors. Williams reported a mean difference of 2.1 μg/m^3 and standard deviation of 3.5 μg/m^3 between the MSP PEM and TEOM. The linear regression coefficient (R^2) was 0.91 with a slope of 1.03 and intercept of 1.4. The mean difference found during our pilot study for the Harvard PEM and TEOM was 4.3 μg/m^3 with a standard deviation of 3.2 μg/m^3 but it is difficult to compare these values due to the slope being much greater than 1. The high slope and low intercept suggest that with higher concentration there would be a greater difference between the concentrations reported by each instrument. The limited range of the majority of the data (0 to 20 μg/m^3) and the relative position of one higher value suggests that the slope of the regression line may be overestimated. Our regression results do show a strong significant correlation similar to the results reported by Williams with an R^2 of 0.93. Although there is a strong correlation between the instruments both the non-parametric Wilcoxon matched pairs test ($p<0.001$) and a simple paired t-test ($p<0.001$) showed a strong significant difference in the medians and means respectively.

2.4.3. Relationship between Absorption Coefficient and Elemental Carbon

The relationship between absorption coefficient and elemental carbon must be quantified to support the use of absorption coefficient as a surrogate measure for elemental carbon concentration. Figure 2-4 and Figure 2-5 show the results of this comparison using linear regression for both of the elemental carbon determination methods.

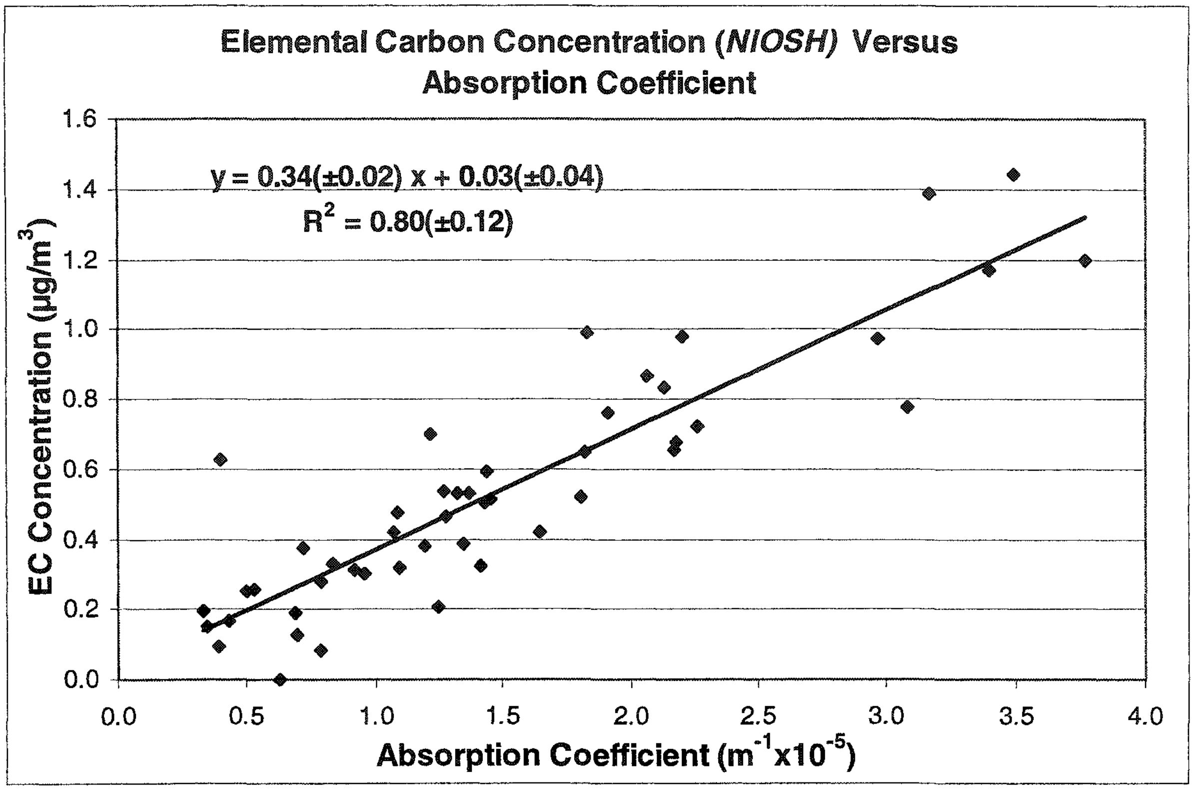


Figure 2-4 Comparison of actual elemental carbon data determined via thermal optical transmittance by Environment Canada and simple absorption coefficient measurements from Teflon filters. The elemental carbon concentrations were calculated using the NIOSH method with one extreme outlier removed (N=48).

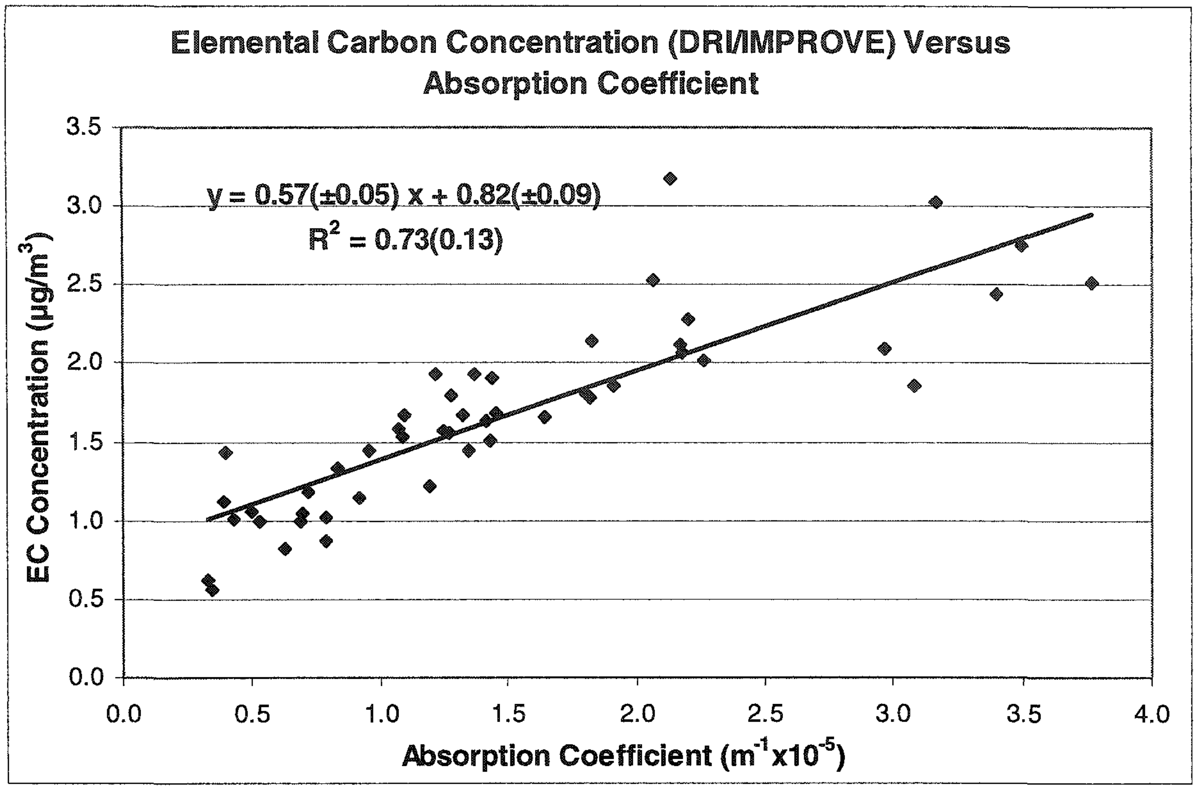


Figure 2-5 Comparison of actual elemental carbon data determined via thermal optical transmittance by Environment Canada and simple absorption coefficient measurements from Teflon filters. The elemental carbon concentrations were calculated using the DRI/IMPROVE method with one extreme outlier removed (N=48).

Elemental carbon concentrations were calculated using two different methods. For both methods one outlier was identified in the data as an erroneous result and was removed from the analysis. This value was more than 3 standard deviations from the mean for the DRI/IMPROVE data and more than 6 standard deviations from the mean for the NIOSH data. The results from both methods have been reported here but use of the NIOSH method will be used in subsequent analysis to describe the site specific relationship between absorption coefficient and elemental carbon levels. This method was chosen based on the blanks, detection limits and co-located samples. The NIOSH method resulted in blank levels closer to zero and a lower detection limit. The co-located samples also had better agreement for the NIOSH method. The linear regression results for both the NIOSH and DRI/IMPROVE elemental carbon levels to absorption coefficient showed strong and significant correlations. This supports the use of the reflectance method and absorption coefficient values as a surrogate measure for elemental carbon concentration. Pearson and Spearman correlations are reported in Table 2-6.

Table 2-6 Summary of correlations between absorption coefficient (ABS), elemental carbon (EC) and total $PM_{2.5}$ concentration ($p<0.000$ for all correlations). Note: One outlier was removed from the analysis.

	Valid N	Spearman r	Pearson R
ABS & EC NIOSH	48	0.88	0.90
ABS & EC DRI	48	0.92	0.85
ABS & $PM_{2.5}$	48	0.87	0.82
$PM_{2.5}$ & EC NIOSH	48	0.67	0.69
$PM_{2.5}$ & EC DRI	48	0.81	0.77
EC DRI & EC NIOSH	48	0.92	0.89

2.5. Conclusion

Results from the analysis of blanks, detection limits and co-located samples confirm acceptable data quality from the pilot study. The pilot study proved to be effective in training research staff regarding the lab and sampling procedures. It also provided an opportunity to test sampling methods in cold weather and make appropriate adaptations where necessary. Using a waterbed heater in a duffle bag proved effective in eliminating problems with the BIOS frictionless piston meter as long as tubing was used to take outside air into the flow meter. The rotameters were only useful as a back up measuring device and should not be relied upon when sampling at temperatures below -5°C. Loss of samples during winter sampling due to frost formation over the sample inlet can be easily prevented by wrapping the sampler with plumber's heat tape to keep the sampler temperature at 0° Celsius. In future studies, researchers must consider that the HPEM's and flow measuring devices were all intended for operation in warmer conditions. Adaptations must be made to accommodate operations in cold weather. Averaging hourly TEOM data over the 24-hour time-frame that HPEM samplers were collecting enabled a comparison of $PM_{2.5}$ concentrations from the two different instruments. As expected the HPEM concentrations were greater with a mean difference of 4.3 ± 3.2 $\mu g/m^3$. The medians and means of the TEOM and HPEM data were both significantly different but there was a high significant correlation with an R^2 of 0.93. This high correlation provides further validation of the data collected using HPEM samplers. Comparison of elemental carbon concentration and absorption coefficient levels showed that using the simple reflectance method was an adequate substitute for assessing elemental carbon levels when sampling can only be performed on one filter type. Both the NIOSH and DRI/IMPROVE elemental carbon concentrations demonstrated a strong and significant correlation with absorption coefficient with Pearson R values of 0.90 and 0.85 respectively. This supports the use of the reflectance method and absorption coefficient values as a surrogate measure for elemental carbon concentration in the Prince George Airshed and greatly reduces cost for supplies and analysis. The relationship described by the regression equation from the NIOSH elemental carbon data will be used to describe the winter-time relationship with absorption coefficient in Prince George (Figure 2.4).

3 Relationship Between Ambient Concentration and Personal Exposure of $PM_{2.5}$, Sulphate and Absorption Coefficient

3.1. Abstract

A field study was undertaken in Prince George, Canada during the winter of 2001. Personal exposure samples were collected from 15 children aged 10 to 12 years and ambient samples were collected at five outdoor monitoring sites located on the roofs of their respective elementary schools. This chapter compares the ambient concentration of $PM_{2.5}$, sulphate and absorption coefficient (an indicator of elemental carbon levels) with personal exposure measurements. The influence of local meteorology and spatial differences in the airshed were also assessed. Inversion conditions were found to be responsible for all high ambient concentrations (>30μg/m^3) and the influence of inversions could also be seen on personal exposures. Although spatial differences in ambient concentrations were found between schools for all three measures, high Spearman correlations (0.83 to 0.97) between the permanent central monitor and the five study sites suggest that the central monitor did adequately represent temporal changes in ambient $PM_{2.5}$ concentration. Comparison of personal exposures to ambient data for the pooled data showed stronger Spearman correlations for sulphate (0.96) and absorption coefficient (0.73) compared to total $PM_{2.5}$ (0.52) and comparable results were found from individual analyses. A large degree of individual variability in the personal-ambient correlation was found for $PM_{2.5}$, while sulphate showed very consistent results supporting its use as an indicator of exposure to sources of ambient origin. Absorption coefficient showed slightly more variability than sulphate due to the

influence of non-ambient sources in a relatively low number of samples. Overall, ambient $PM_{2.5}$ concentrations were high for the study period with levels at the school closest to the downtown core exceeding the Canada-Wide Standard for 2001. High personal $PM_{2.5}$ exposures (greater than 30 $\mu g/m^3$) were only associated with similar ambient levels 30% of the time suggesting that the majority of high total $PM_{2.5}$ personal exposures were due to the presence of non-ambient sources.

3.2. Introduction

There have been many studies performed regarding personal exposure to atmospheric particulate matter in the last decade. A review of personal exposure studies has shown that the relationship between ambient levels of particulate matter and individual exposure can be quite variable between communities (Mage and Buckley, 1995). The use of ambient monitoring data as a surrogate for personal exposure is appropriate for some individuals in some communities but not all. In many places, the ambient concentration does not have a significant influence on average personal exposure and personal activities and indoor sources have a much larger impact. Ozkaynak *et al.* (1996) found that concentrations outside of homes were well correlated with a central site but poorly correlated with both indoor concentrations and personal exposures. Indoor concentrations were found to be more important to personal exposure by Sexton *et al.* (1984) and Spengler *et al.* (1985). Several recent longitudinal PM exposure studies have shown stronger personal-outdoor PM correlations exist when data are analyzed by individual over time but the degree of association varies widely by individual (Ebelt *et* al, 2000;

Janssen *et al*, 1999, 2000; Liu *et* al, 2003; Rojas-Bracho *et al*, 2000; Sarnat *et al*, 2000; and Williams *et al*, 2003). Studies have also confirmed that the correlation between ambient and personal levels is good in the absence of indoor sources (Monn, 2001).

Personal exposure to air pollution can vary greatly between individuals and is impacted by both outdoor and indoor sources. The amount of time that an individual spends in different environments as well as their activity can affect their exposure. Two of the most important factors that may affect the personal-ambient relationship are the presence of indoor sources and variation or differences in ventilation rate. There has been an increasing awareness of the importance of indoor sources on personal exposure but these types of pollutants are difficult to control and reduction is generally the responsibility of the individual. Measures have been taken to improve indoor air quality in public facilities and workplaces but little can be done by regulators to improve air quality in the location where most individuals, especially children, spend the majority of their time - at home. Smoking and cooking have been identified as two of the largest indoor sources of fine particles and there is also a substantial amount of indoor particles generated by unknown sources (Wallace, 1996). Although these indoor sources may be important to the understanding of health impacts of fine particles in general, they are different from ambient particles and their health effects should be dealt with separately. When assessing the relationship between personal exposures and ambient concentrations one must acknowledge that the indoor sources have made a significant contribution to the total personal sample, which may obscure the relationship with ambient particles. Ideally, it is the relationship between personal exposure to particles originating outdoors and outdoor concentrations that is relevant to the results of health studies that have shown an association between ambient concentrations and various health endpoints (Ebelt *et* al, 2003; Mage *et* al, 1999; Wallace, 2000; and Wilson *et al*, 2000). The use of indicators of ambient exposure such as sulphate or elemental carbon may help to better characterize the true personal/ambient relationship. Research has shown that outdoor sources do contribute significantly to fine particle concentrations in the indoor environment. A comprehensive review of exposure literature confirms that outdoor air is the most important source of fine particles to indoor levels when smoking is not a factor (Monn, 2001).

The main factor that affects both the level of particles from indoor sources in a residence or building and the level of particles from ambient sources is ventilation. The impact of ambient particulate matter sources on personal exposures and the role of ventilation have been assessed in several longitudinal panel studies (Lui *et* al, 2003; Rojas-Bracho *et al*, 2000, 2004; Sarnat *et al,* 2000). Sarnat showed that on average 67% of total personal $PM_{2.5}$ exposures were due to ambient sources and this varied with ventilation status: well ventilated residences showed a stronger personal-ambient association compared to moderate and poorly ventilated residences. Rojas-Bracho *et al*, 2004 also found a stronger personal-ambient association with increased ventilation. Lui *et al* (2003) reported that 39% of the outdoor PM contributed to personal $PM_{2.5}$ exposure and this result was improved using a micro-environmental model that included three environments. Lui also found that the longitudinal correlation between personal exposure and ambient concentration was closely related to the particle infiltration efficiency of each residence, which is related to the air exchange rate or ventilation factor. Seasonal variation has also been identified in several of the longitudinal panel studies as impacting both the level of personal exposure and the personal-ambient relationship (Lui *et* al, 2003; Rojas-Bracho *et* al, 2004; and Sarnat *et al*, 2000). This finding may result from differences in ventilation as well as changes in ambient concentration and time-activity patterns (spending less time outdoors). Another study found difference in season to be unimportant (Williams *et al*, 2003).

It is clear from the literature that the relationship between personal exposure and ambient particulate level is dependent on location and individual or residential differences. This relationship should be further investigated in areas where health impacts are suspected in order to understand population exposure for that specific community and the variation due to individual differences.

A field study in Prince George, Canada was undertaken on weekdays during the winter-time from February 5th to March 16th, 2001. Episodes of high PM_{10} and $PM_{2.5}$ levels have been observed during similar periods in past years (Lamble *et al.,* 1998, 1999, 2000*)*. During this study five area monitors and five personal monitors were operated for six weeks to characterize spatial differences in ambient concentrations and investigate the relationship between ambient concentration and personal exposure. This chapter compares ambient data to personal exposure measures of total $PM_{2.5}$, as well as sulphate and absorption coefficient measured on the same $PM_{2.5}$ filters. Sulphate has been shown to be a reliable marker of personal exposure to ambient sources as it does not usually have significant indoor sources (Ebelt et al, 2000 and Sarnat et al, 2000). Absorption coefficient is used as a surrogate measure of elemental carbon and has been suggested as a reliable indicator of exposure to traffic related pollutants and perhaps PM of local origin such as residential wood-smoke (Fisher et al, 2000). Elemental carbon may also result from industrial combustion sources. The local meteorology during the study period is summarized and an assessment of the influence of local meteorological conditions on both ambient concentrations and personal exposures is provided. The spatial variation in both the ambient and personal data is discussed comparing concentration and exposures between the five neighbourhoods in the city. Descriptive statistics and an assessment of the association between personal exposures and ambient concentrations are provided for the pooled data set and for each school. Longitudinal personal to ambient associations for each subject are also investigated in order to assess the variability between individuals.

3.3. Study Design and Methods

3.3.1. Participant Selection

Children were the ideal candidates for this study because they attended a school located in the same area as their home and were generally less mobile than adults. Since one purpose of the study was to look at spatial variation throughout the city, it was best to have the subjects reside in one area the majority of the time that they were being monitored. There are several air pollution exposure studies that have monitored children in a similar manner including Janssen *et al* (1999), Wheeler *et al* (1999) and Geyh *et al* (2000). Five schools were selected to participate in the field study based on support of the school principal, location and access to the roof for an ambient monitoring station. An initial meeting was set up with principals from eight schools across the city. Four other school principals were not interested in learning more about the study and declined an initial meeting. All eight of the schools that were interviewed wanted to participate. The final five schools were selected mainly on the basis of location, to ensure adequate coverage of the study area. Two of the schools were chosen over another in the same area because they were the site of existing PM_{10} monitors in the BC MWLAP network. At each participating school, a meeting was held with a volunteer teacher recommended by the principal at each school. Full support of the teacher was required for participation. Three grade 5 classes, one grade 7 and one grade 6 class were selected for the study. A presentation was made to each class and students filled out a questionnaire asking how they felt about air quality in Prince George, whether they would like to participate in the study and about certain conditions required of study participants. Participants had to live within walking distance of the school and have no family members that smoked. Students were also asked why they would like to participate. A similar questionnaire and information packet was sent home for the parents of children that volunteered to participate. The teacher then considered the students that wanted to participate, had support of their parents and met the required qualifications and ranked them based on her assessment of the student's competency, ability and perceived commitment to complete the study. A meeting was set up with the top 3 candidates at each school to go over the detailed requirements with both the parent and student and to have the parent sign informed consent forms after the final decision was made. Only one of the 15 students that had this family interview was not selected for the study. The next candidate on that teachers list had the family interview, was accepted and agreed to participate in the study. All study participants were between 10 and 12 years old at the start of the study. Study protocol was approved by the University of Northern British Columbia Ethics Committee. All correspondence with the school board, school principals, teachers and families is provided in Appendix 2. Recruitment questionnaires and informed consent forms are in Appendix 3. A household characteristics questionnaire from the Harvard School of Public Health was also used to obtain information about each residence (Appendix 4).

3.3.2. Study Design

Five temporary ambient stations were set up to monitor $PM_{2.5}$ levels on the roof of each elementary school where personal exposure monitoring was being performed. Figure 3-1 shows the city of Prince George and the location of the school ambient monitoring stations and other air quality monitoring sites. All of the study subjects lived within 5 or 6 blocks from their school.

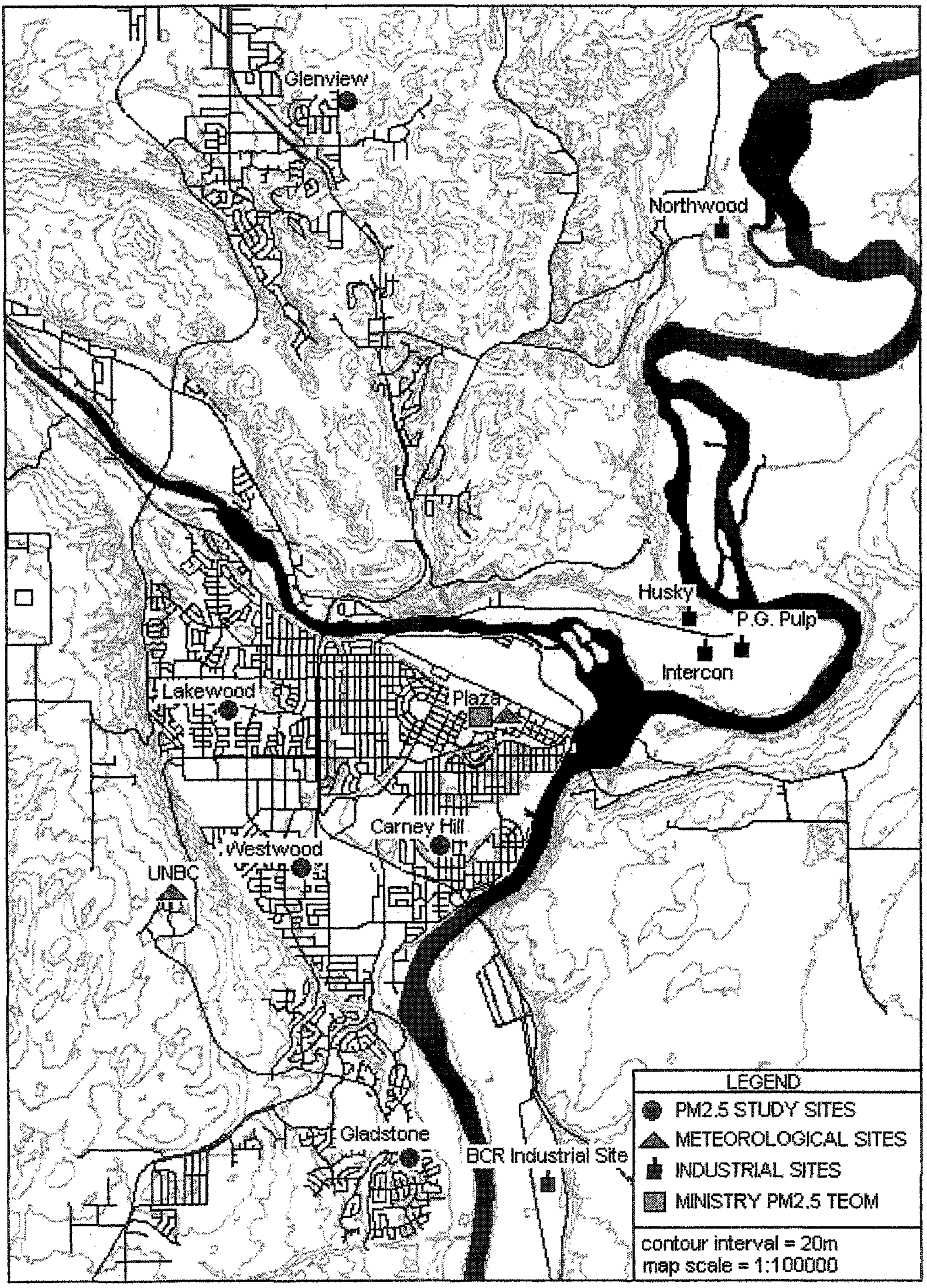


Figure 3-1 Map of the City of Prince George with each ambient monitoring site labeled and contour lines to depict the valley topography of the area.

The set-up and operation of the ambient monitoring sites was identical to that used during the pilot study (chapter 2). $PM_{2.5}$ Harvard personal environment monitors

(HPEMs) were used for both the ambient and personal sampling to collect a 24-hour integrated sample on Teflon filters, 37mm 2 µm pore size (Pall Gelman R2PJ037), suitable for determining total $PM_{2.5}$ concentration, absorption coefficient and sulphate level. Rotameters were used to take all flow measurements (Matheson, 603) and a BIOS frictionless piston meter (BIOS DryCal DC-1) was rotated between the two field workers visiting four of the schools. A flow measurement was taken every other day using this primary standard to confirm the measurements taken by the rotameter. Flow adjustments were rarely needed on the outdoor pumps. At the fifth school (Westwood) a Buck soap bubble instrument was used for all flow measurements (M-5 A.P. Buck Inc. Mini-Buck Calibrator). The Buck is also considered a primary standard. On every other day, a comparison was done between the Buck and the BIOS to confirm accuracy of the measurements at that school. A rotameter equilibrated to outdoor temperatures was used as a back up for the ambient flow measurements.

At the Westwood ambient site co-located samples were collected 10% of the time to ensure accuracy of the sampling method. Temperature and relative humidity data were also recorded daily at each ambient site using a HOBO Pro RH/Temp data logger (H08-032-08). Figure 3-2 shows the Westwood ambient monitoring site.

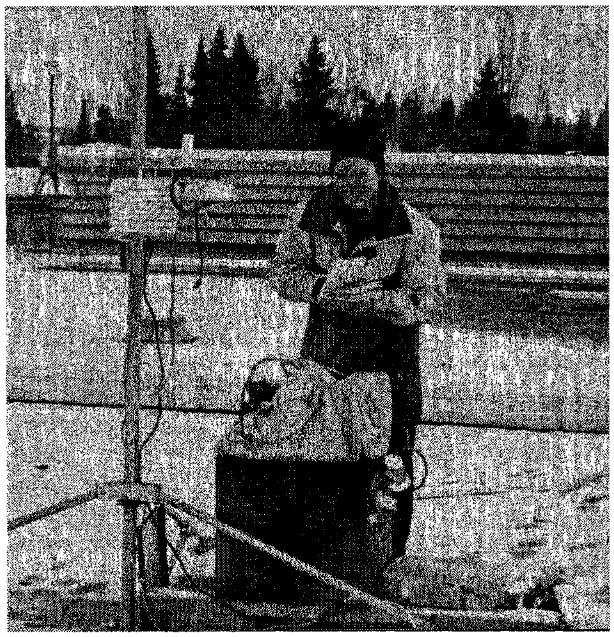


Figure 3-2 Westwood ambient monitoring site located on the roof of Westwood Elementary School. Research assistant Julianne Trelenberg records a flow measurement.

Both ambient and personal exposure samples were collected at each school, 5-days a week for the 6-week duration of the study. Sampling only on weekdays was necessary to reduce confounding that could result from changes in activity patterns on the weekend. No ambient samples were missed and only one personal sample was missed on the second day of the study due to a class ski trip at Glenview Elementary. 299 samples were collected in total, 150 ambient samples and 149 personal samples. The 24-hour personal filters were changed and the samplers restarted at the school immediately before or after the changeover of the ambient sample. All of the ambient and personal samplers were started and then finished the next day between 8:00 and 10:30 am. There was only one exception to this due to a power outage. The sampler was not removed until the power had come back on so that a flow measurement could be taken. This time frame is comparable to other studies. Ebelt *et al* (2000) and Sarnat *et* al (2000) took 3 and 4 hours respectively to change filters and other studies have taken as long as 9 hours (Janssen *et al,* 1998a).

The personal samplers were rotated between the 3 students at each school so that each student carried the monitor 10 times intermittently over the 6-week period. BGI air sampling pumps and battery packs (BGI-400S and BGI-401) were used to draw the sample through the filter for the personal monitoring. The target flow rate for all samplers was 3.8 to 4.2 litres per minute (LPM). Samples not within 10% of 4 LPM were not included in the final data set. The BGI pumps did not seem to maintain flow as well as the outdoor pumps so more adjustment was required on a daily basis. The personal sampling pumps were well insulated to limit the amount of noise emitted and compact to make them easier for the children to carry. The pumps were contained in a child-size backpack and surrounded by foam to further reduce noise from the pump. Latex tubing connected the pump to the sampler. The tubing came out of the pack and over the child's shoulder and then connected to the sampler. The sampler was attached to the strap of the backpack in the breathing zone of the child. Figure 3-3 and Figure 3-4 show the backpack and attached sampler and how the children wore them. The sampler inlet was facing downwards and was protected by a 4-inch piece of plastic tubing to prevent hair, clothing etc. from plugging the sample inlet or preventing air flow. Each morning the backpack was fitted to the appropriate child by research staff. The children were given instructions on carrying the pack and were informed about keeping the inlet clear and not letting the tubing become kinked. The participants were required to wear the pack as much as possible. They were allowed to hang the pack on the back of their chair when sitting at their desk. It was required that the pack be as close to them as possible if they could not wear it and at a minimum in the same room and as close to their breathing height as possible. This was generally only a concern if they were doing physical activity such as gym class.

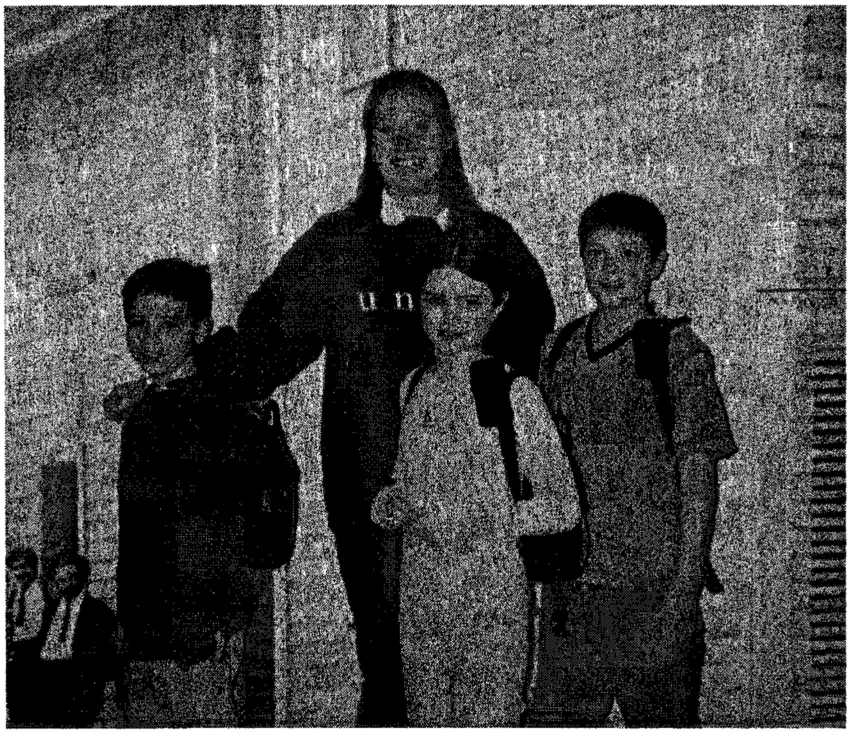


Figure 3-3 Study participants from Gladstone Elementary School wearing the sampler and backpack from the study.

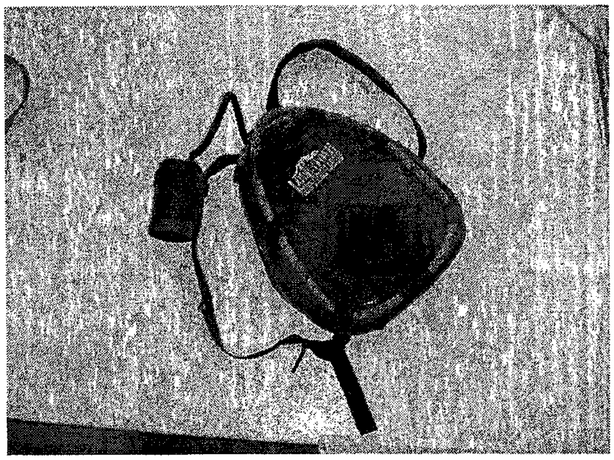


Figure 3-4 Backpack containing pump and battery with tubing coming out of pack and connected to sampler on the shoulder strap.

Each child was required to fill out a time activity diary every 30 minutes on the days that they carried the monitor. The diary asked if they were inside or outside; their location; the pack's location; if they were travelling by car, bus, bike or walking; if they were near a smoker; and what they were doing. A sample of the diary is shown in Appendix 1 along with the checklist given to research staff to ensure a standard review of the diary with each participant at the end of their sampling period. A Hobo motion sensor was also placed in each pack. Data from the sensor was downloaded each morning and then compared to each child's time activity diary as a quality assurance measure. Visual comparisons were made each day between the Hobo log and the time activity diary for every half hour entry and each subject to determine if the sampler was moving or stationary at the appropriate times. Any long periods where the back-pack was stationary were double checked. Any discrepancies were discussed with the appropriate study subject to ensure accuracy of the time activity diary. Initially, the children were not aware of the motion sensor but the data were downloaded from the sensor each day so they were told what it was if they asked.

Since the same staff member went to each school throughout the entire study, they became very familiar with the students and were able to develop a trustworthy relationship with each child. In addition to the morning flow checks and interviews with the students, the head researcher personally visited each participant in the evening at home every time that they carried the sampler. This enabled an additional flow check using the BIOS flow meter and an opportunity to answer any questions and concerns of the student or their parents. It also gave the head researcher an opportunity to review the diary and catch any potential problems. These visits led to a better relationship with the participants, assurance that they were following instructions properly and the ability to assess the home environment on each sampling day.

3.3.3. Lab Preparation and Analysis

Methods used and described here are identical to those used during the pilot study (Chapter 2). The PEMs were cleaned and re-greased daily and underwent a deep-cleaning every 5 sampling days. All cleaning and loading of samplers followed the standard operating procedure (SOP) provided by the Harvard School of Public Health and included in Appendix 5 (Ward-Brown, 2000).

Before and after sampling, Teflon filters were stored at room temperature in sealed, sterile Petri dishes. The Teflon filters were weighed prior to sampling and then again after sampling. Before any gravimetric measurements were performed the filters were equilibrated for 48 hours in a temperature (21.9 ± 0.2°C) and humidity (40±

2%) controlled weighing room at the School of Occupational and Environmental Hygiene of the University of British Columbia in Vancouver. A microbalance (Sartorious M3P; 1 μg resolution) was used to make triplicate measurements of filter weight and agreement was required to be within 5 μg. An external calibration was performed daily using 5, 10 and 20 mg NIST-traceable weights. Frequent internal calibration and triplicate weighing of a test blank filter every 25 filters ensured that accuracy of the instrument was maintained during weighing sessions. Post weighing was performed on the pilot study filters prior to the start of the main field study in order to identify any possible problems. During the field study, filters were stored and then taken to the UBC laboratory in Vancouver for analysis at the end of the study.

After gravimetric analysis and a quality assurance check of the data, Teflon filters underwent reflectometry at the UBC lab. The "blackness" of the $PM_{2.5}$ filter was measured using a reflectometer (M43D Smoke Stain Reflectometer, Diffusion Systems Ltd., London, UK), which measures the reflection of the light incidence in percent. The reflectance analysis followed the standard operating procedure from the ULTRA study to determine absorption coefficient using a reflectometric method and was obtained from researchers at the University of Wageningen in the Netherlands (ULTRA, 1998). This procedure has been used by other researchers (Cyrys *et al,* 2003; Fischer *et al*, 2000; Gotshci *et* al, 2002; Janssen *et al,* 2000; and Kingham *et al*, 2000). Briefly, blank filters were used to set reflectance at 100 percent and then the reflectance was measured on five different spots on each sampled filter. An absorption coefficient (ABS) was then calculated from the average reflectance for each filter using the following formula (International Standard ISO 9835):

(Equation 3.1) $\text{ABS (meters}^{-1}) = 0.5A \ln(R_F/R_S)/V$

where A is the area of the stain on the filter ($7.55\times10^{-4}\ m^2$); R_F is the average reflectance of the field blank filters in percent; R_S is the reflectance of the sample filter in percent; V is the sample volume in cubic meters (ISO, 1993). The absorption coefficient is multiplied by 10^{-5} for the purpose of reporting.

Sulphate analysis was also performed on the samples at the UBC lab after all gravimetric and reflectance measurements were completed. An extraction was done on the filters by wetting the filter with 100 µl of ethanol and sonicating in 5 ml of distilled/deionized water for 15 minutes in polyethylene containers (Ebelt *et al*, 2000 and Koutrakis *et al*, 1988). The extract was then analyzed using an ion chromatograph (Dionex, DX-300) with suppressed conductivity detection.

3.3.4. Data Quality Control/Assurance

Of all of the samples prepared, 10% were lab blanks (N=31) and 10% were field blanks (N=31). Both lab and field blanks were prepared in the lab along with the regular samples. They were loaded into samplers and tested for leakage by attaching to a pump and measuring any change in flow rate. The lab blanks were then stored in a Ziploc bag at room temperature until after sampling was completed. The field blank was taken out into the field with the regular samples, exposed to the air at both a personal and ambient location and then returned to the sealed Ziploc bag used for transport. Both were unloaded with the same batch of samples and the filters were stored for final analysis at room temperature. Co-located samples were also collected for 9% of the total samples to ensure that precision was maintained with the sampling procedures (N=27). Some of the co-located samples were collected with personal samples and some were collected with ambient samples. The co-located personal samples were collected by study staff and not the children in the study. It was not feasible to have the children carry two sampling back-packs at one time to collect duplicate samples.

3.3.5. Data Analysis

All statistics for this chapter were performed using Statistica 5.1 (StatSoft, Inc. 1997). Graphical output was generated using Microsoft Excel 2002.

The impact of wind speed and wind direction on hourly $PM_{2.5}$ concentrations during the study period was assessed using wind rose and pollution rose diagrams. These diagrams provide a graphical means of displaying the percentage of several wind speed and pollution concentration classes that are associated with general wind directions. The relationship between wind and concentration was characterized using $PM_{2.5}$ hourly concentrations from the Ministry of Water, Land and Air Protection $PM_{2.5}$ TEOM at Plaza. Comparison was not suitable for the 24-hour concentration data generated from the study. Rose diagrams for the entire study period were analyzed for all concentrations and for concentrations greater than both

15 and 30 $\mu g/m^3$. The role of wind speed and direction during episodes was also assessed using rose diagrams. The relationship between inversion conditions and hourly $PM_{2.5}$ concentration was investigated by calculating inversion strength from the temperatures recorded at two meteorological sites. Hourly inversion strength was calculated by subtracting hourly temperature data at a valley meteorological site (Plaza) from a higher elevation site (UNBC). Positive values were associated with inversion conditions and all negative values were set to zero. 24-hour inversion strength was calculated by summing the positive temperature differences for a given day and dividing by 24 to normalize, which enabled a comparison to the 24-hour study data. Time-series plots of concentration and inversion strength and Spearman correlations were used to assess the relationship between these variables.

The personal exposure data were summarized by individual subject, by pooling data for each school (or neighbourhood) and by pooling over all subjects. Personal and ambient measurements for $PM_{2.5}$, SO_4^{2-} and absorption coefficient were not normally distributed and were all positively skewed. Log transformation did not improve normality but both the geometric mean and geometric standard deviation for the pooled data are reported for descriptive purposes. The nonparametric Wilcoxon matched pairs test (analogous to a paired t-test) was used to compare medians of the pooled ambient and personal data. Differences in the personal-ambient relationship between schools were described using box plot diagrams showing the median and quartile ranges at each school. Statistical differences in median personal and ambient levels were assessed using the Wilcoxon matched pairs test.

Spatial variation in both ambient concentrations and personal exposures were assessed using straightforward analysis of variance (ANOVA) comparing both medians and means via the non-parametric Kruskal-Wallis ANOVA and a basic parametric ANOVA. These simple analyses were performed because it is often only the means or medians of a time period that are considered when assessing overall air quality in an airshed. The non-parametric Friedman 2-way ANOVA was also used, which provided a more powerful test of spatial differences that accounted for differences over time. For personal exposures a preliminary test for spatial differences was performed using a mixed model ANOVA that accounted for differences between individuals as a random factor in the model. Use of a mixed model ANOVA is the preferred method for analyzing longitudinal data like the personal exposures in this study although much more complex (Verbeke and Molenberghs, 2000).

Finally, both Spearman (r) and Pearson (R) correlations were calculated for the pooled data, the data at each school and for each individual. The regression slope and intercept were also provided for each individual. All analysis involving the personal $PM_{2.5}$ data was repeated with two extreme outliers removed for subject 1002. These two large samples were both collected when the subject's mother cooked the same dinner – fried chicken with curry. There was a noticeable amount of smoke in the residence when visited by research staff during the period when dinner was cooking on one of those occasions. Both samples had a distinct faint yellow colour not seen on any other samples that suggested the high particulate load could be due to cooking with curry, which is clearly an irregular indoor source. The appearance was not consistent with that seen on filters exposed to environmental tobacco smoke. Removal of these two points from the analysis is justified since the main study objective is to assess the impact of ambient sources on personal $PM_{2.5}$ exposure. But even if assessing the non-ambient component, the presence of a rare occurrence of high PM exposure from an unusual situation can seriously bias any regression analysis and does not represent the day-to-day variation of the usual indoor and personal sources of PM that the general public would encounter (Mage *et al*, 1999).

A final section of this chapter compares both ambient concentrations and personal exposures to the Canada-Wide Standard of 30 $\mu g/m^3$ and the Federal Reference Level of 15 $\mu g/m^3$. This comparison is provided only as a means of characterizing the overall air quality for the study period and it should be noted that these benchmarks established by Environment Canada are recommendations for ambient levels only.

3.4. Results and Discussion

3.4.1. Data Quality

The mean difference between pre and post sampling filter weights for the lab and field blanks for the $PM_{2.5}$ samples were 0 and 1 µg respectively with a standard deviation of 5 µg and an average filter weight of 96.738 mg. The average percent change in mass was 0.001%, a slight improvement over the pilot study results of

0.009%. For sulphate the mean mass on the lab and field blanks was 0.04 and 0.05 µg respectively with standard deviations of 0.10 and 0.12 µg. Average reflectance on the blanks was 99.9% and 100.3% for the lab and field blanks. Table 3-1 shows a summary of the descriptive statistics for the field blanks in concentration units calculated by dividing with the mean sample volume (5.748 m^3) for $PM_{2.5}$ and SO_4 and via Equation 1 discussed previously for absorption coefficient. Table 3-2 shows calculated detection limits.

Table 3-1 Summary of field blank levels in concentration units. Arithmetic means, standard deviation (stdev), minimum and maximum values were calculated for total particulate, sulphate and absorption coefficient.

N=31	$PM_{2.5}$ (µg/m^3)	SO_4^{2-} (µg/m^3)	ABS (m^{-1}x10^{-5})
mean	0	0.01	0.0
stdev	1	0.02	0.1
min	-2	0.00	-0.2
max	2	0.08	0.2

Table 3-2 Detection limits based on the field blanks collected during the field study. LOD = 3*standard deviation of the field blanks/mean sample volume. For ABS LOD = 3* standard deviation.

	$PM_{2.5}$ (µg/m^3)	SO_4^{2-} (µg/m^3)	ABS (m^{-1}x10^{-5})
Limit of Detection or LOD	3	0.06	0.2

When data from the actual samples were compared to the calculated limits of detection, we found 8% of the samples below the detection limits for absorption coefficient (no changes were made to the data) and no samples were below the detection limit for either total $PM_{2.5}$ or SO_4^{2-}. The detection limits were comparable or lower than other studies adding confidence to the reliability of the results (Cyrys *et* al, 2003; Janssen *et al*, 2000; Liu *et al*, 2003; Sarnat *et al*, 2000). Summary statistics for the difference in personal and ambient co-located samples are shown in Table 3-3 and Table 3-4 respectively.

Table 3-3 Personal co-located sample difference statistical summary. Arithmetic mean, standard deviation (stdev), minimum and maximum were calculated for total fine particulate, sulphate and absorption coefficient measurements. Precision equals the standard deviation of the differences divided by the square root of 2.

N = 12	$PM_{2.5}$ (µg/m^3)	SO_4^{2-} (µg/m^3)	ABS (m^{-1}x10^{-5})
mean	2	0.04	0.1
stdev	2	0.02	0.2
min	0	0.01	0.0
max	7	0.09	0.8
precision	1.41	0.02	0.16

Table 3-4 Ambient co-located sample difference statistical summary. Arithmetic mean, standard deviation (stdev), minimum and maximum were calculated for total fine particulate, sulphate and absorption coefficient measurements. Precision equals the standard deviation of the differences divided by the square root of 2. *3 outliers (20%) were removed from the analysis.

N = 15	$PM_{2.5}$ (µg/m^3)	SO_4^{2-} (µg/m^3)	*SO_4^{2-} (µg/m^3)	ABS (m^{-1}x10^{-5})
mean	2	0.49	0.08	0.1
stdev	3	1.01	0.11	0.1
min	0	0.01	0.01	0.0
max	14	2.94	0.38	0.6
precision	2.39	0.71	0.08	0.10

The high mean difference found between co-located ambient sulphate samples was likely due to 2 high differences of 2.94 and 2.93 (µg/m^3). When these values were removed from the analysis the mean difference was reduced significantly (0.11±0.17) and these extreme outliers were more than 25 standard deviations from the mean. Removing another high difference of 0.56 resulted in a mean difference (0.08±0.11) more comparable to that obtained during the pilot study (0.06±0.06). This third outlier was 4 standard deviations from the mean. There is no traceable explanation for these large differences, although the one very high difference for co-located $PM_{2.5}$ samples (14) was from the same sample as one of the extreme sulphate differences (2.93). Removal of the extreme $PM_{2.5}$ value did not change the mean difference but it did reduce the standard deviation (1µg/m^3) and subsequently improved the estimate of precision (0.79). A simple paired t-test for dependant samples was performed on the co-located data for each measured variable and showed no significant differences. All duplicates were also highly correlated with each other, with a Pearson's R of 0.99. An additional measure of precision was calculated by dividing the standard deviation of the duplicate differences by the square root of 2. The initial calculated precision for $PM_{2.5}$ from this study of 2.4 µg/m^3 is comparable to that obtained from another study using the same $PM_{2.5}$ HPEM samplers by Liu *et* al (2003) who reported a precision of 2.2 µg/m^3.

For the field study, flow measurements with the ambient sampling pumps were extremely reliable as was found in the pilot study. Only 1 of the measurements taken was outside of the acceptable flow range of 3.6 to 3.8 litres per minute and was consequently removed from the analysis. Six percent of the samples were outside of the target range of 3.8 to 4.2 litres per minute. Again the personal sampling pumps were not as reliable and 6 (4%) of the personal samples had flow measurements outside of the acceptable flow range and were not included in the final data set. For the personal samples, seventeen percent were outside of the target flow range of 3.8 to 4.2 litres per minute but were still within the acceptable range. In total, only one ambient sample and six personal samples were removed due to flow problems. An additional personal sample was removed due to a compliance issue. In total, 97% of the total samples collected were successful and were included in the analysis. An additional problem found with the personal sampling pumps was that cold weather caused the pump flow to increase. This increase was never detected to be outside of the acceptable range although it may have been possible. Only 2 of the study participants were outdoors long enough that a significant increase could have occurred (participant # 4002 and 5003). In order to prevent an increase in pump flow, hand warming packs were used to keep the pump warm. No samples were removed from the analysis due to the possibility of increased flow rates for short periods while outdoors.

3.4.2. Participant Compliance

All of the study participants completed all of the sampling days required of them. There were no dropouts and only one sample was lost due to a problem with compliance. For that one sample the backpack was left by the student in the computer room of the school for most of the day and was retrieved in the evening by study staff when the student realized at the evening check that they had forgotten about the backpack. There were two other times where a pack was forgotten at home while playing at a friend's place but only for a short period. Our evening checks provided further confidence that the children were following study procedures well. At the end of each sampling period a time activity log was reviewed with the student to ensure that what they had written was understood and to question any missing information. Detailed questionnaires were not used although a short list of questions was always reviewed and then missing details were added to the time activity diaries if necessary. Follow-up questionnaires with the parents would have provided useful information to help assess the impact of possible indoor sources at home, especially for cooking which was rarely performed by the children. Results from the daily qualitative checks of the Hobo motion sensor data compared to the time activity diaries provided further evidence that the children did an excellent job of wearing the sampling back-pack throughout each sampling period. There were only a few discrepancies that were checked with the student on the following day and resolved. The selection process that was followed when recruiting study participants was likely the reason that compliance was high.

3.4.3. Time Activity Data

On average the study participants spent 95% of their time indoors and 97% in their own neighbourhood and therefore relatively close to the ambient monitor located on their school roof. Figure 3-5 and Figure 3-6 summarize the information gathered from the time activity diaries for each of the 15 children that participated in the study. Filling in the time activity diary was the most difficult part of the study for the children and there was some missing information. 21% of the diaries had missing information mostly regarding particle generating activity and traveling between locations.

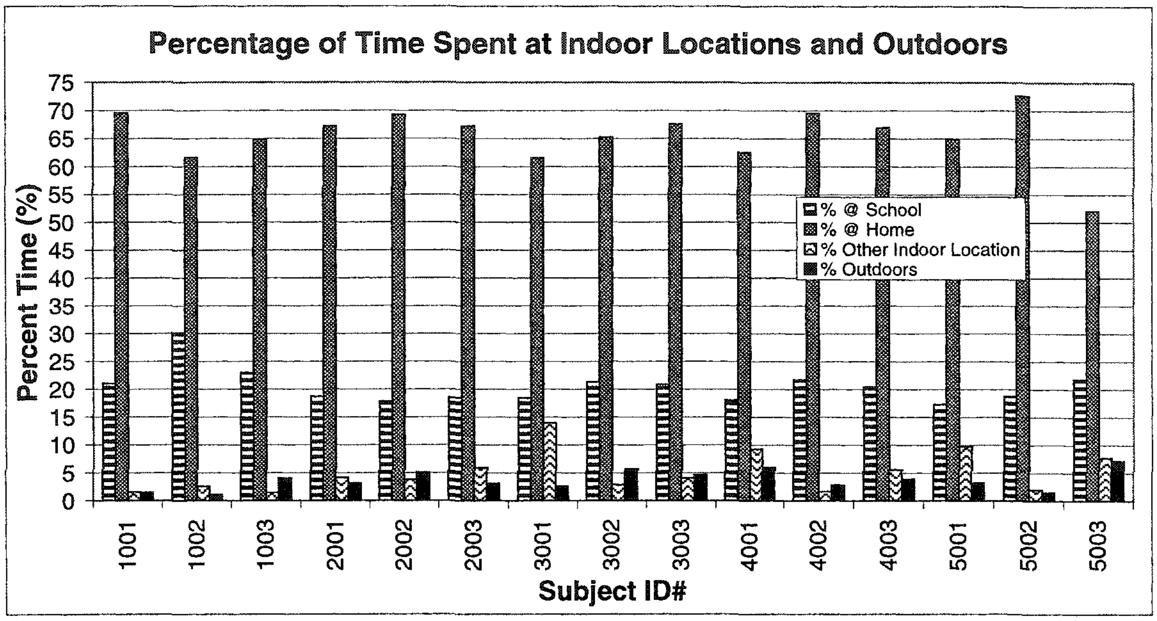


Figure 3-5 Summary of average time spent indoor at home, indoors at school, at other indoor locations and outdoors for each individual subject in the study.

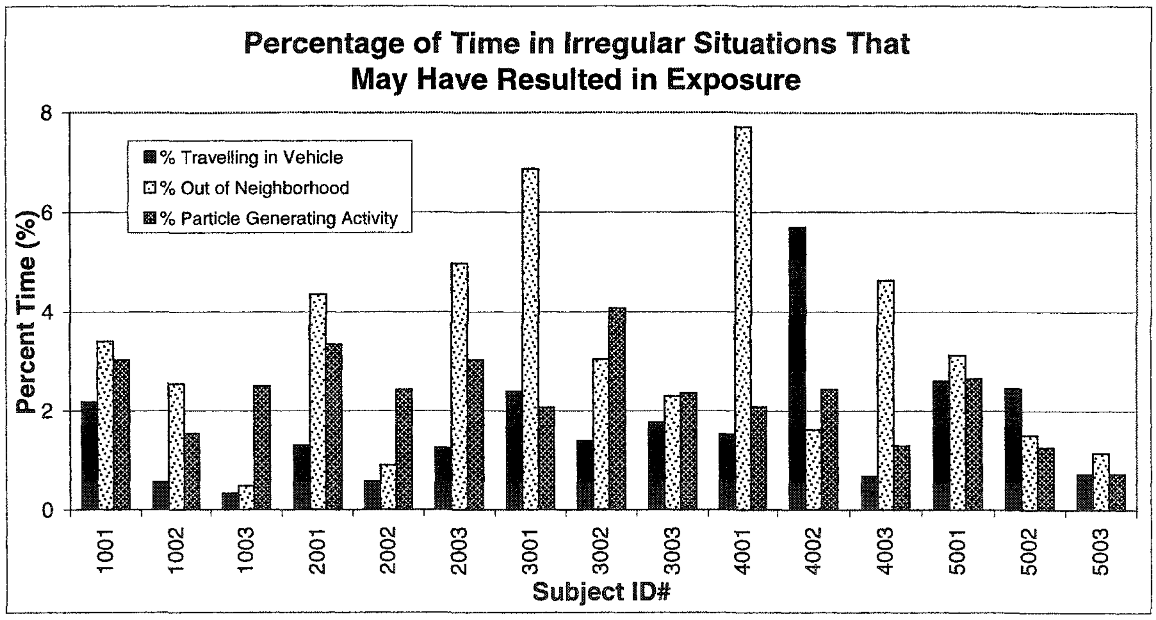


Figure 3-6 Summary of average time spent in irregular situations for each study participant. Due to the possibility of missing information this data can only provide an indication to possible causes of exposure.

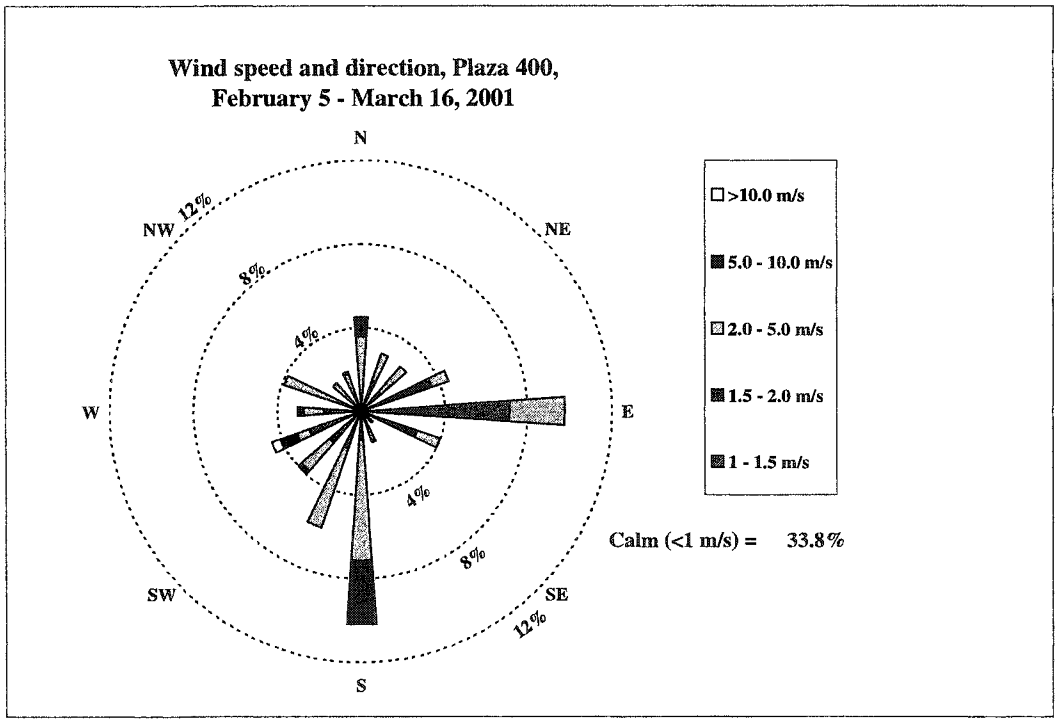
The children did an excellent job of filling out the diaries although more detailed information should have been obtained with the help of the parents regarding particle-generating activities such as cooking and cleaning in the house. There was some ambiguity with the time activity diaries regarding particle-generating activities because the children were often not aware when someone else was doing an activity that was relevant.

A possible source of error with the time activity diaries is that participants might actually change their activities on the days they are sampled. Because the primary researcher got to know each participant quite well and had input from both teachers and parents, it was possible to make a qualitative albeit subjective assessment of whether this might be a problem. The students were all willing participants in the study with relatively high self-esteem. They did not seem bothered by other students about carrying the monitor and seemed to carry on with their regular activities. They were all very excited to be participating in a scientific study and appeared eager to collect a sample that represented their true exposure. There was a study performed in the Netherlands that looked at the possibility of changes in a child's regular activity patterns due to sampling (Janssen *et al*, 1998b). These researchers had students fill out time activity diaries on days that they didn't carry the sampler in addition to the days when they were being monitored. No significant difference was found for the children although it was found that adults spent slightly more time at home and less time outdoors on days that they were being monitored.

3.4.4. Impact of Meteorology

3.4.4.1. Wind Speed and Direction

Wind direction and wind speed are important meteorological variables that could have influenced the level of outdoor concentrations and personal exposures during the study period. Wind and pollution roses for the study period using hourly Plaza data from the Ministry of Water, Land and Air Protection $PM_{2.5}$ TEOM and meteorological station are shown in Figure 3-7 and Figure 3-8 respectively.



Wind speed and direction, Plaza 400, February 5 - March 16, 2001

Figure 3-7 Wind rose showing percentage of each speed classes associated with winds from a given direction during the study period. Hourly wind direction and speed are from the Ministry of Water, Land and Air Protection Omineca-Peace Region Plaza monitoring site.

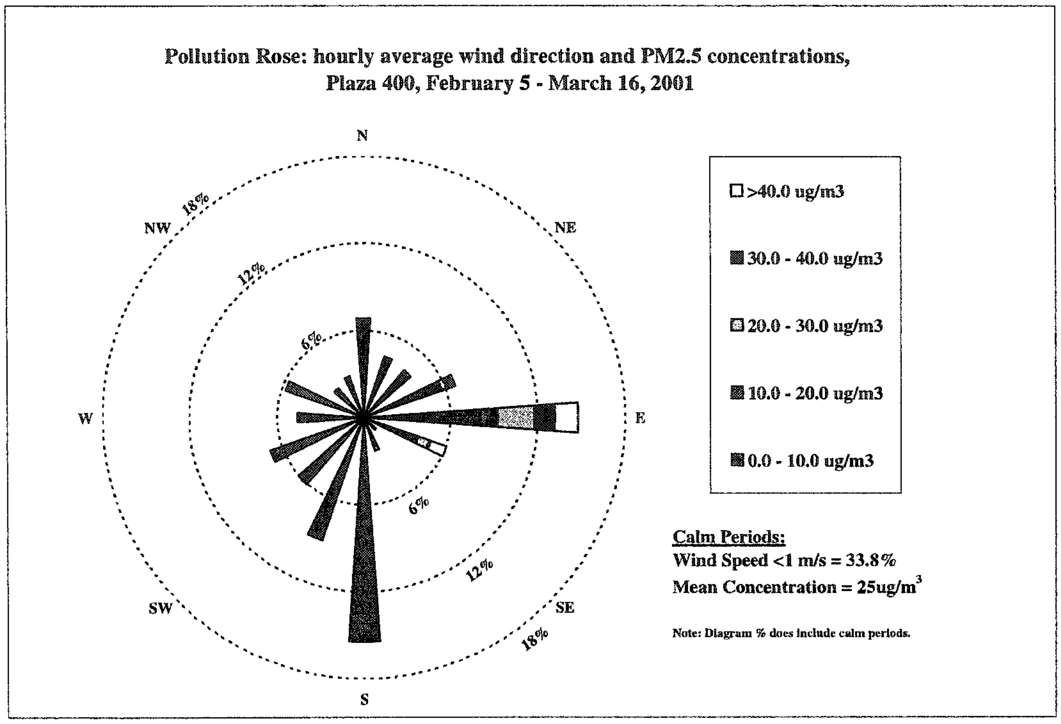
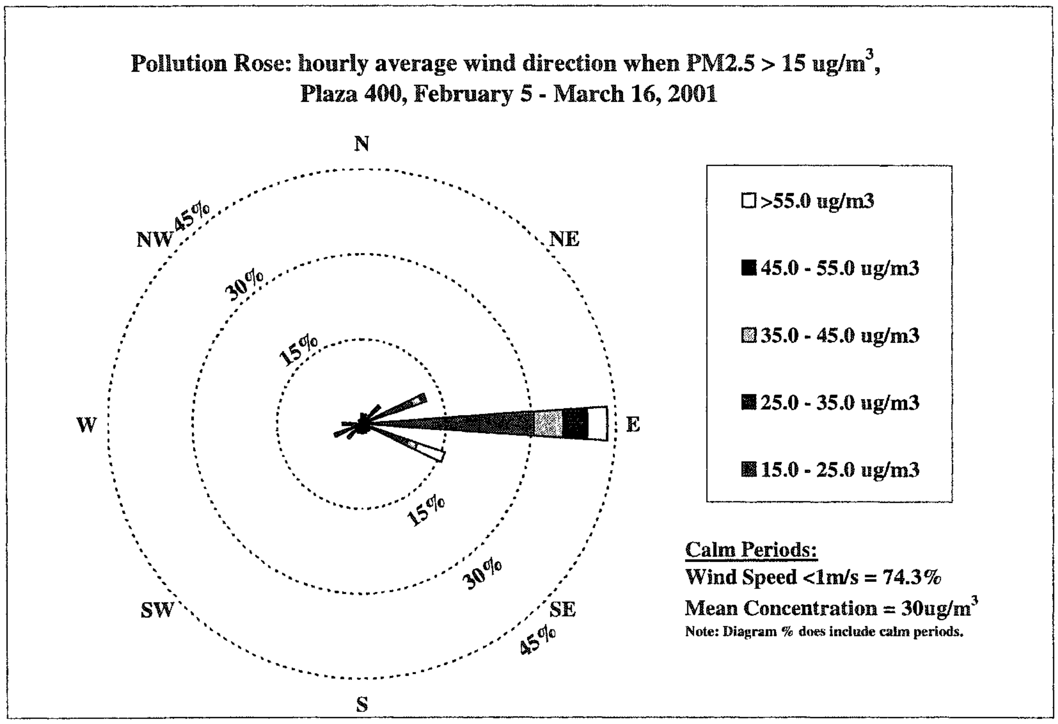


Figure 3-8 Pollution rose depicting the percentage of hourly $PM_{2.5}$ concentrations in each specified concentration range that came from a given direction. Hourly wind direction and $PM_{2.5}$ concentrations are from the Ministry of Water, Land and Air Protection Omineca-Peace Region Plaza monitoring site. Winds below 1 m/s were removed from analysis.

During the study period the highest percentage of winds came from the south and from the east. There was a low proportion of wind speeds over 5 m/s for the period (<10%) and higher winds generally came from the south and occasionally from the north and west. When higher pollution levels were observed the wind was generally coming from an easterly direction. Wind speeds from the east were always less than 5 m/s with a greater proportion of winds below 2 m/s. Winds from an easterly direction would be blowing from the main industrial sources (3 pulp mills and an oil refinery) in the city towards the TEOM monitor when steering from the valley topography is also considered. Analysis of data when the $PM_{2.5}$ concentration was greater than 15 $\mu g/m^3$ showed that approximately 60% of winds were from an easterly direction and 20% were from a south-westerly direction when all winds were included. When winds below 1 m/s were removed to account for the lower limit of sensitivity of the instrument, the remaining winds came for the east. These results can be seen in Figure 3-9 and suggests that generally, the air mass present on days with high concentrations were likely dominated by industrial emissions from the three pulp mills and oil refinery to the east.

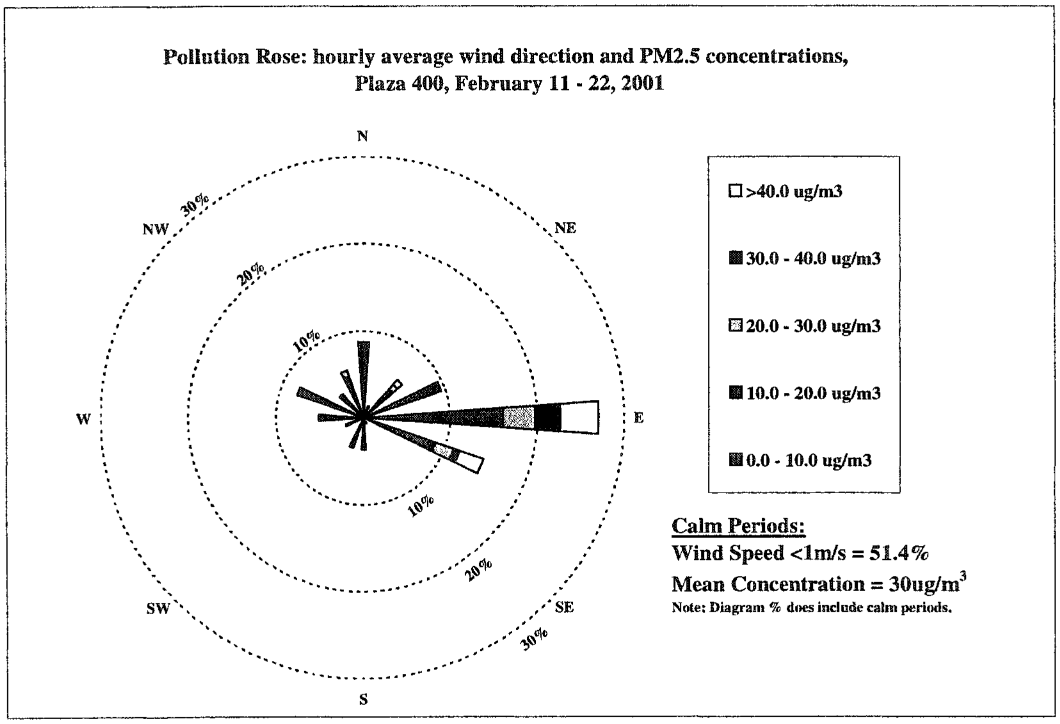


Pollution Rose: hourly average wind direction when PM2.5 > 15 ug/m^3, Plaza 400, February 5 - March 16, 2001

Calm Periods:
Wind Speed <1m/s = 74.3%
Mean Concentration = 30ug/m^3
Note: Diagram % does include calm periods.

Figure 3-9 Pollution rose for study period when concentration was greater than 15 µg/m^3. Diagram shows the percentage of hourly PM$_{2.5}$ concentrations in each specified concentration range that came from a given direction. Hourly wind direction and PM$_{2.5}$ concentrations are from the Ministry of Water, Land and Air Protection Omineca-Peace Region Plaza monitoring site. Winds below 1 m/s were removed from the analysis.

In this study, an episode was defined as any period where there were hourly concentrations greater than 30 µg/m^3 for more than 6 consecutive hours. Five episodes of high PM$_{2.5}$ concentration occurred throughout the study period each associated with high concentrations during part of the day for 2 or more consecutive days. When wind and pollution roses for each of the five episodes were analyzed, it was apparent that higher PM$_{2.5}$ levels measured at the Ministry TEOM were indeed associated with winds from the east and occasionally from other directions suggesting that the high levels were likely due to industrial sources. Figure 3-10 is a pollution rose for the longest episode during the study period.



Calm Periods:
Wind Speed <1m/s = 51.4%
Mean Concentration = 30ug/m^3
Note: Diagram % does include calm periods.

Figure 3-10 Pollution rose depicting the percentage of hourly $PM_{2.5}$ concentrations in each specified concentration range that came from a given direction during an eleven day episode during the study period . Hourly wind direction and $PM_{2.5}$ concentrations are from the Ministry of Water, Land and Air Protection Omineca-Peace Region Plaza monitoring site. Winds below 1 m/s were removed from the analysis.

Meteorological conditions play an important role in any spatial differences that might be present in an airshed. Lower concentrations at the Glenview outdoor monitoring site are expected not only due to distance from local industry but also due to the prevailing wind direction for the period. If winds are blowing from the east, pollution from the main industrial sources would not be blowing towards the Glenview monitoring site. At the beginning of March there was a short episode where $PM_{2.5}$ levels were uniquely higher at the Glenview site. This episode had a high percentage of winds from the south and southwest, which was blowing the main industrial source plumes towards Glenview and away from the other monitoring sites. Wind direction could have also resulted in the main highways having only a minor influence on both the Carney Hill and Glenview absorption coefficient levels. With a prevailing wind from the east (using Plaza as a reference point) the majority of the time during the study, both the Glenview and Carney Hill sites would be up-wind of the major highways.

3.4.4.2. Inversion Strength

Levels of $PM_{2.5}$ during the six-week period of this study were strongly influenced by inversion conditions. An inversion occurs when a layer of warm air resides over a layer of cold air. This causes colder, stable air to be trapped in the valley of Prince George with very little mixing or dispersion of pollution. During the study period, 62% of the days experienced inversion conditions for a portion of the day. Inversion strength, calculated as the temperature difference between UNBC and Plaza, was used to assess the relationship between ambient concentrations and inversions conditions, which typically started between the early to late evening hours and then dissipated by noon the following day. All of the days where hourly ambient $PM_{2.5}$ concentrations exceeded 30 $\mu g/m^3$ were associated with the existence of an inversion on that day. All five episodes of high $PM_{2.5}$ concentration that occurred during the study period were associated with inversion conditions. Figure 3-11 shows the hourly ambient $PM_{2.5}$ concentrations from the Ministry of Water, Land and

Air Protection $PM_{2.5}$ TEOM and inversion strength for the longest episode that occurred during the study period.

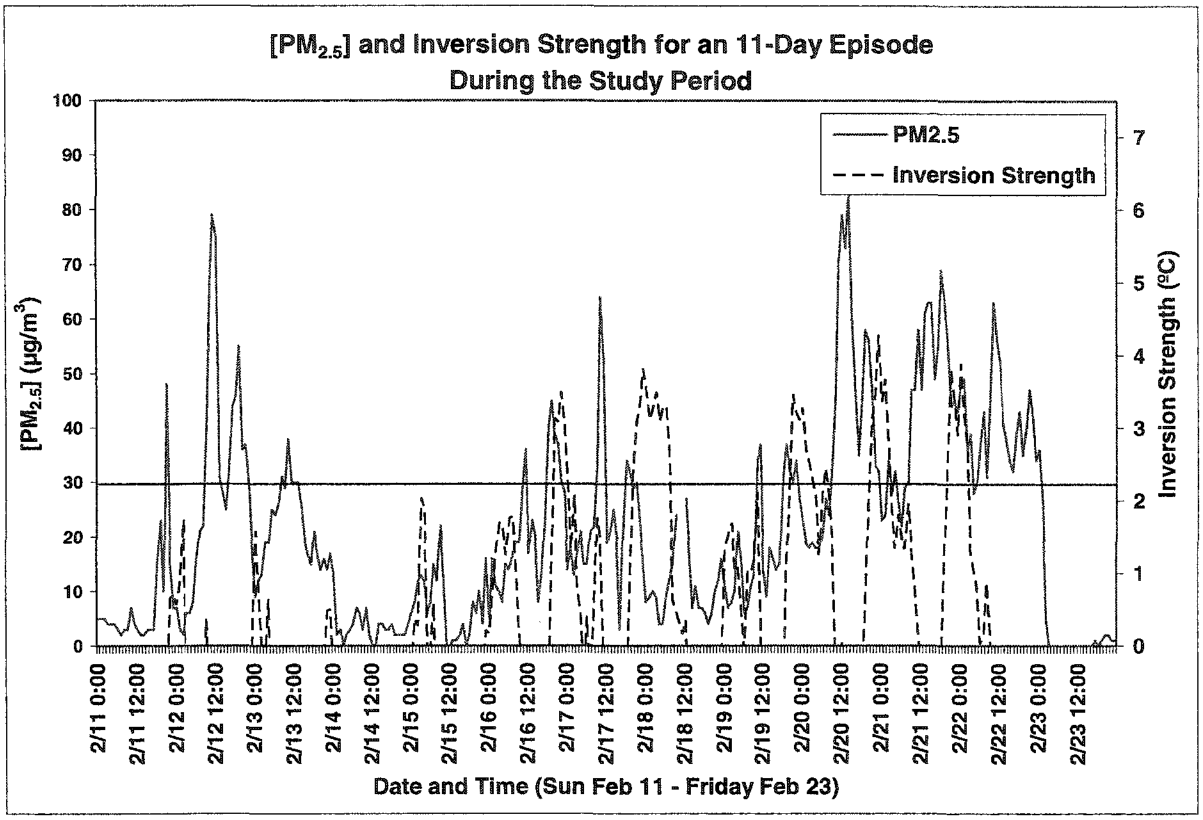


Figure 3-11 Episode during the study period that demonstrates the build-up of $PM_{2.5}$ when inversion conditions occur on consecutive days. *Peak concentrations often occur after the inversion dissipates.* Hourly $PM_{2.5}$ data is from the Ministry of Water, Land and Air Protection permanent TEOM monitor.

Concentrations greater than 30 µg/m^3 appeared to have a diurnal variation related to the existence of an inversion. During several of the episodes, peak $PM_{2.5}$ levels occurred in between daily inversions instead of during the actual inversion period. This is clearly illustrated on the last three days of the episode in Figure 3-11. Although concentrations from lower elevation sources are building up in the valley during an inversion, it is possible that the inversion may actually be protecting the valley from some higher point source emissions. When the inversion dissipates, fumigation of the high point source plume may cause already high pollution levels to increase significantly. The fumigation of point source plumes after an inversion is being investigated by local airshed management (Sutherland and Fudge, 2002). This increase between inversions clearly occurred after 31% of the inversions during the study. These short term high peaks of ambient $PM_{2.5}$ concentration could influence personal exposure. 24 hour exposures may only be changed by 2 to 4 $\mu g/m^3$, depending on the amount of time spent outdoors during this period, but this short term exposure could increase the risk of health effects especially for a susceptible individual such as a child with asthma. For this study group these peaks in high $PM_{2.5}$ concentration often occurred during lunch hour when the students were out in the playground.

The 24-hour data available from each of the outdoor school monitors did not provide similar resolution to enable comparison of concentration and exposure data to the diurnal variation in inversion strength, but an index of 24-hour inversion strength was calculated by adding all of the positive inversion values for a given day together and then dividing by 24 to normalize the value. Time series graphs showing 24-hour inversion strength, $PM_{2.5}$, sulphate and absorption coefficient at each school are shown in Figure 3-12, Figure 3-13 and Figure 3-14 respectively.

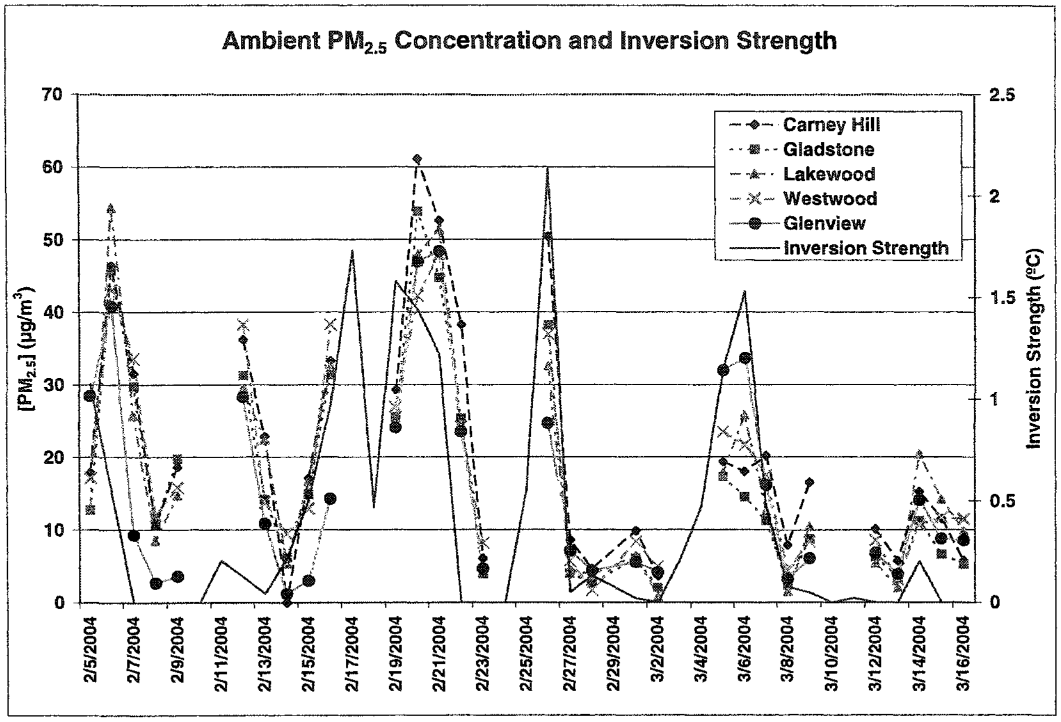


Figure 3-12 Time series of 24-hour inversion strength and ambient $PM_{2.5}$ concentrations at the five school rooftop monitors during the study period. Breaks in the concentration data are due to weekends where no sampling occurred.

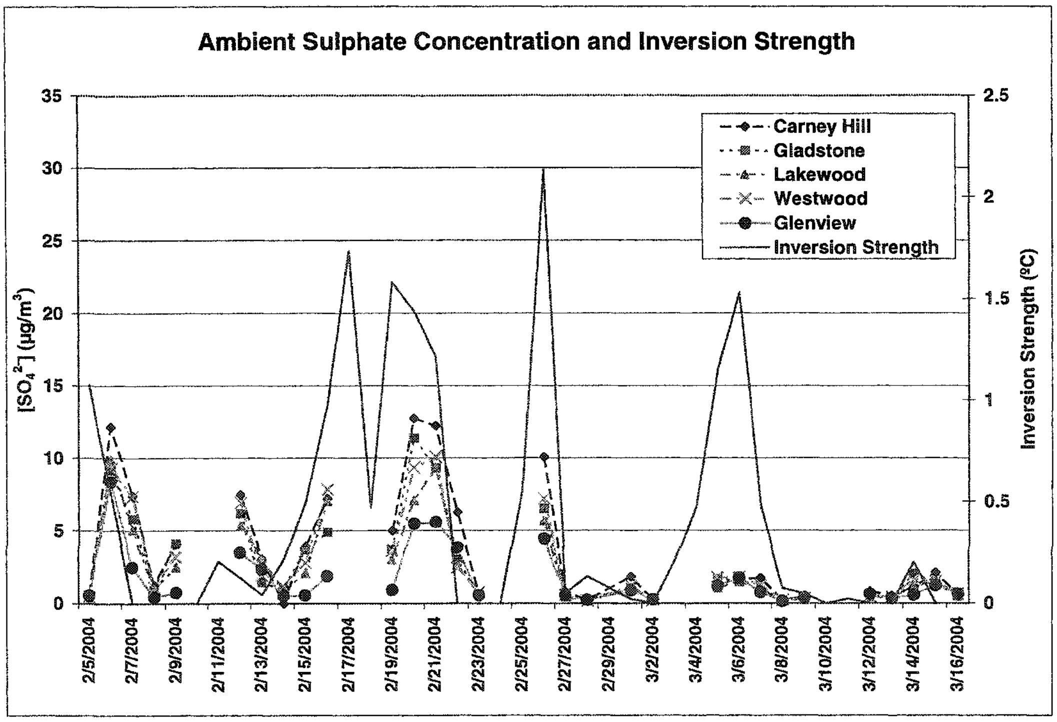


Figure 3-13 Time series of 24-hour inversion strength and ambient sulphate concentrations at the five school rooftop monitors during the study period. Breaks in the concentration data are due to weekends where no sampling occurred.

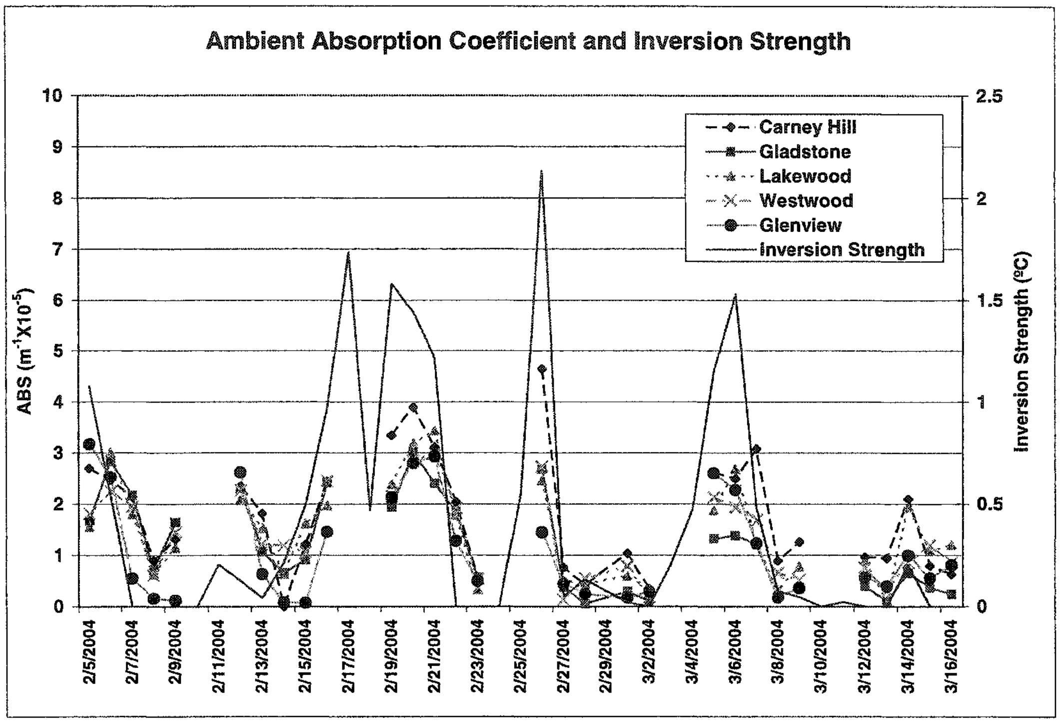


Figure 3-14 Time series of 24-hour inversion strength and ambient absorption coefficient at the five school rooftop monitors during the study period. Breaks in the concentration data are due to weekends where no sampling occurred.

From the time-series above, inversion strength clearly shows the five episodes during the study period that were discussed previously and were identified from hourly $PM_{2.5}$ data collected at Plaza by the Ministry. It is also clear that inversions are the cause of increased concentration for all three measures at all five of the schools including Glenview and Gladstone, which are located at a higher elevation outside of the city “bowl”. Both the sulphate and “elemental carbon” components of $PM_{2.5}$ were affected during each episode except for the one in early March where sulphate was not elevated. This episode also shows higher $PM_{2.5}$ concentrations at Glenview compared to the other schools, when usually Glenview has the lowest $PM_{2.5}$ levels. Analysis of wind and pollution roses for each episode show that the early March episode had the highest percentage of winds from the south and southwest that were also associated with higher pollution levels compared to the other episodes. This suggests that the air mass during this episode was not dominated by industrial emissions from the east that contain precursors of sulphate, which appeared to be the case for the other episodes.

Analysis of the sulphate and elemental carbon content of the total $PM_{2.5}$ also suggested that industrial sources made a significant contribution to total $PM_{2.5}$ levels during periods of high concentrations. During the study period the mean sulphate content of the total ambient $PM_{2.5}$ samples was 13.0±6.9% while the estimate of elemental carbon content (based upon absorption coefficient measurements) was approximately 3.3±1.9%. An analysis of higher pollution days showed that as total $PM_{2.5}$ concentration increased, the mean and median concentrations of sulphate increased by an average amount of 1.0 $\mu g/m^3$ for every 5 $\mu g/m^3$ increase in ambient $PM_{2.5}$ concentration while the elemental carbon concentration only changed slightly by an average of 0.06 $\mu g/m^3$ for every 5 $\mu g/m^3$. On pollution days where ambient concentration exceeded 15 $\mu g/m^3$ and 30 $\mu g/m^3$, the median sulphate content increased to 16.1% and 19.5% and the elemental carbon content decreased to 2.6% and 2.3% respectively. These findings suggest that in general sources of sulphate made a greater contribution to higher pollution levels than sources of elemental carbon such as wood burning or traffic. For the personal samples the mean sulphate and elemental carbon contents were 7.4±6.5% and 2.1±1.9% respectively.

Time series were analyzed to see if a similar pattern existed between 24-hour inversion strength and personal exposures and are shown in Figure 3-15, Figure 3-16and Figure 3-17.

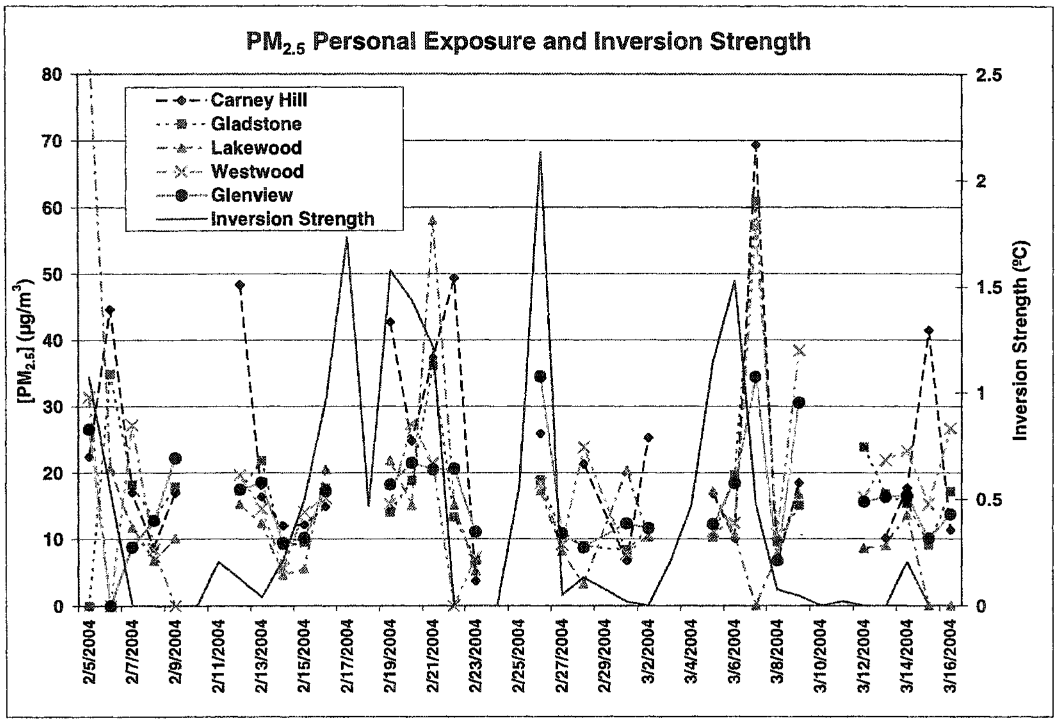


Figure 3-15 Time series of 24-hour inversion strength and $PM_{2.5}$ personal exposure with the data pooled across the 3 individuals at each of the five school. Breaks in the concentration data are due to weekends where no sampling occurred. Two extreme points at Carney Hill are out of the range of the graph.

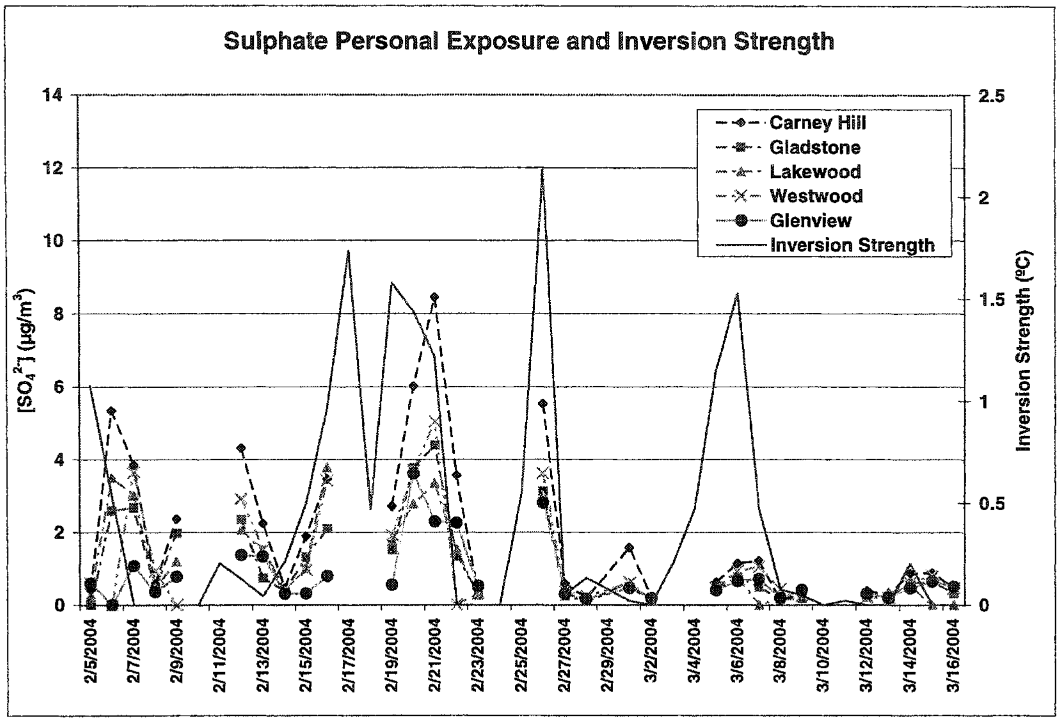


Figure 3-16 Time series of 24-hour inversion strength and sulphate personal exposure with the data pooled across the 3 individuals at each of the five school. Breaks in the concentration data are due to weekends where no sampling occurred.

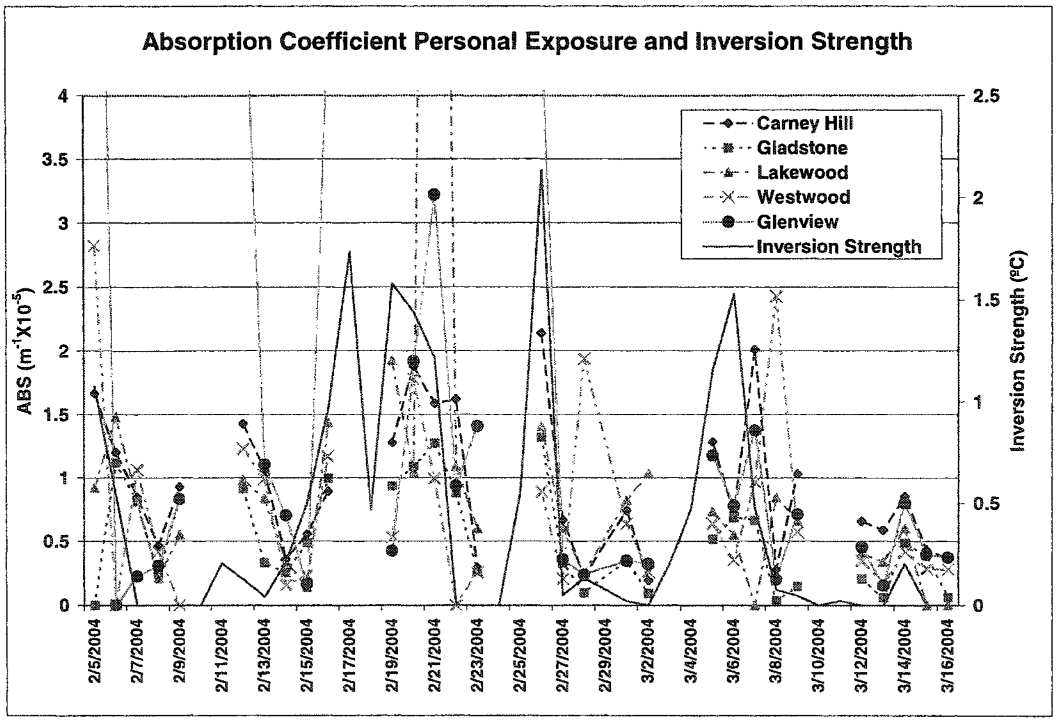


Figure 3-17 Time series of 24-hour inversion strength and absorption coefficient personal exposure with the data pooled across the 3 individuals at each of the five school. Breaks in the concentration data are due to weekends where no sampling occurred. Four extreme points from Glenview students and one from Lakewood are out of the range of the graph.

Personal sulphate exposures show an identical relationship with inversion strength as the ambient sulphate concentrations, although the level of sulphate is proportionately lower. For both $PM_{2.5}$ and absorption coefficient the pattern is still present but it is not as clear from the time series graphs due to the presence of high exposures from non-ambient sources. The correlations in Table 3-5 describe the statistical association between inversion strength and both ambient concentrations and personal exposures.

Table 3-5 Spearman correlations describing association between inversion strength and both ambient concentrations and personal exposures for $PM_{2.5}$, sulphate (SO_4^{2-}) and absorption coefficient (ABS). Correlations are shown for the pooled data and at each school. CH=Carney Hill; GS=Gladstone; LW=Lakewood; WW=Westwood; and GV=Glenview. [a] Two extreme outliers were removed from the data. *Significant at $\alpha = 0.05$ level.

		ALL	CH	GS	LW	WW	GV
Ambient Concentration and Inversion Strength	*N=*	*149*	*29*	*30*	*30*	*30*	*30*
	$PM_{2.5}$	0.67*	0.68*	0.63*	0.65*	0.65*	0.76*
	SO_4^{2-}	0.55*	0.57*	0.54*	0.55*	0.58*	0.60*
	ABS	0.65*	0.73*	0.65*	0.71*	0.61*	0.65*
		ALL	**CH**	**GS**	**LW**	**WW**	**GV**
Personal Exposure and Inversion Strength	*N=*	*141*	*29*	*29*	*27*	*27*	*29*
	$PM_{2.5}$	0.40* (0.41*)[a]	0.41* (0.48*)[a]	0.44*	0.62*	0.21	0.42*
	SO_4^{2-}	0.58*	0.63*	0.59*	0.60*	0.64*	0.51*
	ABS	0.53*	0.66*	0.71*	0.63*	0.32	0.43*

For ambient concentrations, there were moderate correlations with inversion strength across all schools. Interestingly, Glenview showed the highest correlations for both $PM_{2.5}$ and sulphate, which suggests that inversions may be the main reason for high concentrations at that school. At the other four schools there may have been slightly more instances where high concentrations existed without an inversion. The correlations with personal exposure were slightly lower for both $PM_{2.5}$ and absorption coefficient, although still mostly significant, which was likely due to the influence of non-ambient sources. For sulphate the correlations with inversion strength were similar for both ambient concentrations and personal exposure. At Westwood school there was not a statistically significant correlation between inversion strength and personal exposure to both $PM_{2.5}$ and absorption coefficient. It is not clear why there is a difference at this school only but suggests that students at this school may have had a lower proportion of their exposure from ambient sources; therefore, their personal exposures were not influenced as much by a meteorological factor such as inversion strength.

3.4.5. General Analysis of the Pooled Data

The pooled $PM_{2.5}$ ambient and personal data had a mean difference of 3 μg/m^3 with the personal exposures being higher. The personal exposures had a much larger range and a significantly greater standard deviation. When two extreme outliers were removed from the data the means became the same and the standard deviations were within 2 μg/m^3. Removal of the outliers had no effect on the median, geometric mean or geometric standard deviation. Table 3-6 summarizes the data for both ambient and personal total $PM_{2.5}$ and the sulphate (SO_4^{2-}) and elemental carbon components. Elemental carbon level is represented by the surrogate measure absorption coefficient (ABS). Personal and ambient absorption coefficient had a mean difference of 0.4 ($m^{-1}x10^{-5}$). Overall, ambient absorption coefficient was usually higher than personal absorption coefficient with 23% of valid personal samples being greater than the ambient level. Half of these samples were from the students at Glenview school suggesting that there may be an indoor source at the school or in the houses of these students or some sort of local ambient source that was not impacting the ambient monitor on the school roof such as wood-smoke. The mean difference between ambient and personal sulphate was 1.39 $\mu g/m^3$ with ambient sulphate always greater than personal sulphate. Analysis of the time series showed a very strong visual correlation between the personal and ambient sulphate concentrations that was apparent across all schools and subjects.

Table 3-6 Summary statistics for $PM_{2.5}$ and SO_4^{2-} concentrations ($\mu g/m^3$) and absorption coefficient level (10^{-5} m^{-1}). Ambient and personal refer respectively to all outdoor and personal samples collected during the study period. Mean = arithmetic mean; Stdev = standard deviation; GM = geometric mean; GSD = geometric standard deviation. [a] Two extreme outliers were removed from the data.

Measurement	N	Mean	Stdev	Median	Range	GM	GSD
Ambient $PM_{2.5}$	149	18	15	14	1 to 61	13	3
Ambient SO_4^{2-}	149	2.72	3.11	1.23	0.13 to 12.76	1.39	3.39
Ambient ABS	149	1.4	1.0	1.2	0.1 to 4.7	1.0	2.6
Personal $PM_{2.5}$	142 (140)[a]	21 (18)[a]	22 (13)[a]	16 (16)[a]	3 to 179 (3 to 87)[a]	16 (16)[a]	2 (2)[a]
Personal SO_4^{2-}	142	1.33	1.47	0.61	0.11 to 8.44	0.76	2.92
Personal ABS	142	1.0	1.7	0.7	0.0 to 15.3	0.6	2.6

The nonparametric Wilcoxon matched pairs test was performed on the pooled data to see if a difference in medians was significant for the ambient and personal measures. The medians for personal $PM_{2.5}$, SO_4^{2-} and absorption coefficient were compared to the medians of their ambient counterpart. The test results showed that for SO_4^{2-} and absorption coefficient the comparisons resulted in a significant p-value ($p=0.000$) but for the personal to ambient $PM_{2.5}$ comparison the p-value was not significant ($p=0.582$). This shows that there was not enough evidence in the data to suggest that the total $PM_{2.5}$ personal and ambient medians were significantly different from each other. This finding actually supports the use of ambient $PM_{2.5}$ data as a surrogate measure for average personal $PM_{2.5}$ exposure although the degree of correlation between these measures must also be considered and the similarity in values is likely by chance, considering that the subjects spent very little time outdoors or with windows open, and due to the presence of non-ambient sources. The fact that the SO_4^{2-} and absorption coefficient data had personal and ambient medians that were significantly different does not lead to the opposite conclusion that they are not suitable surrogates for total personal exposure. The difference in medians is likely due to the time individuals spent indoors and the low infiltration of these pollutants from ambient sources. The degree of correlation between the personal and ambient measures is a more accurate gauge of a suitable surrogate measure.

3.4.6. Analysis of Spatial Variation

3.4.6.1. Distribution of the Personal and Ambient Data at Each School

The summary statistics in Table 3-6 pool all of the ambient and personal observations together. Because there were five ambient stations collecting data each day and personal exposure data was also collected simultaneously from five individuals, analysis was performed with the data pooled at each of the five schools in the study facilitating an assessment of spatial variation throughout the city. Figure 3-18 shows the distribution of the personal and ambient $PM_{2.5}$ levels at each school enabling a visual assessment of spatial variation between neighbourhoods.

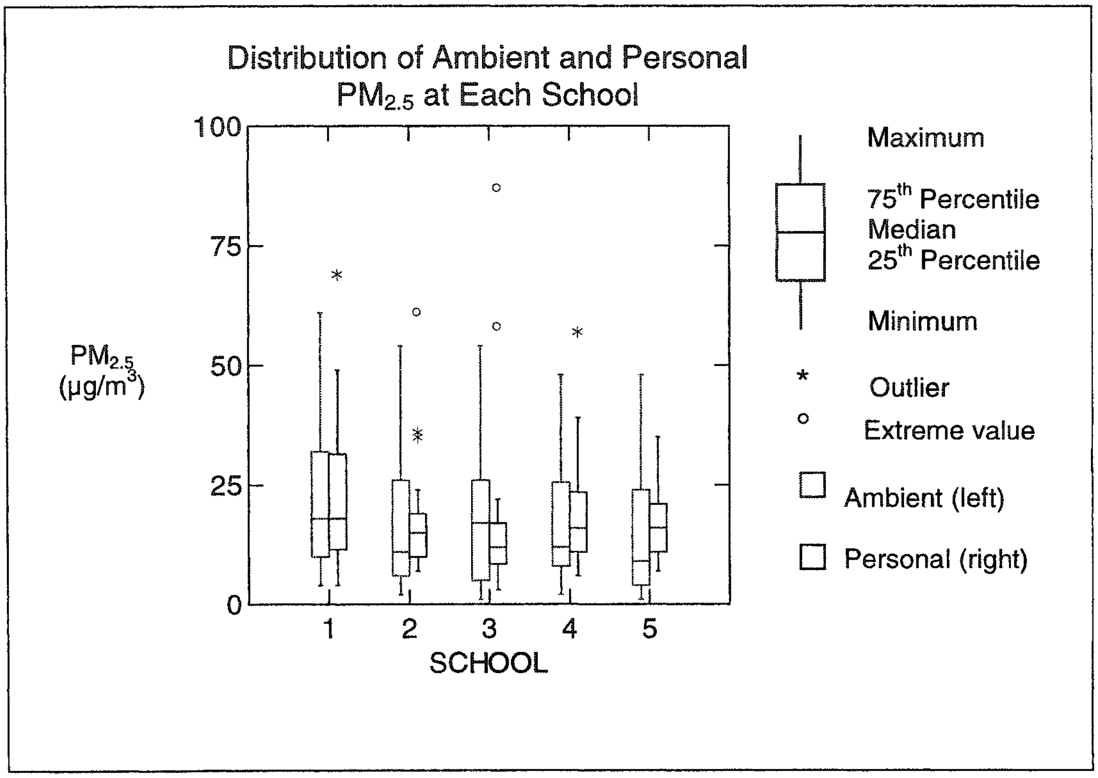


Figure 3-18 Box plots for $PM_{2.5}$ ambient concentration and personal exposures with data pooled by school. Two extreme outliers at Carney Hill are not shown as they were outside of the chosen scale of the figure but they were included in the analysis. 1=Carney Hill, 2=Gladstone, 3=Lakewood, 4=Westwood and 5=Glenview.

A comparison of the medians at each school shows a 0 to 7 µg/m^3 difference between ambient and personal $PM_{2.5}$ at each location. Carney Hill showed the smallest difference between personal and ambient median values while Glenview showed the largest. Although the range in personal data is usually greater for personal exposure, it is general due to rare peaks of high exposure (two extreme values are excluded from the plot at Carney Hill due to the chosen scale). The general variability is actually greater for ambient concentrations compared to personal exposures as indicated by the inter-quartile range of the box plots and the whiskers that show minimum and maximum values without extreme values and outliers. Comparing the medians at each school using the nonparametric Wilcoxon matched pairs test showed that even at Glenview the difference between ambient and personal medians was not statistically significant ($p>0.127$).

It is also important to look at the differences between both SO_4^{2-} and absorption coefficient throughout the city as both of these components of the total $PM_{2.5}$ mass represent different ambient sources. Figure 3-19 and Figure 3-20 show the distribution of the data for both sulphate and absorption coefficient personal and ambient levels at each school.

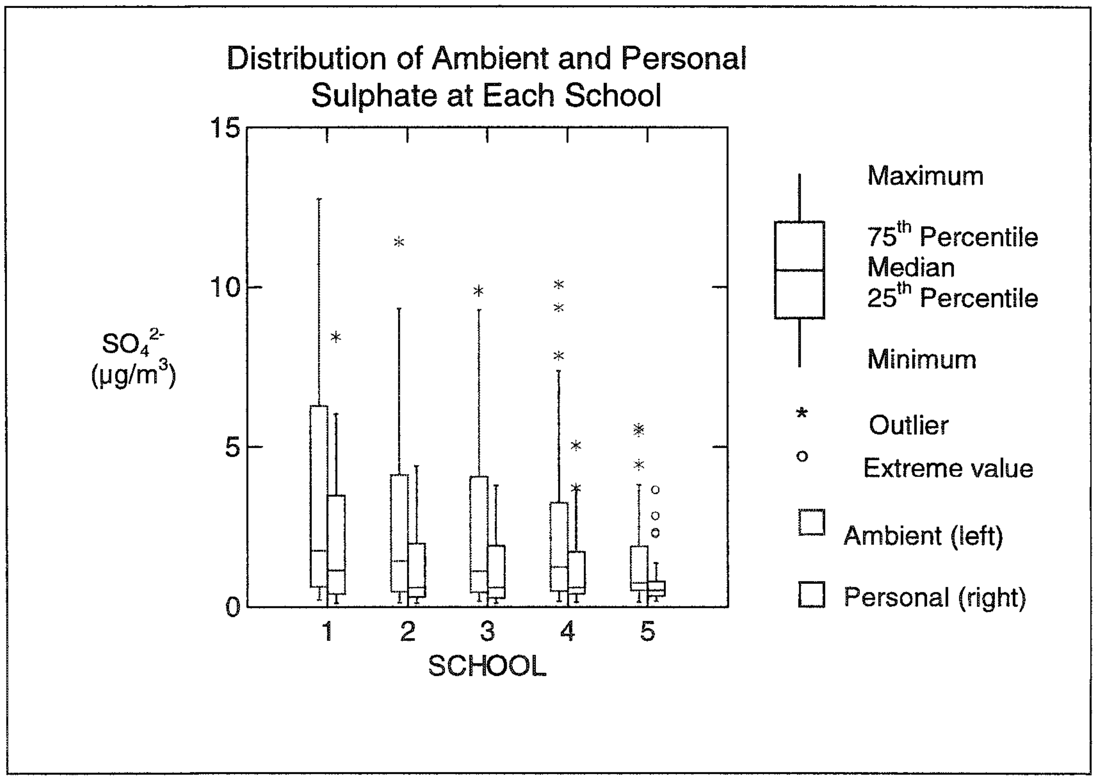


Figure 3-19 Box plots for sulphate ambient concentration and personal exposures with data pooled by school. 1=Carney Hill, 2=Gladstone, 3=Lakewood, 4=Westwood and 5=Glenview.

A visual analysis of sulphate box plots and time series clearly show a strong correlation between the ambient and personal data with higher ambient levels and proportionately lower personal levels in each neighbourhood. The box and whiskers demonstrate that the variability for personal exposure to sulphate is less than for overall ambient concentrations and the outliers in the personal exposure are not as extreme as with personal $PM_{2.5}$ exposure. The Wilcoxon matched pairs test showed that personal and ambient sulphate medians were statistically different from one another at each school and when the data was pooled across all schools ($p<0.001$). This is expected due to the limited amount of time that was spent outdoors. Because there are virtually no indoor sulphate sources, the only source of personal sulphate exposure was due to exposure while outdoors and exposure to ambient sulphate that infiltrates indoors. If the subjects in this study had spent a lot more time outdoors there may have been a smaller difference between ambient and personal sulphate levels.

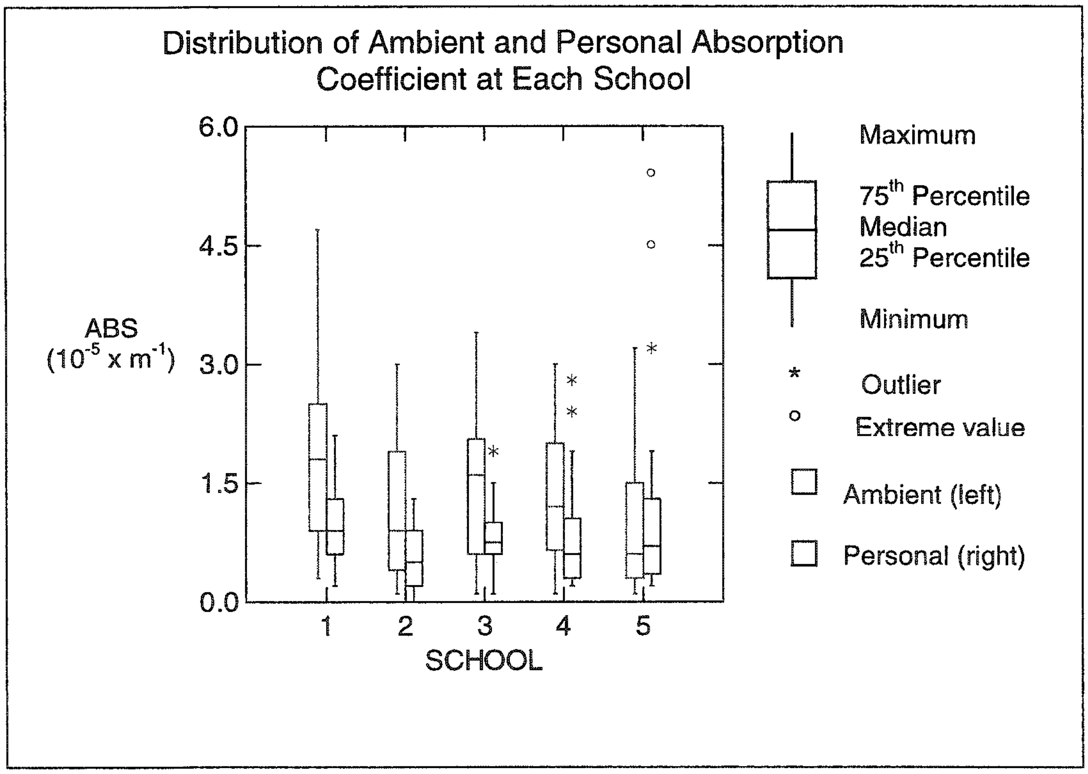


Figure 3-20 Box plots for personal and ambient absorption coefficient with data pooled by school. Two extreme outliers, 1 at Lakewood and 1 at Glenview, are not shown as they were outside of the chosen scale. 1=Carney Hill, 2=Gladstone, 3=Lakewood, 4=Westwood and 5=Glenview.

Visual analysis of the absorption coefficient data showed that the ambient and personal medians appear to correlate well except for at the Glenview location where personal absorption coefficient was slightly higher than ambient. At all other sites the ambient absorption coefficient is higher with the personal proportionately lower. The variation in personal exposure is less than for ambient with a small number of peak exposures shown as outliers and extreme values in the box plots (2 extreme values at Lakewood and Glenview were outside of the chosen scale for the plot). Statistical comparison of the personal and ambient absorption coefficient data using the Wilcoxon matched pairs test showed a significant difference when all of the schools were included in the analysis (p=0.000) and a statistical difference at each school separately (p<0.004) except for Glenview (p=0.443). At most of the schools there were generally only a few personal exposure samples where personal absorption coefficient was higher than the corresponding ambient level suggesting the presence of a non-ambient source impacting the personal sample or a very local ambient source. At Glenview, there were several samples where the personal level was higher than ambient, especially for one subject. This suggests that there may have been a local ambient source in the neighbourhood or perhaps even an indoor source at the school impacting the personal samples but not the ambient monitor on the school roof, which was noted previously. The time activity data did not reveal any relevant information except that the one subject (1002) with the greatest number of high personal exposures had a pellet stove in his home that was used intermittingly throughout the study period. This was not expected to make a difference in personal exposure level. Because there were only 3 subjects at each school it is difficult to determine if this was a neighbourhood effect or simply due to individual differences and varying exposures in their homes. More analysis will be provided in the following sections regarding the effect that location or school has on both the ambient concentrations and personal exposures.

3.4.6.2. *Ambient Spatial Variation*

When looking at the spatial variation in the ambient $PM_{2.5}$ data between the five schools there was a difference in medians of 9 $\mu g/m^3$. Carney Hill experienced the highest levels on average and Glenview had a significantly lower median concentration compared to all the other sites. For sulphate the largest difference between medians was 1.01$\mu g/m^3$, which was also the difference between Carney Hill and Glenview. Due to the non-normal distribution of the data, Kruskal Wallis analysis of variance (ANOVA) was first used to see if the difference in medians for ambient concentration between schools was significant. This analysis showed that there was no significant difference between schools for ambient $PM_{2.5}$ (p=0.378) or sulphate (p=0.361) when all schools were included in the non-parametric analysis. Although the non-parametric test is more appropriate due to the non-normal distribution of the data, a parametric ANOVA was also performed to see if the results were similar. The same conclusion was reached that there was no significant difference between schools for ambient $PM_{2.5}$. For sulphate the parametric ANOVA showed the same result for the pooled analysis of all five schools with no significant difference detected (p=0.177) but when just the schools with highest (Carney Hill) and lowest means (Glenview) were assessed there was significant difference detected ($p<0.024$)

The largest difference between schools for absorption coefficient was again between Carney Hill and Glenview and was 1.2 (10^{-5} m^{-1}). Spatial variation between the schools was detected using Kruskal-Wallis ANOVA ($p=0.043$), which was expected. Individual comparisons between the school with the highest median and the two schools with lowest median values showed that significant differences existed between Carney Hill and both Gladstone ($p<0.016$) and Glenview ($p<0.005$), the two schools located at the highest elevations and furthest distance from Carney Hill. The parametric ANOVA confirmed these findings. Traffic related pollutants, such as absorption coefficient, have been shown to be more spatially variable in an area than total $PM_{2.5}$ measurements (Oglesby *et al,* 2000 and Fischer *et al,* 2000). Another possible explanation for the observed absorption coefficient difference between schools could be the presence of wood-smoke in some neighbourhoods, which is another significant source of elemental carbon (Chow, 1995). Differences in the amount of residential wood-burning between the neighbourhoods were not known but field staff did notice the smell of wood-smoke on two occasions at the Glenview site and this was not reported for any other location. Indicating the presence of wood-smoke was not required of the field staff but generally qualitative comments regarding air quality were noted including the presence of smog or knowledge of an air quality advisory.

An inventory of traffic volume and residential wood-smoke plumes in the respective neighbourhoods would have provided useful information.

The above results show similar findings using simple parametric and non-parametric analysis of variance. These straightforward comparisons of median and mean values between schools illustrate the high degree of spatial variation that exists for absorption coefficient and are important because it is often only the average data that is considered for the purposes of management of an airshed. These results do not conclusively show that a spatial variation does not also exist between schools for total $PM_{2.5}$ and for sulphate. Another analysis (2-factor ANOVA) was also used to investigate the effect of location on ambient concentration while accounting for changing concentrations over time, instead of just comparing the mean or median value from each school for the entire study period. Again both parametric and non-parametric tests were performed producing identical results (slight variation in p-values). The results of the non-parametric Friedman's ANOVA are discussed below.

The 2-factor Friedman's ANOVA showed that there was a spatial difference between schools for all three measures: $PM_{2.5}$, sulphate and absorption coefficient ($p<0.001$). For $PM_{2.5}$ this significant difference between schools no longer existed if Carney Hill was removed from the analysis ($p=0.125$). But the spatial difference for both sulphate and absorption coefficient persisted when both Carney Hill and Glenview were removed from the analysis concurrently ($p\leq0.023$). The finding that sulphate was significantly different between schools was unexpected as sulphate is generally considered to be homogenous throughout an airshed due to formation time. Because Prince George has local industrial sources of sulphur dioxide and complex valley topography, spatial variation in sulphate is plausible. The prevailing wind direction during the study period carried the main industrial plumes in the direction of four of the study monitors and away from the fifth site the majority of the time explaining some of this spatial difference. It is not as clear why a significant spatial difference still exists when both Glenview, which was the site furthest out of the valley and had the lowest concentrations, and Carney Hill, the school closest to the industrial sources and had the highest ambient concentrations, were removed from the analysis. But complex steering of valley winds by the local topography could be responsible for such differences and possibly emissions of primary sulphate particulate matter. Primary emission of sulphate was suggested by Cyrys *et al* (1998) to explain spatially variability in Erfurt, Eastern Germany.

These results suggest that, the permanent monitoring site at Plaza 400, which is closest to Carney Hill relative to the other sites, may not adequately represent actual ambient levels of all three measures when time is considered in the analysis and even when comparing simple mean and median values between some schools for absorption coefficient. However, the correlation between schools was very high with median (range) Spearman correlations of 0.95 (0.71 to 0.96) for $PM_{2.5}$, 0.97 (0.86 to 0.98) for sulphate and 0.85 (0.67 to 0.91) for absorption coefficient. The lowest correlations were always between Glenview and one of the other schools. When wind directions are from the East, the Plaza monitoring site likely overestimates levels at all of the other schools, as was the case for Carney Hill, but due to high correlation between the schools temporal trends would be represented adequately. This may not be the case for different wind patterns. This conclusion is based on sampling with HPEM's and may be different for the continuously operated TEOM, which measures lower concentrations than the HPEM.

3.4.6.3. *Comparison to the MWLAP TEOM*

The TEOM data from the Ministry of Water, Land and Air Protection Plaza monitoring site was compared to Carney Hill (the closet site) and the other schools. A correction was applied to the data using the relationship described in chapter 2 between the HPEM and the TEOM. Due to the higher level of concentrations experienced during the field study this may overestimate higher concentrations. The regression equation from the Carney Hill HPEM concentrations versus adjusted TEOM data does suggest that this may indeed be the case with a slope less than 1 (see Figure 3-21) but deviation from the 1:1 line is minimal.

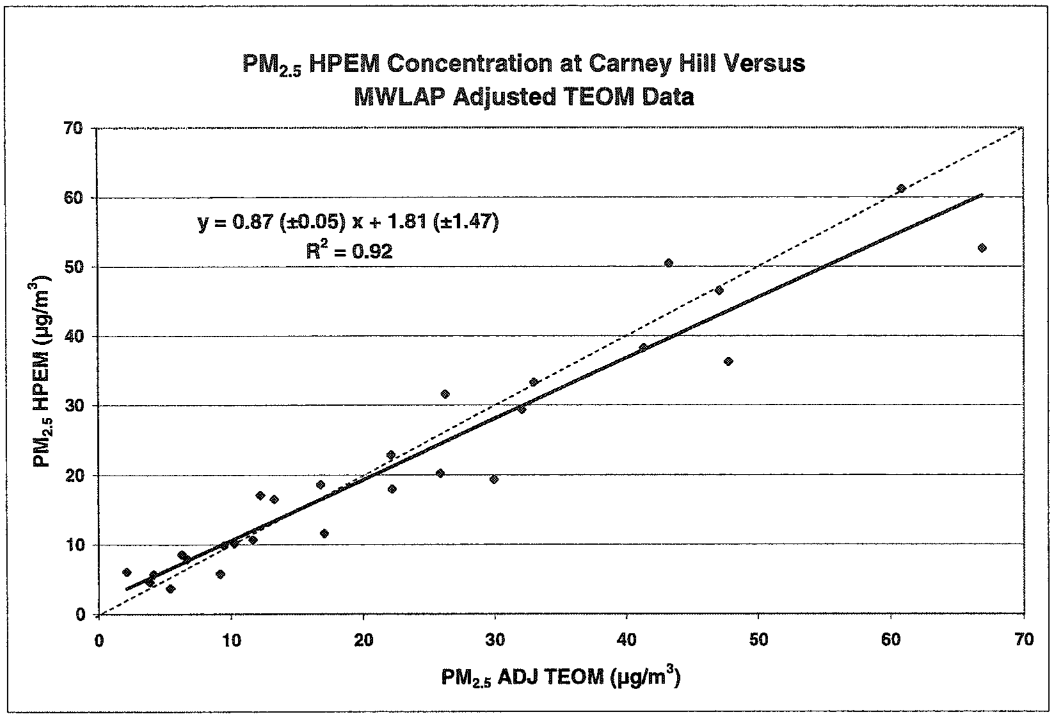


Figure 3-21 Least squares regression between Carney Hill HPEM and Ministry of Water Land and Air Protection TEOM data (adjusted using slope=1.38, intercept=0.62 from pilot study).

The slight difference between the Carney Hill data and the adjusted TEOM data from plaza could be a result of the different location and higher elevation of the TEOM sampler, suggesting that the Plaza monitoring site had slightly higher concentrations than Carney Hill. But the difference could also be due to the higher concentrations during the field study compared to the pilot study causing the scaling factor to be slightly inaccurate. The regression between the non-adjusted TEOM data and Carney Hill concentrations resulted in a slope of 1.21(±0.07) and intercept of 2.36 (±1.44). Comparison of both the non-adjusted and adjusted TEOM data to the other schools using the Friedman ANOVA showed a significant difference between sites for both data sets when all of the sites were included in the analysis ($p<0.000$). Post hoc tests comparing data from individual schools to the TEOM data showed interesting results. There was actually no significant difference found between the non-adjusted TEOM data and Gladstone, Lakewood and Glenview using the Friedman ANOVA, which accounts for both location and time in the analysis. There was a significant difference found for Carney Hill and Westwood ($p<0.01$). The adjusted TEOM data was not significantly different from Carney Hill, which is expected due to that schools close proximity to the Plaza monitoring site. There was a significant difference found between the adjusted TEOM data and all of the other schools. Interestingly, Westwood did not compare well with either TEOM data set, even though concentrations at Westwood were not significantly different than Lakewood (the nearest monitoring site).

There are also strongly significant Spearman correlations between the TEOM data and all of the school ambient monitoring sites. Glenview showed the lowest correlation of 0.83 and correlations ranged across all of the other schools from 0.95 to 0.97. These results suggest that the current MWLAP $PM_{2.5}$ TEOM, with no corrections made to the data, underestimates actual concentrations in the downtown area of Prince George where the monitor is located compared to sampling with HPEMs, but it does accurately represent 3 of the other 4 neighbourhoods monitored during this study. The high correlations suggest that temporal trends are adequately represented at all schools. Table 3-7 shows the regression results between non-adjusted $PM_{2.5}$ TEOM data and HPEM data from each of the school roof-top monitors.

Table 3-7 Regression results for ambient $PM_{2.5}$ from the HPEM samplers at the study sites versus 24-hour average data (non-adjusted) from the MWLAP $PM_{2.5}$ TEOM.

	Slope	Intercept	R^2
Carney Hill	1.21 (±0.07)	2.36 (±1.44)	0.92 (±0.11)
Gladstone	1.09 (±0.08)	-0.05 (±1.56)	0.89 (±0.12)
Lakewood	1.09 (±0.08)	0.386 (±1.63)	0.88 (±0.13)
Westwood	1.02 (±0.07)	2.73 (±1.41)	0.89 (±0.12)
Glenview	0.99 (±0.08)	-0.84 (±1.70)	0.85 (±0.14)

For the HPEM versus TEOM regressions the slopes had significant p-values less than 0.001 and the intercepts were not significantly different than zero. However, at Westwood the intercept was almost significantly different than zero (p=0.064) indicating that overall the HPEM data at Westwood was higher than the TEOM data by a similar offset throughout the period. Higher HPEM concentrations at Carney Hill are indicated by the increased slope which represents a percentage difference between the Carney Hill HPEM and the TEOM. For Carney Hill and Westwood, these equations could be used to calculate HPEM concentrations from the TEOM. For Gladstone, Lakewood and Glenview, use of the TEOM data directly is further confirmed by these equations. However, meteorology different from the conditions observed during the study period could result in increased error associated with these equations and may provide misleading results.

3.4.6.4. Personal Spatial Variation

Looking at spatial differences in the city for personal exposure was much more difficult due to the low number of individuals sampled at each school and the high degree of variation between individuals. Pooling the personal exposure data at each school would enable enough degrees of freedom to perform various statistical tests but any significant differences found between schools could be due to differences in individuals, therefore no firm conclusions could be made. Simple Kruskal-Wallis ANOVA comparing the median personal exposures at each school showed no significant differences for all three measures. Accounting for differences over time using Friedman's ANOVA did show a significant difference between schools for all three measures. If there were more individuals at each school, the effect that different individuals may have had on these results may not have been as important. Post hoc tests comparing individual schools using the Friedman test showed that for $PM_{2.5}$, all of the schools were significantly different from Lakewood only ($p<0.05$) but with Bonferroni correction only the difference between Lakewood and Carney Hill was significant ($p<0.005$). Post hoc tests for both sulphate and absorption coefficient showed different results with significant differences found between several different schools.

A mixed model ANOVA that assesses the amount of variation in a dependant variable due to various fixed and random factors was also used. Using this model, differences between individuals were accounted for as a random factor in the model. Personal exposures were the dependent variable, ambient concentrations were added as a covariate and school was set as a fixed factor. Preliminary analyses using this model suggest that for all three measures school had no significant effect on personal exposures, which is expected due to the large variation between individuals and the small number of subjects at each school. Further investigations with the data set using this modeling approach are planned.

3.4.7. Correlations and Regressions

3.4.7.1. Correlations for the Pooled Data

Pearson and Spearman correlations for the total pooled data set and the data at each school are shown in Table 3-8 and Table 3-9 respectively. Correlations were calculated between personal $PM_{2.5}$, SO_4^{2-} and absorption coefficient and each of the three ambient measures and also within the personal and ambient measures. It is mainly the personal to ambient correlation for each measure that is of interest but looking at the correlation between measures also provides important information. Because the data was not normally distributed Spearman correlations are more appropriate but for comparison purposes with other studies the Pearson correlations and linear regressions are also reported. When a strong correlation existed there was not a large difference between the Pearson and Spearman correlations but when the correlation was low the Spearman correlations showed an improved relationship between the variables. Only the Spearman correlations will be discussed in further detail although the Pearson correlations may be used for comparison to other studies.

Table 3-8 Spearman correlations (r) for all data pooled across subject and schools and pooled at each school. Note: *Significant at the α = 0.05 level. (2 extreme outliers removed for subject 1002). School labels are: CH =Carney Hill, GS = Gladstone, LW = Lakewood, WW = Westwood and GV = Glenview.

	All Data N=141 (139)	CH N=29 (27)	GS N=29	LW N=27	WW N=27	GV N=29
$PM_{2.5\,\text{Personal}}$ & $PM_{2.5\,\text{Ambient}}$	0.50* (0.52*)	0.38* (0.55*)	0.59*	0.66*	0.31	0.57*
$PM_{2.5\,\text{Personal}}$ & $SO_{4\,\text{Ambient}}^{2-}$	0.35* (0.37*)	0.30 (0.43*)	0.46*	0.52*	0.11	0.47*
$PM_{2.5\,\text{Personal}}$ & ABS_{Ambient}	0.48* (0.50*)	0.30 (0.47*)	0.59*	0.66*	0.29	0.55*
$SO_{4\,\text{Personal}}^{2-}$ & $PM_{2.5\,\text{Ambient}}$	0.87*	0.91*	0.94*	0.91*	0.91*	0.71*
$SO_{4\,\text{Personal}}^{2-}$ & $SO_{4\,\text{Ambient}}^{2-}$	0.96*	0.98*	0.98*	0.98*	0.92*	0.91*
$SO_{4\,\text{Personal}}^{2-}$ & ABS_{Ambient}	0.79*	0.73*	0.92*	0.81*	0.88*	0.64*
ABS_{Personal} & $PM_{2.5\,\text{Ambient}}$	0.68*	0.85*	0.93*	0.59*	0.44*	0.71*
ABS_{Personal} & $SO_{4\,\text{Ambient}}^{2-}$	0.57*	0.64*	0.93*	0.55*	0.26	0.61*
ABS_{Personal} & ABS_{Ambient}	0.73*	0.90*	0.94*	0.56*	0.51*	0.73*
$PM_{2.5\,\text{Ambient}}$ & $SO_{4\,\text{Ambient}}^{2-}$	0.88*	0.90*	0.94*	0.93*	0.91*	0.80*
$PM_{2.5\,\text{Ambient}}$ & ABS_{Ambient}	0.95*	0.87*	0.96*	0.96*	0.96*	0.96*
$SO_{4\,\text{Ambient}}^{2-}$ & ABS_{Ambient}	0.78*	0.68*	0.89*	0.84*	0.88*	0.70*
$PM_{2.5\,\text{Personal}}$ & $SO_{4\,\text{Personal}}^{2-}$	0.43* (0.45*)	0.34 (0.48*)	0.54*	0.55*	0.24	0.64*
$PM_{2.5\,\text{Personal}}$ & ABS_{Personal}	0.56* (0.57*)	0.47* (0.61*)	0.50*	0.80*	0.47*	0.68*
$SO_{4\,\text{Personal}}^{2-}$ & ABS_{Personal}	0.66*	0.71*	0.93*	0.55*	0.48*	0.71*

Table 3-9 Pearson correlations (R) for all data pooled across subject and schools and pooled at each school. Note: *Significant at α = 0.05 level. (2 extreme outliers removed for subject 1002.) School labels are: CH = Carney Hill, GS = Gladstone, LW = Lakewood, WW = Westwood and GV = Glenview.

	All Data N=141 (139)	CH N=29 (27)	GS N=29	LW N=27	WW N=27	GV N=29
$PM_{2.5\,\text{Personal}}$ & $PM_{2.5\,\text{Ambient}}$	0.16 (0.36*)	-0.02 (0.46*)	0.36	0.33	0.17	0.42*
$PM_{2.5\,\text{Personal}}$ & $SO_{4\,\text{Ambient}}^{2-}$	0.13 (0.31*)	-0.03 (0.40*)	0.31	0.26	0.10	0.36
$PM_{2.5\,\text{Personal}}$ & ABS_{Ambient}	0.19* (0.39*)	-0.05 (0.44*)	0.38*	0.39*	0.21	0.41*
$SO_{4\,\text{Personal}}^{2-}$ & $PM_{2.5\,\text{Ambient}}$	0.87*	0.94*	0.95*	0.89*	0.94*	0.71*
$SO_{4\,\text{Personal}}^{2-}$ & $SO_{4\,\text{Ambient}}^{2-}$	0.96*	0.97*	0.97*	0.96*	0.98*	0.95*
$SO_{4\,\text{Personal}}^{2-}$ & ABS_{Ambient}	0.74*	0.70*	0.90*	0.77*	0.84*	0.53*
ABS_{Personal} & $PM_{2.5\,\text{Ambient}}$	0.40*	0.78*	0.92*	0.50*	0.24	0.49*
ABS_{Personal} & $SO_{4\,\text{Ambient}}^{2-}$	0.29*	0.61*	0.87*	0.53*	0.21	0.33
ABS_{Personal} & ABS_{Ambient}	0.44*	0.90*	0.96*	0.48*	0.33	0.66*
$PM_{2.5\,\text{Ambient}}$ & $SO_{4\,\text{Ambient}}^{2-}$	0.91*	0.96*	0.97*	0.94*	0.95*	0.78*
$PM_{2.5\,\text{Ambient}}$ & ABS_{Ambient}	0.91*	0.84*	0.95*	0.95*	0.94*	0.94*
$SO_{4\,\text{Ambient}}^{2-}$ & ABS_{Ambient}	0.76*	0.70*	0.88*	0.81*	0.83*	0.62*
$PM_{2.5\,\text{Personal}}$ & $SO_{4\,\text{Personal}}^{2-}$	0.15 (0.34*)	-0.03 (0.40*)	0.33	0.21	0.14	0.47*
$PM_{2.5\,\text{Personal}}$ & ABS_{Personal}	0.20* (0.36*)	0.10 (0.60*)	0.41*	0.51*	0.31	0.47*
$SO_{4\,\text{Personal}}^{2-}$ & ABS_{Personal}	0.26*	0.64*	0.90*	0.44*	0.25	0.31

Almost all of the Spearman correlations in Table 3-8 are significant at an alpha level of 0.05. From the pooled data one can see that a relationship exists between each personal measure and all three of the ambient measures. For personal $PM_{2.5}$ this is expected because both ambient sulphate and absorption coefficient are components of the total ambient $PM_{2.5}$ sample. These significant relationships support the use of ambient measures to represent personal exposure. The higher correlations found between both personal sulphate and absorption coefficient with ambient $PM_{2.5}$ compared to the personal-ambient $PM_{2.5}$ correlation suggest that indeed these measures could be used as tracers of ambient source contribution to personal exposure.

Across schools the sulphate concentrations were the most consistent with relatively high significant correlations between personal sulphate and both ambient sulphate and ambient $PM_{2.5}$. Absorption coefficient also showed a similar pattern but with lower correlations at Lakewood and Westwood. It is not clear from the data why the correlations would be lower at these two schools. The high spatial variability in ambient absorption coefficient could be the reason as well as the large differences observed between individuals. Because there were only three subjects from each school this difference could also be the result of small sample size. More students at each school may have resulted in higher correlations at these schools. Interesting, there were moderately high significant correlations at Glenview between personal absorption coefficient and both ambient absorption coefficient and ambient $PM_{2.5}$ despite the high median personal/ambient ratio at this school for absorption coefficient (1.10) compared to all the other schools (0.48 to 0.56). This suggests that although personal exposures were higher at that school relative to the ambient concentration, they were consistently higher and therefore a stronger correlation existed.

For $PM_{2.5}$ the personal versus ambient Spearman correlations were moderate and consistent between the pooled analysis (slightly lower) and across schools except for at Westwood where the correlation was low and insignificant. The correlations between personal $PM_{2.5}$ and both ambient sulphate and ambient absorption coefficient were also low and insignificant at Westwood. This suggests that there may have been more variance in the personal or indoor sources at this school and less influence from the ambient concentrations, which was also suggested by the correlations with inversion strength. Subjects 4001 and 4003 from this school did spend a significant amount of time out of their neighbourhood compared to most of the subjects in the study (Figure 3-6). The time spent out of the neighbourhood was 7.7 and 4.6% respectively compared to an average of 3.2% across all of the subjects. This could be important, especially since these individuals spent the majority of this time at the same location, an indoor ice rink and gymnastics centre respectively. Table 3-10 shows that these two subjects had the lowest individual correlations for personal-ambient $PM_{2.5;}$ the other subject at this school also had a low insignificant correlation. There appears to be some factor at that school affecting the personal-ambient relationship that is not apparent in analysis of the actual personal and ambient time series or the general pooled descriptive statistics for that school. Analysis of the difference between ambient and personal levels at

Westwood School did not reveal any discrepancy between this school and the others.

Correlations within the ambient and personal data were also assessed to further investigate the relationship between total $PM_{2.5}$ and the sulphate and absorption coefficient components. These correlations are also summarized in Table 3-8 and Table 3-9. Total ambient $PM_{2.5}$ showed strong Spearman correlations with both ambient sulphate (r=0.88) and ambient absorption coefficient (r=0.95). A significant correlation is expected since each of these measures comes from the same sample but the strong correlations also support the use of both sulphate and absorption coefficient as indicators of ambient $PM_{2.5}$. A significant but much lower correlation was found between total personal $PM_{2.5}$ and personal sulphate (r=0.43) and absorption coefficient (r=0.56), which is expected due to the relatively large contribution of non-ambient sources to total $PM_{2.5}$ personal exposures. The corresponding Pearson correlations show an even bigger difference between the ambient and personal comparisons with high ambient correlations of 0.91 for both measures and lower personal correlations of 0.15 and 0.20 for sulphate and absorption coefficient with total $PM_{2.5}$ respectively. The relationship between sulphate and absorption coefficient within the personal and ambient sources is also interesting. For the ambient samples one would not necessary expect a high correlation given that these measures are expected to be from different sources but moderate to high Spearman correlations were found across schools, with a slightly lower correlation at Carney Hill. For the personal exposure samples one would expect these measures to be more highly correlated since they are generally from ambient sources only, although different ambient sources. Moderate to high correlations were found at most of the schools but Lakewood and Westwood had a weaker relationship. When cases with very high personal absorption coefficient relative to the ambient counterpart were removed from the analysis, the personal sulphate versus absorption coefficient correlations improved to some extent. The pooled Spearman correlation increased to 0.76; Lakewood and Westwood increased to 0.69 and 0.78 respectively; while Glenview decreased to 0.63. This could be due to the high personal/ambient ratio at this school for absorption coefficient discussed previously.

3.4.7.2. <u>Individual Longitudinal Correlations and Regressions</u>

Individual Spearman correlations and Pearson correlations with the corresponding regression slope and intercept are reported for the personal to ambient correlation of each measure in Table 3-10 and Table 3-11.

Table 3-10 Spearman correlation (r) between personal exposure and ambient concentration for each pollutant measure by subject. Note: *Significant at $\alpha = 0.05$ level. **Significant at $\alpha = 0.10$ level. (2 extreme outliers removed for subject 1002.) Arithmetic mean, median, standard deviation (Stdev) and inter-quartile range (IQR) of the individual results are also reported.

School	Subject ID #	N	$PM_{2.5}$	SO_4^{2-}	Absorption coefficient
Carney Hill	1001	10	0.66*	0.99*	0.71*
	1002	9 (7)	0.00 (0.64)	0.98*	0.87*
	1003	10	0.55**	0.98*	0.94*
Gladstone	2001	9	0.83*	0.93*	0.80*
	2002	10	0.53	0.99*	0.89*
	2003	10	0.72*	0.98*	0.92*
Lakewood	3001	10	0.50	0.95*	0.62**
	3002	8	0.71*	1.00*	0.60
	3003	9	0.70*	0.93*	0.63**
Westwood	4001	10	0.18	0.85*	0.73*
	4002	7	0.39	1.00*	0.71**
	4003	10	0.35	0.92*	0.27
Glenview	5001	10	0.49	0.93*	0.79*
	5002	9	0.40	0.83*	0.23
	5003	10	0.60**	0.90*	0.81*
Mean			0.51 (0.55)	0.94	0.70
Stdev			0.22 (0.17)	0.05	0.21
Median			0.53 (0.55)	0.95	0.73
IQR			0.28 (0.24)	0.06	0.22

Table 3-11 Summary of linear regression results and Pearson correlation (R) between personal exposure and ambient concentration for each pollutant measure by subject. Note: *Significant at $\alpha = 0.05$ level. **Significant at $\alpha = 0.10$ level. (2 extreme outliers removed for subject 1002.) Arithmetic mean, median, standard deviation (Stdev) and inter-quartile range (IQR) of the individual results are also reported.

ID	$PM_{2.5}$			SO_4^{2-}			Absorption coefficient		
	R	Intercept	Slope	R	Intercept	Slope	R	Intercept	Slope
1001	0.82*	6.12	0.48	0.98*	0.09	0.60	0.85*	0.23	0.39
1002	-0.34 (0.36)	96.19 (31.69)	-1.32 (0.41)	0.98*	0.27	0.45	0.85*	0.23	0.39
1003	0.60**	9.21	0.41	0.99*	0.08	0.49	0.97*	0.01	0.49
2001	0.76*	10.24	0.41	0.99*	-0.04	0.47	0.95*	0.05	0.45
2002	0.26	15.89	0.31	0.96*	0.24	0.30	0.97*	0.13	0.38
2003	0.61**	10.62	0.17	0.98*	0.11	0.35	0.99*	0.04	0.37
3001	0.31	15.66	0.52	0.97*	0.15	0.43	0.72*	-2.85	3.39
3002	0.82*	7.62	0.28	0.97*	0.24	0.35	0.77*	0.32	0.45
3003	0.69*	5.90	0.23	0.96*	0.11	0.42	0.63**	0.31	0.19
4001	-0.10	26.01	-0.11	0.99*	0.28	0.45	0.38	0.54	0.29
4002	0.41	9.71	0.19	0.99*	0.00	0.52	0.68	0.20	0.10
4003	0.36	15.82	0.18	0.98*	0.07	0.42	0.22	0.75	0.20
5001	0.27	18.28	0.14	0.94*	0.17	0.45	0.72*	-0.10	2.03
5002	0.49	12.17	0.46	0.97*	0.08	0.57	0.19	0.57	0.10
5003	0.73*	9.59	0.22	0.95*	-0.02	0.57	0.71*	0.01	1.03
Mean	0.45 (0.49)	17.94 (13.64)	0.17 (0.29)	0.97*	0.12	0.46	0.71	0.03	0.68
Stdev	0.34 (0.26)	22.29 (7.28)	0.44 (0.17)	0.02	0.10	0.09	0.26	0.83	0.89
Median	0.49 (0.49)	10.62 (10.62)	0.23 (0.28)	0.98*	0.11	0.45	0.72*	0.20	0.39
IQR	0.42 (0.38)	6.46 (6.46)	0.24 (0.23)	0.02	0.13	0.09	0.25	0.29	0.23

Analysis of the individual Spearman correlations shows that the significance between personal and ambient $PM_{2.5}$ correlation is not as strong with only 5 of the 15 subjects showing a significant correlation; the small sample size for each individual is probably the main reason for this. The correlation calculated for the pooled data (0.50) is the same as the median of both the school correlations (0.50) and the individual correlations (0.51). When the two extreme outliers were removed the overall pooled correlation and median of the school and subject correlations were improved slightly to 0.52, 0.54 and 0.55 respectively. Sarnat *et al.* (2000) reports the Spearman correlations for summer and winter seasons separately. The median Spearman correlations across individuals for personal and ambient $PM_{2.5}$ were 0.76 (-0.21 to 0.95) and 0.25 (-0.38 to 0.81) for summer and winter respectively. A stronger winter correlation of 0.55 (0.18 to 0.83) was found in the current study. Differences in the sample population could explain this difference as Sarnat was studying senior citizens that probably had very different activity patterns than the children in this study. Four to six subjects were monitored concurrently in the study by Sarnat making the results comparable to this study where five subjects were monitored on each day. A much higher PM_3 Pearson correlation of 0.86 was reported by Janssen *et al* (1999). Although this study was of children of similar age, it took place in the spring. The corresponding Pearson correlation in the current study was 0.49. Other studies have reported even lower individual correlations between personal exposures and ambient concentrations for $PM_{2.5}$. Median individual Pearson correlations (and ranges) of 0.34 (-0.57 to 0.98) and 0.37 (0.01 to 0.87) were reported by Liu *et al* (2003) and Rojas-Bracho *et al* (2000) respectively. Williams *et al* (2003) reported a mean Pearson correlation of 0.39 with a range of 0.00 to 0.65. All three of these studies took place over 1 or 2 years and included data from both winter and summer seasons. The results presented by Sarnat *et al* (2000) suggest that the differences found between studies may be due to a seasonal influence, explaining the high correlations observed by Janssen *et al* (1999) during the spring compared to lower correlations found in this study during the winter. The even lower correlations observed in several of the other studies could be due to the effect of sampling in multiple seasons.

The individual Spearman correlations for sulphate were very strong across all individuals with a median value of 0.95 (0.83 to 1.00). This confirms that there are little non-ambient sources of sulphate and further validates the use of sulphate as an indicator of ambient exposure. Individual absorption coefficient Spearman correlation were also strong (median=0.73) but there was an insignificant correlation for 3 subjects from 3 different schools. This was most likely the result of 1 or 2 high personal exposures that were much greater than the ambient counterpart for each of these individuals. These high exposures were probably due to an indoor combustion source or a local ambient source such as traffic or residential wood-smoke. Use of absorption coefficient as an indicator of exposure to ambient sources is still possible if the small number of samples where non-ambient sources were likely are removed from the analysis.

3.4.8. Comparison to Ambient Standards

In Canada, there are two ambient air quality objectives for $PM_{2.5}$: the recommended Federal Reference Level (FRL) of 15 $\mu g/m^3$ and the Canada-Wide Standard (CWS) of 30 $\mu g/m^3$ both for 24-hour averages. The FRL is an unofficial target level for a 24-hour period and is an estimate of the lowest ambient $PM_{2.5}$ level at which statistically significant increases in health responses can be detected based on available data and current technology (CEPA/FPAC, 1999). This level is not a known threshold of effects below which impacts do not occur but health effects to concentrations above that level have been documented in the literature. Achievement of the CWS is based on the annual 98th percentile averaged over 3 consecutive years (CCME, 2000). This means that the annual average number of days where concentrations are over 30 μg/m^3 must be fewer than 7.3 days (2%) for a 3 year period. These objectives are only recommended levels that provide a benchmark for air quality improvements. Achievement of the CWS standard does not ensure protection of human health and effects at or below this level are possible. The data from this study was compared to these objectives as a means of characterizing the overall air quality in Prince George during the study period for fine particulate matter. It is important to note that February of 2001 did have the highest average concentration compared to all other months of that year. In addition, this monthly average was 4 μg/m^3 higher than the historical average ambient concentration for February over the last 4 years (Fudge *et al*, 2003).

The number of days and percentage of 24-hour samples at each school that exceeded the Reference Level are reported in Table 3-12 for each school and for all of the sites combined. Table 3-13 shows the number of days and the percentage of 24-hour samples that were greater than the level of the Canada Wide Standard as well as the 98th percentiles for the study period. Although these standards are recommended levels for ambient air quality only, a comparison is made to both the ambient and personal samples. Interpretation of this comparison for the personal samples should be performed cautiously and is intended to be for descriptive purposes only.

Table 3-12 Number of days (%) of ambient and personal samples exceeding the Federal Reference Level of 15 μg/m^3 for a 24-hour period.

	All Schools	Carney Hill	Gladstone	Lakewood	Westwood	Glenview

Ambient	68 (46%)	18 (62%)	11 (37%)	14 (47%)	14 (47%)	11 (37%)
Personal	89 (63%)	21 (70%)	17 (59%)	15 (56%)	19 (70%)	17 (59%)

Table 3-13 Number of days (%) of ambient and personal samples exceeding the Canada-Wide Standard of 30 $\mu g/m^3$ for a 24-hour period. Ambient and personal 98th percentiles ($\mu g/m^3$) are shown for the pooled data as well as the data from each school for the 6-week study period. (* Indicates analysis with two extreme outliers removed.)

	All Schools	Carney Hill	Gladstone	Lakewood	Westwood	Glenview
Ambient	32 (21%)	9 (31%)	6 (21%)	5 (17%)	7 (24%)	5 (17%)
Personal	28 (20%)	9 (30%)	4 (13%)	5 (17%)	6 (20%)	4 (13%)
Ambient 98th%	54	61	54	54	48	48
Personal 98th %	87 (61*)	179 (69*)	61	87	57	35

For the personal samples, this comparison provides an indication that the number of exposures greater than the Canada Wide Standard is not much different than for the ambient samples. This result may be due to chance and does not clearly show that when the personal exposures are higher it is due to the high ambient levels. Of the 28 personal exposures greater than 30 $\mu g/m^3$, 8 samples or 29% corresponded to similarly high ambient concentrations. The impact of high ambient concentrations on personal exposure would greatly depend on an individual's activity on that day, including time spent outdoors and the ventilation rate in their home. Because study participants spent the majority of their time indoors with windows closed this low percentage is expected and a much higher value would be expected for a summer study. The increased percentage of personal samples greater than the FRL compared to the ambient samples clearer shows that personal activity and behaviour can greatly affect exposures. Although there is no clear evidence that non-ambient exposures have significant health effects (except for smoking), there may be simple measures that the public can do to reduce their own personal exposure, including use of an exhaust fan while cooking.

Almost half of the ambient samples exceeded the reference level during this 6-week period and the 98th percentile for the ambient data at each school well exceeded the level of the Canada Wide Standard. Interestingly, all of the schools showed a similar ambient 98th percentile except Carney Hill, which is the site closest to the main industrial area in the city. These results do not necessarily represent failure to achieve the standard at all schools because the study period was only for 6-weeks. However, Carney Hill did experience concentrations greater than 30 $\mu g/m^3$ on 9 days during the study period, which resulted in failure to achieve the standard for 2001. Even if concentrations had been zero for the rest of the year the standard would not have been achieved at that location. Although the standard is based on a 3 year average, failure for a single year should still be of concern to both airshed managers and residents of the city. Acute health effects can result from high concentrations over the short-term; therefore standards based on the annual average over several years are more relevant to protection from chronic health effects. The standard for 2001 was nearly exceeded at Westwood and Gladstone, having 7 and 6 days during the study period with levels greater than 30 $\mu g/m^3$ respectively (7.3 days per year > 30 $\mu g/m^3$ is needed to exceed the standard in a given year). The standard was likely exceeded at these schools before the end of the year and possibly by the end of the study due to high levels prior to the start of the study. At Lakewood and Glenview there were 5 days with concentrations greater than 30 $\mu g/m^3$ during the study period.

These results demonstrate the need to reduce ambient $PM_{2.5}$ levels in the city and further investigate the impacts on human health in this airshed, especially in the downtown area of the city close to Carney Hill. The significantly greater number of days exceeding both the FRL and CWS at Carney Hill compared to the other schools suggests that residents of the downtown area of Prince George may have a greater risk of experiencing health effects from fine particle exposure. There also appears to be a slight difference between the other valley schools (Lakewood and Westwood) and the higher elevation schools (Gladstone and Glenview), when considering days where the concentration was greater than 15 $\mu g/m^3$. There was a 10% decrease in days exceeding the FRL for the ambient data at the higher elevation schools. Because the length of the study was short, a 10% decrease only corresponds to 3 days or samples. This does not provide enough evidence to suggest that higher elevation neighbourhoods may have a lower risk to health effects from $PM_{2.5}$, but if this same trend occurred for an entire year this may indeed be the case. This same decrease is not apparent with the CWS suggesting that when levels are higher (above 30 $\mu g/m^3$) these four schools are affected equally.

3.5. Conclusion

The selection process for the study participants went very smoothly and resulted in committed students that were excited to participate. Adding some randomness into the selection process would have added statistical power to the results but would probably not have yielded such high compliance and may have resulted in dropouts from the study. If time had permitted it may have been beneficial to approach a greater number of schools and interviewed more classes. This would have resulted in a bigger pool to select the study participants. Increasing the number of students being monitored and therefore adding to the sample size would have been the main way to improve on this study. Adding more sampling days to each participant would not have been feasible. Monitoring each individual for 10 days over the 6-week period was probably all that the children could handle. By the end of the study the novelty of carrying the sampler had worn off and although they were still committed to completing the study, they were all glad when it was finished. It may have been possible to resample the same individuals for a similar length period during a different season after having a lengthy break from carrying the sampler. Obtaining more detail regarding time activity from the parents and teachers would have provided greater ability to assess the cause of high exposures.

Analyses of the meteorological conditions during the study suggest that diurnal inversions had a significant impact on ambient pollution levels during the study period. Inversion conditions existed on 62% of the study days and pollution levels above 30 $\mu g/m^3$ were always associated with an inversion on the same day. Peak concentrations were generally reached after the dissipation of an inversion and usually occurred at the same time elementary school children were playing outdoors during lunch hour. Time series plots of ambient concentration and 24-hour inversion strength clearly showed that high concentrations occurred with inversions and all five of the study sites experienced elevated concentrations with inversions conditions. Moderate correlations were found between 24-hour inversion strength and the ambient concentrations at each of the five schools. Comparable correlations were also found for personal exposure to sulphate at all schools and slightly lower significant correlations were found for both $PM_{2.5}$ and absorption coefficient exposures for four of the five schools. Ambient pollution levels greater than 15 $\mu g/m^3$ were associated with a prevailing easterly wind direction 60% of the time, which would be blowing the main industrial sources towards four of the study monitoring sites and towards the MWLAP TEOM. Study findings suggest that the air quality present on days with high concentrations during the study was likely dominated by industrial emissions from the east and occasionally from sources to the southwest.

Spatial variation in ambient concentrations was investigated by simple testing of differences between medians and means using analysis of variance. These tests showed that for $PM_{2.5}$ and sulphate there was no significant difference in ambient concentrations between schools. There was a significant spatial difference detected for absorption coefficient which was expected. Post hoc analysis showed that the main differences were between Carney Hill and both Gladstone and Glenview. Accounting for changes over time while looking for differences between schools showed there was actually a significant difference between schools for all three measures. For $PM_{2.5}$ the spatial difference was due to high concentrations at Carney Hill but for both sulphate and absorption coefficient the significant spatial difference persisted even when the school with the highest and lowest concentrations (Carney Hill and Glenview) were removed from the analysis. For absorption coefficient this spatial variation is expected but sulphate is generally considered to be homogenous in most airsheds as it is a secondary pollutant formed through chemical reactions in the atmosphere. The meteorology during the study period and the effect of local valley topography were likely the cause of the observed spatial differences and possibly primary emissions of sulphate. These results suggest that the location of the permanent ambient $PM_{2.5}$ monitor that is located close to the Carney Hill study site may not accurately represent actual levels of $PM_{2.5}$ at the other monitoring sites and may overestimate ambient concentrations throughout the city. However, because the WLAP $PM_{2.5}$ TEOM measures lower than the HPEM samplers used in the study there was actual no significant difference between the TEOM data and 3 of the study sites. There were also very high correlations between the study sites and the TEOM (0.83 to 0.97) suggesting that scaling of concentrations measured at the central monitoring site and consideration of meteorological factors would enable an assessment of levels throughout the airshed.

Analyses of the pooled data showed stronger personal-ambient Spearman correlations for sulphate (0.96) and absorption coefficient (0.73) compared to total $PM_{2.5}$ (0.52). Comparison of the actual levels of personal and ambient $PM_{2.5}$ showed that there was no statistical difference between medians, however, there was a significant difference found for both sulphate and absorption coefficient. Consideration of these results and the correlations suggest that the personal $PM_{2.5}$ exposures were impacted by non-ambient sources causing levels to be close to the ambient conditions when the data was pooled. For sulphate there was virtually no influence of non-ambient sources and the lower personal exposures were due to a low infiltration of ambient sulphate indoors and the limited amount of time spent outdoors. The same results were found for absorption coefficient although there were several samples with exposures significantly higher than the ambient counterpart that resulted in a slightly lower correlation and suggests that a personal, indoor or very local ambient source of elemental carbon was present on some occasions. Similar results were found when the data were analyzed by school. One difference found between schools for absorption coefficient was that ambient and personal levels were not significantly different at Glenview. At this school the ambient absorption coefficient levels were lower than all the other schools but the personal exposures were comparable. A number of personal exposures higher than the ambient level were found at this school suggesting the possibility of a very local ambient source in this neighbourhood, such as wood-smoke, that did not impact the local outdoor monitor, or possibly an indoor source at the school. The low sample size at each school makes it impossible to discern if there is really a neighbourhood or school effect at Glenview or if the high personal exposures are just due to different individual exposures at home.

Analyses of the personal-ambient correlations for each individual showed comparable results to other studies for $PM_{2.5}$ with large differences found between individuals and a median correlation of 0.55 comparable to the correlation found with the pooled data. Sulphate showed very consistent and strong correlations across individuals with a median of 0.95, supporting its use as an indicator of ambient generated exposures. Absorption coefficient demonstrated a lower median correlation of 0.73 with more variability found between individuals possibly due to the influence of non-ambient sources in a relatively low number of samples. Removal of samples where a non-ambient influence was likely would enable absorption coefficient to be used as a marker for ambient generated exposure.

Assessment of the number of ambient and personal samples greater than 15 μg/m^3 and 30 μg/m^3 provided a means of characterizing the overall air quality in Prince George over the study period for total $PM_{2.5}$. High personal exposures, greater than 30 μg/m^3, were associated with similarly high ambient concentrations 29% of the time when days with high personal concentrations were compared to the ambient concentrations. This suggests that the majority of the high $PM_{2.5}$ exposures were not the result of high ambient concentrations but were due to the presence of non-ambient sources. Although the main evidence for a health effect of $PM_{2.5}$ is based on ambient data only, these results suggest that individuals may be able to reduce their own personal exposure level by making different personal choices such as using an exhaust fan while cooking. It is unknown whether or not this would result in a reduction of actually risk to health effects from $PM_{2.5}$. For the ambient data, almost half of the samples were greater than 15 μg/m^3, which is the lowest ambient level at which statistically significant increases in health responses can be detected based on available data and current technology (CEPA/FPAC, 1999). At all of the schools, the 98th percentile for the study period well exceeded the recommended Canada

Wide Standard of 30 µg/m^3 (which is the level of the Canada-Wide Standard based on a 3-year annual average of the 98th percentile). Carney Hill showed a relatively higher 98th percentile than all of the other locations suggesting that residents of the downtown area of Prince George may have a greater risk of experiencing health effects from ambient $PM_{2.5}$. At Carney Hill, there were 9 days during the study period where concentrations were greater than 30 µg/m^3, which was enough to exceed the CWS for 2001. Higher elevation sites appeared to experience levels greater than 30 µg/m^3 close to the same number of days as the valley schools. There were a lower number of days where levels were above 15 µg/m^3 at the higher elevation sites but due to the short study period this difference does not provide enough evidence to suggest that lower elevation neighbourhoods may have a greater risk to health effects from ambient $PM_{2.5}$.

4 Personal Exposure to Ambient and Non-ambient Sources

4.1. Abstract

Sulphate (SO_4^{2-}) and elemental carbon (EC) were used as tracers of ambient fine particulate matter ($PM_{2.5}$) to estimate children's exposure to ambient sources during a 6-week winter period in the city of Prince George, British Columbia. Ambient and non-ambient exposures were estimated from personal/ambient SO_4^{2-} and EC ratios, ambient $PM_{2.5}$ concentrations, measured total personal exposures and time activity data. Estimates for the average infiltration factor (equilibrium fraction of ambient $PM_{2.5}$ found indoors) and air exchange rate were determined for the indoor environment of each individual in the study. Using pooled data across all subjects and SO_4^{2-} as a tracer of ambient PM, the median exposure to ambient PM was 8±1.7 (range: 1 - 36) μg/m^3. Similar values were obtained when EC was used as a tracer of ambient PM; median = 7±2.2 (range: 1 - 33) μg/m^3. A strong association was found between each of these estimates and measured ambient concentrations at both the closest school monitor (r=0.92) and the central site (r=0.88). Individual longitudinal regressions demonstrated that personal exposure to ambient PM was ~45% of ambient concentration based on SO_4^{2-}; 45-60% based on EC. An average indoor infiltration factor for ambient $PM_{2.5}$ of 0.53±0.16 and an air exchange rate of 0.35±0.39 (h^{-1}) were estimated through analysis of the individual data. The permanent central site monitor was found to be suitable for assessing longitudinal ambient exposure

throughout the city. Ambient and non-ambient $PM_{2.5}$ each contributed an almost equal proportion to total personal exposure; the large variation in non-ambient exposure was mainly the result of the rare occurrence of large peaks in exposure. These results indicate that ambient concentrations were highly correlated with exposure to ambient $PM_{2.5}$ and support the use of ambient data for a longitudinal health study in the city.

4.2. Introduction

In the last few years research has been focused on separating the different components of total personal exposure. A 24-hour sample measuring personal exposure is composed of many different types of particles from various source types that may have come from several different indoor and outdoor microenvironments as well as from personal activities unique to the individual being sampled. A personal exposure sample may actually be a confounding factor in investigations dealing with effects of outdoor particles due to the large influence of indoor sources and personal activity (Monn, 2001). It has been determined that ambient concentrations of particulate matter (PM) are not highly correlated with personal exposure to non-ambient PM or total PM (unless non-ambient sources are minimal) but are highly correlated with personal exposure to ambient generated PM (Wilson *et al*, 2000). This finding supports the idea of using ambient concentrations from a community monitoring site in a health study as long as the ambient data is used as a surrogate for personal exposure to ambient generated PM only.

Oglesby *et al* (2000) also concluded that ambient $PM_{2.5}$ levels may be more appropriate exposure estimates than total personal exposure because the total personal exposure reflects a mixture of both indoor and outdoor sources while the ambient measure represents outdoor sources only. They did find that the ambient measure may not be an accurate surrogate for all types of PM including primary traffic-related particles and coarse particles that are more spatially variable in the local environment. Mage *et al* (1999) demonstrated that sources of PM of non-ambient origin operate independently of the ambient PM concentrations; therefore, any mortality effects from either component must be independent and should be investigated separately.

Although it is recognized that PM generated indoors must also be considered when investigating health impacts of particulate pollution, the investigation of ambient generated particulate matter and the impact of outdoor sources is more relevant to air quality management. Governments do not have regulatory authority regarding most indoor generated pollution, although they should be educating the public regarding the hazards; but they are responsible for protecting the public from the hazards of ambient generated air pollution (Wilson *et al,* 2000). Outdoor air is also the second most important source indoors (Monn, 2001). The need for further investigation into exposures attributable to outdoor sources was recognized by the National Research Council in their 1998 report on airborne particulate matter.

Researchers have only started to assess the health impacts of the ambient and non-ambient components of PM pollution. A very recent study found that just the ambient, not the non-ambient component, were associated with decreased lung function, decreased systolic blood pressure, increased heart rate and increased supraventricular ectopic heartbeats (Ebelt *et al*, 2003). Due to the small sample size and limited number of measurements per subject these results are preliminary and need to be replicated in a larger study.

There are several methods available for assessing the contribution of ambient-generated sources to personal exposures (Wilson *et al*, 2000). One of these methods is to use a tracer species, such as sulphate, that has little or no indoor sources. Both Sarnat *et al* (2000) and Ebelt *et al* (2000) suggested the use of sulphate as a tracer for ambient $PM_{2.5}$ and reported higher personal-ambient correlations for sulphate than for total $PM_{2.5}$. Sarnat *et al* (2000) also showed the regression between ambient $PM_{2.5}$ and personal exposure to $PM_{2.5}$ of ambient origin only (based on the personal/ambient ratio for sulphate) and demonstrated the contribution of ambient origin $PM_{2.5}$ to range from 55 to 75% (±13-16%) depending on ventilation status. Leaderer *et al* (1999) used the indoor/outdoor mean sulphate ratio to determine that on average 75% of the fine aerosol indoors during the summer is associated with outdoor sources. The use of sulphur as a tracer of outdoor $PM_{2.5}$ was investigated further by Sarnat *et al* (2002) and showed that sulphur (predominantly in the form of sulphate) could be used as a suitable tracer for all size fractions of total ambient $PM_{2.5}$. Sulphur is more representative of particles in the accumulation mode and more specifically particles in the size range from 0.06 to 0.5 µm due to deposition and penetration differences for both smaller and larger particles but it was shown that indoor/outdoor sulphur ratios were significant predictors of indoor/outdoor ratios for all particle sizes (Sarnat *et al*, 2002). Sarnat *et al* (2000 and 2002) also demonstrated the importance of average air exchange rate for residences in an area and suggested that sulphur may be less suitable as a tracer in areas where there are colder winters and homes are more tightly sealed or where there are hotter summers and air conditioning is used. Elemental or black carbon has also been suggested for use as a tracer of ambient-generated PM if no combustion is allowed indoors (Wilson *et al*, 2000 and Ebelt *et al*, 2003).

Exposure to ambient generated particles is a very useful measure for those managing air quality in an airshed or investigating the health impacts of ambient sources. Although the importance of non-ambient sources is noted, it is important to separate these two exposures in order to fully understand their effects. A tool to predict average exposure to ambient generated particles in an airshed from ambient data is necessary so that expensive and time-consuming personal monitoring is not needed on an on-going basis.

In this chapter, the fraction of sulphate and elemental carbon in the total $PM_{2.5}$ at each temporary outdoor monitoring site is calculated and their average and median contribution to personal exposures is determined. Absorbance is used as a surrogate measure for elemental carbon and is representative of traffic-related $PM_{2.5}$ and residential wood burning. Sulphate and elemental carbon personal-ambient ratios are used to estimate the contribution of ambient-generated $PM_{2.5}$ to total personal exposures. The relationship between total exposure to $PM_{2.5}$ and both ambient and non-ambient sources is characterized. Equations are developed to enable use of past and future ambient data to estimate exposure to ambient generated sources. Data from the permanent Ministry of Water, Land and Air Protection $PM_{2.5}$ TEOM is assessed for representation of average population exposure to ambient-generated sources. The average infiltration factor and air exchange rates are calculated for each individual to enable better comparison with the literature.

4.3. Methods

Study design, methods for sampling and lab analysis have been described in Chapter 3. Least squares linear regression analysis, Pearson correlations (R), Spearman correlations (r) and all descriptive statistics were performed using Statistica 5.1 (StatSoft, Inc. 1997). Graphical output was generated using Microsoft Excel 2002.

4.3.1. Estimation of Ambient Generated Exposure

Using sulphate as a tracer of ambient sources, ambient generated exposures for each individual and every sampling session were estimated from the personal/ambient sulphate ratios and the ambient $PM_{2.5}$ concentrations measured at the nearest school roof. Sarnat *et al* (2000) suggests the use of the following equation to estimate exposure to ambient generated $PM_{2.5}$ (E_{ag}) using sulphate as a tracer:

(Equation 4.1)
$$E_{ag} = \left(\frac{\left[SO_4^{\ 2-}\right]_{Personal\,ij}}{\left[SO_4^{\ 2-}\right]_{Ambient\,jk}} \right) \bullet C_{a\,jk}$$

where $Personal\ ij$ represents the personal exposure to sulphate for subject i on day j and $Ambient\ jk$ represents the ambient sulphate concentration measured on day j at the closest neighbourhood monitoring site k. $C_{a\,jk}$ represents the ambient $PM_{2.5}$ concentration at the closest school site. The personal/ambient sulphate ratio may also be expressed as α_{SO_4}. Using this equation it is assumed that all $PM_{2.5}$ particles had an equal "effective penetration" of ambient $PM_{2.5}$ to personal exposures as sulphate. It is also assumed that the neighbourhood ambient monitor was representative of the trend in ambient levels near where the subject spends time. Least squares linear regression and the corresponding Pearson correlations were used to compare the ambient generated exposure estimates to ambient concentrations at the corresponding school of each subject and to the central MWALP $PM_{2.5}$ TEOM. Crude regression analysis was performed on the pooled data to enable a graphical display of the scatter in the relationship. In order to account for differences between individuals, longitudinal regressions were performed for each subject and the median slope, intercept, R^2 and associated errors are reported to provide a general regression equation. Lower and upper quartiles for the individual analyses were also reported to provide an indication of the distribution across individuals. Spearman correlations (r) were also determined for the data from each individual and the medians reported due to the non-parametric nature of the data. The results of all individual regressions and correlations are reported in Appendix 6.

Identical calculations were performed using elemental carbon (EC) ratios according to the following equation.

(Equation 4.2)
$$E_{ag} = \left(\frac{[EC]_{Personal\,ij}}{[EC]_{Ambient\,jk}} \right) \bullet C_{a\,jk}$$

The personal/ambient elemental carbon ratio may also be expressed as α_{EC}. Elemental carbon (EC) was determined indirectly from absorption coefficients (calculated from measurements of filter reflectance) using the relationship determined from co-located samples during the pilot study and described in detail in Chapter 2. In order to use elemental carbon as a tracer, we limit the analysis to those samples for which the personal levels were less than ambient concentrations. Personal exposure higher than the ambient concentration suggests that an indoor or personal source might have influenced the sample. The high personal samples could have also resulted from a very local ambient source that did not impact the ambient monitor such as car exhaust or fumigation of a nearby residential wood smoke plume but for the purposes of this analysis it was assumed that they were the result of non-ambient sources. In order to be used as a tracer for ambient $PM_{2.5}$, non-ambient sources of elemental carbon must be negligible. A total of 21 pairs (15%) of personal and ambient samples were removed from the analysis to ensure that non-ambient sources of elemental carbon did not influence the results. Samples where the difference between personal and ambient was within 0.05 and could be due sampling error were not removed. Identical statistical analyses were performed with elemental carbon as with sulphate. For subject 5001, only 2 samples could be used for estimating E_{ag}, therefore this subject was removed from all subsequent regression analyses due to sample size being too small.

4.3.2. Estimation of Non-Ambient Generated Exposure

Total personal exposure is composed of contributions from ambient and non-ambient sources. Non-ambient sources are generated indoors or from personal activity. Non-ambient generated exposures (E_{nag}) were estimated by simply subtracting the ambient exposure estimate (E_{ag}) from the total personal exposure (E_T) according to the following equation:

(Equation 4.3) $$E_{nag} = E_T - E_{ag}$$

This approach was also used by Ebelt *et al* (2003). The relationships between non-ambient and ambient exposure as well as the association of each of these components with total personal exposure were investigated using longitudinal least squares linear regression for each individual and both Pearson (R) and Spearman (r) correlations in an identical manner as described above. Individual analyses were performed with the median and lower and upper quartiles reported for the slope, intercept and R^2 for each subject. All individual results are summarized in Appendix 6. Pooled linear regressions and Pearson correlations were affected by two extreme values for subject 1002 in the total $PM_{2.5}$ exposures and the non-ambient exposure estimates. These two data points are described in Chapter 3 and were related to cooking. Analyses where these values affected the results were repeated with these points removed. If there was no effect they were included in the analysis, as was the case for the individual longitudinal analyses where the median and quartile range were reported.

4.3.3. Estimation of Infiltration and Air Exchange Rates

Sarnat et al (2000) showed that association between total personal $PM_{2.5}$ exposure and exposure to ambient generated PM varied by ventilation status with highly ventilated residences showing stronger correlations than those that were poorly ventilated. Air exchange rates can be measured for a residence and they can also be estimated from the sulphate and elemental carbon ratios (Ebelt et al, 2003). An estimation of an average air exchange rate was calculated for each subject in the study based on an estimate of the infiltration factor for each sampling day. The calculation for infiltration factor was derived from an equation provided by Wilson et al (2000), term 1, and the equation provided by Sarnat et al (2000), term 2, shown in more detail above in equation 4.1:

(Equation 4.4) $$E_{ag} = \overbrace{yC_a}^{\text{Term 1}} + \overset{\text{Term 2}\ \downarrow}{(1-y)C_a\frac{Pa}{a+k}} = \alpha_{SO_4}C_a$$

where y is the time spent outdoors or in a vehicle, C_a is the ambient concentration of $PM_{2.5}$, P is the penetration factor, a is the air exchange rate, k is the particle removal rate and both α_{SO_4} and α_{EC} represent the attenuation factor (personal/ambient ratio) for sulphate and elemental carbon respectively. Solving for the attenuation factor leads to the following equation:

(Equation 4.5) $$\alpha_{SO_4} = y + (1-y)\frac{Pa}{a+k}$$

According to Wilson et al (2000), the infiltration factor (F_{inf}) is equal to the following:

(Equation 4.6) $$F_{\text{inf}} = \frac{Pa}{a+k}$$

Therefore, the following equations can be used to estimate the infiltration factor:

(Equation 4.7) $$F_{\text{inf}} = \frac{\alpha_{SO_4} - y}{(1-y)}$$

By assuming values for P and k from the literature, equation 4.6 can then be used to solve for a and determine the air exchange rate on a given sampling day for each individual. P and k for sulphate were assumed to be 1.00 and 0.20 respectively based on data from the PTEAM study (Ozkaynak *et al*, 1996). The infiltration factors and air exchange rates were calculated based on sulphate estimates only and are summarized using the mean, standard deviation, median, inter-quartile range and range for each individual.

4.4. Results and Discussion

4.4.1. Personal Exposure and Ambient Concentration Relationship

For the regression of ambient concentration with personal exposure, the slope gives an indication of the average fraction of the ambient component that is found in personal exposure and the intercept gives the average sum of exposure to non-ambient sources (Wilson et al, 2000). The following regression equation summarizes the individual personal-ambient $PM_{2.5}$ regressions found in Table 3-11 using the median values and including the associated median error for each term from the regressions:

(Equation 4.8) $$E_T = 0.23(\pm 0.17) * C_a + 10.62(\pm 3.78), R^2 = 0.24$$

where E_T is the total personal exposure and C_a is the ambient concentration. Five of the fifteen subjects had significant p-values less than 0.05. This equation suggests that variation in the ambient data explains 24% of the variation in personal exposure and the median slope indicates that approximately 23 ± 17% of ambient levels results in personal exposure. The large intercept also indicates a significant offset suggesting personal exposure will always be greater independent of ambient concentration. The low R^2 and large error suggests that this equation might be unreliable. It is clear that a relationship between personal exposure and ambient concentration exists for $PM_{2.5}$ but it is masked by the large variability in non-ambient sources between individuals and even within individuals. A more accurate assessment of the contribution of ambient sources to total personal exposure is possible using a tracer of ambient $PM_{2.5}$ such as sulphate or elemental carbon that has negligible non-ambient sources.

4.4.2. Personal Ambient Ratios

For the pooled data set the median personal/ambient ratios for total $PM_{2.5}$, sulphate and elemental carbon are presented in Table 4-1.

Table 4-1 Summary statistics for the personal/ambient ratios of $PM_{2.5}$, sulphate (SO_4^{2-}) and elemental carbon (EC). Mean = arithmetic mean; Stdev = standard deviation. [a] EC ratios are also summarized for the reduced data set with samples removed when personal EC was greater than ambient EC.

Measurement	N	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
$PM_{2.5}$ Ratio	141	2.11	2.84	1.14	0.32	0.62	2.17	17.61
SO_4^{2-} Ratio	141	0.60	0.23	0.54	0.27	0.45	0.69	1.89
EC Ratio	141	0.90	0.86	0.59	0.13	0.47	0.89	5.07
EC Ratio [a]	120	0.60	0.26	0.55	0.13	0.42	0.73	1.65

The descriptive statistics for $PM_{2.5}$ ratios all clearly indicate that many of the personal exposures were highly influenced by non-ambient sources and there was large variability in this ratio throughout the study. Median values and an upper quartile less than one for both sulphate and elemental carbon suggest that there was a limited influence of non-ambient sources affecting the personal exposure levels for these measures. For sulphate the influence of non-ambient sources was almost non-existent and personal/ambient ratios were very consistent across individuals supporting the use of sulphate as a tracer of ambient $PM_{2.5}$. There were 7 samples where the personal/ambient ratio was greater than one but these samples were for relatively low concentrations and were very close to the origin of the linear relationship and on or below the 1:1 line. For elemental carbon there were several samples where a non-ambient source was likely. Looking at the raw data revealed that there were a small number of high peaks in elemental carbon personal exposure that exceeded the ambient counterpart for individuals at three schools (8 peaks in total – 1 at Lakewood, 3 at Westwood and 4 at Glenview). Five of these peak exposures were identified as being outliers and were more than 2 standard deviations away from the mean. It appears that if a personal source of elemental carbon is present (one not captured by the ambient monitor) it is often a relatively large exposure and easy to recognize. These large peaks were likely due to personal activity and would therefore violate the assumption of little or no non-ambient sources necessary to use elemental carbon as an indicator for ambient generated exposure. There were also several data points where personal exposure level was only slightly higher then the ambient sample (13 in total). These exposures only came from subjects at Lakewood (5) and Glenview (8). For these samples there may have been a smaller personal or indoor source playing a role. Analysis of the time activity diaries did not reveal any clear explanation for the high personal exposures. There was one individual that had higher elemental carbon personal exposures compared to the ambient level for 8 of 10 samples (3 high peaks) and this was the only individual that had a heating source besides natural gas, in this case a pellet stove. The pellet stove was not always running and it was not clear from the diary whether or not the stove was running on all days where a high exposure was observed for that individual. It was not expected that this type of stove would result in higher exposures. Each of the other 2 subjects at that school (Glenview) also had 2 personal exposures samples that were greater than the corresponding outdoor sample with only one of those 4 samples being a "large" exposure. In order to use elemental carbon as a tracer of ambient generated $PM_{2.5}$ exposure, these samples (15%) had to be removed before further analysis.

A crude linear regression of the pooled ambient concentrations and personal exposures for sulphate is shown in Figure 4-1 providing a general assessment of the scatter in this relationship.

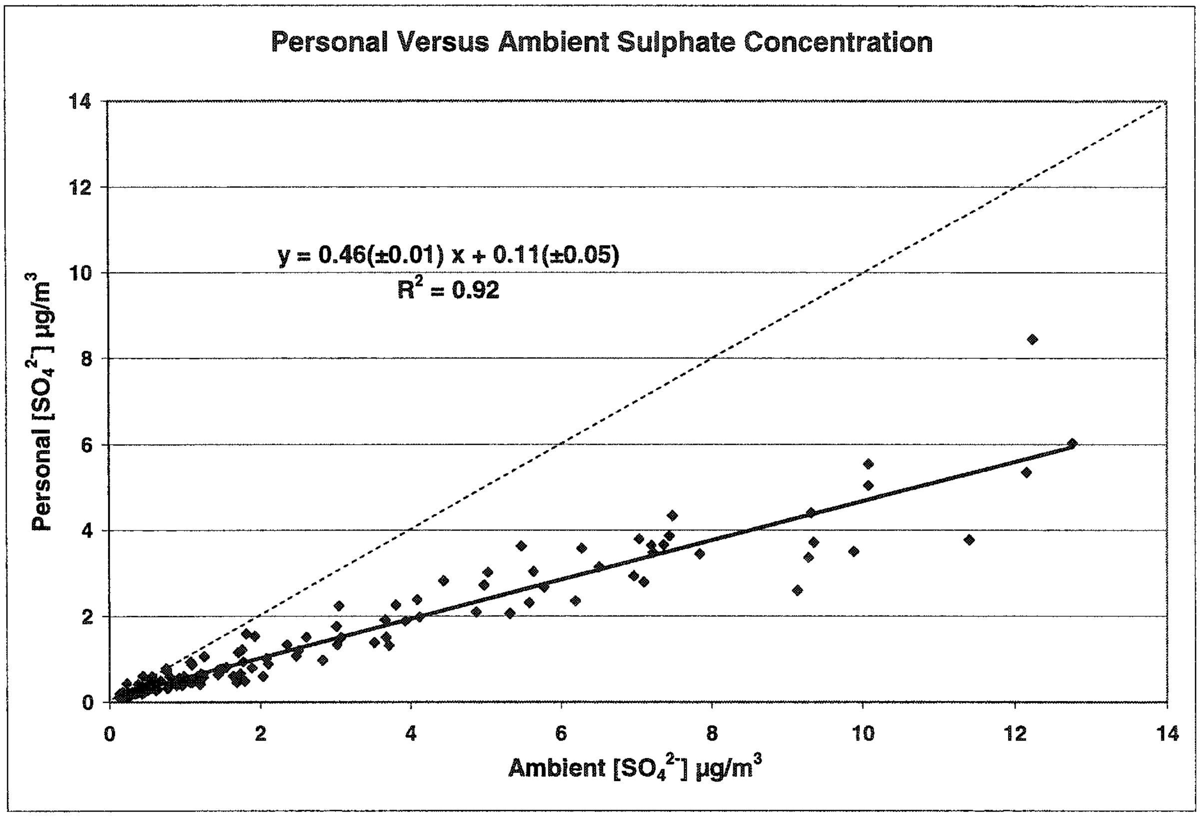


Figure 4-1 Least squares regression analysis of personal and ambient sulphate for the study period pooling data across schools and individuals (N=141, p<0.000).

Assuming that there are no indoor sources of sulphate and there is no actual removal of sulphate when infiltrating indoors, the regression equation between personal and ambient sulphate shows that on average personal exposure to sulphate is 46% of the ambient level. The limited amount of scatter around the regression line and an intercept very close to zero confirm the assumption that non-ambient sulphate is negligible. A similar analysis in Uniontown, PA in the summer reported an R^2 of 0.80 and a slope of 0.69

(Suh *et al*, 1992). Sarnat *et al* (2000) reported the crude R^2 of the pooled data and a slope estimated using mixed models for total personal sulphate exposure versus ambient sulphate. Results were provided for indoor environments that were poorly (slope=0.39, R^2=0.72), moderately (slope=0.40, R^2=0.73) and well ventilated (slope=1.16, R^2=0.88). Indoor ventilation conditions were categorized based on the distribution (mean ± standard deviation) of the fraction of time indoor environments had open windows. This study took place in Baltimore, MD over a summer and winter season and during the winter, ventilation category was assumed to be poor. Interestingly, the current study demonstrated a crude R^2 (0.92) higher than the well-ventilated environments and a slope (0.46) comparable to the moderately ventilated environments. A scripted exposure study in Boston, MA reported a much lower R^2 (0.62) and significantly higher slope (1.38) comparably to the well-ventilated environments in the Baltimore study (Brauer *et al*, 1989). The low regression coefficient was likely due to the low number of paired samples used in the analysis (N=27 samples). A summer study in Saint John, New Brunswick reported an R^2 of 0.79 for personal versus ambient sulphate based on the mean values for each individual (N=21 individuals) (Stieb *et al*, 1998). In the current study, a slightly higher R^2 of 0.83 was found from a similar analysis (N=15).

Figure 4-2 clearly shows that the relationship between personal and ambient elemental carbon was influenced by some non-ambient sources. A significant correlation exists but the relationship was poorer with increased scatter around the regression line.

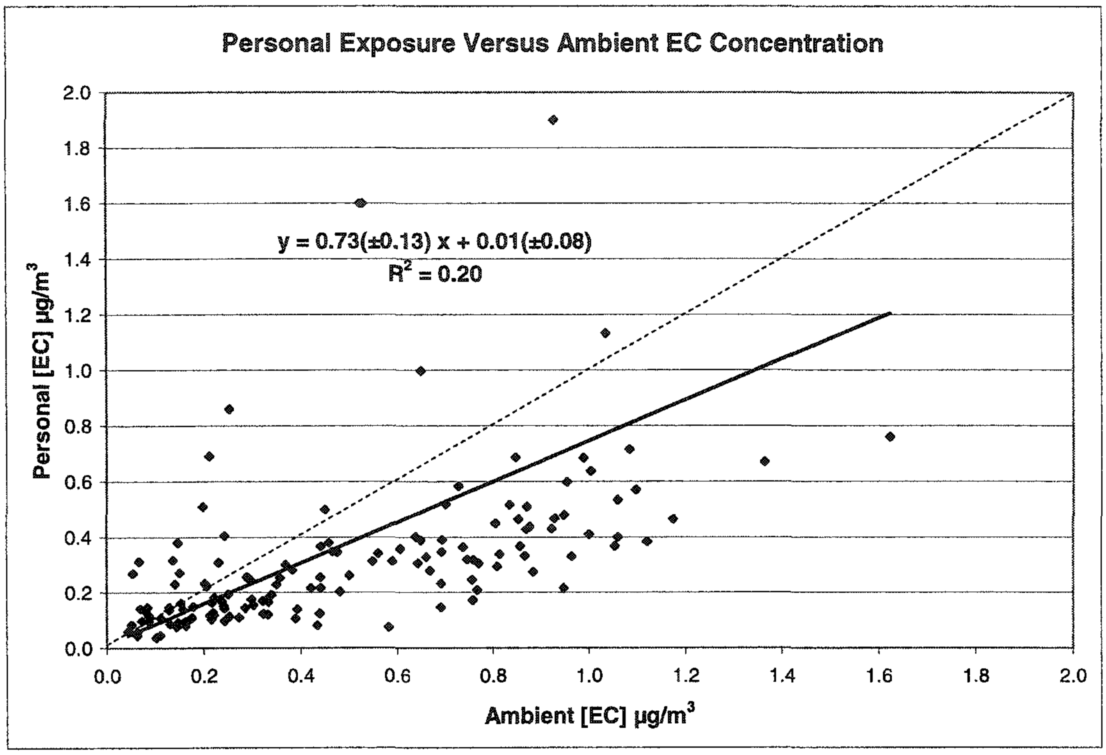


Figure 4-2 Least squares regression analysis of personal and ambient elemental carbon for the study period pooling data across schools and individuals (N=141, p<0.000).

Although the low intercept suggests that there were little non-ambient sources, the low R^2 value indicates that a large portion of the variance remains unexplained by ambient concentrations. All of the samples above the 1:1 line represent samples that are assumed to have been influenced by non-ambient sources and were therefore removed from the analysis as discussed previously. The following equation presents the crude linear regression equation from the pooled analysis of this subset of the data.

(Equation 4.9) $E_{EC} = 0.42(\pm 0.03) * C_{EC} + 0.05(\pm 0.02), R^2 = 0.71, p < 0.000$

E_{EC} is the total personal exposure to elemental carbon and C_{EC} is the ambient concentration of elemental carbon. The slope of the regression equation suggests that on average personal exposure to elemental carbon is 42% of the ambient level when very local and indoor sources are ignored. Because variation in exposure to local sources of elemental carbon does exist and can be very large and unpredictable this equation provides a more accurate tool for assessing average exposure across an airshed for all individuals. Two studies in Amsterdam and Helsinki in the Netherlands measured personal and ambient absorption coefficient as surrogates of elemental carbon over a six month period in the winter and spring (Janssen *et al*, 2000). These studies reported median slopes (R^2) of 0.92 (0.86) and 0.62 (0.66) in Amsterdam and Helsinki respectively for longitudinal individual analyses.

It is very reassuring that the fraction of ambient that results in personal exposure is almost identical for elemental carbon (0.42 ± 0.03) and sulphate (0.46 ± 0.01) when errors in the regressions are considered. This suggests that each of these measures is indeed a reliable tracer of ambient exposure, although to different components of the ambient $PM_{2.5}$ mixture. These slopes represent the average attenuation factor (α) of ambient concentrations to personal exposure across subjects for sulphate and elemental carbon.

4.4.3. Sulphate Based Estimates of Ambient Generated Exposure

Table 4-2 summarizes the descriptive statistics for the estimates of ambient generated exposure (E_{ag}) for the pooled data and for each individual based on the sulphate personal/ambient ratio.

Table 4-2 Summary statistics for personal exposure to ambient generated $PM_{2.5}$ (E_{ag}) derived using the personal/ambient sulphate ratio. Results are provided for the pooled data and for each individual. Values represent concentrations (µg/m^3). Mean = arithmetic mean; Stdev = standard deviation; IQR = inter-quartile range. Summary statistics across individuals are also provided.

E_{ag} (SO_4^{2-})	Valid N	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
Pooled Data	141	10	7	8	1	4	14	36
1001	10	16	9	14	5	12	17	36
1002	9	11	7	11	3	5	16	22
1003	10	10	9	6	3	4	16	29
2001	9	10	7	7	3	5	13	21
2002	10	6	4	5	2	2	10	13
2003	10	6	6	4	2	2	12	18
3001	10	10	6	9	1	4	18	19
3002	8	8	7	5	1	3	14	19
3003	9	9	6	8	2	2	14	19
4001	10	13	7	13	4	6	19	24
4002	7	7	4	4	3	3	12	14
4003	10	8	6	7	1	3	16	17
5001	10	11	9	9	2	6	16	30
5002	9	7	6	4	1	3	14	15
5003	10	8	9	4	2	3	11	31
Mean		9	7	7	2	4	15	22
Stdev		3	2	3	1	3	3	7
Median		9	7	7	2	3	14	19
IQR		3	2	4	1	2	4	9

The mean error associated with these estimates was calculated as 1.7±1.0 µg/m^3. The mean difference in co-located samples was used for the uncertainty of each measure used in the calculation. For ambient sulphate there were two large differences found in the co-located data that were more than 25 standard deviations from the mean. These extreme values, as well as a 3rd outlier (4 standard deviations from the mean), were excluded from the analysis in order to obtain a reasonable uncertainty estimate for ambient sulphate comparable to that obtained during the pilot study. Individuals were not sampled on the same days therefore ambient generated exposure levels are not expected to be identical and differences could result due the temporal change in ambient concentration. Some individuals could have experienced lower exposures to ambient generated $PM_{2.5}$ if they happened to be sampled on lower pollution days. Time activity patterns, including time spent outdoors would also play a crucial role in the level of exposure. Overall, levels of exposure to ambient generated $PM_{2.5}$ were consistent across individuals for the period especially when compared to the large variation in total $PM_{2.5}$ personal exposures reported in Chapter 3 (mean= 21±22 µg/m^3 for the pooled data). Ebelt *et al* (2003) reported a mean estimate and standard deviation of personal exposure to ambient $PM_{2.5}$ based on sulphate ratios of 7.9±3.7 µg/m^3 and a range of 0.9 to 21.3 for a pooled data set of COPD diseased patients in Vancouver, BC during the summer. The current study showed a higher estimate of 10±7 µg/m^3 as well as greater range of 1 to 36 µg/m^3. The corresponding total $PM_{2.5}$ personal exposures for the Vancouver study had a mean of 18.5±14.9 µg/m^3 and a range of 2.2 to 90.9 µg/m^3. In Prince George, the mean (18±13 µg/m^3) and range (3 to 87 µg/m^3) were similar when two extreme values (discussed in Chapter 3) were removed from the data set for subject 1002. Higher personal exposures to ambient were likely the result of higher ambient concentrations in Prince George with a mean concentration of 18±15 µg/m^3 and a range of 1 to 61µg/m^3, although difference in activity patterns between diseased adults in the summer and healthy children in the winter could have also played a role. In Vancouver, the mean ambient concentration was 11.4 ±4.6 μg/m^3 and the range was 4.2 to 28.7μg/m^3.

4.4.4. EC Based Estimates of Ambient Generated Exposure

Table 4-3 summarizes the descriptive statistics for the estimates of ambient generated exposure (E_{ag}) derived from elemental carbon ratios for the pooled data and for each individual.

Table 4-3 Summary statistics for personal exposure to ambient generated $PM_{2.5}$ (E_{ag}) derived using the personal/ambient elemental carbon ratio. Results are provided for the pooled data and for each individual. Values represent concentrations (μg/m^3). Mean = arithmetic mean; Stdev = standard deviation; IQR = inter-quartile range. Summary statistics across individuals are also provided.

E_{ag} (EC)	Valid N	# of Samples Removed	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
Pooled Data	120	21	10	7	7	1	4	13	33
1001	10	0	13	7	12	6	7	14	27
1002	9	0	13	9	12	2	7	14	31
1003	10	0	10	9	6	3	4	13	30
2001	9	0	11	7	8	4	5	13	24
2002	10	0	8	5	7	2	4	13	19
2003	10	0	7	6	5	1	2	12	20
3001	7	3	12	7	9	3	7	19	24
3002	7	1	12	10	8	3	3	21	27
3003	7	2	9	6	7	2	2	15	16
4001	9	1	11	4	10	6	9	13	19
4002	7	0	5	2	5	2	3	7	8
4003	8	2	11	10	6	2	4	20	27
5001	2	8	12	0	12	12	12	12	12
5002	7	2	9	6	6	4	6	18	18
5003	8	2	9	10	4	2	4	10	33
Mean			10	7	8	3	5	14	22
Stdev			2	3	3	3	3	4	7
Median			11	7	7	3	4	13	22
IQR			3	4	4	2	3	4	9

The mean error associated with these estimates was calculated as 2.2±1.2 µg/m^3 using the mean difference in co-located samples as the uncertainty for each measure used in the calculation. The descriptive statistics for the elemental carbon derived estimate of ambient generated exposure are almost identical to those found with the sulphate based estimate with a median range across individuals of 3 to 22 µg/m^3 compared to 2 to 19 µg/m^3 for sulphate. The difference in medians for the pooled data and the average of median values across individuals was only 1 µg/m^3.

4.4.5. Ambient Generated Exposure versus Ambient Concentrations

A crude least squares regression of the pooled data between ambient generated exposure (based on the sulphate) and ambient concentration is shown in Figure 4-3 as a means of displaying the general relationship and scatter between these variables.

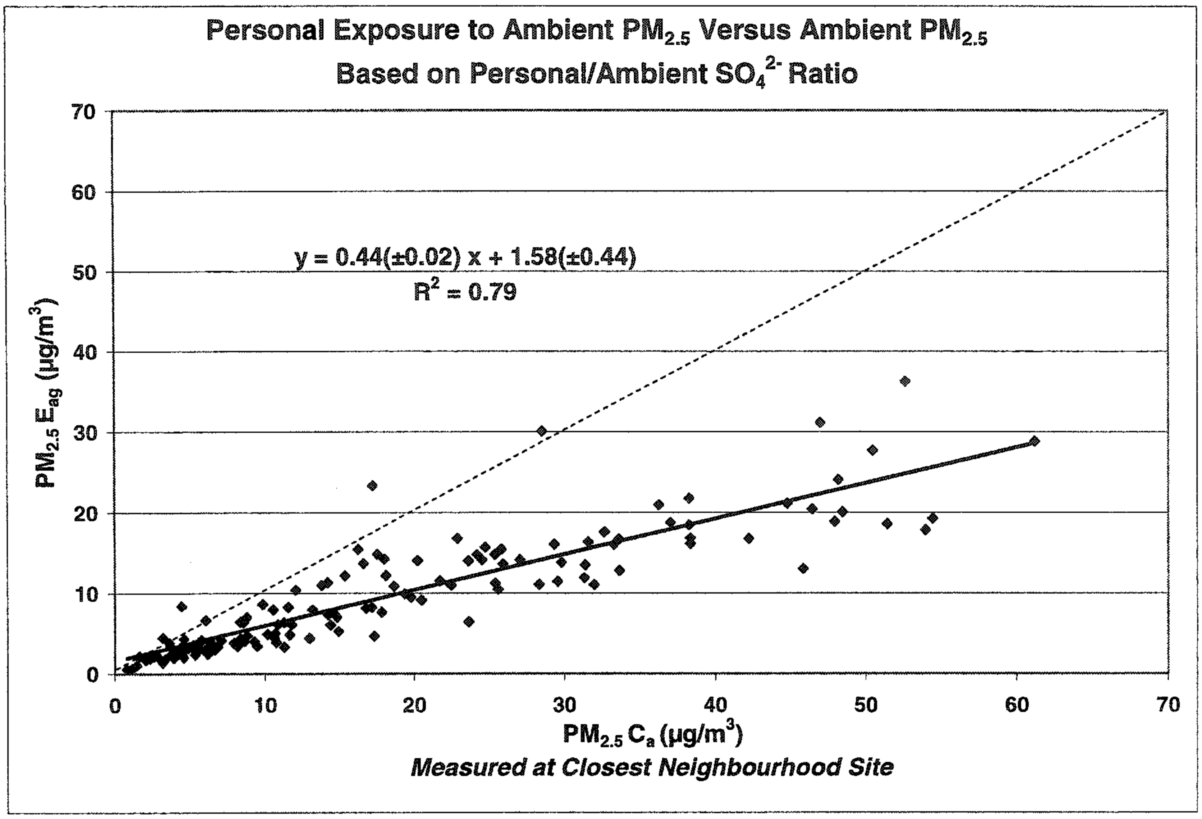


Figure 4-3 Crude least squares regression analysis of personal exposure to ambient generated $PM_{2.5}$ (E_{ag}) and ambient concentration (C_a) measured at the closest outdoor site for the pooled data. E_{ag} is calculated using the SO_4^{2-} personal/ambient ratio as in equation 4.1 and the same ambient data that is used for C_a on the x-axis (N=141 and p<0.000).

The limited number of data points located above the 1:1 line support the assumption of negligible non-ambient sulphate sources. The low scatter and high correlation even when the data is pooled across individuals provides evidence of a strong relationship between ambient generated exposure and actualy ambient concentrations at the closest neighbourhood site. In order to account for the large variability between individuals, the longitudinal regressions were performed for each subject in the study and the median values and errors from the regression equations are shown in the following equation:

(Equation 4.10) $E_{ag} = 0.45(\pm 0.05) * C_a + 1.56(\pm 1.15), R^2 = 0.90, p \le 0.011$

where E_{ag} is exposure to ambient generated $PM_{2.5}$ and C_a is ambient concentration at each student's school. The individual median (range) Pearson correlation was 0.95 (0.76 to 0.99). Similar Spearman correlations were found with a median (range) of 0.92 (0.82 to 0.99). Wilson *et al* (2000) performed a reanalysis of data from the PTEAM study and reported slopes (R^2) for the relationship between E_{ag} and C_a of 0.62 (0. 74) and 0.52 (0.51) using backyard concentrations and ambient data from a central site respectively (E_{ag} was calculated differently by Wilson using measured air exchange rates and time activity information). The larger slope of 0.62 in the analysis by Wilson compared to 0.45 in this study could be related to the difference in season and warmer climate, resulting in more time outdoors and with windows open during the fall in Riverside California. The much stronger correlation in the current study (R^2=0.90) may partly be the result of using a different method to calculate E_{ag} and possible over-estimation of the strength in the association due to the use of the same ambient concentrations in the calculation of E_{ag} and in the regression. However, the high correlation is more likely due to the strong relationship between personal and ambient sulphate that was consistent between and within individuals.

The purpose of describing this relationship is so that both future and past ambient data can be used to generate average exposure information for the study population. In order to understand exposure throughout the entire year and for different sub-populations such as working adults or senior citizens, this relationship should also be investigated in each season and for each different group. Seasonal differences are expected and variables such as increased ventilation and time outdoors in the summertime are expected to change the relationship. To provide a more general and applicable result, analysis was also performed using data from the MWLAP $PM_{2.5}$ TEOM monitor and each individual's estimated exposure to ambient generated $PM_{2.5}$ sources. The resulting equation reporting median values from the individual longitudinal regressions was:

(Equation 4.11) $$E_{ag} = 0.45(\pm 0.08) * C_a + 1.80(\pm 1.63), R^2 = 0.81, p \leq 0.014$$

The range of Pearson correlations found across 14 of the 15 study subjects was 0.77 to 0.98. Significant p-values were found for all individuals except for subject 5001 (p=0.205) whose regression equation had a very high intercept (5.81) and low correlation (R=0.50). A corresponding strong median Spearman correlation of 0.88 (0.64 to 1.00) was found to be significant for all of the study subjects. Equation 4.6 using the ambient data on the student's school roof, and Equation 4.7 are very similar suggesting that the current WLAP $PM_{2.5}$ TEOM is suitable for assessing average exposure throughout the city.

A comparison of elemental carbon based estimates of ambient generated exposure to ambient concentration yielded very similar results to those found for the sulphate based estimates. The crude least squares regression of the pooled data is shown in Figure 4-4 to display the general relationship and demonstrate the scatter in the data. The individual regression results are summarized in Table 4-4 for both elemental carbon and sulphate based comparisons of ambient generated exposure and ambient concentration.

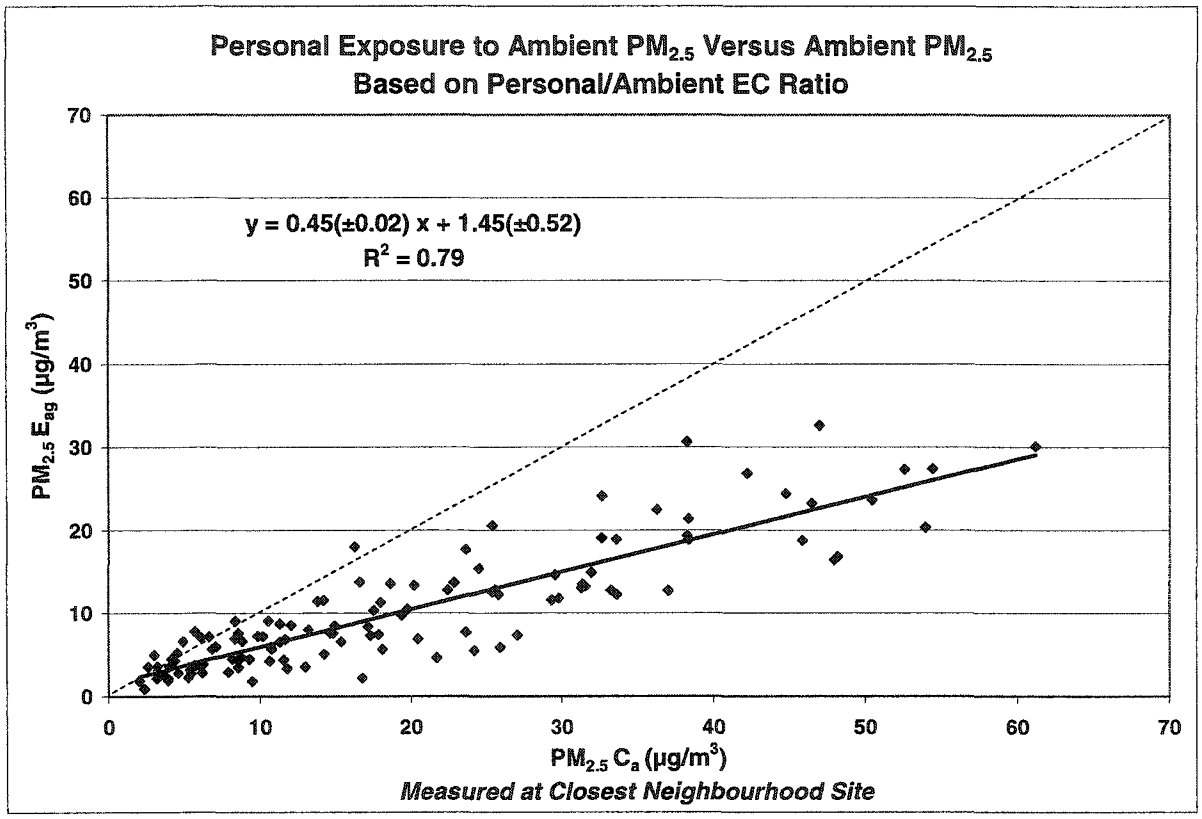


Figure 4-4 Crude least squares regression analysis of personal exposure to ambient generated $PM_{2.5}$ (E_{ag}) and ambient concentration (C_a) measured at the closest outdoor site for the pooled data. E_{ag} is calculated using the EC personal/ambient ratio as in equation 4.2 and the same ambient data that is used for C_a on the x-axis (N=120 and p<0.000).

Table 4-4 Summary statistics for individual least squares regression of ambient generated exposure (E_{ag}) versus ambient concentration measures at the closest school sites and the central MWLAP TEOM. Median, lower quartile and upper quartile of the individual results are shown. All individual results can be found in Appendix 6.

	Results Based on SO_4^{2-}			Results Based on EC		
	Slope	**Intercept**	**R^2**	**Slope**	**Intercept**	**R^2**
E_{ag} vs C_a						
Median	0.45(±0.05)	1.56(±1.15)	0.90	0.46(±0.07)	1.39(±1.76)	0.89
Lower Quartile	0.38(±0.04)	0.65(±0.80)	0.87	0.37(±0.05)	0.32(±1.33)	0.79
Upper Quartile	0.50(±0.07)	2.10(±1.49)	0.94	0.54(±0.10)	2.05(±2.38)	0.93
E_{ag} vs TEOM						
Median	0.45(±0.08)	1.80(±1.63)	0.81	0.50(±0.09)	1.34(±1.88)	0.86
Lower Quartile	0.36(±0.06)	0.85(±1.12)	0.75	0.40(±0.07)	0.93(±1.73)	0.72
Upper Quartile	0.57(±0.10)	3.12(±1.88)	0.92	0.67(±0.14)	2.94(±2.46)	0.87

4.4.6. Ambient Versus Non-ambient Exposure

Table 4-5 and Table 4-6 summarize the pooled and individual descriptive statistics for the estimates of non-ambient generated exposure (E_{nag}) based on the sulphate and elemental carbon personal/ambient ratios respectively.

Table 4-5 Summary statistics for personal exposure to non-ambient generated PM$_{2.5}$ (E_{nag}) obtained from subtracting estimates of ambient generated exposure (derived from SO_4^{2-} ratio) from total personal exposure. Results are provided for the pooled data and for each individual. Values represent concentrations (μg/m^3). Mean = arithmetic mean; Stdev = standard deviation; IQR=inter-quartile range. Summary statistics across individuals are also provided.

E_{nag} (SO_4^{2-})	Valid N	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
Pooled Data	141	11	21	6	-10	3	12	174
1001	10	2	4	0	-2	-2	5	8
1002	9	57	59	28	6	24	55	174
1003	10	7	9	5	-4	1	7	27
2001	9	9	5	9	0	5	12	15
2002	10	14	16	8	2	6	21	55
2003	10	7	4	6	1	4	8	15
3001	10	16	25	6	0	3	16	79
3002	8	4	3	4	1	2	6	10
3003	9	1	5	1	-4	-3	4	10
4001	10	10	16	4	-3	-1	19	42
4002	7	5	4	5	1	1	9	12
4003	10	11	7	10	2	4	18	22
5001	10	9	8	10	-4	5	12	24
5002	9	10	6	9	3	7	12	19
5003	10	5	6	6	-10	2	7	14
Mean		11	12	7	-1	4	14	35
Stdev		13	14	6	4	6	13	43
Median		9	6	6	0	3	12	19
IQR		5	8	4	5	4	10	22

Table 4-6 Summary statistics for personal exposure to non-ambient generated $PM_{2.5}$ (E_{nag}) obtained from subtracting estimates of ambient generated exposure (derived from EC ratio) from total personal exposure. Results are provided for the pooled data and for each individual. Values represent concentrations (µg/m^3). Mean = arithmetic mean; Stdev = standard deviation. Summary statistics across individuals are also provided including.

E_{nag} (EC)	Valid N	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
Pooled Data	120	11	23	5	-11	2	11	172
1001	10	5	4	4	0	2	10	11
1002	9	56	59	31	4	22	56	172
1003	10	7	9	5	-5	4	8	26
2001	9	8	6	9	0	4	12	17
2002	10	13	16	7	1	2	16	54
2003	10	6	5	5	-2	3	9	15
3001	7	13	29	3	-4	-2	8	79
3002	7	1	4	1	-7	0	3	6
3003	7	3	4	1	-1	-1	5	10
4001	9	12	17	5	-3	1	18	47
4002	7	7	4	8	1	4	12	12
4003	8	8	9	6	-2	2	15	22
5001	2	6	1	6	5	5	6	6
5002	7	8	5	7	3	3	13	16
5003	8	3	8	4	-11	0	7	14
Mean		10	12	7	-1	3	13	34
Stdev		13	15	7	4	5	13	43
Median		7	6	5	-1	2	10	16
IQR		5	8	3	4	3	6	25

Due to the residual nature of the estimate for non-ambient exposure, the associated error is unknown and includes the error associated with both the ambient and non-ambient terms. This explains the presence of negative values in the individual summaries. The sulphate and elemental carbon based estimates agree very well with differences of only 1 to 4 µg/m^3 for the pooled data and individual summary statistics. Differences in the statistics for each individual showed slightly more variation with a range in differences of 1 to 9 µg/m^3. Ebelt *et al* (2003) reported a mean estimate of exposure to non-ambient $PM_{2.5}$ based on sulphate of 10.6±14.5 µg/m^3 and the range was -2.6 to 85µg/m^3 for the pooled data sampled from diseased adults in Vancouver, BC during the summer. Comparison to the current study showed similar results when the 2 extreme data points for subject 1002 were removed, with a mean non-ambient exposure of 9±12 μg/m^3 and a range of -10 to 79 μg/m^3 for the pooled data.

Box plots of the pooled data in Figure 4-5 show the overall similarity between the non-ambient exposure estimates for the two tracer species and the relative difference between these estimates and the corresponding estimates of ambient generated exposure.

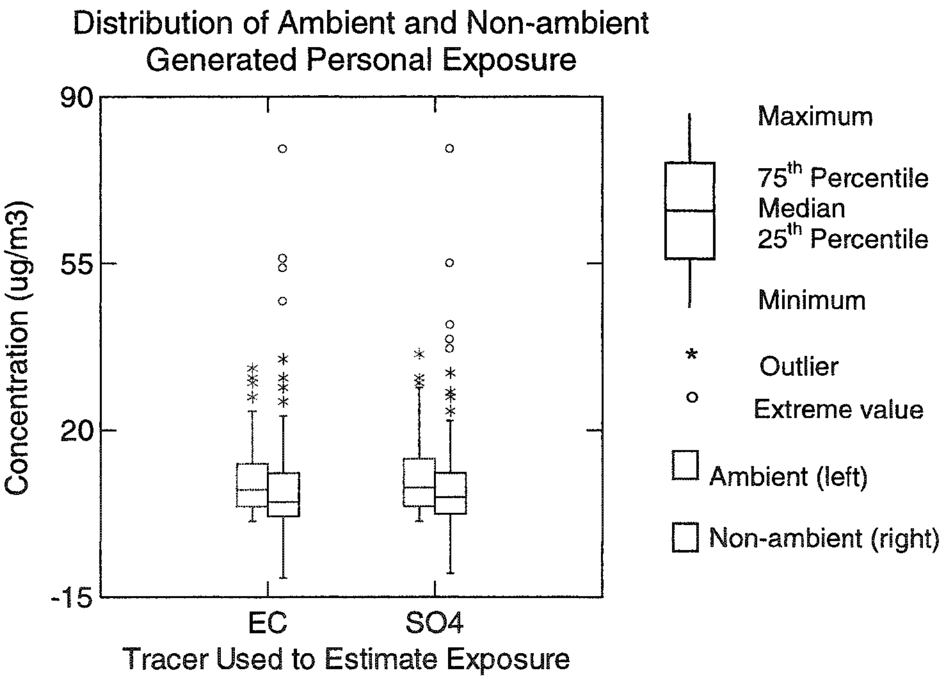


Figure 4-5 Box plots for PM$_{2.5}$ ambient and non-ambient generated exposure based on the sulphate and elemental carbon estimates. Two extreme outliers for each of the non-ambient exposure are not shown as they were outside of the chosen scale. Median, inter-quartile range, range, outliers and extreme outliers are marked by the plot.

Distribution of the pooled estimates for both non-ambient and ambient generated exposure show almost identical results for each of the tracer species. Interestingly, the ambient exposure estimates were slightly greater than the non-ambient estimates overall but with a smaller range and less influence of outliers and extreme values.

Spearman correlations were performed to assess the relationship between total personal $PM_{2.5}$ personal exposure and both the ambient and non-ambient components. For the pooled estimates based on sulphate and elemental carbon respectively, total exposure showed moderate correlation with ambient generated exposure (r=0.57 and r=0.55) and a slightly lower correlation with non-ambient generated exposure (r=0.49 and r=0.50). A much higher correlation between total exposure and the non-ambient component (r=0.84) was found by Ebelt *et al* (2003) and a lower correlation was found with the ambient component (r=0.41). This difference is likely due to the higher ambient concentrations in Prince George and therefore greater outdoor influence, although slightly lower non-ambient exposures may have also had an effect. Individual longitudinal correlations were also determined for each subject in the current study and are summarized with the pooled correlations in Table 4-7 using both Spearman and Pearson statistics.

Table 4-7 Summary of Spearman (r) and Pearson (R) correlations for the pooled data and individual analyses of total personal exposure (E_T) versus ambient generated exposure (E_{ag}) and non-ambient exposure (E_{nag}). Median, quartile range and range of the individual results are shown. All individual results can be found in Appendix 6. (*$p \leq 0.000$)

	Results Based on SO_4^{2-}	Results Based on SO_4^{2-}	Results Based on EC	Results Based on EC
Correlation Type	*Spearman*	*Pearson*	*Spearman*	*Pearson*
Pooled Data				
E_{ag} vs E_T	0.57*	0.21	0.55*	0.19
E_{nag} vs E_T	0.49*	0.94	0.50*	0.95
E_{ag} vs E_T				
Median	0.60	0.62	0.43	0.64
Quartile Range	0.48 to 0.72	0.36 to 0.73	0.22 to 0.64	0.20 to 0.75
Range	0.18 to 0.80	-0.35 to 0.92	0.00 to 0.86	-0.25 to 0.95
E_{nag} vs E_T				
Median	0.40	0.52	0.46	0.63
Quartile Range	0.17 to 0.60	0.19 to 0.80	0.04 to 0.61	0.25 to 0.92
Range	-0.20 to 0.88	-0.31 to 99	-0.64 to 0.93	-0.63 to 0.99

Pearson correlations for the pooled data set were very different from the Spearman correlations showing a very strong correlation between total exposure and the non-ambient component (R=0.94 and R=0.84) and much poorer correlation between total exposure and the ambient component (R=0.21 and R=0.43); R values with two extreme outliers in the analysis and then removed respectively. The sulphate and elemental carbon estimates yielded almost identical results. The median individual longitudinal Pearson correlations were closer to the Spearman correlations compared to the pooled data, although there were still some differences. The Pearson correlations appear to overestimate the association between non-ambient exposure and total exposure for both sulphate and elemental carbon tracers. For the association between the ambient estimate and total exposure, the Pearson and Spearman correlations compare well for sulphate while the elemental carbon tracer still shows a larger Pearson correlation. Due to the skewed nature of the data the Spearman correlations are more accurate and this comparison provides a point of reference for longitudinal regression analyses between these variables.

Table 4-8 summarizes the relative contribution that ambient and non-ambient generated exposures made to total $PM_{2.5}$ personal exposure during the study period by showing the median and quartile range in the slope, intercept and R^2 for individual longitudinal regressions. All individual results can be found in Appendix 6.

Table 4-8 Summary statistics for individual least squares regression of total personal exposure (E_T) versus ambient generated exposure (E_{ag}) and non-ambient exposure (E_{nag}). E_{nag} Median, lower and upper quartile of the individual results are shown. All individual results can be found in Appendix 6. Intercepts were mostly not significantly different than zero across individuals.

	Results Based on SO_4^{2-}			Results Based on EC		
	Slope	**Intercept**	**R^2**	**Slope**	**Intercept**	**R^2**
E_{ag} vs E_T						
Median	0.52(±0.20)	0.55(±4.08)	0.39	0.59(±0.20)	-0.49(±3.91)	0.41
Lower Quartile	0.22(±0.16)	-2.31(±3.24)	0.16	0.08(±0.12)	-1.62(±3.03)	0.05
Upper Quartile	0.82(±0.34)	3.29(±4.94)	0.54	0.75(±0.30)	5.83(±4.94)	0.56
E_{nag} vs E_T						
Median	0.48(±0.20)	-0.55(±4.08)	0.27	0.41(±0.20)	0.49(±3.91)	0.43
Lower Quartile	0.18(±0.16)	-3.29(±3.24)	0.07	0.25(±0.12)	-5.83(±3.03)	0.14
Upper Quartile	0.78(±0.34)	2.31(±4.94)	0.64	0.92(±0.30)	1.62(±4.94)	0.85

The median results suggest that the variation in non-ambient sources explains 27% or 43% of the variation in total personal exposure compared to the 39% or 41% explained by the ambient sources when sulphate and elemental carbon based estimates are used respectively. These values do not add up to 100% due to the error associated with each of the estimates and the large variability between individuals (range in $R^2 = 0.01$ to 0.99). The number of subjects that showed a significant correlation between total exposure and the ambient component was 7 for the sulphate based estimate and 5 for the elemental carbon estimate. For the regression between the non-ambient component and total exposure the sulphate estimate resulted in 4 individuals with a significant correlation and 6 for the elemental carbon estimate. The extreme variability in the correlations is mainly the result of low sample size, with the number of samples for each subject ranging from 7 to 10 samples. It is difficult to obtain more samples per individual for this type of intensive personal monitoring therefore the median results across individuals more accurately represents everyday exposure. The elemental carbon based estimates showed slightly higher R^2 values for regression between non-ambient and total $PM_{2.5}$ personal exposure. This was likely due to the removal of outliers in the measured total exposure with the elemental carbon comparison and not the sulphate comparison. Because some data removal was necessary due to the presence of non-ambient elemental carbon sources, the estimates based on sulphate may be more reliable. However, the similarity in the median results from each tracer species increases the validity of the method used to estimate both the ambient and non-ambient generated exposures. Some differences did result for the two tracer species between individuals, which can be seen from the lower and upper quartile summaries. For the non-ambient generated exposure estimate based on elemental carbon, the correlation and the slopes appear higher than the sulphate based estimate. Although the difference is notable, it is still within the respective error terms.

Comparison of the slopes from the median regression results between total personal exposure and ambient and non-ambient generated exposure yielded interesting results. Both ambient and non-ambient generated exposures contribute equal proportions to total personal exposure. It was unexpected that neither of the source types would have a greater influence on the level of total personal exposure. Due to the large variation in total personal exposure due to peal exposures it was expected that non-ambient sources were playing the dominant role. But the large variation due to the non-ambient sources is mainly the result of the rare occurrence of very large peaks that does not generally represent everyday exposure of the general public. The median and mean results from the individual regression showed very little difference, confirming that the large peaks are uncommon and do not have much influence on overall exposure. It is unknown whether these short-term peaks in exposure have significant health effects.

A main criticism of health findings in the past has been that ambient concentrations used as exposure surrogates in epidemiological studies do not truly represent personal exposures and exposures may therefore be misclassified. It is important to demonstrate that non-ambient exposures are not well correlated with ambient concentrations or with ambient exposure;

therefore, any health impacts found to be associated with ambient concentrations or exposures can not be confounded by or be the result of non-ambient exposure. For this study, the pooled estimate for non-ambient generated exposure had a poor Spearman correlation with both the estimate of ambient generated exposure (r=-0.31) and ambient concentration (r=-0.33) based on sulphate. The pooled Pearson correlations were lower (R=-0.13). The elemental carbon results were almost identical. For the individual longitudinal correlations, the median Spearman correlations were slightly higher for sulphate (r=-0.44 and r=-0.36) and lower for elemental carbon (r=-0.26 and r=-0.28). An association significantly different from zero ($p \leq 0.05$) was found for 3 individuals (1 using the sulphate tracer and 2 with the EC tracer) for the correlation between non-ambient exposure and ambient concentration. For the correlation between non-ambient and ambient exposure, 3 individuals showed a significant association using each of the tracers but only 1 of these individuals showed the significant association using both tracers. The median individual Pearson correlations for both the sulphate and elemental carbon tracers were similar to the Spearman correlations found with sulphate. These results suggest that there may have been a very weak association between the non-ambient generated exposure and both ambient generated exposure and ambient concentration. This is likely the result of the non-ambient estimate being the residual from subtracting ambient exposure from the total exposure. The fact that 1 to 3 individuals did show significant correlations is most likely a result of the limited number of measurements per individual and the 5% chance of finding a significant result when one doesn't exist .

4.4.7. Air Exchange Rates and Infiltration Factors

The descriptive statistics for the calculated infiltration factors and air exchange rates based on the personal/ambient sulphate ratio for each individual are shown in Table 4-9 and Table 4-10 respectively.

Table 4-9 Descriptive statistics for the infiltration factors (F_{inf}) for each subject in the study. Calculated using personal/ambient sulphate ratios. F_{inf} has no units. Mean = arithmetic mean; Stdev = standard deviation; IQR=inter-quartile range. Summary statistics across individuals are also provided.

	Valid N	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
1001	9	0.67	0.15	0.69	0.47	0.54	0.78	0.88
1002	9	0.59	0.17	0.56	0.40	0.45	0.68	0.91
1003	10	0.54	0.12	0.50	0.44	0.45	0.55	0.80
2001	9	0.46	0.14	0.44	0.22	0.42	0.48	0.77
2002	10	0.48	0.17	0.44	0.26	0.38	0.55	0.90
2003	9	0.49	0.22	0.45	0.22	0.32	0.67	0.80
3001	10	0.53	0.13	0.52	0.33	0.46	0.60	0.74
3002	8	0.49	0.08	0.51	0.34	0.45	0.55	0.57
3003	9	0.49	0.17	0.48	0.26	0.38	0.59	0.83
4001	10	0.57	0.22	0.47	0.28	0.42	0.79	0.85
4002	6	0.48	0.09	0.49	0.33	0.46	0.51	0.62
4003	10	0.40	0.14	0.40	0.17	0.37	0.41	0.71
5001	9	0.53	0.19	0.50	0.28	0.41	0.62	0.87
5002	8	0.63	0.15	0.58	0.48	0.56	0.66	0.95
5003	10	0.55	0.22	0.57	0.15	0.42	0.71	0.94
Median		**0.53**	**0.15**	**0.50**	**0.28**	**0.42**	**0.60**	**0.83**
Stdev		**0.07**	**0.04**	**0.07**	**0.10**	**0.06**	**0.11**	**0.11**
Mean		**0.53**	**0.16**	**0.51**	**0.31**	**0.43**	**0.61**	**0.81**
IQR		**0.07**	**0.05**	**0.08**	**0.13**	**0.06**	**0.13**	**0.13**

Table 4-10 Descriptive statistics for air exchange rates (a) for each subject in the study. Calculated using personal/ambient sulphate ratios. All values have units of h^{-1}. Mean = arithmetic mean; Stdev = standard deviation; IQR=inter-quartile range. Summary statistics across individuals are also provided.

	Valid N	Mean	Stdev	Median	Minimum	Lower Quartile	Upper Quartile	Maximum
1001	9	0.56	0.41	0.44	0.18	0.24	0.69	1.42
1002	9	0.47	0.61	0.25	0.13	0.16	0.43	2.06
1003	10	0.28	0.20	0.20	0.16	0.16	0.25	0.78
2001	9	0.20	0.17	0.16	0.06	0.14	0.19	0.66
2002	10	0.32	0.51	0.16	0.07	0.12	0.24	1.76
2003	9	0.30	0.30	0.16	0.06	0.09	0.40	0.81
3001	10	0.27	0.15	0.22	0.10	0.17	0.30	0.58
3002	8	0.20	0.06	0.21	0.10	0.16	0.25	0.27
3003	9	0.27	0.28	0.18	0.07	0.12	0.28	0.97
4001	10	0.44	0.43	0.18	0.08	0.14	0.73	1.17
4002	6	0.20	0.08	0.19	0.10	0.17	0.21	0.33
4003	10	0.16	0.12	0.13	0.04	0.12	0.14	0.49
5001	9	0.36	0.40	0.20	0.08	0.14	0.32	1.32
5002	8	0.73	1.25	0.28	0.18	0.25	0.40	3.81
5003	10	0.55	0.92	0.26	0.04	0.15	0.48	3.13
Median		**0.30**	**0.30**	**0.20**	**0.08**	**0.15**	**0.30**	**0.97**
Stdev		**0.16**	**0.33**	**0.07**	**0.05**	**0.04**	**0.17**	**1.02**
Mean		**0.35**	**0.39**	**0.21**	**0.10**	**0.16**	**0.36**	**1.30**
IQR		**0.22**	**0.30**	**0.07**	**0.05**	**0.04**	**0.17**	**0.97**

For this study the mean infiltration factor for individuals was 0.53±0.16 with the error term being the mean standard deviation reported across individuals. This value is consistent with the F_{inf} obtained during a Seattle study for the heating season (Allen *et al*, 2003). The Seattle study involved very different methods including the use of continuous light scattering data to measure PM as well as mass concentrations and a newly adapted recursive model to model ambient generated PM entering the indoor environment. The modeled infiltration factors determined in Seattle over multiple seasons were 0.65±0.21 based on light scattering data and 0.75 for sulphate. Other studies have reported mean infiltration factors generally over multiple seasons or during the summer ranging from 0.66 to 0.74 for $PM_{2.5}$ and 0.74 to 0.86 for sulphate

(Allen *et al*, 2003). The lower value modeled in the current study using the sulphate ratio is expected due to sampling in the winter only.

The mean air exchange rate across individuals was 0.35±0.39 (h^{-1}). This also compares well to the Seattle study where the mean air exchange rate was reported to be 0.37±0.17 (h^{-1}) for the heating season but with a significantly higher standard deviation. During the Seattle study participants had at least one window open during the heating season 42.1± 38.5% of the time. During this study, there were rarely any windows as it would have been much colder in Prince George than it was in Seattle. The mean temperature during the current study was -4±7°C with a range of -20 to 13°C. As a result of the colder weather and closed windows almost all the time, one would expect to find lower air exchange rates than in Seattle. The distribution of raw air exchange rates was highly skewed and box plots for each individual identified nine outliers in the data. Five of these outliers were more than 2 standard deviations from the mean of the pooled data. This suggests that the median values may provide a more accurate estimate for the air exchange rate for each individual and for all subjects. The median air exchange rate from the individual analyses was 0.20 with a quartile range of 0.15 (h^{-1}).

4.5. Conclusion

Ambient $PM_{2.5}$ shows a very poor relationship with total $PM_{2.5}$ personal exposure and the relationship appears to be masked by the large variability in non-ambient sources between individuals and even within individuals. Using sulphate and elemental carbon as tracers of ambient $PM_{2.5}$ to estimate exposure to ambient sources provided a more accurate assessment of the actually contribution of ambient sources to total personal exposure.

Personal/ambient ratios for sulphate and elemental carbon based on ambient measurements at each school suggested there was a limited influence of non-ambient sources. For elemental carbon, the influence was greater and resulted in the removal of 21 samples (15%) that were suspected to have a significant non-ambient component in the personal exposure measure. Linear regression between the personal and ambient components for each measure confirmed that for sulphate and elemental carbon, when the samples with larger personal/ambient ratios were removed, non-ambient influence was minimal and a strong association existed. The sulphate and elemental carbon regressions between personal and ambient levels showed similar slopes of 0.46 and 0.42 respectively. These slopes represent the average proportion of the ambient concentration that results in personal exposure (attenuation factor (α)) across subjects for sulphate and elemental carbon.

The ratios between personal and ambient measurements for sulphate and elemental carbon provided reliable tracers for ambient generated exposure to outdoor $PM_{2.5}$ pollution in this study. The median sulphate based ambient exposure estimate for the pooled data was 8 μg/m^3 with a calculated error of ±1.7 μg/m^3. For elemental carbon, the median ambient exposure was 7±2.2 μg/m^3. A strong association was found between each of these estimates and ambient concentration at both the closest school monitor (r=0.92) and the central MWLAP TEOM (r=0.88). Individual longitudinal regressions using both sets of ambient data demonstrated that ambient generated personal exposure was 45% of ambient concentration for the study period based on the sulphate estimate. The elemental carbon estimate showed that ambient generated personal exposure was 46% of ambient concentration using ambient data from the closest school site and 50% using the MWLAP TOM. These results suggest that the MWLAP TEOM was suitable for assessing average ambient generated exposure throughout the city during the study period.

Sulphate and elemental carbon based estimates of non-ambient exposure were similar with median estimate of the pooled data of 6 and 5 μg/m^3 respectively. Distribution of the pooled data suggested that overall ambient generated exposures were slightly higher than non-ambient exposures. Moderate median longitudinal associations were found across subjects between total personal exposure and both the ambient (r=0.60) and non-ambient components (r=0.40) based on sulphate. Associations based on elemental carbon showed a lower association for the ambient component (r=0.43) while the association with the non-ambient component was slightly higher (r=0.46). Based on the sulphate and elemental carbon derived estimates respectively, longitudinal regression analyses showed that ambient generated exposure explained 39% or 41% of the variation in total exposure while non-ambient generated exposure explained 27% or 43% of the variation. The ambient component contributed 52% to total personal exposure and the non-ambient component contributed 48% for sulphate. Elemental carbon results showed the ambient contribution to be slightly higher (59%) and the non-ambient contribution slightly lower (41%) than the corresponding sulphate results. For EC the ambient contribution may be overestimated due to accounted for indoor sources or the presence of very local ambient sources. These results indicate that both ambient and non-ambient generated exposures contribute almost equal proportions to total personal exposure. The large variation due to non-ambient sources is mainly the result of the rare occurrence of very large peaks that does not generally represent everyday exposure of the general public.

An average air infiltration factor of 0.53(±0.16) and air exchange rate of 0.35(±0.39) was found through analysis of the individual data. These values compared well with a Seattle study during the heating season (Allen *et* al, 2003). Due to the presence of outliers the median value for the air exchange rate would likely provide a more accurate estimate. The median air exchange rate from the individual analyses was 0.20 with a quartile range of 0.15.

This investigation of the relationship between ambient concentration and exposure to ambient sources clearly shows that 45% of the ambient concentration level results in children's personal exposure to $PM_{2.5}$. These results demonstrate that ambient concentrations are appropriate surrogates of exposure to ambient $PM_{2.5}$ sources and strongly support the use of ambient data for a health study in this city. The regression equations provided here will allow estimation of ambient generated personal exposures based on ambient data for the purpose of a health study looking at impacts of $PM_{2.5}$ pollution on healthy elementary school children during the winter in Prince George. Additional personal monitoring would be required to determine the same information for different sub-groups of the population such as healthy adults, senior citizens or diseased children due to difference in activity patterns. Seasonal differences have also been reported in the literature; therefore the model provided may not give accurate estimates for exposure during different seasons. More extensive modeling of the relationship between these variables and other factors that may affect the personal-ambient relationship is planned. Consideration of infiltration factors, air exchange rates, time activity patterns and meteorology in more complex statistical analyses will provide a more accurate and versatile model to be used for exposure prediction in the Prince George airshed.

5 Summary of Results, Conclusions, Further Research and Recommendations

5.1. Introduction

Exposure assessment is a relatively new field of study that is growing rapidly to meet the demand of society to understand environmental effects on human health. Many factors affect actual personal exposures and in order to completely understand air pollution health effects we must be able to characterize exposure. The findings from this research demonstrate the influence that meteorology has on both ambient concentration and personal exposure for $PM_{2.5}$, the influence of non-ambient sources on personal exposures and the importance of managing ambient levels. This research provides a better understanding of children's exposure to $PM_{2.5}$ and is a stepping stone towards a health study in Prince George.

The primary goal of this thesis was to assess the relationship between outdoor concentration and personal exposure of $PM_{2.5}$ in the city of Prince George and to evaluate whether or not the current ambient monitoring network was representative of both outdoor $PM_{2.5}$ levels at unmonitored locations throughout the airshed and actual personal exposure during the study period. This main goal has been achieved and a summary of conclusions is provided to highlight the findings from this study.

It is important to clarify that these results characterize the relationships that existed between several measures and estimates pertaining to a small sample of elementary school children (15) and include: total personal exposure and ambient concentrations for $PM_{2.5}$, sulphate and elemental carbon (absorption coefficient) and the ambient and non-ambient components of total personal exposure. These measures and estimates were representative of a short six-week period during the winter that was characterized by frequent inversions, high concentrations and an overall easterly wind direction. Study findings are indicative of winter-time exposures for children in the city of Prince George and the relationships provided have an important association with meteorological conditions that existed during the study period.

5.2. Summary of Results and Conclusions

1. In future studies, researchers must consider that HPEM's and possibly other personal samplers and flow measuring devices were not intended for operation in winter conditions. The average temperature (range) during the field study was -4 (-20 to +13). Adaptations made during this study to enable winter sampling are described and include: use of plumber's heat tape to keep frost from forming over the sampling inlet and use of a waterbed heater inside a duffle bag to keep flow measuring devices warm with tubing to intake air from the cold outside environment.

2. Evaluation of the relationship between the $PM_{2.5}$ HPEM samplers and the 1400AB Rupprecht & Patashnick TEOM continuous monitor showed that the HPEM concentrations were greater with a mean difference of 4.3 ± 3.2 $\mu g/m^3$ but a significant R^2 from the linear regression analysis of 0.93 ± 0.04. An equation to provide conversion of 24-hour TEOM data for comparison with data from an HPEM sampler is :

$$HPEM = 1.38\,(\pm 0.04) * TEOM + 0.62\,(\pm 0.67)$$

3. Comparison of elemental carbon concentration and absorption coefficient levels showed that using the simple reflectance method provided an acceptable means of assessing elemental carbon levels. A strong correlation was found between absorption coefficient and elemental carbon concentration. An equation to calculate elemental carbon concentrations ($\mu g/m^3$) from absorption coefficient ($m^{-1}x10^{-5}$) for the Prince George area is:

$$EC = 0.34\,(\pm 0.02) * ABS + 0.03\,(\pm 0.04)$$

4. The importance of inversion conditions and wind direction on high levels of ambient concentrations and personal exposures is indicated by the data from the field study. Study findings suggest that days with high concentrations during the study were likely dominated by

industrial emissions carried to Plaza from the east and occasionally from sources to the southwest. Inversion conditions existed on 62% of the study days and pollution levels above 30 μg/m^3 were always associated with an inversion on the same day. Inversions influenced ambient concentrations and personal exposures at all of the study sites. Moderate Spearman correlations were found between inversion strength and ambient concentration at all of the schools for $PM_{2.5}$ (0.67), for sulphate (0.55) and for elemental carbon (0.65). For personal exposure a similar correlation was found for sulphate (0.58) and elemental carbon (0.53) and a lower correlation compared to ambient was found for $PM_{2.5}$ (0.40).

5. Significant spatial differences in ambient concentrations were found for $PM_{2.5}$, sulphate and absorption coefficient between the temporary study sites but high correlations were also found with median (range) Spearman correlations of 0.95 (0.71 to 0.96) for $PM_{2.5}$, 0.97 (0.86 to 0.98) for sulphate and 0.85 (0.67 to 0.91) for absorption coefficient.

6. The study site closest to the MWLAP permanent $PM_{2.5}$ monitor measured consistently higher concentrations suggesting that the location of the downtown MWLAP site may experience higher levels then the other areas of the city sampled during this study. Because the TEOM measures lower concentrations than the HPEM samplers it

actually represented levels at Gladstone, Lakewood and Glenview adequately. A Friedman ANOVA comparing data from each of these schools separately to unadjusted data from the TEOM showed no statistically difference between HPEM data from these schools and unadjusted TEOM data. Carney Hill did show levels significantly different than the TEOM data but not when the TEOM data was adjusted using the equation provided above from the Pilot study. Westwood did not show levels consistent with either the adjusted or unadjusted TEOM levels. Regression equations between HPEM and unadjusted TEOM data from the field study for each of the schools are provided below:

$\text{Carney Hill HPEM} = 1.21(\pm 0.07)*\text{TEOM} + 2.36(\pm 1.44), R=0.96(\pm 0.06)$

$\text{Gladstone HPEM} = 1.09(\pm 0.08)*\text{TEOM} - 0.05(\pm 1.56), R=0.94(\pm 0.07)$

$\text{Lakewood HPEM} = 1.09(\pm 0.08)*\text{TEOM} + 0.39(\pm 1.63), R=0.94(\pm 0.07)$

$\text{Westwood HPEM} = 1.02(\pm 0.07)*\text{TEOM} + 2.73(\pm 1.41), R=0.95(\pm 0.06)$

$\text{Glenview HPEM} = 0.99(\pm 0.08)*\text{TEOM} -0.84(\pm 1.70), R=0.92(\pm 0.08)$

7. Stronger personal-ambient associations (Spearman correlations) existed for sulphate (0.96) and absorption coefficient (0.73) compared to total $PM_{2.5}$ (0.52). $PM_{2.5}$ exposures were clearly impacted by non-ambient sources and there was a high degree of variability between individuals. For sulphate there was virtually no influence of non-ambient sources and for absorption coefficient there was limited influence (15% of data). The data analyses support the use of both

sulphate and absorption coefficient as indicators of ambient generated exposure.

8. Overall the ambient $PM_{2.5}$ levels in Prince George during the study period were high resulting in poor air quality. Almost half of the ambient samples collected had levels higher than the lowest ambient level at which statistically significant increases in health responses have been detected (15 $\mu g/m^3$). The number of days where concentrations exceeded 30 $\mu g/m^3$ ranged across schools from 5 to 9 days with the highest frequency occurring at Carney Hill. The 9 days at Carney Hill was enough to result in exceedance of the CWS for 2001. The standard was likely exceeded early in 2001 at both Westwood and Gladstone, having 7 and 6 days greater than 30 $\mu g/m^3$ during the study period respectively, due to high concentrations experienced in the city prior to study. The relatively higher concentrations measured at Carney Hill compared to the other schools suggests that residents of the downtown area of Prince George may have a greater risk of experiencing health effects from ambient $PM_{2.5}$.

9. The majority of the really high total $PM_{2.5}$ personal exposures were not the result of high ambient concentrations but were due to the presence of non-ambient sources. High personal exposures corresponded to high ambient concentrations (>30 $\mu g/m^3$) 29% of the time. Although

the main evidence for a health effect of $PM_{2.5}$ in previous studies is based on ambient data only, the results here suggest that individuals may be able to reduce their own personal exposure level by making different personal choices such as using an exhaust fan while cooking. It is unknown whether or not this would result in a reduction of actual risk of health effects from $PM_{2.5}$.

10. The ratios between personal and ambient measurements for sulphate and elemental carbon provided reliable tracers for ambient generated exposure to $PM_{2.5}$ pollution in this study. The median sulphate based ambient exposure estimate for the pooled data was 8 $\mu g/m^3$ with a calculated error of ± 1.7 $\mu g/m^3$. For elemental carbon, the median ambient exposure was 7 ± 2.2 $\mu g/m^3$. Sulphate and elemental carbon based estimates of non-ambient exposure were similar, with median estimate of the pooled data of 6 and 5 $\mu g/m^3$. The distribution of the pooled data suggested that overall ambient generated exposures were slightly higher than non-ambient exposures. Longitudinal regression analyses showed the exposure component of ambient origin contributed 52% to total personal exposure and the non-ambient component contributed 48% for sulphate. Elemental carbon results showed the ambient contribution to be slightly higher (59%) and the non-ambient contribution slightly lower (41%) than the corresponding sulphate results. $PM_{2.5}$ of ambient and non-ambient origin contributed

an almost equal proportion to total personal exposure and large variation due to non-ambient sources was mainly the result of the rare occurrence of very large peaks that does not generally represent everyday exposure of the general public.

11. A strong association was found between personal exposure from ambient origin and ambient concentration at both the closest school monitor (r=0.92) and the central MWLAP TEOM (r=0.88). Individual longitudinal regressions using both sets of ambient data demonstrated that ambient generated personal exposure was 45% of ambient concentration for the study period based on the sulphate estimate. The elemental carbon estimate showed that ambient generated personal exposure was 46% of ambient concentration using ambient data from the closest school site and 50% using the MWLAP TOM. Ambient concentrations were shown to be appropriate surrogates of exposure to ambient $PM_{2.5}$ sources and study results strongly support the use of ambient data for a health study in this city. Use of the regression equation provided will enable future estimation of ambient generated personal exposures for healthy elementary school children during the winter in the city of Prince George based on data from the central TEOM monitor.

$$E_{ag} = 0.45(\pm 0.08) * TEOM + 1.80(\pm 1.63), R^2 = 0.81$$

5.3. Further Research

Additional investigation using data from the time-activity diaries as independent variables in a statistical model could provide important information regarding the source of high exposures and result in a more comprehensive exposure model to further characterize the personal-ambient relationship and possibly extend the information to other sub-populations based on time-activity patterns. Further analyses of the study data may also lead to more information regarding source contributions. Although, the time activity information was helpful in a quantitative assessment of possible sources for high exposures, future studies of children should also obtain detailed information from the parents regarding cooking and other particle generating activities while the child is at home.

More extensive modeling of the relationship between the different exposure variables and other factors that may affect the personal-ambient relationship is planned using a mixed model approach that can account for both fixed and random factors. Consideration of infiltration factors, air exchange rates, time activity patterns and meteorology in more complex statistical analyses will provide a more accurate and versatile model to be used for exposure prediction in the Prince George airshed.

A strong relationship between personal exposure to ambient generated PM and ambient concentrations provides further validation for the observed health effects of ambient fine particles that has been documented extensively throughout the literature. A complete understanding of this relationship in the city of Prince George will facilitate a health study to assess the health impacts of ambient $PM_{2.5}$ levels on the local population.

5.4. Recommendations

The clear association found between the presence of inversion conditions and both high ambient concentrations and personal exposures during this study highlights the need for considering management action during episodes of $PM_{2.5}$ in the Prince George Airshed. Timely reduction of elevated point source emissions during an inversion may reduce the high spikes of ambient concentration that can occur when an inversion dissipates. Reduction of other sources such as residential wood-burning in areas out of the “bowl” are also recommended and supported by the fact that inversion conditions impacted all of the study monitoring sites, even those at a higher elevation. An assessment of the extent to which source reductions would impact ambient concentrations during episodes could be done by atmospheric dispersion modelling. $PM_{2.5}$ modeling could also be validated using the ambient data from this study.

Use of the permanent $PM_{2.5}$ TEOM monitor to represent actual ambient $PM_{2.5}$ levels throughout the city should be performed with consideration of current meteorological conditions. High correlations found between the study sites and the TEOM suggest that this monitor should adequately represent the temporal trend in concentrations but not necessarily actual levels. The regression equations provided can be used to estimate future levels at the schools based on 24-hour TEOM data. The current monitoring site may overestimate concentrations, depending on local wind patterns, providing a margin a safety when issuing air quality advisories. The relatively high concentrations measured at the downtown site compared to the other study sites should be further investigated over a longer period and over multiple seasons to confirm this finding and assess the health risk of people living or working in this area.

The strong association found between personal exposure to ambient generated sources and ambient concentrations supports the use of ambient concentrations in a health study in this city. The regression equation provided will enable estimation of children's personal exposure to ambient generated sources during the winter based on future ambient $PM_{2.5}$ concentrations measured with the central TEOM monitor. These estimates should be used to investigate the impact of ambient $PM_{2.5}$ levels on children's health. The relationship between personal exposure to ambient generated sources and ambient concentrations should be investigated for other sub-populations such as healthy adults, senior citizens and individuals with respiratory or cardiovascular disease. Future exposure studies should also include monitoring of co-pollutants and occur over multiple seasons. Monitoring of various health indicators in conjunction with exposure measurements would provide important information regarding health effects in this airshed.

Glossary of Terms & Acronyms

Abbreviations:

ABS – Absorption Coefficient – A value calculated from the reflectance of a filter that was used to sample fine particles ($PM_{2.5}$ for this study), the area of the filter, volume of the sample and the reflectance of field blank filters.

BC MWLAP – British Columbia Ministry of Water, Land and Air Protection – The local governmental agency responsible for air quality in the city and airshed management. Data obtained from this agency (Omineca-Peace Region) included hourly $PM_{2.5}$ concentrations from the TEOM sampler and wind speed, wind direction and temperature from the Plaza monitoring site.

BIOS – The main flow measuring device used in this study. This frictionless piston meter is considered to be a primary standard suitable for calibrating other flow measuring devices and for taking direct flow measurements from air pollution sampling devices.

BUCK – A different flow measuring device that is also a primary standard but operates by timing the movement of soap bubbles rather than a frictionless piston.

C_a – Concentration of ambient $PM_{2.5}$.

CWS – Canada-Wide Standard – For fine particulate matter less than 2 micrometers, this standard is based on a 3 year average of the 98th percentile. The level of the standard is 30 $\mu g/m^3$. This standard would be exceeded if levels were greater than 30 $\mu g/m^3$ an average of 7.3 days (2%) or more over 3 years.

DRI/IMPROVE – Desert Research Institute/Interagency Monitoring of Protected Visual Environments – One of two common methods used for determination of elemental carbon concentration. The method used in this study (pilot only) enables comparison to both of the standard methods.

E_{ag} – Personal $PM_{2.5}$ **exposure** that is **ambient generated** or originates outdoors. Ambient generated exposure includes exposure to ambient PM while outdoors and exposure to ambient PM that has infiltrated indoors.

E_{nag} – Personal $PM_{2.5}$ **exposure** that is **non-ambient generated** or originates indoors or from personal activity.

E_T – **Total** personal **exposure** to $PM_{2.5}$ including both the ambient and non-ambient components.

EC – Elemental carbon measured in the study via reflectance and the calculation of an absorption coefficient. In the pilot study, elemental carbon was determined in the lab via thermal optical transmittance.

FRL – Federal Reference Level – This level is 15 μg/m^3 and is a target level for a 24-hour period. It is an estimate of the lowest ambient $PM_{2.5}$ level at which statistically significant increases in health responses can be detected based on available data and current technology. This level is not a known threshold of effects below which impacts do not occur but health effects to concentrations above that level have been documented in the literature.

HOBO – Battery-powered data loggers or electronic instruments that record measurements over time. In this study, a HOBO data logger was used to record motion of the personal samplers (0=not moving; 1=moving) and both temperature and humidity at the outdoor monitoring sites.

HPEM – Harvard Personal Environment Monitors – Personal samplers developed by the Harvard School of Public Health to enable the collection of fine particulate matter near the breathing zone of a study subject in order to collect a sample representative of what the subject inhaled over a given period. The inlet is designed to select particles of a specific size range (less than 2.5 micrometers in this study) to deposit on the filter and any larger particles are captured on a greased impactor plate.

IS – Inversion Strength – The difference in temperature between a higher elevation site and a valley site. Hourly inversion strength was calculated by subtracting hourly temperature data at a valley meteorological site (Plaza) from a higher elevation site (UNBC). Positive values were associated with inversion conditions and all negative values were set to zero. 24-hour inversion strength was calculated by summing the positive temperature differences for a given day, which enabled a comparison to the 24-hour study data.

LOD – Limit of Detection – Detection limit or the level below which a species of interest cannot be accurately measured. In this study, the LOD was 3 times the standard deviation of the field blanks divided by the mean sample volume.

LPM – Litres per Minute – The units used for the rate at which air was drawn through personal and ambient samplers by a pump. In this study, the flow rate used was 4 litres per minute.

NIOSH – National Institute of Occupational Safety and Health – The organization that developed one of two common methods used for determination of elemental carbon concentration (method 5040). The method used during this study (pilot only) enables comparison to both of the standard methods.

NIST – National Institute of Standards and Technology – Calibration weights that are traceable standards from this organization were used to ensure accuracy of the microbalance used to measure $PM_{2.5}$ mass on Teflon filters.

PM – Particulate Matter – A mixture of solid particles and liquid droplets that may vary in concentration, composition and size distribution.

$PM_{2.5}$ – Particulate matter fraction that is less than 2.5 micrometers in aerodynamic diameter. More specifically, the 2.5 indicates the inlet cut-off of the sampler for which 50% efficiency is obtained for that size range. This size fraction will be referred to as fine particulate or fine particles throughout this thesis.

PM_{10} – Particulate matter fraction that is less than 2.5 micrometers in aerodynamic diameter. More specifically, the 10 indicates the inlet cut-off of the sampler for which 50% efficiency is obtained for that size range.

r – Spearman correlation coefficient – An indicator between -1 to +1 that reveals how strong and in what direction an association is between two variables that are nonparametric or not normally distributed.

R – Pearson Correlation coefficient – An indicator between -1 to +1 that reveals how strong and in what direction an association is between two variables that are parametric or normally distributed.

R^2 – Coefficient of determination for a linear regression. An indicator from 0 to1 that reveals how strong a linear relationship is between two variables and how close estimated values from a linear equation will correspond to your actual data. It is also the square of the Pearson correlation coefficient.

SO_4^{2-} – Sulphate is a secondary component of particulate matter that forms from chemical reactions in the atmosphere.

TEOM – Tapered Element Oscillating Microbalance – Instrument that measures ambient particulate mass concentration in real time. An inertial mass measurement technique is applied to make continuous direct measurements of particle mass collected on a filter.

TRS – Total Reduced Sulphur – Compounds including mainly hydrogen sulphide, methylmercaptan, dimethyldisulphide and dimethylsulphide.

TSP – Total Suspended Particulate – Particulate matter of all size fractions.

General Definitions:

Absorption Coefficient (ABS) – A value calculated from the reflectance of a filter that was used to sample fine particles ($PM_{2.5}$ for this study), the area of the filter, volume of the sample and the reflectance of field blank filters.

Aerosol – Suspensions of solid or liquid particles in a gas. Although aerosol and particle are different, they are often used interchangeably throughout the literature to refer to the particle only. In this thesis, the words aerosol, particle and particulate matter are used interchangeably.

Ambient – Outdoor measurement of air pollution.

Ambient Generated Exposure (E_{ag}) – Personal $PM_{2.5}$ **exposure** that is **ambient generated** or originates outdoors. Ambient generated exposure includes exposure to ambient PM while outdoors and exposure to ambient PM that has infiltrated indoors.

Attenuation Factor (α) – The fraction of ambient concentration that results in ambient exposure. In this study, both personal/ambient ratios of sulphate and elemental carbon were used as the attenuation factor for total $PM_{2.5}$.

Episode – In this study, an episode was defined as any period where there were hourly concentrations greater than 30 $\mu g/m^3$ for more than 6 consecutive hours. Generally, this occurred on 2 or more consecutive days.

Infiltration Factor (F_{inf}) – Equilibrium fraction of ambient particles found indoors.

Inversion – A meteorological phenomena where a layer of warm air resides over a layer of cold air. In Prince George, this often causes colder, stable air to be trapped in the valley with very little mixing or dispersion of pollution.

Inversion Strength (IS) – The difference in temperature between a higher elevation site (788m) and a valley site (602m). Hourly inversion strength was calculated by subtracting hourly temperature data at a valley meteorological site (Plaza) from a higher elevation site (UNBC). Positive values were associated with inversion conditions and all negative values were set to zero. 24-hour inversion strength was calculated by summing the positive temperature differences for a given day, which enabled a comparison to the 24-hour study data.

Non-ambient – Measurement of indoor air pollution or air pollution that is generated from personal activity.

Non-ambient Generated Exposure (E_{nag}) – Personal $PM_{2.5}$ **exposure** that is **non-ambient generated** or originates indoors or from personal activity.

Particulate Matter (PM) – A complex mixture of solid particles and liquid droplets suspended in the air that may have come from indoor, personal or outdoor sources. Numerous sources of particulate matter can result in large variability in the chemical and physical composition of the particles.

Personal Exposure – In this study, 24-hour composite exposure to fine particles measured in or near the breathing zone of a study subject. Exposure may have come from outdoor sources while outdoors, outdoor sources while indoors, indoor sources, and it may also be generated directly from personal activity.

Statistical Terminology:

Analysis of Variance (ANOVA) – In general, this statistical procedure is used to test for differences in means by comparing variances of multiple groups. There are several different methods for doing this depending on the data. The main assumptions of a basic parametric ANOVA are: normality, equal variance and equal or near equal sample size. The first two assumptions are robust as long as the third assumption is met (Zar, 1984). If the assumptions are not met, non-parametric ANOVA is appropriate. Two non-parametric methods used in this study were the Kruskal Wallis ANOVA and the Friedman's 2-factor ANOVA. The **Kruskal-Wallis ANOVA by ranks** is similar to the basic parametric ANOVA but for data that is not normally distributed, therefore this test assesses the variance in each group and determines if a significant difference exists between groups. **Friedman's 2-Factor ANOVA** is similar to a repeated measures design in that it considers time when assessing variances between groups; the two factors are time and group (school or location in the case of this study). The Friedman's test is especially useful for with data that do not meet the parametric analysis of variance assumptions of normality and equal variance (even if they are seriously violated) and is just as powerful as the parametric ANOVA if the assumptions are met (Zar, 1984). More complex ANOVA's can also be used. For longitudinal data such as an individual's exposure over a given time, **Mixed Model ANOVA** can also be used. With this analysis the model can incorporate data from several subjects and account for differences between individuals as a random factor in the model. The problem with this approach is there are various options available with this model and it is much more complex. More detailed statistical knowledge is required to confidently perform the analysis.

Bonferroni Adjustment – Type I error rate is inflated if multiple tests are done, therefore more stringent alpha levels (p-values) are required. This adjustment requires that the cumulative acceptable alpha level for each individual test be less than the overall acceptable alpha level (Zar, 1985). Example: If data from 5 locations were compared separately to one another, an acceptable alpha would be 0.005 - overall alpha of 0.05 divided by 10 tests.

Correlation – The relationship or association between two variables. If the data is normally distributed the Pearson correlation (R) is used. If the data is non-parametric or not normally distributed than the Spearman correlation (r) is used. The Spearman correlation will not be influenced by outliers in the data. Although the data in this study was highly skewed and therefore not normally distributed, both types of correlations were used to enable better comparison to other studies.

Linear Regression – A statistical procedure that attempts to explain the relationship between two variables by fitting a straight line to the data. A slope and intercept for the line are determined by attempting to make the sum of the square residuals as small as possible. The strength of the relationship is measured by the square of the Pearson correlation (R^2).

Paired T-test – A parametric test for detecting differences between two dependent groups.

Wilcoxon Matched Pairs Test – A non-parametric test for detecting differences between two dependent groups.

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Appendix 1 - Daily Time Activity Diary and Questionnaire

Sample Daily Time Activity Diary

Important Questions and Reminders

Daily Review Checklist

A1.1. Sample Daily Time Activity Diary

	Time	Inside	Outside	Where am I?	Where is the pack?	Travelling	Smoker Nearby?	What am I doing?	Other Notes
	8:00	○	○			○Car ○Bus ○Walk ○Bike	○		
		○	○				○		
	8:30	○	○			○Car ○Bus ○Walk ○Bike	○		
		○	○				○		
M	9:00	○	○			○Car ○Bus ○Walk ○Bike	○		
O		○	○				○		
R	9:30	○	○			○Car ○Bus ○Walk ○Bike	○		
N		○	○				○		
I	10:00	○	○			○Car ○Bus ○Walk ○Bike	○		
N		○	○				○		
G	10:30	○	○			○Car ○Bus ○Walk ○Bike	○		
		○	○				○		
	11:00	○	○			○Car ○Bus ○Walk ○Bike	○		
		○	○				○		
	11:30	○	○			○Car ○Bus ○Walk ○Bike	○		
		○	○				○		

Table A 1 This table shows a sample of the daily time activity diary that each subject was required to fill out throughout their day while carrying the personal exposure monitor. The diary continues until 11am the following day enabling entries every half hour for the entire sampling period. To follow is a list of questions to ask when carrying the sampler, which was provided in a notebook for each subject to help them remember important things that should be recorded in their diary. At the end of each sampling period the diary was reviewed following the Time Activity Diary Review Questionnaire also included in this appendix.

A1.2. Important Questions to Ask Yourself When Carrying the Sampler

1. **Are you wearing the backpack? If not where is it?**
2. **Is there someone smoking nearby?**
3. **Is there someone cooking nearby?**
4. **Is there something (food) burning nearby?**
5. **Is there someone vacuuming or sweeping nearby?**
6. **Is there someone dusting nearby?**
7. **Are there any windows open?**
8. **Is there a fan on (ceiling fan or kitchen stove)?**
9. **Are there candles or a fireplace burning?**
10. **Are you playing with your pet?**

Other Important Things to Remember:

- **Take the pack everywhere!**
- **Fill in the diary every ½ hour.**
- **Wear the pack as much as possible.**
- **When you can't wear it put in safe close location.**
- **Always have sampler pointing downward.**
- **Record any problems in your diary.**
- **Call Melanie if you have questions or there is a problem with the sampler (**
- **Practice being a scientist and have fun!**

A1.3. Time Activity Diary Review Questionnaire

Please Fill In Missing Information in Green Pen on Their TAD

1. If they did not indicate that they were near a smoker ask:
Are you sure you weren't near a smoker?

2. If they indicated they were near a smoker ask:
Approximately how many minutes were you near the smoker?
Did you notice about how many cigarettes were smoker?

3. If they were somewhere other than home or school (restaurant or someone else's house) ask:
Did you notice any one smoking or did you smell any smoke?
Where is that place located...in their neighbourhood, in the bowl or elsewhere?

4. If they were in a car or outside ask:
Approximately how many minutes were you in the car?
Approximately how many minutes were you outside?

5. Always ask about cooking at dinnertime especially if they don't mention eating or cooking in the diary:
When did you eat dinner last night and what did you have...was it fried, baked etc.?
Were you near the kitchen at all when dinner was being cooked?
Did anything get burned and did they notice if the fan was on?
Did your mom or someone else in the family do any other cooking or baking last night?

6. Always ask if there was any fire burning (gas, wood or pellet stove) or any candles being used.
How long was the fire burning?
How many candles were burning and how long were they burning for?

If you notice that they are one place and the pack is somewhere else comment on it and if it is not apparent ask how long the pack was not near them.

Make sure they fill in where they were at home and school...just putting at home or at school is not enough detail.

Don't let them write "filling out diary" as what they did that half hour. Ask them what they were doing before or after they filled in the diary.

If they write in the notebook please remove the pages and stable to the TAD.

Appendix 2 - School and Parent Correspondence

Research Project Information Sheet

Introductory Letter to School Board

Introductory Letter for Principals and Teachers

Curriculum Integration Proposal for Teachers

Mid Study Progress Report

Study Participant Certificate of Completion

Literature Cited in Correspondence

A2.1. Research Project Information Sheet

This information sheet was provided with all initial correspondence and recruitment questionnaires.

Researcher: Melanie Noullett

Home Phone #:
Office Phone #:
Email Address:

Supervisor: Dr. Peter Jackson
UNBC Faculty of Environmental Studies
Prince George, BC
Phone #:
Email Address:

Title of Project: Ambient and Personal Exposure Levels of Fine Particulate Matter Throughout the Prince George Airshed.

Type of Project: Master's Thesis

Research Purpose: To develop a detailed understanding of the relationship between ambient fine particulate matter air pollution levels and actual personal exposure and to characterize the spatial variation of both of these variables throughout the Prince George Airshed.

Start Date: November/2000

Completion Date: November/2001

Potential Benefits: Although there are no direct benefits for participating in the study, indirect benefits include: increased knowledge regarding the relationship between personal exposure and ambient levels of fine particulate air pollution and an assessment of the adequacy of having only one ambient fine particulate monitor in the airshed. Understanding the relationship between exposure and ambient levels is important to determining the impact of fine particulate pollution on human health. Scientific evidence that more monitors are needed may aid in an expedient expansion of the network. This will in-turn result in an increased awareness of the actual level of human exposure to this form of air pollution.

Potential Risks: The main risk to the participant is personal inconvenience during the sampling period from carrying the monitor and frequently filling out a time activity diary. Discomfort may be caused by the weight of the sampler assembly. Another possible risk includes peer pressure, as only 3 students in each class will be carrying a monitor. It is the goal of the researcher to involve the entire class in the project with the help of the teacher and to have those selected to carry monitors feel as though they are being rewarded.

Recruitment: Each participant chosen to take part in the study will be selected based on several criteria. The actual class will be selected based on the interest and support shown by the teacher and the location of the school within the airshed. The students will then be selected based on their interest in participating and recommendation made by the teacher. The participant must also meet several requirements including the following: being from a non-smoking family, living within walking distance from the school and living in a single-family dwelling.

Participant Task: The participant will be required to wear an air pollution monitor that includes a small 2-3 pound pump carried in a small back-pack and a 1 pound sampler that will be attached within the child's breathing zone between the shoulder and chest. This device will be carried 24-hours a day for two 5-day periods. During certain activities, such as swimming and sleeping, the monitor can be set in a nearby, safe location. Participants will also be required to fill out a time activity diary every 30 minutes to show where they are, what they are doing and if they are near a smoker or another obvious source of fine particulate. The researcher will meet with the child and his or her parents prior to their first 5-day monitoring period to explain the time activity diary, demonstrate how the sampler should be carried, record several household characteristics and to answer any questions from the participant or parents. Study staff will also meet with each participant at school each morning to check the flow rate of the sampler, change the filter and collect the previous days time activity diary. The time activity diary will be reviewed and follow-up questions will be asked to ensure that all relevant information is recorded. Each evening a meeting will be arranged with both the parents and child to discuss that day's activity and to ask any follow-up questions that may be necessary. This will also provide an opportunity for the child and parents to ask questions or express concerns.

Consent Form: A parent or guardian of each participant in the study will be asked to sign a form giving their informed consent for the use of all the information provided during the study. Copies of the signed consent form will be available to the participant if requested. A separate optional consent form may also be used to obtain permission to take photos of participants wearing the monitor. With consent the photos will be used to communicate the results and methods used in the study.

Access to Data Collected: Only the researcher and supervisor will have access to the answers obtained during the initial recruitment questionnaire, the first home visit, follow-up interviews and from the time activity diaries. Data from the recruitment questionnaire will only be used to select participants. All other information will be used to aid in the interpretation of exposure level data obtained from lab analysis of the samples collected and will not be reported on an individual basis in any publication. The raw numeric data from lab analysis of the filters will be available to the teachers and their class at the request of the teacher to use as part of a class science project. Individual results will also be available to the participants and their families once the data has been analyzed and an interpretation made by the researcher.

Remuneration: Participants will not receive any remuneration unless a donation is made by an organization within the community prior to the start of the study. Participants will be awarded a certificate of completion.

Anonymity: Participants in the study will be referred to by a number and location code to identify which school they are from. The use of participant's names or any other association will be avoided at all times. The student will be seen carrying the monitor by other members of the class and by people in the community.

Confidentiality: The researcher, Melanie Noullett, her supervisor Dr. Peter Jackson and a research assistant will be the only individuals to review non-numeric raw data from interviews, questionnaires and time activity diaries. The research assistant will primarily help with data collection and not the analysis that will follow.

Data Storage: Data attained from the study will be stored in the secure office of Dr. Peter Jackson for up to two years following the conclusion of the research thesis.

Inquiries: All concerns or questions regarding the research thesis can be directed to Melanie Noullett or Dr. Peter Jackson at the phone numbers or addresses listed above.

Research Results: Research results will be made available to each of the participants of the study at their request. Additional copies can be obtained from Melanie Noullett or Dr. Peter Jackson at any time following the completion of the research thesis.

Information: Requests for additional information on the research study can be directed to Melanie Noullett at the contact information listed above.

Complaints: Complaints regarding the research study can be directed to the Office of Research and Graduate Studies, UNBC,

Participation in this study is voluntary and participants have the right to withdraw from the study at any time.

A2.2. Introductory Letter to School Board

November 15, 2000

School District No. 57
1894 – 9th Avenue
Prince George, BC
V2M 1L7

Attention: Phil Redmond and School Board Members

I am a graduate student at the University of Northern BC conducting a research study of ambient and personal exposure levels of fine particulate air pollution. The purpose of this study is to develop a detailed understanding of the relationship between ambient $PM_{2.5}$ (particulate matter less than 2.5 microns in diameter) levels and actual personal exposure and to characterize the spatial variation of both of these variables throughout the Prince George Airshed. Understanding the relationship between exposure and ambient levels is important to determining the impact of fine particulate pollution on human health. Characterizing spatial variation will help airshed managers assess the adequacy of having only one ambient monitoring station for $PM_{2.5}$ in the airshed and will also provide information needed to decide where additional monitors should be located if needed.

The component of the research that must obtain approval from the School Board will involve personal exposure monitoring of 15 children ages 10 to 11 and outdoor monitoring at school facilities. The reason that children have been selected as subjects for this study is that they often attend schools located in the same area as their home and are generally less mobile than adults. Since one purpose of the study is to look at spatial variation throughout the city, it would be best to have the subjects reside in one area the majority of the time that they are being monitored. Each participant will be asked to carry a monitor 24 hours a day for two 5-day periods. A small backpack containing a flow pump and motion sensor will be attached by latex tubing to a small sampling device. The tubing will run from the backpack over the child's shoulder and the sampling device will be attached to the child's clothing close to their breathing zone (between the shoulder and chest area). Together the sampling apparatus and backpack will weigh approximately 3 to 4 pounds. Participants will also be required to fill out a time activity diary, recording where they are and what they are doing every 30 minutes. The researcher will meet with the child and his or her parents prior to their first 5-day monitoring period to explain the time activity diary, demonstrate how the sampler should be carried, record several household characteristics and to answer any questions from the participant or parents. Each morning of the sampling period the researcher or an assistant will meet with the child at school to check the flow rate of the sampling pump and change the filter. The time activity diary from the previous day will be reviewed with the child and a list of follow-up questions will be asked of the child and the teacher if necessary. Data will also be downloaded from a motion sensor placed in the backpack carrying the sampling pump. Every evening a visit will be made to each participant's house to discuss the activities of the child that day and to administer a follow-up interview with both the parent and the child together. Other researchers have used this method of monitoring personal exposure of children and the results have been published in several academic journals (Geyh *et al*, 2000; Janssen *et al*, 1999; and Wheeler *et al*, 1999). The researcher for this project has visited the Harvard School of Public Health, a leader in the field of personal exposure monitoring, to receive specific training to perform this type of research.

Once approval is given by the school board, participants will be selected by finding schools that are willing to have students participate in the study and teachers that are willing to involve the class in the study as part of a science project. It is important that the entire class be involved to some extent to avoid any peer pressure that could result from only a few selected students actually carrying the personal monitors. A meeting will be set-up with each teacher of grade 5 and 6 classes to discuss the research and assess the interest and support of the teacher. From the teacher interviews the classes will be ranked according to which teachers are more supportive and interested in the research. 5 classes will then be selected based on this ranking and the location of the school in the city. Different areas of the city must be represented and consideration will be given to whether or not local sources of pollution are prominent. A presentation will be given to each of the 5 classes selected outlining the research project and answering any questions. An information letter and questionnaire will be sent home only with students interested in the project and eager to participate. It is important that children are not influenced by their parents when deciding whether or not they want to participate, as they are the ones that will have to carry the monitor. Returned questionnaires will be reviewed and each student will be ranked according to suitability for participation in the study. Students from smoking homes will not be considered and preference will be given to students that live within walking distance to the school. Comments from the teacher regarding the likeliness that a student will commit to the project will also be considered. The top 3 students in this ranking from each of the 5 classes will be asked to participate in the study and a consent form will be sent home. An information sheet will also be sent home with all children in the class to inform their parents about the study. As a class science project the whole class will be involved in some aspect of the study although only 3 students from each class will actually carry monitors. If a student does decide not to participate before or during the study the next student on the ranked list from that class will asked to participate and a consent form sent home.

Several studies in the past have used various forms of compensation as a motivational technique to encourage compliance. Examples of compensation used in other studies involving children include free movie passes, $50 savings bonds, watches, study t-shirts and certificates. Use of one or more of these examples may be used depending on the amount of funds available or if donations can be obtained from a source within the community. Certificates of completion will be awarded, as this is an inexpensive way of rewarding the participants. If possible the researcher would like to give free movie passes and $50 savings bonds to participants that complete the study.

Participants may or may not be aware that the backpacks they carry will contain motion sensors. A data log will be downloaded from the motion sensor on a daily basis and will be used as a means of assessing compliance of the participant in carrying the monitor. Motion data will be compared to the time activity log and problems with compliance will be identified where discrepancies exist. This will only be used as a means of assessing sample validity. If there are several occurrences of non-compliance the participant may be asked to leave the study and a new participant will be recruited.

The actual exposure data obtained from the study will be shared with each class involved once the lab analysis has been completed and the data validated. All of the data will be shared between schools with the names of the students carrying the monitoring remaining confidential. Complete anonymity cannot be guaranteed as participants will be seen carrying monitors by other members of the class and the community. Each individual that participated can request to see their own data and a meeting can be arranged with the researcher to explain the results if the participant wishes. Time activity diaries, questionnaires and household characteristic surveys will be kept confidential. These items are intended to aid in the interpretation of the personal exposure data – weight of sample collected and chemical analysis – and will not be discussed at an individual or even school level. Data will not be available to the participants until the field study and lab analysis are complete and the data has undergone quality assurance by the primary researcher and accepted as valid. Raw data may be shared with the classes at the request of the teacher. Final results of the study will not be available until approximately the end of August.

I hope that this letter has outlined the fundamental elements of the research that would be of interest to the School Board. Attached to this letter is a copy of the information sheet, consent forms and recruitment questionnaires that will be provided to participants and their parents or guardians. Also available is a copy of the academic proposal for this research and a letter demonstrating conditional approval from the UNBC Ethics Committee pending a few minor changes.

The actually monitoring for this project is scheduled to commence at the beginning of February 2001. This start date is based on the availability of equipment being borrowed from the Harvard School of Public Health. Recruitment should be started as soon as possible. I appreciate your time in reviewing this proposal and would be happy to answer any questions by phone, email or in person.

Sincerely,

Melanie Noullett
UNBC MSc Candidate and Project Coordinator
Phone:
Email:

A2.3. Introductory Letter for Principals and Teachers

December 8th, 2000

Attention: School District No. 57 Principals and Teachers

I am a graduate student at the University of Northern BC conducting a research study of ambient and personal exposure levels of fine particulate air pollution. The purpose of this study is to develop a detailed understanding of the relationship between ambient $PM_{2.5}$ (particulate matter less than 2.5 microns in diameter) levels and actual personal exposure and to characterize the spatial variation of both of these variables throughout the Prince George Airshed. Understanding the relationship between exposure and ambient levels is important to determining the impact of fine particulate pollution on human health. Characterizing spatial variation will help airshed managers assess the adequacy of having only one ambient monitoring station for $PM_{2.5}$ in the airshed and will also provide information needed to decide where additional monitors should be located if needed.

Administrators of School District 57 have reviewed a proposal outlining this research and permission has been given by the Superintendent to enter the schools and start the recruitment process. Progress of recruitment and reactions of teachers, parents and children will be reported to the school district on an on-going basis.

The main component of this research will involve personal exposure monitoring of 15 children ages 10 to 11 and outdoor monitoring at school facilities. The reason that children have been selected as subjects for this study is that they often attend schools located in the same area as their home and are generally less mobile than adults. Since one purpose of the study is to look at spatial variation throughout the city, it would be best to have the subjects reside in one area the majority of the time that they are being monitored. Each participant will be asked to carry a monitor 24 hours a day for two 5-day periods. A small backpack containing a flow pump and motion sensor will be attached by latex tubing to a small sampling device. The tubing will run from the backpack over the child's shoulder and the sampling device will be attached to the child's clothing close to their breathing zone (between the shoulder and chest area). Together the sampling apparatus and backpack will weigh approximately 3 to 4 pounds. Participants will also be required to fill out a time activity diary, recording where they are and what they are doing every 30 minutes. The researcher will meet with the child and his or her parents prior to their first 5-day monitoring period to explain the time activity diary, demonstrate how the sampler should be carried, record several household characteristics and to answer any questions from the participant or parents. Each morning of the sampling period the researcher or an assistant will meet with the child at school to check the flow rate of the sampling pump and change the filter. The time activity diary from the previous day will be reviewed with the child and a list of follow-up questions will be asked of the child and the teacher if necessary. Data will also be downloaded from a motion sensor placed in the backpack carrying the sampling pump. Every evening a visit will be made to each participant's house to discuss the activities of the child that day and to administer a follow-up interview with both the parent and the child together. Other researchers have used this method of monitoring personal exposure of children and the results have been published in several academic journals (Geyh *et al*, 2000; Janssen *et al*, 1999; and Wheeler *et al*, 1999). The researcher for this project has visited the Harvard School of Public Health, a leader in the field of personal exposure monitoring, to receive specific training to perform this type of research.

Participants will be selected by finding schools that are willing to have students participate in the study and teachers that are willing to involve the class in the study as part of a science project. It is important that the entire class be involved to some extent to avoid any peer pressure that could result from only a few selected students actually carrying the personal monitors. A meeting will be set-up with each teacher of grade 5 and 6 classes to discuss the research and assess the interest and support of the teacher. From the teacher interviews the classes will be ranked according to which teachers are more supportive and interested in the research. 5 classes will then be selected based on this ranking and the location of the school in the city. Different areas of the city must be represented and consideration will be given to whether or not local sources of pollution are prominent. A presentation will be given to each of the 5 classes selected outlining the research project and answering any questions. An information letter and questionnaire will be sent home only with students interested in the project and eager to participate. It is important that children are not influenced by their parents when deciding whether or not they want to participate, as they are the ones that will have to carry the monitor. Returned questionnaires will be reviewed and each student will be ranked according to suitability for participation in the study. Students from smoking homes will not be considered and preference will be given to students that live within walking distance to the school. Comments from the teacher regarding the likeliness that a student will commit to the project will also be considered. The top 3 students in this ranking from each of the 5 classes will be asked to participate in the study and a consent form will be sent home. An information sheet will also be sent home with all children in the class to inform their parents about the study. As a class science project the whole class will be involved in some aspect of the study although only 3 students from each class will actually carry monitors. If a student does decide not to participate before or during the study the next student on the ranked list from that class will be asked to participate and a consent form sent home.

Several studies in the past have used various forms of compensation as a motivational technique to encourage compliance. Examples of compensation used in other studies involving children include free movie passes, $50 savings bonds, watches to help with time activity diaries, study t-shirts and certificates. Use of one or more of these examples may be used depending on the amount of funds available or if donations can be obtained from a source within the community. Certificates of completion will be awarded, as this is an inexpensive way of rewarding the participants. If possible the researcher would like to give free movie passes and $50 savings bonds to participants that complete the study.

Participants may or may not be aware that the backpacks they carry will contain motion sensors. A data log will be downloaded from the motion sensor on a daily basis and will be used as a means of assessing compliance of the participant in carrying the monitor. Motion data will be compared to the time activity log and problems with compliance will be identified where discrepancies exist. This will only be used as a means of assessing sample validity. If there are several occurrences of non-compliance the participant may be asked to leave the study and a new participant will be recruited.

The actual exposure data obtained from the study will be shared with each class involved once the lab analysis has been completed and the data validated. All of the data will be shared between schools with the names of the students carrying the monitoring remaining confidential. Complete anonymity cannot be guaranteed, as participants will be seen carrying monitors by other members of the class and the community. Each individual that participated can request to see their own data and a meeting can be arranged with the researcher to explain the results if the participant wishes. Time activity diaries, questionnaires and household characteristic surveys will be kept confidential. These items are intended to aid in the interpretation of the personal exposure data – weight of sample collected and chemical analysis – and will not be discussed at an individual or even school level. Data will not be available to the participants until the field study and lab analysis are complete and the data has undergone quality assurance by the primary researcher and accepted as valid. Raw data may be shared with the classes at the request of the teacher and will be available at the end of April. Final results of the study will not be available until approximately the end of August. Data from the existing air pollution monitoring network and from the UNBC weather station will be available for teachers to use in their classes if they choose.

I hope that this letter has outlined the fundamental elements of this research. Attached to this letter is a copy of the information sheet, consent forms and recruitment questionnaires that will be provided to participants and their parents or guardians. Also available on request is a copy of the academic proposal for this research and a letter demonstrating approval from the UNBC Ethics Committee.

The actually monitoring for this project is scheduled to commence at the beginning of February 2001. This start date is based on the availability of equipment being borrowed from the Harvard School of Public Health. Recruitment should be started as soon as possible. I appreciate your time in reviewing this proposal and would be happy to answer any questions by phone, email or in person.

Sincerely,

Melanie Noullett
UNBC MSc Candidate and Project Coordinator
Phone:
Email:

A2.4. Curriculum Integration Proposal for Teachers

Finding interested and supportive teachers is a critical component of this project. It is my intent to disrupt the class as little as possible. But it is also important that the entire class be involved to some extent in order to minimize the potential for peer pressure on the few students selected to actually carry monitors. The extent that this project is integrated with the regular curriculum is at the discretion of each individual teacher. I am willing to provide data to the class and aid the teacher in any way that they require. This project can be an excellent opportunity for children to experience real scientific research and to learn about data collection and what the data can tell us.

The pump that the child carries will make a certain amount of noise, a little louder than the hum of a loud computer. This could be a possible distraction to the class. The student must carry the monitor with them at all times and record their activities every 30 minutes. This will not be an easy task for the child, but could be made easier with reminders from the teacher and parents. Either myself or a research assistant will visit the school every morning before class starts to change the sampler, measure the flow rate and review the child's time activity diary. (It is possible that these visits could be scheduled for lunchtime or after school but it must be the same at all schools.) Some questions may be asked of the teacher if there are uncertainties about what the child has written during school hours. A daily visit will also be made with the parents every evening. I will be available at other times to work with the class if requested by the teacher.

The actually data from the study will not be available until the middle or end of April as lab analysis is being performed at UBC facilities once all sampling is complete. But there will be ambient $PM_{2.5}$ and other air pollution data available on a daily basis from the Ministry of Environment monitoring network if the teacher would like to use it in their class. I would be willing to visit the schools again in May or June when the data set is complete and validated to present preliminary results to the class and to help with an exercise using the data.

This project can have a very small impact on your class with only a minor inconvenience and distraction or it can play an important role in your science curriculum and provide a unique opportunity to your students. The extent of your classes involvement is up to you and I will be available to make any extra work as light as possible.

Please fill free to phone or email me with any questions, concerns or suggestions.

Melanie Noullett
MSc Candidate - Natural Resources and Environmental Studies
University of Northern British Columbia
Phone:
Email:

A2.5. Mid Study Progress Report

February 28, 2001

Attention: Parents, Teachers and Participants of the Prince George Air Quality Exposure Study

It is the middle of week 4 for the air quality exposure study. We are at the halfway point and I just wanted to give everyone an update on how things are going. The kids at all five schools are doing an excellent job. I am very happy with how things are going and impressed with the dedication shown by all of the participants.

Just a few reminders for everyone:

The kids must sleep at their primary residence when they have the backpack - no sleepovers. This is important in order to have some consistency over the sampling days. There are a lot of variables in this type of study and we need to try and control some of them so later we will be able to make firmer conclusions. If there is some kind of family circumstance that requires a child to stay at a different residence over night, please contact me and we will work something out.

Participants should be actually wearing the pack as much as possible. The best sample will be when the sampler is near their breathing zone. This past week we had a small competition at each school. One participant at each school won a free pass to Bubba Baloos for having the best diary and wearing the pack the most. We will be having similar competitions for the remainder of the study with different prizes donated by members of the local community.

From the samples we have collected so far, we suspect that cooking is a very important indoor source. We will be asking the children more questions about what was cooked and how it was cooked when we review their diaries. It would be great if parents could help the children with recording when cooking is done in the household and how and what was cooked. This is important even if the kids are in their room or downstairs.

Another important factor is whether there was anything burning in the house - including a fireplace, candles, food or cigarettes.

I will continue to visit the kids after school or in the early evening. This is necessary to check the flow rate on the sampler. Please contact me if you know you aren't going to be home on the day your child has the pack. Hopefully the next few weeks will go as smoothly as the first three. Thanks to all the kids for helping me collect good data – keep up the good work! Please feel free to contact me anytime if there are any problems or concerns.

Sincerely,
Melanie Noullett

A2.6. Study Participant Certificate of Completion

Certificate of Achievement

This is to certify that

____

Successfully Participated in the Prince George Fine Particulate Air Quality Exposure Study

February 5th to March 16th, 2001

Your hard work and dedication as a UNBC research volunteer has contributed to the success of this project. Thank You.

Certified By ______________________

____

A2.7. Literature Cited in Correspondence

Geyh, Alison S., Jianping Xue, Haluk Ozkaynak and John Spengler. 2000. The Harvard Southern California Chronic Ozone Exposure Study: Assessing Ozone Exposure of Grade-School-Age Children in Two Southern California Communities. *Environmental Health Perspectives,* 108(3): 265-270.

Janssen, Nicole A.H., Gerard Hoek, Hendrik Harssema and Bert Brunekreef. 1999. Personal Exposure to Fine Particles in Children Correlates Closely with Ambient Fine Particles. *Archives of Environmental Health*, 54(2): 95 - 101.

Wheeler, A.J., R. Beaumont, R.S. Hamilton and S. Farrow. 1999. Monitoring Children's Personal Exposure to Airborne Particulate Matter in London, UK – Method Development and Study Design. *The Science of the Total Environment*, 235: 397-398.

Appendix 3 - Recruitment and Consent Forms

Recruitment Questionnaire - Student Portion

Recruitment Questionnaire - Parent or Guardian Portion

Informed Parental or Guardian Consent Form

Photography Consent Form

A3.1. Recruitment Questionnaire – Student Portion

School: ________________________ **Teacher:** ________________________

Student's Name: ________________________

Student Portion:

1. Do you think that air quality is a problem in Prince George? Yes No
2. Would you like to help improve air quality in our city? Yes No
3. Did you find the presentation given to your class interesting? Yes No
4. Would you like to volunteer for the study? Yes No
5. Are you aware that carrying a monitor may be awkward and inconvenient at times? Yes No
6. Do you walk to school? Yes No
7. Does anyone in your family smoke? Yes No
8. Are there often smokers visiting your home? Yes No
9. Do you have any pets? Yes No
10. Do you live in a single-family home (e.g. A house not an apartment)? Yes No

Why would you like to participate in this study?

____

____

____

____

____

____

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____

A3.2. Recruitment Questionnaire – Parental Portion

School: ________________________ **Teacher:** ________________________

Student's Name: ________________________

Hi, my name is Melanie Noullett and I am a graduate student at the University of Northern BC. I am conducting a research study for my master's thesis and would like to monitor your child's personal exposure to fine particulate air pollution. I have given a presentation at your child's school and he/she has shown interest in participating in my study. Attached is an information sheet outlining important details of the project and describing the requirements of the participants and their parents.

Are you interested in having your child participate in the study? Yes No

If so, could you fill out the questionnaire below and have your child return it to their teacher. If your child is selected for the monitoring program an information meeting will be arranged so that you can ask any questions and a consent form can be signed if you decide to have your child participate in the study. Only three students will be monitored from each school selected for the study. Please be honest with your answers and fully consider the inconvenience participation may have to your family. Also consider that the data obtained from this study may lead to improved monitoring of fine particulate pollution in the Prince George area. The information obtained may also help airshed managers assess the potential health effects of this pollutant and determine what actions are necessary to minimize high levels of exposure in our city.

1. Do you think that air quality is a problem in Prince George? Yes No
2. Would you like to help improve air quality in our city? Yes No
3. Did you read the information sheet sent home with your child? Yes No
4. Do you think your child is responsible enough to participate in a scientific study? Yes No
5. Are you aware that carrying a monitor may be awkward and inconvenient for your child? Yes No
6. Does your child walk to school? Yes No
7. Does anyone in your family smoke? Yes No
8. Are there often smokers visiting your home? Yes No
9. Do you have any pets? Yes No
10. Do you live in a single-family home? Yes No

Why would you like your child to participate in this study?

____

____

____

____

A3.3. Informed Parental or Guardian Consent Form

Please circle the appropriate response.

1. Do you understand that your child has been asked to participate in a research study? Yes No
2. Have you read and received a copy of the study Information Sheet? Yes No
3. Do you understand that the research interviews may be tape-recorded? Yes No
4. Do you understand the benefits and risks involved in your child's participation in this study? Yes No
5. Have you had an opportunity to ask questions and discuss this study? Yes No
6. Do both you and your child understand that either of you can refuse to participate or withdraw from the study at any time without giving a reason? Yes No
7. Have you discussed the study with your child and given them the opportunity to decline participation without providing a reason? Yes No
8. Has the issue of confidentiality been explained to you? Do you understand who will have access to the information you provide? Yes No

This study was explained to me by: ______________________________

I agree to let my child take part in this study.

____________________	__________	____________________
Signature of Parent or Guardian	Date	Signature of Student
____________________		____________________
Printed Name of Parent or Guardian		Printed Name of Student

I believe that the person signing this form understands what is involved in the study and voluntarily allows their child to participate.

____________________	__________
Signature of Investigator	Date

A3.4. Photography Consent Form

Title of Project: Ambient and Personal Exposure Levels of Fine Particulate Matter Throughout the Prince George Airshed.

Principal Investigators:
Dr. Peter Jackson
Melanie Noullett - MSc Candidate

Subject's Name: ______________________________

Name of Parent or Guardian: ______________________________

I, ________________________________, give the University of Northern British Columbia permission to photograph my child wearing the personal monitor as part of the study, "Ambient and Personal Exposure Levels of Fine Particulate Matter Throughout the Prince George Airshed," conducted during the Winter of 2001.

Participant ID: ______________________________

Signature of Parent or Guardian: ____________________________ Date: ____________

Witness to Signature: ______________________________ Date: _____________

Appendix 4 - Questionnaire on Household Characteristics

Participant ID:________________

Winter 2001

University of Northern British Columbia

Prince George Fine Particulate Exposure Study
February - March 2001

Questionnaire on Household Characteristics

(Obtained from Harvard School of Public Health: Boston Study Winter 1999-2000)

Household Identification:

1. Address

____
Number Street

____
City Postal Code

2. Telephone

____________________ ____________________
Home Parent or Guardian's Work

Participant ID:

The purpose of this questionnaire is to obtain information about you and your residence. We are asking the same questions from each of the participants in this study. All the information will be kept confidential.

II. HOUSEHOLD CHARACTERISTICS:

I will ask some questions regarding your household.

TYPE OF RESIDENCE:

1. What type of cooking fuel do you use?	Gas.........................1 Electric.....................2 Other........................3
2. Does your house have storm windows?	Yes..........................1 No...........................2 IF YES, ON WHAT % OF WINDOWS ______
2.1 What type of heating do you use during the winter?	Central......................1 Radiators....................2 Kerosene/Space...........3 Wood fireplace...........4 Other ____________....9
2.2 Do you use a wood-burning fireplace?	Yes..........................1 No...........................2 IF YES, HOW FREQUENTLY ______
3. Is there a fan over the cooking stove, range, oven, or elsewhere in the kitchen area?	Yes..........................1 No...........................2 IF NO, GO TO 6
4. How does this fan work?	Kitchen exhaust vented to the outside...............1 Recirculation of indoor air.............................2 Charcoal filter...........3 Other, please specify ____________ 8 Don't know................9 Not applicable............0
5. How often is this fan used?	Most of the time when the stove is used...........1 Occasionally...............2 Rarely.......................3 Never........................4 Don't know..................9 Not applicable..............0

Participant ID:

6. Do you have: **Circle all that apply**	An unvented clothes dryer located in the house or an attached structure, such as a garage?..........1 An unvented kerosene heater in the house or an attached structure?..........2 A fireplace or wood stove in the house or an attached structure?..........3 A whole-house or attic fan?..........4
7. Do you have any air cleaning devices in your home?	Yes..........1 No..........2
8. What type of air cleaning device(s) do you have at home? **Circle all that apply**	Ion generator..........1 Filter..........2 Electrostatic precipitator..........3 Other, please specify...8 ____________ Not applicable..........0
9. What type of garage, if any, do you have?	None, detached or separate carport..........1 Attached..........2 Underneath..........3 Don't Know..........9 **IF NONE, GO TO 12**
10. Is this garage used for:	Parking one car..........1 Parking two cars..........2 Storage only..........3
11. Does this garage:	Have a pull-down door..........1 Electric door..........2 No door..........3
12. How many separate central AC or window/wall units do you have in your home?	______ number of central AC units ______ number of window/wall units
13. Do you have a de-humidifier in your home?	Yes..........1 No..........2
14. How frequently is your home dusted?	Once per day..........1 Once per week..........2 Once every two weeks.....3 Once per month..........4 Less than once per month..........5

15. How frequently is your home vacuumed?	Once per day..........1 Once per week..........2 Once every two weeks..........3 Once per month..........4 Less than once per month..........5
16. Is your vacuum in good working order?	Yes..........1 No..........2 Don't know..........9
17. Do you have a gas-powered lawn mower?	Yes..........1 No..........2 **IF NO, GO TO 20**
18. If yes, how many times a month do you use this mower?	Number of months ________
19. Do you have any pets, such as dogs or cats or other furry animals, which usually spend some time each day in your home?	Yes..........1 No..........2 **IF NO, GO TO 23**
20. If so, how many are there?	One..........1 Two..........2 Three to five..........3 More than five..........4 Not applicable..........0
21. Could you tell me which pets you have? **Circle all that apply**	Cat(s)..........1 Dog(s)..........2 Bird(s)..........3 Other, please specify...8 ________________ Not applicable..........0
22. How many people regularly smoke inside your home?	0..........1 1..........2 2..........3 3 or more..........4
23. Did anyone smoke any tobacco products in your home during the last seven days?	Yes..........1 No..........2 Don't Know..........9

Participant ID:

IV. HOUSEHOLD RESIDENTS:

Finally, I would like to ask you a few questions about people currently living in this house.

24. How many people currently live in this home?	Number of People ( )
25. Do any of the people currently living here smoke at least 1 cigarette, cigar or pipeful per day?	Yes.............................1 No..............................2

THANK YOU VERY MUCH FOR YOUR TIME AND COOPERATION!!!

Participant ID:

INTERVIEWER, RECORD THE FOLLOWING QUESTIONS BY OBSERVATION:

26. How many floors of living space are there in this household?	One.............................1 Two.............................2 Three...........................3 More than three..........4
27. How many units are in the building?	One.............................1 Two.............................2 Three...........................3 Four to six..................4 More than six..............5
28. Is the house located within 100 yards of a busy roadway	YES.............................1 NO..............................2
29. Is there a dirt drive located within 100 yards of this house?	YES.............................1 NO..............................2
30. Are there any other sources of dirt located within 100 yards of the house?	YES.............................1 NO..............................2 If the answer is YES, please specify : __________________

List each room in the house and estimate the percentage of usable floor space covered by rugs or carpets.

	31. Room	32. % of floor covered by rug or carpet
1		
2		
3		
4		
5		
6		
7		

Participant ID:

33. How would you best describe the VENTILATION FACTOR in this unit? **Codes from 0 to 3: 0.5 is fresh** **2.5 is stuffy**	________
34. How would you best describe the DUST FACTOR in this unit? **Codes from 0 to 3: 0.5 is immaculate** **2.5 is clutter**	________

END OF OBSERVATIONS BY INTERVIEWER

Appendix 5 - Field Manual and Lab Operating Procedures

Field Manual for Prince George Study

Standard Operating Procedure for PEM Cleaning, Assembly and Disassembly

A5.1. Field Manual for Prince George Study

(Edited Version of EPA Study Field Manual Obtained for Harvard School of Public Health)

This manual is intended for use by field technicians, each technician has specific tasks to complete and the manual is sub-divided accordingly.

A5.1.1. Prior to Sampling

Set up all pumps to run for 24 hours before set up day, ensure that there is an in-line filter to prevent foreign bodies entering the pump. Continue to run the pump either on-site after set up or in the lab until the first day of sampling.

Charge the BGI pump batteries for at least 16 hours prior to the setup. Also charge a backup battery for each field technician if possible.

A5.1.2. Prior to Visit

Check contents of tool-box for:

Small screwdriver for BGI pump
Wire cutters
Paperclip
Small plastic wire ties
Extra tubing
Ethanol
Buck soap
Pens
Kimwipes
Spare o'ring for PEM
Large screwdriver
Needlenose pliers
Large plastic wire ties
Paper tape
Tubing cutter or razor blade
Buck calibrator
Charger for Buck
Forceps
Calibration caps

Check contents of crate for needed supplies
Batteries
Back-up batteries
Field log book and participant's folder

Pickup laptop and cables if it is your day to check HOBO.

Check that all samplers are in bags.

A5.1.3. Technician for Personal Monitoring Setup

Week 1 – First Sampling Day

Before leaving the lab:

Check for all required equipment, samplers, pumps and batteries.
Pick up the laptop and cables if needed.

At the school;

Proceed to the rooftop monitor or student's classroom according to sampling schedule.

<u>Rooftop monitor:</u>

Pump should be running for the previous 24 hours. Remove dummy PEM and attach new PEM to pump and measure flow rate with equilibrated rotameter. Recheck with Bios or Buck flow meters if available. Adjust flow if too high or low. Record sample ID, flowrate (original and adjusted), pump count and watch time as well as any visual observations of the weather conditions.

Check over log book entries and make sure that all details are completed i.e. date etc.

Launch Temperature Hobo.

<u>Personal Monitor:</u>

Replace the battery for the pump, note the ID in the log book.

Replace the PEM making sure to record the ID number. Remove one label from the back of the PEM and place it into the log book.

Note watch time and reset the pump count using a paperclip, note the pump time.

Check the flow of the PEM, if the overall flow is too high or too low then adjust the BGI pump. Once the PEM is adjusted note the new flow value into the log book.

Check over the log book and make sure that all details are completed i.e. date etc.

Motion Hobos will have been previously launched in the lab.

Remind all study participants that you will be at their school at the designated time each morning they are carrying the sampler. Give the participant and teacher an updated schedule with the dates of the study and try to run through any potential problems with the teacher and students so that alternative times can be arranged.

Talk to the participants to make sure they understand the study and to establish a good relationship.

Fit the personal back-pack to the participant of the day and fix the elutriator within the breathing zone.

Go through the Time Activity Diary and show the participant the sample version, explain the importance of the diary and the kind of information that is required.

Week 1 – Sampling Days 2-4

Check off-flows for PEM and record in logbook along with pump count and time. Repeat same procedure as in day 1.

Go over time-activity diaries with participant from the previous day and go through follow-up questions. Download the Motion Hobo and compare with TAD if you have the laptop, any discrepancy between the two should be discussed with the participant. Reset the Motion Hobo and place into back-pack.

Fit the personal back-pack to the participant of the day and fix the elutriator within the breathing zone.

Again go through the Time Activity Diary and show the participant the sample version, explain the importance of the diary and the kind of information that is required.

At roof-top monitor download Temperature Hobo and reset.

Week 1 to 6 – End of sampling, breakdown day

Check off-flows for all PEMs. Remove the PEMs and attach the dummy PEMs at the correct flow rate.

Go over time-activity diaries with participants.

Download the Motion Hobo from the personal sampler and the Temperature Hobo from outside.

Give out reward to participants for the week completed and extra bonus for the best diary. Remind the first participant for the next week that you will be at the school Monday morning to give them the sampler.

Weeks 2 to 6

Repeat the same procedure outlined for week 1. Make sure to answer any questions and concerns of the participants and try to get to know each individual. Ask them about their day and develop a good relationship. Always be positive and remind them that they are helping in an important research study.

A5.1.4. Laboratory Technician

Make sure pumps are labeled and running for the previous 24 hours.

Load cleaned samplers with filters and leak test all PEMs at their respective flow rate – using a non-flow control pump with a valve, adjust the flow and measure using a Bios calibrator. Then attach the PEM and measure the flow again using a calibration cap. There should be less than 5% difference in flows, given the low pressure drop of the PEMs.

At the end of each week perform a deep-cleaning of all of the PEMs. Follow the attached standard operating procedure from Harvard School of Public Health for PEM cleaning, assembly and disassembly.

A5.2. Standard Operating Procedure for PEM Cleaning, Assembly and Disassembly

Procedures adapted from SOP for PEMs Revision #3 (3/30/00) Obtained from Harvard School of Public Health Environmental Science & Engineering Program

Original Procedure Prepared by Kathleen Ward Brown

A5.2.1. Cleaning and Assembly Procedure

A5.2.1.1. PEMs Deep Cleaning

(at beginning of study and then weekly)

Harvard PEMs (H-PEMs)

Materials

5 large beakers
Distilled water
Non-serrated forceps
Large Kimwipes
H-PEM tops
H-PEM impaction plates
4 to 5 plastic trays
Permanent marker
Powder-free latex gloves
Ethanol
Mild dish detergent
H-PEM O-rings
H-PEM bases
Metal screens
Paper tape

<u>Harvard PEM Tops, Bases and Impactor Plates:</u>

a) Remove any remaining grease from impactor plates using a small spatula.

b) Label two beakers with paper tape using permanent marker to denote contents as "soap and water."

c) Fill both beakers with distilled water and add several drops of a mild dish detergent.

d) Wash bases and tops in one of the beakers, using a brush to ensure that the H-PEM inlets are clear.

e) Wash the impactor plates in the second beaker, using a brush to ensure that as much grease is removed from the impactor plates as possible.

f) Rinse contents of beaker twice (or until water is free of soap) in clean distilled water.

g) To dry H-PEM tops, bases, and impactor plates rapidly, shake excess water from them, then rinse in ethyl alcohol. Place the tops and bases onto trays covered in Kimwipes; cover with an additional layer of Kimwipes. Mark tray as "Clean PEM Parts."

Harvard PEM Screens and O-rings (note screens are same as for SKC PEMs):

a) Place metal screens and Harvard O-rings in a third beaker filled with ethanol. Allow to soak for several minutes. Remove screens from the ethanol and lay flat on a tray. Using a brush and more ethanol, scrub both sides of the screens. Place the scrubbed screens in a fourth beaker filled with distilled water and several drops of mild dish detergent. Carefully agitate contents of beaker using a brush, being careful to avoid bending the screens.

b) To clean O-rings, place in a fifth beaker with distilled water and a few drops of mild dish detergent. Carefully agitate contents of beaker using a brush. (SKC O-rings can be differentiated from H-PEM O-rings, as SKC O-rings are either brown or orange, while H-PEM O-rings are black and smaller than the SKC O-rings.)

c) Rinse contents of beakers twice (or until water is free of soap) in clean distilled water.

d) Remove metal screens and O-rings from the drained beakers using non-serrated forceps and place on trays covered with Kimwipes. Allow O-rings to dry naturally; do not place into alcohol. Screens can be rinsed a final time in ethanol prior to drying on Kimwipes. Cover with additional Kimwipes while drying. Mark tray as "Clean PEM Parts."

A5.2.1.2. PEMs Standard Cleaning

(done daily during study)

Harvard PEMs

Harvard PEM Bases, Tops and Screens:

a.) Wipe off bases and tops with a Milli-Q dampened Kimwipe and allow to dry on a tray between Kimwipes.

b.) Screens and O-rings are cleaned following the same procedures as for deep cleaning, above.

Harvard PEM Impaction Plates:

a.) Using a small spatula, remove the top layer of grease from the impaction surface. Remaining grease does not need to be removed. This prepares the impaction surface for re-greasing.

A5.2.1.5. PEMs Assembly

Harvard PEMs Assembly (for use with 37mm Teflon filters)

Materials:

Plastic or Teflon-coated forceps
Trace metals-grade methanol
Clean and oiled impaction plates
Clean metal screens
37-mm Teflon filters
Drain disks (Whatman #230800)
Drain disk rings
Harvard PEM screws (4 per PEM)
Milli-Q water
Clean tops
Clean bases
Clean O-rings
3 plastic trays
Razor blades
Phillips screwdriver
Large foam swabs

a.) The impaction surface should be re-greased with Dow Corning – High Vacuum Grease, using a small spatula. The grease should be smoothed with a razor blade so that it is even with the impactor surface (for the PM$_{2.5}$ impactor plates, a razor blade cut in half works best and for the PM$_{10}$ impactor plates, ¾ of a razor blade works best). Make sure grease is very smooth. Use large foam swab to clean excess grease from around impactor surface. Greased impaction plates should be taped in stacks with the greased sides facing together and stored in a sealed container until ready for use.

b.) Pair up the Harvard PEM bases and tops. See Figure 2 for details of the components.

c.) Using non-serrated forceps, place the O-ring into the lip on the base of the Harvard PEM. Place a metal support screen for the filter into the base. The screen may be inserted with either side facing the PEM base. Care should be taken to avoid using bent or warped screens.

d.) **For PM$_{2.5}$:** Wash plastic or Teflon-coated forceps with Milli-Q water then rinse with trace-metals grade methanol to dry.

e.) **For PM$_{2.5}$**: Using the clean plastic or Teflon-coated forceps, remove a drain disk from the pack marked "PM$_{2.5}$" and place on top of the metal screen. Then use the plastic forceps to remove a 37-mm Teflon filter from the Petri dish by the plastic outer-ring. Pass both sides of the filter over the ^{210}Po source to remove static. Place the filter on top of the drain disk in the PEM base with the shiny outer-ring side facing up.

f.) **For $PM_{2.5}$**: Using the plastic forceps, place a drain disk ring on top of the Teflon filter so that it just covers the shiny outer ring of the filter.

g.) Inspect the impaction plate to make sure grease is smooth. Then place the impaction ring on top of the Teflon filter with the greased side facing up.

h.) Put a $PM_{2.5}$ inlet on top of the impaction plate, and secure the top and base together using four screws and a Phillips screwdriver, ensuring that all four are tightened evenly.

A5.2.2. QA/QC

A5.2.2.1. Leak Test Procedures

SKC PEM, Harvard PEM and Mini-PEMs

a.) After running for 30 minutes, the initial flow of a non-flow-controlled pump is measured using a NIST-traceable Buck Soap Bubble Calibrator. The valve should be adjusted to set the flow rate to which the PEM or mini-PEM will be operated. This flow is measured twice and recorded.

b.) Attach a calibration cap to the PEM with the other end attached to the Buck calibrator. Then attach the PEM directly to the pump. Check the flow rate for each of the PEMs after assembly. If there are no leaks, the flow should be within 5% of the initial pump flow. If not, check the tightness of the PEM screws; if they are too loose, leakage will occur. If this does not work, then open the PEM and check the O-ring placement, as this may have moved. For mini-PEMs, try tightening the connector ring or use the wrench to tighten the inlet.

c.) For mini-PEMs, leaks can most often be remedied by using a small amount of Teflon tape on the threaded ring for a better seal.

A5.2.2.2. After Deployment in the Field

a.) In the field, the initial flows should be within 5% of target limits. If the flow rate is not within the required range, then valves must be adjusted.

b.) PEMs are transported in resealable bags to and from the field.

c.) PEMs tops and bases are unscrewed and the filter removed with clean forceps—non-serrated, flat forceps for ECOC or sulfate, and plastic or Teflon-coated forceps for PM2.5 (washed with Milli-Q and trace-metals-grade methanol).

d.) Each sample is placed in a Petri dish that is taped closed.

e.) Petri dishes containing Teflon filters are stored in the refrigerator; Petri dishes containing quartz filters are wrapped in aluminum foil and stored in the freezer until ready for analysis.

f.) Shipping of samples is done in coolers with blue ice by priority overnight mail.

A5.2.3. References

MSP Corporation, Model 200 Personal Environmental Monitor Instruction Manual Rev. October 1992.

Appendix 6 – Individual Regression Summaries

Summary of Individual Longitudinal Regression Results (Based on Sulphate Estimates)

Summary of Individual Longitudinal Regression Results (Based on Elemental Carbon Estimates)

Summary of Individual Longitudinal Spearman Correlations (Sulphate and Elemental Carbon Based Results)

A6.1. Summary of Individual Longitudinal Regression Results (SO_4^{2-})

Table A6.1-1 $PM_{2.5}$ ambient generated personal exposure versus $PM_{2.5}$ ambient concentration at the closest school site: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI=95.

E_{ag} vs C_a (SO_4^{2-})	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	10	0.56	0.07	2.41	1.91	0.95	0.11	0.000
1002	9	0.46	0.05	1.80	1.27	0.96	0.11	0.000
1003	10	0.48	0.02	0.81	0.59	0.99	0.05	0.000
2001	9	0.46	0.04	0.40	0.87	0.98	0.08	0.000
2002	10	0.29	0.04	1.85	0.73	0.94	0.12	0.000
2003	10	0.33	0.03	1.03	0.62	0.97	0.08	0.000
3001	10	0.39	0.05	2.20	1.15	0.95	0.11	0.000
3002	8	0.38	0.05	1.56	1.25	0.95	0.13	0.000
3003	9	0.40	0.05	1.26	1.10	0.96	0.11	0.000
4001	10	0.39	0.12	5.12	2.92	0.77	0.23	0.010
4002	7	0.52	0.04	0.05	0.63	0.98	0.08	0.000
4003	10	0.36	0.06	2.00	1.40	0.90	0.15	0.000
5001	10	0.45	0.14	2.92	3.19	0.76	0.23	0.011
5002	9	0.64	0.09	0.32	1.15	0.94	0.13	0.000
5003	10	0.52	0.08	0.49	1.59	0.92	0.14	0.000
Mean		0.44	0.06	1.61	1.36	0.93	0.12	
SD		0.09	0.03	1.29	0.78	0.07	0.05	
95%CI		0.05	0.02	0.65	0.40	0.04	0.03	
Median		0.45	0.05	1.56	1.15	0.95	0.11	
25th Percentile		0.38	0.04	0.65	0.80	0.93	0.09	
75th Percentile		0.50	0.07	2.10	1.49	0.97	0.13	

Table A6.1-2 $PM_{2.5}$ ambient generated personal exposure versus ambient concentration from the Ministry of Water, Land and Air Protection $PM_{2.5}$ TEOM monitor: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI=95.

E_{ag} vs TEOM (SO_4^{2-})	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	8	0.69	0.08	3.81	1.99	0.96	0.12	0.000
1002	9	0.61	0.07	1.80	1.39	0.95	0.12	0.000
1003	10	0.59	0.05	1.81	1.01	0.97	0.08	0.000
2001	7	0.45	0.06	1.59	1.36	0.96	0.12	0.000
2002	10	0.36	0.06	0.97	1.03	0.91	0.14	0.000
2003	10	0.36	0.06	1.43	1.21	0.90	0.15	0.000
3001	9	0.42	0.08	2.50	1.78	0.89	0.17	0.001
3002	8	0.57	0.05	-0.50	0.86	0.98	0.08	0.000
3003	8	0.35	0.08	2.65	1.78	0.87	0.20	0.005
4001	9	0.41	0.13	6.47	2.92	0.77	0.24	0.014
4002	6	0.56	0.06	0.69	0.66	0.98	0.10	0.001
4003	10	0.34	0.08	3.60	1.63	0.83	0.20	0.003
5001	8	0.35	0.25	4.41	5.81	0.50	0.35	0.205
5002	9	0.57	0.11	-0.11	1.73	0.88	0.18	0.002
5003	10	0.51	0.11	0.74	2.16	0.86	0.18	0.002
Mean		0.48	0.09	2.12	1.82	0.88	0.16	
SD		0.12	0.05	1.85	1.24	0.12	0.07	
95%CI		0.06	0.03	0.93	0.63	0.06	0.04	
Median		0.45	0.08	1.80	1.63	0.90	0.15	
25th Percentile		0.36	0.06	0.85	1.12	0.87	0.12	
75th Percentile		0.57	0.10	3.12	1.88	0.96	0.19	

Table A6.1-3 PM$_{2.5}$ ambient generated personal exposure versus total PM$_{2.5}$ personal exposure: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI=95. *(2 extreme outliers removed for subject 1002.)

E_{ag} vs E_T (SO_4^{2-})	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	10	0.93	0.14	-0.35	2.81	0.92	0.14	0.000
1002	9	-0.04	0.04	14.38	3.82	-0.35	0.35	0.360
	(7)*	(0.19)*	(0.17)*	(4.99)*	(7.43)*	(0.46)*	(0.40)*	(0.366)*
1003	10	0.49	0.18	1.78	3.81	0.69	0.26	0.027
2001	9	0.67	0.21	-2.82	4.08	0.78	0.24	0.014
2002	10	0.08	0.09	4.80	2.31	0.29	0.34	0.425
2003	10	0.85	0.32	-4.87	4.52	0.68	0.26	0.029
3001	10	0.06	0.08	8.29	2.95	0.26	0.34	0.476
3002	8	1.04	0.20	-5.01	2.77	0.90	0.18	0.002
3003	9	0.78	0.37	0.94	4.22	0.62	0.30	0.073
4001	10	0.08	0.17	11.56	4.72	0.16	0.35	0.662
4002	7	0.50	0.45	0.55	5.70	0.45	0.40	0.317
4003	10	0.35	0.25	1.67	5.15	0.44	0.32	0.199
5001	10	0.58	0.36	-0.75	7.90	0.50	0.31	0.143
5002	9	0.52	0.19	-1.80	3.52	0.72	0.26	0.028
5003	10	1.42	0.45	-10.00	6.03	0.74	0.24	0.014
Mean		0.55	0.23	1.23	4.29	0.52	0.28	
SD		0.41	0.13	6.42	1.47	0.33	0.07	
95%CI		0.21	0.07	3.25	0.75	0.17	0.04	
Median		0.52	0.20	0.55	4.08	0.62	0.30	
25th Percentile		0.22	0.16	-2.31	3.24	0.36	0.25	
75th Percentile		0.82	0.34	3.29	4.94	0.73	0.34	

Table A6.1-4 $PM_{2.5}$ non-ambient generated personal exposure versus total $PM_{2.5}$ personal exposure: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI=95. *(2 extreme outliers removed for subject 1002.)

E_{nag} vs E_T (SO_4^{2-})	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	10	0.07	0.14	0.35	2.81	0.17	0.35	0.633
1002	9	1.04	0.04	-14.38	3.82	0.99	0.04	0.000
	(7)*	(.91)*	(0.19)*	(-4.99)*	(7.43)*	(0.91)*	(0.19)*	(0.005)*
1003	10	0.51	0.18	-1.78	3.81	0.71	0.25	0.022
2001	9	0.33	0.21	2.82	4.08	0.52	0.32	0.151
2002	10	0.92	0.09	-4.80	2.31	0.96	0.09	0.000
2003	10	0.15	0.32	4.87	4.52	0.16	0.35	0.655
3001	10	0.94	0.08	-8.29	2.95	0.97	0.09	0.000
3002	8	-0.04	0.20	5.01	2.77	-0.09	0.41	0.838
3003	9	0.22	0.37	-0.94	4.22	0.22	0.37	0.575
4001	10	0.92	0.17	-11.56	4.72	0.89	0.16	0.001
4002	7	0.50	0.45	-0.55	5.70	0.44	0.40	0.319
4003	10	0.65	0.25	-1.67	5.15	0.67	0.26	0.034
5001	10	0.42	0.36	0.75	7.90	0.38	0.33	0.282
5002	9	0.48	0.19	1.80	3.52	0.69	0.27	0.040
5003	10	-0.42	0.45	10.00	6.03	-0.31	0.34	0.382
Mean		0.45	0.23	-1.23	4.29	0.49	0.27	
SD		0.41	0.13	6.42	1.47	0.40	0.12	
95%CI		0.21	0.07	3.25	0.75	0.20	0.06	
Median		0.48	0.20	-0.55	4.08	0.52	0.32	
25th Percentile		0.18	0.16	-3.29	3.24	0.19	0.21	
75th Percentile		0.78	0.34	2.31	4.94	0.80	0.35	

A6.2. Summary of Individual Longitudinal Regression Results (EC)

Table A6.2-1 $PM_{2.5}$ ambient generated personal exposure versus $PM_{2.5}$ ambient concentration at the closest school site: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI= confidence interval.

E_{ag} vs C_a (EC)	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	10	0.45	0.06	1.76	1.82	0.93	0.13	0.000
1002	9	0.55	0.10	1.51	2.56	0.90	0.17	0.001
1003	10	0.50	0.03	0.29	0.88	0.98	0.07	0.000
2001	9	0.48	0.06	0.99	1.37	0.95	0.11	0.000
2002	10	0.39	0.03	2.10	0.62	0.98	0.08	0.000
2003	10	0.36	0.02	1.27	0.46	0.99	0.05	0.000
3001	7	0.67	0.14	-1.44	3.09	0.91	0.19	0.005
3002	7	0.51	0.07	1.89	1.83	0.95	0.13	0.001
3003	7	0.36	0.10	0.06	2.66	0.85	0.23	0.014
4001	9	0.25	0.05	5.66	1.32	0.88	0.18	0.002
4002	7	0.03	0.12	4.74	1.75	0.11	0.44	0.807
4003	8	0.63	0.07	-2.42	1.78	0.97	0.11	0.000
5001	2							
5002	7	0.41	0.26	3.80	3.99	0.57	0.37	0.181
5003	8	0.61	0.07	0.44	1.41	0.96	0.11	0.000
Mean		0.44	0.08	1.47	1.82	0.85	0.17	
SD		0.17	0.06	2.19	0.98	0.24	0.11	
95%CI		0.08	0.03	1.11	0.49	0.12	0.06	
Median		0.46	0.07	1.39	1.76	0.94	0.13	
25th Percentile		0.37	0.05	0.32	1.33	0.89	0.11	
75th Percentile		0.54	0.10	2.05	2.38	0.97	0.19	

Table A6.2-2 $PM_{2.5}$ ambient generated personal exposure versus ambient concentration from the Ministry of Water, Land and Air Protection $PM_{2.5}$ TEOM monitor: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI= confidence interval.

E_{ag} vs TEOM (EC)	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	8	0.49	0.08	4.90	1.97	0.92	0.16	0.001
1002	9	0.71	0.15	1.68	2.79	0.88	0.37	0.002
1003	10	0.62	0.05	1.20	0.95	0.98	0.25	0.000
2001	7	0.51	0.07	0.89	1.69	0.96	0.26	0.001
2002	10	0.47	0.07	1.07	1.21	0.93	0.34	0.000
2003	10	0.40	0.05	1.57	1.07	0.93	0.29	0.000
3001	6	0.74	0.24	0.15	4.47	0.84	0.44	0.036
3002	7	0.73	0.13	-0.28	2.55	0.93	0.14	0.002
3003	6	0.37	0.07	1.29	1.90	0.93	0.29	0.007
4001	8	0.23	0.08	7.30	1.84	0.77	0.38	0.026
4002	6	0.06	0.16	4.62	1.82	0.19	0.44	0.712
4003	8	0.58	0.10	1.39	2.18	0.92	0.35	0.001
5001	2							
5002	7	0.42	0.19	3.36	3.22	0.69	0.26	0.084
5003	8	0.69	0.10	-0.39	1.87	0.94	0.28	0.000
Mean		0.50	0.11	2.05	2.11	0.84	0.30	
SD		0.20	0.06	2.21	0.93	0.20	0.09	
95%CI		0.10	0.03	1.12	0.47	0.10	0.05	
Median		0.50	0.09	1.34	1.88	0.93	0.29	
25th Percentile		0.40	0.07	0.93	1.73	0.85	0.26	
75th Percentile		0.67	0.14	2.94	2.46	0.93	0.36	

Table A6.2-3 PM$_{2.5}$ ambient generated personal exposure versus total PM$_{2.5}$ personal exposure: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI= confidence interval. *(2 extreme outliers removed for subject 1002.)

E_{ag} vs E_T (EC)	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	10	0.73	0.13	-0.31	2.63	0.89	0.16	0.001
1002	9	-0.04	0.06	15.68	5.00	-0.25	0.37	0.509
	(7)*	(0.20)*	(0.24)*	(6.28)*	(10.56)*	(0.35)*	(0.42)*	(0.443)*
1003	10	0.53	0.18	0.96	3.88	0.71	0.25	0.021
2001	9	0.67	0.24	-1.86	4.76	0.73	0.26	0.027
2002	10	0.07	0.12	6.49	2.99	0.22	0.34	0.537
2003	10	0.76	0.39	-2.90	5.49	0.57	0.29	0.088
3001	7	-0.03	0.12	12.62	4.20	-0.11	0.44	0.822
3002	7	1.38	0.21	-5.87	2.94	0.95	0.14	0.001
3003	7	0.83	0.32	-0.67	3.93	0.76	0.29	0.048
4001	9	-0.02	0.10	11.50	2.87	-0.06	0.38	0.888
4002	7	0.11	0.25	3.85	3.14	0.19	0.44	0.690
4003	8	0.62	0.44	-0.90	9.07	0.50	0.35	0.205
5001	2							
5002	7	0.57	0.18	-0.76	3.51	0.81	0.26	0.027
5003	8	1.53	0.61	-9.99	7.97	0.72	0.28	0.045
Mean		0.55	0.24	1.99	4.46	0.47	0.30	
SD		0.50	0.15	7.27	1.93	0.40	0.09	
95%CI		0.25	0.08	3.68	0.98	0.20	0.05	
Median		0.59	0.20	-0.49	3.91	0.64	0.29	
25th Percentile		0.08	0.12	-1.62	3.03	0.20	0.26	
75th Percentile		0.75	0.30	5.83	4.94	0.75	0.36	

Table A6.2-4 PM$_{2.5}$ non-ambient generated personal exposure versus total PM$_{2.5}$ personal exposure: individual regression summary showing slope, intercept, Pearson R and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI= confidence interval. *(2 extreme outliers removed for subject 1002.)

Enag vs Et	N	Slope	Slope Error	Intercept	Intercept Error	R	R Error	p
1001	10	0.27	0.13	0.31	2.63	0.58	0.29	0.077
1002	9	1.04	0.06	-15.68	5.00	0.99	0.05	0.000
	(7)*	(0.80)*	(0.24)*	(-6.28)*	(10.56)*	(0.83)*	(0.25)*	(0.020)*
1003	10	0.47	0.18	-0.96	3.88	0.67	0.26	0.033
2001	9	0.33	0.24	1.86	4.76	0.46	0.34	0.211
2002	10	0.93	0.12	-6.49	2.99	0.94	0.12	0.000
2003	10	0.24	0.39	2.90	5.49	0.21	0.35	0.552
3001	7	1.03	0.12	-12.62	4.20	0.97	0.11	0.000
3002	7	-0.38	0.21	5.87	2.94	-0.63	0.35	0.127
3003	7	0.17	0.32	0.67	3.93	0.23	0.44	0.626
4001	9	1.02	0.10	-11.50	2.87	0.96	0.10	0.000
4002	7	0.89	0.25	-3.85	3.14	0.85	0.24	0.015
4003	8	0.38	0.44	0.90	9.07	0.33	0.38	0.418
5001	2							
5002	7	0.43	0.18	0.76	3.51	0.72	0.31	0.066
5003	8	-0.53	0.61	9.99	7.97	-0.34	0.38	0.413
Mean		0.45	0.24	-1.99	4.46	0.50	0.26	
SD		0.50	0.15	7.27	1.93	0.50	0.12	
95%CI		0.25	0.08	3.68	0.98	0.25	0.06	
Median		0.41	0.20	0.49	3.91	0.63	0.30	
25th Percentile		0.25	0.12	-5.83	3.03	0.25	0.15	
75th Percentile		0.92	0.30	1.62	4.94	0.92	0.35	

A6.3. Summary of Individual Longitudinal Spearman Correlations

Table A6.3-1 $PM_{2.5}$ ambient generated personal exposure versus $PM_{2.5}$ ambient concentration at the closest school site and the Ministry of Water, Land and Air Protection TEOM: individual correlation summary showing Spearman r and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI=confidence interval. *(2 extreme outliers removed for subject 1002.)

Spearman r Summary	SO_4^{2-} Based Estimate						EC Based Estimate					
	Eag vs Ca			Eag vs TEOM			Eag vs Ca			Eag vs TEOM		
	N	r	p	N	r	p	N	r	p	N	r	p
1001	10	0.94	0.000	8	0.90	0.002	10	0.67	0.033	8	0.76	0.028
1002	9	0.92	0.001	9	0.87	0.002	9	0.85	0.004	9	0.77	0.016
1003	10	0.92	0.001	10	0.98	0.000	10	0.88	0.001	10	0.90	0.000
2001	9	0.93	0.000	7	1.00	0.000	9	0.82	0.007	7	0.79	0.036
2002	10	0.98	0.000	10	0.94	0.000	10	0.98	0.000	10	0.93	0.000
2003	10	0.94	0.000	10	0.83	0.003	10	0.88	0.001	10	0.82	0.004
3001	10	0.99	0.000	9	0.93	0.000	7	0.86	0.014	6	0.77	0.072
3002	8	0.98	0.000	8	0.95	0.000	7	0.86	0.014	7	0.82	0.023
3003	9	0.90	0.001	8	0.81	0.015	7	0.86	0.014	6	0.89	0.019
4001	10	0.88	0.001	9	0.88	0.002	9	0.88	0.002	8	0.90	0.002
4002	7	0.89	0.007	6	0.94	0.005	7	-0.14	0.760	6	-0.14	0.787
4003	10	0.82	0.004	10	0.78	0.008	8	0.90	0.002	8	0.81	0.015
5001	10	0.85	0.002	8	0.69	0.058	2			0		
5002	9	0.90	0.001	9	0.80	0.010	7	0.43	0.337	7	0.54	0.215
5003	10	0.82	0.004	10	0.64	0.048	8	0.62	0.102	8	0.90	0.002
Mean		0.91			0.86			0.74			0.75	
SD		0.05			0.11			0.29			0.27	
95%CI		0.03			0.05			0.15			0.14	
Median		0.92			0.88			0.86			0.81	
25th Percentile		0.89			0.80			0.71			0.77	
75th Percentile		0.94			0.94			0.88			0.90	

Table A6.3-2 $PM_{2.5}$ ambient and non-ambient generated personal exposure and versus total $PM_{2.5}$ personal exposure: individual correlation summary showing Spearman r and p-value. Summary statistics for the individual results are also reported. Mean=arithmetic mean; SD=standard deviation; CI= confidence interval. *(2 extreme outliers removed for subject 1002.)

Spearman r Summary		SO_4^{2-} Based Estimate					EC Based Estimate			
		Eag vs ET		Enag vs ET			Eag vs ET		Enag vs ET	
	N	r	p-value	r	p-value	N	r	p-value	r	p-value
1001	10	0.77	0.009	0.42	0.229	10	0.76	0.011	0.52	0.128
1002	9	0.18	0.637	0.88	0.002	9	0.08	0.831	0.72	0.030
	(7)*	(0.68)*	(0.094)*	(0.75)*	(0.052)*	(7)*	(0.46)*	(0.294)*	(0.39)*	(0.383)*
1003	10	0.78	0.008	0.21	0.556	10	0.62	0.054	0.41	0.244
2001	9	0.80	0.010	0.57	0.112	9	0.57	0.112	0.55	0.125
2002	10	0.60	0.067	0.62	0.054	10	0.43	0.214	0.66	0.038
2003	10	0.67	0.033	-0.20	0.580	10	0.65	0.043	-0.04	0.907
3001	10	0.44	0.200	0.57	0.112	10	0.50	0.253	0.55	0.125
3002	8	0.74	0.037	0.02	0.955	8	0.86	0.014	-0.64	0.119
3003	9	0.52	0.154	0.13	0.732	9	0.71	0.071	0.00	1.000
4001	10	0.20	0.580	0.67	0.033	10	0.07	0.865	0.87	0.002
4002	7	0.64	0.119	0.32	0.482	7	0.04	0.939	0.93	0.003
4003	10	0.42	0.229	0.68	0.029	10	0.36	0.385	0.12	0.779
5001	10	0.71	0.022	0.26	0.467	10	0.00	0.000	0.00	0.000
5002	9	0.60	0.088	0.40	0.286	9	0.39	0.383	0.46	0.294
5003	10	0.54	0.108	0.07	0.855	10	0.36	0.385	0.07	0.867
Mean		0.57		0.38			0.43		0.34	
SD		0.19		0.30			0.14		0.21	
95%CI		0.10		0.15			0.28		0.42	
Median		0.60		0.40			0.43		0.46	
25th Percentile		0.48		0.17			0.22		0.04	
75th Percentile		0.72		0.60			0.64		0.61	