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1. INTRODUCTION

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 and residential and commercial heating (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 Canada-Wide Standard for PM_{2.5} (3-year average of 98th percentile greater than 30 µg/m³) was almost exceeded 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 µg/m³ respectively (Lamble *et al*, 2002 and Fudge *et al*, 2003). In both 2000 and 2001, the 30 µg/m³ level was exceeded with single-year annual 98th percentiles of 32.1 and 32.5 µg/m³.

A multitude of epidemiological studies have demonstrated that a significant health effect exists for atmospheric fine particulate matter (Pope, 2000 and Vedal, 1997). There have also been many studies performed regarding personal exposure to particulate matter and it has been shown that the relationship between ambient levels of particulate matter and individual exposure can be quite variable between individuals and communities (Mage and Buckley, 1995). The relationship between personal exposure and ambient particulate level is site specific and should be further investigated in areas where high ambient levels are observed and health impacts are suspected.

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. Longitudinal PM_{2.5} exposure studies have shown stronger personal-outdoor correlations do exist when data are analyzed by individual over time but the degree of association varies widely by individual (Ebel *et al*, 2000; Janssen *et al*, 1999, 2000; Liu *et al*, 2003; and Sarnat *et al*, 2000). Personal-outdoor associations have been shown to be even stronger for the sulphate (SO₄²⁻) component of fine

particles indicating that it may be a more appropriate measure of exposure to ambient generated particles due to the absence of indoor and personal sources (Ebel *et al*, 2000 and Sarnat *et al*, 2000). Use of the absorption coefficient of PM_{2.5} filters, a measure of light absorbing carbon and a surrogate measure for the elemental carbon of PM, has also lead to a stronger association between personal exposure and ambient concentrations (Janssen *et al*, 2000). The use of indicators of ambient exposure such as sulphate or light absorbing carbon may help to better characterize the true personal/ambient relationship.

A field study to assess the personal-ambient relationship, spatial variation and meteorological influence for fine particulate matter took place in Prince George during the winter of 2001 from February 5th to March 16th. This paper compares ambient data to personal exposure measures from elementary school children for total PM_{2.5}, as well as the sulphate and light absorbing components. Spatial variation in ambient levels is discussed and the influence of local meteorology is investigated including the role of thermal inversions on high ambient concentrations and the subsequent impact on personal exposures. Longitudinal personal to ambient associations for each subject are determined to account for the large variability between individuals and estimates of ambient generated exposure are calculated and compared to both ambient concentrations and total personal exposures. An extensive description of methods, data analysis and results can be found in Noullett, 2004.

2. METHODS

Five temporary ambient stations were set up to monitor PM_{2.5} levels on the roof of five elementary schools where personal exposure monitoring was being performed. Figure 1 shows the city of Prince George and the location of the school ambient monitoring stations and other air quality monitoring and meteorological sites. Both ambient and personal exposure samples were collected at each school, on week days only for the 6-week duration of the study. The personal samplers were rotated between 3 students (aged 10 to 12 years) at each school so that each student completed 10 monitoring sessions intermittently over the 6-week period. PM_{2.5} Harvard Personal Environment Monitors (HPEM_{2.5}) were used for both the ambient and personal sampling to collect a 24-hour integrated sample on Teflon filters. For each personal exposure sample the children recorded their activity in a diary every 30 minutes. Data from the BC Ministry of Water, Land and Air Protection TEOM PM_{2.5} sampler were also used in this study.

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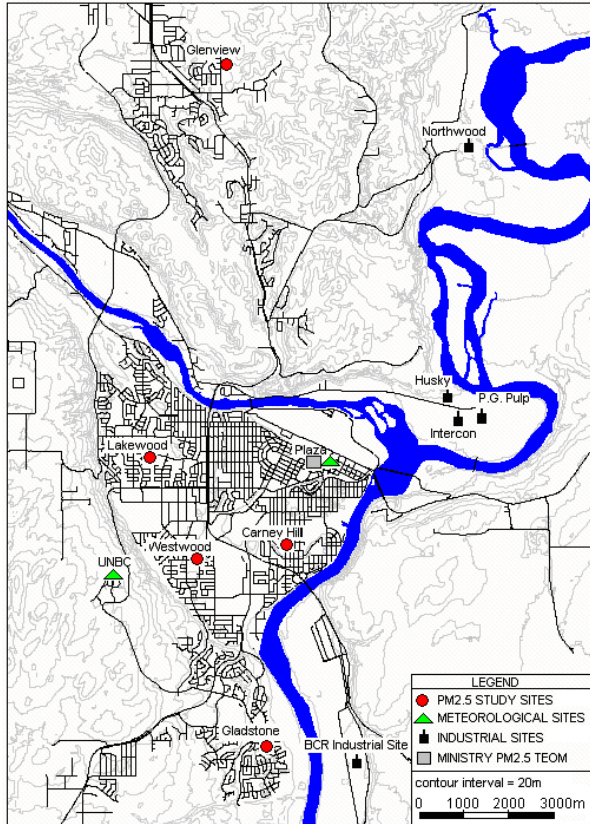


Figure 1 Map of the City of Prince George with each ambient monitoring site labelled and contour lines to depict the valley topography of the area.

All cleaning and loading of the HPEM_{2.5} samplers followed the standard operating procedure (SOP) provided by the Harvard School of Public Health (Ward-Brown, 2000). Gravimetric analysis, reflectometry and sulphate analysis were performed on all personal and ambient samples. Reflectometry measures the “blackness” of the PM_{2.5} filter using a Smoke Stain Reflectometer following standard operating procedure from the ULTRA study (ULTRA, 1998). This analysis provides the reflection of light incidence in percent, which is then converted to an absorption coefficient and is a measure of light absorbing carbon and surrogate for elemental carbon. Sulphate analysis was performed using an ion chromatograph (Dionex, DX-300) with suppressed conductivity detection following the procedure used by Ebel *et al* (2000).

Using sulphate and absorption coefficient as tracers of ambient sources, ambient generated exposures for each individual and every sampling session were estimated from the personal/ambient sulphate and absorption coefficient 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:

$$(Equation 1) \quad E_{ag} = \left(\frac{SO_4^{2-}}{SO_4^{2-}} \right)_{Personal ij} \cdot C_{a jk}$$

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. 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. Identical calculations were performed using light absorbing carbon ratios determined indirectly from absorption coefficients but the analysis was limited 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 influenced the sample. Higher 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.

3. RESULTS AND DISCUSSION

3.1. Ambient Spatial Variation

Ambient spatial variation in total PM_{2.5}, sulphate and absorption coefficient was detected using the non-parametric 2-factor Friedman’s ANOVA ($p < 0.001$). This analysis tests for differences between locations while accounting for changing concentrations over time. For PM_{2.5} this significant difference between schools no longer existed when the downtown school closest to the industrial sources (Carney Hill) was removed from the analysis ($p = 0.125$). But the spatial difference for both sulphate and absorption coefficient persisted even when both Carney Hill and Glenview (furthest out of the valley) were removed from the analysis concurrently ($p \leq 0.023$). Significant spatial variation in absorption coefficient was expected as traffic related pollutants have been shown to be more spatially variable in an area than total PM_{2.5} measurements (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). 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 and limited dispersion during periods of calm winds could be responsible for such differences as well as possible emissions of primary sulphate particulate matter.

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 over time of all three measures. 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, but due to high correlation between the schools temporal trends would be represented adequately. Comparison of HPEM_{2.5} total PM_{2.5} data to unadjusted TEOM PM_{2.5} data suggested that the current MWLAP central monitor underestimates actual concentrations in the downtown area of Prince George where the monitor is located, which was expected due to differences in the sampling methods, but it did accurately represent 3 of the other 4 neighbourhoods monitored during this study. There were also strong, significant Spearman correlations ($r_s=0.83$ to 0.97) between the TEOM data and all of the school ambient monitoring sites suggesting that temporal trends are adequately represented at all schools by the TEOM.

3.2. Meteorological Influence

Wind speed, direction and the presence of a thermal inversion were all important meteorological variables that influenced the level of outdoor concentrations and personal exposures during this study. The highest percentage of winds came from the south and the east and there was a low proportion of wind speeds over 6 m/s for the period (~8%). Higher winds generally came from the south and occasionally from the north and south-west. When higher pollution levels were observed the wind was generally coming from an easterly direction. Winds from an easterly direction would be blowing from the main industrial sources (3 pulp mills and an oil refinery) in the city towards 4 of the 5 study sites and directly to the permanent central TEOM monitor operated by the Ministry of Water, Land and Air Protection. Analysis of data when the PM_{2.5} concentration was greater than 15 µg/m³ showed that all winds with speeds greater than 1 m/s (the lower limit of sensitivity for the anemometer) came from the east. These results suggest 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.

During the study period, 62% of the days experienced inversion conditions for a portion of the

day. All of the days where hourly ambient PM_{2.5} concentrations exceeded 30 µg/m³ were associated with the existence of an inversion on that day. Five main episodes were identified during the study as periods where there were hourly concentrations greater than 30 µg/m³ for more than 6 consecutive hours on 2 or more consecutive days. During inversions there is very limited dispersion and emissions likely linger close to their source. High point source emissions collect beneath the inversion layer and smaller local sources, such as residential wood-smoke, would remain within a neighbourhood. This could explain the significant spatial variation that existed for all three measures.

Inversion strength, calculated as the temperature difference between UNBC and Plaza, was used to assess the relationship of both ambient concentration and personal exposures with inversion conditions. An index of 24-hour inversion strength was calculated by adding all of the positive inversion strength values for a given day together and then dividing by 24 to normalize the value. Time series graphs showing 24-hour inversion strength and both ambient and personal PM_{2.5} at each school are shown in Figure 2. Ambient sulphate and absorption coefficient showed identical patterns as seen for total PM_{2.5} and it is 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". The relationship between inversion strength and personal exposures is not as clear due to the presence of indoor and personal sources. However, similar time-series plots for ambient and personal sulphate with inversion strength were almost identical to the ambient PM_{2.5} plot. For absorption coefficient the patterns were similar to the ambient and personal PM_{2.5} plots with a clear pattern between inversion strength and ambient levels and personal exposures being affected by local ambient, indoor or personal sources.

For ambient concentrations, there were moderate Spearman correlations with inversion strength across all schools for PM_{2.5} (0.63 to 0.76), sulphate (0.54 to 0.60) and absorption coefficient (0.61 to 0.73). 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 more instances where high concentrations existed without an inversion. The correlations with personal exposure were lower for both PM_{2.5} (0.21 to 0.48) and absorption coefficient (0.32 to 0.71), although still mostly significant, which was likely due to the influence of non-ambient sources or very local ambient sources. For sulphate the correlations with inversion strength were similar for both ambient concentrations and personal exposure (0.51 to 0.63). 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 this does suggest that students at this school may have had a lower proportion of their exposure from ambient

sources; therefore, their personal exposures to total PM_{2.5} and absorption coefficient were not influenced as much by a meteorological factor such as inversion strength.

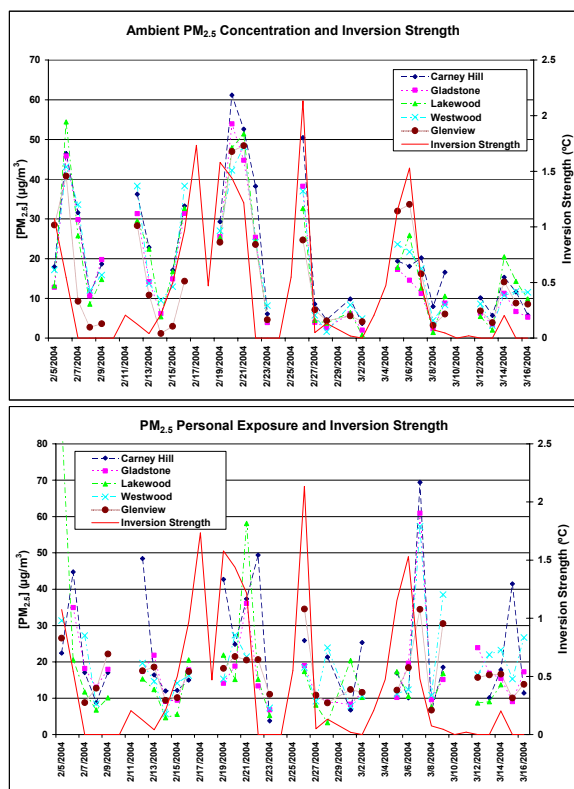


Figure 2 Time series of 24-hour inversion strength and both ambient PM_{2.5} concentrations (top) and personal exposures (bottom) from the five schools. Personal exposure data is pooled across the 3 individuals at each school. Two extreme personal exposures at Carney Hill are out of the range of the graph.

3.3. Personal and Ambient Associations

Individual Spearman correlations were calculated for the personal to ambient association of each measure. The mean correlations and standard deviations show that personal and ambient PM_{2.5} associations (0.51 ± 0.22) were not as strong as that found between personal and ambient sulphate (0.94 ± 0.05) and absorption coefficient (0.70 ± 0.21). The individual Spearman correlations for sulphate were very strong across all individuals with a range of 0.83 to 1.00. This confirms that there were limited non-ambient sources of sulphate and further validates the use of sulphate as an indicator of ambient exposure. Individual absorption coefficient correlations were also strong but more variable across individuals with a range of 0.23 to 0.94 and 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. Although there was similar variability between total PM_{2.5} and absorption coefficient personal-ambient

associations there were higher correlations for absorption coefficient overall suggesting that a smaller number of samples were influenced by personal, indoor or local ambient sources. Use of absorption coefficient as an indicator of exposure to ambient sources is still possible if these samples are removed from the analysis. The overall strong correlations for sulphate and moderate correlation for absorption coefficient make these components suitable candidates for use as a tracer of ambient PM_{2.5}.

3.4. Ambient Generated Exposure

Estimates of ambient generated exposure for both sulphate ($r_s=0.92$) and light absorbing carbon ($r_s=0.86$) showed a stronger median Spearman correlation with ambient concentrations at the nearest school site than that found for total PM_{2.5} ($r_s=0.53$) across individuals. The median slope from the linear regression between ambient generated exposure based on sulphate ratios and ambient concentration showed that individual exposure to ambient levels was approximately 45% of actual ambient levels. This ranged across individuals from 0.29 to 0.64. Almost identical results were found using light absorbing carbon, with a median slope of 0.46 and slightly larger range from 0.03 to 0.67 (only one individual had a correlation below 0.25). Comparison between the estimates of ambient generated exposure and the central TEOM monitor showed a similar median Spearman correlation for both sulphate ($r_s=0.88$) and light absorbing carbon ($r_s=0.81$) suggesting that data from this monitor also adequately represents the general temporal trend in exposure to ambient generated PM_{2.5}. The TEOM comparison resulted in a linear regression slope of 0.45 for the sulphate based estimates and 0.50 for the elemental carbon estimates; both comparable to the regression slope using the closest school ambient monitor.

The slope from the linear regression between ambient generated exposure and total personal exposure showed that 52% of the total exposure sample was from ambient sources when the sulphate estimates were used and 59% for the light absorbing carbon estimates. The possibility of non-ambient sources of light absorbing carbon affecting some personal exposure samples or the presence of a very local ambient source would explain the higher slope, although there is no significant difference in results when the errors associated with each term are considered. Because of the almost equal contribution of ambient and non-ambient sources to total personal exposure there was only low significant Spearman correlations between total personal exposure and estimates of both ambient (0.60 and 0.43) and non-ambient (0.40 and 0.46) generated exposure for the sulphate and light absorbing carbon estimates respectively.

4. CONCLUSION

A combination of topography, meteorological conditions and ambient sources resulted in episodic levels of fine particulate matter during the short study period in the winter of 2001. Thermal inversions were

associated with both high ambient levels and personal exposures and were likely responsible for the spatial variation found in ambient concentrations throughout the city. Management of ambient sources during periods of limited dispersion could reduce both ambient levels and personal exposures in the airshed and should be further investigated. An assessment of the extent to which source reductions would impact ambient concentrations during episodes could be done by atmospheric dispersion modelling and validated using the ambient data from this study.

Both sulphate and absorption coefficient showed a stronger personal-ambient association than total PM_{2.5} and were found to be suitable tracers of ambient generated exposure to PM_{2.5}. The strong relationship between ambient generated exposure and ambient concentrations supports the findings of epidemiological studies that show a health effect for ambient PM_{2.5} and use of ambient data for a longitudinal health study in this city. 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 as well as education regarding the role of indoor and personal sources such as cooking.

5. ACKNOWLEDGEMENT

This research was made possible through funding from the Natural Sciences and Engineering Research Council, Science Council of BC, the Canadian Petroleum Products Institute Clean Air Fund, UNBC Northern Land Use Institute, BC Ministry of Water, Land and Air Protection and Canadian Forest Products Ltd. In kind contributions were made by the Harvard School of Public Health, UBC School of Occupational and Environmental Hygiene and the Air Quality Research Branch of the Meteorological Service of Canada.

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