AN ASSESSMENT OF HOPANES IN SETTLED DUST AND AIR AS INDICATORS OF EXPOSURE TO TRAFFIC-RELATED AIR POLLUTION IN WINDSOR, ONTARIO

by

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ABSTRACT

Traffic-related air pollution (TRAP) has been linked with several adverse health effects. We investigated hopanes, markers of primary particle emissions from gasoline and diesel engines, in house dust as an alternative approach for assessing exposure to TRAP in Windsor, Ontario.

Settled house dust was collected from the homes of 28 study participants (10 – 13 yrs). The dust was then analyzed for a suite of hopanes by gas chromatography-mass spectrometry. We calculated correlations between dust hopane concentrations and estimates of annual average NO2 concentrations derived from an existing LUR model.

Hopanes were consistently present in detectable quantities in house dust. Annual average outdoor NO2 estimated was moderately correlated with hopanes in house dust ($r = 0.46; p<0.05$). The correlations did not vary by infiltration efficiency or the presence of an attached garage. Hopanes measured in settled house dust show promise as an indicator of long-term exposure to traffic-related air pollution.

Keywords: hopane; air pollution; traffic; dust; exposure; TRAP.
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# TABLE OF CONTENTS

Approval ........................................................................................................................................... ii
Abstract ............................................................................................................................................... iii
Acknowledgements ........................................................................................................................ iv
Table of Contents .............................................................................................................................. v
List of Figures ....................................................................................................................................... vii
List of Tables ........................................................................................................................................ ix

1: Background and public health significance ................................................................................. 1
   1.1 Introduction .................................................................................................................................. 1
   1.2 Traffic emissions .......................................................................................................................... 1
   1.3 Adverse health effects of traffic-generated air pollution ............................................................ 2
      1.3.1 Traffic exposure and mortality ................................................................................................. 3
      1.3.2 Asthma ..................................................................................................................................... 3
      1.3.3 Lung function and COPD .......................................................................................................... 4
      1.3.4 Allergy ..................................................................................................................................... 4
      1.3.5 Cardiovascular morbidity ........................................................................................................ 5
      1.3.6 Reproductive effects and birth outcomes ................................................................................ 5
      1.3.7 Cancers ................................................................................................................................... 6
      1.3.8 Cognitive effects ..................................................................................................................... 7
   1.4 Exposure assessment .................................................................................................................. 7
      1.4.1 Introduction ............................................................................................................................. 7
      1.4.2 Current models used to assess levels of exposure .................................................................... 9
      1.4.3 Other considerations in assessing exposure to TRAP .............................................................. 13
   1.5 Emerging TRAP exposure tools and methods ........................................................................... 15

2: Methods ......................................................................................................................................... 17
   2.1 Study area .................................................................................................................................... 17
   2.2 Study population and design ...................................................................................................... 17
   2.3 Data sources ................................................................................................................................ 18
   2.4 Data analysis ................................................................................................................................ 23

3: RESULTS ....................................................................................................................................... 25
   3.1 Summary statistics ..................................................................................................................... 25
   3.2 Exposure metric correlations ...................................................................................................... 28
   3.3 Modifiers of Hopane-NO₂ Relationships ................................................................................... 31

4: Discussion ....................................................................................................................................... 37
   4.1 Findings ....................................................................................................................................... 37
      4.1.1 Presence of an attached garage ............................................................................................... 38
      4.1.2 Infiltration efficiency .............................................................................................................. 39
4.1.3 Time-location patterns ................................................................. 39
4.2 Limitations .................................................................................. 40
4.3 Validations .................................................................................. 43
4.4 Conclusion .................................................................................. 43

5: Appendix ....................................................................................... 45

6: Reference List ............................................................................... 49
LIST OF FIGURES

Figure 1.1.1 The elements of a land use regression model displaying monitoring locations for NO\textsubscript{2} as the dependent variable and land use characteristics within buffers as the predictor variables (Jerrett et al., 2007). ................................................................. 11

Figure 1.2 $F_{\text{det}}$ detached model results in the heating season and predicted indoor ambiet PM\textsubscript{2.5} from outdoor ambient concentrations (15 ug/m\textsuperscript{3}) (Hystad et al., 2009). ................................................................. 14

Figure 1.3 General structure of a hopane. ................................................................. 16

Figure 2.1 Personal monitoring device. ................................................................. 19

Figure 2.2 High volume surface sampling system (HVS3) vacuum. ......................... 20

Figure 2.3 Type and number of hopane sampling events. ...................................... 21

Figure 2.4 Land-use regression NO\textsubscript{2} map of Windsor, Ontario (2005) (Wheeler et al., 2008). ................................................................. 22

Figure 3.1 Summary web of exposure indicator correlations. .............................. 29

Figure 3.2 Total dust hopanes vs. IUR NO\textsubscript{2}. .............................................. 30

Figure 3.3 Total personal hopanes vs. LUR NO\textsubscript{2}. ........................................... 31

Figure 3.4 Dust hopane concentrations versus land-use regression NO\textsubscript{2} concentration stratified by presence of “attached garage”. .................. 32

Figure 3.5 Dust hopane concentrations versus land-use regression NO\textsubscript{2} concentration – stratified by absence of “attached garage”. .................... 32

Figure 3.6 Dust hopane concentrations versus land-use regression NO\textsubscript{2} concentration – stratified by infiltration efficiency greater than the median of 0.24 ......................................................... 34

Figure 3.7 Dust hopane concentrations versus land-use regression NO\textsubscript{2} concentration – stratified by infiltration efficiency greater than the median of 0.24 ......................................................... 34

Figure 3.8 Personal hopane concentrations versus land-use regression NO\textsubscript{2} concentration – stratified by the percentage of time spent at home greater than the median of 74.45%. ......................................................... 36

Figure 3.9 Personal hopane concentrations versus land-use regression NO\textsubscript{2} concentration – stratified by the percentage of time spent at home less than the median of 74.45%. ......................................................... 36

Figure 4.1 Ratio of residential indoor to residential outdoor concentration of selected PAHs and hopanes. The box defines the interquartile range
and median of the distribution; the whiskers are the 10th and 90th percentiles of the distribution (adapted from Olson et al., 2008).
LIST OF TABLES

Table 1.1 Comparison and evaluation of exposure assessment models (HEI, 2010; Jerrett et al., 2005). .............................................................. 13
Table 2.1 Hopanes analyzed in dust samples. ......................................................... 21
Table 3.1 Summary of Windsor, Ontario data including sampling periods, exposure indicators and select modifiers. .................................................. 27
Table 3.2 Summary statistics of exposure indicators and potential modifiers. ............. 28
Table 3.3 Correlations between summed hopane-related variables and two outdoor TRAP indicators. ................................................................. 30
1: BACKGROUND AND PUBLIC HEALTH SIGNIFICANCE

1.1 Introduction

Increases in urban populations, the number of vehicles and miles traveled, and traffic congestion have combined to raise the relative contribution of traffic-related air pollution (TRAP) to the urban air pollution mix (Zmirou et al., 2004). Urban land-use practices have also resulted in population increases near traffic, suggesting that motor vehicle emissions should be considered in the context of their proximity to populated areas (HEI, 2010). Gradient studies indicate exposure zones for TRAP in the range of 50 to about 1500 metres from roads. Studies in two large North American cities, Los Angeles, California and Toronto, Ontario, suggest that between 30% and 45% of the total urban population lies within this range (Gilbert et al., 2005; Jerrett et al., 2007). TRAP, in short, affects a large percentage of the urban population. To fully understand the effect of motor vehicle emissions on human health, it is necessary to accurately estimate exposure to TRAP for epidemiology and risk assessment. A number of studies showing associations between TRAP and negative health outcomes have further highlighted the importance of developing more accurate methods for characterizing exposure to TRAP (HEI, 2010). This study examines a novel tool for assessing chronic exposure to TRAP.

1.2 Traffic emissions

Traffic-related emissions comprise numerous organic and inorganic compounds in both gas and particle phases, including nitrogen oxides (NOx), carbon monoxide (CO), particulate matter (PM), sulphur dioxide (SO2), and volatile organic compounds (VOCs). The organic compounds, found in both the particle and gas phases, include aldehydes, alkenes, alkanes, aromatic hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), nitro-PAHs, and heterocyclic compounds. Many of these inorganic and organic traffic-related pollutants are regulated individually as toxic air pollutants and each may be associated with health outcomes (Laumbach & Kipen, 2010). Epidemiological evidence suggests that long-term exposure to traffic may be an important risk factor for
certain respiratory, cardiovascular, and reproductive morbidities (HEI, 2010). A summary of the adverse health effects associated with TRAP is presented in section 1.3.

An important consideration in the assessment of exposure to TRAP involves proximity to source. Traffic-related contaminants have an effect on outdoor air quality on a great range of spatial scales, from local roadsides and urban scales to broad, regional background scales. Gradient studies characterizing distributions of vehicular exhaust have documented that levels are elevated near roadways, but decay with increasing distance from the source. For example, NO2 concentrations are significantly elevated within 50 to 100 metres of major roads (Jerrett, et al., 2008; Gilbert et al., 2005). In contrast to other outdoor air pollutants, evaluation of particular health risks associated with TRAP is challenging due to exposures to complex mixtures with physical and chemical characteristics that change rapidly over short distances from their sources (Laumbach & Kipen, 2010).

1.3 Adverse health effects of traffic-generated air pollution

In North America, cohort studies such as the Harvard Six Cities study (Dockery et al., 1993), the American Cancer Society studies (Krewski et al., 2003; Pope et al., 1995), and the Adventist Smog and Health study (Abbey et al., 1999), have consistently shown associations between long-term air pollution exposure and mortality, and have subsequently played a pivotal role in the formation of national air quality standards/objectives in both the United States and Canada (Greenbaum et al., 2001). However, these early studies relied primarily on exposure gradients between cities, and therefore they do not provide information related specifically to TRAP. Recent studies have focused on chronic exposure contrasts within cities, with a particular focus on TRAP. Some of this research links proximity to major roads with adverse health outcomes, including wheezing, incident asthma, adverse birth outcomes, and cardiopulmonary morbidity and mortality (Brunekreef et al., 2009; Hoek et al., 2002; Venn et al., 2001; Wilhelm & Ritz, 2003). Even in areas with satisfactory regional air quality, such as Vancouver, BC, proximity to traffic has been linked with incident asthma (Clark et al., 2010), cardiovascular disease (Gan et al., 2010), and both low birth weight and premature birth (Brauer et al., 2008).
1.3.1 Traffic exposure and mortality

Hoek et al. reported that long-term residential exposure to TRAP in the Netherlands was associated with a nearly two-fold increase in risk of cardiopulmonary mortality (RR=1.95) (Hoek et al., 2002). In another study, long-term average concentrations of TRAP markers black smoke and NO2, as well as PM2.5 were related to respiratory mortality (Brunekreef et al., 2009). Jerrett and colleagues reported an association between NO2 and both all cause and cardiovascular mortality in a Toronto, Ontario cohort (Jerrett et al., 2009). A U.K. study reported the risk of stroke mortality for men and women combined was 5% higher in U.K. men and women living within 200 metres of a main road compared with men and women living 1,000 metres away (Maheswaran & Elliott, 2003).

1.3.2 Asthma

Asthma is a chronic inflammatory condition of the airways characterized by recurrent symptoms of variable airflow limitation. The prevalence of allergy and asthma has risen considerably over the past few decades, particularly in the younger age groups and in developing countries (Asher et al., 2006). The substantial increases in the incidence of asthma over recent decades and the geographic variation in prevalence support the hypothesis that environmental changes play a significant role in the current asthma epidemic (Subbarao et al., 2009). Environmental triggers may affect asthma differently at various times of an individual’s life, and the relevant risk factors may change temporally (Subbarao et al., 2009). According to the Health Effects Institute review on TRAP, there is sufficient evidence to infer a causal role for TRAP in the exacerbation of asthma in children (HEI, 2010).

Traffic pollution exposures may also contribute to the incidence of asthma (Brauer et al., 2002; Kim et al., 2008; Sarnat & Holguin, 2007). A recent analysis from a study on traffic, susceptibility, and childhood asthma indicates that lifetime asthma risk is related to air pollution from traffic sources (McConnell et al., 2006). In one of the few studies to investigate the effects of in utero air pollution exposure on pediatric asthma risk, higher exposure to outdoor air pollution in early life was associated with elevated risks of asthma diagnosis in preschool-age children (Clark et al., 2010). The researchers determined that traffic-derived pollutants, including NO, NO2, CO, and black carbon (BC), were associated with the highest risk estimates (Clark et al., 2010).
1.3.3 Lung function and COPD

Due to the paucity of studies on the subject, a meaningful assessment of the role of traffic-related air pollution in the occurrence of chronic obstructive pulmonary disease (COPD) is difficult (HEI, 2010). Cesaroni and colleagues compared the respiratory health of adults with several different indices of exposure of TRAP, including: self-reports of traffic intensity; area-based emissions of particulate matter (PM); distance from busy roads derived from geographical information system (GIS); and estimated concentrations of NO2 using a land-use regression model (Cesaroni et al., 2008). The study found no association between traffic exposure and chronic bronchitis or emphysema and asthma in a population of adults comprised of both non-smokers and smokers (Cesaroni et al., 2008). Conversely, Schikowski and colleagues assessed the long-term effects of air pollution on the development of COPD through the combination of broad- and small-scale spatial exposure gradients (Schikowski et al., 2005). They determined that chronic exposure to PM10, NO2 and living near a major road may increase the risk of developing COPD and can have a detrimental effect on lung function (Schikowski et al., 2005). Overall, chronic respiratory disease in adults is heterogeneous and has numerous risk factors, such as personal smoking and occupational exposure, which do not directly affect children. This larger variety of risk factors may complicate research and contribute to inconclusive results in adults (Lindgren et al., 2009).

In terms of long-term exposures to TRAP, there is some coherence in the data to suggest that long-term exposure is associated with changes in lung function in adolescents and young adults. In the southern California Children’s Health Study, Gauderman and colleagues showed that local exposure to traffic on a freeway is associated with substantial deficits in children’s lung development, which could result in important deficits in attained lung function in later life, including obstructive lung disease. (Gauderman et al., 2007).

1.3.4 Allergy

A study by Krämer and colleagues, involving children from three areas of Düsseldorf, reported consistent results for an association between skin-prick-test reactivity and allergy history in relation to NO2 at children’s residences as a marker of traffic (Kramer et al., 2000).
Cesaroni and colleagues reported consistent associations between self-reported rhinitis (or irritation and inflammation of internal areas of the nose) in adults and several measures of traffic exposure including traffic density, length of high-traffic road within various buffers, distance to high-traffic roads, traffic-generated PM emissions derived from an emission model, and NO2 concentrations estimated from land-use regression (Cesaroni et al., 2008). The associations appeared to be confined exclusively to non-smokers. A study of children living in four Swedish municipalities indicated that the effect of air pollution on sensitization to inhalant allergens, particularly to pollen, is present by the age of four years, and that TRAP plays an important role (Nordling et al., 2008). Kramer and colleagues reported the prevalence of physician-diagnosed eczema in a child’s sixth year of life was associated with TRAP exposure (Kramer et al., 2009). In a large cohort study in the Netherlands, Brauer and colleagues described associations between TRAP and some measures of asthma and allergy, including specific sensitization to common food allergens (Brauer et al., 2002).

### 1.3.5 Cardiovascular morbidity

Several studies have reported associations between chronic TRAP exposure and adverse cardiovascular health outcomes, including the increased prevalence and incidence of coronary heart disease (Hoffmann et al., 2006; Rosenlund et al., 2008), subclinical atherosclerosis (Hoffmann et al., 2007), stroke (Hoffmann et al., 2007; Maheswaran & Elliott, 2003), and myocardial infarction (MI) (Rosenlund et al., 2006; Rosenlund et al., 2009; Tonne et al., 2007; 2009).

In addition to the chronic effects of TRAP, short-term exposure to traffic may trigger acute cardiovascular events. Peters and colleagues reported that a first MI was significantly associated with being in traffic one hour prior to the event (Peters et al., 2004).

### 1.3.6 Reproductive effects and birth outcomes

Exposure to TRAP has been linked to adverse pregnancy outcomes such as low birth weight (LBW), small for gestational age (SGA), pre-term deliveries, and perinatal or postnatal mortality (Brauer et al., 2008; Slama et al., 2007). Using spatiotemporal exposure metrics, Brauer et. al evaluated the impacts of air pollution on small for gestational age (SGA) birth weight, LBW, and preterm birth in Vancouver. The results
suggest a general association between traffic-derived air pollutants such as NOx and CO, and adverse birth outcomes, in particular an increased risk of SGA birth weight (Brauer et al., 2008). The study also observed an association between PM2.5 and risk for preterm birth (Brauer et al., 2008). In models adjusted for background air pollution concentrations, all measured SES variables, and presence of one or more freeways near the residence, Wilhelm and Ritz showed the risk of term LBW and preterm birth increased by 19% and 11%, respectively, per 1 ppm increase in annual average background CO concentration in Los Angeles.

1.3.7 Cancers

Two European studies have reported that urban air pollution is associated with lung cancer risk and that vehicle emissions may be particularly important (Nafstad et al., 2003; Nyberg et al., 2000). As well, investigators have found that living in close proximity to heavy-traffic roads, or exposure to NO2, is associated with elevated risk of lung cancer (Vineis et al., 2006). Evidence that air pollution is at least a moderate risk factor for lung cancer mortality has been found in several case-control studies (Barbone et al., 1995; Jedrychowski et al., 1990), with effect estimates of the same order of magnitude as those estimated from cohort studies (Vineis et al., 2004). Several animal studies have evaluated the potential carcinogenicity of exposure to whole exhaust and to constituents of exhaust from diesel- and gasoline-fuelled internal combustion engines (IARC, 1998). Diesel engine exhaust is considered a Group 2A (probable) carcinogen by the International Agency for Research on Cancer (IARC). Gasoline engine exhaust is noted as a Group 2B (possible) carcinogen to humans (IARC, 1998).

Additional supporting evidence of the carcinogenic potential of TRAP in humans is provided by epidemiologic studies of workers highly exposed to TRAP. Palli and colleagues suggested that traffic-exposed workers, such police officers and bus drivers, were exposed to higher levels of genotoxic agents, related to vehicle emissions, in a DNA adduct level assay, in comparison to the general population in Florence, Italy (Palli et al., 2001). Another study of U.S. trucking industry workers found lung cancer hazard ratios were elevated in workers with jobs associated with regular exposure to vehicle exhaust from diesel and other types of vehicles on loading docks, city streets, and highways (Garshick et al., 2008).
Beelen and colleagues found evidence for an association of exposure to black smoke and traffic with lung cancer incidence in people who had never smoked (Beelen et al., 2008). No clear associations were found for former or current smokers, however. The researchers noted that one significant limitation of their exposure assessment method was that only outdoor concentrations were assessed and factors related to infiltration of outdoor air pollution into the home, such as air exchange rate, were not accounted for (Beelen et al., 2008).

1.3.8 Cognitive effects

The issue of air pollution leading to cognitive deficits and brain structural changes in vulnerable populations is an emerging area of research for public health investigators. A study by Calderón-Garciduenas and colleagues proposed that exposure to significant concentrations of air pollutants, including PM2.5, is associated with neuroinflammation – as measured by mRNA cyclooxygenase-2, interleukin-1beta, and CD14 in target brain regions – and altered innate immune responses in crucial brain target anatomical areas in children and young adults (Calderón-Garciduenas et al., 2008). Suglia and colleagues examined cognition in children and exposure to black carbon as a major component of TRAP and found children who were exposed to higher levels of BC during their life showed decreased cognitive function across assessments of verbal and nonverbal intelligence and memory (Suglia et al., 2008). In a study using a validated spatiotemporal land-use regression model for BC to assess long-term exposure to TRAP, Power and colleagues showed that outdoor TRAP was associated with decreased cognitive function in older men (Power et al., 2011).

1.4 Exposure assessment

1.4.1 Introduction

Exposure assessment issues for TRAP are complex and need to be considered before undertaking investigations of health effects (Morgenstern et al., 2007). Several factors that add to this complexity include the fact that traffic pollutants vary spatially and temporally, that no single metric of traffic exists, that other non-traffic sources contribute some of the same pollutants to the outdoor and indoor air mix, and individuals spend the majority of their time indoors.
Although government-run air pollution monitoring networks adequately capture temporal concentration trends, and are therefore appropriate for estimating exposure in time-series epidemiologic studies, these networks generally do not have adequate spatial density to capture the small-scale spatial variations in TRAP. Common schemes to characterize TRAP exposure include the use of individual pollutants as surrogates or markers, land use regression (LUR) models, and/or the use of proximity to major roads as an exposure surrogate.

Traffic surrogates can aid in the assessment of temporal and spatial distributions of outdoor pollution related to motor vehicles, personal exposures, and exposures in epidemiologic studies (HEI, 2010). Some of the most commonly employed pollutant markers include carbon monoxide (CO), nitrogen oxides (NOx), benzene, and elemental carbon (EC) or BC. These air pollutants infiltrate indoors and can therefore also contribute to indoor exposures. The fact that TRAP constituents tend to be correlated in space and time makes it difficult to identify the specific component(s) that is/are responsible for the observed associations with health effects.

While not often included in exposure models, additional factors such as time-activity patterns can affect personal exposure. Exposures to pollutants in the transport microenvironment, for example, are often highly elevated compared to other microenvironments (Kaur & Nieuwenhuijsen, 2009). Data from U.S. travel surveys indicate that in 2001 commuters spent on average 81 minutes per day in vehicles, and that children spent 48 minutes per day in vehicles (Hu & Reuscher, 2004). Individuals are most likely to be exposed to the highest concentrations of TRAP when traveling in or near vehicles (relative to outdoor concentrations measured at monitoring sites) and to receive as much as half their share of daily exposure to TRAP in this microenvironment (HEI, 2010). An understanding of factors that influence personal exposures is vital in the development of targeted control strategies in urban air quality management and useful in developing a better understanding of the health risks posed by air pollutants in various conditions (Kaur & Nieuwenhuijsen, 2009). Personal monitors are sometimes employed in epidemiologic studies relating to TRAP. These monitors usually consist of a small, portable, personal sampling device that a participant would wear and carry with them over a finite sampling period. Given appropriate resources and depending on the ultimate goal, measurement error in epidemiologic studies can be reduced significantly with the use of personal exposure measurements or prediction models developed from
indoor measurements and survey data. However, due to the influence of indoor sources and personal activities, personal exposure measurements are generally more variable than outdoor measures. Zipprich and colleagues showed that up to 68 per cent of the variation in personal exposure to NO₂ could be explained by introducing an indoor measurement into statistical models (Zipprich et al., 2002).

Several modeling techniques have been developed to estimate traffic-pollutant exposures and these hold promise for use in large epidemiologic studies of TRAP in which home or personal measurements for every study participant are not feasible. Several of these are discussed below.

1.4.2 Current models used to assess levels of exposure

Several advances in exposure modeling for TRAP have occurred over the last decade, facilitated in part by the increased use of geographic information systems (GIS) and associated modeling techniques (Briggs, 2007). In their review, Jerrett and colleagues highlight five major exposure modeling approaches that may be useful for specific traffic-generated pollutants that display substantial gradients within areas downwind of major highways or congested urban centres (Jerrett et al., 2005) (Table 1.1).

1.4.2.1 Proximity models:

This relatively straightforward approach to exposure assessment assigns exposure values of a subject or population based on how near to traffic a subject resides, assuming that the closer an individual is to an emission source, the higher the exposure (Jerrett et al., 2005). As a limitation, however, proximity models may be overly simplistic, as they often ignore factors that may affect exposure, such as time-activity patterns, which may lead to misclassification of exposure (Jerrett et al., 2005). Also, the characteristics of traffic, such as the proportion of cars versus trucks, are often ignored, though these factors may have a large impact on emissions (Jerrett et al., 2005). Wind patterns and topography are unlikely to correspond to the assumption of isotropic dispersion, or equal dispersion in all directions, inherent in this method, particularly if predominant wind conditions blow pollutants towards one direction more than others (Jerrett et al., 2005). The proximity method remains a useful tool in research areas in the exploratory stage, as it is relatively inexpensive compared with other methods, and this
method is attractive because of its clear policy relevance. However, systematic validation of proximity models has been limited (HEI, 2010; Jerrett et al., 2005).

1.4.2.2 Geostatistical interpolation models:

Interpolation models rely on deterministic and stochastic geostatistical techniques (Jerrett et al., 2005). Measurements of a particular pollutant are obtained at a network of monitoring stations distributed across the study area that were chosen based on factors such as the local emissions, topography of area, and degree of variability of the measured pollutant. Based on these factors, the goal is to generate estimates of the concentration of pollutant in areas other than the location of monitoring stations, thereby establishing a continuous surface of pollution concentration. Two of the most typical geostatistical techniques used in air pollution research are inverse distance weighting and kriging (Jerrett et al., 2005). Though relatively simple to implement and often appropriate for more spatially homogenous pollutants such as O3, these techniques are of limited value in studies of TRAP because they generally do not adequately capture small-scale spatial concentration variations.

1.4.2.3 Land-use regression (LUR) models:

This methodology predicts pollution concentrations at a particular site based on surrounding land use, traffic, and topographic characteristics (Jerrett et al., 2005). LUR models consider the air pollutant of interest as the dependent variable and traffic, proximate land-use, and physical environmental variables as predictor variables (Figure 1.1). NO2 is the traffic pollutant that has been most frequently used for LUR modeling because it can be measured relatively inexpensively using passive monitors. LUR modeling generally requires a minimum of 40 to 80 monitoring sites, depending on the size and complexity of the study area (Hoek et al., 2008). The benefit of this method is that it can assign local values through regression mapping, without the need for further monitoring, and can identify areas that require specific monitoring (Jerrett et al., 2005). LUR models have been developed for several Canadian cities, including Windsor (Wheeler et al., 2008), Montreal (Gilbert et al., 2005), Edmonton (Allen et al., 2011), Winnipeg (Allen et al., 2011), Vancouver (Henderson et al., 2007) and Toronto (Jerrett et al., 2007).
Validation studies have indicated that LUR models generally perform as well as or better than dispersion models in European areas for NO$_2$ estimates (Briggs et al., 2000; Hoek et al., 2008). Another study reported that LUR outperformed proximity models, kriging, and dispersion models for predicting PM$_{10}$ (Briggs, 2007). In North American studies, LUR has typically achieved predictions for NO$_2$ in the range of $R^2 = 0.56$ to 0.79 (HEI, 2010). Hoek and colleagues reviewed 25 studies using land-use regression approaches, and reported that LUR models generally performed well in explaining spatial variation and describing pollution hotspots (Hoek et al., 2008). They also found that LUR models for NO$_2$ had generally higher $R^2$ values than NO models (Hoek et al., 2008).

Limitations associated with LUR estimates include the fact that these models are only capable of estimating outdoor exposure. As well, other challenges may arise when transporting a model based on one study area to an area with very different land use characteristics (Jerrett et al., 2005). Changes in land uses, population densities, traffic patterns, or other factors over time may make these models less reliable for assessing exposure. LUR methods can also benefit from models that include both a spatial and...
temporal component for studies that require exposure variables on a more detailed scale (Hoek et al., 2008). Additional attention to a more systematic selection and description of monitoring locations and periods was also highlighted as an important consideration (Hoek et al., 2008). To account for time activity patterns, LUR models need to be validated with personal monitoring (Briggs, 2005; Hoek et al., 2008).

1.4.2.4 Dispersion models:

Another approach for estimating exposures is based on dispersion models and local emission rates for specific pollutants and sources. This method requires a detailed inventory of meteorological data, emission sources, and a series of outdoor measurements with which to calibrate the model (Lipfert & Wyzga, 2008). This method relies on Gaussian plume equations to make assumptions about the transport of emissions based on meteorological conditions and topography, in order to estimate air pollution concentrations (Jerrett et al., 2005). Used in combination with GIS, information from monitoring systems, traffic characteristics, and point sources, the model is calibrated to compute pollution concentrations for a given time period (Jerrett et al., 2005). As a result, the ability of this model to incorporate both spatial and temporal differences ranks as a major advantage (Jerrett et al., 2005). This model can be used to describe episodes of heightened air pollution, and the movement of pollution between areas (Jerrett et al., 2005). Disadvantages of the model include its high costs and data demands, and unrealistic assumptions about dispersion patterns (Jerrett et al., 2005). Furthermore, an extensive proficiency in GIS programming is necessary to construct dispersion models (Jerrett et al., 2005).

1.4.2.5 Hybrid models

Hybrid models combine personal or regional monitoring with other air pollution exposure methods. With accurate data inputs, these hybrid personal exposure models have the potential to incorporate the various components of personal exposure (e.g., in a vehicle compared with outdoors at home) for inclusion in health-effects assessments (HEI, 2010). As with many of the preceding models, the underlying data quality and accuracy are constraints that limit the accuracy of the resulting model output. The difficulty in implementing hybrid models hinges on the combination of models being used.
Table 1.1 Comparison and evaluation of exposure assessment models (HEI, 2010; Jerrett et al., 2005).

<table>
<thead>
<tr>
<th>Model</th>
<th>Data Requirements</th>
<th>Expertise</th>
<th>Software</th>
<th>Utility to Health Studies</th>
<th>Extrapolation</th>
<th>Limitations</th>
<th>Marginal Benefits</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proximity models</td>
<td>Traffic counts, density, distances of the measurement point, questionnaire data</td>
<td>GIS, statistics</td>
<td>GIS (Arcinfo) or equivalent</td>
<td>Low, crude, error prone</td>
<td>Low</td>
<td>Possibility of large errors in complex meteorology and terrain</td>
<td>Base case</td>
</tr>
<tr>
<td>Geostatistical interpolation</td>
<td>Topography, socioeconomic data, location of monitors, monitoring data</td>
<td>GIS, statistics, monitor experts</td>
<td>Spatial statistics, GIS</td>
<td>Depends on the density of the monitoring network</td>
<td>Low</td>
<td>Monitoring networks rarely available to support model estimation</td>
<td>Quantification of error structure</td>
</tr>
<tr>
<td>Land use regression</td>
<td>Traffic volumes, land cover, meteorologic data, monitoring data</td>
<td>GIS, statistics, monitor experts</td>
<td>GIS, statistical software</td>
<td>Depends on the density of monitoring and land-use data</td>
<td>Medium</td>
<td>Interpretation of traffic effects complicated by inclusion of other variables (e.g., population density)</td>
<td>Highly resolved exposure prediction if data are available</td>
</tr>
<tr>
<td>Dispersion</td>
<td>Meteorologic data, traffic volumes, topography, background concentrations, emission data from point and line sources</td>
<td>GIS, statistics, operators for measurement equipment</td>
<td>GIS, statistical software, specialized dispersion software</td>
<td>Often lacks adequate data to support; depends on data input</td>
<td>Medium</td>
<td>Rarely have data support required to calibrate model</td>
<td>Easily transferable and conceptually well matched</td>
</tr>
<tr>
<td>Hybrid (personal monitoring and one of the preceding methods)</td>
<td>Questionnaire (socioeconomic data, activities), monitoring data, other data depending on the combination</td>
<td>Depends on the method combined with personal monitoring</td>
<td>Depends on the combination</td>
<td>Very good</td>
<td>Low</td>
<td>Usually very expensive and limited to small samples of subjects, making it difficult to use in epidemiologic studies</td>
<td>Higher validity in epidemiologic studies</td>
</tr>
</tbody>
</table>

1.4.3 Other considerations in assessing exposure to TRAP

1.4.3.1 Assessing infiltration

Since individuals in North America spend an average of 87 per cent of their time indoors and outdoor pollutants readily penetrate indoors, a significant proportion of total exposure to outdoor-generated pollutants occurs indoors (Klepeis et al., 2001; Leech et al., 2002). Particle infiltration is the key determinant of indoor concentrations of outdoor particles.
Quantifying the infiltration efficiency ($F_{\text{inf}}$) of particulate matter (PM) in residences may be helpful to characterize indoor concentrations and limit exposure misclassification (Allen et al., 2007). In combination with time-location information, $F_{\text{inf}}$ may be used to estimate separate exposures to outdoor-generated and indoor-generated pollution (Ebel et al., 2005; Koenig et al., 2005). Allen and colleagues showed that residential infiltration efficiencies exhibited substantial variation between residences and within residences over time (Allen et al., 2003). A study by Clark and colleagues found that infiltration estimates found in Toronto homes were aligned with previous estimates from similar climates and homes (Clark et al., 2010). The study also study confirmed a high degree of variation in infiltration across different residences. As a result, exposure to outdoor TRAP can be substantially varied even for individuals residing in close proximity and experiencing similar outdoor levels (Clark et al., 2010). Figure 1.2 shows an example of the $F_{\text{inf}}$ model results for the heating seasons mapped by residential parcels in Seattle, Washington and Victoria, British Columbia (Hystad et al., 2009). The $F_{\text{inf}}$ model shows substantial variability in infiltration between homes, and indoor residential $PM_{2.5}$ estimates ranged 5.4 \( \mu g/m^3 \) to 9.0 \( \mu g/m^3 \). Indoor residential estimates may better trace actual exposure levels, as this is where individuals spend the majority of their time, and therefore improve epidemiological studies of the health effects of TRAP. (Hystad et al., 2009).

![Figure 1.2](image)

**Figure 1.2** $F_{\text{inf}}$ detached model results in the heating season and predicted indoor ambiet $PM_{2.5}$ from outdoor ambient concentrations (15 \( \mu g/m^3 \)) (Hystad et al., 2009).

Therefore, accurate assessment of the risks posed by particle exposures is dependent on our ability to characterize indoor concentrations of outdoor particles (Sarnat et al., 2006). The use of continuous and semi-continuous particle monitors allow for the estimation of infiltration efficiencies of outdoor pollution and requires neither the collection of a physical sample nor any composition analysis, making the use of such
monitors practical in epidemiologic studies that rely on outdoor concentrations to assign exposure (Allen et al., 2007).

1.5 Emerging TRAP exposure tools and methods

Assessing indoor levels of traffic-generated pollution through the collection and analysis of settled house dust is a new area of study. Pollutants in house dust are an important contributor of indoor pollution and dust is a commonly used tool for assessing environmental exposures to biological pollutants in epidemiologic studies of asthma and allergy (Roberts et al., 2009). Settled house dust is a sink and repository for particle-bound matter and semi-volatile organic compounds. While the accumulation of house dust depends on several factors (e.g. infiltration efficiency, pollutant source, cleaning practices, sampling surface), dust concentrations and loadings of pollutants show less variation over time than do air concentrations and may therefore be a better indicator of long-term conditions inside residences (Egeghy et al., 2005).

In addition to the commonly measured antigens, researchers have begun to measure dust concentrations of phthalate esters and evaluate relationships with allergic symptoms and asthma (Bornehag et al., 2004; Kolarik et al., 2008). Should traffic-related chemical compounds be readily detected in house dust, the quantification of tracers in house dust may prove a useful exposure assessment approach that incorporates both the spatial patterns of outdoor traffic pollution concentrations and the infiltration of those pollutants into the indoor environment.

One such group of tracers is the hopane class of organic compounds. Hopanes are pentacyclic triterpanes commonly containing 27 to 35 carbon atoms in a naphthenic structure composed of four six-membered rings and one five membered ring (Figure 1.2) (Omar et al., 2006). Hopanes are not found in gasoline and diesel fuel because they are in the higher boiling fraction of petroleum, but are present in engine oil lubricants (Omar et al., 2006; Pakbin et al., 2009; Zakaria et al., 2001). As a result, these compounds are commonly used as tracers for diesel and gasoline emissions, and are particularly useful on account of their relative stability and involatile nature in the atmosphere (Kleeman et al., 2008; Turlington et al., 2010). Rogge and colleagues showed that approximately 85% of all particle-associated hopanes emitted to the atmosphere in Los Angeles originate from internal combustion (Rogge et al., 1993).
Hopanes are commonly used in receptor model source apportionment studies (Linet al., 2010). According to Zielinska and colleagues, the three most abundant hopanes are 17α(H),21β(H)-29-norhopane, 17α(H),21β(H)-hopane, and 22,29,30-trisnorneohopane (Zielinska et al., 2004). Olson and McDow quantified spatial differences in organic source markers near a highway in North Carolina (Olson & McDow, 2009). Among the 33 commonly used source markers, the researchers found significant differences in concentrations sampled in air between near (10 metres) and far (75 metres) locations for elemental carbon (EC) and three hopanes (17α(H),21β(H)-30-norhopane, 17α(H),21β(H)-hopane, 22S-17α(H),21β(H)-30-homohopane). Still, the potential for indoor dust as a tool for assessing exposure to outdoor-generated pollution has not been extensively evaluated. Future research aimed specifically at traffic-pollution constituents likely to have few indoor sources and with the capacity to discriminate near-source traffic and background traffic is needed to interpret the epidemiologic studies attempting to link traffic to adverse health outcomes (HEI, 2010).

The overall objective of this study is to evaluate hopanes as a tool for assessing chronic exposure to traffic-related air pollution (TRAP). Sub-objectives were to:

a) Estimate NO₂ exposure from a LUR model in Windsor and compare that with measured personal and dust hopane concentrations.

b) Examine potential modifiers that could alter the LUR-dust hopane relationship.

c) Examine potential modifiers that could alter the LUR-personal hopane relationship.

d) Evaluate the micronenvironments and activities contributing to personal TRAP exposure as indicated by personal hopane concentrations.
2: METHODS

2.1 Study area

Windsor is the 20th largest city in Canada, with a 2006 population of approximately 216,473 (Statistics Canada, 2007). The city is impacted by both domestic and international sources of air pollution including; local industrial and municipal point sources, commercial and residential area sources, and transportation sources that contribute to both domestic and transboundary air quality issues on both sides of the border (Wheeler et al., 2008). There are border crossings at the Ambassador Bridge and the Detroit-Windsor Tunnel. These crossings, along with the Bluewater Bridge, which connects Sarnia and Port Huron, are the busiest international crossings between Canada and the United States. Collectively, they represent nearly half of the traffic volume crossing between Canada and the United States, with more than 75,000 vehicles crossing the border each day (Wheeler et al., 2008).

2.2 Study population and design

In 2005 and 2006, Health Canada and the University of Windsor conducted a personal exposure study in Windsor, in which study participants were recruited from the larger Windsor Children’s Respiratory Health Study (Dales et al., 2009). Forty-eight adults (parents of the study participants in the (Dales et al., 2009) study) participated in 2005, and 48 asthmatic children (ages 10-13 years) participated in 2006. Because some participants were monitored in both seasons, we define a “monitoring event” as a 5-day monitoring session for an individual participant. From this study, we analyzed environmental samples from a total of 42 monitoring events among 28 of the children who participated in 2006. Residential indoor and outdoor exposures were assessed over a period of 10 days, with a total of 5 sampling days each in the winter (January-March) and summer (July-August). Fourteen of 28 participants’ homes received both summer and winter measurements. Eleven participants were monitoring only during summer, and 3 participants were monitoring only in winter.
The Health Canada REB and the University of Windsor REB approved the study protocol and material.

2.3 Data sources

Home outdoor NO$_2$ monitoring was conducted using Ogawa passive samplers (Ogawa & Co., Pompano Beach, FL, USA) for five days. Ogawas were deployed for five 24-hr periods, and each home’s outdoor NO$_2$ mean required at least 4 valid 24-hour samples to be included in the analysis.

The outdoor monitors were mounted on a tripod at a height of 1.5 metres in each participant’s backyard and sheltered under a stainless steel rain shelter to prevent damage from excess sun or precipitation. Sampling times were approximately 24 ± 3 hours for each NO$_2$ badge (Wheeler et al., 2011).

Four continuous 2-week outdoor NO$_2$ measurements were also collected for each home as part of a spatial monitoring program that made use of the participant’s back yard for security. Not all participants have this measurement because the initial purpose of this outdoor NO$_2$ measurement was for the construction of the LUR model. Hence not every home constituted a “spatial monitoring” site for the purpose of LUR model construction. These two-week measurements were collected simultaneously at approximately 50 locations in Windsor in February (winter), May (spring), August (summer), and October (fall). In the case where participants declined to participate at a given time period, sampling was not always completed for NO$_2$ for all sites in each of the 4 sampling sessions.

Indoor 24-hr PM$_{2.5}$ filter samples were collected using personal environmental monitors (PEM) and continuous personal PM$_{2.5}$ concentrations were measured using DustTraks operated actively with size-selective inlets. During summer 2006, an additional pre-fired quartz 37mm fibre filter (Pall-Gelman, Missisauga, Ontario) was placed in another PEM to collect the pre-separated particulates (PM$_{2.5}$) over five consecutive 24-hour periods at a flow rate of 1.8 lpm. Samplers were checked daily to ensure that they were within 10% of the target 1.8 lpm target flow rate.

Participants also carried an active sampling personal DataRAM (pDR) (Thermo Scientific, Waltham, MA, USA) to measure continuous PM$_{2.5}$ over a 5-day period (Figure 2.1). The personal sampler was contained within a backpack and participants were
asked to carry it with them wherever they went, leaving the backpack close to them to capture their exposure to air pollution when they were not able to carry it, such as during play, showering, and sleep. Similar to the filter used in the indoor sampling, during the winter and summer a pre-fired quartz 37mm fibre filter (Pall-Gelman, Mississauga, Ontario) was placed in line after the pDR to collect the pre-separated PM$_{2.5}$ that passed through the inlet and the pDR unit. The filter was in place for over the entire 5 days of each season at a flow rate of 1.8 litres per minute. A separate PEM with a Teflon filter was used to collect PM$_{2.5}$ particulates on a daily basis for 5 x 24-hr periods.

Figure 2.1 Personal monitoring device.

Settled dust was collected from the floor in the main activity room of each participant’s home using the High Volume Surface Sampling System (HVS3) vacuum (Figure 2.2). Samples were typically collected at the conclusion of the PM$_{2.5}$ monitoring sessions to ensure that the vacuuming did not interfere with those measures. Dust sampling occurred between the months of February and June.

As a result, the settled dust was not collected during the same time period as the personal and indoor air samples (Table 3.1). Technicians vacuumed a 4m$^2$ section of floor in the most used living area for a period of 4 minutes. Technicians were asked to ensure the collection of at least one gram of dust for analysis; a small weighing scale was used to pre- and post-weigh the amber glass bottle used to collect the sample in the field. In the event that less than 1 gram of dust was collected during this time period, technicians would sample another 4m$^2$ section of floor until a total of 1 gram of dust was
collected. Technicians noted any deviation in the total sample area. Samples were stored in the amber glass jars and a brown paper bag for shipping. From the field, the samples were shipped with a cold pack in a cooler. They were then initially stored for approximately 18 months at -80°C before being transferred (in a cold cooler) to a regular freezer set at a temperature of -4°C.

On receipt of the samples, the dust was sieved before extraction with a non-metallic, 300µm sieve to remove any coarse material. Dust samples were further sieved into <150µm size fractions for analysis.

Figure 2.2 High volume surface sampling system (HVS3) vacuum.

The 5-day indoor and personal PM2.5 quartz fibre filters and the dust samples were analyzed for a suite of organic compounds, including several hopanes, by gas chromatography/mass spectrometry (GC/MS). From the field, the samples were shipped with a cold pack in a cooler. They were then initially stored for approximately 18 months at -80°C before being transferred (in a cold cooler) to a regular freezer set at a temperature of -4°C. The samples were then thawed for sorting, refrozen (at -18°C) and shipped cold to a -10°C freezer before being analyzed. For analysis, samples were prepared in an isoctane solution and passed through an ASE 200 (Accelerated Solvent Extractor), Zymark TurboVap. The six hopanes analyzed are included in Table 2.1.
Table 2.1 Hopanes analyzed in dust samples.

<table>
<thead>
<tr>
<th>Hopane abbreviations</th>
<th>Full chemical name</th>
</tr>
</thead>
<tbody>
<tr>
<td>H17a</td>
<td>17α(H)-22, 29, 30-Trisnorhopane</td>
</tr>
<tr>
<td>a_b_nor</td>
<td>17α(H), 21β(H)-30-Norhopane</td>
</tr>
<tr>
<td>a_b_hop</td>
<td>17α(H), 21β(H)-Hopane</td>
</tr>
<tr>
<td>b_a_hop</td>
<td>17β(H), 21α(H)-Hopane</td>
</tr>
<tr>
<td>H22S</td>
<td>17α(H), 21β(H)-22S-Homohopane</td>
</tr>
<tr>
<td>H22R</td>
<td>17α(H), 21β(H)-22R-Homohopane</td>
</tr>
</tbody>
</table>

Figure 2.3 summarizes the number of hopane sampling events collected using indoor air, personal air, and dust sampling methods.

Figure 2.3 Type and number of hopane sampling events.

Time-activity patterns of each participant were recorded daily in time activity diaries (TADs). For each period, they noted their activities in open text and selected one of six microenvironments: indoors at home, outdoors at home, in transit, at work/school, indoors away from home and outdoors away from home. Field technicians using a questionnaire also collected additional data relating to housing characteristics, such as the presence or absence of an attached garage and approximate age of the home. All home surveys and diaries were independently entered twice and compared electronically to each other to identify any discrepancies in the data entry.
A LUR model (2005) of annual average NO\textsubscript{2} concentration was previously developed for Windsor. A total of 75 variables were generated in 4 categories to characterize road network, population density, land use and point sources at different radii around each sampling site (Figure 2.4). The LUR modelling made use of the 2-week integrated NO\textsubscript{2} measurements obtained through Ogawa passive samplers and described earlier. The final predictors of NO\textsubscript{2} used in the LUR model were proximity to the Ambassador Bridge (the main Canada—U.S. border crossing point), and proximity to highways and major roads. The model predicted NO\textsubscript{2} concentrations with an $R^2=0.77$ (Wheeler et al., 2008).

![Figure 2.4 Land-use regression NO2 map of Windsor, Ontario (2005)](image)

Using the continuous DustTrak data, we estimated for each season and home the residential PM\textsubscript{2.5} $F_{inf}$, defined as the fraction of the outdoor concentration that penetrates indoors and remains suspended, using a censoring algorithm and recursive mass balance model applied to the continuous indoor and outdoor PM\textsubscript{2.5} data. (Allen et al., 2003; Allen et al., 2007).
2.4 Data analysis

We used correlation coefficients and scatterplots to assess the relationship between hopanes in indoor dust, indoor air, and personal air and two indicators of outdoor TRAP. Because our main interest was in assessing hopanes in house dust as a tool for assessing chronic exposure to TRAP, our primary comparison was with annual average NO\(_2\) concentration modeled outside the home using LUR. As a secondary comparison, we also compared hopane concentrations with NO\(_2\) concentrations measured with Ogawas outside each home. These were selected as the indicators of outdoor TRAP because NO\(_2\) is a commonly used indicator of TRAP in epidemiologic studies and these two measures represent both annual average (LUR) and short-term (Ogawa) averaging times for comparison. While the indoor and personal air hopanes were compared with the corresponding 5-day outdoor NO\(_2\) measurements, dust hopanes were compared with one of four 2-week outdoor NO\(_2\) measurements in 2006 that most closely matched the indoor dust sampling date.

Dust hopane concentrations were normalized to total dust collected (ng/mg). We assessed both the individual hopanes, as well as the sum of all hopanes, and made the following comparisons:

**Primary comparison:**
- indoor dust hopane concentration and the annual average NO\(_2\) concentration modeled outside the home using LUR

**Secondary comparisons:**
- personal air 5-day hopane concentration and the annual average NO\(_2\) concentration modeled outside the home using LUR;
- indoor air 5-day hopane concentration and the annual average NO\(_2\) concentration modeled outside the home using LUR;
- indoor dust hopane concentration and the two-week NO\(_2\) concentration measured outside the home that most closely matches the date of dust collection;
- personal air 5-day hopane concentration and the 5-day NO\(_2\) concentration measured outside the home and collected during the same 5-day period;
- indoor air 5-day hopane concentration and the 5-day NO\(_2\) concentration measured outside the home and collected during the same 5-day period.
To ensure that measurements reflect only spatial variability, LUR measurements were adjusted for the impact of temporal variability (Hoek et al., 2002). In this study, the LUR model was temporally adjusted using data from the two National Air Pollution Surveillance (NAPS) monitors in Windsor (NAPS 2006) to adjust for the mean NO2 concentrations 7 days, 14 days, 28 days, 56 days, 84 days, 112 days, and 140 days before the date of dust sampling, and 5-days after the start of the personal and indoor monitors. The mean NO2 concentrations at the NAPS sites over these time periods were then divided by the annual average to produce a temporal adjustment factor. This factor was multiplied by the LUR-derived annual estimate to create a temporally adjusted NO2 concentration representing various time periods. In addition, because differences in $F_{inf}$ and housing characteristics may modify the relationships between outdoor concentrations and personal exposures to TRAP, we stratified the above comparisons into two groups based on the median infiltration efficiency, and based on the presence of an attached garage. We also stratified the personal monitoring data based on time spent at home, away from home, and while commuting (above/below the median). Interaction models were constructed to test for differences in model slopes and intercepts by infiltration efficiency and presence of an attached garage (for the comparisons involving dust hopane concentrations), and by time-location patterns (for the comparisons involving personal hopane concentrations).

To account for outliers and to test the effect of certain influencing factors on our original hopane relationships, we plotted the residuals of those relationships against the following covariates:

- Dust hopanes vs. LUR NO\textsubscript{2} residuals vs. “Age of Home”
- Personal hopanes vs. LUR NO\textsubscript{2} residuals vs. “Time Spent in Transit”
- Personal hopanes vs. LUR NO\textsubscript{2} residuals vs. “Time Spent frying, grilling, sautéing and broiling”

Note: The residuals of the “dust hopanes vs. LUR NO\textsubscript{2}” relationship were not plotted against the “Time spent frying and cooking” because the time periods in which the data was collected did not coincide.

Data analysis was performed with statistical application package SAS Version 9.2 (SAS Inc., Cary, NC).
3: RESULTS

3.1 Summary statistics

The 28 participants had a mean (± SD) age of 10.9 (± 0.79) years and 39% were female (Table 3.1). A total of 42 monitoring events included at least two hopane-related measurements (dust, indoor air and/or personal air). The study sample size varied among the exposure indicators because of missing data (Figure 2.3). With one exception, sampling start dates were the same for 5-day personal air hopanes, 5-day indoor air hopanes (winter only), and 5-day outdoor NO$_2$. The lone exception (home #1 – Table 3.1) was included as the start dates differed by only one day.

The homes were single-family detached homes spatially distributed across Windsor. In the 15 homes that included summer and winter 5-day outdoor NO$_2$ measurements, 14 of the samples had the higher NO$_2$ concentrations in the winter. Conversely, among the 14 participants with personal PM$_{2.5}$ samples in both seasons, there was no clear seasonal pattern in personal exposures (Table 3.1).

Less than 1% of the individual hopane concentrations fell below the limit of detection and these (a total of 4 values) were limited to the H22S and H22R compounds. The values below the limit of detection in both cases were corrected using LOD/2. On average, a_b_nor registered the highest concentration of all 5 personal and indoor air hopanes sampled.

Variation across samples is observed for the majority of the exposure indicators and potential modifying factors. Infiltration efficiency, however, had an inter-quartile range of only 0.13 (Table 3.2). Average infiltration efficiencies measured in summer months (0.29 ± 0.16) were similar to those measured in the winter (0.22 ± 0.09). The average infiltration value across all homes and seasons was 0.27 (Table 3.2). In other words, on average, 27% of outdoor fine particles infiltrate indoors. Limited variation was also observed for the LUR-derived NO$_2$ estimates (IQ range of less than 3.0 ppb). As well, participants spent the majority of their time (roughly 74.6%, on average) indoors at home.
The suite of individual dust hopanes correlated strongly with each other with a maximum correlation of \( r=0.95 \) (\( p<0.01 \)) between a_b_nor and H17a (\( n=27 \)) and a minimum correlation of \( r=0.73 \) (\( p<0.01 \)) between H22R and H17a (\( n=27 \)).

The suite of individual personal hopanes also correlated strongly with each other with a maximum correlation of \( r=0.95 \) (\( p<0.01 \)) between H22S and H22R (\( n=42 \)) and a minimum correlation of \( r=0.50 \) (\( p<0.01 \)) between H22R and H17a (\( n=42 \)).

The suite of individual indoor hopanes registered moderate to high correlations with each other with a maximum correlation of \( r=0.90 \) (\( p<0.01 \)) a_b_hop and H22S (\( n=25 \)) and a minimum correlation of \( r=0.37 \) (\( p=0.07 \)) between H22R and H17a.
Table 3.1 Summary of Windsor, Ontario data including sampling periods, exposure indicators and select modifiers.

<table>
<thead>
<tr>
<th>Sex</th>
<th>Age</th>
<th>Season</th>
<th>Personal air concentration (µg/m³) with sampling start date</th>
<th>Indoor air concentration (µg/m³)</th>
<th>Dust concentration (µg/m³)</th>
<th>LUR NO₃ estimate (2005) (ppb)</th>
<th>Mean (1-week) outdoor NO₂ concentration (ppb)</th>
<th>Mean (2-week) outdoor NO₂ concentration (ppb)</th>
<th>Garbage (attached, detached or none)</th>
<th>Percentage time spent in the home</th>
<th>Percentage time spent in transit</th>
<th>Infiltration efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Female</td>
<td>11</td>
<td>summer</td>
<td>386.76 (22/06/2006)</td>
<td>237.87 (22/06/2006)</td>
<td>4.30</td>
<td>13.47</td>
<td>19.80</td>
<td>75.40</td>
<td>7.90</td>
<td>5.88</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Male</td>
<td>12</td>
<td>summer</td>
<td>802.44 (14/06/2006)</td>
<td>389.34 (14/06/2006)</td>
<td>8.87</td>
<td>16.82</td>
<td>24.40</td>
<td>99.10</td>
<td>15.90</td>
<td>2.68</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Female</td>
<td>10</td>
<td>winter</td>
<td>652.66 (27/02/2006)</td>
<td>780.96 (19/07/2006)</td>
<td>5.69</td>
<td>14.46</td>
<td>20.79</td>
<td>23.90</td>
<td>1.49</td>
<td>0.39</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Male</td>
<td>10</td>
<td>winter</td>
<td>652.66 (27/02/2006)</td>
<td>780.96 (19/07/2006)</td>
<td>5.69</td>
<td>14.46</td>
<td>20.79</td>
<td>23.90</td>
<td>1.49</td>
<td>0.39</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>Female</td>
<td>12</td>
<td>summer</td>
<td>415.75 (25/07/2006)</td>
<td>399.96 (25/07/2006)</td>
<td>14.81</td>
<td>21.60</td>
<td>25.10</td>
<td>84.90</td>
<td>0.00</td>
<td>0.17</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Female</td>
<td>11</td>
<td>summer</td>
<td>1966.54 (13/06/2006)</td>
<td>689.96 (13/06/2006)</td>
<td>8.62</td>
<td>16.00</td>
<td>20.50</td>
<td>74.00</td>
<td>0.90</td>
<td>0.16</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Male</td>
<td>10</td>
<td>summer</td>
<td>961.52 (20/06/2006)</td>
<td>771.64 (20/06/2006)</td>
<td>12.98</td>
<td>24.50</td>
<td>28.80</td>
<td>88.50</td>
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<td>389.93 (21/06/2006)</td>
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<td>9</td>
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<td>12</td>
<td>winter</td>
<td>342.89 (26/06/2006)</td>
<td>592.81 (03/07/2006)</td>
<td>2.92</td>
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<td>9.90</td>
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<td>winter</td>
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<td>629.90 (03/07/2006)</td>
<td>12.29</td>
<td>24.30</td>
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<tr>
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<td>11</td>
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<td>438.88 (03/07/2006)</td>
<td>426.88 (03/07/2006)</td>
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<td>8.00</td>
<td>12.10</td>
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<tr>
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<td>winter</td>
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<td>winter</td>
<td>358.64 (21/06/2006)</td>
<td>389.93 (21/06/2006)</td>
<td>13.90</td>
<td>21.20</td>
<td>25.10</td>
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<tr>
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<td>2.92</td>
<td>5.90</td>
<td>9.90</td>
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<tr>
<td>20</td>
<td>Male</td>
<td>11</td>
<td>winter</td>
<td>651.70 (03/07/2006)</td>
<td>629.90 (03/07/2006)</td>
<td>12.29</td>
<td>24.30</td>
<td>28.80</td>
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<td>0.24</td>
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</tr>
<tr>
<td>21</td>
<td>Male</td>
<td>11</td>
<td>winter</td>
<td>438.88 (03/07/2006)</td>
<td>426.88 (03/07/2006)</td>
<td>4.00</td>
<td>8.00</td>
<td>12.10</td>
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<td>968.23 (03/07/2006)</td>
<td>434.80 (03/07/2006)</td>
<td>2.56</td>
<td>5.70</td>
<td>9.70</td>
<td>75.70</td>
<td>2.30</td>
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<td>23</td>
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<td>11</td>
<td>winter</td>
<td>968.23 (03/07/2006)</td>
<td>434.80 (03/07/2006)</td>
<td>2.56</td>
<td>5.70</td>
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<tr>
<td>24</td>
<td>Female</td>
<td>11</td>
<td>winter</td>
<td>968.23 (03/07/2006)</td>
<td>434.80 (03/07/2006)</td>
<td>2.56</td>
<td>5.70</td>
<td>9.70</td>
<td>75.70</td>
<td>2.30</td>
<td>0.53</td>
<td></td>
</tr>
<tr>
<td>AVG</td>
<td>10.9</td>
<td>315.90</td>
<td>273.65</td>
<td>5.08</td>
<td>14.12</td>
<td>19.82</td>
<td>74.61</td>
<td>3.85</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>STD</td>
<td>0.79</td>
<td>207.09</td>
<td>133.79</td>
<td>2.98</td>
<td>2.06</td>
<td>6.61</td>
<td>5.99</td>
<td>9.12</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MAX</td>
<td>12</td>
<td>1566.70</td>
<td>780.96</td>
<td>12.55</td>
<td>18.02</td>
<td>30.30</td>
<td>27.10</td>
<td>100.00</td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>MIN</td>
<td>10</td>
<td>116.09</td>
<td>237.87</td>
<td>1.13</td>
<td>10.47</td>
<td>4.40</td>
<td>2.70</td>
<td>58.50</td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

AVG = Average
STD = Standard Deviation
MAX = Maximum
MIN = Minimum
Table 3.2 Summary statistics of exposure indicators and potential modifiers.

<table>
<thead>
<tr>
<th>Variable</th>
<th>N</th>
<th>Mean</th>
<th>Standard Deviation</th>
<th>25th Percentile</th>
<th>Median</th>
<th>75th Percentile</th>
</tr>
</thead>
<tbody>
<tr>
<td>LUR NO2 – 2005 (ppb)</td>
<td>40</td>
<td>14.12</td>
<td>2.06</td>
<td>12.79</td>
<td>13.88</td>
<td>15.15</td>
</tr>
<tr>
<td>5-day Outdoor NO2 (ppb)</td>
<td>42</td>
<td>16.05</td>
<td>6.61</td>
<td>11.10</td>
<td>16.05</td>
<td>21.30</td>
</tr>
<tr>
<td>2-week Outdoor NO2 (ppb)</td>
<td>25</td>
<td>19.82</td>
<td>5.99</td>
<td>17.90</td>
<td>21.70</td>
<td>23.90</td>
</tr>
<tr>
<td>Infiltration efficiency - Finf</td>
<td>41</td>
<td>0.27</td>
<td>0.14</td>
<td>0.18</td>
<td>0.24</td>
<td>0.31</td>
</tr>
<tr>
<td>Dust hopanes (ng/mg)</td>
<td>27</td>
<td>5.56</td>
<td>2.80</td>
<td>2.80</td>
<td>5.62</td>
<td>7.44</td>
</tr>
<tr>
<td>Personal air hopanes (pg/m³)</td>
<td>42</td>
<td>518.95</td>
<td>257.00</td>
<td>356.75</td>
<td>446.76</td>
<td>631.52</td>
</tr>
<tr>
<td>Indoor air hopanes (pg/m³)</td>
<td>25</td>
<td>445.86</td>
<td>133.79</td>
<td>345.17</td>
<td>430.83</td>
<td>529.11</td>
</tr>
<tr>
<td>Time spent in home (%)</td>
<td>42</td>
<td>74.61</td>
<td>9.12</td>
<td>67.40</td>
<td>74.45</td>
<td>79.00</td>
</tr>
<tr>
<td>Time spent in transit (%)</td>
<td>42</td>
<td>3.85</td>
<td>2.98</td>
<td>1.40</td>
<td>3.45</td>
<td>5.40</td>
</tr>
<tr>
<td>Time spent at school (%)</td>
<td>42</td>
<td>7.80</td>
<td>9.74</td>
<td>0.00</td>
<td>0.00</td>
<td>18.90</td>
</tr>
</tbody>
</table>

3.2 Exposure metric correlations

The total concentration of hopanes in dust was moderately correlated (Pearson’s \( r = 0.46 \); Spearman’s \( r = 0.58 \); \( n = 26 \)) with the annual average outdoor NO₂ concentration estimated from the LUR model (Table 3.3; Figures 3.1 and 3.2). Temporal adjustments of the LUR NO₂ (to shorter averaging times hypothesized to match the period captured by dust samples) using the two fixed, government monitoring sites in Windsor decreased the dust-LUR correlations slightly (Appendix; Table A.1). Dust hopanes were not correlated (Pearson’s \( r = 0.07 \); Spearman’s \( r = 0.05 \)) with the measured outdoor NO₂ concentrations, though the correlations are based on only 16
observations (Table 3.3). Correlations between dust hopanes and both indoor air and personal air hopanes were also very weak (Table 3.3).

The LUR NO\textsubscript{2} also showed a moderate positive correlation with personal air hopanes (Pearson’s $r=0.50$; Spearman’s $r=0.43$; $n = 40$), but a weaker positive, non-significant correlation with indoor air hopanes (Pearson’s $r=0.20$; Spearman’s $r=0.23$; $n = 24$) (Figure 3.3; Table 3.3). A moderate correlation was seen between personal air hopanes and indoor air hopanes (Pearson’s $r=0.44$; Spearman’s $r=0.63$; $n=25$).

Correlations with LUR NO\textsubscript{2} for individual hopanes in the house dust (ranges: Pearson’s $r = 0.38 – 0.52$; Spearman’s $r = 0.44 – 0.58$), indoor air (ranges: Pearson’s $r = 0.08 – 0.27$; Spearman’s $r = 0.10 – 0.27$) and personal air (ranges: Pearson’s $r = 0.33 – 0.50$; Spearman’s $r = 0.30 – 0.42$) were very similar to the correlations for total hopane concentrations (Table A.2). For dust and personal air, the hopane most correlated with long-term outdoor NO\textsubscript{2} was a\_b\_nor, while for indoor air the highest correlation was found for a\_b\_hop.

We found that after accounting for LUR NO\textsubscript{2}, "time spent cooking" did not explain any additional variance in personal hopane concentrations.
Table 3.3 Correlations between summed hopane-related variables and two outdoor TRAP indicators.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Dust hopane concentration</th>
<th>Personal air hopane concentration **</th>
<th>Indoor air hopane concentration **</th>
<th>Outdoor NO2 concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pearson</td>
<td>Spearman</td>
<td>Pearson</td>
<td>Spearman</td>
</tr>
<tr>
<td>Personal air hopane concentration **</td>
<td>0.21     (p=0.28)</td>
<td>0.25     (p=0.21)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>n=27</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Indoor air hopane concentration **</td>
<td>0.07     (p=0.84)</td>
<td>0.12     (p=0.75)</td>
<td>0.44     (p&lt;0.05)</td>
<td>0.63     (p&lt;0.01)</td>
</tr>
<tr>
<td></td>
<td>n=10</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Outdoor NO2 concentration</td>
<td>0.07     (p=0.81)</td>
<td>0.05     (p=0.86)</td>
<td>0.21     (p=0.18)</td>
<td>0.28     (p=0.07)</td>
</tr>
<tr>
<td></td>
<td>n=16</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LUR NO2 estimate</td>
<td>0.46     (p&lt;0.05)</td>
<td>0.58     (p&lt;0.01)</td>
<td>0.50     (p&lt;0.01)</td>
<td>0.43     (p&lt;0.01)</td>
</tr>
<tr>
<td></td>
<td>n=26</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Two-week average outdoor NO2 concentrations that most closely approached the dust sampling period were used in calculating these correlations.

** Five-day average outdoor NO2 concentrations that matched the personal and indoor air sampling periods were used in calculating these correlations.

Figure 3.2 Total dust hopanes vs. LUR NO2.
3.3 Modifiers of Hopane-NO₂ Relationships

The presence of an attached garage and infiltration efficiency were evaluated as potential modifiers of the dust hopane – LUR NO₂ correlations. Though based on small numbers of samples, homes with an attached garage had a higher dust hopane – LUR correlation (r = 0.62, n = 10) than homes without an attached garage (r = 0.33, n = 16) (Figures 3.4 and 3.5). The slopes of the two stratified relationships were very similar in magnitude (0.63 vs. 0.66; p=0.96).

A similar effect was observed between the LUR NO₂ and the personal hopanes in the presence of an attached garage (r = 0.59 vs. 0.47) (Appendix; Table A3). Neither the slopes (p=0.51) nor the intercepts of the two relationships were different (p=0.16).

We found that after accounting for LUR NO₂, "Age of Home" did not explain any additional variance in dust hopane concentrations.
Figure 3.4 Dust hopane concentrations versus land-use regression NO$_2$ concentration stratified by presence of “attached garage”.

Figure 3.5 Dust hopane concentrations versus land-use regression NO$_2$ concentration – stratified by absense of “attached garage”.

32
Similar relationships after stratifying by infiltration efficiency were observed. The correlations between dust hopanes and LUR NO₂ were similar for homes above and below the median $F_{inf}$ of 0.24. Homes with an infiltration efficiency greater than the median had moderate to strong correlations (Pearson’s $r = 0.52$; Spearman’s $r = 0.60$), while homes below the median $F_{inf}$ had moderate correlations (Pearson’s $r = 0.42$; Spearman’s $r = 0.60$) (Figures 3.6 and 3.7; Appendix; Table A.4). When comparing the regression results for homes with an infiltration efficiency greater than the median to homes with an infiltration efficiency below the median, neither the slopes ($p=0.86$) nor the intercepts were different ($p=0.88$).
Figure 3.6 Dust hopane concentrations versus land-use regression NO2 concentration – stratified by infiltration efficiency greater than the median of 0.24.

Figure 3.7 Dust hopane concentrations versus land-use regression NO2 concentration – stratified by infiltration efficiency greater than the median of 0.24.
The correlations between personal hopanes and LUR NO2 were similar among participants whose percentage time spent at home was above the median of 74.45% and those whose percentage time spent at home was below the median (Figures 3.8 and 3.9; Table A.5). The regression slope among participants who spent more than 74.5% of their time indoors at home and the slope of participants who spent less than 74.5% of their time indoors at home were different (p<0.05) with the former having a much steeper slope. The intercepts of the two relationships were marginally different (p=0.06).

We found that after accounting for LUR NO₂, “time spent in traffic” did not explain any additional variance in personal hopane concentrations.
Figure 3.8 Personal hopane concentrations versus land-use regression NO₂ concentration – stratified by the percentage of time spent at home greater than the median of 74.45%.

Figure 3.9 Personal hopane concentrations versus land-use regression NO₂ concentration – stratified by the percentage of time spent at home less than the median of 74.45%.
4: DISCUSSION

4.1 Findings

In this analysis we evaluated hopanes as a tool for assessing chronic exposure to traffic-related air pollution (TRAP) and compared hopane concentrations to two indicators of outdoor TRAP.

Hopanes have been cited in apportionment studies and reviews as indicators for mobile source emissions (Lin et al., 2010), as well as studies examining spatial differences in organic source markers near a highway (Olson & McDow, 2009). However, our study marks the first time hopanes in settled house dust have been used to assess exposure to TRAP.

The results show that hopanes measured in dust correlated better with the annual average (LUR) NO$_2$ than with the outdoor (5-day and 2-week) NO$_2$. This suggests that hopanes measured in dust may be more representative of long-term conditions (Table 3), although the small sample sizes limit our ability to draw firm conclusions. Adjustment of the LUR estimates to time periods thought to be more consistent with the period captured by dust samples did not improve the dust-NO$_2$ correlations. Settled house dust, depending on the location being sampled, has a deposition rate of 0.0022-0.08 g/m$^3$ per day, and can act as a reservoir for organic compounds (Maertens, Bailey, & White, 2004). The rate of removal would of course depend on, among other things, the cleaning habits of the occupants, the activity of the occupants, the surface on which the dust has collected, as well as the composition of the dust itself, which varies. It seems highly likely that in homes with heavy carpet, where regular vacuuming does not remove all contaminants, dust has the potential to build up over longer periods. Upon inspection of the regression plots, several potential confounders including age of home, and presence or absence of an attached garage could not explain outliers in this data.

Personal hopanes also correlated moderately well with the annual average (LUR) NO$_2$ ($r = 0.50; p<0.01$). In line with other studies examining the time-location patterns of North Americans (Klepeis et al., 2001; Leech et al., 2002), our study participants spent
the majority of their time (approximately 75%, on average) indoors in the home (Table 2). This further confirms that in order to accurately assess exposure to TRAP, indoor TRAP indicators, such as hopanes in dust, may be valuable. It also would suggest that the personal and indoor air measurements should then be relatively well aligned. In our study, personal and indoor hopanes were weak-to-moderately correlated (Pearson’s $r = 0.44; p<0.05$; Spearman’s $r = 0.63; p<0.01$). Given the relatively small sample size ($n=25$), non-parametric analysis may be more appropriate in this case.

Moderate and low correlations in comparing the dust and air hopanes to both the outdoor NO$_2$, and the LUR NO$_2$ might also be explained by the natural incongruence between comparing particles (hopanes) and gases (NO$_2$). Nethery and colleagues found that LUR models showed the strongest ability to predict personal measures for NO$_2$ while outdoor NO$_2$ monitor estimates were less predictive (Nethery et al., 2008).

As we reported, the hopanes in air and dust showed greater variation than the LUR estimates. First, indoor sources of hopanes could have contributed to the increase in variation (indoor sources is discussed in more detail in the Limitations section below). Secondly, LUR models are assumed to represent long-term measures based upon how they are constructed. However, LUR models depend upon the NO$_2$ measurements used as independent variables in the model development. Should these measurements be atypical of the long-term conditions, the model will then be less likely to be a good long-term exposure predictor. Fortunately, the Windsor LUR model incorporated all four seasons into the model, which adds to its precision. The LUR model constructed for this study only modeled NO$_2$. In a study on the application of land use regression to estimate certain long-term TRAP contaminants, Henderson et al. suggested that the distribution of NO, for example, is more heterogeneous than that of NO$_2$ (Henderson et al., 2007). We must also not assume that a LUR model represents the best indicator of TRAP. Hopanes in dust could in fact be tracing indoor exposure to TRAP more effectively and accurately than the LUR model.

4.1.1 Presence of an attached garage

Vehicle emissions in attached garages do infiltrate and ultimately influence the indoor air quality of houses with attached garages (Graham et al., 2004; Hun, 2011). In our study, homes with an attached garage showed a stronger correlation for dust hopanes versus annual average (LUR) NO$_2$ ($r= 0.62$, $n=10$), and personal hopanes
versus annual average (LUR) NO\textsubscript{2} ($r = 0.59$, n=16). This result is not expected, as the garage source of hopanes should weaken the correlation with the LUR NO\textsubscript{2} since these localized sources are not captured in LUR models. Possible explanations include the small sample size following stratification, as well as that a home with an attached garage does not explicitly mean that vehicles are stored or started in the garage.

### 4.1.2 Infiltration efficiency

Since the penetration of outdoor particles into the indoor environment has been shown to be a significant source of indoor particles (Maertens et al., 2004), and infiltration efficiencies vary between homes (Hystad et al., 2009) it is important to consider infiltration efficiency of each home as a potential modifier. Our study found that infiltration efficiency, however, did not significantly affect the outdoor-indoor or outdoor-personal correlations (Figure 4). A steeper slope in “leaky” (greater than the median) homes was expected, but not observed. It is important to note, however, that there was little variation in $F_{\text{inf}}$ in Windsor compared to other cities. For example, Allen et al. found that 10-day average $F_{\text{inf}}$ ranged between 0.24 and 1.00 in Seattle, WA, a city with a mild climate where relatively few homes have air conditioning (Allen et al., 2007). The use of air conditioners could influence residential PM\textsubscript{2.5} $F_{\text{inf}}$, and requires further studies examining $F_{\text{inf}}$ in different climate regions during the full range of seasons (Hystad et al., 2009).

### 4.1.3 Time-location patterns

When we stratified the personal hopane versus LUR NO\textsubscript{2} relationship by the amount of time spent indoors at home, we found two different relationships (Figure 5). While the resulting correlation coefficients were nearly identical ($r = 0.63$), the significant difference in the two slopes confirmed that the unadjusted personal hopanes versus annual average (LUR) NO\textsubscript{2} relationship masks two separate relationships. Further analysis showed that “time spent in transit” did not alter the personal hopane vs. LUR relationship. We would expect it to strengthen the relationship (i.e. the amount of personal variation we can explain with LUR and time in transit should be greater than the amount of variation we can explain with LUR alone). A possible explanation is that time-location patterns derived from a questionnaire are relatively crude. As well, differences in
types of roads and modes of transport are not accounted for in the “time spent in transit” period.

### 4.2 Limitations

Some important limitations of this study should be noted. First and foremost, the small sample size and restricted number of monitoring events over seasons limited the generalizability of our results. As well, it prevented further stratifications and analysis. Future studies with larger sample sizes and complete sets of indicators in at least heating and non-heating periods of the year would allow for a more robust assessment of factors that modify the hopane-TRAP relationships.

While dust does integrate exposures over time, the averaging time is not constant, presenting a challenge in these types of indoor exposure assessment studies. As well, while the LUR model and dust represent relatively long averaging times, indoor and personal air samples are limited to a five-day period. We attempted to account for some this temporal variability in the data by adjusting the annual average land use regression estimates (NO$_2$) with daily National Air Pollution Surveillance (NAPS) data for NO$_2$ (Tables A.1.a, b, and c). The adjustment was found to have little effect on the correlation between the hopane predictor variables and the LUR estimate. There is also a paucity of data available that accurately describes the heterogeneous distribution of dust and its major constituents within a home (Lioy et al., 2002). A greater understanding of how settled dust and its constituents (particularly hopanes) are distributed temporally and spatially within a home could provide further insight. Certain housing characteristics known to influence dust deposition, such as floor covering type, were not included in this analysis. One significant limitation included the fact that the settled house dust was collected independently and outside the time period associated with personal and indoor air – making comparisons of the hopanes among the different media difficult.

The fact that the LUR method used in Windsor models a gas (NO$_2$) and not a particle, which is what the hopanes are tracing, poses some additional limitations. Land-use regression methods have generally been applied successfully to model annual mean concentrations of TRAP particles (e.g. BC) in different settings (Hoek et al., 2008).

Another important consideration is whether hopanes infiltrate with the same efficiency as other traffic-related particulate matter (PM) constituents. PM infiltration varies with particle size, with a maximum infiltration efficiency at approximately 0.2-0.3
µm (Sarnat et al., 2006), while hopane particles in heavy duty diesel emissions peaked in size ranges larger than 0.18 µm particle diameter under light load conditions and less than 0.10 µm particle diameter under heavier load conditions (Riddle et al., 2007a). Hopane particles released from light duty gasoline vehicles had a bimodal size distribution of between 0.1 and 0.18 µm diameter (with a catalyst at operating temperature), and between 0.18 and 0.32 µm diameter (from visibly smoking vehicles) (Riddle et al., 2007b). An analysis of the how different PM constituents infiltrate homes, as well as their suspension and deposition, warrant further examination. As well, a critical assumption underlying the use of hopanes molecular tracers in this type of study is that the compound is chemically stable. Weitkamp and colleagues measured the heterogeneous oxidation of hopanes in a laboratory setting (Weitkamp et al., 2008). Aerosolized motor oil was exposed to OH inside a smog chamber; filter samples were analyzed for changes in the hopanes’ molecular composition. Hopanes were found to react at atmospherically significant rates across the entire range of experimental conditions. Chemical mass balance (CMB) analysis was performed to illustrate the effects of oxidation and it was found that as the level of oxidation increases, CMB analysis increasingly underestimates the contribution of gasoline vehicles, while the diesel estimates are largely unaffected (Weitkamp et al., 2008). This finding could reduce the relative utility of the hopane compound in assessing exposure to both gasoline and diesel powered emissions.

Another critical assumption made in this study is that hopanes have few sources beyond engine oil lubricants. We tested for indoor sources such as frying, broiling, grilling and sautéing, but found no significant effect on the hopane and land use regression relationship. A limited number of studies suggest that this may not be the case. In a 2008 study, three hopanes, amongst a suite of inorganic and other organic compounds, were measured from residential indoor, residential outdoor, and other outdoor microenvironments in Tampa Bay, Florida (Olson et al., 2008). The resulting indoor/outdoor ratio of the hopanes among nine pilot homes suggests a great deal of variability, and indoor/outdoor ratios > 1 in some homes suggests the potential for indoor sources of the three hopanes. However, the authors note that the highest indoor/outdoor hopane ratios were observed when outdoor concentrations were near the limit of detection, thus measurement error may be driving some of the high indoor-outdoor ratios. Populin et al. found hopane concentrations within samples of vegetable oils, mussels and clams as well as of human milk (Populin et al., 2004). It is thought that
hopanes migrate into foodstuffs via mineral oil and mineral paraffin products found in food packaging (cans, polysterene containers, waxed paper, for example) (Populin et al., 2004).

Figure 4.1 Ratio of residential indoor to residential outdoor concentration for selected PAHs and hopanes. The box defines the interquartile range and median of the distribution; the whiskers are the 10th and 90th percentiles of the distribution (adapted from Olson et al., 2008).

A continuation of this study is already underway in Vancouver, British Columbia, as part the Vancouver pilot project of the Canadian Healthy Infant Longitudinal Development (miniCHILD) study. In addition to LUR NO2 estimates, LUR models have been constructed for NO and BC. One advantage this presents is the ability to compare particles with particles (in the case of BC), as well as the ability to examine correlations between NO and hopanes. Nethery and colleagues found that while both the NO and NO2 models were developed using the same number of samples and possess similar R^2 values, only NO demonstrated a strong relationship with personal measurements in the study (Nethery et al., 2008). The Vancouver study also includes hopane sampling using
window dust wipes and settled dust collection. As well, this study contains a larger sample size with two sampling visits, twice the amount of dust collected, housing characteristics (including floor type) and direct infiltration measurements.

4.3 Validations

To validate and extend the conclusions of this study for future research, we recommend the following:

a) Construct similar studies with larger sample sizes, which would allow additional stratifications based on housing types, personal activity measures and seasonal variations, for example.

b) Assess spatial variation within the home. The indoor monitor and settled dust were collected from the most used living area and presume that the participant spends the majority of his/her time in that room. Activities and time spent in one particular area of the home may vary temporally.

c) To account for the potential confounding of indoor sources of hopanes, we recommend continuous indoor and outdoor measurements to assess the contribution of indoor sources (Olson et al., 2008).

d) Loading of dust has been used in several lead/dust studies and should be considered in future research (Kumar et al., 2009; Petrosyan et al., 2006; Roberts et al., 2005; Roberts et al., 2009). In this study, we opted to use the concentration of the hopanes in dust because the sampling surface in each home varied – from hardwood floor to short, shag carpet. By applying a metric that included sample area, such as loading – as opposed to total dust volume – we would be introducing a sampling bias based on the understanding that dust collects differently on different surfaces.

e) Future studies should incorporate NO (more variability) and BC (modeling particles) land use regression models for comparison.

4.4 Conclusion

There is growing evidence that traffic-related air pollution (TRAP) is related to a wide range of adverse health effects including asthma incidence, other respiratory and cardiovascular morbidities, and mortality (Brauer et al., 2002; Clark et al., 2010).
However, exposure assessment issues for TRAP are complex and need to be considered to reduce exposure misclassification and improve the power of investigations of health effects (Morgenstern et al., 2007; Arrandale et al., 2010). Our analysis demonstrated limited variation in infiltration and LUR NO$_2$ in Windsor, Ontario. In the former, we assume homes are relatively tightly sealed on account of the local climate. We would expect increasing variability in a city such as Vancouver. As well, we have shown traffic-related organic compounds, such as hopanes, can be readily detected in house dust and may incorporate both the spatial patterns of outdoor traffic pollution concentrations, and the infiltration of those pollutants into the indoor environment. Hopanes found in settled dust may trace TRAP as a long-term metric, hence its stronger correlation with the annual NO$_2$ average observed in Windsor.

Because house dust samples are routinely collected for assessing environmental exposures to biological pollutants in epidemiological studies of asthma and allergy, our study shows the potential for enhanced utility from dust samples already collected in other studies. However, storage and stability of hopane samples originating from PM$_{2.5}$ and settled dust remain an area of further investigation. Overall, improving the TRAP exposure estimates will increase the precision of epidemiologic studies and thereby provide a better scientific basis for minimizing the health effects of TRAP exposure.
Table A.1.a Correlations between summed dust hopane concentrations and land-use regression NO2 – LUR adjusted for temporal variability using fixed location government (NAPS) monitors.

<table>
<thead>
<tr>
<th>LUR NO2 concentration</th>
<th>Pearson</th>
<th>Spearman</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unadjusted n=26</td>
<td>0.46 (p&lt;0.05)</td>
<td>0.58 (p&lt;0.01)</td>
</tr>
<tr>
<td>7-day adjustment n=25</td>
<td>0.27 (p=0.19)</td>
<td>0.34 (p=0.09)</td>
</tr>
<tr>
<td>14-day adjustment n=25</td>
<td>0.22 (p=0.29)</td>
<td>0.24 (p=0.25)</td>
</tr>
<tr>
<td>28-day adjustment n=25</td>
<td>0.26 (p=0.20)</td>
<td>0.32 (p=0.11)</td>
</tr>
<tr>
<td>56-day adjustment n=25</td>
<td>0.33 (p=0.11)</td>
<td>0.40 (p&lt;0.05)</td>
</tr>
<tr>
<td>84-day adjustment n=25</td>
<td>0.40 (p&lt;0.05)</td>
<td>0.45 (p&lt;0.05)</td>
</tr>
<tr>
<td>112-day adjustment n=24</td>
<td>0.29 (p=0.17)</td>
<td>0.41 (p&lt;0.05)</td>
</tr>
<tr>
<td>140-day adjustment n=24</td>
<td>0.34 (p=0.10)</td>
<td>0.46 (p&lt;0.05)</td>
</tr>
</tbody>
</table>
Table A.1.b Correlations between summed personal hopane concentrations and land-use regression NO2 – LUR adjusted for temporal variability using fixed location government (NAPS) monitors.

<table>
<thead>
<tr>
<th>LUR NO2 concentration</th>
<th>Personal hopanes</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Unadjusted n=24</td>
<td>Pearson</td>
<td>0.20 (p=0.36)</td>
</tr>
<tr>
<td></td>
<td>Spearman</td>
<td>0.23 (p=0.27)</td>
</tr>
<tr>
<td>5-day adjustment n=24</td>
<td>Pearson</td>
<td>-0.10 (p=0.65)</td>
</tr>
<tr>
<td></td>
<td>Spearman</td>
<td>0.01 (p=0.96)</td>
</tr>
</tbody>
</table>

Table A.1.c Correlations between summed indoor hopane concentrations and land-use regression NO2 – LUR adjusted for temporal variability using fixed location government (NAPS) monitors.

<table>
<thead>
<tr>
<th>LUR NO2 concentration</th>
<th>Indoor hopanes</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Unadjusted n=40</td>
<td>Pearson</td>
<td>0.50 (p&lt;0.01)</td>
</tr>
<tr>
<td></td>
<td>Spearman</td>
<td></td>
</tr>
<tr>
<td>5-day adjustment n=40</td>
<td>Pearson</td>
<td>0.27 (p=0.09)</td>
</tr>
<tr>
<td></td>
<td>Spearman</td>
<td></td>
</tr>
</tbody>
</table>

Table A.2.a Correlations between individual dust hopanes* and two outdoor TRAP indicators.

<table>
<thead>
<tr>
<th>Variable</th>
<th>H17a</th>
<th></th>
<th>a, b, nor</th>
<th></th>
<th>a, b, hop</th>
<th></th>
<th>b, a, hop</th>
<th></th>
<th>H22S</th>
<th></th>
<th>H22R</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pearson</td>
<td>Spearman</td>
<td>n</td>
<td>Pearson</td>
<td>Spearman</td>
<td>n</td>
<td>Pearson</td>
<td>Spearman</td>
<td>n</td>
<td>Pearson</td>
<td>Spearman</td>
<td>n</td>
</tr>
<tr>
<td>Outdoor NO2 concentration</td>
<td>-0.10 (p=0.72)</td>
<td>-0.01 (p=0.97)</td>
<td>n=16</td>
<td>0.03 (p=0.92)</td>
<td>0.13 (p=0.64)</td>
<td>n=16</td>
<td>-0.13 (p=0.64)</td>
<td>-0.11 (p=0.69)</td>
<td>n=16</td>
<td>0.26 (p=0.33)</td>
<td>0.40 (p=0.13)</td>
<td>n=16</td>
</tr>
<tr>
<td>LUR NO2 estimate</td>
<td>0.38 (p=0.05)</td>
<td>0.49 (p=0.05)</td>
<td>n=26</td>
<td>0.38 (p=0.05)</td>
<td>0.44 (p=0.05)</td>
<td>n=26</td>
<td>0.45 (p=0.05)</td>
<td>0.58 (p=0.01)</td>
<td>n=26</td>
<td>0.42 (p=0.05)</td>
<td>0.51 (p=0.01)</td>
<td>n=26</td>
</tr>
</tbody>
</table>
Table A.2.b Correlations between individual personal hopanes* and two outdoor TRAP indicators.

<table>
<thead>
<tr>
<th>Variable</th>
<th>H17a Pearson</th>
<th>H17a Spearman</th>
<th>a_b_nor Pearson</th>
<th>a_b_nor Spearman</th>
<th>a_b_hop Pearson</th>
<th>a_b_hop Spearman</th>
<th>H22S Pearson</th>
<th>H22S Spearman</th>
<th>H22R Pearson</th>
<th>H22R Spearman</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outdoor NO2 concentration</td>
<td>0.30 (p=0.05)</td>
<td>0.31 (p=0.05)</td>
<td>0.25 (p=0.12)</td>
<td>0.24 (p=0.13)</td>
<td>0.19 (p=0.22)</td>
<td>0.0 (p=0.0.)</td>
<td>0.12 (p=0.46)</td>
<td>0.0 (p=0.0.)</td>
<td>0.11 (p=0.47)</td>
<td>0.22 (p=0.17)</td>
</tr>
<tr>
<td>LUR NO2 estimate</td>
<td>0.33 (p=0.06)</td>
<td>0.30 (p=0.06)</td>
<td>0.50 (p=0.01)</td>
<td>0.41 (p=0.01)</td>
<td>0.48 (p=0.05)</td>
<td>0.0 (p=0.05)</td>
<td>0.48 (p=0.01)</td>
<td>0.42 (p=0.01)</td>
<td>0.47 (p=0.01)</td>
<td>0.40 (p=0.05)</td>
</tr>
</tbody>
</table>

Table A.2.c Correlations between individual indoor hopanes* and two outdoor TRAP indicators.

<table>
<thead>
<tr>
<th>Variable</th>
<th>H17a Pearson</th>
<th>H17a Spearman</th>
<th>a_b_nor Pearson</th>
<th>a_b_nor Spearman</th>
<th>a_b_hop Pearson</th>
<th>a_b_hop Spearman</th>
<th>H22S Pearson</th>
<th>H22S Spearman</th>
<th>H22R Pearson</th>
<th>H22R Spearman</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outdoor NO2 concentration</td>
<td>-0.35 (p=0.08)</td>
<td>-0.34 (p=0.09)</td>
<td>-0.03 (p=0.90)</td>
<td>-0.12 (p=0.57)</td>
<td>-0.11 (p=0.60)</td>
<td>-0.07 (p=0.75)</td>
<td>-0.08 (p=0.49)</td>
<td>-0.14 (p=0.74)</td>
<td>-0.19 (p=0.74)</td>
<td>-0.07 (p=0.74)</td>
</tr>
<tr>
<td>LUR NO2 estimate</td>
<td>0.00 (p=0.70)</td>
<td>0.10 (p=0.63)</td>
<td>0.15 (p=0.50)</td>
<td>0.13 (p=0.54)</td>
<td>0.27 (p=0.19)</td>
<td>0.27 (p=0.20)</td>
<td>0.22 (p=0.30)</td>
<td>0.22 (p=0.30)</td>
<td>0.09 (p=0.68)</td>
<td>0.11 (p=0.62)</td>
</tr>
</tbody>
</table>

* Hopane legend
H17a 17α(H)-22, 29, 30-Trisnorhopane
a_b_nor 17α(H), 21β(H)-30-Norhopane
a_b_hop 17α(H), 21β(H)-Hopane
b_a_hop 17β(H), 21α(H)-Hopane
H22S 17α(H), 21β(H)-22S-Homohopane
H22R 17α(H), 21β(H)-22R-Homohopane

Table A.3 Correlations between summed hopane-related variables and land-use regression NO2 – Adjusted by presence or absence of attached garage.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Unadjusted LUR NO2 Concentration</th>
<th>[LUR NO2] for homes with attached garages</th>
<th>[LUR NO2] for homes without attached garages</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pearson</td>
<td>Spearman</td>
<td>Pearson</td>
</tr>
<tr>
<td>Dust hopanes</td>
<td>0.46 (p&lt;0.05)</td>
<td>0.58 (p&lt;0.01)</td>
<td>0.62 (p=0.06)</td>
</tr>
<tr>
<td></td>
<td>n=26</td>
<td></td>
<td>n=10</td>
</tr>
<tr>
<td>Personal hopanes</td>
<td>0.50 (p&lt;0.01)</td>
<td>0.43 (p&lt;0.01)</td>
<td>0.59 (p&lt;0.05)</td>
</tr>
<tr>
<td></td>
<td>n=40</td>
<td></td>
<td>n=16</td>
</tr>
<tr>
<td>Indoor hopanes</td>
<td>0.20 (p=0.36)</td>
<td>0.23 (p=0.27)</td>
<td>0.22 (p=0.64)</td>
</tr>
<tr>
<td></td>
<td>n=24</td>
<td></td>
<td>n=7</td>
</tr>
</tbody>
</table>
Table A.4 Correlations between summed hopane-related variables and land-use regression NO2 – Adjusted by infiltration efficiency relative to the median.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Unadjusted LUR NO2 Concentration</th>
<th>[LUR NO2] for homes with Infiltration Efficiency (Finf) &gt; median (0.24)</th>
<th>[LUR NO2] for homes with Infiltration Efficiency (Finf) &lt; median (0.24)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pearson</td>
<td>Spearman</td>
<td>Pearson</td>
</tr>
<tr>
<td>Dust hopanes</td>
<td>0.45</td>
<td>(p&lt;0.05)</td>
<td>0.52</td>
</tr>
<tr>
<td></td>
<td>n=25</td>
<td></td>
<td>n=13</td>
</tr>
<tr>
<td>Personal hopanes</td>
<td>0.51</td>
<td>(p&lt;0.01)</td>
<td>0.41</td>
</tr>
<tr>
<td></td>
<td>n=39</td>
<td></td>
<td>n=21</td>
</tr>
<tr>
<td>Indoor hopanes</td>
<td>0.20</td>
<td>(p=0.36)</td>
<td>0.23</td>
</tr>
<tr>
<td></td>
<td>n=24</td>
<td></td>
<td>n=15</td>
</tr>
</tbody>
</table>

Table A.5. Correlations between summed personal hopane concentrations and land-use regression NO2 – Adjusted by percent time spent in home and in transit relative to the median.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Unadjusted LUR NO2 Concentration</th>
<th>[LUR NO2] for participants who spend % time in home &gt; median (74.45)</th>
<th>[LUR NO2] for participants who spend % time in home &lt; median (74.45)</th>
<th>[LUR NO2] for participants who spend % time in transit &gt; median (3.45)</th>
<th>[LUR NO2] for participants who spend % time in transit &lt; median (3.45)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pearson</td>
<td>Spearman</td>
<td>Pearson</td>
<td>Spearman</td>
<td>Pearson</td>
</tr>
<tr>
<td>Personal hopanes</td>
<td>0.50</td>
<td>(p&lt;0.01)</td>
<td>0.63</td>
<td>(p&lt;0.01)</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>n=40</td>
<td></td>
<td>n=20</td>
<td></td>
<td>n=20</td>
</tr>
</tbody>
</table>


6: REFERENCE LIST


