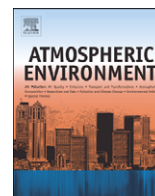




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Predicting personal exposure of Windsor, Ontario residents to volatile organic compounds using indoor measurements and survey data

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ABSTRACT

As part of a multi-year personal exposure monitoring campaign, we collected personal, indoor, and outdoor levels of 188 volatile organic compounds (VOCs). In 2005, data were obtained for 48 non-smoking adults from Windsor, Ontario in order to assess their exposure to VOCs based on their daily routines and characteristics of their homes. During the 8-week winter and summer sampling sessions, five repeated 24-h measurements were obtained for each home. This paper focuses on the analysis of 18 VOCs: 11 have been declared toxic as defined under the Canadian Environmental Protection Act, [1999. Statutes of Canada. Act assented to September 14, 1999. Ottawa: Queen's Printer. Available at Canada Gazette (Part III) 22(3): (Chapter 33). <http://canadagazette.gc.ca/partIII/1999/g3-02203.pdf>], and seven are commonly found in household and personal care products. Results of mixed effects models indicate that personal exposure to these VOCs can be largely predicted by indoor concentrations, with models including indoor concentrations found to have an r^2 value for the fixed effects ranging from 58.4% to 87.2% for the CEPA toxic VOCs and from 41.7% to 90.1% for the commonly found VOCs. Given that people spend the majority of their time inside their home, characteristics of the home such as air exchange rates, type of garage, and type of stove have a greater potential to impact personal exposures.

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1. Introduction

To date, many studies have relied upon ambient fixed-site monitoring stations such as Environment Canada's National Air Pollution Surveillance (NAPS) network to characterize population air pollution exposures. However, research has consistently shown that ambient concentrations of volatile organic compounds (VOCs) are often

much lower than corresponding personal exposure levels (Wallace et al., 1985; Anderson et al., 2001; Edwards et al., 2001; Kim et al., 2002; Adgate et al., 2004; Sexton et al., 2004a,b). Therefore, those epidemiological studies employing outdoor measurements may result in exposure measurement error and biased risk estimates (Payne-Sturges et al., 2004). This in turn may constitute a substantial source of uncertainty for risk-based regulatory decision making.

Few studies such as this one obtain simultaneous repeated measurements for personal, indoor, and outdoor concentrations of air pollutants using SummaTM canisters,

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as well as collect information relevant to characterizing exposure based on participant's daily activities, housing characteristics, and proximity to point sources (Edwards et al., 2001; Adgate et al., 2004; Sexton et al., 2007). Additionally, this study is particularly unique as it collected a total of 10 repeated measurements within two seasons per participant.

Windsor, Ontario was selected, as several local industries, including automotive manufacturing plants, long lines of idling trucks at the border between Canada and the US, and long-range transportation of air pollution from the north eastern US (Diamond and Parker, 2004; Ontario Ministry of Environment, 2007) have made poor air quality a concern for both residents and authorities. Comparison of Canadian and US time activity databases by Leech et al. (2002) also revealed that Canadian adults spend on average 2% less time outdoors in the winter and over 4% less time indoors at home in the summer than their US adult counterparts, which could result in small differences in exposure to air pollutants. However, in general, like in most industrialized countries, Canadians, spend between 80% and 90% of their time indoors, which includes at home, at work, in restaurants and in shopping centers (Klepeis et al., 2001; Leech et al., 2002; Schweizer et al., 2007).

The objectives of the study were to:

- understand exposure patterns of individuals residing in a community impacted by a range of different pollutant sources;
- describe indoor, outdoor and personal VOC levels within a Canadian city and study seasonal differences;
- investigate the relationships between indoor, outdoor and personal concentrations of different VOCs to identify environmental sources;
- identify reasons for differences in exposure through the development of personal prediction models, thereby identifying contributing factors captured by the participant time activity diary, the daily monitoring questionnaire (use of household and personal care products) and the baseline survey (housing characteristics).

This paper presents descriptive results for 18 VOCs with detailed personal modeling for 14 of them. Results of this study provide relevant Canadian information on exposure to several VOCs that are on the Canadian Environmental Protection Act (CEPA, 1999) Toxic Substances List.

2. Methods

2.1. Collection of data

Over 8 successive weeks per session, six of the 48 homes were sampled concurrently for 5 consecutive days, beginning on a Monday (day 0) and ending on a Saturday (day 5). At 24 ± 3 h intervals, teams of two technicians visited each home to change samplers, check equipment and administer questionnaires. Air samples were collected using cleaned and evacuated SummaTM canisters. At each home, three VOC canisters were deployed every 24 h. One 6.0L canister was placed inside the participant's home, typically within

the family or living room where participants spent a substantial amount of time, and another 6.0L canister was located in the backyard, several meters away from the home and away from combustion sources such as barbecues and automobiles. The participants also carried a 1.0L canister within a padded backpack and took it wherever they went in order to monitor their personal exposure to VOCs. The VOC canisters sampled at flow rates of 3.5 and 0.5 mL min⁻¹ for the 6.0 and 1.0L canisters, respectively. It was anticipated that 1395 VOC canisters would be deployed into the field; however, due to scheduling conflicts with participants, 1383 VOC canisters were actually set up within their respective locations. Of these, 82 samples were considered invalid due to VOC canisters that had flow gauge failures or that had collection duration of < 18 h or > 30 h, leaving a total sample size of 1301.

In order to prevent losses due to physical adsorption, chemical reactions of VOCs with co-collected ozone or other reactive pollutants, aqueous hydrolysis or biological degradation, all canisters were analyzed within thirty days of preparation. Once at the lab, the VOC canisters were placed on an automated cryogenic pre-concentrator coupled to a high-resolution GC-MS, operated in selected ion monitoring mode. One hundred and eighty-eight polar and non-polar VOCs were identified by using GC retention time. Laboratory equipment was cleaned every 2 months, analytical procedures remained constant over the duration of the study and MDLs were assessed once per calendar year.

Air exchange rates (AERs) were obtained for each home during both seasons using perfluorocarbon tracer gas (Dietz and Cote, 1982). Four sources of the tracer gas and one receptor were deployed on the main floor of the homes, away from potential sources of draft such as kitchens, doors and windows. Every 24 h for the five day sampling period, the receptors were collected and new ones were deployed. Using the tracer gas absorbed on the receptors and the square footage of each home, daily average air exchange rates were obtained.

Three questionnaires were administered to capture factors contributing to participants' personal exposure to various substances, including VOCs. An extensive baseline survey obtained characteristics about the home environment such as age and square footage of the house, number of occupants, type and use of garage, method of heating, stove type, and recent renovations. A daily monitoring questionnaire described participant use of personal care and household products. And a time activity diary kept track of an individual's activities in 15 min segments as they passed through one of six microenvironments (indoors at home, indoors away from home, in transit, at work, outdoors at home, outdoors away from home), as well as any exposure to second hand environmental tobacco smoke. Overall, excellent compliance was achieved on the questionnaires.

2.2. Recruitment of participants

An initial group of participants was identified from a larger Windsor Children's Respiratory Health Study. In

order to facilitate comparison between homes, adults were considered eligible for inclusion into our study if individuals were non-smoking, living in a detached home, and were not occupationally exposed to VOCs. Using these criteria, a pool of 90 eligible volunteers was established, and preference was subsequently given to households that were spatially distributed across Windsor. Forty-eight adults were originally recruited for both the winter and summer 2005 sampling sessions. However, as five participants withdrew from the study after the winter session due to moving, renovating homes, or summer travel plans, two additional participants were recruited for the summer. Therefore, the total sample size was 48 and 45, in winter and summer, respectively, with 43 of the same homes participating in both seasons.

2.3. Selection of VOCs

Due to the large number of chemicals collected using the VOC canister method, it was decided to focus on 18 of the 188 measured VOCs. Eleven of them have been declared toxic under CEPA, 1999 because they enter the environment in a quantity that may have immediate or long-term harmful effects on the environment, or may constitute a danger in Canada to human health (i.e. carcinogens) or to the environment upon which life depends: 1,1,1-trichloroethane, 1,2-dichloroethane, 1,3-butadiene, acetaldehyde, acrolein, acrylonitrile, benzene, dichloromethane, ethylene oxide, hexachlorobutadiene, and vinylchloride. The other seven VOCs are routinely found in the scientific literature to originate from household and personal care products and due to their use indoors, may be important sources of personal exposure: α -pinene, acetone, ethanol, isobutane, limonene, *m,p*-xylene, and toluene.

2.4. Statistical analysis

Preliminary analysis revealed that the distributions for the concentrations of the 18 VOCs were right skewed and a Shapiro–Wilks *W* test of normality indicated that the data were not normally distributed. The natural logarithm was therefore used for all remaining analysis with the transformed data approximating a Gaussian distribution. Non detectable VOC concentrations were assigned one-half of their respective method detection limit (MDL). Statistical analysis was conducted using SAS 9.1 (SAS Institute, 1999) and figures were produced using Statistica 7 (SoftStat, 2004).

A generalized mixed linear model, using a first-order autoregressive (AR(1)) correlation structure, was applied to the repeated measurements in order to account for the correlations existing within the data. Analysis was completed on a combined winter and summer dataset as it was assumed that major predictors of exposure would be relatively constant, and that intermittent exposure to VOCs created during activities such as use of cleaning products, would be better captured using this increased sample size.

To test for significant microenvironmental and seasonal differences, a complex model with a total of 14 covariance estimates was derived. In this model, heterogeneity in the covariance structure *R* was specified to indicate that measurements within each season and measure of exposure (i.e. indoor winter, outdoor summer) would be more correlated. The basic model used to perform tests of significance is

$$(\ln)Y_{ijkl} = \beta + \beta_1(\text{season})_{ijk} + \beta_2(\text{environment})_{ijl} + \beta_3(\text{environment*season})_{ij} + b_{il} + \varepsilon_{ijkl} \quad (1)$$

where Y_{ijkl} represents an observed VOC exposure for subject *i*, on day *j*, in environment *k* for season *l*; α signifies the regression intercept; season_{ijk} represents the season for subject *i* on day *j* in environment *k*; environment_{ijl} signifies the environment for subject *i* on day *j* in season *l*; $\text{environment*season}_{ij}$ represents interaction between season and environment for subject *i* on day *j*; β denotes the fixed effect of *X* (season, environment or environment*season) on *Y*; b_{il} represents the random subject effect $\sim N(0, \sigma^2 b_l)$, and ε_{ijkl} denotes the random error associated with each subject *i*, on day *j* in season *l* in environment *k*. Additionally, annual geometric means, which were used to create indoor/outdoor (I/O), personal/outdoor (P/O) and personal/indoor (P/I) ratios, were generated by applying similar mixed-effects models as presented in Eq. (1).

Four VOCs having a significant number of non-detectable samples (acrylonitrile, ethylene oxide, hexachlorobutadiene and vinylchloride) were excluded from further analysis. The fixed effects of dichotomous, categorical, and continuous predictors were tested for the remaining 14 VOCs using the mixed-effects model. Baseline survey data and daily monitoring questionnaire data were either categorical or dichotomous, while VOC concentrations, AERs, exposure to environmental tobacco smoke from the time activity diary and land use variables generated from Geographic Information System (GIS) ESRI Arc View 9.0 (Redlands, CA, USA) were continuous. As six households were sampled during each week, tests for spatial clustering were conducted. Moran's *I* for the summer and winter sampling were close to 0 indicating an absence of clustering or dispersion, and therefore that the sites were randomly distributed. Model random effects were study participants within each season and on an annual basis. A generic equation capturing the model used for these analyses is presented below:

$$(\ln)Y_{ijl} = \alpha + \beta X_{ijl} + b_{il} + \varepsilon_{ijl} \quad (2)$$

where Y_{ijl} represents an observed VOC exposure for subject *i*, on day *j*, in sampling session *l*; α signifies the regression intercept; X_{ijl} denotes either baseline survey, daily questionnaire, or time activity diary data, air exchange rates, indoor or outdoor VOC concentrations, or land use variables for subject *i*, day *j* and season *l*; β represents the fixed effect of *X* on *Y*; b_{il} denotes the random subject effect $\sim N(0, \sigma^2 b_l)$, and ε_{ijl} represents the random error. Variance components allowed for differences between and within subject error for each seasons.

Inclusion of indoor VOC concentrations in the different models prevented the inclusion of most other variables due to collinearity. As a result, two models were created, one including corresponding indoor VOC concentrations and one excluding them. The latter models were created in order to explore potential sources of exposure (other than indoor concentrations) and to provide insight into the predictive ability of survey data and ambient measurements.

Each fixed effect was tested individually and variables with a very low r^2 value or a non-significant p -value were eliminated from further analysis. All fixed effects tested in the model were identified from a comprehensive review of the literature and hypothesized to contribute to personal exposure to VOCs. A manual stepwise procedure was then followed, and the model that yielded the best Akaike's Information Criterion (proximity to 0 indicating the best model fit) and statistical significance of the variables was then chosen.

As statistically significant seasonal differences were observed for many VOCs, effect modification for season was tested within each model. Furthermore, given recent findings by Dodson et al. (2007), it was hypothesized that garage type may have an effect on AER. As a result, the effects of garage type on AERs were tested for, and given that a significant relationship was found, all models including garage type were adjusted for the confounding effects of AER.

Final models were evaluated by correlating predicted with observed values. This was done only for the fixed effects as the alternative predicted values would include the Bayes estimates for each subject and therefore would result in an r^2 predicted value that was too high. By squaring the correlation coefficient obtained, an r^2 for the fixed effects was obtained. Residual analysis was also conducted to test the assumption of a normal distribution and homoscedasticity.

3. Results and discussion

3.1. Description of VOC concentrations, and air exchange rates

Table 1 summarizes MDLs, detection frequencies and geometric means for the 18 VOCs in both seasons, within all three exposure categories. As previously stated, acrylonitrile, ethylene oxide, hexachlorobutadiene and vinylchloride were removed from further analysis due to a significant number of non-detectables.

Seasonal variations in VOC concentrations are associated with differences in emission sources and rates, temperature, and to the rate of reactions with other chemicals. Not surprisingly, concentrations of the majority of the VOCs were higher in summer than in winter. Nine of the 14 VOCs had statistically significantly higher concentrations in summer than in winter for all three measures of exposure. Exceptions included 1,1,1-trichloroethane, 1,3-butadiene, benzene, dichloromethane and isobutane, which did not show significant seasonal differences ($\alpha = 0.05$) in at least one measure of exposure. 1,3-butadiene was the only VOC to consistently demonstrate lower geometric mean concentrations in summer than winter across all three exposure measures. In addition to similar findings at Environment Canada's outdoor National Air Pollution Surveillance sites (Curren et al., 2006), this trend was also observed within the complementary US Environmental Protection Agency's Detroit Exposure Assessment Research Study (US EPA, 2007). The US study reported higher outdoor benzene levels as well as higher personal, indoor and outdoor 1,3-butadiene levels in winter than in summer ($p < 0.05$) (Vette et al., 2006). Curren et al. (2006) attributed this finding to increased atmospheric reactivity of 1,3-butadiene outdoors in the summer due to higher ambient temperatures than those observed in winter.

Table 1
Geometric mean of VOCs calculated using generalized mixed linear models

| VOC | MDL | % Below detection limit | | Indoor GM ($\mu\text{g m}^{-3}$) | | Outdoor GM ($\mu\text{g m}^{-3}$) | | Personal GM ($\mu\text{g m}^{-3}$) | | I/O | P/O | P/I |
|-----------------------|-------|-------------------------|--------|------------------------------------|---------|-------------------------------------|--------|--------------------------------------|---------|--------|--------|------|
| | | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | | | |
| 1,1,1-Trichloroethane | 0.006 | 0 | 0 | 0.22* | 0.30* | 0.13* | 0.13* | 0.20 | 0.30 | 1.99 | 1.90 | 0.96 |
| 1,2-Dichloroethane | 0.006 | 0 | 0.2 | 0.10 | 0.19 | 0.04 | 0.04 | 0.10 | 0.23 | 3.47 | 3.93 | 1.13 |
| 1,3-Butadiene | 0.006 | 0.2 | 0 | 0.13* | 0.11* | 0.07 | 0.05 | 0.16* | 0.14* | 2.14 | 2.70 | 1.26 |
| Acetaldehyde | 0.009 | 0 | 0.3 | 18.44 | 39.63 | 3.51 | 6.64 | 19.97 | 39.46 | 5.60 | 5.81 | 1.04 |
| Acrolein | 0.027 | 3.3 | 0.6 | 1.29 | 5.02 | 0.14 | 0.58 | 1.16 | 4.04 | 8.97 | 7.67 | 0.85 |
| Acrylonitrile | 0.031 | 96.4 | 93.1 | <0.031 | <0.031 | <0.031 | <0.031 | <0.031 | <0.031 | – | – | – |
| Benzene | 0.014 | 0 | 0 | 1.68* | 1.95* | 0.97* | 0.79* | 1.69* | 1.96* | 2.08 | 2.08 | 1.00 |
| Dichloromethane | 0.004 | 0 | 0 | 0.93 | 1.56 | 0.32 | 0.39 | 1.30* | 1.69* | 3.42 | 4.22 | 1.23 |
| Ethylene oxide | 0.062 | 94.9 | 59.4 | <0.062 | <0.062 | <0.062 | 0.11 | <0.062 | 0.07 | – | – | – |
| Hexachlorobutadiene | 0.014 | 92.0 | 86.1 | <0.014 | <0.014 | <0.014 | <0.014 | <0.014 | <0.014 | – | – | – |
| Vinylchloride | 0.001 | 15.8 | 41.2 | 0.01 | <0.001 | 0.01 | <0.001 | <0.001 | 0.01 | – | – | – |
| α -Pinene | 0.005 | 2.0 | 0.2 | 4.51 | 26.69 | 0.04 | 0.33 | 6.88 | 29.75 | 96.72 | 126.15 | 1.30 |
| Acetone | 0.009 | 0 | 0.6 | 32.15 | 137.01 | 4.04 | 10.01 | 38.74 | 115.83 | 10.43 | 10.53 | 1.01 |
| Ethanol | 0.027 | 0.2 | 0.2 | 713.15 | 1168.70 | 4.30 | 6.70 | 661.42 | 1022.71 | 170.00 | 153.16 | 0.90 |
| Isobutane | 0.007 | 0 | 0 | 18.54 | 35.94 | 1.39* | 1.10* | 23.51* | 34.39* | 20.81 | 22.92 | 1.10 |
| Limonene | 0.01 | 7.7 | 1.2 | 9.91 | 19.80 | 0.03 | 0.09 | 22.14 | 47.90 | 287.63 | 668.62 | 2.32 |
| <i>m,p</i> -Xylene | 0.005 | 0.2 | 0 | 4.43 | 11.15 | 0.85 | 1.63 | 5.04 | 10.06 | 5.98 | 6.06 | 1.01 |
| Toluene | 0.003 | 0 | 0 | 11.95 | 30.14 | 2.51 | 4.61 | 13.90 | 24.90 | 5.58 | 5.46 | 0.98 |

* Not a statistically significantly difference ($p < 0.05$) between winter and summer VOC concentrations.

Ratios between personal, indoor residential and outdoor residential concentrations are also presented in Table 1. Commonly found VOCs such as α -pinene, ethanol, isobutane and limonene had *I/O* and *P/O* ratios ranging from 20.81 to 668.92, indicating predominantly indoor sources. In contrast, traffic related VOCs (1,3-butadiene, benzene, acetaldehyde, *m,p*-xylene, and toluene) all had *I/O* and *P/O* ratios of <10 , with 1,3-butadiene and benzene having the lowest *I/O* and *P/O* ratios between 2.08 and 2.70. We would have expected 1,3-butadiene and benzene, which have no indoor sources in non-smoking households, to have an *I/O* and *P/O* ratios of less than one, indicating that they are being generated outdoors. However, the relatively large *I/O* and *P/O* ratios found in this study suggest that 1,3-butadiene and benzene originate from outdoor sources and permeate the home via infiltration, and that they appear to have built up an indoor concentration due to little loss from subsequent reaction or sorption. With the exception of limonene with a *P/I* ratio of 2.32, all *P/I* ratios were very close to 1.0 (0.85–1.30) indicating a large contribution of indoor concentrations to personal exposure levels.

Due to the late arrival of the perfluorocarbon tracer gas, AER data were collected for 32 homes in winter and 42 homes in summer. In winter, the geometric mean for AERs was 0.32 h^{-1} (CI: $0.26\text{--}0.40 \text{ h}^{-1}$), which was significantly ($\alpha = 0.05$) different from that of summer AERs, 0.19 h^{-1} (CI: $0.15\text{--}0.24 \text{ h}^{-1}$). As recent evidence has indicated that residential air flows may differ based on the location of garage (Dodson et al., 2007), mean AERs were also calculated stratifying by garage type. It was found that significant mean AER differences exist in households with attached garages, when compared with those who had a detached garage ($p = 0.0019$). This was taken into account in subsequent modeling by including AER in models where garage type was a significant predictor.

3.2. Determinants of personal exposure

Table 2 summarizes the results of the mixed-model analysis, which essentially indicate that indoor concentrations were highly predictive of personal exposures. This is not surprising since the results of our time activity diary resemble those of a large Canadian Human Activity Pattern Survey (Leech et al., 2002), and are similar to time activity patterns found within the US (Klepeis et al., 2001) and in other developed countries (Schweizer et al., 2007). Windsor participants spent approximately 80% of their time indoors at or away from home, 10% of their time at work, 5.5% of their time in transit, and about 4.5% of their time outdoors at or away from home. Therefore, given that participants spent the majority of their time inside their home, there is a greater potential for their exposure to possible environmental hazards within this microenvironment.

Overall, inclusion of indoor concentrations in the exposure modeling resulted in an r^2 value for the fixed effects ranging from 57.8% to 87.2% for CEPA toxic VOCs and from 41.7% to 90.1% for commonly found VOCs.

Exclusion of indoor VOC concentrations within the modeling reduced the r^2 value for the fixed effects to 0.01–73.1% for CEPA toxic VOCs and 0.01–43.4% for commonly found VOCs. Although in some cases, such as that of isobutane, exclusion of indoor concentrations resulted in an uninformative model (r^2 for the fixed effects = 0.01%), other predictors in 8 of the 14 VOC models could explain over 30% of the variability in personal exposures, yielding valuable insights into sources of exposure.

One important source of exposure that was identified for traffic-related VOCs was garage type; individuals residing in homes with attached garages had the highest levels of exposure. This categorical variable was defined as having four levels (attached with a connecting door, attached no door, detached garaged, no garage) and proved to be a significant predictor for benzene ($p < 0.0001$), 1,3-butadiene ($p = 0.0204$), *m,p*-xylene ($p = 0.0036$), and toluene ($p = 0.0013$). Mean concentration of benzene in homes generated using mixed models adjusted for season, were 2.58, 1.23, 1.48, and $1.48 \mu\text{g m}^{-3}$ for homes with an attached garage having a connecting door, those with a detached garage, those with attached garages having no connecting door, and those having no garage, respectively. The higher benzene concentrations observed in homes with attached garages was likely due to infiltration of VOCs indoors from the garage, either continuously from cracks in the foundation and/or intermittently when the connecting doors were opened. Batterman et al. (2007) report that average air flows from attached garages to houses are $6.5 \pm 5.3\%$ of the houses' overall air exchange, and that garage sources of benzene contribute to approximately 50% of indoor concentrations. In addition to vehicle exhaust, evaporative gasoline from vehicles, lawnmowers, and storage containers are important contributors to total VOC compositions in garages (Batterman et al., 2007). Decreasing the exposure of people living in homes with attached garages to gasoline-related VOCs could include: not idling the cars inside the garage, reducing the amount of stored gasoline inside the garage, sealing the cracks in the foundations between the house and the garage, providing ventilation in the garage to the outdoors, as well as maintaining a positive pressure differential between the house and garage (Batterman et al., 2007). Fig. 1 illustrates personal exposure levels to benzene based on presence of attached (with connecting door), detached and no garage for each participant in both seasons combined.

In addition to garage type, garage use was also a significant predictor for 1,3-butadiene ($p = 0.0228$) with those who used their garage for parking showing a higher mean concentration (mean = $0.22 \mu\text{g m}^{-3}$, as compared to $0.15 \mu\text{g m}^{-3}$ (no garage), $0.14 \mu\text{g m}^{-3}$ (storage), $0.14 \mu\text{g m}^{-3}$ (park and storage) and $0.16 \mu\text{g m}^{-3}$ (other)). No significant interaction was detected between garage type and garage use.

Personal exposure to traffic-related VOCs (1,3-butadiene, acetaldehyde, benzene, *m,p*-xylene, and toluene) were also predicted by outdoor concentrations, which is not surprising given that these pollutants typically originate outdoors. However, when indoor concentrations

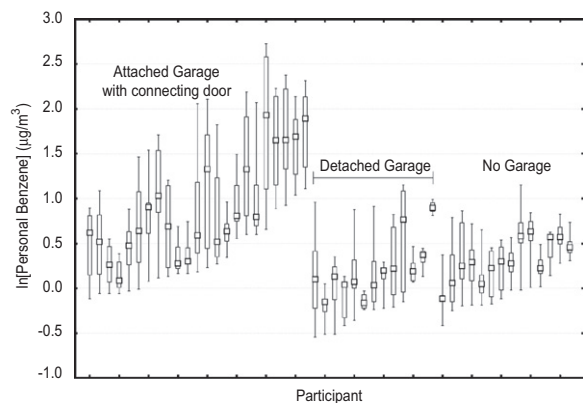


Fig. 1. Participant personal benzene levels categorized by type of garage (10 repeated measurements).

were included in the model, the significance of outdoor concentrations was lost with the exception of 1,3-butadiene. Therefore, although outdoor concentrations can be informative regarding pollutants originating outside the home, indoor levels are a much better estimate of personal exposure when available.

Presence of a gas stove was found to be a significant predictor of personal levels of acrolein in both the model including and the model excluding indoor concentrations. Homes having a gas stove had a significantly different ($p = 0.0113$) mean personal exposure to acrolein ($2.68 \mu\text{g m}^{-3}$) than homes that had electric stoves ($2.03 \mu\text{g m}^{-3}$). Due to incomplete combustion of fuels, gas stoves have been identified as a source of exposure to aldehydes such as acrolein, and therefore this trend is not unexpected (Health Canada, 2004). Including stove type and AERs in the season adjusted model, allowed for the creation of a good predictive model for acrolein (r^2 for the fixed effects = 73.1%). Unfortunately for the non-traffic-related VOCs, the models were relatively uninformative when indoor concentrations were excluded as very few variables gained significance at $\alpha = 0.05$. Generally, the model relied primarily on AERs with an adjustment for season; as a negative association between concentration of VOCs and AERs was found, increasing the AERs in homes could provide a means to decrease personal exposure to VOCs. One exception to this is limonene, which, as expected, also included use of air fresheners in the model. Results indicated that when air fresheners were used, the mean personal limonene concentration was $39.95 \mu\text{g m}^{-3}$ as compared to $29.79 \mu\text{g m}^{-3}$ when they were not (significantly different: $p = 0.0066$).

One variable that may have improved these models was major renovations within the last month. Unfortunately, this data could not be included in the annual model as this question was only asked during the winter months when the baseline survey was usually administered. Analysis of only the winter data revealed that this variable was a significant predictor for personal exposure to acetaldehyde at the $\alpha = 0.05$ level and at the $\alpha = 0.1$ level for α -pinene, 1,2-dichloroethane, and ethanol (results not shown). It is expected that if this question had been

asked in both winter and summer, the increased sample size would have allowed for significant results in the annual model as it is believed that major renovations in the last month could be a large source of exposure for these VOCs.

It was hoped that the model for 1,1,1-trichloroethane would yield some interesting insights into sources of exposure. It was expected that 1,1,1-trichloroethane would have a substantial number of non-detects since, with the exception of military use, it was phased out in Canada by 1996. In addition, the atmospheric half-life of 1,1,1-trichloroethane is about 26 weeks (Mackay et al., 2006), and its estimated atmospheric lifetime is 6 years (Agency for Toxic Substances and Disease Registry (ATSDR), 2006). Although it is anticipated that outdoor sources are contributing to the personal exposure levels observed, outdoor concentrations of 1,1,1-trichloroethane did not prove to be a significant predictor of personal exposures ($p = 0.5008$). A model excluding indoor concentrations did not provide any insights into sources of exposure as only AER proved to be predictor of personal exposure and accounted for only 11.9% of the variability. Instead, a model including indoor concentrations could predict approximately 83.9% of the fixed effects. According to the Agency of Toxic Substances & Disease Registry's Toxicological Profile, about 300 million pounds of 1,1,1-trichloroethane was produced in the US in 2000, but less is being made today (ATSDR, 2006). Although Michigan's releases into air from facilities that produce, process or use 1,1,1-trichloroethane are relatively small ($56 \text{ pounds year}^{-1}$ in 2003), those of nearby Ohio are much greater ($3231 \text{ pounds year}^{-1}$) (ATSDR, 2006) and may be a contributing factor to concentrations observed.

Although Windsor, Ontario has numerous industrial VOC point and fugitive sources, land use variables input into the model, such as distance to Detroit, distance to Ambassador Bridge, and the number of VOC point sources within a 4000 m radius were not significant predictors of personal exposure to VOCs. It is anticipated that these results are a consequence of the relatively small intra-urban variability in outdoor concentrations (Wheeler et al., 2008), as well as the inability of outdoor concentrations to strongly predict personal exposures.

These results are based on results from non-smoking, non-occupationally exposed adults living in single family homes in Windsor, Ontario. This study does not capture all sources of exposure and therefore cannot draw conclusions based on other, larger, contributors. For example, the ATSDR, 2005, benzene ToxGuide estimates that approximately 50% of the entire nationwide exposure to benzene is a result of tobacco smoke. Although we captured exposure to environmental tobacco smoke in the time activity diary, the amount of time exposed was so small and intermittent that it did not gain significance in the models.

4. Conclusions

SummaTM canisters were used to obtain personal, indoor and outdoor concentrations of VOCs allowing us to observe the relationships between seasons, microenvironments and

a select number of VOCs. Not surprisingly, the concentrations of the majority of VOCs were higher in the summer than in the winter with indoor concentrations contributing more to personal exposures than outdoor concentrations. Results of mixed effects models indicate that personal exposure to these VOCs can be largely predicted by indoor concentrations, with models including indoor concentrations found to have an r^2 value for the fixed effects ranging from 58.4% to 87.2% for the CEPA toxic VOCs and from 41.7% to 90.1% for the commonly found VOCs. Given that people spend the majority of their time inside their home, there is a much greater potential for exposure to VOCs indoors.

The collection of VOCs in indoor and outdoor micro-environments also occurred in 2006 with the participation of asthmatic children. Results for this monitoring campaign will be forthcoming.

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