Windsor, Ontario Exposure Assessment Study: Design and Methods Validation of Personal, Indoor and Outdoor Air Pollution Monitoring.

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ABSTRACT

The Windsor, Ontario Exposure Assessment Study (WOEAS) evaluated the contribution of ambient air pollutants to personal and indoor exposures of adults and asthmatic children living in Windsor, Ontario, Canada. Additionally, the role of personal, indoor and outdoor air pollution exposures upon asthmatic children's respiratory health was assessed. Several active and passive sampling methods were applied, or adapted, for personal, indoor and outdoor residential monitoring of nitrogen dioxide (NO_2), volatile organic compounds (VOC), particulate matter ($PM_{2.5}$ and PM_{10}), elemental carbon (EC), ultrafine particles (UFP), ozone (O_3), air exchange rates, allergens in settled dust and particulate associated metals. Participants completed five consecutive days of monitoring during the winter and summer of 2005 and 2006. During 2006 in addition to undertaking the air pollution measurements asthmatic children completed respiratory health measurements including peak flow meter tests and exhaled breath condensate, as well as tracking of respiratory symptoms in a diary. Extensive quality assurance and quality control steps were implemented including the collocation of instruments at the National Air Pollution Surveillance (NAPS) site operated by Environment Canada and at the Michigan Department of Environmental Quality site in Allen Park, Detroit. During field sampling duplicate and blank samples were also completed and these data are reported.

In total, 50 adults and 51 asthmatic children were recruited to participate resulting in 922 participant days of data. When comparing the methods employed in the study with standard reference methods, field blanks were low, bias was acceptable with most methods being within 20% of reference methods. Duplicates were typically within less than 10% of each other, indicating that study results can be used with confidence.

This manuscript covers study design, recruitment, methodology, time activity diary, surveys, and quality assurance and control results for the different methods employed.

INTRODUCTION

The challenges of assigning exposure on an individual basis have received increasing commentary in the literature with several studies being conducted to better assess the impact of exposure misclassification in health effects research.[1-3] The impact of air pollutants on the health of susceptible populations such as the elderly,[4] diabetics,[5] children[6] and asthmatics[7-10] indicate that exposure sources and baseline health are important factors for guiding regulatory decisions related to ambient or indoor air quality guidelines or standards.

Human exposure to air pollution is influenced by indoor and outdoor sources, as well as personal activities; the complex interplay of these factors complicates the interpretation of personal exposures. This research will address these challenges by assessing exposures for adult populations and asthmatic children.

There are a number of air pollutants known to have potential human health impacts including particulate matter ($PM_{2.5}$ and PM_{10}), nitrogen dioxide (NO_2), allergens in settled dust, volatile organic compounds (VOC), and ozone (O_3), as well as constituents of $PM_{2.5}$ such as elemental carbon (EC), ultrafine particles (UFP) and metals. It is important to identify all potential sources of personal exposures to these pollutants in an effort to understand source-specific impacts as well as the potential for misclassification of exposure and the role that this may have on discerning health impacts. Outdoor sources include traffic emissions, industrial emissions, long range transport, secondary formation of pollutants in the atmosphere, and personal activities such as refuelling vehicles and using a barbecue. Indoor environments are also affected by outdoor generated pollutants via infiltration;[11-13] therefore housing characteristics that affect infiltration are important to consider. Indoor sources of exposure also include off-gassing of building materials, combustion processes such as cooking, use of personal care and cleaning products, cigarette smoke and presence of pets.[14] Several studies that have included personal, indoor and outdoor pollutant measurements have found that personal exposures can exceed both indoor and outdoor concentrations.[15-18] These findings suggest that not all personal exposures are captured by residential indoor and outdoor measurements. In the case of particulate air pollution, simple movement by residents in the home can resuspend particles from clothes or carpeting. Exposure to nitrogen dioxide indoors may be increased by being close to gas stoves. Recent literature suggests that time spent in traffic can contribute significantly to personal exposures for a variety

of air pollutants.[19-21] In addition, locations such as work, school, restaurants and other indoor locations are also known to contribute to personal exposures.[22,23]

As part of the Border Air Quality Strategy (BAQS) a personal exposure assessment study was conducted in 2005 and 2006 in Windsor, Ontario to understand air quality issues in this area. One objective was to examine the relationships between indoor and outdoor concentrations and personal exposures to a variety of air pollutants, including PM, O_3, NO_2 and VOC. The goal of this research was to develop a better understanding of factors affecting these relationships for adults and asthmatic children. A second objective was to examine the role of personal, indoor and outdoor air pollution exposures upon asthmatic children's respiratory health with the goal of identifying which sources of exposure had the greatest impact upon health.

Windsor is a relatively small geographical area (120.6 km^2) that is impacted by a variety of ambient air pollution sources. It is the site of one of the major border crossings to the United States and is therefore impacted by large volumes of commercial truck traffic. Additionally, Windsor also has several local industries such as automobile manufacturing. Finally, there is long-range transportation of pollutants from the United States.[24,25]

This manuscript presents the study design and methodology as well as important data for method validations including quality assurance and control. It includes summary results for recruitment, time activity diaries and surveys.

MATERIALS AND METHODS

Study Design

Health Canada and the University of Windsor conducted a personal exposure study with Windsor adults in 2005 and asthmatic children in 2006. Personal, 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) of each year. Pollutants included in the study were NO_2, VOC, O_3, UFP, EC, $PM_{2.5}$, PM_{10} and components of PM including EC, nitrate and metals. Ancillary measurements included air exchange rates (AER), temperature, relative humidity, and settled dust, as well as respiratory health measures collected for asthmatic children in 2006. Sampling began Monday evenings at approximately 4:00 pm, and ended on Saturday evenings at approximately the same time. At the end of each 24 ± 3 hour interval, teams of two technicians visited each home to refurbish sampling equipment, check for mechanical malfunctions, and administer questionnaires. All data were collected over 8 consecutive weeks

per season, with a total of six homes being sampled concurrently. Personal sampling was conducted by having participants carry a small backpack housing air pollution monitoring equipment. Participants were asked to keep the backpack with them throughout their daily activities, although no direct methods were applied to assess compliance. If they were in one location for an extended period of time (i.e., at school or work) they were instructed to place the backpack close to them with the sampling inlet facing up. Indoor residential monitoring was conducted by placing equipment inside the participant's home (i.e., typically within the family or living room where participants spent a substantial amount of time). Outdoor residential monitoring was located in the backyard, several meters away from the home and away from any combustion sources such as barbecues and driveways. Both indoor and outdoor residential sampling was conducted at breathing height (1.5m). The Windsor study design paralleled that of the United States Environmental Protection Agency's (EPA) Detroit Exposure and Aerosol Research Study (DEARS) with respect to sampling periods, days of the week, and survey and questionnaire design[18] so that environmental data from both cities could be used to investigate border air quality issues in the region. Table 1 provides details for the methods and instrumentation employed in the Windsor study.

Approval was obtained from Health Canada and the University of Windsor Research Ethics Boards to conduct this study and all personal information is protected according to the Canadian Access to Information Act and the Privacy Act.

Sample Population – Recruitment and Retention

In 2005, an initial pool of potential volunteer participant families was identified from the Windsor Children's Respiratory Health Study.[7] From the pool of potential participants, homes meeting inclusion criteria were randomly selected and their adult residents were approached for participation. Adults were considered eligible for study inclusion if they were non-smoking, living in a detached home, had an asthmatic child, were not occupationally exposed to VOC, and did not have any workplace restrictions on carrying the personal monitoring equipment. Using these criteria a pool of 90 eligible volunteers was established. Among the eligible volunteers preference was subsequently given to households that were spatially distributed across Windsor. In 2006, eligible participants included physician-diagnosed asthmatic children between the ages of 10 and 13 years. Of the available pool of candidates drawn from a previous study of 186

asthmatic children[4] further consideration was given to ensuring an approximately even spatial distribution of homes across Windsor. Figure 1 identifies residential locations.

Given the above mentioned eligibility criteria and the need to ensure that there would be sufficient statistical power to assess the role of personal, indoor and outdoor air pollution exposures upon asthmatic children's respiratory health a power calculation was conducted. Statistical power estimates were calculated and applied for both years by taking into account the repeated measures design of the study. Due to correlations within the data, standard power calculations were adapted to account for dependencies. Using a methodology described by Killip et al.[26] the effective sample size was first calculated. This involved estimating the intraclass correlation coefficient (ICC) for the study participants using personal monitoring data collected in previous publications; their values of the variance of FEV_1 scores in asthmatic children were also used.[27,28]

The ICC is a ratio of the variability between subjects to the total variability:

$$ICC = \rho = \frac{s_b^2}{s_b^2 + s_w^2} \quad (1)$$

where s_b^2 is the variance between individuals and s_w^2 is the variance within individuals.

The effective sample size was then estimated by dividing the total number of planned observations (n=480) each year by the design effect. As outlined by Killip et al.[26] the design effect was estimated as:

$$DE = 1 + \rho(M - 1) \quad (2)$$

where M represents the number of observations taken in each cluster. PASS software 2007 (NCSS, Utah, USA) was then applied to estimate the percent change in FEV_1 for a 10 $\mu g/m^3$ increase in $PM_{2.5}$ that could be detected with a power of 80%.

Based on these calculations, 48 participants were recruited in each of the years. The study design was not intended to recruit a representative selection of the population but rather to identify homes of susceptible individuals.

157 Participant retention was encouraged through several techniques. Prior to recruitment, two
158 technicians visited each residence to demonstrate the monitoring equipment as well as to answer
159 any questions. During this visit, consent was obtained, the baseline housing questionnaire was
160 completed, visit schedules were discussed, and suitable locations for the indoor and outdoor
161 monitoring equipment were identified. At the end of each season participants were provided with
162 a personalised report describing their individual data in comparison to others who were
163 monitored during the same week. These reports contained guidance material from the Canadian
164 Mortgage and Housing Corporation and Health Canada on different air pollutants. These were
165 provided at a meeting where the principal investigator discussed study findings and was
166 available to answer participants' questions. Between sampling seasons in 2006, children were
167 invited to a pizza party with their parents to provide them with the opportunity to meet other
168 participating children and discuss their experiences.

169 **Passive Samplers**

170 During 2005 and 2006, personal, indoor and outdoor Ogawa passive samplers (Ogawa &
171 Company, Pompano Beach, FL, USA) were used to measure exposures to O_3 and NO_2. The O_3
172 badge was only used for personal and outdoor monitoring. The sampler used a single nitrite-
173 coated quartz-fibre filter purchased from the manufacturer; when O_3 is present in the sampled air
174 it diffuses into the filter and oxidizes the nitrite to nitrate on an equimolar basis. The NO_2 badge
175 uses a single carbonate-coated quartz-fibre filter, also purchased from the manufacturer, to trap
176 NO_2.[18] Sampling times were approximately 24 ± 3 hours for each badge. The badges were
177 located within the breathing zone for the personal samples and were placed in a manifold-type
178 device that housed all the active and passive samplers thus ensuring constant air flow across the
179 face of the passive badges. Figure 2 demonstrates the personal, indoor and outdoor monitoring
180 equipment setup. All Ogawa badges were refrigerated during storage and shipping.
181 The Ogawa filters were analysed according to the Ogawa Standard Operating Protocols (SOP),
182 the only deviation from the O_3 method was that the protocol assumes two filters were used for
183 measurements and in the Windsor situation there was only one filter. The sampling rate was
184 therefore half the rate cited in the protocol. After exposure, the O_3 nitrite-coated filter was
185 extracted with ultra-pure (Milli-Q) water, whereas the NO_2 carbonate-coated filter was extracted
186 with 0.09% (v/v) hydrogen peroxide. Both extracts were analyzed by ion chromatography (IC)
187 using Dionex DX-300 or DX-600 IC systems (Dionex, Sunnyvale, CA, USA). Nitrate from the

188 extracts of the O_3 filter was analyzed using a Dionex IonPacAS15 column and gradient elution,
189 whereas nitrate and other anions extracted from the nitrogen dioxide filter was analyzed using a
190 Dionex-AS4A column with carbonate/bicarbonate eluent. Calibration checks were performed
191 daily before analysis of the field samples and once in every 10-15 samples using standards
192 prepared from NIST-traceable standards.
193 Selected VOC in air were collected using cleaned and evacuated Summa canisters. During both
194 years indoor and outdoor measurements were made at each of the residences using 6.0 L
195 canisters deployed every 24 hours. During 2005, the adult participants also carried a 1.0 L
196 Summa canister within the padded backpack to monitor their personal exposures. The canisters
197 sampled at flow rates of 3.5 mL/min and 0.5 mL/min for the 6.0 L and 1.0 L canisters,
198 respectively. The passive canister sampling systems included four basic components: an in-line
199 Swagelok™ filter with 2 μm stainless-steel sintered filter to eliminate particulates, a restrictor, a
200 Veriflow SC423XL back-pressure flow regulator and a vacuum gauge. The back-pressure flow
201 regulator ensured that approximately a 0.5 to 1 psi pressure drop across the restrictor was
202 maintained until the canister was within 1 to 2 psi of reaching atmospheric pressure, after which
203 the regulator no longer maintained a 1 psi differential across the orifice, resulting in a drop in
204 flow rate. The flow controllers were assembled in the laboratory and leak tested. United States
205 EPA Compendium Method TO-15 requires that the flow controllers be certified clean prior to
206 use. The flow controllers were certified as clean by passing a humidified, high-purity air through
207 the flow controller into evacuated canisters which were then analyzed by GC-MS; if no VOC
208 concentrations were greater than 0.2 ppbv the flow controllers were determined to be clean. The
209 certified flow controllers were then capped with Swagelok fittings and shipped to the site for
210 sampling. The Summa canister analysis methods followed the US EPA method TO-15. The 2005
211 VOC analytical methods and quality assurance data have been published elsewhere.[17]

212 **Continuous Measurements**

213 Continuous measurements of $PM_{2.5}$, UFP, EC, and temperature / relative humidity (RH) were
214 collected indoors and outdoors at each residence using DustTrak (Model 8520, TSI, ST Paul,
215 MN, USA), PTrak (Model 8525, TSI, ST Paul, MN, USA), Aethalometer (AE-42, Magee
216 Scientific Company, Berkley, CA, USA), and Smart ReaderPlus 2 (ACR Systems Inc., Surrey,
217 BC, Canada) monitors, respectively. These methods and their validation are included in a

218 companion manuscript detailing their performance including precision and Limits of Detection
219 (LOD) calculations[29] methods are summarised briefly below.
220 Two DustTrak instruments (one indoors and one outdoors) were deployed at each residence;
221 these are optical instruments capable of measuring particles from about 0.3 micrometers (μm) in
222 diameter up to 2.5 μm. The PTrak measurements of UFP number concentrations were also
223 conducted indoors and outdoors but for only 10 minutes per hour in each location (20 x 30-
224 second averages) due to their limited alcohol storage reservoir volume. Although PTraks count
225 all particles from 20 nm to 1 μm, the instrument is considered to monitor mainly UFP because
226 approximately 80-99% of these particles are below 0.1 μm.[30]
227 The Aethalometer measures light absorption from particles collected on a quartz fibre tape.
228 Because only a limited number of Aethalometers were available, only one unit was operated at
229 each residence to sample EC both indoor and outdoor; the intake was programmed to switch
230 between indoor sampling and outdoor sampling every 30 minutes during the day and hourly at
231 night. The PTrak instruments were programmed to synchronize with the Aethalometer as it
232 switched from an indoor to an outdoor air intake.
233 In 2006, the asthmatic children carried an active sampling personal DataRAM (pDR) (Thermo
234 Scientific, Waltham, MA, USA) to measure continuous $PM_{2.5}$; the cut size was ensured by
235 pumping the intake air through a 1.8 Lpm $PM_{2.5}$ personal environmental monitor (PEM)
236 (Chempass System R&P/Thermo, Waltham, MA, USA) with no filter present. Like the
237 DustTrak, the pDR uses optical means to measure particles smaller than 2.5 μm, and is
238 laboratory-calibrated to a NIST particle standard.[22] The pDR uses a laser at higher frequency
239 (i.e. lower wavelength) than the DustTrak; therefore the highest sensitivity regions for these
240 instruments occurred at somewhat different diameters.
241 **Active Samplers**
242 Particulate matter ($PM_{2.5}$ and PM_{10}) was measured using the R&P Chempass multi-pollutant
243 PEM (Chempass System R&P/Thermo, Waltham, MA, USA) as described in Demokritou et al.[31]
244 Teflon filters were pre-conditioned for 24-hr before mass measurement, following US EPA
245 quality assurance guidelines,[32] at Health Canada's Archimedes M3™ Buoyancy-Corrected
246 Gravimetric Analysis Facility.[33] Average daily standard deviation of blank reference filter
247 measurements was typically ± 0.5-0.6 μg (n=10-14), resulting in daily laboratory detection
248 limits of 1.5-1.6 μg based on 3 times standard deviation.[34] Method detection limits for the

249 present study are based on variability introduced throughout all pre-weighing and post-weighing
250 steps, including storage for the entire time elapsed from removal of the filter from its packaging
251 to the final post-weighing (typically 1-2 months).
252 $PM_{2.5}$ and PM_{10} measurements were collected in all three locations (indoor, outdoor, and
253 personal) during the 2005 monitoring. During 2006, PM_{10} was measured only indoors and
254 outdoors using the PEM; the personal PM_{10} inlet was replaced by an active personal DataRam
255 (pDR) as described above. Technicians ensured that the $PM_{2.5}$ and PM_{10} target flow rates were
256 4.0 Lpm, with an acceptable range of ± 20%. Flow rates were assessed pre and post sampling
257 every 24-hr using a soap bubble flow meter (AP Buck, Orlando, FL, USA).
258 In addition to the PEM samplers there were two low flow particulate samplers operating at 0.8
259 Lpm. One collected elemental carbon / organic carbon (EC/OC) onto a pre-fired quartz fibre
260 15mm filter (Pall-Gelman, Missisauga, ON, Canada) and the other collected particulate-
261 associated nitrate through a denuder which scrubbed gas-phase nitrates prior to collecting the
262 particulates on pre-coated 15mm glass fibre filters (Pall Gelman, Missisauga, Ontario). The
263 EC/OC filter analysis has yet to be completed and will not be included in this paper. Due to
264 problems with field laboratory protocols the nitrate denuders were unreliable in 2005 and part of
265 the winter 2006 and only the acceptable duplicate data will be included in this paper.
266 During the winter and summer of 2006 a pre-fired quartz 37mm fibre filter (Pall-Gelman,
267 Missisauga, Ontario) was placed in line after the pDR to collect the pre-separated particulates
268 ($PM_{2.5}$) that passed through the PEM inlet and the pDR unit. This filter was in place for the entire
269 5 days of each season at a flow rate of 1.8 Lpm. During summer 2006 the same filter type was
270 also located indoors for the same five day period. The filters will be analysed for traffic markers
271 (hopanes) in an attempt to understand longer term exposure to traffic related air pollutants.
272 Analysis of these filters is still pending so details are not included in this paper.
273 Settled dust was sampled by technicians using the High Volume Surface Sampling System
274 (HVS3) vacuum.[35] Samples were typically collected during visits scheduled one week after the
275 conclusion of the winter monitoring to ensure that the request not to vacuum for a week prior to
276 the settled dust collection did not interfere with the air pollution monitoring. The sampling
277 required a minimum of two square meters of the floor in the living area to be vacuumed for a
278 four minute period. Technicians were asked to ensure the collection of at least one gram of dust;
279 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 one gram of dust could not be collected from the two square meters, for example on hardwood floors, further living space was sampled to ensure sufficient sample for all intended analyses; technicians noted any increase in sample area. Samples were stored in amber glass jars and a brown paper bag for shipping.

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 300-150µm and <150µm size fractions and recombined for analysis of house dust mite allergen (DerP1 and DerF1), cat allergen (Feld1), endotoxin and (1→3)-β-D-Glucan.

Allergens were assayed by monoclonal enzyme immunoassays (ELISA). Endotoxin analyses were completed in accordance with the 1996 version of the American Industrial Hygiene Association protocol described in the Field Guide for the Determination of Biological Contaminants in Environmental Samples. (1→3)-β-D-Glucan was analysed using the limulus ameobocyte lysate based method.

Metals in airborne PM and in the settled dust were determined using ICP-MS.[36-38]

Fixed-Site Monitors

Environment Canada maintains two outdoor National Air Pollution Surveillance (NAPS) sites in the city of Windsor. These sites monitor a variety of pollutants, including particulate matter and criteria gases such as NO_2, O_3 and VOC. The College Road East site, located to be representative of urban ambient air pollution found in Windsor, was used for conducting the majority of the WOEAS duplicate sampler deployment and instrument comparisons to assess bias and precision. Duplicate 24-hr samples for all WOEAS active and passive samplers, excluding the continuous instruments, were collocated at this NAPS site in the summer of 2005 and 2006 and in the winter of 2006; the site was being refurbished during winter 2005 and was therefore not available for that sampling season.

The Environment Canada method for measuring continuous $PM_{2.5}$ at the Windsor NAPS sites was a tapered element oscillating microbalance (TEOM) (Thermo Fisher Scientific, MA, USA) with an inlet heater temperature of 40 °C using the Sample Equilibrium System (SES) sample dryer. The SES dryer contains specially-designed Nafion tubing inlets on the main flow to minimize potential for particle loss. The dryer lowers the relative humidity in the main flow, and allows for mass transducer operation at 5 °C above the peak air monitoring station temperature. NO_2 was measured by Environment Canada using the chemiluminescence method (Thermo

311 Model 42 Nitrogen Dioxide Analysers, TEI Inc, Franklin, MA). O_3 was measured by
312 Environment Canada using the ultraviolet photometric method (Thermo Model 49C, TEI Inc,
313 Franklin, MA). There were no Environment Canada NAPS instruments available to measure
314 UFP, PM_{10}, or EC. All NAPS instruments operated on a continuous basis and time-averaged
315 values were calculated to correspond directly with the WOEAS instrument measurements.
316 The WOEAS duplicate passive and active samplers included the Chempass PM system and the
317 Ogawa badges. These were set up by technicians at the Windsor NAPS site and timed to
318 correspond with the rest of the study's personal sampling periods, typically 4pm to 4pm.[39]
319 Duplicate indoor and personal samples were also deployed for PM_{10} for five 24-hr periods in
320 winter 2006.[38] Duplicate personal samples were collected by technicians who were located in
321 Windsor and replicated typical participant activities.
322 The WOEAS Chempass filter-based particulate samples were also collected at the Michigan
323 Department of Environmental Quality (MDEQ) site at Allen Park, Detroit. These measurements
324 were undertaken to ensure the comparability of WOEAS and DEARS methods, and to enable
325 comparison of WOEAS particulate samplers with a dichotomous sampler located at this site.[18]
326 All samples were collected for 24-hr from 9am to 9am, for two days per week on Tuesday and
327 Wednesday. Data from this location were used to examine possible bias between the PEM used
328 in this study and the dichotomous sampler measuring $PM_{2.5}$ and PM_{10}-$PM_{2.5}$, which is an
329 equivalent Federal Reference Method (FRM). This represents the only comparison of the PEM
330 used in this study with a FRM. The filters used in the FRM dichotomous sampler underwent
331 gravimetric analysis using a Mettler UT20 balance. The filters were placed in Petri dishes in a
332 controlled environmental chamber for a minimum of 24-hr to allow the filters to equilibrate. The
333 temperature remained between 19 - 23 °C +/- 2 °C and the humidity was kept between 30 - 40%
334 +/- 5% for a minimum of 24-hr. The balance was warmed up for a minimum of 1 hour prior to
335 use. Following the internal balance calibration procedure, a manual audit of the balance was
336 performed using a 100 mg mass (ASTM Class 1 or NBS Class P weight). The balance was
337 viewed as operational if the audit indicated the result was within 10 µg of the expected value. In
338 addition, a laboratory reference filter was then analyzed; reference filter mass had to be within 20
339 µg of the expected value. At the end of every 5 measurements, the precision for the balance had
340 to be less than 1 µg; the precision test was repeated until this was achieved. Further tests
341 involved analyzing a laboratory filter blank for long and short-term drift of filter weights.

342 Following the analysis of every 25th filter, a random re-weigh of at least one filter was
343 performed. The original and re-weigh values had to differ by less than ± 6 μg of the original
344 weight. If not, all of the 25 filters were re-weighed until the required precision was achieved.
345 Comparison of the personal monitors to a reference method is a crucial requirement in
346 determining the reliability of the data. We used two regression methods to compare the
347 samplers. Ordinary least squares (OLS) regression minimizes the vertical distance between the
348 data points and the line of best fit. This type of regression is sometimes said to assume no error
349 in the x-axis measurements. However, it is also the best estimate of the y-axis values given the
350 x-coordinate even though the x-coordinate is in error.[40] Orthogonal regression recognizes that
351 error may occur in both measurement methods, and therefore minimizes the perpendicular
352 distance to the line of best fit. However, this approach assumes equal variance in the two
353 measurement methods, which is seldom the case. Reduced major axis (RMA) regression is a
354 form of weighted orthogonal regression in which the ratio of the variances is used to modify the
355 angle between a data point and the line of best fit. The RMA method results in the best estimate
356 of the "true" relationship between the two methods. It has been recommended as the best way to
357 determine whether candidate methods can be certified as reference methods for environmental
358 measurements.[41] Therefore, we carried out both OLS and RMA regression methods on the
359 $PEM_{2.5}$ and PEM_{10} monitors versus the dichotomous sampler.

360 **Air Exchange Rates**

361 A daily assessment of air exchange was undertaken in each of the homes using perfluorocarbon
362 tracer (PFT) gas.[42] Four sources of the tracer gas were set up on the main floor of the home at the
363 beginning of the first day to allow the gas to equilibrate for 2 hours during equipment setup. The
364 emitters were deployed for the duration of the 5 day sampling period in each season. One
365 receptor (capillary adsorption tube) was installed daily at a location away from any potential
366 ventilation or heating sources. The total amount of the tracer gas absorbed by the receptors
367 combined with the participant reported square footage of each home and the emission rate of the
368 emitters was used to calculate a daily average air exchange rate (AER) for each home.

369 **Questionnaires and Time Activity Diaries**

370 Adult participants in 2005 and the parents of asthmatic children in 2006 completed two different
371 questionnaires to assess potential sources of exposure: (1) a baseline questionnaire to obtain
372 information on housing characteristics that did not change over time and (2) a daily questionnaire

373 to obtain information on daily activities such as cooking and cleaning. During the second season
374 a shorter baseline survey was administered by the recruitment technician to assess any new
375 renovations and changes undertaken in the residence since the last season of sampling. In both
376 years participants completed a time activity diary throughout the day, noting their activities as
377 well as their presence in various locations in 15 and 30 minute intervals for the adults and
378 children, respectively. The adult diary included details on whether the individual cooked or
379 cleaned; this was deemed less likely for the children and was removed from their diaries. Both
380 diaries included information on whether the participants were in close proximity to any smokers,
381 and for how long, as this was deemed important in terms of increased exposure to air pollutants.
382 Key locations noted on the diaries included; at home, outside at home, in transit, at work /
383 school, outside away from home and inside away from home. Multiple responses could be
384 included if activities or locations changed within the time interval. The activities were then
385 coded for all diaries and coding was confirmed by manually assessing 10% of all diaries to
386 assure consistency in interpretation of the descriptions. All surveys and diaries were
387 independently entered twice and compared electronically to each other to identify any
388 discrepancies in the data entry.

389 **Health Measurements**

390 During the 2006 sampling period asthmatic children also conducted respiratory health
391 assessments, including forced expiratory volume in one second (FEV_1) which was estimated
392 using a PiKo-1 electronic peak expiratory flow/FEV_1 meter (Ferraris Medical, Louiseville, CO,
393 USA); these instruments have comparable responsiveness with pneumotachographs.[43] Twice
394 daily (first thing in the morning, again at bedtime), three consecutive FEV measurements were
395 made prior to taking any asthma medications. During the technician visit in the evenings an
396 exhaled breath condensate sample was collected using an RTube (Respiratory Research Inc.,
397 Charlottesville, VA, USA). Participants sat and breathed at tidal volumes orally into a
398 mouthpiece attached to a cold condenser for 10 min. Approximately 1 mL of breath condensate
399 was collected. The condensed breath was then transferred to several microtubes and stored at –
400 20°C and then –80°C until analyses. The sample was first analysed for amylase to test for saliva
401 contamination. This was followed by the measurement of the oxidative stress biomarkers,
402 thiobarbituric acid reactive substances (TBARS) and 8-isoprostane, and the inflammatory
403 cytokine Interleukin-6 (IL-6); all laboratory methods are described in detail in Liu et al.[4]

Children's time activity diaries included space to report any symptoms, including cough, wheeze, chest tightness and difficulty breathing. These symptoms were scored from 0 – 4 where 0 indicated no symptom and 4 indicated the worst symptoms. Scoring was explained to the participants at the start of the study and technicians reviewed the diaries each day to assess completeness.

Quality Assurance

Laboratory detection limits (LDL) were estimated as three times the standard deviation of the laboratory blanks, with field detection limits (FDL) being defined as 3 times the standard deviation of the field blanks. Field blanks comprised approximately 10% of all samples. Duplicate outdoor samples were collected at the NAPS sites.

The quality assurance program included the calibration of flow rates, leak tests, collection of routine field blanks and determination of precision and accuracy during sampling as well as for the chemical analyses. Various quality control samples were used to determine accuracy and precision of the chemical analyses and to diagnose any sources of contamination.[36,38]

Blank corrections were applied when more than 50% of the field blanks were greater than the lab detection limit (LDL). In these situations, a field detection limit (FDL) was then calculated as being three times the standard deviation of the field blanks. Sample data were then adjusted by subtracting the median of the field blanks. Any resulting values which were lower than the LDL were substituted with ½ LDL. Samples that were above the LDL but below the FDL were not changed.

All data were assessed for validity using the following criteria. Any samples requiring a specific flow rate were tested at the beginning and end of each 24-hr sampling period; if the end flow rate was operating at a flow more than 20% above or below the target flow rate, they were deemed invalid. Samples were also deemed invalid if they were deployed for more than 30 or less than 18 hours. Other criteria for invalidating samples included: presence of insects found during assembly, evidence that filters were mishandled in the field or laboratory, or noted sources of contamination either in the field or the laboratory.

Duplicate comparisons for all methods, where these data exist, were used to assess precision estimates. Data from standardised methods used at the NAPS and MDEQ were used to yield estimates of bias.

The following bias definition, also frequently referred to as the fractional or percent difference was utilised:

$$\text{Bias} = \frac{A-T}{T} \tag{3}$$

where A is the instrument value and T is the true value. This returns a positive or negative number, which can be multiplied by 100 to produce a "percent bias" normally reported.

A precision calculation of two identical instruments is often defined as the absolute value of the difference between one instrument reading and the mean of the two, divided by that mean, which works out to be the difference divided by the sum:

$$\text{Abs [A-B]/(A+B)} \tag{4}$$

The idea in this definition is that when the true value is unknown, it can be assumed that it is near the average of the two instruments.

In many cases in this study it was not just the comparison of two instruments, but rather several, as all continuous instruments underwent pre and post side by side intercomparisons. It was determined that a reasonable approach would be to compare the continuous instruments to the median of their readings for any given 3-minute sampling period. In this case, the median was assumed to be close to the "true" reading. The resulting formula for the bias-corrected precision is:

$$\frac{Abs[A'-T]}{T} \tag{5}$$

where A´ is the bias-corrected value for instrument A and T is the median. For each collocation session, the correction factor for each instrument was calculated as the ratio of the mean of all the instruments' medians to the mean of each instrument. The correction for bias was then calculated by multiplying this correction factor in that particular session by the mean (or median) of each individual instrument. This approach was typically applied to the continuous instrument data.

For each pollutant, the laboratory results were combined with log sheet data to calculate concentrations. Several coding flags were included to address any field or sample specific issues that arose during the sample collection; each sample was coded as being valid, flagged or invalid. All analyses were conducted using SAS v. 9.1 (SAS Institute Inc, NC, USA).

RESULTS

Participant Retention Rates

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 new 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. There were 5 male and 45 female adult participants in total.

During the winter and summer of 2006 the total sample size for the asthmatic children was 48 and 48 respectively, with 45 individuals participating in both seasons. In total, 51 asthmatic children were recruited for this study with one winter and two summer sets of siblings being followed. The children were between 10 and 13 years of age with the majority of them (n=31) being male. This age range was selected because children of this age are able to participate in personal monitoring and complete time activity diaries with minimal supervision. Table 5 shows participant data.

A total of 922 participant days were completed. This was only 4% less than the intended 960 days, and was due to the small number of participants who were unavailable in the second season. The compliance for the data collection, including wearing the backpack and completing the diaries, was high. Participants reported to the technicians daily highlighting any non-compliance due to restrictive activities.

Nitrogen Dioxide

The mean WOEAS NO_2 level over two years of sampling at the central NAPS site was 20 ppb for the Ogawa badges, compared to 16 ppb for the collocated Environment Canada chemiluminescence method (Table 2). The Ogawa badges had a median bias compared to the NAPS method of 17% and a median precision of 7%.

Ozone

493 Ozone sampling was only undertaken during the summer periods. The mean value for both years
494 was 36 ppb for the Environment Canada UV photometric method compared to 26 ppb for the
495 WOEAS O_3 collocated at the NAPS site (Table 2), resulting in a median bias of -24%. The
496 Ogawa O_3 badges had a median precision of 9%.

497 **Volatile Organic Compounds (VOC)**

498 VOC sampler deployment was intended to be 1395 samplers in 2005 (total number of indoor,
499 outdoor, and personal samplers for all participants) and 930 samplers in 2006 (total number of
500 indoor and outdoor samplers for all participants). However, the few participant retention issues
501 described previously resulted in slightly lower numbers of deployed canisters. In addition, a
502 small number of deployed VOC canisters were deemed invalid and therefore excluded from the
503 analysis due to flow gauge failures, which was determined as a canister sampling time of less
504 than 18 hr and/or the canister not being collected within 30 hr of deployment. As a result, the
505 total sample size for VOC canisters was 1294 in 2005 and 872 in 2006. A Health Canada report
506 including the full descriptive statistics for the 2005 and 2006 VOC data is available upon
507 request.[44]

508 **Continuous Instruments**

509 Results from the continuous instruments are presented in a companion paper.[29] Briefly, for both
510 years and both seasons there were 902 and 834 person days of indoor and outdoor data collected
511 for the DustTraks, 656 and 657 person days for the Aethalometers, 666 and 659 person days for
512 the PTraks, and during 2006 personal pDR data resulted in 358 person days of data. The
513 DustTrak and pDR had positive biases of a factor of about 2.5 and 1.6, respectively, compared to
514 the PEM. However, their average bias-corrected precisions were within 10%, indicating that a
515 proper correction for bias would bring them into very good agreement with standard methods.
516 Both instruments had limits of detection of approximately 5 $\mu g/m^3$. Although no standard
517 methods exist to establish the bias of the Aethalometer and P-Trak, their precision estimates
518 were within 20% for the Aethalometer and within 10% for the P-Trak.

519 **Chempass $PM_{2.5}$ and PM_{10} PEM**

520 *Allen Park Site.* Intercomparisons between the two different size fractions of the PEM $PM_{2.5}$ and
521 PM_{10} with the FRM dichotomous sampler were conducted over the four seasons. Due to missing
522 data in either of the two size fractions of the FRM dichotomous sampler a total of 38 days were
523 available for comparison of both methods. The slopes and intercepts of the two lines of best fit

524 are provided in Figures 3a and 3b. As can be seen, because of the excellent agreement of both
525 PEMs with the two dichotomous fractions, the OLS and RMA lines virtually overlap. The PEM
526 $PM_{2.5}$ showed very good agreement ($R^2 = 0.98$) with the collocated Allen Park FRM
527 dichotomous sampler (Table 3, Figure 3a), although with a small positive median bias of 9%.
528 The bias was significant (95% confidence interval 1.07-1.15) and only 3 of the 38 paired samples
529 were below the 1:1 line. The $PM_{2.5}$ PEM showed good precision (median 7%). The PM_{10} PEM
530 sampler also showed excellent agreement ($R^2 = 0.97$) with the reference dichotomous sampler,
531 with a median positive bias of 9% and a median precision of 6% (Table 3, Figure 3b). The PEM
532 estimate of coarse particle concentrations (PM_{10}-$PM_{2.5}$) agreed well with the dichotomous
533 sampler, with a small positive bias of 9% and a slightly worse median precision of 12% (Table
534 3).
535 Details of the method evaluation for metals in airborne PM in the Windsor study have been
536 described previously, detailing quality assurance procedures during sampling, handling and
537 analysis, analytical method comparisons and collocated duplicates[36,37].
538 *NAPS site*. Based on its excellent performance in comparison with the FRM dichotomous
539 sampler, the decision was made to use the PEM as the standard method and then compare it with
540 the TEOM at the Windsor NAPS site. As found in other studies,[45] the TEOM displayed strong
541 losses in the one winter season with collocated data having a median negative bias of 52% (Table
542 4). The summer median bias was still negative but less so at -26%.
543 **Particulate Associated Nitrate**
544 The mini-PEM samplers for measuring particulate-associated nitrate required the coating of a
545 denuder each time the samplers were used. Due to field laboratory problems the consistency of
546 the coating was unreliable and all data from 2005 and part of winter 2006 had to be discarded.
547 The median bias for the NAPS based WOEAS duplicate samplers was 7% there was no
548 reference method available at the NAPS site for bias calculations.
549 **Settled Dust**
550 A single dust sample was collected from each of the residences in 2005 and 2006 resulting in a
551 total of 93 samples. Samples from the separated settled dust of grain size <300um were analysed
552 for dust mite allergens Der p1 and Der f 1, cat allergen Fel d 1, endotoxin and (1,3)-D-Glucan.
553 For all of these analyses only the Der p1 samples were found to have the majority of the samples
554 below the LDL, 35 and 23 of all of the samples collected in 2005 and 2006 and as such, the

555 replacement of ½ the LDL was not applied. For the remainder of the allergen analyses, ½ of the
556 LDL was applied when any were below LDL.

557 **Air Exchange Rate**

558 The average daily AER for each residence was calculated using the estimated house volume and
559 the known amount of PFT that was emitted and trapped on the corresponding daily capillary
560 adsorption tube (CAT). A total of 30 and 45 homes in the winter and summer of 2005 and 33 and
561 46 homes in winter and summer 2006 had valid daily AER calculations. Homes missed in the
562 winter season of 2005 and 2006 were due to difficulties in financial contracting that resulted in
563 the late start of the AER sampling.

564 **Questionnaire and Time Activity Diary**

565 The majority of the homes were detached, single family dwellings (n=92) with electric stoves
566 (n=77). Approximately half of the homes had either an attached garage or no garage at all.
567 Summary statistics are provided in Table 5.
568 Data obtained via the daily questionnaire on activities that occurred in the residence included
569 information on daily cooking, cleaning, presence of smokers, ventilation use, and candle use. Of
570 all the homes included in the study only four homes had a smoker present at any point in the
571 study, 257 sampling days had open windows, and on 55 of the sampling days candles were used.
572 The adults' activity patterns did not alter significantly between winter and summer therefore both
573 seasons were combined in Table 5. Adults spent approximately 80% of their time indoors at
574 home or indoors away from home, 10% of their time at work, 5.5% of their time in transit, and
575 about 4.5% of their time outdoors. The children spent significantly more time indoors at home in
576 the summer than in the winter (77.4% vs. 68.7%) and indoors away from home (9.8% vs. 5.8%).
577 In winter, the children spent significantly more time at school (18.5%), compared with summer
578 (0.6%). These children were also found to spend significantly more time outdoors in the summer,
579 than during the winter, likely due to more favourable weather conditions (23.5°C versus 0.5°C
580 mean temperatures in each season). The children spent 4.8% of their time in the yard at home or
581 close to home in the summer compared with only 0.7% in winter. No significant differences in
582 the average time spent in transit were observed (3.7% in winter and 3.4% in summer).

583 **Health Measurements**

584 During 2006, the asthmatic children completed peak flow measures, provided exhaled breath
585 condensate samples and noted any symptoms in their diaries. The best three forced expiratory

586 flows in one second (FEV_1) and forced vital capacity (FVC) trials were used. The total number
587 of morning and evening FEV_1 (422 and 425 measurements) and FVC (424 and 428
588 measurements) were completed. The exhaled breath condensate sample was only completed each
589 evening with technician supervision, resulting in 458 samples.
590 The children used the time activity diary to record symptom prevalence and this indicated that
591 only a small percentage of children reported any cough (2.6%), wheeze (1.6%), chest tightness
592 (1.7%) or difficulty breathing (1.3%) at any point during the 10 days of sampling.

593 ## DISCUSSION

594 There are several personal exposure studies that have included healthy adults and asthmatic
595 children, and of these studies the EPA DEARS conducted at the same time as WOEAS is the
596 most comparable in terms of the adult population and the methods employed.[18] The main
597 difference in the study designs was that DEARS included a randomly selected population. The
598 WOEAS 2006 asthmatic children represent a susceptible population with a similar age group to
599 other personal monitoring studies that have been conducted.[28,46,47] The sample size for the 2006
600 WOEAS health measurements was determined using a power calculation which was based on
601 results obtained from Delfino et al.[28] The Delfino et al. [28] study had a population of 19
602 participants which completed a total of 710 FEV_1 manoeuvres; this is comparable with the
603 Windsor results where there were a total of 847 FEV_1 manoeuvres available for analyses.
604 Children in the age range of 10 – 13 years have been shown to be capable of carrying out study
605 activities and complying with study requirements. Retention of participants in both years was
606 high (43 out of 48 in both 2005 and 2006 completed 2 seasons of data collection); the small
607 numbers of losses were due to home renovations and scheduling, with no losses attributed to
608 study fatigue. Spatial representativeness was more difficult to ensure seasonally due to
609 participants' scheduling requirements.
610 The Ogawa NO_2 badge median precision of 7% in this study is slightly higher than the findings
611 by Mukherjee et al.[48] who reported that 8 paired duplicate samples collected at two different
612 locations (4 pairs at each site) over 3, 4 and 7 days had percent relative standard deviation values
613 less than 3.6%. The Quebec City study conducted by Gilbert et al.[49] found 7-day Ogawa samples
614 had an average precision of 4.5%.
615 Similarly, the Ogawa O_3 method had a precision of 9%, although it underestimated the
616 concentrations in comparison to the NAPS measurements with a median bias of -24%. Possible

617 reasons for this bias are under investigation. Varns et al.[50] also used the Ogawa samplers for
618 ozone monitoring and conducted duplicate analyses as well as an estimate of bias compared to
619 Dasibi model 1008 continuous monitors. Their duplicate data indicated that there was a median
620 absolute difference of 1.38ppb and a negative bias that ranged from 2.7 – 4.7 ppb over the four
621 locations used. Typical ozone concentrations were 20 – 70 ppb in Texas, which is similar to the
622 values found in Windsor. Another paper by Gibson et al.[51] showed their duplicate analyses had
623 an overall precision of 5.4% while comparisons at three locations with Thermo Electron
624 Instruments Inc. model 49C continuous ozone analysers had R^2 values ranging from 0.82-0.95.
625 The VOC Summa Canisters proved to be an acceptable method of obtaining 24-hr indoor and
626 outdoor residential measurements in both years of sampling. Personal VOC measurements were
627 restricted to the 2005 adult population as the combined weight of the canister along with the
628 personal pump and battery was too heavy for the children to carry. The total sample size for
629 VOC canisters was 1294 in 2005 and 872 in 2006, which was 93% and 94% of the intended
630 1395 and 930 samples, respectively, and represents one of the largest VOC datasets using this
631 method.
632 The Chempass $PM_{2.5}$ PEM compared well with the FRM dichotomous sampler over two years of
633 side-by-side measurements at the Allen Park MDEQ site in Detroit. The PEM had an overall
634 bias of +11% compared to the FRM sampler. A regression of the PEM on the FRM showed a
635 small intercept and an R^2 of 97%. The positive bias of 11% for the Windsor PEM $PM_{2.5}$ is an
636 improvement upon the 18% positive bias noted for the Marple $PEM_{2.5}$ in Ozkaynak et al.[15]
637 However, Liu et al.[52] after initially using oiled impactors and noting a large positive bias
638 averaging 7.7 $\mu g/m^3$, switched to greased impactors and reported a negligible positive bias of 0.4
639 $\mu g/m^3$. Williams et al.[53] reported that their PEM had a 10 - 20% higher mass concentration than
640 the FRM, probably as a result of retention of semi-volatile organic compounds (SVOC) by the
641 PEMs that are blown off by high filter face velocity samplers such as the dichotomous sampler.
642 Winter and summer comparisons for the current study found that the PEM was approximately
643 52% and 26% higher respectively than the TEOM. Differences between the PEM and the TEOM
644 have been previously attributed to evaporation of PM volatiles in the TEOM measurements.[45]
645 The TEOM reads lower than other filter-based methods due to its elevated inlet temperature,
646 which causes a proportion of the volatiles in the particulates to be vaporized on intake. This bias
647 will vary depending on the proportion of volatiles in the particulates.[54] During the winter, when

the temperature difference between ambient air and the filter is greater, there may be greater volatilization than during the summer.[45] The DustTrak and pDR $PM_{2.5}$ agreed well with the gravimetric PEM $PM_{2.5}$ method, (R^2 of 87% and 71%, respectively). These continuous data are important indicators of peak exposures which can be identified through the location and activity data available from the time activity diaries.

Time activity patterns are similar to those reported in the Canadian Human Activity Pattern Survey,[55] and those reported within the US[56] and other developed countries.[57] Some of the biggest seasonal differences were due to the children being in school during the winter sampling period compared to the summer when school was not taking place; the adult population did not have equivalent differences. Time spent outdoors was also influenced by season and both populations spent more time outdoors in summer (approximately 10% of their total day).

CONCLUSIONS

The majority of the WOEAS data can be used with confidence to examine the relationships between personal, indoor and outdoor concentrations for the range of pollutants listed. Predictors of these relationships can be determined using the questionnaires and time activity diary data which were reviewed by the technicians on a daily basis with the participants to ensure accuracy and compliance. The study can also be used to understand the impact that ambient air pollution has upon personal and indoor residential exposures. When the data are combined with the health effects data collected in 2006 it will be possible to investigate the effects of personal, indoor and outdoor air pollution exposures upon respiratory health. These WOEAS data can be used with confidence for developing risk management policies to reduce personal and indoor exposures to air pollutants.

ACKNOWLEDGEMENTS

The participants and their families are gratefully thanked for their contributions to these two years of data collection; without them none of this research would have been possible. The careful field work undertaken by the numerous field technicians from Health Canada and the University of Windsor is appreciated. The authors would like to acknowledge contributions from Dr. Paul Villeneuve (Health Canada) for the power calculation, Alice Grgicak-Mannion (GLIER, University of Windsor) for the map of residential locations, Mark Davey (University of Washington), Steve Ferguson and Dr. Mike Wolfson (Harvard School of Public Health) for their guidance on methods and laboratory analyses, Sandy Benetti of Environment Canada for her

help in the initial coordination of the laboratory activities and sample analyses, Don Fugler of Canadian Mortgage and Housing Corporation and Dr. Russell Dietz of Brookhaven National Laboratory for their support with air exchange rate measurements, Environment Canada and Michigan Department of Environmental Quality for permitting us to locate our instruments at the College Road and Allen Park sites and access to their data for the method comparisons, and Dr. Markey Johnson and Dr. Scott Weichenthal from Health Canada for conducting the internal review. Funding was provided by the Border Air Quality Strategy (BAQS) through Health Canada.

Although this work was reviewed by the US EPA and approved for publication, it may not necessarily reflect official Agency policy.

IMPLICATIONS

It is important to obtain data to identify any factors that can influence the relationships between personal, indoor and outdoor concentrations for a range of air pollutants. Ensuring that the methods employed are valid and comparable to reference methods used in typical air pollution monitoring is crucial for data to be of use to regulators. These exposure data can then be used for developing risk management policies to reduce personal and indoor exposures to air pollutants.

695 **Table 1.** Target variables and instrumentation

Variable	Instrument / Equipment	Principle	Frequency	Location
$PM_{2.5}$ and PM_{10} (mass)	Impactor	Gravimetric	Daily	Indoors, outdoors; personal ($PM_{2.5}$ 2005 personal only)
$PM_{2.5}$ associated nitrate	Impactor and denuder		Daily	Indoors, outdoors, personal
$PM_{2.5}$ (mass)	TEOM	Piezoelectric	Daily	Central site (NAPS)
$PM_{2.5}$ and coarse particles (PM_{10}-$PM_{2.5}$)	Dichotomous (virtual impactor)	Gravimetric	Daily	Outdoors only at Allen Park, Detroit
NO_2	Ogawa	Diffusion	Daily	Indoors, outdoors, personal
O_3	Ogawa	Diffusion	Daily (Summer only)	Personal, outdoors
Fine particles (>0.1 μm - <2.5 μm)	DustTrak	Optical	Every 3 minutes	Indoors, outdoors
	Personal DataRAM (pDR)	Optical	Every 3 minutes	Personal (2006 only)
Ultrafine particles (Number) (20 nm to about 1 μm)	P-Trak	Condensation particle counter (CPC)	Every 30 secs for 10 mins each hour	Indoors, outdoors
Elemental carbon	Aethalometer	Absorption at 880 nm	Every 3 minutes; alternating location every half hour during day & hourly at night	Indoors, outdoors
Air change rate	Perfluorocarbon tracer	Tracer gas collection	Daily	Indoors
Temperature / Relative humidity	Smart Reader Plus 2	Thermistor Thin film	Every 3 minutes	Indoors, outdoors
Household characteristics & personal activities	Questionnaire, diary		Once per household, daily for activities	N/A
Settled Dust	HVS3 Vacuum		Once after the completion of the air pollution measurements	Indoors

Lung function	PiKo-1 peak expiratory flow meter		Morning and evening pre-medication use	Personal (2006 only)
Exhaled Breath Condensate (EBC)	R Tube		Evening daily	Personal (2006 only)

Ancillary meteorological variables (wind speed and direction, atmospheric pressure, visibility, and weather conditions) were also obtained from Environment Canada and added to the dataset.

700 **Table 2.** Relative bias and precision of collocated samplers at NAPS.

Pollutant	Method	Descriptive Statistics						Bias			Precision		
		N	Mean	Std. Dev.	p25	Median	p75	p25	Median	p75	p25	Median	p75
NO_2 (ppb)	Ogawa	113	20	10	13	18	26	0.01	0.17	0.35	0.03	0.07	0.17
	EC	113	16	6	12	15	20						
O_3 (ppb)	Ogawa	76	26	9	21	25	34	-0.15	-0.24	-0.39	0.05	0.09	0.17
	EC	76	36	11	29	36	42						
Nitrate ($\mu g/m^3$)	Mini-PEM	56	1.9	1.6	1	1.5	2.1	-	-	-	0.03	0.07	0.13

701

702 **Table 3.** Comparison of gravimetric PEM to dichotomous reference sampler at Allen Park location: bias
703 and precision.

Pollutant ($\mu g/m^3$)	Method	Descriptive Statistics						Bias			Precision		
		N	Mean	Std.Dev.	p25	Median	p75	p25	Median	p75	p25	Median	p75
$PM_{2.5}$	Dichot	38	15.8	8.8	9.0	13	21	1.04	1.11	1.19	0.03	0.07	0.12
	PEM	38	17.6	10.3	9.8	16	24						
PM_{10}	Dichot	38	26.6	11.3	17	24	34	1.02	1.10	1.15	0.03	0.06	0.10
	PEM	38	29.0	12.6	20	27	38						
$PM_{2.5-10}$	Dichot	38	10.4	3.2	8.2	9	13	0.94	1.09	1.19	0.05	0.12	0.24
	PEM	38	10.9	3.5	8.9	11	13						

704

705 **Table 4.** Comparison of gravimetric $PM_{2.5}$ PEM with collocated TEOM at NAPS location: TEOM bias.

$PM_{2.5}$ ($\mu g/m^3$)	Descriptive Statistics						TEOM Bias		
	N	Mean	Std.Dev.	p25	Median	p75	p25	Median	p75
Winter									
PEM	39	12.0	6.6	6.5	9.8	16.6	-0.59	-0.51	-0.47
TEOM	39	6.0	3.8	2.8	5.0	7.4			
Summer									
PEM	77	17.9	10.0	10.1	15.2	22.9	-0.32	-0.26	-0.16
TEOM	77	13.7	8.7	6.9	12.0	18.8			

706

707 **Table 5.** Descriptive Statistics of the Study Population by Year.

Study Population		**2005**	**2006**
Gender (N)	Male	5	31
	Female	45	20
Ethnicity (N)	Caucasian (other)	N / A	48 (3)
Age (N)	10	N / A	20
	11		20
	12		10
	13		1
Baseline Housing Characteristics			
Home Type (N)	Detached	50	42
	Row House	0	4
	Duplex/triplex	0	2
	Other	0	1
Home Size (m^2)	Mean	207.5	172
Stove Type (N)	Gas / Electric	41 / 3	12 / 36
Air Cleaning Device on Furnace (N)	Yes (No)	14 (30)	34 (12)
Garage Type (N)	No Garage	14	21
	Detached	12	10
	Attached	23	16
	Attached no door	1	1
Daily Questionnaire Variables (No. of Sampling days)			
Season	Summer / Winter	229 / 239	240 / 240
Candles Used in the Home Today	Yes (No)	28 (420)	27 (438)
Windows Open Today	Yes (No)	134 (315)	123 (338)
Cooking	Yes (No)	324 (128)	365 (99)
Cleaning	Yes (No / Don't Know)	226 (236)	254 (206 / 2)
Presence of smokers	Yes (No)	3 (445)	1 (463)
Presence of pets	Yes (No)	310 (139)	260 (204)
Time Activity Data			
% of time spent indoors at home	Summer / Winter	74.6 / 76.5	77.4 / 68.7
% of time spent indoors away from home	Summer / Winter	3.8 / 0.5	9.8 / 5.8
% of time spent at work / school	Summer / Winter	5.6 / 5.4	0.6 / 18.5
% of time spent in the yard or nearby	Summer / Winter	8.7 / 10.4	4.8 / 0.7
% of time spent outdoors away from home	Summer / Winter	2.4 / 10.4	5.6 / 5.2
% of time spent in transit	Summer / Winter	4.8 / 6.3	3.4 / 3.7

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Figure 1. Map of participant locations and NAPS site.

Figure 2. Picture of equipment set up.

Figure 3a. Comparison of the PEM2.5 sampler with the Allen Park Dichotomous sampler.

Figure 3b. Comparison of the PEM10 sampler with the Allen Park Dichotomous sampler.

Figure 1. Map of participant locations and NAPS site.

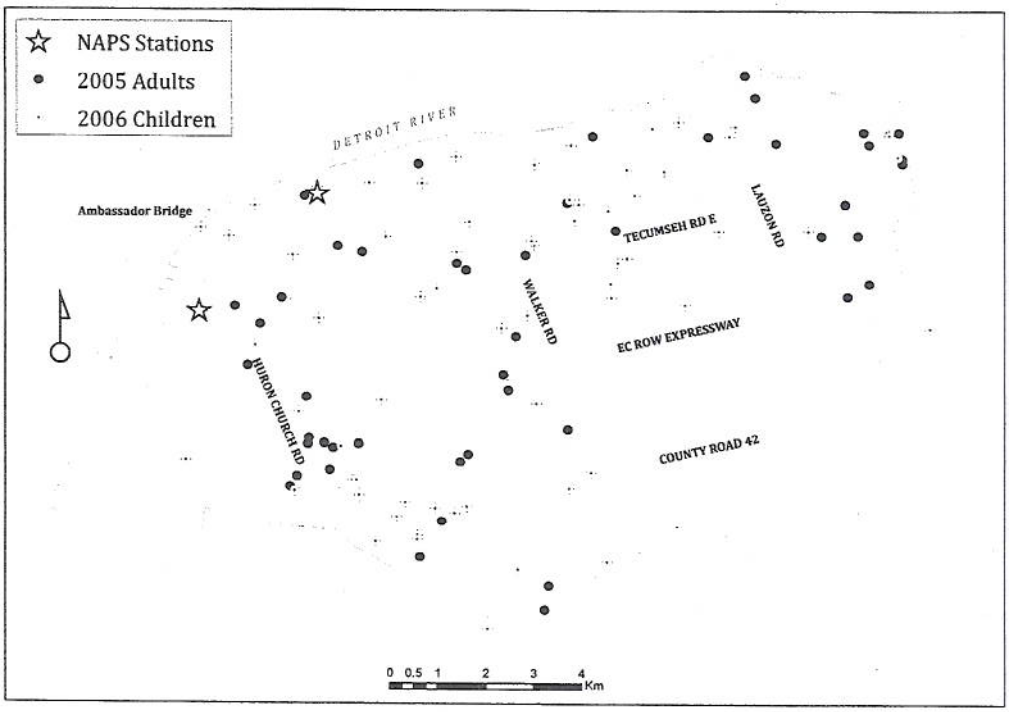
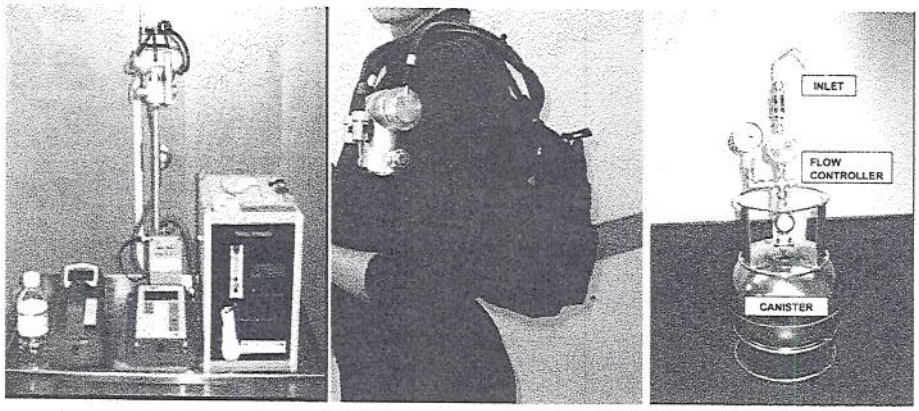


Figure 2. Picture of equipment set up



a) Indoor & outdoor monitors b) Personal set up c) VOC canister

Figure 3a. Comparison of the $PEM_{2.5}$ sampler with the Allen Park Dichotomous sampler.

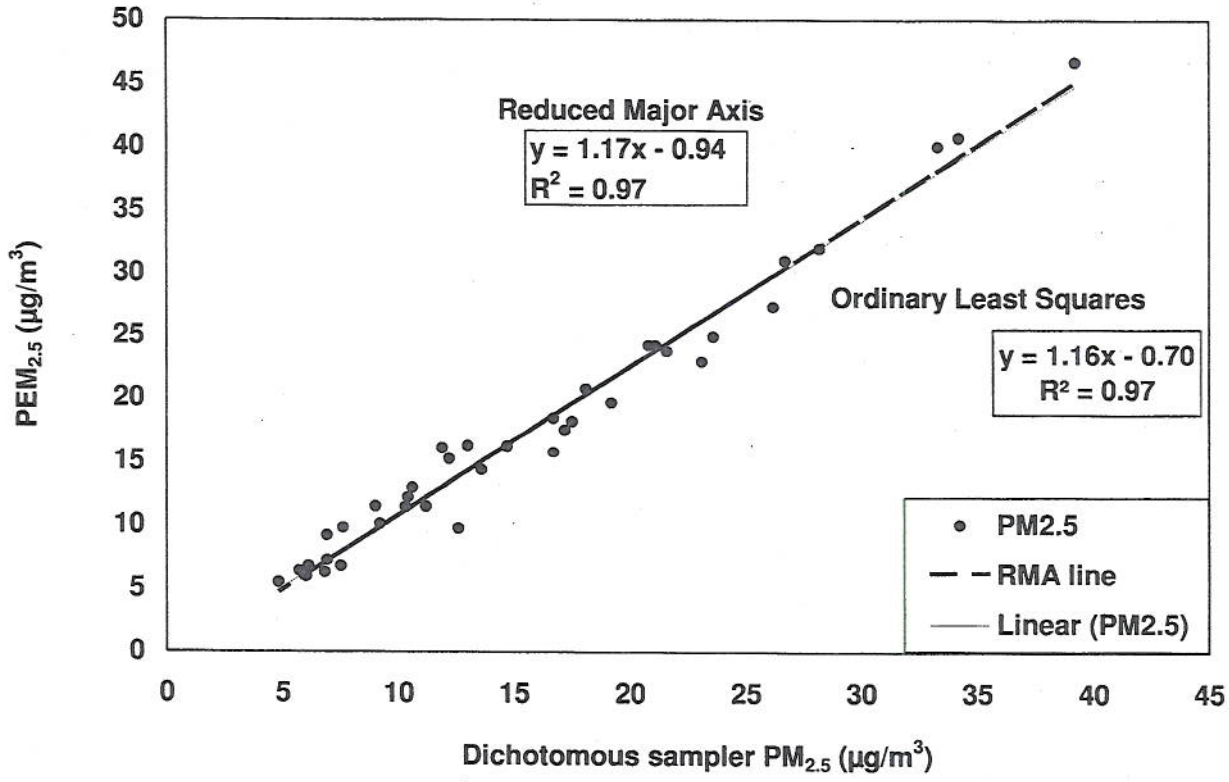


Figure 3b. Comparison of the PEM_{10} sampler with the Allen Park Dichotomous sampler.