WOODSMOKE SOURCE APPORTIONMENT AND HOME INFILTRATION STUDY IN THE RURAL ANNAPOLIS VALLEY, NOVA SCOTIA, CANADA.

Air & Waste Management Association's
103rd Annual Conference, Calgary, June 22, 2010.
Expert panel presentation and paper in the conference proceedings. Paper **#142**

Mark D. Gibson,[1] Tony J. Ward,[2] Amanda J. Wheeler,[3] Judy R. Guernsey,[1] Matt P. Seaboyer,[1] Patrick Bazinet,[1] Gavin H. King,[1] Neil K. Brewster,[1] James Kuchta,[1] Ryan Potter,[1] Dave M. Stieb[4]

[1]Atlantic RURAL Centre, Community Health and Epidemiology, 5790 University Avenue, Dalhousie University, Halifax, Nova Scotia, Canada, B3H 1V7.
[2]Center for Environmental Health Sciences, 32 Campus Drive Missoula, University of Montana, Missoula, Montana, USA. 59812
[3]Air Health Science Division, Health Canada, 269 Laurier Avenue West, Ottawa, Ontario, Canada. K1A OK9
[4]Healthy Environments and Consumer Safety Branch, Health Canada, 269 Laurier Avenue West, Ottawa, Ontario, Canada. K1A OK9

INTRODUCTION

Woodsmoke has recently received increasing attention as an important source of particulate matter, particularly in rural areas. [1-7] Indoor woodstoves and outdoor wood boilers are widely used in the Annapolis Valley, in rural Nova Scotia, and the area's topography is conducive to trapping emissions especially during thermal inversions (typically 2 to 5 per winter). In an effort to quantify the impact of these factors on local air quality, the source contribution of residential wood burning to ambient $PM_{2.5}$ at one fixed site on the Annapolis Valley floor was determined during the winters of 2008/2009 (phase 1) and again in 2009/10 (phase 2). Running concurrently with the $PM_{2.5}$ ambient monitoring in 2009/10 was an investigation of woodsmoke home infiltration into 30 homes, together with an evaluation of the effectiveness of high efficiency particle air (HEPA) cleaners at reducing indoor $PM_{2.5}$ concentrations (phase 2).

EXPERIMNTAL METHODS

Phase 1 monitoring (winter 2008/09)

The ambient monitoring site used to conduct the woodsmoke source apportionment in phase 1 was located 3 km South of the small town of Middleton on the Annapolis Valley floor (LAT 44° 54' 48.78" LONG -65° 03' 37.36", elevation 26.8 m). The ambient monitoring equipment used during phase 1 included: 4x SKC leyland legacy deployable particle samplers (DPS) used for 24-hr sampling of $PM_{2.5}$, at a flow rate of 10 l/min. 1x DPS contain a teflon filter, 2x DPS contained a quartz filter in each and the 4th DPS housed a nylon filter. 2x Dust Trak (TSI, Shoreview, MN) nephelometers used to measure PM, one measuring $PM_{2.5}$ and one measuring PM_{10}. The Dust Trak's were housed in a heated environmental enclosure. The difference between the two Dust Trak instruments providing an estimate of the course PM fraction (2.5 – 10 μm). Volatile organic compounds (VOC) were sampled passively over 24-hrs and 7-days using three different types of Perkin-Elmer thermal desorption tubes (TDT) that included Tenax TA, Unicarb and Carbotrap tubes. These three TDT were deployed simultaneously to capture the entire range of VOCs from low boilers, e.g. vinyl chloride, to high boilers, e.g. naphthalene. The VOC TDT samples were analyzed using a Perkin-Elmer TurboMatrix thermal desorption system coupled to a Perkin-Elmer Clarus 500 GC equipped with a Restek Rtx-1 60 m, 0.53 mm ID, 3 μm df capillary column with ECD and FID detectors. 24-hr and 7-day nitrogen dioxide (NO_2) and ground-level ozone (O_3) samples were collected using Ogawa passive samplers. The ambient site during phase 1 also contained a Thermo, model 49, auto-analyzer that provided 15 min time integrated O_3 measurements. Weather parameters (wind, T, RH, rain, UV, SR, mb) were measured using a Davis Vantage Pro II weather station. All samples were transported by on ice and returned the same way, with the exception of the VOC TDT which were sealed with brass swage lock end caps, wrapped in tin foil and then placed in a Ziploc bag. Each collected $PM_{2.5}$ speciation filter from phase 1 was analyzed for target analytes that included 40 metals (XRF & ICP-MS), anions and cations (IC), organic and elemental carbon (DRI thermo-optical method). Briefly, levoglucosan, and other woodsmoke markers, were extracted using ethyl acetate and trimethylamine, spiked with 4 deuterated woodsmoke makers, followed by derivitization using N-O-bis(trimethylsilyl)trifluoroacetamide (BSTFA), trimethylchlorosilane (TMCS), and trimethylsilylimidazole (TMSI). After

sample prepartion the woodsmoke markers were analyzed using an Agilent 6890N GC equipped with a HP-5MS, 30m x 0.25 mm ID, 0.25 μm df capillary column with an Agilent 5973 MS detector. Quartz filter $PM_{2.5}$ specimens were sent to University of Arizona's Accelerator Mass Spectrometry Laboratory facility for ^{14}C analysis.

The $PM_{2.5}$ chemical species were used in the US Environmental Protection Agency (EPA) chemical mass balance model (CMB) v8.2 model to apportion the sources of $PM_{2.5}$ measured at the ambient monitoring site in phase 1. ^{14}C was used to determine the old carbon (fossil fuel) versus new carbon (woodsmoke) contributions to the $PM_{2.5}$ at the ambient monitoring site, as well as further validate the results of the CMB modeling. CMB v8.2 compares $PM_{2.5}$ sample chemical species with source profile chemical species. The CMB model attributes the contribution of the identified sources to the total $PM_{2.5}$ sampled at a given receptor, in our case the ambient monitoring site at Middleton. Source profiles obtained from the USEPA Speciate v4.2 database were used in the CMB model. Speciate v4.2 source profiles from many different wood burning appliances and types of wood fuel were used in the CMB model, e.g. woodstoves, outdoor woodboilers, hard woods and soft woods. We also collected our own source profile from an outside woodboiler with plans to collect street sand and a vehicle profile in the Annapolis Valley for use in the CMB model.

The censoring algorithms, multiple linear regression and recursive mass balance models used by Barn et al 2008 will be applied to the indoor/outdoor continuous $PM_{2.5}$ data to determine the infiltration rate and effectiveness of the HEPA filters.[7]

Phase 2 monitoring (winter 2009/10)

For phase 2 monitoring in the winter of 2009/10, the ambient monitoring site was improved considerably and was equipped with a Thermo 2025 Federal Reference Monitor (FRM) used to collect $PM_{2.5}$ mass/elements. A Thermo 2300 fitted with four ChemComb cartridges (including a sodium carbonate denuder to removed SO_2 in the nylon filter air stream) was also located at the ambient monitoring site. The ChemComb's, housing 47 mm diameter filters, included 1x Quartz (16.7 l/min), 1x

Quartz (10 l/min), 1x nylon (10 l/min) and 1x teflon (16.7 l/min). As in phase 1, these filters, provide further samples for CMB analysis, ^{14}C and woodsmoke marker analysis. 90x 24-hr, 5x simultaneous $PM_{2.5}$ filters specimens were collected in phase 2. In phase 2 the weather metrics (wind, T, RH) at the ambient site near Middleton were obtained from the Partisol 2025 onboard meteorological sensors. The ambient monitoring coincided with monitoring of $PM_{2.5}$ infiltration and HEPA filter evaluation in 30 homes in the Annapolis Valley.

The indoor monitoring equipment used in phase 2 included a Dust Trak (1-min average $PM_{2.5}$), VOC (passive TDT and active TDT *via* FLEC air pumps and triple bed VOC tubes), NO_2 (Ogawa passive sampler), $PM_{2.5}$ mass/elements (Harvard Impactor containing a Teflon filter), $PM_{2.5}$ chemical species (ChemComb containing a quartz filter [wood smoke chemical markers]), CO_2, RH and temperature (YES monitor), CO (Langan T15n monitor). Air exchange rate was measured using a perfluorocarbon tracer emitted (PFT) with a corresponding capillary adsorption tubes (CAT). 3M Filtrete HEPA filters were used and their placement in the home noted. 2g of house dust was also collected from each home to determine the presence an abundance of allergens, e.g. dust mites, endotoxin, mold, cat and cockroach.

The outdoor air monitoring equipment included a Dust Trak, VOCs, NO_2, $PM_{2.5}$ mass/elements, $PM_{2.5}$ woodsmoke markers using the identical equipment as for indoor sampling. Non-continuous samplers inside (including the CAT) and outside the home were swapped out every 24-hr. A Davis II weather station (wind, T, RH, SR, UV, mb) was deployed outside each home. Blanks and duplicates accompany 10% of passive and filter based samples taken both indoor and outdoor homes and at the ambient monitoring site.

Sampling in each home lasted for 3 days. On the first day the participants used their internal woodstove and or woodfurnace as normal but with the HEPA filter switched off. This allowed us to gain a baseline of indoor air quality impacted from both indoor and outdoor sources. On the second day the participant did not use their indoor

woodstove/woodfurnace and the HEPA was randomly assigned to be switched on/off. An outdoor woodboiler (where present) was allowed to remain in operation over the 3-days. On the third day the participants still did not use their woodstove/woodfurnace and the HEPA was in the opposite mode to the previous day. When the HEPA was off, we were able to determine $PM_{2.5}$ infiltration into the home. When the HEPA was switched on, we were able to evaluate the HEPA's effectiveness at reducing indoor $PM_{2.5}$.

Results and Discussion

The acquisition of the 50 days of 4x simultaneously sampled $PM_{2.5}$ filter specimens and associated gases was successfully collected in the winter of 2008/09 with a further 90 sets of 5 $PM_{2.5}$ filter specimens and associated gases from the Middleton ambient site for the winter of 2009/10. 24-hr averaged Dust Trak $PM_{2.5}$ data showed that there were no exceedances of the 24-hr, 30 $\mu g\ m^{-3}$ (average of the 99th percentile over three years), Canada Wide Standard for $PM_{2.5}$ in the winter of 2008/09 but at least one exceedance during the winter of 2009/10 (currently being investigated). A number of short-term excursions over 100 $\mu g\ m^{-3}$ were observed from the real-time $PM_{2.5}$ data in both winters. It was seen that there was no significant difference ($P = 0.524$) between the Dust Trak $PM_{2.5}$ and Dust Trak PM_{10} data suggesting that the PM_{10} size fraction is dominated by the fine PM mode.

The $PM_{2.5}$ chemical composition data will hopefully confirm the sources of the $PM_{2.5}$ found at this receptor. CMB conducted to date on 16x 24-hr $PM_{2.5}$ samples has yielded an average woodsmoke source contribution to $PM_{2.5}$ of 56.2% (range 32.6% - 73%), with 12.4 % NH_4NO_3 (range 5.0% - 20%), 24.1% SO_4 (range 11.0% - 47%), 9.2% $(NH_4)_2SO_4$ (range 2% - 19.8%), 3.9% unknown mass (range 0.6% - 14%) with no statistically significant input from vehicle emissions. The 16 days chosen from phase 1 were due to the samples having sufficient mass to yield complete chemical species data with which to run the CMB model. The remaining 34 days did not have sufficient mass to run the model. Further CMB will be conducted on the $PM_{2.5}$ filter specimens obtained from the 2009/10 campaign.

The mean, min and max levoglucosan from phase 1 to date is 234.2, 154.9 and 274.3 ng/m^3 respectively.

It was observed from phase 1 (winter 2008/09) that OC and EC show a strong association (R^2 = 0.61) implying they share the same source. It was also observed that the OC and EC are also strongly associated with $PM_{2.5}$ (R^2 0.54 and 0.52 respectively) suggesting that the $PM_{2.5}$ share the same source as OC and EC.

In phase 2, the preliminary $PM_{2.5}$ woodsmoke home infiltration into the 22 homes sampled thus far (target 30) demonstrated infiltration of ambient $PM_{2.5}$ into the homes. The mean, min and maximum $PM_{2.5}$ found in the 22 homes sampled when the indoor woodstove was in use (HEPA switched off) were 17.9, 1.0 and 1917 μg m^{-3}. For sampling when the HEPA filter was switched on (indoor woodstove off) the mean, min and maximum $PM_{2.5}$ observed was 5.0, 0.0 and 641 μg m^{-3}. For sampling when the HEPA filter was switched off (indoor woodstove off) the mean, min and maximum $PM_{2.5}$ observed was 15.7, 0.0 and 3578 μg m^{-3}. This preliminary data shows significantly ($p<0.05$) lower $PM_{2.5}$ concentration when the HEPA filter was in use compared to when not in use. Censoring algorithms, multiple linear regression and recursive mass balance models will be applied to the indoor/outdoor continuous $PM_{2.5}$ data to determine the effectiveness of the HEPA filters and will be presented. The mean, min and max CO concentrations found in the 22 homes over the entire sampling period was 1.3, 0.03 and 11.8 ppm respectively.

SUMMARY

The mean woodsmoke contribution to ambient $PM_{2.5}$ in the winter 2008/09 was found to be 56.2% (range 32.6% - 73%). Infiltration of ambient $PM_{2.5}$ was clearly observed in the 22 homes sampled to date. This preliminary data shows significantly ($p<0.05$) lower $PM_{2.5}$ concentration when the HEPA filter was in use compared to when not in use providing tentative evidence that they are indeed effective at reducing indoor $PM_{2.5}$. Further analysis is required to calculate the actual infiltration rate in each of home and also to produce an accurate assessment of the effectiveness of the HEPA filters ability to

reduce indoor $PM_{2.5}$. This study provides new insights into the woodsmoke contribution to ambient $PM_{2.5}$ in a Rural, constrained Valley, impacted by strong sources of woodsmoke compounded by occasional wintertime thermal inversions. Additionally, this study also provides new information on the infiltration of ambient woodsmoke into homes and the effectiveness of cheap, portable HEPA filters at reducing indoor $PM_{2.5}$. This study provides valuable new data related to woodsmoke exposure in rural environments.

ACKOWLDEGEMENTS

The authors would like to thank the Clean Annapolis River Project (CARP) and the Annapolis Valley Community Health Board for their significant efforts in helping to recruit homes for this study. Many thanks to Richard Bridge for generously allowing the use of his land to establish the ambient monitoring site near Middleton, Nova Scotia. Finally, many thanks to Environment Canada for their continued support and for providing air pollution data from Aylesford Mountain and the Kentville National Air Pollution Surveillance (NAPS) stations.

REFERENCES

1. Moore, D.; Copes, R.; Fisk, R.; Joy, R.; Chan, K.; Brauer, M., Population health effects of air quality changes due to forest fires in British Columbia in 2003: estimates from physician-visit billing data. *Canadian Journal of Public Health* **2006,** *97*, 105-108.
2. Ward, T. J., The Missoula, Montana $PM_{2.5}$ Source Apportionment Research Study. *The University of Montana Report – Missoula, Center for Environmental Health Sciences* **2007**.
3. Ward, T. J.; Hamilton, J., Raymond F.; Smith, G. C., The Missoula, Montana $PM_{2.5}$ speciation study--seasonal average concentrations. *Atmospheric Environment* **2004,** *38* (37), 6371-6379.
4. Ward, T. J.; Rinehart, L. R.; Lange, T., The 2003/2004 Libby, Montana $PM_{2.5}$ Source Apportionment Research Study. *Aerosol Science and Technology* **2006,** *40*, 166-177.
5. Naeher, L. P.; Brauer, M.; Lipsett, M.; Zelikoff, J. T.; Simpson, C. D.; Koenig, J. Q.; Smith, K. R., Woodsmoke Health Effects: A Review. *Inhalation Toxicology* **2007,** *19* (1), 67-106.
6. Su, J. G.; Buzzelli, M.; Brauer, M.; Gould, T.; Larson, T. V., Modeling spatial variability of airborne levoglucosan in Seattle, Washington. *Atmospheric Environment* **2008,** *42* (22), 5519-5525.

7. Barn, P.; Larson, T.; Noullett, M.; Kennedy, S.; Copes, R.; Brauer, M., Infiltration of forest fire and residential wood smoke: an evaluation of air cleaner effectiveness. *Journal of Exposure Science and Environmental Epidemiology* **2008,** *18* (DOI 1559-0631/08), 503-511.