The daily and seasonal variability in residential concentrations of aldehydes in two Canadian cities

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SUMMARY

Indoor air concentrations of formaldehyde, acetaldehyde and acrolein were measured in Halifax, Nova Scotia and Edmonton, Alberta. Seven consecutive 24-hr measurements were made in 50 non-smoking homes in winter and summer in each city. In addition, data on relative humidity, temperature, air exchange rates, housing characteristics and occupants' activities were collected. Determinants of indoor levels for formaldehyde and acetaldehyde were examined using linear mixed-effects regression models. The housing characteristics and occupants' activities investigated in this study explained between 26% and 50% of the variability in pollutant concentrations, with air exchange rates, year of construction, indoor temperature and indoor relative humidity being important predictors for both formaldehyde and acetaldehyde.

IMPLICATIONS

Aldehydes are known irritants of the eye and upper airways, especially the nasal cavity, and formaldehyde is suspected of causing allergic sensitization. Several organizations are developing residential indoor air quality guidelines for formaldehyde, and there is an interest in getting recent information on residential concentrations and their contributing sources.

KEYWORDS

Indoor air exposure, field studies, sources, formaldehyde, ventilation

INTRODUCTION

Aldehydes are highly reactive compounds released from several sources into the indoor environment, including off-gassing from building materials, paints and varnishes, and combustion appliances. Because of the many indoor sources, aldehyde concentrations are typically higher in indoor air than in ambient air.

The current studies in Halifax and Edmonton were undertaken by Health Canada, in collaboration with Dalhousie University and the University of Alberta, as part of a series of indoor air exposure studies that have been conducted across Canada. The main objectives were to determine the concentrations of various air pollutants in a sample of Canadian homes, and investigate the associations between these pollutants and the housing characteristics and activities of occupants.

METHODS

Indoor concentrations of the three aldehydes (formaldehyde, acetaldehyde and acrolein) were measured for seven consecutive 24-hr periods in 50 non-smoking homes in winter and summer in both Halifax, Nova Scotia and Edmonton, Alberta. In addition, data on relative humidity, temperature, air exchange rates, housing characteristics and occupants' activities were collected.

For both cities, the residences were selected based on the *a priori* hypotheses that the age of the home, as well as types of heating and cooking appliances, would impact indoor air quality. Sampling was therefore stratified by age of home, and the residences were grouped into five subsets of construction year (1945 and before, 1946–1960, 1961–1980, 1981–2000, and 2001 and after) and presence or absence of gas stoves.

Measurements were usually taken at breathing height (1.5 m) within the family or living room where participants spent a significant amount of time. Air pollution measurements were completed in the winter (January to March/April) and summer (June to September) seasons in 2009 (Halifax) and 2010 (Edmonton). Nine seven-day sampling periods were conducted per season per city, with six homes being measured concurrently per sampling period. Participants were asked to continue with their normal activities throughout the sampling period in order to ensure that the concentrations measured were representative of typical indoor exposures.

Two different types of questionnaires were completed in order to identify possible sources of indoor exposure and factors that may influence concentrations. Participants were interviewed by a field technician to obtain information about housing characteristics that would not change over the course of a season of sampling, including: age and type of home, heating and cooking systems, recent addition of furniture, recent painting or varnishing, storage of paints and chemicals and presence of an attached garage. A daily questionnaire was also completed by the study participants for every corresponding 24-hr air measurement. The questionnaire responses described activities in the home during sampling, such as: cleaning and cooking, use of personal care products and chemicals, opening and closing of windows, and car idling.

Concentrations of aldehydes were measured using UMEX 100 Passive Samplers (SKC Inc. Eighty Four, PA, USA), with 2,4-dinitrophenylhydrazine (DNPH) as the reagent. The samples were analyzed by high performance liquid chromatography (HPLC) using US EPA Compendium Method TO-11A.

Home air exchange rates (AER) were determined for the corresponding 24-hr periods by the perfluorocarbon tracer (PFT) technique (Dietz and Cote, 1982). The samples were taken on the same floor that the indoor air measurements were made. After exposure, the capillary absorption tubes (CATs) were shipped to the laboratory and analyzed by gas chromatography with electron capture detection (GC/ECD). For each home, the air exchange rate, expressed as air changes per hour (h^{-1}), was calculated by dividing the infiltration rate by the measured house volume.

Temperature and relative humidity were recorded continuously for the full 7-day sampling period at each house at 10 minute intervals using a YES-206 Falcon data logger (Young Environmental).

Ten percent field blanks and duplicate samples were also collected for quality assurance purposes. The minimum reporting limit (MRL) of the method was 40 ng per sample, corresponding to a concentration of 1.0 $\mu g/m^3$. Blank corrections were applied when more than 50% of the field blanks were greater than the MRL. The field detection limit (FDL) was defined as three times the standard deviation of the field blanks. Concentrations were then blank-corrected using the field blank median values. Any resulting values that were lower than the MRL were substituted with half of the MRL. Samples that were above the MRL but below the FDL were unchanged. Samples were deemed invalid if they were deployed for more or less than 20% of the target time of 24-hr.

Statistical analyses were performed using SAS software version 9.2 (SAS Institute, Cary, NC, USA). Seasonal differences in pollution levels were examined using t-tests. Within home differences were examined by calculating the coefficient of variation (CV) for each 7-day measurement session. Determinants of indoor pollution levels were examined using linear mixed-effects regression models (PROC MIXED in SAS) using a first order autoregressive covariance structure. Pollutant levels were right-skewed and therefore log-transformed for analysis (including seasonal t-tests). Housing characteristics that were hypothesized to influence pollution levels were entered as fixed effects and house ID was fitted as a random intercept term. Season was entered as a group effect to allow for different parameters in the two seasons. Variables for the final models were chosen using a p-value cutoff of 0.05 and were based on *a priori* expectations gleaned from the literature.

RESULTS

After blank correction, 0% of the formaldehyde samples, and between 0% (Edmonton winter) and 8.5% (Halifax winter) of the acetaldehyde samples were below the MRL, respectively. For acrolein, between 77% (Edmonton winter) and 92% (Halifax winter) of the samples were below the MRL; therefore, descriptive statistics are not presented and predictive models have not been developed for acrolein.

Descriptive statistics for formaldehyde and acetaldehyde for both cities in summer and winter are presented in Table 1. Due to operational difficulties in the field, not all homes had valid results for the winter sampling in both cities. Results show statistically higher formaldehyde concentrations in summer than in winter for both cities (Halifax: p=0.03; Edmonton: p=0.001). This seasonal trend was not observed for acetaldehyde. Acetaldehyde showed higher variation within homes than formaldehyde, and both pollutants showed more variation in summer than in the winter.

Table 1. Descriptive statistics for formaldehyde and acetaldehyde ($\mu g/m^3$) in both cities

Season	City	# homes	Mean	S.D.	Min	Max	Geo Mean	Geo S.D.	Mean CV*
	Formaldehyde								
Summer	Edmonton	48	31.42	21.36	1.41	109.76	24.95	2.03	24%
	Halifax	50	33.38	30.24	5.18	234.08	25.50	2.03	27%
Winter	Edmonton	32	23.54	11.84	1.68	60.56	20.68	1.70	12%
	Halifax	33	29.03	24.34	2.01	141.27	22.36	2.04	15%
	Acetaldehyde								
Summer	Edmonton	48	10.68	7.26	0.43	39.68	8.34	2.15	39%
	Halifax	50	8.12	14.52	1.04	255.43	5.95	2.04	44%
Winter	Edmonton	32	9.63	4.94	0.49	31.07	8.40	1.75	27%
	Halifax	33	8.63	8.30	0.48	57.33	5.66	2.80	40%

* Mean within-home coefficient of variation (%)

Air exchange rates (mean ± S.D.) in both cities were higher in the summer than in the winter (T-test p<0.0001 in both cases): summertime AER averages were 0.51 ± 0.51 h^{-1} in Edmonton and 0.65 ± 0.60 h^{-1} in Halifax, while wintertime averages were 0.33 ± 0.27 h^{-1} in Edmonton and 0.33 ± 0.16 h^{-1} in Halifax. As expected, indoor temperature and relative humidity (mean ± S.D.) were also higher in summer (T-test p<0.0001 for all seasonal comparisons): 25 ± 3ºC with 37 ± 5% humidity (RH) in Edmonton, and 24 ± 2ºC with 46 ± 5% RH in Halifax, compared with wintertime measurements of 21 ± 1ºC and 32 ± 4% RH in Edmonton, and 20 ± 2ºC and 31 ± 5% in Halifax.

Table 2 presents the results for mixed-effects regression models. The percent change per unit (% change/unit) indicates the modelled change in pollutant level with a one unit change in the independent variable. In the case of dichotomous variables, the % change reflects the change in pollutant level associated with the presence *vs*. the absence of the factor. Models predicted an estimated 41% and 50% of the variability in indoor levels for formaldehyde, and 26% and 28% in indoor levels for acetaldehyde, for Edmonton and Halifax respectively. Air exchange was a significant predictor in all models, with higher air exchange rates decreasing pollutant levels. Higher indoor temperature and relative humidity were generally associated with increased aldehydes levels. Another important predictor was the year of construction of the home, with newer homes having higher concentrations. Additional predictors included storage of paints and solvents in the garage, which increased formaldehyde levels (Halifax only), and the use of the oven, which increased acetaldehyde levels in both Halifax and Edmonton.

Table 2. Results for fixed effects of mixed-effects regression models

Edmonton				Halifax			
Independent variable	Coefficient	P-value	% change /unit	Independent variable	Coefficient	P-value	% change /unit
Formaldehyde							
Intercept	-11.1	0.003		Intercept	-16.6	0.001	
Air exchange (h^{-1})	-0.53	<0.001	-41%	Air exchange (h^{-1})	-0.51	<0.001	-40%
Indoor temperature (ºC)	0.05	<0.001	5%	Indoor temperature (ºC)	0.10	<0.001	11%
Indoor relative humidity (%)	0.01	0.004	1%	Indoor relative humidity (%)	0.01	<0.001	1%
Year home built	0.01	<0.001	1%	Year home built	0.01	<0.001	1%
				Paints/solvents stored in garage	0.34	0.008	41%
Acetaldehyde							
Intercept	1.62	<0.001		Intercept	-15.1	0.001	
Air exchange (h^{-1})	-0.84	<0.001	-57%	Air exchange (h^{-1})	-0.52	<0.001	-40%
Indoor temperature (ºC)	0.04	0.003	4%	Indoor temperature (ºC)	0.07	<0.001	7%
Oven used on monitoring day	0.15	0.002	16%	Indoor relative humidity (%)	0.02	0.014	2%
				Year home built	0.01	0.001	1%
				Oven used on monitoring day	0.189	<0.0001	21%

DISCUSSION

Formaldehyde levels measured in these two cities are within the range of levels measured in other cities, where average levels ranged from 20.9 to 39.0 μg/m^3 (Bell et al., 1994; Dingle and Franklin, 2002; Gilbert et al., 2005, 2006; Héroux et al., 2010; Hun et al., 2010; Marchand et al., 2006). However, acetaldehyde levels are comparable to levels recently measured in Regina, Saskatchewan, Canada (Héroux et al., 2010), but lower than what was measured in other studies referenced in the literature (Stocco et al., 2008; Bell et al., 1994; Gilbert et al., 2005; Marchand et al., 2006), where average concentrations ranged from 18.1 to 39.6 μg/m^3. This could be explained in part by the differences in study design, or sampling methodologies that were used for which method performances are not necessarily comparable.

Ten percent of homes in summer in both cities and 3% (Edmonton) and 9% (Halifax) of homes in winter had an average formaldehyde level above Health Canada's long-term exposure guideline of 50 μg/m^3 (Health Canada, 2006). This is consistent with what was found in Regina, Canada, where 16% and 3% of the homes were above that same guideline value in summer and winter seasons, respectively (Héroux et al., 2010). However, this is considerably lower than what was reported by Gilbert et al. (2006), where 16% of the homes in winter in Quebec City, Canada, had concentrations above the guideline value. Summer measurements were not conducted as part of the Quebec City study, and therefore no comparison can be made with our results. Higher mean formaldehyde concentrations (mean ± S.D. = 32.7 ± 15.3 μg/m^3) and lower air exchange rates (mean ± S.D. = 0.2 ± 0.1 h^{-1}), due to differences in building characteristics such as the high prevalence of electric heating in Quebec City compared to Regina, Halifax, and Edmonton may explain these differences.

Both formaldehyde and acetaldehyde were significantly associated with air exchange rates in our models. Since indoor concentrations are usually higher than outdoor concentrations, due to the many possible indoor sources, an increase in air exchange can therefore significantly lower concentrations. Common indoor sources of aldehydes include building materials such as wood-based products, flooring materials, insulation, and paints and coatings (Salthammer et al., 2010; Loh et al., 2006) that off-gas at a greater rate when they are newly installed or recently applied. The year of construction of the home, as a general proxy for newer building materials and products, was consequently associated with higher concentrations of formaldehyde and acetaldehyde (Halifax only). In both cities, there were about ten homes per season that were built after 2001. Formaldehyde was also associated with the storage of paints and solvents in the garage in our Halifax sample. The number of homes where paints and solvents were stored in the garage was much higher in Halifax (32%) than in Edmonton (6%), which may explain why the association is only significant for the Halifax dataset.

Acetaldehyde was strongly associated with the use of the oven during sampling. Thirty-seven percent of sampling days (196 days out of 532) in Halifax, and 30% of sampling days in Edmonton (155 days out of 513) had participants report the use of the oven during sampling. Acetaldehyde has been associated in the literature with cooking activities involving oil (Fullana et al., 2004) and meat charbroiling/grilling (Schauer et al., 1999). It is possible that these cooking activities occurred in the homes during sampling and could explain the increased acetaldehyde emissions.

CONCLUSIONS

Indoor concentrations of aldehydes were measured as part of two large indoor air quality studies in Halifax and Edmonton, Canada. The characteristics of houses and occupants' activities investigated in this study explained 41-50% and 26-28% of the variability in indoor concentrations of formaldehyde and acetaldehyde, respectively. Air exchange, the year of construction of the home, indoor temperature and indoor relative humidity were important predictors for formaldehyde and acetaldehyde levels measured in this study.

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