

Selected Volatile Organic Compounds in Residential Air in the City of Ottawa, Canada

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Airborne levels of selected volatile organic chemicals (VOCs) that are priorities for exposure assessment under the Canadian Environmental Protection Act (CEPA) 1999 were measured in both indoor air and outdoor air of 75 residential houses, in the city of Ottawa, Canada, during the winter of 2002/2003. The houses were randomly selected using Ottawa 2001 population census data. VOCs were collected on adsorbent tubes and measured by thermal desorption GC/MS. Among 37 chemicals monitored, 17 were detected with a frequency greater than 80% in indoor air; 9 were between 30% and 80%; 7 were between 1% and 30%; and 4 were not detected. Concentrations of VOCs in both indoor and outdoor air are presented. Virtually all of the target VOCs were detected more frequently and were present at significantly higher levels, in indoor air than in outdoor air. As an indication of the contribution of indoor levels of these chemicals, ratios of the concentration found in indoor air to outdoor air (I/O) and the indoor source strength expressed in estimated emission rate per house are also presented. Compared with earlier published studies including a 1991/1992 Canadian national survey of VOCs in residential air, levels of target analytes in indoor air in this study were lower for a number of chemicals, indicating a possible trend toward decreased inhalation exposure to these chemicals in residential environments. This study has yielded up-to-date information on levels of a variety of priority airborne chemicals in residential air, which is being used to estimate current exposure to these substances as input to health risk assessments and risk management actions under CEPA 1999.

Introduction

The assessment of public exposure to harmful substances is an integral part of both health risk assessment and risk management in Canada. The Canadian Environmental Protection Act (CEPA), which was first established in 1988 and renewed in 1999, requires the Government of Canada to conduct risk assessments for substances that may be present in the environment and cause adverse effects to the environment or to human health in Canada (1). Among the substances identified in the first and second Priority Substances Lists (PSL1 and PSL2, respectively) under the CEPA

1988, in-depth risk assessments were conducted for almost 70 discrete chemicals, chemical classes, effluents, and emissions (2). The renewed CEPA 1999 also requires the Government of Canada to conduct environmental and health screening assessments for those substances on the Domestic Substances List (DSL) of almost 23 000 substances that are persistent, bioaccumulative, and inherently toxic or that present the greatest potential for exposure to Canadians (3).

For many of the chemicals being assessed or managed under CEPA, there are little or no data available on the levels of these substances, particularly in indoor environments where Canadians typically spend most of their time. Most indoor air studies in the past two decades have been focused on evaluating indoor air quality by measuring dominant volatile organic compounds (VOCs) found in indoor air (4–6) or chemicals that are related to the use of construction materials (7). Although some of the CEPA chemicals were already measured in these studies, many were not included.

Chemical-specific indoor air studies for risk assessment and risk management framework can provide monitoring data on the chemicals that are otherwise not the focus of more general indoor air quality studies. For example, a study designed to measure aniline in indoor air has provided critical exposure data to aid the risk assessment of this chemical (8). An early study conducted in 1991/1992 in which selected VOCs were measured in a national survey of over 700 randomly selected Canadian homes has provided information on baseline levels for several VOCs that were on PSL1 and PSL2 (9, 10). To provide updated inhalation data for use in the exposure assessment for certain PSL chemicals and for some of those DSL chemicals that are currently undergoing health screening assessments, levels of 37 VOCs have recently been measured in both indoor and outdoor air of 75 randomly selected residential houses in the city of Ottawa, and the results are presented in this paper. Statistical analyses relating the levels of measured VOCs to questionnaire information on sources of these chemicals will be presented elsewhere.

Methods

Study Design and Home Selection. To ensure that a random and representative sample was chosen, a two-stage stratified sampling process was used to randomly select (i) dissemination areas (DAs) and (ii) dwellings (houses and apartments) to be representative of Ottawa residences. In the first stage, the city was divided into three geographical areas based on urbanicity (urban core, urban fringe, rural fringe), using data from the 2001 Census (11). To determine the number of DAs to choose in each area, a stratified random sample selection was carried out. Historical data from nonsmoking homes in the Windsor Air Quality Study (12) were used to give an estimate of variance. The concentration range of acetaldehyde (1.7–61.9 $\mu\text{g m}^{-3}$), which was the largest among all analytes in the Windsor study was used as maximum anticipated concentration range for this study to guide the determination of the number of dwellings required for this study. The mean value for acetaldehyde was 21.7 $\mu\text{g m}^{-3}$, with a standard deviation of 16.0 $\mu\text{g m}^{-3}$ (24 observations). On the basis of the Neymann allocation (13) and historical data, it was estimated that 31 (of 1024) DAs were needed in the urban core, 4 (of 124) in the urban fringe, and 3 (of 79) in the rural fringe (approximately 3–4% in each geographical area) in order to obtain satisfactory precision in the overall estimate of the mean level of VOCs for the city. Satisfactory precision indicates that the bound on the 95% confidence interval (CI) for the mean is approximately 7 $\mu\text{g m}^{-3}$. A simple random

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sample was then carried out to choose the DAs from each geographical area.

In the second stage, the number and type of dwellings (houses and apartments) was recorded by driving through the selected DAs, using 2001 Census maps (purchased from Statistics Canada) for DA boundaries. It was estimated that a total of 120 dwellings (approximately 90 homes and 30 apartments) would be required from the Ottawa region. The number of dwellings to be sampled in each DA was again determined using Neymann allocation, stratified by type of dwelling. On average, approximately 4% of houses and 2% of apartments were selected within each DA to a total of 306 dwellings for the study, based on the assumption of a 40% response rate achieved in the 1991/1992 study (14). Dwellings from each DA were then selected by a simple random sample.

Residents of 306 selected dwellings were sent a letter describing the study and asking them to telephone for more information and/or to indicate whether they were willing to participate. A reminder letter was sent if no response was received within 2 weeks. Nonresponse to the second letter was considered a refusal. Of a total of 306 residences approached, 75 agreed to participate and were sampled, resulting in an overall response rate of 25%. The response rate for houses (39%) was close to the 40% assumed in developing the sampling frame. None of 110 apartment-dwellers contacted agreed to participate. Since proportional allocation was used to determine DA and no apartments participated in the study, samples were self-weighted; therefore, survey weights were not used in the analysis.

Sample Collection. The field work was conducted between November 2002 and March 2003 in the city of Ottawa. One residence was sampled each day with an average of five residences per week. In each home, indoor and outdoor samplers were deployed for the determination of VOCs. Sampling tubes were located in the middle of either the living room or family room in the house, while for outdoor sampling they were located on the front porch or on the driveway of the house, both at a height of about 1.5 m above the ground. Ten liters of air was sampled at a rate of 100 mL min⁻¹ for 100 min. Battery operated Gilian 5 personal samplers (Sensidyne, Florida) were used for collecting indoor air, while mass flow controlled pumps were used for outdoor samples. The flow rate was precalibrated in the laboratory and was measured before and after each sample collection in the field, and the average flow rate was used as the sampling flow rate. The temperature and relative humidity (RH) in both indoor and outdoor air during sampling were also recorded. A questionnaire was administered to collect data on the building characteristics and on the presence of potential sources of the target analytes in the house including tobacco smoke, proximity to vehicle emissions, use and storage of consumer products in the house, home renovations and decorating activities, etc. At least one resident was present and was encouraged to engage in their routine activities during sample collection.

Analytical Method. The gaseous standard (1,3-butadiene, 50 ppm or 121 mg m⁻³ in nitrogen) was custom-ordered from Praxair Specialty Gases and Equipment (Brampton, ON, Canada). Other individual neat compounds were purchased from various suppliers with the highest purity available (at least 98%). Diluted gaseous compound (1,3-butadiene) was prepared by injecting a predetermined volume of the gaseous compound into a fixed volume static dilution bottle filled with ultrapure nitrogen. A composite mixture of other standards was made by first weighing the liquid chemicals and then solid ones directly into a 2-mL amber vial. The composite mixture was then diluted in methanol to desired concentrations. Thermal desorption tubes (Air Toxics, Supelco) used for the study were the standard 3 1/2 in. long stainless steel tubes with brass caps and Teflon ferrules on

both ends. The tubes were packed with Carboxen B and Carboxen 1000 adsorbent material. They were thoroughly conditioned and sealed prior to sample collection. A gastight syringe and a 5- μ L GC injection syringe were used to transfer gaseous and liquid standards onto the sampling tube. The tube was then flushed with nitrogen gas for 1 min at a flow rate of 130 mL min⁻¹ and sealed with stainless steel caps immediately after purging.

An Agilent gas chromatograph (model 6890) coupled with an Agilent mass spectrometer (model 5973N) (Agilent Technologies, Palo Alto, CA) was used to analyze the target chemicals. The analytical column for separating targets was SP-624 (60 m, 0.25 mm i.d., 250 μ m film thickness) (Supelco). The targets in the thermal desorption tubes were released to the GC/MS through a Perkin-Elmer Turbomatrix thermal desorber with an autosampler and an internal standard assembly (Perkin-Elmer, Inc., Norwalk, CT). The instrument operation was controlled by an Agilent Chemstation data system. The thermal desorption condition was set as follows: Initial purge: 3 min at room temperature. Primary desorb: 12 min at 350 °C. trap temperature: -30 °C. Desorb gas: helium at 20 psi. Inlet split: 7 mL min⁻¹. Outsplit: 14 mL min⁻¹. Desorb flow: 60 mL min⁻¹. Secondary desorb from the trap: 3 min at 300 °C. The GC oven temperature started at 40 °C for 5 min, increased at 5 °C/min to 220 °C and was held there for 10 min. The analytes were analyzed under the full-scan detection mode. The mass spectrum of each peak was used for the identification, while the quantification ion (Q-ion) was used for the quantification.

QA/QC. Residues of target analytes in laboratory blank tubes were checked throughout the study.

The method detection limits (MDLs) based on seven low spike sample tubes, expressed in nanograms per tube, were estimated according to eq 1:

$$\text{MDL (ng tube)} = 3.143 \times \text{SD} + B \quad (1)$$

where SD is the standard deviation of the results from 7 sample tubes that were spiked with low level standards, *B* is the mean amount of the chemical in blank tubes, and 3.143 is the Student *t*-value at 99% confidence level for 7 replicates. At least one field blank and one travel blank were included each week, resulting in a total of 17 field blanks and 17 travel blanks for the whole study.

Precision of the method was evaluated by analyzing 5 replicate samples, at both low (10–30 ng per compound) and high spiking levels (200–600 ng per compound). Evaluation of sample breakthrough volume was carried out by assessing the sampling efficiency with a sampling volume of 15 L. The sample tubes were spiked at 300–900 ng per compound and flushed afterward with 15 L of clean air. Storage stability was examined by storing the tubes that were spiked with target analytes for up to 7 days prior to analysis. At least one field blank and one travel blank were included each week, resulting in a total of 17 field blanks and 17 travel blanks for the whole study.

Results and Discussion

Analytical Method Performance. The performance of the analytical method was evaluated for each of the target analytes (Table 1). The majority of the analytes were not present in detectable amounts in the blank tubes. Only a few analytes that are common laboratory solvents (carbon disulfide, benzene, xylene, and acetone) were detected in the blank tubes with mean values between 0.1 and 0.2 ng per tube with the exception of dichloromethane, which had the blank level at 1.01 ng per tube. MDLs of the majority of analytes were below 0.5 ng per sample tube, which translates into MDLs of less than 0.05 μ g m⁻³ based on 10 L of air sampled (values listed in Table 3). Only a few analytes had

TABLE 1. List of Target Analytes and Method Performance for These Analytes

analyte	CEPA class ^b	CAS number	GC/MS		tube blank (n = 9)		low level spiking (n = 7)			precision (n = 5)		sampling efficiency (%)	duplicate (Diff) ^a mean (n) ^g RSD ^f
			RT (min)	Q-ion ^c (m/z)	mean (ng)	SD ^d	mean (ng)	SD ^d	MDL ^e (ng)	low RSD ^f	high RSD ^f		
acetone	SA	67-64-1	9.05	43	0.08	0.04	0.24	0.06	0.26	16	12	96	15 (7)
benzene	PSL1	71-43-2	16.07	78	0.16	0.09	0.40	0.11	0.50	25	2	88	40 (7)
dichloromethane	PSL1	75-09-2	10.27	49	1.01	1.21	0.55	0.06	1.2	4	3	88	17 (7)
toluene	PSL1	108-88-3	21.43	91	0.03	0.02	0.23	0.08	0.29	45	3	90	23 (7)
chloroform	PSL2	67-66-3	14.51	83	0.00	0.00	0.79	0.05	0.17	3	3	85	40 (7)
cyclohexane	SA	110-82-7	15.25	56	0.00	0.00	0.66	0.10	0.32	3	3	88	29 (6)
2-propanol	SA	67-63-0	9.40	45	0.00	0.00	0.88	0.14	0.44	8	3	90	36 (7)
styrene	PSL1	100-42-5	27.59	104	0.00	0.00	0.48	0.16	0.50	5	3	90	21 (7)
decane	SA	124-18-5	30.42	43	0.00	0.00	0.31	0.04	0.12	6	3	90	44 (7)
o-xylene	PSL1	95-47-6	27.57	91	0.02	0.07	0.23	0.06	0.20	6	2	89	31 (7)
m/p-xylene	PSL1	h	26.29	91	0.10	0.19	0.45	0.11	0.43	6	3	89	29 (7)
1,2,4-trimethylbenzene	SA	95-63-6	31.77	105	0.02	0.05	0.29	0.08	0.28	2	2	88	49 (7)
2-butanone	SA	78-93-3	13.65	43	0.00	0.00	0.14	0.03	0.10	4	3	72	17 (7)
naphthalene	SA	91-20-3	40.45	128	0.00	0.00	0.60	0.07	0.22	3	2	90	39 (7)
tetrachloroethylene	PSL1	127-18-4	23.08	166	0.00	0.00	0.80	0.08	0.26	3	2	89	29 (7)
ethylbenzene	SA	100-41-4	25.94	91	0.00	0.00	0.35	0.05	0.14	4	3	89	29 (7)
1,4-dichlorobenzene	PSL1	106-46-7	33.07	146	0.00	0.00	0.31	0.04	0.13	8	3	88	44 (7)
phenol	PSL2	108-95-2	34.29	94	0.00	0.00	0.21	0.06	0.20	3	3	85	32 (7)
carbon disulfide	PSL2	75-15-0	9.62	76	0.20	0.07	0.58	0.04	0.33	5	3	88	31 (4)
1-butanol	SA	71-36-3	17.45	56	0.00	0.00	2.43	0.37	1.15	19	2	86	53 (5)
acrylonitrile	PSL2	107-13-1	10.88	53	0.00	0.00	0.32	0.08	0.25	7	4	90	27 (5)
4-methyl-2-pentanone	SA	108-10-1	20.78	43	0.00	0.00	0.29	0.05	0.16	3	3	76	19 (7)
1,1-dichloroethylene	SA	75-35-4	8.97	61	0.00	0.00	0.60	0.04	0.11	6	6	90	9 (4)
2-butoxyethanol	PSL2	111-76-2	28.70	57	0.00	0.00	2.28	0.90	2.8	41	10	91	25 (6)
trichloroethylene	PSL1	79-01-6	17.81	95	0.00	0.00	0.51	0.06	0.19	5	2	91	23 (6)
1,3-butadiene	PSL2	106-99-0	5.50	39	0.00	0.00	11.08	1.01	3.2	10	3	99	33 (5)
methyl methacrylate	PSL1	80-62-6	18.61	41	0.00	0.00	0.39	0.03	0.10	4	2	85	2 (1)
methyl tert-butyl ether	PSL1	1634-04-4	10.98	73	0.00	0.00	0.99	0.16	0.51	5	3	74	31 (2)
chlorobenzene	PSL1	108-90-7	25.67	112	0.00	0.00	0.45	0.04	0.12	4	2	90	na
3,5-dimethylaniline	PSL1	108-69-0	40.10	121	0.00	0.00	19.96	3.82	12	35	8	139	38 (3)
1,2-dichloroethane	PSL1	107-06-2	16.09	62	0.00	0.00	0.70	0.05	0.17	3	3	86	50 (1)
1,2-dichlorobenzene	PSL1	95-50-1	34.31	146	0.00	0.00	0.57	0.07	0.23	5	3	89	14 (1)
2-ethoxyethanol	PSL2	110-80-5	19.67	59	0.00	0.00	3.13	0.42	1.3	24	5	83	16 (1)
2-methoxyethanol	PSL2	109-86-4	16.22	45	0.00	0.00	2.48	0.73	2.3	12	3	78	na
1,2-dichloropropane	SA	78-87-5	18.43	63	0.00	0.00	1.26	0.12	0.38	4	3	85	na
ethylene dibromide	SA	106-93-4	24.21	107	0.00	0.00	0.84	0.06	0.18	2	3	93	na
1,1,2,2-tetrachloroethane	PSL1	79-34-5	29.61	83	0.00	0.00	0.77	0.07	0.23	2	2	91	na

^a Diff = percentage difference between duplicate pairs, defined as $|d_1 - d_2| / (d_1 + d_2) \times 2 \times 100$. ^b CEPA class: PSL = Priority Substance List; SA = screening assessment. ^c Quantification ion (Q-ion) was used for quantifying the peak. ^d SD = standard deviation. ^e MDL = method detection limit, in ng per tube. ^f RSD = relative standard deviation, defined as $(SD/mean) \times 100$. ^g Mean values of Diff of all duplicate pairs. ^h The CAS number for *m*-xylene is 108-38-3 and for *p*-xylene is 106-42-3.

TABLE 2. House Characteristics and Sampling Conditions

study period	November 20, 2002–March 11, 2003
indoor temp (°C)	mean: 19 ± 2, range: 14.5–26.2
outdoor temp (°C)	mean: -10 ± 9, range: -26 to 12
indoor RH	mean: 32 ± 9, range: 12–53
outdoor RH	mean: 61 ± 15, range: 30–90
type of dwelling	62 single, 8 row unit, 4 semi-detached, 1 mobile home
age of house (yr)	mean: 37 ± 26
floor area (ft ²)	mean: 1776 ± 614
heating type	61 natural gas, 10 oil, 4 others
location of home	48 quiet residential area, 22 main residential area, 5 major commercial area
smoking indoors	10 homes, range from less than 1 h before sampling to several days

MDLs above 0.05 $\mu\text{g m}^{-3}$. They were 1,3-butadiene (MDL = 0.32 $\mu\text{g m}^{-3}$), which is the only gaseous analyte in this study; dichloromethane (MDL = 0.12 $\mu\text{g m}^{-3}$), a common laboratory solvent with high levels in blank tubes; 3,5-dimethylaniline (MDL = 1.2 $\mu\text{g m}^{-3}$), an alkaline chemical with poor peak shape on the GC chromatogram; and three polar glycol ethers (2-methoxyethanol, MDL = 0.23 $\mu\text{g m}^{-3}$; 2-ethoxyethanol, MDL = 0.13 $\mu\text{g m}^{-3}$; and 2-butoxyethanol, MDL = 0.28 $\mu\text{g m}^{-3}$) also due to their poor peak shape.

Good precision was demonstrated at two spiking levels. At the high spiking level, the relative standard deviation (RSD) was in the range of 2–3% for the majority of the analytes. Only two analytes had an RSD of 10% or more (acetone, RSD = 12%; 2-butoxyethanol, RSD = 10%). At the low spiking level, the RSDs for the majority of the analytes were still under 25%. The analytes that had large RSDs were toluene (RSD = 45%), 2-butoxyethanol (RSD = 41%), and 3,5-dimethylaniline (RSD = 35%).

Since the sampling volume of 10 L was anticipated based on the detection requirement, an evaluation of sampling efficiency at a sampling volume of 15 L was carried out. The sampling efficiencies were all greater than 70%, indicating no significant breakthrough. In general, the sampling efficiency of oxygen-containing targets (2-butanone, 72%; methyl *tert*-butyl ether, 74%; 4-methyl-2-pentanone, 76%; and 2-methoxyethanol, 78%) was low as compared to the others. 3,5-Dimethylaniline had a rather abnormal value (139%). There was also no significant loss of analytes for the storage time up to 7 days with recoveries all over 80% (data not shown).

Duplicate samples were collected in about 10% of the participating homes. The mean values of the percentage difference (%Diff) between each pair of duplicate samples

TABLE 3. Levels of Target Analytes ($\mu\text{g m}^{-3}$) in Indoor and Outdoor Air of Randomly Selected Dwellings in Ottawa^a

analyte	indoor air (n = 75)							outdoor air (n = 74)							I/O correlation
	MDL	concn range ^b	AM	50th	75th	90th	det freq	concn range ^b	AM	50th	75th	90th	det freq	P value ^c	
acetone	0.03	(0.015–455.87)	44.44	28.48	46.85	76.4	99	(0.015–15.25)	1.22	0.2	1.21	3.56	68	<0.0001	
benzene	0.05	(0.025–20.99)	2.85	2.15	3.43	5.21	97	(0.025–16.88)	1.19	0.29	0.79	2.38	62	<0.0001	
dichloromethane	0.12	(0.06–408.37)	14.98	1.87	8.15	43.21	95	(0.06–3.49)	0.32	0.06	0.23	0.58	45	<0.0001	
toluene	0.03	(0.015–112.93)	11.54	5.53	12.25	25.47	95	(0.015–30.10)	2.48	0.45	1.43	9.58	73	<0.0001	
chloroform	0.02	(0.01–8.23)	1.72	1.19	2.49	4.39	93	(0.01–0.88)	0.06	0.01	0.01	0.13	22	<0.0001	
cyclohexane	0.03	(0.015–54.12)	6.58	4.51	7.86	15.1	93	(0.015–32.80)	1.98	0.015	1.13	2.37	43	<0.0001	
2-propanol	0.04	(0.02–238.17)	18.14	3.32	21.35	68.76	92	(0.02–4.89)	0.14	0.02	0.02	0.18	24	<0.0001	
styrene	0.05	(0.025–6.53)	0.69	0.46	0.87	1.49	88	(0.025–4.80)	0.14	0.025	0.06	0.17	34	<0.0001	
o-xylene	0.02	(0.01–205.11)	5.08	1.22	3.37	6.48	87	(0.01–14.44)	0.83	0.1	0.33	1.39	73	<0.0001	
decane	0.01	(0.005–84.60)	5.28	2.17	4.68	8.09	87	(0.005–3.44)	0.31	0.085	0.3	0.8	72	<0.0001	
m/p-xylene	0.04	(0.02–138.97)	7.5	3.59	6.93	16.35	85	(0.02–30.92)	1.78	0.24	0.88	3.78	74	<0.0001	
1,2,4-trimethylbenzene	0.03	(0.015–56.60)	3.97	2.21	3.38	6.73	85	(0.015–11.43)	0.79	0.15	0.41	2.92	68	<0.0001	
2-butanone	0.01	(0.005–16.45)	2.54	1.48	2.82	6.66	84	(0.005–4.23)	0.37	0.12	0.29	0.6	61	<0.0001	
tetrachloroethylene	0.03	(0.015–9.23)	1.15	0.47	1.4	3.25	83	(0.015–2.44)	0.19	0.015	0.08	0.31	38	<0.0001	
ethylbenzene	0.01	(0.005–201.41)	4.71	1.05	1.98	4.76	83	(0.005–9.35)	0.58	0.1	0.26	2.23	73	<0.0001	
naphthalene	0.02	(0.01–144.44)	3.87	0.39	1.1	4.75	83	(0.01–3.85)	0.18	0.02	0.07	0.25	54	<0.0001	
1,3-dichlorobenzene	0.01	(0.005–16.19)	0.77	0.15	0.29	1.05	81	(0.005–1.46)	0.06	0.005	0.03	0.08	27	<0.0001	
phenol	0.02	(0.01–5.16)	0.7	0.42	1.14	1.67	69	(0.01–1.41)	0.06	0.01	0.04	0.13	28	<0.0001	
carbon disulfide	0.03	(0.015–3.29)	0.34	0.13	0.46	0.86	67	(0.015–0.38)	0.04	0.015	0.015	0.11	22	<0.0001	
1-butanol	0.12	(0.06–139.66)	4.25	0.4	2.02	5.96	59	(0.06–0.80)	0.09	0.06	0.06	0.06	7	<0.0001	
4-methyl-2-pentanone	0.02	(0.01–1.40)	0.26	0.16	0.38	0.8	53	(0.01–0.61)	0.05	0.01	0.01	0.01	9	<0.0001	
acrylonitrile	0.03	(0.015–8.89)	0.27	0.06	0.13	0.26	53	(0.015–0.18)	0.02	0.015	0.015	0.015	9	<0.0001	
1,1-dichloroethylene	0.01	(0.005–4.05)	0.27	0.005	0.37	0.83	45	(0.005–0.83)	0.05	0.005	0.005	0.12	18	<0.0001	
2-butoxyethanol	0.28	(0.14–41.44)	2.85	0.14	1.89	7.06	40	(0.14–3.91)	0.23	0.14	0.14	0.14	7	<0.0001	
trichloroethylene	0.02	(0.01–0.87)	0.06	0.01	0.08	0.19	33	(0.01–1.49)	0.08	0.01	0.01	0.13	19	0.1083	
1,3-butadiene	0.32	(0.16–3.65)	0.5	0.16	0.47	1.64	32	(0.16–0.16)	0.16	0.16	0.16	0.16	0	<0.0001	
methyl methacrylate	0.01	(0.005–1.12)	0.05	0.005	0.005	0.06	11	(0.005–0.33)	0.009	0.005	0.005	0.005	1	0.0332	
methyl tert-butyl ether	0.05	(0.025–3.32)	0.17	0.025	0.025	0.025	9	(0.025–1.65)	0.05	0.025	0.025	0.025	3	0.0348	
chlorobenzene	0.01	(0.005–0.04)	0.006	0.005	0.005	0.005	8	(0.005–0.09)	0.008	0.005	0.005	0.005	5	0.8082	
3,5-dimethylaniline	1.2	(0.60–4.71)	0.77	0.6	0.6	0.6	8	(0.60–0.60)	0.6	0.6	0.6	0.6	0	0.0146	
1,2-dichloroethane	0.02	(0.01–0.71)	0.03	0.01	0.01	0.01	5	(0.01–0.69)	0.04	0.01	0.01	0.01	4	0.4309	
1,2-dichlorobenzene	0.02	(0.01–0.11)	0.01	0.01	0.01	0.01	5	(0.01–0.04)	0.01	0.01	0.01	0.01	3	0.3207	
2-ethoxyethanol	0.13	(0.065–27.14)	0.43	0.065	0.065	0.065	3	(0.065–0.065)	0.065	0.065	0.065	0.065	0	0.1602	
2-methoxyethanol	0.23	(0.115–0.115)	0.115	0.115	0.115	0.115	0	(0.115–0.115)	0.115	0.115	0.115	0.115	0	na	
1,2-dichloropropane	0.04	(0.02–0.02)	0.02	0.02	0.02	0.02	0	(0.02–0.23)	0.03	0.02	0.02	0.02	3	0.1602	
ethylene dibromide	0.02	(0.01–0.01)	0.01	0.01	0.01	0.01	0	(0.01–0.01)	0.01	0.01	0.01	0.01	0	na	
1,1,2,2-tetrachloroethane	0.02	(0.01–0.01)	0.01	0.01	0.01	0.01	0	(0.01–0.01)	0.01	0.01	0.01	0.01	0	na	

^a na: Test was not performed due to all values being non-detects. AM = arithmetic mean. ^b VOC levels that were below the detection limit or were nondetected were replaced with 1/2 MDL. ^c The statistical significance was tested using a Wilcoxon Signed Rank Test, since the assumptions for a paired t-test were not satisfied. The test is based on 74 matched pairs as the outdoor level for one of the houses was missing.

are presented in Table 1. The majority of the mean values were in the range of 10–40%.

Levels of Target Analytes in Residential Air. Sample collection was conducted during the winter season, when the outdoor temperature ranged from –26 to +12 °C. The indoor temperature remained relatively constant (19 ± 2 °C) as houses in Ottawa are heated during the winter season. The majority of participating homes were single houses located in residential areas and used natural gas as the heating source. The average age of the houses was 37 years with a range from newly constructed to over 100 years old. The house characteristics and sampling conditions (temperature and relative humidity) are summarized in Table 2. The roughly 13% smoking homes (10 homes) in this study is similar to the reported 17% of the smoking population aged 12 years and older in the city of Ottawa (15).

Table 3 summarizes the levels of target analytes in indoor air and outdoor air of the 75 selected homes. One of the outdoor air samples was invalid due to the failure of the sampling instrument; therefore, the number of outdoor air samples was 74. The distribution of the levels of target analytes in both indoor and outdoor air were right skewed; they were neither normally nor log-normally distributed as indicated by the Anderson–Darling test. Therefore, concentrations at different population percentiles (50th, 75th, 90th, and range) are listed in Table 3 along with the values of arithmetic means for each target analyte. The target analytes in Table 3 are

ordered according to their detection frequency in indoor air. The first 17 analytes were detected in indoor air with detection frequency greater than 80%. The next nine analytes were detected with a moderate detection frequency between 30% and 80%. The remaining 11 analytes in the third group were either detected with very low frequency or not detected at all in indoor air. In calculating the arithmetic mean values reported in Table 3, a value of 1/2 MDL was assumed for those samples that did not contain detectable levels of a given target analyte. A sensitivity analysis was carried out for target analytes with detection frequency lower than 80% (results not shown). Zero and MDL value were used as the minimum and maximum numbers to replace the nondetected concentrations for the analysis. It was observed that the mean value for the target analytes with less than 80% detection frequency was not highly affected by the value used for imputation. Of course the results changed little for analytes with more than 80% of samples above the detection limit.

In general, both the detection frequency and the concentrations were lower in outdoor air. For all of the compounds in the first two groups, except trichloroethylene, the concentrations in indoor air were significantly higher than those outdoors with $p < 0.0001$. For less frequently detected compounds (third group), the difference in levels in indoor and outdoor air was often not statistically significant.

TABLE 4. 95% Confidence Intervals (CI) for Target Analytes ($\mu\text{g m}^{-3}$) with Detection Frequency Greater than 80% in Indoor Air, Based on Bootstrap Method ($n = 75$)

analyte	mean ^a ± SD ^b	95% CI	det freq
acetone	44.44 ± 65.88	(31.51–60.99)	99
benzene	2.85 ± 2.86	(2.27–3.56)	97
dichloromethane	14.98 ± 49.6	(6.65–27.94)	95
toluene	11.54 ± 17.54	(8.04–15.88)	95
chloroform	1.72 ± 1.83	(1.33–2.15)	93
cyclohexane	6.58 ± 7.8	(5.00–8.49)	93
2-propanol	18.14 ± 35.34	(11.05–26.78)	92
styrene	0.69 ± 0.93	(0.51–0.92)	88
o-xylene	5.08 ± 23.67	(1.82–10.96)	87
decane	5.28 ± 12.05	(3.03–8.31)	87
m/p-xylene	7.5 ± 17.14	(4.52–11.96)	85
1,2,4-trimethylbenzene	3.97 ± 8.57	(2.33–6.16)	85
2-butanone	2.54 ± 3.28	(1.85–3.33)	84
tetrachloroethylene	1.15 ± 1.75	(0.78–1.57)	83
ethylbenzene	4.71 ± 23.52	(1.34–10.74)	83
naphthalene	3.87 ± 17.25	(1.14–8.54)	83
1,4-dichlorobenzene	0.77 ± 2.48	(0.29–1.40)	81

^a VOC levels that were below the detection limit or were nondetected were replaced with 1/2 MDL. ^b SD = standard deviation.

The concentration range in indoor air of most compounds varied over 3–4 orders of magnitude. For example, the range for acetone in indoor air was 0.015–456 $\mu\text{g m}^{-3}$, and for benzene it was 0.025–21 $\mu\text{g m}^{-3}$. Some concentration ranges were much larger than the most variable compound (acetaldehyde) in the Windsor Air Quality study (12), whose results were used in determining the required numbers of DAs and dwellings for the current study. As a result, compounds with a larger variability compared to acetaldehyde in the Windsor study will consequently have a larger bound in their respective 95% CI. The Bootstrap method (16) was used to estimate 95% confidence limits for analytes with detection frequency greater than 80%, since the concentration levels of the analytes did not follow either a normal or log-normal distribution (Table 4). For those analytes with a detection frequency less than 80%, the Bootstrap method could not produce reliable results. The 95% confidence limits for all these VOCs except acetone, dichloromethane, and 2-propanol were estimated within the target bound of 7 $\mu\text{g m}^{-3}$.

Indoor/Outdoor (I/O) Ratio and Indoor Source Strength.

The extent of the contribution of indoor sources to indoor air levels was investigated using the I/O ratio and indoor source strength. The concentration levels in indoor air and outdoor air at the 50th, 75th, and 100th percentiles were used (MDL values were used for non-detectables) to generate the indoor air to outdoor air concentration ratios. I/O ratios for the analytes that had detection frequency of greater than 30% in indoor air (the first two groups) are summarized in Table 5. The remaining 11 compounds that were detected less frequently in indoor air (Table 2) were not included in Table 5 because with such a low detection frequency, the I/O ratio becomes less meaningful. The I/O ratios were greater than 1 for virtually all analytes and were as high as 150, indicating the presence of strong indoor sources for some of these chemicals. The I/O ratio of trichloroethylene at the 100th percentile (I/O = 0.6) was the only value below 1.

The indoor source strength, expressed as estimated emission rate per home (E , mg h^{-1}) can be calculated using eq 2:

$$E = (C_i - C_o) \times \text{ach} \times V/1000 \quad (2)$$

where V (m^3) is average house volume, ach is the average air change rate (h^{-1}), and C_i and C_o are the concentrations ($\mu\text{g m}^{-3}$) of a chemical in indoor and outdoor air, respectively (17). The emission rates at three percentiles were calculated

TABLE 5. I/O Ratios and Estimated Emission Rates (mg h^{-1}) of Measured Chemicals That Had a Detection Frequency in Indoor Air Greater than 30%

analyte	I/O ratio			emission rate per home (mg h^{-1})		
	50th	75th	100th	50th	75th	100th
acetone	148	38	30	3.9	6.0	60
benzene	7.7	4.3	1.2	0.26	0.35	0.6
dichloromethane	16	38	117	0.26	1.1	55
toluene	13	10	3.8	0.70	1.4	11
chloroform	59	122	9.4	0.16	0.33	1.0
cyclohexane	150	7.2	1.7	0.62	0.92	2.9
2-propanol	83	417	49	0.45	2.3	32
styrene	9.2	14	1.4	0.06	0.11	0.2
decane	27	16	25	0.29	0.59	11
o-xylene	13	9.4	14	0.15	0.34	26
m/p-xylene	16	9.0	4.5	0.46	0.78	15
1,2,4-trimethylbenzene	16	8.5	5.0	0.28	0.41	6.2
2-butanone	13	9.2	3.9	0.19	0.32	1.7
naphthalene	22	17	38	0.05	0.14	19
tetrachloroethylene	16	16	3.8	0.06	0.16	0.9
ethylbenzene	11	7.2	22	0.13	0.22	26
1,4-dichlorobenzene	15	16	11	0.02	0.03	2.0
phenol	21	41	3.7	0.06	0.15	0.5
carbon disulfide	4.1	14	8.8	0.02	0.06	0.4
1-butanol	3.3	15	174	0.05	0.25	19
acrylonitrile	2.1	4.3	49	0.01	0.02	1.2
4-methyl-2-pentanone	7.9	19	2.3	0.02	0.05	0.1
1,1-dichloroethylene	nd ^a	34	4.9	0.05	0.4	
trichloroethylene	nd	3.6	0.6	0.01	0.0	
1,3-butadiene	nd	1.4	11	0.06	0.5	
2-butoxyethanol	nd	6.5	11	0.25	5.1	

^a nd = not detected.

using the indoor and outdoor air concentrations at these respective percentiles (Table 2).

The air change rate of homes was not measured in this study. Average air change rates measured in 24 selected homes in 1991/1992 national survey of VOCs in Canadian homes and in 44 Canadian homes during wintertime (January–February 1991) both were 0.34 h^{-1} (17). This value was considered a reasonable estimate to apply to the current study. Although the current study was conducted more than 10 years later, the dwellings in the two studies were built at roughly the same year on average. In addition, although the current study was restricted to the winter, Davis and Otson (17) reported in 1991/1992 study that the average air exchange rates for Canadian dwellings was similar in available studies regardless of whether they were restricted to winter or not.

TABLE 6. Comparisons of Indoor Air Concentrations from This Study to Two Earlier Published Studies^a

analyte	present study			Canadian survey ^b	Brown et al. study ^c
	AM ^d		GM ^d	AM ^d	WAGM ^d
	(0 MDL) ^e	(1/2 MDL) ^e	(1/2 MDL) ^e	(0 MDL)	not indicated
acetone	44.44	44.44	24.02		20–50
benzene	2.85	2.85	1.83	6.4	5–10
dichloromethane	14.98	14.98	2.18	20	10–20
toluene	11.54	11.54	4.74	18	20–50
chloroform	1.72	1.72	0.80	1.3	1–5
cyclohexane	6.58	6.58	3.28		1–5
2-propanol	18.13	18.14	3.43		
styrene	0.69	0.69	0.35	0.1	15
<i>o</i> -xylene	5.08	5.08	0.74	4.1	5–10
decane	5.27	5.28	1.08	25	5–10
<i>m/p</i> -xylene	7.50	7.50	1.71	16	10–20
1,2,4-trimethylbenzene	3.97	3.97	1.07	6.9	5–10
2-butanone	2.54	2.54	0.71		1–5
tetrachloroethylene	1.15	1.15	0.36	2.1	5–10
ethylbenzene	4.71	4.71	0.47	6.5	5–10
naphthalene	3.87	3.87	0.33	3.2	<1
1,4-dichlorobenzene	0.77	0.77	0.11	36	5–10
phenol	0.69	0.70	0.19		
carbon disulfide	0.33	0.34	0.11		
1-butanol	4.22	4.25	0.42		<1
4-methyl-2-pentanone	0.26	0.26	0.07		
acrylonitrile	0.26	0.28	0.05		
1,1-dichloroethylene	0.27	0.27	0.04		1–5
2-butoxyethanol	2.76	2.85	0.49		
trichloroethylene	0.06	0.06	0.02		5–10
1,3-butadiene	0.39	0.50	0.28		
methyl methacrylate	0.04	0.05	0.01		
methyl <i>tert</i> -butyl ether	0.14	0.17	0.04		
chlorobenzene	0.002	0.006	0.01		<1
3,5-dimethylaniline	0.22	0.77	0.67		
1,2-dichloroethane	0.02	0.03	0.01	nd	<1
1,2-dichlorobenzene	0.003	0.01	0.01		<1
2-ethoxyethanol	0.37	0.43	0.07		
2-methoxyethanol	0.00	0.115	0.115		
1,2-dichloropropane	0.00	0.02	0.02		<1
ethylene dibromide	0.00	0.01	0.01		1–5
1,1,2,2-tetrachloroethane	0.00	0.01	0.01	nd	

^a Concentrations in $\mu\text{g m}^{-3}$. ^b 1991/1992 Canadian indoor air study, see refs 9, 10, and 17. ^c A review by Brown et al., see ref 4. Treatment of nondetectables was not indicated. ^d AM = arithmetic mean, GM = geometric mean, WAGM = weighted average geometric mean. ^e For below MDL data, zero (coded as 0 MDL) or half of the MDL (1/2 MDL) value was used.

Moreover, since the indoor source strength was primarily used to rank substances with respect to their estimated quantity of indoor emissions, (i.e., it was used as a relative measure), the value assumed for the average air exchange rate was less critical. The average house volume of the present study was estimated to be 402 m³, based on the information obtained through the questionnaire administered in each home during the study (the average floor area of 165 m² times a height of 2.44 m). This value is larger than the one (265 m³) derived from Canadian national statistics (17) at least in part because the dwellings in the present study did not include the apartment units.

Estimated emission rates of various chemicals at 50th, 75th, and 100th percentiles are presented in Table 5. At the 50th percentile, or median, the emission rate ranged from 0.01 mg h⁻¹ for acrylonitrile to 3.9 mg h⁻¹ for acetone in residential dwellings. Acetone had the highest emission rates at all three percentiles, with its maximum emission rate (100th percentile) estimated at 60 mg h⁻¹.

The I/O ratios and the estimated emission rates both confirm the important contribution of indoor sources to the indoor levels measured for a number of these compounds. However, it is noted that the ranking that would be indicated by these two measures differs substantially. For example, phenol had the third highest I/O ratio at 75th percentile but ranked 17th in corresponding emission rate, and toluene

had the 15th in I/O ratio but ranked 2nd in emission rate at 50th percentile.

Comparing Levels of VOCs Measured in This Study with Others. It is of interest to compare the indoor air concentrations measured in this study to similar studies conducted at earlier time periods. The key study for comparison is a 1991/1992 Canadian study by Otson and colleagues, in which levels of 26 VOCs were determined in 754 dwellings selected at random (using a three-stage sampling design) to be representative of the general population in all 10 provinces in Canada (9, 10, 14, 17). Fellin and Otson (9) reported the arithmetic mean (AM) concentrations of the individual VOCs for each season. For comparison to the results from the present study, only data from the 185 dwellings sampled during the winter were used, since this was the season when the present study was conducted.

Another source of data for comparison is a 1994 review by Brown et al. (4) of studies from the 1980s and 1990s in which concentrations of VOCs in the indoor air of dwellings were measured. In the review, the authors provided weighted average geometric means (WAGM) for a number of VOCs reported in established dwellings, based on several studies from the United States and several European countries.

Table 6 presents the comparison of the summary results for the individual VOCs from these studies to those from our

study. To facilitate comparison, the AM concentration for each VOC measured in our study has been calculated in a fashion similar to that used by Fellin and Otson (9) for the 1991/1992 data (i.e., assuming a value of zero for those samples that were not detected (nd)) as well as using the more conventional assumption of one-half of the detection limit for values less than the MDL. Although the concentration levels from the present study were not normally or log-normally distributed, the geometric mean (GM) values were calculated for the purpose of comparison with the WAGM values reported by Brown et al. (4).

The AM levels of the present study were substantially lower for most chemicals than those reported for the 1991/1992 Canadian survey (9). The greatest apparent decrease was observed for 1,4-dichlorobenzene (a component of some formulations of toilet bowl deodorizers and mothballs) from $36 \mu\text{g m}^{-3}$ observed in 1991/1992 study down to $<1 \mu\text{g m}^{-3}$. Other chemicals with lower mean levels in indoor air in our study included benzene, toluene, *m/p*-xylene, decane, ethylbenzene, 1,2,4-trimethylbenzene, dichloromethane, and trichloroethylene. AM levels of styrene, chloroform, and naphthalene were higher in the present study than those reported from the 1991/1992 study. However, this apparent increase is likely the result of the bias introduced by assuming a value of zero for nd samples, as the 1991/1992 study had a much higher frequency of nd in samples as a consequence of its higher detection limits ($1.6\text{--}5.9 \mu\text{g m}^{-3}$) (9). The GM values from the present study were generally lower than the WAGM calculated by Brown et al. (4) as well, with most chemicals having GM values several times below the WAGM range calculated by Brown et al. The largest decrease was for 1,4-dichlorobenzene (0.11 vs $5\text{--}10 \mu\text{g m}^{-3}$) and tetrachloroethylene (0.36 vs $5\text{--}10 \mu\text{g m}^{-3}$).

Hence, for most of the VOCs studied, the values determined in the present study were lower than those from earlier studies. A more rigorous analysis is not possible because of differences in study design, sampling, and analytical methods and the limited reporting of the results from earlier studies. However, this simple comparison suggests that typical residential exposures to a number of VOCs may be decreasing over time. This is likely the combined result of a number of factors, including a growing concern with residential indoor air quality issues, the establishment of U.S. national VOC emission standards for certain categories of consumer products in 1998 (18), increased availability of products that release lesser amounts of these substances, and a decrease in the prevalence of current smokers (from roughly 30–35% of Canadians aged 15+ years at the time of the earlier studies to about 20% currently) (19). The decrease in indoor air concentrations of a number of VOCs from historical levels was also noted in a recent review of U.S. studies (20). Decreased levels would also be expected if the houses in the 1991/1992 survey were much newer than those in the Ottawa study (emissions from building materials decrease over time) or if houses constructed in Ottawa in the interim were better ventilated than those in the older studies. However, emissions from building materials, while initially high, fall off to very low background levels within several months to a year, whereas the average age of the homes was roughly 30 years or more in both studies. In addition, only 4 of the 75 homes in our study were constructed since the time of the 1991/1992 survey. Hence, it is unlikely that the large apparent decrease in VOC levels over time is due to differences in the age or ventilation of the houses between our study and the earlier ones.

The present study has yielded up-to-date information on the levels of a number of airborne chemicals in indoor air and outdoor air of representative houses in the city of Ottawa. A number of the target analytes have not traditionally been measured in indoor air research but are instead priorities

from the perspective of health risk assessment and risk management programs under CEPA 1999. These data highlight the importance of indoor sources of these chemicals in residential environments and suggest that levels of many of these chemicals in indoor air may be decreasing over time. The results can be used to estimate population inhalation exposure to these chemicals in a residential environment based on the inhalation rate and time people spend indoors and outdoors.

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