

Differences in source emission rates of volatile organic compounds in inner-city residences of New York City and Los Angeles

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The Toxics Exposure Assessment Columbia-Harvard (TEACH) Project characterized personal, indoor, and outdoor concentrations of a suite of volatile organic compounds (VOCs) for high school students living in New York City (NYC) and Los Angeles (LA). This paper presents the analysis of VOC measurements collected indoors and outdoors for 46 students' homes in NYC and for 41 students' homes in LA across two seasons. Dual-sorbent thermal desorption tubes were used for the collection of 15 VOCs and C₁₈ 2,4-dinitrophenylhydrazine-coated cartridges were used for the collection of seven aldehydes. Air-exchange rates (AERs) were also measured using a perfluorocarbon tracer gas method. The AERs were lower in the winter in both cities, averaging 1 h⁻¹ in NYC and 1.4 h⁻¹ in LA, compared with 1.8 h⁻¹ in NYC in the summer and 2.5 h⁻¹ in LA in the fall. Higher AERs were generally associated with lower indoor-outdoor ratios with significant differences for the compounds with indoor sources, including chloroform, 1,4-dichlorobenzene, and formaldehyde. Using a mass-balance model to account for AER and other housing parameters, effective source emission rates (SER) were calculated for each compound. Based on I/O ratios and source emission rates, VOCs could be divided into: (1) indoor-source-influenced compounds, (2) those with contributions from both indoor and outdoor sources, and (3) those with mostly outdoor sources. Significant indoor sources were found for the following six compounds (mean emission rates presented): chloroform (0.11 mg/h), 1,4-dichlorobenzene (19 mg/h), formaldehyde (5 mg/h), acetaldehyde (2 mg/h), benzaldehyde (0.6 mg/h), and hexaldehyde (2 mg/h). Although chloroform had variable I/O ratios across seasons, SERs, which accounted for AER, were similar in both cities for both seasons (e.g., LA means 0.12 and 0.11 mg/h in winter and fall, respectively). Formaldehyde had substantially higher indoor emission rates in the summer in NYC compared to winter (3.8 vs. 1.6 mg/h) but lower in the fall in LA compared to winter (4.3 vs. 5.0 mg/h). Uncertainty analysis determined that source strength calculations were not sensitive to measurement error for a subset of homes in LA.

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Introduction

In the 1990, Clean Air Act (CAA) amendments, 189 compounds were designated as hazardous air pollutants (HAPs).² Furthermore, EPA (2000) identified 33 of these 188 compounds as high-priority HAPs in their integrated urban air toxic strategy. A majority of these priority HAPs are volatile organic compounds (VOCs) that may be associated with a variety of adverse health effects ranging from acute effects, such as irritation and headaches to more chronic effects, such as liver and kidney damage and cancer (Molhave, 1991; Wallace, 1991; Lippmann, 1992; Hodgson et al., 1994; Caprino and Tonga, 1998). For example,

formaldehyde is both an irritant and a suspected human carcinogen (Kerns et al., 1983; Woutersen et al., 1987; Nielsen et al., 1999; Clausen et al., 2001). Benzene is a known human carcinogen (IARC, 1982; Rinsky, 1989; Hayes et al., 1997; Rinsky et al., 2002), and several chlorinated compounds have been shown to cause liver and kidney damage in animal studies (Klaassen and Plaa, 1966).

As part of the US EPA's HAPs regulatory strategy, the Cumulative Exposure Study was conducted to estimate outdoor VOC concentrations across the US and determine the risks associated with their exposures (Woodruff et al., 1998, 2000; Rosenbaum et al., 1999). Estimates of outdoor concentrations, however, may underestimate actual exposures to many of these VOCs due to the fact that people spend, on average, over 85% of their time indoors, and hence personal exposure to VOCs occur largely in the home and other indoor microenvironments (Klepeis et al., 2001).

Several compounds are expected to have a significant portion of exposure from indoor sources. Tetra-chloroethylene is emitted from dry-cleaned clothes (Moschandreas and

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2. Note that only 188 HAPs are now listed, caprolactam was removed.

Odea, 1995; Thompson and Evans, 1997; Evans et al., 2000); chloroform from chlorinated water (McKone, 1987; Wallace, 2001); formaldehyde from consumer products, building materials, and indoor chemical and surface reactions with ozone (Cooke, 1991; Weschler et al., 1992; Reiss et al., 1995b); 1,4-dichlorobenzene from air fresheners and moth cakes (Wallace, 1991); benzene, styrene, and 1,3-butadiene from cigarette smoke (Hodgson et al., 1996; Daisey et al., 1998; Singer et al., 2002); and BTEX compounds (benzene, toluene, ethylbenzene, and xylenes) from solvents and other craft and building supplies.

Indoor-outdoor relationships are explored by assessing differences between indoor and outdoor concentrations, often using ratios. One of the first studies to explore these relationships for VOCs was the EPA TEAM study conducted in the early 1980s (Wallace et al., 1985, 1988; Wallace, 1987). This study clearly established the importance of indoor exposures to VOCs. However, the EPA TEAM study is over 15 years old. More recent exposure data on a limited number of VOCs were collected in various states as part of the NHEXAS study (Lebowitz et al., 1995; Gordon et al., 1999; Robertson et al., 1999; Whitmore et al., 1999). A similar study was conducted in Elizabeth, New Jersey, Houston, Texas, and Los Angeles, California of a suite of VOCs and aldehydes (Weisel, 2002; Weisel et al., in preparation). In addition, studies have been conducted in Europe (Seifert et al., 1989, 2000; Jantunen et al., 1999; Edwards et al., 2001). A few small-scale studies have been carried out to assess indoor aldehyde concentrations in US homes (Zhang et al., 1994a; Reiss et al., 1995b).

However, many studies have been conducted to both model emission rates (Little et al., 1994; Cox et al., 2002; Zhao et al., 2002) or quantify emissions of VOCs from building materials using chamber studies (Tichenor et al., 1991; Bouhamra and Elkilani, 1999; Sparks et al., 1999a; Meininghaus et al., 2000; Elkilani et al., 2001; Won et al., 2001) or special test homes with controlled release of specific sources (Sparks et al., 1999b). However, there is still a need for updated information on indoor exposures to a wide range of VOCs in actual residential environments, including outdoor measurements and a measure of air exchange rate in order to better characterize indoor-outdoor relationships. Few studies have calculated source emission rates (SERS) to determine the contributions to personal exposures from indoor sources (Lewis and Zweidinger, 1992; Reiss et al., 1995b; Sarwar et al., 2002).

To address some of these needs, the Toxics Exposure Assessment Columbia-Harvard (TEACH) project was designed to characterize indoor and outdoor concentrations for 15 VOCs and 7 aldehydes, along with air exchange rates (AERs) in homes of inner-city teenagers in two distinct urban centers: New York City (NYC) and Los Angeles (LA) (Kinney et al., 2002). In each city, data were collected over two seasons. Compounds measured include the suspected

carcinogens benzene, 1,3-butadiene, styrene, tetrachloroethylene, trichloroethylene, 1,4 dichloro benzene, chloroform, methylene chloride, carbon tetrachloride, formaldehyde, and acetaldehyde. Additional compounds measured were toluene, ethylbenzene, *o*- and *m,p*-xylene, chosen because they are found in high concentrations in the environment; MTBE, chosen as a marker of mobile sources; and 1,1,1-trichloroethane and a suite of five additional aldehydes, chosen because little information exists on indoor concentrations of these compounds.

This paper reports home outdoor and indoor concentrations of the measured VOCs. Measurements were conducted in about 40 homes in each of the two cities across two seasons. Indoor-outdoor relationships as well as SERs were calculated for each home and sources of variability in the data were examined. Between homes, variability may be due to differences in housing characteristics, building materials, use and storage of household products, and AERs. Between cities, variability can be associated with differences in ambient emission sources and meteorological patterns. Also, seasonal variability within each city can be due to different meteorological patterns in different seasons, which in turn affect AER, environmental chemistry, emission rates, and environmental dispersion rates. By determining the variability in both indoor-outdoor relationships and SERs, we can gain a better understanding of indoor contributions to human exposures. The degree of uncertainty associated with measurement error was also calculated for the estimated emission rates and this uncertainty was compared to the inherent variability. We discuss the implication of this uncertainty on predicting emission rates of VOCs in homes.

Methods

Study Design

Nonsmoking participants (ages 14–19 years) were recruited from the A. Philip Randolph High School located in Harlem in NYC and from Jefferson High School in South Central, LA. Indoor and outdoor VOC measurements were taken at each participant's nonsmoking homes for one 48-h period in either or both of two seasons. A total of 46 homes were sampled in NYC, 38 in the winter and 41 in the summer. In all, 33 of the homes were monitored in both seasons. In LA, 41 homes were sampled, 40 in the winter and 35 in the fall. A total of 34 homes were sampled in both winter and fall in LA. The majority of the NYC homes were located in the upper Manhattan and the Bronx (>80%) and the rest in Brooklyn and Queens. In LA, all homes were located in South Central LA. In both cities, the teenagers lived in inner-city neighborhoods and represent an understudied population.

A home characteristics and inspection form was completed by a study technician for each home, including data on home volume. In addition, air exchange, temperature, and humidity measurements were taken in each home coinciding with VOC sampling.

Sampling was conducted in two seasons to explore the potential seasonal effects on concentrations of air pollutants in each city. In NYC, winter (February–April 1999) and summer (June–August 1999) were chosen to maximize the temperature differences. In LA, sampling was done in the winter (February–March 2000) and fall (September–October 2000) as higher VOC concentrations may be found in the fall due to a higher occurrence of thermal inversions in this season (AQMD, 1999; Holzworth, 1974).

Field Monitoring

In each city and season, 3–5 different homes were sampled simultaneously each week for one 48-h period (typically Tuesday through Thursday) over a period of 6–9 weeks. Indoor samplers were generally placed in the living room of each home, while outdoor samplers were set up through a window in the home using a custom-built PVC tube containing the sampling lines. An adjustable PVC spacer was placed securely in the opening of the window, to both hold the sample line tubes and to enable normal opening and closing of the window by residents. Outdoor samplers extending 0.6 m from the edge of the building and were shielded from wind and rain with stainless-steel buckets.

Two sampling systems, each containing three 7 l/min (LPM) pumps (Medo Inc.), timers, and time counters, were used in each home, one for indoor and one for outdoor sampling. One of the pumps was designated to sample VOCs and aldehydes using a three-way split flow design. Low-flow needle valves (SKC, Inc.) were used to control the flow through VOC tubes (ca. 1.8 sccm/min) and aldehyde samplers (ca. 100 sccm/min), and a vent line with a restriction tube was also used. The other pumps were used to collect duplicate samples and simultaneous PM_{2.5} samples.

Pumps were programmed using timers to allow for simultaneous sampling of indoor and outdoor air, and multiple homes. Counters were used to determine the elapsed time. The on and off flow rates were determined using two calibrated digital low-flow meters (Alltech).

Analytical Methods

Concentrations of target VOCs were determined using multisorbent “air toxics” samplers (Perkin-Elmer), stainless-steel tubes approximately 90 mm (3.5 in) long and 6.35 mm (0.25 in) in diameter containing 35 mm of Carbo-pack B (a medium-strength hydrophobic sorbent) and 10 mm of Carboxen 1000 (a strong sorbent, slightly hydrophilic). The sampling and analytical methods are described in US EPA’s Compendium Method TO-17 (Woolfenden and McClenny, 1997). A diffusion barrier

was placed upstream of the sampling tubes to prevent oversampling by diffusion, which may occur because of the low flow rates and the significant lag times between equipment setup and sampling start times. The diffusion barrier consisted of a small ID (0.5 mm) stainless-steel tube 200 mm in length. A similar diffusion barrier was tested and used by the European EXPOLIS team (Jantunen et al., 1999; Jurvelin et al., 2001). Analysis of VOC tubes was carried out using a Perkin-Elmer automatic thermal desorber (ATD), Model 400 connected to a Hewlett-Packard (HP) 5890II GC/5971 MSD.

Aldehydes were sampled using a C₁₈ silica cartridge coated with acidified 2,4-dinitrophenylhydrazine (DNPH). The coated samplers were obtained from ATMAA (Calabasas, CA, USA). The DNPH-derivatives (hydrazones) were eluted with acetonitrile and then analyzed using an HPLC (Hewlett Packard 1100) with a UV detector (360 nm).

The effects of ozone on aldehyde sampling with DNPH-coated cartridges have been a source of debate (Arnts and Tejada, 1989; Reiss and Roberts, 1997; Kleindienst et al., 1998). In NYC in the summer, to examine ozone interferences with aldehyde sampling, we used copper tubes (8-cm long) coated with a saturated solution of potassium iodide and dried with pure nitrogen gas, as an ozone scrubber. Co-located samples with and without the ozone scrubber did not show significant differences in concentrations of formaldehyde or the other aldehydes analyzed in this study. Consequently, the C₁₈ samplers were used without an ozone scrubber. This decision was also supported by several studies that compared C₁₈ samplers with silica-gel samplers (Fung and Wright, 1990; Zhou and Mopper, 1990; Sirju and Shepson, 1995).

AER was measured using the perfluorocarbon (PFT) technique (Dietz et al., 1986). The PFT technique is based on a continual release of tracer gas and diffusion samplers (capillary absorption tubes or CATs). The sources were placed in the subject’s home 24–72 h prior to placement of CATs, to allow PFT concentrations to equilibrate inside the home. Two to three CATs were placed per home, typically in the main living area and in the subject’s bedroom. AER measurements took place while air sampling was underway in each home. Analysis of CATs was performed following the procedure given by Dietz et al. (1986). It is worth noting that for apartments, air exchange measurements cannot distinguish between air entering from the outdoors and air entering from the corridor and other apartments in the building.

Quality Assurance

Field and laboratory blanks were collected, with each totaling at least 10% of the number of samples. Field blanks were transported and handled like regular samples, but were not attached to pumps. Field blanks were used to determine background contamination and for calculation of method limits of detection (LODs), calculated as the standard

deviation of the field blank concentration multiplied by the Student's *t*-value at the 99% confidence level. Instrument detection limits were also calculated based on the standard deviation of multiple injections of the lowest calibration standard multiplied by the Student's *t*-value at the 99% confidence level. Whichever value was highest was used as the LOD. Precision of the method was determined using duplicate samples and was equal to the mean relative difference (RD), calculated by taking the absolute difference of a pair of duplicates divided by the mean of the pair. For most compounds, the mean RD was below 25%, indicating reasonable method precision. LOD and precision are shown in Table 1.

VOC and aldehyde breakthrough was tested by connecting two sorbent tubes in series. A minimum of 10% of the samples had breakthrough tubes attached. All breakthrough tubes had levels of VOCs that were consistent with the field blank levels. All field and analytical logs were examined for completeness and unusual events; suspect sampling or analytical data were flagged. Samples that were subject to

equipment or analytical problems were excluded from data analysis.

Data Analysis

All concentrations were blank corrected using the mean blank value from field blanks, with resulting negative values set to zero. All values were included in the analysis, even if the concentration fell below the method detection limit. Concentrations of VOCs were non-normally distributed for most compounds; thus, nonparametric statistical methods were used when possible. Indoor-outdoor ratios (I/O ratios) were calculated for each home and the effects of air exchange rate on the I/O ratios was assessed by dichotomizing the data into high and low AER, at approximately the median value for both LA and NYC (1.1 h⁻¹). Wilcoxon rank sum tests were used to determine statistically significant differences in the I/O ratios between the two AER groups.

Effective SERs were estimated by assuming a single well-mixed compartment where

$$S_{in} = C_{in}(k + R)V - RVC_{out} \quad (1)$$

Table 1. LOD values and precision estimates for VOCs and aldehydes

Compounds	New York				Los Angeles			
	LOD (µg/m ³)		RD ^a – both seasons		LOD (µg/m ³)		RD – both seasons	
	Winter	Summer	N pairs	Mean	Winter	Fall	N pairs	Mean
<i>Indoor</i>								
Chloroform	0.11	0.11	17	0.33	0.11	0.11	21	0.18
1,4-Dichlorobenzene	1.43	1.43	26	0.29	1.43	1.43	28	0.20
Formaldehyde	1.10^b	0.34	43	0.08	0.52	1.16	42	0.12
Acetaldehyde	1.54	0.64	43	0.05	0.35	0.98	42	0.08
Benzaldehyde	0.71	0.72	39	0.21	0.32	0.89	34	0.37
Hexaldehyde	0.46	0.88	42	0.18	0.46	0.46	37	0.26
<i>Indoor+Outdoor</i>								
Valeraldehyde	0.29	0.80	43	0.36	0.29	0.29	40	0.36
Propionaldehyde	0.29	0.33	43	0.15	0.29	0.29	42	0.16
Styrene	0.41	0.41	26	0.20	0.41	0.41	28	0.23
1,3-Butadiene	0.06	0.06	5	0.51	0.06	0.06	6	0.42
Methylene chloride	0.22	1.63	25	0.36	0.27	0.22	26	0.37
Toluene	1.76	0.28	26	0.20	2.54	3.10	30	0.19
<i>n</i> -Butyraldehyde	1.20	0.31	43	0.21	1.28	0.60	41	0.29
Trichloroethylene	0.13	0.13	16	0.43	0.13	0.13	17	0.19
<i>Outdoor</i>								
Benzene	1.47	1.69	26	0.25	1.09	0.83	29	0.24
Ethylbenzene	0.27	0.27	26	0.19	0.27	0.27	30	0.13
<i>o</i> -Xylene	0.33	0.33	26	0.18	0.33	0.33	29	0.15
<i>m,p</i> -Xylene	0.70	0.83	26	0.19	0.72	0.79	30	0.15
MTBE	0.12	0.12	26	0.24	0.12	0.12	30	0.15
Tetrachloroethylene	0.15	0.15	26	0.15	0.15	0.15	28	0.12
1,1,1-Trichloroethane	0.11	0.11	26	0.16	0.11	0.11	26	0.15
Carbon tetrachloride	0.09	0.09	26	0.19	0.09	0.09	27	0.25

^aRD—relative difference calculated as the absolute difference between a duplicate pair divided by the mean of the pair.

^bItalics/bold values indicate LOD based on field blank values, all others are based on the instrument detection limit.

where S_{in} is the effective indoor SER ($\mu\text{g}/\text{h}$), C_{in} is the indoor VOC concentration ($\mu\text{g}/\text{m}^3$), k is the reaction rate in air (h^{-1}), R is the air exchange rate (h^{-1}), V is the volume of the well-mixed compartment (m^3), and C_{out} is the outdoor VOC concentration ($\mu\text{g}/\text{m}^3$). Values of C_{in} , C_{out} , R , and V were measured in each home.

In most cases, the well-mixed compartment was the whole house. In some cases, the teenager's room was excluded because tracer gas (PFT) measurements in the room showed a large deviation from measurements in the rest of the home. The variability of tracer gas concentrations in each home was analyzed, and a coefficient of variation (CV) was calculated. A cutoff CV of 0.35 was established, below which we considered the well-mixed assumption to be valid, based on the measurement error of the method. In NYC, about 60% of the homes fit into this category. Whereas in LA, about 35% of the homes fit into this category. In cases where the teenager's room had a tracer gas concentration more than 20% different from the average, it was assumed that this room was closed off from the rest of the house, the room was excluded from the volume of the home and the AER recalculated. This was the case in 10% of the homes in NYC and about 40% of the homes in LA. Still, about 30% of the homes in NYC and LA were classified as not well-mixed due to the large variability in tracer gas concentrations across locations within the home. We explored the effect on the source emission terms if these homes were excluded from the analysis and found no significant changes for compounds with predominantly indoor sources, and only isolated shifts in the statistics for compounds with outdoor sources. These shifts appeared to be due to removal of outliers in the data, and are probably unrelated to the well-mixed status of the home.

Little information exists on the penetration efficiencies of VOCs. One study found penetration efficiencies equal to 1 (Lewis and Zweidinger, 1992). Carbon tetrachloride and MTBE, both compounds with no known indoor sources, were found to have I/O ratios close to or equal to 1, supporting the use of a penetration efficiency for VOCs equal to unity.

The reaction rates of individual VOCs with hydroxyl radicals (OH) were also included in the calculation of emission rates for completeness (Singh et al. 1979; Atkinson, 1989) using an estimated indoor OH concentrations of 3×10^4 molecules/ cm^3 (Sarwar et al., 2002). However, sensitivity analysis showed that this removal process had little effect on the modeled results. SER estimates for the most reactive compound, styrene, changed by about 10% for only a subset of homes.

The calculated SER effectively includes the gains and losses through sorption and desorption on indoor surfaces. The interaction of VOCs with interior surfaces is a complex process that is not fully understood. Materials in the home can serve as temporary sinks for most VOCs, and these

VOCs can then be re-emitted over time and contribute to indoor concentrations long after the original source is gone. Thus, even though the sink material may temporarily remove the VOC and reduce indoor concentrations, over a long term these VOCs will continue to contribute to indoor concentrations. The partitioning of VOCs to materials is dependent on the compound, the material, the velocity of air in the room, temperature, and humidity (Jorgensen and Bjorseth, 1999). Any of the processes may be occurring during the sampling period in the home and these will be reflected in the calculated SER. We also note that the calculated SERs reflect the 48-h period that was sampled.

Equation (1) can be rearranged to estimate the indoor concentration due solely to indoor source emissions:

$$C = S_{in} / (R + k) V \quad (2)$$

The percent contribution from indoor sources, can then be calculated as $(C/C_{in}) * 100$ for each home.

To determine the uncertainty of calculated SERs, a propagation of measurement error was conducted using Crystal Ball software. A total of 3000 Monte Carlo simulations were run to obtain distributions of SERs on 32 single-family homes in LA, 18 of which were sampled in both winter and fall, to yield a total of 50 samples. Only LA single-family homes were included in the analysis due to the added uncertainty in the AER measurements associated with apartment buildings, which comprise most of the NYC homes. Measurements taken on a home in both winter and fall were treated as independent measurements since most parameters except for volume differed across the two seasons. Normal distributions were defined for each parameter in Eq. (1), except for the rate constant, which was defined as a log-normal distribution. For AER, the mean RD of duplicate samples was used as the measure of uncertainty. For volume, we assumed a 10% CV because we did not have a precision estimate. Indoor and outdoor concentrations were assigned the precision estimates obtained from duplicate measurements, also calculated as the mean RD. The rate constant (κ) was assigned a CV of 1, which results in the 10th and 90th percentile varying by an order of magnitude. Distributions of SERs were obtained for each home and each compound. The CV for each home was chosen to describe the uncertainty associated with the calculated SER for each home. We also determined the 95th percent confidence that each home had a positive SER by assessing if the 5th percentile of the uncertainty distribution was a positive value.

Data from home questionnaires were examined to determine if indoor-outdoor relationships or emission rates were associated with specific housing characteristics or household product use. Median emission rates were compared across different housing characteristics and Wilcoxon rank sum tests were performed to determine statistical significance at a P -value of 0.05. Univariate regression analyses were also explored.

Results and discussion

Housing Characteristics

Housing characteristics were determined using a questionnaire administered during the setup of monitoring equipment. We were interested in assessing differences in heating and cooling characteristics, types of building materials, and consumer product use. In NYC, 78% of the homes were apartments, while in LA 68% of the homes were either attached or detached single-family homes. The average size and range of the homes were higher, on an average, in LA than in NYC (mean: 900 ft², range: 500–2500 in LA and mean: 775 ft², range: 450–1800 in NYC). In NYC most homes (63%) used fuel oil, while in LA either gas (51%) or electric heating (24%) were used. Few homes in both cities had central air conditioning and only in NYC, 65% of the homes had window AC units, which were typically not used. Gas was used for cooking in most homes, both in LA and NYC. The types of building materials used in each of the cities were fairly similar. The largest differences were in flooring. LA had notably more wall-to-wall carpeting (NYC: 30% and LA: 88%), and NYC has more wood flooring (NYC: 56% and LA: 18%). Attached garages were only found in approximately 20% of the homes in both cities. Renovations, mostly painting, were done in about half of the homes over the previous year in both cities. The use of moth repellents was generally low in both cities (<20%), but air fresheners were commonly used (NYC: 65%, and LA: 75%).

AER measurements in NYC and LA homes showed significant differences across cities and seasons (Figure 1). The mean AERs (\pm SD) in the winter in NYC were approximately half the AER in the summer (0.99 ± 0.69 vs. 1.81 ± 1.14). In LA, mean AERs were higher than in NYC, but with a similar seasonal pattern; lower winter AERs (1.36 ± 1.17) than fall AERs (2.45 ± 2.22).

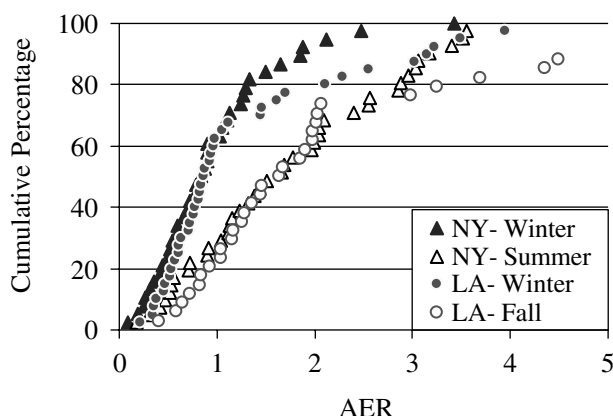


Figure 1. Cumulative distributions of AERs in NYC and LA homes.

Overview of VOC Concentrations

The results from the sampling campaigns are summarized in Tables 2 and 3 for outdoor and indoor home VOC concentrations, respectively. Compounds were divided into groups based on the I/O ratios and the calculated SERs shown in Table 4 to facilitate discussion. Compounds with predominantly indoor sources had the highest I/O ratios and SERs. These include chloroform, 1,4 dichlorobenzene, formaldehyde, acetaldehyde, benzaldehyde, and hexaldehyde. Compounds with dominant outdoor sources had median I/O ratios close to 1, and small SERs. These included benzene, ethylbenzene, xylenes, MTBE, tetrachloroethylene, trichloroethylene, and carbon tetrachloride. The rest of the VOCs fell in the middle category with contributions from both indoor and outdoor sources.

Compounds with Indoor Sources

Of the compounds with indoor sources, chloroform had the highest I/O ratios, with markedly higher ratios in NYC than in LA (Table 4), and also greater seasonal differences in NYC (15 in winter vs. 5 in summer). Figure 2 shows that in NYC I/O ratios of chloroform were almost twice as high for homes with a lower AER. Unlike the I/O ratios, the estimated SERs for chloroform showed little seasonal variability (Table 4). Thus, seasonal differences in chloroform levels are clearly

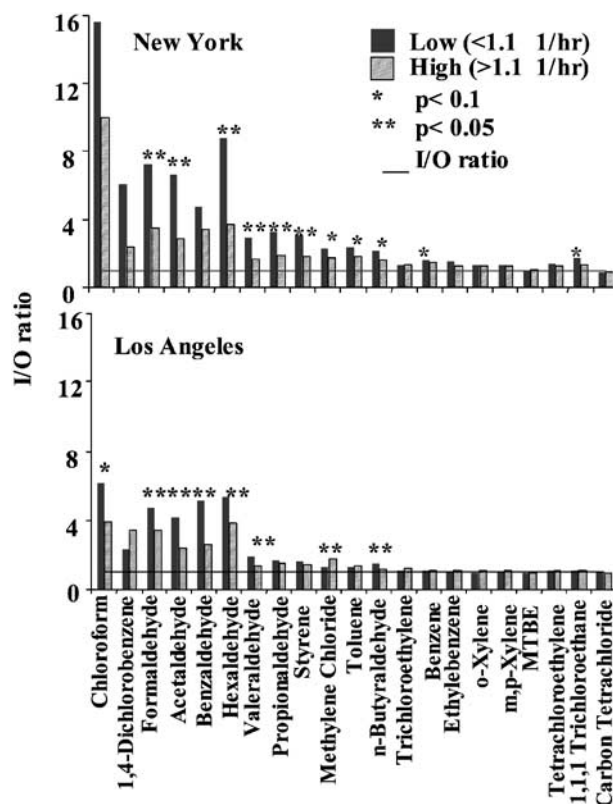


Figure 2. Effects of air exchange on I/O ratios in New York and Los Angeles.

Table 2. New York and Los Angeles outdoor VOC concentrations in two seasons

Compound	Outdoor concentrations ($\mu\text{g}/\text{m}^3$)																			
	New York, 1999									Los Angeles, 2000										
	Winter				Summer					W-S ^b	Winter				Fall					
	N (% > LOD ^a)	MED	Mean (SD)	Range	N (% > LOD)	MED	Mean (SD)	Range	N (% > LOD)		MED	Mean (SD)	Range	N (% > LOD)	MED	Mean (SD)	Range	W-F ^b		
<i>Indoor</i>																				
Chloroform	31 (75)	0.2	0.2 (0.2)	ND–1.0	27 (50)	ND	0.3 (0.7)	ND–3.3	**	35 (29)	0.1	0.1 (0.1)	ND–0.2	32 (22)	ND	0.1 (0.1)	ND–0.2			
1,4-Dichlorobenzene	31 (67)	1.8	4.2 (6.6)	0.5–34	26 (74)	1.9	4.1 (5.6)	0.2–27		35 (53)	1.4	2.0 (1.4)	0.5–6.1	32 (32)	1.7	3.5 (4.7)	0.4–22			
Formaldehyde	36 (89)	2.2	2.1 (0.9)	0.5–4.1	36 (100)	4.6	5.3 (2.3)	1.9–13	*	40 (100)	3.7	3.9 (1.3)	2.3–8.4	35 (100)	3.6	4.4 (1.6)	2.5–7.8			
Acetaldehyde	36 (100)	2.7	2.8 (0.9)	1.5–5.4	36 (100)	4.1	4.2 (1.5)	1.4–10	*	40 (100)	3.5	3.7 (1.2)	1.8–7.4	35 (100)	3.6	4.1 (1.6)	2.3–8.5			
Benzaldehyde	35 (14)	0.1	0.4 (0.7)	ND–3.6	36 (39)	0.6	0.8 (0.6)	0.2–3.2	*	40 (60)	0.4	0.5 (0.3)	ND–1.4	35 (29)	0.4	0.6 (0.6)	ND–1.9			
Hexaldehyde	35 (91)	1.1	1.1 (0.5)	0.1–2.6	36 (83)	1.5	1.7 (1.3)	ND–7	*	40 (98)	1.5	1.7 (1.0)	ND–5.5	35 (100)	1.5	1.5 (0.4)	0.9–2.5			
<i>Indoor+outdoor</i>																				
Valeraldehyde	35 (91)	0.7	0.8 (0.5)	0.1–3.0	36 (83)	1.7	1.8 (1.0)	0.2–5.7	*	40 (93)	1.2	1.3 (1.1)	ND–5.2	34 (100)	1.7	2.2 (1.4)	0.8–7.0	*		
Propionaldehyde	35 (97)	0.9	1.2 (1.1)	0.1–7.4	36 (94)	0.7	0.8 (0.4)	0.3–2.3	*	40 (100)	1.1	1.2 (0.5)	0.6–3.1	35 (100)	1.1	1.3 (0.6)	0.6–3.6			
Styrene	31 (42)	0.3	0.4 (0.2)	0.1–0.9	27 (4)	0.3	0.2 (0.1)	ND–0.5	*	35 (82)	0.6	0.7 (0.4)	0.2–1.8	32 (69)	0.6	0.6 (0.3)	0.2–1.3			
1,3-Butadiene	31 (14)	ND	0.1 (0.2)	ND–0.7	27 (11)	ND	0.1 (0.4)	ND–2.0		35 (24)	ND	0.2 (0.4)	ND–1.7	32 (3)	ND	0.01	ND–0.3	*		
Methylene chloride	31 (94)	1.0	1.9 (3.5)	0.4–19	27 (14)	0.5	1.0 (1.4)	ND–6.6	*	35 (97)	1.2	1.5 (1.3)	0.2–7.3	32 (78)	0.4	0.6 (0.6)	ND–1.9	*		
Toluene	31 (94)	5.5	5.7 (2.7)	1.4–10	27 (100)	6.2	6.9 (3.5)	1.5–14		35 (100)	14	16 (9.4)	6.0–55	32 (100)	9.9	11 (3.8)	4.3–19	*		
n-Butyraldehyde	35 (57)	1.3	1.3 (0.6)	0.2–3.1	36 (75)	0.8	0.7 (0.5)	ND–2.1	*	40 (25)	1.0	1.3 (1.0)	0.6–6.2	35 (97)	1.2	1.3 (0.4)	ND–2.3			
Trichloroethylene	31 (78)	0.3	0.3 (0.2)	ND–1.0	27 (32)	ND	0.1 (0.2)	ND–0.7	*	35 (47)	0.1	0.1 (0.1)	ND–0.4	32 (31)	0.1	0.1 (0.2)	ND–0.8			
<i>Outdoor</i>																				
Benzene	31 (72)	2.4	2.4 (1.2)	ND–4.9	27 (25)	0.8	1.1 (1.1)	0.2–4.5	*	35 (100)	4.3	4.3 (1.7)	2.2–9.0	32 (97)	2.2	2.3 (0.8)	0.8–4.3	*		
Ethylbenzene	31 (100)	1.0	1.1 (0.5)	0.3–2.0	26 (100)	1.3	1.7 (1.6)	0.5–8.9	**	35 (100)	2.7	2.9 (1.4)	1.0–7.3	32 (100)	2.1	2.1 (0.7)	0.9–3.4	*		
<i>o</i> -Xylene	31 (100)	1.1	1.3 (0.6)	0.4–2.4	26 (96)	1.5	1.8 (1.7)	0.4–9.5		35 (100)	3.8	3.9 (1.9)	1.2–9.7	32 (100)	2.9	2.8 (0.9)	1.2–4.5	*		
<i>m,p</i> -Xylene	31 (100)	3.3	3.7 (1.7)	1.1–6.8	27 (96)	4.0	5.0 (5.4)	ND–30		35 (100)	10	11 (5.1)	3.2–26	32 (100)	7.9	7.8 (2.5)	3.4–13	*		
MTBE	31 (100)	10	11 (5.3)	2.9–24	27 (93)	10.9	12.7 (14)	1.5–73		35 (100)	16	19 (8.7)	7.9–45	31 (100)	13	13 (4.8)	4.7–22	*		
Tetrachloroethylene	31 (100)	1.4	2.2 (1.8)	0.7–7.8	27 (79)	1.4	9.2 (32)	ND–167		35 (100)	1.7	1.9 (0.7)	1.0–3.7	32 (100)	1.1	1.4 (0.6)	0.7–2.9	*		
1,1,1-Trichloroethane	31 (100)	0.4	0.5 (0.3)	0.3–1.7	27 (86)	0.3	0.6 (1.7)	ND–9.1	*	35 (100)	0.4	0.4 (0.1)	0.3–0.5	32 (100)	0.3	0.3 (0.1)	0.3–0.6	*		
Carbon tetrachloride	31 (100)	0.6	0.7 (0.1)	0.5–1.0	27 (86)	0.5	0.5 (0.2)	ND–0.8	*	35 (100)	0.5	0.5 (0.1)	0.4–0.7	32 (100)	0.6	0.6 (0.1)	0.2–0.7	*		

^aLOD values are given in Table 1.

ND – nondetected level, no analyte detected.

^bWilcoxon rank sum test between seasons, differences are considered significant if $P < 0.05$ (*) or $P < 0.1$ (**).

Table 3. New York and Los Angeles indoor VOC concentrations in two seasons

Compound	Indoor concentrations ($\mu\text{g}/\text{m}^3$)																		
	New York, 1999									Los Angeles, 2000									
	Winter				Summer					<i>W-S</i> ^a	Winter				Fall				
	<i>N</i> (% > LOD)	MED	Mean (SD)	Range	<i>N</i> (% > LOD)	MED	Mean (SD)	Range	<i>N</i> (% > LOD)		MED	Mean (SD)	Range	<i>N</i> (% > LOD)	MED	Mean (SD)	Range	<i>W-F</i> ^a	
<i>Indoor</i>																			
Chloroform	36 (100)	2.6	3.4 (2.8)	0.5–13	30 (100)	1.7	2.3 (1.8)	0.5–9.2	*	40 (88)	0.5	0.8 (0.9)	ND–4.2	32 (88)	0.3	0.4 (0.3)	ND–1.5	*	
1,4-Dichlorobenzene	36 (92)	8.9	49 (93)	0.4–390	30 (97)	6.1	111 (489)	1.2–2684		40 (85)	4.7	40 (71)	0.7–245	32 (78)	3.2	52 (109)	0.7–423		
Formaldehyde	37 (100)	12	12 (4.7)	5.2–22	41 (100)	19	21 (11.0)	5.8–51	*	40 (100)	18	21 (11)	7.9–59	33 (100)	15	16 (6.2)	8.2–32		
Acetaldehyde	37 (100)	14	16 (9.5)	5.4–54	41 (100)	11	15 (16.7)	2.9–92	*	40 (100)	15	15 (7.2)	4.1–36	33 (100)	8.6	9.6 (4.2)	3.5–23	*	
Benzaldehyde	37 (100)	2.5	3.5 (4.3)	0.5–26	41 (100)	1.5	2.3 (2.8)	0.6–14		40 (98)	1.9	2.1 (1.2)	ND–6.6	33 (91)	1.7	1.9 (1.0)	0.6–4.4		
Hexaldehyde	37 (100)	2.2	3.0 (2.6)	0.6–14	41 (100)	2.9	3.8 (3.9)	ND–23		40 (100)	7.9	10 (7.2)	2.5–35	33 (100)	5.9	6.4 (2.8)	1.8–15	**	
<i>Indoor+outdoor</i>																			
Valeraldehyde	37 (100)	8.0	11 (13.6)	2.2–82	41 (100)	5.8	9.7 (12.5)	1.6–60		40 (100)	2.4	3.1 (2.5)	0.3–10	33 (100)	2.6	2.8 (1.5)	0.6–6.8		
Propionaldehyde	37 (100)	2.1	2.2 (1.2)	0.5–6.3	41 (100)	1.3	1.7 (1.7)	ND–10	*	40 (100)	2.1	2.4 (1.4)	0.6–6.6	33 (100)	1.7	2.0 (0.9)	0.9–4.9		
Styrene	36 (94)	1.0	1.1 (0.6)	0.2–3.0	30 (78)	0.5	0.8 (0.7)	0.2–3.2	**	40 (100)	1.0	1.3 (0.8)	0.4–4.1	32 (88)	0.8	0.9 (0.5)	0.3–2.0	*	
1,3-Butadiene	36 (64)	0.7	1.0 (1.4)	ND–5.8	30 (44)	ND	1.2 (2.6)	ND–12		40 (60)	0.5	0.5 (0.6)	ND–1.8	32 (38)	ND	0.2 (0.3)	ND–1.5	*	
Methylene chloride	36 (97)	2.2	5.5 (12.3)	0.2–69	30 (28)	1.4	10 (32.9)	ND–176	*	40 (95)	1.9	2.4 (2.0)	ND–8.7	32 (100)	1.1	1.4 (1.2)	0.2–4.3	*	
Toluene	36 (100)	12	16 (12.8)	3.0–69	30 (91)	10	15 (18.9)	ND–93		40 (100)	17	24 (17)	4.6–69	32 (100)	13	15 (12)	4.3–68	*	
n-Butyraldehyde	37 (81)	2.1	2.4 (2.6)	0.3–14	41 (100)	2.2	2.7 (1.8)	0.8–9.2		40 (68)	1.6	2.0 (1.3)	ND–5.9	33 (100)	1.4	1.6 (0.7)	0.7–3.8		
Trichloroethylene	36 (92)	0.4	1.1 (3.2)	ND–19	30 (44)	0.1	0.3 (0.5)	ND–2.6	*	40 (68)	0.2	0.2 (0.3)	ND–1.2	32 (47)	0.1	0.2 (0.2)	ND–0.8		
<i>Outdoor</i>																			
Benzene	36 (100)	3.6	5.3 (6.5)	1.7–39	30 (38)	1.5	1.7 (1.2)	ND–6.3	*	40 (100)	4.3	4.9 (2.8)	1.6–17	32 (100)	2.3	2.5 (1.3)	1.0–6.3	*	
Ethylbenzene	36 (100)	1.6	3.2 (6.1)	0.5–34	30 (100)	1.6	1.8 (1.0)	0.4–5.1		40 (100)	2.8	3.0 (1.5)	1.4–7.5	32 (100)	1.8	2.5 (2.6)	0.8–15	*	
<i>o</i> -Xylene	36 (100)	1.7	3.0 (5.4)	0.6–31	30 (100)	1.7	2.0 (1.2)	0.4–6.5		40 (100)	3.7	4.1 (2.0)	1.8–10	32 (100)	2.3	3.1 (2.3)	1.1–12	*	
<i>m,p</i> -Xylene	36 (100)	5.1	9.2 (17.8)	1.8–106	30 (100)	4.9	5.7 (3.7)	1.1–20		40 (100)	10	11 (5.3)	4.9–27	32 (100)	6.9	9.4 (10.5)	3.1–62	*	
MTBE	36 (100)	11	19 (28.9)	3.8–170	30 (100)	14	23 (51.5)	3.2–293		40 (100)	14	18 (9.8)	7.7–47	32 (100)	11	13 (10)	4.6–61	*	
Tetrachloroethylene	36 (100)	3.5	6.7 (13.1)	0.8–78	30 (78)	2.0	5.3 (8.7)	ND–43		40 (100)	1.9	2.3 (1.6)	0.7–11	32 (100)	1.3	1.8 (1.4)	0.6–6.8	*	
1,1,1-Trichloroethane	36 (100)	0.6	4.8 (20.2)	0.3–122	30 (81)	0.5	1.3 (3.1)	ND–16	**	40 (100)	0.4	0.6 (0.6)	0.2–2.8	32 (100)	0.4	0.5 (0.4)	0.2–2.7		
Carbon tetrachloride	36 (100)	0.6	0.7 (0.1)	0.5–1.2	30 (97)	0.5	0.5 (0.1)	ND–0.8	*	40 (100)	0.5	0.5 (0.1)	0.3–1.0	32 (100)	0.6	0.6 (0.1)	0.5–1.0	*	

LOD values are given in Table 1; ND – nondetected level, no analyte detected.

^aWilcoxon rank sum test between seasons, differences are considered significant if $P < 0.05$ (*) or $P < 0.1$ (**).

Table 4. I/O ratios and effective SERs (mg/h) for New York and Los Angeles homes

Compound	New York										Los Angeles													
	Winter					Summer					Winter					Fall								
	I/O		Emission rate			I/O		Emission rate			W-S comp ^c		I/O		Emission rate			I/O		Emission rate			W-F comp ^c	
	MED ^a (N)	CV ^b	MED (N)	CV		MED (N)	CV	MED (N)	CV	I/O	ER	MED (N)	CV	MED (N)	CV	MED (N)	CV	MED (N)	CV	I/O	ER	I/O	ER	
<i>Indoor</i>																								
Chloroform	15 (30)	0.8	0.3 (30)	1.0	5.3 (13)	1.6	0.4 (17)	0.6	*		5.1 (26)	0.8	0.1 (35)	1.2	3.4 (14)	0.7	0.1 (29)	0.8						
1,4-Dichlorobenzene	6.1 (35)	1.1	1.6 (30)	1.7	2.3 (24)	3.2	1.4 (16)	2.4	**		3.4 (38)	2.0	0.6 (35)	1.9	2.0 (30)	1.9	0.5 (29)	2.1						
Formaldehyde	6.6 (35)	0.5	1.4 (34)	0.7	3.4 (36)	0.7	3.4 (33)	0.7	*	*	4.5 (40)	0.6	2.7 (40)	1.2	3.6 (32)	0.3	4.0 (32)	0.6	*	*				
Acetaldehyde	5.2 (35)	0.6	1.7 (34)	0.8	2.6 (36)	1.1	1.6 (33)	0.9	*		3.9 (40)	0.7	1.9 (40)	0.7	2.4 (32)	0.3	1.8 (32)	0.5	*	*				
Benzaldehyde	4.7 (21)	3.0	0.2 (33)	1.2	3.4 (36)	1.0	0.3 (33)	0.8			4.9 (39)	0.9	0.3 (40)	0.8	2.5 (24)	0.9	0.5 (32)	0.5	*	*				
Hexaldehyde	7.3 (34)	1.3	1.0 (33)	0.9	3.4 (35)	1.1	1.1 (33)	2.2	*		5.0 (39)	0.9	1.3 (40)	1.2	4.0 (32)	0.4	1.5 (32)	0.6						
<i>Indoor+outdoor</i>																								
Valeraldehyde	3.1 (34)	1.8	0.2 (33)	0.9	1.8 (36)	1.3	0.2 (33)	1.9	*		1.6 (39)	1.5	0.1 (40)	1.6	1.4 (31)	0.4	0.3 (31)	0.9						
Propionaldehyde	2.6 (34)	0.9	0.2 (33)	0.9	2.0 (36)	1.6	0.2 (33)	2.5			1.7 (40)	0.7	0.2 (40)	1.5	1.5 (32)	0.3	0.3 (32)	1.0						
Styrene	2.7 (35)	0.7	0.1 (30)	1.2	1.9 (21)	0.7	0.1 (17)	0.7			1.6 (38)	0.5	0.1 (35)	0.9	1.4 (30)	1.0	0.1 (29)	1.6						
Methylene chloride	1.9 (33)	2.2	0.1 (30)	3.1	2.2 (22)	2.5	0.1 (17)	2.2			1.4 (38)	2.0	0.1 (35)	1.8	1.7 (25)	0.7	0.1 (29)	1.4						
Toluene	2.3 (35)	1.2	1.0 (30)	1.1	1.6 (25)	1.9	1.4 (17)	1.9			1.3 (38)	0.7	1.2 (35)	1.6	1.3 (29)	1.3	0.7 (29)	2.1						
<i>n</i> -Butyraldehyde	1.7 (34)	0.6	0.1 (33)	1.1	1.7 (31)	3.8	0.1 (33)	1.3			1.5 (40)	0.7	0.1 (40)	2.0	1.3 (32)	0.3	0.1 (32)	1.0						
Trichloroethylene	1.5 (30)	3.2	0.01 (30)	4.4	0.9 (9)	0.5	— (17)	5.6	*	*	1.2 (29)	1.0	0.01 (35)	1.8	1.2 (18)	0.9	0.01 (29)	1.5						
<i>Outdoor</i>																								
Benzene	1.7 (33)	1.9	0.2 (30)	1.3	1.1 (19)	0.7	0.1 (17)	2.4	*	*	1.1 (38)	0.5	0.1 (35)	2.4	1.1 (29)	0.4	0.1 (29)	2.1						
Ethylbenzene	1.4 (35)	1.5	0.1 (30)	3.7	1.2 (24)	1.2	0.1 (16)	3.1			1.1 (38)	0.3	0.04 (35)	1.6	1.0 (29)	0.6	0.04 (29)	4.0						
<i>o</i> -Xylene	1.3 (35)	1.2	0.1 (30)	3.8	1.2 (24)	1.6	0.04 (16)	3.7			1.0 (38)	0.3	0.03 (35)	1.5	1.0 (29)	0.5	0.03 (29)	3.4						
<i>m,p</i> -Xylene	1.3 (35)	1.4	0.3 (30)	3.9	1.3 (24)	1.6	0.2 (17)	3.4			1.1 (38)	0.3	0.1 (35)	1.4	1.0 (29)	0.7	0.1 (29)	4.2						
MTBE	1.0 (35)	1.1	0.03 (30)	4.9	1.1 (23)	0.5	0.2 (17)	2.5		*	1.0 (38)	0.2	— (35)	2.1	1.0 (29)	0.5	— (28)	2.5						
Tetrachloroethylene	1.5 (35)	1.8	0.2 (30)	1.9	1.1 (22)	0.9	0.02 (17)	4.2	*	*	1.1 (38)	1.1	0.02 (35)	2.2	1.0 (29)	0.8	0.02 (29)	2.0						
1,1,1-Trichloroethane	1.5 (35)	4.3	0.02 (30)	4.2	1.4 (23)	1.3	0.01 (17)	2.3			1.1 (38)	0.9	0.003 (35)	2.7	1.0 (29)	0.6	0.003 (29)	2.4						
Carbon tetrachloride	0.9 (35)	0.4	— ^d (30)	2.2	0.9 (22)	0.3	— (17)	7.6			1.0 (38)	0.3	— (35)	2.2	1.0 (29)	0.4	— (29)	2.7						

^aMedian I/O ratio or emission rate.^bCoefficient of variation calculated as the standard deviation of the I/O ratio or emission rate divided by the mean of the ratio or ER.^cWilcoxon rank sum test between seasons, differences are considered significant if $P < 0.05$ (*) or $P < 0.1$ (**).^dA dash (—) indicates that the emission rate was zero or negative.

explained by the differences in AERs, with high I/O ratios being a function of low AER in the winter. By accounting for AER in the calculation of SER, the differences are attenuated.

Chloroform SERs in NYC were higher than in LA. Indoor sources of chloroform have been shown to be associated with use of chlorinated water in the home, indicating that possible differences in drinking water treatment (such as chlorination, filtration to reduce organic matter, and ozonation) or water usage may account for the differences in SER between the two cities (Principe et al., 2000; City of Los Angeles Water Services, 2002). Figure 3a shows the cumulative distributions of chloroform in NYC and LA homes, depicting clearly the between city variability in the SER. The percent contribution from outdoor air to

indoor concentrations was calculated using the estimated SERs, and median percents are shown in Figure 4. For chloroform, over 90% of the indoor concentrations come from indoor sources.

1,4-Dichlorobenzene also had significant contributions from indoor sources. The percent contribution to indoor concentrations, however, was more variable than chloroform (Figures 3b and 4). There was a large variability in indoor concentrations of 1,4-dichlorobenzene across homes, with some homes that had concentrations up to 3 mg/m³. Consequently, I/O ratios and SER were equally variable with typical CVs near 2. Potential indoor sources of 1,4-dichlorobenzene include the use of products such as moth cakes and air fresheners (Wallace, 1991). Thus, the observed variability may be due to the presence of products containing this compound in some homes and not in others. I/O ratios were higher in the winter, but only slightly significant in NYC ($P < 0.1$) as shown in Table 4. When comparing the I/O ratios for high and low AERs as shown in Figure 2, differences were not significant in both cities; however, in NYC higher I/O ratios were associated with low AER and in LA higher I/O ratios were associated with higher AER. SERs were somewhat higher in the winter in LA, but not significantly.

Both univariate regression analysis and Wilcoxon rank tests failed to show a significant correlation between use of these products and indoor concentrations or SERs of 1,4-dichlorobenzene. This may be a function of the small sample size in some of the comparison groups, or the failure to properly identify the sources of 1,4-dichlorobenzene with the survey instrument used in this study. Also, it is worth noting that for homes with the highest 1,4-dichlorobenzene, high outdoor levels were also present. This may be an indication that we failed to sample effectively outside of the building envelope and that we may have "leakage" of indoor concentrations to the outdoors. For these cases, we would be over-representing the outdoor contributions and reducing the I/O ratios and SERs.

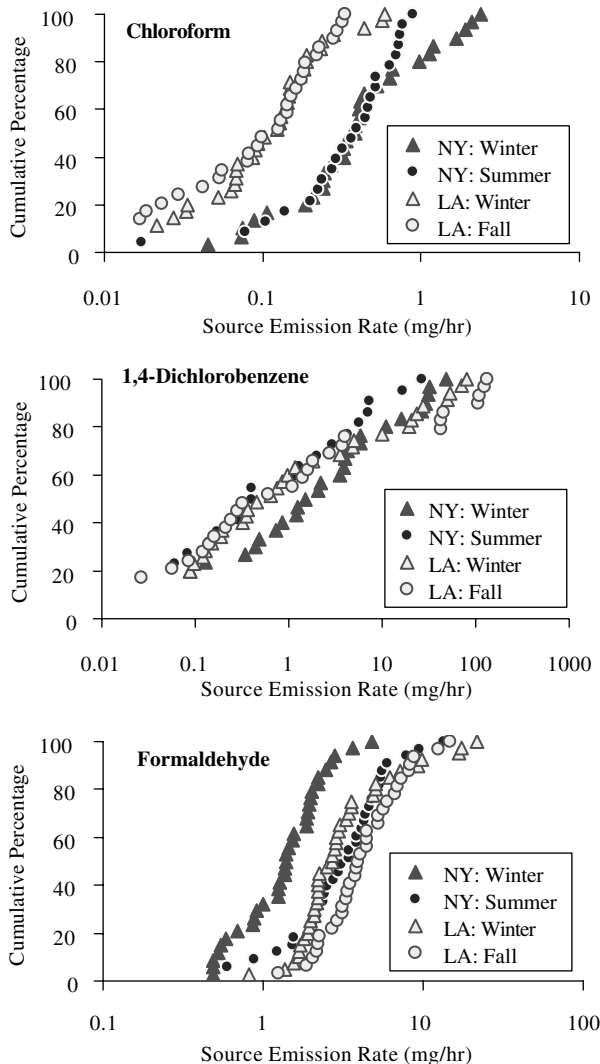


Figure 3. Cumulative distributions of SERs of chloroform, 1,4-dichlorobenzene, and formaldehyde in NYC and LA.

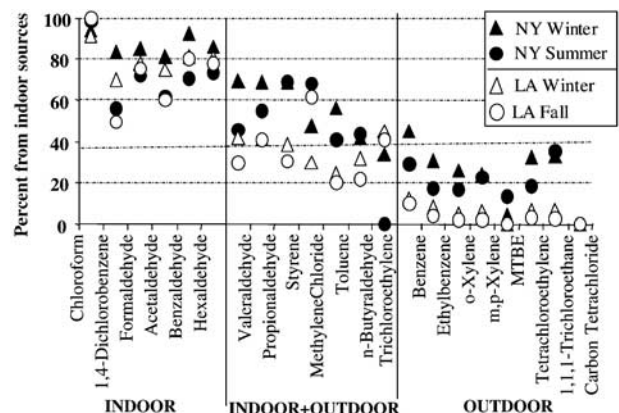


Figure 4. Percent contributions from indoor sources for NYC and LA homes.

Another group of compounds with important indoor sources were some of the aldehydes, namely formaldehyde, acetaldehyde, hexaldehyde, and benzaldehyde. Aldehydes, in particular formaldehyde, can be formed from reactions of ozone with various surfaces, such as walls painted with latex paint (Reiss et al., 1995a) and carpets (Weschler et al., 1992; Morrison and Nazaroff, 2002a), or with reactions with various unsaturated hydrocarbons in air (Grosjean and Grosjean, 1996, 1997; Weschler and Shields, 1997; Fan et al., 2003). Additionally, there are direct emissions from cabinetry, doors, plywood subfloor, and particleboard (Brown, 1999).

The contributions to indoor concentrations from indoor sources of these aldehydes (Figure 4) varied significantly across seasons, and more so for NYC. The cumulative distributions shown in Figure 3c for formaldehyde also illustrate this point. This can be explained by the fact that ambient levels in NYC were significantly higher in the summer than in the winter, likely driven by greater photochemistry in the summer. In LA, this seasonal variability in ambient concentrations was less. Indoor concentrations were also significantly different across seasons for formaldehyde in NYC (summer > winter), for acetaldehyde in both cities (winter > summer), and marginally for hexaldehyde in LA (winter > summer). I/O ratios were always greater in the winter than in the summer, but only significantly for formaldehyde and acetaldehyde in both NYC and LA, for hexaldehyde in NYC, and for benzaldehyde in LA. The effects of AER on the I/O ratios can be seen in Figure 2, where significantly higher ratios were associated with low AERs for most of these aldehydes. Accounting for AER in the estimated SERs, a different seasonal pattern emerges, where SERs are higher in the summer than in the winter, statistically significant for formaldehyde in both cities and benzaldehyde in LA only (Table 4).

Past studies have shown that emission rates of formaldehyde increase with higher ozone concentrations, temperature, and relative humidity (Hawthorne and Matthews, 1987; Reiss et al. 1995b). On an average, both cities had higher ambient ozone concentrations and a greater number of exceedence days in summer than winter (NYC, 25 vs. 14 p.p.b.; LA, 18 vs. 12 p.p.b; EPA Airs data), indicating that there were higher peak concentrations associated with summertime. Reiss et al. (1995b) sampled a limited number of homes in Boston MA in winter and summer of 1993, and used a similar steady-state model to calculate SERs. He found that higher indoor emission rates for aldehydes in the summer were consistent with higher concentrations of reactants such as ozone indoors. Zhang et al. (1994a) found similar results in his study of six homes in New Jersey.

Compounds with Predominantly Outdoor Sources

The compounds that were considered to be predominantly from outdoor sources include benzene, ethylbenzene, xylenes,

MTBE, tetrachloroethylene, 1,1,1-trichloroethane and carbon tetrachloride. Mobile sources are the primary ambient source of benzene, ethylbenzene, xylenes, and MTBE. In LA, all of these compounds had significantly higher indoor and outdoor concentrations in the winter, but in NY there was little seasonal variability except for benzene (Tables 2 and 3). There was a clear city-to-city difference as well, with LA median indoor and outdoor concentrations of most compounds a factor of 2 higher than in NYC. Higher concentrations in LA may be associated with greater emissions of mobile sources (AQMD, 1999). Median I/O ratios were close to unity for these compounds, indicating the influence of outdoor sources, similarly SER were low with medians close to 0 especially in LA. Previous studies have found higher I/O ratios for homes with garages. Approximately 20% of our LA homes had attached garages, but no correlations were found between a garage and indoor levels or SERs of benzene or other BTEX compounds. I/O ratios were slightly higher in NYC when compared to LA, indicating potential presence of indoor sources in NYC. Univariate and Wilcoxon rank sum analysis showed significant correlations with indoor concentrations of ethylbenzene and xylenes associated with repairs in the last year in NYC only.

Carbon tetrachloride, tetrachloroethylene, and 1,1,1-trichloroethane also had low or no indoor sources, particularly in LA. Carbon tetrachloride is a compound that was banned from the residential and commercial markets. Its chemical stability, however, makes it very persistent in the environment and probably explains the consistent concentrations across cities and seasons.

In LA and NYC, I/O ratios for tetrachloroethylene and 1,1,1-trichloroethane, solvents typically used for dry cleaning, were close to 1 in both seasons, indicating that few homes had significant indoor sources of these pollutants. Other studies have shown I/O ratios greater than 1 for both of these compounds (Brown et al., 1994). The high CVs do suggest that some homes had significant sources. Also, it should be noted that tetrachloroethylene particularly in NYC in the winter had higher I/O ratios and source emissions. This may indicate the presence of sources either within the apartments NYC or in the building. The reductions in 1,1,1-trichloroethane are consistent with the EPA's CAA mandated reductions of this chemical.

Compounds with Indoor and Outdoor Sources

Among the BTEX compounds, toluene had the highest I/O ratios. Seasonal patterns in I/O ratios and SER of toluene differed between the two cities. In LA, I/O ratios of toluene were similar in both seasons and did not vary by AER, whereas in NYC significant differences were related with AER. On the other hand, SERs were consistent across seasons in NYC, but were lower in the fall in LA, although

not statistically different. In NYC, univariate analysis showed significantly higher indoor concentrations and SERs associated with home renovations conducted in the past year.

While 1,3-butadiene and styrene had low detection rates outdoors, styrene was detectable in most homes and 1,3-butadiene in about half of the homes suggesting indoor sources. Environmental tobacco smoke (ETS) is a potential source of styrene, 1,3-butadiene and benzene. Although our homes were all nonsmoking (self-reported), visitors or possible intrusion of ETS from other apartments could explain the elevated SERs for these compounds in NYC apartments. In these cases, the air exchange may have come, in part, from within the apartment building. To illustrate this point, SERs were calculated for apartments and compared to SERs calculated for single-family homes. The mean SERs for apartments were 34%, 18%, and 17% higher than for single-family dwellings for benzene, 1,3-butadiene, and styrene, respectively. NYC had a higher percentage of apartments, and LA a higher percentage of single-family homes, which might account for the systematic differences seen for all the percent contributions from indoor sources (Figure 4).

Methylene chloride had very consistent SERs across homes. These results were problematic because of high background level of methylene chloride in the field and lab blanks.

Indoor sources of *n*-butyraldehyde include both direct emissions from furnishings (Brown, 1999) and secondary reactions between alkenes, namely 4-octene and 1-pentene, and ozone (Grosjean and Grosjean, 1997). However, we found little evidence of indoor sources based on the low median SER, unlike formaldehyde and acetaldehyde, which are both associated with less specific sources.

Comparison with Other Studies

The EPA TEAM Studies collected indoor and outdoor concentrations in the early 1980s in New Jersey and Los Angeles (Wallace, 1987). All the VOC concentrations from these studies were, on an average, higher than the results from the TEACH study, except for concentrations indoors and outdoors of 1,4-dichlorobenzene. For the compounds dominated by outdoor sources, benzene was the compound with the greatest reductions compared with the TEACH results, for both indoor and outdoor concentrations. The TEAM study results are over 15 years old, and these reductions could be attributable to steps EPA has taken to reduce levels of HAPs from industrial and mobile sources in this period. Concentrations of most chlorinated compounds were found to be higher in the TEAM study, with the largest differences for 1,1,1-trichloroethane.

The results from the NHEXAS study, which also provided indoor and outdoor concentrations data for Arizona and states within EPA's Region V for a limited number of VOCs were much more comparable to the results found in NYC

and LA, including formaldehyde (Lebowitz et al., 1995; Gordon et al., 1999). In addition, for aldehydes, indoor and outdoor concentrations were consistent with the results from Reiss et al. (1995b) and Zhang et al. (1994a).

Uncertainty Analysis

Uncertainty propagation for all parameters from Eq(1) was performed to estimate the uncertainty in SER calculations. Table 5 shows the median SERs for the 50 samples, the uncertainty estimates based on the median CV across all homes, variability estimates based on the CV, and a 95% confidence interval that a source is present across all homes. Sensitivity analysis showed that indoor concentrations and air exchange contributed the most uncertainty to the calculation of SERs.

Larger uncertainty (higher CV) was associated with compounds that have smaller SERs. Although not shown, this trend was also seen across homes for several compounds, especially those that had highly variable SERs across homes. For example, 1,4-dichlorobenzene had CVs that ranged from 0.25 to 11.5, and SERs that ranged from 0 to 111; whereas formaldehyde had CVs that ranged from 0.21 to 0.23 and SERs that ranged from 1.2 to 25. As the indoor and outdoor concentrations approach each other, the uncertainty in the RD between them increases. Formaldehyde, acetaldehyde, benzaldehyde, hexaldehyde, chloroform, and 1,4-dichlorobenzene, all had CVs less than 1. These compounds also tended to have the most consistent CV across all homes.

Present-day models for predicting population-based human exposure are increasingly differentiating between the uncertainty and the variability in exposures (Burke et al., 2001; Zartarian et al., 2000). The variability across homes (given as the CV) was compared with the uncertainty estimates. The only compound with greater uncertainty, compared to variability was 1,1,1-trichloroethane; however, for this cohort, indoor concentrations for this compound were dominated by outdoor sources.

We also determined what percent of the homes had a positive indoor SER with a 95th percent confidence interval. The compounds in the group dominated by indoor sources all had a greater than 80% probability of having an indoor source. 1,4-Dichlorobenzene had the lowest probability, as expected, due to the home-to-home variability in the SERs for this compound.

Conclusions

Six compounds were found to have dominant indoor sources in the NYC and LA homes sampled in this study: chloroform, 1,4-dichlorobenzene, formaldehyde, acetaldehyde, benzaldehyde, and hexaldehyde. Among these, formaldehyde had variable indoor SERs across season. Temperature and

Table 5. Uncertainty analysis of estimated emission rates in TEACH Los Angeles single-family homes in winter and fall

	SER (mg/h), ^a Median	Uncertainty, Median CV ^b	Variability CV ^c	Homes with 95% probability of an indoor source (total $N = 50$) ^d	
				N	Percent of homes
<i>Indoor</i>					
Chloroform	0.09	0.43	1.2	44	88
1,4-Dichlorobenzene	0.36	0.31	2.1	40	80
Formaldehyde	2.9	0.21	0.98	50	100
Acetaldehyde	1.7	0.22	0.72	50	100
Benzaldehyde	0.33	0.26	0.69	50	100
Hexaldehyde	1.2	0.25	1.1	50	100
<i>Indoor+outdoor</i>					
Valeraldehyde	0.12	0.81	4.6	19	39
Propionaldehyde	0.20	0.68	1.4	27	54
Styrene	0.07	0.77	1.5	20	40
Methylene chloride	0.12	1.3	1.8	15	30
Toluene	0.64	1.2	2.6	12	24
<i>n</i> -Butyraldehyde	0.08	0.78	12.0	19	38
Trichloroethylene	0.01	1.3	2.4	15	30
<i>Outdoor</i>					
Benzene	0.04	2.2	8.7	1	2
Ethylbenzene	0.03	1.8	6.4	6	12
<i>o</i> -Xylene	0.03	1.8	6.9	4	8
<i>m,p</i> -Xylene	0.08	2.0	6.9	3	6
MTBE	0.00	3.5	7.9	1	2
Tetrachloroethylene	0.02	1.8	4.0	6	12
1,1,1-Trichloroethane	0.00	5.7	2.9	5	10
Carbon tetrachloride	0.00	2.7	7.8	2	4

^aMedian SER for 50 LA single-family homes for both winter and fall.

^bMedian of CVs across all homes, obtained from individual distributions of each home.

^cStandard deviation of emission rate estimates divided by the mean of the estimates.

^dA positive emission rate at the 5% of the uncertainty distribution was counted as a source.

humidity as well as secondary formation of these pollutants may be important contributors to increased concentrations indoors in the summertime. 1,4-Dichlorobenzene had highly variable concentrations across the sampled homes and is the only compound that showed higher concentrations in NYC and LA compared to the earlier TEAM studies results. Chloroform had very consistent SERs across seasons, but higher SER were evident in NYC compared to LA. Both 1,4-dichlorobenzene and chloroform are potential carcinogens.

Modeling of the SER in this sample of NYC and LA showed that I/O ratios may in some cases be inappropriate estimates of indoor exposures, especially where underlying patterns of exposure may be obscured by the influence of AERs, as was the case for chloroform and formaldehyde. This method is not appropriate, however, for compounds that do not have significant indoor sources, as was the case for benzene, ethylbenzene, xylenes, and MTBE, as well as some of the chlorinated compounds, tetrachloroethylene, 1,1,1-trichloroethane, and carbon tetrachloride, all of which had significant contributions from outdoor sources. In these

cases, I/O ratios may be more appropriate to express the contributions to indoor exposures.

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