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Levels and sources of volatile organic compounds in homes of children with asthma

Abstract Many volatile organic compounds (VOCs) are classified as known or possible carcinogens, irritants, and toxicants, and VOC exposure has been associated with the onset and exacerbation of asthma. This study characterizes VOC levels in 126 homes of children with asthma in Detroit, Michigan, USA. The total target VOC concentration ranged from 14 to 2274 $\mu g/m^3$ (mean = 150 $\mu g/m^3$; median = 91 $\mu g/m^3$); 56 VOCs were quantified; and *d*-limonene, toluene, *p*, *m*-xylene, and ethyl acetate had the highest concentrations. Based on the potential for adverse health effects, priority VOCs included naphthalene, benzene, 1,4-dichlorobenzene, isopropylbenzene, ethylbenzene, styrene, chloroform, 1,2-dichloroethane, tetrachloroethene, and trichloroethylene. Concentrations varied mostly due to between-residence and seasonal variation. Identified emission sources included cigarette smoking, solvent-related emissions, renovations, household products, and pesticides. The effect of nearby traffic on indoor VOC levels was not distinguished. While concentrations in the Detroit homes were lower than levels found in other North American studies, many homes had elevated VOC levels, including compounds that are known health hazards. Thus, the identification and control of VOC sources are important and prudent, especially for vulnerable individuals. Actions and policies to reduce VOC exposures, for example, sales restrictions, improved product labeling, and consumer education, are recommended.

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Practical Implications

Total target VOC concentrations in the Detroit homes ranged from 14 to 2274 $\mu g/m^3$, generally lower than found in earlier studies. However, a subset of houses had elevated concentrations, and levels of 1,4-dichlorobenzene, naphthalene, and benzene reached levels commensurate with excess individual cancer risks of 10^{-2}, 10^{-3}, and 10^{-4}, respectively. VOC concentrations varied mostly due to between-residence and season effects. The most important sources included cigarette smoking, vehicle-related emissions, building renovation, solvents, household products, and pesticides.

Introduction

Many volatile organic compounds (VOCs) are classified as known or possible human carcinogens, irritants, and toxicants (U.S. EPA, 2012). In addition, several studies have linked VOC exposure with asthma exacerbation in children, although the evidence has been judged inadequate or insufficient (National Research Council, 2000). These studies include a cross-sectional survey in Sweden of children's exposure (ages 13–14 years, $n = 627$) to VOC levels in schools (Smedje et al., 1997); a case–control study in Australia of children (aged 6 months to 3 years; 88 cases; 104 controls) implicating benzene, toluene, and dichlorobenzene (Rumchev et al., 2004); a cross-sectional study in Buffalo, New York, of children (ages 1–17 years; $n = 3008$) using self-reported indoor chemical odors (Lin et al., 2008); and a case–control study (33 cases, 40 controls) in Korea (Hwang et al., 2011). As an example of a study with contrasting results, a case–control analysis of children (ages 9–11 years; 193 cases; 223 controls) in the United Kingdom found no relationship between total VOC levels and persistent wheeze (Venn et al., 2003). Other indoor and environmental factors known to trigger asthma symptoms include temperature, humidity, mold, allergens, and other air pollutants (Mendell, 2007; Sly et al., 2011; Weichenthal et al., 2007).

Most exposure to VOCs occurs indoors, a result of numerous indoor emission sources, low ventilation

rates, and the length of time spent indoors, especially in homes. Important VOC sources include cleaning products, tobacco smoke, cooking, heating appliances, VOC outgassing (from furniture, floor and wall coverings, and other building products), paints, glues, polishes, waxes, pesticides, personal care products, and the migration of emissions from attached garages (Batterman et al., 2006b, 2007; Dunagan et al., 2011; Schlink et al., 2010; Wallace, 2001; Wieslander et al., 1997). Indoor concentrations can be affected by air exchange rates (AERs), building renovations, house age and size, door and window opening, and outdoor VOC levels (Breen et al., 2010; Brown et al., 1994; Jia et al., 2008b; Jo and Sohn, 2009; Weisel et al., 2005).

The objective of this study is to characterize VOC levels in living rooms and bedrooms of a large group of children with asthma living in Detroit, Michigan, USA. We examine the spatial and temporal variability in the homes of these children over multiple seasons, evaluate factors that may affect VOC concentrations, and identify culpable sources. Results are compared with several other recent studies.

Materials and methods

Participant recruitment and study design

Families in Detroit, Michigan, with a child ages 6–12 years having symptoms or medication use consistent with persistent asthma were recruited as part of an intervention study examining ways to reduce triggers of asthma in the home by the use of air filters and air conditioners (ACs). The study used a community-based participatory research approach. Recruitment used questionnaires distributed to caregivers at schools, community fairs, and other venues (Parker et al., 2008), and ultimately, a total of 126 households were recruited following informed consent and other procedures approved by our community-based steering committee and the University of Michigan Institutional Review Board. Families entered the study on a rolling basis between March 2009 and February 2010, and the field study was concluded in September 2010. Households were randomized to a control group receiving only community health worker (CHW) home education visits ($n = 37$); the 'standard' intervention group receiving a stand-alone filter (Whispure 510, Whirlpool Corporation, Benton Harbor, MI) installed in the child's bedroom and the CHW visits ($n = 47$); and an 'enhanced' intervention group receiving the filter, the CHW visits, and a room AC (FAA062P7A, Frigidaire Augusta, GA) also installed in the child's bedroom ($n = 42$).

A total of 356 visits to the households were conducted for environmental assessments, of which 325 were considered to yield valid VOC data. Six to 10 homes were typically visited each week. Most visits occurred in spring and summer seasons (105, 102, 65, and 53 visits occurred in spring, summer, fall, and winter, respectively; Table S1). Most (90%) households had valid visits in at least two seasons (13, 38, 66, and 9 homes had 1, 2, 3, and 4 visits, respectively). Visits included physical/chemical measurements (described below), inspection of building characteristics (documenting the type of structure, house/room size, heating and cooling systems, etc.), health measurements, and surveys administered to the child's primary caregiver (to be reported elsewhere) on health, demographic, and personal factors (education, race, etc.), and indoor activities (e.g., smoking, cooking, and cleaning practices). Building features, AERs, particulate matter concentrations, filter use, and characteristics of the study homes and population have been previously reported (Batterman et al., 2012, 2013; Du et al., 2011). This study reports on the over 1300 VOC samples collected in the study homes during the study.

Sample collection and analysis

VOCs were measured as integrated 7-day samples using tube-type passive samplers (thermal desorption tubes, stainless steel, 10 cm × 4 mm id; Scientific Instrument Services, Inc., Ringoes, NJ, USA) packed with 160 mg of Tenax GR and 70 mg of Carbosieve S-III adsorbents. Duplicate samplers were deployed in the child's bedroom along with continuous monitors for temperature and relative humidity (T/RH). A third VOC sampler (and sometimes a fourth as a duplicate) was placed in the main living area, along with a second T/RH monitor. A field blank was also deployed on each visit.

The protocols and performance of the sampling and analysis methods are described elsewhere (Batterman et al., 2002; Jia et al., 2006; Peng and Batterman, 2000). In brief, each sample was spiked with 2 μL of the internal standard (containing 1 ng/μL each of fluorobenzene and p-bromofluorobenzene) and analyzed using a short-path thermal desorption/cryofocusing system (Scientific Instrument Services, Inc., Ringoes, NJ, USA) and gas chromatography/mass spectrometry (GC 5973/MS 6890, Agilent, Palo Alto, CA, USA) for over 100 target compounds including alkane, aromatic, terpenoid, halogenated, and phenolic compounds. VOC targets also included two tobacco smoke (ETS) tracers: 2,5-dimethylfuran (DMF) and 3-ethenylpyridine (3-EP) (Charles et al., 2008). ETS is very likely present if these tracers are detected, although some false positives are possible (burnt food and coffee roasting can contain DMF) (Powrie et al., 1986; Wang et al., 1983). Method detection limits (MDLs) were determined for each VOC as the standard deviation of 7 replicate low concentration measurements, using standard solutions spiked into the thermal desorption tubes, multiplied by 3.14 (Peng and Batterman, 2000).

AERs were measured using two perfluorocarbon tracers (PFTs), hexafluorobenzene and octafluorotoluene, that are inert and do not exist naturally (Batterman et al., 2006a). These compounds were measured with high sensitivity. Emitters were individually calibrated, periodically checked, and refilled as needed. AERs for the house's living area and child's sleeping area, and interzonal flows between these areas have been reported elsewhere (Du et al., 2012).

Quality assurance (QA) measures taken to ensure reproducibility and data quality included the use of standard operating protocols, collection and analysis of blanks at each visit, quarterly calibrations, and duplicate samples. Blank-field samples showed little if any contamination. MDLs for individual VOCs ranged from 0.02 to 1.8 $\mu g/m^3$, depending on the compound. Duplicate samples had precisions below 20% in most cases, thus most analyses averaged duplicates. Overall measurement uncertainty was considered to be low, and the sampling and analytical methods were appropriate for the indoor application.

Data analysis

Description statistics were computed for each VOC and home (average of multiple visits), including the detection frequency (DF; fraction of observations exceeding the MDL). We checked for outliers in the 13 homes that had only one valid visit and whether statistics fell within the expected range. To further ensure that results were representative, we restricted the analysis to those homes that had valid measurements in at least 2 or 3 seasons, and repeated the descriptive analysis. To understand sources of variation, random-effect models were used to apportion the variance of each VOC's concentration to four sources: between-residence variability, within-residence variability (i.e., living room and bedroom), season (variance between seasonal visits), and measurement uncertainty (variance between replicates) (Jia et al., 2012). Apportionments were performed individually for the 35 VOCs with DF above 50%. (Apportionments with low DF may not reliable.) Because this analysis assumes normality, log-transformed data were used for this analysis. (VOC concentrations were approximately lognormally distributed.)

As mentioned, the presence of ETS was determined by the detection of the DMF and 3-EP tracers, as well as survey responses regarding indoor smoking. Group differences in VOC levels were compared using Kruskal–Wallis (K-W) tests with $\alpha = 0.05$ to examine effects of season, AER, temperature, traffic measures, and other variables. The effect of vehicle traffic within 100- and 300-m buffers of each home on benzene and ΣBTEX (sum of benzene, toluene, ethylbenzene, and xylene) levels was investigated using generalized linear models that used quartiles of vehicle-miles travelled per day within each buffer as a potential explanatory variable.

Potential VOC sources and associations among VOCs were identified using factor analyses and varimax rotations. Factors with eigenvalues exceeding 1 that included at least one variable with a loading over 0.3 were extracted. These analyses included those VOCs that had measurable levels (>MDL) for at least 10% of the homes (40 target VOCs). Measurements below MDLs were set to one-half of MDL.

VOCs were prioritized based on screening-level health risk estimates. The excess lifetime individual cancer risk of the ith VOC was calculated as $Risk_i = EC_i \times CSF_i$ where EC_i = exposure concentration, estimated as the average indoor concentration ($\mu g/m^3$), and CSF_i = cancer slope factor (per $\mu g/m^3$). The non-cancer risk hazard quotient (HQ_i) was calculated as $HQ_i = EC_i/RfC_i$, where RfC_i = reference concentration ($\mu g/m^3$) relevant to adverse (non-cancer) outcomes. CSF and RfC values were taken from the integrated risk information system (IRIS) and, if unavailable, from the initial risk screening levels (IRSLs) used in Michigan's air toxic screening program (IARC, 2012; MDEQ, 2012; U.S. EPA, 2012). (CSF, RfC, and IRSL values, and other information are listed in Table S2.) The risk and HQ calculations represent screening-level estimates that assume indoor measurements are representative of lifetime exposures and that exposures outside the home, for example, at school or work, are low or comparable with levels in homes. These assumptions are supported by many studies showing that indoor settings, particularly residences, are the dominant VOC microenvironment for most individuals (Su et al., 2013; U.S. EPA, 2011), especially for children and the elderly who spend most of their time at home. This assumption may not apply to adults who spend less time at home, and particularly to those working in settings with high VOC exposure.

Descriptive and factor analyses used SPSS 17 (IBM Corp., Somers, NY, USA). The traffic analysis used PROC GLM, and the variance component analyses used PROC NESTED, both in SAS (v9.1.3, SAS Institute, Cary, NC, USA). Data and results were organized using Microsoft Excel (Microsoft, Seattle, WA, USA).

Results and discussion

Indoor concentrations

A total 56 VOCs were detected in the 126 homes, which were distributed across the more populated areas of Detroit (Figure 1). A total of 35 compounds were detected in at least half of the homes (Table 1). The household average total target VOC (ΣVOC) concentration was 150 $\mu g/m^3$ (median of 91 $\mu g/m^3$). Concentrations varied considerably among households, from 14 to 2274 $\mu g/m^3$, and distributions were strongly

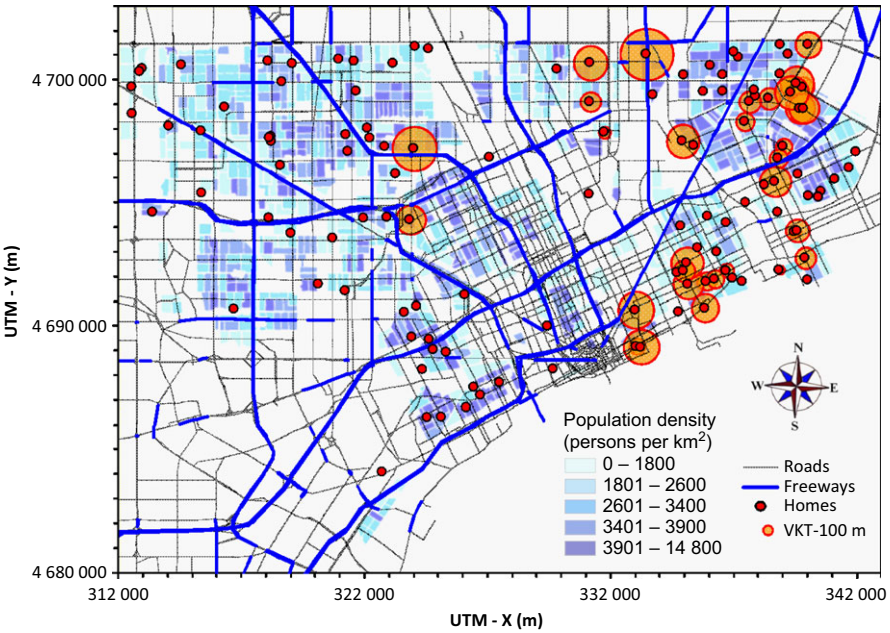

Fig. 1 Locations of the 126 study homes (red dots) in Detroit, along with major roads and the population density. The area of the orange circle around each home is proportional to the vehicle-kilometers-driven (VKT) per day within a 100-m buffer of the home

right-skewed. VOCs with the highest average concentrations included aromatics (benzene, toluene, and xylenes), alkanes (n-$C_{7–13}$ and methyl cyclohexane), terpenes (d-limonene and α-pinene), and tetrachloroethene. No VOC differed significantly among the three intervention groups, thus subsequent analyses use pooled data.

Because the sample was unbalanced with respect to number of visits made to each home, we checked to whether the group of homes with fewer visits (13 homes had 1 visit) was representative. In this subset, the VOC data showed no outliers, and means and other statistics were within the seasonal range of the larger sample. Analyses restricted to the 75 households that had valid measurements during at least three seasons (Table S3) also showed results very similar to those obtained using all of the household data (Table 1). For example, most 25th, 50th, and 75th percentile concentrations were within a few percent, and means were within 25% (and usually much closer) with the exceptions of 1,2,3-trimethyl benzene (37% higher), 1,4-dicholorobenzene (64% lower), ethyl acetate (34% higher), and ethyl methacrylate (31% lower).

Many VOCs were found in all or nearly all homes, including d-limonene and α-pinene, which are constituents of cleaning products, air fresheners, and fragrances (Aronson et al., 2007; Nazaroff and Weschler, 2004), and toluene and other aromatics, which are in household products, paints, adhesives, synthetic fragrances, evaporated fuel, vehicle emissions, and many other products. Ethyl acetate, detected in 71% of visits, exceeded 10 $\mu g/m^3$ in 20 households (16%); this VOC is used in nail polish remover, glue, and other products (U.S. National Library of Medicine, 2012). Naphthalene and 1,4-dichlorobenzene, two VOCs used as pest repellents and deodorants, were found at very high levels in a few homes (e.g., the highest household levels were 556 and 4249 $\mu g/m^3$, respectively, which occurred in different homes); such high levels likely result from inappropriate use of these products (Batterman et al., 2012; Chin et al., 2013; Jia and Batterman, 2010).

Comparison with other studies. Although concentrations of several VOCs in a number of houses appeared elevated, VOC levels measured in other North American studies tend to be higher. Figure 2 contrasts results with four other relevant studies. The Three Cities Study (TCS) used 1-week samples in living rooms (n = 161 houses) measured in 2005 and 2006 in three southeast Michigan cities: Dearborn, a densely populated and industrialized city, in many ways similar to Detroit; Ypsilanti, a more urban and commercial city; and Ann Arbor, a largely suburban and affluent community (Jia et al., 2008a). The Mechanistic Indicators of Childhood Asthma Study (MICAS) collected 7-day samples (participant-deployed) in Detroit homes of children (ages 9–13 years, n = 41) with and without asthma in November and December, 2006 (Johnson et al., 2009, 2010). The Windsor, Ontario (Canada) Exposure Assessment Study (WOEAS) (Health Canada, 2006) was conducted just south of Detroit in non-smoking homes with asthmatic children (aged 9–12 years, n = 47) in summer and winter of 2006. In WOEAS, caregivers were not occupationally exposed, and 5-day samples collected in summer, and winter results were averaged. The fourth study is a compilation of 77 reports examining residences in the US and

Table 1 VOC concentrations ($\mu g/m^3$) by household average ($n = 126$), except 'Max event' which shows highest household average in a single visit based on 325 visits

	MDL	DF(%)	Mean	Std. Dev.	Min	25th	Median	75th	90th	95th	Max	Max event
Aromatics												
Benzene	0.06	100	2.27	3.25	0.33	0.85	1.45	2.50	4.14	4.89	27.93	71.59
Toluene	0.04	100	11.62	23.22	1.08	4.17	6.02	9.30	19.88	30.56	179.84	448.05
Ethylbenzene	0.03	100	1.72	2.29	0.43	0.75	1.08	1.85	3.71	4.85	22.47	59.16
p-Xylene, m-Xylene	0.04	100	6.34	10.01	1.39	2.48	3.29	6.34	13.71	19.98	98.48	260.36
o-Xylene	0.04	100	2.02	3.15	0.34	0.80	1.14	1.81	4.11	6.18	30.46	80.65
4-ethyl toluene	0.17	100	1.92	3.91	0.29	0.59	0.79	1.28	4.02	6.58	30.81	71.22
1,3,5-Trimethylbenzene	0.02	100	0.70	1.09	0.10	0.25	0.33	0.55	1.55	2.48	9.59	23.61
1,2,4-Trimethylbenzene	0.03	100	2.18	3.61	0.29	0.77	1.03	1.72	4.92	8.41	34.29	83.89
Naphthalene	0.09	100	7.88	25.93	0.19	0.65	0.97	1.64	9.34	37.60	200.58	556.24
1,2,3-trimethyl benzene	0.03	99	0.80	1.12	0.01	0.29	0.40	0.83	1.98	2.56	9.52	22.43
Styrene	0.04	98	0.70	0.54	0.02	0.35	0.53	0.82	1.51	1.92	2.72	6.46
2-ethyl toluene	0.02	98	0.74	1.43	0.01	0.25	0.34	0.54	1.44	2.46	12.51	21.24
p-Isopropyltoluene	0.02	98	1.58	1.65	0.01	0.69	1.09	1.71	2.95	4.86	10.75	17.94
1,4-Dichlorobenzene	0.02	98	40.01	212.41	0.01	0.38	0.64	3.06	23.86	88.15	2125.94	4249.23
n-Propylbenzene	0.02	98	0.52	0.97	0.01	0.18	0.25	0.38	1.06	1.82	7.88	16.48
2-methylnaphthalene	0.10	94	0.30	0.24	0.05	0.17	0.23	0.35	0.49	0.79	1.60	3.06
1-Methylnaphthalene	0.08	79	0.16	0.11	0.02	0.09	0.13	0.19	0.25	0.39	0.75	1.45
Isopropylbenzene	0.05	67	0.13	0.18	0.02	0.04	0.07	0.15	0.31	0.45	1.23	2.91
n-Butylbenzene	0.03	49	0.11	0.28	0.02	0.02	0.03	0.07	0.26	0.37	2.30	6.83
Chlorobenzene	0.05	7	0.03	0.03	0.02	0.02	0.02	0.02	0.02	0.08	0.35	0.67
2-Chlorotoluene	0.02	2	0.01	0.04	0.01	0.01	0.01	0.01	0.01	0.01	0.34	0.67
sec-Butylbenzene	0.03	2	0.02	0.04	0.01	0.01	0.01	0.01	0.01	0.01	0.36	0.73
1,2-Dichlorobenzene	0.02	2	0.01	0.05	0.01	0.01	0.01	0.01	0.01	0.01	0.51	1.52
Bromobenzene	0.06	2	0.03	0.01	0.03	0.03	0.03	0.03	0.03	0.03	0.16	0.43
1,2,4-Trichlorobenzene	0.07	1	0.04	0.04	0.03	0.03	0.03	0.03	0.03	0.03	0.43	0.63
Alkanes												
n-Heptane	0.06	100	4.77	15.49	0.35	1.13	1.85	3.53	6.42	12.33	167.73	498.68
n-Octane	0.07	100	1.52	1.56	0.32	0.72	1.10	1.81	2.62	3.24	13.59	35.77
n-Nonane	0.58	97	4.01	8.10	0.29	1.02	1.70	3.35	6.68	16.19	62.92	124.31
n-Decane	0.31	92	3.19	7.16	0.11	0.58	1.02	2.10	6.06	13.54	47.37	93.47
n-Undecane	0.07	99	2.04	6.36	0.04	0.34	0.53	1.08	2.85	6.71	56.38	161.77
n-Dodecane	0.07	99	1.20	2.25	0.04	0.41	0.57	1.08	2.18	3.35	16.96	48.51
n-Tridecane	0.13	97	1.86	3.35	0.06	0.36	0.80	1.55	3.43	9.80	20.18	54.15
n-Tetradecane	0.26	98	5.22	12.36	0.13	1.03	1.83	3.64	9.65	25.53	107.71	310.12
n-Pentadecane	0.19	97	2.02	3.58	0.09	0.55	0.97	1.81	4.57	7.16	25.51	73.49
n-Hexadecane	0.44	90	1.32	0.96	0.22	0.69	1.06	1.69	2.50	3.14	5.27	11.13
Methyl cyclohexane	0.02	99	1.48	3.58	0.01	0.30	0.60	1.15	2.14	4.53	31.99	95.42
Cyclohexane	0.59	53	1.41	2.57	0.20	0.36	0.62	1.19	2.27	7.55	16.40	48.18
Terpenes												
a-Pinene	0.03	99	4.44	7.23	0.01	1.26	1.84	4.22	9.15	16.65	44.46	82.98
d-Limonene	0.56	99	22.56	23.44	0.38	8.34	16.23	27.25	43.30	60.58	172.96	305.68
Halogenated												
Tetrachloroethene	0.09	91	0.71	1.66	0.05	0.15	0.26	0.53	1.35	2.39	13.70	21.80
Chloroform	0.16	35	0.36	0.74	0.08	0.08	0.08	0.39	0.96	1.22	7.07	14.05
1,2-Dichloroethane	0.39	13	0.34	0.51	0.18	0.20	0.20	0.21	0.48	1.01	3.93	7.68
Trichloroethylene	0.09	6	0.07	0.14	0.04	0.04	0.04	0.04	0.04	0.10	1.48	2.70
Methylene chloride	0.71	6	0.54	0.91	0.35	0.35	0.35	0.35	0.38	0.91	7.85	12.43
1,1,1-Trichloroethane	0.39	6	0.26	0.41	0.19	0.19	0.19	0.19	0.19	0.48	4.47	5.79
Dibromochloromethane	0.21	4	0.11	0.04	0.10	0.10	0.10	0.10	0.13	0.18	0.43	0.65
1,1,1,2-Tetrachloroethane	0.21	2	0.13	0.12	0.11	0.11	0.11	0.11	0.11	0.11	1.04	2.28
Bromodichloromethane	0.73	1	0.38	0.05	0.37	0.37	0.37	0.37	0.37	0.37	0.87	1.25
Ketones, ethers, others												
Ethyl acetate	1.64	71	7.34	15.57	0.51	1.47	3.13	6.90	12.96	24.79	117.38	345.52
2-Hexanone	0.38	13	0.52	1.47	0.19	0.19	0.19	0.19	0.64	1.60	12.40	5.82
Ethyl methacrylate	0.65	4	1.22	6.87	0.33	0.33	0.33	0.33	0.33	0.58	73.74	36.82
2-Butanone	0.38	3	0.43	1.50	0.19	0.19	0.19	0.19	0.19	0.19	13.09	2.77
Phenol	1.15	56	1.32	0.60	0.36	0.88	1.23	1.66	1.92	2.44	3.28	12.53
Tetrahydrofuran	1.85	6	1.05	0.78	0.27	0.93	0.93	0.93	0.93	2.58	6.73	1.25
3-Ethenylpyridine	0.38	10	0.24	0.16	0.10	0.19	0.19	0.19	0.38	0.57	1.48	86.00
2,5-Dimethyl furan	0.36	5	0.21	0.09	0.13	0.18	0.18	0.18	0.26	0.35	0.82	25.99
SVOC	–	–	150	233	14	53	91	161	249	442	2274	4520

MDL: method detection limits; DF: detection frequency.

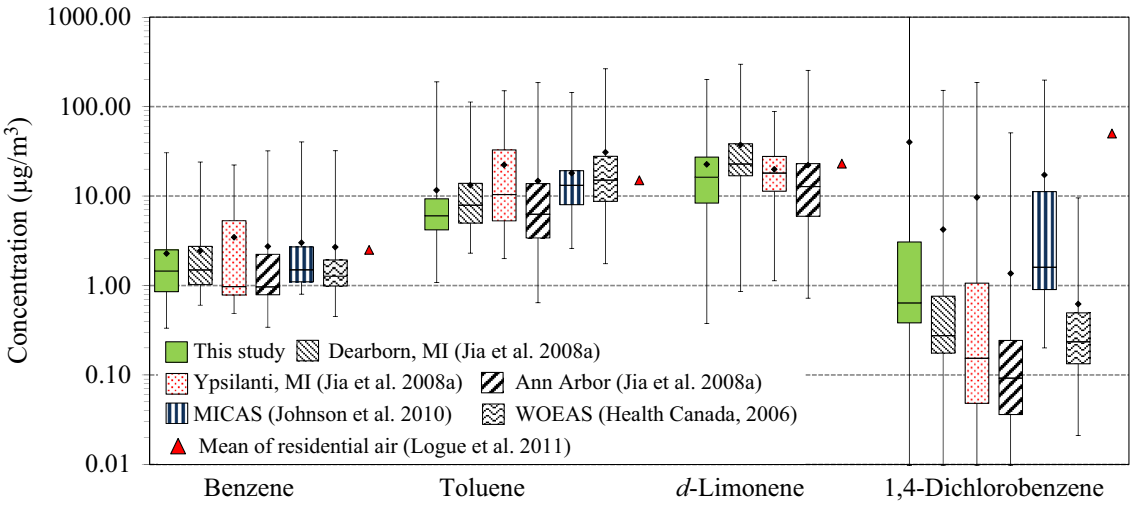

Fig. 2 VOC concentrations in this study compared with results from the 3 city study in southeast Michigan (Jia et al., 2008a); Canada (Health Canada, 2006), Detroit (Johnson et al., 2010), and a meta-study (Logue et al., 2011). Box plots show minimum, maximum, 25th, 50th, and 75th percentile concentration; points show means

countries with similar lifestyles from 1995 to 2010 (Logue et al., 2011). While there is significant variability within each study, median concentrations of benzene and d-limonene levels among the studies were similar; 1,4-dichlorobenzene levels varied widely (probably dependent on occupant use of deodorant and pesticide products (Chin et al., 2013); and toluene levels were lowest in the present study.

Outside of North America, much higher VOC levels have been reported in homes of individuals with asthma (or with asthma-like symptoms). As examples, in Sweden, toluene averaged 120 $\mu g/m^3$ (range of 1–2330 $\mu g/m^3$; $n = 88$) in living rooms and bedrooms sampled in 1991 and 1992 (Norbäck et al., 1995); in Australia, median and maximum benzene concentrations were 20 and 82 $\mu g/m^3$, respectively, and toluene levels were 17 and 154 $\mu g/m^3$ in living rooms of children's homes (ages 6 months to 3 years, $n = 88$) sampled in 1997 through 1999 (Rumchev et al., 2004); and in Korea, toluene, formaldehyde, *p, m*-xylene, and ethylbenzene had geometric mean concentrations of 46, 31, 14, and 9.2 $\mu g/m^3$, respectively, in homes of children (ages 8–13 years, $n = 33$) sampled in 2008 (Hwang et al., 2011). The Swedish, Australian, and Korean studies show higher, and sometimes much higher, VOC levels than the present study and most of the North American studies.

Concentrations of VOCs measured in different studies can differ for many reasons. Levels in more recent studies (after 2000 or so) are often lower due to progress made in controlling indoor and outdoor emissions, as well as the reduction or elimination in indoor smoking (Batterman et al., 2012; McCarthy et al., 2007; Su et al., 2011). Because outdoor VOC levels provide a 'floor', indoor concentrations can be affected by urbanization, proximity to industry, traffic and other emission sources, and meteorology. Regional differences in building design (including attached garages), building materials, and climate can influence both AERs and emissions. In comparison with most other North American studies, few Detroit homes have attached garages, eliminating a common source of BTEX and other fuel-related VOCs (Batterman et al., 2006c; Chin and Batterman, 2012). The use of VOC-emitting products (air fresheners, scents, pesticides, etc.) can depend on social and cultural practices and jurisdictional controls. Finally, sampling and analytical methods can affect results, for example, studies using short sampling periods and without seasonal averaging generally show greater variability, a result of temporal variability (described below).

Variation in VOC levels

The variance components analysis apportioned the variability in VOC concentrations to between-household variation (49 ± 11% of the total variance), seasonal variation (34 ± 12%), within-household variation (between living rooms and bedrooms; 10 ± 6%), and measurement variation (among replicates; 7 ± 6%). While the apportionments varied by VOC (Figure 3, Table S4), the largest source of variation was household-to-household differences, a result of the different VOC products used/stored in each house, variation in smoking and AERs, and other factors that varied by household. A few studies have reported large variation in VOC concentrations among households (Jia et al., 2012; Sexton et al., 2004; Zhu et al., 2005). As discussed below, however, seasonal variation was nearly as important as variation among household.

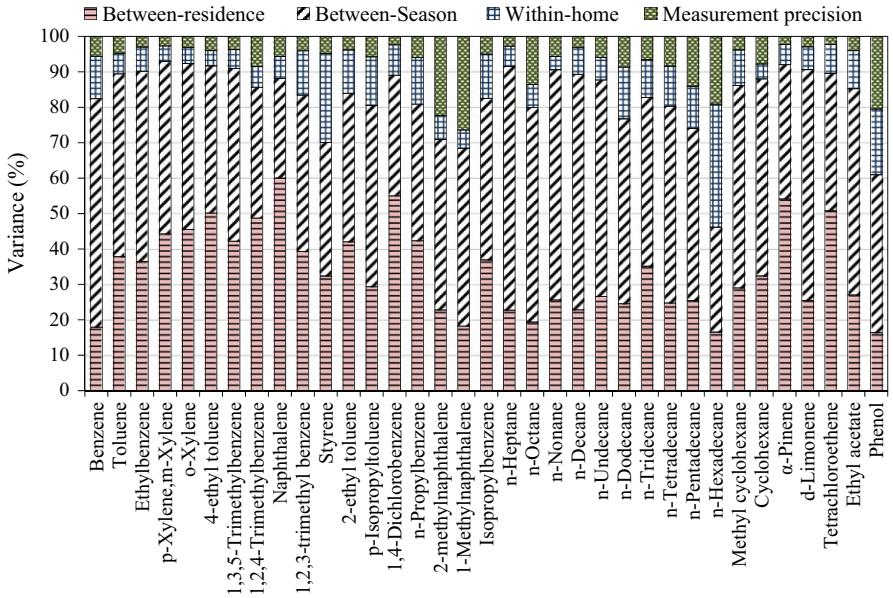

Fig. 3 Variance components for indoor VOCs with detection frequencies above 50% (35 VOCs) showing fraction of variance attributable to four sources: between-residence, between-season, within-residence, and measurement precision. Based on 325 visits with valid data from 126 homes

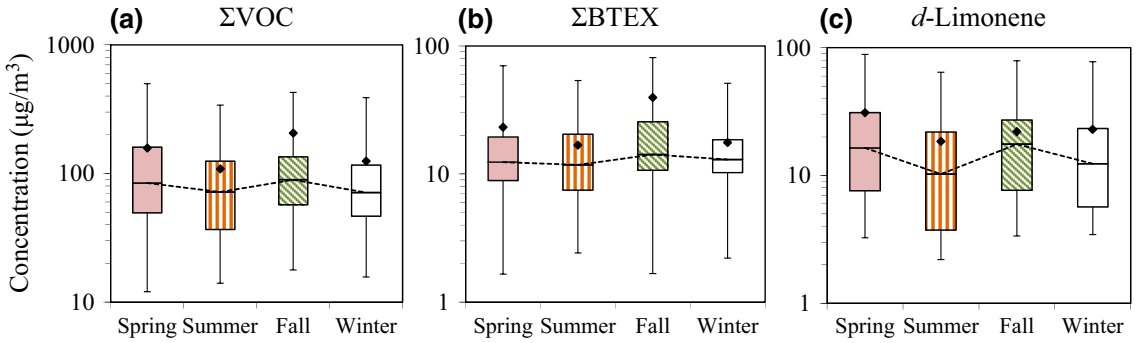

Fig. 4 Season average concentrations for ΣVOCs, ΣBTEX, and *d*-limonene. Sample sizes were 105, 102, 65, and 53 for spring, summer, fall, and winter, respectively. Plots show 10th, 25th, 50th, 75th, and 90th percentile concentrations; points show means

Table 2 Air exchange rates (AER, h^{-1}) and indoor temperature (Temp, °C) in the living area by season

	n	Mean	Std. Dev.	Median	Min	Max
AER (h^{-1})						
Spring	87	0.81	0.71	0.59	0.14	4.47
Summer	85	1.17	1.28	0.85	0.15	9.79
Fall	53	1.04	0.71	0.91	0.11	3.63
Winter	52	1.11	0.62	0.98	0.20	2.47
Temp (°C)						
Spring	87	21.4	2.6	21.7	14.1	27.9
Summer	85	25.4	2.6	25.7	19.0	29.9
Fall	53	21.7	2.8	21.7	15.6	31.3
Winter	52	20.3	3.6	19.9	12.7	31.6

Seasonal variation. Overall, VOCs levels were highest in spring and fall, and lowest in summer and winter (Figure 4). Seasonal changes can be caused by many factors including the AER, which depend on the indoor–outdoor temperature difference, the presence and use of air conditioners, window opening, wind speed, age and condition of the house, and other factors (Breen et al., 2010; Du et al., 2012). In the Detroit homes, the AER in the living area averaged 1.02 ± 0.92 1/h (median = 0.80 1/h, n = 277) and varied seasonally (KW test, P = 0.002; median test, P = 0.009) (Table 2) (Du et al., 2012). The higher AERs observed in winter reflect large indoor–outdoor temperature differences and high wind speeds; in summer, temperature differences as well as opened windows also increase the AER. AERs are lower in other seasons (seen especially in spring) as windows are closed, and the driving forces (primarily the temperature difference) are small (Breen et al., 2010). AERs were negatively correlated with VOC concentrations, especially for toluene, styrene, α-pinene, and limonene (Spearman rank correlation coefficients from −0.3 to −0.4; Table S5), as expected for VOCs with strong

indoor sources (Gilbert et al., 2008; Haghighat and Huang, 2003; Lin et al., 2009; Stocco et al., 2008).

Several studies have reported seasonal variation and similar trends in VOC levels as found in Detroit (Ilgen et al., 2001; Jia et al., 2012; Rehwagen et al., 2003; Schlink et al., 2010). Importantly, this variability means that measurements in multiple seasons are needed to estimate long-term VOC levels in households.

Within-house variation. Within-household variation was relatively small, based on the variance components analysis, and also based on relative absolute concentration differences between the concentrations in living rooms and bedrooms, which showed a median difference of 30% across the 41 VOCs (range from 8 to 69%; $n = 279$ pairs). The variation within a building depends on the degree of mixing, airflows and locations of VOC sources. Given that most (87%) Detroit homes had forced air heating/cooling systems, indoor air is often well-mixed and within-home concentration gradients were small, especially when averaged over longer periods (1 week in the present case) (Batterman et al., 2007; Dodson et al., 2008). However, this conclusion is limited to differences between living rooms and bedrooms, which were selected as the locations where children spent most of their time. VOC levels may show greater differences in kitchens, bathrooms, and possibly other settings.

Temperature. Indoor temperatures averaged 22.5 ± 3.5 °C (median = 22.3 °C, $n = 227$), varied seasonally (KW test $P < 0.001$; median test, $P < 0.001$), and increased by 5 °C in summer compared with winter (Table 2). Concentrations of most VOCs were negatively correlated with indoor temperature ($-0.1 \leq r \leq -0.3$; Table S5). AERs and indoor temperatures were positively (although weakly) correlated ($r = 0.17$).

Environmental tobacco smoke. Based on ETS tracers (detected in 35 homes) and occupant surveys (positive responses in 37 homes), smoking may have occurred in 53 homes. Both indicators were positive in 19 homes. Homes with smoking during the sampling period are defined using the ETS tracers, which provided stronger evidence of smoking based on particulate matter (PM) concentrations (Batterman et al., 2012). Homes with ETS detected during the sampling period had higher levels of benzene, tetrachloroethene, styrene, phenol, n-butylbenzene, naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, and n-pentadecane, but lower levels of α-pinene (Table S6). Most of these VOCs are found in cigarette smoke (Chambers et al., 2011; Charles et al., 2007). While the differences noted were statistically significant, concentration changes were small, for example, median levels changed by only 10–20%.

While smoking and ETS have been associated with large increases in PM concentrations (Batterman et al., 2012), effects on VOC levels tend to be small. As examples: in Regina, Saskatchewan, Canada, benzene concentrations averaged 3.4 $\mu g/m^3$ in homes with at least one smoker ($n = 31$) compared with 2.5 $\mu g/m^3$ ($n = 158$) in non-smoking homes (Health Canada, 2007); and in MICAS, homes with smokers had higher indoor/outdoor ratios of benzene, ethylbenzene, and xylene (Johnson et al., 2010). Smoking causes larger differences when personal sampling is used (which gives exposures), as compared to area sampling used in the present (and most) studies (Edwards et al., 2001).

Other factors. Additional factors that have been associated with indoor VOC concentrations include the use and storage of certain household products, building renovations, attached garages, and vehicle emissions (Brown et al., 1994; Chin and Batterman, 2012; Diez et al., 2000; Jia and Batterman, 2010; Rumchev et al., 2004). Elevated concentrations (>50 $\mu g/m^3$) of *n*-alkanes (n-C_{7-13}) were found in 14 Detroit homes, which may indicate recent building renovation (Rumchev et al., 2004). In this study, only nine houses had an attached garage. Although the amount of traffic exposure varied considerably among homes, the amount of traffic within either 100- or 300-m buffers showed only weak and statistically insignificant effects on indoor benzene and ΣBTEX levels (using correlations and GLMs). The ability to discern the influence of outdoor VOC levels and sources on indoor concentrations is limited because many indoor sources emit the same compounds, for example, the BTEX compounds associated with vehicle exhaust and gasoline (Chin and Batterman, 2012) are also found in ETS, paints, adhesives, solvents, oils, and incense (Alexopoulos and Bakeas, 2011; Charles et al., 2007; Diez et al., 2000). Similarly, identifying sources by VOC profiles is rarely definitive, as further discussed in Section 3.3.

Measurement uncertainty. As noted, measurement variation was small, averaging only 8 ± 12%. Measurement variation increased for VOCs with low detection frequencies, for example, 1-methylnaphthalene, 2-methylnaphthalene, n-hexadecane, and phenol.

Source identification using factor analysis

Factor analyses using the long-term (multiseason) averages at the homes ($n = 126$) resolved 11 factors, a large number. Frequently, several VOCs were present in multiple factors, indicating multiple sources or effects on that VOC (demonstrated in the loading matrix, Table S7). Factor 1 contained mostly aromatics

(BTEX, trimethylbenzenes, etc.) and likely represents solvents emissions (pens, inks, coatings, paint, perfume, adhesives, and varnish) and vehicle-related emissions. Factor 2 contained $C_{12–16}$ n-alkanes and likely reflects oil and diesel engine exhaust. Factor 3 with *d*-limonene, phenol, and α-pinene reflects the use of household products, fragrances, air fresheners, and building materials. Factor 4 included $C_{9,10,12}$ n-alkanes, which are constituents of oils, lubricants, and solvents. The remaining factors explained from 3.1 to 4.8% of the total variance. Factor 5 (cyclohexane, n-heptane, methyl cyclohexane) may indicate paints, adhesives, and solvents. Factor 6 (ethyl acetate, 2-hexanone, tetrachloroethene) may reflect glues, nail polish removers, solvents, and paints. Factor 7 (chloroform) likely reflects disinfection by-products associated with water use. Factor 8 (1,2-dichloroethane, 1,4-dichlorobenzene) includes degreasers, paint remover, and moth crystals. Factor 9 ($C_{11–12}$ n-alkanes and tetrachloroethene) may reflect solvents. Factor 10 (3-EP) represents ETS sources. Lastly, factor 11 (1,4-dichlorobenzene, naphthalene) is the pest repellent discussed earlier.

Factor analyses based on the seasonal visits (n = 325) resolved 10 factors (Table S8). In this case, ETS was not resolved (the detection frequency of 3-EP was only 8%). Analyses performed for each season might better identify sources that vary by season and can separate periods (e.g., summer) when children are more likely to be home. However, most results of seasonally stratified analyses were comparable with that for household averages, for example, again, 10 factors were identified, and the first four or five factors explained over half of the total variance with similar VOC sources (Tables S9–S12).

Risk-based rankings

Ten of the target VOCs have been identified as possible or known carcinogens (Table S3). Of these, household average concentrations of six VOCs exceeded an individual excess lifetime cancer risk level of 10^{-5} in some or many homes (73, 68, 40, 29, 17, and 2 homes for naphthalene, benzene, 1,4-dichlorobenzene, chloroform, 1,2-dichloroethane, and isopropylbenzene, respectively), and all ten VOCs exceeded the 10^{-6} level (exceeded in 126, 126,124, 126, 126, 41, 17, 10, 4, and 3 homes for naphthalene, benzene, 1,4-dichlorobenzene, chloroform, 1,2-dichloroethane, isopropylbenzene, ethylbenzene, styrene, trichloroethylene, and tetrachloroethene, respectively) (Figure 5). 1,4-dichlorobenzene and naphthalene presented the highest risks, which reached 10^{-2} in one home and 10^{-3} in 10 homes. Benzene risk exceeded a risk level of 10^{-4} in two homes, and 10^{-5} in half (n = 68) of the homes, based on the lower limit of slope factor range (1.3–4.5 $\mu g/m^3$ for a 10^{-5} risk) (U.S. EPA, 2012). Another known carcinogen, chloroform, exceeded a risk level of 10^{-5} in 31 homes and 10^{-4} in one home. The probable human carcinogen, 1, 2-dichloroethane, exceeded a risk level of 10^{-5} in 16 homes. Isopropylbenzene exceeded risks of 10^{-5} in two homes.

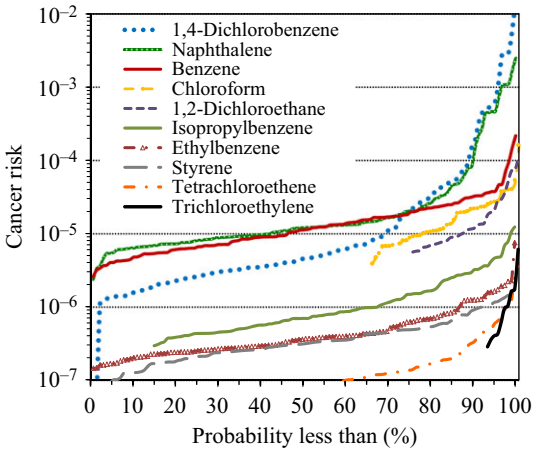

Fig. 5 Distribution of cancer risks for selected VOCs in Detroit homes (n = 126). Lines may start at high probabilities (not 0) as VOC concentrations in some homes fell below method detection limits (MDLs)

Based on 75th percentile concentrations, the risk ranking of the ten carcinogenic VOCs is 1,4-dichlorobenzene > naphthalene > benzene > (chloroform) > (1,2-dichloroethane) > isopropylbenzene > ethylbenzene > styrene > (tetrachloroethene) > (trichloroethylene). In this ranking, VOCs in parentheses have a detection frequency below 50%, and thus reflect some uncertainty regarding the robustness of this ranking.

Hazard quotients (HQ) for chronic non-cancer health effects exceeded one in 21 homes for naphthalene (maximum HQ = 67) and in 2 homes for 1,4-dichlorobenzene (maximum HQ = 3). Concentrations of other VOCs were far below reference concentrations, indicating that non-cancer health effects are unlikely.

Calculations of excess health risks have been used to prioritize VOCs in several studies. A recent review identified seven VOCs (acetaldehyde, acrolein, benzene, 1,3-butadiene, 1,4-dichlorobenzene, formaldehyde, and naphthalene) as priority indoor air pollutants (Logue et al., 2011). Three VOCs (formaldehyde, benzene, and naphthalene) were identified as priorities in European homes (Koistinen et al., 2008). WHO identified five priority VOCs (benzene, styrene, toluene, trichloroethylene, and tetrachloroethene) (WHO, 2000). The VOCs identified in these rankings are similar to those in the Detroit homes.

VOCs that can pose health risks arise from many indoor and outdoor sources. Identifying and controlling VOC sources are important and prudent, particularly for vulnerable individuals. Individuals can minimize exposure by eliminating or minimizing use of

VOC-emitting products (e.g., air fresheners), choosing VOC-free or low-VOC consumer products, minimizing migration indoors of VOCs from attached garages and outdoor sources, and using ventilation and other controls when VOCs are used indoors. Policies that can reduce exposure include sales bans and restrictions on toxic chemicals, improved labeling, building and ventilation standards, and consumer education.

Study strengths and limitations

This study characterized VOC concentrations in a large sample of Detroit homes with asthmatic children using measurements repeated in multiple seasons. Sampling used a one-week sampling period and included VOC measurements in both living rooms and bedrooms, as well as blanks, replicates, and other means to ensure quality results. In addition, AERs and other variables were measured to help to interpret results. We recognize several limitations. Target VOCs did not include very volatile compounds, for example, aldehydes and carbonyls. Only homes of asthmatic children in Detroit were sampled, and results may not be applicable to other homes. While home inspections were conducted, not all sources that might emit VOCs were noted. Outdoor VOC concentrations were not measured. Both VOC and AER measurements were time averaged, and short-term effects (hourly to daily) were not characterized. Because a single location was monitored in each room, we do not have direct information on within-room variation in VOC levels; however, this is unlikely to be significant for the 1-week averaging period given the small between-room differences observed. The factor analysis provides only tentative identification of sources. The risk estimates, which used available measurements and recommended parameters, are screening-level estimates of excess risk that assume that VOC measurements are representative of long-term exposure, and they do not account for exposures in environments outside the home, time budgets, and mixture effects. While we used robust statistical tests, repeated measures, and a large sample size, many results are VOC-specific, and some may be affected by interactions, outliers, sampling schedules, or other factors that could not be identified, in part due to sample size limitations. Most of these limitations result from cost and logistical factors. Lastly, the study design does not permit a direct investigation of whether short- or long-term VOC levels are associated with asthma.

Conclusions

A total of 56 VOCs were detected in the 126 homes sampled of children with asthma living in Detroit, Michigan, and 35 VOCs were found in at least half of the homes. The predominant compounds included aromatics, alkanes, terpenes, and tetrachloroethene. Although many compounds were detected, concentrations were generally lower than levels reported in other recent studies in North America. The analysis of temporal and spatial variability indicates the need to obtain repeated measurements in multiple seasons to characterize indoor levels; this is generally more important than sampling in multiple locations within a home. Emission sources, identified using tracers, high concentrations of several VOCs, and factor analysis, included cigarette smoking, vehicle-related emissions, renovation, solvents, household product, and pesticides. As expected, concentrations were negatively associated with air exchange rates, indicating the significance of indoor sources. VOCs posing cancer risks that exceed guideline levels in many homes included naphthalene, benzene, 1,4-dichlorobenzene, isopropylbenzene, ethylbenzene, and styrene; a small number of homes had high risks due to chloroform, 1,2-dichloroethane, tetrachloroethene, and trichloroethylene. High levels of naphthalene and 1,4-dichlorobenzene, associated with non-cancer effects in humans, were found in a subset of residences.

This study provides information regarding VOC exposures of a vulnerable population, children with asthma. While evidence supporting the role of VOC exposure in asthma exacerbation is limited, this study identifies VOCs and emission sources that might pose hazards, and thus can help to facilitate actions and policies to decrease exposure and reduce possible triggers of asthma.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Table S1 Number of visits and samples collected among the 126 households by season.

Table S2 Unit risks and chronic inhalation reference concentration (RfCs) for selected VOCs.

Table S3 Summary of VOC concentrations ($\mu g/m^3$) for the 75 households that had at least three valid seasonal measurements.

Table S4 Variance components for indoor VOCs on 325 visits, showing fraction of variance attributable to four sources: between-residence, between-season, within-residence, and measurement precision.

Table S5 Spearman correlation coefficients between air exchange rates (AER, h^{-1}) and indoor temperature (°C), and target VOCs, respectively (n = 277).

Table S6 VOC concentrations stratified by environmental tobacco smoke (ETS) tracers.

Table S7 Factor loadings and variance apportionments for VOCs in Detroit homes (N = 126).

Table S8 Factor loadings and variance apportionments for VOCs in Detroit homes based on 325 seasonal visits.

Table S9 Factor loadings and variance apportionments for VOCs in Detroit homes during spring (n = 105).

Table S10 Factor loadings and variance apportionments for VOCs in Detroit homes during summer (n = 102).

Table S11 Factor loadings and variance apportionments for VOCs in Detroit homes during fall (n = 65).

Table S12 Factor loadings and variance apportionments for VOCs in Detroit homes during winter (n = 53).

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