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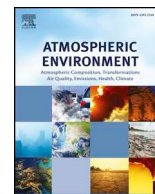
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Assessment of indoor volatile organic compounds in Head Start child care facilities



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ABSTRACT

Exposure to volatile organic compounds (VOCs) in child care environments has raised a public concern. This study aimed to characterize indoor VOCs in four facilities of Head Start programs in Kansas city, Missouri, investigate seasonal and spatial variations in VOC levels, and assess health risks associated with children's VOC exposure. In total, 49 VOCs including aromatic and aliphatic hydrocarbons, aldehydes, glycol ethers, esters and chlorinated hydrocarbons were identified and quantified in the facilities. Significant differences were noted for the VOC concentrations among the facilities. Toluene was the most abundant aromatic hydrocarbon detected in all the air samples, with a narrow median concentration range of 2.17–3.07 $\mu\text{g}/\text{m}^3$. 2-(2-Methoxyethoxy)ethanol never reported in prior research was detected in only one facility in this study, with the median (range) concentration of 0.83 (< LOD, 5.64) $\mu\text{g}/\text{m}^3$. The VOC concentrations in ground-floor classrooms differed significantly from those in basement classrooms. The VOC profiles varied substantially between fall and winter. Identified emission sources of VOCs included vehicle-related emission, solvent-related emission, building materials, personal care products and household products. Through health risk assessment, potential carcinogenic compounds (i.e., benzene, ethylbenzene, naphthalene, 1,4-dichlorobenzene, tetrachloroethylene and trichloroethylene) were of concern as the total cancer risk exceeded 10^{-6} . Future research on children's chronic exposure to these pollutants needs to further assess their possible additive and/or synergistic effects on children's health and development.

1. Introduction

Child care and preschool centers accommodate children aged up to five years for 5–10 h per day, and 5 days per week, making them the most time-spent indoor environments outside of residential buildings. The time spent in such centers hints at the importance of indoor exposure in those settings. Recent studies have provided strong evidence that children's potential exposure to hazardous volatile organic compounds (VOCs) is occurring in child care centers (Roda et al., 2011; Hoang et al., 2016; Hwang et al., 2017; Jia et al., 2019). Given the relatively large amount of time children spend indoors in early education centers, and the presence of hazardous VOCs in those centers, research aimed at identifying and examining VOCs in indoor air is

important.

Prior research has characterized various major constituents of indoor air, such as benzene, toluene, ethylbenzene, and xylenes (collectively known as BTEX), dichlorobenzenes, nitrogen dioxide, and formaldehyde in child care facilities (St-Jean et al., 2012; Hoang et al., 2016). Exposure to these indoor air contaminants has been linked to adverse effects on health. For example, BTEX often used as solvents in consumer products including cleaning agents, paint thinners and vanishes can cause eye and skin irritation, central nervous system depression and effects on respiratory system (ATSDR, 2004). Dichlorobenzenes often used as ingredients in moth balls and bathroom deodorizers have been associated with eye and respiratory tract irritation and increases in diagnosed asthma (Rumchev et al., 2004). Vehicle-

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related nitrogen dioxide at high levels near child care centers was shown to lead to development of wheezing bronchitis, diagnosed asthma, ear, nose and throat infections (Pershagen et al., 1995; Brauer et al., 2002). Exposure to formaldehyde which is often released from rigid polyurethane foams, adhesives, coatings and inks has been correlated with increased airway inflammation and decreased lung function in asthmatic children (McGwin et al., 2009). In addition, a growing body of scientific evidence indicates that exposure to many of these chemicals could negatively affect child development through altered endocrine function (Meeker, 2012). Compared to adults, children can be exposed to more air pollution because they breathe more air per pound body weight and surface area of the respiratory tract. Their exploratory behaviors may also put them at increased risk from contaminants in the environment. Children's heightened susceptibility to environmental assaults is attributable to their immunological, physiological, and neurological immaturity, which also translates into greater health risks.

Prior studies conducted in the U.S. have been focused on several hazardous indoor VOCs including BTEX and brominated flame retardants in early education and child care facilities (Bradman et al., 2014; Quirós-Alcalá et al., 2016). Recently, Hoang et al. (2016) reported the presence of 38 VOCs in center-based and home-based child care facilities located in California. In the present study, we monitor a wide range of VOCs using a thermal desorption sampling and analytical method developed for determination of a wide range of volatility and polarity of VOCs. To our knowledge, the present study is one of the most comprehensive studies that document the presence of a wide range of VOCs in child care facilities (i.e., Head Start facilities) in the U.S., identify VOC sources, and report the estimation of VOC-associated human health risks. Head Start programs provide free learning and development services to children aged up to five years from low-SES (socioeconomic status) families in the U.S. Evidence has begun to emerge that exposure to VOCs is correlated with SES (Adamkiewicz et al., 2011; Tyrrell et al., 2013). Despite increasing awareness and publicity of environmental threats to children associated with VOC exposure, limited data are available on indoor air quality in Head Start child care settings and its influence on young low-SES children's health, development, and learning ability. The objectives of this study were to 1) identify and quantify indoor VOCs in four Head Start facilities located in Kansas City, Missouri, 2) compare the targeted VOC levels within and between the facilities, and 3) estimate human health risks associated with the targeted VOCs.

2. Materials and methods

2.1. Site information

In the present study, we worked closely with Mid-America Regional Council (MARC, Kansas City, Missouri) and Missouri Head Start Programs to select representative child care facilities. Facilities were selected based on building size and type (good vs. poor ventilation; proximity to low/high traffic or industrial area), use of cleaning supplies purchased for the facilities (e.g., public school-used cleaning supplies vs. center-specific cleaning supplies). Additionally, information including age, building materials and renovations of the facilities was obtained (Table S1, Supplementary data). Four facilities of Head Start programs were selected, and their locations are shown in Fig. S1 (Supplementary data). Our sampling was designed to sample the eligible facilities that are as different as possible to capture the gradient of potential indoor air exposure.

2.2. Air sampling and GC-MS analysis of VOCs

The indoor air sampling was carried out in ten classrooms at the four Head Start facilities in Kansas City, Missouri. The air-sampled classrooms include one playground, seven ground-floor learning rooms

and two basement learning rooms. All the air samples were collected in the presence of children in the classrooms. The VOC sampling was performed by passing approximately 70 L of air at flow rate of 0.18 L/min through pre-conditioned sampling tubes (part no. MX062131, SKC Inc., Pennsylvania, USA) containing three layers of sorbent material (20:35 mesh Tenax-TA/Carboxen-1000/Carbosieve). The air pump (SKC 222-3, SKC Inc., Pennsylvania, USA) was deployed at a location inside the classroom, not covered or obstructed by furniture and other teaching supplies. The air sampling tube connected to the pump was positioned at 1 m above the floor (near the heights of children's noses). After 6 h of sampling between 9:00 a.m. and 3:00 p.m., the tube was disconnected from the pump, and carefully sealed in a clean glass container, stored at -20°C until analysis. The analysis of VOCs was to be conducted within seven days. Each classroom was sampled during the period from mid-October in 2014 to late January in 2015. The field blanks were included for every sampling date for quality control purpose.

The analysis of VOCs was performed using CDS Dynatherm 9300 thermal desorption system (CDS, Oxford, PA) which is interfaced to Agilent 6890N gas chromatograph coupled with 5973 quadrupole mass spectrometer (Agilent Technologies, Santa Clara, CA) as previously described by Vu et al. (2018). The analytical method previously developed was optimized for 73 VOCs (Table S2, Supplementary data). Calibrations prepared using seven concentrations (1.0, 2.5, 5.0, 10, 25, 50, 100 $\mu\text{g}/\text{mL}$) of the mixed standard solutions were employed to quantify the 73 targeted analytes. The preparation of calibrations was performed by spiking 1 μL of each mixed standard solution into pre-conditioned sampling tubes followed by the thermal desorption and GC-MS procedure. The correlation coefficients (R^2) of the calibrations ranged from 0.982 to 0.999. The method validation was performed as described in Vu et al. (2018). Field blank and laboratory blank samples were used to determine background contamination during the transportation of the samples and sample-to-sample carryover. Volatile organic compounds were identified by comparison of their mass spectra and retention times with those of authentic reference standards.

2.3. Estimation of health risks

The risk assessment process assists in estimating the likelihood of occurrence of adverse child health effects resulting from exposure to chemical pollutants in child care facilities. The process is focused on chronic exposure to VOCs that may cause cancer or other health effects, rather than on acute toxicity owing to abrupt exposure to VOCs. The adverse health effects of inhalation exposure to hazardous VOCs for non-cancer and cancer risks in the four investigated Head Start facilities were estimated by following the U.S Environmental Protection Agency (EPA) approach. The Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual - Part F was employed for the risk assessment process (EPA, 2009). The evaluation of non-cancer and cancer risks for inhalation pathway was detailed in Table S3 of Supplementary data.

2.4. Statistical analysis

The chromatographic data obtained in the analysis of VOCs were processed with MSD Chemstation (Agilent Technologies, Santa Clara, CA). Statistical analysis was performed with XLSTAT Premium 19.5 (Addinsoft, Paris, France). The nonparametric Kruskal – Wallis test was used to compare volatile compositions of the four Head Start facilities in order to test the null hypothesis that all the four facilities originate from the same distribution against the alternative hypothesis that at least two of the facilities vary in regard to location. Multiple pairwise comparisons were determined using the Dunn test. The Mann – Whitney U test was applied to test whether two independent samples (collected from ground floors, basement, teaching rooms, playing rooms) are from the same population (whether medians vary significantly). A significant

Table 1
Descriptive statistics of indoor VOC concentrations ($\mu\text{g}/\text{m}^3$) in the studied Head Start facilities.

VOC	School A										School B										School C										School D									
	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF										
123TMB	0.09	0.03	0.04	0.10	0.15	100	0.13	0.05	0.05	0.14	0.21	100	0.09	0.02	0.06	0.09	0.14	100	0.15	0.13	0.05	0.11	0.49	100																
124TMB	0.33	0.13	0.14	0.32	0.56	100	0.50	0.21	0.17	0.52	0.81	100	0.30	0.07	0.22	0.28	0.46	100	0.63	0.64	0.17	0.40	2.15	100																
135TMB	0.08	0.03	0.04	0.07	0.15	100	0.11	0.05	0.04	0.12	0.18	100	0.06	0.03	0.04	0.06	0.13	100	0.16	0.18	0.04	0.08	0.60	100																
MeS	0.01	0.01	nd	0.01	0.02	85	0.01	0.00	nd	0.01	0.02	88	0.01	0.01	nd	0.01	0.02	91	0.01	0.01	nd	0.01	0.03	80																
Benzene	0.08	0.10	nd	0.01	0.25	62	0.06	0.11	nd	0.01	0.43	69	0.29	0.69	nd	0.01	2.27	55	0.07	0.16	nd	0.01	0.53	70																
Cumene	0.12	0.11	0.02	0.07	0.40	100	0.09	0.09	0.02	0.06	0.40	100	0.12	0.16	0.02	0.07	0.51	100	0.08	0.05	0.02	0.05	0.14	100																
DEtB	0.27	0.09	0.13	0.29	0.46	100	0.35	0.12	0.17	0.37	0.54	100	0.25	0.06	0.18	0.24	0.37	100	0.38	0.28	nd	0.34	1.02	90																
EB	0.41	0.23	0.12	0.38	0.96	100	0.36	0.25	0.08	0.33	1.06	100	0.38	0.39	0.13	0.27	1.50	100	0.34	0.14	0.17	0.36	0.58	100																
EtT	0.33	0.12	0.16	0.31	0.51	100	0.29	0.17	0.11	0.23	0.61	100	0.29	0.12	0.19	0.24	0.57	100	0.40	0.22	0.16	0.37	0.92	100																
MPX	0.35	0.14	0.15	0.38	0.54	100	0.39	0.17	0.15	0.48	0.58	100	0.23	0.10	0.10	0.24	0.40	100	0.38	0.18	0.17	0.34	0.67	100																
NAP	0.45	0.45	0.11	0.35	1.82	100	0.16	0.05	0.08	0.15	0.27	100	0.17	0.06	0.11	0.16	0.31	100	0.32	0.27	0.06	0.22	0.94	100																
PtB	0.17	0.11	0.05	0.15	0.44	100	0.16	0.12	0.05	0.15	0.55	100	0.16	0.17	0.05	0.11	0.56	100	0.17	0.11	0.09	0.13	0.44	100																
OXY	0.27	0.10	0.12	0.30	0.39	100	0.30	0.13	0.12	0.37	0.44	100	0.19	0.08	0.08	0.20	0.31	100	0.31	0.13	0.13	0.31	0.55	100																
Styrene	1.08	0.86	0.25	0.63	2.83	100	0.64	0.85	0.08	0.36	3.63	100	1.04	1.36	0.13	0.54	4.39	100	0.65	0.42	0.17	0.52	1.15	100																
Toluene	2.78	1.30	1.37	2.39	5.70	100	2.74	1.05	0.87	3.07	4.15	100	3.65	3.80	0.94	2.17	13.36	100	3.29	1.35	1.61	2.94	5.45	100																
Decane	0.38	0.16	0.13	0.40	0.60	100	1.19	0.66	0.32	1.12	2.58	100	0.55	0.33	0.15	0.57	1.27	100	0.49	0.36	0.15	0.41	1.30	100																
Dodecane	0.92	1.17	0.33	0.64	4.74	100	1.49	0.86	0.38	1.72	3.04	100	1.03	0.64	0.44	0.78	2.23	100	0.41	0.19	0.17	0.35	0.77	100																
HexD	0.57	0.15	0.33	0.55	0.98	100	0.35	0.08	0.20	0.33	0.50	100	0.41	0.08	0.30	0.41	0.53	100	0.42	0.13	0.30	0.39	0.72	100																
Nonane	0.25	0.23	nd	0.31	0.66	69	0.76	0.52	nd	0.71	1.96	94	0.25	0.23	nd	0.25	0.71	73	0.38	0.41	nd	0.30	1.38	80																
Octane	0.89	0.78	nd	0.87	2.39	69	0.62	0.71	nd	0.34	2.16	56	0.27	0.54	nd	nd	1.50	18	1.37	2.35	nd	0.47	7.68	50																
TetD	1.86	0.72	0.88	1.52	3.01	100	1.70	0.73	0.63	1.57	3.46	100	0.83	0.28	0.56	0.78	1.60	100	1.25	0.61	0.79	0.98	2.71	100																
Tridecane	0.85	0.98	0.16	0.49	3.95	100	0.64	0.48	0.00	0.73	1.49	94	0.69	0.36	0.31	0.55	1.29	100	0.19	0.18	nd	0.17	0.60	70																
Undecane	0.51	0.32	nd	0.46	1.30	92	1.26	0.64	0.19	1.19	2.70	100	1.14	0.99	0.36	0.82	3.43	100	0.66	0.40	0.24	0.59	1.26	100																
α -Pinene	0.68	0.79	0.01	0.40	2.69	69	0.21	0.34	nd	nd	1.11	38	1.18	0.92	nd	1.39	3.10	73	1.87	2.88	nd	0.58	9.22	60																
Limonene	2.43	3.19	0.62	1.66	12.91	100	1.70	1.60	0.31	0.90	5.06	100	10.38	11.36	1.68	4.20	31.32	100	15.51	11.67	0.78	20.18	30.60	100																
2BE	1.27	1.72	nd	nd	4.76	38	1.67	2.79	nd	0.44	9.18	50	0.40	0.53	nd	nd	1.29	36	0.34	0.66	nd	nd	1.97	20																
2EtH	1.76	1.69	nd	0.98	4.82	77	1.45	1.54	nd	1.24	6.50	88	2.27	2.08	nd	1.55	7.72	91	0.85	0.98	nd	0.42	2.50	60																
22EE	1.00	0.85	nd	0.74	2.09	77	nd	-	nd	nd	nd	0	0.28	0.26	nd	0.25	0.59	55	0.55	0.60	nd	0.24	1.68	70																
22MEE	1.51	1.56	nd	0.83	5.64	77	nd	-	nd	nd	nd	0	nd	-	nd	nd	nd	0	nd	-	nd	nd	nd	0																
BuE	0.06	0.10	nd	nd	0.27	46	0.01	0.01	nd	nd	0.05	19	0.06	0.13	nd	nd	0.33	27	0.04	0.09	nd	nd	0.24	30																
BZALD	3.07	1.94	1.44	2.08	6.83	100	1.43	0.64	0.77	1.15	2.72	100	1.75	1.02	0.74	1.47	4.09	100	1.50	0.83	0.89	1.19	3.43	100																
Decanal	0.65	0.40	nd	0.57	1.38	92	0.45	0.18	0.29	0.37	0.95	100	0.94	0.26	0.64	1.01	1.47	100	0.70	0.19	0.46	0.67	1.10	100																
Heptanal	nd	-	nd	nd	nd	0	nd	-	nd	nd	nd	0	0.21	0.15	nd	0.24	0.44	73	0.02	0.06	nd	nd	0.19	10																
Nonanal	1.45	0.62	nd	1.41	2.60	92	0.91	0.33	0.33	0.97	1.67	100	2.21	0.54	1.27	2.27	3.02	100	1.32	0.65	nd	1.47	2.53	90																
Octanal	0.22	0.15	nd	0.20	0.53	85	0.20	0.17	nd	0.19	0.52	69	0.41	0.09	0.30	0.42	0.58	100	0.21	0.09	0.10	0.18	0.36	100																
ACPH	0.38	0.13	0.20	0.34	0.62	100	0.29	0.17	nd	0.33	0.54	81	0.34	0.10	0.20	0.33	0.50	100	0.28	0.16	nd	0.30	0.49	90																
2EtHS	0.28	0.12	0.11	0.26	0.48	100	0.20	0.19	0.05	0.14	0.81	100	0.50	0.26	0.19	0.47	1.14	100	0.29	0.12	0.18	0.29	0.59	100																
BuA	0.22	0.16	nd	0.26	0.45	77	0.26	0.20	nd	0.26	0.61	75	0.47	0.49	nd	0.30	1.51	82	1.03	1.19	0.23	0.49	3.97	100																
HMS	0.04	0.03	0.02	0.03	0.12	100	0.11	0.10	0.04	0.09	0.45	100	0.50	0.26	0.22	0.46	1.03	100	0.08	0.04	0.02	0.07	0.17	100																
14DCB	0.30	0.29	0.08	0.13	1.02	100	1.55	1.01	0.18	1.61	3.85	100	0.11	0.05	0.05	0.11	0.23	100	0.31	0.18	0.08	0.27	0.62	100																
PERC	0.15	0.21	0.03	0.09	0.84	100	0.70	0.50	0.09	0.63	1.87	100	0.11	0.06	0.04	0.09	0.22	100	0.12	0.07	0.05	0.11	0.30	100																

(continued on next page)

Table 1 (continued)

VOC	School A						School B						School C						School D					
	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF	Mean	SD	Min	Med	Max	%DF
TCE	0.12	0.24	nd	nd	0.82	46	0.45	0.43	nd	0.24	1.40	81	0.16	0.26	nd	nd	0.65	36	0.20	0.30	nd	nd	0.77	40
HA	0.45	0.50	nd	nd	1.16	38	0.72	0.48	nd	0.91	1.45	69	0.52	0.44	nd	0.79	1.06	55	0.28	0.44	nd	nd	1.19	20
BZTZ	0.13	0.07	0.06	0.11	0.29	100	0.08	0.03	0.03	0.07	0.12	100	0.14	0.05	0.08	0.14	0.26	100	0.11	0.06	0.07	0.08	0.27	100

SD stands for standard deviation.

Med stands for median value.

%DF is the detection frequency of a VOC.

nd: below LOD value. LOD and LOQ values are shown in Table S2 (Supplementary data).

"-": not calculated.

Abbreviations: 1,2,3-trimethylbenzene, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene, MeS = α -methylstyrene, DEtB = m-,o-,p-diethylbenzenes; EtB = ethylbenzene, MPX = m/p-xylenes, EtT = m-,o-,p-ethyltoluenes, HexD = hexadecane, TetD = tetradecane, Limonene = D-limonene, 2BE = 2-butoxyethanol, 2EtH = 2-ethylhexanol-1, 22EE = 2-(2-ethoxyethoxy)ethanol, 22MEE = 2-(2-methoxyethoxy)ethanol, BuE = butyl ether, BZALD = benzaldehyde, ACPH = acetophenone, 2EtHS = 2-ethylhexyl salicylate, BuA = butyl acetate, HMS = homosalate, 14DCB = 1,4-dichlorobenzene, PERC = tetrachloroethylene, TCE = trichloroethylene, HA = heptanoic acid, BZTZ = benzothiazole.

level of $p < 0.05$ was used in all the statistical tests. Box and whisker plots were employed to graphically summarize the differences in VOC concentrations at the four facilities. The boxes represent the interquartile ranges (25th and 75th), and the whiskers indicate the minimum and maximum values.

Principal component analysis (PCA) with Varimax rotation (Kaiser normalization) was employed for the VOC dataset to identify potential sources of VOCs. Factors with eigenvalues greater than one that consisted of at least one variable with a loading factor over 0.5 were considered for further analysis. The PCA was implemented using XLSTAT.

3. Results and discussion

3.1. Indoor air concentrations of VOCs

In total, 49 VOCs including aromatic and aliphatic hydrocarbons, aldehydes and ketones, glycol ethers, esters and halocarbons were identified and quantified in the investigated child care facilities. A total of 50 air samples were collected in 10 classrooms in the four Head Start facilities, which accommodated approximately 680 children. The descriptive statistics for the indoor concentrations of the VOCs is demonstrated in Table 1. Comparison of VOC profiles among the four Head Start facilities using the nonparametric Kruskal–Wallis test was made. Significant differences were noted for the concentrations of 25 VOCs among these facilities ($p < 0.05$) (Fig. S2, Supplementary data).

3.1.1. Aromatic hydrocarbons

Among the chemical classes, aromatic hydrocarbons (i.e., benzene derivatives) accounted for 41% (20 out of 49) of the total detected VOC compositions (Table 1). Toluene was detected in all the indoor air samples collected, with a narrow median concentration range of 2.17–3.07 $\mu\text{g}/\text{m}^3$ among the facilities. Also, the toluene was the most abundant aromatic hydrocarbon targeted in all the facilities. This is in agreement with prior investigations into VOCs in early education centers and elementary schools in the U.S. (Hoang et al., 2016; Quirós-Alcalá et al., 2016; Raysoni et al., 2017). Along with toluene, the other aromatic hydrocarbons except benzene, α -methylstyrene and diethylbenzenes were detected in all the indoor samples. Benzene was found in approximately two thirds of the indoor samples. In the present study, the contribution of each BTEX component to the total amount of BTEX was also calculated to evaluate the determinants of BTEX exposure. Fig. 1 displays the contribution percentage of benzene, ethylbenzene, xylenes and toluene to this total amount in the four facilities. The proportion of each BTEX component was found to be similar among the facilities. Prior research on indoor BTEX in a child care center in Spain revealed a similar pattern (Esplugues et al., 2010). The comparison of VOC profiles among the facilities showed that the concentrations of 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene and naphthalene varied significantly ($p < 0.05$) (Table 1, Fig. S2). The median concentration of naphthalene in School A was twice as much as those in School B and School C.

Through Spearman correlations, most of the aromatic hydrocarbons were found to be positively correlated with each other ($r > 0.4$, Table S4, Supplementary data). The highest correlations obtained were between m/p-xylenes and o-xylene ($r = 0.98$, $p < 0.0001$), 1,2,3-trimethylbenzene and 1,2,4-trimethylbenzene ($r = 0.96$, $p < 0.0001$), styrene and cumene ($r = 0.95$, $p < 0.0001$).

3.1.2. Aliphatic hydrocarbons

Eight alkanes listed as the aliphatic hydrocarbons were found in the Head Start facilities. These VOCs are petroleum-associated hydrocarbons, and can also be found in organic solvents, fuels, paints, and detergents (Lagoudi et al., 1996). All the aliphatic hydrocarbons reported were present in more than 90% of the indoor samples collected, except octane and nonane. Significant differences were noted for the

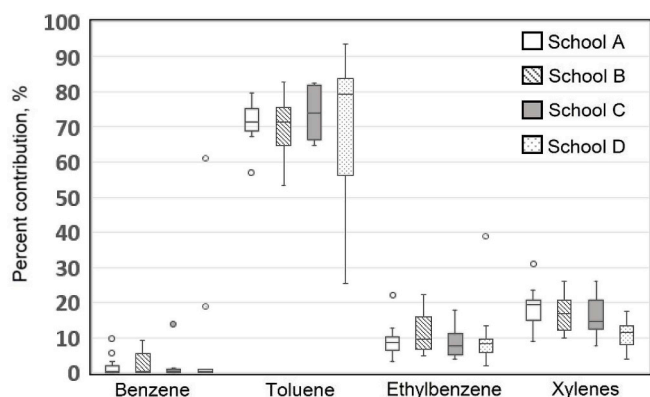


Fig. 1. Contribution percentage of BTEX components among the four Head Start facilities.

concentrations of all the aliphatic hydrocarbons targeted among the facilities, with an exception of octane ($p < 0.05$). The highest levels of decane, dodecane, nonane and undecane in School B among the selected facilities may be due to its proximity to traffic, gas stations (Table S1) or commercial sources.

3.1.3. Terpenes

The results showed the presence of cyclic terpenes, such as α -pinene and D-limonene in the Head Start classrooms. Significant variations were found for the concentrations of these two compounds ($p < 0.05$). As shown in Table 1, D-limonene was detected with a wide range of concentrations. Particularly, School D had 22 times as much of D-limonene ($20.18 \mu\text{g}/\text{m}^3$) as School B ($0.9 \mu\text{g}/\text{m}^3$). These compounds are often used in cleaning products and air fresheners for pine scent (α -pinene) and citrus scent (D-limonene) (Bennett et al., 2011). The differences in use frequency of limonene and pinene-containing products could be responsible for the significant variations in concentrations of these compounds among the facilities.

3.1.4. Glycol ethers and alcohols

The results also yielded evidence of several glycol ethers and alcohols. 2-Butoxyethanol, 2-ethylhexanol-1, 2-(2-ethoxyethoxy)ethanol (22EE) and 2-(2-methoxyethoxy)ethanol (22MEE) are widely used as ingredients in liquid soaps, cosmetics, household cleaners, dry-cleaning agents, paint thinners, coatings, inks and spot removers (Bennett et al., 2011). The former two VOCs were detected in all the facilities, and no significant variations were noted for these compounds across the four facilities ($p > 0.05$). One prior study reported the presence of 2-butoxyethanol and 2-ethylhexanol-1 at median concentrations of 2.9 and $1.6 \mu\text{g}/\text{m}^3$, respectively, in child care facilities in California (Hoang et al., 2016). 22MEE was found only in School A (77% of the indoor samples collected), with the median (range) concentration of $0.83 (< \text{LOD}, 5.64) \mu\text{g}/\text{m}^3$. The presence of 22MEE-containing products related to the maintenance activity outside School A during the sampling time may explain the detection of this VOC (Table S1). 22EE was found in three of the facilities not including School B. The results showed that the indoor concentrations of 22EE and 22MEE varied significantly among the four facilities ($p < 0.05$). To our knowledge, the presence of 22EE and 22MEE has not been reported in Head Start or early education facilities, child care facilities, or schools in prior research.

3.1.5. Aldehydes and Ketones

Acetophenone, benzaldehyde and aliphatic aldehydes, such as octanal, nonanal and decanal were detected in the four facilities. Heptanal was detected in School C and School D. These compounds are often used as components in flavors, perfume and cosmetic products. Prior research suggested positive correlations between aliphatic aldehydes

and use of air fresheners (Hoang et al., 2016). The Spearman correlation showed that these aliphatic aldehydes were positively correlated with each other ($r > 0.4$, Table S5, Supplementary data). The results also showed that the four facilities differed significantly on the indoor concentration levels of benzaldehyde, heptanal, octanal, nonanal and decanal ($p < 0.05$).

3.1.6. Esters

We also detected three esters, namely 2-ethylhexyl salicylate, butyl acetate and homosalate (homomenthyl salicylate), which were found in all four facilities. Particularly, 2-ethylhexyl salicylate and homosalate often used in personal care products, such as sunscreens, were detected in 100% of the samples collected. Significant differences were found in the indoor concentrations of these esters among the facilities ($p < 0.05$). The median indoor concentration of homosalate in School C ($0.46 \mu\text{g}/\text{m}^3$) was 4–14 times higher as compared to the other facilities. The results also showed that School C had three times as much level of 2-ethylhexyl salicylate as School B. A growing concern about skin cancer has resulted in increased use of sunscreens in children, with many early education programs, child care centers and schools having policies requiring application of sunscreen for outdoor play (Geller et al., 2003; Reynolds et al., 2012). Despite an effective chemical ultraviolet filter, homosalate has been shown to be an antiandrogen and an estrogen agonist in vitro (Ma et al., 2003; Schreurs et al., 2004).

3.1.7. Chlorinated hydrocarbons

Among the chlorinated hydrocarbons detected in the present study, PERC (tetrachloroethylene) is known to interfere with endocrine systems, and is probably carcinogenic to humans (Carney et al., 2006; Guha et al., 2012). The compound was detected in 100% of the indoor air samples collected. The median indoor concentration of PERC in School B ($0.63 \mu\text{g}/\text{m}^3$) was found to be approximately seven times as high as those in the other Head Start facilities. PERC has long been recognized as an effective dry-cleaning chemical, and it is widely used in dry-cleaning facilities in the U.S. Among the Head Start facilities that participated in this study, School B is located opposite a dry-cleaning business (Table S1, Supplementary data). This could explain the elevated indoor concentration levels of PERC observed in School B. Another chlorinated hydrocarbon included in our study is 1,4-dichlorobenzene which was also found in 100% of the indoor samples. As described in Table 1, the median indoor concentration of this compound in School B ($1.61 \mu\text{g}/\text{m}^3$) was 12-fold higher than those in School A ($0.13 \mu\text{g}/\text{m}^3$) and School C ($0.11 \mu\text{g}/\text{m}^3$). Prior research revealed the presence of 1,4-dichlorobenzene at a median concentration of $0.81 \mu\text{g}/\text{m}^3$ in an elementary school located in Texas (Raysoni et al., 2017).

3.1.8. Comparison with prior research

The pooled VOC dataset was used to compare with prior research in child care facilities and elementary schools in North America (Fig. 2). BTEX were selected for the comparison because these VOCs were reported in all the previous studies. We observed that toluene and ethylbenzene levels in the present study were comparable to those in the studies conducted by Hoang et al. (2016) and Raysoni et al. (2017), while lower than those recorded by St-Jean et al. (2012) and Quirós-Alcalá et al. (2016). In addition, the levels of benzene and xylenes were substantially lower than those documented in the prior investigations. Multiple factors were ascribable to the differences in BTEX levels among the studies, including meteorological parameters, location, methods of air sampling and VOC analysis, characteristics of school supplies and examined buildings (structure, materials, age, ventilation and heating systems), and housekeeping activities.

3.2. Comparison of indoor VOC levels between classrooms

In the present study, the indoor air samples were collected in classrooms located either on ground floors or in basements. Comparison

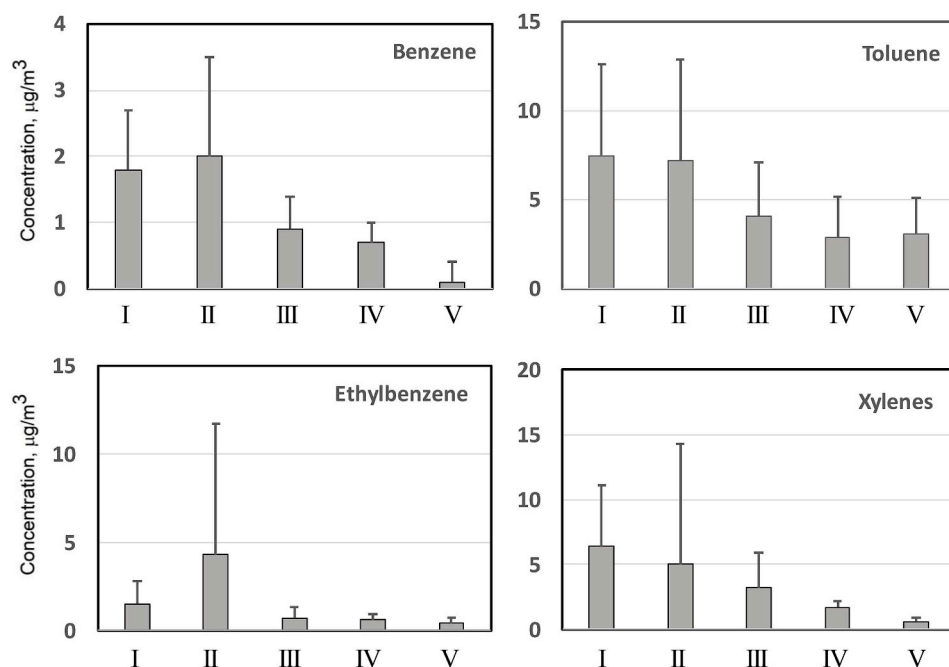


Fig. 2. Comparison of mean BTEX levels between studies in North America. Error bars represent standard deviation of the mean. I, II, III, IV, and V stand for studies conducted by St. Jean et al. (2012) in Canada, Quirós-Alcalá et al. (2016) in Washington D.C., Hoang et al. (2016) in California, Raysoni et al. (2017) in Texas, and the present study in Missouri, respectively. The IV and V results were reported as pooled means and standard deviations.

of VOC levels between the rooms in each Head Start facility was drawn using the nonparametric Mann – Whitney U test. Among the Head Start facilities investigated in this study, both School A and D have the two types of classrooms.

In School A, in total, 48 VOCs not including heptanal were detected in the classrooms. Of these, 2-butoxyethanol, 2-ethylhexanol-1, benzene and naphthalene were the compounds with concentrations measured in the ground-floor and basement rooms differing significantly ($p < 0.05$) (Fig. 3A). The numbers of indoor air samples collected from the investigated ground-floor and basement rooms in this facility were 7 and 6, respectively. The results demonstrated significantly higher concentrations of 2-butoxyethanol measured in the basement classrooms. Unlike 2-butoxyethanol, the other three VOCs were present in the ground-floor classrooms at higher levels as compared to the basement classrooms. The median concentration of 2-ethylhexanol in the ground floor ($3.32 \mu\text{g}/\text{m}^3$) was approximately 10 times as high as that in the basement ($0.36 \mu\text{g}/\text{m}^3$). We observed that benzene and 2-ethylhexanol-1 were present in 85–100% of the samples collected in the ground-floor rooms while not more than half of those collected in the basement rooms were found to contain the two compounds. Naphthalene was detected in all the samples in the two types of classrooms. In School D, five indoor air samples were collected for each type of classrooms, resulting in the detection of 48 VOCs not including 22MEE.

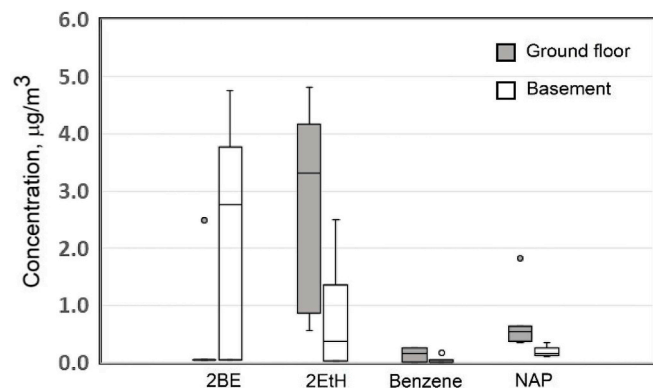


Fig. 3A. Comparison of VOC levels between ground floor ($n = 7$) and basement ($n = 6$) classrooms in School A.

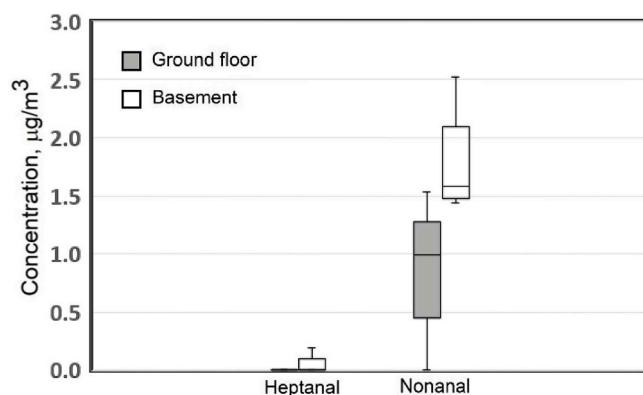


Fig. 3B. Comparison of VOC levels between ground floor ($n = 5$) and basement ($n = 5$) classrooms in School D.

The two types of classrooms varied significantly on the concentrations of heptanal and nonanal ($p < 0.05$). As depicted in Fig. 3B, the median levels of these compounds were higher in the basement than in the ground-floor rooms. Prior research on the influence of basements on indoor air quality of residential places indicated that VOC levels in basements often exceeded those in living areas, such as ground floors (Du et al., 2015). This is because basements in residential places are often used for storage of household cleaners, air fresheners, gasoline-powered equipment and solvents. In the present study, basements are tantamount to ground floors for their use purposes. This suggests that the higher levels of these VOCs in the ground-floor or basement classrooms in the present study could be due to the extensive use and storage of household cleaning products, deodorizers and solvents in this area.

3.3. Seasonal variation of VOC concentrations in the facilities

In our study, the air sampling was performed in fall (October 17 and 23, November 6) and winter (November 20, January 15 and 29). Comparison of VOC levels in the two seasons in each Head Start facility was made using the nonparametric Mann – Whitney U test. Fig. 4 displays seasonal variations of VOC levels in the four facilities.

In School A, five compounds including 22EE, butyl ether, octane,

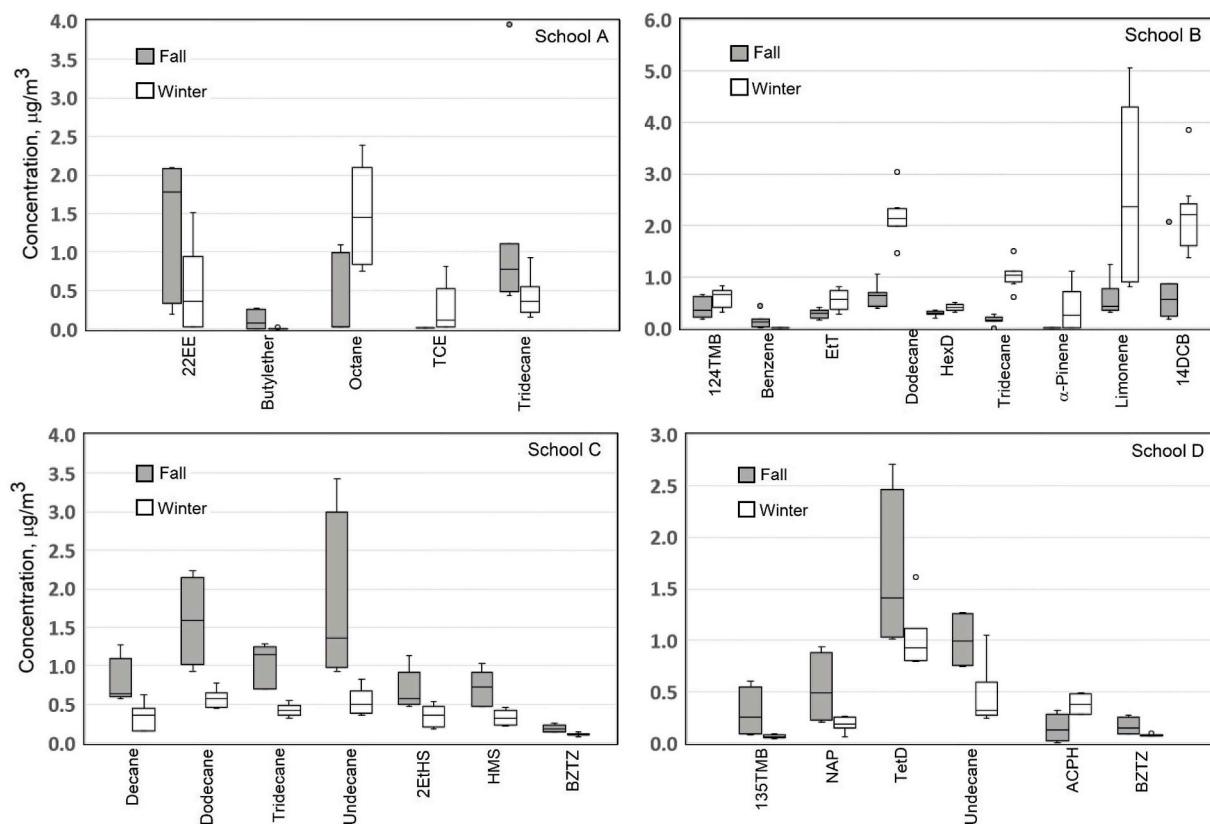


Fig. 4. Seasonal variations of VOC levels in the four Head Start facilities.

trichloroethylene (TCE) and tridecane had the median concentrations that substantially varied between the fall and winter ($p < 0.05$). While 22EE, butyl ether and tridecane were found at higher levels in the fall, octane and TCE were more abundant in the winter. The results also showed that the infrequent detection of octane and the absence of TCE in the indoor air samples collected in the fall may have resulted from higher ventilation in the building. In addition, the detection of TCE in all the samples collected during the winter suggests the presence of an emission source of this chlorinated compound (e.g., spot removers or cleaning solvents) in School A.

In School B, significant differences were noted for the levels of nine compounds (i.e., 1,2,4-trimethylbenzene, benzene, ethyltoluenes, dodecane, hexadecane, tridecane, α -pinene, D-limonene and 1,4-dichlorobenzene) measured in the two seasons ($p < 0.05$). With an exception of benzene, all the other compounds were present at higher median concentrations in the winter than in the fall. One possible reason for higher levels of VOCs is lower ventilation rates in buildings during winter. Prior research has indicated that reduced ventilation rates often resulted in increased concentrations of VOCs associated with indoor sources, such as D-limonene and 1,4-dichlorobenzene (Adgate et al., 2004; Sofuoğlu et al., 2011). Low ventilation rates may also influence aliphatic hydrocarbons (Zuraimi and Tham, 2008; Hoang et al., 2016), suggesting the higher concentrations of dodecane, hexadecane and tridecane measured in the winter in the present study. We observed that no α -pinene was detected in the samples collected in the fall while two-thirds of those in the winter contained the compound. This could be due to the use of pine-scented cleaning products in the winter, which did not occur in the fall.

In School C and School D, in total, 13 compounds had the median concentrations differing significantly between the two seasons ($p < 0.05$). Unlike the pattern demonstrated by the VOC levels in School B, the VOC concentrations in School C and School D were more abundant in the fall, with an exception of acetophenone in School D. As

seen in Fig. 4, several aliphatic hydrocarbons including decane, dodecane, hexadecane, tetradecane, tridecane and undecane contributed to the seasonal variations of VOCs in the two facilities. Among the studied Head Start facilities, School C is the only facility that has an outdoor playground. The application of sunscreen during outdoor play time in the fall may have resulted in the substantially higher levels of 2-ethylhexyl salicylate and homosalate (sunscreen ingredients as described earlier) in the indoor air samples as illustrated in Fig. 4.

3.4. Source identification

The PCA, which has been widely used in atmospheric data processing, was applied to the pooled VOC dataset for source identification. Among the VOCs reported earlier, 37 major compounds were selected for the PCA. Through the analysis, nine factors with eigenvalues exceeding one were extracted (Table S6, Supplementary data). Based on the factor loadings after Varimax rotation (Kaiser normalization), multiple possible sources were identified as contributing to the VOC concentration values at the Head Start facilities (Table S7, Supplementary data). Marker compounds linked to specific sources were determined based on prior research into indoor/outdoor VOC profiles. The source contribution for each factor is summarized in Table 2 while the description of these sources is discussed in S8 of the Supplementary data. The PCA also shows that all these factors explain 82.3% of the total data variance (Table S7, Supplementary data).

3.5. Potential health risks

In the present study, health risk assessment for non-cancer (chronic toxic effects) was performed for trimethylbenzenes, benzene, ethylbenzene, cumene, xylenes, naphthalene, styrene, toluene, 2BE, 14DCB, PERC and TCE. Risk assessment of carcinogenic effects was carried out for benzene, ethylbenzene, naphthalene, 14DCB, PERC and TCE. As

Table 2
Sources of indoor VOCs in the Head Start facilities identified by PCA.

Factors	Sources	Associated VOCs	Reference
1	Vehicle-related emissions and solvent emissions	Trimethylbenzenes, diethylbenzenes, ethyltoluenes, xylenes	Watson et al. (2001) Chin et al. (2014) EPA (2016)
2	Dry-cleaning solvents	PERC, trichloroethylene, decane, nonane	McDermott et al. (2005) Sherlach et al. (2011)
3	Floor materials, computers, and printers	Cumene, ethylbenzene, styrene, 2-ethylhexanol-1	Kagi et al. (2007) Destailats et al. (2008) Kowalska et al. (2015)
4	Personal care products, oil-based paints and adhesives	2-Ethylhexylsalicylate, homosalate, toluene, undecane	Namiński et al. (1992) EPA (2006a)
5	Consumer cleaning products	22EE, 22MEE, benzaldehyde, naphthalene, hexadecane	EPA (2006b)
6	Fragrance products	D-Limonene, α -pinene, butyl acetate	Uhde and Schulz (2015)
7	Off-gassing of building materials, and perfumes, odorants	Octanal, nonanal, decanal	Hodgson et al. (2002) Uhde and Salthammer (2007)
8	Vehicle-related emissions	Benzene	ATSDR (2007)
9	Lubricants, degreasers, paint removers, and mothballs	Dodecane, tridecane, 1,4-dichlorobenzene	Weisel et al. (2008)

Table 3A
Mean cancer risks ($\times 10^{-6}$) for the individual VOCs in the Head Start facilities.

Centers	Benzene	Ethylbenzene	Naphthalene	14DCB	PERC	TCE
School A	0.20	0.08	1.25	0.27	0.08	0.02
School B	0.14	0.07	0.45	1.41	0.35	0.07
School C	0.69	0.09	0.48	0.09	0.05	0.03
School D	0.18	0.06	0.93	0.30	0.06	0.03

Table 3B
Total non-cancer and cancer risks for exposure to the indoor VOCs in the Head Start facilities.

Risk assessment	Child care centers	Mean risk value	95% Confidence interval of the mean	
			Lower bound	Upper bound
Non-cancer _s	School A	0.05	0.03	0.07
	School B	0.07	0.04	0.09
	School C	0.04	0.02	0.05
	School D	0.05	0.03	0.07
Cancer _{s*} ($\times 10^{-6}$)	School A	1.90	0.94	2.85
	School B	2.51	2.08	2.93
	School C	1.43	0.33	2.52
	School D	1.55	0.82	2.27

* Total non-cancer risks were assessed for exposure to trimethylbenzenes, benzene, ethylbenzene, cumene, xylenes, naphthalene, styrene, toluene, 2BE, 14DCB, PERC, and TCE.

** Total cancer risks were assessed for exposure to benzene, ethylbenzene, naphthalene, 14DCB, PERC, and TCE.

seen in Table 3A, the cancer risk values obtained for the individual VOCs in all the centers were lower than 10^{-6} , except for 14DCB in School B and naphthalene in School A. However, total cancer risk for exposure to these chemicals in each facility was found to be higher than 10^{-6} (Table 3B). No significant differences were noted for these total mean values among the facilities ($p > 0.05$). When evaluating the values calculated for Hazard Quotient (Table S3, Supplementary data), we observed that the total non-cancer risk for children in each Head Start facility was found to be lower than one (Table 3B).

Notably, health risks could be underestimated due to the fact that inhalation was merely one of the pathways for exposure to chemicals in child care environments. Other exposure scenarios including ingestion are also important for health risk assessment. Concurrent or sequential exposures to a mixture of VOCs in early education and child care environments could also induce synergistic or antagonistic effects besides the additive effect as assumed for the estimate of health risks.

3.6. Study strengths and limitations

One of the strengths of this study is the simultaneous identification and quantification of a wide range of VOCs, revealing several compounds which have not been characterized previously in early education environments. In addition, our study, which was performed during a period of four months, allowed for an extensive evaluation of potential VOC emissions and seasonal variation of VOC compositions in indoor Head Start facilities. Through health risk assessment, the potential carcinogenic compounds (i.e., benzene, ethylbenzene, naphthalene, 14DCB, PERC, and TCE) were of concern as the associated total cancer risk exceeded the acceptable level of 10^{-6} . Additional research on chronic exposure to the potential carcinogenic compounds should be focused on full assessment of their possible additive and/or synergistic effects. The present study was conducted in the facilities for Head Start programs, which provide early childhood education, health, and nutrition to low-income children and their families (eligible for federal assistance). These facilities reside in diverse neighborhoods within an urban area. Although future research is necessary to examine indoor air quality in early childhood education centers located in middle- or high-SES neighborhoods, the present study represents a unique but rarely studied context.

As described above and in Table S3 (Supplementary data), inhalation Reference Concentrations used for calculating Hazard Quotient for 16 VOCs were found in the EPA's online IRIS system, while those for many others including endocrine disruptors (22MEE, 2-ethylhexanol-1, benzaldehyde) and aliphatic alkanes are not available in the IRIS database and literature. Therefore, limited information about toxicity values has restricted the estimate of non-cancer risks for several VOCs. Another limitation is that detailed information about on-site storage and use frequency of household products, as well as application of pesticides was not obtained. Those data would likely be helpful for identifying variations of VOC compositions and health risk estimates among the studied Head Start facilities. Also, due to limited resources, the air sampling was conducted only during the fall and winter seasons. As seasonal use of household products likely differs, year-round VOC profiling of indoor child care facilities would give a better understanding of indoor VOC sources, and help enable effective reduction of VOC emissions.

In this study, the identification of VOC sources was based on indoor VOC assessment. Future research should consider measurement of not only indoor, but also outdoor VOC concentrations to examine outdoor VOC sources and their contributions to indoor air. This would provide a better characterization of contributors to indoor sources. Exposure to VOCs in home-based child care facilities, which was not assessed in this study, could be different from that in Head Start facilities located in

school buildings. Thus, future studies should identify indoor VOC exposure in different types of buildings.

4. Conclusions

A wide range of VOCs in Head Start facilities located in Kansas City, Missouri were characterized. The findings show that the VOC profiles substantially varied among the facilities, and VOC levels were significantly different between ground-floor and basement classrooms. In addition, VOC levels measured in the fall differed significantly from those in the winter. The identification of VOC emission sources indicated that the VOCs were linked to vehicle-related emission, solvent-related emission, building materials, personal care products and household products. Potentially harmful effects are expected as a result of the exposure to benzene, ethylbenzene, naphthalene, 14DCB, PERC and TCE in the child care facilities. Exposure to the other harmful VOCs including endocrine disruptors (22MEE, 2-ethylhexanol-1, benzaldehyde) and aliphatic alkanes with toxicological information not available in the IRIS database and literature warrants additional health assessment. Based on our data, strategies such as restricted use and proper storage of chemicals and appropriate ventilation should be taken into consideration to improve indoor air quality in child care facilities. Future research should be focused on evaluating the influence of indoor air quality in Head Start child care facilities on children's development and learning ability.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2019.116900>.

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