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Relationships of TVOC with Several Aromatic Hydrocarbon Constituents at Preschool Facilities

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요 약

이 연구의 목적은 유치원에서 총 휘발성 유기화합물의 농도를 평가하고, 총 휘발성 유기화합물 농도와 대표적인 8개 방향족 화합물의 상관관계를 조사하는데 있다. 도시에 위치한 11개 유치원의 실내와 실외에서 각각 30개, 11개의 지역시료를, 시골에 위치한 4개 유치원에서는 각각 10개, 4개의 시료를 테낙스 튜브를 이용하여 오전에 1-2시간 채취하였다. 채취한 시료는 열탈착하여 가스크로마토그래피-질량분석기로 분석하였다. 13가지 물질을 각각의 표준물질로 개별 정량하여 이중 빈번히 발견되는 8가지 방향족 유기화합물은 상관관계 평가에 사용하였다. 총 휘발성 유기화합물은 톨루엔을 기준으로 정량하였다. 도시에 위치한 유치원 실내의 총 휘발성 유기화합물 농도가 높았고, 조사 건수의 50%가 환경부 및 교육인적자원부의 가이드라인(400 μg/m³)을 초과하였다. 도시지역의 유치원 실내 및 실외의 기하평균은 각각 387.9 μg/m³과 134.9 μg/m³이었고, 시골지역 유치원에서는 각각 189.6 μg/m³, 74.4 μg/m³이었다. 톨루엔, 크실렌, 에틸벤젠, 정량한 유기화합물 총합, 총 휘발성 유기화합물은 기하정규분포를 하였다. 벤젠, 톨루엔, 에틸벤젠, 크실렌(BTEX)은 도시에 위치한 유치원에서 농도도 높고, 총 휘발성 유기화합물중 함량도 높았고, 시골지역에서는 농도와 상대적 함량이 낮았다. 도시지역에서는 총 휘발성 유기화합물 중 BTEX의 비중이 25.2%였고 정량한 13가지 유기화합물 중에서는 35.6%를 차지하였다. BTEX 각각 개별물질은 미국 환경보호청이 제시하는 일일 노출 기준량(Reference Concentration; RfC) 보다는 현저히 낮았다. 총 휘발성 유기화합물원 농도는 실내가 실외 보다 높았다(I/O ratio 2.5). BTEX의 상대적 함량도 실내가 실외보다 높아 실내에도 발생원이 있음을 암시하고 있다. 자료 분석결과 유치원 실내의 벤젠은 실외로부터 유입되고 있었고, 톨루엔, 에틸벤젠, 크실렌은 실외뿐 아니라 실내에서도 발생하고 있었다. 정량한 8개 화합물 각각과 총 휘발성 유기화합물의 스피어만 상관계수는 벤젠을 제외하고는 모두 유의하였다. 이중 톨루엔과 크실렌은 총 휘발성 유기화합물과 좋은 상관성(톨루엔 0.76, 크실렌, 0.87)을 나타내었다. 이 연구는 톨루엔과 크실렌이 총 휘발성 유기화합물의 좋은 지표를 사용될 있고, 톨루엔, 에틸벤젠, 크실렌 등 많은 휘발성 유기화합물의 발생원은 실외뿐 아니라 실내에도 있음을 나타내고 있다.

Keywords: benzene, BTEX, indoor air quality, IAQ, indoor, preschool, TVOC, volatile organic hydrocarbons, VOC

I. Background

Environmental contaminants are hazardous to all lives, but especially more detrimental to the children. In regarding to the exposure, physiology, pharmacokinetics and health effect, children differ from adults.[1,2] Also, children are more susceptible to hazardous agents than adults because of their immature immune system.[3] Consequently, children's exposure to environmental contaminant and health effects are great concern.[4-6]

Because of so many volatile organic materials in indoor air, identification and characterization of all constituents seems to be very complicated and unnecessarily makes a burden to the health regulatory agency. So, total parameters or reliable surrogates are often used for the evaluation of indoor air quality (IAQ) instead of determining individual

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concentration of environmental contaminants. One of such parameters is total volatile organic compounds (TVOC) and BETX can be used as surrogates.[7] Aromatic hydrocarbons are a category of TVOC and their health effects are much more studied. Inhalation is considered as the main intake route to aromatic hydrocarbons which accounts for 95% of the total daily intake.[8]

In Korea, Both Ministry of Environment and Ministry of Education and Human Resources Development had developed the guideline as 400 μg/m^3 for the concentration of TOVC in indoor air and class room. Though TOVC rather than individual compounds is regulated by the government, it is necessary for the scientists to examine the relationships between TVOC, surrogates and individual compounds. Furthermore, some kinds of individual compound, for example aromatic benzene, must be considered more importantly than other compounds because of its carcinogenicity.

Recently, there are so many papers on the concentration of TVOC and each compound. But a limited number of studies have been reported for the relationship among individual compounds. For example, Total Exposure Assessment Methodology (TEAM) studies in the eighties[9] and the National Human Exposure Assessment Survey (NHEXAS) in the early nineties[10] were performed in the US. In Europe, EXPOLIS (Exposure Distributions of Adult Urban Populations in Europe) was conducted in 1996-1998.[11,12]

The purposes of this article were to evaluate and test the distributions of the concentration of TVOC and eight aromatic hydrocarbons at the indoor air and the outdoor air of preschool facilities and to examine the relationships between the TVOC and some aromatic hydrocarbons.

II. Methods

Microenvironment concentrations of total volatile organic compound (TVOC) and its constituents were measured in 15 preschool facilities. The participant facilities were randomly drawn from Korean Peninsula. Sampling was done simultaneously at the indoor air and outdoor of the classroom. Total 55 microenvironments were successfully measured; 11 urban located preschools were surveyed for 30 indoor samples and 11 outdoor samples, 4 rural area located preschool classrooms were surveyed for 10 indoor and 4 outdoor samples.

Indoor and outdoor air samples were collected simultaneously using pre-cleaned Tenax tube (60-80 mesh, 250 mg/tube) connected to the low flow rate sampling pumps (Gilian LFS-113DC, Sensidyne, USA). The flow rates were 70~100 ml/min and calibrated before and after the sampling. Most of sampling was done before noon and sampling periods were 60~100 minutes to prevent the possible breakthrough. Tenax tubes were placed at breathing height at a distance of at least 1 m from walls to guarantee sufficient air circulation. At the end of the sampling period they were closed with an impermeable cap and stored at 4-8°C until analyzed.

TVOC and its constituents were analyzed with a gas chromatograph (GC, Hewlett-Packard 6890 plus, USA) - the mass spectrometer detector (MSD, Hewlett-Packard MSD 5973, USA) after thermal desorption (Tekmar 6000, USA) at 250°C and cold condensation at −150°C. The GC was equipped with non-polar columns (HP-5MS, 30 m × 0.25 mm, 0.25 μm phase thickness, Hewlett Packard, USA) with the split ration 20:1 and the flow rate of pure helium carrier gas was 1 ml/min.

The concentration of TVOC was calculated using toluene as a surrogate. Each VOC compound in the sample was identified from the MS total ion chromatogram by reference spectra of the Wiley 275 spectra library (Hewlett-Packard, USA). Each VOC was quantified by linear regression using calibration curves based on calibration solution analysis. Over hundreds of VOC constituents were identified in the samples, but calibration was focused on the set of 13 target VOCs (benzene, toluene, ethyl benzene, xylene, styrene, bromobenzene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, isopropyltoluene, butylbenzene, 1,2,4-trichlorobenzene, 1,2,3-trichlobenzene, naphthalene). Among these VOCs, 8 constituents (benzene, toluene, ethyl benzene, xylene, styrene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, naphthalene) were quantified because of their frequent existence in the field samples. For xylene, meta-xylene was used as standard material of three isomers(Ortho-, meta-, para-).

Airborne concentrations were summarized as an arithmetic mean and a geometric mean. For statistical analysis for ANOVA, correlation and distribution type testing, SigmaStat® 3.0 was run. For the correlation between the surveyed VOCs, Person product moment correlation was calculated. In case of a lognormality testing, data sets were transformed because there was no function to test lognormal distribution directly in SigmaStat®. So, measured concentrations were log-transformed and tested with the 'Kolmogorov-Smirnov test' about the normal distribution. It could be said that the data was lognormal distributed if log-transformed data sets were passed normal distribution test. T-test with the normal distribution or Mann-Whitney Rank Sum test with the non-normal distribution was run for the differences of BTEX concentration between indoor air and outdoor air.

III. Results and Discussion

1. Concentration of TVOC and Distribution Test

TVOC concentrations at urban and rural area were summarized in Table 1. Both the arithmetic mean and the geometric mean (GM) were represented because the distribution patterns were varied as shown in Table 2. Also, the percentages for exceeding the government guidelines (Both IAQ guidelines by Ministry of Environment and Ministry of Education and Human Resources were 400 μg/m^3) were shown. TVOC concentrations at the urban preschool indoor remained the highest level and 50% of surveyed results exceeded the Korean government regulatory limit. The rural indoor air concentration as GM was 189.8 μg/m^3 and 40%

Table 1. Concentration of total volatile organic compounds (TVOC) in preschool facilities

		No. of samples	Concentration, μg/m^3			Percentage of exceeding guideline	GM I/O ratio
			GM(GSD)	AM(SD)	Range		
Urban	Indoor	30	387.9(2.4)	548.0(465.6)	73.34-1927.7	50	2.9:1
	Outdoor	11	134.9(3.4)	276.9(404.4)	13.21-1322.9	18	
Rural	Indoor	10	189.6(5.2)	480.9(581.4)	16.07-1783.0	40	2.5:1
	Outdoor	4	74.4(6.6)	278.3(470.0)	16.94-981.2	25	

note) Indoor air quality standard of Ministry of Environment and Ministry of Education in Korea : 400 μg/m^3

Table 2. Kolmogorov-Smirnov test for normal and lognormal distribution

Tested VOC and/or site			Normal distribution	Lognormal distribution
Non-classified by sampling site	Toluene		Failed (P < 0.001)	Passed (P > 0.200)
	Xylene		Failed (P = 0.001)	Passed (P > 0.200)
	Ehtylbenzene		Failed (P < 0.001)	Passed (P > 0.200)
	Benzene		Failed (P < 0.001)	Failed (P < 0.001)
	1,2,4-trimethylbenzene		Failed (P < 0.001)	Failed (P < 0.001)
	Styrene		Failed (P = 0.004)	Failed (P < 0.001)
	Naphthalene		Failed (P < 0.001)	Failed (P < 0.001)
	1,3,5-trimethylbenzene		Passed (P = 0.051)	Failed (P < 0.001)
	Quantified VOC		Failed (P = 0.002)	Passed (P = 0.153)
	TVOC		Failed (P < 0.001)	Passed (P > 0.200)
Classified by sampling site	TVOC	Urban-Indoor	Passed (P = 0.058)	Passed (P > 0.200)
		Urban-Outdoor	Failed (P < 0.001)	Passed (P = 0.064)
		Rural-Indoor	Passed (P > 0.200)	Passed (P > 0.200)
		Rural-Outdoor	Failed (P = 0.024)	Passed (P > 0.200)
	Quantified VOC	Urban-Indoor	Passed (P = 0.092)	Passed (P > 0.200)
		Urban-Outdoor	Passed (P = 0.056)	Passed (P > 0.200)
		Rural-Indoor	Failed (P = 0.033)	Failed (P = 0.030)
		Rural-Outdoor	Passed (P > 0.200)	Passed (P > 0.200)

was above the Korean regulatory limit.

TVOC concentrations at the urban facilities were higher than those of rural facilities. TVOC concentration as GM in the urban indoor was 387.9 μg/m^3 and the outdoor was 134.9 μg/m^3. TVOC in the rural area was 189.6 μg/m^3, 74.4 μg/m^3 respectively. Both of urban indoor and outdoor air concentration was about 2 times higher those that of rural area. Outdoor air concentration in urban area was 1.8 times higher than that of rural area. Indoor/Outdoor ratio (I/O ratio) in urban preschool was 2.9:1 and 2.5:1 in rural area respectively.

The largest study was carried out from 1980 to 1987 by the US EPA, the so-called 'total exposure assessment methodology' (TEAM) study[13)] which was recently reviewed by Wallace.[14)] According to the EPA TEAM study, Indoor TVOC concentration was 2~5 times higher than that of outdoor in urban and rural area, and the concentration maintained high if a VOC emitting material were used.

Distributions of exposures to individual VOC and corresponding concentrations measured in different microenvironments have frequently been described as log normally distributed or more highly skewed than normal.[15,16)] Distributions of this survey results were generally shown as lognormal but some data subsets were also satisfied with normal distribution criteria as shown in Table 2. A test that passes indicates that the survey data matches the pattern expected if it was drawn from a population with a normal distribution or lognormal distribution whereas a test that fails indicates that the survey data varies significantly from the pattern expected if it was drawn from a population with a normal distribution or lognormal distribution.[17)]

Each of toluene, xylene, ethylbenzene, quantified VOC and TVOC was lognormal distribution rather than normal distribution but 1,2,4-trimehtylbenzene was normally distributed.

If samples were classified by site, TVOC and quantified VOC at every site were lognormal distribution except at rural area. Also, TVOC at urban indoor, rural indoor, quantified VOC at every site except rural indoor satisfied with normal distribution criteria. Fig. 1 illustrates some examples of lognormal distribution.

2. Airborne Concentration of Quantified Aromatic Hydrocarbons

Eight quantified aromatic compounds were shown as bar graphs in Fig. 2. Among these compounds, BTEX concentration and the percentage were higher in urban than in rural area, and higher in indoor air than in outdoor air. The concentration and the

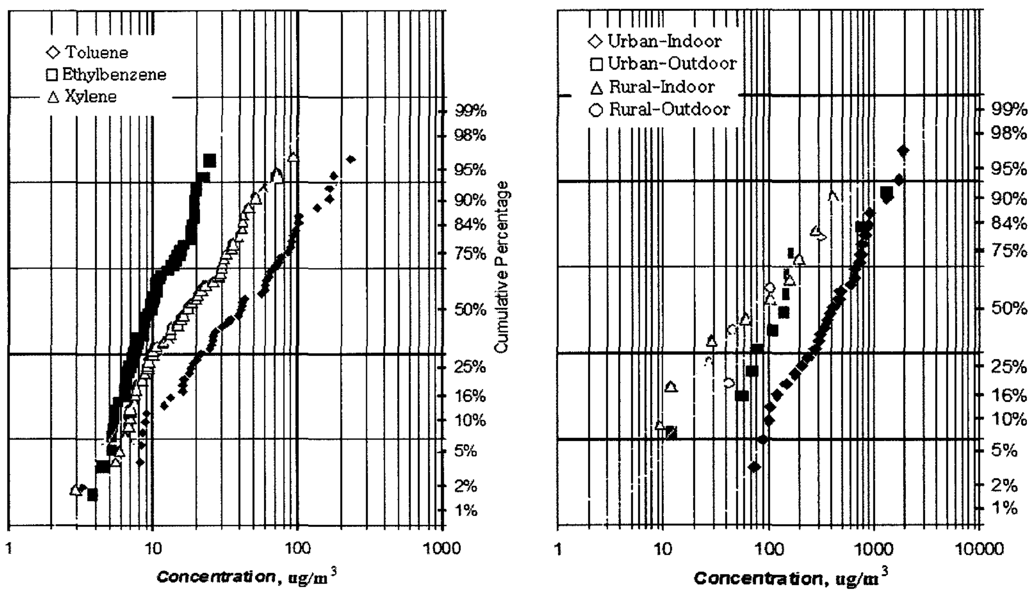

Fig. 1. Illustrations of lognormal distribution of TVOC by site and individual compounds (toluene, ethylbenzene, xylene).

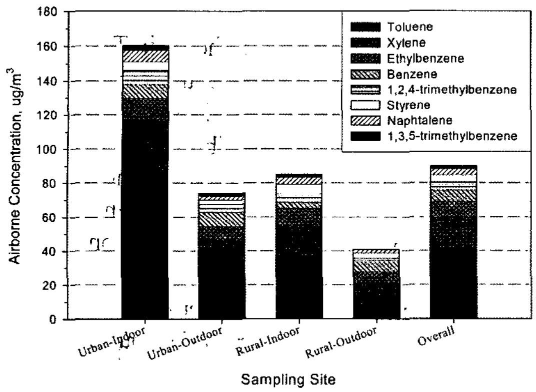

Fig. 2. Quantified aromatic volatile organic compounds by sampling site.

percentage of BTEX occupied the highest portion in urban indoor air and the lowest in rural outdoor air. BTEX occupied 25.2% of TVOC and 35.6% of quantified VOC in urban indoor whereas it was 22.8%, 31.2% in urban outdoor air, respectively. It occupied 14.3% of TVOC and 17.4% of quantified VOC in rural indoor whereas it was 12.1%, 14.2% in urban outdoor air, respectively. The mean percentage of BTEX was 19.2% of TVOC and 24.8% of quantified VOC.

Toluene occupied the highest concentration regardless of sampling site; 86.1 μg/m³, 31.1 μg/m³ at the urban located indoor classroom and outdoor, respectively. 35.6 μg/m³, 13.2 μg/m³ at the rural located indoor classroom and at the outdoor, respectively. The I/O ratio was 2.8:1 at the urban preschool facilities and 2.7:1 at the rural preschool facilities. These results were much lower than the US EPA's Reference Concentration (RfC) for toluene, 5 mg/m³, which was based on the numerous human and animal chronic and subchronic studies (EPA, 2005).[18]

Xylene was the second highest and occupied around a half of that of toluene; 30.3 μg/m³, 15.1 μg/m³ at the urban indoor and outdoor, respectively. 19.1 μg/m³, 7.7 μg/m³ at the rural indoor and out door, respectively. The US EPA's Reference Concentration (RfC) for xylene is 100 μg/m³ based on the impaired motor coordination.[19]

Ethylbenzene was the third highest concentration; 13.5 μg/m³, 8.4 μg/m³ at the urban indoor and outdoor, respectively. 10.6 μg/m³, 6.7 μg/m³ at the rural indoor and out door, respectively. The US EPA's Reference Concentration (RfC) for ethylbenzene is 100 μg/m³ based on the developmental toxicity.[20]

Benzene, a well known carcinogen, was the next; 8.1 μg/m³, 8.6 μg/m³ at the urban indoor and outdoor, respectively. 3.5 μg/m³, 6.1 μg/m³ at the rural indoor and out door, respectively. Contrary to other aromatic substances, outdoor benzene concentration was high in urban and rural area. But there was no statistical differences between sampling site ($p=0.34$). In the TEAM study,[14] benzene concentration in general indoor was up ~10 μg/m³ and outdoor air was up to ~6 μg/m³. The general average personal exposure to benzene of 800 non-occupationally exposed persons was reported as about 15 μg/m³. EPA estimated that continuously breathing air containing 1.3 to 4.5 μg/m³ would result in not greater than a one-in-a-hundred thousand increased chance of developing cancer.[21] EPA estimated using extrapolation method that a range of 2.2×10^{-6} to 7.8×10^{-6} is the increase in the lifetime risk of an individual who is exposed for a lifetime to 1 μg/m³ benzene. Air concentration at specified risk level is 1/10,000 with the 13.0 to 45.0 μg/m³, 1/100,000 with the 1.3 to 4.5 μg/m³ and 1/1,000,000 with the 0.13 to 0.45 μg/m³.[22] EPA has established Rfc for benzene as 30 μg/m³ in relation to decreased lymphocyte count. The California Environmental Protection Agency (CalEPA) has established a chronic reference exposure level as 60 μg/m³ for benzene based on hematological effects in humans.[23]

Above 4 aromatic hydrocarbons, benzene, toluene, ethylbenzene and the isomeric xylenes (BTEX), are among the main components of automobile exhaust, if gasoline engines are used. Evaporation of solvents, i.e. in lacquers and paints, represents another important source in particular for toluene and C8-aromatic hydrocarbons.[7] These possible emitting sources may explain why these compounds were found in particularly high concentrations in the indoor air of the preschool facilities, particularly in urban located classroom. This study shows that the BTEX concentrations measured in indoor air were generally higher than the outdoor air concentrations in both urban and rural area as shown in Fig. 1, confirming that besides their presence in

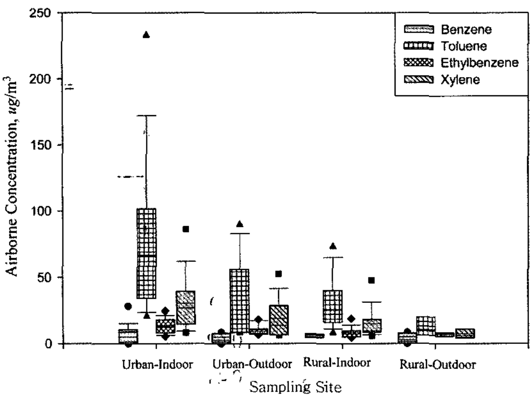

Fig. 3. BTEX compounds (benzene, toluene, ethylbenzene and xylene) in indoor and outdoor air in preschool facilities.

outdoor air, BTEX compounds also have material- and human-activity-based sources.[24]

For each BTEX compounds, statistical differences between indoor and outdoor were tested by sampling site for the purpose of checking outdoor BTEX would be the possible source for BTEX detected in indoor air. T-test was run if testing compound were normal distributed, otherwise, Mann-Whitney Rank Sum test was run. The benzene concentration was not statistically significantly different in indoor air and outdoor air (P=0.780 in urban area, P=0.357 in rural area, Mann-Whitney Rank Sum Test). Toluene concentration was significantly higher in indoor air in urban preschool but not significant in rural area (p=0.001 in urban area with Mann-Whitney Rank Sum Test, p=0.189 in rural area with t-test). Ethylbenzene presented the same pattern with toluene (p=0.022 in urban area with t-test, p=0.088 in rural area with t-test). Xylene also presented the same pattern with toluene (p=0.008 in urban area with Mann-Whitney Rank Sum test, p=0.178 in rural area with t-test). These facts indicated that benzene in preschool indoor air could be entered from the contaminated outdoor air but toluene, ethylbenzene and xylene were originated not only from outdoor source but also from indoor source. There were many sources in preschool facilities that emitted volatile organic compound, including formaldehyde. Examples include paint, lacquers, paint stripers, cleaning supplies, pesticide, building materials, new furnishings, spraying repellents, waxes, adhesives, glues and compressed wood furnishings. It is important that increasing ventilation when using products that emit TVOC.[6] In the previous study, out door VOC compounds should be considered as one of important sources of indoor air pollution indicated that petroleum hydrocarbons might have contributed significantly to the indoor levels.[24,25] Several efforts to identify and control the emissions from the possible sources were done.[26] Various modeling could be used to predict the possible sources.[27,28]

3. Relationship between 8-quantified VOC and TVOC

Fig. 4 illustrates the arithmetic mean concentrations of eight quantified aromatic VOC and the non-quantified VOC. The concentrations and percentages of the quantified aromatic VOC in urban preschool facilities were higher than those of rural area. The concentration of TVOC was presented in Table 1. As shown in Fig. 3, quantified VOC occupied 29.3% at indoor, 26.3% at outdoor in urban area whereas it was 17.7%, 14.6% in rural area, respectively. The mean percentage of quantified VOC was 22.8% of TVOC.

This study investigated the strength of association among each VOC compounds. The Spearman correlation coefficient was presented in Table 3. The Spearman correlations were significant ($p < 0.05$) among all the target VOC compounds except benzene. This means that all the target VOC compounds except benzene tend to have positive

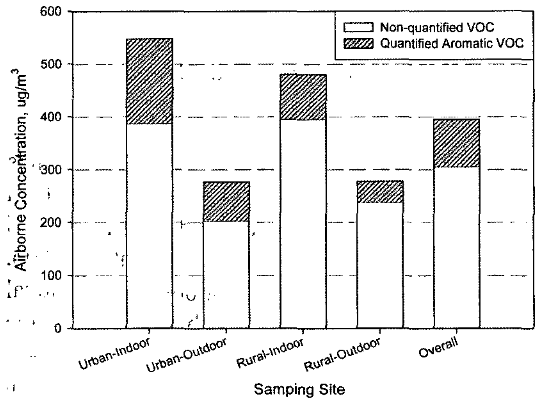

Fig. 4. Concentration distribution of eight quantified aromatic hydrocarbons and TVOC.

Table 3. Correlation coefficient among the VOC compounds (Spearman rank order correlation)

	Xylene	Ethyl-benzene	Benzene	1,2,4-trimethyl benzene	Styrene	Naphthalene	1,3,5-trimethyl benzene	Quantified VOC	Non-quantified VOC	TVOC
Toluene	0.86	0.68	0.15*	0.50	0.33	0.55	0.44	0.96	0.72	0.76
Xylene		0.83	0.14*	0.62	0.31	0.61	0.52	0.93	0.84	0.87
Ehtylbenzene			0.09*	0.72	0.52	0.71	0.64	0.79	0.70	0.72
Benzene				0.004*	0.10*	-0.18*	0.21*	0.19*	0.11*	0.11*
1,2,4-trimethylbenzene					0.46	0.64	0.69	0.62	0.53	0.55
Styrene						0.46	0.28	0.41	0.34	0.35
Naphthalene							0.56	0.62	0.51	0.53
1,3,5-trimethylbenzene								0.54	0.32	0.34
Quantified VOC									0.80	0.84
Non-quantified VOC										0.99

*P value is above 0.05, P values of all others are less than 0.05

relationships between the pairs. Benzene, on the contrary, has no significant relationship with other target VOC including TVOC though the reason is uncertain.

Toluene, xylene and ethylbenzene have high correlation coefficients ($r \geq 0.7$) with quantified VOC, non-quantified VOC and TVOC. So, these substances could be used as the index of TVOC. Xylene shows higher correlation coefficient of 0.86 for toluene, 0.83 with ethylbenzene, 0.93 with quantified VOC, and 0.76 with TVOC. Quantified VOC in this study also represents higher correlation coefficient of 0.80 with non-quantified VOC, and 0.84 with TVOC.

IV. Conclusion

This study showed that indoor air quality in respect to TVOC and several aromatic compounds in urban located classrooms were higher than those of rural area. Also, we found that indoor level was higher than outdoor level (I/O ratio 2.5>1). Though, the mean concentrations of individual BTEX constituents were much lower than the EPA's Reference Concentration (RfC), high proportion of TVOC exceeded the level of the Korean government's regulatory limit (400 $\mu g/m^3$).

Higher concentration and higher proportion of BTEX in indoor air than outdoor air implicated that indoor level was affected not only from outdoor sources but also from the possible indoor sources. To decrease the level of TVOC and individual compounds in class room, we have to endeavor our efforts to diminish possible indoor source and to make outdoor air clean. For example, introduction of water-based paints instead of organic-based paint could lead to a reduction of the pollution level by these compounds in the indoor environment. Toluene and Xylene were proved as possible surrogates of TOVC.

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