The Relationship Between Indoor, Outdoor and Personal VOC Concentrations in Homes, Offices and Schools in the Metropolitan Region of Kocaeli, Turkey

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Abstract Human exposure to volatile organic compounds (VOCs) and residential indoor and outdoor VOC levels had hitherto not been investigated in Turkey. This study details investigations of indoor, outdoor, and personal exposure to VOCs conducted simultaneously in 15 homes, 10 offices and 3 schools in Kocaeli during the summer of 2006 and the winter of 2006–2007. All VOC concentrations were collected by passive sampling over a 24-h period and analyzed using thermal desorption (TD) and a gas chromatography/flame ionization detector (GC/FID). Fifteen target VOCs were investigated and included benzene, toluene, m/p-xylene, o-xylene, ethylbenzene, styrene, cyclohexane, 1,2,4-trimethylbenzene, n-heptane, n-hexane, n-decane, n-nonane, n-octane and n-undecane. Toluene levels were the highest in terms of indoor, outdoor, and personal exposure, followed by m/p-xylene, o-xylene, ethylbenzene, styrene, benzene and n-hexane. In general, personal exposure concentrations appeared to be slightly

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higher than indoor air concentrations. Both personal exposure and indoor concentrations were generally markedly higher than those observed outdoors. Indoor target compound concentrations were generally more strongly correlated with outdoor concentrations in the summer than in winter. Indoor/outdoor ratios of target compounds were generally greater than unity, and ranged from 0.42 to 3.03 and 0.93 to 6.12 in the summer and winter, respectively. Factor analysis, correlation analyses, indoor/outdoor ratios, microenvironment characteristics, responses to questionnaires and time activity information suggested that industry, and smoking represent the main emission sources of the VOCs investigated. Compared with the findings of earlier studies, the level of target analytes in indoor air were higher for several target VOCs, indicating a possible trend toward increased inhalation exposure to these chemicals in residential environments.

Keywords Indoor air quality. VOCs . Personal exposure . Seasonal variability . Traffic . Industry

1 Introduction

In recent years, research has shown that indoor air quality (IAQ) is an important determinant in terms of human health. People spend as much as 90% of their time indoors, comprising a microenvironment which may contain higher levels of pollution than outdoor air even in the largest and most industrialized cities. The amount of time an individual spends in any one microenvironment impacts directly on the health of that individual. Although indoor concentrations and personal exposure to pollutants are strongly correlated in the case of most organic air pollutants, the extent of personal exposure shows great variability in terms of outdoor air concentrations (Namieśnik et al. [2002](#page-15-0); Payne-Sturges et al. [2004](#page-15-0); Ohura et al. [2006](#page-15-0)). Urban air compounds consist of volatile organic compounds (VOCs) and many of these, such as benzene, styrene and toluene, are known or suspected of being toxic or carcinogenic.

Under Title III Section 112 of the 1990 Clean Air Act (CAA) Amendments, numerous VOCs have been identified as hazardous air pollutants (HAPs; Sexton et al. [2004](#page-15-0)). This list was amended to the CAA under Title I, Part A, Section 112 (CAA [1991](#page-16-0)). Ambient VOCs contribute to the formation of ozone and photochemical oxidants, which in turn determine the formation and fate of airborne toxic chemicals (Finlayson-Pitts and Pitts [1997](#page-15-0)). Vehicular emissions and industrial sources represent major sources of ambient VOCs, while indoor environment VOC sources derive from cigarette smoke, combustion byproducts, cooking fumes, construction materials, furnishings, paints, varnishes and solvents, adhesives and caulks, office equipment, and consumer products (Vega et al. [2000](#page-16-0); Chan et al. [2002](#page-15-0); Ho et al. [2002](#page-15-0); Guo et al. [2003](#page-15-0)).

Passive sampling is generally just as accurate as active sampling and obviates the use of expensive and impractical active sampling equipment such as pumps and flow meters. One of the few disadvantages associated with the use of passive sampling methodologies is the comparatively low sampling rate, which requires long sampling times for low concentration VOC environments. However, this may also be an advantage since this approach readily facilitates determinations of time-weighted average (TWA) analyte concentrations. As a general evaluation of the effects of pollutants on human health, TWA concentrations are more useful than short-term concentrations since they show the long-term action of the compounds under investigation (Zabiegala et al. [2002](#page-16-0)).

Examination of the levels of 15 target VOCs was performed in relation to personal, indoor and outdoor air of randomly selected homes (15), offices (10) and schools (3) in the city of Kocaeli.

2 Materials and Methods

2.1 Sampling

In the first stage, three areas (urban, industrial and rural) were selected in an effort to determine whether pollutants generated by industrial and urban areas influenced personal, indoor and outdoor VOC exposure in Kocaeli. Samples were taken from 15 homes, 10 offices and 3 schools during the summer (May 31, 2006 to June 29, 2006) and winter months (December 16, 2006 to January 20, 2007) in Kocaeli (Fig. [1](#page-2-0)). Details concerning the sampling sites are shown in Table [1](#page-3-0).

VOCs were collected simultaneously from indoor, outdoor and personal environments for 24 hours using Radiello (Fondazione Salvatore Maugeri, Padova, Italy) passive samplers containing 350 mg of graphitized charcoal (Carbograph 4) with 35–50 mesh particle size. Prior to the main study, several pilot experiments were conducted to evaluate the suitability of the sampling and analytical procedures that were intended for use in the main study. These included an estimation of detection limits using actual field and blank samples and the determination of appropriate sampling times.

Passive samplers were placed in the living room, kitchen and bedroom of homes during both seasons. The sampling equipment was placed in the center of each microenvironment at a height of ca. 1.5 m above floor level in order for the sampling to occur within the breathing zone of a seated individual. At the same time, an outdoor sampler was placed away from exhaust vents and heat sources of the house and protected from rain and direct sunlight. Passive samplers were also placed within the breathing zone of one participant at each microenvironment. Following completion of the sampling, samplers were stored in a freezer at −20°C until analysis. Active charcoal filters were stored in the freezer with the samplers to prevent contamination.

Thermal comfort parameters (temperature and relative humidity) were measured simultaneously using the Langan Model L76n Indoor Air Quality Measurer (Langan Products, Inc. CA, USA) to

Fig. 1 Location of Kocaeli province in Turkey

produce a general profile of both indoor and outdoor air. The 24-h average values of temperature and relative humidity were obtained from measurements recorded using a data logger during each sampling event.

2.2 Extraction and Analysis

Prior to sampling, each passive sampler was conditioned by passage of an ultra-pure stream of nitrogen at 350°C for 6-h. Conditioned samplers were then capped with Swagelok end-caps and PTFE ferrules and stored in sealed glass jars in a container at 4°C immediately prior to use. A Unity*™* thermal desorber (Markes International Limited, UK) unit coupled to an Agilent gas chromatograph (Model 6890) and two independent flame ionization detectors (FID; Agilent Technologies, Inc. Santa Clara, CA, USA), one to measure total hydrocarbons and the other to measure methane and non-methane components, were used to analyze the target compounds. The analytical columns used for separating targets comprised a DB-1 capillary column (60 m, 0.25 mm i.d., 1 μ m film thickness) and an HP-PLOT Al_2O_3 "S" deactivated capillary column (50 m, 0.32 mm i.d., 8 μm film thickness). Operation of the instrument was controlled using an Agilent Chemstation data system. The oven temperature program was initially set to 40°C for 5 min., which then increased at a rate of 5° C min⁻¹ up to 195°C, and was then finally maintained for 10 min at 195°C. The FID temperature was set to 300°C. The ultra-pure nitrogen carrier gas flow rate was 2 ml min⁻¹. Fifteen target VOCs that included benzene, toluene, m/p-xylene, o-xylene, ethylbenzene, styrene, cyclohexane, 1,2,4-trimethylbenzene, n-heptane, nhexane, n-decane, n-nonane, n-octane and n-undecane were investigated. Spectra (Spectra Gases, Inc. Branchburg, NJ, USA) EPA PAMS (Photochemical Assessment Monitoring System) calibration standard, which comprises a mixture of 56 volatile organic compounds each at a concentration of 1 ppm, was used. Sampling rate values of the passive sampler for 15 volatile organic compounds were taken from Radiello's literature.

Blank samples, limits of detection (LOD), reproducibility and recovery were assessed for quality assurance of the analysis of the target pollutants. A field blank sample was also determined on each sampling day. The amount of VOCs in blank samples ranged from below LOD to 1.22 μ g m⁻³ for toluene.

Characteristics	Summer		Winter					
	$Mean \pm SD^a$	Range	$Mean \pm SD$	Range				
Time spent in microenvironment (h)								
Indoors (all locations)	19.6 ± 2.0	$0.15 - 22.30$	21.5 ± 1.6	$0.50 - 21.30$				
At home	16.6 ± 4.6	$9.40 - 22.30$	16.9 ± 5.8	$1.50 - 21.30$				
At work/school	2.6 ± 3.7	$0.20 - 10.45$	4.0 ± 5.4	$1.10 - 11.30$				
At other	0.40 ± 0.83	$0.15 - 3.00$	0.6 ± 0.8	$0.50 - 2.35$				
Outdoors (all locations)	3.5 ± 1.8	$0.15 - 7.40$	1.2 ± 1.4	$0.20 - 9.30$				
At home	1.4 ± 1.1	$0.30 - 4.30$	0.2 ± 0.5	$0.50 - 2.50$				
At work/school	0.5 ± 0.7	$0.15 - 2.50$	0.2 ± 0.4	$0.35 - 2.00$				
At other	1.6 ± 1.2	$0.35 - 7.40$	0.8 ± 1.3	$0.20 - 9.30$				
In transit	0.9 ± 0.6	$0.10 - 2.00$	1.3 ± 0.9	$0.15 - 2.10$				
Study period	May 31, 2006–June 29, 2006			Dec 16, 2006–Jan 20, 2007				
Sampling site region	15 urban areas, 9 industrial areas, 4 rural areas							
Age of building (years)	18 ± 13							
Floor area (m^2)	71 ± 40							
Thermal comfort parameters								
Indoor temperature $(^{\circ}C)$	25 ± 2	$17 - 34$	20 ± 2	$3 - 25$				
Outdoor temperature $(^{\circ}C)$	24 ± 4	$14 - 39$	9 ± 3	$1 - 30$				
Indoor relative humudity $(\%)$	54 ± 6	$23 - 76$	$48 + 5$	$28 - 85$				
Outdoor relative humidity $(\%)$	$58 + 9$	$17 - 86$	65 ± 6	$20 - 87$				

Table 1 Microenvironment characteristics and sampling conditions

^a Standard deviation

LOD values, which are listed in Table [3](#page-5-0), were defined as three times the standard deviation $(n=6)$ of the lowest concentration of the blank samples. The coefficients of variation in the reproducibility test $(n=8)$ ranged from 2.8% for benzene to 8.8% for ethylbenzene. The recovery test $(n=7)$ was performed by spiking the compounds in the corresponding samplers. The recoveries were 93.5% for n-undecane to 97.3% for n-hexane.

2.3 Questionnaires

Participants in each microenvironment surveyed completed a time activity diary and a questionnaire pertaining to the sites, surroundings, personal activity and commuting behavior during the course of the study. The activity diary consisted of half-hour time intervals during the day and 1-h time intervals from midnight to 7 A.M. The questionnaire dealt with microenvironment characteristics, cleaning and ventilation habits and living conditions. On average, participants spent only 10% of their time outdoors during the sampling and almost 85% indoors; 70 and 13% of their time in the home and workplace, respectively (Table 1). Samples were collected during the summer when the outdoor temperature ranged from 14 to 39°C and during the winter when the outdoor temperature ranged from 1 to 30°C. The indoor temperature remained relatively constant $(20\pm$ 2°C) given that most of the selected microenvironments were heated with natural gas (occasionally by fuel oil and electricity) during the winter. The average age of the buildings was 18 years, and ranged from newly constructed buildings to buildings greater than 50 years old. The selected microenvironment characteristics and sampling conditions (temperature and relative humidity) are summarized in Table 1. Table [2](#page-4-0) summarizes the categorization of the 15 homes, 10 offices and 3 schools in relation to the main VOC sources.

2.4 Statistical Analysis

Statistical analyses were performed using the SPSS 11 software (SPSS Inc., Chicago, USA) for Windows. Values less than the detection limit were substituted by one-half of the detection limit.

The data were log-normally distributed in the Kolmogorov-Smirnov statistical test, which was employed to assess the normality of the data. The

Table 2 Categorization of the 15 homes, 10 offices and 3 schools in relation to three main VOCs sources

Categories	Number of microenvironments sampled	Percent of the total number of microenvironments sampled
Homes	15	
Smokers' homes	9	60
Non-smokers' homes	6	40
Homes in roads with a moderate and high traffic level	10	67
Homes in roads with a low traffic level	5	33
New and renovated homes	15	100
Old homes	Ω	Ω
Offices	10	
Smokers' offices	7	70
Non-smokers' offices	3	30
Offices in roads with a moderate and high traffic level	$\mathbf{8}$	80
Offices in roads with a low traffic level	2	20
New and renovated offices	10	100
Old offices	Ω	θ
Schools	3	
Smokers' schools	θ	Ω
Non-smokers' schools	3	100
Schools in roads with a moderate and high traffic level	$\overline{2}$	67
Schools in roads with a low traffic level	1	33
New and renovated schools	2	67
Old schools	1	33

distribution of indoor air quality data and air pollutants was generally found to be typical (Baek et al. [1997](#page-15-0)). Therefore, we used a nonparametric Friedman test in an effort to determine whether there were significant differences between indoor, outdoor and personal exposure in different microenvironments and between summer and winter. Statistical significance was set to a level of 5% $(P<0.05)$.

Factor analysis (FA) is a multivariate statistical tool widely employed for several years to assign source in the field of air pollution control (Chen et al. [2002](#page-15-0); Miller et al. [2002](#page-15-0)). To this end, a correlation matrix is initially calculated and then a matrix of "factor loadings" of species included in the FA is generated by a rotating correlation matrix. In the factor loading matrix, species are grouped into factors and the factor loading of a parameter is a measure of the association of a particular parameter with that factor. Factor analysis is simple to use and, although the mathematics behind it is fairly

complicated for most users, it does not require any prior information relating to the composition of source emissions of any measured species, and as such meteorological parameters can be included in the factor analysis (Hopke et al. [1976](#page-15-0); Henry [1997](#page-15-0)).

3 Results and Discussion

3.1 Indoor and Outdoor Levels and Personal Exposure to VOC in Different Microenvironments

Although some VOCs have been identified as a significant risk factor associated with the onset of cancer and shown to involve the formation of photochemical oxidants (Baek et al. [1997](#page-15-0)), these analytes have not been measured in Turkey and no national ambient air quality standard has been established for VOCs. Little or no data is available

Table 3 VOC mean concentrations of indoor, outdoor and personal exposure in different environments in Kocaeil

Components		Summer mean conc. $(\mu g m^{-3})$			I/O/P corr. ^a	Winter mean conc. (μ g m ⁻³)			I/O/P corr.	S/W corr. ^b
		Indoor $(n=61)$	$(n=28)$	Outdoor Personal $(n=28)$	P value ^{c}	Indoor $(n=61)$	$(n=28)$	Outdoor Personal P value $(n=28)$		P value
Benzene (LOD ^d :	Home	8.88	10.03	10.06	0.627	13.06	7.82	12.07	0.097	0.297
0.2)	Office	11.95	7.83	9.75	0.122	15.09	9.25	13.98	0.497	0.028
	School	7.50	4.77	6.80	0.097	19.77	16.41	31.25	0.717	1.000
Toluene (LOD:	Home	44.19	30.72	55.28	0.007	72.44	19.96	81.62	< 0.001	0.011
1.1)	Office	53.98	33.73	62.27	0.050	104.37	25.07	122.43	0.001	0.144
	School	55.05	18.15	45.09	0.097	77.77	41.96	74.79	0.368	0.021
m/p -xylene (LOD:	Home	12.30	12.76	14.78	0.247	27.46	13.43	28.34	< 0.001	< 0.001
(0.4)	Office	14.37	9.24	15.13	0.002	34.50	17.34	40.42	0.007	< 0.001
	School	9.55	6.80	9.55	0.097	21.47	20.19	24.31	0.717	0.004
o-xylene (LOD:	Home	5.73	6.89	7.13	0.282	16.24	7.61	17.69	< 0.001	< 0.001
(0.3)	Office	10.80	6.88	9.95	0.020	20.75	9.78	23.97	0.006	0.003
	School	5.88	5.62	7.11	0.368	11.19	10.02	10.91	0.717	0.004
Ethylbenzene	Home	13.07	4.49	14.82	0.007	16.90	5.31	17.11	< 0.001	0.053
(LOD: 0.2)	Office	11.13	5.57	15.46	0.002	19.75	5.69	17.88	0.008	0.144
	School	11.11	6.10	9.45	0.097	12.40	8.81	12.97	0.368	0.248
Styrene (LOD: 0.3) Home		9.39	4.19	9.51	0.005	11.65	4.80	12.93	0.001	0.002
	Office	8.99	3.90	7.97	0.001	16.51	5.72	15.74	0.050	0.001
	School	7.20	5.71	6.94	0.097	11.80	8.20	8.11	0.097	0.248
Cyclohexane	Home	6.58	5.03	7.82	0.024	9.29	5.33	10.02	0.002	0.180
(LOD: 0.3)	Office	6.01	3.77	6.55	0.001	8.28	4.82	10.08	0.014	0.003
	School	4.65	3.87	3.81	0.717	11.55	12.30	13.08	0.717	0.004
1,2,4-TMB ^e (LOD: Home		2.35	2.47	2.51	0.282	4.20	1.45	5.22	< 0.001	0.101
0.2)	Office	2.34	5.54	8.07	0.122	5.88	2.31	4.95	0.045	0.715
	School	7.95	15.57	9.81	0.717	1.97	0.51	1.34	0.097	0.004
n-heptane (LOD:	Home	5.49	2.42	6.35	< 0.001	8.73	2.67	8.75	< 0.001	0.025
0.2)	Office	9.40	4.92	9.87	0.045	16.85	2.88	17.82	< 0.001	0.273
	School	11.65	5.68	7.34	0.264	14.14	9.72	16.46	0.097	0.248
n-hexane (LOD:	Home	9.31	9.72	9.73	0.819	12.63	5.97	11.58	0.097	0.763
0.3)	Office	20.93	7.16	11.95	0.150	14.68	6.75	13.86	0.122	0.068
	School	5.02	6.40	14.89	0.264	16.42	17.66	30.44	0.368	0.564
n-decane (LOD:	Home	4.74	2.70	5.25	0.005	5.09	1.57	6.15	< 0.001	0.763
0.2)	Office	3.92	3.83	6.09	0.273	7.05	2.03	5.91	0.014	0.705
	School	5.42	6.09	5.28	1.000	2.00	0.60	2.79	0.097	0.021
n-nonane (LOD:	Home	9.05	5.30	10.99	0.007	13.53	3.56	15.56	< 0.001	0.101
(0.2)	Office	9.75	5.23	10.16	0.003	21.47	4.32	19.49	0.007	0.068
	School	9.31	4.49	6.02	0.097	12.06	5.37	10.48	0.717	1.000
n-octane (LOD:	Home	4.18	2.67	4.80	0.011	6.51	2.54	5.94	< 0.001	0.180
(0.3)	Office	6.53	3.44	7.61	0.001	10.00	3.45	9.70	0.007	0.068
	School	4.25	3.17	3.10	0.264	4.36	3.25	5.35	0.368	0.248
n-undecane (LOD:	Home	4.26	4.31	4.74	0.214	5.59	1.37	6.26	< 0.001	0.881
0.2)	Office	6.05	3.13	7.07	0.122	7.46	1.88	5.47	0.025	0.095
	School	3.14	2.60	2.39	0.717	2.38	0.39	1.87	0.097	0.083

a Indoor/Outdoor/Personal correlation

^b Summer/Winter correlation

^c Friedman test significance

^d Limit of detection (μg m⁻³)

^e Trimethylbenzene

on the levels of VOCs, particularly in relation to personal exposure and indoor environments where Turkish people typically spend most of their time. Consequently, this study is the first to measure personal exposure and microenvironmental concentrations of VOCs in different microenvironments in a Turkish city. The 15 target VOC compounds investigated in this study are listed in Table [3](#page-5-0) with their limit of detection (LOD) and arithmetic mean concentration of indoor, outdoor and personal exposure samples in the summer and winter.

The living room, kitchen and bedroom values for the target VOCs were highly correlated (Pearson correlation coefficients $0.6-0.9$, $P<0.05$), where indoor air values of the homes were defined as the mean of the living room, kitchen and bedroom daily concentrations.

Table [3](#page-5-0) shows that the mean indoor, outdoor and personal exposure levels of the target compounds measured in this study varied widely among the three types of environments. Toluene had the highest mean indoor, outdoor and personal exposure concentration, followed by m/p-xylene, o-xylene, ethylbenzene, styrene, benzene and n-hexane. The results of the nonparametric Friedman test (Table [3](#page-5-0)) show that personal exposure concentrations are significantly higher than indoor and outdoor air concentrations for toluene, ethylbenzene, styrene, cyclohexane, n-heptane, n-nonane and n-octane in homes and offices in the summer and winter. There was no significant difference observed for all target compounds in schools during either season. There was also no significant difference observed for benzene and n-hexane in the three microenvironments during either season, or for benzene, 1,2,4-trimethylbenzene, n-hexane and n-undecane during the summer season. A significant difference between indoor, outdoor and personal exposure levels suggests an indoor source for these compounds, whereas the lack of a significant difference suggests that outdoor sources are the main contributors to indoor and personal exposure levels.

It is apparent from Table [3](#page-5-0) that there is a general increase of nearly all the compounds measured in this study from summer to winter and when comparing outdoor to indoor air. For nearly all compounds, indoor VOC concentrations were significantly greater in winter than in summer. Indoor activity, ventilation and the duration of human occupancy all influence indoor air quality during the winter, as do meteorological factors and the outdoor air. One possible explanation for elevated winter VOC concentrations is a decrease in ventilation during the winter since windows are opened less frequently and air conditioners are seldom used. VOCs may accumulate in higher concentrations as a result of lower ventilation rates when there are persistent VOC sources indoors, together with VOCs entering from outside when present in higher concentrations in the outdoor air. The Friedman test was performed in an effort to determine whether the difference between summer and winter VOC levels was significant for individual compounds. The results of this test show that there is a significant seasonal influence on both indoor and outdoor values for toluene, m/p-xylene, o-xylene, styrene and n-heptane in homes, for benzene, m/p-xylene, o-xylene, styrene and cyclohexane in offices, and for toluene, m/p-xylene, o-xylene, cyclohexane, 1,2,4-trimethylbenzene and n-decane in schools.

Results of the nonparametric Friedman test concerning seasonal differences indicated marked differences in some outdoor VOC levels. The possible causes of these increases in outdoor VOC concentrations during the winter have been ascribed to source variation, fuel consumption, poor operation of catalytic converters, meteorology, limited vertical mixing in the atmosphere, and limited or decreased photochemical reactivity (Baek et al. [1997](#page-15-0)). During both seasons, the indoor concentration of most of the target compounds measured in the office was higher than those found in the home and school, highlighting the effect of office activity on indoor air quality.

It is of interest to compare the indoor, outdoor and personal exposure concentrations measured in this study with similar studies conducted in earlier years. Table [4](#page-7-0) compares the results for individual VOCs from the current study with those of previous studies. In general, the levels of benzene, toluene, ethylbenzene, xylenes and styrene (BTEXS) in the present study were higher than the values reported in other studies. Personal exposure, outdoor and indoor BTEXS concentrations were higher than values measured at urban sites in Minnesota, USA (Adgate et al. [2004](#page-15-0)) and in Europe (Ilgen et al. [2001](#page-15-0); Schneider et al. [2001](#page-15-0)), and comparable with values reported for Mexico (Tovalin-Ahumada and Whitehead [2007](#page-16-0)) and Europe (Zuraimi et al. [2006](#page-16-0)), but lower than those values reported for Korea (Son et al. [2003](#page-15-0)) and Singapore (Zuraimi et al. [2006](#page-16-0)).

Table 4 VOC concentrations (μ g m⁻³) from various sources

Concentration (μ g m⁻³)

^a p-xylene, n.d.: analyte was not determined, n.m.: analyte was not measured

3.2 Indoor, Outdoor and Personal Exposure in Smoking and Non-smoking Microenvironments

Table [5](#page-8-0) shows personal exposure concentrations of the target compounds in relation to non-smoking and smoking participants. Participants who smoked cigarettes at any time during the 24-h sampling period had

significantly elevated personal exposure to benzene, styrene, cyclohexane, n-hexane and n-undecane. Benzene has been identified as a component of tobacco smoke by the International Agency for Research on Cancer (IARC; IARC [1986](#page-15-0)). Toluene, m/p-xylene, o-xylene, styrene, and ethylbenzene have also been reported to be components of tobacco

Styrene 13.31 8.78 11.61 6.12 4.08 5.52 0.050 Cyclohexane 10.21 5.78 9.31 5.37 1.92 5.56 0.050 1,2,4-TMB 6.13 7.83 3.17 2.58 2.62 1.74 0.637 n-heptane 11.26 7.47 9.35 8.17 9.30 5.37 0.637 n-hexane 15.98 13.70 12.27 5.91 4.55 4.41 0.005 n-decane 6.60 3.91 5.23 3.58 2.05 2.86 0.157 n-nonane 15.85 12.15 13.48 7.89 5.75 5.40 0.157 n-octane 7.45 4.25 6.88 4.25 4.45 2.73 0.059 n-undecane 6.05 5.00 4.55 3.53 2.59 2.84 0.018

Table 5 Differences in VOC personal exposure concentrations for participants by cigarette smoke exposure

a Smokers/Non-Smokers correlation

^b Friedman test significance

smoke (Wallace et al. [1987](#page-16-0)). The arithmetic mean and median VOC concentrations for participants that smoked were approximately twofold higher compared with non-smoking participants. This observation may suggest that smoking is a major source of exposure to VOCs.

3.3 Indoor, Outdoor and Personal Exposure in Urban, Industrial and Rural Areas

Another aim of this study was to compare VOC concentrations in regions possessing different characteristics. Urban areas with heavy traffic, areas with heavy industry, and rural areas far removed from industry and heavy traffic were deliberately chosen as sample sites. The results of the indoor and outdoor environment samples of the selected areas for the summer and winter seasons are presented in Table [6](#page-9-0). The results obtained from the three areas showed that the contribution of each compound examined differed, and that industrial and urban areas with heavy industry and traffic had significantly higher toluene, styrene, cyclohexane, nheptane and n-octane concentrations compared with rural areas. The concentration of VOCs which represent industrial and urban area markers was 2 ∼ 4-fold higher compared with rural areas, and reflects the magnitude of the contribution by motor vehicles and industry. The higher level of VOCs found in urban areas compared

with industrial areas does not necessarily indicate that there is no contribution by industry to the total VOCs found. When the concentration of VOCs obtained from these two regions was measured, it was found that some compounds were present at high levels in urban areas with heavy traffic, while others were present at high levels in industrial areas. Benzene, a motor vehicle marker, as well as toluene, ethylbenzene, cyclohexane, n-decane, n-nonane and n-octane concentrations were higher in urban areas. Whereas styrene and n-heptane concentrations were higher in industrial areas where petrochemical plant and gas filling stations are located. Statistical analyses revealed that winter concentrations were significantly higher than summer concentrations for toluene and xylenes in urban areas and for styrene and cyclohexane in industrial areas.

One proposed method of determining VOC sources in urban areas is to examine the relative ratio of different aromatic compounds. The ratio of benzene to toluene, measured in a variety of studies in different parts of the world, usually falls in the range 1:2 to 1:4 for ambient air. These values are close to the levels reported for vehicle emissions (Baek et al. [1997](#page-15-0)). In this study, ambient benzene to toluene ratios were fairly constant at 1:3. The similarity of the ratio measured for Kocaeli to that found in other regions implies that vehicle emissions are most likely to contribute to outdoor VOCs monitored in the urban areas of Kocaeli.

Components		Summer Conc. $(\mu g \ m^{-3})$	Winter Conc. $(\mu g \text{ m}^{-3})$	R/I/U corr. ^a P value ^c	S/W corr. b P value
Benzene	Rural $(n=4)$	5.95	3.77	0.417	0.067
	Industrial $(n=9)$	6.17	6.09		0.317
	Urban $(n=15)$	7.36	10.35		0.439
Toluene	Rural	9.01	6.80	0.030	0.097
	Industrial	14.37	16.05		0.317
	Urban	18.79	28.02		0.039
$\sum X \rm{y}$ lenes ^d	Rural	7.62	7.27	0.197	1.000
	Industrial	16.42	17.77		0.317
	Urban	15.63	29.81		0.020
Ethylbenzene	Rural	2.11	2.42	0.197	0.317
	Industrial	4.53	4.94		0.739
	Urban	6.40	5.73		0.796
Styrene	Rural	2.86	2.02	$0.010\,$	0.067
	Industrial	4.29	8.65		0.003
	Urban	3,84	3.75		0.796
Cyclohexane	Rural	2.60	3.04	0.042	$1.000\,$
	Industrial	2.38	4.27		0.020
	Urban	6.37	6.06		0.796
1,2,4-TMB	Rural	1.34	0.22	0.303	0.317
	Industrial	2.15	0.41		0.096
	Urban	1.47	1.07		0.796
n-heptane	Rural	1.96	2.15	0.050	$1.000\,$
	Industrial	3.49	3.19		0.339
	Urban	3.13	2.68		0.197
n-hexane	Rural	3.54	3.30	0.417	$1.000\,$
	Industrial	6.32	8.87		0.739
	Urban	8.96	5.51		0.796
n-decane	Rural	3.48	0.89	0.417	0.067
	Industrial	2.71	1.20		0.120
	Urban	3.36	1.82		0.197
n-nonane	Rural	4.19	2.76	0.607	0.096
	Industrial	2.64	3.43		0.739
	Urban	5.10	4.10		0.796
n-octane	Rural	1.03	1.61	0.020	0.317
	Industrial	2.92	2.35		0.739
	Urban	3.16	3.10		0.439
n-undecane	Rural	2.98	0.42	0.417	0.096
	Industrial	2.14	1.61		0.739
	Urban	3.45	0.89		0.071

Table 6 Outdoor air concentrations of VOCs (μ g m⁻³) in rural, industrial and urban areas in summer and winter in Kocaeli

^a Rural/Industrial/Urban correlation

^b Summer/Winter correlation

c Friedman test significance

^d Sum of the three xylene isomers (m,p,o-xylene)

3.4 Indoor/Outdoor Ratio and Correlation

In an effort to evaluate the relationship between indoor and outdoor environments, correlation analyses and residential indoor/outdoor (I/O) ratios were calculated using the indoor and outdoor concentrations of each target compound in the three environments studied.

The I/O ratios of each pollutant and their r^2 values are summarized for the summer and winter seasons in Table 7. An I/O ratio greater than unity indicates the presence of strong indoor sources for some of these compounds. The ratio of target compounds ranged from 0.42 to 3.03 and 0.93 to 6.12 for summer and winter, respectively. For almost all of the compounds, the I/O ratio was greater than unity during the summer and winter sampling seasons, and many ratios exceeded 2 or 3. Whereas indoor VOC levels may increase due to the entry and accumulation of VOCs from outdoor sources (Lewis [1991](#page-15-0)), the presence of dominant VOC sources is illustrated by the magnitude of the I/O ratio. The indoor concentration of VOCs in the sampled microenvironments (Table [3](#page-5-0)) was found to be similar, which suggests that exposure to VOCs is significant in indoor environments. When the same evaluation is made in relation to the indoor/outdoor environment and personal/outdoor concentration ratios the ratio is close to 3 despite some seasonal differences, which shows that indoor pollutants play a greater role in terms of human exposure to VOCs than outdoor pollutants.

Pearson correlation coefficients (r) were calculated in an effort to investigate the relationship between indoor and outdoor air quality in different microenvironments (home, office and school) for the summer and winter data sets (Table 7). In general, strong correlations were shown for the air quality in homes during both the summer and winter. For most of the compounds examined, correlations related to the offices and schools displayed greater variability for the two seasons.

In general, indoor target compound concentrations were more strongly correlated with outdoor concentrations in the summer than in winter. Statistically significant correlations were found between indoor and outdoor environment concentrations for benzene, toluene, m/p-xylene, o-xylene, cyclohexane, n-heptane, n-hexane, n-decane and n-nonane compounds in homes, especially during the summer period, while statistically weak correlations were found for the different microenvironments during the winter period. This shows that the contribution of these compounds to indoor environments is higher during the summer than winter months as these represent motor vehicle and industrial markers and windows are more likely to be open during the summer than in winter.

For styrene, no statistically significant correlation was found between I/O values in homes and offices during both seasons (Table 7). This shows that dominant indoor environment sources have a stronger impact than outdoor environment sources on indoor styrene concentrations. Tobacco smoke has been

Table 7 I/O ratios and Pearson correlation matrix between indoor and outdoor VOC concentrations in Kocaeli

	Summer					Winter						
	Indoor and outdoor correlation coefficients			I/O concentration ratios		Indoor and outdoor correlation coefficients			I/O concentration ratios			
Components	Home	Office	School	Home	Office	School Home		Office	School	Home	Office	School
Benzene	$0.75^{\rm a}$	0.34	0.97	0.89	1.53	1.57	0.24	0.31	0.94	1.67	1.63	1.20
Toluene	$0.74^{\rm a}$	0.57	0.45	1.44	1.60	3.03	-0.01	$0.74^{\rm a}$	-0.96	3.63	4.16	1.85
m/p -xylene	0.61 ^a	0.61	1.00 ^a	0.96	1.55	1.40	0.12	-0.02	0.97	2.04	1.99	1.06
o-xylene	$0.56^{\rm a}$	$0.83^{\rm a}$	0.78^{a}	0.83	1.57	1.05	0.23	0.22	0.99 ^a	2.13	2.12	1.12
Ethylbenzene	0.37	0.56	0.70	2.91	2.00	1.82	$0.91^{\rm a}$	-0.37	0.98	3.18	3.47	1.41
Styrene	0.22	0.49	0.99	2.24	2.31	1.26	0.20	-0.08	$0.99^{\rm a}$	2.43	2.89	1.44
Cyclohexane	$0.54^{\rm a}$	$0.84^{\rm a}$	0.89	1.31	1.59	1.02	0.37	0.11	0.99 ^a	1.74	1.72	0.94
$1,2,4$ -TMB	0.45	-0.14	0.24	0.95	0.42	0.51	0.88 ^a	0.09	0.73	2.90	2.54	3.85
n-heptane	$0.65^{\rm a}$	0.55	0.63	2.27	1.91	2.05	-0.24	$0.73^{\rm a}$	0.99	3.27	5.85	1.45
n-hexane	$0.91^{\rm a}$	-0.16	-0.09	0.96	2.92	0.78	0.06	0.18	0.99	2.11	2.18	0.93
n-decane	$0.64^{\rm a}$	-0.25	0.93	1.75	1.02	0.89	0.81 ^a	0.15	-0.58	3.24	3.48	3.35
n-nonane	$0.76^{\rm a}$	$-0.80^{\rm a}$	-0.25	1.71	1.87	1.43	$0.61^{\rm a}$	-0.10	-0.54	3.79	4.97	2.24
n-octane	0.45	$0.91^{\rm a}$	$0.99^{\rm a}$	1.56	1.90	1.34	$0.72^{\rm a}$	0.40	0.83	2.57	2.89	1.34
n-undecane	0.06	0.25	-0.06	0.99	2.32	1.21	0.68 ^a	0.23	0.39	4.09	3.96	6.12

 a $P < 0.05$

shown to have an impact on styrene concentrations obtained indoors as indicated by time-activity forms completed by individuals in locations where samples were taken. Moreover, the I/O ratios obtained for styrene were around unity (1.26–1.82) in schools where samples were taken in classrooms (non-smoking), and around 3 (2.00–3.47) in homes and offices inhabited by smokers (Table [7](#page-10-0)). This demonstrates the relationship between the styrene concentration obtained from the evaluated microenvironments and tobacco smoke.

3.5 VOC Sources

A varimax rotated FA was performed separately on indoor, outdoor and personal exposure data sets for homes and offices in an effort to identify possible VOC sources. Given that use of a small sample size can lead to unexpected associations observed in factor profiles, FA was not performed for the school data sets. The eigenvalue was set to 1.0 as a threshold in order to limit the number of extracted factors. Factor loadings equal to or greater than 0.3 were presented since those less than 0.3 were considered as insignificant. Source profiles were obtained from EPA SPECIATE v4.0 [\(2006](#page-16-0)) and the literature.

3.5.1 Homes

Table [8](#page-12-0) shows the varimax rotated FA results for homes in Kocaeli. Four factors were extracted for indoor, outdoor and personal exposure to VOCs.

For the outdoor data, the four factors obtained accounted for ca. 79% of the total variance, with the first factor F1-out accounting for 42.01%, the second factor F2-out 14.58%, the third factor F3-out 14.13% and the fourth factor F4-out 8.38%. Two traffic-related factors (diesel exhaust and gasoline exhaust) were differentiated in this analysis. F1-out was correlated most strongly with n-decane, 1,2,4-trimethylbenzene and n-undecane, and less strongly with cyclohexane, toluene, n-octane, ethylbenzene and n-nonane. F2-out was correlated most strongly with m/p-xylene and o-xylene, and less strongly with benzene, n-heptane, toluene, n-octane and ethylbenzene. These compounds have traditionally been associated with motor vehicle emissions (Scheff et al. [1989](#page-15-0); Barrefors and Petersson [1993](#page-15-0); Watson et al. [2001](#page-16-0)). The explosive growth in traffic observed in the last decade has had a noticeable impact on air quality. In the center of the city there are

several congested national highways (used by 300,000 cars/day in 2005; Cetin et al. [2007](#page-15-0)). Thus, F1-out and F2-out correspond to diesel exhaust and gasoline exhaust factors, respectively. F3-out correlated moderately with cyclohexane, n-heptane, ethylbenzene, styrene, n-nonane and n-decane, while F4-out correlated heavily with hexane and benzene. Kocaeli is one of the most urbanized, economically-developed and industrialized regions in Turkey. Air quality in Kocaeli is largely determined by emissions from some 1,000 industrial plants, including a hazardous and medical waste incinerator, three major tire factories, an automotive plant, a pulp and paper factory, several petrochemical plants, manufacturing facilities, Liquefied Petroleum Gas filling plants, storage tanks for solvents used by petroleum, paint, plastics and pesticide companies, population-oriented sources and a petroleum refinery that supplies more than 30% of Turkey's fuel (Pekey et al. [2004](#page-15-0); Karademir [2006](#page-15-0)). Therefore, F3-out and F4-out were identified as industrial and petroleum refinery factors, respectively. The FA outdoor results showed that near 57 and 22% of ambient VOC impacts are due to traffic-related sources and industrial sources, respectively.

For the indoor data, the four factors obtained accounted for ca. 77% of the total variance, with the first factor F1-in accounting for 39.37%, the second factor F2-in 19.08%, the third factor F3-in 10.58% and the fourth factor F4-in 8.06%. F1-in correlated with the volatile alkanes (n-heptane, n-nonane, noctane, n-decane and n-undecane) and ethylbenzene, m/p-xylene, o-xylene and 1,2,4-trimethylbenzene. These compounds have been associated with indoor cleaning products and consumer products. F2-in correlated with n-hexane, benzene, cyclohexane and styrene. As previously mentioned, F2-in of the VOCs was associated with indoor smoking. F3-in correlated with cyclohexane, toluene, n-octane, m/p-xylene, o-xylene, n-decane and 1,2,4-trimethylbenzene. F3-in of the VOCs was associated with motor vehicles, as with the F1-out and F2-out profiles. F4-in was associated with styrene and n-decane, and its profile was similar to the F3-out profiles and hence was identified as an industrial factor.

For the personal exposure data, the four factors obtained accounted for ca. 78% of the total variance, with the first factor F1-per accounting for 46.20%, the second factor F2-per 13.78%, the third factor F3-per 9.42% and the fourth factor F4-per 8.84%. F1-per

correlated with n-octane, ethylbenzene, m/p-xylene, oxylene, n-nonane, n-decane and 1,2,4-trimethylbenzene. F1-per and F1-in represent indoor-related factors related to personal exposure and indoor data sets, respectively. The composition of these two factors was similar. These two factors are probably identical and represent similar indoor-related components (indoor cleaning products and consumer products) in relation to the personal and indoor data sets. Similar factors were obtained from the indoor and personal FA analyses for F2-per and F3-in, F3-per and F2-in, and F4-per and F4-in FA solutions. Thus, F2-per was identified as a motor vehicle factor, F3-per as a smoking factor and F4-per as an industrial factor.

3.5.2 Offices

Table [9](#page-14-0) shows the varimax rotated FA results for offices in Kocaeli. Four factors were extracted for indoor, outdoor and personal exposure to VOCs.

For the outdoor data, the four factors obtained accounted for ca. 85.9% of the total variance. F1-out (variance, 54.66%) correlated with n-hexane, benzene, n-heptane, toluene, n-octane, ethylbenzene, o-xylene, nnonane, n-decane and 1,2,4-trimethylbenzene and was identified as a diesel exhaust factor. F2-out (variance, 12.99%) correlated with benzene, cyclohexane, toluene, n-octane, ethylbenzene, m/p-xylene, o-xylene and nnonane and was identified as a gasoline exhaust factor. F3-out (variance, 9.88%) correlated with n-hexane, benzene and toluene and was identified as a petroleum refinery factor. F4-out (variance, 8.36%) correlated with cyclohexane, n-heptane and styrene and was identified as an industrial factor.

For the indoor data, the four factors obtained accounted for ca. 83.2% of the total variance. F1-in (variance, 48.52%) correlated with cyclohexane, noctane, ethylbenzene, m/p-xylene, o-xylene, n-nonane, n-decane, 1,2,4-trimethylbenzene and n-undecane. This factor was identified as representing a variety of product emissions from office indoor environments. F2-in (variance, 14.57%) correlated with cyclohexane, n-heptane, n-octane, styrene, o-xylene and n-nonane and was identified as representing industrial sources derived mostly from three major tire factories, Liquefied Petroleum Gas filling plants, and storage tanks for solvents used by petroleum, paint, plastics and pesticide companies. F3-in (variance, 10.33%) correlated with n-hexane, benzene, cyclohexane and styrene. These VOCs were

mainly derived from smoking sources. F4-in (variance, 9.79%) correlated with toluene, n-octane, m/p-xylene and o-xylene and this source profile corresponds to motor vehicle emissions.

For the personal exposure data, the four factors obtained accounted for ca. 81.0% of the total variance. F1-per (variance, 45.94%) correlated with cyclohexane, toluene, n-octane, m/p-xylene, o-xylene and n-nonane and was identified as a factor representing office equipment and consumer products. F2-per (variance, 17.45%) correlated with benzene, n-octane, ethylbenzene, o-xylene, n-nonane, n-decane, 1,2,4 trimethylbenzene and n-undecane derived from motor vehicle emissions. F3-per (variance, 9.54%) correlated with benzene, n-heptane, n-octane, ethylbenzene, styrene, o-xylene, n-nonane and n-decane and was identified as representing industrial emissions. Finally, F4-per factor (variance, 8.10%) correlated with nhexane, benzene and styrene and was identified as a smoking factor.

4 Conclusions

The present study has yielded up-to-date information concerning the level of several airborne chemicals in different microenvironments in the city of Kocaeli. The effect of outdoor air pollution on indoor air pollution was evaluated. The I/O values of target compounds ranged from 0.42 to 3.03 and 0.93 to 6.12 during the summer and winter, respectively. The multivariate receptor modelling technique based on FA was applied to the data sets in an effort to identify the VOC sources contributing to indoor, outdoor and personal exposure in homes and offices. The FA results earmarked industrial, motor vehicle and smoking factors as major sources VOCs. Participants exposed to cigarette smoke showed significantly higher personal exposure concentrations of benzene, sytrene, cyclohexane, n-hexane and n-undecane. Motor vehicles were identified as a potential source of all target VOCs investigated, with the exception of styrene. Although, the VOC composition of personal exposure samples reflected the complex mixture of microenvironments in which participants had spent their time, there were important similarities between indoor and personal exposure FA results for homes and offices. The combination of seasonal activity and VOC sources markedly influenced microenvironment

and personal exposure concentrations. The indoor and outdoor environments investigated possess VOC sources that increased indoor VOC concentrations to varying degrees, resulting in an increase in personal exposure to different pollutants. The data derived from this study is expected to be of value in efforts related to the identification of pollutants for the purposes of risk assessment and management.

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