BMC Public Health

Research article

Open Access

An analysis of factors that influence personal exposure to toluene and xylene in residents of Athens, Greece

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Published: 28 February 2006

*BMC Public Health*2006, **6**:50 doi:10.1186/1471-2458-6-50

Received: 28 July 2005
Accepted: 28 February 2006

This article is available from: http://www.biomedcentral.com/1471-2458/6/50

Abstract

Background: Personal exposure to pollutants is influenced by various outdoor and indoor sources. The aim of this study was to evaluate the exposure of Athens citizens to toluene and xylene, excluding exposure from active smoking.

Methods: Passive air samplers were used to monitor volunteers, their homes and various urban sites for one year, resulting in 2400 measurements of toluene and xylene levels. Since both indoor and outdoor pollution contribute significantly to human exposure, volunteers were chosen from occupational groups who spend a lot of time in the streets (traffic policemen, bus drivers and postmen), and from groups who spend more time indoors (teachers and students). Data on individual and house characteristics were obtained using a questionnaire completed at the beginning of the study; a time-location-activity diary was also completed daily by the volunteers in each of the six monitoring campaigns.

Results: Average personal toluene exposure varied over the six monitoring campaigns from 53 to 80 $\mu g/m^3$. Urban and indoor concentrations ranged from 47 – 84 $\mu g/m^3$ and 30 – 51 $\mu g/m^3$, respectively. Average personal xylene exposure varied between 56 and 85 $\mu g/m^3$ while urban and indoor concentrations ranged from 53 – 88 $\mu g/m^3$ and 27 – 48 $\mu g/m^3$, respectively. Urban pollution, indoor residential concentrations and personal exposures exhibited the same pattern of variation during the measurement periods. This variation among monitoring campaigns might largely be explained by differences in climate parameters, namely wind speed, humidity and amount of sunlight.

Conclusion: In Athens, Greece, the time spent outdoors in the city center during work or leisure makes a major contribution to exposure to toluene and xylene among non-smoking citizens. Indoor pollution and means of transportation contribute significantly to individual exposure levels. Other indoor residential characteristics such as recent painting and mode of heating used might also contribute significantly to individual levels. Groups who may be subject to higher exposures (e.g. those who spent more time outdoors because of occupational activities) need to be surveyed and protected against possible adverse health effects.

Background

Toluene and xylene (o-, m- and p-isomers) are widespread in the environment because they have been used in many commercial products. Vehicle exhaust is considered to be the main source but non-professional use of paints, glues, adhesives, varnishes, lacquers, shoe polishes and cigarette smoke contribute significantly, especially to levels in the internal environment and to personal exposure [1-3].

At high concentrations, toluene and xylene have serious adverse effects on human health. Toluene vapor is heavier than air and may travel along the ground, but harmful air contamination levels can be reached rather quickly by evaporation of toluene at 20°C. Toluene has a lower clearance rate in winter than in summer. The acute and chronic effects of toluene, and to a lesser degree of xylene, on the central nervous system are of most concern. In addition, there is concern about potential effects on reproduction and hormone balance in women, and hormone imbalances in exposed males have also been reported [4-6].

In occupational settings, official agencies (OSHA, ACGIH, HSE, DFG) have established an 8-hour time-weighted average permissible exposure limit (TWA PEL) of 100 ppm (375 mg/m^3 and 435 mg/m^3 for toluene and xylene isomers, respectively). Levels only slightly above the 8-hour TWA may cause lack of coordination and amnesia. Even levels lower than 50 ppm cause drowsiness, moderate fatigue and headaches [7]. The World Health Organization has recently recommended a guideline for ambient toluene exposure to protect against developmental neurotoxicity: 260 μg/m^3 (68 ppb) as a weekly average concentration.

Athens, like most large cities all over the world, has air pollution problems. These were made worse by poor town layout, location (surrounded by mountains), intense sunlight and low wind speed. Emission of toluene has been calculated as 20 million tons per year, one of the largest among European towns [8]. In Athens, fixed point monitoring stations have very recently started to measure hydrocarbon concentrations, but very little comparative research has been conducted to give consistent estimates of exposures of individuals in the general population or of indoor pollution [9-13]. The main goal of the present study was to estimate personal exposures to toluene and xylene by monitoring fifty volunteers, their homes and various urban sites over one year. Another goal was to examine the relationship between toluene and xylene pollution in indoor, urban and personal air. Benzene measurements included in the study have already been published elsewhere [14].

Methods

Sampling

A radial symmetry sampler, Radiello® Model 3310 Passive Sampling System, was used. This sampler works by spontaneous transfer of gas molecules through a diffusive barrier. It comprises a cylindrical microporous diffusive body and an absorbing cartridge, also cylindrical, placed coaxially inside the diffusive body. When the assembled apparatus is exposed, it is necessary only to note the days and times when exposure begins and ends [15]. Subjects and their homes were also equipped with radial path diffusive samplers (Radiello) to measure the time-weighted average (TWA) concentrations of toluene and xylene in the breathing zone over a 108-hour period. At night, personal samplers were set on the volunteers' bedside tables. The samplers were desorbed with carbon disulfide, shaken for 30 minutes and analyzed by gas chromatography coupled with mass spectrometry. This analytical method has been described in detail elsewhere [15]; its detection limit is 0.2 μg/m^3. The overall reliability of the sampling device has been judged as excellent by the European Reference Laboratory for Air Pollution (ERLAP) of the Joint Research Centre of Ispra and the sampler has been widely used [16,17]. Measurements with these samplers performed contemporaneously with ours gave comparable results [9,13].

The whole sampling campaign was repeated every two months over one year (September 1997 to September 1998). Each monitoring campaign lasted from Monday morning to Friday evening and used Radiello samplers in 100 sites around Athens. Fifty volunteers and their homes were monitored over the same period. The campaign included 2400 measurements and losses did not exceed 7% in any sampling period. The experimental database comprised 1140 environmental data and 556 and 564 personal and home pollution data, respectively.

The 100 sampling sites chosen for urban monitoring were distributed on the intersections of a multi-scale grid drawn over the town map. The mesh size was approximately 400 m. Eighty percent of the sites lay in the city center and its periphery (i.e. almost 13 squares kilometers). The other 20% lay in two background zones (northeast and southeast) with sparse traffic and parks. Each sampling site was uninterruptedly monitored from Monday morning (6 to 8 am) to Friday afternoon (6 to 8 pm) on six occasions through the year. On site, each sampler was placed inside a shelter hung on a lamppost about 3 m high. Personnel employed to place the samplers at the beginning of the campaign and collect them at the end were trained and supervised by members of the research project (CC & ECA). The volunteers were selected on the basis of expected exposure level. People who for occupational reasons spend a lot of time in the street, such as bus drivers, policemen and postmen, composed the first group. The lower expected exposure group comprised teachers and students. Volunteers were selected on the basis of two criteria: non-smoking and job located in the city centre. Ten persons were selected in each of the following categories: teachers, students, bus-drivers, postmen and policemen. All volunteers gave their written informed consent. After the 50 volunteers were selected, personal exposure was determined directly over the series of six 108-hour (4.5 day) sampling periods. The personal sampler used was attached to the volunteer's lapel and during the night was set on the bedside table. At the end of each day in each sampling period, volunteers were instructed to complete a diary stating their activities during the sampling period.

The home microenvironments monitored were located in the greater Athens basin. Although all volunteers worked in the city centre, about two thirds of their houses were located outside the city centre in less urban regions, and a few houses were located in the Athens suburbs. These areas had lower population and traffic densities and therefore were typically less congested, and ambient air concentrations were expected to be lower.

Statistical analysis

Univariate analyses were performed to examine the covariates age, sex, room-mates' smoking habits, job title, time spent outdoors, transportation mode and house characteristics (floor, location, recent painting, heating mode, proximity to gasoline station and proximity to busy road). We applied linear mixed-effects regression models to estimate the significant prognostic factors for toluene and xylene exposure levels using maximum likelihood and restricted/residual maximum likelihood methods [18]. Mixed effects models had the advantage of adjusting for invariant factors by fixed-effects models and accounting for individual differences by random- effects models [19]. In our mixed-effects models, we treated subjects' personal and home characteristics as fixed effects and each subject as a random effect. The measurement period was used to identify repeated observations. Type III sums of squares were used to calculate the effects in the models. The multivariate linear mixed model included all the variables that contributed significantly to the final model (Wald statistics, criterion $p < 0.05$). For each factor, the regression coefficient and 95% confidence interval (95% CI) were calculated. All statistical analyses were performed with SPSS 11.0 software.

Results

The study population had an average age of 36.3 years and consisted predominantly of men (66%). Each sampling campaign lasted 108 hours. In each measurement period, subjects spent an average of 26.3 hours outdoors (sd 13.8). Time spent outdoors differed markedly between occupational groups. Teachers and students spent, as an annual average, less than 15 hours per sampling period outdoors, 50% during their free time, while bus drivers and traffic policemen exceeded 40 hours outdoors, almost 80% during work activities. In Table 1, the job and house characteristics of volunteers are presented along with the corresponding measurements of toluene and xylene (see also additional file 1, Table 1, for further residential characteristics). An arterial road with traffic density of more than 5000 vehicles per day witch judged independently by two professional drivers as busy road.

Urban levels, indoor residential concentrations and personal exposures to toluene and xylene during the six sampling periods are presented in Tables 2 and 3. Table 4 shows average wind speed, humidity, amount of sunlight and temperature for all six measurements. As shown in these tables, personal exposures varied between 53 – 80 and 56 – 85 μg/m^3 for toluene and xylene, respectively. The higher values were observed mainly during the first two periods i.e. Autumn and Winter, when the wind speed did not exceed 0.5 m/s. Indoor residential concentrations varied between 30 and 51 μg/m^3 and 27 and 48 μg/m^3 for toluene and xylene, respectively. Urban levels ranged from 47 to 84 and 53 to 88 μg/m^3 for toluene and xylene, respectively. Personal exposures followed similar trends to urban and indoor pollution throughout the study period (Figures 1 and 2).

Correlations between personal exposures and indoor concentrations were weaker for xylene than for toluene, and, as expected, for other seasons than for winter. The correlation coefficient (r) between personal exposure and residential level varied between 0.33 and 0.82 (all campaigns 0.57) for toluene and between 0.20 and 0.65 (all campaigns 0.39) for xylene (see additional file 1, Table 2).

Personal exposures to and indoor concentrations of toluene and xylene are presented in Figures 3 and 4 according to occupational group. Teachers and students differed significantly from other groups in respect of personal exposure to both toluene and xylene ($p < 0.01$).

Indoor pollution

For xylene, indoor pollution was influenced by location (centre or suburb), proximity to busy road and proximity to gasoline station (Table 1). Heating mode, recent painting, and type/floor of house also affected indoor concentrations, but owing to small numbers did not reach a conventional level of significance (Figure 5; see also additional file 1: Table 1). The use of oil and natural gas ovens during the 2nd and 4th sampling periods (the colder ones) raised indoor concentrations (mainly for xylene, $p = 0.065$) and personal exposures, but there is a high likeli-

Table 1: Job and house characteristics of volunteers (n = 50) andcorresponding annual (median) toluene and xylene measurements (μg/m³)

			Toluene (μg/m³)		Xylene (μg/m³)	
	n	%	Personal	House	Personal	House
Transportation means						
By foot	7	14	46.0		52.4	
By foot and vehicle	35	70	56.0		55.9	
By vehicle	8	16	71.6[b]		74.9[b]	
Use of vehicle in work						
Yes	28	56	72.5[b]		75.9[b]	
No	22	44	52.0		55.4	
Smoker room-mate						
Yes	15	30	64.0	40.2	65.9	42.3
No	35	70	63.0	38.5	67.7	35.8
City area						
Center	42	84	64.0	39.8[a]	67.7	38.9[a]
Suburbs	8	16	55.2	26.5	65.3	23.2
Proximity to gasoline station						
Yes (< 50 m)	3	6	91.9[a]	60.9	98.1[a]	52.0[a]
No	47	94	61.7	38.1	65.9	35.2
Proximity to busy road						
Yes (< 50 m)	16	32	65.5	42.0[a]	67.2	46.0[a]
No	34	68	63.0	37.1	67.4	33.8

Significant differences, [a] $P < 0.05$, [b] $P < 0.01$

hood of effects related to other sources of exposure since few homes had such types of heating. The five houses with fireplaces exhibited low indoor concentrations because they were all located in the less polluted suburbs. Finally, multivariate analysis of indoor residential concentrations showed that only seasonal or climate variation, mainly wind speed, remained an important factor for both toluene and xylene; indoor concentrations decreased by 40–50 μg/m³ per 1 m/s increase in wind speed.

Personal exposure

Occupational group, transportation mode and the use of vehicle during work had significant impacts on personal exposure to toluene and xylene (Table 1, Figures 3 and 4). Heating mode and recent painting had effects of borderline significance ($0.05 < p < 0.10$) on personal exposure to toluene and xylene (Figure 6).

Table 5 shows the important prognostic factors in the multivariate analysis of personal exposure to toluene and xylene. Adjusted for seasonal/climate variation, personal exposure was determined by indoor residential concentration and time spent outdoors in the city center; and, for toluene, the means of transportation also contributed.

Discussion

In this study, non-smoking citizens of Athens were selected among people who, owing to occupational duties, spent a lot of time in the streets, and people who spent more time indoors in schools or offices. Those who

Table 2: Personal, indoor and outdoor concentrations of toluene (μg/m³) in the six periods

	Personal			Indoor			Urban		
Period	Mean	Median	10th–90th percentiles	Mean	Median	10th–90th percentiles	Mean	Median	min – max
1st – SEP	90.7	79.6	53.0–130.8	58.4	50.6	23.1–81.6	99.8	84.1	37.3–256.8
2nd – DEC	72.0	69.6	46.1–120.7	57.2	41.0	21.5–131.3	76.7	68.2	22.7–225.6
3rd – FEB	74.9	63.0	35.2–123.1	46.6	39.6	13.8–68.4	74.8	65.5	34.2–143.8
4th – APR	80.1	58.9	34.7–147.8	48.2	37.0	15.9–79.9	59.6	53.9	19.5–125.4
5th – JUN	58.2	53.3	26.2–97.8	44.9	30.4	16.2–97.0	53.3	46.9	18.2–113.1
6th – SEP	69.4	59.0	30.7–109.1	40.5	33.1	17.5–67.6	77.7	66.3	29.0–241.1
Annual	74.3	63.0	34.1–115.5	49.2	38.9	19.4–82.5	73.6	63.3	18.2–256.8

Table 3: Personal, indoor and outdoor concentrations of xylene (μg/m³) in the six periods

	Personal			Indoor			Urban		
Period	Mean	Median	10th–90th percentiles	Mean	Median	10th–90th percentiles	Mean	Median	min–max
1st – SEP	89.4	84.6	53.5–141.9	49.6	48.4	20.3–79.6	101.5	88.3	41.4–263.0
2nd – DEC	75.9	71.6	49.1–113.8	56.4	39.7	22.1–117.0	85.6	74.6	28.0–180.5
3rd – FEB	74.5	63.8	39.7–141.3	42.7	39.2	16.9–67.9	85.6	75.8	41.2–197.9
4th – APR	83.1	63.2	30.7–147.7	45.7	37.1	18.7–92.9	71.5	63.0	23.2–167.4
5th – JUN	60.7	56.0	26.6–97.5	32.3	27.4	16.2–49.4	64.0	53.3	21.1–160.9
6th – SEP	75.5	63.9	38.2–122.8	36.5	31.9	20.9–51.5	88.8	76.4	35.4–229.2
Annual	76.6	67.4	37.8–123.3	43.7	36.2	19.5–72.9	82.8	72.3	21.1–263.0

Table 4: Climate parameters (means) during the six monitoring campaigns

		Temperature (°C)	Wind speed (m/s)	Humidity (%)	Sunlight (hours)
PERIOD	1st – SEP	23.2	0.5	57	9.9
	2nd – DEC	13.0	0	70	4.2
	3rd – FEB	12.5	2.4	81	2.7
	4th – APR	10.4	2.2	62	7.6
	5th – JUN	23.6	4.0	54	10.2
	6th – SEP	21.8	3.7	69	7.2

spent more working or leisure time in the heavily polluted city center were expected to be highly exposed, given the pattern of pollution of Athens (i.e. pollution comes from outdoors). Although inferences about a population are problematic when volunteers are used because of the potential for selective participation, the use of volunteers is suitable for investigating the relationships among predictors of personal exposure [20].

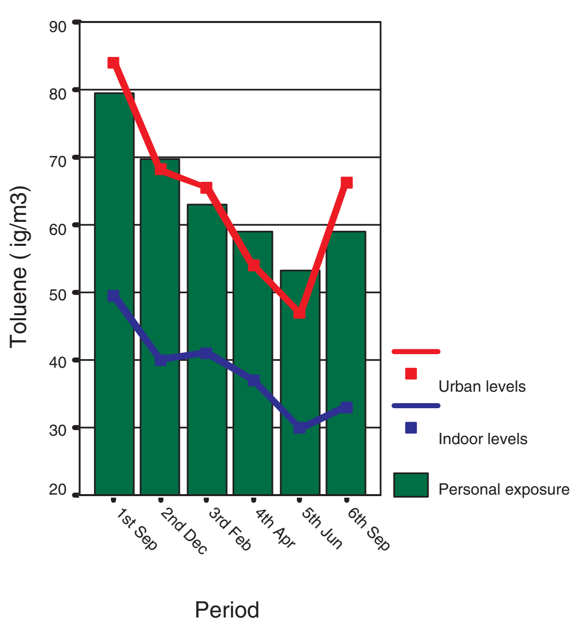

Figure 1
Toluene levels (median values) during monitoring periods.

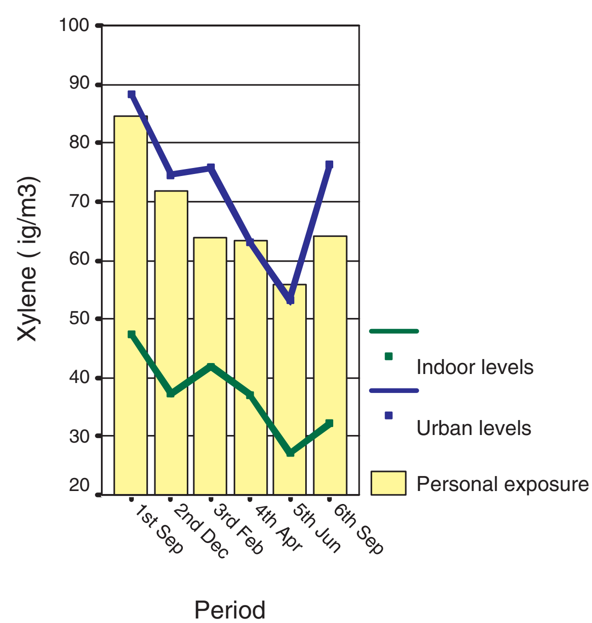

Figure 2
Xylene levels (median values) during monitoring periods.

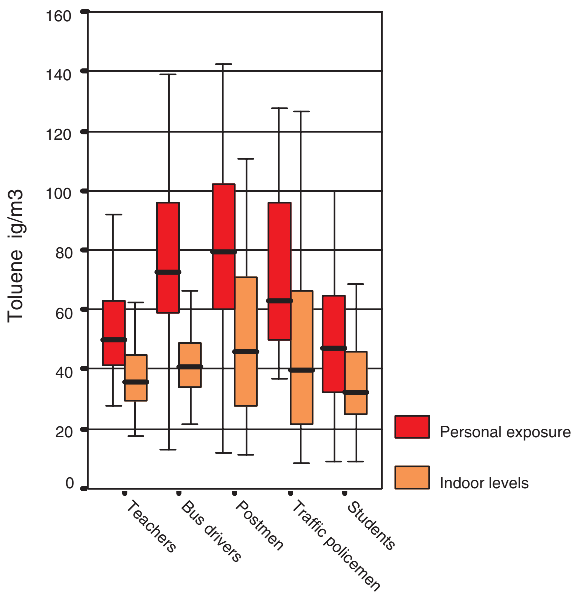

Figure 3
Average annual toluene levels among occupational groups.

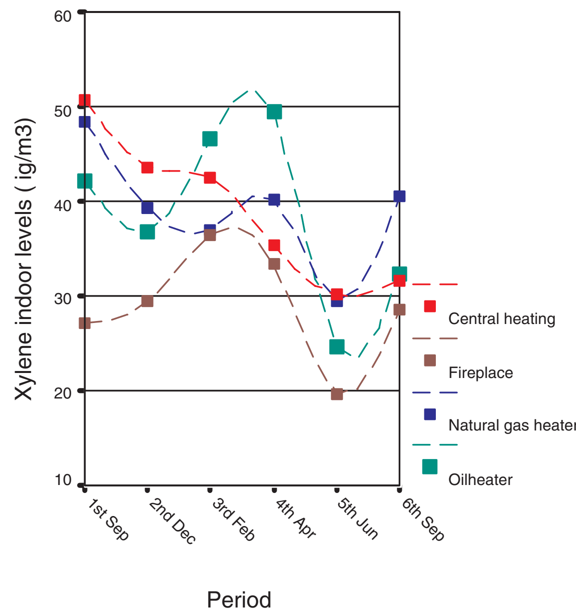

Figure 5
Heating mode and indoor residential xylene concentrations during monitoring periods.

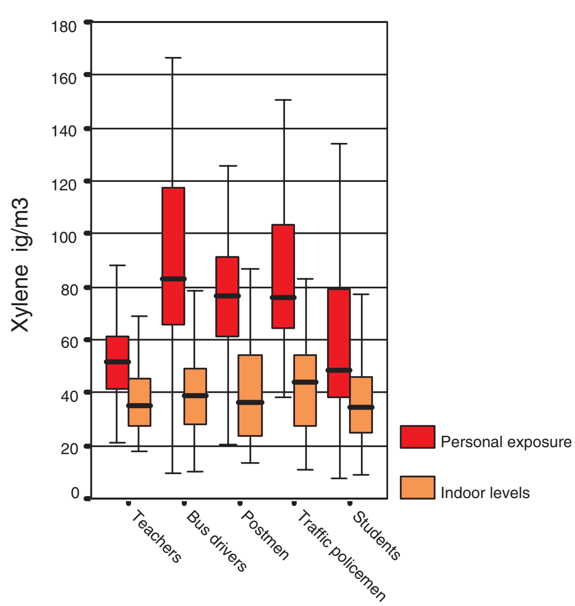

Figure 4
Average annual xylene levels among occupational groups.

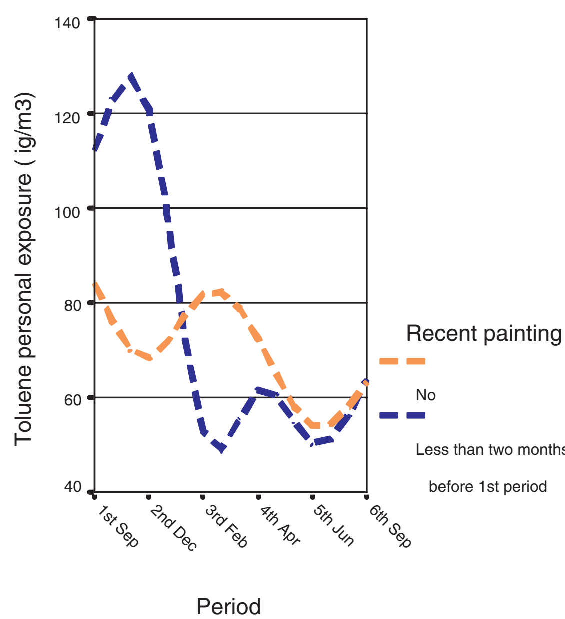

Figure 6
Recent painting in participants' houses and personal exposure to toluene.

Table 5: Multivariate associations between personal exposure to toluene and xylene (μg/m³) and characteristics of study population

	TOLUENE beta (95% CI)	P	XYLENE beta (95% CI)	P
Transportation means				
Foot	RC			
Vehicle	19.13 (4.6, 33.6)	0.012	NS	
Both	5.44 (-12.6, 23.5)	0.541		
Time spent outdoors (hours)	0.582 (0.22, 0.94)	0.003	1.184 (0.78, 1.59)	$<10^{-3}$
Home levels (μg/m³)	0.554 (0.45, 0.65)	$<10^{-3}$	0.561 (0.38, 0.74)	$<10^{-3}$
Wind speed (m/s)	-54.15 (-89.9, -18.4)	0.004	-41.03 (-68.1, -13.9	0.004
Humidity (%)	2.96 (1.3, 4.7)	$<10^{-3}$	2.53 (0.9, 4.2)	0.003
Sunshine (hour)	10.46 (4.4, 16.5)	0.001	9.23 (3.5, 14.9)	0.002
R^2	0.43		0.33	

Beta: regression coefficient; CI: confidence interval; RC: reference category; NS: not significant

Urban levels of toluene and xylene were generally high. The average annual indoor residential concentrations were found to be 39 μg/m³ and 36 μg/m³ for toluene and xylene, respectively. These levels are in the same order as those found in other studies in Athens and cities with similar characteristics, and lower than some other Mediterranean cities [9-13,21-28]. The average annual personal exposures were found to be 63 μg/m³ and 67 μg/m³ for toluene and xylene, respectively. Urban toluene pollution very rarely (<2% of all measurements) exceeded the limit of 260 μg/m³ proposed by WHO as a weekly average. In this particular setting, the toluene and xylene measurements probably represented the expected levels of exposure of the vast majority of non-smoking citizens of Athens who were not occupationally exposed.

Urban and indoor air pollution and personal exposure exhibited the same pattern of variation among the measurement periods. This variation among monitoring campaigns might be explained by differences in climate parameters, namely wind speed, humidity and amount of sunlight. It was also anticipated that changes in transport and modernization of the bus fleet during the study period would to some extent have influenced atmospheric pollution levels, especially in the city centre. The most important change was a project for replacing the old buses entirely with new ones equipped with anti-pollution devices. More than 100 buses were replaced during the study period; in addition, increased numbers of dedicated bus lanes improved running conditions.

Indoor concentrations of toluene and xylene are often significantly higher than external air levels [29,30]. Lebret found an internal/external ratio of 8 in Holland, while Gilli found a ratio of 3 in Torino, Italy [31,32]. In contrast, our results showed an indoor/outdoor toluene air pollution ratio of 0.64 for houses within the environmental sampling areas, although the samples were not obtained directly outside the homes. The ratio for xylene concentrations was 0.54. Probably poor emission materials and characteristics related to the Athens climate, such as frequent ventilation and use of non-absorbent materials for wall and floor coverings, account for this result. The frequent ventilation of houses in Athens lowered indoor pollution because such ventilation usually takes place when urban pollution is lower (not rush hour) and there are putative indoor emission sources, so the infiltration of outdoor air is beneficial. This argument is supported by the finding that differences between outdoor and indoor levels were lower during the spring and summer periods (continuous infiltration of outdoor air). Compared to others studies, we found weak but not significant effects on indoor air pollution from environmental smoke, heating mode, type and floor of house, and recent painting [2,3,29-34].

Indoor air contributes significantly to human exposure. In poorly-ventilated buildings, indoor emission sources have a more significant influence on hydrocarbon concentrations than infiltration of outdoor air [[26,35] and [1]]. In our study, correlations between personal exposure and indoor concentrations were higher for toluene than for xylene and lower in summer than other seasons, probably because factors outside houses have a predominant effect on the personal exposure pattern. Adjusted for climate variation, significant factors influencing personal exposure were indoor pollution, total time spent in the polluted city center, and means of transportation used.

In contrast to other studies, we found no differences among urban BTX ratios during the measurement periods, but there were differences in personal exposure and especially in indoor concentrations (see additional file 1, Table 3) [14,18,24]. This finding might be explained by the putative effects of indoor sources. The correlation between toluene and xylene was quite good (coefficient

0.59–0.86), similar to those reported in other studies [23,29]. Comparing the factors that influence exposure to benzene, toluene and xylene, it was found that proximity to a gasoline station or a busy road, and a smoker roommate, had a much greater impact on both personal exposure and indoor residential levels of benzene (though volunteers were instructed to avoid exposure to environmental smoke and gasoline refueling). On the other hand, the factors that had some degree of influence on toluene and/or xylene concentrations, such as heating mode and recent painting, had no impact on benzene levels [14,18].

Conclusion

In Athens, Greece, the time spent outdoors during work or leisure is a major contributor to hydrocarbon exposure among non-smoking citizens. Indoor pollution and means of transportation contribute significantly to individual levels. Other factors such recent painting, smoking room-mate and mode of heating might contribute significantly to individual levels of toluene and xylene exposure. Identification of specific groups of people who may be subject to higher exposure is an important step towards preventing possible adverse health effects and a prerequisite for evidence-based intervention.

Competing interests

The author(s) declare that they have no competing interests.

Authors' contributions

ECA participated in data collection, carried out the statistical analysis and drafted the manuscript. CC participated in the design, coordination of the study, data collection and helped in the revision. AL participated in the design and coordination of the study and helped in the revision. All authors read and approved the final manuscript.

Additional material

Additional File 1

***Sample contains Tables** 1, **2and** 3 Table 1 shows house characteristics in relation to annual (median) toluene and xylene measurements (µg/ m³)Table 2 shows correlation coefficients between personal exposure and indoor residential concentration during the six monitoring campaignsTable 3 shows Benzene:Toluene:Xylene (BTX) ratios during the measurement periods*

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Acknowledgements

The authors are grateful to reviewers for useful comments.

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Pre-publication history

The pre-publication history for this paper can be accessed here:

http://www.biomedcentral.com/1471-2458/6/50/prepub