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An approach for the evaluation of exposure patterns of urban populations to air pollution

P. Pérez Ballesta[a,*], R.A. Field[a], R. Fernández-Patier[b], D. Galán Madruga[b], R. Connolly[a], A. Baeza Caracena[a], E. De Saeger[a]

[a]*European Commission—Joint Research Centre, Institute for Environment and Sustainability, TP-441, 21027 Ispra (VA), Italy*
[b]*Instituto de Salud Carlos III, Ministerio de Sanidad y Consumo, Majadahonda, Madrid, Spain*

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Abstract

Exposure concentrations of aromatic compounds were correlated with variables derived from time-microenvironment-activity (TMA) diaries to understand the relationship between exposure patterns and commuting behaviour of the population in a case study in Madrid. Approximately 200 air pollution samples were taken during a one-day campaign by means of diffusive samplers. An approach to determine the importance of selected activities and locations relative to a baseline condition, defined by a sample of approximately 100 commuters is described. A regression model is applied to determine the relative importance of identified situations, whether related directly to transport behaviour or through a labelled situation. The regression defines a baseline exposure concentration level where activities act as multiplying factors. The relationship of this baseline level and the set of activity factors, assigned to the population considered, to the ambient background is explored for its application to future studies. The calculation of exposure concentration gradients from the regression provides a means to characterise of the relative importance of different activities. A good level of agreement, in particular for benzene, was evident between the observed exposure concentrations and those calculated using the regression model.

Keywords: Air quality monitoring; Personal exposure; Exposure assessment; Diffusive samplers; Aromatic compounds; Benzene

1. Introduction

In recent years the influence of even low concentrations of air pollutants on human health has re-emerged as an important scientific issue (Dockery et al., 1993; Pope et al., 1995; Laden et al., 2001). Numerous studies have linked various acute and chronic health impacts to air pollution (Clancy et al., 2002; Goodman et al., 2004). Regardless of the pollutant considered, a major concern has been the use of various measures of ambient air quality as a surrogate for pollution exposure, either due to underestimating the importance of indoor air quality (Roosbroeck et al., 2007) or overestimating the applicability of background sites (Field et al., 2005). Epidemiological studies that relate air pollution with the health of human populations often rely upon measurements undertaken to

*Corresponding author. Tel.: +39 0332 78 5322; fax: +39 0332 78 9364.
E-mail address: pascual.ballesta@jrc.it (P. Pérez Ballesta).

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determine compliance of limit values required for air quality legislation (Jerrett et al., 2005). Such compliance monitoring sites are situated within areas of higher population density, usually away from the influence of local emission sources. This approach is necessary given air quality monitoring network design, as background levels allow for an understanding of longer term trends related to the relationships between emission sources, meteorology and pollution control policies.

The application of air quality standards, through limit values, has been successful in reducing urban concentrations (WHO, 2000). Nevertheless, in recent years the scientific community has become increasingly concerned with the determination of actual levels of exposure for the general population to air pollutants (WHO, 1999). In order to address this central question in the field of urban air quality the European Commission instigated the Population Exposure to Air Pollutants in Europe (PEOPLE) project. A previous paper reported on the broad relationships observed between ambient background, hot spot and commuting populations for six European capital cities. For these cities, the commuting population was exposed to air pollution levels 1.5 times that of background levels and 0.6 times that of hot spot levels (Pérez Ballesta et al., 2006). While the afore-mentioned relationship reinforces the importance of background monitoring, it does not explain the causes for the elevation of pollution levels of the monitored volunteers above the urban background level.

The purpose of this study is to introduce an approach that determines the relative importance of time spent in a variety of labelled activities and locations measured through time-microenvironment-activity (TMA) diaries. This was achieved through a regression analysis approach that estimates the importance of these different situations relative to a baseline condition. This approach is different from other methodologies that aim to either define factors that attribute sources with exposure concentrations or estimate population exposure. The former approach, attribution, relies upon statistical methods such as principal components analysis (Kim et al., 2002). The latter approach, estimation, can be broadly categorised as: (i) surrogate or (ii) microenvironmental. Both of these indirect approaches expand measured or modelled situations to apply to wider populations. Surrogate approaches are often represented through extension of air quality networks or other base data through land use or other GIS-based transformations (Madsen et al., 2007) that often focus upon the importance of traffic emissions (Buliung and Kanaroglou, 2006; Beelen et al., 2007). The approach adopted depends on the pollutant and spatial scale considered, and is often based upon dispersion modelling (Bellander et al., 2001) although statistical models (Brauer et al., 2003) and even satellite imaging techniques have also been applied (Liu et al., 2005). Nitrogen dioxide has been evaluated in a number of different urban areas, due to the large amount of available network monitoring data and the ease of measurement through diffusive sampling (Stedman et al., 1997; Kanaroglou et al., 2005). Despite confounding factors, namely; indoor emission sources related to cooking and heating and outdoor stationary combustion sources, a number of studies have developed good relationships between predicted and measured levels of population exposure through both dispersion and emission modelling approaches (Bartonova et al., 1999; Kousa et al., 2002). A number of studies have focused upon the applicability of urban air quality networks as an indicator of air pollution exposure (Baldauf et al., 2001; Chow et al., 2002; Kanaroglou et al., 2005). Microenvironmental approaches developed from total human exposure methodologies (Wallance et al., 1986) often combine activity profiles concentration data (Edwards et al., 2001, 2005) This approach is analogous to building an air pollution inventory that represents the total emissions into a given area. The methodological limitations of both of these approaches, due primarily to the uncertainty associated with input data or applied factors, are accepted given the high financial and organisational costs associated with the wide scale direct monitoring of population exposure.

In this paper, we do not attempt to predict population exposure, through extrapolation of data or use of surrogate information. Instead, we describe an approach to determine the importance of activities and locations relative to a baseline condition, defined by a sample of approximately 100 commuters. Commuters were selected as they are representative of a large portion of the urban population. The exposure of urban populations is known to be affected predominately by emissions from transportation. This study differs from previous transnational European studies, MACBETH and EXPOLIS in that it considers commuters. While all three of these studies considered the relationship between background air quality and

human exposure the former study focused on groups that represented high and low exposure conditions (Cocheo et al., 2000) while the latter study centred upon indoor situations (Jantunen et al., 1998). Rather than attempting to relate emission factors to ambient background (Latella et al., 2005) or estimate the contribution of different transport sources to exposure (Kanaroglou and Buliung, 2008), we attempt to understand the relative importance of identified situations, whether related directly to transport behaviour or through a labelled position. Through commuting and living in cities people are exposed to ambient air concentrations that are different primarily because of the dispersion of mobile sources. This is widely recognised in a range of scientific communities and has led to a wide variety of strategies to reduce the influence of transport emissions primarily through urban planning and emission control (COM, 2004, 2005). In this paper the relationship of ambient background to a baseline condition, set by the characteristic of the city and population behaviour, is then explored to determine the utility of this approach for future studies. For pollutants with a variety of emission sources, whether indoor or outdoor, ambient data are less likely to give a good estimate of actual exposure. Understanding the main factors that influence the exposure of people is important for pollution control policy. The presented data are from a campaign carried out in Madrid on 3 December 2003 as part of the European Commissions transnational PEOPLE project.

2. Methodological design

The monitoring campaign consisted of outdoor, microenvironment and personal exposure measurements of benzene and other aromatic hydrocarbons by diffusive sampling. Outdoor and microenvironmental sampling was carried out for a period of 24 h, while personal monitoring was restricted to 12 h. Outdoor sampling was performed at 36 urban background sites that had the characteristics required for compliance monitoring of air quality directives and as such are considered to represent the background exposure of the population. Microenvironment sampling was performed at typical indoor locations. A total of 36 diffusive samplers were assigned to indoor locations and particular microenvironments, which included homes, offices, shops, schools, bars and taxis. Volunteers for personal sampling were approached through media advertisements; approximately 300 people responded whose data were entered into a coded database. Over 100 citizens were then selected through the completion of a screening questionnaire designed to define commuting behaviour. These volunteers were trained in the correct handling, operation and subsequent storage of the diffusive sampler. Transport choices included private cars, buses, metro, bicycles and walking. The majority of the selected volunteers were sorted into a variety of different groups that reflected commuting behaviour. A separate group of commuting smokers was also derived from the volunteer database. A number of people that stayed inside their homes and were not directly exposed to transport sources or cigarette smoke were also chosen as a control group. The personal sampling volunteers were further instructed on how to record daily activities with a simple TMA diary. This one page diary included the four main categories: transport (walk, car, bus and metro/train), inside (home, work/office, cafe/bar, shop and restaurant), outside (street) and smoking (self and same room). Other possible variables defined as necessary by the participant could be included under comments. The TMA diary was capable of defining a 5-min resolution of these variables by circling pre-defined locations and activities. Most of the volunteers commuted from home to the office and then back home again, with movements generally during peak traffic periods. The set-up and implementation of the sampling required co-ordination between the European Commission's Joint Research Centre, national government departments, municipal authorities, media representatives and volunteer citizens. Field et al. (2005), describe the finer details of these procedures. After receipt of the samplers, a number were rejected due to either incomplete TMA diaries or incorrect sampler storage leaving 112 valid personal monitoring samples. The high recovery level of the samplers was a testament to the study design. This strategy should have utility in future studies as it allows cost effective assessments of large groups of people with a simple and lightweight device.

2.1. Sampling and analysis

Aromatic air pollutants were sampled by diffusive techniques, using a Radiello® diffusive sampler (Cod. RAD1202-Sigma-Aldrich) with Carbopack-X 40/60 mesh (SUPELCO Cod. 10436) as the adsorbent,

for periods of 12 and 24 h for personal and ambient measurements, respectively. Analyses were performed with a Gas Chromatograph AGILENT 6890 Series II connected to a thermal desorption unit Perkin-Elmer TURBOMATRIX ATD 50. The methodology has been extensively tested and validated according to protocols from the European Committee for Standardisation, CEN (EN 13528-1–3, 2003), as described in previous publications (Baldan et al., 1999; Bruno et al., 2005; Pennequin-Cardinal et al., 2005; Strandberg et al., 2005, 2006; Pérez Ballesta et al., 2006; González-Flesca et al., 2007). Diffusive sampling has been normalised by CEN to achieve the EC benzene Directive data quality objective for fixed measurements (EN 14662-5, 2005; EU, 2000).

3. Results

Fig. 1 shows the concentrations of benzene, toluene, ethyl-benzene and *m*,*p*-xylene in a number of microenvironments together with the outdoor city background measurements. The ambient air quality measurements were suitable as defining city background for two main reasons. First the sampling positions were either at European urban background network air quality Directive monitoring sites or at locations with equivalent characteristics. Such sampling sites are representative of the general area rather than that of a specific location. Secondly, a determination of the minimum number and most appropriate spatial positions was undertaken to ensure adequate coverage of the size of this

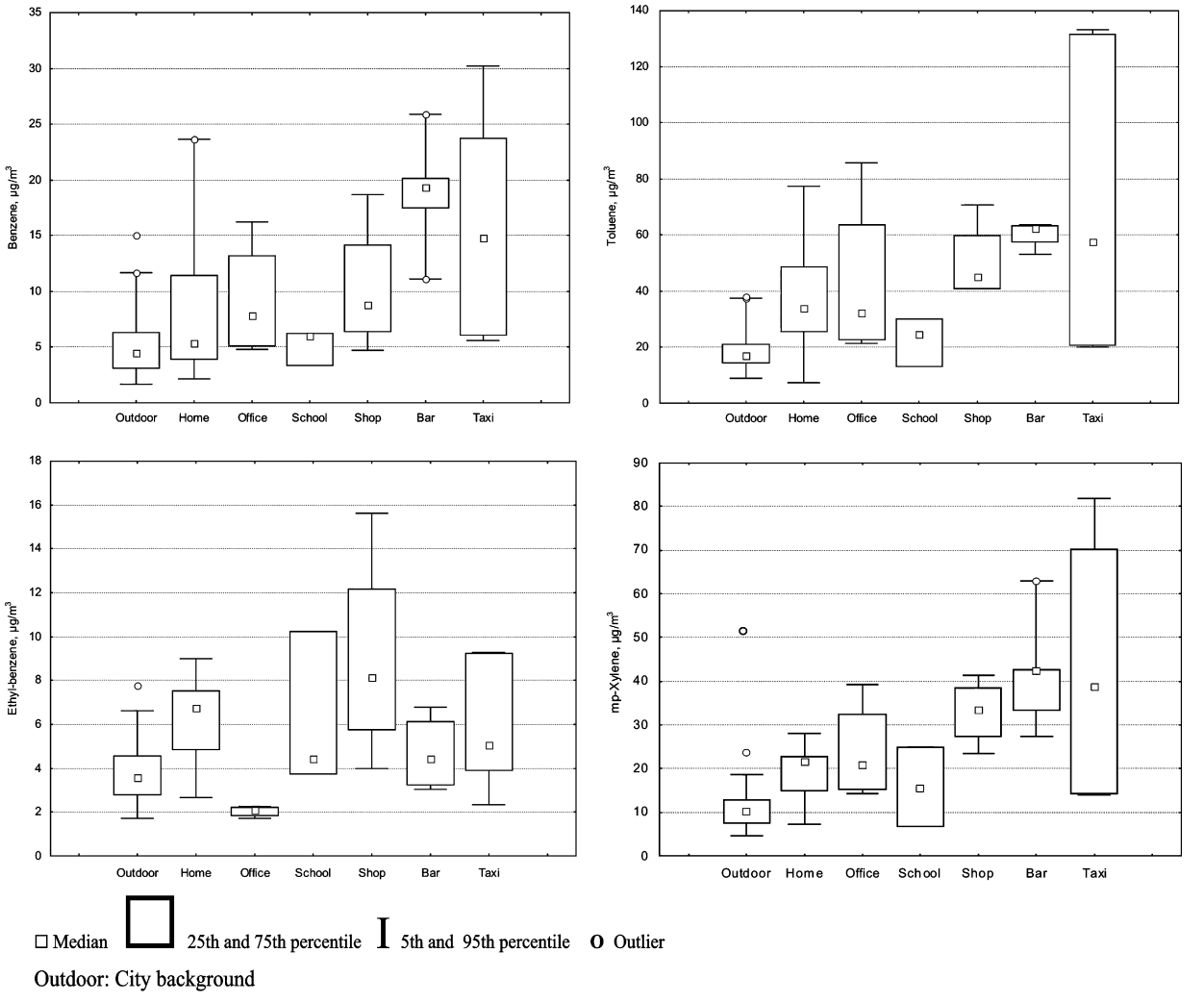

Fig. 1. Levels of the aromatic compounds measured in microenvironments in Madrid (3 December 2003).

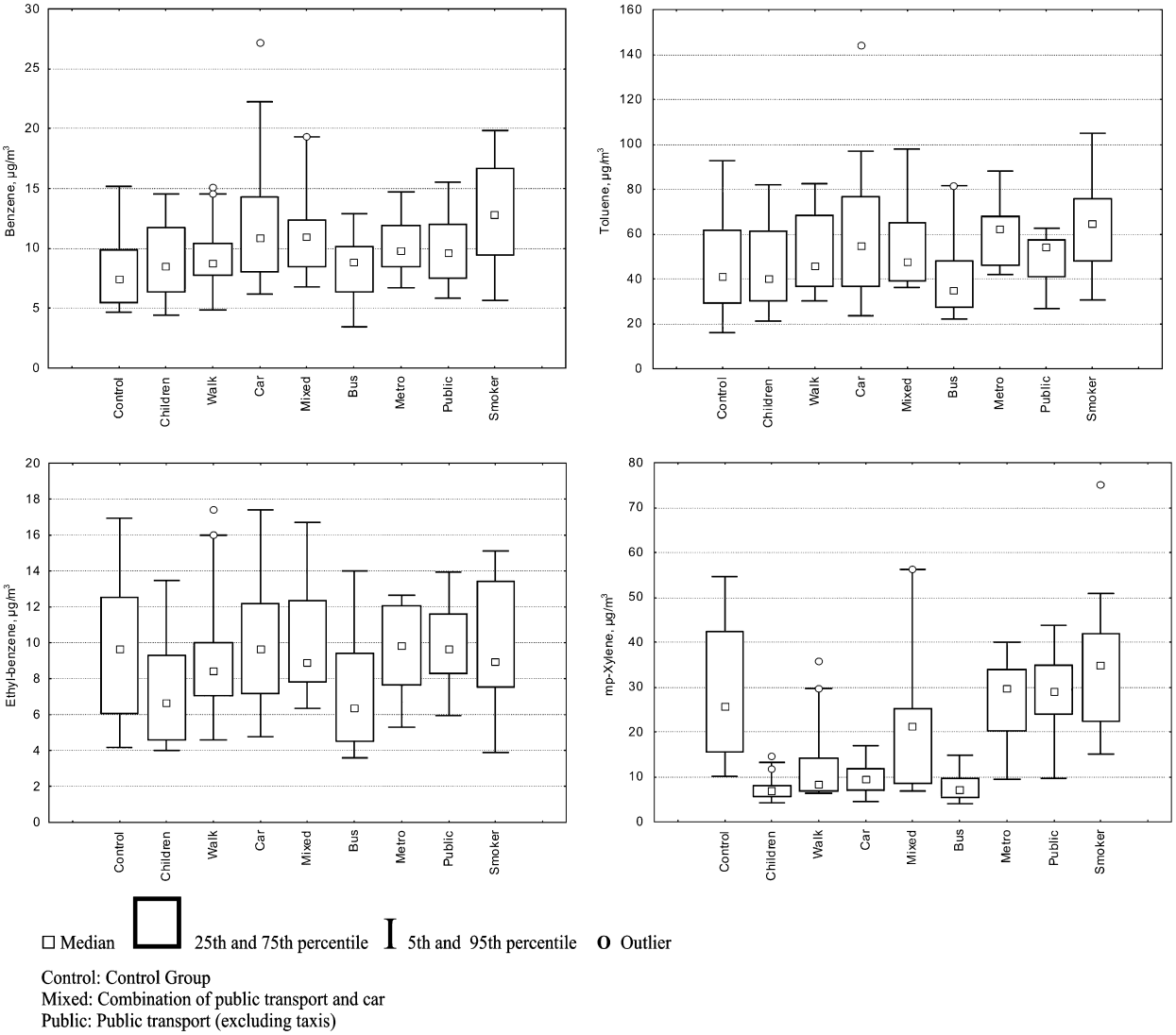

Fig. 2. Personal exposure levels on the day of the campaign in Madrid (3 December 2003).

city (Pérez Ballesta et al., 2001; Wright, 2002; Galán Madruga, 2004).

Thirteen homes, seven taxis, five bars, four offices, four shops and three schools were monitored. The pollutants showed similar relative levels in these microenvironments that consistently exceeded those of the city background air. The increase in concentrations observed was least pronounced for benzene. In general, schools had the lowest concentrations while bars had the highest. Concentrations inside taxis were relatively high, and comparable to those obtained for bars. The behaviour of ethyl-benzene was, however, different for places where tobacco smoke was present, such as offices and bars; the mean ratio benzene/ethyl-benzene being around 4, i.e. 2–4 times the ratio determined in city background levels.

Fig. 2 shows the results of the personal monitoring of aromatic compounds. Personal exposure was measured for 112 citizens, who were classified according to their commuting behaviour. The higher individual values were associated with groups that included travelling by car. The concentrations are presented as different exposure groups based upon the actual commuting behaviour as determined from the completed microenvironment activity diaries. When diaries were not completed properly the sample was removed from the data set. The following sampler numbers were assigned to these categories: 13 control, 28 smoker, 30 car, 17

mixed transport (car and bus), 10 public transport (bus, metro or train) 8 metro and 6 bus. A group of 27 children was extracted from the commuting categories for comparison with the adult working population.

The values for each measured species in the control group were comparable to those obtained for homes in Fig. 1. This was anticipated as these participants spent most of their sampling time at home. On the other hand, the mean exposure concentrations of the control group were generally higher than those of the commuter groups for *m*, *p*-xylene and ethyl-benzene, comparable for toluene and lower for benzene, as expected from their source origins. In general, ambient air is characterised by ratios of benzene/ethyl-benzene from 0.8 to 2 depending on the proximity to motor vehicle sources, while emissions of tobacco smoke have much higher ratios of about 8 (Darrall et al., 1998). Benzene and aromatic compounds are present in gasoline vapours and tobacco smoke, although, benzene is absent from building materials and carpeting, which do contain the heaviest aromatic fraction (Ilgen et al., 2001). This can be observed in the ratios of aromatic compounds determined for the different microenvironments, where ratios *m*, *p*-xylene/benzene and toluene/benzene tend to be higher for indoor locations, when compared to outdoors. The ratio ethyl-benzene/benzene was much higher for those offices and bars where tobacco smoke was expected.

In broad terms, the commuting groups reported equivalent concentration levels. The higher individual values were associated with groups that included travelling by car. The concentration values for school children were, in general, comparable to the results obtained from the commuter categories. In Fig. 2, the differences in levels between the personal monitoring control group with the highest amount of time indoors, and the other categories reinforces the relative importance of indoor locations for exposure, in particular, to the heaviest aromatic compounds.

4. Discussion

Pérez Ballesta et al. (2006) described the broad relationship between population exposure and ambient air quality for the six cities in the PEOPLE project. Population exposure to benzene for commuting groups was approximately 1.5 times the city background and 0.6 times the maximum "hot spot" values. These comparisons were only possible due to the appropriate derivation of city background with a common approach on the given campaign days and between cities for measurements of the commuting populations. The city background for the campaign day is best represented by this spatial approach while the urban network background network air quality data is essential to place the campaign day within the annual distribution of the pollution levels of the city. While extrapolation into longer time frames is important, that is not necessarily the approach of this study. The one-day campaign focuses upon short-term sampling to allow a better sensitivity to the identification of exposure factors.

To understand human exposure to air pollution it is necessary to consider activities and locations. A 12-h sampling period was selected to include transport to and from work but exclude night time. Sampling of the latter would reduce the possibility of detecting differences due to daytime activities. As a first approach categorisation according to the mode of transport indicates the influence of behaviour on pollution exposure. The exposure concentration represents the average value of the concentration to which an individual has been exposed during the sampling period. This classification, however, does not include transport time in the evaluation of the results. A comparative analysis requires a more detailed consideration of the TMA diary. The first step is the creation of a matrix that includes activities and time durations for the sampling period (walk, car, bus, metro, home, work, bar, shop, restaurant, street, smoking and passive smoking). Table 1 shows the statistics of the TMA diary data collected in Madrid for the aforementioned variables.

4.1. Parameterisation of the regression model

The exposure concentration can be related to this matrix through the use of polynomial equations. Pollutant concentrations, whether measured at the same place or during the same activity, changes with time. Furthermore, the definition of a given activity encompasses different exposure conditions. A journey in a bus circulating within a city centre is generally shorter and through more polluted areas compared to a journey from the outskirts of the city. Exposure concentrations for each pollutant and time spent in the identified activities can be correlated by a multi-polynomial (second order)

Table 1
Median, 10th and 90th percentile of time spent by the sampled population in different locations or activities

Activity/location	No. of inputs[a]	Median time (min)	10th percentile (min)	90th percentile (min)
Walk	83	67.5	22.50	169.00
Car	50	62.5	21.75	122.00
Bus	30	30	15.00	60.00
Metro	30	47.5	30.00	83.25
Home	94	120	26.50	354.75
Work	87	415	256.50	540.00
Bar	65	30	15.00	74.00
Shop	32	52.5	15.00	126.75
Restaurant	26	60	30.00	97.50
Street	28	33.75	12.75	92.25
Smoking[b]	23	35	16.00	99.00
Passive smoking	55	45	15.00	324.00

[a]No. of inputs corresponds to the number of people who indicated a time spent in the corresponding actitity/location.

[b]Smoking time is calculated according to the number of cigarettes that the person smoked (5 min cigarette^{-1}). Passive smoking is considered a subjective variable as it is linked to the sensitivity of the individual in detecting the presence of tobacco smoke.

regression. The introduction of a logarithmic link function in the model prevents the estimation of negative values for exposure. The proposed fitting equation is expressed as follows:

$$EC_i = \exp\left[A_i + \sum_j (a_{i,j}t_j + b_{i,j}t_j^2)\right], \quad (1)$$

where EC_i is the exposure concentration of the compound "i"; t_j is the time spent in the activity "j", and A_i, $a_{i,j}$ and $b_{i,j}$ are the regression coefficients corresponding to the compound "i" and the activity "j". Eq. (1) can be considered as the product of two main factors: a baseline exposure concentration level, BS_i, represented by $\exp[A_i]$ and an overall activity factor, AT_i, resulting from the contribution of all the considered activities: $AT_i = \prod_j AT_{i,j}$, being $AT_{i,j} = \exp[a_{i,j}t_j + b_{i,j}t_j^2]$.

The greater and more appropriate the number of activities selected for the characterisation of the exposure, the closer the estimated values should be to the observations. In the hypothetical case where no activities were present, i.e. $t = 0$, the overall AT_i factor would be 1 and the baseline would represent the average exposure concentration. Therefore, these baseline exposure concentrations would be set to a large extent by the ambient background air quality while also being influenced by the general behaviour of the population, the environmental conditions and the number of activity factors, $AT_{i,j}$, considered in the regression model.

Furthermore, the exclusion in the model of activities that represent an important contribution to the exposure would imply a subsequent increase of the baseline concentration level and decrease of the overall activity factor, compensating the lack of information introduced in the regression. The opposite assumption is also valid, i.e. activities which reduce the overall exposure to the pollutant will decrease the baseline concentration level, when removed from the regression, and increase the overall activity factor. This can be clearly observed in Fig. 3, which represents the baseline exposure concentration level and the overall activity factor for the correlation between benzene exposure data and TMA.

In Fig. 3, the central position corresponds to the output of the correlation considering all the TMA variables, the AT values have been calculated for the corresponding 10th percentile of the corresponding activity time calculated from the sampled population (see Table 1). Variables are consecutively removed towards both directions of the midpoint in different orders to show that the relative increase or decrease of BS or AT after removing a specific activity is also influenced by the variables that have been previously excluded. Fig. 3 also includes the coefficient of correlation, R^2, for each regression. As expected, the coefficient of correlation improves with the number of correlated activities and with their importance for the exposure. The regression model is consequently considering the full set of activities previously indicated and in which Fig. 3 shows the highest coefficient of correlation.

As an experimental correlation the parameters fit the data within the range of experimental observations. Therefore, the result of the model cannot be extrapolated to time activities that are not similar to the actual observed environments. The results may be extended to a wider population on the sampling day, but beyond that, extrapolation is less appropriate. Fig. 4 represents the correlation between observed and predicted values with the corresponding coefficients of correlation (R^2) for all the considered pollutants. Points outside of the 95% interval of the prediction range of the regression line are also shown in Fig. 4. These have activities that are known to have the possibility of high

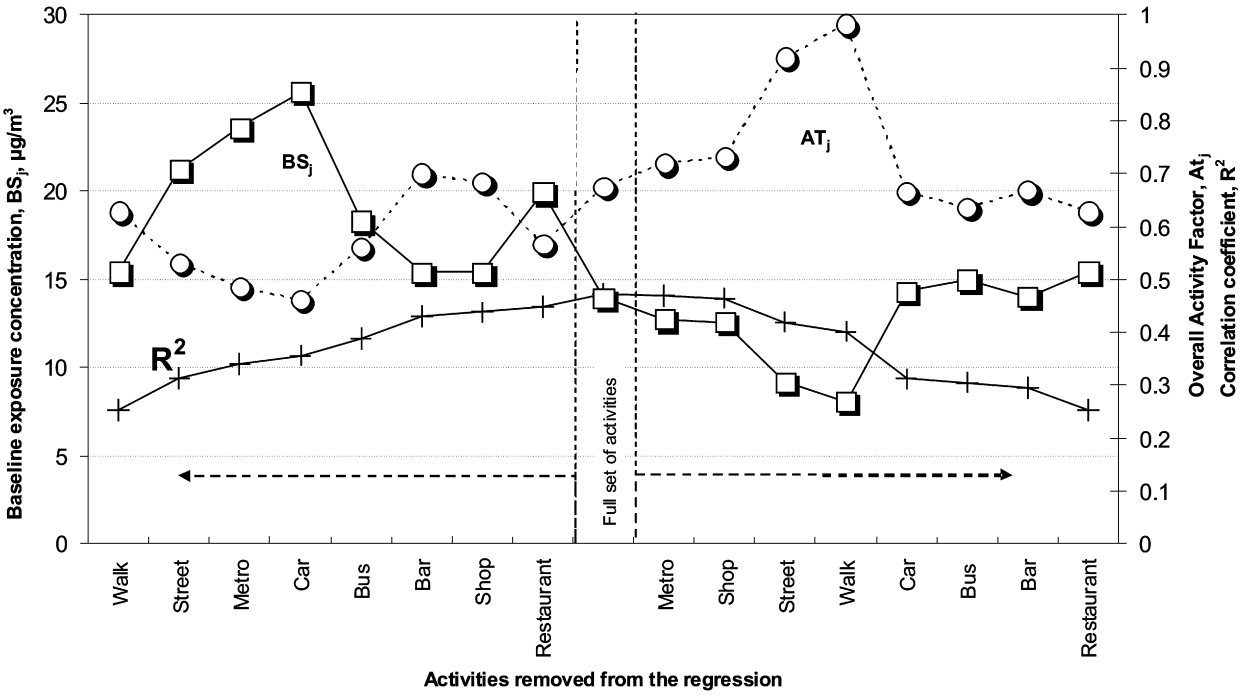

Fig. 3. BS, AT factor and correlation coefficient, R^2 of benzene, at the 10th percentile of the corresponding activity time, when removing successive variables (j) from the regression.

concentration peaks. Benzene and *m*,*p*-xylene have better correlation (i.e. higher R^2) compared to toluene or ethyl-benzene.

The compositional differences between sources affect the correlation coefficient, as not all the input variables of the model cover all compounds equally and exposure factors can differ between compounds for equivalent activities and locations.

For a better understanding of the different activities and their influence on the exposure concentration with time, an exposure gradient, $EG_{i,j}$, which would represent the gradient of the exposure concentration level with time for the corresponding compound (i) and described activity (j), is calculated. This can be defined as the partial derivative of the exposure concentration with respect to a specific time and activity:

$$EG_{i,j} = \frac{\partial EC_{i,j}}{\partial t_j} = EC_{i,j}(a_{i,j} + 2b_{i,j}t_j), \quad (2)$$

where $EC_{i,j}$ is the calculated exposure concentration for the compound "i" related to the activity "j".

Therefore, exposure concentration gradients are related to a time activity by a non-linear relationship. Gradients of exposure can become positive or negative, which should be interpreted as situations that increase or reduce the exposure concentration with time compared to the baseline.

Fig. 5 represents the exposure concentration gradient, $EG_{i,j}$, for the aromatic compounds calculated according to Eq. (2) at the 10th, 50th and 90th percentile of the time spent by the sampled population for each activity (see Table 1). Fig. 5 also shows at the secondary Y-axis the corresponding exposure concentration, $EC_{i,j}$, at the aforementioned percentile of time for each activity.

For outdoor activities classified as 'walk' and 'street', the exposure gradients differ due to the differing situations that describe the two categories. For toluene, ethyl-benzene and *m*,*p*-xylene 'street' starts with positive exposure gradients. For the other aromatic compounds, the exposure gradients become positive after a 1-h period. On the other hand, 'walk' had a negative exposure gradient for all the measured species and only with times close to 3 h does a positive exposure gradient appear. Only in the case of toluene and *m*,*p*-xylene for 'street' are the exposure concentrations significantly higher than those for the baseline. While both categories cover a wide range of situations, 'streets' are more associated with short-term exposure to traffic

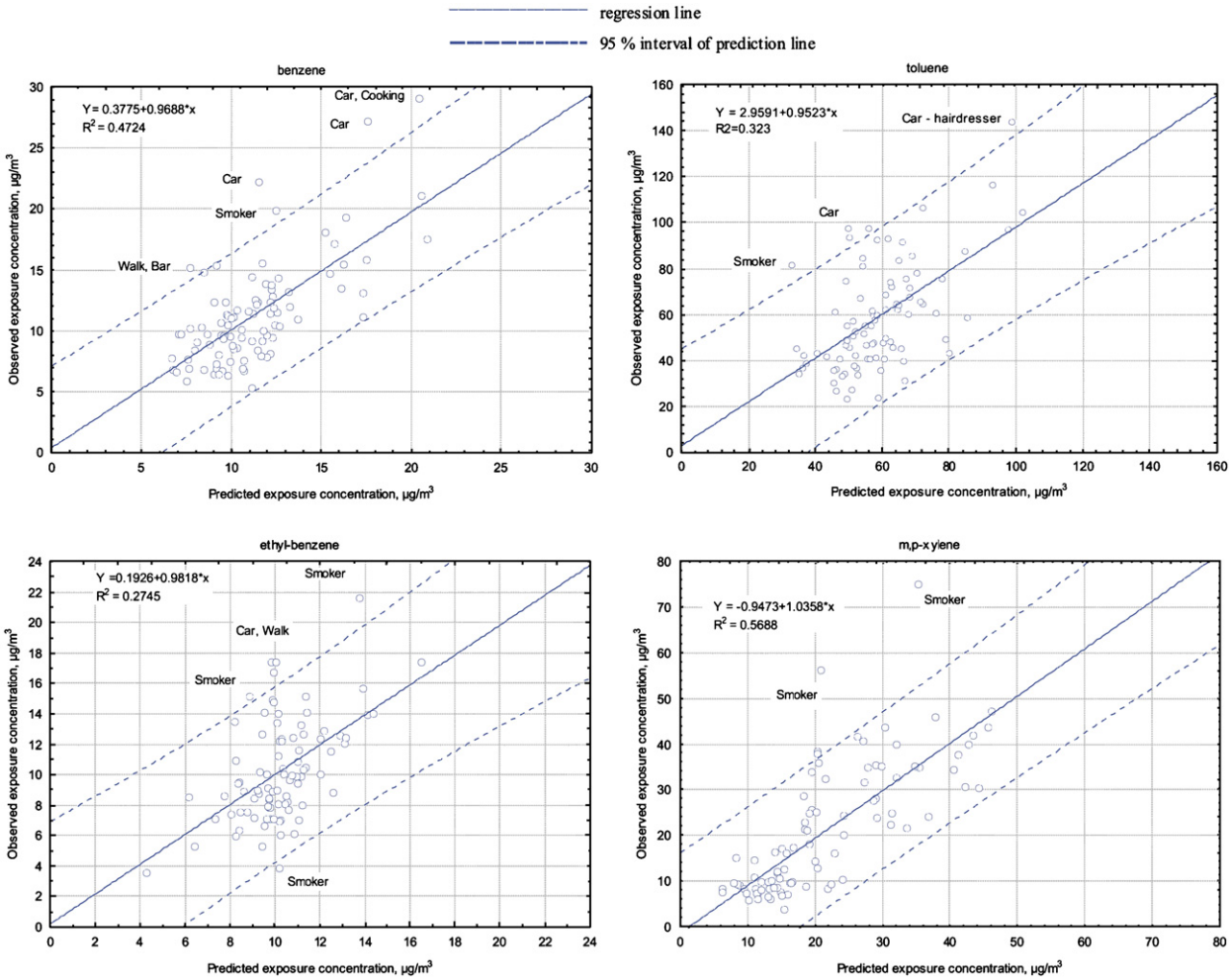

Fig. 4. Observed versus predicted exposure concentrations for the aromatic compounds.

emissions whereas 'walking' is more related to longer term exposure to background air.

Positive exposure gradients are related to the use of cars and buses for toluene and benzene. The exposure concentration remains higher than the baseline over a wide time range. For ethyl-benzene and *m*,*p*-xylene exposure factors become definitively positive for reported times >1 h, although the exposure concentration were below the baseline. Exposure concentrations in the metro for all aromatics are close to the baseline and negative exposure gradients are apparent after the transport has been used longer than the corresponding median time, with the exception of *m*,*p*-xylene, which starts to decrease after a longer reported time.

Activities linked to indoor locations: home, work, bars, shops and restaurants show strong positive exposure gradients for toluene, ethyl-benzene and *m*,*p*-xylene. These compounds are frequently present in paints, solvents and glues and are often associated with indoor pollution (Kotzias et al., 2005). The exposure concentration level of benzene for indoor locations is, however, around the baseline or lower. The elevation of toluene, ethyl-benzene and *m*,*p*-xylene exposure concentration above the baseline is most pronounced for work and shop microenvironments.

As expected, smoking is always characterised by a positive exposure gradient and represented by an exposure concentration higher than the corresponding baseline of each compound. An average number of 10 cigarettes was smoked per person. The results obtained for passive smoking are conditioned by the subjective response of people to smoke. For toluene and *m*,*p*-xylene a positive exposure gradient, as well as, exposure concentrations higher than the

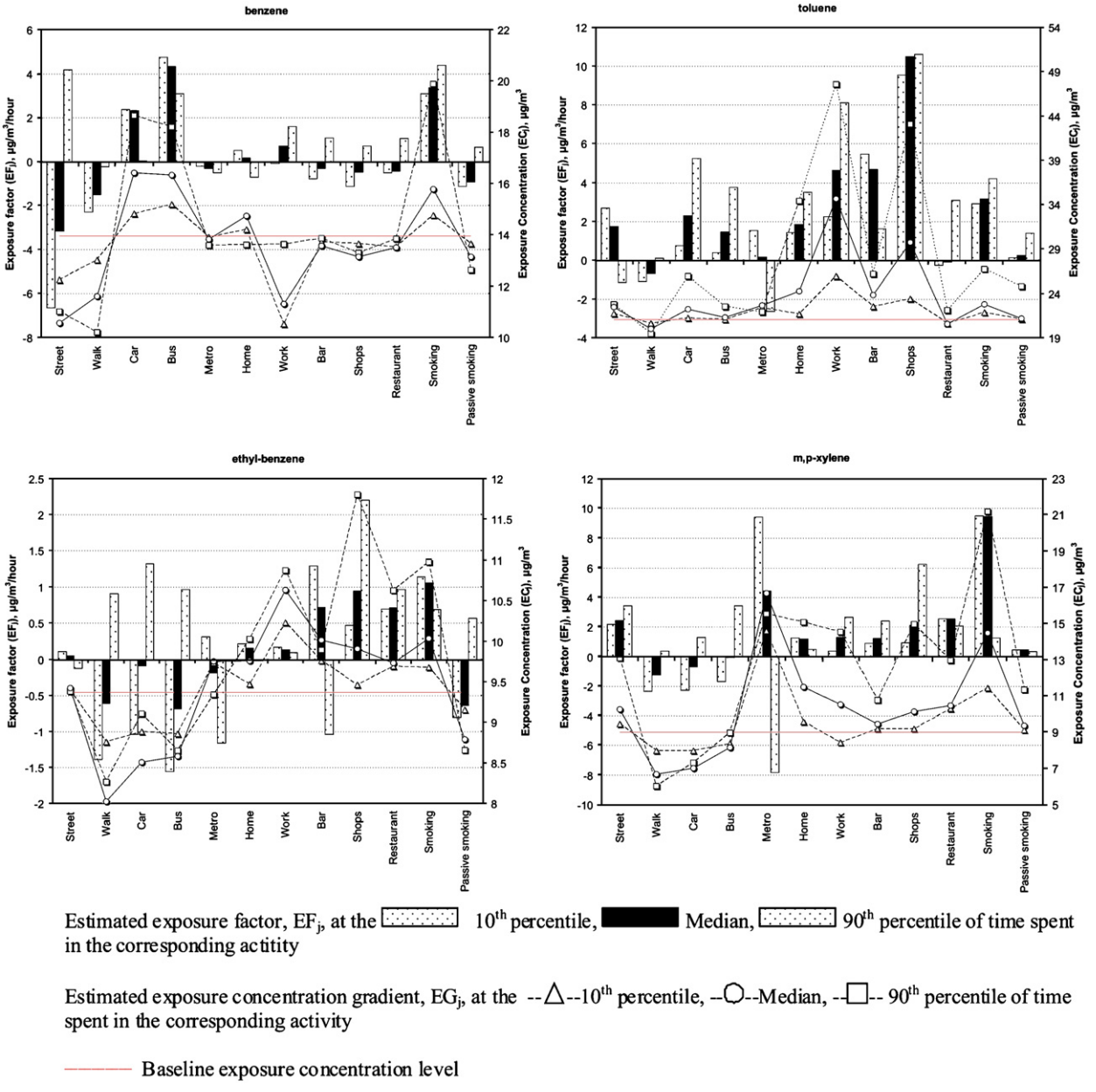

Fig. 5. Exposure gradients and exposure concentrations for aromatic compounds according to activity and environments.

respective baseline are evident. Nevertheless, benzene and ethyl-benzene have exposure concentrations slightly below the respective baselines although their corresponding exposure gradients became positive with longer reported times.

4.2. Uncertainty of the regression model

As this is not a predictive model but a correlation between exposure data and activity time, the uncertainty of the estimated exposure data are calculated as those derived from the uncertainty of the regression. Therefore, the variance that characterises the overall uncertainty of the estimated exposure concentration, $u_{EC_{i,j}}$, is the sum of the variance of the random uncertainty of the estimated exposure concentration, EC_i, and the variance associated with the bias between estimated and measured exposure concentration, MEC_i. The first term can be calculated according to the following

expression (Dietrich, 1991):

$$u_{\mathrm{EC}_i}^2 = \sum \left(\frac{\partial \mathrm{EC}_i}{\partial x_{i,j}}\right)^2 u_{x_{i,j}}^2, \quad (3)$$

where $x_{i,j}$ is representing the corresponding parameters described in Eq. (1): A_i, $a_{i,j}$, $b_{i,j}$ and t_j; $u_{x_{i,j}}$ being the corresponding associated uncertainties or standard deviations. Eq. (3) does not include the covariance between parameters, which, as a conservative approach, is considered negligible to the overall uncertainty contribution. Therefore, the uncertainty of the estimated exposure concentration can be derived:

$$u_{\mathrm{EC}_i} = \mathrm{EC}_i \sqrt{u_{A_i}^2 + \sum_j t_j^2 u_{a_{i,j}}^2 + \sum_j t_j^4 u_{b_{i,j}}^2 + \sum_j a_{i,j}^2 u_{t_j}^2 + 4\sum_j b_{i,j}^2 t_j^2 u_{t_j}^2}. \quad (4)$$

However, the bias contribution to the uncertainty is calculated as follows:

$$u_{\mathrm{bias}} = \frac{\mathrm{EC}_i - \mathrm{MEC}_i}{k}, \quad (5)$$

where k corresponds to the coverage factor for the calculation of the expanded uncertainty.

The input parameters for Eq. (4) estimated from the multi-polynomial correlation between experimental and measured exposure concentrations are given in Table 2. The overall uncertainty of the correlation model for a 95% confidence interval ($k = 2$) is determined by the following expression:

$$U_{\mathrm{model}_i} = k\sqrt{u_{\mathrm{EC}_i}^2 + u_{\mathrm{bias}}^2}. \quad (6)$$

Expanded uncertainties calculated according to Eq. (6) are represented as a percentage of exposed population in Fig. 6. According to these calculations about 70–80% of the investigated population can be correlated within 50% of expanded uncertainty for benzene and ethyl-benzene, whilst this percentage rises to 60% for *m*,*p*-xylene and toluene. For benzene, toluene and ethyl-benzene 90% of the population is correlated with a 70% of expanded uncertainty. Only *m*,*p*-xylene shows an asymptotic

Table 2
Estimated parameter and standard deviation associated with the multi-polynomial regression

	Benzene		Toluene		Ethyl-benzene		*m*,*p*-Xylene	
	Estimated parameter	Standard deviation	Estimated parameter	Standard deviation	Estimated parameter	Standard deviation	Estimated parameter	Standard deviation
A	2.636719	0.512917	3.05003	0.585086	2.237421	0.534202	2.198823	0.707068
a_{Walk}	−0.003311	0.001477	−0.00101	0.001873	−0.003320	0.002536	−0.005780	0.002451
b_{Walk}	0.000009	0.000007	0.00000	0.000008	0.000015	0.000015	0.000020	0.000012
a_{Car}	0.002829	0.001500	−0.00001	0.001808	−0.002899	0.002249	−0.006390	0.002483
b_{Car}	−0.000004	0.000007	0.00001	0.000007	0.000022	0.000016	0.000039	0.000010
a_{Bus}	0.006008	0.002629	−0.00052	0.003630	−0.004522	0.003428	−0.006535	0.004141
b_{Bus}	−0.000026	0.000025	0.00003	0.000033	0.000053	0.000030	0.000108	0.000034
a_{Metro}	−0.000024	0.003749	0.00289	0.004834	0.002016	0.003962	0.021499	0.005793
b_{Metro}	−0.000004	0.000048	−0.00003	0.000059	−0.000025	0.000049	−0.000180	0.000069
a_{Home}	0.000722	0.000925	0.00105	0.001102	0.000421	0.001014	0.002335	0.001505
b_{Home}	−0.000002	0.000002	0.00000	0.000002	−0.000001	0.000002	−0.000002	0.000003
a_{Work}	−0.002055	0.001178	0.00017	0.001270	0.000406	0.001346	−0.001299	0.001829
b_{Work}	0.000004	0.000001	0.00000	0.000001	−0.000000	0.000001	0.000004	0.000002
a_{Bar}	−0.001506	0.002727	0.00481	0.003344	0.003226	0.002962	0.001117	0.004233
b_{Bar}	0.000019	0.000020	−0.00003	0.000024	−0.000034	0.000028	0.000018	0.000028
a_{Shop}	−0.001682	0.002355	0.00716	0.002312	0.000533	0.002169	0.001035	0.002863
b_{Shop}	0.000010	0.000015	−0.00001	0.000016	0.000010	0.000015	0.000023	0.000019
$a_{\mathrm{restaurant}}$	−0.001461	0.001997	−0.00133	0.002314	0.001060	0.002012	0.004816	0.003068
$b_{\mathrm{restaurant}}$	0.000014	0.000008	0.00002	0.000010	0.000002	0.000008	−0.000011	0.000015
a_{street}	−0.011495	0.004386	0.00252	0.005358	0.000268	0.004734	0.003765	0.007508
b_{Street}	0.000097	0.000043	−0.00002	0.000058	−0.000003	0.000048	0.000003	0.000081
$a_{\mathrm{Smoking\ time}}$	0.003476	0.003139	0.00213	0.003954	0.002154	0.003934	0.016276	0.004637
$b_{\mathrm{Smoking\ time}}$	0.000001	0.000026	0.00000	0.000032	−0.000006	0.000036	−0.000077	0.000039
a_{passive}	−0.001492	0.001078	0.00006	0.001236	−0.001589	0.001084	0.000879	0.001541
b_{passive}	0.000004	0.000002	0.00000	0.000003	0.000004	0.000002	−0.000001	0.000003

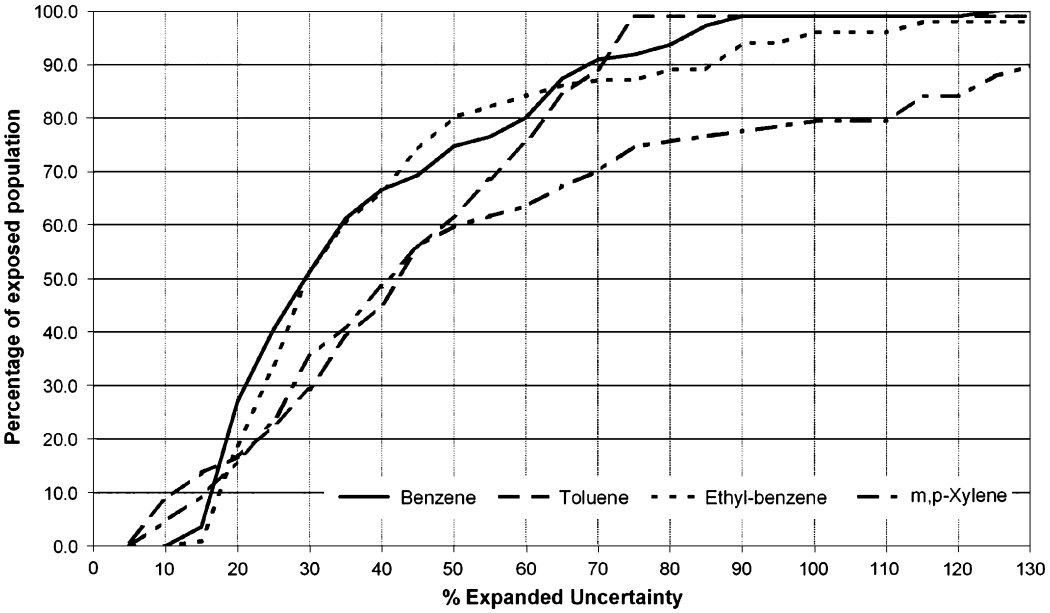

Fig. 6. Relative expanded uncertainty for the estimated exposure concentration versus percentage of exposed population.

behaviour with a 10% of residual population with expanded uncertainties over 130%. This percentage of population mainly corresponds to the smoker subgroup, where the regression seems to underestimate the exposure to this pollutant.

5. Conclusions

The described methodology enabled both a broad comparison between general categories of behaviour and environmental situations as well as a more specific assessment of factors that determine exposure concentration levels. This assessment was built upon the application of a regression model that used variables from a TMA diary. The relative importance of activities and locations that contribute to exposure concentrations was assessed. While movement through different environments is known to influence overall exposure concentration levels it is important to understand the relative impact.

The relationship of the baseline condition, set by the activities of the population considered, to the ambient background is important to understand the interaction of the model. In a hypothetical case where no activities are present, the baseline would be an average exposure concentration level equivalent to ambient background, which characterises the sampled population. While it is likely that the ambient background is close to the baseline condition it should be recognised that both represent mixed air that is a combination of emission sources.

The ability of the model to illustrate the influence of variables upon a baseline condition is influenced by the appropriate identification of the behaviour patterns of the population. The regression optimises the correlation parameters according to the number of identified activities. This increases or decreases the baseline level as an overall reflection from the activity factors. Although, the greater and more appropriate the number of activities selected for the characterisation of the exposure, the closer the estimated values should be to the observations.

Exposure concentration gradients have been identified as an optimum tool for the characterisation of the relative importance of the identified activities and to see how these activities affect the exposure with time.

In broad terms, the dynamics of the city set a background upon which personal behaviour is superimposed. The link between exposure of the population, outdoor concentration and pollution in microenvironments requires the use of activity profiles. These profiles are able to define locations and activities that have different influences upon exposure concentration. The application and interpretation of the regression model to the Madrid campaign allows the identification of the main exposure activity factors for each pollutant. Benzene, excluding tobacco smoke, is mainly associated with

transport activities (car and bus). For the heaviest pollutants, in particular ethyl-benzene and *m*, *p*-xylene, indoors activities (home, work, bar, shop and restaurant) are more important. By contrast, exposure to toluene seems to be affected by significant contributions from indoor and outdoor situations.

Finally, the environmental conditions of the sampling day and the number of activity factors considered in the regression model should influence the output. Nevertheless, in relative terms, the conclusions with respect to the identification and relevance of the exposure activities during a one-day campaign are correct as far as this approach identifies exposure sources associated with the characteristics of the city and population behaviour. Meteorological conditions during other days of the year could enhance or hide exposure factors related to outdoor or indoor activities by influencing the baseline exposure concentration level of the model, but this will not modify the emission sources that were identified during this snapshot campaign.

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