EXPOLIS-INDEX:

Work Package 2

Final Report

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VOC personal exposures in EXPOLIS - relationships to indoor, outdoor and workplace concentrations

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ABSTRACT

Evaluation of relationships between median residential indoor, indoor workplace and population exposures may obscure potential strategies for exposure reduction. Evaluation of participants with personal exposures above median levels in the EXPOLIS study in Athens, Helsinki, Oxford and Prague illustrated that these participants frequently showed a different relationship to indoor and workplace levels than that shown by the population median. Further, personal exposures at the upper end of the distribution may exceed the US EPA Rfc, illustrated here using hexane, naphthalene and benzene. Thus, prioritization of environments for control measures based on median exposures may exclude important areas where effectively focused control measures are possible, and may therefore have little impact on the highest and most harmful exposures. Strategies to reduce exposures to individual compounds, therefore, may benefit from focus on the high end of the distribution to identify activities and behaviors that result in elevated exposures. Control strategies targeting activities that lead to exposures in the upper end of the distribution would reduce the variability associated with population median values by bringing the upper end of the exposure distribution closer to median values. Thus, compliance with health-based standards would be more protective of the higher exposed fraction of the population, in whom health effects would be more expected.

1. INTRODUCTION

Current distributions of exposures to individual VOC and corresponding concentrations measured in different microenvironments have frequently been described as log normally distributed or more highly skewed (Brown et al 1994). A fundamental question, therefore, is whether there are effective control methods that may be employed by reducing exposures in the upper end of the distribution and reducing the variability associated with population median or mean exposures. Directing control measures across the population are likely to be difficult to implement, requiring significant effort and regulation in reducing emissions. Reduction of the upper end of exposures, thus narrowing the distribution would allow health based standards to be more protective of a higher exposed fraction of the population, while also reducing mean and median values.

Control measures directed at the upper end of the exposure distribution relies on characterizing sources and activities that lead to greater exposures, and the microenvironments in which the exposures occur. Frequently median values are used to evaluate relationships between microenvironment concentrations and personal exposures. While such an approach may identify more general relationships for the majority of the population, these may not be indicative of the relationships in the upper end of the exposure distribution. Prioritization of environments based on median exposures may therefore exclude important areas where effectively focused control measures are possible, and may therefore have little impact on the highest and most harmful exposures. This is especially relevant if multiple sources are present in different environments, and exposures represent the combined contributions from multiple environments.

Many indoor sources have been largely ignored in regulation, as sources are individually relatively small. They may contribute disproportionately to personal exposures, however, as they are emitted in close proximity to where people spend significant portions of their time. A greater fraction of the pollutant emitted may come into contact with an individual relative to the mass emitted into the environment. This concept is encompassed by the term “Intake fraction” (Bennett et al 2002). This concept is especially relevant to VOC exposures as emissions from multiple indoor sources may result in indoor levels that are often higher than outdoor levels (Edwards et al 2001a). Thus careful attention to the emissions profile of items and products for residential indoor use, which have traditionally not been considered as environments where pollution should be controlled, may achieve significant reduction in exposures and control measures may be more cost effective.

A further potential area for control measures occurs through activities or locations in a small number of individuals that do not appear as major sources on a population basis. They may, however, be significant sources of exposure for those involved. For example home workshops have been associated with elevated levels of benzene (Edwards and Jantunen, 2001). Others may be related to specific hobbies, or product uses. Greater controls over the content of products that are used in these locations could therefore significantly reduce exposures in these individuals. Such reductions may be related to individual compounds that are only detected in a few individuals and are thus usually excluded from statistical analysis, or they may occur as superimposition of exposure on sources prevalent in the majority of the population (e.g. exposure to ETS or automobile exhaust), and may be easily overlooked in relation to the more prevalent sources. It is important, therefore, that both the common population sources and specific sources affecting small sub-populations are evaluated in the context of control measures

The EXPOLIS project is well placed in being able to evaluate some of these effects within adult populations of 4 European cities Athens, Helsinki, Oxford and Prague. In the EXPOLIS centers personal exposures were measured with concurrent measures of both inside and outside the home environment and inside the work environment during the time that the participant reported they would spend in the residential or workplace environment. Thus the contribution of VOC concentrations in each microenvironment to the personal exposure concentration could be assessed in each location. This enables comparison of personal exposures and microenvironment concentrations, and the implications for control measures across the distribution.

The purpose of the current paper is to examine if different implications about the patterns of personal exposure and areas for control strategies may be derived from looking at relationships between microenvironmental concentrations and personal exposures in the upper end of the distribution rather than median levels across populations. Since the distributions are highly skewed to the right (upper end), we attempt to identify activities and microenvironments associated with elevated exposures that do not become apparent in investigating population median levels. We focus in particular on naphthalene, hexane, benzene, butoxyethanol and cyclohexane.

2. METHODS

The different centers

The EXPOLIS study focused on air pollution exposures for active working age adults between 25-55 years old (Jantunen et al., 1998; Jantunen et al., 1999). In the current manuscript we include EXPOLIS populations from Athens, Helsinki, Oxford and Prague. Although other centers were included in the EXPOLIS project they have not been included in current analyses either because populations were restricted to specific groups e.g Milan was selected from 15-55 year old office workers and Grenoble was selected from half asthmatics and half controls, or because Basel used a different sampling and analysis procedure for VOCs (Jurvelin et al 2001). For the four centers included in the current analysis there were slightly different population sample selection procedures used, and the effect of sample selection bias has been comprehensively discussed in Rotko et al (2000). Briefly, in Helsinki the initial questionnaire was followed by a reminder mailing and subsequently by a computer assisted telephone interview to achieve a response rate of 74%. In Athens, a private opinion polling company was used to find 2000 individuals and visit the homes to administer the baseline questionnaire. Only non-smokers were selected for further contact and 1 in 8-10 agreed to participate. In Prague the population was selected from District V in the city center. Response rates to the initial mailing were very low and cannot be considered representative of the population of the city. The exposure group consisted of those who responded to the initial mailing and were willing to participate when contacted by telephone. In Prague the age distribution was skewed toward the younger ages and the more educated. The distribution of workplaces (within the following categories: one building, outdoors, multiple daily locations, home or not working) was similar to the other centers, however, home locations were more uniform (only downtown) as it was a specific district in the city.

Sampling and Analysis

Participants carried an aluminum briefcase for 48 hours to estimate VOC personal exposures (Jurvelin et al 2001). Residential indoor, residential outdoor and indoor workplace microenvironments were sampled during periods the participant reported they would be in that microenvironment during the sampling period. In addition to the active measurements, participants were asked to respond to questionnaires and time activity diaries.

VOC samples were adsorbed onto Perkin Elmer Tenax TA adsorbent tubes (chrompack, Middleburg, Netherlands). Identical standard operating procedures and sampling equipment were used in each EXPOLIS center in the current analysis. All samples were analyzed at VTT Chemical Technology (Espoo, Finland). Calibration and the results of comprehensive quality assurance tests including comparisons of personal and microenvironment sampling techniques, duplicates, blanks and performance evaluations are presented in Jurvelin et al (2001). VOCs were desorbed from tubes with helium at 50mlmin-1 at 260°C into a cold trap. Subsequently flash desorption was followed by a 1:1 split into two non-polar capillary columns (PONA, length 50m, internal diameter, 0.2mm, phase thickness 0.5µm) of a Hewlett Packard 5890 series II+ gas chromatograph (GC) (Hewlett Packard GmbH, Waldbronn, Germany) with flame ionization (FID) and mass selective detection (MSD - Hewlett-Packard MSD 5972). VOCs were identified from MSD total ion chromatogram by a Wiley 275 software library. Peaks on FID chromatograms were identified on the basis of retention times of standard reference materials (high purity). Detection limits for Tenax TA were 1-5 $\mu g/m^3$ depending on the compound with a mean of 2 $\mu g/m^3$. Tenax TA showed no significant contamination apart from benzaldehyde a known artifact with this absorbent.

Target compounds

A total of 323 different volatile organic compounds were identified in EXPOLIS samples, however calibration and quality assurance and control (QA/QC) measures, reported in Jurvelin *et al* (2001), were directed at a group of 30 target volatile organic compounds on the basis of frequency of detection in previous VOC studies (Brown et al., 1994), quantification using current methods, environmental and health significance of some of the compounds and utility of one or group of few compounds as markers of pollution sources (Jantunen *et al* 1998). 11 of the 30 target compounds were HAPs incorporated into the U.S. Clean Air Act Amendments (CAA 1991). Benzene and styrene are IARC carcinogens (class I and class IIB respectively). In addition, although health effects have been observed only at levels far in excess of those expected in indoor environments, 2-butoxyethanol and d-limonene are skin contact allergens (class III and IIB respectively - NKB 1994) and styrene, 2-ethylhexanol, phenol, 2-butoxyethanol, hexanal and benzaldehyde are mucous membrane irritants.

Statistical treatment of data

SPSS for windows version 10.0 was used for all analyses. The sensitivity of the analytical method varied for each compound measured and the number of samples in which each compound was measured reflects the sensitivity of the analytical method in addition to the prevalence of each compound. Treatment of non-detects, therefore, was handled on an individual compound basis and limits of detection (LOD) were computed for each individual compound (Jurvelin *et al* 2001). Half of the respective LOD for each compound (Hornung and Reed, 1990) was used in analyses for samples in which the compound was not detected.

Smoking

Personal exposure to environmental tobacco smoke (ETS) was defined as those participants who reported coming into contact with tobacco smoke in any microenvironment during the 48-hour sampling period. This included active smokers, those with partners that smoke, those who work with people that smoke inside the workplace and those that briefly shared the same microenvironment as a smoker during the sampling period. Edwards et al (2001b) demonstrated that many of the compounds associated with emissions from motor vehicles, other combustion sources and some indoor products are also associated with tobacco smoke. For those exposed to tobacco smoke, the most dominant source of exposure to these compounds, however, is tobacco smoke.

3. RESULTS AND DISCUSSION

The following sections illustrate that relationships between median microenvironment concentrations and median personal exposures may not be the same as similar relationships in the upper end of the exposure distribution. Our focus on the high end of the distribution reveals relevant exposure patterns not detected in the total population sample. Thus, prioritization of environments for control measures based on median exposures may exclude important areas where effectively focused control measures are possible, and may therefore have little impact on the highest and most harmful exposures.

Napthalene: Simple measures may be available to reduce exposures of naphthalene in Athens, where US EPA Rfc (Inhalation reference concentration) of 3 μg/m^3 were exceeded in every personal exposure concentration and mean and median concentrations were 54.0 μg/m^3 and 22.6 μg/m^3, respectively. In Prague naphthalene personal exposures exceeded the Rfc in 29% of cases, but both mean and median concentrations (2.4 μg/m^3 and 1.8 μg/m^3 respectively) were below the Rfc. In other EXPOLIS centers naphthalene concentrations were lower and in Helsinki and Oxford, naphthalene was detected in less than 10% of personal exposures. If median values in Athens and Prague are analyzed (Figure 1) median personal exposure concentrations in Prague were higher than indoor levels, which in turn were higher than workplace levels. In Athens, however, median indoor levels were considerably higher than median personal levels, which in turn were higher than workplace levels. Clearly on this basis, it would appear that a different control strategy would be merited in Athens compared to Prague. Analysis of individual cases in Athens that reported indoor and personal exposure concentrations, and those in Prague where the personal exposure concentration was above the limit of detection (Figure 1) revealed that the median approach would be limited, however, and that there are further strategies for reduction that would not be apparent. In Athens median concentrations were driven by 5 participants whose personal exposures were considerably higher than the rest of the population and are shown in Figure 1A. Personal exposure concentrations for these five individuals ranged from 74 to 469 μg/m^3. Indoor concentrations were even higher and ranged from 114 to 989 μg/m^3, respectively. If calculation of risks to human health are based on median values, as used in the US EPA 1996 national scale air toxics assessment, elimination of these elevated values would result in reduction of the median indoor concentrations from 25.6 to 21.8 μg/m^3 and reduction of median personal exposures from 23.1 to 21 μg/m^3. Clearly from a control strategy standpoint, reduction of these elevated indoor concentrations that lead to the highest personal exposure concentrations would be desirable. What is not apparent, however, is that the majority of the personal exposures above the median personal exposure level are driven by naphthalene exposures during personal activities or non-measured microenvironments, rather than indoor levels as suggested by the median values (Figure 1B). This is somewhat similar to the situation in Prague (Figure 1C), where a couple of elevated indoor concentrations drive the top end of the distribution, and subsequently personal activities or non-measured microenvironments drive personal exposures. This similarity in personal exposure patterns between centers, therefore, would suggest that similar reduction strategies might be employed between centers, which would not be suggested by evaluation of median concentrations.

The principal end use for naphthalene is as an intermediate in the production of phthalate plasticizers, resins, phthaleins, dyes, pharmaceuticals, insect repellents, and other materials. It is also used in the production of the insecticide carbaryl used in home yards and gardens, and in paints, dyes and resins. Crystalline naphthalene is also used as a moth repellent and as a solid block deodorizer for toilets (ATSDR 1995). Wood smoke, fuel oil and gasoline also contains naphthalene. For Athens, multiple linear regression was used to identify predictors for personal exposures to naphthalene using questionnaire information as independent variables. The 5 elevated values that were dominated by high indoor values were omitted from the analysis in order to explore predictors for naphthalene exposure driven by personal activities and non-measured environments. Table 1 shows multiple linear regression was able to account for 44% of the variance between personal exposures. The strongest predictor identified with the highest standardized coefficient (Beta) was *time actively smoking*. The second highest standardized coefficient was for presence of *attached garage*. Although this may be related to either storage of moth repellents or fuel oil and gasoline, which contain naphthalene, there were insufficient attached garages in this sample to draw conclusions. The third predictor identified was *home location in the downtown area*, which appears to identify emissions from automobiles as a source. The final predictor was *time using a gas stove*. Naphthalene has been identified as being emitted from stationary natural gas fired turbines. We were not able to locate emission factors for naphthalene from gas stoves in the literature, however, and this merits further evaluation.

2-Butoxyethanol: Although median personal exposure concentrations of 2-butoxyethanol were below the US EPA Rfc of 20 $\mu g/m^3$ in each EXPOLIS center, there were a few participants that exceeded the Rfc in each center. Median concentrations in each center were 8 $\mu g/m^3$ in Athens, 1.6 $\mu g/m^3$ in Helsinki, 1.6 $\mu g/m^3$ in Oxford and 2.2 $\mu g/m^3$ in Prague. Considering that 2-butoxyethanol was detected in 14%, 22% and 20% of personal exposure samples in Helsinki, Oxford and Prague respectively, median values have little meaning as they represent the limits of detection. In Helsinki, however, there was one participant with a personal exposure concentration of 943 $\mu g/m^3$, corresponding to an elevated workplace concentration of 2422 $\mu g/m^3$. This participant also showed elevated levels of m,p-xylene (1933 $\mu g/m^3$) , o-xylene (858 $\mu g/m^3$) and ethylbenzene (631 $\mu g/m^3$) corresponding to similarly elevated levels in the workplace (1390, 2779 and 1384 $\mu g/m^3$ respectively). In Athens 2-butoxyethanol was detected in greater than 50% of the personal exposure samples (67%), and a greater number of participants had personal exposure concentrations that exceeded the Rfc. The participants with personal exposure concentrations above the Rfc differed from the participant in Helsinki in that correspondingly elevated 2-butoxyethanol concentrations were in the indoor environment rather than the workplace, and they were not associated with elevated ethylbenzene or xylenes. Concentrations in Athens therefore, likely represent the contribution of household cleaners, although it is also present in liquid soaps, cosmetics and dry-cleaning compounds. In Helsinki although wall painting or renovation had occurred in the workplace of this participant during the last year it is not known how soon before the sampling period. 2-butoxyethanol is present as a solvent in spray lacquers, enamels, varnishes, and latex paints and as an ingredient in paint thinners, paint strippers, and varnish removers (ATSDR 1999).

Hexane: A similar pattern may be observed for Hexane exposures. While personal exposures to cyclohexane were below health-based guidelines, however, personal exposures of some individuals to hexane exceeded US EPA Rfc (Inhalation reference concentration) of 0.2 mg/m^3. In particular, maximum personal exposure concentrations were 0.83 mg/m^3 in Oxford, 0.67 mg/m^3 in Athens, 0.18 mg/m^3 in Prague and 0.12 mg/m^3 in Helsinki. Detection of hexane in personal exposure samples, however, was 44% in Athens, 39% in Oxford, 27% in Prague and 13% in Helsinki. Median exposure levels for the Oxford population indicated workplace concentrations exceeded indoor concentrations, which were higher than personal exposures. Indeed, highest concentrations measured in each EXPOLIS center were in the workplace microenvironment and the 4 highest workplace concentrations in Oxford were 2.9, 1.8, 1.2 and 0.4 mg/m^3, considerably above the Rfc of 0.2 mg/m^3. When those individuals with personal exposure above the median concentration were examined, however, it was clear that there were other areas where exposure reduction strategies could be implemented. Figure 2 shows participants in Oxford with personal exposure concentrations above the median concentration. In several cases personal exposure concentrations were equal to or above workplace concentrations, while indoor concentrations remained low, indicating that personal behaviors and activities were significant contributors to exposure. In 3 cases indoor concentrations were significantly elevated, and in a further 3 cases elevated workplace concentrations were associated with elevated personal exposures that were considerably higher than indoor levels. Hexane was found in a wide range of household product classes from 37% of adhesive related products, 26 % of oils greases and lubricants, 13% of automobile products, 5% of paint related products, 3% of household cleaners/polishes and other less detected classes of household products (Sack et al., 1992).

Benzene: The US Rfc of 30 μg/m^3 was exceeded quite dramatically by personal exposure of one participant in Athens to 217 μg/m^3. This participant reported they spent 567 minutes in the car during the measurement period, indicating that they were employed as a driver. This participant also reported high exposures to other traffic related VOC compounds hexane, m,p and o Xylenes and trimethylbenzene. Interestingly this participant also showed similarly high exposure to d-limonene, possibly from air fresheners used in the car. Lawrk et al (1995) reported elevated exposures to volatile organic compounds in passenger compartments of automobiles during commutes in New Jersey. While the US and much of the EU have mandated reduced benzene content in gasoline, those that spent long periods in the car occupationally are likely to be exposed to elevated levels of traffic related VOC compounds. For those areas, especially in the developing world, that have not mandated lower benzene levels in fuel, benzene exposures for occupationally exposed drivers likely represent a considerable public health concern. Other personal exposures to benzene that exceeded the Rfc were for active smokers in Helsinki (1) and Athens (2). In Prague the Rfc was exceeded by two participants that spent 300 minutes in a car and 225 minutes in a car and bus, respectively, and also spent 45 minutes and 245 minutes in a home workshop.

Figure 3 shows personal exposures to benzene in Helsinki above the median for participants not exposed to ETS. Median population levels indicated that personal exposures were above workplace levels, which in turn were significantly above indoor levels. Clearly, however, in the upper end of benzene personal exposures there are a number of participants with indoor benzene concentrations that are more elevated than in the rest of the population.

Cyclohexane: In Helsinki the 90 percentile workplace cyclohexane concentration level was 2.93 ug/m^3 with detection in 4% of workplace environments, yet the maximum level was 1512 ug/m^3 leading to the corresponding personal exposure level of 1484 ug/m^3 in that individual. While these values are lower than health based guidelines for cyclohexane, they illustrate that significant exposures may occur to specific individuals. Clearly programs to reduce exposure would be directed at these individuals and activities, as cyclohexane was only detected in 8% of personal exposures, rather than more general control measures within the population of Helsinki. There were clear differences between EXPOLIS centers, however, and cyclohexane was detected in a much greater fraction of microenvironments and personal exposure samples in Prague, with 67% detection in personal exposure samples followed by 33% in Athens and 17% in Oxford. The distribution of exposures was still highly skewed, however and non-ETS exposed arithmetic mean personal exposure concentrations in Prague were 27.6 ug/m^3 compared to median values of 6.3 ug/m3. Still the median was significantly above levels measured in different centers (Athens = 1.9ug/m3, oxford =1.4 ug/m3, Helsinki = 1.4 ug/m3) in both Kruskal Wallis and Median Tests. If control measures were directed at the two individuals with personal exposures above 50 ug/m3, however, the arithmetic mean concentration would be reduced to 8.9 and median concentration to 4.7 ug/m3 in Prague.

Median values for cyclohexane in Prague indicate that personal exposures are similar to indoor levels, which are both significantly higher than workplace levels. This would suggest that efforts to reduce exposures should be focused on reducing indoor levels, and that focusing on personal exposures and workplace concentrations would not be merited. If individual cases that are above the median concentration are analyzed, however, a different picture emerges which would lead to alternative exposure reduction strategies. Figure 4 shows residential indoor, workplace and personal exposure concentrations of cyclohexane for participants in Prague with personal exposure above median values, and the median for the center. Participants with missing values for one microenvironment were excluded. The personal exposures above the median value illustrated in figure 4 A, B and C, indicate that there are several distinct patterns among exposures as might be expected. In figure 4A personal exposures and other microenvironments dominate and personal exposures are more elevated than both workplace and residential indoor levels. In figure 4B workplace concentrations are considerably above indoor and personal exposure levels. Figure 4C shows participants with personal exposure concentrations that are similar to indoor levels with workplace concentrations considerably below the indoor levels. Clearly in each of these three types A, B and C, a different exposure control strategy would be recommended. Since these are exposures that are above median levels for the center, these are individuals where exposure reduction strategies would be most warranted, especially if sample populations are selected from susceptible individuals.

4. CONCLUSIONS

The examples above illustrate that often the relationship between median indoor, median outdoor and median personal exposure may not reflect the patterns observed in the upper end of the personal exposure distribution. Thus, prioritization of environments for control measures based on median exposures may exclude important areas where effectively focused control measures are possible, and may therefore have little impact on the highest and most harmful exposures. Control strategies targeting activities that lead to exposures in the upper end of the distribution would reduce the variability associated with population median values by bringing the upper end of the exposure distribution closer to median values. Thus, compliance with health-based standards would be more protective of the higher exposed fraction of the population, in whom health effects would be more expected, while also reducing mean and median values.

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7. TABLES

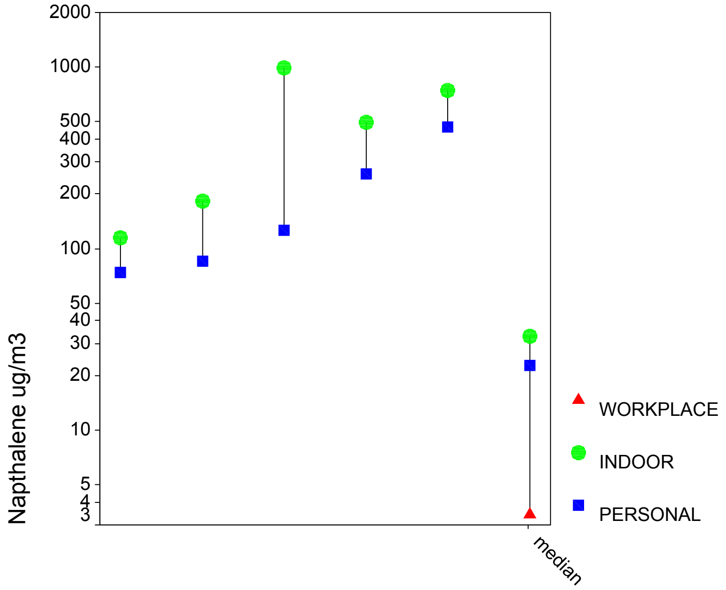
Table 1. Predictors for naphthalene in Athens

Dependent variable	***n***	***Adjusted r^2***	***Predictor variables***	***Unstandardized Coefficients***		***Standardized coefficients***	***Colinearity diagnostic***	
				B	*Std. Error*	*Beta*	*Condition Index*	*VIF*
naphthalene	36	0.44	(Constant)	18.2	2.2		1.00	
			Time actively smoking	0.10	0.03	0.48	1.26	1.013
			garage	37.5	11.7	0.40	1.30	1.009
			home location downtown	12.8	5.3	0.31	1.53	1.061
			Time using gas stove	0.07	0.04	0.25	1.82	1.053

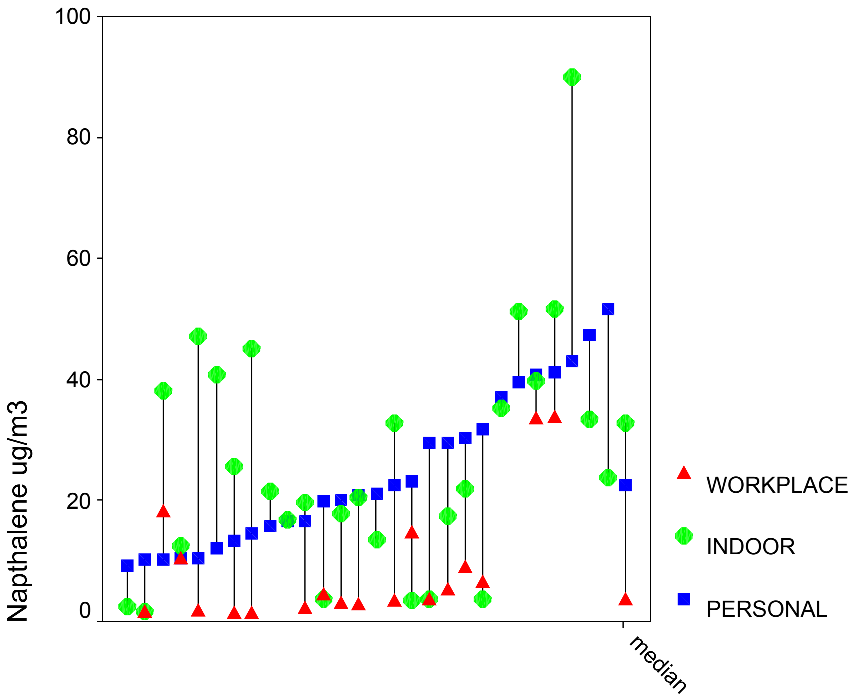
8. FIGURES

Figure 1. Residential indoor, workplace and personal exposure concentrations of Naphthalene for participants in Athens and Prague

A) Personal exposures to 5 elevated indoor Naphthalene concentrations in Athens



B) Residential indoor, workplace and personal exposure concentrations of Naphthalene in Athens – omitting 5 elevated values in Figure 1A



C) Residential indoor, workplace and personal exposure concentrations of Naphthalene for participants in Prague with personal exposure above median values.

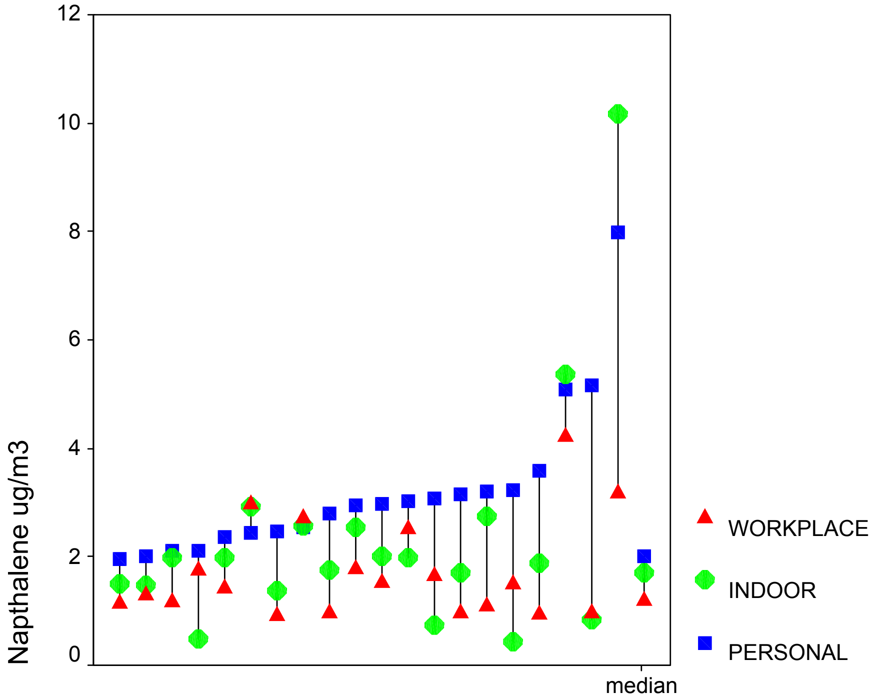


Figure 2. Residential indoor, workplace and personal exposure concentrations of Hexane for participants in Oxford with personal exposure above median values.

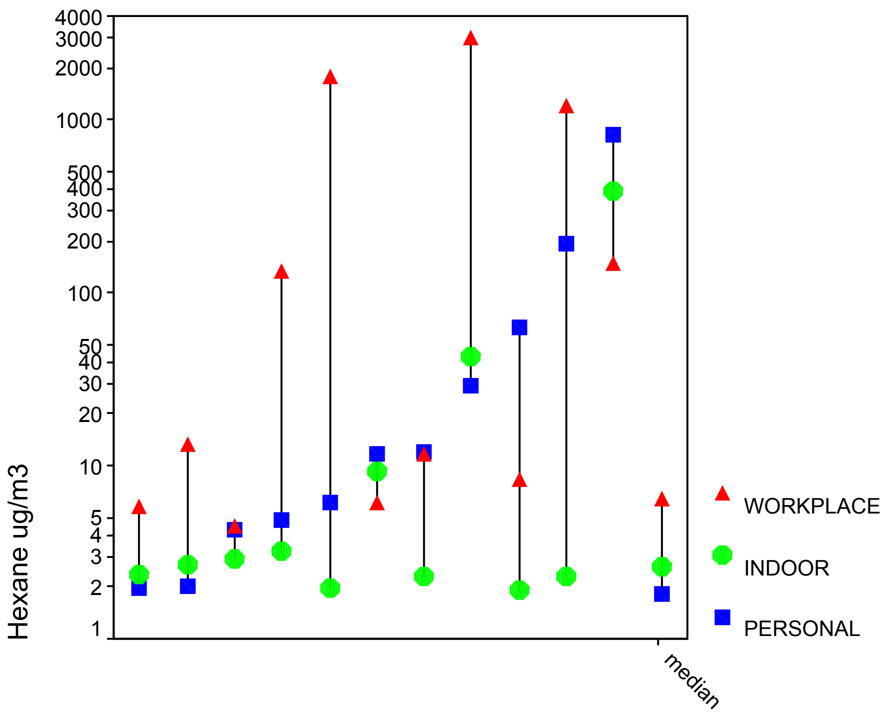


Figure 3. Residential indoor, workplace and personal exposure concentrations of Benzene in Helsinki for participants not exposed to ETS with personal exposure above median values.

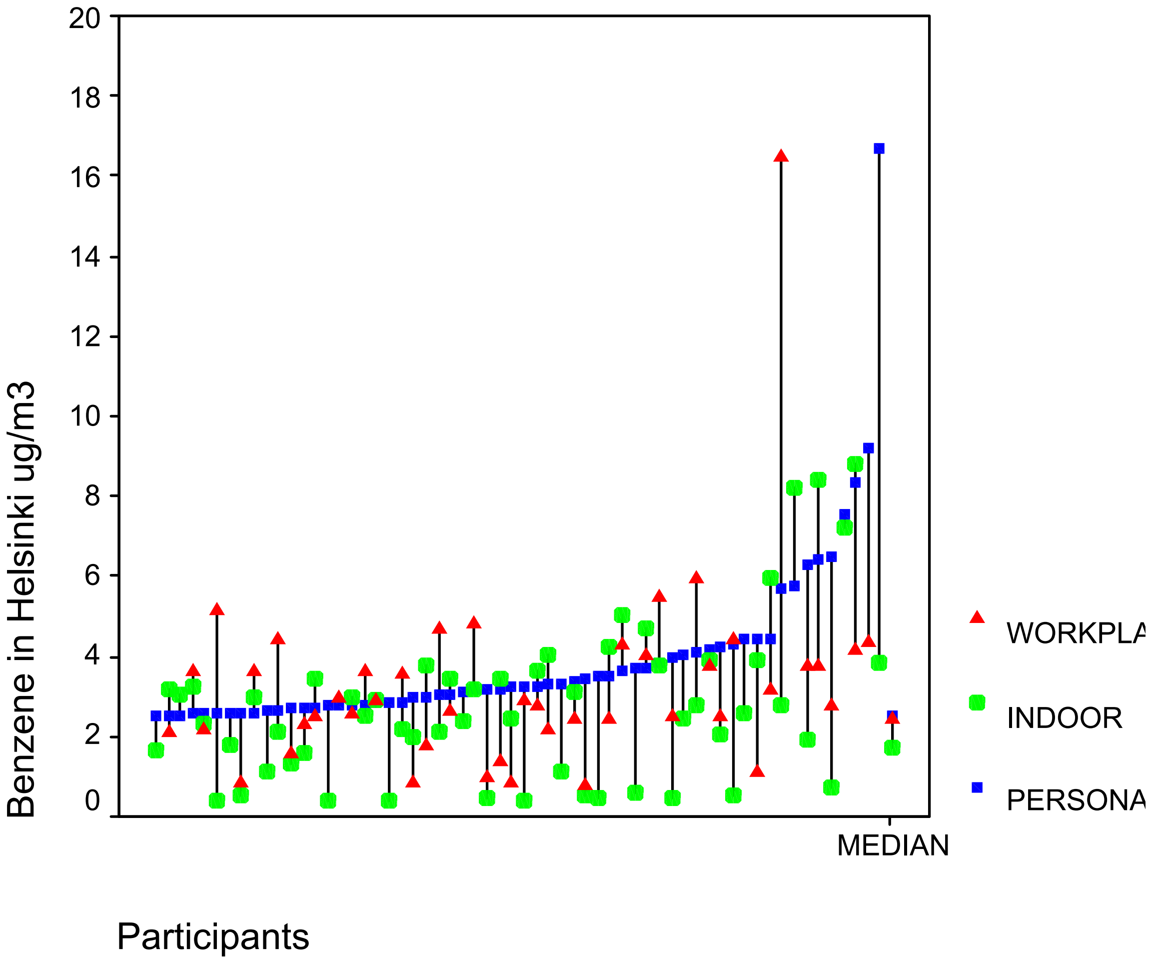
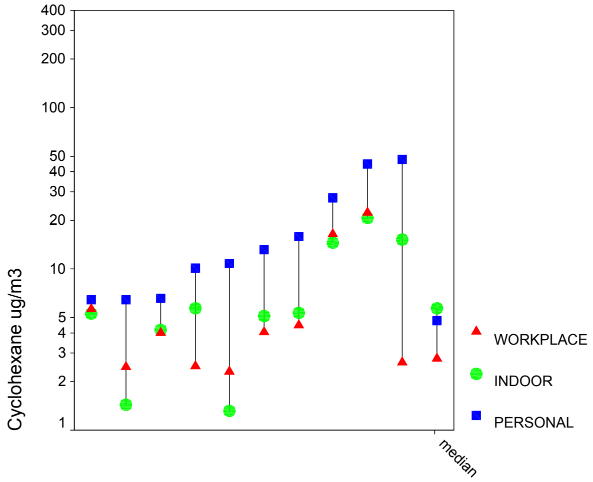
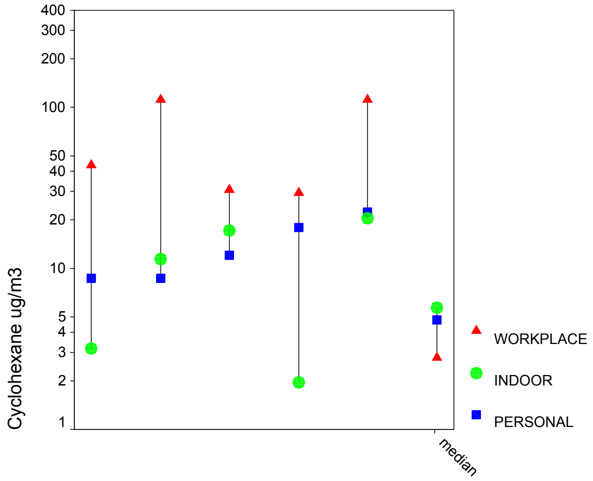


Figure 4. Residential indoor, workplace and personal exposure concentrations of cyclohexane for participants in Prague with personal exposure above median values.

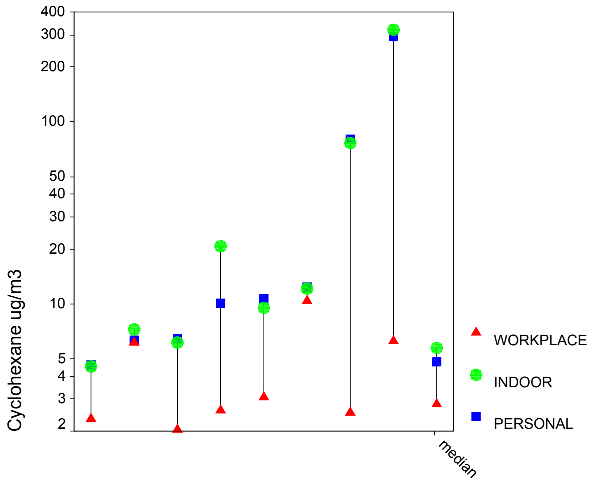
A) Personal activities and other microenvironments - personal exposures are more elevated than both workplace and residential indoor levels



B) Elevated workplace in comparison to personal exposures and indoor levels



C) Indoor concentrations dominate



VOC personal exposures in EXPOLIS populations in Athens, Helsinki, Oxford and Prague

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Introduction

The EXPOLIS study was a European representative population based study of adult air pollution exposures, where personal exposure and workplace, indoor residential and outdoor residential environments were measured for participating adults. Objectives of the VOC component of the study were to determine background exposures to 30 VOCs selected for their relevance to health or as markers of pollution sources. Due to the highly skewed nature of these distributions, in the current paper we wish to expand on this approach to identify activities and sub-populations with more elevated exposures and examine differences in personal exposures of EXPOLIS populations in Athens, Helsinki, Oxford and Prague in relation to questionnaire information and 48-hour time activity diaries.

Sampling design

- Participants carried an aluminum briefcase, containing VOC sampling apparatus and other sampling equipment, at all times during the 48-hour sampling period
- VOCs were actively sampled using a modified Buck IH Pump (A.P.Buck Inc. Orlando, Florida) and absorbed onto Perkin Elmer Tenax TA absorbent tubes
- Analysis was performed by VTT (Espoo, Finland) using a Hewlett-Packard 5890 Series II gas chromatograph with flame ionization (FID) and mass selective detection (Hewlett-Packard MSD 5972). VOCs were identified from MSD total ion chromatogram by a Wiley 275 software library. Peaks on FID chromatograms were identified on the basis of retention times of standard reference materials (high purity).
- Further details of the VOC sampling and analysis including comparisons of PEM and MEM measurements, duplicates and performance evaluations may be found in Jurvelin et al (2000) (1).

Smoking

Many of the compounds that are associated with automobile emissions and other combustion processes are also present in tobacco smoke, which is such a dominant localized source that variation due to more distant sources would be overwhelmed. Thus to avoid such confounding of the source identification, the analysis is restricted on participants not exposed to environmental tobacco smoke (ETS).

Step 1: Factor Analysis

- Principal component analysis with VARIMAX rotation on Natural Log transformed VOC personal exposure concentrations
- Factor analysis is used to identify underlying patterns that explain common variations among a set of variables. Principal component analysis (PCA) relies on a slightly different mathematical model where unique factor loading and scores are left out of the analysis, and uses linear combinations of element concentrations to characterize or account for the variation of each dimension in a multivariate space.
- Linear recombination of eigenvectors of the correlation matrix of element concentrations by applying a VARIMAX rotation produces the source vectors (3). Thus, the rotated factors represent major sources or meteorological effects to explain common variations in VOC concentrations in personal exposure samples.

Rotated Component Matrix[a]

	Component 1	2	3	4	5
LN 1-Butanol	.456			.491	-.445
LN 2- Ethylhexanol				.805	
LN 2-Methyl-1-propanol			.456		
LN 3-Carene			.875		
LN a-Pinene			.847		
LN Benzene	.633				.457
LN Decane		.883			
LN d-Limonene					.789
LN Ethylbenzene	.882				
LN Hexanal			.608	.477	
LN m,p-Xylene	.864				
LN Nonane		.802			
LN Octanal				.678	
LN o-Xylene	.857				
LN Propylbenzene	.531	.690			
LN Toluene	.825				
LN Trimethylbenzene	.553	.659			
LN Undecane		.817			

Extraction Method: Principal Component Analysis.
Rotation Method: Varimax with Kaiser Normalization.
a. Rotation converged in 14 iterations.

Identification Of Similar Source Factors in Helsinki (Edwards et al., 2001) (4)

Factor 1	Traffic/combustion emissions long range transport
Factor 2	localized traffic emissions
Factor 3	Cleaning and household products
Factor 4	product emissions from indoor environments and mould
Factor 5	d-limonene indoor sources

STEP 2: Stepwise Linear regression

Factor scores for each participant were saved for all factors. The sizes of the factor score coefficients for each case correspond to the loading for that factor. In other words the scores represent the strength of the source factor for each individual. The next approach was to use a stepwise linear regression with the factor scores for each factor or source category with the following variables as independent variables.

Input variables to models

Continuous	Binary	Categorised
minutes walking	HELSINKI	home location
minutes riding motorcycle	ATHENS	home building type
minutes in car	PRAGUE	location height of home
minutes in bus	OXFORD	kitchen fan or vent
minutes home indoors	attached garage	year home was built
minutes home outdoors	linoleum floor	traffic volume on nearby street
minutes at work indoors	pvc floor	heavy traffic volume on nearby street
minutes at work outdoors	wood floor	use of vacuum cleaner
minutes at other indoors	wood panelling	
minutes at other outdoors	plaster board walls/ceiling	
minutes cooking	chip board walls	
hours using gas stove	wallpaper	
hours using coal stove	wall painting/paper renovation in last year	
hours using fireplace	floor renovation in last year	
hours using fan	District heating	
hours	central heating	
hours using humidifier	electric heating	
hours using air conditioning	gas heating	
hours using electric dryer	fireplace home heating	
hours windows were open	air conditioning in home	
hours painting	use of air fresheners	
hours using glue	use of parfume	
hours in workshop	home location downtown	
hours used to wash car	Home location: Suburban high rise	
hours in gas station	Home location: Suburban small buildings	
hours used to grill		
hours used in garage		
hours exercising outdoors		
number of cats		
number of dogs		

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Dependent variable	Source Factor Identification	n	Adjusted r^2	Predictor variables	Standardized coefficients Beta	Colinearity diagnostic Condition Index
Factor score 1	Traffic and combustion long range transport	123	0.46	(Constant)		1
				PRAGUE	0.6	1.448
				ATHENS	0.294	1.548
				Time in other indoors	-0.162	1.631
				Time washing car	0.179	1.872
				Attached garage	0.168	1.963
				Home location: Suburban high rise	0.149	3.164
Factor score 2	localized traffic emissions	129	0.32	(Constant)		1
				Time exercising outdoors	0.373	1.563
				HELSINKI	-0.278	1.601
				Time in workshop	0.261	1.696
				Time cooking	-0.229	1.896
				Air conditioner in home	0.179	2.103
				Air fresheners in home	0.176	3.683
Factor score 3	Cleaning and household products	123	0.22	(Constant)		1
				electric heating	0.154	1.902
				District heating	-0.505	2.016
				HELSINKI	0.498	2.172
				chip board walls	-0.247	2.384
				paint/wallpaper renovation	0.179	4.111
				Time in car	-0.159	5.733
Factor score 4	indoor product emissions and mould	132	0.25	(Constant)		1
				OXFORD	-0.332	1.251
				minutes at work outdoors	0.262	1.499
				gas heating	0.389	1.832
				Time using gas stove	-0.293	2.137
				floor renovation	-0.208	2.596
Factor score 5	d-limonene indoor sources	123	0.27	(Constant)		1
				HELSINKI	-0.361	1.727
				Time windows were open	-0.258	1.952
				Time at work indoors	-0.224	2.506
				Time exercising outdoors	-0.171	4.422
				PRAGUE	-0.199	5.483

Activities and behaviors related to personal exposures to source factors

Factor 1: (Traffic / combustion long range)

- The variables included are compatible with the less volatile components of automobile emissions. In Helsinki this factor was clearly associated with long range transport, as wind vectors showed directional dependency of this factor although participants were spread over the whole metropolitan area and monitored during the whole year (4).
- Prague is the most dominant factor clearly identifying a more exposed population. The second more exposed population is Athens, both showed considerably elevated concentrations of these compounds relative to other centers. This is perhaps not surprising given that the exposure sample from Prague was selected from municipality employees, in other words downtown office workers, and in Athens indoor levels of these compounds were elevated and greater time was spent in the car. This is related to the third factor where more time spent in other indoor (not home or workplace) was related to reduced exposure to this factor.
- Secondary sub-populations with greater exposure and sources appear to be those who spend greater time in a carwash, whose homes have an attached garage and those who live in high-rise suburban neighborhoods. It is interesting to note attached garage appearing as these variables have also been identified as leading to higher exposures to the compounds in this factor in other studies.

Factor 2 (local traffic emissions)

- This factor was associated with the more volatile components of vehicle emissions and related to localized sources.
- The strongest predictor was the amount of time spent exercising outdoors
- A negative coefficient for those in Helsinki indicated that they were less exposed to this factor (but not because they exercised outdoors less).
- Subsequently the regression models identified those with a home workshop, a factor that has previously been identified with elevated exposures.
- Interestingly the next factor identified is those participants whose homes had an air conditioner, which allow penetration of outdoor air to indoors, followed by use of air fresheners. Use of air fresheners, however was associated with having an attached garage ($r = 0.35$, $p<0.001$) and those living downtown ($r=0.16$, $p=0.06$)

Factor 3: (Cleaning and household products)

- This factor appears to identify different types of homes and socioeconomic levels, associated with different product use.
- Helsinki participants were associated with increased exposure to this factor, and demonstrated the largest standardized co-efficient
- Electric heating – more prevalent in suburban single family homes – was associated with this factor. Interestingly district heating was less associated with this source, which is associated with apartment buildings, and suburban areas with high-rise buildings. This could reflect socioeconomic differences in the use of consumer products.
- Chipboard was also associated with suburban areas with single family homes but not associated with electric heating, and was negatively correlated with this source.
- Time spent in the car was negatively associated with this factor. Although time spent in the car was inversely correlated with time spent home indoor ($r=-0.25$, $p=0.003$) and time spent at work indoors ($r=-0.19$, $p<0.03$), removal of car did not allow these variables to enter the model.

Factor 4: (Product emissions and mould)

- Factor 4 was associated with product emissions from the indoor environment and mould. Interestingly Oxford, whose homes were more associated with the periphery and lower traffic, was less associated with this factor. More importantly, however, Oxford homes mostly had central heating, which may reduce damp and mould.
- Work outdoors was also positively associated with this factor, possibly due to products used or mould.
- Gas heating was also positively associated with this factor, but was mainly associated with homes in Prague ($r=0.55$, $p<0.001$). Time using a gas stove, however, was inversely associated with this factor.
- Floor renovation or repairs during the past year were also less associated with this factor, supporting association of this factor with mould.

Factor 5: (d-Limonene source)

- This appears to be a residential indoor source of d-limonene associated with product use.
- Predictors for this factor were all negative and associated with reduced exposure to this source. Exposures to this factor were negatively associated with participants in Helsinki, time windows were open at home, time spent at work indoors, time spent outdoors exercising and participants in Prague.

Shown as a poster at the 13th Annual Conference of the International Society of Exposure Analysis ISEA, Stresa (Italy), September 2003