



The German Environmental Survey 1990/92 (GerES 11): Sources of personal exposure to volatile organic compounds

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In the framework of the second German Environmental Survey carried out in the Western part of Germany in 1990/91 (GerES IIa) 113 adults aged 25–69 years were selected at random from the total study population of about 2500 to investigate personal exposure to about 70 volatile organic compounds (VOCs). Each subject wore a diffusive badge-type sampler for 1 week. The VOCs determined included alkanes, aromatics, aliphatic halocarbons, terpenes, and oxygen-containing compounds. Multivariate regression analysis was carried out to determine and quantify the major sources of personal exposure to various VOCs. In this paper, results are given for benzene, and C8- and C9-aromatics. Being subject to environmental tobacco smoke was found to be the most important determinant of benzene exposure, but automobile-related activities such as driving a car or refuelling, were also associated with significantly increased levels of benzene. The major determinant of C8- and C9-aromatics concentrations was occupational exposure. Emissions from paints, lacquers, newspapers, magazines and print-works were also important contributors to C8-aromatics exposure. Renovation, painting and smoking were associated with a significant increase of the exposure to C9-aromatics. *Journal of Exposure Analysis and Environmental Epidemiology* (2000) 10, 115–125.

Keywords: exposure assessment, GerES, multivariate statistical analysis, personal monitoring, volatile organic compounds.

Introduction

The number of studies on the occurrence of various volatile organic compounds (VOCs) in indoor air has increased in recent years (Miksch et al., 1982; Mølhave, 1982; Girman et al., 1986; Lebrecht et al., 1986; Seifert et al., 1989; Krause et al., 1991; Brown et al., 1993; Crump and Madany, 1993; Fellin and Otson, 1993; Schreiber et al., 1993; Heavner et al., 1995, 1996). Although people tend to spend most of their time indoors, indoor VOC concentrations do not necessarily reflect personal exposure adequately. Even modelling personal exposure using indoor monitoring and time-activity data is not without problems. At the present stage, the most reliable information on personal exposure will be obtained by personal monitoring in which the pollutants of interest are measured close to the breathing zone of a person. In previous studies dealing with personal monitoring (Wallace et al., 1988, 1991; Proctor et al., 1991) only 24-h exposures were analysed. Wallace et al. (1994) stated that such short-term measurements are not

suitable to estimate long-term distributions of exposure to most of the analysed VOCs. The sampling period should be at least 1 week including the weekend, because exposure may partly be due to activities that people do only occasionally. In fact, longer-term exposure measurements are important to generate as accurate as possible annualised (or even lifetime) exposure distributions. Long-term sampling has not only been used in GerES I (Krause et al., 1987) but also in the NHEXAS study (Pellizzari et al., 1995).

In the framework of the second German Environmental Survey (GerES IIa) conducted in the Western part of Germany (old states before reunification) a study was conducted to characterise population exposure to about 70 VOCs (Hoffmann et al., 1996). Here we report and discuss the results obtained for benzene and a number of its homologues.

Methods

Selection of the Study Participants

In GerES IIa, 2524 adults aged 25 to 69 years were randomly selected using a two-stage procedure stratified according to community size, gender and age (Seifert et al., 2000). From this population sample a subsample of 113 subjects was drawn at random to study personal exposure to VOCs. The participating subjects were distributed over 36 sample points to cover different areas of the Western part of

1. Abbreviations: GerES, German Environmental Survey; VOC, volatile organic compounds; TVOC, total volatile organic compounds; ETS, environmental tobacco smoke; CI, confidence interval; GM, geometric mean; LOQ, limit of quantification.

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Table 1. Comparison between study population and total population.

Category	Study population (%) (N=113)	Total population (%) (N=35,841,000)*
Sex		
female	48.7	51.0
male	51.3	49.0
Age (years)		
25–29	15.9	13.6
30–39	24.0	23.2
40–49	22.1	20.0
50–59	19.5	23.5
60–69	17.7	18.9

*West German resident population, aged 25–69 years, on the basis of the micro-census 1991.

Germany. The distribution by gender and age in the subsample was similar to that in the population (Table 1). The number of persons engaging in exposure related activities like smoking and refuelling can be found in Tables 9, 11 and 13.

Personal Sampling

OVM-3500 diffusive samplers (3M, St. Paul, MN, USA) were used as personal samplers to collect VOCs. A member of the field staff instructed the subjects how to use the personal sampler. The subjects then wore the monitor close to the breathing zone over seven days placing it at the bedside when sleeping. At the end of the sampling period, the sampler's collecting surface was covered with the lid provided by the manufacturer for this purpose. The sampler was then wrapped tightly in an aluminium foil and sent to the laboratory for analysis.

Subjects were asked to complete a special questionnaire and to note the time they stayed in the living room, bedroom, kitchen, bathroom, and in other rooms out of home as well as in traffic and outdoors. The three rooms they stayed in most of the time had to be described more precisely with regard to floor space, height, ventilation habits, type of heating system, presence of tobacco smoke, type of windows as well as type and age of wallpapers, carpets and furniture. Moreover, activities like renovation and painting during the 7-day period had to be reported. Besides these information, each subject completed the basic questionnaire of GerES IIa which included detailed questions about the household, characteristics of the residential quarter, occupation, smoking habits and other lifestyle characteristics.

Analysis and Quality Control

The diffusive samplers were analysed by a gas chromatographic procedure according to German VDI guideline

3482/4 (VDI, 1984). This procedure includes sampling on activated carbon, desorption with CS₂ and separation using a nonpolar capillary gas chromatographic column. To confirm the identification of compounds which was done via the retention indices and retention index differences, another aliquot of the sample was analysed using a second gas chromatograph equipped with a column of slightly different polarity. The total procedure used has been described in more detail by Seifert et al. (1989). Seventy-four VOCs, including *n*- and iso-alkanes, aromatic hydrocarbons, halocarbons, terpenes, esters, ketones and alcohols were analysed. Each VOC was calibrated individually using standard compounds. The peak area obtained from the instrument's computer system was checked manually for correct baseline setting. The baseline was corrected in case of need. Quantification was carried out by means of an internal standard method (ISM) using cyclooctane and 1,2,3-trichloropropane as internal standards. The ISM included correction for potential dilution errors, varying solvent desorption efficiency and errors in gas chromatographic injection volumes.

A field blank was taken with every 10th sample in order to test for correct sample handling, transportation and storage conditions. Furthermore, laboratory blanks were run continuously. In one case interfering compounds were found in the field blank, and their concentrations were markedly higher than those of the laboratory blanks. Since this blank sample exhibited a specific VOC pattern which was also found in the accompanying real sample, the latter was excluded from further treatment. Consequently, all results are based on 113 data sets.

In the evaluation of the results of a European inter-laboratory experiment on the performance of diffusive samplers (De Bortoli et al., 1986) in which eight laboratories participated, the mean relative interlaboratory standard deviation has been calculated for a variety of compounds representing different chemical classes, namely 1-butanol, 1,1,2-trichloroethane, 1-octene, butylacetate, 3-heptanone, *o*-xylene, alpha-pinene, and *n*-decane. The mean relative interlaboratory standard deviation was found to be about 13% for concentration levels between 20 and 1000 µg/m³ and a sampling period of four days. In addition, it was found that for nonpolar compounds (*o*-xylene and *n*-decane) the passive sampling results were generally in close agreement with those of active sampling whereas for the six polar compounds named above the results of active sampling using thermal desorption after collection on Tenax exceeded those of passive sampling by 13% to 56%. In the interlaboratory comparison mean desorption efficiencies of 78% to 105% were obtained for loadings of 10 to 100 µg/sampler. In our own laboratory, desorption efficiencies were between 93% and 105%. Only in the case of 1-butanol was the desorption efficiency much lower and more variable, namely between about 30% and 65%. This variability led us

to abstain from correcting the alcohol concentration levels. It was felt, however, that even the **uncorrected data**, although underestimating the real concentrations, should be included in Table 5 because they represent useful information.

To check the validity of the passive samplers results, a test series was conducted in a test chamber using test gases at low concentration levels. The model compounds, 1,1,1-trichloroethane, *m*-xylene, *n*-heptane, benzene, toluene, and *n*-hexane were studied at concentrations below 25 µg/m³ (Ullrich and Nagel, 1996). In these tests passive sampling was carried out in parallel to active sampling which was used as a reference. Passive samplers were exposed to these concentrations between 3 days and 3 weeks. If a mean uptake rate of 27.9 cm³/min was used to calculate the results for the passive samplers, the overall mean deviation between active and passive sampling was -8.5% with a range between +2% and -19%. For high-boiling compounds (*n*-decane to *n*-dodecane) it has to be anticipated that the deviation is larger and may attain about 40%.

In another study which has been completed since (Ullrich et al., 1999) relative standard deviations of concentrations obtained by the passive sampling procedure including the sampling step were determined from 18 duplicate samples. Relative standard deviations varied between 5% (benzene) and 31% (*n*-dodecane).

In addition, quality assurance was achieved by participation in external intercomparison experiments (Hoffmann et al., 1996). From the information obtained in all these quality control activities it can be concluded that for the reported results a measurement error between about 10% and 50% has to be assumed, depending on the compound and the concentration level.

Statistical Data Treatment

The statistical data analysis was performed using SPSS software (SPSS, 1993) and included regression analysis as well as descriptive methods. As usual, regression models were derived on the basis of unweighted data. To be compatible with the regression results, tables with data across categories of model predictors were also based on unweighted records. The Shapiro-Wilk test was applied to test the distribution type of the data (for more details see Results and Discussion section).

Results and discussion

Statistical parameters that describe the concentration distribution of most of the determined VOC are given in Tables 2-5. Not included are 4-methylheptane, 1,2,5-trimethylhexane, 2-ethyltoluene, 1,2,4,5-tetramethylbenzene, tetraline, 1,4-dichlorobenzene, trichloromethane,

tetrachloromethane, bromodichloromethane, 1,1,2-trichloroethane, chlorodibromomethane, tribromomethane, α -terpinene, isobutylacetate, 4-methyl-2-pentanone, hexanal, 2-methoxyethylacetate and 2-ethoxyethylacetate as more than two thirds of the respective concentrations were below the LOQ. Rather than determining the LOQ for each VOC individually, it was felt that it was more appropriate to choose one LOQ value for all VOCs. The LOQ was set to 1 µg/m³. This is an estimate and higher than statistically derived LOQ. The estimate takes into account some variability of the blank value and uncertainties of the "real" baseline of small chromatographic peaks. Corrections for blanks had only to be applied in the case of benzene and *n*-hexane which were contained even in pure CS₂.

Table 6 gives the results for different groups of VOCs. The concentration of a group was calculated by summation of the concentrations of all analysed compounds belonging to the respective group. In a similar way, the total volatile organic compounds (TVOC) concentration was obtained as the sum of the concentrations of all 74 individually calibrated VOCs. In the summation, concentrations of compounds below LOQ were set at 0.7 LOQ. The factor 0.7 was chosen since in the case of asymmetric distributions it is a better choice than the normally used factor 0.5 (Hallez and Derouane, 1982).

As could be expected from theoretical considerations (Ott, 1990) and the results of previous studies (Lebrete et al., 1986; Krause et al., 1991; Wallace et al., 1991) the concentration of most of the VOCs had asymmetric distributions with a small portion of extremely high values which can be well approximated by logarithmic normal distributions. Statistical tests applied to the present data confirmed the lognormality of the distributions. Tests for lognormality were performed as tests for normality using the logarithmically transformed data. Since the sample size was moderate, the Shapiro-Wilk test was preferred to the Kolmogorov and the χ^2 test. Using the Shapiro-Wilk test, the null hypothesis of normality was not rejected for the large majority of the logarithmically transformed VOC concentrations. Tables 2-6 give the summary statistics for the different VOCs. Besides geometric mean (GM), arithmetic mean (AM), 10th, 50th, and 90th percentiles, the 95th percentile was presented to focus on persons with high exposure. The geometric mean seems to be the preferred statistical location measure for the given data since it is robust against single elevated values. To describe the estimation error, a 95% confidence interval (CI-GM) was added. The standard deviation of the GM can be easily calculated from the confidence interval and the given sample size if wanted.

Toluene was the most frequently observed compound in the present study. This is in accordance with the results of other studies of personal and indoor air (e.g., Krause et al., 1991; Proctor et al., 1991) and can be explained by both its

Tabk 2. Summary statistics: Alkanes, including cycloalkanes ($\mu\text{g}/\text{m}^3$) [$N=113$].

compound	$n < \text{LOQ}$	P10	P50	P90	P95	AM	GM	CI-GM
<i>n</i> -Hexane	0	6	9	20	40	13.4	10.3	(9.2–11.5)
<i>n</i> -Heptane	0	3	5	22	49	14.7	6.1	(5.1–7.3)
<i>n</i> -Octane	26	<1	2	8	20	4.3	2.1	(1.7–2.5)
<i>n</i> -Nonane	9	1	3	14	26	6.0	3.3	(2.7–4.0)
<i>n</i> -Decane	3	2	5	23	41	9.7	5.2	(4.3–6.3)
<i>n</i> -Undecane	1	2	4	21	29	9.9	5.2	(4.4–6.2)
<i>n</i> -Dodecane	2	3	5	12	21	8.0	5.6	(4.9–6.5)
<i>n</i> -Tridecane	12	<1	2	6	10	3.2	2.2	(1.9–2.5)
<i>n</i> -Tetradecane	0	2	3	5	7	3.6	3.3	(3.1–3.5)
<i>n</i> -Pentadecane	0	2	3	5	5	3.5	3.4	(3.2–3.5)
2-Methylpentane	0	7	13	39	150	29.2	16.3	(13.9–19.1)
3-Methylpentane	5	2	6	30	53	11.6	6.3	(5.2–7.6)
2,3-Dimethylpentane	2	2	4	11	41	8.1	4.1	(3.4–4.8)
2-Methylhexane	34	<1	2	6	19	4.3	1.7	(1.4–2.1)
3-Methylhexane	1	2	4	14	45	10.3	4.8	(4.0–5.7)
2-Methylheptane	60	<1	<1	4	9	2.2	1.3	(1.1–1.5)
3-Methylheptane	71	<1	<1	3	7	1.7	1.1	(1.0–1.2)
Iso-nonane I	8	1	2	4	6	2.4	2.0	(1.7–2.2)
Iso-nonane II	62	<1	<1	3	5	1.5	1.1	(1.0–1.3)
Methylcyclopentane	0	2	3	8	23	5.6	3.9	(3.5–4.5)
Cyclohexane	0	2	3	11	20	20.4	3.8	(3.1–4.6)
Methylcyclohexane	2	2	3	20	54	11.5	4.4	(3.6–5.4)

N =sample size; $n < \text{LOQ}$ =number of values below the limit of quantification (LOQ; for details see text); P10, P50, P90, P95=percentiles; GM=geometric mean; CI-GM=95% confidence interval for GM. Values below LOQ are set to 0.7. LOQ for calculation purposes.

presence in outdoor air and its frequent use indoors as a solvent in paints, lacquers, printing inks, adhesives and other household products (Kumai et al., 1983; Sack et al., 1992). Somewhat unexpected was that 2-propanol and

limonene were next in rank with mean concentrations higher than $30 \mu\text{g}/\text{m}^3$. All together, the geometric mean of the TVOC concentration was $584 \mu\text{g}/\text{m}^3$. If VOCs are considered by chemical class, aromatics were the most

Tabk 3. Summary statistics: Aromatic compounds ($\mu\text{g}/\text{m}^3$) [$N=113$].

Compound	$n < \text{LOQ}$	P10	P50	P90	P95	AM	GM	CI-GM
Benzene	0	5	11	21	32	13.5	10.5	(9.3–11.9)
Toluene	0	32	69	208	382	130.2	13.9	(62.7–87.0)
Ethylbenzene	0	3	7	21	106	24.0	8.5	(7.1–10.2)
<i>m</i> -lp-Xylene	0	7	16	55	283	50.5	19.9	(16.4–24.1)
<i>o</i> -Xylene	1	2	5	17	67	13.6	6.5	(5.4–7.7)
Iso-/ <i>n</i> -propylbenzene	12	1	3	8	12	4.6	3.5	(3.1–3.9)
3-/4-Ethyltoluene	1	3	8	21	35	12.5	8.3	(7.2–9.7)
1,2,3-Trimethylbenzene	5	2	4	8	12	4.9	3.8	(3.4–4.3)
1,2,4-Trimethylbenzene	0	3	7	22	44	11.8	7.3	(6.2–8.5)
1,3,5-Trimethylbenzene	2	2	3	7	13	4.8	3.7	(3.2–4.1)
Styrene	24	<1	2	7	8	5.1	2.1	(1.8–2.5)
Naphthalene	4	1	2	3	4	2.3	2.1	(2.0–2.3)
4-Phenylcyclohexene	0	3	5	6	7	4.8	4.1	(4.4–4.9)

N =sample size; $n < \text{LOQ}$ =number of values below LOQ (for details see text); P10, P50, P90, P95=percentiles; AM=arithmetic mean; GM=geometric mean; CI-GM=95% confidence interval for GM. Values below LOQ are set to 0.7. LOQ for calculation purposes.

Tabk 4. Summary statistics: Aliphatic halocarbons and terpenes ($\mu\text{g}/\text{m}^3$) [$N=113$].

Compound	$n < \text{LOQ}$	P10	P50	P90	P95	AM	GM	CI-GM
1,1,1-Trichloroethane	39	<1	1	15	20	6.6	2.1	(1.7–2.7)
Trichloroethylene	75	<1	<1	6	8	2.7	1.2	(1.0–1.4)
Tetrachloroethylene	33	<1	2	6	22	3.8	2.0	(1.7–2.4)
α -Pinene	1	2	5	34	74	18.3	6.7	(5.4–8.3)
β -Pinene	3	3	4	12	20	8.1	4.6	(4.0–5.4)
3-Carene	12	<1	3	17	30	9.8	3.4	(2.8–4.2)
Limonene	0	10	32	124	155	53.5	34.1	(28.7–40.6)
Terpinene-artefact	22	<1	3	9	15	5.0	3.0	(2.5–3.6)
γ -Terpinene	25	<1	2	5	6	2.5	1.9	(1.7–2.2)

N =sample size; $n < \text{LOQ}$ =number of values below LOQ (for details see text); P10, P50, P90, P95=percentiles; AM=arithmetic mean; GM=geometric mean; CI-GM=95% confidence interval for GM. Values below LOQ are set to 0.7. LOQ for calculation purposes.

prevalent compounds, followed by oxygen-containing compounds and alkanes (Table 6).

Correlation between VOCs

To detect correlations between VOCs of one and the same chemical class the Pearson correlation coefficient was calculated for different pairs of VOCs. Since the Pearson correlation coefficient is highly influenced by one or two high values in a data set, logarithmic concentrations were used as the data basis. In this way, the influence of outliers is reduced without the loss of information that would have occurred in the ranking process needed to calculate Spearman's rank correlation coefficient. The higher the correlation coefficient between logarithmic concentrations of two compounds the less the ratios of the compound levels in all samples vary. It can be assumed that high correlations

of VOCs are due to emissions of the same sources. As a result of an extensive correlation analysis subclasses of highly correlated compounds could be derived (Table 7).

Compounds belonging to the same subclass all showed correlations that were significantly different from zero at the 0.001 level of significance. The Pearson correlation coefficients within the classes varied between 0.67 and 0.99. Very high correlation coefficients were obtained for C8-aromatics. VOCs not belonging to any of the more frequently encountered classes were, in general, not highly correlated with other VOCs as, very likely, they have some specific emission sources or their own source pattern. The result of the correlation analysis was checked by factor analysis, another multivariate statistical method. The classes of compounds obtained by factor analysis coincide largely with the classes given in Table 7.

Tabk 5. Summary statistics: Oxygen-containing compounds ($\mu\text{g}/\text{m}^3$) [$N=113$].

Compound	$n < \text{LOQ}$	P10	P50	P90	P95	AM	GM	CI-GM
Ethylacetate	1	5	12	46	180	69.8	15.4	(12.4–19.1)
n -Butylacetate	29	<1	4	31	135	35.3	4.4	(3.2–5.9)
Methyl ethyl ketone	16	<1	4	44	92	24.1	5.1	(3.9–6.7)
3-Heptanone	60	<1	<1	4	6	1.7	1.2	(1.1–1.4)
Methyl benzoate	35	<1	2	5	7	3.1	1.8	(1.6–2.1)
n -Butanol ¹	60	<1	<1	15	31	6.9	2.0	(1.5–2.5)
Iso-butanol ¹	67	<1	<1	7	23	22.0	1.6	(1.3–2.1)
Iso-amyl alcohol ¹	60	<1	<1	2	3	1.4	1.1	(1.0–1.2)
2-Ethyl-1-hexanol ¹	25	<1	4	8	14	4.4	3.0	(2.5–3.5)
2-Propanol ¹	1	7	40	188	326	80.0	39.1	(30.8–49.6)
t -Butyl methylether	57	<1	<1	7	16	4.1	1.8	(1.4–2.2)

N =sample size; $n < \text{LOQ}$ =number of values below LOQ (for details see text); P10, P50, P90, P95=percentiles; AM=arithmetic mean; GM=geometric mean; CI-GM=95% confidence interval for GM. Values below LOQ are set to 0.7. LOQ for calculation purposes.

The concentrations given underestimate the real concentrations (see text for details).

Table 6. Summary statistics: Sums of VOC concentrations by chemical class ($\mu\text{g}/\text{m}^3$) [$N=113$].

Chemical class	P10	P50	P90	P95	AM	GM	CI-GM
Alkanes, including cycloalkanes	56	107	336	641	187	122	(105–140)
Aromatic hydrocarbons	78	150	608	944	286	180	(155–208)
Aliphatic halocarbons	7	13	38	68	21	16	(14–18)
Terpenes	23	57	188	303	98	64	(55–75)
Oxygen-containing compounds	44	106	438	739	308	127	(105–152)
Total volatile organic compounds (TVOC)	275	511	1480	2810	901	584	(509–671)

N =sample size; P10, P50, P90, P95=percentiles; AM=arithmetic mean; GM=geometric mean; CI-GM=95% confidence interval for GM; TVOC=sum of the concentrations of all 74 VOC.

Sources of Exposure to Benzene

To determine and quantify the main factors that influence the benzene level, regression analysis was applied. About 100 variables comprising relevant questionnaire data were considered for entry into the regression model. Using a stepwise selection procedure a model with five predictors was derived (Table 8). The model function is a product of factors since it was obtained by exponential transformation from a linear model derived for the logarithmic benzene concentration. This model transformation was necessary because the benzene concentration did not meet the requirement of regression analysis to be normally distributed, whereas the logarithmic benzene concentration fulfilled this condition.

The two most important predictors in the model relate to the presence of environmental tobacco smoke (ETS) indoors. They account for 20% of the observed variance of the benzene concentration. Two automobile-related activities, namely refuelling and the time spent in automobile traffic, were further predictors. Together they explain 12% of the variance of benzene. The fifth predictor is a variable expressing population density in the area which accounted for 7% of the observed variance. The presence of this predictor in the model is likely to be due to elevated

emission rates of benzene from automobile exhaust gases in areas with high population density. Altogether, 39% of the observed variance of benzene are explained by the model. The remaining 61% of the variance includes errors in measuring the benzene concentration and incompleteness in allowing for all possible sources of exposure. As a consequence of the latter point some specific but unknown sources of exposure are not considered in the model. On the other hand, the two known main determinants of benzene concentration, ETS and automobile-related activities, are subject to a number of variations details of which are unknown, however. For example, the effect of 1 h spent daily in automobile traffic varies depending on, e.g., traffic density, type and age of vehicle, ventilation conditions, and temperature.

In contrast to usual linear regression models, the interpretation of a multiplicative regression model is based on a percentage rather than an absolute change. An increase or decrease of the value of any predictor given in Table 8 always implies a percentage change of the benzene level. For instance, an increase by 1 h of the daily time spent in automobile traffic, which means that predictor D (see Table 8) is increased by 1, results in a relative increase of the expected benzene concentration by 21% since the corres-

Table 7. Subclasses of highly correlated VOC.

Subclass	Compounds	Correlation coefficients ^a
C6-alkanes	<i>n</i> -hexane, iso-hexane, methylcyclopentane	0.75–0.93
C7-alkanes	<i>n</i> -heptane, iso-heptane, methylcyclohexane	0.82–0.94
C8-alkanes	<i>n</i> -octane, isooctane	0.89
C9–C11-alkanes	<i>n</i> -nonane, iso-nonane, <i>n</i> -decane, <i>n</i> -undecane	0.67–0.93
C12-, C13-alkanes	<i>n</i> -dodecane, <i>n</i> -tridecane	0.67
C14-, C15-alkanes	<i>n</i> -tetradecane, <i>n</i> -pentadecane	0.76
C8-aromatics	ethylbenzene, <i>m</i> -/p-xylene, o-xylene	0.97–0.99
C9-aromatics	iso-/n-propylbenzene, 3-/4-ethyltoluene, 1,2,3-trimethylbenzene, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene	0.71–0.96
selected terpenes	α -pinene, β -pinene, 3-carene	0.63–0.80

^aPearson correlation coefficients for the logarithmically transformed concentrations.

Tabk 8. Predictors and regression model for the benzene concentration in inhaled air.

Symbol	Predictor	Percent variance explained
O	Subject to occasional or frequent smoking indoors (no=0, yes=1) ^a	8%
F	Subject to frequent smoking indoors (no=0, yes=1) ^a	12%
D	Daily time in automobile traffic (in hours)	6%
P	Refuelling (no=0, yes=1)	6%
L	Living in or next to, an area with apartment houses (no=0, yes=1)	7%
		R²=39%
Regression model for the benzene concentration (Y): $Y = 5.11 \times 1.38^O \times 1.66^F \times 1.21^D \times 1.28^P \times 1.40^L$ ($\mu\text{g}/\text{m}^3$)		

R²=Coefficient of determination.

^aPersons that are subject to frequent smoking indoors are characterised by O=1 and F=1, whereas persons that are subject to occasional smoking indoors are characterised by O=1 and F=0.

ponding factor in the model equation is 1.21. Refuelling once in a week, i.e., an increase of predictor **P** by 1 causes an increase of the predicted benzene concentration by 28%. A person staying mainly in rooms where he/she or others are frequently smoking has to expect a benzene level in air that is 129% higher than a person who does not stay in such rooms. This relative change follows from multiplication by both factors concerning the presence of ETS indoors. The constant 5.11 $\mu\text{g}/\text{m}^3$ of the model equation represents the basic level of exposure to benzene which is not due to smoking indoors or any other of the benzene-related predictors.

The influence of the different predictors on the benzene level could also be demonstrated by bivariate statistical analysis. In Table 9 the mean benzene concentrations were given for subgroups defined by the predictors of the regression model. However, it should be noted that bivariate description is less appropriate to predict exposure than is multivariate statistical analysis, such as regression analysis. For instance, Table 9 would suggest that refuelling activities cause an increase of the benzene level by about 50% while the regression model indicates only an increase of 28%. In this case the result of bivariate analysis is erroneous because it does not take into account that there exists a correlation between the two important automobile-related variables. Obviously, people who refuel their car are drivers who spend more time in automobile traffic than others.

The finding of the present study that the presence of tobacco smoke indoors and automobile-related activities are the main sources of personal exposure to benzene supports earlier results on different levels of benzene concentrations in the indoor air of smokers' and nonsmo-

kers' homes (Krause et al., 1987). It is also in agreement with the results of the TEAM study (Wallace et al., 1988; Wallace, 1990). In the present study roughly half of the subjects refuelled their car during the sampling week. Although this activity needs only a short time, its contribution to the cumulative exposure during the sampling period is considerable because high benzene concentrations can be observed at gas stations. As an example, 500 $\mu\text{g}/\text{m}^3$ were found by Römmelt et al. (1989) in Munich.

While close to 40% of the variance of the benzene concentration could be explained, this percentage was much lower for toluene (18%). This may be due to the large number of possible sources of toluene indoors and outdoors. The only significant predictors of toluene obtained in the regression analysis were the occurrence of paints or lacquers at the workplace and the use of adhesives. A larger sample size than 113 may be required to achieve a higher explanation of variance and a larger number of significant predictors.

Sources of Exposure to C8-Aromatics

Ethylbenzene, *m*-/*p*-xylene and *o*-xylene were highly correlated (see Table 7), which means that the three compounds occurred in characteristic proportions. The concentration ratio of ethylbenzene to *o*-xylene was approximately 1.3 with a 95% confidence interval from 0.8 to 2.1, while the estimated ratio of *m*-/*p*-xylene to *o*-xylene was 3 with a 95% confidence interval from 2.2 to 4.4. These data reflect the proportions of the C8-aromatics concentrations in crude oil. For comparison purposes it may

Table 9. Benzene concentrations in inhaled air ($\mu\text{g}/\text{m}^3$) across categories of the model predictors.

Subgroup	N	Geometric mean
<i>Subject to smoking indoors</i>		
never	41	7.8
occasional	54	10.8
frequent	18	19.6
<i>Daily time in automobile traffic</i>		
≤20 min	31	8.3
21–60 min	45	10.4
>60 min	37	13.0
<i>Refuelling</i>		
no	56	8.6
yes	57	12.8
<i>Living in or next to, an area with apartment houses</i>		
no	62	9.0
yes	51	12.8

be mentioned that Scheff et al. (1989) found a ratio of 1.5 form-*p*-xylene to *o*-xylene in auto exhaust while the ratio between ethylbenzene and *o*-xylene was 0.5. From published data (Wallace et al., 1987; Hajimiragha et al., 1989) it may be concluded that the ratio of ethylbenzene to the other C8-aromatics is somewhat higher in cigarette smoke than in auto exhaust.

Again, stepwise regression analysis was applied to identify the main sources of exposure. The predictors selected from more than 100 variables were identical for the three individual C8-aromatics and their sum. In Table 10 the five predictors and the model for the sum of C8-aromatics are given. The models for the individual compounds differed only slightly from the equation given and, therefore, are omitted here.

The regression model accounted for 60% of the observed variance of the total C8-aromatics concentration. Spending time in workshops and warehouses and the Occurrence of paints or lacquers at the work place were the major contributors to the model. The three corresponding predictors explained 44% of the observed variance. Reading newspapers or magazines and living or working near print-works or printing shops accounted for 10% and 6% of the variance, respectively.

The model allows to quantify percent changes of C8-aromatics levels as a function of the predictor values. The cumulative exposure to C8-aromatics increases by 22% if the time spent daily in workshops and warehouses is prolonged by 1 h. Workers who occasionally are subject to the influence of paints or lacquers at work have to expect C8-aromatics concentrations in the air they breathe that are higher by 69% than persons not subject to such conditions. The lowest C8-aromatics level obtained from the regression

Table 10. predictors and regression model for C8-aromatics in inhaled air.

Symbol	Predictor	Percent variance explained
D	Daily 'time in workshops and warehouses (in hours)	20%
O	Occasional or frequent occurrence of paints or lacquers at workplace (no=0, yes=1) ^a	9%
F	Frequent occurrence of paints or lacquers at workplace (no=0, yes=1) ^a	15%
T	Daily time spent on reading newspapers and magazines (in hours)	10%
P	Living or working near print-works or printing shops (no=0, yes=1)	6%
		R ² =60%
Regression model for the SUM of C8-aromatics concentrations (Y): Y=19.3×1.22 ^D ×1.69 ^O ×6.45 ^F ×1.55 ^T ×1.94 ^P (µg/m ³)		

R²=Coefficient of determination.

^aFrequent occurrence of paints or lacquers is characterised by O=1 and F=1, whereas occasional occurrence of paints or lacquers is characterised by O=1 and F=0.

Table 11. C8-aromatics concentrations in inhaled air (µg/m³) across categories of the model predictors.

Subgroup	N	Geometric means			
		Ethylbenzene	m-lp Xylene	o-Xylene	C8-Aromatics
<i>Time in workshops and Warehouses</i>					
≤3 h/day	100	6.8	15.9	5.3	28.1
>3 h/day	13	48.0	114	30.4	195
<i>Paints or lacquers at work place</i>					
no or not employed	98	6.9	16.2	5.4	28.6
occasional	8	14.8	38.4	12.7	66.2
frequent	7	79.5	171	41.8	298
<i>Reading newspapers and magazines</i>					
≤45 min/day	86	8.0	18.7	6.1	32.9
>45 min/day	27	10.5	24.2	8.0	42.9
<i>Living or working near to print-works or printing shops</i>					
no	104	7.7	18.1	5.9	28.4
yes	8	30.9	67.3	21.0	88.6

model is given by the model constant, 19.3 µg/m³. This minimum will result if all predictor values are zero.

In Table 11 the mean concentrations of ethylbenzene, m-lp-xylene, o-xylene and their sum are given for subgroups defined by the predictors of the regression model. There is a striking uniformity of the three compounds in the percentage changes of the geometric means between subgroups. It is clear that in some cases, e.g., for those exposed to paints or lacquers at work, the size of the subsample is not large enough to permit the use of the geometric mean as a reliable estimate of the mean exposure of the corresponding subpopulation.

The main result of the multivariate statistical analysis was that exposure to C8-aromatics is substantially influenced by emissions from paints, lacquers and printing inks. This is consistent with the results of Kumai et al. (1983) and Sack et al. (1992) on the content of organic solvent components in various products. The authors found that ethylbenzene, m-*p*-xylene and o-xylene were contained in more than half of the paints and lacquers under study, partially in considerable concentrations. Wadden et al. (1986) suggested that 90% of the solvents in paints and lacquers are C8-aromatics and toluene.

It was somewhat surprising that other known emission sources like smoking and automobile exhaust did not appear in the final regression model. A possible explanation which, however, is a purely statistical one is that the sample size in the present study does not allow a model with more significant predictors. In fact, if the three dominant predictors covering occupational exposure were removed

Table 12. Predictors and regression model for C9-aromatics in inhaled air.

Symbol	Predictor	Percent Variance explained
<i>D</i>	Daily time in workshops and warehouses (in hours)	28%
<i>R</i>	Renovation or painting in the person's surroundings (no=0, yes=1)	13%
<i>N</i>	Number of cigarettes smoked per day	5%
<i>L</i>	Living in or next to, an area with apartment houses (no=0, yes=1)	3%
		$R^2 = 49\%$

Regression model for the sum of C9-aromatics concentrations (*Y*):
 $Y = 18.7 \times 1.24^D \times 1.64^R \times 1.02^N \times 1.25^L$ ($\mu\text{g}/\text{m}^3$)

R^2 = Coefficient of determination.

from the model, some variables concerning smoking and automobile-related activities would enter the regression model. However, such a model would only explain about 30% of the observed variance of the C8-aromatics. This degree of explained variance is similar to that of the regression models derived in the **TEAM** study (Wallace et al., 1988).

It should be noted that in the **case** of competing models which explain different portions of the variance of the VOC concentration by considering different pathways of exposure, the percentage of the explained variance could be increased by combining the predictors of the competing

models. However, to ascertain the statistical significance of all the predictors in the combined model more subjects would have to be studied.

Sources of Exposure to C9-Aromatics

For the individual compounds, iso-/*n*-propylbenzene, 3-J 4-ethyltoluene, 1,2,3-trimethylbenzene, **1,2,4-trimethylbenzene**, and **1,3,5-trimethylbenzene**, and for the sum of C9-aromatics including 2-ethyltoluene, stepwise regression analysis was carried out to determine the most important **factors** that influence human exposure. The **final** regression models for the individual compounds contained three or four predictors and did not differ very much one from the other. Hence, only one model equation is given in Table 12 for the **sum of C9-aromatics**, together with the corresponding predictors.

Almost half of the variance of the total C9-aromatics concentrations **can be** explained by the regression model. **Spending time in workshops** and warehouses was the major contributor to the model with a contribution of 28%. Renovation activities in the person's surroundings (**at home, at work place**) accounted for 13% of the observed variance. Intensity of smoking and population density explained 5% and 3% of the variance, respectively.

The model can be used to predict levels of C9-aromatics in a person's breathing **air** and to **quantify the effects** of changing predictor values. Prolonging the **time** spent daily in workshops and warehouses by 1 h is connected with an increase of the expected level of C9-aromatics by 24%

Table 13. C9-aromatics concentrations in inhaled air ($\mu\text{g}/\text{m}^3$) across categories of the model predictors.

Subgroup	N	Geometric means					
		Iso- + n - propylbenzene	3- + 4- Ethyltoluene	1,2,3- Trimethylbenzene	1,2,4- Trimethylbenzene	1,3,5- Trimethylbenzene	C9- Aromatics
<i>Time in workshops and warehouses</i>							
≤3 h/day	100	3.1	7.2	3.4	6.2	3.3	25.1
>3 h/day	13	9.1	26.4	9.5	24.4	9.2	84.7
<i>Renovation/painting in the surroundings</i>							
no	91	3.0	7.0	3.4	6.0	3.2	24.6
yes	22	6.3	17.4	6.3	15.6	6.2	56.8
<i>Number of cigarettes smoked</i>							
0 (nonsmoker)	61	3.3	7.5	3.6	6.3	3.3	26.2
1–20 cigarettes/day	19	3.9	10.0	4.0	9.4	4.1	34.1
>20 cigarettes/day	7	5.8	18.8	6.5	20.5	6.5	61.8
<i>Living in or next to, an area with apartment houses</i>							
no	62	3.2	7.1	3.3	6.3	3.2	25.3
yes	51	3.8	10.2	4.6	8.6	4.4	33.9

since the corresponding model factor is **1.24**. Renovation activities during the sampling period increases the cumulative exposure to C9-aromatics by **64%**. The effect of smoking is less pronounced. The sum of the C9-aromatics concentrations increases by **2%** if the number of cigarettes smoked per day is increased by one. According to the model given in Table 12 this means that a person who smokes, e.g., 10 cigarettes daily has to expect a C9-aromatics level in breathing air that is about 22% higher than that of a nonsmoker. The basic level of the sum of C9-aromatics is $18.7 \mu\text{g}/\text{m}^3$. It should be noted that the Occurrence of paints or lacquers at work is an additional predictor for the two compounds 1,2,3-trimethylbenzene and 1,3,5-trimethylbenzene.

To illustrate and confirm the results of regression analysis, Table 13 describes mean C9-aromatics concentrations for subgroups defined by the predictors of the model. Persons who stay more than 3 h in workshops and warehouses daily have elevated C9-aromatics concentrations in the air they breathe. Also, the effect of renovation activities is remarkable and almost equal to the increase prognosticated by the model which indicates its independence of the other predictors.

Elevated C9-aromatic levels in a person's breathing air connected with a long stay in workshops and warehouses can be explained by emissions from solvent-containing industrial products like paints, lacquers and glues. The influence of renovation activities on human exposure to C9-aromatics is probably due to emissions from solvents. The predictor of smoking intensity reflects the exposure to C9-aromatics contained in tobacco smoke. Thus, two known emission sources of C9-aromatics, namely emissions from solvents and tobacco smoke can be shown to be relevant for human exposure by regression analysis. A third known emission source, C9-aromatic compounds in fuels and exhaust gases (Sigsby et al., 1987; Scheff et al., 1989) had only a negligible influence on a person's exposure and was therefore not considered in the regression model. Also, bivariate statistical considerations did not show associations between C9-aromatics levels and automobile-related activities like travelling in an automobile and refuelling.

Conclusions

Passive sampling over 1 week is a suitable tool to measure personal exposure. The results of such personal monitoring, in combination with information on time-activity patterns can be used to establish population exposure distributions for VOCs if a large enough number of individuals has been studied. Multivariate regression analysis was applied successfully to determine major sources of personal exposure to a number of

aromatic VOCs. For benzene, **40%** of the total variance could be explained with exposure to environmental tobacco smoke and automobile-related activities having the highest influence on exposure. In the case of C8-aromatics, even **60%** of the variance could be explained. Here, work place conditions proved to be the essential influencing factor for exposure followed by the contribution of handling printed matter in one way or the other. Besides the time spent daily in workshops and warehouses, renovation and painting activities, and — to a lesser extent — smoking were associated with a significant increase of the exposure to C9-aromatics. For these compounds almost half of the variance could be explained by the regression model.

The results of this study underline that personal exposure to aromatic VOCs is to a large extent influenced by both individual activities, especially those that relate to the automobile and to tobacco smoke, and work place conditions.

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