French permanent survey on indoor air quality—microenvironmental concentrations of volatile organic compounds in 90 French dwellings

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ABSTRACT
Before starting a French nationwide survey, a pilot study was conducted in 2001 on 90 dwellings in three geographical areas (North, East and South of France). This survey included measurements on 40 priority indoor parameters (VOC, NO_2, CO, CO_2, bacteria, moulds, allergens, MMMF, temperature, humidity) and questionnaires on building characteristics, occupants' description and time activity diaries. The paper focuses on the chemical levels found in dwellings. Volatile organic compounds (30 compounds including formaldehyde, acetaldehyde, BTEX) were measured by passive samplers, during 7 days, in kitchen, bedroom and outdoor from selected homes. Indoor concentrations were low, with 40% of values under the quantification limit. The range of values was wide. The maximum indoor concentrations measured in the kitchen were 400 $\mu g/m^3$ (alpha-pinene, *o*-xylene) and 293 $\mu g/m^3$ (1,4-dichlorobenzène) in the bedroom. Formaldehyde was frequently found varying form 2 to 75 $\mu g/m^3$. The outdoor concentrations were lower than indoor concentrations. Measurements in the kitchen and bedroom were significantly correlated.

INDEX TERMS
Indoor air; VOC; Aldehydes

INTRODUCTION
Decided by the French government in 1999 and funded by the Ministries in charge of Building, Health, Environment as well as the Energy Control and Environment Agency (ADEME) and CSTB, the Permanent Survey on indoor air quality (in French: Observatoire de la Qualité de l'Air Intérieur) is one of the major part of a multiyear governmental programme on 'building and health'. It aims to provide continuously the necessary data for risk assessment and management related to indoor air pollution exposure, by better understanding of: (1) environmental and behavioural determinants of indoor exposures; (2) population exposure levels. It is also designed to collect useful data for setting guidelines on ventilation systems as regards health and environment.

The French permanent survey aims to conduct experimental campaigns on the general population living in France and collect data issued from other projects conducted in France on indoor pollution. The survey's design (sampling, analytical methods, questionnaire's development, etc.) is elaborated and standardized as a peer validation basis with participation of experts from the field of public health, environment and building coordinated by CSTB (Kirchner *et al*., 2002).

Before starting the first nationwide survey on 710 dwellings (to be conducted in 2003), a pilot study has been conducted in 2001 on 90 dwellings (main residence) located in three geographical regions (North, East and South of France). This survey included measurements on 40 priority indoor parameters (VOC, NO_2, CO, CO_2, bacteria, moulds, allergens, MMMF, temperature, humidity) and questionnaires on building characteristics, occupant's description

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and time activity diaries. The paper focuses on the chemical measurements results found in dwellings.

METHODS

Dwelling Sampling Procedure

Ninety dwellings were chosen by surveyors with the objective to get various typologies of buildings (single house, multi-family houses, etc.). Volunteers were thus chosen using newspaper information, leaflets leaved in medical centres or direct contact with multi-family building owners.

Measurements of VOCs and Aldehydes

Thirty compounds (see details in Table 1) were measured during 7 days, in the kitchen and bedroom of selected homes and outdoors. Field blank tubes (transported, opened and closed and stored as other sampling tubes) were put in 10% of the dwellings. Moreover, replicate samplings (five tubes exposed at the same time) have been conducted in one-third of the dwellings.

Analysis of VOCs and Aldehydes

VOC sampling was performed using a radial passive sampler with Carbograph adsorbents (Cocheo *et al.*, 1996). Analyses were performed on an automatic thermal desorption apparatus (turbomatrix, Perkin-Elmer) linked to an autosystem XL gas-chromatograph (Perkin-Elmer) equipped with mass spectrometry detector (MSD) for identification and flame ionization detector (FID) for quantification and an apolar column (CP-SIL PONA CB, 100 m, 0.25 mm i.d., 0.5 µm). Aldehydes were sampled by a radial passive sampler with DNPH and desorbed with acetonitrile. The analysis was performed on a HPLC (Alliance 2695/2487, Waters) equipped with a UV/VIS detector operated at 360 nm. The separation was carried out on a Novapack C-18 column (Waters, 150 mm, 3.9 mm i.d., 4 µm). Five percent of tubes of each batch were used as laboratory blank tubes.

Table 1 Target compounds measured in the 90 dwellings

Aromatic hydocarbons	Benzene, toluene, ethylbenzene, *m*/*p*-xylenes, *o*-xylene, 1,2,4-trimethylbenzene, styrene
Aliphatic hydrocarbons (C6–C16)	*n*-Decane, *n*-undecane
Cycloalcanes	Cyclohexane
Terpenes	alpha-Pinene, limonene
Alcohols	2-Ethyl-1-hexanol
Glycol ethers	2-Ethoxyethanol, 2-butoxyethanol, 1-methoxy-2-propanol
Halogens	Trichloroethylene, tetrachloroethylene, 1,1,1-trichloroethane, 1,4-dichlorobenzene
Esters	Butyl acetate, isopropyl acetate, 2-ethoxyethyl acetate
Aldehydes	Formaldehyde, acetaldehyde, benzaldehyde, hexaldehyde, isobutyraldehyde/butyraldehyde, isovaleraldehyde, valeraldehyde

Statistical Analysis

The statistical data analysis was performed using SAS software release 8.2 (SAS Institute Inc., Cary, NC, USA).

We present the geometric mean as the central tendency, because the concentrations of most of the target VOCs had asymmetric distributions, with some higher values. Correlations between compounds were studied using a Principal Component Analysis (PCA). PCA was performed on the logarithmic transformed concentration of VOCs detected for more than 80% of the dwellings. After the factor extraction by PCA, we used an orthogonal Varimax rotation with a Kaiser normalization. The Wilcoxon rank sum test and the spearman rank correlation coefficient were used for comparisons of indoor and outdoor concentrations and correlation of VOCs between rooms respectively.

RESULTS

The 90 investigated dwellings were multi-family houses for 69% with 28% of owners. Smokers were present in 40% of the households. Building age varied from 1 year to more than 130 years.

The results were validated for 62 sites for VOCs and 88 sites for aldehydes. The main causes of invalidated data were broken tubes and delayed analysis caused by analytical equipment incidents. Statistical parameters describing the concentration distribution of target VOCs and aldehydes in the kitchen are presented in Table 2. The quantification limit for VOCs and aldehydes ranged from 1 to 4 $\mu g/m^3$. Corrections of concentrations with laboratory blanks were applied for aldehydes but not for VOCs.

The indoor concentrations were generally low, with 40% of values under the quantification limit. Nevertheless, the range of values was wide. The maximum indoor concentrations measured in the kitchen were 400 $\mu g/m^3$ for alpha-pinene and *o*-xylene and 293 $\mu g/m^3$ for 1,4-dichlorobenzene in the bedroom. Formaldehyde was frequently found varying form 2 to 75 $\mu g/m^3$ (geometric mean: 21 $\mu g/m^3$ in the kitchen and 23 $\mu g/m^3$ in the bedroom). The outdoor concentrations were lower than indoor concentrations: 69% of values were lower than the quantification limit. Depending on compounds, the 90th percentile outdoors varied from 1 to 9 $\mu g/m^3$. The median value of the kitchen/outdoor ratios were all equal or above 1 with values ranged from 1 (isopropyl acetate, 2-ethoxyethyl acetate, 1,1,1-trichloroethane, 2-ethoxyethanol, cyclohexane, isovaleraldehyde) to 13.3 (limonene) and 22 (hexaldehyde). Concentrations were homogeneous between kitchens and bedrooms. VOCs with spearman correlation coefficient higher than 0.90 were *m,p*-xylene, benzene, ethylbenzene, *o*-xylene, 1,4 dichlorobenzene, undecane and decane.

The concentration of most VOCs presents an asymmetric distribution with a small portion of extremely high values. This is in agreement with other studies (Hoffmann *et al.*, 2000). Formaldehyde and hexaldehyde, however, show different distributions quasi-symmetric with a higher number of dwellings exposed to high values (P90 = 43 and 42 $\mu g/m^3$ for formaldehyde and hexaldehyde, respectively).

Table 3 displays the matrix of correlations between compounds and the rotated factors for VOCs. The analysis identified seven factors that accounted for 75% of the inertia. After rotation, these factors explained 25.1, 17.2, 8.1, 6.7, 6.2, 6.0 and 5.4% of the total inertia. Factor 1 was correlated with 1,2,4-trimethylbenzene, benzene, ethylbenzene, toluene, *m,p*-xylene, *o*-xylene, decane and undecane. Factor 2 was highly correlated with the aldehydes (acetaldehyde, formaldehyde, hexaldehyde, isobutyraldehyde/butyraldehyde and valeraldehyde) and styrene. The third factor was highly correlated with alpha-pinene and limonene. Factor 4 was correlated with 1,4-dichlorobenzene and to a less extent with butyl acetate. Factor 5 was correlated with trichloroethylene. Factor 6 was correlated with 1-methoxy-2-propanol, and factor 7 with tetrachloroethylene.

Table 2 Summary statistics: concentration of VOCs and aldehydes in the kitchen (μg/m^3) and median value of the kitchen/oudoor ratio

Compounds	*N*	*N* < LQ	P10	P50	P90	Max.	GM	Kitchen/oudoor ratio
alpha-Pinene	62	4	1	4	26	400	5.1	5.3
1,4-Dichlorobenzene	62	28	1	1	33	174	2.4	1.5
1-Methoxy-2-propanol	62	22	1	2	11	48	2.0	2.1
1,1,1-Trichloroethane	62	60	1	1	1	10	0.7	1.0
1,2,4-Trimethylbenzene	62	6	1	2	9	45	2.9	2.5
2-Butoxyethanol	62	51	1	1	4	23	1.0	1.0
2-Ethoxyethanol	62	56	1	1	1	8	0.8	1.0
2-Ethoxyethyl acetate	62	61	1	1	1	1	0.7	1.0
2-Ethyl-1-hexanol	62	34	1	1	4	16	1.3	1.0
Benzene	62	4	1	2	5	13	2.1	1.4
Butyl acetate	62	20	1	2	11	36	2.0	2.4
Cyclohexane	62	54	1	1	1	3	0.8	1.0
Decane	62	1	2	8	22	85	7.4	2.6
Ethylbenzene	62	7	1	2	5	29	2.1	1.8
Isopropyl acetate	62	59	1	1	1	2	0.7	1.0
Limonene	62	1	4	12	49	112	12.9	13.3
Styrene	62	46	1	1	2	4	0.9	1.0
Toluene	62	1	6	15	36	370	15.8	3.7
Trichloroethylene	62	32	1	1	4	107	1.3	1.0
Tetrachloroethylene	62	23	1	1	4	70	1.4	1.1
Undecane	62	2	3	7	23	126	7.6	2.2
m+*p*-Xylenes	62	0	2	5	14	91	5.1	2.0
o-Xylene	62	8	1	2	5	400	2.0	1.6
Acetaldehyde	88	0	7	14	36	48	14.1	6.8
Benzaldehyde	88	67	1	1	1	2	0.8	1.0
Formaldehyde	88	0	11	22	42	60	20.8	9.0
Hexaldehyde	88	3	6	19	43	106	16.6	22.0
Iso- + *n*-Butyraldehyde	88	3	6	9	15	28	8.8	3.0
Isovaleraldehyde	88	0	2	2	2.1	5	2.2	1.0
Valeraldehyde	88	6	1	5	11.4	27	4.3	4.0

N = sample size; *N* < LQ = number of values below the quantification limit (LQ); P10, P50, P90 = percentiles; Max. = maximum; GM = geometric mean; Kitchen/outdoor = median value of the kitchen/outdoor ratio.

DISCUSSION

BTEX, alkanes, aldehydes (except benzaldehyde) and terpenes (a-pinene and limonene) are found in quite all the investigated homes. On the contrary, 1,1,1-trichloroethane, isopropyl acetate, cyclohexane and target glycol ethers (except 1-methoxy-2-propanol) are not significant in this survey. These results are in accordance with other reviews or surveys, especially for BTEX (see compared data in Wolkoff and Nielsen, 2001) and terpenes (Reitzig *et al.*, 1998). A major difference here comes from 1,1,1-trichloroethane which is reported frequently in Australian (Brown, 1999) and US studies (Holcomb and Seabrook, 1995; Girman *et al.*, 1999). However, this compound does not appear in other European studies (Bernhard *et al.*, 1995; Bornehag and Stridh, 2000).

Table 3 Principal component analysis of VOCs and aldehydes indoor concentrations (n = 62)

	Rotated factors*						
	1	2	3	4	5	6	7
alpha-Pinene			0.808				
1,4-Dichlorobenzene				0.749			
1-Methoxy-2-propanol						0.752	
124-Trimethylbenzene	0.774						
2-Ethyl-1-hexanol					–0.609		
Benzene	0.555			–0.509			
Butyl acetate				0.622		0.474	
Decane	0.729						–0.377
Ethylbenzene	0.934						
Limonene		0.449	0.734				
Styrene	0.350	0.611		–0.349			
Toluene	0.781					0.320	
Trichloroethylene					0.817		
Tetrachloroethylene							0.742
Undecane	0.717						–0.481
m,p-Xylene	0.926						
o-Xylene	0.948						
Acetaldehyde		0.614					
Formaldehyde		0.701					
Hexaldehyde		0.826					
Isobutyraldehyde/butyraldehyde		0.775					
Isovaleraldehyde		0.354	–0.488				
Valeraldehyde		0.779					

*Correlation coefficients less than 0.30 have been omitted.

Indoor and outdoor concentrations were significantly different except for those with indoor and outdoor concentrations close to the quantification limit (1µg/m^3) and having a median value of the indoor/outdoor ratios close to 1: isopropyl acetate, 2-ethoxyethyl acetate, 1,1,1-trichloroethane, 2-ethoxyethanol and cyclohexane. Except for these compounds, the concentration level found in the dwellings may thus mainly be caused by indoor sources.

PCA results identify correlations between compounds. In such analysis, each factor is a linear combination of VOC concentrations explaining sample variations. A high correlation on a same factor suggests a strong relationship between compounds and indicates a potential common source. It is clear from Table 3 that there are common sources for some VOCs. Factor 1 seems to be associated with BTEX, decane and undecane. These compounds have been meanly associated with urban traffic pollution (Edwards, 2001). The second and the third factors are correlated, respectively, with aldehydes and terpenes. Theses compounds are associated with indoor equipments and cleaning products. The others factors have to be interpreted with caution.

CONCLUSION AND IMPLICATIONS

Although the sample was not drawn randomly, the results of this pilot study gave some indications on the variability of various VOC levels in France. BTEX, alkanes, aldehydes and terpenes were ubiquitous in the investigated homes. However, this study shows differences between VOCs according to outdoor or indoor sources. Results of this study were used to assess the feasibility of a nationwide survey planed in 2003/2004.

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