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Commission of the European Communities

environment and quality of life

MEASUREMENTS OF INDOOR AIR QUALITY AND COMPARISON WITH AMBIENT AIR

A STUDY ON 15 HOMES IN NORTHERN ITALY



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A STUDY ON 15 HOMES IN NORTHERN ITALY

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This report contains the results of a survey carried out on 15 homes in Northern Italy to evaluate the occurrence of 35 selected volatile organic compounds and of total volatile organics in indoor and in ambient (outdoor) air. Measurements were carried out to determine also respirable suspended particulates and ventilation rates.

The following results were obtained:

Single pollutant concentrations were found almost always higher indoors than outdoors, frequently by an order of magnitude. Mean concentration of total volatile organics was about 3 mg/m³ indoors, compared to about 0.4 mg/m³ outdoors. The large variations from home to home observed for total volatile organic compounds in indoor air could not be correlated with the measured values of the minimum ventilation rate. A more detailed analysis by GC-MS of 6 indoor samples led to the identification of a much larger number of compounds (more than 100 in two cases). The presence of some compounds could be positively associated with building materials and household products. Most identified compounds are of the solvent type.

Questo rapporto contiene i risultati di un'indagine condotta su 15 abitazioni situate in Italia settentrionale, con la misura di 35 composti organici e del totale dei composti organici volatili nella aria simultaneamente all'interno ed all'esterno delle case. Sono state anche eseguite misure del pulviscolo (frazione respirabile) e della velocità di ricambio d'aria.

I risultati principali di questa serie di misure sono i seguenti. Le concentrazioni dei singoli composti all'interno sono quasi sempre più alte che all'esterno, spesso di un ordine di grandezza. La concentrazione media del totale dei composti organici volatili risulta 3 mg/m³ all'interno, contro 0,4 mg/m³ all'esterno. Le grandi variazioni da casa a casa, osservate per quest'ultimo parametro, non sono correlabili con i valori misurati della velocità minima di ricambio dell'aria. Un'analisi dettagliata, mediante gas-cromatografia-spettrometria di massa, effettuata su sei campioni prelevati all'interno, ha permesso di identificare un numero molto maggiore di composti (più di 100 in due casi). La presenza di alcuni composti ha potuto essere attribuita a materiali da costruzione/arredamento ed a prodotti di uso casalingo. La maggior parte dei 35 composti misurati è del tipo "solvente".

INTRODUCTION

During the last years, among those actively involved in the measurement of air pollution, the awareness has grown that the pollutant concentrations observed in the outdoor atmosphere are not representative for the indoor air, at least for many pollutants and particularly for volatile organic compounds. The fact that people spend most of their time indoors (at home or work or school) indicates that "to understand (...) health effects of air pollutants, total human exposure - including indoor as well as outdoor exposure - must be considered" [1].

with the exception of formaldehyde for the time being only few data exist on the indoor occurrence of a wide range of volatile organic compounds (VOC's) and the indoor/outdoor concentration ratios [2-6]. All these data have been collected in Northern Europe. Very recently further data on selected volatile organic compounds measured in the USA have been published [7,8]. All these measurements show that for most compounds indoor/outdoor concentration ratios greater and even considerably greater than one are found. More data and on a wider geographical basis have to be collected for an assessment of population exposures and of the relative importance of related pollution sources.

Work reported in this paper is intended to contribute to this assessment in a geographical area, Northern Italy, for which data on indoor air pollution are not yet available. Five apartments and nine detached (single family) houses have been selected for this study and include residences in urban areas (four), suburban areas (seven) and rural areas (three) and exposed to high (three), intermediate (six) and low (five) traffic density. The houses were built in part before 1973 (seven) and in part

after 1973 (seven); one of the latter has been finished less than two months before sampling. Measurements and sampling were performed in the living room, keeping, as far as practicable, the sampling devices apart from known or suspect sources. One sample was collected in an attic and another one in an administration building of the JRC. The latter was the only case with complaints about irritation by indoor air pollution.

The survey took place during 1983 and beginning of 1984

II. EXPERIMENTAL

Sampling and analysis of volatile organic compounds (VOC)

a. Carbonyl compounds

Carbonyl compounds were determined through their reaction with dinitro-phenylhydrazine (DNHP) as described in reference [9]. The only modification was the substitution of the proton donor acid: The proposed 1 N HClO4 was replaced by 10 N H3PO4 to increase the velocity of the derivatization of ketones.

DNPH was dissolved in purified acetonitrile (625 μ g DNPH/ml) and stored at -20°C. Immediately before analysis, two impingers were filled with 15 ml of the DNPH solution and 250 ul of 10 N H3PO4. An air volume of up to 50 l was sucked by a personal pump at a flow rate of 0.8 l/min through the two impingers in series. The content of each impinger was reduced to 3 ml by evaporation under vacuum and 5 μ l of the concentrated solution injected directly in a LC column. The detailed analytical conditions are the following:

- Beckman chromatograph (two solvent delivery pumps mod. 112, mod. 421 controller, Rheodyne 7410 sampling valve);
- Hewlett Packard 1040 A LC UV detection system;
- Two HPLC columns, 25 x 0.46 cm each, filled with Supelcosil RP-18 5 μ (Supelco);
- Liquid gradient starting with 50% CH3CN in water and increasing up to 90% CH3CN in 40 min.

The detection limit is 2 ppb.

b. Volatiles sampled on Tenax

The first four homes were sampled with 3 M type OVM-3500 passive samplers which were eluted with 2 ml of CS2 and the eluate used both for FID and ECD-GC analysis. These samplers were successively replaced by Tenax cartridges to increase sensitivity for the detection of the very low outdoor concentrations and also because of some unresolved difficulties connected with the use of passive samplers in low pollution level (compared to work place levels) studies, which resulted from

test experiments performed in the meantime [10].

Two samplers were exposed for periods between 4 and 7 days simultaneously indoors and outdoors; in the case of Tenax cartridges (1 g), small constant flow personal pumps were used with a total sample volume of 3 - 15 l indoors and 15 - 30 l outdoors. The sampled vapours were eluted thermally (250°C), condensed in an intermediate cryogenic trap (glasslined tube immersed in liquid N₂) and injected by flash heating of the latter. This sampling procedure is quantitative for compounds with boiling point $\geq 70^{\circ}$ C (n-hexane) when the air volume is ≤ 10 l; in the case of outdoor samples, therefore, the n-hexane concentrations may be underestimated.

The GC-FID analysis was carried out by means of a Hewlett Packard 5880 GC, under programmed temperature condition (4° C/min) from 0 to 280° C, with a 25 m, 0.3 mm id/glass column, coated with 1.1 μ m of OV-1 phase. Qualitative and quantitative results were based, besides analysis of appropriate reference mixtures, also on internal addition of four bromo-alkanes (C_3 , C_6 , C_9 and C_{12}) which were used as markers for the calculations of retention indices. Previous experience [11] had shown that this is a valuable identification tool; however GC-MS confirmation was used whenever a reasonable doubt arose. The value of "total volatile organics" was obtained expressing the total integrated FID signal in hydrocarbon equivalents.

Tenax sampling and thermal elution was also used for GC-MS analysis. In this case a split interface at the outlet of the analytical GC column divided the sample flow between a FID and the mass spectrometer. A Varian model 2700 gas chromatograph, equipped with a constant flow regulator and a 25 m persilylated, OV-1 coated (1.1 µm film thickness) glass capillary column, was linked to a Ribermag R 10-10 quadrupole mass spectrometer. A Finnigan Incos data system controlled the MS scan and data acquisition. The FID was connected to a Hewlett Packard model 3390 recording integrator. The analysis report of the integrator was after each run - transferred via a modem interface to the Incos data system.

Compound identification was based on mass spectra and retention indices, calculated by means of a method described elsewhere [11], based on internal standards and a reference mixture chromatogram.

Quantitative determinations were performed by means of the FID signal using internal standards (1-bromohexane, -nonane, -dodecane) and response factors for different compounds or compound classes. In case of not resolved GC peaks the MS signal was used to estimate the relative contributions of the different unresolved components, if their mass spectra were sufficiently different, as is the case for overlapping alkanes and alkylbenzenes. Equations and response factors used for quantification are reported in Annex I.

c. Halogenated hydrocarbons

In parallel with Tenax samplers, other samplers were run to collect organic vapours for the analysis of halogenated hydrocarbons. The samplers used for this purpose were charcoal tubes manufactured by SKC following NIOSH recommendations, containing 150 mg of adsorbant; air flow rate and total volume were similar to those adopted with Tenax sampling. However, due to insufficient number of low flow pumps, part of these samples were collected using small personal pumps with a flow rate of about 30 cm³/min; to keep the total filtered air volume within a safe limit, i.e. avoiding breakthrough for the more volatile compounds, these pumps were activated intermittently (e.g. 15 minutes each hour). The charcoal tubes were eluted with 2 ml CS, and 0.5 μ l of the solution injected in the GC through an on-column injector. The instrument used was a Carlo Erba mod. 4160, equipped with a Ni-63 ECD, a constant flow device and with a column identical to that described in the last paragraph. The temperature was programmed linearly from 35 to 280°C (4°C/ min). The detector was connected with a Hewlett Packard mod. 3390 recording integrator. Peak identification and quantitation were carried out as described for volatiles sampled on Tenax.

Measurement of respirable suspended particulate (RSP)

when visiting the homes to install samplers, RSP measurements were carried out by means of a portable instrument manufactured by TSI Inc., St. Paul, Mn., USA, and named Piezobalance mod. 3500. The instrument basically consists of an impactor which cuts off the particles larger than 2.5 µm and of an electrostatic precipitator which deposits the particles on the sensing crystal; this converts the weight into a change of its basic frequency. The sensitivity of this measurement is 0.01 mg/m³ at an integration time of 2 minutes.

The response of this instrument was compared with that of a portable nephelometer, the RAM-1 manufactured by the GCA Corp. (Bedford, Mass., USA). The comparison was made measuring with both instruments the RSP in different days in our laboratory and outdoors. Within the concentration range found (0 - 100 $\mu g/m^3$) the discrepancy was on the average 30%, with fluctuations in both directions, which can be considered acceptable at this concentration level.

3. Measurement of ventilation rate

Since the sampling of organic volatiles was carried out over 4 - 7 day periods under real life conditions, i.e. doors and windows were opened and closed in an irregular way, ventilation rates varied also throughout the sampling period. No means were, however, available to monitor ventilation rates as a function of time. Considering the important influence of ventilation on indoor air quality it was decided to measure at least the minimum ventilation rates, i.e. the ventilation rates at closed windows and doors, in all homes included in the study.

About 1 cm 3 of SF $_6$ (already diluted in 1 - 10 l of pure air) was released throughout the house and after a delay for complete mixing, air samples were taken at 20 - 30 minute intervals for 2 hours in the room where sampling for indoor pollutants was under way. SF $_6$ concentration in the samples was successively determined by GC-ECD using a molecular sieve column: the slope of a semilogarithmic plot of the concentration

decrease (due to dilution with outdoor air) versus time corresponds !mmediately to the ventilation rate, in terms of air changes/hour.

III. RESULTS

1. Indoor and outdoor concentrations of VOC

Results of the measurements of indoor and outdoor concentrations of the 35 compounds selected for this part of the study are reported for each sampling site in Table 1 and are summarized in Tables 2 and 3. As mentioned in the analytical section, reported values are 4 - 7 day averages with the exception of RSP, which is an instantaneous value, and of the carbonyl compounds formaldehyde, acetaldehyde, butanal, acetone and butanone-2, the values of which are 1 hour averages.

a. Carbonyl compounds

Three compounds were observed in all homes (see Table 1), i.e. formaldehyde, acetaldehyde and acetone, with mean indoor concentrations of 29, 16 and 40 µg/m³, respectively, and mean indoor/outdoor ratios of 4.9, 4.9 and 6.4 (see Table 2). Higher aldehydes, in particular hexanal and nonanal, and ketones (butanone-2) were observed in the majority of the homes, even at concentrations higher than those of formaldehyde. Moreover, in some homes the occurrence of propanal, hexanone and cyclohexanone was observed.

b. Hydrocarbons

The complete series of n-alkanes from C_6 to C_{13} has been measured in almost all homes (see Table 1), with mean concentrations in the range of 10 - 100 $\mu g/m^3$. There seem to be two types of compound distributions and hence presumably two distinct sources: one with prevalence of the lighter alkanes (C_6 - C_8), and the other with prevalence of the fraction between C_9 and C_{11} . The mean indoor/outdoor ratio (see Table 2) is between 5 and 12.

Benzene and the 6 alkyl derivatives selected for this study, have been measured in all the homes (with few exceptions) at levels sometimes higher than the alkanes (see Tables 1 and 2). Toluene is in general the highest in this group of compounds and the sum of meta- and para-xylene is generally second in the ranking.

A third group of miscellaneous hydrocarbons observed in this study includes limonene and a-pinene, most frequent and with highest concentrations on the average, and naphtalene which is found in about 50% of the homes at a much lower mean concentration. β -pinene has been only occasionally found.

c. Halogenated hydrocarbons

Eight compounds are listed in Table 1; four out of these have been observed in all the homes: 1.1.1.trichloroethane, carbon tetrachloride (which is below detection limit only in the JRC administration building), trichloro-ethylene, tetrachloroethylene. In addition, trichlorofluoromethane (F-11) and dichloromethane are found in some cases at rather high (the latter even at very high) concentrations, due to its use as a propellant in spray products. Also 1.4 dichlorobenzene, constituent of antiodorant and antimoth products, is found in most homes. Except dichloromethane, the mean indoor concentrations of the compounds measured range between 10 and 100 $\mu g/m^3$; the mean indoor/outdoor ratio, in the cases where both values were above the detection limit, is between 1.3 and 22.

2. Detailed GC-MS analysis of volatile organic indoor pollutants

In parallel to the indoor/outdoor measurements of selected compounds six indoor air samples have been analysed in detail by GC-MS with the scope of obtaining a more complete picture of organic indoor air pollution and also to confirm uncertain identifications in particularly complex samples encountered during the indoor/outdoor study.

Due to the applied sampling procedure (Tenax adsorption columns, thermal elution) these measurements are limited to thermally stable compounds with boiling points $\sqrt[5]{70^{\circ}}$ C (n-hexane). Moreover, very polar and in particular basic compounds tend to be not eluted as a sharp peak from the OV-1 GC column and, hence, may be not quantitatively detected or even overlooked. The results of these measurements are summarized, in Table 4. Compounds are grouped in eight classes: alkanes, olefins

and cycloalkanes, terpenes, benzene and alkylbenzenes, other aromatic hydrocarbons, chlorinated hydrocarbons, aliphatic aldehydes, and other oxygen containing compounds.

Alkanes

Concentration values are given only for normal (straight chain) alkanes and the sum of all isomers with a given carbon number. No effort has been made to identify the exact configuration of branched alkane isomers. Alkane concentrations vary within a wide range (more than two orders of magnitude). In the case of high concentrations C9 to C12 isomers are predominant, which probably originate from petroleum distillate fractions used as solvents. Whereas in general the n-alkanes are the most prominent single compounds, in space A (JRC office building) only branched alkanes were present. So long no source for these branched alkanes could be identified.

Olefins and cycloalkanes

Cyclohexanes are the most frequently occurring unsaturated hydrocarbons. Concentrations of unsaturated hydrocarbons are always lower than those of saturated ones but show similar molecular weight distributions pointing to similar sources.

Terpenes

As already evidenced in the indoor/outdoor study, alpha-pinene and limonene are frequently found in indoor air. Delta-3-carene found in space B (which is identical with home Nr. 6 in the indoor/outdoor study) is, together with alpha-pinene, a major emission product of pine trees [12] and confirms the finding that freshly introduced pinewood furniture was an important pollution source in this space (see section 6).

Benzene and alkylbenzenes

Alkylbenzenes like alkanes, occur generally in complex mixtures, which print also to petroleum distillate fractions as their origin. Different distributions have been found with maximum concentrations of

toluene, of the groups of C_2 - C_3 and of C_4 -substituted benzenes. The total concentration of alkylbenzenes may even exceed that of alkanes.

Other aromatic hydrocarbons occur only in concentrations which are an order of magnitude smaller than those of alkylbenzenes. There appears to be a correlation between these two groups of compounds which points to a common source. Naphtalene and related compounds (tetralin and methyl-naphtalenes) are the most important members of this group.

Chlorinated hydrocarbons occur consistently as indoor air pollutants. Among the more frequently found compounds are 1,2-dichloropropane, tri- and tetrachloroethylene, which are typical solvents used for cleaning purposes, and 1,4-dichlorobenzene, which has different applications in household products (see section 6).

Aliphatic aldehydes $(\geq C_5)$

Among the higher aliphatic aldehydes hexanal is the most recurrent one. Heptanal through decanal appears to be emitted together with an emission maximum for nonanal.

Other (oxygen containing) compounds

A number of further compounds which all contain oxygen has been detected, some of them only sporadically. Most of them (12 out of 19) are alcohols or alkoxy-alcohols and their acetic acid esters. Out of them 2-ethoxy-ethyl-acetate is the most frequently occurring one. Also these compounds are all used as solvents. The same is true for cyclohexanone. Cyclic siloxanes are decomposition products of silicon oils and greases of widespread use. Since they may be also released from the GC stationary phase a contribution from this source cannot be entirely excluded. Benzaldehyde may be produced by interaction of ozone and u.v. light with Tenax [13]. Since neither ozone nor u.v. light have been monitored during sampling, an artifact cannot be excluded, though both in general do not occur indoors in appreciable concentrations. On the other hand, it is not known whether NO₂ may have a similar effect.

3. Indoor and outdoor concentration of respirable particulate (RSP)

These data are less representative than those referring to organic vapours because they are quasi instantaneous values and do not indicate weekly means. The RSP concentrations observed (see Tables 1 and 2) range between 40 and 140 $\mu g/m^3$ in indoor air and between 20 and 210 outdoors. This highest value was measured in the centre of a town (home 1 - Gallarate). The only other case of outdoor concentration significantly higher than the indoor concentration is that of a home located in a rural area where the measurement was made during meadow mowing.

The effect of cigarette smoking which has been reported to be a relatively important source of indoor particulate does not appear in our sample. Although smoking is practised in 2/3 of the homes, the mean concentration of RSP is the same in homes with smokers (75 \pm 37 $\mu g/m^3$) and without smokers (78 \pm 28 $\mu g/m^3$). This fact can most probably be explained considering that RSP measurements were made (quasi instantaneously as said above) during working hours, when part of the inhabitants were absent.

4. Ventilation rates

The results of this type of measurements are reported in Tables 1 and 4. The values range between 0.16 and 1.4 air changes/hour, with a mean of 0.63. As already pointed out in the experimental section, these values are not representative of mean conditions where more or less frequent opening of windows and doors occurs, but indicate the value of minimum exchange (at the climatologic conditions of the measurements) between indoor and outdoor air, due to the so-called "le kage area", i.e. the sum of all small openings in and along the border of windows and doors, in the walls, etc.

In Table 5 the indoor-outdoor temperature difference is given for each home. These values were obtained from the mean of the continuous record indoors and from the mean of two readings outdoors (on the first

and on the last day of the sampling period). Despite this approximation and especially despite the fact that different homes are dealt with, a positive correlation appears between temperature difference and ventilation rate (see Fig. 2). The least-square line of this correlation is

$$y(h^{-1}) = 0.13 + 0.042 \times (^{\circ}C)$$

with a correlation coefficient r = 0.663.

5. Complementary characterization of sampling spaces

For each home visited, a questionnaire was filled in with the scope of collecting information which could be useful in interpreting the analytical data. This questionnaire, which is reproduced in Annex 2, was developed within a working group of scientists from different countries of the European Community, experienced in indoor air quality, who meet periodically for cooperation in this field. From the questionnaires, the following data are worth reporting here (in addition to those already given in the Introduction):

Cigarette smoking: is practised in 65% of the homes (10 out of 15).

Unvented gas appliances (for cooking): are used in all the homes (excluded the JRC administration building).

Source identification

The identification of materials or activities releasing specific compounds has not been undertaken systematically in the framework of this investigation. However, an effort was made to identify a few sources, giving rise to the occurrence in indoor air of some compounds observed in certain homes at particularly high concentrations. The attempt to identify sources was based on the one side on the information gathered through the questionnaires, in some cases complemented by ad hoc interviews, and on the other side on head-space GC and GC-MS analyses of the suspected materials or household products.

The results of this work are summarized in Table 6. Dichloromethane which was observed in two homes at concentrations of 3 and 5 mg/m 3 was positively identified as the major constituent (> 90%, propellant) of a hair spray which was daily used in the homes. The compound is a substitute for traditional propellants (fluorotrichloromethane or F-11 and difluorodichloromethane or F-12), which are being abandoned because of their suspected effects on the stratospheric ozone layer. The presence of 1.4-dichlorobenzene was found associated with anti-moth tablets and the same is true for naphtalene.

The high value of alpha-pinene in home 6 (Table 1), respectively in space B (Table 4) and in home 13 (Table 1) was due to new furniture and wall panels made of conifer wood. Head space analyses of different wood samples showed that alpha-pinene may account for up to 80% of the emitted volatile organic compounds. Another contribution to alpha-pinene in indoor air originates from certain products for the preservation and polishing of wooden furniture. Delta-3-carene, also found in space B (Table 4) is a major emission product of living pine tree [12] and was probably emitted by a new pine wood table. Limonene is emitted only to a minor degree by most conifer trees. An important source of this terpene compounds, which was frequently detected at concentrations up to 0.5 mg/m^3 , are kitchen detergents with addition of lemon flavour, as strongly advertised on the product labels. Head-space sampling on Tenax and GC analysis of such detergents indicated that limonene may represent from 50 to 80% of the volatile organic compounds released by these materials.

Complex mixtures of Cg to C12 alkanes and C2 to C5 alkylbenzenes are emitted by waxes and polishes for floor and furn; ture treatment. They probably originate from petroleum distillate fractions used as solvents in these products.

IV. DISCUSSION

The main results of this survey have been synthesized in Table 7 and in Figures 3 to 5, aggregating the data into compound classes. In Table 7 rough estimates of population and traffic density at the different sampling sites are also reported. No clear correlation appears between these parameters and the outdoor concentrations of the various compound classes. However, this finding is not surprising if the following facts are considered:

- a) the home sample is small;
- b) no large urban area is represented, Varese and Gallarate having less than 100 thousand inhabitants. Even some of the samples collected in these two centers refer to homes located in the suburbs (Nr. 10 and 14) or at the 9th floor of one of the rare high rise buildings (Nr.3);
- c) the samples have been collected in different seasons, so that the effects of winter heating and of meteorological dispersion contribute to the variability of outdoor concentrations.

The most important finding of this study is that indoor concentrations of a wide range of volatile organic compounds are consistently higher or even much higher than outdoor concentrations. This is a clear indication of the presence of important indoor sources for these pollutants.

It is interesting to look at the nature of these IAP sources. For the time being, mostly two groups of sources have been taken into consideration: building materials and combustion processes including smoking.

Historically, formaldehyde, a pollutant originating from building materials, has first raised the issue of indoor air quality in Northern Europe. Formaldehyde has been emitted in acutely disturbing quantities from particle board and from urea-formaldehyde fome, widely used in the early 70's in energy efficient buildings in these countries. As a consequence, attention was focused on building materials as major sources of indoor air pollution, which - as a further consequence - was considered essentially a problem of new buildings or of buildings refitted

in an energy efficient way.

In the second half of the 70's a new concern was raised about combustion processes as important indoor sources of $\rm NO_2$ and particles. This concern originated in the U.S.A., where many laboratories had been involved in outdoor air monitoring of these criteria pollutants and then, in part, switched to indoor air monitoring.

Looking at the results of the present study focused on volatile organic compounds, a third group of sources appears to be of importance: consumer products. This group of sources is characterized by two major features:

- they emit mainly organic pollutants;
- they occur in all types of buildings, old as well as new ones.

The first feature is the major reason why until now little attention has been paid to these sources: analysis of organic pollutants is complex and expensive. Results of this study show that most of the organic indoor pollutants can be grouped under the broad denomination of "solvent type compounds", which includes alkanes, benzene and its alkyl derivatives, halogenated hydrocarbons, alcohols, alkoxy-alcohols and their acetic acid esters, and ketones. Some of their major sources are paints, wood impregnants, glues, cleaning agents, liquid waxes and polishes.

All these sources are consumer products of large use. In fact, paints, wood impregnants and glues might also be classified as building materials. We prefer, however, their classification as consumer products since

- they are widely used in old buildings;
- they are also used for "do it yourself" and hobby activities, and we prefer to limit in this context the term building materials to products used for new constructions or basic modifications of buildings only, but not for ordinary maintenance or renovation.

Building materials according to this definition differ much more in different regions, e.g. in Northern and Southern Europe, than consumer products, which in general are distributed all over the Common Market. Consequently, indoor air pollution by emissions of consumer products can be expected to show less variation between different regions than that by emission of building materials. This is confirmed by two findings of the present study:

- a) The measured indoor levels of formaldehyde (see Table 1) are all well below 120 µg/m³, a concentration which is considered the limit for acute effects (irritation), but which has often been exceeded in Northern Europe. In fact, particle board and urea formaldehyde foam, the major sources of high formaldehyde levels, are not used to an appreciable extent as building materials in Northern Italy. Anyway, formaldehyde concentrations measured in this study are indoors still consistently higher than outdoors, with a mean indoor/outdoor ratio of 4.9 (see Table 3), indicating the presence of indoor sources. However, it was not yet possible to identify all related sources. In some cases combustion processes (gas appliances) made some contribution but did not account for a major portion of the detected concentrations.
- b) All other volatile organic compounds reported as indoor pollutants in the four studies published until now [2-6] and performed in Northern Europe have also been detected in this study at comparable or higher concentrations. In addition to solvent constituents, limonene and 1,4-dichlorobenzene had been reported earlier. Limonene has been shown in this study to originate mainly from detergents to which it is added as odorant. 1,4-dichlorobenzene is used in anti-moth products and in deodorant products. All these sources are typical consumer products.

Further sources identified in this study and reported in Table 6 are also consumer products with the exception of conifer wood, which has been used for furnishing and wall panels.

In view of the predominant use of classical materials like bricks and concrete for residential constructions in Northern Italy, no major

contribution of building materials in the above defined sense may be expected. Since building materials will make the highest contribution to IAP in new houses, the only new house (Nr. 13 in Table 1) included in this study was sampled twice, immediately after the occupants moved in and two months later. However, besides benzene, alkylbenzenes and alkanes, which are typical solvent compounds contained in paints and wood impregnants, only terpenes showed a remarkable concentration decrease. This points to the wood of conifers as source which had been used in the construction of windows and doors, and to cover the ceilings of two rooms. The concentrations of other compounds like most halocarbons and hexanal, increased however, pointing clearly to consumer products as sources. It should be mentioned that, anyway, only the concentration of alpha pinene exceeds the range of concentrations measured in the older buildings.

In Fig. 6 the values of total volatile organic compounds (TVOC) and of respirable suspended particulates (RSP) have been plotted for each home versus the minimum air exchange rates measured with the SF6 dilution technique. The lack of correlation which appears looking at the figure was also confirmed by calculations: in the case of TVOC a correlation equation with a negative slope is obtained (i.e. decrease of the concentration increasing the exchange rate), but the correlation coefficient is very small (r = 0.164). This finding suggests that for TVOC the influence of source variability and of human behaviour (e.g. opening of windows and doors) on indoor pollution prevails over that of the minimum air exchange rates due to infiltration. In the case of RSP the correlation is higher (r = 0.514), and the equation obtained has a positive slope (i.e. increase of the concentration increasing the exchange rate). The latter fact can be interpreted in the sense that outdoor sources are more important for RSP than for TVOC in determining indoor concentrations; this appears also from the indoor/outdoor ratios (see Table 3) which are much smaller for RSP than for TVOC.

A particularly interesting case is that of home Nr. 15, located in the centre of Varese at the 3rd floor, along a street with heavy traffic:

the highest outdoor concentration observed in this study, the indoor concentration was much higher, even for compounds typical for car exhaust. Since the outdoor sample had been taken on a balcony looking out on a yard, whereas the indoor sample had been collected in a room on the street side, the former might have been less influenced by the traffic than the latter. In order to test this hypothesis new measurements of volatile organic compounds (using Tenax sampling) were performed in September 1984. In contrast to the previous measurements performed in January, however, two indoor and two outdoor samples were taken over 50 hr periods, one of each type at the front (street) side and one at the rear (yard) side of the house. The analytical results of these samples are presented in Table 8, together with the results obtained in January. Some evident features are:

- the street concentrations are 2-3 times higher than the yard concentrations, except for heavier alkanes which are at the same level;
- the two indoor samples, substantially identical, show concentrations intermediate between the two outdoor samples, except for alkanes heavier than octane and for terpenes, which are higher than both outdoor samples.

If the data of September are compared with those of January, it appears that in the September samples the concentrations both indoors and outdoors (considering only the yard values) were much lower than in January: if the concentrations of total volatile organic compounds are compared the ratio January/September is 7 indoors and 4 outdoors. But if we examine the single compounds we find indoor ratios between 10 and 15 for some of them (even 76 for limonene), whereas the outdoor ratios fluctuate between roughly 3 and 5.

This complex pattern can be partly unravelled considering the following aspects. During the September sampling the windows remained open part of the time, but this, of course, was not the case in January. A liquid floor wax is applied about once per month: this probably happened shortly before the January sampling and certainly not in September.

The two facts can explain the difference between the indoor concentrations and the particularly large difference observed for the heavier alkanes and terpenes, typical wax constituents. The contribution of heating to outdoor pollution in January and a probably smaller dilution of car exhaust gases at the same time could explain the large difference in outdoor concentrations between January and September.

V. SOME COMMENTS ON POTENTIAL HEALTH IMPLICATIONS

For the time being nearly no data are available which would allow an assessment of the findings of this study with respect to potential health implications. In principle, three major effects are conceivable:

- acute effects, in particular eye and airway irritation;
- chronic intoxication;
- induction or promotion of genetic damage, in particular of cancer.

Acute effects

For none of the detected compounds acute effects are known at the concentration levels reported here, though many of them may act as irritants at higher concentrations (e.g. aldehydes, terpenes). However, recent human exposure studies on the irritation by typical indoor mixtures of organic pollutants indicate that such mixtures may influence performance of susceptible individuals already at levels of total concentration as low as 5 mg/m³ [14], a level which has been exceeded in five of the 15 houses studied (see Table 1). It is, however, not yet clear how far the composition of the mixture influences the irritation potential.

Chronic intoxication

The only data which may give hints to potential chronic effects of the compounds detected in indoor air are occupational exposure limits. The highest indoor concentrations observed in this study are at least 1000 times lower than the Occupational Exposure Limits for all the compounds, except formaldehyde, dichloromethane and benzene, for which the maximum concentrations are roughly 100 times lower than the corresponding limits. However, these limit values have been established:

- for healthy adults, whereas the population exposed to indoor air pollution includes children, pregnant women, aged and ill persons;
- for an exposure time of 40-45 hours per week, or about 25% of the total time, whereas recent studies on the time budget of the population in industrialized countries have shown that the time spent indoors is between 21 and 23 hours/day or about 90% of the total [15];

- taking into account only effects of each compound on its own, whereas indoor environments are characterized by the presence of complex mixtures of many pollutants. An appropriate exposure evaluation in these cases would require not only the addition of all the single contributions, but also a careful investigation of the possible synergistic effects, i.e. of the mechanisms through which the adverse effect of one compound is enhanced by the simultaneous occurrence of others.

For all these reasons, Occupational Exposure Limits are not an adequate reference for the evaluation of the risks due to indoor pollution.

Cancer induction or promotion

Some of the compounds observed most frequently and at higher concentrations in indoor air have been demonstrated to be carcinogenic in animals and one, benzene, also in man. Table 9 summarizes some information on this effect and on mutagenicity, which is considered a sort of index of potential carcinogenic properties of a compound.

VI. CONCLUSIONS

The work described in this report on indoor air pollution by volatile organic compounds leads to the following conclusions:

- a) Indoor concentrations are always higher and frequently much higher than outdoor concentrations.
- b) Most detected compounds are of the solvent type (released from paints, wood impregnants, glues, cleaning agents, liquid waxes and polishes, hobby products). Others are deodorants or disinfectants.
- c) Besides building materials and combustion processes consumer products are an important source of volatile organic compounds in indoor environments. In fact, in the cases included in this study, consumer products (including paints, wood impregnants and glues) appear to be the most important source.
- d) Pollutant concentrations essentially do not correlate with minimum ventilation rates; this suggests that for volatile organic compounds, the influence of source variability and of human behaviour on indoor pollution is dominating. Consequently, any effort to reduce indoor air pollution by VOC's should concentrate on the control of their sources rather than of minimum ventilation rates.

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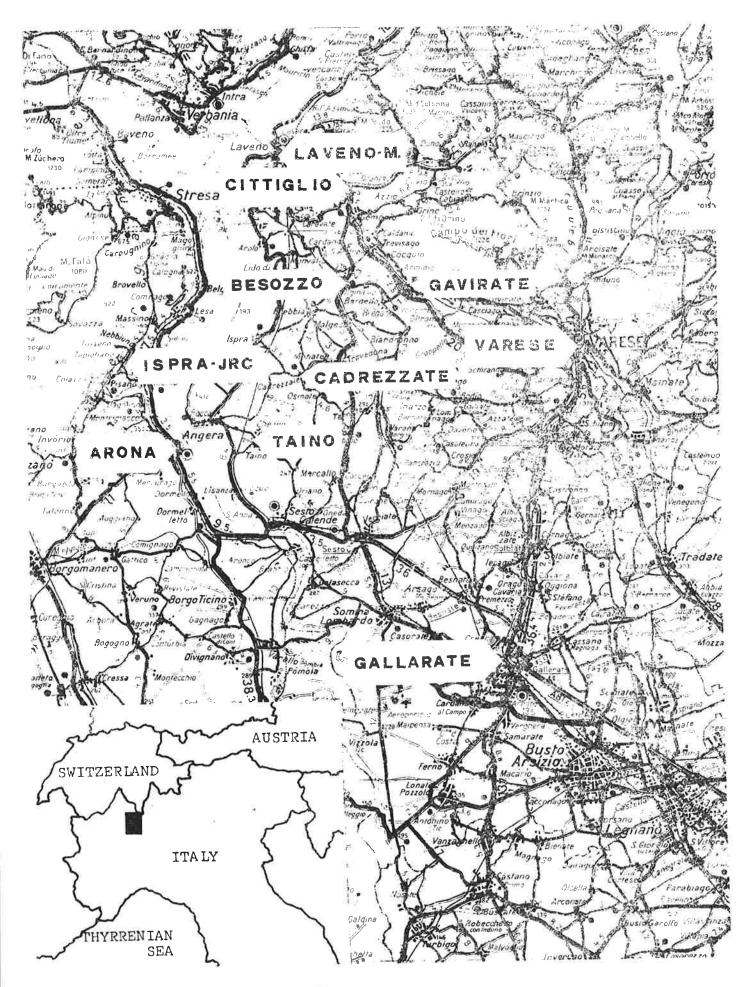


Fig. 1 - Map of the area where this study has been performed (1 cm = 2 km)

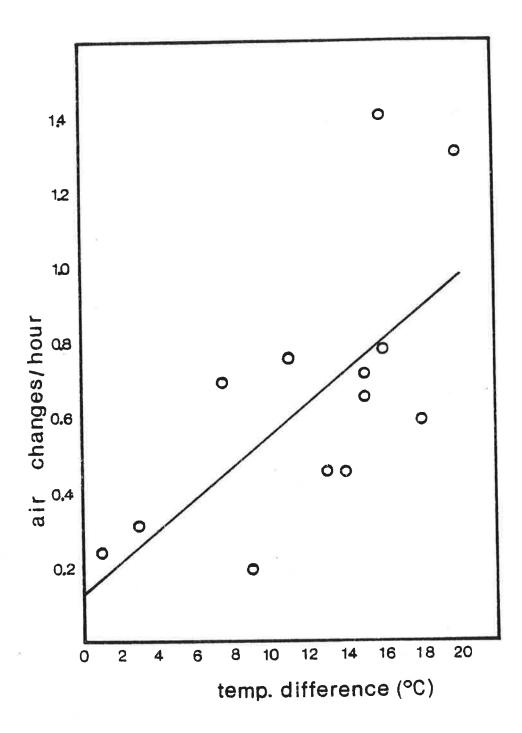
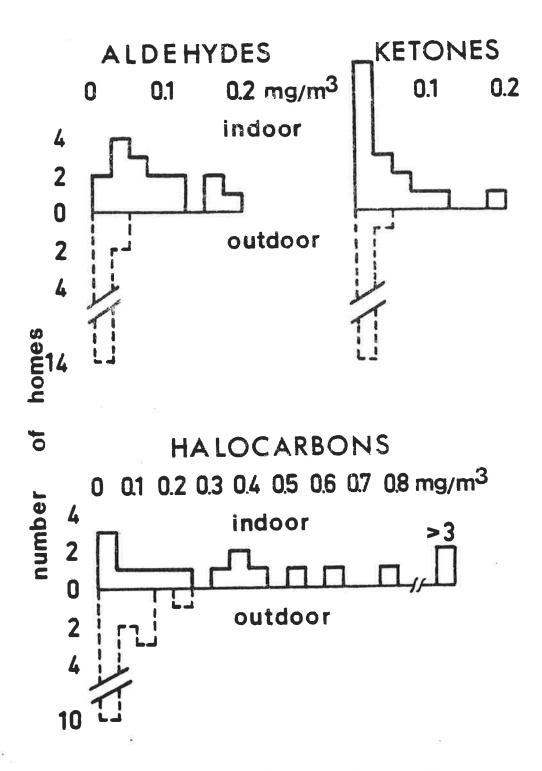
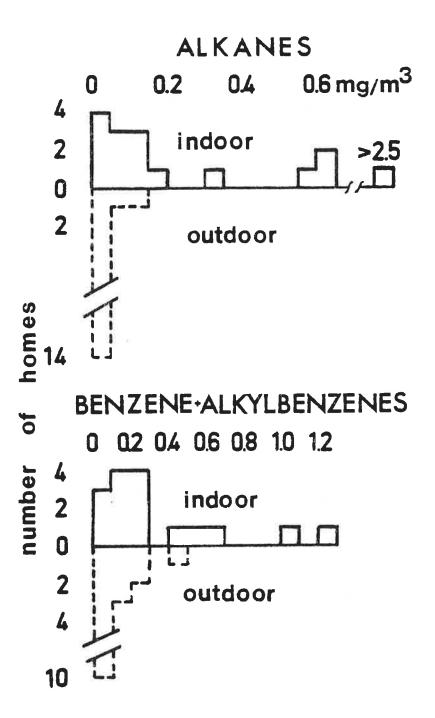


Fig. 2 - Correlation between ventilation rate (air changes/hour) and indoor-outdoor temperature difference (°C)





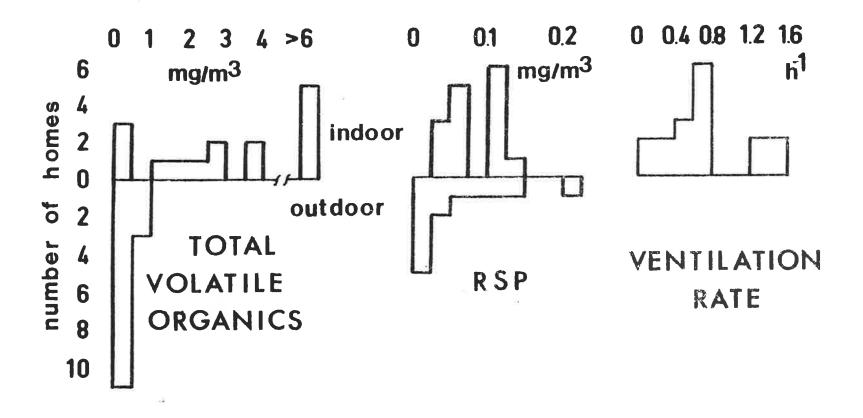


Fig. 5 - Indoor and outdoor distribution of total volatile organics and respirable suspended particules (RSP).

Distribution of ventilation rate

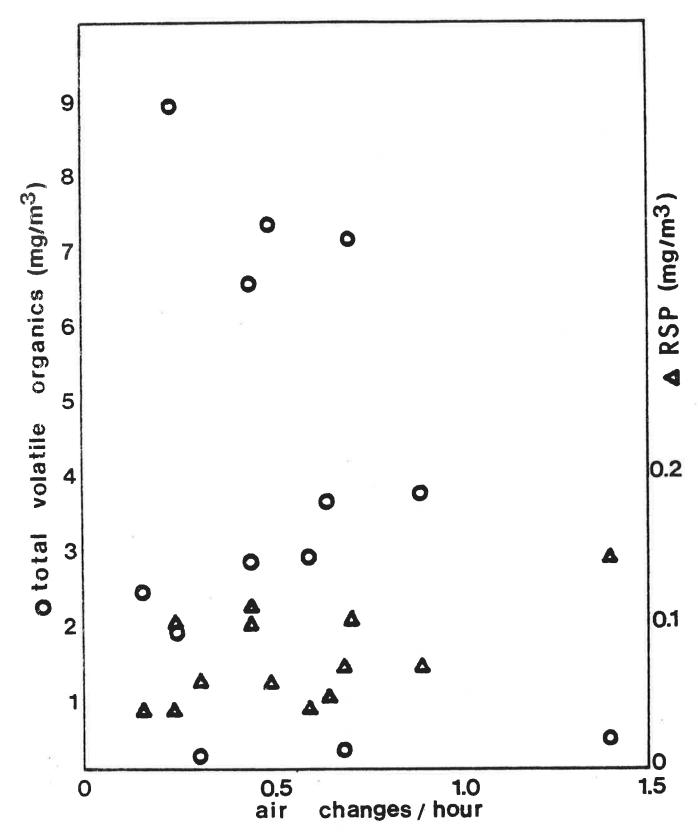


Fig. 6 - Indoor concentrations of total volatile organics and RSP vs ventilation rate, showing absence of correlation

TABLE 1 - Results of indoor-outdoor measurements $(\mu g/m^3)$ (for each home the first column refers to indoor air and the second column to outdoor air)

home number home type location (month of sampling)	_	tment arate (5)		ched house	-	tment se (3)	4 apartment Arona (6)		
air exchange rate (h ⁻¹)	1.3		1.	4	0.69		0.31		
respirable susp. part.	110	210	140	-	70	-	60	20	
total volatile organicsa	960	400	320	220	240	180	190	180	
formaldehyde acetaldehyde butanal hexanal nonanal	52 21 < 1 28 82	3 2 < 1 2 9	20 22 < 1 < 2 < 2	8 4 < 1 < 2 < 2	9 2 < 1 < 2 17	2 4 < 1 < 2 < 2	27 9 7 5 6	10 19 10 < 2 < 2	
acetone butanone-2	16 5	8 2	55 4	< 2 < 2	3 < 2	< 2 < 2	14 < 2	7 < 2	
trichlorofluoromethane dichloromethane chloroform 1.1.1. trichloroethane carbontetrachloride trichloroethylene tetrachloroethylene 1.4-dichlorobenzene	19 390 < 1 20 4 14 33 23	8 10 < 1 3 < 1 1 4 2	- <10 < 1 6 3 1 8 <10	- <10 < 1 2 2 1 4 <10	- <10 < 1 6 2 8 12 <10	<pre><10 < 1 7 3 8 16 <10</pre>	- <10 < 1 4 4 4 9 200	- <10 < 1 4 5 5 5 5 <10	
n-hexane n-heptane n-octane n-nonane n-decane n-undecane n-dodecane n-tridecane	9 3 2 4 4 5 3	10 4 3 2 3 3 2 1	30 12 14 3 5 2 5 < 1	25 3 < 1 < 1 2 < 1 < 1 < 1	14 8 2 < 1 3 < 1 < 1	17 * 8 1 < 1 2 < 1 < 1 < 1	14 11 < 1 1 2 1 < 1 < 1	20 10 < 1 < 1 < 1 < 1 < 1 < 1	
ben zene toluene ethylbenzene 1.3-xylene + 1.4-xylene 1.2-xylene 1.3.5-trimethylbenzene 1.2.4-trimethylbenzene	9 55 5 36 17 4 10	13 47 7 23 11 4	11 82 < 2 14 6 < 1	7 14 < 2 4 2 -	16 36 < 2 29 13 < 2	14 25 < 2 20 7 < 2	17 38 6 17 10 2	13 31 4 11 4 < 1 < 1	
α-pinene β-pinene limonene naphtalene	4 < 1 300 9	< 1 < 1 < 1 < 1 < 1 < 1 < 1	- - 30 -	- - < 1	- - 95	- - 11 -	- 13 1	- < 1 < 1	

a: hydrocarbon equivalent (FID response) of volatile organics trapped on Tenex (i.e. with boiling point $\stackrel{>}{\sim}~70^{\rm o}{\rm C})$ b: with air conditioning

c: branched alkanes

home number home type location (month of sampling)	5 office JRC-Isp	ora (5)				ched house o (10)		ched house a (10)
air exchange rate (h-1)	0.69 (1	.1) ^b	0.2	24	0.4	15	0.	16
respirable susp. part.	130 1	.0	40	20	110	70	40	30
total volatile organics ^a	9400 21	10	7100	200	5200	340	1900	180
formaldehyde acetaldehyde butanal hexanal nonanal	35 1 29 < 1 < 6 < 2	_	50 1 34 53 < 2	5 < 1 6 < 2 < 2	31 9 < 1 5 19	3 1 < 1 < 2 < 2	41 48 < 1 < 2 < 2	4 2 < 1 < 2 < 2
acetone butanone-2	9 < 2 <	3 2	24 < 2	11 < 2	5 6	< 2 12	42 30	6 4
trichlorofluoromethane dichloromethane chloroform 1.1.1. trichloroethane carbontetrachloride trichloroethylene tetrachloroethylene 1.4-dichlorobenzene	1 < < 10 < 1 < < 1 < < 1 < < 1 < < 1 < < 1 < < 1 < < 1 < < 1 < < 1 < < 5 < < 5 < < 5 < < 5 <	1 O 1 1 1	- <10 1 4 11 3 3 130	- <10 < 1 5 4 3 3 <10	230 5000 15 60 10 12 20 67	4 <10 3 25 8 6 8 < 2	12 360 < 1 7 7 7 10 < 10	6 < 5 < 1
n-hexane n-heptane n-octane n-nonane n-decane n-undecane n-dodecane n-tridecane	200 ^C 340 ^C 104 ^C 4 <		45 3 6 165 1100 950 220 20	25 < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5 < 5 <	590 15 3 12 5 9 1 < 1	20 7 3 4 4 2 < 1 < 1	10 3 2 4 4 3 3 < 1	4 1 < 1 2 2 1 < 1 < 1
benzene toluene ethylbenzene 1.3-xylene + 1.4-xylene 1.2-xylene 1.3.5-trimethylbenzene 1.2.4-trimethylbenzene	6 8	3 3 1 3 1 1 2	67 150 33 120 28	11 24 .9 8 < 5	63 120 7 17 11 3 7	46 56 6 19 < 1 3	23 93 8 22 4 5	11 16 3 7 3 2 3
α-pinene β-pinene limonene naphtalene	< 1 < 170	1 1 3 1	350 - 45 -	< 5 - < 5 -	23 9 40 4	12 2 1 1	47 12 43 < 1	2 1 < 1 < 1

Table 1 - cont.

home number home type location (month of samplin	9 detached hou g)Varese (10)			12 use detached house 11 Gavirate (12)
air exchange rate (h^{-1})	0.19	0.75	0.45	0.65
respirable susp. part.	100 140	70 –	100 100	50 30
total volatile organicsa	1500 260	3000 140	2200 100	2900 690
formaldehyde acetaldehyde butanal hexanal nonanal	26 10 22 7 < 1 < 1 < 2 < 2 < 2 < 2	15 8 7 4 < 1 < 1 14 < 2 < 2 < 2	17 4 7 2 < 1 < 1 < 2 < 2 < 2 < 2	25 12 37 6 < 1 1 12 < 2 27 < 2
acetone butanone-2	23 8 10 6	30 5 < 2 5	8 2 < 2 < 2	77 8 11 9
trichlorofluoromethane dichloromethane chloroform 1.1.1.trichloroethane carbontetrachloride trichloroethylene tetrachloroethylene 1.4-dichlorobenzene	21 2 290 < 5 2 1 56 7 6 5 20 11 13 7 230 < 2	26 5 240 < 5 1 < 1 9 6 9 6 10 4 34 8 < 5 < 2	13 6 < 5 < 5 8 < 1 12 11 5 10 7 8 8 13 < 5 < 2	41 4 3000 5 2 1 42 5 8 4 86 5 31 7 50 < 2
n-hexane n-heptane n-octane n-nonane n-decane n-undecane n-dodecane n-tridecane	13 5 4 2 34 1 11 1 24 1 16 1 3 1 < 1 < 1	100 2 40 1 21 < 1 12 1 10 1 6 < 1 3 < 1 < 1 < 1	57 2 21 8 12 < 1 7 < 1 5 < 1 5 < 1 2 < 1 < 1 < 1	4 14 7 6 15 6 16 5 27 6 15 4 5 2 4 1
benzene toluene ethylbenzene 1.3-xylene + 1.4-xylene 1.2-xylene 1.3.5-trimethylbenzene 1.2.4-trimethylbenzene	19 11 61 23 10 6 32 14 10 9 8 3 18 6	90 5 200 8 43 2 120 3 40 1 22 < 1 51 1	106 8 230 10 53 2 140 6 49 2 12 < 1 38 2	35 30 82 70 14 14 46 51 13 19 6 10 28 22
α -pinene β -pinene limonene naphtalene	21 1 3 - 10 1 16 1	14 1 10 9 4 < 1	39 < 1 2 - 176 < 1 4 < 1	20 < 1 6 - 187 1 8 4

Table 1 - cont.

home number	13	13	14	15
home type	detached house	detached house	apartment	apartment
location (month of sampling)	Besozzo (12)	Besozzo (2)	Varese (1)	Varese (1)
air exchange rate (h ⁻¹)	0.78	0.66	0.59	0.71
respirable susp. part.	50 20	70 80	40 20	100 80
total volatile organics ^a	5800 340	2700 290	2300 650	5700 1500
formaldehyde	29 5	26 14	8 5	16 9
acetaldehyde	18 5	10 9	6 3	18 5
butanal	<1 < 1	<1 < 1	< 1 < 1	< 1 < 1
hexanal	20 < 2	58 < 2	< 2 < 2	10 < 2
nonanal	20 < 2	22 < 2	< 2 < 2	10 < 2
acetone	157 6	15 5	21 16	98 12
butanone-2	38 8	2 1	4 11	10 < 2
trichlorofluoromethane dichloromethane chloroform 1.1.1. trichloroethane carbontetrachloride trichloroethylene tetrachloroethylene 1.4-dichlorobenzene	20 19 < 5 < 5 < 1 < 1 22 23 12 20 42 24 25 38 24 < 2	43 38 < 5 < 5 < 1 < 1 125 31 12 29 112 11 64 32 10 < 2	16 16 225 < 5 < 1 < 1 34 35 2 22 21 18 10 38 73 2	29 39 590 100 < 1 < 1 21 21 11 11 26 14 47 48 28 4
n-hexane n-heptane n-octane n-nonane n-decane n-undecane n-dodecane n-tridecane	48 8	9 2	12 17	114 42
	30 3	7 2	5 7	76 17
	21 2	29 3	4 3	65 15
	84 2	21 2	16 3	67 9
	94 3	25 3	26 3	72 10
	35 2	20 2	14 3	134 5
	7 1	6 1	3 1	35 5
	3 < 1	4 < 1	< 1 < 1	15 2
henzene toluene ethylbenzene 1.3-xylene + 1.4-xylene 1.2-xylene 1.3.5-trimethylbenzene 1.2.4-trimethylbenzene	81 26	39 16	31 35	204 67
	300 40	165 25	72 80	378 156
	109 7	41 5	19 17	96 34
	390 22	141 14	62 59	280 111
	108 7	39 5	23 22	132 42
	38 3	14 4	14 9	59 16
	76 8	38 7	36 27	150 44
α-pinene	605 < 1	322 < 1	71 < 1	29 3
β-pinene	104 < 1	60 < 1	8 < 1	29 < 1
limonene	240 < 1	57 < 1	480 < 1	268 4
naphtalene	11 3	10 2	5 4	70 11

TABLE 2 - Minima, Maxima and Means of Indoor and Outdoor Concentrations

		Pollutar	t concen	trations	$[\mu g/m^3]$	
		value outdoor	highes indoor	t value outdcor		value ^a outdoor
RSP	40	10	140	210	81	63
TVO ^b	190	100	9400	1500	3250	370
Formaldehyde	8	2	52	12	27	6.6
Acetaldehyde	1	< 1	48	19	17	4.5
Butanal	< 1	< 1	34	10	2.7	< 2.0
Hexanal	< 2	< 2	53	2	10	< 2.0
Nonanal	< 2	< 2	82	< 2	12	< 2.0
Acetone	3	< 2	157	16	39	6.1
Butanone-2	< 2	< 2	38	12	7.9	3.8
Trichlorofluoromethane (F-11) 12	< 1	230	39	39	10
Dichloromethane	< 5	< 5	5000	100	670	< 14
Chloroform	< 1	< 1	15	3	1.9	< 1
1.1.1-Trichloroethane	4	< 1	60	35	21	11
Carbon tetrachloride	< 1	< 1	12	22	6.3	7.0
Trichloroethylene	1	< 1	86	24	18	7.5
Tetrachloroethylene	3	1	47	48	18	14
1.4-Dichlorobenzene	< 5	< 2	230	4	55	< 5
n-Hexane	3	2	590	4.2	71	14
n-Heptane	1	< 1	76	17	16	5.1
n-Octane	< 1	< 1	65	15	14	2.4
n-Nonane	< 1	< 1	165	9	27	2.1
n-Decane	2	< 1	1100	10	92	3.1
n-Undecane	< 1	< 1	950	5	80	< 2.0
n-Dodecane	< 1	< 1	220	5	20	< 1.7
n-Tridecane	< 1	< 1	20	2	3.1	< 1.3
Benzene	5	3	204	67	52	20
Toluene	17	3	378	156	128	40
Ethylbenzene	< 2	< 1	109	34	27	7.4
1.3-Xylene + 1.4-Xylene	6	3	390	111	89	24
1.2-Xylene	4	< 1	132	42	26	8.7
1.3.5-Trimethylbenzene	< 1	< 1	59	16	13	3.8
1.2.4-Trimethylbenzene	< 1	< 1	150	44	36	11
α-Pinene	< 1	< 1	605	3	102	< 1.6
β-Pinene	< 1	< 1	104	2	17	< 1.1
Limonene	10	< 1	480	11	140	2
Naphthalene	< 1	< 1	70	11	11	2

For the calculation of means all values below the detection limit have been assumed to be 'zero', unless the resulting mean value was smaller than two times the detection limit. In this last case an upper estimate for the mean value has been given using the detection limits as measured values.

^{&#}x27;Total Volatile Organics', i.e. total FID integrator signal expressed as hydrocarbon equivalents.

TABLE 3 - Minima, Maxima, Means and Medians of the Indoor/Outdoor ratios

ACTION AND AND AND AND AND AND AND AND AND AN	lowest	highest	meanb	medi	anb
RSP	0.5	13	2.6 (12)	1.6	(12)
TVO ^a	1.1	45	13 (15)	5.8	(15)
Formaldehyde	1.6	17	5.3 (15)		(15)
Acetaldehyde	0.5	24	6.0 (14)	3.6	(15)
Butanal	0.7	5.7	С	C	(0.)
Hexanal	>2.5	>26	C	>6	(9)
Nonanal	<1	>13	С	>8.8	(8)
Acetone	1.3	>28	6.2 (12)	3	(15)
Butanone-2	0.4	7.5	2.7 (7)	>1.8	(10)
Trichlorofluoromethane (F-11	0.7	58	9.3 (10)	2	(11)
Dichloromethane	5 , 9	>600	C	>53	(8)
Chloroform	>1	5	С	>2	(6)
1.1.1-Trichloroethane	0.8	8.4	2.7 (14)	1.4	(15)
Carbon tetrachloride	0.1	>4	1.3 (13)	1.4	(14)
Trichloroethylene	0.8	17	3.4 (14)	1.8	(15)
Tetrachloroethylene	0.3	8.3	2.2 (15)	1.8	(15)
1.4-Dichlorobenzene	7	>115	19 (3)	>20	(9)
n-Hexane	0.3	50	8.7 (15)	1.8	(15)
n-Heptane	0.7	40	6.5 (13)	2.0	(15)
n-Octane	1	34	6.9 (8)	2.5	(13)
n-Nonane	1	42	9.7 (9)	5.3	
n-Decane	0.7	>220	7.9 (12)	>4.7	(14)
n-Undecane	>1	>190	9.8 (8)	4.7	(13)
n-Dodecane	>1	>44	4.3 (6)	>3	(13)
n-Tridecane	>1	7.5	4.8 (3)	>3.5	(6)
Benzene	0.7	18	3.8 (15)	1.7	(15)
Toluene	0.9	25	6.1 (15)	2.6	(15)
Ethylbenzene	0.7	27	6.8 (12)	>2	(13)
1.3-Xylene + 1.4-Xylene	0.9	40	7.8 (15)	2.3	(15)
1.2-Xylene	0.7	40	8.0 (13)	3.0	(15)
1.3.5-Trimethylbenzene	0.6	>22	3.3 (8)	2.6	(12)
1.2.4-Trimethylbenzene	<0.5	51	9.5 (10)	3.2	(12)
α-Pinene	>4	>605	17 (4)	23	(10)
β-Pinene	4.5	>104	C	12	- (5)
Limonene	1.1	>480	53 (7)	>43	ं (15)
Naphthalene	>1	16	5.6 (6)	4	(11)

a see footnote on Table 1; b in parentheses number of values a see footnote on Table 1,

c excessively small number of defined values

	Det	ected cond	entrations	(μg/m ³):	in space	
Identified compounds or compound classes	A	В	С	D	Е	F
Total identified organics	14,000	8,800	3,400	2,000	510	570
Total non identified (alkane equivalent)	390	1	150	560	100	32
(Number of identified compounds)	(63)	(105)	(118)	(47)	(68)	(66)
Σ n-Alkanes and isomers (C ₆ - C ₁₆) n-Hexane n Heptane Σ Heptane isomers n-Octane Σ Octane isomers n-Nonane Σ Nonane isomers n-Decane Σ Decane isomers n-Undecane Σ Undecane isomers n-Dodecane Σ Dodecane isomers n-Tridecane n-Tetradecane	13,000 13 - - - 325 8,190 - 3,020 1,590 - 14	3,900 3 3 6 6 165 200 1,100 1,450 950 1,720 220 530 20 2	920 7 22 53 43 120 70 155 83 194 150 250 36 66 14 22	110 - 3 3 - - 14 60 60 13 13 19 19	130 - - - 1 1 40 70 15 36 5 16 - 2 2	34 4 - 12 3 7 1 7 3 4 - - - -
n-Pentadecane n-Hexadecane	-	-	22 13			.=.
Σ Olefins and cycloalkanes Methylcyclopentane Methylcyclohexane Ethylcyclohexane cis-1-Ethyl-3-methylcyclohexane n-Propylcyclohexane Σ Nonene isomers n-Butylcyclohexane Σ Decene isomers Σ Undecane isomers	- - - - - - -	650 - 8 25 16 40 145 330 270	90 - 25 9 12 9 38 - 4	- - - - - - -	27 - - 2 2 10 - 16 1	12 1 1
Σ Terpenes (Mol.W. = 136) alpha-Pinene delta-3-Carene beta-Pinene Limonene	- - - -	680 400 210 - 65	360 13 - 17 325	16 7 - - 9	5 	9 2 - - 7

Identified compounds or compound classes	Detected concentrations (µg/m³) in space								
*	А	В	С	D	E	F			
Benzene and alkylbenzenes	140	2,900	1,500	510	20	110			
Benzene	105	3	38	117	5	11			
Toluene	30	1 25	215	270	6	42			
Ethyl-benzene	5	25	90	21	1.	7			
m-,p-Xylene		7 5	255	85	4	22			
0-Xylene	trace	21	120	10	2	8			
C C2-Alkylbenzenes	5	120	465	116	7	37			
Isopropylbenzene (cumene)		5	16	3	12	-			
n-Propylbenzene	_	24	42	-	5 4	2			
1-Ethyl-3methyl-and-4methylbenzene	_	105	150	6	1	6			
1.3.5-Trimethylbenzene	-	140	46	-	< 1	2			
1-Ethyl-2-methylbenzene	-	160	45	-	_	3			
1.2.4-Trimethylbenzene	_	245	150	-	1	7			
1.2.3-Trimethylbenzene	-	140	35	-	_	==			
C3-Alkylbenzenes	_	820	480	9	2	20			
sec-Butylbenzene	_	4	16	-	=	-			
1-Propyl-4-isopropylbenzene (p-cymene)	_	22	42	_	-	-			
1-Methyl-3-propylbenzene	_	280	40	-	-	**			
1-Methyl-4-propylbenzene	_	100	17	-	-	< 1			
1,3-Dimethyl-5-ethylbenzene	_	200	34	_	_	-			
1-Methyl-2-Propylbenzene	_	180	19	-1/	_	-			
1,4-Dimethyl-2-ethylbenzene	_	130	24	-	_	-			
1,3-Dimethyl-4-ethylbenzene	_	60	-	-	_	-			
1,2-Dimethyl-4-ethylbenzene	_	160	27	_	_	-			
1,3-Dimethyl-2-ethylbenzene	_	20		_	-	-			
1,2-Dimethyl-3-ethylbenzene	_	28	16	_	-	-			
1.2.3.5-Tetramethylbenzene	_	225	25	_	_	-			
1.2.3.4-Tetramethylbenzene	_	34	15	-	-	-			
	_	1,440	275	-	-	-			
Σ C ₄ -Alkylbenzenes Σ C ₅ -Alkylbenzenes	-	350	47	_	-	=			
Σ Other aromatic hydrocarbons		130	100	14	1	9			
Styrene	trace	9	1.	7	< 1	7			
Indane	_	2 = 1	20	_	-	-			
Teţralin	_	20	10	-	-	· 			
Naphthalene	-	80	40	7	1	2			
Dime thylindane	_	7	7	-	-	-			
2-Methylnaphthalene	_	10	14	_	-	-			
1-Methylnaphthalene	_	2	6	_	_	_			

Identified compounds or compound classes	Dete	cted conce	ntration	is $(\mu g/m^3)$ i	n space	
	А	P	C	D	E	F
Chlorinated hydrocarbons	260	245	145	920	36	250
Dichloromethane	¥	<u>=</u>	trace	(25,000)*	=	200
1,1.1-Trichloroethane	70	-	·	7,000	-	-
1.2-Dichloropropane	=	45	84	150	-	90
Trichloroethylene	190	-	29	50	-	1
Tetrachloroethylene		trace	32	720	19	20
1,4-Dichlorobenzene	=	200	1	trace	17	140
E Aliphatic aldehydes (≥ C ₅)	-	72	17	79	39	16
n-Pentanal	-	8	-	15	120	-
n-Hexanal	-	64	11	24	6	7
n-Heptanal	_	_	~	5	1	-
n-Octanal	-	_	-	6	trace	_
n-Nonanal	_	_	6	19	22	7
n-Decanal	-	-		10	10	2
C Other (oxygen containing) compounds	390	200	310	380	250	139
Ethylacetate	-	_	26	-	-	9
2-Methyl-1-propanol	-	-	23	60	8	4
1-Butanol	- ,	-	20	-	•	-
2,3-Epoxy-4-methylpentane	97	-	S#	70	-	-
2-Methoxy-ethanol	-	-	: **	(, -)	-	70
1-Methoxy-2-propanol	-	_	60	9 .00 .0	7.7	•
Acetic acid**	244	_	40	S X	200	3.00
2-Ethoxy-ethanol	9 4 8	6	60	2-2	-	10
iso-Butylacetate	=	-	34	100	-	_
Butylacetate			34	14	-	5
2-Methoxy-ethylacetate	-	-	244	○ -):	-	4
Hexamethyl-cyclo-trisiloxane	-	61	::=	60	21	3
Cyclohexanone	120	=	-	6	-	1
2-Ethoxy-ethylacetate	50	130	2	21	-	7
2-Butoxy-ethanol	-	_	3 = 1	127)=	8
Benzaldehyde	-	_	30	2	7	7
1,8-Cineol (eucalyptol)	_	-	-	35	-	1
Octamethyl-cyclo-tetrasiloxane		-	_	10	13	
Camphor	_	-	_ 14	227	3#4	3

^{*} This high value is due to a hair spray used accidentally during sampling and has not been included in the sum values.

^{**} Values are lower limits.

TABLE 5 - Ventilation rate and indoor-outdoor temperature difference

Home Nr.	Туре	Ventilation rate (air exch/hr)	<pre>Indoor-outdoor temp. difference</pre>
1	apartment	1.3	20
2	detached house	1.4	16
3	apartment	0.69	7.5
4	apartment	0.31	3
6	detached house	0.24	1
7	detached house	0.45	14
8	detached house	0.16	-
9	detached house	0.19	9
10	detached house	0.75	11
11	detached house	0.45	13
12	detached house	0.65	15
13	detached house	0.78	16
13	detached house	0.66	15
14	apartment	0.59	18
15	apartment	0.71	15
Mean and	standard deviation:	0.63 ± 0.35	

TABLE 6 - Compounds and identified sources

Compound	Jource
dichloromethane	propellant of spray cans
1.4-dichlorobenzene	anti-moth or deodorant products
a-pinene	conifer wood, products for furniture maintenance
limonene	lemon flavoured detergents
naphtalene	anti-moth products
n-hexanal	printed paper
n-pentanal to n-decanal	paints applied to radiators
C ₉ - C ₁₂ alkanes	waxes and polishes
C ₂ - C ₅ alkylbenzenes	waxes and polishes

TABLE 7 - Population and traffic indices and summary of concentrations of RSP and compound classes

							Sum of compound classes (µg/m³)										
Home	Population	Traffic	RSP	(µg/m ³)	Total	L vol.					halog.				benz.+ alkyl		
Nr.	density	density			org.	(μg/m ³)	aldeh	ydes	keto		4	car.	alka			enz.	
112.		-	ind.	outd.	ind.		ind.	outd.	ind.	outd.	ind.	outd.	ind.	outd.	ind.	outd.	
1	town	heavy	110	210	960	400	183	<18	21	<12	503	< 30	33	28	136	117	
2	village	medium	140	_	320	220	42	< 18	59	< 6	18	< 30	71	<35	113	<29	
2	town	heavy	70	-	240	180	29	<12	10	< 6	28	<55	27	< 32	94	< 70	
4	village	weak	60	20	190	180	54	< 44	14	< 11	238	<40	28	<38	97	65	
5	rural area	weak	125	10	9400	210	170	<19	9	< 7	14	<21	658	<17	41	< 14	
6	village	medium	40	20	7100	200	166	<19	24	< 15	151	< 36	2510	<60	278	<49	
7	village	medium	110	70	5200	340	64	<10	11	< 16	5414	<66	635	<42	228	134	
8	village	medium	40	30	1900	180	89	<12	72	<12	403	<42	29	<13	165	45	
9	rural area	weak	100	140	1500	260	48	<23	33	<16	638	<40	105	13	158	72	
10	village	weak	70	_	3000	140	36	<18	30	<12	329	< 37	192	< 9	566	21	
11	rural area	weak	100	100	2200	100	24	<12	8	< 6	53	< 56	109	<16	628	< 31	
12	village	medium	50	30	2900	690	101	<24	88	<19	3260	<33	93	44	224	216	
13	rural area	weak	50	20	5800	340	87	< 16	195	<16	145	<132	322	22	1102	113	
13bis	rural area	weak	70	80	2700	290	116	<29	17	< 8	366	<149	121	16	477	76	
14	village	medium	40	20	2300	650	14	< 14	25	<29	381	<137	80	38	257	249	
15	town	heavy	100	80	5700	1500	54	<20	108	< 16	752	238	578	115	1299	470	

TABLE 8 - Results of an additional sampling and analysis in home Nr. 15, Varese (see the text for explanation) $(\mu g/m^3)$

	Januar (from T	y 1984 able 1)	September 1984			
	indoor (street)	outdoor (yard)	ir (yard)	ndoor (street)	ou (yard)	tdoor (street)
Total volatile organics*	5700	1500	780	790	380	1070
n-hexane	114	42	26	18	10	33
n-heptane	76	17	11	9	5	13
n-octane	65	15	7	- 8	3	8
n-nonane	67	9	6	9	< 1	3
n-decane	72	10	7	9	3	3
n-undecane	134	15	7	9	4	4
n-dodecane	35	5	2	- 3	1	1
n-tridecane	15	2	1	2	< 1	< 1
benzene	204	67	46	39	23	63
toluene	378	156	115	109	56	124
ethylbenzene	96	34	16	18	11	26
1.3-xylene + 1.4-xylene	280	111	63	64	37	88
1.2-xylene	132	42	24	24	14	33
1.3.5-trimethylbenzene	59	16	10	11	5	12
1.2.4-trimethylbenzene	150	44	30	35	18	42
alpha-pinene	29	3	3	4	< 1	1
beta-pinene	29	< 1	1	3	< 1	< 1
limonene	268	4	3	4	< 1	< 1
naphtalene	70	11	5	6	4	7

^{*} see footnote in Table 1 for meaning of this denomination.

TABLE 9 - Carcinogenicity and mutagenicity of the compounds reported in Table 1a

Compound	Carcinogenicity Man Animal		Mutagenicity	
Formaldehyde	inadequate ^b	+ (c)	+	
Dichloromethane	inadequate	inadequate	+/- (d)	
1.1.1-trichloroethane	inadequate	inadequate	+	
Carbon tetrachloride	inadequate	+	inadequate	
Trichloroe thylene	inadequate	+	inadequate	
Tetrachloroethylene	inadequate	+	inadequate	
1.4-dichlorobenzene	inadequate	inadequate	inadequate	
Benzene	+	+/-	+/-	

a: Data taken from the International Agency for Research on Cancer.

b: Inadequate: data available in the literature are inadequate for an assessment.

c: positive effect in all experiments.

d: positive effect in some experiments and no effect in others.

ANNEX 1

CALCULATION METHODS FOR (SEMI-) QUANTITATIVE GC-MS ANALYSIS



Calculation Methods for (Semi-) Quantitative GC-MS Analysis

A. Quantitative determinations in case of resolved GC peaks.

The quantity Q(i) of a compound i is determined by comparison of its FID peak area AREA(i) with the peak area of an internal standard AREA(IS):

(1)
$$Q(i) = AREA(i) \cdot \frac{Q(IS)}{AREA(IS)} \cdot RFID(i)$$

where RFID(i) is a response factor which is determined by means of the external standard method, i.e. introducing known amounts of the compound i and of the internal standard into the gas chromatograph:

(2) RFID =
$$\frac{Q(i)/AREA(i)}{Q(IS)/AREA(IS)}$$
.

RFID values determined in this study are reported in table 1.

B. Semi-quantitative determination in case of non-resolved GC peaks.

In case of non-resolved GC peaks, each component i contributes an amount AREA(i) to the total area of the GC peak \nearrow AREA(j), which can be expressed as a fraction B_i of this area:

(3) AREA(i) =
$$\beta_i \cdot \sum_{j} AREA(j)$$

(4) $\beta_i = \frac{AREA(i)}{\sum_{j} AREA(j)}$

 $B_{\mathbf{i}}$ can be determined by means of the total ion current (TIC) of component i at its GC elution maximum. This TIC value can, however, only be determined to the degree to which the mass spectra of the co-eluting substances differ from each other. In the case of a co-eluting alkane and alkylbenzene, e.g., a fairly good separation is possible whereas coeluting alkanes or alkylbenzenes in general cannot be separated.

The determination of β_i by means of the TIC value is based on the proportionality of the number of moles M(i) of substance i and the corresponding TIC value:

(5)
$$M(i) = k \cdot RTIC(i) \cdot TIC(i)$$

where k is the TIC response factor of the internal standard

$$M(IS) = k \cdot TIC(IS)$$

and RTIC(i) is the relative TIC response factor of substance i

(6) RTIC(i) =
$$\frac{M(i)/TIC(i)}{M(IS)/TIC(IS)}$$

RTIC values are determined by the external standard method and table 1 reports the values measured during this study.

with

$$Q(i) = MW(i) \cdot M(i)$$

(MW = molecular weight), eq. (1) can be re-written:

(7) AREA(i) =
$$c \cdot MW(i) \cdot M(i) / RFID(i)$$

$$c = \frac{AREA(IS)}{O(IS)}.$$

Substituting eq. (5) in eq. (7) yields

and
$$\beta_{i} = \frac{\text{MW(i)} \cdot \text{TIC(i)} \cdot \text{RTIC(i)} / \text{RFID(i)}}{\text{MW(j)} \cdot \text{TIC(j)} \cdot \text{RTIC(j)} / \text{RFID(j)}}$$

C. Practical quantification procedure

The measured GC peak areas are transformed into relative quantities

GC peaks are then analysed by means of the reconstructed ion chromatogram for not resolved peaks, for which TIC(i) values are determined as far as possible and $\beta_i\text{-values}$ are calculated. Quantification of sample components is performed by means of equation

(9)
$$Q(i) = \beta_i \cdot QREL \cdot RFID(i)$$

with β_i = 1 for resolved GC peaks and calculated by means of eq. (8) otherwise.

		VI
Table 1 SUBSTANCES or SUBSTANCE CLASSI	ES RFID	RTIC
n-alkanes (C ₇ - C ₁₂) alkyl-benzenes (C ₁ - C ₆)	0.71 0.67	1.10 1.81
1-bromo-alkanes (C ₆ , C ₉ , C ₁₂)	1.00	1.00
p-dichloro-benzene l-butanol n-aldehydes (C5 - C9) l,3,-dichloro-propane 2-ethoxy-ethanol ethyl-acetate trichloro-ethene	1.02 1.03 1.26 2.13 2.42 3.26 4.40	1.41 1.56 2.17 1.87 11.1 1.36 2.16
tetrachloro-ethene	4.35	1.88

ANNEX 2

INDOOR POLLUTANT INVENTORY SAMPLE DESCRIPTION FORM



INDOOR POLLUTANT INVENTORY - SAMPLE DESCRIPTION FORM SAMPLE NUMBER USER DEFINED NUMBER LABORATORY/OPERATOR IDENTIFICATION BIBLIOGRAPHY CODE LABORATORY NAME (USE STAMP IF AVAILABLE) OPERATOR IDENTIFICATION SAMPLING DATE® NO INFO* HINDH YEAR IF NO, GIVE SAMPLING DURATION IN NO SAMPLING DURATION TES NO INFO* GRAN SAMPLE ? TINUTES HOURS DAYS 5. SAMPLING PROCEDURE NO INFO* CODE DESCRIPTION : NO CODE AVAILABLE SAMPLING SITE 5. POSTAL CODE (AS USED ON CARS) NO INFO+ IDENTIFICATION SAMPLING SITE CHARACTERIZATION LOCATION NO INFO* SUBURBAN MABRE RURAL TRAFFIC DENSITY NO INFO*LIGHT TRAFFIC/MODERATE TRAFFIC/HEAVY TRAFFIC INDUSTRY NO INFO* NO INCUSTRY / SINGLE END. SOURCE/LIGHT INCUSTRY/ HEAVY INCUSTRY TYPE OF BUILDING 8. THER NO INFO#RESIDENTIAL SCHOOL OFFICE IF 'OTHER', PLEASE GIVE DESCRIPTION 9. AGE OF BUILDING CONSTRUCTE ON COMP NO INFO* AFTER 1973 1945 - 1973 WITHIN LAST 3 MONTHS 3EFORE 1945 ROOM TYPE 10. BATH/WC LIVING DINING BEDROOM STUDIO/OFFICE NO INFO# CLASSROOM OTHER IF 'OTHER' GIVE DESCRIPTION HOBBY CORRIDOR 11. FLOOR LEVEL PLEVEL/STOREY(D = GROUND LEVEL) NO INFO* ABOVE GROUND BELOW GROUND 42 ROOM SIZE/HEIGHT 12. APPROXIMATE SURFACE AREA APPROXIMATE ROOM HEIGHT NO INFO* ONLY IN CASE OF SAMPLING IN A 1 M2 13. PRIVATE RESIDENCE R. OF STRATIBARKI VR. OF ROOMS NO INFO* TOTAL SURFACE AREA APPROX. 14. VENTILATION TYPE + MIXED NATURAL FORCED NO INFO* 15. VENTILATION STATUS DURING SAMPLING* INTERMEDIATE/NORMAL NO INFO* MINIMUM+ MUMIXAM DURING LAST TWO HOURS BEFORE SAMPLING + 1 NO INFO* INTERMEDIATE/ NORMAL *MUMIKIN MUMIXAR 7°C (MEAN) TEMPERATURE/ 16. YIJGIMUE NO INFO* TEMPERATURE HUMIDITY DURING SAMPLING

17.	PEOPLE:	Were people press sampling (except sampling device)	during install		
18.	PETS:	No INFO* Are there usuall NO INFO*	y pets in the s	ampling space?	
19.	RENOVATION:	sampling space w	ithin the last		
20.	CONSUMER PRODUCTS:				release volatile day(s) of sampling?
21.	SMOKING:	Did anybody smok hours before sam		ng space during	or within two
22.	APPLIANCES:	Has any applianc within one hour	e been used in		ace during or
23.	COMFORT COMP	NO INFO* UNVENTED IN CASE OF 'OTHER VOI LAINTS: Have com		PLEASE, SPECIFY APP	LIANCE:
		sampling space? NO INFO* If YES, were the	NO	YES	
	•	CLIMATE? ODOUR?	NO INFO*	NO NO	YES YES
24.	HEALTH COMPL	AINTS: Have heal space?	th complaints t	een associated	with the sampling
9C 	•	If YES, were the EYES?	y related to	NO	YES
Rc of	•	UPPER AIRWAYS?	NO INFO*	NO	YES
	•	LOWER RESPIRA- TORY TRACT? SKIN?	NO INFO*	NO	YES
	•	OTHER?	NO INFO*	NO	YES
			NO INFO#	,10	YES

INDOOR POLLUTANT INVENTORY - SAMPLE DESCRIPTION FORM

- EXPLANATIONS AND COMMENTS -

Four types of data are requested in the sample description form:

- data on the sample itself including sampling and analysis procedures (items 1 - 6);
- data describing physically the indoor space in which the sample has been taken including some information on its surroundings (items 7 - 17);
- data describing potential indoor sources of pollution (items 18 23) and
- data related to comfort and health disturbances observed in the sampling space (items 24 25).

Jata are in general supplied by crossing one or several options or by filling in numbers or pames

Only for items 5 and 6 information should be supplied in a coded form. Codes are defined on the following pages. If no appropriate code is found, a free description of the sampling procedure and/or the method(s) applied in the analysis of the sample should be given in the appropriate spaces.

If for any item no information is available, the respective 'NO INFO' field should be crossed.

Figures for hours and days (item 4), surface areas (items 13 and 14) and temperature and humidity (item 17) should be rounded to nearest integers.

jae na saatta saatta ka saatta

SAMPLING PROCEDURE

Please specify which method has been used for sampling by inserting into the "CODE" field one of the following codes:

blank - no information available

- 01 direct introduction of an air sample into the analytical apparatus
- 02 air sample taken in a bag and subsequent direct introduction into the analytical apparatus after transfer to the laboratory
- 10 passive sampling on carbon / solvent extraction
- 20 adsorption on TENAX / thermal elution
- 21 adsorption on Carbon Molecular Sieve / thermal elution
- 22 adsorption on TENAX + Carbon Molecular Sieve / thermal elution
- 23 adsorption on active charcoal / solvent extraction
- 24 adsorption on Polyurethane foam / solvent extraction
- 31 low volume sampler / filter extraction
- 32 low volume sampler / direct filter analysis
- 33 high volume sampler / filter extraction
- 34 high volume sampler / direct filter analysis
- 10 low volume sampler/filter + polyurethane foam/
 solvent extraction
- 41 high volume sampler / filter + polyurethane foam / solvent extraction
- 99 other

ANALYTICAL METHOD(S)

Please specify the analytical method(s) by which the sample has been analysed using the codes defined below. Insert the appropriate code(s) in the "CODE" field(s).

blank no information available

- 01 colorimetric analysis
- 02 spectrophotometric analysis
- 03 fluorescence photometry (fixed wave length)
- 04 spectrofluorimetry
- 05 infrared spectroscopy (IR)
- 06 Fourrier Transform Infrared Spectroscopy (FTIR)
- 07 chemiluminescence analysis
- 10 paper chromatography
- 11 thin layer chromatography (TLC)
- 20 yas chromatography (GC)
- 21 GC-flame ionization detection (FID)
- 22 GC-electron capture detection (ECD)
- 23 GC-flame photometric detection (FPD)
- 24 GC-FID-ECD
- 30 liquid chromatography (LC).
- 31 high performance liquid chromatography (HPLC)
- 32 HPLC UV detection (UV)
- 33 HPLC fluorescence detection (FD)
- 34 HPLC FD UV
- 40 mass spectrometry (MS)
- 41 00-MS
- 42 GC-FID-MS
- 43 HPLC-MS
- 99 other