FLIES (Flanders Indoor Exposure Survey): The influence of contaminants in ambient air on the indoor air quality and children’s exposure

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SUMMARY

This study presents the first Flemish monitoring campaign on indoor air quality in children’s living environment. The main aim of the study was to determine for sensitive groups of children the indoor environment exposure a result of contaminants that occur in the outdoor and indoor air, and to quantify the relative importance of ambient and indoor-source related exposure to total personals exposure of children.

The monitoring campaign was performed in January – February 2006 in 50 dwellings, and 24 other children’s micro-environments. Indoor and outdoor concentrations (7-day averages) of 15 pollutants were measured: MTBE, benzene, trichloroethene, tetrachloroethene, ethylbenzene, m+p xylene, styrene, o-xylene, 1,2,4 trimethylbenzene, p-dichlorobenzene, TVOC, NO2, formaldehyde, acetaldehyde and PM.

Since MTBE has no indoor source (petrol additive), the slope of indoor versus outdoor MTBE ($F_{INF} = 0.86; 95\% \text{ C.I.}: 0.59-1.13$) could be used to assess the infiltration of other gases. Using this indirect technique, it was estimated that, on average, the relative importance of indoor sources to total indoor concentrations indoor varied from 85 % for formaldehyde to 34 % for traffic related components such as benzene. The MTBE infiltration technique was not suitable for PM and NO2.

Typical children’s personal exposure was modeled based on typical time patterns and median indoor concentrations of different micro-environments. It revealed that on average, the indoor-source exposure attributed from 32 % (benzene) to 80 % (formaldehyde) to personal exposure, whereas ambient exposure accounted for 66 % (benzene) to personal exposure. Exposure experienced during transport consisted of 2 % (tetrachloroethene) to 15 % (MTBE). It is concluded that the relative importance of indoor sources or ambient sources is pollutant-dependent and it is advised to take this pollutant-dependent behaviour into account when stipulating policies aiming at reducing exposure to children.

INTRODUCTION

Whereas ambient air quality in Flanders has been monitored for years, indoor air quality in Flanders has not been investigated systematically in a quantitative way in the past. However, people spend up to 90 % of their time indoors, which indicates the relevance of indoor air quality to respiratory health.

Air pollutants present in the indoor environment are the result of (1) product emissions such as furniture, consumer products, cleaning materials,… and (2) infiltration of ambient substances. Knowledge on the relative importance of these two terms is crucial for stipulating a good indoor air quality policy.

The aim of the study was (1) to make an inventory of indoor concentration in Flemish dwellings and other micro-environments in which people spend a large part of their time, (2) to assess if traffic density affected indoor and outdoor concentrations and (3) to determine the
personal exposure of children and, (4) to differentiate the personal exposure in an ambient and non-ambient fraction. It is hypothesized that traffic density affects the concentration of typically traffic-related compounds such as MTBE and benzene and not for other compounds.

Children were selected as sensitive groups because they experience a higher air pollutant load given their 50% larger body-weight rescaled or 35% larger long-surface rescaled exposure than adults. The selection of the investigated pollutants was based on 3 criteria: 1) only pollutants with possible outdoor sources were selected (biological agents and ozone not retained) and 2) only pollutants for which health effects have been proven in the past (e.g., based on WHO health criteria). Terpenes, pesticides and bromated flame retardants were not measured in the first Flemish Indoor Exposure Survey because of too specific and demanding sampling techniques.

METHODS

Selection of measuring locations

The monitoring campaign was performed in January-February 2006 in the eastern part of Flanders in 50 dwellings of volunteers. The dwellings were selected based the proximity to traffic: 12 houses in rural background (RB) areas (< 50 cars passing/day), 24 houses in urban background (UB) area (< 500 cars/day) and 12 houses in urban hot spot (HS) area (>15000 cars/day). The emphasis on dwellings in UB areas is because of 75% of Flemish houses are of this type.

In the houses, 2 indoor locations, namely living room and bedroom, and 2 outdoor locations, namely front door and back door (if applicable) concentrations were sampled. The set of 50 houses existed of a heterogeneous mix of dwelling types: detached dwellings (12), semi-detached dwellings (n = 3), connected dwellings (n = 23), flats at ground floor (n = 1) and flats above the ground floor (n = 7). Additionally, indoor and outdoor air was sampled at schools or daycares (n = 10), transport (n = 9; car, public transport, wandering) and sport and leisure infrastructures (n = 7). The number of sampled dwellings was smaller for PM (10 living rooms, 34 bedrooms and 19 dwelling outdoor locations) because of logistical reasons.

Questionnaires

Information regarding possible indoor sources (presence of consumers products, cleaning products, furniture, heating devices, ...), dwelling characteristics (ventilation types and frequency, isolation, single or double glass windows, presence of an attached garage …) was obtained by means of a detailed questionnaire. The second part of the questionnaire investigated the children’s time patterns (7-day diary with 1 hours intervals for each child).

Sampling and analysis techniques

Except for particulate matter, all pollutants were measured by diffusive sampling techniques. For VOC’s Radiello samplers were used. After 7-days sampling, the samplers were extracted with carbon disulfide containing an internal standard. Analysis were performed on a HP6890 gas chromatograph hyphenated with a HP5975 mass spectrometer. Total volatile organic compounds (TVOC) was measured in full scan-modus, while the selected individual compounds were measured in selected ion monitoring mode. External standards were used for calibration. Aldehydes were sampled with diffusive dosimeters manufactured by SKC.

Mechanism of the dosimeters is chemisorption with dinitrophenylhydrazine. After the sampling period, the aldehydes were desorbed with acetonitrile and analyzed by LC-UV. NO2...
was sampled with diffusive monitors IVL (Swedish Environmental Institute) and analysed by IVL. Particulate matter was measured by Grimm dust, Buck or Aeromini monitors. The latter was provided with a weather proof enclosure and was used outdoors. Grimm and Buck monitors were used indoors.

Infiltration of ambient pollutants

The total indoor concentration of a substance can be expressed with equation 1 [1] which included a left term related to outdoor concentration and a right term representing the contribution of indoor sources:

\[
C_{\text{indoor}} = \frac{P a}{a + k} C_{\text{outdoor}} + C_{\text{indoor sources}}, \quad (1)
\]

The infiltration factor (FINF) can be derived in the absence of indoor sources, assuming the following relationship between concentrations indoor (Ci) and outdoor (Co) [2,3]:

\[
F_{\text{INF}} = \frac{C_i}{C_o} = \frac{P a}{a + k}, \quad (2)
\]

with P: penetration factor (-); a is the air exchange rate (/h) and k is the deposition, removal or sorption rate (/h). In this study, MTBE was selected as a tracer with only outdoor sources to calculate FINF and thus, in a next step, to discriminate the outdoors generated from indoor generated fractions of other pollutants:

\[
C_{ig,x} = C_{i,x} - F_{\text{INF}} \times C_{o,x}, \quad (3)
\]

with ig = indoor generated and x = substance x
This method is analogous to the principle used in other studies using other tracers (e.g. SO4 as tracer for PM2.5) to determine outdoor to indoor infiltration of PM [4,5]

Personal exposure

The typical children’s personal exposure was modeled based on time activity patterns obtained through the questionnaires and concentrations of each micro-environment. Total personal exposure can be split up in 1) ambient exposure (directly and infiltrated), 2) indoor sources and 3) exposure experienced during transport. The latter micro-environment is difficult to separate in a ambient and indoor generated fraction under the current set-up. Thus, personal exposure to substance x was calculated as:

\[
T_x = A_x + N_x + TR_x, \quad (4)
\]

Ambient (A_x) and non-ambient (N_x) exposure were respectively calculated as :

\[
A_x = \sum_j t_{o,j} \times C_{o,j,x} + \sum_j t_{i,j} \times F_{\text{INF}} C_{o,j,x}, \quad (5)
\]

\[
N_x = \sum_j t_{i,j} \times C_{i,j,x}, \quad (6)
\]

with t_{o,j} and t_{i,j}: time fraction spent respectively outdoor and indoor at µ-environment j.

Exposure experienced during transport (TR_x) (I or O) was calculated as a separate term. Total personal exposure to x is the sum of A_x, N_x and TR_x.

The variability in exposure was assessed based on Monte Carlo simulations (Crystall Ball software) by accounting for variability in dwelling indoor concentrations, which is the micro-
environment with the highest variation in concentrations and the most important compartment with respect to time budget. Variations in other micro-environments were not accounted for because they affected to a much lesser extent the exposure and because a distribution on concentrations could not be established because of the limited dataset for micro-environments other than dwellings.

Statistical analyses

The statistical analyses were performed using the statistical tool package Statistica (version 7). The non-parametric Kruskal-Wallis Anova test and non-parametrical correlation analyses were applied because of not normally distributed datasets.

RESULTS

A summary of indoor and outdoor concentrations in 50 Flemish dwellings is given in Table 1. Concentrations of gases show a very high variability between different houses (n=50), both indoors and outdoors. The most abundant gases in both indoor and outdoor environments were toluene, NO₂, formaldehyde and acetaldehyde.

Indoor concentrations generally exceeded corresponding outdoor concentrations (except for NO₂ and PM10). The target value of the Flemish Indoor Decree of 11/06/2004 [6] of 200 µg/m² is exceeded in 95 % of the investigated indoor environments. The Flemish target values for formaldehyde (10 µg/m³) and for benzene (2 µg/m³) were exceeded in nearly 50 % of the cases. The Flemish intervention limits for benzene and formaldehyde were exceeded in respectively 3 and 1 indoor dwelling locations. The Flemish Indoor Decree does not define intervention limits for TVOC’s.

Table 1: Indoor (living room and bedroom) and outdoor (front door and backdoor) concentrations in 50 Flemish dwellings

<table>
<thead>
<tr>
<th></th>
<th>Dwelling Indoor (n=100)</th>
<th>Dwelling Outdoor (n=86)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>median</td>
<td>min</td>
</tr>
<tr>
<td>MTBE</td>
<td>0,54</td>
<td>0,12</td>
</tr>
<tr>
<td>Benzene</td>
<td>2,14</td>
<td>0,70</td>
</tr>
<tr>
<td>Trichloroethene</td>
<td>0,16</td>
<td>0,03</td>
</tr>
<tr>
<td>Toluene</td>
<td>8,13</td>
<td>1,29</td>
</tr>
<tr>
<td>Tetrachloroethene</td>
<td>0,26</td>
<td>0,06</td>
</tr>
<tr>
<td>Ethylbenzene</td>
<td>1,09</td>
<td>0,20</td>
</tr>
<tr>
<td>m-p-Xylene</td>
<td>2,33</td>
<td>0,43</td>
</tr>
<tr>
<td>Styrene</td>
<td>0,21</td>
<td>0,01</td>
</tr>
<tr>
<td>o-Xylene</td>
<td>0,86</td>
<td>0,14</td>
</tr>
<tr>
<td>1,2,4-Trimethylbenzene</td>
<td>1,99</td>
<td>0,17</td>
</tr>
<tr>
<td>p-Dichlorobenzene</td>
<td>0,07</td>
<td>0,03</td>
</tr>
<tr>
<td>TVOC</td>
<td>491</td>
<td>138</td>
</tr>
<tr>
<td>NO₂</td>
<td>21,3</td>
<td>7,1</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>23,7</td>
<td>1,4</td>
</tr>
<tr>
<td>Acetaldehyde</td>
<td>21,8</td>
<td>1,1</td>
</tr>
<tr>
<td>PM10</td>
<td>11,7</td>
<td>2,3</td>
</tr>
</tbody>
</table>
Statistics in Table 1 include both living room and bedroom concentrations and front door and backdoor concentrations. On average, the ratio of bedroom to corresponding living room concentration was near 1 (average ratio varied from 0.80 for acetaldehyde to 1.51 for styrene). In general, backdoor concentrations were slightly (±0.9) fold lower than frontdoor concentrations. This suggests that the dwellings act as a barrier for pollutants that are mainly formed at the street.

For some gases, outdoor concentrations were affected by the traffic density, concentrations for HS locations being highest, followed by urban background, and by rural background (Table 2). As expected, traffic related compounds (MTBE, benzene, toluene and NO2) are higher in outdoor environment of HS compared to RB. For some traffic pollutants, higher indoor concentrations in HS than in RB and UB (e.g. NO2) were observed. Compared to the outdoor concentrations traffic-related pollutants like toluene are no longer significantly different in the different locations, indicating an additional contribution from indoor sources. For other, more indoor generated pollutants (xylenes, …) no effect of traffic density indoor concentrations was observed. The distribution of PM sampling sites was not balanced enough over HS, UB and RB classes to allow a statistical comparison (data not shown).

Table 2: Mean outdoor and indoor concentrations for 3 traffic density classes (UB: urban background; HS: hot spot; RB: rural background) Statistical different values between location type classes (Anova, P<0.05) are marked with different letters (for gases without differences between any of the 3 groups are not marked with letters)

<table>
<thead>
<tr>
<th></th>
<th>UB</th>
<th>HS</th>
<th>RB</th>
<th>UB</th>
<th>HS</th>
<th>RB</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>µg/m³</td>
<td>µg/m³</td>
<td>µg/m³</td>
<td>µg/m³</td>
<td>µg/m³</td>
<td>µg/m³</td>
</tr>
<tr>
<td>MTBE</td>
<td>0.4A</td>
<td>0.6A</td>
<td>0.3B</td>
<td>1.53</td>
<td>0.75</td>
<td>1.52</td>
</tr>
<tr>
<td>Benzene</td>
<td>1.6A</td>
<td>2.0B</td>
<td>1.4A</td>
<td>2.75</td>
<td>4.16</td>
<td>2.15</td>
</tr>
<tr>
<td>trichloroethene</td>
<td>0.13A</td>
<td>0.12A</td>
<td>0.07B</td>
<td>0.22</td>
<td>0.28</td>
<td>0.12B</td>
</tr>
<tr>
<td>Toluene</td>
<td>3.5AB</td>
<td>4.3A</td>
<td>2.5B</td>
<td>13.31</td>
<td>10.58</td>
<td>19.03</td>
</tr>
<tr>
<td>ethylbenzene</td>
<td>0.6A</td>
<td>0.5A</td>
<td>0.1B</td>
<td>0.76</td>
<td>4.13</td>
<td>0.23C</td>
</tr>
<tr>
<td>styrene</td>
<td>0.1</td>
<td>1.5</td>
<td>1.1</td>
<td>3.23</td>
<td>2.97</td>
<td>3.5</td>
</tr>
<tr>
<td>m+p xylene</td>
<td>0.5</td>
<td>5.7</td>
<td>8.8</td>
<td>34.5A</td>
<td>36.9AB</td>
<td>21.1B</td>
</tr>
<tr>
<td>NO2</td>
<td>38.4A</td>
<td>47.5B</td>
<td>27.5A</td>
<td>24.3A</td>
<td>31.7B</td>
<td>17.6C</td>
</tr>
<tr>
<td>formaldehyde</td>
<td>11.8</td>
<td>5.7</td>
<td>8.8</td>
<td>17.9</td>
<td>24.4</td>
<td>16.4</td>
</tr>
</tbody>
</table>

The MTBE infiltration method (see above) can only be used under the assumption of absence of indoor sources of the tracer. However, exceptionally high indoor/outdoor MTBE ratios were observed for 2 dwellings. In these two dwelling, MTBE indoor concentrations 7.6-33 µg/m³ were measured, while concentrations were as low as 0.146 - 0.195 µg/m³. The only potential MTBE indoor source was the presence of gasoline indoors. Indeed, the presence of a garage in the dwelling, or adjacent to the dwelling (with passage between garage and house) in these 2 houses suggests the presence of MTBE indoor source. In 6 other houses, indoor concentrations above 1 µg/m³ were measured; although the MTBE ratio was I/O remarkably lower than of the 2 above mentioned dwellings, I/O of these 6 dwellings was largely above
the remainder part, and 5 of 6 of these houses had a garage in the dwelling (directly or adjacent with passage), suggesting MTBE indoor sources. Therefore, these houses were excluded and based on the filtered dataset (n = 42), the average $F_{\text{INF}}$ factor was derived from the slope of indoor MTBE versus outdoor MBTE.

\[ y = 0.8625x + 0.1565 \]

\[ R^2 = 0.5061 \]

Figure 1: Indoor versus outdoor MTBE concentrations in 42 houses without indoor sources of MTBE. The average infiltration factor FINF is defined as the slope of indoor versus outdoor MTBE.

The indoor generated concentrations of pollutants ($C_{ig}$) were calculated for each dwelling individually, using an average $F_{\text{INF,MTBE}}$ factor (0.86; 95% CI: 0.59-1.13). It should be kept in mind that the individual $F_{\text{INF,MTBE}}$ values may differ between houses. However, dwelling type and ventilation frequencies, building properties (isolation, single versus double glass windows) did not statistically affect I/O MTBE ratio’s. It was thus not feasible to stratify $F_{\text{INF,MTBE}}$ for building type and ventilation classes due to the limited dataset of houses in this study.

The pollutant with the largest median $C_{ig}/C_i$ ratio was formaldehyde ($C_{ig}/C_i = 0.85$) and the pollutant with the lowest $C_{ig}/C_i$ was benzene ($C_{ig}/C_i = 0.34$). For each component, rather large deviations from this median value was observed (e.g. formaldehyde P25 of $C_{ig}/C_i = 0.71$ and P75 of $C_{ig}/C_i = 0.91$; for benzene: P25 of $C_{ig}/C_i = 0.22$ and P75 of $C_{ig}/C_i = 0.55$). This is not surprising given the wide spread in possible indoor sources between individual dwellings. Notwithstanding the dominance of indoor sources to indoor formaldehyde concentrations pointed out by this indirect MBTE infiltration method, the statistical analyses between indoor sources, building and ventilation characteristics and measured concentrations did not reveal significant relationships for formaldehyde. Also for other pollutants, the statistical relations between indoor sources and measured concentrations were generally poor. The most important significant relations were the significant effects of (1) smoking on $C_i$ and $C_{ig}$ of toluene, (2) use of glue and stain removers on $C_i$ and $C_{ig}$ TVOC and (3) use of sealing products on $C_i$ and $C_{ig}$ benzene.

The MTBE infiltration method was not successful to estimate $C_{ig}$ for NO2 and PM10.

The typical exposure for children is calculated based on median concentration in each micro-environment and average time patterns. Typical children’s exposures attributed $0.52 - 0.71 \mu g$ MBTE/m³, $1.9 - 2.7 \mu g$ benzene/m³, $0.17 - 0.23 \mu g$ trichloroethylene/m³, $6.3 - 7.5 \mu g$ toluene/m³, $0.24 - 0.48 \mu g$ tetrachloroethene/m³, $0.9 - 1.4 \mu g$ ethylbenzene/m³, $2.0 - 2.9 \mu g$ m+p xylene/m³, $0.10 - 0.70 \mu g$ styrene/m³, $0.8 - 1.1 \mu g$ o-xylene/m³, $1.6 - 2.3 \mu g$ 1,2,4-trimethylbenzene, $0.1 - 0.9 \mu g$ p-dichlorobenzene, $410 - 522 \mu g$ TVOC/m³, $19 - 34 \mu g$ NO2/m³, $15 - 25 \mu g$ formaldehyde/m³, $10 - 27 \mu g$ acetaldehyde/m³ and $8 - 13 \mu g$ PM10/m³. These ranges of typical exposure refer to
variation in typical exposure for different age classes (0-2.5 years, 2.5-6 years, 6-12 years, 12-18 years) and proximity of the houses to traffic density (UB, RB, HS). The differences in typical exposure between different age classes and traffic density classes were rather small. It was assessed that the highest exposed children (P95) were 2-4 fold higher exposed than the median, typical exposed children for most gases. Exceptions were tetrachloroethene (x11), and p-dichlorobenzene (x 50). These extremes were related to the extremely large range of indoor concentrations (Table 1).

The distribution of exposure over ambient (A: $A_i + A_{o,i}$ with $A_{o,i}$ exposure to ambient pollutants directly in outdoor environment and $A_i$, exposure to ambient pollutants that have infiltrated indoors)), indoor source-related (N) and exposure during transport (TR) is given in Figure 2 for TVOC and for the 2 components with the most extreme A,N, TR distribution profiles (except for MTBE, NO2 and PM).

For other pollutants, either ambient exposure dominated typical personal exposure (for MTBE, trichloroethene, tetrachloroethene), or indoor sources exposure (toluene, ethylbenzene, m+p xylene, styrene, o-xylene, 1,2,4-trimethylbenzene, p-dichlorobenzene) dominated the personal exposure. Exposure during transport was for most components below 5 % to the total personal exposure and varied from < 2% (tetrachloroethene) to 15 % (MTBE).

Figure 2: Distribution of typical exposure to formaldehyde, TVOC and benzene over ambient, non-ambient (indoor sources) exposure and exposure during transport.

**DISCUSSION**

Indoor concentrations of benzene, toluene, o-xylene, ethylbenzene, trimethylbenzene,… in Flemish dwellings were generally within ranges reported previously elsewhere [7,8]. In contrast, the low indoor-outdoor PM ratio of this study (mean I/O ratio: 0.3) are not in line with previous studies [9] and could not be explained.

The use of MTBE as infiltration tracer is relatively new, and this MTBE based infiltration method gives realistic values of $F_{inf}$ for dwellings, in accordance with infiltration factors reported by others using other tracers (e.g. $F_{inf}$ based on sulphate = 0.7 (90 % CI: 0.5-0.9) [5]. The intercept of the MTBE indoor versus outdoor graphs refers to small residual (background) MTBE concentrations in houses. Similar background concentrations were also found for SO$_4$ [10]. However, the MTBE based infiltration factor was not applicable to discriminate indoor and outdoor generated NO$_2$ and PM$_{10}$ concentrations in this study. The inappropriateness of MTBE to estimate the infiltration of PM and NO$_2$ probably lies in different penetration factors and/or removal/sorption rates or, in the extreme low indoor-outdoor ratio’s for PM$_{10}$. The 95 % CI of $F_{INF}$ (0.59-1.13) indicates that there is a further need to refine $F_{INF}$ for different dwelling types with respect to ventilation. A larger dataset than the one of this study is needed hereto.
Children’s exposure is mainly determined by indoor dwelling exposure given the majority of time spent indoors. Variation in exposure between children is mainly due to variations in indoor dwelling concentrations (up to 100-fold), rather than variations in children’s time budgets, which are limited.

This study shows that the contribution of infiltrated outdoor air pollution to personal exposure is different among investigated pollutants. This is a point of attention in ambient air quality policies, to include the indoor exposures more explicit. Recommendations for precautionary measures to reduce or avoid exposure indoor exposure to certain gases, for example for formaldehyde, are difficult to make based on this study because only few clear source-concentrations-exposure relationships were found. For this, work on short-term and long-term emission sources and their relation to concentrations, using various time average measurements should be performed. This is best placed in the context of product policy.

ACKNOWLEDGEMENT

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