(Contract n° 041689)

The influence of contaminants in ambient air on the indoor air quality – part 1: exposure of children

Report of work package 3: interpretation and policy recommendations

Katleen De Brouwere, Christa Cornelis, Eddy Goelen, Gudrun Koppen, Maarten Spruyt, Rudi Torfs

Work performed under contract with Environment, Nature and Energy Department, Environment and Health Unit (former AMINAL)

2007/IMS/R/62

VITO

February 2007

TABLE OF CONTENTS

0 SUMMARY AND OUTLINE

This report is the third and final part of the study "The Influence of Contaminants in Ambient Air on the Indoor Air Quality, part 1: Exposure of Children".

In this report, data of the measuring campaign (workpackage 2) in 50 dwellings is used to develop an exposure assessment of children. In this work package, a stepwise approach was used to convert air concentrations of gases and particulate matter (measured in WP 2) in different micro-environments in which typically Flemish children spend their time into children's exposure to air pollutants.

Firstly, indoor and outdoor concentrations are evaluated relative to the type of micro-environment type (1.1.1 and 1.1.2). Concentrations in dwellings, schools and other micro-environments are summarized. Differences between micro-environments in dwellings (bedroom versus living room) and differences between front door and backdoor concentrations are assessed. It is also tested whether traffic density near dwellings affects indoor and outdoor concentrations (1.1.3). Finally, correlations between indoor occupant activities or product use indoors and these indoor concentrations are calculated (1.1.4).

To evaluate the contribution of indoor and outdoor sources of air pollution separately, the measured indoor concentrations are broken down into a fraction that is related to outdoor air pollution that has infiltrated indoors, and a fraction that is related to indoor sources (1.1.5). The latter indoor concentrations were then again analysed for relationships with indoor activities, product use, building materials, …(1.1.6)

In a second part (1.2), children's exposure to air pollutants is determined for different age categories and for different locations, with traffic density as the important indicator for location specific differences in air pollution. Typical exposure scenarios were set up using the median concentrations in different micro-environments and typical time activity patterns. The children's exposure to air pollutants was allocated to either the outdoor sources and the indoor sources.

Finally, in addition to the typical (median) exposure, the distribution of exposure was also assessed using the range of concentrations across the examined micro-environments. Such distributions are in particular useful to estimate the exposure of high exposed children.
Exposure data were also converted into pollutant doses to children.

A discussion of the results is given in chapter 2 together with some policy recommendations.

The main conclusions of WP 3 are:

High indoor concentrations, large variations and exceedance of limit values
Among the 14 measured gases the most abundant gases in both indoor and outdoor environment were formaldehyde (up to 124 µg/m³) , acetaldehyde (up to 65 µg/m³), NO_2 (up to 122 µg/m³) and toluene (up to 122 µg/m³). These upper values are all for indoor environments (living rooms and bedrooms). Concentrations of gases show a very high variability between different houses (n=50), both indoors and outdoors. Especially for some gases like formaldehyde and toluene (for which the concentrations in bedrooms varied with a

factor of 50) that can be associated with building materials and product use, there is a need to assess how widespread this problem is. Product standards, ventilation and prevention information are needed and their efficiency tested. In more than 85 % of the investigated indoor environments, the guideline values of the Flemish Indoor Decree for TVOC (200 µg/m³), formaldehyde and benzene were exceeded. In addition, in 3 houses the intervention values for benzene and in 1 house the intervention value for formaldehyde was exceeded. It is recommended to measure selected gases (TVOC, benzene, formaldehyde) in a larger dataset of Flemish houses in order the evaluate the magnitude of problem in Flanders.

Outdoor pollution contributes to indoor pollution
On the basis of MTBE as the indicator of infiltration indoor, an assessment was made for the dwellings of the fraction of the indoor concentration attributable to indoor sources. The relative contribution of outdoor generated and indoor sources was dependent on the pollutant type: typically, 85 % of the total indoor formaldehyde concentration was attributed to indoor sources, whereas for benzene, trichloroethene, and tetrachloroethene the infiltration of outdoor generated compounds dominated (by 70 %) the indoor concentration. Variability of the infiltration factor results in a variability of the contribution of indoor sources to the indoor concentrations of about a factor 2.

Measurements of concentrations do not show clear relations with indoor sources and activities.
In general, only few significant correlations between indoor concentrations and indoor/building properties were present. Indoor concentrations of toluene and PM were affected by presence of smokers. Indoor concentrations of xylenes and TVOC's (combustion products) were associated with stove use. However, for most of the expected source-concentration analyses, no significant relationships between source and concentration were present. This can be explained by the large averaging time (7 days) of the measurements for the 50 dwellings.

Exposure is dominated by time spent indoor
Exposure of children is dominated by the time spent indoor at home, basically in the living room (on average 4h/day) and bedroom (on average 11h/day) at home and in the school or day care (on average 4h/day). Other micro-environments, namely transport, are less important in an average exposure pattern, although they give rise to high concentrations. If health effects from exposure to air pollution is dominated by the long-term average exposure than our attention should go to the micro-environments where most time is spent. But at the same time acute effects from peak exposure cannot be excluded, keeping other micro-environments like motorised traffic, like leisure indoors in the picture.

The typical exposure of children to the selected pollutants does not vary significantly across ages and across locations. Typical exposures to traffic related pollutants are slightly (but not significantly) higher in hot spot areas compared to urban or background areas. Exposure indoors dominates the total exposure. Using the range of concentrations at home results in a highly exposed group of children whose exposure is 2 times (for benzene) higher than the median or typical exposure.

Exposure to air pollution is widespread and difficult to avoid. Ambient air quality policies will result in lower outdoor concentrations and a lower exposure, but at the same time the

relative importance of indoor air pollution due to indoor sources will increase. In ambient air quality standard setting the exposure indoor to outdoor pollution that has infiltrated is implicit. This study shows that the contribution of this infiltrated outdoor air pollution is different for the different pollutants studied. This is a point of attention in ambient air quality policies, to include the exposure indoors more explicit.

Exact recommendations for precautionary measures to reduce or avoid exposure to certain gases are difficult to make at the moment because no 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 policies. Currently, the federal product policy only regulates bulk concentrations of a product, and no emissions, nor does it link to typical and high exposures. There is limited evidence on the health relevance of these exposures. This requires further toxicological and epidemiological evidence of indoor exposure and effects.

Finally a continued effort to inform the public on good product use to the public is welcomed. A good cooperation and communication with industry to appeal for better labelling and to stimulate the development of innovative and safe product, especially to avoid exposure of children, is the best way forward.

1 DATA INTERPRETATION

1.1 Interpretation indoor and outdoor concentrations

1.1.1 Indoor and outdoor concentrations of gases

1.1.1.1 Dwellings indoor

Bedroom concentrations

Concentrations in bedrooms of 50 houses are presented in Figure 1. These have been reported before (in WP2). Note that the Y-axis has been cut of at 70 µg/m³ because of the visibility of concentrations below 10 µg/m³. Some points of attention:

- Two extreme values are not shown: 1 toluene result (116 µg/m³) and 1 formaldehyde result (124 µg/m³). The extreme value for toluene is above the range for indoor toluene concentrations (20-74 µg/m³) reported in the literature review of work package 1 (Table 6). The extreme value for formaldehyde is within the range of indoor concentrations found in the literature review (10-350 µg/m³).
- The upper values for acetaldehyde and 1,2,4-trimethylbenzene measured in this study (Figure 1) are also above literature values (acetaldehyde: 20-50 µg/m³; 1,2,4-trimethylbenzene: 0-4 µg/m³).
- Concentrations of other gases were within indoor ranges reported in the literature review of WP1 (MTBE: 0,6-40 µg/m³; benzene: 2-30 µg/m³; trichloroethene: 1-10 µg/m³; tetrachloroethene: 0-5 µg/m³; ethylbenzene: 1-138 µg/m³; xylenes: 8-37 µg/m³; styrene: 1-6 µg/m³; p-dichlorobenzene: 2-240 µg/m³; NO_2: 30-100 µg/m³; formaldehyde: 10-350 µg/m³).
- A more than 50-fold range of bedroom concentrations for some substances (e.g. formaldehyde, toluene,…) were found in the 50 investigated dwellings.

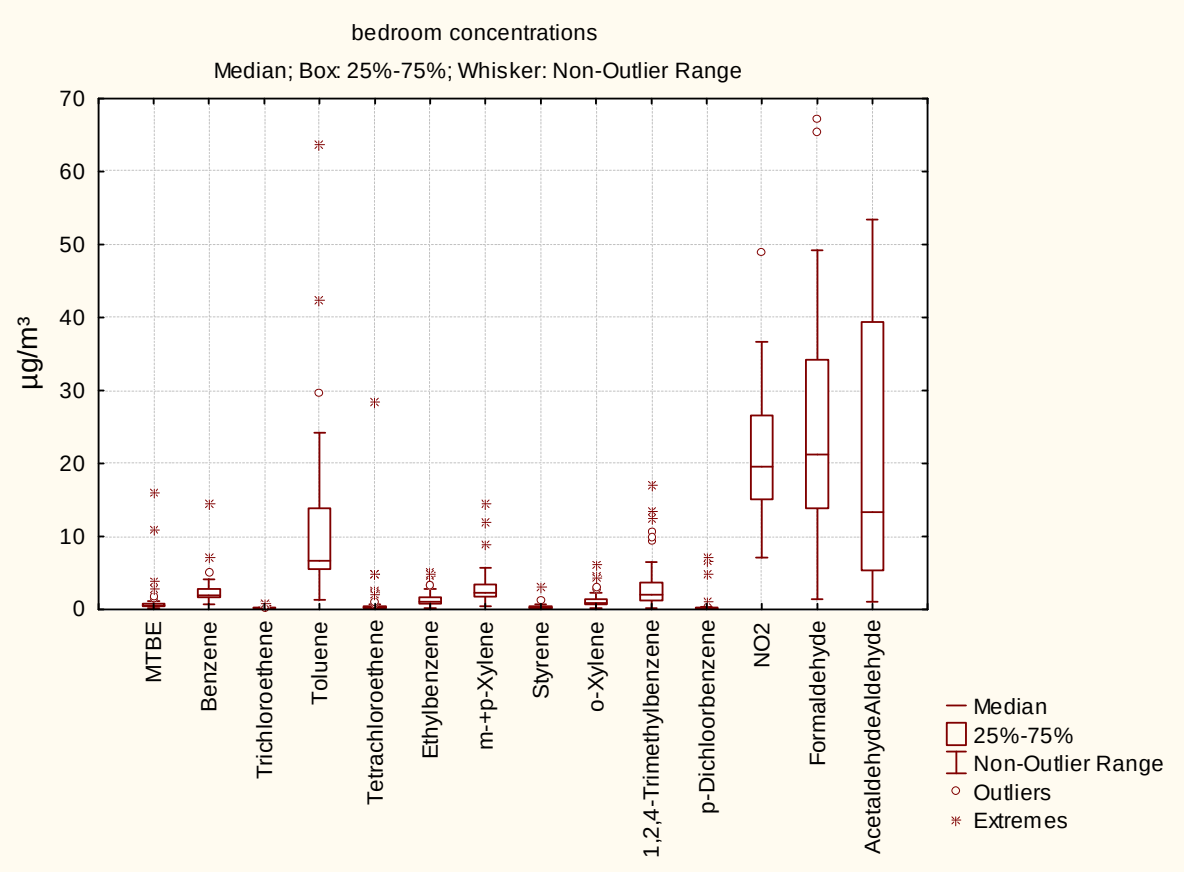

Figure 1: Boxplots of concentrations (7-days averages; expressed as µg/m³) in bedrooms in 50 dwellings

Living room concentrations

An analogous graph for living room concentrations is shown
Figure *2*. To keep the graphs comparable, the Y-axis was also scaled to 70 µg/m³.

- Four values are hence not shown (1 for toluene (122 µg/m³) ; 3 for formaldehyde (82; 90 and 91 µg/m³) and 1 for NO_2 (122 µg/m³).
- More or less the same gases were abundant in bedrooms as in living rooms (i.e. toluene, NO_2, formaldehyde and acetaldehyde).
- The upper concentrations of gases in living rooms exceeded indoor concentrations reported in literature for toluene, tetrachloroethene, 1,2,4-trimethylbenzene, NO_2 and acetaldehyde.
- For other gases, measured concentrations were within ranges reported in the literature review of work package 1.

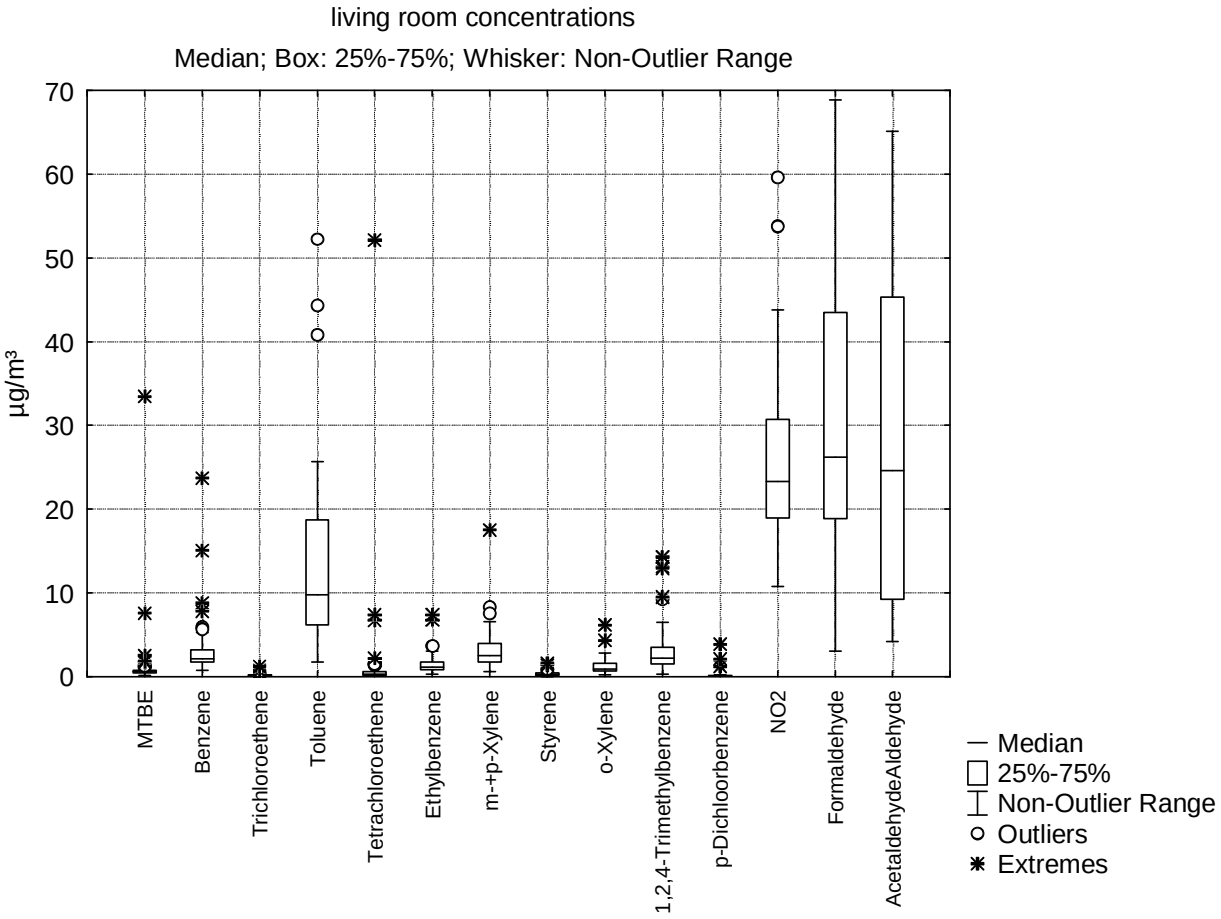

Figure 2: Boxplots of concentrations (7-days averages; expressed as µg/m³) in living rooms in 50 dwellings.

TVOC concentrations: living room and bedroom concentrations

Because of larger values than for individual gases, the box plots for the total sum of volatile organic compounds (TVOCs) are plotted in a separate graph (Figure 3). The values for the sum parameter TVOC are often much larger than the sum of the individual compounds reported in Figure 1 and
Figure *2*. This points out that other, not identified compounds,
also contribute significantly to TVOC.
The TVOC concentration measured in this study fall, except for one measurement (see Figure 3) within the range of literature data (300-1700 µg/m³).

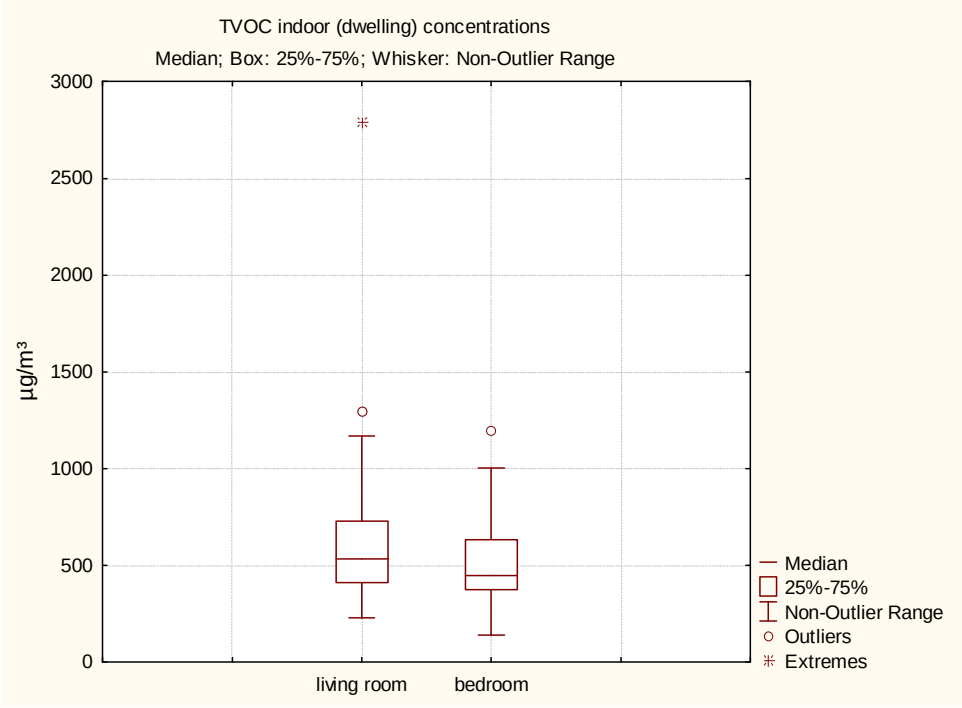

Figure 3: Boxplots of TVOC concentrations (7-days averages; expressed as µg/m³) in living rooms and bedrooms in 50 dwellings

Ratio bedroom -living room concentrations

The ratio of pair wise bedroom and living room concentrations is shown in Table 1. Typical bedroom/living room concentrations were close to 1. This is expressed by the median value in Table 1. These bedroom/living room ratios close to 1 are somewhat surprising because living rooms are expected to be more influenced by indoor sources (heating, cooking, product use,..) and are less ventilated than bedrooms. Hence, one would expect lower bedroom than living room concentrations; however, this was not observed. The data suggest that pollution indoors, measured as a 7-day average concentration, is rather homogenous across different rooms.

Table 1: Ratio's of bedroom to living room concentrations in 50 dwellings ($C_{bedroom}/C_{living\ room}$)

gas	average	P25	P75	median	min	max
MTBE	1,02	0,84	1,10	0,95	0,27	2,91
benzene	0,92	0,81	1,03	0,98	0,08	1,89
trichloroethene	0,99	0,77	1,10	0,95	0,28	2,96
tetrachloroethene	1,03	0,88	1,11	1,00	0,25	2,55
ethylbenzene	1,08	0,78	1,10	0,97	0,29	6,25
m+p xylene	1,01	0,84	1,08	0,99	0,22	2,80
styrene	1,51	0,49	1,11	0,78	0,08	13,08
o-xylene	1,08	0,82	1,15	0,97	0,20	5,13
1,2,4trimethylbenzene	1,02	0,76	1,14	1,01	0,19	2,95
p-dichlorobenzene	4,18	0,93	1,36	1,13	0,66	74,66

TVOC	0,88	0,71	1,00	0,87	0,33	2,54
NO_2	0,86	0,64	0,98	0,81	0,30	1,70
formaldehyde	1,11	0,54	1,41	0,80	0,06	6,14
acetaldehyde	0,80	0,48	1,02	0,70	0,05	2,00

1.1.1.2 Dwellings: outdoor

Dwelling front door concentrations

NO_2, formaldehyde, acetaldehyde and toluene have the highest concentrations in the outdoor environment near dwellings, as could be expected. Similar to indoor concentrations, large variations between the 50 locations were observed. Generally, outdoor concentrations of the gases considered were lower than indoor concentrations.

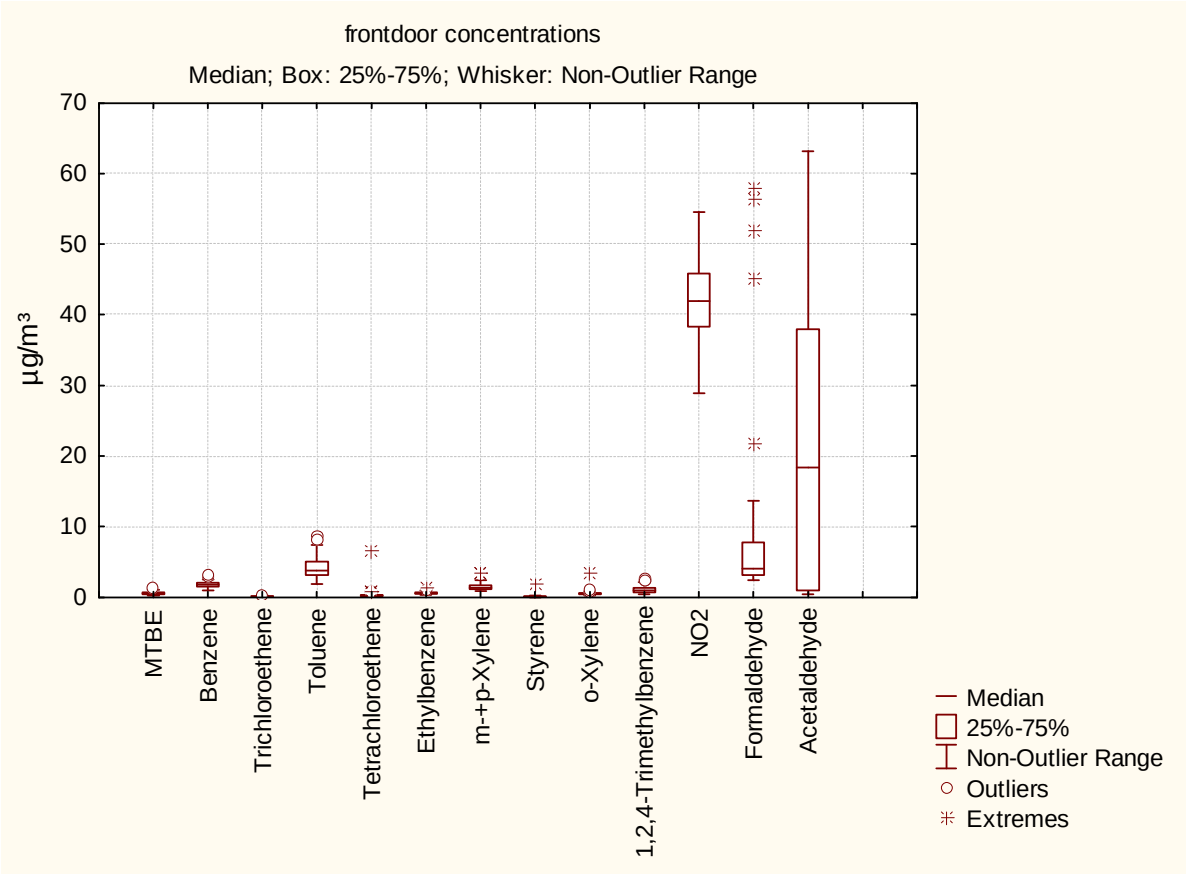

Figure 4: Boxplots of concentrations (7-days averages; expressed as µg/m³) in the front door outdoor environment of 50 dwelling

Dwelling backdoor concentrations

Also backdoor concentrations of NO_2, acetaldehyde and formaldehyde are the highest among the measured gases. The maximal values for outdoor tetrachloroethene, and especially for formaldehyde and acetaldehyde measured in this study (Figure 4 and Figure 5) exceed literature values for outdoor concentrations (tetrachloroethene: generally less than 5 µg/m³; acetaldehyde: average 5 µg/m³; formaldehyde: 1-20 µg/m³). Concentrations of other gases

were within outdoor ranges reported in the literature review of WP1 (MTBE: 0,6-7 µg/m³; benzene: 1-20 µg/m³; trichloroethene: 1-10 µg/m³; toluene: 5-150 µg/m³; ethylbenzene: 0,02-14 µg/m³; xylenes: 2-20 µg/m³; styrene: 1-10 µg/m³; p-dichlorobenzene: <0,6 µg/m³; NO_2: 19-80 µg/m³).

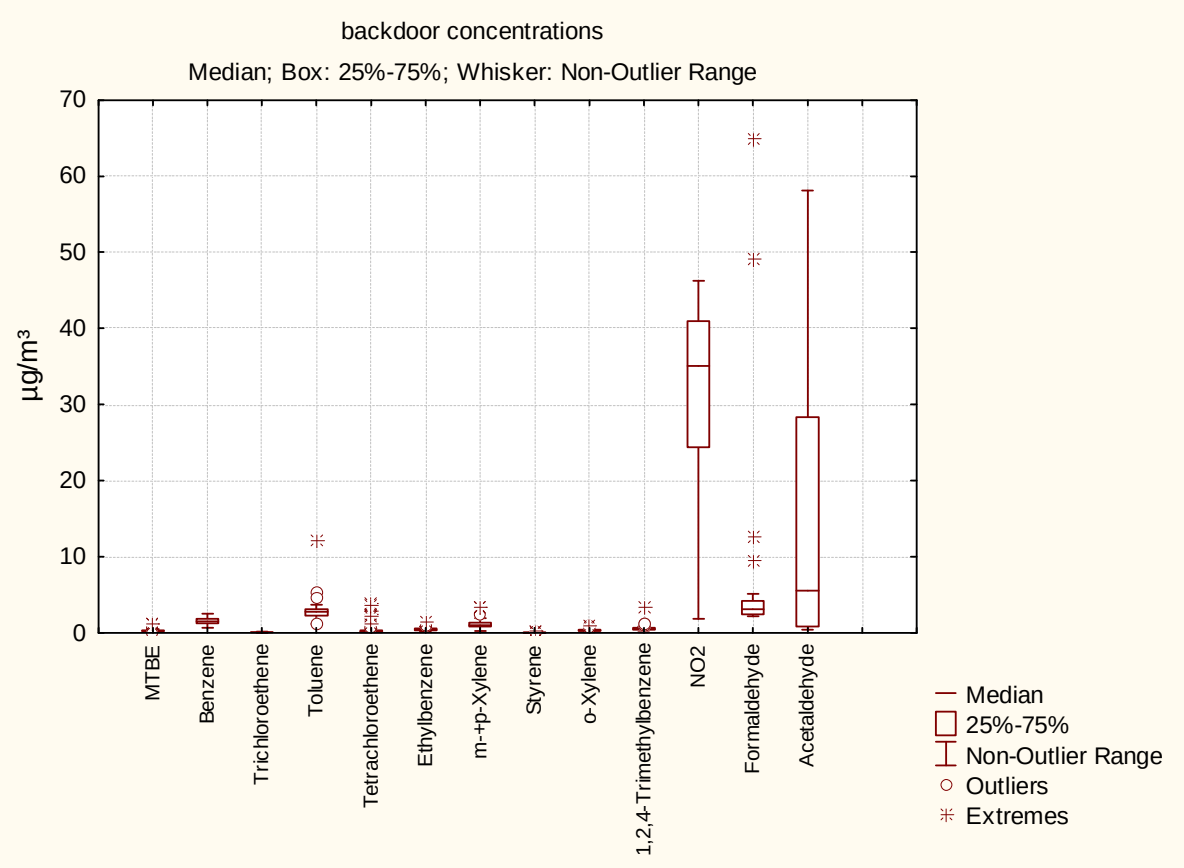

Figure 5: Boxplots of concentrations (7-days averages; expressed as µg/m³) in the backdoor outdoor environment of 50 dwelling

Because of larger values than for individual gases, the box plots for TVOCs are plotted in a separate graph (
Figure 6). Analogously to indoor TVOC concentrations, the values for the sum parameter TVOC are in many cases much larger than the sum of the individual compounds reported in Figure 4 and Figure 5. The outdoor TVOC concentrations are smaller than indoor TVOC concentrations.
The outdoor TVOC concentration measured in this study fall within the range of literature data (20-650 µg/m³).

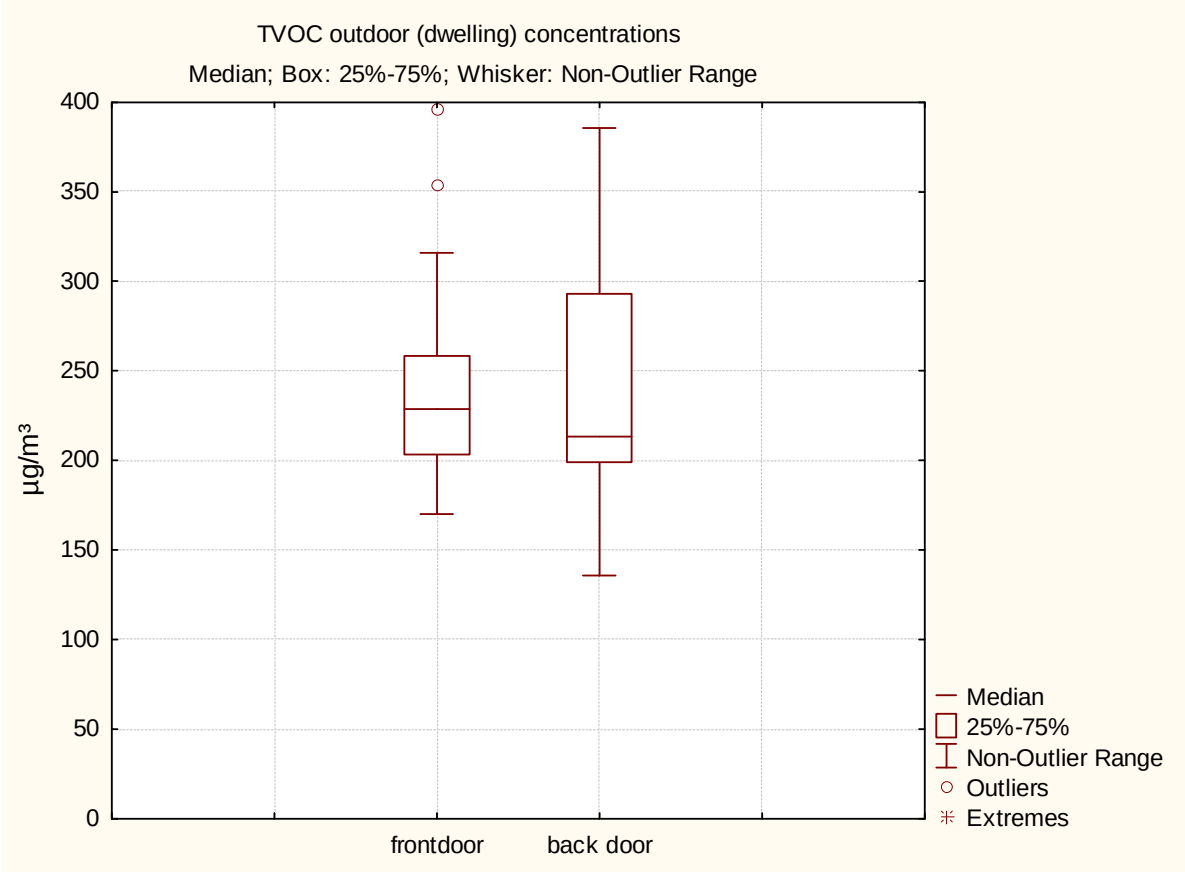

Figure 6: Boxplots of TVOC concentrations (7-days averages; expressed as µg/m³) in the front door and back door environment of 50 dwellings

Ratio front door-back door concentrations (dwellings)

In general, backdoor (BD) concentrations were slightly lower than front door (FD) concentrations (with some exceptions on gas/dwelling combinations). This suggests that the dwelling acts as a barrier for pollutants that are mainly formed at the street. However, the gradient from front door to backdoor depends on the gas. Median BD/FD ratio's were between 0,6-0,8 for MTBE, toluene, ethylbenzene and, 1,2,4 trimethylbenzene; between 0,8-1 for benzene, trichloroethene, tetrachloroethene, m+p xylene, styrene, TVOC, NO_2 and formaldehyde; and slightly greater than 1 for acetaldehyde.

Table 2: Backdoor/front door ratio's of pollutants in 50 dwellings.

	mean	min	P25	median	P75	max
MTBE	0,77	0,26	0,45	0,58	0,75	3,80
benzene	0,91	0,59	0,75	0,86	0,92	2,25
trichloroethene	0,99	0,54	0,90	0,98	1,04	1,60
toluene	0,87	0,32	0,57	0,69	0,82	4,24
tetrachloroethene	2,41	0,51	0,87	0,97	1,03	23,51
ethylbenzene	0,88	0,46	0,65	0,76	0,88	3,11
m+p xylene	0,94	0,30	0,67	0,80	0,89	3,57

styrene	1,13	0,04	0,66	0,95	1,43	4,29
o-xylene	0,89	0,11	0,63	0,79	0,92	3,01
1,2,4 trimethylbenzene	0,92	0,20	0,47	0,61	0,84	7,09
p-dichlorobenzene	-	-	-	-	-	-
TVOC	0,97	0,55	0,90	0,95	1,01	1,71
NO_2	0,91	0,76	0,88	0,93	0,98	1,02
formaldehyde	0,89	0,11	0,73	0,92	1,05	1,49
acetaldehyde	2,41	0,21	0,82	1,10	1,32	14,92

-: below detection limit

1.1.1.3 Other micro-environments

Schools

Schools demonstrated lower ranges of air pollutant concentrations than dwellings. For example, maximum formaldehyde concentrations of 34 µg/m³ for indoor school environments compared to the maximum of 124 µg/m³ in dwellings. The lower maximal concentrations in schools compared to dwellings, is off course partly related to the lower numbers of measured schools (n = 5) than the number of dwellings (n=50). The median values for formaldehyde however were in the same range for schools as for dwellings.
Analogously to dwellings, indoor/outdoor ratio's were above unity for schools. This is in accordance with an earlier study on indoor and outdoor environments in 27 primary schools in Flanders (Stranger, 2005). Stranger (2005) found I/O ratio's exceeding unity for benzene, toluene, ethylbenzene, xylenes, formic acid and acetic acid.

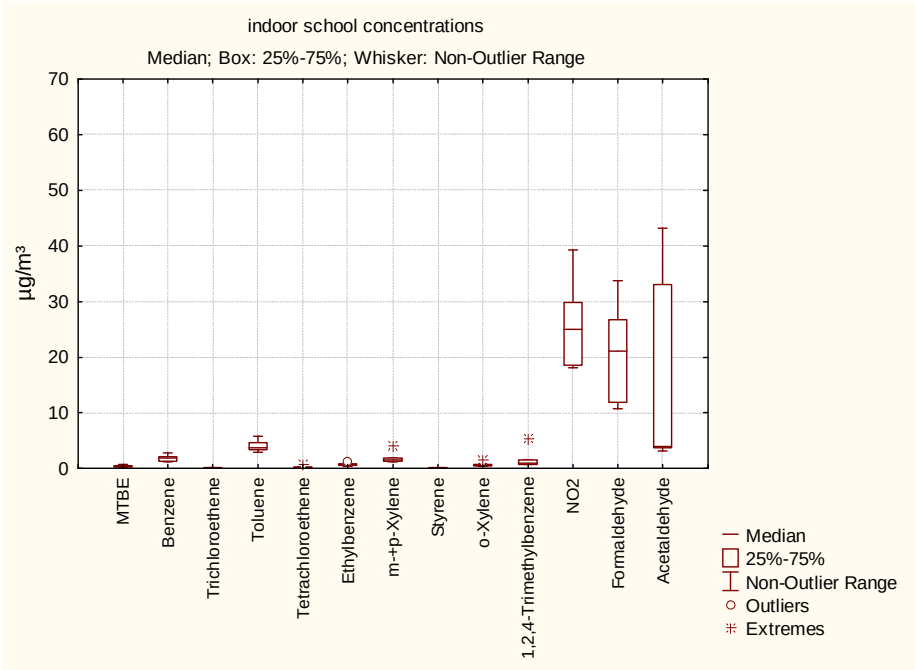

Figure 7: Boxplots of concentrations (7-days averages; expressed as µg/m³) in indoor school environment (5 schools)

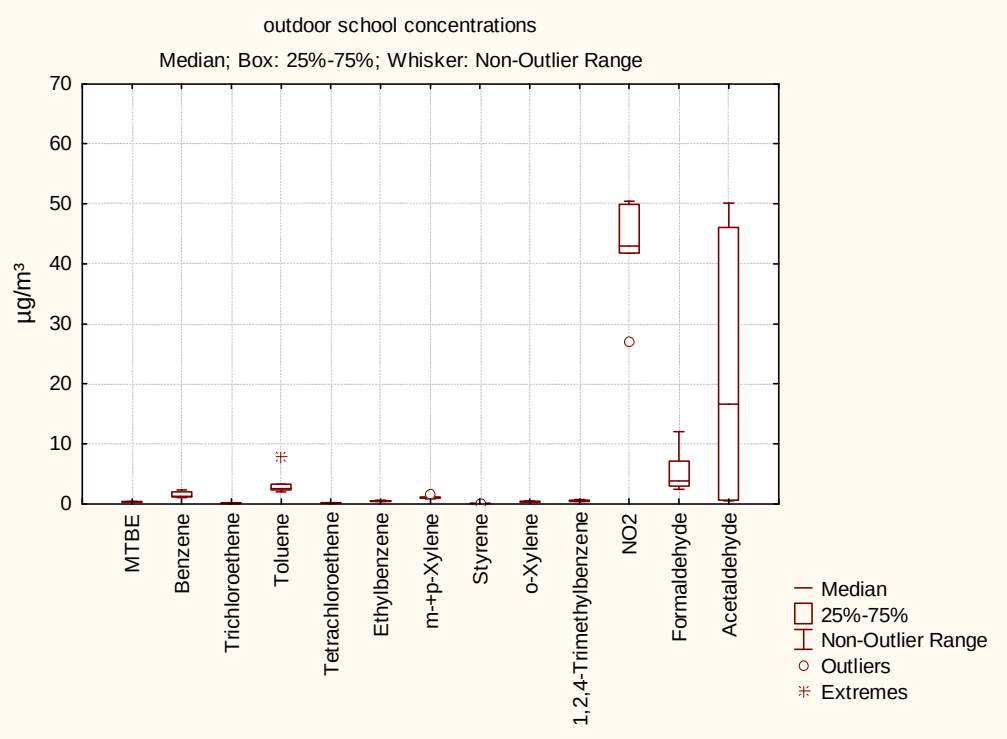

Figure 8: Boxplots of concentrations (7-days averages; expressed as µg/m³) in outdoor school environment (5 schools)

Transport

The range of concentrations indoors of 5 transport modes (car and public transport) are shown in
Figure *9*. In transport indoor environments, much larger NO_2 concentrations were measured than in dwellings and schools.

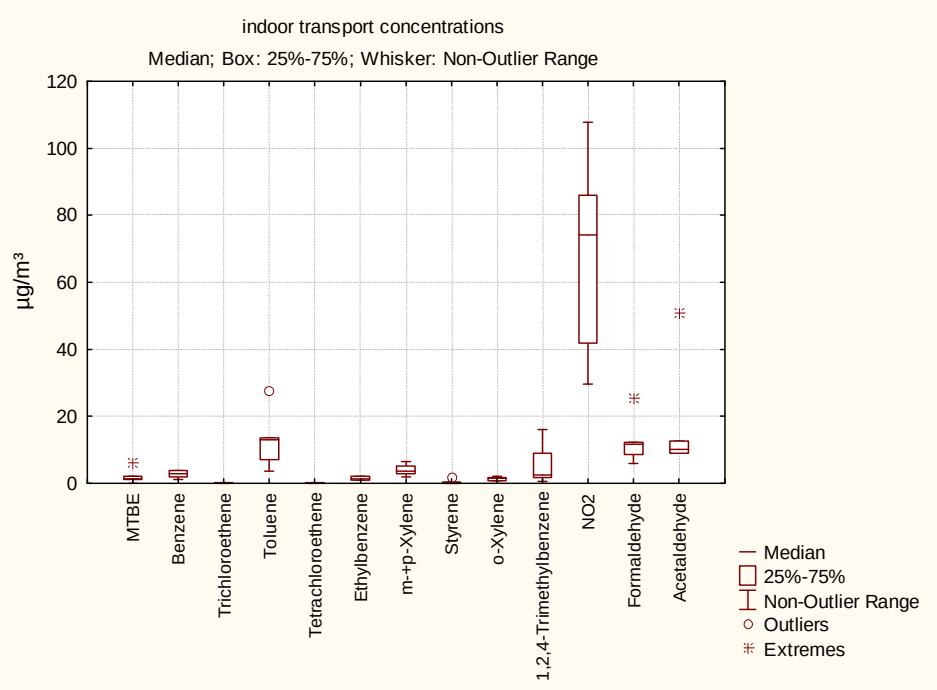

Figure 9: Boxplots of concentrations (7-days averages; expressed as µg/m³) in indoor transport (car and public transport) environments (n=5).

Outdoor transport includes walking and cycling. Here also, only a few sampling points (n = 4) were measured. Larger acetaldehyde concentrations (a marker for diesel traffic) were measured in outdoor transport environments than in any of the other micro-environments (dwellings, schools,…). The comparison of
Figure *9* and
Figure 10 shows that larger NO_2, toluene and benzene concentrations were observed for indoor transport than for than outdoor transport. The measurements were not performed simultaneously nor at the same locations/trajectories. So no further conclusion can be drawn from this observation. It is merely an indication that exposure to these pollutants can vary accordingly.

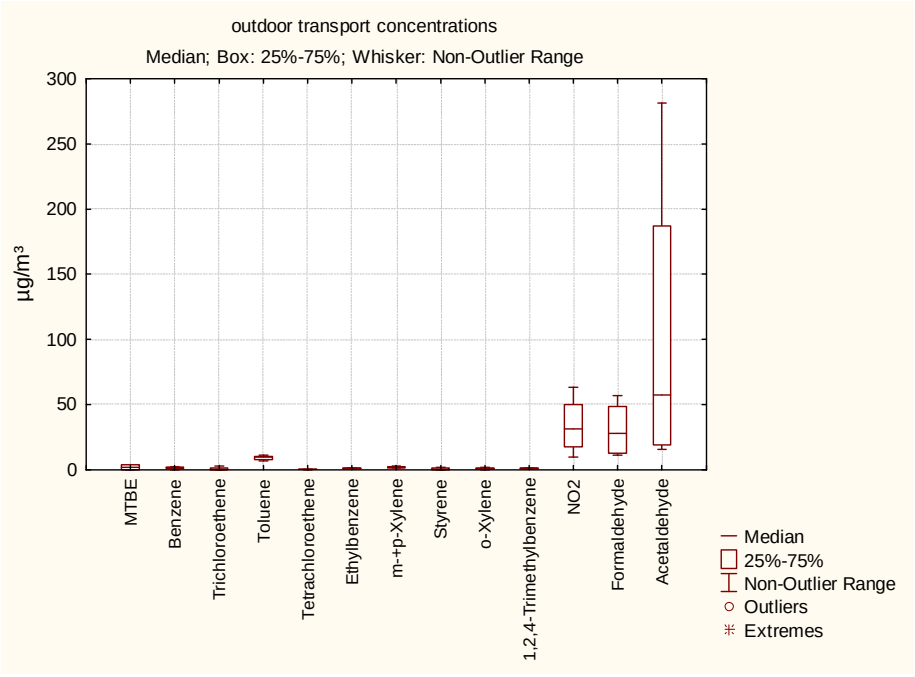

Figure 10: Boxplots of concentrations (7-days averages; expressed as µg/m³) in outdoor transport (walking and cycling) environments (n=4).

Leisure

The maximum value of the Y-axis in
Figure 11 was set at 70 µg/m³. As a consequence one value for NO_2 (143 µg/m³) is not shown. It is noticed that rather large amounts of toluene, xylenes and 1,2,4-trimethylbenzene were measured in indoor leisure environments. This is mainly attributable to one indoor leisure environment (a room in a youth club) and is not systematically for all indoor leisure environments. Given the composition of the air in the youth club it is assumed that smoking (or use of paints) was the source of the gases.

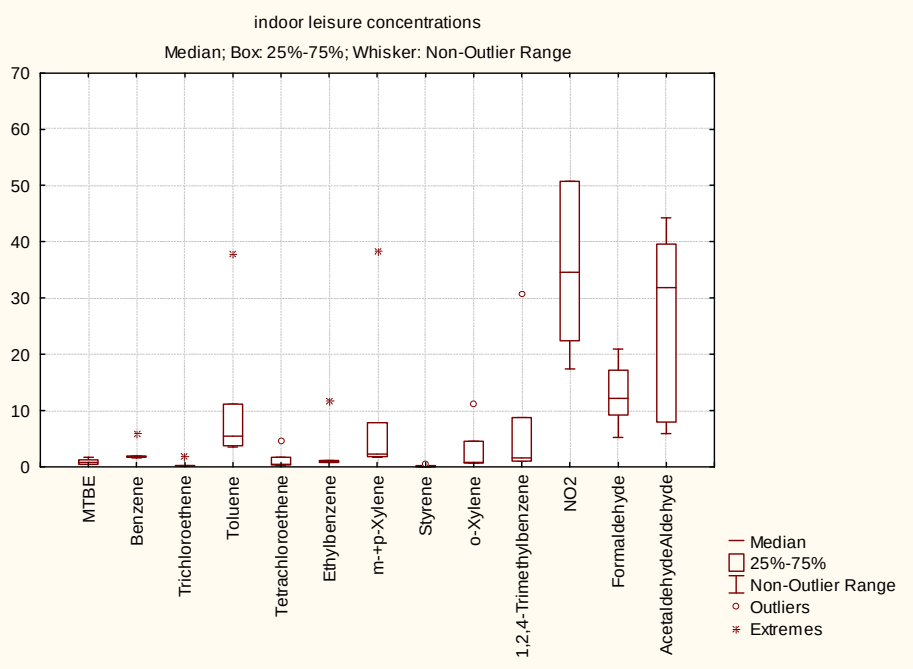

Figure 11: Boxplots of concentrations (7-days averages; expressed as µg/m³) in indoor leisure (including indoor sports) environments (n = 6).

1.1.2 Indoor and outdoor concentrations particulate matter (PM)

Measurements of living room and outdoor PM10 were limited to 16 dwellings; in 32 houses, various PM fractions (PM1, PM10, PM2.5 and TSP) were measured in bedrooms. In bedrooms, PM1 constitutes the largest fraction of fine particles (relative to PM2.5 and TSP). Among all settings, outdoor PM10 concentrations were higher than indoor PM10 levels (indoor levels were calculated as the average of living room and bedroom concentrations).

On average, indoor PM levels were 3-fold lower indoors than outdoors (see also in 1.1.5.1). From experience and literature these results are difficult to interpret. In well ventilated houses, an equilibrium exists between indoor and outdoor. In the presence of indoor sources and in poorly ventilated buildings PM concentrations are generally higher than outdoors. The fact that measurements were mainly performed in winter, when ventilation is low, and in the absence of PM sources can explain the low PM concentrations. Moreover, from the time-resolved GRIMM data (e.g. Figure 12), it can be seen that re-suspension of PM cause TSP concentrations to peak very briefly, while during the day when all occupants are gone to work, to school or to the day care concentrations remain stable but low. More research, calibrated equipment per location and longer time series are needed to explain these findings.

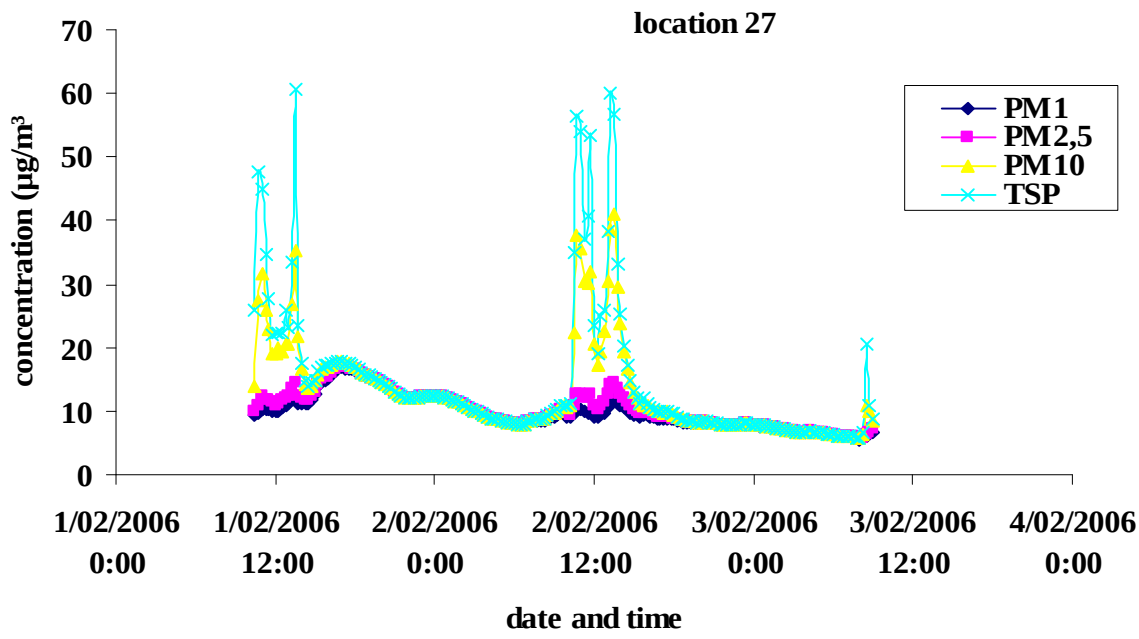

Figure 12: Time-resolved GRIMM PM data show a strongly fluctuating pattern, with peaks around the noon.

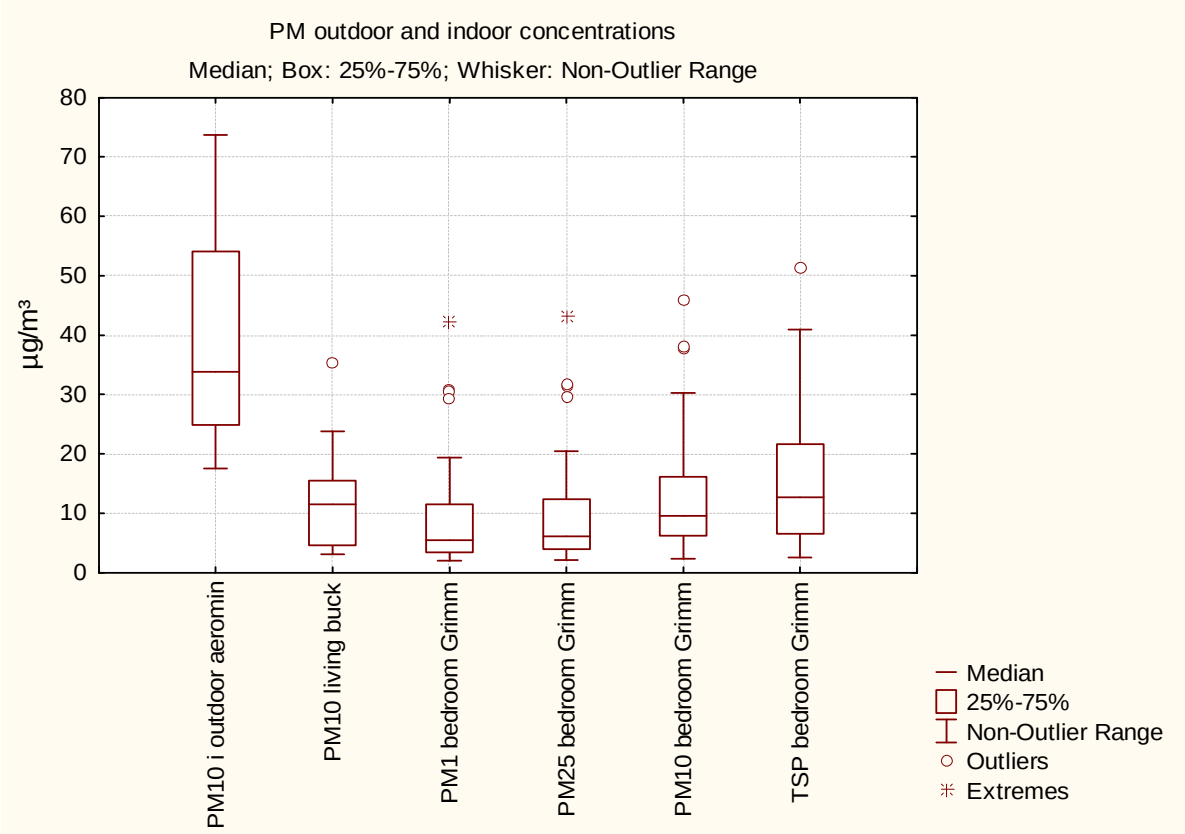

Figure 13: Boxplots of PM (PM1, PM2.5, PM10 and TSP) concentrations in dwellings (bedroom, living room and outdoors)

1.1.3 Comparison between different location types (hotspot, urban background, rural background)

The statistical comparison of concentrations between different location types was performed using the statistical tool Statistica (version 7). The non-parametric Kruskal-Wallis Anova test was used because the data were not normally distributed. Only dwellings were included in this analysis.

1.1.3.1 Gases

Comparison of outdoor concentrations

Anova was performed to test whether location type (urban background (UB), hotspot (HS), rural background (RB)), as a proxy for traffic density, affected outdoor concentrations. Mean outdoor concentrations by location type and significance of the location type on outdoor concentrations are given in Table 3.

Table 3: mean outdoor concentrations by location type (expressed as µg/m³). 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)*

	UB	HS	RB
MTBE	**0,4^{AB}**	**0,6^{A}**	**0,3^{B}**
benzene	**1,6^{A}**	**2,0^{B}**	**1,4^{A}**
trichloroethene	**0,13^{A}**	**0,12^{A}**	**0,07^{B}**
toluene	**3,5^{AB}**	**4,3^{A}**	**2,5^{B}**
tetrachloroethene	**0,6^{A}**	**0,5^{A}**	**0,1^{B}**
ethylbenzene	0,5	0,7	0,5
m+p xylene	1,3	1,5	1,1
styrene	0,1	0,1	0,0
o-xylene	0,5	0,5	0,4
1,2,4trimethylbenzene	**0,8^{A}**	**1,0^{A}**	**0,4^{B}**
p-dichlorobenzene	0,0	0,0	0,0
TVOC	229,6	244,6	263,4
NO_2	**38,4^{A}**	**47,5^{B}**	**27,5^{A}**
formaldehyde	11,8	5,7	8,8
acetaldehyde	18,5	23,1	15,4

* the following example for MTBE explains the coding system: the letter A is attributed to UB and HS which means no significant differences between UB en HS; the letter B is attributed to UB and RB, thus no significant differences between UB and RB. HS and RB are significantly different from each other since they are not marked with a common letter.

For some gases, concentrations were affected by the location type, concentrations for HS locations being highest, followed by urban background, and by rural background. As expected, traffic related compounds (MTBE, benzene, toluene and NO_2) are significantly higher in HS compared to RB. 1,2,4-trimethylbenzene might also be associated with traffic, giving significantly lower concentrations in rural background locations. Both trichlorethene and tetrachloroethene are not typically associated with traffic proximity, and the significantly lower results in rural background locations cannot be explained.

The summary statistics are given in Table 3 and graphs in Figure 14 for a few selected pollutants are presented for the sake of illustration.

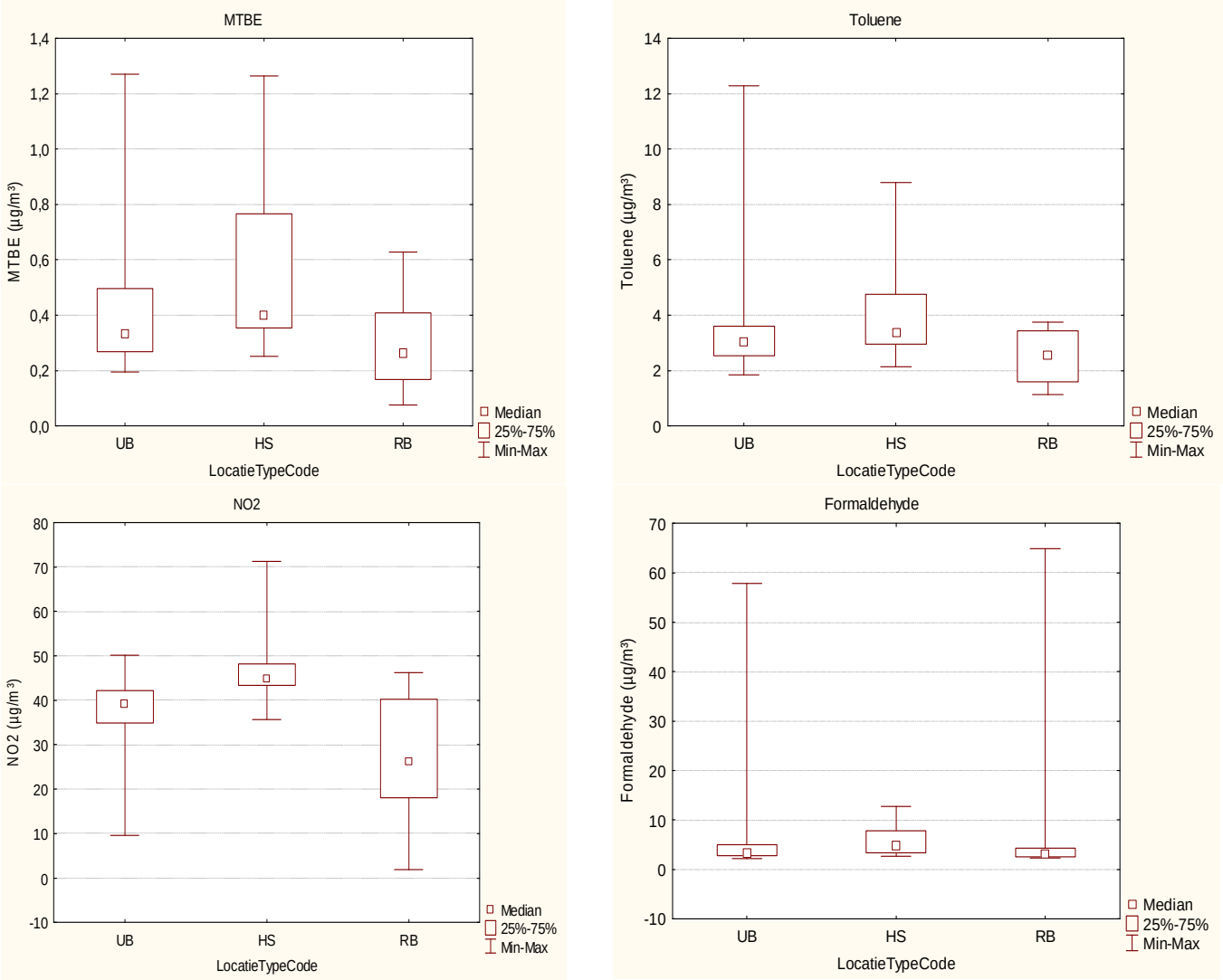

Figure 14: Comparison of outdoor concentrations (MTBE, toluene, NO_2 and formaldehyde) between urban background (UB), hot spot (HS) and rural background (RB) regions.

Comparison of indoor concentrations

Anova analyses were also performed to test if the location type (UB, HS, RB) affected indoor concentrations. Mean indoor concentrations by location type and significance of the location type on indoor concentrations are given in Table 4.

Table 4: Mean indoor concentrations (bedroom and living room) by location type (expressed as µg/m³). 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)

	UB	HS	RB
MTBE	1,53	0,75	1,52
benzene	**2,75^{A}**	**4,16^{B}**	**2,15^{A}**
trichloroethene	**0,22^{A}**	**0,28^{A}**	**0,12^{B}**
toluene	13,31	10,58	19,03
tetrachloroethene	**0,76^{A}**	**4,13^{B}**	**0,23^{C}**
ethylbenzene	1,45	1,47	1,66
m+p xylene	3,23	2,97	3,50
styrene	0,30	0,23	0,36
o-xylene	1,27	1,24	1,31
1,2,4trimethylbenzene	3,47	3,37	3,99
p-dichlorobenzene	**0,61^{A}**	**0,10^{B}**	**0,06^{B}**
TVOC	510,5	605,5	700,7
NO_2	**24,3^{A}**	**31,7^{B}**	**17,6^{C}**
formaldehyde	**34,5^{A}**	**36,9^{AB}**	**21,1^{B}**
acetaldehyde	17,9	24,4	16,4

For some traffic pollutants, higher indoor concentrations in HS than in RB and UB (e.g. NO_2) were observed. Although indoor sources of NO_2 could be present, this does not alter the significantly higher 'hot-spot' result. 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 MTBE the absence of a significant difference indoors, for a pollutant that is only generated outdoors (by petrol cars) is puzzling. This might be an indication of indoor MTBE sources.
Trichlorethene and tetrachloroethene are still significantly lower in rural background locations, but this can now be explained partly by a few very high results for tetrachloroethene indoors.

For other, more indoor generated pollutants (xylenes, …) no effect of location type on indoor concentration levels was observed.
For a few selected gases, the comparison of indoor concentrations between location types is shown in Figure 15.

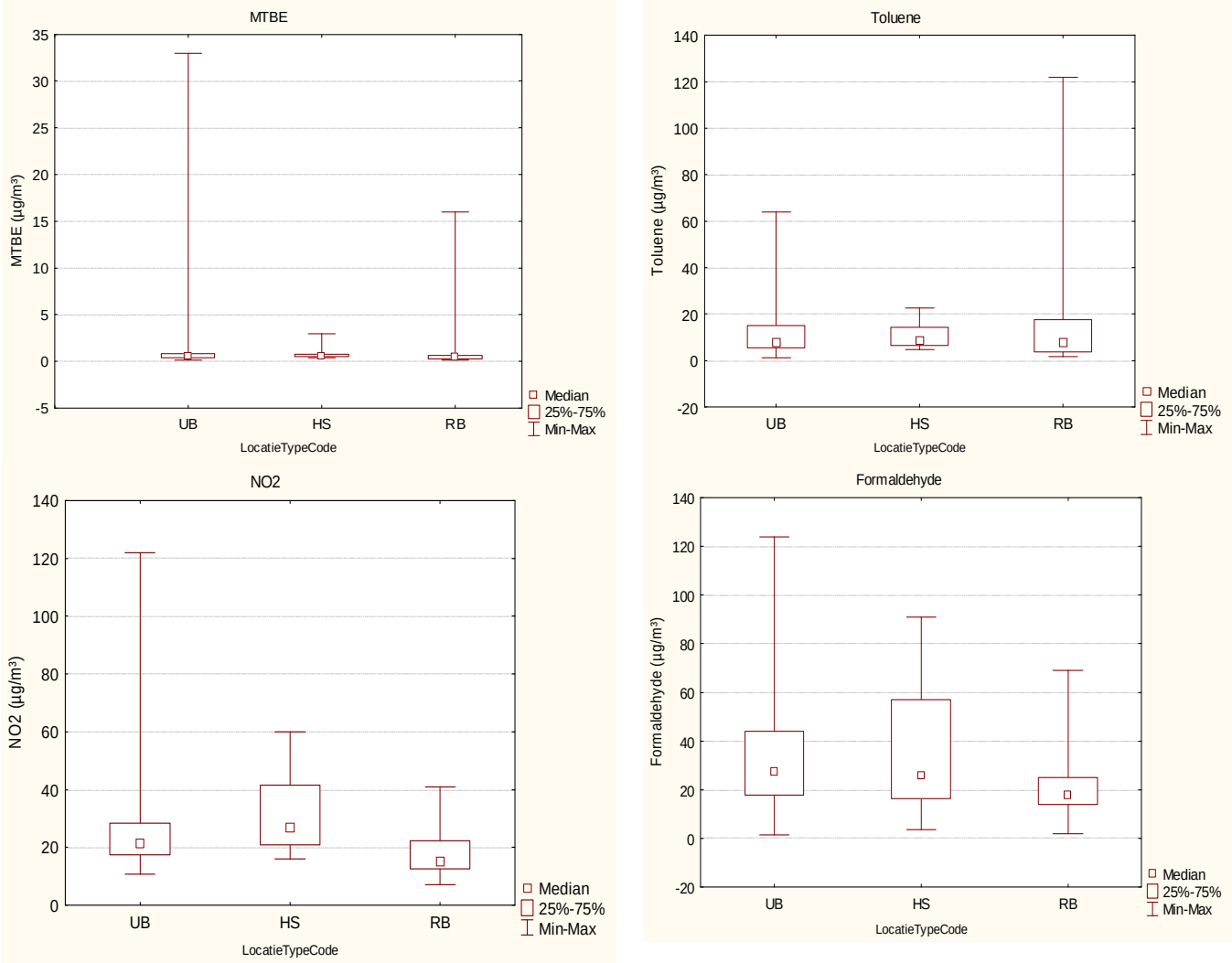

Figure 15: Comparison of indoor concentrations (MTBE, toluene, NO_2 and formaldehyde) between urban background (UB), hot spot (HS) and rural background (RB) regions

1.1.3.2 Particulate matter

Comparison of outdoor concentrations

Only 18 locations were sampled for PM, and the distribution over the 3 categories was rather poor (n = 13 for UB; n = 4 for HS and n = 1 for RB), which makes it difficult to detect statistical significant differences. Mean outdoor PM10 concentrations decreased as expected: concentrations were highest for HS (43,6 µg/m³), followed by UB (38,3 µg/m³), and then by RB (24,9 µg/m³) (Figure 16). However, differences between location types were not statistically significant.

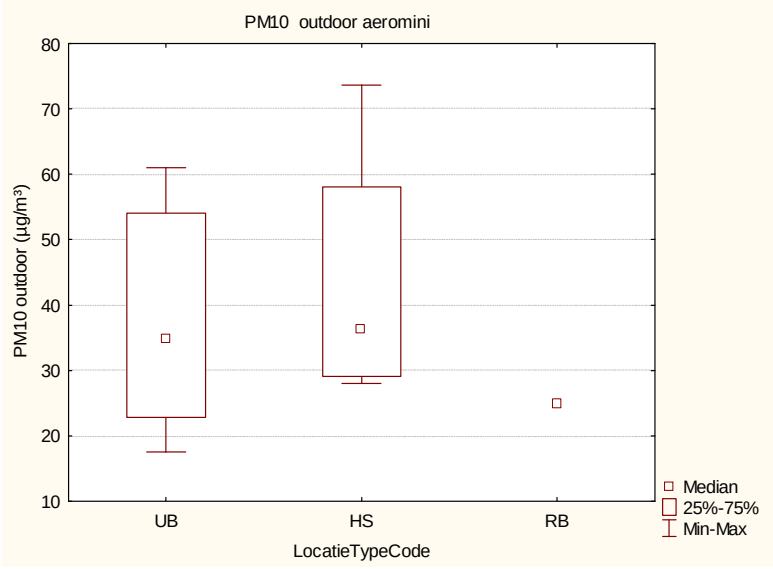

Figure 16: Comparison of outdoor concentrations (PM10) between urban background (UB), hot spot (HS) and rural background (RB) regions

Comparison of indoor concentrations

Analogously to outdoor concentrations, PM indoor concentrations were higher in HS areas than in UB areas (Figure 17), though none of the differences were statistically significant. The higher PM levels for RB in this analysis probably rely in the very limited numbers of RB cases (n=1 for living rooms and n = 2 for bedrooms) in the dataset, and thus might poorly represent the 'real' average RB concentrations. (Representativeness of UB (n = 15 for bedroom concentrations and n = 2 for living rooms) and HS (n = 10 for bedrooms and n = 7 for living rooms))

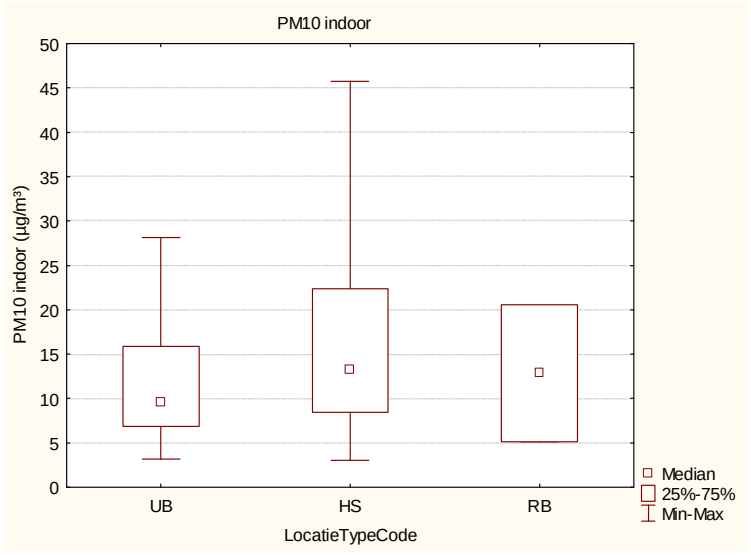

Figure 17: Comparison of indoor (bedroom + living room) concentrations (PM10) between urban background (UB), hot spot (HS) and rural background (RB) regions

The data are summarized in Table 5.

Table 5: Mean indoor (bedroom/living room) PM (PM1, PM2.5, PM10 and TSP) concentrations in different location types.

location	PM fraction	UB	HS	RB
bedroom	PM1 Grimm	7,2	13,8	16,7
	PM10 Grimm	10,8	17,9	21,6
	PM2.5 Grimm	7,7	14,4	17,6
	TSP Grimm	12,7	20,7	22,9
living room	PM10 Buck	15,3	9,6	3,1

1.1.4 Correlation between total indoor concentrations and indoor activities/home characteristics

1.1.4.1 Correlation matrix for ventilation habits and (combustion) appliances

A correlation analysis was done to detect relationships between indoor concentration and indoor activities, indoor sources or building properties. Data were derived form questionnaires. A correlation matrix was calculated between indoor activities, sources or properties and indoor concentrations (Table 6). Indoor concentrations in this analysis are the average of bedroom and living room concentrations. This correlation analysis was based on the number of filled in questionnaires for the (building/source/activity) parameter that is assessed. For example, out of the 44 received questionnaires, the parameter "hours of total central heating use" was filled in 37 times. The correlation analysis was then performed on these 37 cases. Six dwellings (# measured dwellings = 50) were not included in this analysis because of the absence of questionnaires.

It is likely that participants only filled in the questionnaires when the answer was different from zero. For example, only for 3 houses, the parameter "# hours fireplace use" was filled in. Probably, the number of hours fireplace use in the other houses was zero. For the houses without fireplace (n = 38), we can be sure of that. But for 3 out of 6 houses with a fireplace; the variable "# hours fireplace" use was not filled in, and we assume it to be zero. However, we cannot be sure of that.

To illustrate the importance of either (1) ignoring the not answered questions and (2) attributing zero value to not filled in answers, the correlation between indoor toluene concentrations and hours of woodstove use is plotted in Figure 18.

Omitting not-filled in answers, only 3 valid cases remain, resulting in an extreme high correlation coefficient (r = 1,00). This correlation coefficient drops to r = 0,19 (and the correlation is not significant at the p=0,05 level) if in the remaining cases a value of zero is attributed (0 hours fireplace use/week).

The comparison of analysis (1) and (2) shows, first, that fireplace use might be, at least partly, explanatory for indoor toluene concentrations. However, this analysis is based on too few cases to validate this hypothesis. Second, other variables than fireplaces use are stronger influencing factors for indoor toluene concentrations given the larger variations in indoor toluene concentrations in houses without fireplace than in houses with a fireplace.

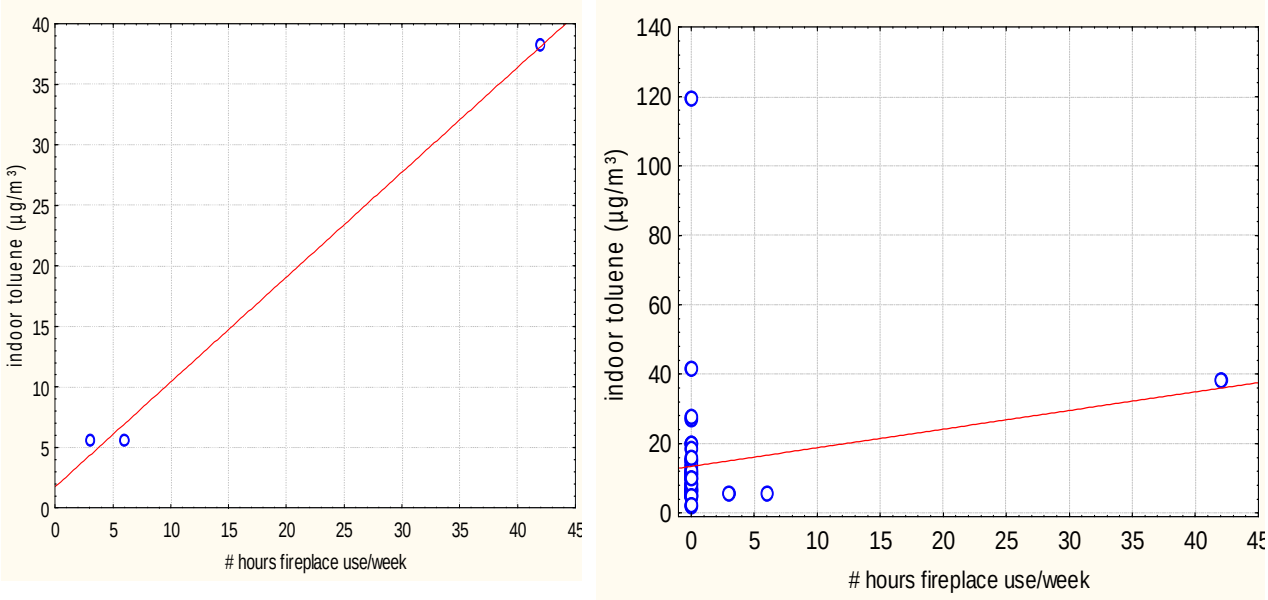

Figure 18: Correlation between answered # hours fireplace use (h/week) with indoor toluene concentrations omitting not answered questionnaires (left graph) and attributing 0 for # hours fireplace use for not filled in answers (right graph).

It was decided to perform the correlation analysis by omitting not-filled in answers (Table 6). This is the most likely method to detect source- or activity-concentrations relationships. In a few cases the correlation is significant, meaning that a higher use of a specific source, or a more frequent activity is correlated with a higher concentration indoors. It does not mean that the correlation is causal.

*Table 6: Correlation matrix between indoor concentrations (expressed as µg/m³) and indoor activity frequencies (expressed as hours/week). Non-filled in answers (#: in the first column) were omitted in the analysis. Significant (p<0,05) correlations are marked in **bold italic**.*

	# answered (total : 44)	MTBE	Benzene	Trichloroethene	Toluene	Tetrachloroethene	Ethylbenzene	m-+p-Xylene	Styrene	o-Xylene	1,2,4-Trimethylbenzene	p-Dichloorbenzene	TVOC	NO_2	Formaldehyde
Central Heating	37	-0,30	-0,05	-0,09	-0,33	0,09	-0,20	-0,31	0,08	-0,32	-0,18	-0,12	-0,15	-0,1	-0,15
Stove	9	0,22	0,25	0,24	0,53	0,75	0,07	0,17	***0,83***	0,74	0,37	***0,92***	0,41	-0,3	0,76
Open Fire	7	***1,00***	0,99	-0,95	***1,00***	-0,65	***1,00***	***1,00***	0,31	***1,00***	***1,00***	-0,49	0,99	-1,0	1,00
Other heating	8	0,64	0,11	0,20	0,30	0,54	0,62	-0,21	0,73	-0,20	-0,01	-0,60	0,91	-0,9	-0,40
hot water production with gas	27	0,02	-0,15	-0,19	-0,03	-0,06	0,02	0,01	0,60	0,10	-0,17	0,16	0,19	-0,4	0,01
other hotwaterproduction type	8	-0,49	-0,59	-0,40	-0,22	-0,18	0,72	***-0,82***	-0,10	-0,68	-0,33	-0,43	-0,48	-0,6	0,01
cooker	38	0,02	0,01	0,18	-0,21	0,24	-0,05	-0,05	-0,13	-0,06	-0,05	***0,56***	-0,01	0,2	0,03
oven	18	-0,12	-0,09	0,18	-0,27	-0,18	-0,04	-0,07	-0,14	-0,04	0,14	***0,90***	0,00	0,3	0,03
kitchen fan	34	-0,04	-0,07	-0,08	-0,05	-0,02	-0,02	-0,07	0,31	-0,08	-0,12	-0,05	0,05	-0,2	-0,08
Open Windows Or Doors at The Front Side	19	-0,20	0,12	-0,17	-0,12	-0,17	-0,20	-0,25	-0,21	-0,24	0,18	-0,09	-0,11	0,4	0,07
Open Windows Or Doors at The Back Side	33	-0,07	-0,05	-0,12	***0,66***	-0,11	0,28	0,07	0,17	0,09	***0,46***	-0,07	0,29	-0,2	-0,12
Ventilation Grids or Ventilation Fans in Use: Ground Floor	9	-0,59	-0,29	0,38	-0,17	-0,28	0,18	0,10	0,05	0,21	0,64	-0,21	-0,14	-0,4	0,28
Ventilation Grids or Ventilation Fans in Use: First Floor	5	0,10	0,63	0,63	0,29	0,25	0,67	0,66	0,27	0,47	0,32	0,25	0,56	-0,7	-0,63
Other Means of Ventilation	6	0,18	-0,42	-0,55	0,41	-0,73	0,45	0,72	0,08	0,54	-0,05	-0,44	0,19	-0,2	-0,34
# persons in the dwelling	33	0,07	-0,16	-0,15	-0,01	0,02	0,03	-0,03	-0,02	-0,03	-0,05	0,06	-0,05	0,1	-0,17

The main conclusions of this correlation analysis are:

- Central heating systems are unlikely to contribute strongly to indoor pollutants given the weakly negative (not-significant) relations between central heating use duration and indoor pollutants.
- Indoor concentrations of tetrachloroethene, m+p-xylene and o-xylene and TVOC's (and p-dichlorobenzene) are positively ($p<0,05$) correlated with stove use. The latter 3 compounds are combustion products.
- Open fire place use contributes positively to indoor concentrations of MTBE, ethylbenzene, m+p-xylene, o-xylene, formaldehyde and 1,2,4-trimethylbenzene. It is however noted that the correlation might be largely influenced by one house with high fireplace use duration. Such correlations that are mainly based on single points should be interpreted with care.

  In addition, the correlations do not always reflect a direct source-concentration effect. For example, open fireplace use is commonly accompanied by high air exchange rates. These high air exchange rates are probably explanatory for the outdoor to indoor movement of MTBE rather than MTBE being emitted due to wood burning.
- Furnaces and oven use durations are positively correlated with indoor p-dichlorobenzene concentrations, although these are not sources of p-dichlorobenzene.
- Ventilation durations were generally slightly negatively (not-significantly) correlated with indoor concentrations, as expected for indoor generated pollutants. However, for a few (generally indoor generated) pollutants, i.e. toluene, tetrachloroethene and styrene, a positive ($p<0,05$) correlation between concentrations and ventilation duration was observed. No explanation for this could be found.

The corresponding correlation matrix for indoor PM concentrations and indoor activity durations is listed in
Table 7.

Table 7: : Correlation matrix between indoor PM concentrations (expressed as µg/m³) and indoor activity frequencies (expressed as hours/week). Non-filled in were omitted in the analysis. Significant (p<0,05) correlations are marked in bold italic.

	PM10 living buck	PM1 bedroom Grimm	PM2.5 bedroom Grimm	PM10 bedroom Grimm	TSP bedroom Grimm
Central Heating	-0,28	-0,36	-0,38	***-0,47***	***-0,50***
Stove		***1,00***	***1,00***	0,92	0,76
Open Fire					
Other heating	-1,00	***1,00***	***1,00***	***1,00***	***1,00***
hot waterproduction with gas	-0,61	0,33	0,32	0,24	0,19
other hotwaterproduction type	-0,34	0,79	0,80	0,80	0,83
cooker	-0,07	-0,26	-0,26	-0,25	-0,24
oven	-0,39	-0,09	-0,10	-0,15	-0,18
kitchen fan	-0,47	0,36	0,37	0,40	0,37
Open Windows Or Doors at The Front Side	0,89	0,24	0,24	0,22	0,20
Open Windows Or Doors at The Back Side	0,22	0,11	0,11	0,10	0,08
Ventilation Grids or Ventilation Fans in Use: Ground Floor	0,55	-0,37	-0,38	-0,51	-0,61
Ventilation Grids or Ventilation Fans in Use: First Floor		0,86	0,94	0,25	0,05
Other Means of Ventilation	1,00	-0,44	-0,44	-0,52	-0,57

# persons in the dwelling	-0,06	-0,02	-0,02	-0,04	-0,05

No significant correlations were observed for living room PM concentrations and indoor activity durations. The highly significant and positive correlations between 'other heating' or 'stove use' and bedroom PM are driven by only 3 data points.

1.1.4.2 Significance test for indoor characteristics.

Based on the information retrieved from the questionnaires, statistical analyses (Anova) were done to detect if there was an effect of indoor characteristics on concentrations. Basically, the indoor characteristics were dichotomic (present or not present), and statistical comparison (Anova) was used to test if concentrations between the 2 classes were significantly different.

Results are presented in
Table 8. This table is limited to combinations of indoor-characteristics and pollutants that are hypothetically related. The potential indoor and outdoor sources listed in Table 6 of work package 1 were used to select the combinations.
Results are expressed as the ratio of mean concentration in homes where the specific source was present to the mean concentration in homes where that source was not present. A ratio larger than 1 points out that the indoor source contributes to indoor levels.

Table 8: Ratio's of mean concentration of pollutant x in dwellings in the presence of the indoor source to the mean concentration of pollutant x in dwellings in absence of the indoor source. Ratio's are marked in bold if the concentration between the two groups (absence versus presence) was statistically significant (Anova, P<0,05).

factor	**PM1**	**PM2.5**	**PM10**	**formaldehyde**	**acetaldehyde**	**NO_2**	**TVOC**	**benzene**
chipboard, parquet, fibreboard				0,77			1,05	
cavity wall insulation				1,15			1,14	
gas furnace				0,91	0,71		0,89	
natural gas heating system				0,94	0,79	1,36	0,88	
hot water system on natural gas				0,99	0,56	0,97	1,07	
glue				0,81			**2,20**	1,00
stain remover				0,81			**2,20**	1,00
gasoline, exhaust gases							1,02	1,02
vinyl wallpaper				1,07				
sealing products								**5,41**
carpets				**0,62**				
curtain material								
varnish				0,69				0,76
printer								1,13
photocopier								0,84
insecticide								-
smoking	**2,89**	**2,84**	**2,51**				0,78	
heating	-	-	-			-		

Table 8: continued

	toluene	**ethylbenzene**	**o-xylene**	**m+p xylene**	**styrene**	**1.2.4 trimethylbenzene**	**p-dichlorobenzene**	**trichloroethene**	**tetrachloroethene**
varnish	0,64		1,56	1,47					
cork floor	0,70		0,57	0,60					
parquet	1,54		1,22	1,25					
linoneum			0,95	0,95					
paint	0,67	1,26	1,06	1,02				0,81	
glue		1,56							
gasoline, motor exhaust		1,20							
furniture		0,95							
polishing wax		1,05							
vinyl wallpaper						0,45			
sealing products		0,54	0,61	0,54					
carpet					0,93	0,48			0,38
chipboard, parquet, fibreboard		0,95							
curtain material	2,18								3,98
computer	1,70	1,24	1,11	1,02					
printer	1,17	0,77	0,89	0,90	0,99			0,86	
photocopier	0,54	0,65	0,74	0,78	0,49			0,66	
smoking	**3,68**	0,78	0,74	0,78	1,91				
insecticide							-		
maintenance products								0,64	

For only very few indoor characteristics, there was a statistical difference in indoor concentration between the presence and absence of the indoor characteristic. Particulate matter concentrations in bedrooms (PM1, PM10 and PM2.5) were significantly elevated in houses where people had been smoking compared to houses without smoking inhabitants. The same pattern was observed for indoor toluene concentrations. Effects of other indoor sources on indoor pollutants were less clear:

- For most pollutants, the effect of indoor sources on indoor concentrations was insignificant. A possible reason is that the indoor concentrations in this study reflect 7-day averaged sampling periods, that do not reveal peak concentrations released at the moment of product use (e.g. heating, painting, …).
- The significant effects of glue and stain remover on TVOC should also be interpreted with caution: this ratio is derived from one dwelling where glue and stain remover was used compared to 43 dwellings without use of stain remover and glue. The same imbalance between absence and presence of sealing products might explain the significant effect of sealing products on indoor benzene concentrations.
- A larger dataset, and a balanced distribution of absence/presence of indoor sources is desirable to validate these observations.
- The distance of the dwelling to the road did not affect significantly indoor PM concentrations.

1.1.5 Infiltration of outdoor generated pollutants to the indoor environment

1.1.5.1 Ratio indoor/outdoor (I/O) concentrations in dwellings

Gases

Indoor concentrations were, irrespective of the substance, generally higher indoors than outdoors (Figure 19). The lowest I/O ratio's were observed for NO_2, followed by MTBE.

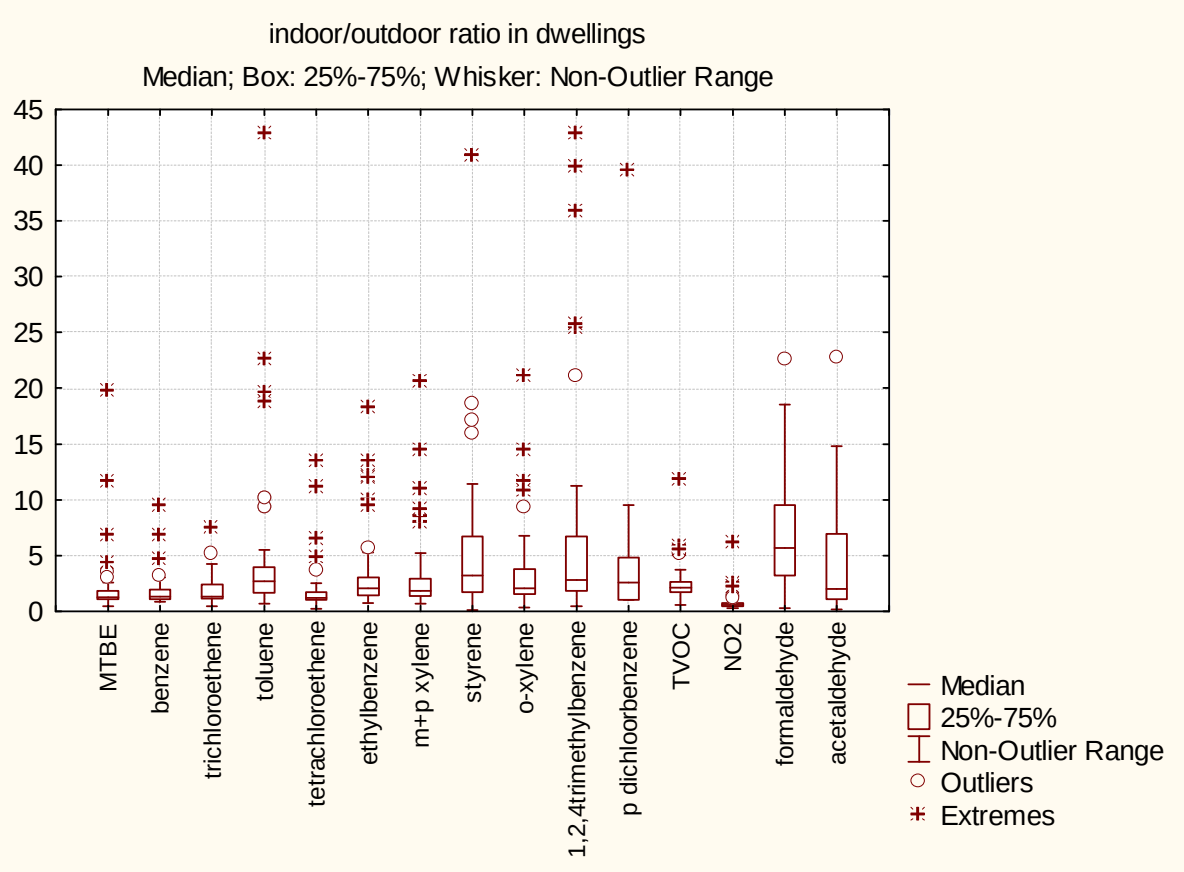

Figure 19: Ratio's indoor-outdoor concentrations in dwellings (50) . Due to the cut-off at 45 on the Y-axis, 2 extreme values for MBTE (up to 115), 1 for tetrachloroethene (110) and 3 values for p-dichlorobenzene (up to 170) are not visible in the graph.

The overall trend of higher indoor than outdoor concentrations suggests that indoor sources contribute significantly to indoor concentrations.

PM10

In contrast to I/O ratios of above mentioned gases, the PM10 indoor concentrations were significantly lower than outdoor PM10 concentrations.

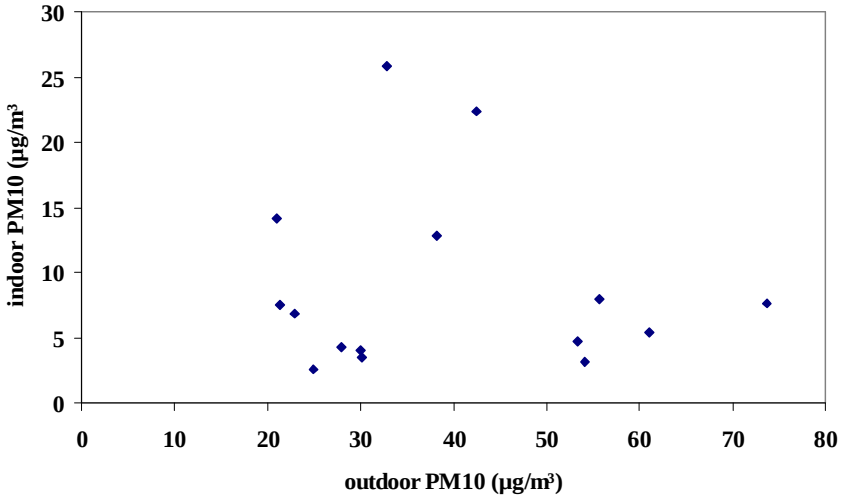

Figure 20: Indoor versus outdoor PM10 concentrations in 16 dwellings

Table 9 : Indoor/outdoor ratio's PM10 in 16 dwellings

	I/O PM 10
mean	0,32
median	0,29
minimum	0,09
maximum	0,79
P25	0,19
P75	0,34

These I/O ratios are rather high compared to PM I/O ratio's reported in the literature. In homes without indoor sources (including human activity) indoor PM10 concentrations are typically 70 % of outdoor concentrations (Monn et al., 1997). The highest indoor/outdoor (I/O) concentration ratios, i.e. I/O = 2, were recorded for homes with smoking inhabitants. Occurrence of human activities and gas cooking resulted in I/O ratios of respectively 1,4 and 1,2 (Monn et al.,1997). Cao et al. (2005) found I/O ratios of 1,0, 1,5 and 1,0 for residential homes in Hong Kong near roadsides, in urban areas and rural areas respectively. Mean residential indoor concentrations of ambient PM2.5 particles ranged from 7 (Helsinki) to 21 µg/m³ (Athens) in the EXPOLIS study. In the EXPOLIS study I/O PM2.5 concentration ratios vary from 0,90 (Athens) to 1,04 (Prague) (Götschi et al., 2002).

1.1.5.2 contribution of indoor sources to total indoor concentrations

The total indoor concentration of a substance can be expressed with equation 1 (Hänninen et al., 2004) which included a left term related to outdoor concentration and a right term representing the contribution of indoor sources:

$$C_{indoor} = \frac{Pa}{a+k} C_{outdoor} + C_{indoor\ sources} \qquad Equation\ 1$$

The infiltration factor (known as F_{INF}) can be derived in the absence of indoor sources, assuming the following relationship between concentrations indoor (C_{in}) and outdoor (C_{out}) (see also Wallace, 1996; Allen et al., 2003 and Yeh et al., 2002):

$$F_{INF} = \frac{C_{indoor}}{C_{outdoor}} = \frac{Pa}{a+k} \qquad Equation\ 2$$

with P: penetration factor (-); a is the air exchange rate (/h) and k is the deposition, removal or sorption rate (/h). The penetration factor P is the dimensionless fraction of the pollutant in ambient air that penetrates into the indoor environment.

In this study, MTBE was selected as a tracer with only outdoor sources to calculate F_{INF} and thus, in a next step, to discriminate the outdoors generated from indoor generated fractions of other pollutants:

$$C_{ig,x} = C_{indoor\ total\ x} - F_{INF,MTBE} \times C_{outdoor,x} \qquad Equation\ 3$$

with ig = indoor generated
and x = substance x

The fraction of indoor concentration that can be attributed to indoor sources can then be calculated as:

$$\%C_{ig,x} = {C_{ig,x}}/{C_{indoor\ total,x}} \qquad Equation\ 4$$

This method is analogous to the principle used in other studies using other tracers (e.g.SO_4 as tracer for PM2.5) to determine outdoor to indoor infiltration of pollutants (Wilson et al.,2000; Wilson et al., 2006).

The tracer method can only be used under the assumption of absence of indoor sources of the tracer. In case of MTBE this was expected as it is a typical traffic exhaust pollutant, an additive of petrol. This assumption is valid if there is a clear relationship between indoor and outdoor MTBE, and the intercept low. The indoor MTBE concentrations versus outdoor MTBE concentrations are illustrated in Figure 21.

In a first step, the relationship between indoor and outdoor MTBE is plotted for the overall dataset of 50 dwellings ((Figure 21; graph A). It reveals from that graph that the indoor/outdoor relationships is very weak. However, the weakness of this relationship is mainly driven by 2 outliers with extreme high MTBE indoor concentrations.

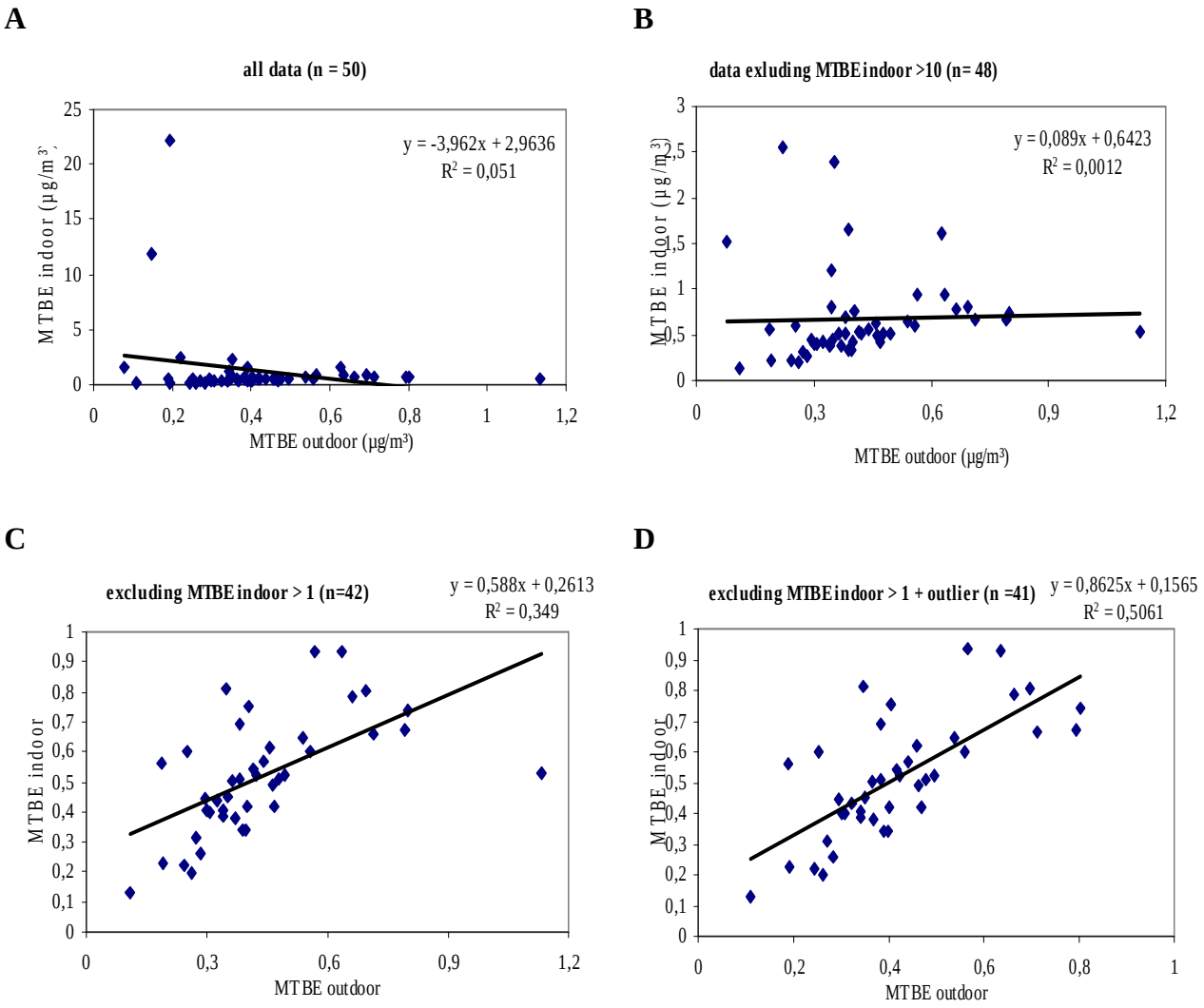

Figure 21: Indoor versus outdoor MTBE concentrations in dwellings

Exceptionally high indoor/outdoor MTBE ratios were observed for 2 dwellings. In these two dwelling, MTBE concentrations in living rooms were 33 and 7,6 µg/m³, in bedrooms 10,9 and 16 µg/m³ respectively, and outdoor (backdoor) concentrations were as low as 0,195 and 0,146 µg/m³ respectively. The only potential MTBE indoor source of which we could think of was the presence of gasoline indoors. The presence of a garage in the dwelling, or adjacent to the dwelling (with passage between garage and house), as an indication for possible indoor gasoline sources was verified. Indeed, in the two dwellings with high MTBE concentrations, such a garage was present. However, in 8 other dwellings, a garage in or with passage to the house was present, without exceptional high $F_{inf,MTBE}$ values. Among these 8 houses, 7 had an I/O_{MTBE} ratio below the mean values of 4,1 and 4 had an I/O_{MTBE} below the median of 1,25. Ventilation frequencies in those 10 houses were not explanatory for difference in $F_{inf,MTBE}$ (almost all these dwellings were ventilated once a day).
The lower I/O_{MTBE} in some houses with garages is not surprising since the presence of a garage is obviously not a good substitute for the presence of petrol cars; there are probably at least some diesel cars (no MTBE sources) in the sample. Unfortunately, the questionnaires

did not enquire for car fuel type. Thereby, the hypothesis of gasoline cars in cars responsible for extreme high MTBE concentrations can not be confirmed nor refuted.

In a second step, these 2 cases with exceptional high MTBE indoor concentrations were omitted (Figure 21; graph B). Still, the relationship between indoor and outdoor MTBE concentrations remains weak. Additionally, 6 points have elevated indoor MTBE concentrations, which were in 4 cases potentially explained by the presence of a garage in or connected to the dwelling (3 with direct connection to the remainder part of the house, and 1 without passage to the house). For the 2 other houses, no explanation for elevated indoor MTBE concentrations was found. For one house, no information was available since the occupants did not filled in the questionnaires; for the other house, no possible MBTE sources were present in that house (no garage, no storage of motor fuel in the house,… was reported by the occupants). Excluding these 6 dwellings with elevated MTBE indoor concentrations (probably due to indoor sources) in a third step, gives a much better relationship (graph C of Figure 21).

The exclusion of 1 extra outlier with very high MTBE outdoor concentrations in a last step (graph D in Figure 21) improves the regression equation (from $R^2 = 0,30$ for graph C to $R^2 = 0,50$ for graph D). That outlier is a dwelling situated in a hotspot area, and that dwelling was not ventilated. Since this can be considered as rather exceptional situation which influences largely the slope in Figure 21, it was decided to omit this outlier for the derivation of an average infiltration factor.

The resulting infiltration factor ($F_{INF, MTBE}$), is derived from the slope of the MTBE indoor versus outdoor graphs (F_{inf} = 0,86 95 % CI: 0,59- 1,14) based on this filtered dataset (n = 41 out of 50; graph D in Figure 21) and 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) Wilson et al., 2006).

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 by Ebelt et al. (2006).

Alternatively to a calculated F_{inf} based on MTBE concentrations, it was also verified if NO_2 could be used as a tracer for outdoor to indoor infiltration. NO_2 has even lower indoor-outdoor ratio's than MTBE. However from Figure 22 it is learned that there is no relationship between indoor and outdoor NO_2 concentrations, and thus NO_2 as tracer to estimate infiltration from outdoor to indoor environment is useless.

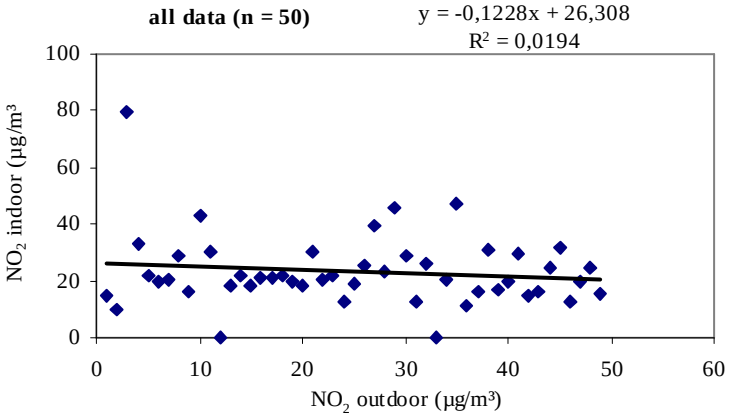

Figure 22: Indoor versus outdoor NO_2 concentrations in dwellings

The indoor generated concentrations of pollutants were calculated for each dwelling individually, using an average $F_{INF, MTBE}$ factor (0,86; 95% CI: 0,59-1,13) (Table 10). It is important to keep in mind that the individual $F_{INF, MTBE}$ values differ between houses and depend of dwelling types (flats, connected or detached dwellings), ventilation frequencies, building properties, … However, it was not feasible to stratify $F_{INF, MTBE}$ for building type and ventilation classes due to the limited dataset of houses in this study. For example, the 7 points that are obviously above the regression line in Figure 21 D (i.e. that have higher infiltration) are all related to flats or connected houses. However, other flats and connected houses are below the line in that graph. In addition, one would expect a lower and not a higher infiltration factor for flats and connected houses compared to detached dwellings.

Table 10: Fractions of total indoor concentrations that are attributable to indoor sources (% C_{ig}). The distributions of % C_{ig} are based on a general $Finf_{,MTBE}$ for all dwellings combined with individual indoor and outdoor concentrations.

substance	average	P5	P25	P50	P75	P95	P50 HS	P50 RB	P50 UB
Benzene	38%	7%	21%	34%	55%	78%	43%	29%	33%
Trichloroethene	40%	13%	23%	34%	63%	79%	49%	32%	34%
Toluene	64%	36%	48%	68%	78%	96%	67%	70%	65%
Tetrachloroethene	22%	<0	18%	29%	50%	90%	34%	43%	25%
Ethylbenzene	56%	24%	40%	57%	71%	93%	42%	68%	57%
m-+p-Xylene	54%	19%	38%	53%	70%	91%	45%	67%	53%
Styrene	22%	<0	51%	73%	87%	95%	67%	79%	73%
o-Xylene	54%	29%	44%	57%	77%	92%	46%	70%	60%
1,2,4-Trimethylbenzene	67%	38%	54%	69%	87%	97%	54%	80%	70%
p-Dichloorbenzene	57%	14%	19%	66%	82%	99%	66%	14%	69%
TVOC	57%	36%	50%	59%	67%	84%	56%	58%	61%
NO_2	<0	<0	<0	<0	<0	53%	<0	<0	<0
Formaldehyde	63%	<0	71%	85%	91%	95%	83%	85%	87%
Acetaldehyde	34%	<0	20%	56%	88%	93%	39%	85%	58%
PM10	<0	<0	<0	<0	<0	<0	43%	29%	33%

For some pollutants, the median indoor concentrations are typically driven by outdoor generated sources (%C_{ig} < 50 %, e.g. benzene, trichloroethene, tetrachloroethene, NO_2, PM), while for most pollutants the largest contribution to indoor concentrations comes from indoor sources (%C_{ig} > 50 %; e.g. for toluene, styrene, 1, 2,4-trimethylbenzene, p-dichlorobenzene, TVOC, formaldehyde). Other pollutants have a typical 50/50 indoor/outdoor source distribution (m+p xylene, o-xylene, ethylbenzene, TVOC, acetaldehyde) (see Table 10).
It is noted that the %C_{ig} varies greatly among dwellings. This is not surprising since some dwellings have indoor sources for a given pollutants while other do not.

The median %C_{ig} for data grouped according to location types (hot spot, rural background and urban background) is given in the 3 right columns of Table 10. The statistical analysis (non-parametrical Kruskal-Wallis Anova) revealed that the %C_{ig} were not different between the 3 location types, for none of the substances. Therefore, in further calculations of exposure related to indoor versus outdoor sources (1.2.2) one general median %C_{ig} will be used.

For some combinations of dwellings and pollutants, a negative C_{ig} value was obtained, which is physically impossible. This probably relies in the overestimation of $F_{inf,MTBE}$ of those dwellings when using the average $F_{inf,MTBE}$ instead of home-specific F_{inf} values.

It is noted that %C_{ig} in Table 10 was calculated using an average $F_{inf,MTBE}$ factor of 0,86. The uncertainty on this average (95 % CI: 0,59 – 1,13) value could also be taken further into account. The effect of this uncertainty on C_{ig} is illustrated in the next example: calculating with the lower end of this distribution ($F_{INF,MTBE}$ = 0,59) would lead to %C_{ig} = 55% for P50 of benzene, and calculating with the higher end ($F_{inf,\ MTBE}$= 1,13) leads to %C_{ig} = 12% for P50 of benzene. Thus, it should be kept in mind that the uncertainty of $F_{inf,MTBE}$ introduces roughly a factor of 2 uncertainty on %C_{ig}. The uncertainty range of %C_{ig} is in principle equal for all substances.

The MTBE infiltration method was not successful to estimate C_{ig} for NO_2 and PM10. This is basically because indoor/outdoor ratios for NO_2 and PM10 were lower than for MTBE. As mentioned above, rather exceptional low I/O PM10 ratios were found in this study compared to literature data.
Thus, notwithstanding MTBE, NO_2 and PM10 probably all have little indoor sources, their I/O ratio's differ from each other. This inappropriateness of MTBE to estimate the infiltration of PM and NO_2 probably lies in different penetration factors and/or removal/sorption rates between these compounds (see Equation 4). Indeed, it was assumed that these factors are equal between MTBE and the target pollutant for which one wants to know the infiltrated fraction.

1.1.6 Correlation between indoor generated concentrations and indoor activities/home characteristics

1.1.6.1 Correlation matrix

Alternatively to statistical analysis of indoor characteristics on total indoor concentrations (1.1.4), the effect of indoor characteristics on indoor **generated** concentrations was analysed.

Theoretically, this should lead to better correlations since the uncorrelated factor of outdoor generated substances is filtered out. Unfortunately, this analyses cannot be performed for NO_2 and PM because the $F_{inf,MTBE}$ method was inapplicable for these substances.

Results of the correlation analysis between indoor **generated** concentrations and indoor activity durations (fireplace, heating, ventilation) were very similar to the correlation with **total** indoor concentrations. Apparently, the influence of outdoor generated concentrations was rather minimal and correlations between indoor activity durations and indoor concentrations were generally rather very weak.

*Table 11: Correlation matrix between indoor generated concentrations (expressed as µg/m³) and indoor activity frequencies (expressed as hours/week). Non-filled in answers were omitted in the analysis. Significant correlations are marked in **bold italic**.*

	# answered (total : 44)	MTBE	Benzene	Trichloro ethene	Toluene	Tetrachloro ethene	Ethylbenz ene	m-+p-Xylene	Styrene	o-Xylene	1,2,4-Trimethyl benzene	p-Dichloor benzene	TVOC	Formalde hyde	Acetald ehyde
Central Heating	37	-0,30	-0,07	-0,10	***-0,35***	0,09	-0,24	***-0,36***	0,11	***-0,35***	-0,20	-0,12	-0,17	-0,04	0,23
Stove	9	0,18	0,21	0,07	0,37	0,73	-0,06	0,06	***0,84***	0,68	0,22	***0,92***	0,36	-0,76	0,12
Open Fire	7	***1,00***	***1,00***	***1,00***	***1,00***	0,26	***1,00***	***1,00***	0,29	***1,00***	***1,00***	-0,46	0,98	0,98	
Other heating	8	0,05	-0,55	-0,39	0,24	0,49	0,15	-0,44	0,72	-0,35	-0,68	-0,60	0,72	-0,72	-0,64
hot waterproduction with gas	27	-0,10	-0,19	-0,22	-0,08	-0,07	-0,07	-0,18	***0,61***	0,01	-0,20	0,13	0,08	-0,22	-0,08
other hotwaterproduction type	8	-0,42	-0,34	-0,34	-0,20	0,15	0,67	-0,47	-0,06	-0,53	-0,31	-0,39	-0,41	0,03	-0,48
cooker	38	0,02	0,00	0,18	-0,20	0,25	-0,05	-0,06	-0,13	-0,06	-0,04	***0,56***	0,00	0,07	0,01
oven	18	-0,10	-0,10	0,19	-0,24	-0,13	-0,03	-0,05	-0,16	-0,03	0,16	***0,90***	0,01	0,09	-0,08
kitchen fan	34	-0,04	-0,11	-0,11	-0,05	-0,01	-0,05	-0,11	0,31	-0,11	-0,12	-0,05	0,01	-0,21	-0,18
Open Windows Or Doors at The Front Side	19	-0,12	-0,07	-0,14	-0,10	-0,12	-0,20	-0,23	-0,20	-0,22	0,22	-0,09	-0,20	-0,38	-0,18
Open Windows Or Doors at The Back Side	33	-0,06	-0,04	-0,06	***0,66***	-0,10	0,30	0,11	0,19	0,13	***0,49***	-0,07	0,26	-0,08	0,03
Ventilation Grids or Ventilation Fans in Use: Ground Floor	9	-0,39	-0,35	0,41	-0,14	-0,28	0,12	0,03	0,05	0,16	0,65	-0,21	-0,24	0,31	0,20
Ventilation Grids or Ventilation Fans in Use: First Floor	5	-0,49	-0,15	0,37	0,16	0,25	0,38	0,44	0,28	0,42	0,30	0,24	0,35	-0,77	0,26
Other Means of Ventilation	6	0,32	-0,01	0,00	0,42	-0,20	0,71	0,74	0,14	0,61	0,31	-0,44	0,21	0,12	0,73
# persons in the dwelling	33	0,07	-0,17	-0,10	-0,02	0,11	0,01	-0,05	-0,03	-0,04	-0,06	0,07	-0,09	-0,18	0,14

1.1.6.2 Influence of indoor characteristics on indoor **generated** concentrations

The same trends as for the analysis of indoor characteristics on total indoor concentrations were present (Table 12). Notwithstanding that confounding factors of outdoor borne concentrations were filtered out, no clearer effects of indoor sources on indoor concentrations were obtained (Table 12).

Table 12: Ratio's of indoor generated concentration of pollutant x in dwellings in the presence of the indoor source to the average concentration of pollutant x in dwellings where the investigated indoor source is absent. Ratio's are marked in bold if the concentration between the two groups (absence versus presence) was statistically significant (P<0,05).

indoor source	**formaldehyde**	**acetaldehyde**	**TVOC**	**benzene**
chipboard, parquet, fibreboard	0,78		1,08	
cavity wall insulation	1,09		1,26	
gas furnace*	1,08	0,91	1,00	
natural gas heating system	0,89	0,66	0,84	
hot water system on natural gas	0,89	0,66	1,08	
glue	0,80		**2,89**	0,47
stain remover	0,80		**2,89**	0,47
gasoline, exhaust gases			3,79	1,21
vinyl wallpaper	1,24			
sealing products				**9,85**
carpets	0,70			
varnish	0,72			0,27
printer				1,29
photocopier				0,61
insecticide				-
smoking			0,68	

Table 12: continued

	toluene	**ethylbenzene**	**o-xylene**	**m+p xylene**	**styrene**	**1,2,4-trimethylbenzene**	**p-dichlorobenzene**	**trichloroethene**	**tetrachloroethene**
varnish	0,45		1,74	1,65					
cork floor	0,69		0,51	0,51					
parquet	0,53		1,54	1,75					
linoneum			0,89	0,81					
paint	0,56	1,29	1,01	0,92				0,66	
glue		1,59							
gasoline, motor exhaust		1,24/0,92							
furniture		0,92							
polishing wax		1,16							
vinyl wallpaper						0,29			
sealing products		0,30	0,39	0,31					
carpet					0,91	0,37			13,58
chipboard, parquet, fibreboard		-							
curtain material	2,72								384
computer	2,09	1,54	1,33	1,24					
printer	1,21	0,70	0,89	0,89	1,06			0,85	
photocopier	0,42	0,49	0,61	0,55	0,44			0,50	
smoking	**4,61**	0,80	0,81	2,22					
insecticide							-		
maintenance products								0,59	

1.2 Exposure assessment: methodology and interpretation

1.2.1 Total personal exposure for Flemish children (by age category and location type)

A child's total personal exposure to an air pollutant (*T*) is composed of exposure fractionated over different micro-environments, and consists of the concentration of the micro-environment in which that child spend its time, rescaled with the time fractions that the child spends in the corresponding micro-environment:

$$T = \sum_{micro-environment\ j} E_j = \sum_{micro-environment\ j} (t_j * C_j) \qquad \text{Eqn. 3}$$

with t_j: time spent in micro-environment *j* and C_j: concentration in micro-environment *j*.
In a first assessment, point estimates for total 'typical' exposure for children for all age categories (0-2,5 y; 2,5-6 y; 6-12 y; 12-18 y) and location types (HS, UB or RB) were made. It is assumed that a child who lives in a hotspot area has also his other micro-environments (day care, school) located in a hotspot region. Exception herein is transport; the questionnaires contain information on the perceived traffic intensity of the street of residence (calm versus busy). A typical children's exposure is defined as the exposure of a child with an average time activity pattern, in combination with median concentrations for the different micro-environments in which the child spends its time. In cases where (median) concentrations for a given micro-environment in a given location type are lacking (e.g. concentrations for day care in hot spot areas), the daily (24h) exposures is calculated from all available exposures (in the micro-environment for which concentrations were available) and then rescaled to 24 hours.

It was preferred here to use the median instead of the average concentrations since extreme high concentrations in a few locations distort the average concentration, which cannot be considered as 'typical'. In contrast, activity patterns in different dwellings were calculated as the average because if one uses the median durations in different micro-environments, the sum of times spend in different micro-environments can deviate substantially from 24 hours. The average is much more robust in this respect (24h ± 0,5 h).

After identifying the micro-environments that contribute the most to total exposure, the variations in concentration in those environments will be taken into account to determine the variation in the total personal exposure.

1.2.1.1 Time activity patterns for Flemish children

The average children's time patterns were derived from questionnaires for 4 age categories: babies and toddlers (0-2,5 years), infants (2,5-6 years), primary school children (6-12 years) and secondary school children (12-18 years). It is assumed that the time activity patterns do not depend on location (Antwerp or elsewhere) or location type (hotspot, urban background, rural background). Average time patterns for Flemish children were calculated for different age categories. As mentioned in the report of WP2, for a relative high number of days (25

%), only 12 hours (8 am- 20 pm) activities were filled. Apparently, some participants mist the back side of the questionnaires. These data were omitted from the analysis.

A summary of the time activity patterns for Flemish children is given in Table 13.

Table 13: Average time activity patterns for Flemish children

location	age categories			
	0-2,5	2,5-6	6-12	12-18
Number of Children	*15*	*9*	*8*	*3*
	h/day spent in micro-environment			
Dwelling – Bedroom	11,22	11,79	11,23	11,57
Dwelling – Kitchen	0,71	1,32	0,71	1,81
Dwelling – Living Room	5,89	3,41	4,07	2,43
Dwelling - Bad Room	0,56	0,68	0,7	0,41
Dwelling – Other	0,08	0,27	0,03	0,57
Dwelling – Outside	0,02	0,08	0,08	0
Day Care – Inside	4,18	0,08	0,04	0
Day Care – Outside	0,05	0,00	0	0
School - Class Room	0,10	3,68	4,11	4,76
School - Play Ground	0,00	1,04	1,31	0,24
Leisure – Inside	0,36	0,57	0,66	0,44
Leisure – Outside	0,08	0,20	0,34	0,85
Other Inside	0,00	0,00	0	0
Transport (busy) - Walking	0,05	0,10	0,01	0,03
Transport (busy) - Cycling	0,11	0,14	0,12	0,32
Transport (busy) - Motorised	0,37	0,26	0,31	0,34
Transport (calm) - Walking	0,04	0,07	0,01	0,03
Transport (calm) - Cycling	0,00	0,00	0,13	0
Transport (calm) - Motorised	0,01	0,15	0,05	0
total recorded time	23,8	23,8	23,9	23,8

Some remarkable findings from this table:

- Main outdoor activities happen at school (play ground) and other outdoor leisure activities.
- Hardly any time was spent outdoors home. This corresponds well with estimates based on enquiries performed by the Belgian National Institute for Statistics (NIS). This source reports on average one minute per day outdoor residence at home.

In Table 13, time spent at school is 4-5 hours per week. On a weakly basis, this corresponds to a school duration of 28-35 hours, which is a normal average. The same consideration is true for babies and toddlers who go to daycares or crèches.

The time activity patterns in Table 13 are in accordance with time patterns of a Belgian study (for 12-18 years) and with foreign studies (the Netherlands and the USA) reviewed in WP1 (section 2.1). One exception is the reported time spent outdoors. This time amounts to 4 hours/day for children (0-12 y) according to the study of Kruize et al. (2000). The time outdoors probably depends on the season in which the studies were performed. The measuring campaign of this study is not representative for the whole year. The summer

period is not included in this study. Especially during summer time, children are more outside than in the period of this campaign.
Based on data of NIS (Belgian National Institute of Statistics), it was estimated that children's outdoor residence amounts to 2h42 in summer days. Summer days are defined as days with temperatures above 25°C (on average 24,3 days/year). On a yearly basis, this would correspond to 11 minutes per day (Maes, 2006).

Thus, it should be kept in mind that the exposure for children calculated with this dataset of time patterns and concentrations is not representative for the whole years, since summer is not included. In summer exposure is different given different time patterns of children, and also concentrations are different, e.g. the absence of heating related pollution.

1.2.1.2 Concentrations in different micro-environments and location types

Median concentrations for the different micro-environments are listed in
Table 14 (PM) and Table 15 (gases). As mentioned above, not for all micro-environments for which time activity patterns are recorded, concentrations were measured (e.g. kitchen). These exposures were neglected (not set to zero); exposures were recalculated for the time for which concentrations were available. In case where micro-environments (e.g. kitchen) are omitted that possibly have higher or lower concentrations compared to other micro-environments, this might lead to an under-or overestimation of the real personal exposure.
The time spent in motorized transport is not differentiated between public and car transport. To match with the concentrations categories, ¼ of the time spent in motorized transport is attributed to public transport and ¾ is attributed to transport by car. This ratio is derived from a study on transport behaviour for in Flanders (2000-2001; www.uitweg.be).

Table 14: Median concentrations (in µg/m³) of PM10 in different micro-environments, location types and sampling location, taken forward to exposure calculations

LocatieType	Micro-environment	location	PM10 (µg/m³)
HS	dwelling	bedroom	13,3
HS	dwelling	living	9,6
HS	dwelling	outdoor	36,3
HS	day care	indoor	no data
HS	day care	outdoor	no data
HS	school		no data
HS	school		no data
HS	indoor leisure		no data
HS	outdoor leisure		no data
RB	dwelling	bedroom	9,0
RB	dwelling	living	3,1
RB	dwelling	outdoor	24,9
RB	day care	indoor	no data
RB	day care	outdoor	no data
RB	school		no data
RB	school		no data
RB	indoor leisure		no data
RB	outdoor leisure		no data
UB	dwelling	bedroom	9,1

UB	dwelling	living	12,9
UB	dwelling	outdoor	32,9
UB	day care	indoor	12,8
UB	day care	outdoor	38,2
UB	school		no data
UB	school		no data
UB	indoor leisure		no data
UB	outdoor leisure		no data
traffic calm	walk		no data
traffic calm	bike		no data
traffic calm	motorized (3/4 car / 1/4 public)		22,5
traffic busy	walk		25,1
traffic busy	bike		no data
traffic busy	motorized (3/4 car / 1/4 public)		27,1

Table 15: Median concentrations (in µg/m³) of gases in different micro-environments, location types and sampling location, taken forward to exposure calculations

Location Type	MicroType	location	MTBE	Benzene	Trichloroethene	Toluene	Tetrachloroethene	Ethylbenzene	m-+p-Xylene	Styrene	o-Xylene	1,2,4-Trimethylbenzene	p-Dichlorobenzene	TVOC	NO_2	Formaldehyde	Acetaldehyde
HS	D	B	0,56	2,86	0,18	8,09	0,54	0,96	2,24	0,13	0,86	1,93	0,096	425	26,1	21,8	22,9
HS	D	L	0,61	1,71	0,13	3,58	0,21	0,55	1,35	0,07	0,46	0,89	0,027	236	38,5	5,0	26,9
HS	D	O	0,40	2,01	0,12	3,36	0,28	0,57	1,25	0,06	0,47	0,78	0,027	222	44,8	4,7	19,9
HS	DC	L	0,41	2,10	0,49	7,94	0,19	1,27	2,48	3,01	1,67	1,59	0,027	444	48,0	11,9	40,4
HS	DC	O	0,55	2,64	0,63	12,31	0,28	1,61	3,53	4,42	1,31	2,70	0,108	1066	11,8	41,4	12,3
HS	S	I	0,80	2,85	0,14	5,79	0,68	1,36	4,03	0,01	1,62	5,28	3,015	766	39,3	11,9	3,9
HS	S	O	0,29	1,03	0,15	1,97	0,13	0,35	0,86	0,01	0,29	0,43	0,027	287	42,7	2,5	0,5
HS	IL	I	no data														
HS	OL	O	no data														
RB	D	B	0,47	1,91	0,11	6,60	0,14	1,24	2,38	0,13	0,78	1,60	0,027	492	14,6	16,6	7,4
RB	D	L	0,47	1,96	0,14	6,37	0,21	0,80	1,84	0,12	0,71	1,40	0,027	377	28,9	20,8	26,9
RB	D	O	0,26	1,22	0,07	2,55	0,08	0,40	1,02	0,04	0,34	0,43	0,027	300	26,2	3,1	1,4
RB	DC	I	no data														
RB	DC	O	no data														
RB	S	I	0,34	2,18	0,10	3,40	0,24	0,89	1,89	0,15	0,69	0,76	0,096	316	18,1	33,7	43,2
RB	S	O	0,30	2,26	0,08	2,60	0,12	0,49	1,06	0,06	0,36	0,43	0,027	207	27,1	7,1	50,1
RB	IL	L	0,39	1,65	1,82	37,69	4,68	11,64	38,23	0,07	11,15	8,71	38,018	943	17,3	21,0	7,9
RB	OL	O	no data														
UB	D	B	0,54	1,89	0,16	6,42	0,25	1,07	2,16	0,16	0,85	2,31	0,085	442	19,8	23,3	9,5
UB	D	L	0,59	2,08	0,16	9,09	0,26	1,00	2,28	0,25	0,86	2,59	0,074	524	23,3	32,1	17,7
UB	D	O	0,33	1,53	0,13	3,02	0,21	0,47	1,07	0,07	0,38	0,64	0,027	213	39,2	3,2	5,3
UB	DC	I	0,55	1,85	0,10	4,68	0,20	0,62	1,37	0,16	0,55	1,59	0,027	381	29,8	21,1	33,1
UB	DC	O	0,44	1,97	0,09	3,24	0,18	0,54	1,11	0,12	0,41	0,68	0,027	202	49,9	12,0	46,1
UB	S	I	0,34	1,27	0,16	3,33	0,18	0,52	1,35	0,07	0,45	0,87	0,075	332	20,2	18,8	3,4
UB	S	O	0,32	1,22	0,13	2,50	0,16	0,41	1,01	0,04	0,38	0,50	0,027	293	43,2	3,6	1,2
UB	IL	I	1,15	5,88	0,13	11,11	0,47	1,09	2,29	0,01	0,82	1,94	0,085	339	143,1	17,1	18,3
UB	OL	O	no data														

traffic busy	TW	O	0,03	0,01	0,03	10,99	0,03		1,67	0,01	0,01	1,10	0,027	1500	36,8	11,0	15,5
traffic busy	TB	O	1,58	2,02	0,10	6,47	0,25	0,73	1,35	0,41	0,41	0,37	0,027	1250	63,0	14,5	22,3
traffic busy	TM		4,88	3,43	0,16	12,34	0,20	1,24	3,41	0,33	1,00	2,77	0,039	1089	54,1	11,7	9,6
traffic calm	TW	O		1,76	2,60	9,42	0,28	1,53	2,84	1,72	1,79	1,26	0,097	5880	9,7	40,8	281,3
traffic calm	TB	O	3,49	1,77	0,14	9,64	0,46	1,00	2,08	0,01	0,78	0,91	3,570	3310	25,3	56,6	92,7
traffic calm	TM		1,65	3,15	0,16	21,69	0,08		4,39	0,01	1,77	7,13	0,115	6049	43,7	10,8	19,5

UB: urban background; RB: rural background; HS: hot spot

D: dwelling; DC: day care; S: school; IL: indoor leidure; OL; outdoor leisure

TW: traffic walking; TB: traffic bike; TM: traffic motorized (3/4 car / 1/4 public)

1.2.1.3 Children's daily exposure

Total children's exposure was calculated according to:

$$T = \sum_{micro-environment\ j} E_j = \sum_{micro-environment\ j} (t_j * C_j)$$

with t_j = average time fraction (h/24h) spent in micro-environment j (Table 13)
and C_j = median concentration in micro-environment j (
Table 14 and Table 15).

Table 16: Typical daily exposure (µg/m³) to pollutants for children of different age classes and for different location of their homes

location	age	MTBE	Benzene	Trichloro ethene	Toluene	Tetrachloro ethene	Ethylbenze ne	m-+p- Xylene	Styrene
HS	0-2,5	0,62	2,40	0,23	6,93	0,38	0,91	2,07	0,10
HS	2,5-6	0,65	2,55	0,17	6,74	0,48	0,92	2,35	0,10
HS	6-12	0,68	2,51	0,16	6,48	0,47	0,92	2,34	0,09
HS	12-18	0,71	2,68	0,16	6,96	0,52	1,00	2,55	0,10
RB	0-2,5	0,55	2,01	0,22	7,52	0,25	1,31	2,89	0,69
RB	2,5-6	0,52	2,04	0,17	7,02	0,30	1,37	3,20	0,15
RB	6-12	0,54	2,06	0,17	6,98	0,32	1,39	3,29	0,14
RB	12-18	0,54	2,05	0,16	6,70	0,28	1,34	3,03	0,14
UB	0-2,5	0,65	2,05	0,16	7,08	0,25	0,98	2,09	0,19
UB	2,5-6	0,59	1,96	0,17	6,61	0,25	0,94	2,06	0,16
UB	6-12	0,62	1,97	0,17	6,57	0,25	0,94	2,04	0,16
UB	12-18	0,61	1,91	0,17	6,32	0,24	0,94	2,02	0,16

location	age	o-Xylene	1,2,4- Trimethylbenze ne	p- Dichloro benzene	TVOC	NO_2	Formal dehyde	Acetaldehyde
HS	0-2,5	0,9	1,6	0,1	410	34,2	15,2	27,4
HS	2,5-6	0,9	2,3	0,6	522	32,0	16,0	19,6
HS	6-12	0,9	2,3	0,7	490	32,6	15,4	18,8
HS	12-18	1,0	2,6	0,8	517	32,1	16,9	18,7
RB	0-2,5	1,1	1,7	0,7	495	26,0	17,2	19,7
RB	2,5-6	1,0	1,6	1,1	527	19,9	20,3	20,5
RB	6-12	1,1	1,6	1,2	493	20,3	20,9	21,6
RB	12-18	1,0	1,6	0,9	485	19,2	21,5	19,8
UB	0-2,5	0,8	2,3	0,1	485	25,9	25,2	17,1
UB	2,5-6	0,8	2,1	0,1	513	26,6	23,1	11,0
UB	6-12	0,8	2,0	0,1	484	27,3	23,3	10,7
UB	12-18	0,8	2,0	0,1	466	25,1	23,1	10,0

location	age	PM10
HS	0-2,5	12,3
HS	2,5-6	13,0
HS	6-12	12,7
HS	12-18	13,0
RB	0-2,5	7,4

RB	2,5-6	8,3
RB	6-12	7,9
RB	12-18	8,4
UB	0-2,5	11,2
UB	2,5-6	10,6
UB	6-12	10,5
UB	12-18	10,1

Typical exposure to pollutants is slightly higher for some pollutants (MTBE, benzene, tetrachloroethene, NO_2 and PM10) for children living in HS areas compared to RB or UB areas (Table 16). These differences are relatively small. In addition, for the majority of the investigated pollutants (e.g. trichloroethene, toluene, ethylbenzene, m-+p-xylene, styrene, o-xylene, 1,2,4-trimethylbenzene, p-dichlorobenzene, TVOC, formaldehyde, acetaldehyde) there is hardly any difference in children's exposure between HS, RB and UB. There was almost no difference in external exposure among different age classes (Table 16). This is a consequence of the rather similar time-activity patterns for children of different age classes.
One exception is the 5-fold larger styrene exposure to the youngest children in RB areas compared to older ages in RB areas and compared to other location types. This high value is attributable to elevated styrene concentrations (both indoor and outdoor) for one day care centre (the only one that was located in RB areas). Thus for this micro-environment, the median is derived from only one single measuring point and thus must be taken with caution.

The majority of exposure to pollutants (>80 %) is related to exposure in the indoor environment. Exposure during time spent outdoors or in transport contributes generally less than 10 % to total exposure (except for MTBE and TVOC). In Table 17, an example of exposure distribution indoor/outdoor/transport for children from 6 to 12 years in hot spot is given. The same trend, i.e. mainly (>80 %) indoor exposure was also found for other ages categories and location types.

Table 17: Fractions of total personal exposure related to indoor exposure, outdoor exposure and exposure during transport (here for children 6-12 years in hot spot areas)

	indoor exposure/total exposure	outdoor exposure/total exposure	transport exposure/total exposure
MTBE	82%	3%	15%
Benzene	94%	3%	3%
Trichloroethene	91%	6%	3%
Toluene	93%	2%	5%
Tetrachloroethene	97%	2%	2%
Ethylbenzene	94%	3%	3%
m-+p-Xylene	94%	2%	3%
Styrene	91%	1%	8%
o-Xylene	95%	2%	3%
1,2,4-Trimethylbenzene	96%	1%	3%
p-Dichlorobenzene	96%	0%	3%
TVOC	85%	4%	12%
NO_2	87%	9%	4%
Formaldehyde	95%	1%	4%
Acetaldehyde	95%	1%	5%

PM10	93%	2%	6%

The above exposure calculations are based on median concentrations in all micro-environments. This is however only representative for the exposure of a 'median' person. From a public health perspective, we're also interest in the higher (e.g. P95) exposed persons. Monte Carlo simulations (Crystall Ball software) were used to estimate the variation of exposure for children within each age group or location. With bedroom and living room being the largest contribution of exposure in indoor dwellings (bedroom + living room) (>80 %) for the 'typical' (=median) scenarios, variation in these micro-environments was taken forward to the Monte Carlo analysis. For example, for children living in urban areas, the distribution/variation in air concentrations within the 23 dwellings in urban areas was taken into account. Variations in other compartments were not accounted for. Firstly, because they affect to a much lesser extent the total exposure, and secondly, because, for some micro-environments, a distribution on concentrations could not be established because of the limited dataset (e.g. only measurements in one school in the urban background areas).
The variation in time patterns wasn't taken into account. This was technically impossible because distributions of time use in different micro-environment were not independent from each other. Using distributions of time use in different micro-environments would lead for some combinations to total daily time far below or above 24 hours. Obviously, this should be avoided. But the variation in time patterns between different children is smaller than variation in concentrations in different micro-environments.
Therefore, the point estimates for time activity patterns were combined with the variation of concentrations in indoor environments in dwellings (bedroom + living room) and point estimates of concentrations in other micro-environments (e.g. school, transport). This results in distribution of exposure estimates. A graphical example of exposure distributions is given in
Figure 23.

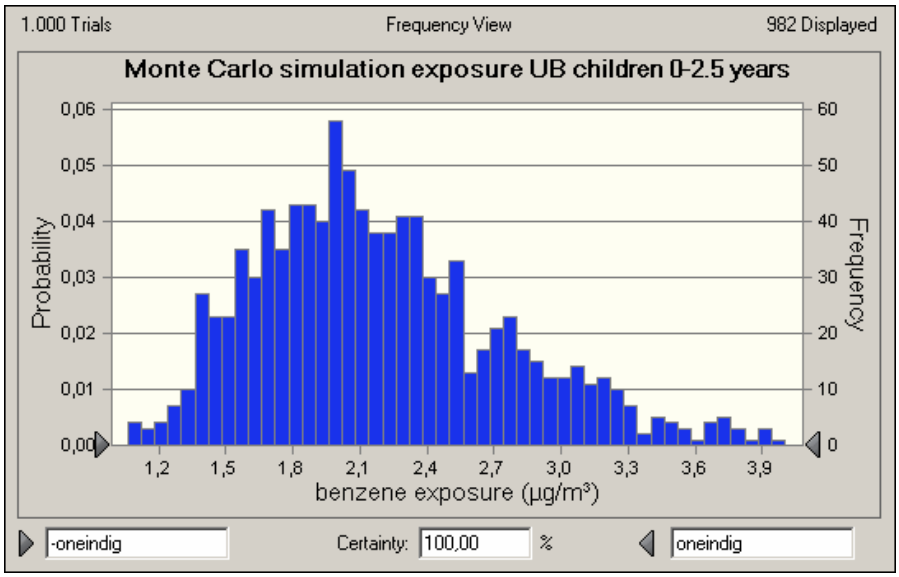

Figure 23: Distribution of children (0-2,5 years; urban background) exposures to benzene using Monte Carlo simulations

The predicted distributions for children's exposure are illustrated for urban background areas and summarized in Table 18.

For most gases, high exposed children (P95) are up to factor 2-4 more exposed than the median exposure (Table 18). Exceptions are tetrachloroethene (x 11) and p-dichlorobenzene (x 50). These extremes are related to the extremely large range of indoor concentrations.

Table 18: Distributions (5^{th} ,50^{th} and 95^{th} percentile) of total children's exposure (µg/m³) accounting for spreading in indoor dwellings (living room + bedroom) concentration using Monte Carlo simulations

	age	percentile	MTBE	Benzene	Trichloroethene	Toluene	Tetrachloroethene	Ethylbenzene	m-+p-Xylene	Styrene	o-Xylene	1,2,4-Trimethylbenzene	p-Dichloorbenzene	TVOC	NO2	Formaldehyde	Acetaldehyde
UB	0-2,5	P5	0,4	1,4	0,1	3,5	0,1	0,5	1,1	0,1	0,4	1,1	0,0	374	21,9	15,3	11,8
UB	0-2,5	P50	0,7	2,1	0,1	8,4	0,3	1,0	2,2	0,1	0,8	2,5	0,1	497	26,0	27,1	18,4
UB	0-2,5	P95	2,1	3,3	0,4	27,4	3,5	2,3	5,4	0,4	2,5	7,1	4,2	673	32,2	56,7	37,8
UB	2,5-6	P5	0,3	1,4	0,1	3,2	0,1	0,5	1,1	0,1	0,4	1,0	0,0	404	22,3	13,0	6,2
UB	2,5-6	P50	0,6	2,0	0,1	7,5	0,3	1,0	2,1	0,1	0,8	2,2	0,1	518	26,7	24,4	11,9
UB	2,5-6	P95	2,2	3,2	0,4	26,1	3,1	2,3	5,4	0,4	2,7	7,2	4,1	704	33,3	55,9	30,4
UB	6-12	P5	0,3	1,4	0,1	3,3	0,1	0,5	1,1	0,1	0,4	1,0	0,0	378	23,4	14,0	6,0
UB	6-12	P50	0,6	2,0	0,1	7,5	0,3	1,0	2,1	0,1	0,8	2,2	0,1	492	27,3	24,7	11,9
UB	6-12	P95	2,1	3,1	0,4	25,1	3,1	2,2	5,4	0,4	2,5	6,8	3,9	666	33,7	54,1	29,4
UB	12-18	P5	0,3	1,3	0,1	3,0	0,1	0,5	1,0	0,1	0,3	0,9	0,0	357	21,0	13,3	5,3
UB	12-18	P50	0,6	2,0	0,1	6,9	0,3	1,0	2,1	0,1	0,8	2,1	0,1	473	25,2	23,9	10,8
UB	12-18	P95	2,2	3,1	0,4	25,8	2,9	2,3	5,7	0,5	2,7	7,1	4,1	664	32,1	57,1	29,8

1.2.2 Indoor exposure related to outdoor generated substances

Children's exposure as calculated in 1.2.1 can be broken down into a fraction that is related to (1) outdoor sources, (2) indoor sources and (3) transport related exposure. The exposure related to outdoor sources (i.e. the ambient exposure) is here defined as the sum of the exposure in the outdoor environment and the exposure in the indoor environment that is attributable to outdoor pollution that has infiltrated into the indoor environment. The $\%C_{ig}$ for each pollutant (see 1.1.5) is used (P50, or median values of Table 10, and not stratified for location type). Calculation of exposure related to outdoor generated NO_2 and PM was not possible due to the unsuitability of $F_{inf,MTBE}$ for this purpose; hence data for PM and NO_2 are not presented.

Transport is here considered as a separate fraction (see above) because the indoor generated sources in transport and infiltration factors were not determined in this study.

Table 19: Contribution of outdoor, transport, indoor exposure from indoor sources, indoor exposure from outdoor sources, and the derived total outdoor related exposure to the total the exposure (median values) of children's (6-12 years) living in hotspot areas.

	outdoor exposure/total exposure	**transport exposure/total exposure**	**indoor exposure from outdoor sources / total exposure**	**indoor exposure from indoor sources /total exposure**	**total outdoor related* exposure/ total exposure**
MTBE	2,8%	15%	70%	11%	73%
Benzene	2,8%	3%	63%	32%	65%
Trichloroethene	6,1%	3%	60%	31%	66%
Toluene	2,0%	5%	30%	63%	32%
Tetrachloroethene	1,9%	2%	68%	28%	70%
Ethylbenzene	2,5%	3%	40%	54%	43%
m-+p-Xylene	2,4%	3%	45%	50%	47%
Styrene	0,7%	8%	25%	67%	25%
o-Xylene	2,2%	3%	40%	55%	43%
1,2,4-Trimethylbenzene	1,3%	3%	30%	66%	31%
p-Dichloorbenzene	0,3%	3%	33%	64%	33%
TVOC	3,7%	12%	35%	50%	39%
NO2	9,0%	4%	87%	0%	96%
Formaldehyde	1,1%	4%	15%	80%	16%
Acetaldehyde	0,6%	5%	42%	53%	42%

*total outdoor related exposure is defined as the exposure in the outdoor environment and the fraction of indoor exposure that is due to infiltration of outdoor generated pollutants into the indoor environment

In the above table, results for children 6-12 y in HS area are presented. The same exercise was repeated for other age and location categories, and summarized in Table 20.

Table 20: Variations (minima and maxima) on the contribution from indoor/outdoor sources across all age categories (0-2,5 y; 2,5-6y; 6-12y, 12-18y) and location categories (hot spot, rural background, urban background).

	outdoor exposure/total exposure		**transport exposure/total exposure**		**indoor exposure from outdoor sources / total exposure**		**indoor sources exposure indoors/total exposure**		**total outdoor (= in indoor + outdoor env) related exposure/ total exposure**	
	min	max	min	max	min	max	min	max	min	max
MTBE	0,2%	4%	12,6%	20%	67%	74%	10%	12%	69%	76%
Benzene	0,3%	7%	3,0%	5%	60%	66%	28%	32%	64%	68%
Trichloroethene	0,2%	6%	2,6%	7%	58%	73%	21%	33%	62%	76%
Toluene	0,1%	2%	3,8%	7%	29%	41%	52%	65%	30%	43%
Tetrachloroethene	0,2%	4%	1,2%	3%	67%	81%	15%	29%	70%	83%
Ethylbenzene	0,2%	3%	1,8%	4%	40%	55%	40%	56%	41%	58%
m-+p-Xylene	0,2%	3%	2,5%	5%	44%	64%	31%	51%	46%	66%
Styrene	0,1%	3%	1,5%	15%	23%	26%	62%	71%	23%	28%
o-Xylene	0,2%	3%	2,0%	4%	40%	59%	36%	56%	42%	61%
1,2,4-Trimethylbenzene	0,1%	2%	2,2%	6%	29%	41%	54%	68%	30%	42%
p-Dichloorbenzene	0,0%	2%	0,1%	23%	27%	97%	2%	66%	29%	98%
TVOC	0,1%	4%	8,1%	18%	33%	40%	44%	53%	36%	41%
NO2	0,2%	10%	3,9%	11%	84%	96%	0%	0%	89%	96%
Formaldehyde	0,1%	2%	1,4%	4%	15%	17%	78%	82%	15%	19%
Acetaldehyde	0,0%	14%	2,8%	13%	36%	43%	45%	54%	41%	50%

Main conclusions from Table 19 and Table 20 are:

- indoor exposure to outdoor generated substances that have infiltrated indoors is much larger (at least 10-fold higher) than the exposure to these substances in the outdoor environment itself.
- for some pollutants the largest fraction of the indoor exposure is related to indoor sources (toluene, styrene, 1,2,4-trimethylbenzene, TVOC, formaldehyde), while for other pollutants, indoor exposure is mainly attributable to outdoor generated pollutants. The latter is typically the case for traffic related substances (MTBE, benzene, NO_2).
- total outdoor related exposure forms the largest contribution to total personal exposure for NO_2, MTBE, benzene, trichloroethene, tetrachloroethene.

Based on calculations with (1) median concentrations in different micro-environments, aggregated by location type, and (2) general $F_{inf,\ MTBE}$ and $\%C_{ig}$ (for each substance), (3) typical time activity patterns of children per age category, and independent of location type, there was within each substance only limited variation in the contribution of outdoor related exposure to total exposure (Table 20), except for p-dichlorobenzene. For the latter component, the exposure is driven by a few outliers, and concentrations below the detection limit for the majority of the measurements.

This relatively small range (Table 20), is partly related to the use of one $F_{inf, MTBE}$ value, irrespective of dwelling type, ventilation profile,… If we knew the F_{inf} of each individual house or the relationship between $F_{inf, MTBE}$ and ventilation profile, a more realistic, but probably larger distribution of the ratio "total outdoor related exposure/ total exposure" between different location types than given in (Table 20) would be obtained. As mentioned above, the dataset was too small to break up $F_{inf, MTBE.}$

1.2.3 Interpretation of exposure in function of health effects

1.2.3.1 Guideline values

At present, no guidelines, limit values or thresholds in terms of exposure exist for the substances on which we focused in this study. Instead, guidelines, limit values or thresholds in terms of concentrations in the air are available (at least for some of the compounds). These values are generally based on exposure and health effects of the substance on humans, or if not available, such limits are derived from animal or in vitro tests. Without going into detail whether the concentration limits include considerations of exposure (i.e. accounting for time activity patterns), the most appropriate way currently available to elucidate if there is a potential harmful effect of the substance on the health, is referencing the concentrations in the air against limits for concentrations.

This task was partly reported in work package 2 (tables 13-15) (testing concentrations against the Flemish Indoor Decree), and is completed and summarized in Table 21.

Table 21: testing measured concentrations in 119 indoor environments in Flanders against guideline and intervention values for indoor concentrations regulated by the Flemish Indoor Decree (Belgian Law Gazette, 19/10/2004).

pollutant	**guideline value(µg/m³)**	**# exceedings guideline value**	**locations exceeding**	**intervention value(in µg/m³)**	**# exceedings intervention value**	**averaging time**
acetaldehyde	4600	0/119				
benzene	2	60/119	various	10	3/119	
formaldehyde	10	102/119	various	100	1/119	30 minutes
NO2	135	1/119	leisure	200	0/119	1 hour
tetrachloroethene	100	0/119				
toluene	260	0/119				
trichloroethene	200	0/119				
TVOC	200	116/119				
PM10	40	1/44				1 year

In more than half of the sampled indoor environments, the measured indoor concentrations exceeded the guideline values for benzene, formaldehyde and TVOC. For formaldehyde TVOC, the percentage guideline value exceedances are respectively 85 % and 97%. The intervention values were exceeded in 3 dwellings for benzene and in 1 dwelling for formaldehyde. For TVOC, no intervention limit is available. For NO_2 and PM10, the

guideline values were exceeded in 1 case for each (NO_2: 1/119; PM10: 1/44). For NO_2, none of the measurements exceeded the intervention limit.

The comparison of measured concentrations with threshold values is extended for other pollutants that are not regulated in the Flemish Indoor Decree but for which other guidelines (e.g.WHO) exist in Table 22. It is mentioned that the time span to which the guideline values refer are not always the same as the 7-days periods of the current measurements. This complicates the evaluation. For example, if 7-days average concentrations are below limits (based on e.g. 1 hour period), it cannot be excluded that the limits would also be met (and no health effects are to be expected) if measurements on 1 hour periods would be performed.

Table 22: Evaluation of measured indoor concentrations against guideline values

Pollutant	**Health Effects**		**IARC Carcinogenic Class[(b)]**	**Guideline Value**	**Time***	**# exceeding**	**micro-environment with exceeding**
	Acute	**Chronically**					
PM	Respiratory and Cardiovascular Disorders	Cardiopulmonary Disorders/ Bronchopulmonar Cancer	-	PM2.5 ALTER: 40 µg/m³ (Canada) PM2.5 ASTER: 100 µg/m³ (Canada)	1h (ASTER)		
NO_2	Lung function	20 % increase in risk of childhood respiratory illness corresponding to an increase of 30 µg/m³ NO2 level	-	200 µg/m³ (WHO)	1 hour	0/119	
TVOC	no specific effects, might be used as an indicator			200 µg/m³ (Flemish indoor Decree) (guideline value)		116/119	
Benzene	Neurotoxic/ Immunotoxic	Leukaemia	1	5 µg/m³ (EC) $4 \times 10^{-6(a)}$	annual	12/119	living (8), bedroom (3), indoor leisure (1)
Toluene	Neurotoxic	Neurotoxic	3	0,26 mg/m³ (WHO)	1 week	0/119	
Ethylbenzene	-	Under Development	-	22 mg/m³ (WHO)	1 year	0/119	
Xylenes	Neurotoxic	Neurotoxic	3	0,87 mg/m³ (WHO)	1 year	0/119	

1,2,4-Trimethylbenzene	Irritating, headache	Neurotoxic , Asthmatically Bronchitis, Anaemia	-	PEL 120 mg/m³ (OSHA)	8h	0/119	
Styrene	Neurotoxic	Neurotoxic/lung cancer	2B	0,26 mg/m³ (WHO)	1 week	0/119	
p-dichlorobenzene	Respiratory Disorders	Kidney Disorders	-	134 µg/m³ (WHO)	1 year	0/119	
Trichloroethene	Neurotoxic	Disorders of Liver/Kidney/Endocrine Systems and Immunity/ Testicle- Lymph- and Oesophageal Cancer	2A	5000 µg/m³ (WHO) $4{,}3 \times 10^{-7(a)}$	long-term	0/119	
Tetrachloroethene	Kidney Disorders	Neurotoxic/Cancer	2A	0,26 mg/m³ (WHO)	annual	0/119	
MTBE	Neurotoxic/Irritating/ Respiratory Disorders	Liver Disorder	-	TLV 180 mg/m³ (ACGIH)	8h	0/119	
Formaldehyde	Respiratory Disorders	Nasal- and Pharynx Cancer	1	0,1 mg/m³ (WHO)	30 minutes	1/119	bedroom
Acetaldehyde	Respiratory Disorders	Nasal- and Larynx Cancer	2B	50 µg/m³ (WHO) $(1{,}5–9) \times 10^{-7(a)}$	1 year	6/119	living (4), bedroom (1), public transport (1)

* time period to which limits refer; for measurements, the concentrations always referred to 7-days averages
(a) Lifetime cancer risk at 1 µg/m³

(b) IARC carcinogenic class (IARC):

- Class 1: proven to cause cancer;
- Class 2A: probably carcinogenic for humans;
- Class 2B: possibly carcinogenic for humans

Class 3: not classifiable as carcinogenic for humans.

1.2.3.2 From exposure to inhalation dose

Notwithstanding that current limits are expressed in concentration per unit air, it is a useful exercise to express the exposure in terms of exposure at the human body level.

The exposure as calculated in the above section (Table 16) refers to exposure in terms of concentrations expressed per units of air. Translating this external exposure to the doses to which children's lungs are exposed, probably brings us closer to 'real exposure' and thus to the relation with health effects.

The external exposure (calculated in 1.2.1), in terms of concentrations expressed per unit air needs to be translated in terms of exposure that a person experiences at the body level. This includes a conversion from external to internal exposure, or potential dose, based on air inhalation rates and body weights.

Most air guidelines are based on a daily inhalation rate of 20 m³ for a 70-kg day adult (= 0,286 m³/kg-day). However, infants and children inhale 2-4 times less air than adults.
Recently, Brochu et al. (2006) advised to use the 99th percentile inhalation rate of 0,725 m³/kg-day for boys (< 2,6 y) to calculate air quality criteria and standards for non-carcinogenic compounds pertaining to individuals of any age or gender.

Table 23: Average inhalation rates for children (data from Brochu et al., 2006)

age (year)	inhalation rate
	m³/day
males	
0,22-0,5	3,38
0,5-1	4,22
1-2	5,12
2-5	7,6
5-7	8,64
7-11	10,59
11-23	17,23
females	
0,22-0,5	3,26
0,5-1	3,96
1-2	4,78
2-5	7,06
5-7	8,22
7-11	9,84
11-23	13,28

No inhalation data for Flemish children are available to our knowledge, and therefore, the data for the Canadian children are used.
Body weights for Flemish children are available, and used for the calculations of internal exposure.

Table 24: Typical pollutant dose (µg/d/kg) to children of different age classes and split up by location of their homes

location	age	MTBE	Benzene	Trichloroethene	Toluene	Tetrachloroethene	Ethylbenzene	m-+p-Xylene	Styrene
UB	0-2,5	0,29	1,12	0,11	3,24	0,18	0,43	0,97	0,04
UB	2,5-6	0,31	1,21	0,08	3,19	0,23	0,43	1,11	0,05
UB	6-12	0,28	1,02	0,06	2,62	0,19	0,37	0,95	0,04
UB	12-18	0,21	0,79	0,05	2,06	0,15	0,30	0,76	0,03
RB	0-2,5	0,25	0,94	0,10	3,51	0,11	0,61	1,35	0,32
RB	2,5-6	0,24	0,97	0,08	3,32	0,14	0,65	1,51	0,07
RB	6-12	0,22	0,84	0,07	2,83	0,13	0,56	1,33	0,06
RB	12-18	0,16	0,61	0,05	1,98	0,08	0,40	0,90	0,04
HS	0-2,5	0,30	0,96	0,07	3,31	0,12	0,46	0,98	0,09
HS	2,5-6	0,28	0,93	0,08	3,13	0,12	0,45	0,97	0,08
HS	6-12	0,25	0,80	0,07	2,66	0,10	0,38	0,83	0,06
HS	12-18	0,18	0,57	0,05	1,87	0,07	0,28	0,60	0,05

location	age	o-Xylene	1,2,4-Trimethylbenzene	p-Dichlorobenzene	TVOC	NO2	Formaldehyde	Acetaldehyde
UB	0-2,5	0,42	0,75	0,04	191,37	15,99	7,11	12,77
UB	2,5-6	0,42	1,09	0,28	246,86	15,11	7,55	9,26
UB	6-12	0,36	0,92	0,27	198,32	13,22	6,26	7,61
UB	12-18	0,29	0,76	0,23	153,23	9,50	4,99	5,53
RB	0-2,5	0,52	0,79	0,30	231,09	12,14	8,05	9,21
RB	2,5-6	0,50	0,77	0,51	249,09	9,42	9,60	9,70
RB	6-12	0,43	0,65	0,50	199,56	8,24	8,45	8,74
RB	12-18	0,29	0,46	0,26	143,61	5,69	6,38	5,87
HS	0-2,5	0,38	1,06	0,03	226,71	12,10	11,78	7,96
HS	2,5-6	0,37	0,98	0,04	242,62	12,58	10,91	5,21
HS	6-12	0,31	0,82	0,04	196,16	11,06	9,45	4,34
HS	12-18	0,23	0,59	0,02	137,92	7,45	6,85	2,96

In general, the youngest children are subjected to the largest dose (which is rescaled for lung volume and body weight). The dose to children slightly decreases with age.
The age group of 0-2,5 year, i.e. the most exposed group, is a heterogeneous age group with respect to inhalation rates and body weight. However, the ratio of body weight to inhalation rate was fairly constant in the Canadian children in the study of Brochu et al. (2006), i.e., on average, 0,50 for 0,22-0,5 year children; 0,48 for 1-2 year children and 0,50 for 2-5 year children. Therefore, the average internal exposure for the group 0-2,5 can be considered as representative for the whole group.

2 POLICY RECOMMENDATIONS

2.1 Interpretation

High indoor concentrations, large variations and exceedance of limit values
Among 14 measured gases (MTBE, benzene, trichloroethene, toluene, tetrachloroethene, ethylbenzene, m+p xylene, styrene, o-xylene, 1,2,4-trimethylbenzene, p-dichlorobenzene, NO_2, formaldehyde, acetaldehyde) the most abundant gases in both indoor and outdoor environment were formaldehyde (up to 124 µg/m³) , acetaldehyde (up to 65 µg/m³), NO_2 (up to 122 µg/m³) and toluene (122 µg/m³). These upper values are all for indoor environments (living rooms and bedrooms). Concentrations of gases show a very high variability between different houses (n=50), both indoors and outdoors.
Especially for some gases like formaldehyde and toluene, for which the concentrations in bedrooms varied with a factor of 50, and that can be associated with building materials and product use, there is a need to assess how widespread this problem is. Product standards, ventilation and prevention information are needed and their efficiency tested.

In more than 85 % of the investigated indoor environments, the guideline values of the Flemish Indoor Decree for TVOC (200 µg/m³), formaldehyde and benzene were exceeded. In addition, in 3 houses the intervention values for benzene and in 1 house the intervention value for formaldehyde was exceeded. The exceedance of the intervention values demonstrate that indoor air quality policy merits attention, firstly, because of the health risks for the occupants, and secondly, because of the possible drastic consequences of these exceedances for the occupants and house owners: intervention values exceedances imply in principle that the house can be declared unfit for human inhabitation.

The relative high frequency (4 exceedings in 50 houses) of intervention limit exceedances in this relative small set of houses in Flanders, shows that bad indoor air quality is not a negligible issue. It is recommended to measure selected gases (TVOC, benzene, formaldehyde) in a larger dataset of Flemish houses in order the evaluate the magnitude of problem in Flanders.

From our dataset it is concluded that more attention is needed for TVOC, benzene, formaldehyde, acetaldehyde and PM.

On average, using 7 day measurement results, indoor concentration in dwellings are quite homogenous. In most dwellings, concentrations in bedrooms were very similar to the concentrations in living rooms (e.g. the median ratio of bedroom to living room concentrations were typically near 1 (0,78-1,13). Measuring the 7 day average concentration probably misses out on the high, acute peak concentrations due to product or combustion source use indoors. A more detailed assessment is needed to establish the exact role of ventilation for example.

TVOC concentrations indoors are high, in the order of several 100 µg/m³ indoors. The selected VOC only explain a small fraction of the TVOC. A first assessment of the TVOC spectra shows a much higher heterogeneity of VOC indoors compared to outdoors.

Gas concentrations were generally higher in the indoor environment of dwellings than in the corresponding outdoor environment (except for NO_2). Near all p-dichlorobenzene concentrations were below the detection limit in the outdoor environment.

In contrast to for most gases, indoor PM levels were lower (on average a factor 3) indoors than outdoors. From experience and literature these results are difficult to interpret. In well ventilated houses, an equilibrium exists between indoor and outdoor. In the presence of indoor sources and in poorly ventilated buildings PM concentrations are generally higher than outdoors. The fact that measurements were mainly performed in winter, when ventilation is low, and in the absence of PM sources can explain the low PM concentrations. Moreover, from the time-resolved GRIMM data, it can be seen that re-suspension of PM cause concentrations to peak very briefly, while during the day when all occupants are gone to work, to school or to the day care concentrations remain stable but low. More research, calibrated equipment per location and longer time series are needed to explain these findings.

The measurements for other indoor environments (day care and schools (n=5), indoor transport, i.e. car and public transport modes (n=5, indoor leisure facilities) are indicative of the possible peak exposures for children. Schools demonstrated lower ranges of air pollutants than dwellings (both indoor and outdoor); though the median values of indoor concentrations in schools were comparable to that of houses. In motorized transport, peak exposure to NO_2 (up to 122 µg/m³) is common. Transport by cycling or walking mode can be accompanied by peak exposure to acetaldehyde (up to 283 µg/m³), which is 3-fold above concentrations in any other environment. In one indoor leisure location, namely a room in a youth club, high concentrations of toluene (38 µg/m³) and xylenes (m+p xylenes: 38 µg/m³; o-xylene: 11 µg/m³) were measured. It is likely that part of these concentrations are due to smoking.

As expected, traffic related compounds (MTBE, benzene, toluene and NO_2) measured outdoors are significantly higher in HS (hot spot) compared to RB (rural background). For some traffic pollutants, higher indoor concentrations in HS than in RB and UB (urban background) (e.g. NO_2) were observed. Although indoor sources of NO_2 could be present, this does not alter the significantly higher 'hot-spot' result. 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 MTBE the absence of a significant difference indoors, for a pollutant that is only generated outdoors (by petrol cars) is puzzling. To improve our knowledge of outdoor pollution leaking indoors, we should test the validity of MTBE as a tracer further. The role of an adjacent garage with connection to the living compartments of a house should be further investigated.

Outdoor pollution contributes to indoor pollution

On the basis of MTBE as the indicator of infiltration indoor, an assessment was made for the dwellings of the fraction of the indoor concentration attributable to indoor sources. Based on a filtered dataset of 41 dwellings (excluding houses with high indoor MBTE related to indoor sources), an average infiltration factor for MBTE of 0,86 (95 % CI: 0,59-1,13) was used. A break down between indoor and outdoor generated PM and NO_2 concentrations could not be made according to this approach. For most gases indoor concentrations are mainly generated indoors. For indoor median concentrations in 50 dwellings, 68 % toluene, 53 % m+p xylene, 73 % styrene, 57 % o-xylene, 69 % 1,2,4 trimethylbenzene, 67 % TVOC,

91 % formaldehyde and 88 % acetaldehyde concentrations are attributable to indoor sources. Only for benzene (34%), trichloroethene (34%), tetrachloroethene (29%) the indoor generated fraction of indoor concentrations is smaller than the fraction that is generated outdoors and infiltrated into the indoor environment in our dataset of 50 dwellings. Variability of the infiltration factor results in a variability of the contribution of indoor sources to the indoor concentrations of about a factor 2.
It is recommended to make the infiltration factor more dwelling-specific, to reduce the uncertainty of the contribution of outdoor generated substances to total indoor concentrations. Due to the limited number of houses in this study, this was not possible in this study. Though, based on a wider dataset of houses, is should be possible to differentiate the infiltration factor between dwelling types (flats, attached houses, detached houses) or/and isolation or ventilation degree (e.g. double glass versus single glass).

Measurements of concentrations do not show clear relations with indoor sources and activities.
It was attempted to elucidate relationships between total indoor concentrations or indoor generated concentrations and possible sources, indoor activities or building properties. The information hereto was collected by means of questionnaires filled in the occupants of the houses of measuring campaign.
For example, relationship between ventilation rates, specific sources (heating fuel type mode, duration, various product uses, …) and gases that are known to deliberate gases were investigated by means of statistical tools (using the software package Statistica). In general, only few significant correlations between indoor concentrations and indoor/building properties were present. Indoor concentrations of toluene and PM were affected by presence of smokers. Indoor concentrations of xylenes and TVOC's (combustion products) were associated with stove use. However, for most of the expected (based on literature) source-concentration analyses, no significant relationships between source and concentration were present. The absence of source – concentration relationships probably can be explained by the large time span (7 days) of the measurements for the 50 dwellings. In these 7-day averages, short-time peak concentrations (due to product use) are averaged out. The 7-day average concentrations are not sensitive enough to identify specific sources indoors.

Exposure is dominated by time spent indoor
Exposure of children is dominated by the time spent indoor at home, basically in the living room (on average 4h/day) and bedroom (on average 11h/day) at home and in the school or day care (on average 4h/day). Other micro-environments, namely transport, are less important in an average exposure pattern, although they give rise to high concentrations. If health effects from exposure to air pollution is dominated by the long-term average exposure than our attention should go to the micro-environments where most time is spent. But at the same time acute effects from peak exposure cannot be excluded, keeping other micro-environments like motorised traffic, like leisure indoors in the picture.

On the basis of average time activity patterns for children, and using median concentrations in the different micro-environments the typical exposure of children to the selected pollutants does not vary significantly across ages and across locations. Typical exposures to traffic related pollutants are higher in hot spot areas, but not significantly higher. Exposure indoors dominates the total exposure. Using the range of concentrations at home results in a highly exposed group of children whose exposure is 2 times (for benzene) higher than the median or

typical exposure. We lack the health data however to draw clinically relevant conclusions from this. More research is needed to establish clear concentration-response relationships from which the relevance of high and low exposures can be deduced. Depending on the pollutant considered indoor exposure is either dominated by indoor generated pollution, or outdoor generated pollution infiltrating indoors. The latter is the case for NO2, MTBE, benzene, trichloroethene, tetrachloroethene.

inhalation exposure dominates the overall human exposure for most of the investigated pollutants

For the major part of the investigated gases, human exposure via air inhalation is the most important exposure pathway. Other exposure pathways were out of the scope of the current study. In Table 25 a general overview is made of the contribution of different environmental media to the overall human exposure to that compound (e.g. ingestion via food and water intake) (source data: Environmental Health Criteria, WHO, available at http://www.inchem.org). For most gases, air inhalation is the dominant exposure pathway. For formaldehyde and acetaldehyde, important contributions to the total human exposure arise also due to food intake. For acetaldehyde, food intake is the dominant exposure pathway.

Table 25: overview of contribution of different exposure pathways (air, water and food) to the total human exposure

gas	exposure via air	details	exposure via water	exposure via food
MTBE	>70 %		<30%	minimal
benzene	>90 %	cigarettes	minimal	minimal
		gasoline		
trichloroethene	main		minimal	minimal
toluene	main	cigarettes	minimal	minimal (fish)
		gasoline		
tetrachloroethene	main	near dry-cleaning shops	minimal	minimal
ethylbenzene	main		minimal	minimal
xylenes	main		minimal	minimal
styrene	main		minimal	minimal (migration of packaging material)
1,2,4 trimethylbenzene	?		?	?
p-dichlorobenzene	main		minimal	minimal
NO2				
formaldehyde	main		minimal	main (natural occuring in food, in bound and unavailable forms)
acetaldehyde	minimal		minimal	main

2.2 Recommendations

Exposure to air pollution is widespread and difficult to avoid. Ambient air quality policies will result in lower outdoor concentrations and a lower exposure, but at the same time the relative importance of indoor air pollution due to indoor sources will increase. In ambient air quality standard setting the exposure indoor to outdoor pollution that has infiltrated is implicit. This study shows that the contribution of this infiltrated outdoor air pollution is different for the different pollutants studied. This is a point of attention in ambient air quality policies, to include the exposure indoors more explicit.

From this study it is clear that exposure to indoor air pollution is similar for children in different age groups and not very much influenced by location. Traffic density seems to increase the indoor pollution and exposure. Policies that will influence the general average exposure, either through emission limits, information about use of consumer products and alternatives and product standards and labelling are best suited. However, the variation in concentrations is large and hence there is a group of highly exposed children that needs more attention. This high exposed group has no different time-activity pattern from the average group, but here certain products, activities or habits generate high concentrations indoor. There is a need to develop a better understanding of the emissions of products, appliances and building materials, of the use and probable misuse of these products and of the insufficient or uncontrolled ventilation of homes.

In four cases the intervention limit in Flanders was exceeded, in three cases for the carcinogenic benzene and in one case for the carcinogenic formaldehyde. For these cases it is first of all necessary to reassess the situation with new measurements, and in case of reproducibility an inquiry into the source of the pollution is needed to remediate the problem. Apart from this acute intervention it is necessary to perform a wider screening of dwellings in Flanders for these pollutants, and perhaps also for some other pollutants with high maximum values in this study, to assess whether the problem is widespread. Toluene, acetaldehyde, MTBE are some examples.

TVOC concentrations are also very high (>95% exceeding the guideline value) and the spectrum of organic compounds contributing to TVOC is much wider than outdoor. There is a need to standardize TVOC measurements and a need for extensive emission and exposure data to develop new standards.

Exact recommendations for precautionary measures to reduce or avoid exposure to certain gases are difficult to make at the moment because no clear source-concentrations-exposure relationships were found. For this, work on short-term and long-term emission sources their relation to concentrations, and using on various time average measurements should be performed. This is best placed in the context of product policies. Currently, the federal product policy only regulates bulk concentrations of a product, and no emissions, nor does it link to typical and high exposures. There is limited evidence on the health relevance of these exposures. This requires further toxicological and epidemiological evidence of indoor exposure and effects.

Finally a continued effort to inform the public on good product use to the public is welcomed. A good cooperation and communication with industry to appeal for better

labelling and to stimulate the development of innovative and safe product, especially to avoid exposure of children, is the best way forward.

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