



**Ministry of Environment
and Food of Denmark**
Environmental
Protection Agency

Hydrocarbon background levels in Denmark - Outdoor and indoor air

Environmental Project
No. 2019

June 2018

Publisher: The Danish Environmental Protection Agency

Text:

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ISBN: 978-87-93710-37-5

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1. Introduction

1.1 Study background

The term used for volatile hydrocarbons throughout this report is VOCs (Volatile Organic Compounds). In Denmark, this term refers to the compounds detected in the analytical procedure for quantifying hydrocarbons by GC-MS and GC-FID following desorption from (i) carbon tubes (active sampling) or (ii) ORSA- or ATD-samplers (passive sampling). The compounds quantified in the chemical analyses are BTEXN (benzene, toluene, ethylbenzene, xylenes and naphthalene), C9 and C10 aromatics and TVOC (total volatile organic compounds).

VOCs can originate from both outdoor and indoor ambient sources and constitute "background levels". When assessing the potential for VOCs from a subsurface contamination to migrate to the indoor or outdoor air, the resulting concentration will contain a contribution from ambient sources as well as from the subsurface contamination. The Danish regulatory limit for both indoor and outdoor air relates to the contribution from a subsurface source and not to the actual concentration in the indoor or outdoor air.

At sites with a subsurface petroleum hydrocarbon contamination, many of the specific compounds (e.g. BTEX) detected in the indoor air can originate both from ambient sources and from the contaminated subsurface through vapor intrusion. Hence, vapor intrusion studies at hydrocarbon contaminated sites are challenged by the task of distinguishing between the contribution from ambient sources, and the contribution from vapor intrusion. Knowledge of background levels can help in this assessment.

The currently available data on background levels of volatile hydrocarbons in Danish houses and outdoor air are based on older studies (2000-2008) comprising a limited number of data (44-74) from sites where soil or groundwater contamination might have affected the supposed background levels /1/. Furthermore, the current Danish regulatory limit for TVOCs is related to hydrocarbons in the range C6-C35, whereas the background levels in /1/ are based on measurements from active sampling on carbon tubes (C6-C25) and passive sampling on ATD-samplers (C6-C18).

The current standard method for measuring indoor air concentrations of hydrocarbon VOCs in studies for the Danish Regions is by using passive sampling on ORSA-samplers (C6-C35).

1.2 Study objectives

The primary objective of this study is to provide well-documented background hydrocarbon levels in both outdoor and indoor air at uncontaminated Danish residential sites. The study has been funded and conducted by Region Midtjylland (the Central Denmark Region) and DMR A/S.

Secondary objectives of the study are to investigate if there are significant seasonal variations in background levels in outdoor or indoor air, and to determine if other factors influence background levels such as population density, tobacco smoking, wood-burning stoves, ventilation habits/methods, house renovation, etc.

To investigate the secondary objectives, a number of hypotheses concerning cause and effect have been proposed and investigated in subset data studies.

2. Hydrocarbon VOCs in relation to residential buildings

2.1 Danish regulatory limits

The Danish regulatory limits for VOCs in the indoor air in residential buildings are not air quality standards per se, but are specified as acceptable contribution levels from soil and groundwater contamination. Since benzene is listed as a carcinogenic compound (Carc1A), it is associated with a particularly low regulatory limit value in air. The Danish regulatory limits are shown in the table below. The EU limit value for ambient (outdoor) air, based on /32/, is 5 $\mu\text{g}/\text{m}^3$, /8/.

TABLE 2.1. Danish regulatory limits for VOC contribution to residential buildings from contaminated soil and groundwater /23/.

Chemical Name	Regulatory limit ($\mu\text{g}/\text{m}^3$)
Benzene	0,13
Toluene	400
Xylenes*	100
Naphthalene	40
C9/C10-aromatics (Sum)	30
TVOC (Sum of C6-C35)	100

* = sum of *o*-xylene, *m*-xylene, *p*-xylene and ethylbenzene.

2.2 Sources and sinks of ambient hydrocarbon VOCs

Most studies of sources and sinks for ambient hydrocarbons (i.e. processes that emit or remove compounds from ambient air) are related to single compounds, such as BTEX, while fewer studies are dedicated to the study of TVOC as a sum of hydrocarbons. Due to the ubiquitous occurrence and carcinogenic nature of benzene, this compound has received special attention in studies of ambient VOC levels. Many of the sources of benzene are identical with the sources for toluene, ethylbenzene and xylene, often leading to a high correlation between ambient concentrations of these compounds /4/.

2.2.1 Sources

There are both natural and anthropogenic sources of ambient hydrocarbon VOCs, i.e. sources of VOCs unrelated to soil and groundwater contamination.

Natural sources of VOCs, including benzene, include emissions from various biological systems and processes, such as vegetation /1/, /13/; decomposition /1/, /14/ and biomass burning, e.g. forest fires /3/, as well as emissions related to geothermal activity /15/.

The most often cited anthropogenic outdoor sources of VOCs, including benzene, are emissions from traffic, industry, burning of waste and high-temperature incineration /1/, /2/, /4/.

Indoor sources can be building materials, furniture, paint, tobacco smoking, cooking, burning, and emissions from a wide range of household chemicals and products. /1/, /2/, /11/, /21/. One of the most cited indoor sources of ambient VOCs is tobacco smoking.

Several references cite biomass burning (e.g. residential wood-burning stoves) as a significant source of VOCs, including benzene, in ambient air /1/, /2/, /10/. It has, however, been reported

that indoor levels of BTEX are not affected by wood-burning stoves, but emissions affect outdoor levels /25/.

Due to internal sources in houses, several studies have reported that the median ambient benzene concentrations are about 1,7-1,8 times higher in indoor air than in outdoor air /1/, /2/.

2.2.2 Sinks

The most cited sinks for atmospheric levels of VOCs, including benzene, are photochemical degradation /4/, /5/, /6/ and precipitation /4/, /18/.

Photochemical half-lives for benzene are reported in the range of 3-10 days /6/.

As a consequence of atmospheric deposition by precipitation (rain and snow), studies have shown that benzene can be detected at low levels in both unpolluted/clean ocean waters and in mountain snow /7/, /16/, /17/ and /18/.

Several studies have documented seasonal variations in ambient concentrations in both outdoor and indoor air, with a general trend towards lower ambient VOC concentrations in the summer than in the winter /1/ - /4/. Some studies have also documented diurnal variations (day and night variation) in ambient concentrations /5/. Temporal variations are related to variations in both emission rates and photochemical degradation.

2.3 Danish background levels

2.3.1 Background levels, 2000-2008 (outdoor and indoor levels)

The most comprehensive collection of Danish VOC background levels is presented in /1/. The data was collected in the period 2000-2008 in connection with site investigations carried out at suspected polluted sites. Sampling methods included passive sampling by ATD tubes and active pumped sampling on carbon tubes. Data cleaning was carried out to remove values that were suspected of being influenced by soil contamination. The number of samples for hydrocarbon VOCs in outdoor and indoor air was 44 and 74, respectively.

The background levels for hydrocarbon VOCs in outdoor and indoor air from /1/ are presented in table 2.2-2.6. The number of naphthalene data in the study was too low for statistical analyses (a total of seven points in outdoor and indoor air).

TABLE 2.2. Summary of outdoor and indoor background levels from /1/. The intervals indicate that the statistics are influenced by non-detects (NDs); the low-end of the interval includes NDs = 0 and the high-end of the interval includes NDs = the detection limit.

	No. Data Points	Min	Percentiles			Estimated max value
			25%	50%	75%	
Outdoor air						
µg/m³						
Benzene	51	0-0,04	0,16-0,22	0,48	0,83	1,4
Toluene	60	0-0,11	0,16-0,22	1,8	4,0	15
Ethylbenzene	53	0-0,02	0-0,14	0,18-0,30	0,56	1,0
Xylenes	53	0-0,10	0,42-0,45	0,80-0,87	2,5	4,9
C9/C10-aromatics	38	0-0,10	0-0,62	0-1,0	1,1-2,1	2,5
TVOC	44	0-25	0-50	0-61	0-80	97-100
Indoor air						
µg/m³						
Benzene	74	0-0,15	0,49	0,80	1,2	2,0
Toluene	66	1,2	0,49	7,3	9,5	20
Ethylbenzene	62	0-0,16	0,51	0,75	1,2	1,8
Xylenes	67	0-0,64	2,0	3,0	5,0	9,8
C9/C10-aromatics	47	0-0,50	0,80-1,1	1,5-1,9	2,6	4,9
TVOC	70	0-49	150	275	430	650

Comparing the ambient levels of BTEX and TVOC to the regulatory limit values in table 2.1, it is easily seen that the hydrocarbons of interest when assessing potential vapor intrusion based on direct indoor measurements are benzene and TVOC.

The practical application of the data from /1/ has been limited because the data includes sites where vapor intrusion from contaminated soil and groundwater might have contributed to the measured levels.

2.3.2 Benzene and toluene in Copenhagen (outdoor levels)

Benzene and toluene has been measured on two streets with heavy traffic in Copenhagen as a part of the Danish Air Quality Monitoring Programme. Measurements have been made at the oldest of the monitoring stations (Jagtvej), since the late nineties /19/, /20/.

As shown in figure 2.1, the yearly averages from this study show a steadily decreasing trend for both benzene and toluene. Over the monitoring period, ambient benzene levels have decreased from an average of 5,83 $\mu\text{g}/\text{m}^3$ to 0,74 $\mu\text{g}/\text{m}^3$, while toluene levels have decreased from an average of 23,1 $\mu\text{g}/\text{m}^3$ to 1,95 $\mu\text{g}/\text{m}^3$. The decline is associated with a reduction of aromatics in automotive gasoline /1/, more strict emission standards for personal vehicles (EURO-norms), including increased use of catalytic converters /20/ and a relative increase in the use of diesel cars for personal transport in the period 2000-2011 /9/, /20/.

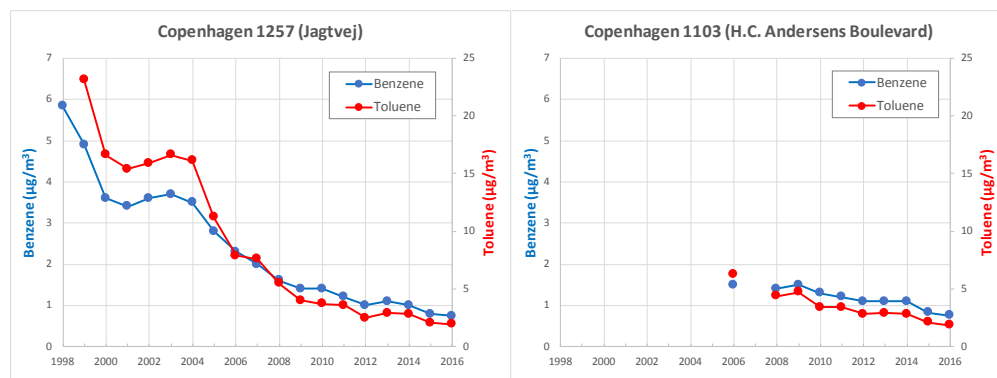


FIGURE 2.1. Annual average curbside concentrations of benzene and toluene at two monitoring stations in Copenhagen (Jagtvej and H.C. Andersens Boulevard) /8/.

For the 2016 data, it is estimated that the contribution of benzene from traffic amounts to 39% of the measured curbside concentration whereas the contribution from traffic accounts for 58% of the toluene concentration /19/. Besides the contribution from traffic, combustion in residential wood-burning stoves is an important source of benzene in ambient air /19/.

2.3.3 Benzene and TVOC in Danish railroad towns/villages (outdoor levels)

A study published in 2012 from the Central Denmark Region investigated the possible contribution of benzene and TVOC from railroad traffic in three smaller Danish railroad towns/-villages (Studsgård, Hjerm and Struer) with populations of about 470, 1.100 and 10.400 /21/.

A total number of 80 passive samples were collected and analyzed over two sampling campaigns in June and November 2011, by deploying ATD- and ORSA-samplers. To test the hypothesis that the railroad traffic contributed with benzene and TVOC to the ambient air in the towns, sampling positions were placed at different distances from the railroad line and in locations representing background levels.

The results from /21/ indicated no significant contribution to the ambient air from the railroad traffic, and the data can be considered representative of Danish background levels in general.

The results show that >98% of the benzene values were above the detection limit of 0,2 µg/m³, whereas only 6,3% of the TVOC values were above the detection limit of 80 µg/m³. The detected ranges of benzene and TVOC were <0,2-1,7 µg/m³ and <80-270 µg/m³, with median concentrations of 0,91 µg/m³ and <80 µg/m³, respectively.

2.3.4 BTEX and TVOC in Danish children's bedrooms (indoor levels)

In a Danish study published in 2016, a wide range of VOCs were measured in children's bedrooms in 19 Danish homes /10/. The highest values in the study were associated with emissions from an adjacent shed used as a store for a lawn mower, gasoline, paints, household chemicals etc.

The average levels of toluene, sum of BTEX and TVOC were 9,1, 14,6 and 338 µg/m³, respectively, discounting the results from the bedroom with very high levels, mentioned above. For the other bedrooms, the highest levels were associated with a house renovating project (emissions from fresh paint), glue from model airplanes and plastic/rubber figurines /10/.

2.3.5 Benzene, C9/C10-aromatics and TVOC from smoking (indoor levels)

Based on data from several international studies, a recent Danish study reported smoking as a major contributor to indoor levels of hydrocarbon VOCs, especially benzene /24/. It was concluded that smoking of five cigarettes in an average living room could lead to maximum concentrations of benzene, C9/C10-aromatics and TVOC of 24, 185 and 600 µg/m³, respectively. For the given scenario, daily average indoor concentrations of 6,3, 49 and 160 µg/m³, were calculated.

2.3.6 Wood-burning stoves

No Danish studies have been reported on the use of wood-burning stoves, but the following pie charts show the results of a survey from Bolius, a home owner's organization, demonstrating the distribution of Danish residences with/without wood-burning stoves across the five Danish regions /12/.

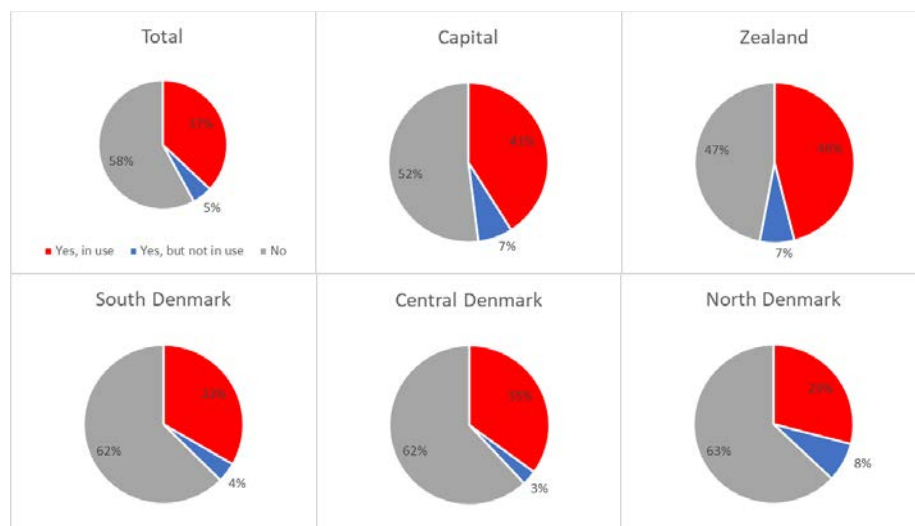


FIGURE 2.2. Regional distribution of Danish residences with wood-burning stoves in use (red), not in use (blue) and without a wood-burning stove (grey) in Denmark (top left). Data from /12/.

As shown in the figure, 42% of Danish residences have a wood-burning stove, and 37% use them, predominately in the Capital and Zealand Regions, whereas in Jutland, the percentage of residences without wood-burning stoves is higher (62-63% vs. 47-52% in Zealand).

3. Methods

3.1 Study design

Based on the need for reliable and up-to date Danish background levels for hydrocarbon VOCs, the Central Denmark Region and DMR A/S have conducted a study with the following primary and secondary objectives.

The primary objective of the study is to provide well-documented hydrocarbon background levels in outdoor and indoor air at uncontaminated Danish residential sites, and to identify which hydrocarbon VOCs are of significance in relation to the Danish regulatory limits associated with vapor intrusion studies. The new updated VOC levels are compared to previous levels, cf. /1/.

Secondary objectives of the study are to investigate if there are significant seasonal variations in the background levels, and to investigate certain hypotheses regarding the background levels; e.g.:

- Higher outdoor VOC levels in cities than in less densely populated urban areas and in rural areas.
- Higher indoor VOC levels in homes of smokers who smoke indoors.
- Higher indoor VOC levels in homes with wood-burning stoves.
- Lower indoor VOC levels resulting from ventilation.
- Higher indoor VOC levels after house renovation.
- Higher indoor VOC levels with higher occupancy.
- Lower indoor VOC levels with older building.
- Higher outdoor VOC levels due to traffic related emissions.

In the course of the investigation of the secondary study objectives, several explanatory hypotheses were put forth and investigated by a number of side studies.

Prior to the main study, a pre-study was performed in order to investigate the best outdoor sampler deployment strategy for passive sampling on Dräger ORSA-samplers; either protected in a plastic cup or unprotected. The results indicated that the most representative VOC levels would be obtained by unprotected sampler deployment and this strategy was therefore used throughout the main study, see Appendix 1.

3.1.1 Main study

The primary and secondary study objectives were investigated in the main study as follows.

The strategy for the main study has been to rely on voluntary sampler deployment in the homes of employees of the Central Denmark Region and DMR A/S, as well as their friends and family members. This has been done to maximize the number of samples collected within the financial frame of the project and in order to achieve a good geographical coverage for measurements.

At each residential building, passive samples have been collected by deploying Dräger ORSA-samplers for approx. 14 days in the living room (indoor sample) and under the eaves (outdoor sample). A package containing two ORSA-samplers, two deployment clips, a questionnaire, a deployment guide document and a pre-paid return envelope was sent to each home owner. Each residence was given a unique number and the ORSA-samplers and questionnaire was pre-filled with this number.

The questionnaire and deployment guide document are shown in Appendix 2.



FIGURE 3.1. Two ORSA-samplers and deployment clips, questionnaire and deployment guidance document sent to each home owner in the main study.

After completion of the sampling period, the ORSA-samplers were returned in the pre-paid envelope to DMR A/S, and the information in the filled-out questionnaires was digitized.

Two sampling campaigns were conducted; November 2015 (131 residences) and May 2016 (142 residences). The geographical distribution of the residences is shown in the figure below. Seven outdoor samples were lost, three from the first round and four from the second round.

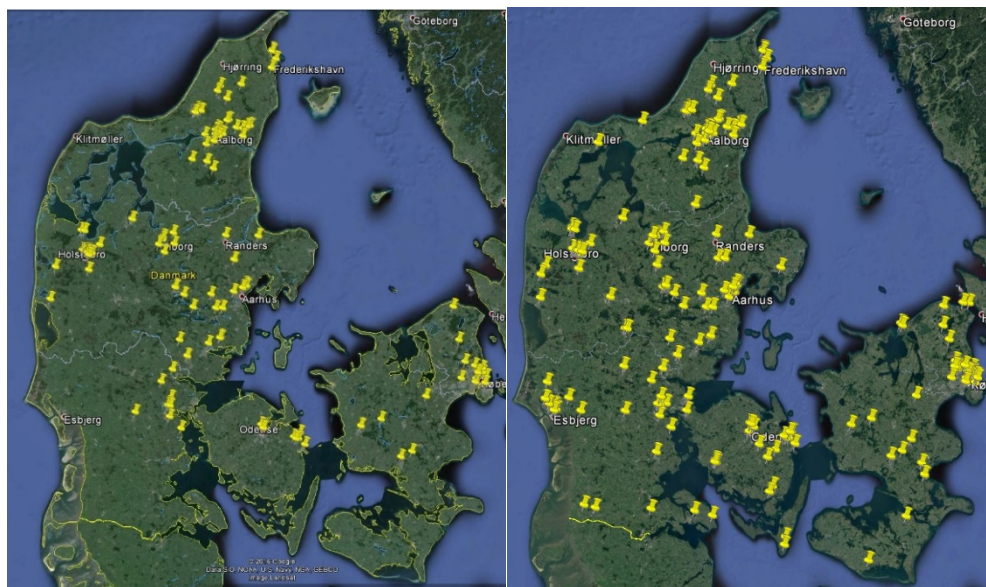


FIGURE 3.2. Geographical distribution of study residences; 131 residences in November 2015 (left), and 142 residences in May 2016 (right).

The ORSA-samplers were analyzed by Eurofins Denmark A/S for quantification of benzene, toluene, ethylbenzene, o-/m-/p-xylene, naphthalene, C9/C10-aromatics and TVOC (C6-C35). The results were subsequently matched to metadata in an Excel spread-sheet using the unique number for each residence. The specific addresses related to each number are known only by DMR A/S, but results are shown in anonymized form in Appendix 3. Residents with

very high indoor benzene concentrations were contacted by DMR A/S regarding potential actions to identify indoor sources of benzene, and to initiate follow-up measurements.

3.1.2 Additional side studies

Study of Variability

The results from the main study raised questions and three Jutland residences (A, B and C), included in the pre-study and the two measuring campaigns of the main study were selected for further investigation. The objective of these investigations was to evaluate the seasonal variability in more detail, as well as to assess inter-duplicate variability in both outdoor and indoor levels and inter-position variability in outdoor levels. The location of the three residences is seen in the following figure.

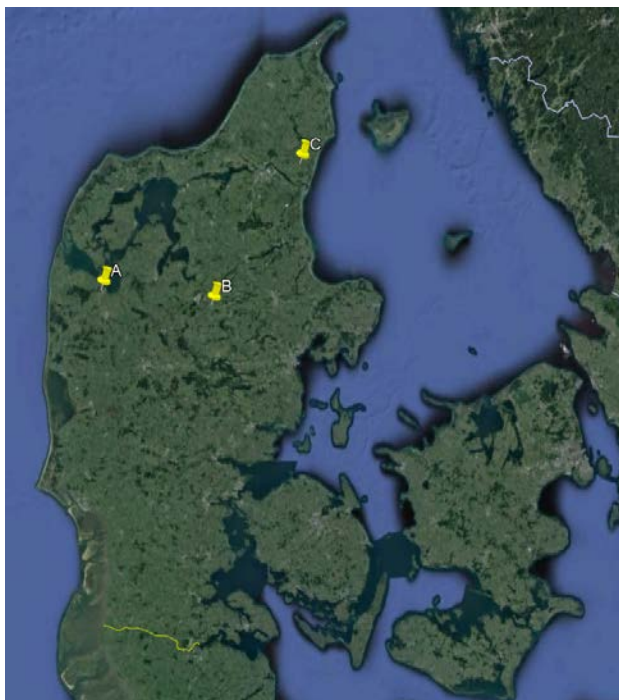


FIGURE 3.3. Location of residences A, B and C included in the study of variation.

TABLE 3.1. Information on the three residential buildings, A, B and C, included in the study.

Residence	Population density	Construction year	No. of residents	Ventilation	
				Method	Frequency
A	Town	1979	5	Manual	< 1 day ⁻¹
B	Village	1856	2	Manual	2-4 day ⁻¹
C	Village	1912	5	Manual	1-2 day ⁻¹

All the three residences are homes of non-smokers. Residence A and C are heated by district heating plants, whereas residence B has a wood-burning stove and is partially heated by a residential wood gasification boiler.

Eight sampling campaigns were performed: September, November, December of 2016 and January, March, May, June and September of 2017. For each sampling campaign, outdoor sampling duplicates were placed on the eastern, western, northern and southern side of each of the residences, and a set of duplicates were placed indoors.

Detailed Study of Variability

At one of the residences (C), an increased frequency of sampling was commenced prior to the beginning of the variability study. At this residence, five additional indoor sampling campaigns (without duplicates) were performed: November 2015, February, April, May and July of 2016.

Chimney Study

One of the residences (B) included in the study of variability is partially heated by a wood gasification boiler located in the garage. At this residence, an additional two sampling points were established on the east and west side of the chimney as shown in the figure below, to evaluate if the emissions from the wood gasification boiler were a major source for TVOC and/or BTEX in local ambient air.

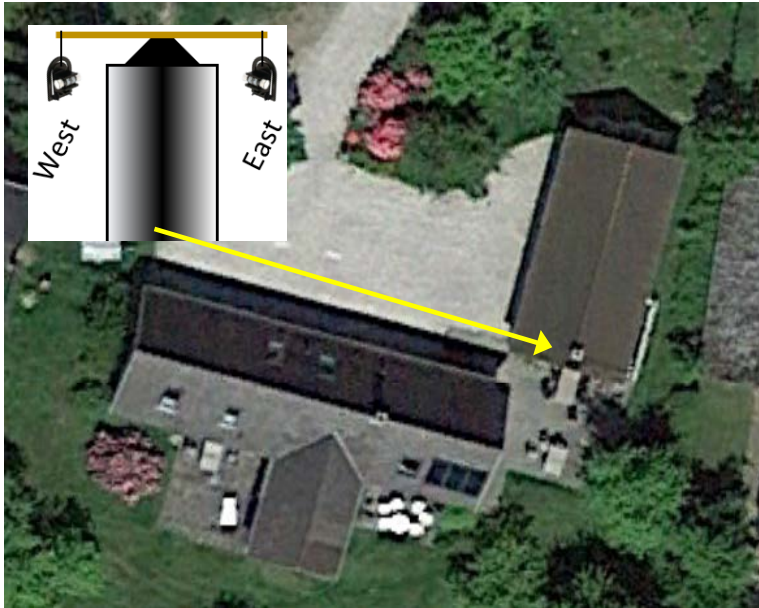


FIGURE 3.4. Schematic of the chimney positions included in the Chimney Study, residence B.

Chimney measurements were included in six of the measuring campaigns: November of 2016 and January, March, May, June and September of 2017. Measurements were done in duplicate at each of the West and East chimney positions.

Coastal Study

Due to the ubiquitous presence of benzene discovered in the main study, an additional side study was initiated to test the presence of VOCs (mainly benzene) on the west and east coast of the Central Denmark Region. Hence, a total of 17 sampling positions were established along the coast lines, four in duplicate, according to the figure below. The samplers were deployed in December 2016.



FIGURE 3.5. Locations of 17 sampling positions in the Coastal Study.

Wind Turbine Study

As an extension of the Coastal Study, six sampling points were established on three wind turbines (turbines 02, 07 and 74) in the Horns Rev 1 offshore wind turbine park, approx. 14 kilometers west of Blåvands Huk. The samplers at each of the three wind turbines were deployed at two different heights; at the base of the tower (9 m DVR) and at the top of the turbines (71 m DVR). One of the samplers was lost (turbine 07, 71 m DVR). The samplers were deployed in January 2017.

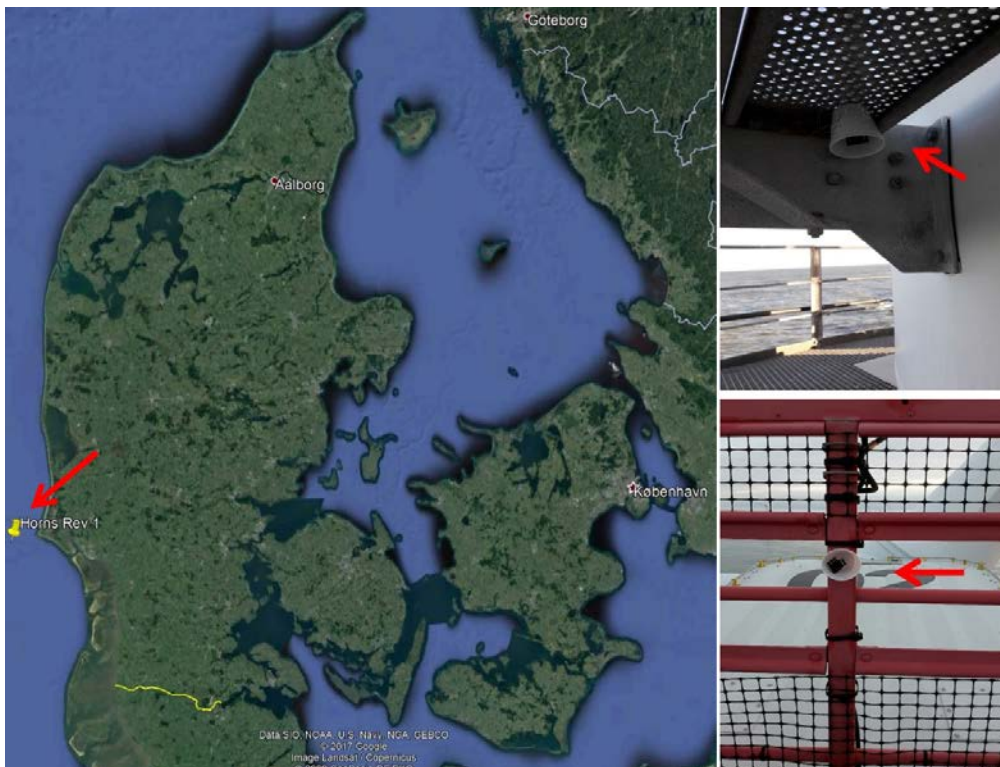


FIGURE 3.6. Location of Horns Rev 1 offshore wind turbine park, 14 km west off the western most point in Denmark. Inserted photos are bottom and top positions on one of the turbines.

Fir Study

Based on the hypothesis that Christmas trees can be a significant indoor source of VOCs, including terpenes (C₅H₈)_n, during the Christmas month, a few branches of Nordmann fir (*Abies Nordmanniana*) were enclosed in a double layer bag of Nalophane with two ORSA-samplers. The study was carried out in February 2017 with a sampler exposure of 14 days, about 1,5 months after the tree had been felled.

The results of this study can only be interpreted as “indicative” measurements of potential emission from fir trees since the prerequisite movement of air around the deployed ORSA-samplers is not met and therefore the results cannot be interpreted as a quantitative measure of emission (see section 3.2).



FIGURE 3.7. Nalophane bag containing Nordmann fir branches and two ORSA-samplers for the Fir Study.

3.2 Sampling media and analytical procedure

The sampling method applied in the present study is deployment of Dräger ORSA 5 diffusive samplers, containing 400 mg of activated coconut shell carbon /26/. Sampler deployment has been approx. 14 days for most samples; and between 7-18 days for some samples. The ORSA-samplers have a sampling rate of approx. 5-10 mL/min and require free air movement around the diffusive ends of the sampler, with air velocities of >1 cm/s /27/.

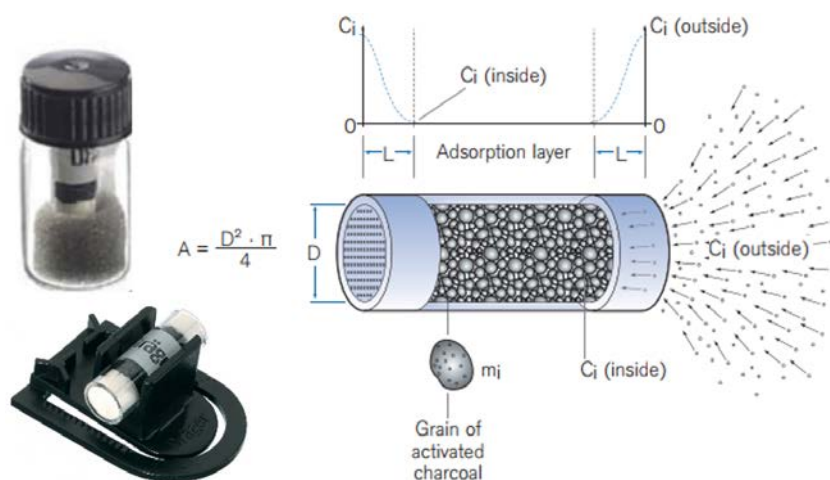


FIGURE 3.8. Dräger ORSA 5 diffusive sampler in pre- and post-deployment packaging (top left), in clip-holder during deployment (bottom left) and diffusive uptake principle (right) /26/.

The analytical procedure used by Eurofins Denmark A/S is based on NIOSH 1500/1501 /28/, /29/, with carbon disulfide extraction and subsequent analysis on a Gas Chromatograph fitted with a FID and MS detector. A summary is given in the table below.

TABLE 3.2. Specifications of methods and analytical detection limits.

Chemical name	Method	Detection limit		
		µg/sampler	* µg/m ³	
Benzene	GC-MS	0,001	0,01	
Toluene	GC-MS	0,05	0,5	
Ethylbenzene	GC-MS	0,01	0,1	
o-xylene	GC-MS	0,01	0,1	
m+p-xylene	GC-MS	0,01	0,1	
C9-aromatics	GC-MS	0,01	0,1	
C10-aromatics	GC-MS	0,01	0,1	
Naphthalene	GC-MS	0,04	0,4	
TVOC	C6-C10	GC-FID	5	43
	C10-C15	GC-FID	4	35
	C15-C20	GC-FID	4	35
	C20-C25	GC-FID	6	52
	C25-C35	GC-FID	8	69
TVOC	C6-C35	GC-FID	5	43

* = With a sampler deployment time of 14 days.

3.3 Data treatment

The raw data collected in this study are presented in Appendix 3.

Aspects of the statistical methods applied throughout this study are explained in Appendix 4; i.e. construction of empirical distribution functions and extraction of statistical properties from these, as well as exploration of study hypothesis through a visual comparison of empirical distribution functions and application of the Mann-Whitney test.

Some data-sets contain a large number of results below the analytical detection limit (non-detects), e.g. TVOC in outdoor air. For some datasets, the content for some single compounds cannot be quantified due to analytical interference in the lab sample; e.g. C9-aromatics.

For the statistical analyses, non-detects are replaced by the detection limit, and therefore some of the statistical measures (e.g. median levels) might actually be lower than reported. Results with analytical interference are omitted and distribution functions for such compounds might comprise a lower number of samples than for other compounds with a full data-set.

For sum values of groups of compounds such as xylenes and C9/C10-aromatics where some compounds are below the analytical detection limit, the sum is calculated for the compounds above the detection limit.

This data treatment for non-detects, analytical interference and group summation with some individual compounds below the detection limit does not affect the general conclusions concerning background levels for use in vapor intrusion studies, since either the results are below the level of interest (the regulatory limit values) or the fraction of results affected is low. However, the results should be reviewed carefully if the statistics for values much lower than the Danish regulatory limit values are utilized for other purposes. In this case, the raw data provided in Appendix 3 can be applied as required with respect to non-detects etc.

Since the ORSA-samplers in this project were deployed for approx. 14 days, data points are plotted in relation to the midpoint in the deployment period for data evaluations involving seasonal variation (e.g. in section 6.1.1).

4. Results – primary study objectives

Three outdoor samples from the first sampling campaign (November 2015) and four outdoor samples from the second sampling campaign (May 2016) were lost during deployment. Hence, the final data sets of the main study consisted of 128 outdoor and 131 indoor samples for November 2015, and 138 outdoor samples and 142 indoor samples for May 2016.

Only one of the naphthalene results was above the analytical detection limit (indoor November 2015). Therefore, results for naphthalene are not included in the data evaluations.

4.1 Background levels – November 2015 (autumn)

The results from the November 2015 measuring campaign are shown as box-and-whiskers plots in the following figures. Sum-parameters are treated in categories corresponding to the Danish regulatory limit values for vapor intrusion contribution to the indoor air, cf. section 2.1. Hence, TVOC is the sum of C6-C35 hydrocarbons and xylenes represent the sum of o/m/p-xylene and ethylbenzene.

For C9-aromatics, four outdoor and 54 indoor samples showed component interference in the analysis. For C10-aromatics, two indoor samples showed component interference in the analysis. Therefore, only 124 outdoor samples and 75 indoor samples are included in the statistical analysis for the C9/C10-aromatics.

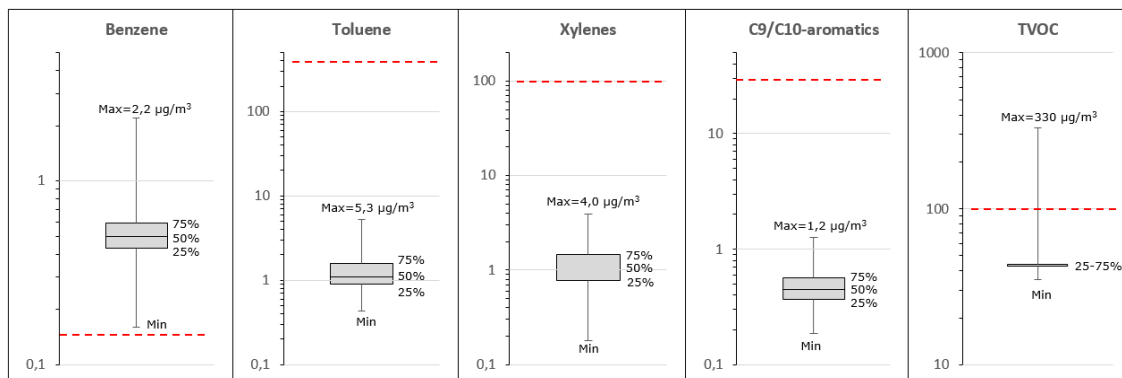


FIGURE 4.1. Box-and-whiskers plots for the outdoor VOC concentrations from the November 2015 sampling campaign (128 residences). Regulatory limits are shown as red dashed lines.

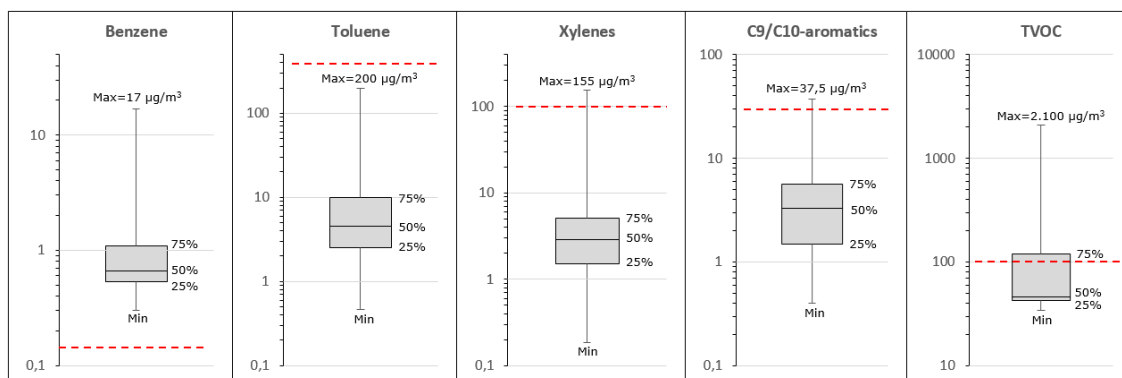


FIGURE 4.2. Box-and-whiskers plots for the indoor VOC concentrations from the November 2015 sampling campaign (131 residences). Regulatory limits are shown as red dashed lines.

For TVOC, the number of non-detects is 125 (approx. 98%) in outdoor air and 60 (approx. 46%) in indoor air.

All the measured benzene levels (November 2015) in both outdoor and indoor air are above the Danish regulatory limit of $0,13 \mu\text{g}/\text{m}^3$, while all the toluene levels are below the regulatory limit of $400 \mu\text{g}/\text{m}^3$. For outdoor air, all the xylene and C9/C10-aromatics measurements are below the regulatory limits of 100 and $30 \mu\text{g}/\text{m}^3$, respectively, while only one TVOC value is above the regulatory limit of $100 \mu\text{g}/\text{m}^3$. For indoor air, two values of xylene and one of C9/C10-aromatics are above the regulatory limits, while 38 measurements of TVOC (29%) are above the regulatory limit.

Summary statistics for the first measurement round are presented in table below. Min and Max are the lowest and highest measured values in the data-sets, respectively.

TABLE 4.1. Summary statistics for outdoor and indoor VOC levels (November 2015).

	No. data points	Min	Percentiles					Max
			10%	25%	50%	75%	90%	
Outdoor air		$\mu\text{g}/\text{m}^3$						
Benzene	128	0,16	0,36	0,43	0,50	0,59	0,70	2,2
Toluene	128	0,43	0,60	0,90	1,1	1,6	2,5	5,3
Xylenes	128	0,18	0,53	0,78	1,0	1,5	2,0	4,0
C9/C10-aromatics	124	<0,19	0,27	0,37	0,45	0,57	0,66	1,2
TVOC	128	<35	<40	<43	<43	<44	<45	330
Indoor air		$\mu\text{g}/\text{m}^3$						
Benzene	131	0,30	0,40	0,53	0,66	1,1	2,5	17
Toluene	131	0,46	1,4	2,6	4,5	9,9	20	200
Xylenes	131	0,19	0,93	1,5	2,9	5,1	15	155
C9/C10-aromatics	75	0,41	1,2	1,5	3,3	5,7	10	38
TVOC	131	<34	<41	<43	<46	120	300	2.100

<DL = Below the detection limit (value specified).

With median benzene levels of $0,50$ and $0,66 \mu\text{g}/\text{m}^3$ and all measured benzene values above the regulatory limit ($0,13 \mu\text{g}/\text{m}^3$), it is clear that ambient benzene can confound conclusions for risk assessments of vapor intrusion in residential buildings based (in part) on measured indoor concentrations.

Although the median value of TVOC is below the analytical detection limit (non-detects), indoor TVOC measurements also have the potential to confound interpretation of vapor intrusion studies, with approx. 29% of the measured values above the regulatory limit of $100 \mu\text{g}/\text{m}^3$.

Distribution plots for benzene (outdoor and indoor) and TVOC (indoor) are presented in the figure below, while distribution plots for all compounds are presented in Appendix 5.

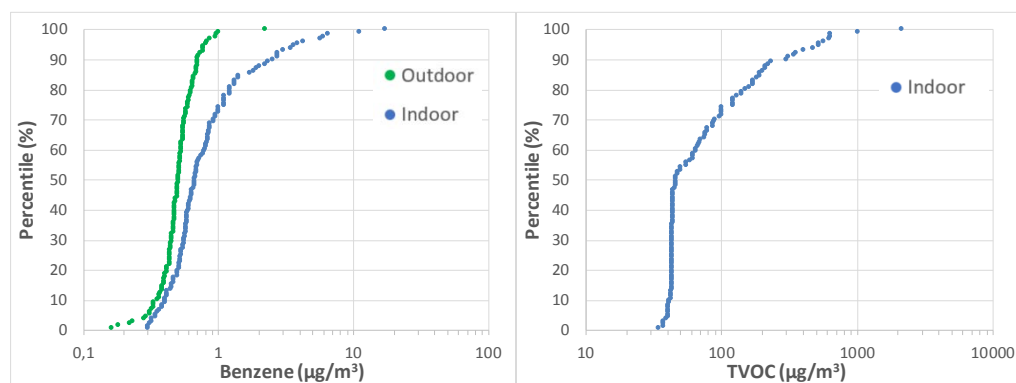


FIGURE 4.3. Distribution functions for benzene (left) and TVOC (right) for the November 2015 sampling campaign (autumn).

4.2 Background levels – May 2016 (spring)

The outdoor and indoor results from the May 2016 measuring campaign are shown as box-and-whiskers plots in the following figures. Sum-parameters are treated in categories corresponding to the Danish regulatory limit values for vapor intrusion contribution to the indoor air, cf. section 2.1. Hence, TVOC is the sum of C6-C35 hydrocarbons and xylenes represent the sum of o/m/p-xylene and ethylbenzene.

For C9-aromatics, 36 outdoor and 52 indoor samples showed component interference in the analysis. Hence, only 102 outdoor samples and 90 indoor samples are included in the statistical analysis for C9/C10-aromatics.

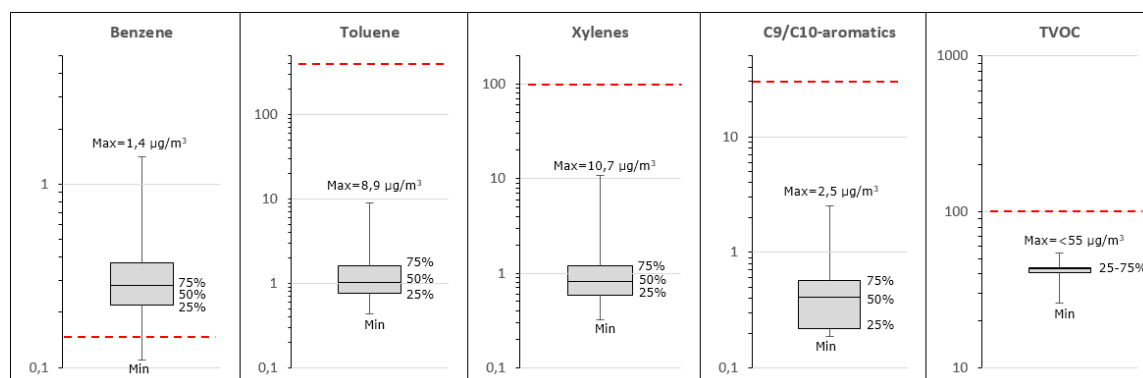


FIGURE 4.4. Box-and-whiskers plots for the outdoor VOC concentrations of the May 2016 sampling campaign (138 residences). Regulatory limits are shown as red dashed lines.

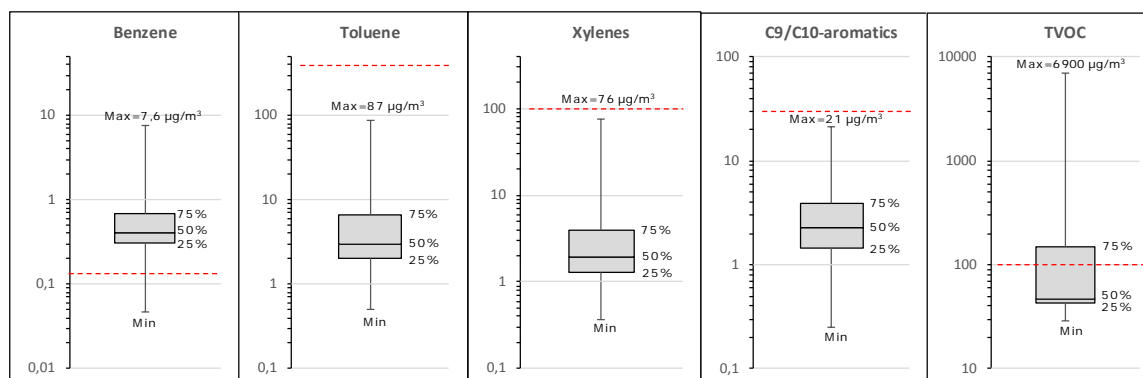


FIGURE 4.5. Box-and-whiskers plots for the indoor VOC concentrations of the May 2016 sampling campaign (142 residences). Regulatory limits are shown as red dashed lines.

All the measured benzene levels (May 2016) in both outdoor and indoor air (except three) are above the Danish regulatory limit of $0,13 \mu\text{g}/\text{m}^3$, while all the toluene, xylene and C9/C10-aromatics levels are below the regulatory limits in both outdoor and indoor air.

For TVOC, the number of non-detects is 138 (100%) in outdoor air and 95 (approx. 67%) in indoor air. For TVOC in indoor air, 47 (33%) measurements are above the regulatory limit of $100 \mu\text{g}/\text{m}^3$.

Summary statistics for the second measurement campaign are presented in table below. Min and Max are the lowest and highest measured values in the data-sets, respectively.

TABLE 4.2. Summary statistics for outdoor and indoor VOC levels (May 2016).

	No. data points	Min	Percentiles					Max
			10%	25%	50%	75%	90%	
Outdoor air		µg/m³						
Benzene	138	0,11	0,19	0,22	0,28	0,37	0,51	1,4
Toluene	138	<0,43	0,61	0,77	1,0	1,6	2,8	8,9
Xylenes	138	<0,33	0,48	0,58	0,81	1,2	2,2	11
C9/C10-aromatics	102	<0,19	<0,21	<0,22	0,41	0,57	0,79	2,5
TVOC	138	<26	<38	<41	<43	<44	<45	<55
Indoor air		µg/m³						
Benzene	142	0,044	0,23	0,30	0,41	0,68	1,9	7,6
Toluene	142	0,50	1,3	2,0	3,0	6,6	13	87
Xylenes	142	<0,38	0,89	1,3	2,0	4,0	9,2	76
C9/C10-aromatics	90	<0,25	0,87	1,4	2,3	3,9	6,6	21
TVOC	142	<29	<40	<43	<47	148	230	6.900

<DL = Below the detection limit (value specified).

With median benzene levels of 0,28 and 0,41 µg/m³ and more than 98% of the measured benzene values above the regulatory limit (0,13 µg/m³), it is clear that ambient benzene can confound conclusions for risk assessments of vapor intrusion in residences based (in part) on measured indoor concentrations. The same is true for indoor ambient TVOC levels, where approx. 33% of the measured values are above the regulatory limit of 100 µg/m³.

Distribution plots for benzene (outdoor and indoor) and TVOC (indoor) are presented in the figure below, while distribution plots for all compounds are presented in Appendix 5.

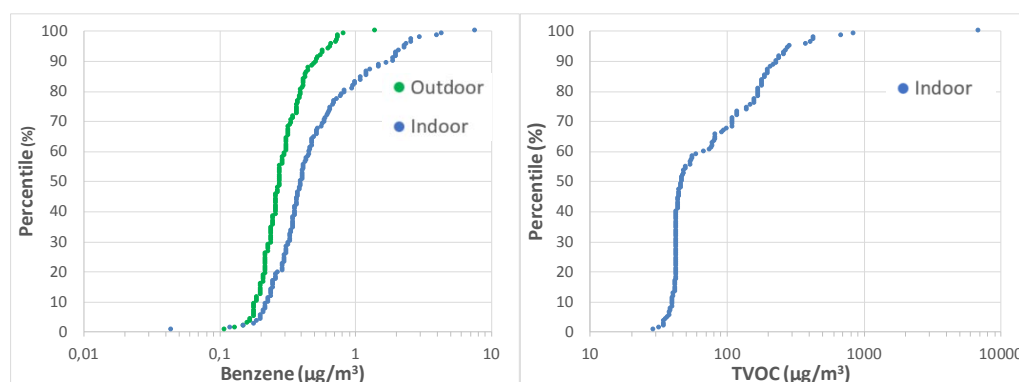


FIGURE 4.6. Distribution functions for benzene (left) and TVOC (right) for the May 2016 sampling campaign (spring).

4.3 Comparison to previous Danish background levels

In table 4.3, the new background levels (November 2015 and May 2016) are summarized and compared to the previous levels (2000-2008 from /1/). From the table, it is seen that there is a tendency towards lower median levels in the spring (May 2016) than in the autumn (November 2015).

Comparing the new background levels from November 2015 to the previous Danish VOC background levels, there is quite a good correspondence between both outdoor and indoor levels of benzene, xylenes and C9/C10-aromatics, with a tendency towards slightly lower values in the new study. For toluene, however, the new background levels tend to be significantly lower with median outdoor and indoor values of 1,1 and 4,5 µg/m³ (November 2015), compared to 1,8 and 7,3 µg/m³ (2000-2008). Also, the indoor TVOC level is significantly lower in the new data set with a median level of <46 µg/m³ (November 2015), compared to 275 µg/m³ (2000-2008).

In the May 2016 data, both outdoor and indoor levels are significantly lower than the previous data set (2000-2008), except for indoor C9/C10-aromatics levels. Especially the outdoor and indoor benzene levels and the indoor TVOC levels are lower in the May 2016 data set, with median levels of 0,28, 0,41 and <47 µg/m³ (May 2016), respectively, compared to 0,48, 0,80 and 275 µg/m³ (2000-2008).

TABLE 4.3. Summary of median outdoor and indoor background VOC levels from the present study, compared to the previous levels from /1/ and the Danish regulatory limits for VOC contribution to residential buildings from contaminated soil and groundwater. Intervals indicates that the statistic is influenced by non-detects (NDs); the low end of intervals is with NDs = 0 and the high end with NDs = the detection limit /1/.

	Regulatory limit	Median VOC levels		
		Nov. 2015	May 2016	2000-2008 /1/
Outdoor air		µg/m³		
Benzene	0,13	0,50	0,28	0,48
Toluene	400	1,1	1,0	1,8
Xylenes*	100*	1,0*	0,81*	0,80-0,87 [£] 0,18-0,30 [§]
C9/C10-aromatics	30	0,45	0,41	0-1,0
TVOC	100	<43	<43	0-61
Indoor air		µg/m³		
Benzene	0,13	0,66	0,41	0,80
Toluene	400	4,5	3,0	7,3
Xylenes*	100*	2,9*	2,0*	3,0 [£] 0,75 [§]
C9/C10-aromatics	30	3,3	2,3	1,5-1,9
TVOC	100	<46	<47	275

* = sum of o-xylene, m-xylene, p-xylene and ethylbenzene. [£] = xylenes; [§] = ethylbenzene.

<DL = Below the detection limit (value specified).

Bold = Median levels above the regulatory limit.

While the tendency for lower BTEX values might be explained by a reduction in traffic related emissions, cf. section 2.3.2, a hypothesis concerning different measurement techniques has been proposed to explain the apparent reduction in TVOC levels. The samples in /1/ were collected on ATD-samplers and carbon tubes using active sampling, whereas passive sampling on ORSA samplers has been used in this study. This hypothesis has been explored and supported by a collaborative study by the Zealand Region and DMR A/S /30/.

4.4 Indoor internal contribution

In the Danish regulatory framework for vapor intrusion, the VOC contribution to residential buildings from contaminated soil and groundwater is viewed as the average contribution (µg/m³), resulting from a number of underlying and possibly dynamic conditions. Most often, indoor measurements are based on passive samples (i.e. ORSA- or ATD-samplers for which a sampling period of 14-day is usually recommended) so that the assessment is not overtly influenced by short-term dynamic conditions during sampling.

Based on the assumption that indoor air is outdoor air which - dependent on the air exchange rate in the building - has received an additional VOC contribution due to emission from materials or activities within the home, then the internal background contribution from the building can be estimated.

This means that the indoor internal contribution of VOCs can be estimated by subtracting the outdoor level from the indoor level; both based on 14-day passive samples in uncontaminated Danish residential buildings.

4.4.1 Indoor internal contribution of benzene

In the following section, the indoor internal contribution of benzene is estimated at each residence and the distribution functions for the indoor internal contribution across all residences are constructed for the November 2015 and May 2016 measuring campaigns.

The results of the indoor internal contribution for benzene are presented below.

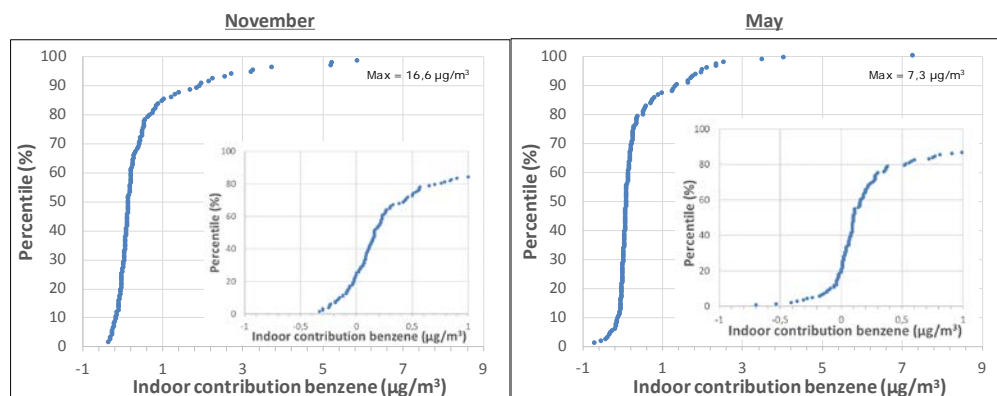


FIGURE 4.7. Distribution functions for the contributions to the indoor benzene concentration, November 2015 (left) and May 2016 (right). Inserted graphs are a zoom on indoor internal contributions of -1 to 1 $\mu\text{g}/\text{m}^3$.

Based on this analysis, the median indoor internal contributions are 0,16 $\mu\text{g}/\text{m}^3$ (November 2015) and 0,10 $\mu\text{g}/\text{m}^3$ (May 2016), while the median for the indoor air is 0,66 $\mu\text{g}/\text{m}^3$ and 0,41 $\mu\text{g}/\text{m}^3$ respectively, cf. table 4.3. Hence, approximately 24% of the indoor benzene concentration is due to internal emissions, in both measurement campaigns.

It should be noted that for approx. 20% of residences (23% and 19%, respectively), the indoor internal contribution of benzene is negligible (≤ 0 when calculated in this manner).

4.4.2 Indoor internal contribution of TVOC

In the following, the indoor internal contribution of TVOC is estimated at each residence and the distribution functions for the indoor internal contribution across all residences are constructed for the November 2015 and May 2016 measuring campaigns. In cases where the indoor concentrations of TVOC were below the detection limit, the indoor internal contribution is set to zero.

The results of indoor internal contribution for TVOC are shown below.

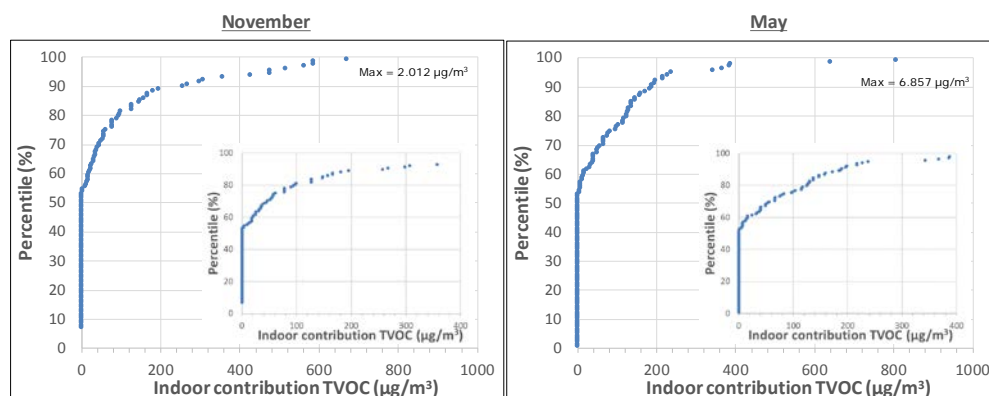


FIGURE 4.8. Distribution functions for indoor internal contributions to the TVOC concentration, November 2015 (left) and May 2016 (right). Inserted graphs are a zoom on indoor internal contributions of up to 400 $\mu\text{g}/\text{m}^3$.

At approx. 53% of the residences in the November 2015 sampling campaign and 51% in the May 2016 sampling campaign, there is no measurable indoor internal contribution of TVOC.

The indoor internal contribution is less than 50 µg/m³ in 70% and 68% of the residences in November 2015 and May 2016, respectively. The indoor internal contribution is less than 100 µg/m³ in 81% and 76% of the residences in November 2015 and May 2016, respectively.

4.4.3 Summary statistics for Indoor internal contribution of all VOCs

Distribution functions for indoor internal contributions of toluene, xylene and C9/C10-aromatics concentrations are shown in Appendix 5.3 and 5.4. Summary statistics for each of the VOCs are shown in the following table.

TABLE 4.4. Statistical summary for indoor internal contribution, November 2015 and May 2016. Median levels (µg/m³), median % of indoor concentration, and % of residences with an indoor internal contribution ≤ zero.

	November 2015			May 2016		
	Median	≤ zero		Median	≤ zero	
Benzene	0,16 µg/m³	24%	23%	0,10 µg/m ³	24%	19%
Toluene	3,4 µg/m³	76%	5%	1,8 µg/m ³	60%	10%
Xylenes	1,5 µg/m ³	52%	11%	1,0 µg/m ³	50%	14%
C9/C10-aromatics	2,9 µg/m³	88%	3%	1,6 µg/m ³	75%	4%
TVOC	<DL	-	-	<DL	-	-

<DL = Median indoor concentration less than the detection level.

Bold = Median indoor internal contribution above the regulatory limit.

From the table, it is seen that the internal contribution of toluene (60-76%), xylenes (50-52%) and C9/C10-aromatics (75-88%) are considerably higher than the indoor internal contribution of benzene (24%). Furthermore, the percentage of residences with an indoor internal contribution of ≤ zero for these compounds is lower (3-14%) than for benzene (19-23%). These statistics indicate that a majority of the indoor concentrations of toluene, xylenes and C9/C10-aromatics are due to indoor sources whereas a majority of the indoor concentrations of benzene are due to outdoor sources.

4.5 Seasonal variation

The results presented in section 4.1 and 4.2 indicate that there is a tendency for the VOC background levels to be slightly lower in May (spring 2016) than in November (autumn 2015). Seasonal variation in the VOC levels is analyzed in more detail in the following.

Seasonal variations are analyzed graphically by comparing the distribution functions of the outdoor and indoor concentrations of individual compounds for the November 2015 and May 2016 measuring campaigns. Mann-Whitney tests at the 5% significance level ($\alpha = 0,05$) have been performed, see Appendix 4.4, whereby p-values of <0,05 indicate that the hypothesis that the two data sets are similar can be accepted (with less than 5% risk of accepting a wrong hypothesis).

Since benzene in both outdoor and indoor air and TVOC in indoor air are the compounds of special interest in vapor intrusion investigations, cf. sections 4.1 and 4.2, these are explored in further detail, while graphs for toluene, xylenes and C9/C10-aromatics are displayed in Appendix 5.5. The results for all compounds are summarized in section 4.5.3.

4.5.1 Seasonal variation in outdoor and indoor benzene levels

As shown in figure 4.9, there is an apparent significant seasonal variation for both indoor and outdoor levels of benzene, with lower concentrations in May 2016 than in November 2015.

The p-values of the Mann-Whitney tests for testing equality of the distributions for outdoor and indoor benzene distributions (November 2015 vs. May 2016) are both <0,001. Hence, the seasonal variability in benzene levels is statistically significant at more than the 99,9% level.

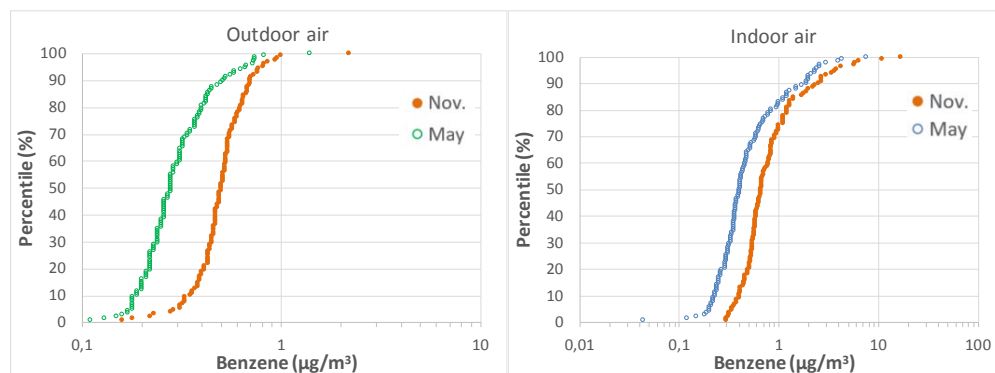


FIGURE 4.9. Benzene distribution functions for November 2015 and May 2016 plotted together for visual exploration of seasonal variation, outdoor levels (left) and indoor levels (right).

4.5.2 Seasonal variation in indoor TVOC levels

In contrast to the benzene results, there is no apparent seasonal variation in the indoor TVOC data, as seen in the figure below.

The p-value of the Mann-Whitney tests for testing equality of the distributions for the indoor TVOC distributions (November 2015 vs. May 2016) is 0,26. Hence, the seasonal variability in the indoor TVOC levels is not statistically significant.

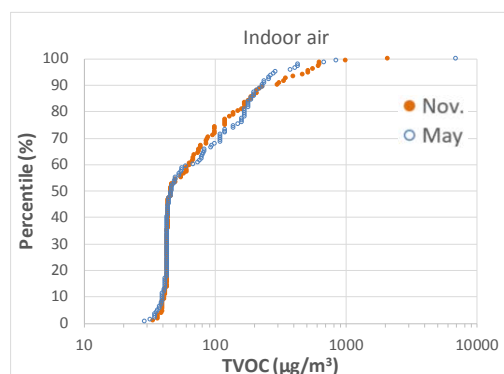


FIGURE 4.10. Distribution functions for TVOC indoor levels for November 2015 and May 2016 plotted together for visual exploration of seasonal variation.

4.5.3 Seasonal variations – summary statistics for all VOCs

Summary statistics for seasonal variations in outdoor and indoor VOC levels, between the November 2015 and May 2016 measuring campaigns, are shown in the table below.

TABLE 4.5. Summary of seasonal variation in VOC levels between November 2015 (autumn) and May 2016 (spring) data.

	Outdoor			Indoor		
	Seasonal variation		p-value	Seasonal variation		p-value
Benzene	Yes	High in autumn	<0,001	Yes	High in autumn	<0,001
Toluene	No	-	0,10	Yes	High in autumn	<0,001
Xylenes	Yes	High in autumn	0,0028	Yes	High in autumn	0,0043
C9/C10-aromatics	Yes	High in autumn	0,029	Yes	High in autumn	0,0032
TVOC	No	-	0,12	No	-	0,26

As seen in the table, there is a significant seasonal variation in the outdoor levels of benzene, xylenes and C9/C10-aromatics, and a significant seasonal variation in the indoor levels of benzene, toluene, xylenes and C9/C10-aromatics.

5. Results – secondary study objectives

The secondary study objectives, cf. section 3.1, are explored in the following sections. Since benzene in both outdoor and indoor air, and TVOC in indoor air, are the compounds of special interest in a vapor intrusion investigation, cf. sections 4.1 and 4.2, only these combinations of compound and outdoor/indoor placement are evaluated in relation to the hypotheses proposed in this study.

The hypotheses are explored graphically by comparing the distribution functions of the individual compounds in outdoor and indoor air and for data sets representing residential buildings with differing metadata (e.g. residences in cities vs. residences in less densely populated areas).

5.1 Effect of population density (outdoor air - benzene)

As mentioned in 3.1.2, it is hypothesized that outdoor levels of VOCs are higher in cities than in less densely populated urban areas (town or village) and in rural areas (countryside). This hypothesis is explored for benzene. All graphs for outdoor air are shown in Appendix 5.6, and the results for benzene are shown below.

“City” is specified as one of the four largest cities in Denmark (Copenhagen, Aarhus, Odense and Aalborg with between 1.300.000 and 113.000 inhabitants), whereas the division between other categories is based on responses given to the questionnaires. For data sampled in November 2015, the percentage of residences in each group (city, town, village and countryside) is approx. 20, 38, 25 and 17%. For data sampled in May 2016, the percentage of residences in each group is approx. 12, 53, 20 and 15%.

In both campaigns (November 2015 and May 2016), the fraction of residences in the “town” category is so high, that these distributions mostly coincide with the distributions for the entire data-set (with “median” distributions). Hence, the study focusses on differences in distributions from cities, villages and the countryside in the following evaluations.

As indicated by the figures below, benzene concentrations are generally lower in the cities than in both villages and the countryside in the November 2015 (autumn) campaign, whereas the tendency is reversed in the May 2016 (spring) campaign. This surprising result is explored in more detail in the following, and in sections 5.8 and 6.3.

The tendency for a reversal of the order between the two measurement campaigns could be because the measured outdoor air benzene concentrations (in the majority of cases) are affected by (i) traffic emissions and (ii) emissions from wood-burning stoves and biomass furnaces, and (iii) photochemical degradation. And that the balance between emission sources and sinks are different for the different seasons and for the different kinds of urban and rural areas.

Whereas traffic emissions can be assumed to be constant throughout the seasons, emissions from domestic heating through residential biomass burning (including wood-burning stoves) and district heating plants can safely be assumed to be higher in the autumn than in the spring. Also, the sink (removal) associated with photochemical degradation is assumed to be more important in the spring than in the autumn.

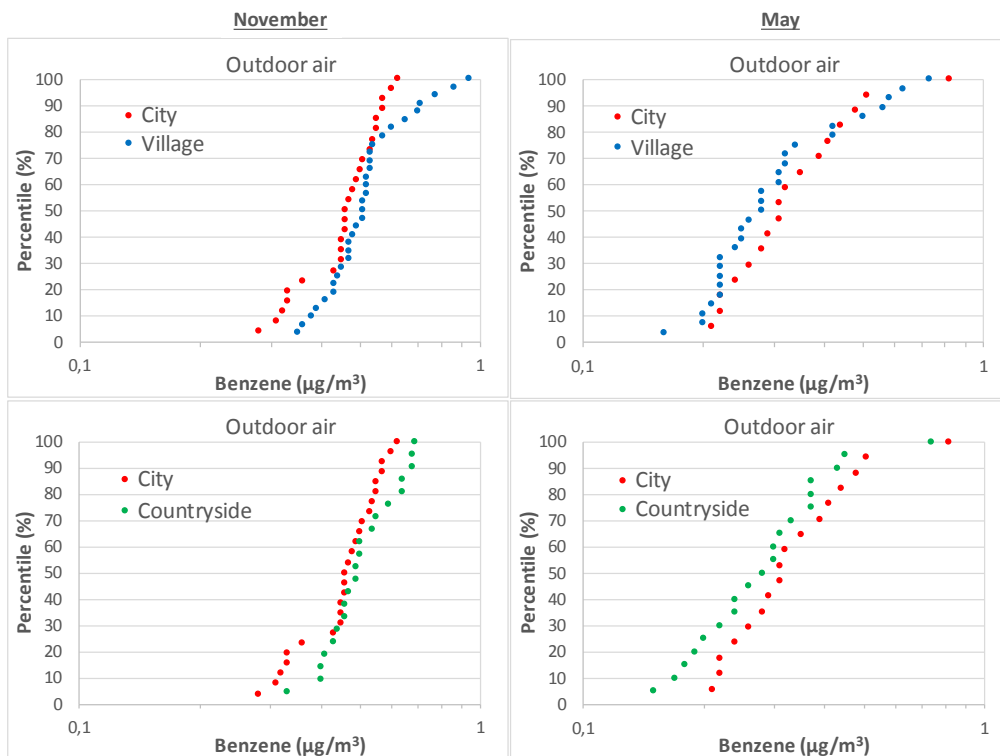


FIGURE 5.1. Distribution functions for outdoor benzene concentrations for city, village and countryside data. Results for November 2015 (left) and May 2016 (right). Log scale.

To explore the sub-hypothesis that the balance between emission sources and sinks are different for the different seasons and for areas with different population densities, the seasonal variations in the outdoor benzene levels are illustrated in the graphs below.

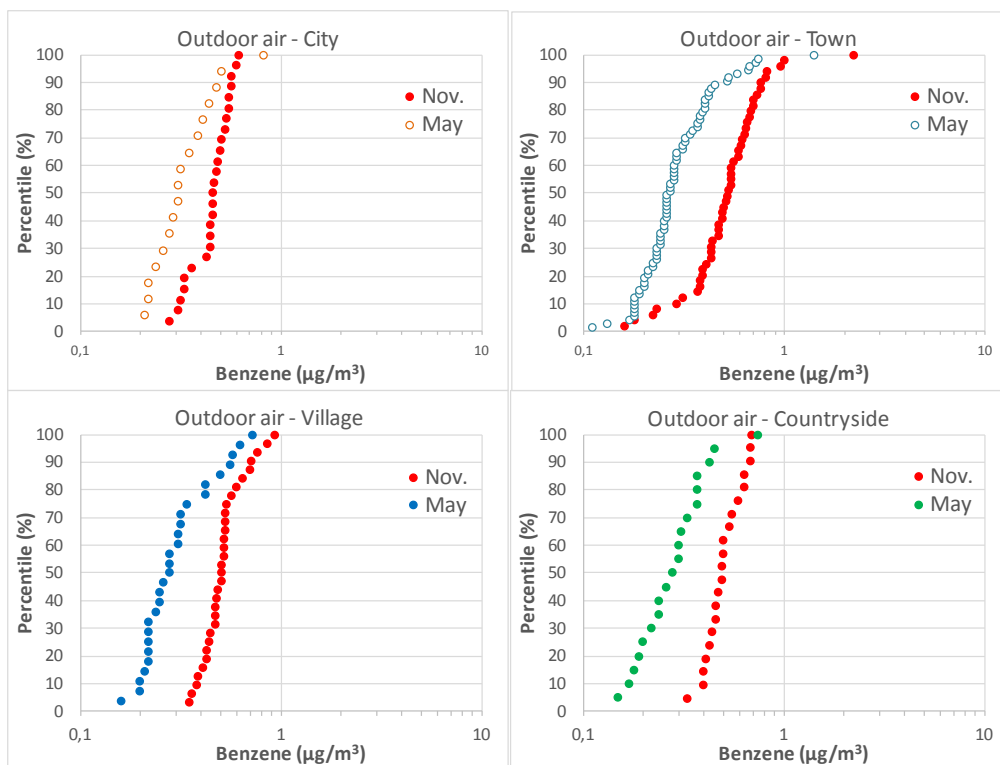


FIGURE 5.2. Seasonal variations in the distribution functions for outdoor benzene concentrations measured in cities (top left), towns (top right), villages (bottom left) and in the countryside (bottom right).

As seen in the graphs, the seasonal effects are the same in all the environments: The outdoor benzene concentrations are lower in May 2016 than in November 2015. And while the overall level of benzene in outdoor air is of the same order of magnitude in urban and more rural areas, it is quite clear that the seasonal variability is lower in the city than in the other environments.

These results could indicate that the outdoor benzene level in the cities is relatively more affected by traffic (a constant emission source), while the levels in villages and the countryside, especially in the heating season (autumn) is relatively more affected by domestic heating by biomass burning (a variable emission source).

As the results above can be of significance with respect to interpretation of vapor intrusion at contaminated sites, the variability in outdoor benzene levels in relation to seasonal variation and density of population can easily be assessed by including an outdoor sampling point when otherwise sampling at the location.

5.2 Effect of tobacco smoking (indoor benzene and TVOC)

As mentioned in 3.1.2, it is hypothesized that indoor levels of VOCs are higher in residences where smokers smoke indoors than in residences without indoor smoking. This hypothesis is explored for benzene and TVOC.

The number of residences with indoor tobacco smoking are 11 (8,4%) and 10 (7,0%) for the November 2015 and May 2016 sampling campaigns respectively.

To investigate the significance of tobacco smoking with respect to indoor levels of benzene, the residences with indoor tobacco smoking have been marked by red circles in the total distribution functions for indoor benzene (November 2015 and May 2016, respectively).

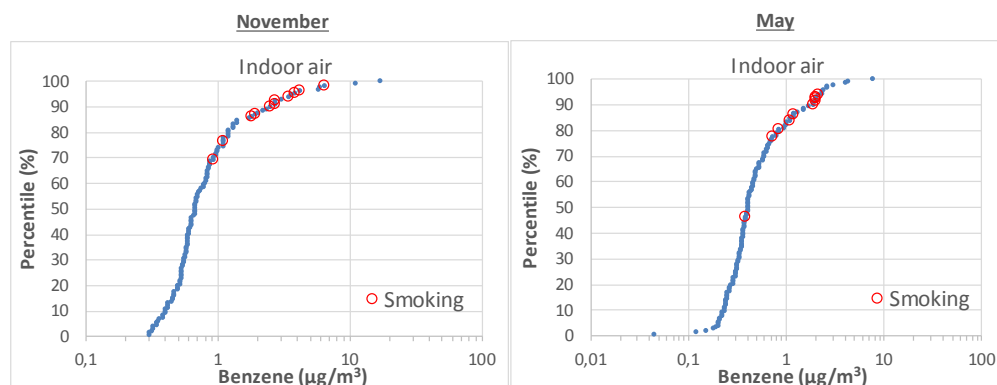


FIGURE 5.3. Total distribution functions for indoor benzene concentrations, November 2015 (left) and May 2016 (right). Data points from residences with indoor smoking are circled in red.

As seen in the graphs, it is apparent that benzene concentrations measured in residences with indoor smoking are located at the high end of the distribution functions; hence it can be inferred that indoor smoking has a marked effect on the indoor benzene concentration. Whereas the median indoor concentrations in the total data sets for benzene in November 2015 and May 2016 are respectively 0,66 and 0,41 $\mu\text{g}/\text{m}^3$, the corresponding median levels in residences with indoor smoking are 2,7 and 1,6 $\mu\text{g}/\text{m}^3$ (4,1 and 3,9 times higher).

When looking at the highest 10% of the total data sets (i.e. approx. 13 and 14 measurements respectively for November 2015 and May 2016), residences with indoor smoking represent respectively 7 and 5 measurements for November 2015 and May 2016. This means that 54% and 36% of the residences with the 10% highest benzene concentrations are residences with

indoor smoking in the November 2015 and May 2016 data sets. This reveals that residences with indoor smoking are statistically over-represented in the residences with the 10% highest benzene concentrations.

Hence, the results indicate that indoor concentrations of benzene are significantly higher in residences with indoor smoking than in residences in general. The median value is approx. 4 times higher for residences with indoor smoking than residences in general. However, as seen in figure 5.3, the very highest values of benzene concentrations are not measured in homes with indoor smoking.

To investigate the significance of tobacco smoking with respect to indoor levels of TVOC, the residences with indoor tobacco smoking have been marked by red circles in the total distribution functions for indoor TVOC (November 2015 and May 2016 respectively).

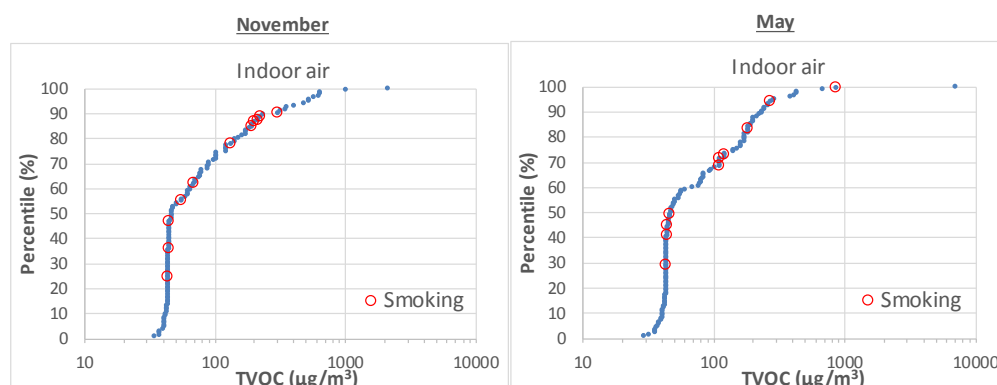


FIGURE 5.4. Total distribution functions for indoor TVOC concentrations, November 2015 (left) and May 2016 (right). Data points from residences with indoor smoking are circled in red.

As opposed to the benzene results, the TVOC concentrations are not all located at the high end of the distribution functions, but are more evenly distributed. Whereas the median indoor concentrations of TVOC in November 2015 and May 2016 in the total data sets are <46 and $<47 \mu\text{g}/\text{m}^3$, respectively, the corresponding median levels in residences with indoor smoking are 130 and $110 \mu\text{g}/\text{m}^3$ ($>2,8$ and $>2,3$ times higher).

When looking at the highest 10% of the total data sets (i.e. approx. 13 and 14 measurements respectively for November 2015 and May 2016), residences with indoor smoking represent 1 and 2 measurements respectively for November 2015 and May 2016. This means that respectively only 7,7 and 13% in the November 2015 and May 2016 data sets of the residences with the 10% highest TVOC concentrations are residences with indoor smoking. Hence, residences with indoor smoking are not statistically over-represented in the residences with the 10% highest TVOC concentrations.

Hence, the results indicate that indoor concentrations of TVOC are slightly higher in residences with indoor smoking than in residences in general. The median value is $>2,3$ - $2,8$ times higher for residences with indoor smoking than residences in general. However, as seen in figure 5.3, the very highest values of TVOC concentrations are not measured in homes with indoor smoking.

The effects of indoor tobacco smoking on VOCs in indoor air are highly significant, and indoor measurement of benzene and TVOC could easily lead to erroneous conclusions regarding vapor intrusion, unless smoking is accounted for in the data interpretation. Hence, indoor sampling in homes of smokers should only be included in vapor intrusion studies when absolutely necessary and interpreted with care.

5.3 Effect of wood-burning stoves (indoor benzene and TVOC)

Based on a literature review, it is hypothesized that indoor levels of VOCs are higher in residences with wood-burning stoves than in homes without wood-burning stoves. This hypothesis is explored for benzene and TVOC.

The number of residences with wood-burning stoves are 43 (33%) and 40 (28%) respectively for the November 2015 and May 2016 sampling campaigns.

To investigate the significance of wood-burning stoves with respect to indoor levels of benzene, the distribution functions for homes with and without wood-burning stoves are plotted next to each other (for November 2015 and May 2016 respectively).

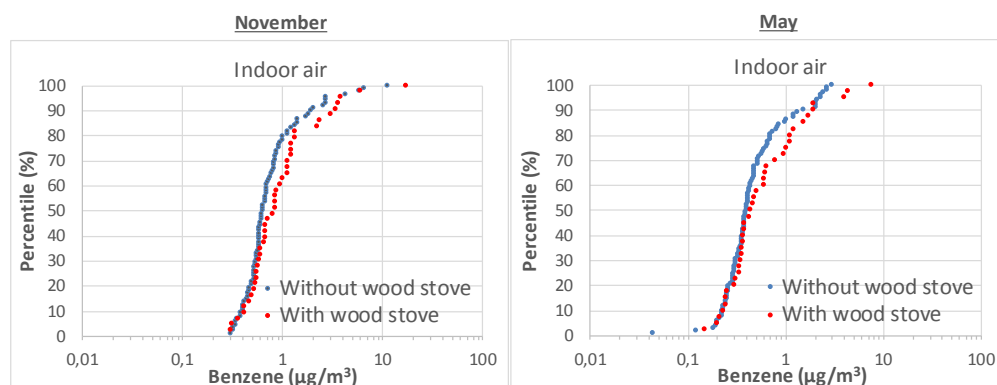


FIGURE 5.5. Distribution functions for indoor benzene concentrations with and without wood-burning stoves, November 2015 (left) and May 2016 (right).

From the graphs, it is apparent that benzene concentrations measured in residences with wood-burning stoves tend to be slightly higher than in residences without stoves. The median indoor concentrations of benzene in November 2015 and May 2016 in the residences without wood-burning stoves are respectively 0,63 and 0,40 µg/m³. The corresponding median levels in residences with wood-burning stoves are 0,83 and 0,45 µg/m³ (32 and 13% higher).

To investigate the significance of wood-burning stoves with respect to indoor levels of TVOC, the residences with and without stoves are plotted next to each other (for November 2015 and May 2016 respectively).

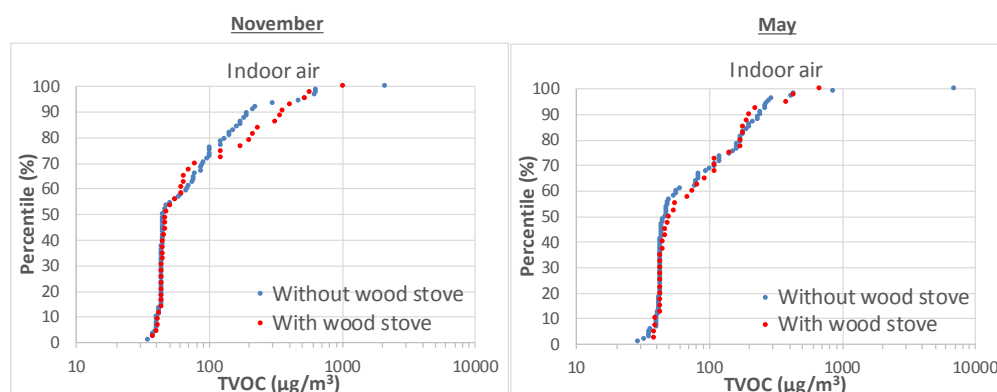


FIGURE 5.6. Distribution functions for indoor TVOC concentrations with and without wood-burning stoves, November 2015 (left) and May 2016 (right).

From the graphs, it is apparent that the median TVOC concentrations in residences with wood-burning stoves are not higher than those measured in homes without stoves. However, there is a tendency for the TVOC concentrations to be higher in homes with stoves (at the high end

of the distribution) in the November 2015 measuring campaign, indicating that wood-burning stoves can act as an internal source of TVOC in some residences in the cold season.

Overall, the slight tendency for higher indoor levels of benzene and TVOC due to wood-burning stoves is of minor consequence with respect to vapor intrusion studies.

5.4 Effect of ventilation (indoor benzene and TVOC)

As mentioned in 3.1.2, it is hypothesized that indoor levels of VOCs are lower in residences with a higher frequency of ventilation (by manual airing or mechanical ventilation) than in residences with a lower frequency. This hypothesis is explored for benzene and TVOC.

Based on the responses to the questionnaire, the data are divided into groups of residences that report airing out of the house <once a day, once or twice a day and >twice a day. The group of more than twice a day includes residences with mechanical ventilation systems. For the November 2015 data set, the percentage of residences in each group is approx. 24, 48 and 28% respectively. For the May 2016 data set, the percentage of residences in each group is approx. 14, 43 and 43% respectively.

Figure 5.7 shows the distribution functions of indoor benzene and TVOC concentrations for both the November 2015 and May 2016 sampling campaigns.

Based on the visual trends of the graphs, there are only minor differences between the benzene and TVOC distributions based on the frequencies for airing the house. Basically, the results are inconclusive and ambiguous.

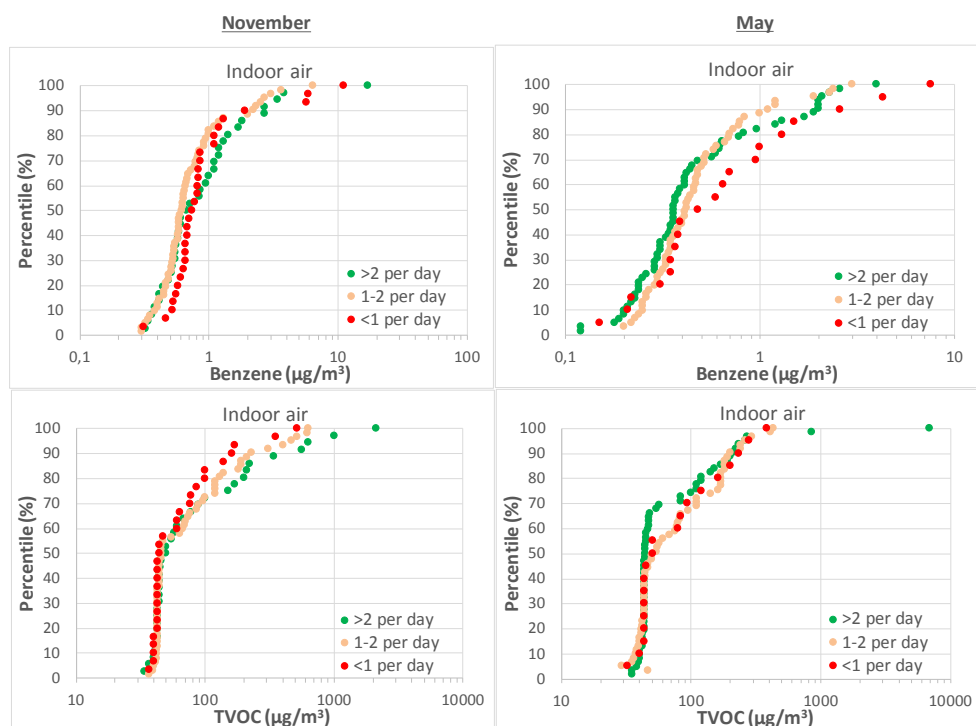


FIGURE 5.7. Distribution functions of indoor benzene concentrations (top) and TVOC concentrations (bottom) according to the frequency of airing out the house, November 2015 (left) and May 2016 (right).

5.5 Effect of home improvement (indoor benzene and TVOC)

As mentioned in 3.1.2, it is hypothesized that indoor levels of VOCs are higher in residences that have recently been renovated. This hypothesis is explored for benzene and TVOC.

Based on the responses to the questionnaire, the data are divided into groups of residences where renovation/home improvement or remodeling activities have been carried out within a year before measurements. The fraction of residences with renovation activities is approx. 43 and 37% for the November 2015 and May 2016 measurement campaigns. Home improvement, renovation or remodeling can cover a wide variety of activities such as painting, home extensions, new furniture, carpets or kitchen elements etc.

Figure 5.8 shows the distribution functions of indoor benzene and TVOC concentrations for both the November 2015 and May 2016 sampling campaigns.

For benzene, there is a minor tendency for higher concentrations after home improvements, especially at lower concentrations. For TVOC, there is a significant tendency for higher indoor concentrations around the median in November 2015, but neither at the low or the high end of the distribution. Basically, there are no significant effects on the indoor benzene or TVOC levels arising from recent home improvement activities.

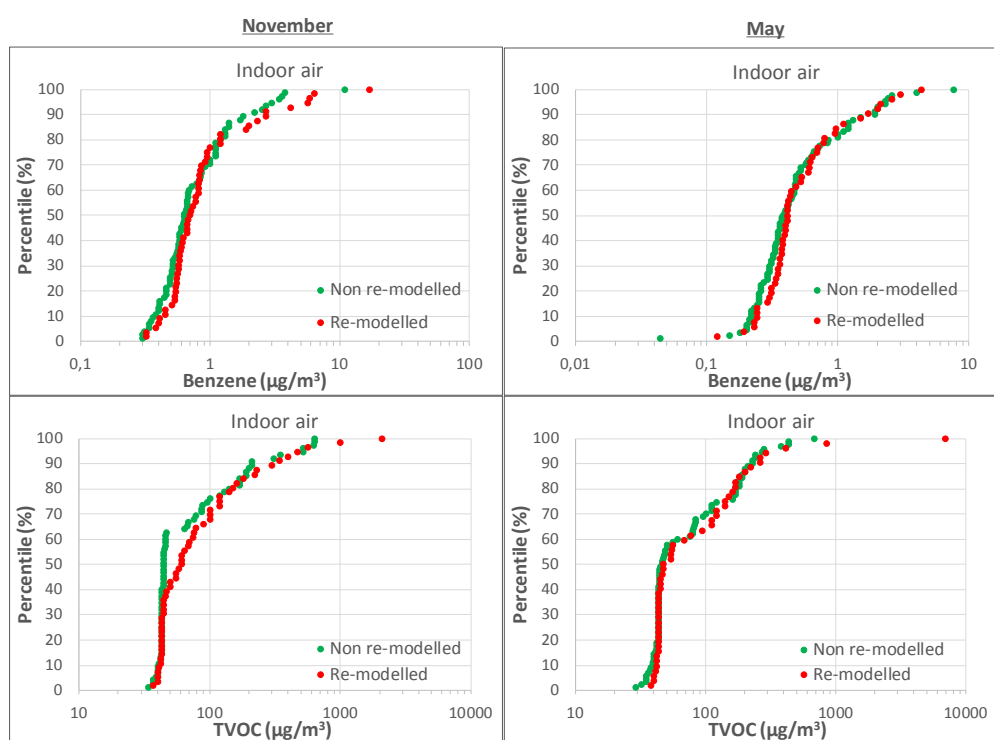


FIGURE 5.8. Distribution functions of indoor benzene concentrations (top) and TVOC concentrations (bottom) and the effect of home improvement/remodeling, November 2015 (left) and May 2016 (right).

5.6 Effect of occupancy (indoor benzene and TVOC)

It is hypothesized that indoor levels of VOCs are higher in residences that have high occupancies due to higher emissions. This hypothesis is explored for benzene and TVOC.

Based on the responses to the questionnaire, the data are divided into groups of residences with occupancy of 1, 2, 3, 4 and 5/6 people per household. For the November 2015 data set, the percentage of residences in each group is approx. 11, 37, 15, 27 and 10% respectively. For the May 2016 data set, the percentage of residences in each group is approx. 11, 38, 13, 29 and 9%.

As seen from the graphs in figure 5.9, there are no significant and consistent trends for the indoor benzene and TVOC levels to be higher with higher occupancy. On the contrary, resi-

dences with 5 or 6 occupants seem to be associated with lower concentrations than most of the other categories. Homes with 2 occupants seem to be associated with high levels of both benzene and TVOC.

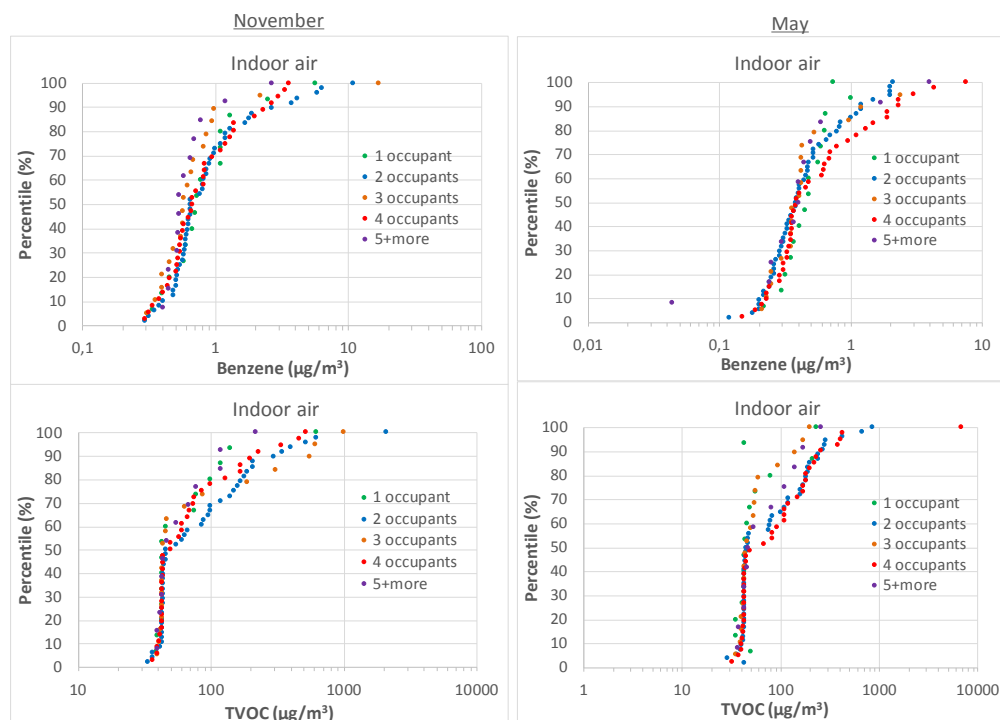


FIGURE 5.9. Distribution functions of indoor benzene concentrations (top) and TVOC concentrations (bottom) and the effect of number of occupants per household, November 2015 (left) and May 2016 (right).

5.7 Effect of building age (indoor benzene and TVOC)

As mentioned in 3.1.2, it is hypothesized that indoor levels of VOCs are lower in older residences due to less energy efficient constructions and a higher natural air exchange rate. This hypothesis is explored for benzene and TVOC. Building age is related to the construction year.

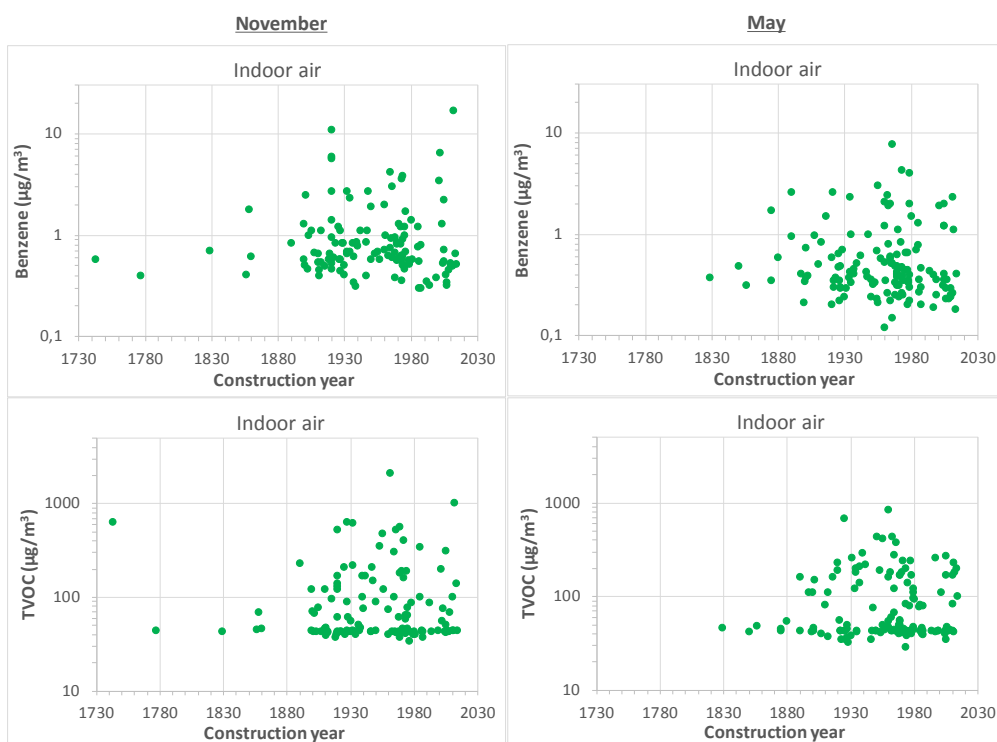


FIGURE 5.10. Indoor concentrations of benzene (top) and TVOC (bottom) plotted versus building construction year, November 2015 (left) and May 2016 (right).

The graphs in figure 5.10 indicate a slight tendency towards lower indoor benzene and TVOC concentrations for buildings constructed before 1880, but there are very few data points for such old buildings, and one high data value for TVOC in the November 2015 data set for the oldest building indicate that the trend cannot be assumed to be generally valid.

For the rest of the data, a slight tendency for higher values in residences constructed in 1950-1970 is indicated.

5.8 Effect of traffic related emissions (outdoor benzene)

The exploration of traffic related emissions is divided into three sub-studies; street type/traffic load, distance to traffic lights, and distance to gas station. The hypotheses are that a higher traffic density (related to the street type), or shorter distance to either traffic lights or a gas station will lead to higher benzene concentrations in the outdoor air.

5.8.1 Street type/traffic density (outdoor benzene)

Based on the responses to the questionnaire, the data are divided into the following groups, with a descending order of traffic density: Motorway/highway, urban street and suburban street. For the November 2015 data set, the percentage of residences in each group is approx. 15, 31 and 54% respectively. For the May 2016 data set, the percentage of residences in each group is approx. 6, 37 and 57% respectively.

The effect of street type/traffic density on the outdoor benzene concentration is explored in the figure 5.11.

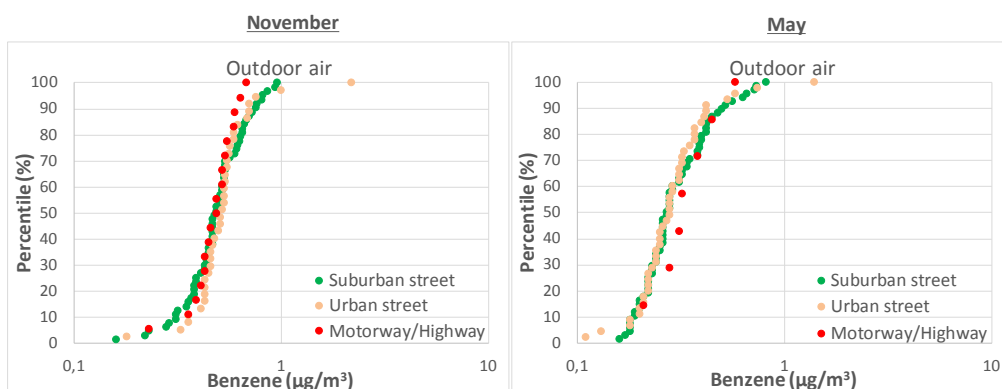


FIGURE 5.11. Distribution functions of outdoor benzene concentrations and effect of street type/traffic density, November 2015 (left) and May 2016 (right).

As seen in the above graphs, there are no significant effects of street type/traffic density on the outdoor benzene concentrations.

5.8.2 Distance to traffic lights (outdoor benzene)

Based on the responses to the questionnaire, 13 residences (approx. 10%) are located with less than 100 meters to traffic lights in the November 2015 campaign and 7 residences (approx. 5%) in the May 2016 campaign.

The outdoor results for benzene related to the distance to traffic lights are presented in the figure below.

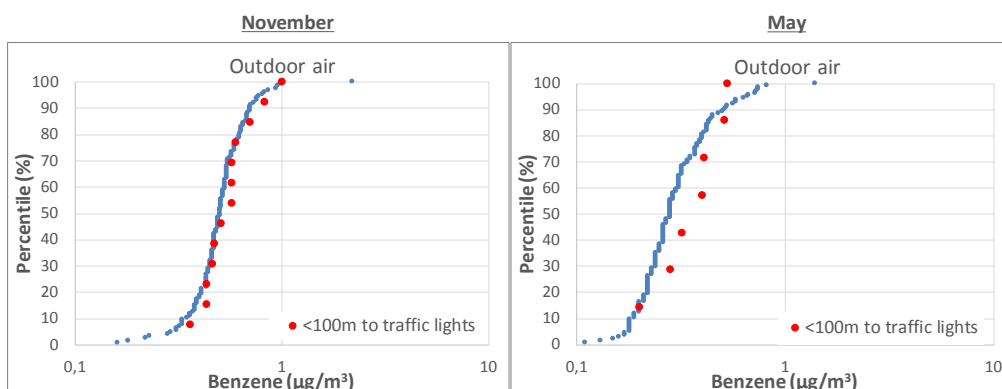


FIGURE 5.12. Distribution functions of outdoor benzene concentrations and effect of distance to traffic lights, November 2015 (left) and May 2016 (right).

Only a few data fit the parameter for distance to traffic lights, but the distribution functions show a slight tendency for outdoor benzene concentrations to be higher for the data sets with less than 100 meters to traffic lights, especially for the May 2016 data set.

5.8.3 Distance to gas station (outdoor benzene)

Based on the responses to the questionnaire, 13 residences (approx. 10%) are located with less than 100 meters to a gas station in the November 2015 campaign and 8 residences (approx. 6%) in the May 2016 campaign.

The outdoor results for benzene related to the distance to a gas station are presented in the figure below.

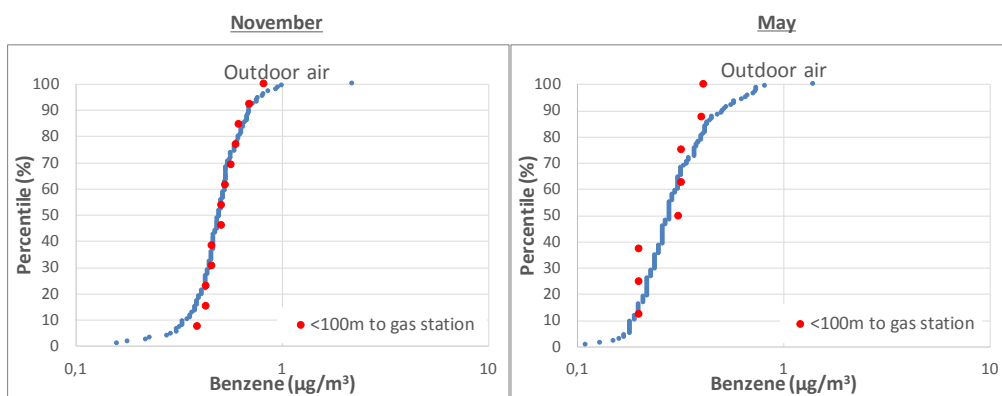


FIGURE 5.13. Benzene outdoor air concentrations distribution functions (total – blue dots) and residences with distances <100m to gas station (red circles) for the November 2015 (left) and May 2016 (right) sampling campaign.

Only a few data fit the parameter for distance to the nearest gas station, but as seen in the above graphs, there are no significant effects on the outdoor benzene concentration in relation to distance to the nearest gas station.

6. Results – side studies

The results of the side studies listed in section 3.1.2 are presented in the following sections.

6.1 Study of variability

In this side study, seasonal variability in benzene (outdoor and indoor) as well as TVOC (indoor) levels is investigated in more detail in three homes. We also investigate inter-duplicate variability in both outdoor and indoor levels, and inter-position variability in outdoor levels.

Eight sampling campaigns were carried out over a one-year period (August 2016 to September 2017) with placement of outdoor ORSA-samplers on the North, South, East and West sides of each residence, and indoor sampling in one position. All positions were sampled in duplicate.

6.1.1 Seasonal variability (benzene and TVOC)

The results of benzene concentrations in both outdoor and indoor air are shown below.

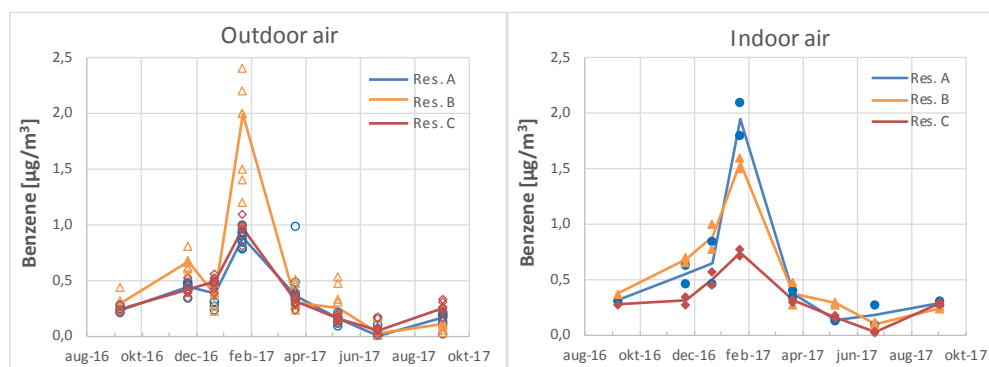


FIGURE 6.1. Seasonal variability in outdoor (left) and indoor (right) benzene concentrations at residence A, B and C included in the study of variability. Symbols show individual measurements and lines show median levels.

As seen from the graphs, there is a common tendency for both outdoor and indoor benzene concentrations to peak in January, and be at their lowest in June. However, there are also differences between the three residential buildings, where outdoor benzene levels tend to be higher at residence B than at A and C, while the indoor benzene concentration tend to be lower at residence C than at the other residences. The individual residences are explored in further detail in the figure below.

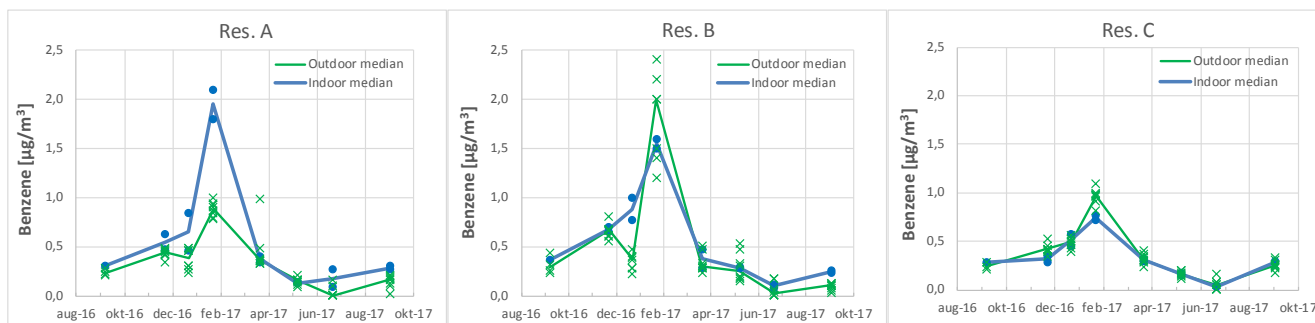


FIGURE 6.2. Seasonal variability in outdoor (green) and indoor (blue) benzene concentrations at residence A, B and C included in the study of variability. Symbols show individual measurements and lines show median levels.

From the figure above, it is apparent that although the overall tendency for benzene concentrations to be high in January and low in June, there are notable differences between the residences. Hence, the data from residence A indicate a higher indoor internal contribution than residence B and C. Residence B seems to be influenced by a strong outdoor source of benzene in the heating season. Both residence B and C have a negative indoor internal contribution in January 2017.

From the metadata presented in table 3.1, it is noted that residence A has the lowest ventilation frequency whereas residence B has a wood-burning stove and is partially heated by a biomass furnace. These factors could explain the differences seen in figure 6.2. Especially the high outdoor values of benzene at residence B is in correspondence with literature information about biomass burning being a significant source of outdoor benzene levels, cf. section 2.2.

The results of TVOC concentrations in indoor air are shown in the figures below.

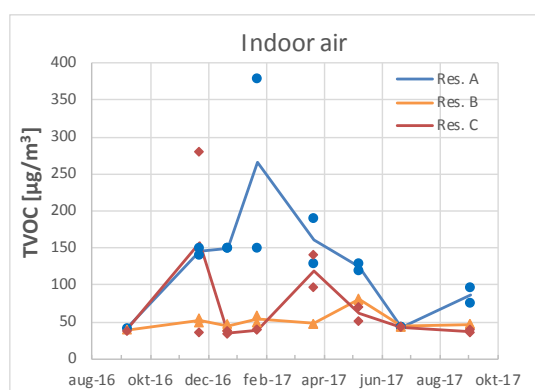


FIGURE 6.3. Seasonal variability in indoor TVOC concentrations at residence A, B and C included in the study of variability. Symbols show individual measurements and lines show median levels.

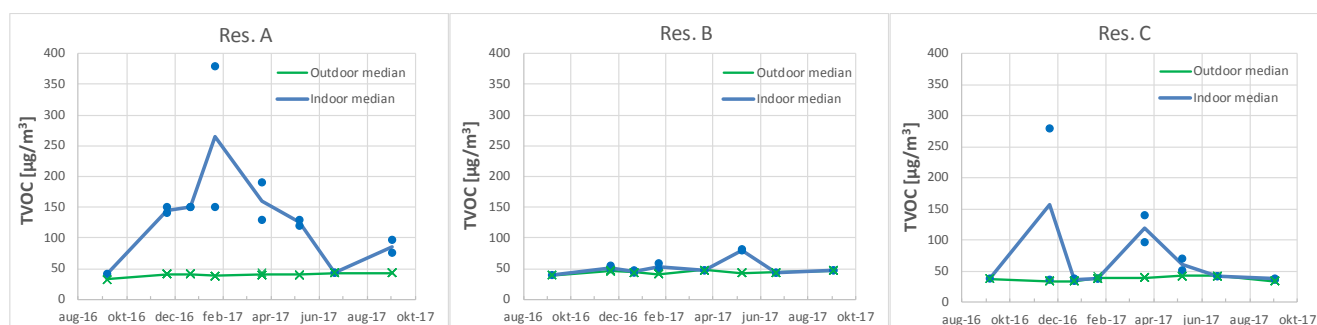


FIGURE 6.4. Seasonal variability in outdoor (green) and indoor (blue) TVOC concentrations at residence A, B and C included in the Three-Residence Study. Symbols show individual measurements and lines show median levels.

As seen in the graphs above, most of the TVOC measurements (both outdoor and indoor) are below the detection limit, and except for residence A. Indoor TVOC occurrence seems to be sporadic. In contrast, residence A seems to have a significant indoor source of TVOC, corresponding to the pattern for benzene at this residence, cf. figure 6.2. Higher indoor levels at this residence might also be influenced by a lower frequency of airing at this residence, cf. table 3.1.

Graphs for toluene, xylenes, C9- and C10-aromatics are shown in Appendix 5.7. These graphs support the statistics in section 4.4.3, inferring that indoor benzene levels are mostly influenced by outdoor sources, while indoor concentrations of the other compounds are mostly

influenced by indoor sources. Hence indoor levels of toluene, xylenes, C9/C10-aromatics and TVOC tend to be more sporadic than the benzene levels.

6.1.2 Inter-duplicate variability (benzene and TVOC)

All the sampling positions in the study of variability have been sampled in duplicate, and, as indicated in figures 6.1-6.4, there are indications of inter-duplicate variability between measurements from the same position for the same sampling period. The magnitude of inter-duplicate variability is explored in the following.

For the data treatment, all duplicates are paired, and the ratio between the high and the low value is calculated for each position and sampling period. Hence, a max/min-ratio of 1 means that the duplicate measurements are equal. Both indoor and outdoor air concentrations are considered and the results for the three residences are grouped together.

Figure 6.5 shows the distribution function for the inter-duplicate variability for benzene. Since some of the relatively high inter-duplicate variabilities are seen for data sets with values close to the analytical detection limit, two graphs are constructed; one with all data pairs and one with data pairs where both values are above the vapor intrusion relevant level of $0.13 \mu\text{g}/\text{m}^3$.

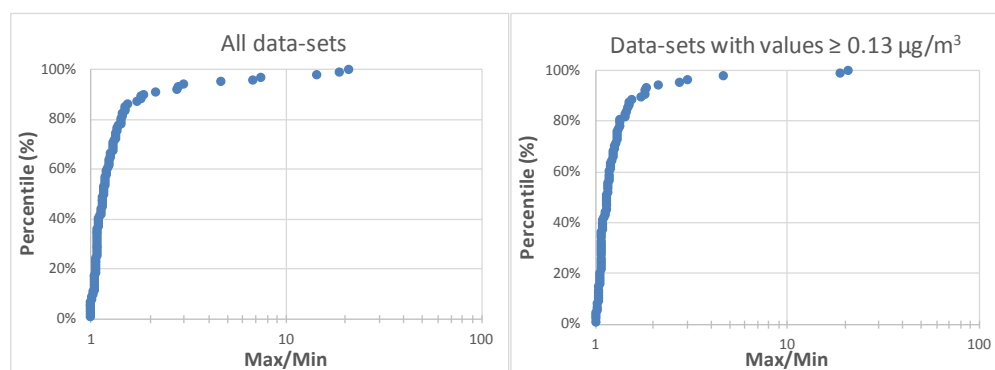


FIGURE 6.5. Distribution functions of max/min-ratios (log scale) for duplicate measurements of benzene in the study of variability (outdoor and indoor samples); all data, N=102 (left) and data-sets with both values $\geq 0,13 \mu\text{g}/\text{m}^3$, N=88 (right).

For the entire data-set, the median inter-duplicate variability of benzene is 1,2 with a 90% confidence interval of 1,0-4,6, and 90% of duplicates with a ratio ≤ 2 .

For the reduced data-set, the median inter-duplicate variability of benzene is 1.1 with a 90% confidence interval of 1,0-2,5, and 96% of duplicates with a ratio ≤ 2 .

Hence, for 50% of benzene samples, the inter-duplicate variability is <10-20% (a max/min-factor of 1,1-1,2), and for 90% of duplicate samples, inter-duplicate variability is less than 100% (a max/min-factor of 2). Inter-duplicate variability is more than 100% (a max/min-factor of 2) for only approx. 4% of vapor intrusion relevant benzene concentrations ($\geq 0,13 \mu\text{g}/\text{m}^3$).

The figure below shows the distribution function for the inter-duplicate variability for TVOC. Since most of the outdoor values are below the analytical detection limit, two graphs are constructed; one with all data pairs and one with values above the analytical detection limit.

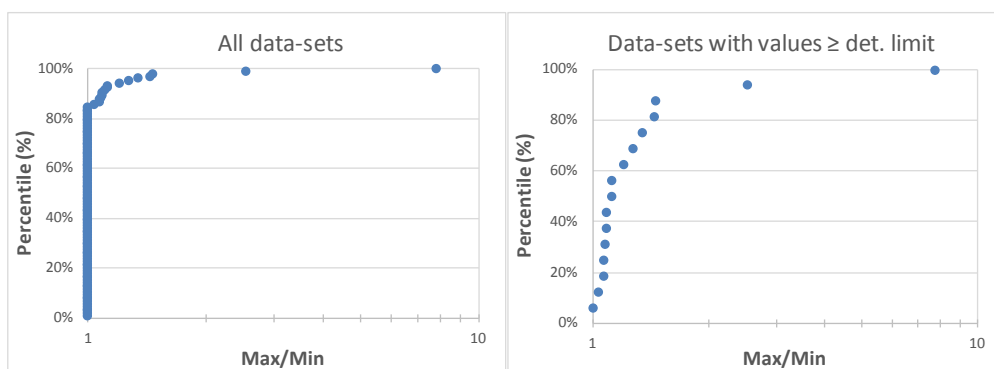


FIGURE 6.6. Distribution functions of max/min-ratios (log scale) for duplicate measurements of TVOC in the study of variability (outdoor and indoor samples); all data, N=105 (left) and data-sets with values above the analytical detection limit, N=16 (right).

For the entire data-set, the median inter-duplicate variability of TVOC is ≤ 2 for 98,6% of the sampling positions. For the reduced data-set, the median inter-duplicate variability of TVOC is 1,1 with a 90% confidence interval of 1,0-3,8, and 91% of duplicates has a ratio ≤ 2 .

Hence, for 50% of TVOC samples, the inter-duplicate variability is $<10\%$ (a max/min-factor of 1,1), and for $>91\%$ of duplicate samples, inter-duplicate variability is less than 100% (a max/min-factor of 2).

6.1.3 Inter-position variability (outdoor benzene)

The outdoor positions in the study of variability have been sampled on four different sides of each residence. The common practice in Danish vapor intrusion studies of measuring outdoor levels is to place an ORSA-sampler in one (random) outdoor sampling position. Only one outdoor measurement of TVOC in this side study is above the analytical detection limit, and therefore inter-position variability is only evaluated for benzene as follows.

To extract the “true” inter-position variability (i.e. not influenced by inter-duplicate variability), the mean values for duplicate results at each combination of sampling position and sampling period is used in the analysis. As in the previous section, the max/min-ratio is calculated by pairing the different sampling locations around each residence for each sampling campaign (6 data pairs per residence per campaign; W-N, W-E, W-S, N-E, N-S and E-S). The results for the three residences are grouped together and are shown in the figure below.

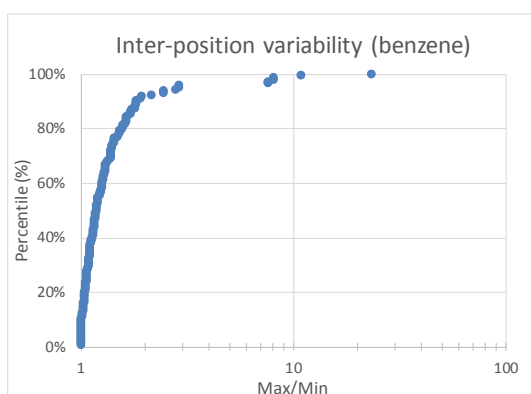


FIGURE 6.7. Distribution of the ratio between the maximum and the minimum values (log scale) of benzene concentrations for 6 different positioning combinations at the three residences (N=144).

The median of the ratios is 1,2 $\mu\text{g}/\text{m}^3$ ([1,0-2,9] $\mu\text{g}/\text{m}^3$ for 90% confidence interval), which demonstrates a rather insignificant difference of 20% between the positions. It is also observed that at about 92% of the cases, the difference is less than 100% (a ratio of 2).

6.2 Detailed study of variability (benzene and TVOC)

At residence C (see section 6.1 for the study of variability), an increased frequency of sampling was carried out prior to the study of variability. At this residence, five additional indoor sampling campaigns (without duplicates) were measured, so almost two full years of monitoring results are available: November 2015 to September 2017. The results are presented in the figure below.

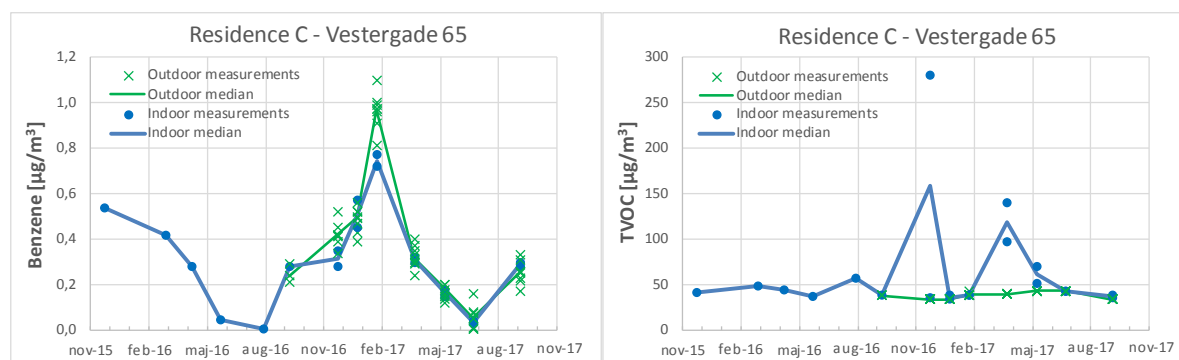


FIGURE 6.8. Benzene (left) and TVOC (right) concentrations for indoor (blue line) and outdoor (green line) air in residence C. Symbols show individual measurements and lines show median levels.

The extended monitoring period as depicted in figure 6.8 clearly illustrates that there is a cyclic structure to the indoor benzene concentrations. Hence, medium-high concentrations are seen in November (0,4-0,6 $\mu\text{g}/\text{m}^3$) and February-March (0,3-0,4 $\mu\text{g}/\text{m}^3$), and low concentrations are seen in May-July ($< 0,1 \mu\text{g}/\text{m}^3$). Unfortunately, no measurements were made in January 2016, to confirm the very high level in January 2017 (0,75 $\mu\text{g}/\text{m}^3$).

The extended TVOC monitoring period confirms that the indoor TVOC levels in residence C are indeed low, and only sporadically exceed the analytical detection limit.

Combined with the benzene results from the study of variability, it appears that benzene levels can be expected to be relatively low in March to August (spring and summer) and relatively high in September to February (autumn and winter).

6.3 Chimney Study (outdoor VOCs)

At residence B (see section 6.1 for the study of variability), additional outdoor sampling positions were established on the west and east side of the chimney from the wood gasification boiler to investigate the local scale emissions from biomass burning. The results from the study of variability (cf. section 6.1.1) indicate that biomass burning can lead to higher local scale concentrations of benzene concentrations in outdoor air.

Figure 6.9 shows the concentrations for all VOCs from the Chimney Study. The data are divided into a period with the wood gasification boiler in use and a period without use. All data points are median values, and "empty" symbols indicate results below the analytical detection limit.

From the graphs in figure 6.9, it is apparent that outdoor chimney measurements are significantly higher for most of the compounds when the wood gasification boiler is in use than when not in use. Furthermore, the outdoor chimney measurements are comparable to the other

outdoor measurements when the gasification boiler is not in use. It is also clear, however, that there can be a significant difference between the West and East outdoor chimney positions (probably depending on the predominant wind direction during the sampling period).

For benzene, the outdoor chimney measurements are an order of 100 times the general outdoor level when the wood gasification boiler is in use. For toluene, xylenes and naphthalene, the outdoor chimney measurements are on the order of 10-100 times the general outdoor level. For C9/C10-aromatics and TVOC, the outdoor chimney measurements are an order of 1-10 times the general outdoor level. This effect is apparent even though the biomass burning in residence B is carried out in a modern wood gasification boiler, which should minimize the VOC emissions.

Also, there is a clear tendency for the local outdoor concentrations of benzene to be higher during use of the wood gasification boiler than when the boiler is not in use. Hence, the results indicate that local biomass burning can lead to higher local scale concentrations of benzene concentrations in outdoor air.

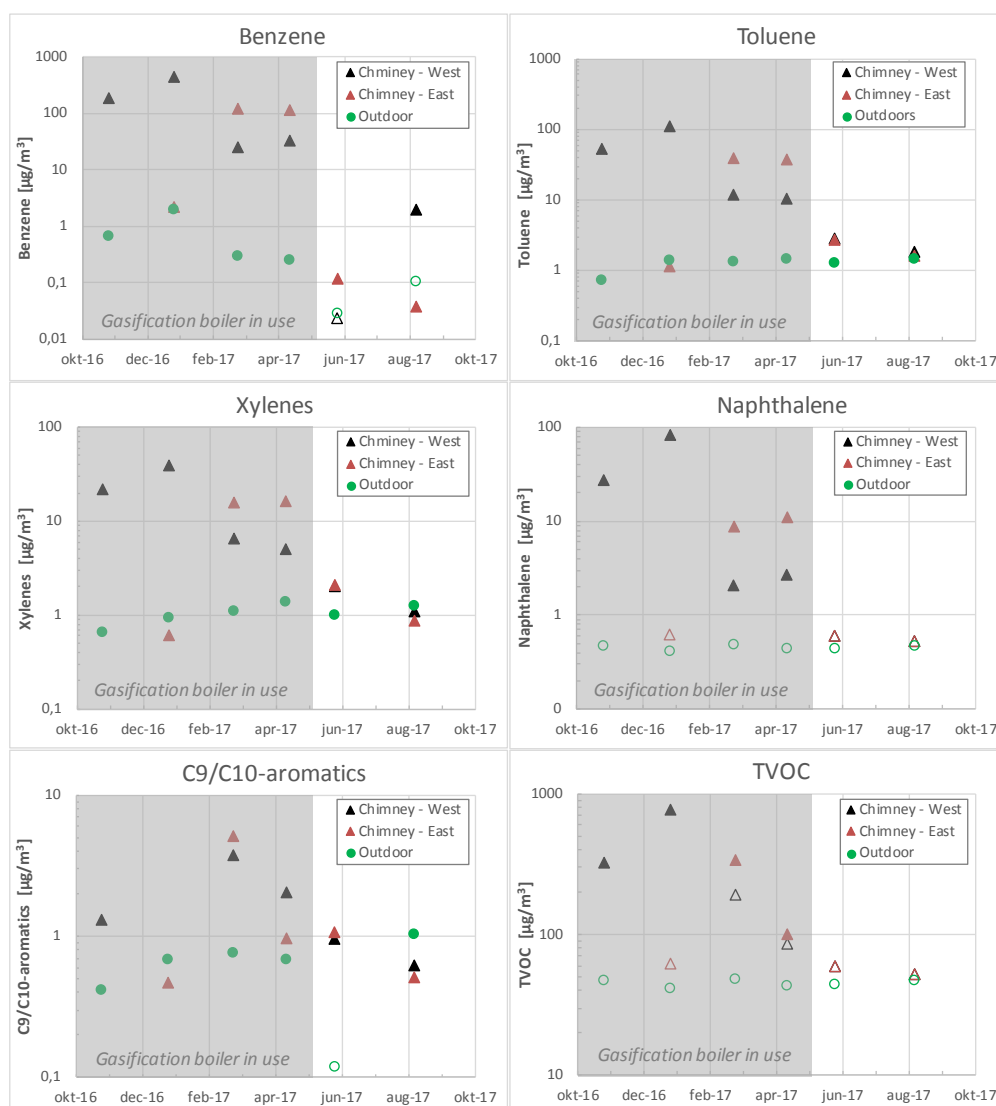


FIGURE 6.9. Median concentrations of VOCs in outdoor air at chimney sampling positions (west and east; triangles), and outdoor positions (circles) at residence B. Solid symbols indicate levels above the analytical detection limits, open symbols indicate levels below the analytical detection limit.

6.4 Coastal Study (outdoor benzene)

The results of the Coastal Study show very low levels of VOCs with no concentrations of toluene, naphthalene, C9/C10-aromatics or TVOC above the analytical detection limits. Xylenes (m-/p-xylene) is present in all the samples at a maximum concentration of $0,68 \mu\text{g}/\text{m}^3$ (i.e. well below the regulatory limit of $100 \mu\text{g}/\text{m}^3$).

Only benzene is detected in concentrations relevant for vapor intrusion studies, and the benzene results are presented in the figure below. One of the measurements in each coastal data series is suspected of being influenced by humidity causing desorption, and these measurements are not included in the median values calculated.

In the measurement period, the wind direction has been predominantly from the west (7 days), east (3 days) and south (6 days). Of the 6 days with winds from the south, 4 are from a more westerly direction (than easterly), and 1 day is from a more easterly direction.

As shown in figure 6.10, the median benzene level is $0,17 \mu\text{g}/\text{m}^3$ along the west coast and $0,23 \mu\text{g}/\text{m}^3$ along the east coast, and the benzene concentrations are generally higher on the east coast. If compared to the general background levels for November 2015, the west coast median corresponds to the minimum value in the data set, and the east coast median corresponds to less than the 5% percentile (cf. section 4.1). So, although the coastal measurements are indicative of the ubiquitous presence of benzene in outdoor air, the levels are also quite low.

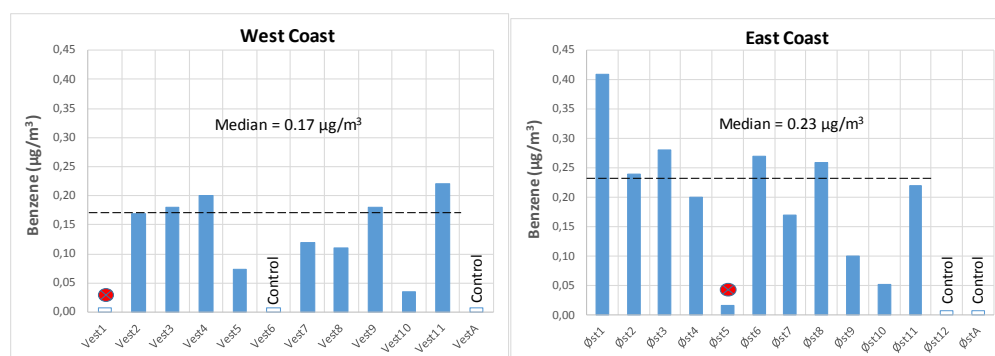


FIGURE 6.10. Benzene concentrations measured along the west and east coast of Denmark. Empty columns correspond to values below the analytical detection limit and red circles correspond to measurements that have not been included in the median levels. December 2016.

The slightly, but significantly, higher benzene level on the east coast corresponds to a predominant wind direction from the west and known land emissions e.g. biomass and traffic.

6.5 Wind Turbine Study (outdoor benzene)

Off-shore concentrations of VOCs were investigated on three different wind turbines at the Horns Rev 1 wind turbine park. The samplers were deployed at two different heights; at the base of the tower (close to the water at 9 m DVR) and at the top of the wind turbine (at 71 m DVR). The results for benzene are shown in the figure below. The measurement "Vest 2" (Blåvands Huk Lighthouse) from the coastal study is also illustrated, since this measurement point is the closest sampling point to the wind turbine park (and also represents the median benzene level on the west coast).

In the measurement period, the wind direction has been predominantly from the west (9 days), east (1 day) and south (5 days). Of the 5 days with winds from the south, 5 are from a more westerly direction (than easterly), and 1 day is from a more easterly direction.

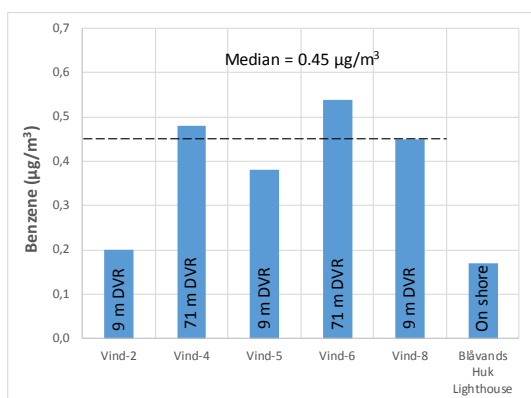


FIGURE 6.11. Benzene concentrations measured at the Horns Rev 1 off-shore wind turbine park, and at the Blåvands Huk Lighthouse (Vest 2 in the Coastal Study). January 2017.

With a median value of benzene of $0,45 \mu\text{g}/\text{m}^3$, (corresponding to the 30 percentile of the background levels of November 2015) at the wind turbines 14 km off the west coast of Jutland, compared to a value of $0,17 \mu\text{g}/\text{m}^3$ on the west coast, a possible benzene contribution from the oil fields in the North Sea, or perhaps the British Isles cannot be overlooked. There is, however, a slight tendency for higher benzene values at the 71 meter points than at the 9 meter points on the wind turbines. Also, low levels of toluene and xylenes are present in the high sampling positions and not in the low positions. This could be an indication of local emissions from the wind turbine.

6.6 Fir Study

Duplicate “potential” measurements of VOC emissions from fir branches were obtained by the procedure described in section 3.1.2. Since the requirements for free air movement around the samplers were not met during the deployment period, the absolute values cannot be used directly. The results are presented in the table below.

TABLE 6.1. Measurements of the potential for VOC emission from fir trees.

	Potential emission ($\mu\text{g}/\text{m}^3$)	
	Gran A	Gran B
Benzene	0,74	0,73
Toluene	310	310
Xylenes	21	21
C9/C10-aromatics	1.700	1.700
TVOC	190.000	210.000

As seen from the results, there is a potential for VOC emissions from fir trees (Christmas trees), although the benzene contribution is low compared with the potential contribution of C9/C10-aromatics and TVOC.

7. Summary and conclusions

The present study was initiated to address a need for well-documented Danish background/ambient levels of hydrocarbon VOCs in outdoor and indoor air in residences, that can be used for vapor intrusion studies. Besides providing well-documented background levels, aspects concerning seasonal variation, measurement variability, spatial variability in outdoor measurements and the sources of hydrocarbon VOC background/ambient levels in indoor and outdoor air have been investigated.

7.1 General hydrocarbon VOC background levels

Well-documented hydrocarbon background levels for benzene, toluene, xylenes, naphthalene, C9/C10-aromatics and total volatile organic carbon (TVOC), in both outdoor and indoor air, have been provided. The results are based on two measurement campaigns in November 2015 (autumn) and May 2016 (spring) at respectively 131 and 142 uncontaminated Danish residential buildings. The measurements are based on passive sampling with Dräger ORSA 5 diffusive samplers with an approx. 14-day sampling deployment.

Median background levels – for each combination of compound, measurement campaign and outdoor/indoor-position – are presented in table 7.1, together with the Danish regulatory limits associated with vapor intrusion from soil and groundwater contamination /23/. The EU limit value for benzene in ambient (outdoor) air, based on /32/, is $5 \mu\text{g}/\text{m}^3$ /8/.

TABLE 7.1. Danish regulatory limits for VOC contribution to residential buildings from contaminated soil and groundwater, and median background levels of hydrocarbon VOCs in uncontaminated Danish residences ($\mu\text{g}/\text{m}^3$).

Compound	Regulatory limit	November 2015		May 2016	
		Outdoor	Indoor	Outdoor	Indoor
Benzene	0,13	0,50	0,66	0,28	0,41
Toluene	400	1,1	4,5	1,0	3,0
Xylenes*	100	1,0	2,9	0,81	2,0
Naphthalene	40	<DL	<DL	<DL	<DL
C9/C10-aromatics	30	0,45	3,3	0,41	2,3
TVOC (C6-C35)	100	<DL	<DL	<DL	<DL

* = sum of *o*-xylene, *m*-xylene, *p*-xylene and ethylbenzene.

<DL = Below the analytical detection limit.

Bold = Median levels above the regulatory limit.

The percentages of samples above the regulatory limit – for each combination of compound, measurement campaign and outdoor/indoor-position – are presented in table 7.2.

TABLE 7.2. Percentage of samples above the Danish regulatory limit (RL).

Compound	November 2015		May 2016	
	Outdoor Above RL	Indoor Above RL	Outdoor Above RL	Indoor Above RL
Benzene	100%	100%	99%	99%
Toluene	0%	0%	0%	0%
Xylenes*	0%	1,5%	0%	0%
Naphthalene	0%	0%	0%	0%
C9/C10-aromatics	0%	1,3%	0%	0%
TVOC (C6-C35)	0,8%	29%	0%	33%

* = sum of *o*-xylene, *m*-xylene, *p*-xylene and ethylbenzene.

From tables 7.1 and 7.2, it is apparent that the compounds of general interest in relation to vapor intrusion in the Danish regulatory setting are benzene and TVOC.

Based on this study, it is concluded that both outdoor and indoor background concentrations of benzene exceed the Danish regulatory limit for vapor intrusion contribution to residential buildings from contaminated soil and groundwater ($0,13 \mu\text{g}/\text{m}^3$) in >99% of uncontaminated Danish residences. Based on the results, the average expectation for the background indoor concentration of benzene can be expected to be $0,66 \mu\text{g}/\text{m}^3$ in November and $0,41 \mu\text{g}/\text{m}^3$ in May. For benzene in outdoor air the average background concentration of benzene can be expected to be $0,50 \mu\text{g}/\text{m}^3$ in November and $0,28 \mu\text{g}/\text{m}^3$ in May.

The indoor background concentration of TVOC can be expected to exceed the Danish regulatory limit of $100 \mu\text{g}/\text{m}^3$ in about 30% of uncontaminated Danish residences, but the general background concentration of TVOC in outdoor air cannot be expected to exceed the regulatory limit.

7.2 Indoor internal contribution

The indoor internal contribution of VOCs, at a given residence, can be estimated by subtracting the outdoor level from the indoor level. This allows the background level in indoor air to be divided into a contribution from outdoor air and a contribution from internal background sources in ordinary Danish homes, which can be presented as a percentage. If the indoor concentration is \leq the outdoor concentration the indoor internal contribution is ≤ 0 (zero). Summary statistics for indoor internal contributions in uncontaminated Danish residences are shown in table 7.3.

TABLE 7.3. Statistical summary for indoor internal contribution, November 2015 and May 2016. Median levels ($\mu\text{g}/\text{m}^3$), median % of indoor concentration, and % of residences with an indoor internal contribution \leq zero.

Compound	November 2015			May 2016		
	Median	\leq zero	\leq zero	Median	\leq zero	\leq zero
Benzene	$0,16 \mu\text{g}/\text{m}^3$	24%	23%	$0,10 \mu\text{g}/\text{m}^3$	24%	19%
Toluene	$3,4 \mu\text{g}/\text{m}^3$	76%	5%	$1,8 \mu\text{g}/\text{m}^3$	60%	10%
Xylenes*	$1,5 \mu\text{g}/\text{m}^3$	52%	11%	$1,0 \mu\text{g}/\text{m}^3$	50%	14%
C9/C10-aromatics	$2,9 \mu\text{g}/\text{m}^3$	88%	3%	$1,6 \mu\text{g}/\text{m}^3$	75%	4%
TVOC (C6-C35)	<DL	-	-	<DL	-	-

* = sum of o-xylene, m-xylene, p-xylene and ethylbenzene.

<DL = Median indoor concentration less than the detection level.

From the table, it is seen that the median for indoor internal contribution of benzene is an order of $0,10$ - $0,16 \mu\text{g}/\text{m}^3$, as compared with the background levels in table 7.1 of $0,41$ and $0,66 \mu\text{g}/\text{m}^3$. In other words, the background levels of benzene in homes comprises a 24% contribution from sources within the home and a 76 % contribution from sources in the outdoor air. The indoor internal contribution of benzene is below zero in 19-23% (about 20%) of uncontaminated Danish residences, meaning that the outdoor concentration is higher than the indoor concentration.

For a given indoor concentration of benzene in uncontaminated residences an average of 1/4 stems from indoor internal sources and 3/4 from outdoor air. In approximately 1/5 of the residences there is no indoor internal contribution of benzene. For toluene, xylenes and C9/C10-aromatics the indoor internal contribution to the total concentration is significantly higher (50-88 %).

7.3 Seasonal variation

From table 7.1 and 7.3 it is apparent that there are differences in the median concentrations for the various VOCs between the November 2015 and May 2016 measurement campaigns. These differences indicate a seasonal variation in the background levels.

Seasonal variations in the distribution functions of the individual compounds in the outdoor and indoor air samples in the November 2015 and May 2016 measuring campaigns are explored by performing statistical tests at the 5% significance level ($\alpha = 0,05$), cf. appendix 4. The results are summarized in table 7.4.

TABLE 7.4. Summary of seasonal variation between November 2015 (autumn) and May 2016 (spring) data. **Bold** = statistically significant, **grey** = not statistically significant at $\alpha = 0,05$.

Compound	Outdoor			Indoor		
	Seasonal variation		p-value	Seasonal variation		p-value
Benzene	Yes	High in autumn	<0,001	Yes	High in autumn	<0,001
Toluene	No	-	0,10	Yes	High in autumn	<0,001
Xylenes*	Yes	High in autumn	0,0028	Yes	High in autumn	0,0043
C9/C10-aromatics	Yes	High in autumn	0,029	Yes	High in autumn	0,0032
TVOC (C6-C35)	No	-	0,12	No	-	0,26

* = sum of *o*-xylene, *m*-xylene, *p*-xylene and ethylbenzene.

As seen in the table, there is a significant seasonal variation in the outdoor levels of benzene, xylenes and C9/C10-aromatics, and a significant seasonal variation in the indoor levels of benzene, toluene, xylenes and C9/C10-aromatics.

Detailed studies of benzene at three residences (section 6.1.1 and 6.2) support the conclusion that there is a significant seasonal variability in both outdoor and indoor benzene levels. As seen in figure 7.1, both outdoor and indoor benzene levels are relatively high in the autumn and winter months and relatively low in the spring and summer months.

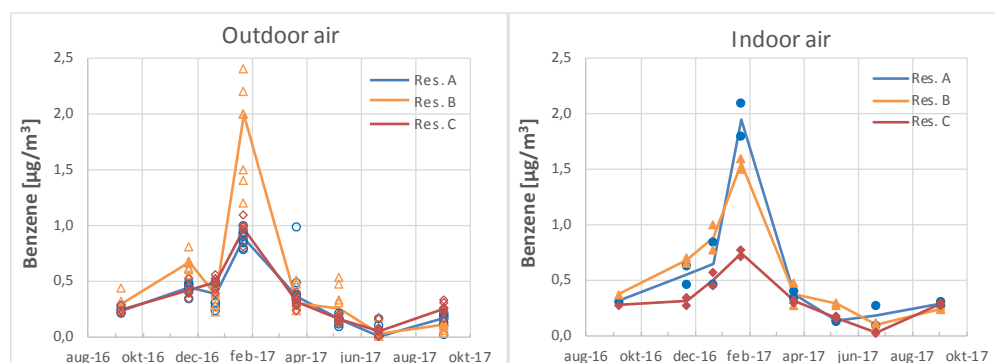


FIGURE 7.1. Seasonal variability in outdoor (left) and indoor (right) benzene concentrations at three residences. Symbols show individual measurements and lines show median levels.

The measurements show clear seasonal variability in the benzene concentrations. The detailed study in three residences indicate that the median concentrations for benzene in both outdoor and indoor air can be significantly higher in January-February than in November.

7.4 Factors of consequence

The secondary objectives of this study were to determine if other factors such as population density, tobacco smoking, wood-burning stoves, ventilation habits/methods, house renovation, etc., could influence background levels. Hence, a questionnaire was filled out at each residence, and metadata was paired with the compounds of special interest in a vapor intrusion context; i.e. levels of benzene and TVOC in outdoor and/or indoor air.

The results are presented in detail in chapter 5, and the qualitative conclusions are summarized in table 7.5.

TABLE 7.5. Summary of conclusions regarding consequential factors in relation to the background levels of benzene and TVOC. The results are divided into qualitative categories of: **High**, Medium, Low or No effect.

	Outdoor	Indoor		Section
	Benzene	Benzene	TVOC	
Population density	Low	-	-	5.1
Indoor tobacco smoking	-	High	Low	5.2
Wood-burning stoves	-	Medium	Low	5.3
Frequency of airing in the house	-	No	No	5.4
Home improvements/ renovation	-	Low	Low	5.5
Occupancy	-	No	No	5.6
Building age	-	Low	Low	5.7
Street type/traffic density	No	-	-	5.8.1
Distance to traffic lights	Low	-	-	5.8.2
Distance to gas stations	No	-	-	5.8.3

A few of the more pronounced results are highlighted in the following sections.

7.4.1 Population density

The study of the effect of population density as related to city, towns, villages or rural countryside (cf. section 5.1) revealed some surprising results regarding the outdoor benzene background levels. Based on the literature review in chapter 2, it was expected that the benzene levels in cities would be significantly higher than in towns, villages and in the countryside.

These expectations were met to a lesser degree in the May 2016 (spring) measurement campaign, but the tendency was reversed in the November 2015 (autumn) measurement campaign. In the November 2015 measurement campaign, the outdoor benzene concentrations were significantly higher in both villages and the rural countryside than in the towns and cities.

A more detailed investigation of the results revealed that the seasonal variability in the outdoor concentration of benzene was much more pronounced in villages and in the countryside than in the cities. The results indicate that the outdoor benzene level in cities is relatively more affected by traffic (a constant emission source), while the levels in villages and the countryside, especially in the heating season (autumn), are relatively more affected by domestic heating by biomass burning (a variable emission source).

In general, the differences in the outdoor concentrations of benzene between city/town and village/countryside, for both the May and November measurement campaigns were small, and the trend between high and low benzene concentration for the different population densities was opposite in the two campaigns. In general, the outdoor benzene concentration is expected to be independent of the population density.

7.4.2 Indoor smoking

Indoor smoking is the single most influential factor investigated, as far as the indoor benzene concentration is concerned. The results indicate an approx. four-fold increase in the indoor benzene level in the homes of smokers who smoke indoors as compared with general background level; 2,7 $\mu\text{g}/\text{m}^3$ compared with 0,66 $\mu\text{g}/\text{m}^3$ in November 2015, and 1,6 $\mu\text{g}/\text{m}^3$ compared with 0,41 $\mu\text{g}/\text{m}^3$ in May 2016 (spring). Very high levels of benzene are also related to indoor smoking.

For TVOC, the results indicate higher concentrations in residences with indoor smoking than in residences in general, with a median value approx. 2,5 times higher for residences with indoor smoking than residences in general. Very high levels of TVOC are related to other factors than smoking.

7.4.3 Wood-burning stoves and residential biomass burning

Higher indoor benzene concentrations were observed in residences with wood-burning stoves in both the November 2015 (autumn) and May 2016 (spring) measurement campaigns. The results indicate that the median indoor benzene concentration is approx. 58 and 84% higher in residences with wood-burning stoves than in residences without; 0,63 $\mu\text{g}/\text{m}^3$ compared to 0,40 $\mu\text{g}/\text{m}^3$ in November 2015 and 0,83 $\mu\text{g}/\text{m}^3$ compared to 0,45 $\mu\text{g}/\text{m}^3$ in May 2016.

A tendency toward higher indoor TVOC concentrations (at the high end of the distribution) was observed in residences with wood-burning stoves in the November 2015 (autumn) measurement campaign. This tendency was not observed in the May 2016 (spring) measurement campaign. The results indicate that wood-burning stoves can be associated with an increased indoor internal contribution of TVOC in the heating season in some homes.

The emissions of VOCs from the chimney of a wood gasification boiler was investigated in section 6.3. In this sub-study, it was documented that biomass burning is associated with local-scale emissions of VOCs. It was also documented that the local outdoor concentration of benzene is higher in the heating season than when the wood-gasification boiler was not in use. Hence, it can be expected that local biomass burning leads to higher local outdoor concentrations of benzene.

7.5 Outdoor sources

The results in table 7.3 indicate that about 50-80% of the indoor concentration of toluene, xylenes and C9/C10-aromatics is due to indoor sources, while only approx. 25% of indoor benzene concentration is due to indoor internal sources and 75% to outdoor sources.

To investigate a hypothesis that a significant part of the outdoor benzene is due to biomass burning (wood-burning stoves, residential biomass furnaces and local biomass powered district heating plants), detailed studies of VOC emissions from biomass burning at the residential scale were investigated at one residence by sampling around the chimney from a wood gasification boiler (section 6.3).

From this study, it is clear that biomass burning affects the local outdoor VOC concentrations during the heating season. For benzene, the chimney measurements are an order of 100 times the general outdoor level. For toluene, xylenes and naphthalene, the chimney measurements are an order of 10-100 times the general outdoor level. For C9/C10-aromatics and TVOC, the chimney measurements are an order of 1-10 times the general outdoor level.

Living plants have also been investigated as a source of hydrocarbon VOCs (section 6.6), and Fir trees (Christmas trees) are identified as a potential contribution source of VOCs to the indoor air (except benzene).

Sampling at 21 positions along the east and west coast of Jutland (December 2016) with a predominant wind direction from the west (11 days, vs. 4 days from the east) yielded median benzene level of 0,17 $\mu\text{g}/\text{m}^3$ on the west coast and 0,23 $\mu\text{g}/\text{m}^3$ on the east coast (section 6.4). The results underline the ubiquitous presence of benzene in outdoor air at levels above the regulatory limit (0,13 $\mu\text{g}/\text{m}^3$), but also support the fact that there are many land based sources of benzene emission, e.g. traffic and biomass burning.

Sampling at the off-shore wind turbine park Horns Rev 1 (section 6.5) 14 km from the shoreline showed benzene concentrations of 0,20-0,54 $\mu\text{g}/\text{m}^3$, indicating that there might be a source of benzene, perhaps related to the oil fields in the North Sea or the British Isles, that give rise to benzene background concentrations in Denmark above the regulatory limit.

7.6 Inter-duplicate and inter-position variability

In the study of variability (cf. section 6.1.2 and 6.1.3) four outdoor and one indoor measurement positions have been sampled in duplicate in eight measurement campaigns over one year of sampling. Inter-duplicate and inter-position variability in benzene and TVOC concentrations were explored for this data set.

The results for benzene, show that the inter-duplicate variability is <10-20% for 50% of the measurements, and less than 100% for 90% of measurements. For approx. 4% of vapor intrusion relevant benzene concentrations ($\geq 0,13 \mu\text{g}/\text{m}^3$), inter-duplicate variability is more than 100%. Hence, for half of the benzene measurements the difference between results for two samplers in the same position is less than 10-20%, but for 10% of the measurements the difference is a factor of two or more.

The results for TVOC, show that the inter-duplicate variability is <10% for 50% of the samples, and less than 100% for 90% of duplicate samples. Hence, for half of the TVOC measurements the difference between results for two samplers in the same position is less than 10%, but for 10% of the measurements the difference is a factor of two or more.

The results for benzene, show that the outdoor inter-position variability is <20% for 50% of the positions, and less than 100% for 92% of duplicate positions. Hence, for half of the benzene measurements the difference between results for two samplers in different positions around the residence is less than 10%, but for 8% of the measurements the difference is a factor of two or more.

7.7 Conclusions related to vapor intrusion studies

7.7.1 Overall conclusions

Overall, the background levels (2015-2016) documented in this study, and based on a 14-day passive sampling on ORSA-tubes, are comparable to the previous levels derived in 2000-2008 when different measurement methods were generally applied /1/. While the new background levels for BTEX and C9/C10-aromatics are only slightly lower than the previous levels, the new background levels of TVOC are significantly lower.

The background benzene level can be expected to exceed the Danish regulatory limit of 0,13 $\mu\text{g}/\text{m}^3$ in both outdoor and indoor air at uncontaminated Danish residences. The results show that >99% of both outdoor and indoor benzene measurements in Danish uncontaminated residences are above the regulatory limit (100% in November 2015 and 98,9% in May 2016). Hence, the Danish regulatory limit can be expected to be exceeded everywhere in Denmark, even in the most rural areas along the Danish coast line.

Indoor background levels of TVOC can be expected to exceed the Danish regulatory limit of 100 $\mu\text{g}/\text{m}^3$ in about 30% of uncontaminated Danish residences (29% in November 2015 and 33% in May 2016), while the TVOC background concentration is below the analytical detection limit (of approx. 43 $\mu\text{g}/\text{m}^3$) in approx. 50% of the uncontaminated Danish residences, irrespective of the measurement campaign.

Concentrations of toluene, sum of xylenes (including ethylbenzene), naphthalene and C9/C10-aromatics are not expected to exceed the Danish regulatory limits at uncontaminated resi-

dences, neither in outdoor nor indoor air. For these compounds, only very few measurements (<2%) were above the regulatory limit values.

Based on the new background levels derived in this study, significant background level interference is not expected for vapor intrusion studies involving indoor measurements of toluene, xylenes (including ethylbenzene), naphthalene and C9/C10-aromatics. For benzene, the indoor background concentration can be expected to be above the regulatory limit in >99% of uncontaminated Danish Residences, and for TVOC, about 30% of uncontaminated residences will have background levels exceeding the regulatory limit.

This study has documented that outdoor sources are responsible for approx. 75% of the background concentrations of benzene in indoor air in uncontaminated residences.

7.7.2 Seasonal variation

Based on two measurement campaigns (November 2015 and May 2016) and the variability study (section 6.1 and 6.2), a significant seasonal variation in the outdoor levels of benzene, xylenes and C9/C10-aromatics has been identified. The highest levels are measured in the autumn and winter, and the lowest in spring and summer.

However, only ambient benzene levels are associated with significant seasonal variations with the potential to interfere with vapor intrusion studies using indoor measurements.

7.7.3 Significant local factors

Indoor smoking dramatically increases the indoor concentration of benzene, and the background benzene level in residences with indoor smoking can be expected to increase four-fold in comparison with the overall median background level. Indoor smoking also influences the indoor TVOC concentration, and the median level in residences with indoor smoking can be expected to increase to approx. 2,5 times the general background level.

Residences with wood-burning stoves are expected to be associated with approx. 60-80% higher indoor concentrations of benzene than residences without wood-burning stoves, irrespective of the season. In some residences with a wood-burnings stove, higher indoor TVOC concentrations can be expected in the heating season. Higher local outdoor benzene concentrations can be expected in residences heated by residential biomass burning.

Effects of population density, ventilation practices, home improvements/renovation/-remodeling, occupancy, building age and local traffic related emissions are only expected to influence background levels to a lesser degree – or have no effect at all.

7.7.4 Use of outdoor reference measurements

In vapor intrusion studies, the effects of seasonal variations, local biomass burning and other local factors of consequence, the outdoor variations should be accounted for by always including outdoor reference measurements.

For half of the benzene and TVOC measurements, the difference between two samplers in the same position (benzene and TVOC), or measurements in two different outdoor positions (benzene), is less than 10-20%, but for approx. 10% of the measurements the difference is a factor of 2 or more. It is noted that 20% variation/uncertainty on the median outdoor benzene concentration in November 2015 corresponds approx. to the Danish regulatory limit for benzene (20% of 0,50 $\mu\text{g}/\text{m}^3$).

Based on the results, it is recommended that the outdoor VOC level is measured in at least 2 positions, since the outdoor level in many (for benzene almost all) instances will be of great importance in the interpretation of the subsurface contaminant contribution to the indoor air.

8. Sammenfatning og konklusioner (dansk)

Dette studie er igangsat med henblik på at fremskaffe veldokumenterede danske baggrunds-niveauer for flygtige kulbrinter i udeluft og indeklima i boliger, som kan bruges i sammenhæng med indeklimaundersøgelser, relateret til forurenede jord og grundvand. Sideløbende hermed er en række aspekter undersøgt, herunder sæsonvariation i baggrunds-niveauerne, variation på dobbeltbestemmelser og på flere udeluftreferencer, samt forskellige kilder til baggrunds-niveauer i udeluft og indeklima.

8.1 Generelle baggrunds-niveauer for flygtige kulbrinter

Via studiet er der frembragt nye veldokumenterede baggrunds-niveauer for benzen, toluen, xylener, naphthalen, C9/C10-aromater og totalkulbrinter (TVOC), både i udeluft og indeklima. Resultaterne er baseret på to målerunder, november 2015 og maj 2016, foretaget i og omkring hhv. 131 og 142 uforurenede danske boliger. Alle målinger er baseret på passiv prøveopsamling med Dräger-5 ORSA-rør over ca. 14 dages perioder.

Median baggrunds-niveauer, for hver kombination af stof, prøverunde og udeluft/indeklima-position, er opsummeret i tabel 8.1, sammen med de danske afdampningskriterier /23/. EU's grænseværdi for benzen i udeluft, baseret på /32/, er $5 \mu\text{g}/\text{m}^3$, /8/.

TABEL 8.1. Danske afdampningskriterier og median baggrunds-niveauer for flygtige kulbrinter i uforurenede danske boliger ($\mu\text{g}/\text{m}^3$).

Stof	Afdampnings-kriterium	November 2015		Maj 2016	
		Udeluft	Indeklima	Udeluft	Indeklima
Benzen	0,13	0,50	0,66	0,28	0,41
Toluen	400	1,1	4,5	1,0	3,0
Xylener*	100	1,0	2,9	0,81	2,0
Naphthalen	40	<DG	<DG	<DG	<DG
C9/C10-aromater	30	0,45	3,3	0,41	2,3
TVOC (C6-C35)	100	<DG	<DG	<DG	<DG

* = sum af o-xylen, m-xylen, p-xylen og ethylbenzen.

<DG = Under detektionsgrænsen.

Fed = Medianniveauer over afdampningskriterierne.

Procentdelen af prøver over afdampningskriterierne, for hver kombination af stof, prøverunde og udeluft/indeklima-position er opsummeret i tabel 8.2.

TABEL 8.2. Procentdel af prøver over afdampningskriteriet (ADK).

Stof	November 2015		Maj 2016	
	Udeluft >ADK	Indeklima >ADK	Udeluft >ADK	Indeklima >ADK
Benzen	100%	100%	99%	99%
Toluen	0%	0%	0%	0%
Xylener*	0%	1,5%	0%	0%
Naphthalen	0%	0%	0%	0%
C9/C10-aromater	0%	1,3%	0%	0%
TVOC (C6-C35)	0,8%	29%	0%	33%

* = sum af o-xylen, m-xylen, p-xylen og ethylbenzen.

Af tabel 8.1 og 8.2 ses det, at de komponenter, der er af speciel interesse ift. danske indeklimaundersøgelser, relateret til forurenede jord og grundvand, er benzen og TVOC.

Baseret på nærværende studium kan det konstateres, at baggrundsniveauet for benzen i både udeluft og indeklima overstiger afdampningskriteriet på $0,13 \mu\text{g}/\text{m}^3$ i >99% af de undersøgte uforurenede danske boliger. Baseret på dette studium kan der i gennemsnit forventes et baggrundsniveau for benzen i indeklima på $0,66 \mu\text{g}/\text{m}^3$ i november og $0,41 \mu\text{g}/\text{m}^3$ i maj. For benzen i udeluft er baggrundsniveauerne tilsvarende $0,50 \mu\text{g}/\text{m}^3$ i november og $0,28 \mu\text{g}/\text{m}^3$ i maj.

Baggrundsniveauet i indeklima for TVOC kan forventes at overstige afdampningskriteriet på $100 \mu\text{g}/\text{m}^3$ i ca. 30% af uforurenede danske boliger, men det generelle baggrundsniveau i udeluft ikke kan forventes at overstige afdampningskriteriet.

8.2 Bidrag fra interne kilder i indeklimaet

Det interne bidrag af flygtige kulbrinter i indeklimaet, i en given bolig, kan estimeres ved at trække udeluftkoncentrationen fra indeklimakoncentrationen. Dette muliggør at indeklimakoncentrationen (baggrundsniveauet) kan opdeles i et bidrag fra udeluften og et bidrag fra interne kilder i indeklimaet, i uforurenede danske boliger. Det interne bidrag kan udtrykkes som en procentdel af den samlede indeklimakoncentration. Hvis indeklimakoncentrationen er \leq udeluftkoncentrationen, vil det interne bidrag være ≤ 0 (nul). Opsummerende statistik for de interne bidrag til indeklimaet i uforurenede danske boliger er vist i tabel 8.3.

TABEL 8.3. Sammenstilling af interne bidrag, november 2015 og maj 2016. Median niveauer ($\mu\text{g}/\text{m}^3$), median % af indeklimakoncentration, og % af boliger med et internt bidrag \leq nul.

	November 2015			Maj 2016		
	Median		\leq nul	Median		\leq nul
Benzen	$0,16 \mu\text{g}/\text{m}^3$	24%	23%	$0,10 \mu\text{g}/\text{m}^3$	24%	19%
Toluen	$3,4 \mu\text{g}/\text{m}^3$	76%	5%	$1,8 \mu\text{g}/\text{m}^3$	60%	10%
Xylener*	$1,5 \mu\text{g}/\text{m}^3$	52%	11%	$1,0 \mu\text{g}/\text{m}^3$	50%	14%
C9/C10-aromater	$2,9 \mu\text{g}/\text{m}^3$	88%	3%	$1,6 \mu\text{g}/\text{m}^3$	75%	4%
TVOC (C6-C35)	<DG	-	-	<DG	-	-

* = sum af o-xylen, m-xylen, p-xylen og ethylbenzen.

<DG = Median af internt bidrag under detektionsgrænsen.

Af tabellen ses det, at medianen for det interne bidrag af benzen er i størrelsesordenen $0,10$ - $0,16 \mu\text{g}/\text{m}^3$. Til sammenligning er baggrundsniveauerne i tabel 8.1 på $0,41$ og $0,66 \mu\text{g}/\text{m}^3$. Dvs. at det interne bidrag af benzen til indeklimaet i uforurenede boliger kun udgør 24%, mens 76% af indeklimakoncentrationen skyldes bidrag fra udeluften. Det interne bidrag af benzen er mindre end nul i 19-23% (ca. 20%) af uforurenede danske boliger. I disse tilfælde er udeluftkoncentrationen altså højere end koncentrationen i indeklimaet.

For en given indeklimakoncentration af benzen i uforurenede boliger kommer i gennemsnit ca. 1/4 fra interne kilder og ca. 3/4 fra udeluften. I ca. 1/5 boligerne er der intet internt bidrag for benzen. For toluen, xylener og C9/C10-aromater er det interne bidrags andel af den totale koncentration betydelig højere (50-88%).

8.3 Sæsonvariationer

I tabel 8.1 og 8.3 ses der forskelle i medianværdierne for de forskellige flygtige kulbrinter hvis resultaterne for november 2015 og maj 2016 målerunderne sammenlignes. Disse forskelle indikerer, at der er en sæsonvariation i baggrundsniveauerne.

Sæsonvariation i fordelingsfunktionerne for de forskellige komponenter/stofgrupper i udeluft og indeklima fra november 2015 og maj 2016 målerunderne er undersøgt ved at gennemføre statistiske tests på 5% signifikansniveau ($\alpha = 0,05$), jf. appendix 4. Resultaterne er opsummeret i tabel 8.4.

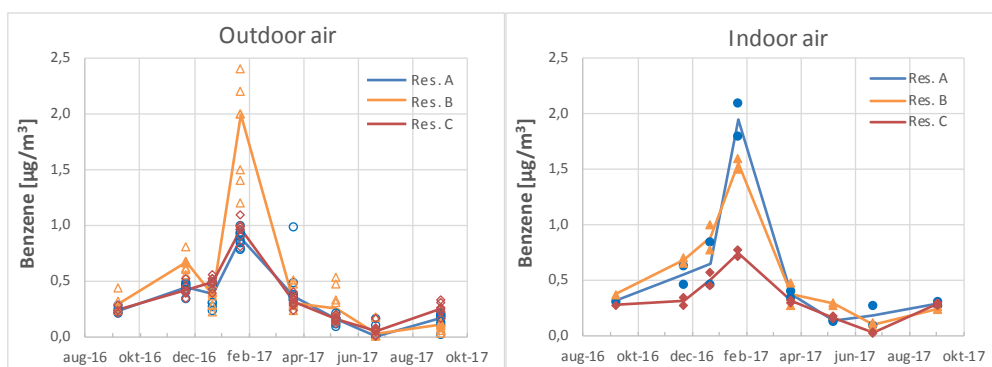
TABEL 8.4. Opsummering af statistiske test for sæsonvariation imellem data fra målerunderne i november 2015 (efterår) og maj 2016 (forår). **Fed Ja** = statistisk signifikant, grå Nej = ikke statistisk signifikant ved $\alpha = 0,05$.

Stof	Udeluft			Indeklima		
	Sæsonvariation		p-værdi	Sæsonvariation		p-værdi
Benzen	Ja	Høj om efteråret	<0,001	Ja	Høj om efteråret	<0,001
Toluen	Nej	-	0,10	Ja	Høj om efteråret	<0,001
Xylener*	Ja	Høj om efteråret	0,0028	Ja	Høj om efteråret	0,0043
C9/C10-aromater	Ja	Høj om efteråret	0,029	Ja	Høj om efteråret	0,0032
TVOC (C6-C35)	Nej	-	0,12	Nej	-	0,26

* = sum af o-xylen, m-xylen, p-xylen og ethylbenzen.

Som det ses i tabel 8.4, så er der tale om signifikante sæsonvariationer i udeluftkoncentrationen af benzen, xylener og C9/C10-aromater, samt signifikante sæsonvariationer i indeklimatekoncentrationerne af benzen, toluen, xylener og C9/C10-aromater.

Detaljerede studier i tre boliger (jf. afsnit 6.1.1 og 6.2) understøtter en konklusion om at der er betydelige sæsonvariationer i benzenniveauet i både udeluften og indeklimaet. Som det ses i figur 8.1 er både udeluft- og indeklimateniveauet for benzen relativt højt i efterårs- og vintermånedene og relativt lave i forårs- og sommermånedene.



FIGUR 8.1. Sæsonvariation i benzenkoncentrationerne i udeluft (venstre) og indeklima (højre) for tre boliger. Symbolerne viser enkeltmålinger og linjerne viser medianniveaue.

De udførte målinger viser betydelige sæsonvariationer for benzen. Detailstudiet i tre boliger tyder på, at medianværdierne for benzen i indeklima og udeluft kan være betydelig højere i januar-februar end i november.

8.4 Betydende faktorer

Sekundære formål med projektet var at undersøge om andre faktorer, f.eks. befolkningstæthed/bystørrelse, tobaksrygning, brændeovne, ventilationsvaner/-metoder, renovering m.m. har indflydelse på de forventede baggrunds niveauer. Derfor blev et spørgeskema udfyldt for hver bolig i undersøgelsen, og metadata om forskellige forhold blev sammenholdt med indeklimatekoncentrationerne af de stoffer/stofgrupper, der har speciel interesse i indeklimatestudier, relateret til forurenede jord og grundvand; dvs. benzen og TVOC i udeluft og/eller indeklima.

Resultaterne er præsenteret i detaljer i kapitel 5, og de kvalitative resultater er opsummeret i tabel 8.5.

TABEL 8.5. Opsummering af konklusioner vedr. betydende faktorer ift. baggrunds niveauer for benzen og TVOC. Resultaterne er inddelt i de kvalitative kategorier: **Stor**, Medium, Lav eller Ingen betydning.

	Udeluft	Indeklima		Rapport- afsnit
	Benzen	Benzen	TVOC	
Befolkningstæthed/bystørrelse	Lav	-	-	5.1
Indendørs tobaksrygning	-	Stor	Lav	5.2
Brændeovn	-	Medium	Lav	5.3
Udluftningsvaner	-	Ingen	Ingen	5.4
Renovering	-	Lav	Lav	5.5
Antal beboere	-	Ingen	Ingen	5.6
Bygningsalder	-	Lav	Lav	5.7
Veitype/trafiktæthed	Ingen	-	-	5.8.1
Afstand til trafiklys	Lav	-	-	5.8.2
Afstand til tankstation	Ingen	-	-	5.8.3

Nogle af de mere markante resultater er trukket op i det følgende.

8.4.1 Befolkningstæthed/bystørrelse (udeluft)

Undersøgelsen af befolkningstætheden, relateret til kategorierne storby, mindre by, landsby eller på landet (jf. afsnit 5.1) afslørede nogle overraskende resultater mht. baggrunds niveauerne for benzen i udeluften. Baseret på litteraturstudiet i kapitel 2 var det forventet, at benzen-niveauet var væsentligt højere i store og mindre byer end i landsbyer og på landet.

Disse forventninger blev i mindre grad opfyldt i maj 2016 målerunden (forår), mens tendensen var omvendt i november 2015 målerunden (efterår). I november 2015 målerunden var benzenkoncentrationen signifikant højere både i landsbyer og på landet end i storbyer og mindre byer.

En mere detaljeret undersøgelse af resultaterne viste at sæsonvariationen i udeluftkoncentrationen af benzen var væsentligt mere udtalt i landsbyer og på landet end i storbyerne. Resultaterne indikerer, at udeluftkoncentrationen af benzen i storbyer er relativt mere påvirket af trafik (en konstant kilde), mens udeluftkoncentrationen af benzen i landsbyer og på landet, specielt i fyringssæsonen (efteråret), er relativt mere påvirket af emissioner fra boligopvarmning og biomasseforbrænding (en variabel kilde).

Generelt var der kun mindre forskelle i udeluftkoncentrationerne for benzen i storby/by kontra landsby/land for både maj og november målingerne, og forskellen var modsat rettet i de 2 målerunder. Generelt må forventes samme baggrunds niveau for benzen i udeluft uafhængig af befolkningstæthed.

8.4.2 Indendørs tobaksrygning

Indendørs tobaksrygning er den mest betydende faktor undersøgt i projektet mht. indeklima-koncentrationen af benzen. Resultaterne indikerer ca. 4 gange højere median indeklimakoncentration af benzen i boliger med indendørs rygning end i boliger generelt; $2,7 \mu\text{g}/\text{m}^3$ sammenlignet med $0,66 \mu\text{g}/\text{m}^3$ i november 2015, og $1,6 \mu\text{g}/\text{m}^3$ sammenlignet med $0,41 \mu\text{g}/\text{m}^3$ i maj 2016. Meget høje benzenkoncentrationer er også relateret til indendørs rygning.

For TVOC er der forhøjede koncentrationer i rygerboliger end i andre boliger, med en median-værdi der er ca. 2,5 gange højere i rygerboliger end i datamaterialet som helhed. De højeste koncentrationer af TVOC skyldes andre faktorer end indendørs rygning.

8.4.3 Brændeovn og fastbrændselsfy

I hjem med brændeovn blev der observeret højere indeklimakoncentrationer af benzen både i november 2015 målerunden (efterår) og i maj 2016 målerunden (forår). Resultaterne indikerer ca. 58 og 84% højere median indeklimakoncentration af benzen i boliger med brændeovn end

i boliger uden brændeovn; $0,63 \mu\text{g}/\text{m}^3$ sammenlignet med $0,40 \mu\text{g}/\text{m}^3$ i november 2015, og $0,83 \mu\text{g}/\text{m}^3$ sammenlignet med $0,45 \mu\text{g}/\text{m}^3$ i maj 2016.

For TVOC er der i november 2015 målerunden (efterår) observeret en tendens til højere indeklimakoncentrationer (i den høje ende af distributionen) i hjem med brændeovn end i hjem uden brændeovn. Denne tendens ses ikke i maj 2016 målerunden (forår). Resultaterne indikerer at brændeovne, i nogle hjem, kan være forbundet med et øget internt bidrag af TVOC i fyringssæsonen.

I afsnit 6.3 er afgasningen af VOC'er fra skorstenen på et fastbrændselsfyr undersøgt. Her er det dokumenteret, at biomasseforbrænding er forbundet med afgasning af VOC'er. Det er ligeledes dokumenteret, at den lokale udendørs koncentration af benzen er højere i fyringssæsonen, og det forventes således at en lokal biomasseforbrænding medfører højere lokale udeluftkoncentrationer af benzen.

8.5 Kilder til kulbrinter i udeluften

Resultaterne i tabel 8.3 indikerer, at mellem ca. 50 og 80% af indeklimakoncentrationen af toluen, xylener og C9/C10-aromater skyldes interne kilder, mens kun ca. 25% af benzenkoncentrationen skyldes interne kilder, og 75% udendørs kilder.

For at undersøge en hypotese om at en betydelig andel af benzen i udeluften skyldes forbrænding af biomasse (brændeovne, pillefyr og lignende samt lokale biomassefyrede varmeværker) er der udført detaljerede studier af kulbrinteemissionerne fra skorstenen på et fastbrændselsfyr ved en enkelt bolig (jf. afsnit 6.3).

På baggrund af resultaterne fra dette studie står det klart, at forbrænding af biomasse påvirker udeluftkoncentrationerne af kulbrinter i fyringssæsonen. For benzen viser skorstensmålingerne ca. 100 gange højere koncentrationer end det generelle udeluftniveau omkring boligen. For toluen, xylener og naphthalen viser skorstensmålingerne ca. 10-100 gange højere koncentrationer end det generelle udeluftniveau omkring boligen. For C9/C10-aromater og TVOC viser skorstensmålingerne ca. 1-10 gange højere koncentrationer end det generelle udeluftniveau omkring boligen.

Levende planter er også undersøgt som kilde til kulbrinter (afsnit 6.6) og grene fra et juletræ (Nordmannsgran) er identificeret som en potentiel kilde til kulbrinter (undtaget benzen).

Prøvetagning i 21 positioner langs den jyske øst- og vestkyst (december 2016), med en overvejende vindretning fra vest (11 dage, imod 4 dage med en overvejende vindretning fra øst), gav median benzen koncentrationer på $0,17 \mu\text{g}/\text{m}^3$ på vestkysten og $0,23 \mu\text{g}/\text{m}^3$ på østkysten (afsnit 6.4). Resultaterne understreger at benzen stort set findes i al dansk udeluft i koncentrationer over afdampningskriteriet ($0,13 \mu\text{g}/\text{m}^3$), men underbygger også konklusionen om at der er mange landbaserede emissioner af benzen, f.eks. trafik og forbrænding af biomasse.

Prøvetagning i hav vindmølleparken Horns Rev 1 (afsnit 6.5) 14 km fra kysten viser benzenkoncentrationer mellem $0,20$ - $0,54 \mu\text{g}/\text{m}^3$. Dette indikerer, at der kan være udendørs kilder til benzen, som er relateret til olieflerterne i Nordsøen eller at kilder på de britiske øer evt. kan give anledning til et baggrunds niveau for benzen i Danmark, der er over afdampningskriteriet.

8.6 Duplikatvariation og variation på udeluftpositioner

I variationsstudiet (jf. afsnit 6.1.2 og 6.1.3) er der, i tre boliger, ophængt to ORSA-rør i fire udendørs positioner og i én indeklimaposition, over otte målerunder over et helt år. For dette datasæt er variationen på duplikater undersøgt for benzen og TVOC, og variationen på forskellige udeluftpositioner undersøgt for benzen.

For benzen viser resultaterne, at duplikatvariationen er mindre end 10-20% for 50% af målingerne, og mindre end 100% for 90% af målingerne. For positioner med benzenkoncentrationer, der overstiger afdampningskriteriet ($\geq 0,13 \mu\text{g}/\text{m}^3$), er duplikatvariationen kun $>100\%$ i ca. 4% af tilfældene. Dvs. at for halvdelen af benzen-målingerne er forskellen mellem 2 målerør på samme position mindre end 10-20%, men for 10% af målingerne er forskellen en faktor 2 eller mere.

For TVOC indikerer resultaterne, at duplikatvariationen er mindre end 10% for 50% af målingerne, og mindre end 100% for 90% af duplikaterne. Dvs. at for halvdelen af TVOC-målingerne er forskellen mellem 2 målerør på samme position mindre end 10%, men for 10% af målingerne er forskellen en faktor 2 eller mere.

Resultaterne for benzen i fire forskellige udeluftpositioner viser, at variationen på udeluftpositionerne er mindre end 20% for 50% af positionerne, og mindre end 100% for 92% af positionerne. Dvs. at for halvdelen af benzen-målingerne er forskellen mellem forskellige udeluftpositioner omkring en bolig mindre end 20%, men for 8% af målingerne er forskellen en faktor 2 eller mere.

8.7 Konklusioner relateret til indeklimaundersøgelser

8.7.1 Overordnede konklusioner

I store træk er de nye baggrundsniveauer (2015-2016), som er baseret på 14 dages ORSA-rørsmålinger, sammenlignelige med de tidligere niveauer fra 2000-2008, hvor andre opsamlingsmetoder generelt blev benyttet /1/. Mens de nye niveauer for BTEX og C9/C10-aromater kun er lidt lavere end de tidligere niveauer, så er de nye baggrundsniveauer for TVOC væsentligt lavere.

Baggrundsniveauet for benzen kan forventes at overskride afdampningskriteriet på $0,13 \mu\text{g}/\text{m}^3$ i både udeluften og i uforurenede danske boliger. I studiet er der konstateret indhold over afdampningskriteriet i $>99\%$ af udeluft- og indeklimatemålinger ved uforurenede danske boliger (100% i november 2015 og 98,9% i maj 2016). Det kan således forventes at afdampningskriteriet overskrides overalt i Danmark, selv på de mest øde strækninger af de danske kystlinjer.

Det kan forventes, at baggrundsniveauet for TVOC i indeklimate overskrider afdampningskriteriet på $100 \mu\text{g}/\text{m}^3$ i ca. 30% af uforurenede danske boliger (29% i november 2015 og 33% i maj 2016), mens det ligger under detektionsgrænsen (på ca. $43 \mu\text{g}/\text{m}^3$) i ca. 50% af de uforurenede danske hjem, uafhængigt af målerunden.

Koncentrationen af toluen, sum af xylener (inklusive ethylbenzen), naphthalen og C9/C10-aromater forventes ikke at overskride afdampningskriterierne i uforurenede danske boliger, hverken i udeluft eller i indeklimate. For disse komponenter er der således kun konstateret ganske få indhold ($<2\%$) over de respektive afdampningskriterier.

Baseret på de nye baggrundsniveauer fra dette studium, forventes det således ikke at indeklimateundersøgelser ifm. jord- og grundvandsforurening, hvor der foretages indeklimatemålinger, vil blive uheldigt påvirket af baggrundsniveauerne for toluen, xylener (inklusive ethylbenzen), naphthalen og C9/C10-aromater. For benzen kan det forventes, at indeklimate baggrundskoncentrationen overstiger afdampningskriteriet i $>99\%$ af uforurenede boliger, og for TVOC kan det forventes at indeklimate baggrundskoncentrationen overskrider afdampningskriteriet i ca. 30% af uforurenede boliger.

I studiet er det dokumenteret, at ca. 75% af indeklimatekoncentrationen af benzen i uforurenede boliger stammer fra udendørs kilder.

8.7.2 Sæsonvariationer

Baseret på to målerunder (november 2015 og maj 2016) og variationsstudiet (afsnit 6.1 og 6.2) er der identificeret signifikante sæsonvariationer i udeluftniveauet for benzen, xylener og C9/C10-aromater. De højeste niveauer findes i efterårs- og vintermånedene, og de laveste niveauer i forårs- og sommermånedene.

Det er dog kun baggrundsniveauerne af benzen, der har betydende variationer og dermed potentiale til at forstyrre tolkningen af resultaterne af indeklimaundersøgelser, baseret på indeklimamålinger.

8.7.3 Lokale faktorer af betydning

Indendørs tobaksrygning påvirker indeklimakoncentrationen af benzen i hjem hvor der ryges, og medianniveauet kan forventes at stige til ca. fire gange det generelle niveau for uforurenede danske boliger, når der ryges indendørs. Indendørs rygning påvirker også indeklimakoncentrationen af TVOC, og medianniveauet kan forventes at stige til ca. 2,5 gange det generelle niveau for uforurenede danske boliger, når der ryges indendørs.

I hjem med brændeovn kan der forventes ca. 60-80% højere indeklimakoncentrationer af benzen end i hjem uden brændeovn, uanset årstiden. I nogle hjem med brændeovn kan der, i fyringssæsonen, forventes en højere indeklimakoncentration af TVOC. I hjem med fastbrændselsfyr kan der forventes højere lokale udeluftkoncentrationer af benzen.

Effekten af befolkningstæthed, udluftningsvaner, renovering, antal beboere, bygningsalder og lokale trafikrelaterede emissioner forventes kun at påvirke baggrundsniveauet af kulbrinter i mindre grad – eller slet ikke.

8.7.4 Brug af udereferencer

I indeklimaundersøgelser, relateret til jord- og grundvandsforurening, bør effekterne af sæsonvariationer, lokal biomasseforbrænding og andre lokale variationer i udeluftkoncentration indtages ved altid at inkludere udeluftreferencer.

For halvdelen af benzen og TVOC-målinger er forskellen mellem 2 målerør i samme position (benzen og TVOC), eller målinger i 2 forskellige udeluftpositioner (benzen), mindre end 10-20%, men for ca. 10% af målingerne er forskellen en faktor 2 eller mere. Det bemærkes, at 20% variation/usikkerhed på median udeluftkoncentrationen af benzen i november 2015 målerunden er på niveau med afdampningskriteriet for benzen (20% af 0,50 µg/m³).

Det anbefales at der udføres mindst 2 udeluft referencemålinger i forbindelse med indeklimamålinger for kulbrinter, da udeluftkoncentrationer i mange (for benzen næsten alle) tilfælde vil være af stor betydning ved vurdering af risikoen for en uacceptabel afdampning fra en jordforurening til indeklimaet.

9. References

- /1/ Prioriteringsniveauer for indeklimasager på kortlagte vejendomme. Videncenter for Jordforurening. Teknik og Administration, nr. 2, 2010.
- /2/ WHO guidelines for indoor air quality: Selected pollutants. ISBN 978 92 890 0213 4. WHO, 2010.
- /3/ The influence of biomass burning on the global distribution of selected non-methane organic compounds. A. C. Lewis, M. J. Evans, J. R. Hopkins, S. Punjabi, K. A. Read, S. Andrews, S. J. Moller, L. J. Carpenter, J. D. Lee, A. R. Rickard, P. I. Palmer and M. Parrington. Atmos. Chem. Phys. Discuss.: 10 Sep 2012.
- /4/ Levels and sources of BTEX in ambient air of Ahvaz metropolitan city. Rad, H. D., Babaei, A. A., Goudarzi, G., Angali, K. A., Ramezani, Z., and Mohammadi, M.M. Air Quality, Atmosphere & Health, 7(4), 515-524, 2014.
- /5/ Ambient levels of volatile organic compounds in the vicinity of petrochemical industrial area of Yokohama, Japan. Tiwari, V., Hanai, Y. and Masunaga, S. Air Qual. Atmos. Health, 3, 65-75, 2010. DOI 10.1007/s11869-009-0052-0
- /6/ Elevated Atmospheric Levels of Benzene and Benzene-Related Compounds from Unconventional Shale Extraction and Processing: Human Health Concern for Residential Communities. Rich, A. L., and Orimoloye, H. T. Environmental Health Insights, 10, 75–82, 2016. <http://doi.org/10.4137/EHI.S33314>
- /7/ Clinical Environmental Health and Toxic Exposures. Second Edition. Sullivan, J. B., and Krieger, G. R. (Eds.). Lippincott Williams & Wilkins. 2001. ISBN 9780683080278.
- /8/ DCE – Danish Center for Environment and Energy, Aarhus University. Actual air quality in Denmark. Data based on several annual reports retrieved from:
<http://envs.au.dk/videnuveksling/luft/maaling/aarsrapporter/>
[http://www2.dmu.dk/1_Viden/2_miljoe-tilstand/3_luft/4_litteratur/LmpRap.asp?a=\(DMUA.TIT%20Like%20%27%25Air%20Quality%20Monitoring%20Programme%25%27\)](http://www2.dmu.dk/1_Viden/2_miljoe-tilstand/3_luft/4_litteratur/LmpRap.asp?a=(DMUA.TIT%20Like%20%27%25Air%20Quality%20Monitoring%20Programme%25%27))
- /9/ Diesebiler taber terræn. Artikel på fdm.dk, 4. november 2015.
<https://fdm.dk/nyheder/2017-09-diesebiler-taber-terraen>
- /10/ Kortlægning og risikovurdering af toluen og andre neurotoksiske stoffer i børneværelset. Kortlægning af kemiske stoffer i forbrugerprodukter nr. 145, 2016. Danish Environmental Protection Agency (Miljøstyrelsen). ISBN 978-87-93435-41-4.
- /11/ Status og perspektiver på indeklimaområdet. Danish Environmental Protection Agency (Miljøstyrelsen). Project no. 1097, 2006.
- /12/ Bolius Boligejeranalyse, Boligejernes Videncenter. Kantar Gallup. May 2017.
- /13/ Emissions of volatile organic compounds from vegetation and the implications for atmospheric chemistry. Fehsenfeld, F., Calvert, J., Fall, R., Goldan, P., Guenther, A. B., Hewitt, C. N., Lamb, B., Liu, S., Trainer, M. Westberg, H. and Zimmerman, P. Global Biogeochemical Cycles, 6(4), 389-430, 2991.
- /14/ Emissions of volatile organic compounds during the decomposition of plant litter. Gray, C. M., Monson, R. K. and Fierer, N. Journal of Geophysical Research, 115, G03015, 2010. <http://doi:10.1029/2010JG001291>.
- /15/ Volatile organic compounds (VOCs) in air from Nisyros Island (Dodecanese Archipelago, Greece): Natural versus anthropogenic sources. Tassi, F., Capecchiacci, F., Gianini, L., Vougioukalakis, G. E. and Vaselli, O. Environmental Pollution, 180, 111-121, 2013.

- /16/ Guidelines for managing water quality impacts within UK European marine sites. Cole, S., Codling, I. D., Parr, W. and Zabel, T. UK Marine SAC Project, 1999.
- /17/ Air Pollution and Water Quality. Atmospheric Deposition Initiative. What is atmospheric deposition and how does it occur? USEPA. Office of Water.
http://itepsrv1.itep.nau.edu/itep_course_downloads/Ecosystems%20_Resources/Air_Pollution_Water_Quality.pdf
- /18/ Benzene, alkylated benzenes, chlorinated hydrocarbons and monoterpenes in snow/ice at Jungfrauoch (46.6 degrees N, 8.0 degrees E) during CLACE 4 and 5. Fries E., Sieg K., Püttmann W., Jaeschke W., Winterhalter R., Williams J. and Moortgat G. K. Sci Total Environ. 391(2-3):269-277, 2008. DOI: 10.1016/j.scitotenv.2007.10.006
- /19/ The Danish Air Quality Monitoring Programme. Annual Summary for 2016. Scientific Report from DCE – Danish Centre for Environment and Energy. No. 234, 2017. ISBN 978-87-7156-273-6.
- /20/ Danish Emission Inventories for Road Transport and Other Mobile Sources. Inventories until the year 2013. Scientific Report from DCE – Danish Centre for Environment and Energy. No. 148, 2015. ISBN 978-87-7156-138-8.
- /21/ NIRAS (2012). Togtrafikkens påvirkning af udeluften med udgangspunkt i stationsbyerne Hjerm, Studsgård og Struer (Report nr. 14.490.00).
- /22/ Background Indoor Air Concentrations of Volatile Organic Compounds in North America Residences (1990-2005): A Compilation of Statistics for Assessing Vapor Intrusion. USEPA. OSWER. EPA 530-R-10-001, 2011.
- /23/ Liste over kvalitetskriterier i relation til forurenede jord og kvalitetskriterier for drikkevand. Danish Environmental Protection Agency (Miljøstyrelsen), June 2015. Retrieved from: <http://mst.dk/media/90004/kvalitetskriterier-jord-og-drikkevand-juni-2015.pdf>
- /24/ Cigaretrøg kan være en væsentlig kilde til benzen i indeklimaet. Larsen, P., Loll, P., Larsen, C. and Østergaard, H. K. Miljø og Ressourcer nr. 3, 20-28, 2016.
- /25/ Personal Monitoring of Selected VOCs: The Contribution of Woodsmoke to Exposure. Department of the Environment and Heritage. Natural Heritage Trust. CSIRO Atmospheric Research, 2004. ISBN 0642550255.
- /26/ Dräger-Tubes & CMS-Handbook. 16th edition. Soil, water, and air Investigations as well as Technical Gas Analysis. 2011. ISBN 3-926762-06-3.
- /27/ Diffusion Sampler ORSA.
<http://www.afcintl.com/pdfs/Draeger%20pdfs/draeger/6728891.pdf>
- /28/ Hydrocarbons, BP 36-216°C: Method 1500, Issue 3. NIOSH Manual of Analytical Methods (NMAM). Fourth Edition. 15. March 2003.
- /29/ Hydrocarbons, Aromatic: Method 1501, Issue 3. NIOSH Manual of Analytical Methods (NMAM). Fourth Edition. 15. March 2003.
- /30/ ATD-rør og ORSA-rør giver forskellige resultater for TVOC. Loll, P. and Jannerup, M. Miljø og Ressourcer, 3, 3-12, 2017.
- /31/ Practical nonparametric statistics. Conover, W. J. John Wiley and Sons, 1999. ISBN 0-471-16068-7.
- /32/ Directive 2008/50/EC of the European Parliament and of the Council of 15 December 2004 on ambient air quality and cleaner air for Europe. Official Journal of the European Union L152/1.

Appendix 1. Pre-study of outdoor sampler deployment

A pre-study of outdoor sampler deployment was conducted in June 2015. The pre-study aimed at testing two possible strategies for outdoor sampler deployment to be used in the main study: ORSA-samplers sheltered from rain by plastic cups and ORSA-samplers without cups. The cups were intended to protect the ORSA-samplers from external factors, but at the same time it was a concern that they would lead to higher concentrations due to emission of VOCs.

The pre-test was conducted at 6 residences, located in a city (Aalborg, 113.000 inhabitants), two towns (Viborg, 40.400 inhabitants and Struer, 10.400 inhabitants), a village (Ulsted, 1.100 inhabitants) and a residence in the countryside (Land). The experimental set-up is shown in the schematic below.

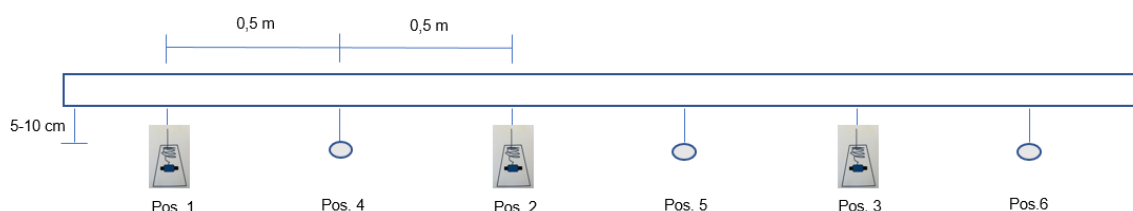


FIGURE A1.1. Illustration of the pre-study outdoor sampler deployment set-up.

The results for naphthalene, C10-aromatics and TVOC were below the analytical detection limits in all samples. The average values of each set-up (with and without cups) are plotted against each other, along with their min-max error bars to show the extent of the values of each data-set.

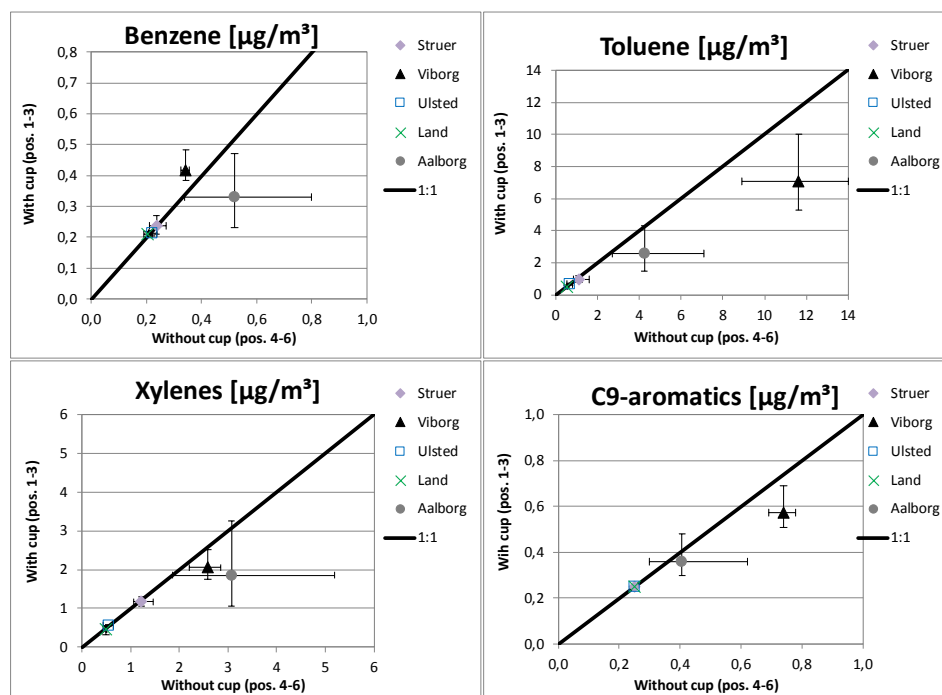


FIGURE A1.2. Average concentrations and min-max error bars for each location with and without a protective plastic cup are shown for benzene (top - left), toluene (top – right), xylenes (bottom – left) and C9-aromatics (bottom right).

As shown in figure A1.2, higher concentrations of toluene, xylenes and C9-aromatics are generally observed for sampler deployment without protective cups. For benzene however, the data pairs are ambiguous, since data for four of the locations rest on the 1:1 line (which means that they are equal), one data pair (the Viborg location) gives slightly higher benzene concentrations with sampler deployment with protective cups than without cups, and another (Aalborg) shows higher concentration with sampler deployment without cups than when with cups.

Overall, it is concluded that the set-up without protective plastic cups yields more representative results than the deployment with cups. Hence sampler deployment for the further studies was conducted without cups, with exception of the wind turbine study.

Appendix 2. Questionnaire and deployment guidance document (in Danish)

2015-0564 Baggrunds niveauer af olie kulbrinter

Medarbejder: _____

Information om ORSA-rør:

Prøvenr.	Ophængt		Nedtaget	
	Dato	Kl	Dato	Kl
-inde				
-ude				

Centralt i stuen
Beskyttet udenfor (f.eks. under udhæng)

Adresse: _____

Boligen er opført: _____ Antal beboere: _____

Etageejendom Rækkehus Villa Landejendom

Boligtipe:

Etage: _____

Hvilken zone ligger boligen i ?

by (≥ Aalborg) Mindre by Landsby På landet

Hvordan opvarmes boligen?

Oliefyr Fjernvarme Naturgas Elvarme Brændeovn

Hvis oliefyr. Er tanken indenfor? Ja Nej

Hvis brændeovn, hvor ofte benyttes den (i gennemsnit)?

-3 x pr. md. 1-2 x pr. uge 3-7 x pr. uge Hvor mange gange har den ca. været benyttet i måleperioden? _____

Bor der rygere i boligen? Ja Nej

Hvis ja, bliver der røget indendørs?

Er der blevet renoveret rum i boligen indenfor det seneste år? Ja Nej
(inkl. maling af rum)

Hvis ja, hvilken form for renovering?

Ligger boligen ved?

<100 meter til:

Motorvej Hovedvej Villavej Vej/gade i by Lyskryds Tankstation

Hvordan udluftes boligen?

Ventilationsanlæg (f.eks. Genveks) Manuel udluftning (evt. med alm. emhætte og badeværelsesventilator)

Hvis manuel udluftning, hvor ofte i gennemsnit (≥5-10 min varighed)?

<1 x pr. d. 1-2 x pr. d. 2-4 x pr. d. Ofte eller "kontinuerligt" (f.eks. åbent vindue længere ad gangen)

Har I hobbyer, som indebærer brug af produkter, som kan afgive kulbrinter (brug af oliemaling, lim, lak o.lign.) Ja Nej

Hvis ja, foregår brugen af produkterne indendørs

Og er produkterne brugt i måleperioden

Hvis ja, hvilke: _____

Findes der industri i omkreds af 1 km fra boligen? Ja Nej

Hvis ja, hvilke(n) form(er) for industri:

Bemærkninger om andre forhold der kunne have betydning for resultaterne:
(interne bidrag, ventilationsforhold eller andet)

Inspiration ift. ophængning af ORSA-rør

Udenfor:

Målingen skal repræsentere "frisk luft", der hvor boligen er – overvej dette ved valg af placering. Sæt gerne røret lidt beskyttet (det må helst ikke få direkte regn, men skal sidde i fri luft. Placer f.eks. røret under et tagudhæng eller lignende. **Husk at notere tidspunkt for ophængning og nedtagning i infoarket!**



Indenfor:

Målingen skal repræsentere middelluften i stuen.

Placeres derfor helst centralt i stuen. *Ikke* lige ved et vindue, en ventilationskanal eller en varmekilde (f.eks. *ikke* der hvor der kommer direkte strålevarme fra en lampe eller en radiator). Det er ok at lægge den på toppen af en høj reol i stedet for at hænge op, men placér den da længst fremme på et hjørne, så luften har fri bevægelse omkring rør-enderne. **Husk at notere tidspunkt for ophængning og nedtagning i infoarket!**



144	1920	2	Countryside	X	X	3-7 per week	11	Manual	<1 per day	X	Painting							
145	1901	1	Village	X				Manual	1-2 per day	X								X
146	1934	4	Town		X	1-3 per month	1	Manual	1-2 per day	X	New plaster walls, insulation and painting.							X
147	1971	1	Town		X	3-7 per week	5-6	Manual	<1 per day	X								
148	1920	4	Countryside		X			Manual		X								
149	1911	1	Village		X			Manual	1-2 per day	X								
150	1915	2	Countryside		X			Manual	1-2 per day	X								
151	1920	4	Countryside	X				Manual	1-2 per day	X								
152	1923	3	Town		X			Manual	<1 per day	X								
153	1950	3	City		X			Manual	1-2 per day	X								Ventilation strips always open
154	1930	5	Town		X	1-3 per month		Mechanical		X	Painting of living room, bathroom downstairs, new kitchen, bathroom fixtures							
155	1971	2	City		X	1-2 per week	0	Manual	1-2 per day	X	Renovation of kitchen, ceiling and walls (painted)							
157	1981	4	Town		X	3-7 per week	approx. 7	Manual	<1 per day	X	Painted ceiling, walls inside, 1-4 months.							
159	1980	4	Countryside		X			Mechanical		X								
161	2010	5	Village		X			Manual	1-2 per day	X								
162	1986	2	Village		X			Manual	1-2 per day	X								X
163	2014	2	Village		X			Manual	1-2 per day	X								X
164	1829	5	Countryside		X	1-2 per week	1	Manual	<1 per day	X	Replacement of doors and windows							
																		Gas stove (open living room/ kitchen) not used in the summer period. Often candles.

111	1928	4	Town	X			Manual	<1 x pr. d.	X						
113	2004	4	Village	X			Mechanical	<1 x pr. d.	X	Painted walls				X	
114	2001	4	Town	X		2	Manual	More often	X					X	There is grill directly under 113-ude sample (gas grill).
115	1922	1	Countryside	X			Manual	More often	X						
116	1980	4	Town	X	3-7 per week	2	Manual	<1 x pr. d.	X	Bedroom is watercrafted and painted. ORSA tubes are located in the living room.				X	Agriculture within 1 km.
117	1969	1	Town	X			Manual	1-2 x pr. d.	X	Painting of kitchen and dining room (partly).				X	
118	1964	4	Town	X	3-7 per week	3	Manual	1-2 x pr. d.	X	Painting of living room				X	
119	1987	5	Village	X	3-7 per week	0	Manual	1-2 x pr. d.	X					X	
121	1912	2	City	X			Manual	1-2 x pr. d.	X				X	X	Outdoor sample may be broken. Indoor sample is from home with smoking, so it is forwarded anyway.
123	1935	16	Town	X			Manual	<1 x pr. d.	X					X	
124	1986	3	Village	X	3-7 per week		Manual	More often	X					X	There is no fire at the stove during the hanging period.
125	1962	2	Countryside	X			Manual	1-2 x pr. d.	X						Work clothes can deliver oil / paint / fuel odors at the residence
126	1974	2	Town	X			Manual	<1 x pr. d.	X					X	
128	1925	4	Village	X	3-7 per week	2	Manual	More often	X	Painting of ceiling in the living room. Ca. 42 m ² , min volume 145 m ³ .				X	
129	1927	1	Town	X			Manual	1-2 x pr. d.	X					X	
130	1979	3	Town	X			Manual	1-2 x pr. d.	X					X	1 time application of nail polish.
132	1927	2	Countryside	X			Manual	1-2 x pr. d.	X						
133	1970	1	City	X			Manual	1-2 x pr. d.	X						
134	1926	1	Villa	X			Manual	1-2 x pr. d.	X				X		
135	2012	4	Countryside	X	1-3 per month		Manual	1-2 x pr. d.	X						
136	1937	1	Countryside	X			Manual	More often	X						
137	1953	2	City	X	3-7 per week	0	Manual	More often	X					X	Venting approx. every other day in the living room. Daily on the 1st floor (all night).
138	1897	4	City	X				1-2 x pr. d.	X	New floor, linoleum, laminate, painting, jointing, new bathroom, oiled wood flooring.				X	Have just got new linoleum flooring in progress.
139	around 1950	2	Countryside	X	3-7 per week	0	Manual	More often	X	Painting					In the forest. Wood stove not in use in the summer period.
140	2007	4	Village	X			Mechanical		X	Painting and new carpets				X	
141	2006	2	Town	X			Manual	More often	X					X	
142	1947	2	Town	X	3-7 per week	1	Manual	<1 x pr. d.	X	Painted walls				X	
144	1856	2	Village	X		0	Manual	2-4 x pr. d.	X					X	
145	1960	2	Town	X			Manual	1-2 x pr. d.	X					X	
146	1977	2	Town	X			Manual	More often	X					X	
147	1927/1992	2	Town	X			Manual	2-4 x pr. d.	X	Hallway (painting, ceiling, walls, new carpet. Living room (painting of walls and new floor))				X	
149	1973	4	Town	X	1-2 per week	0	Manual	1-2 x pr. d.	X					X	
150	1979	5	Town	X			Manual		X	Painting and laying of new floor in two bedrooms, no change in the living room				X	
152	1973	4	Town	X	1-3 per month	0	Manual	<1 x pr. d.	X	Total renovation of the toilet and office: new floors, new ceilings, new walls, painted etc.				X	
153	1977	2	Village	X			Mechanical		X					X	
154	1985	4	Village	X	1-3 per month	0	Mechanical	1-2 x pr. d.	X	New wooden floor, painting, new ceilings, new kitchen				X	
155	1965	2	Town	X			Manual		X					X	
157	1901	1	Village	X			Manual	1-2 x pr. d.	X					X	Has a separate smoking oven in the garden.
159	1960	2	City	X			Manual	More often	X	Painting and new carpet				X	
160	2005	4	Town	X	1-3 per month			2-4 x pr. d.	X	Painting				X	One week on vacation during sampling = limited ventilation.
161	1912	5	Village	X				1-2 x pr. d.	X					X	

109	0,36	3,5	0,34	0,23	0,50	1,1	< 0,44	interference	1,1	41	109	0,29	0,80	0,13	0,15	0,42	0,7	< 0,44	0,26	< 0,12	0,26	< 44	
110	0,77	10	0,26	0,25	0,82	1,3	< 0,44	interference	1,0	100	110	0,31	0,89	0,11	0,12	0,41	0,64	< 0,44	0,21	< 0,12	0,21	< 43	
111	3,0	35	5,6	5,3	21	32	< 0,44	6,6	3,2	9,8	520	111	0,43	0,97	0,15	0,16	0,48	0,79	< 0,44	0,25	< 0,11	0,25	< 43
112	0,68	10	0,68	0,58	2,2	3,5	< 0,43	interference	1,2	< 43	112	0,45	0,93	0,12	0,15	0,41	0,68	< 0,41	0,24	< 0,11	0,24	< 41	
113	0,46	0,96	0,24	0,16	0,49	0,89	< 0,43	0,41	1,1	1,5	67	113	0,40	0,69	0,10	0,12	0,37	0,59	< 0,43	0,20	< 0,11	0,20	< 42
114	0,63	3,0	0,58	0,50	1,6	2,7	< 0,44	interference	1,7	< 43	114	0,54	1,1	0,17	0,19	0,60	0,96	< 0,44	0,25	< 0,12	0,25	< 43	
115	0,52	12	0,78	0,55	2,0	3,3	< 0,41	interference	2,0	< 40	115	0,41	0,84	0,12	0,13	0,41	0,66	< 0,41	0,13	< 0,11	0,13	< 40	
116	3,6	32	3,8	4,0	14	22	< 0,44	3,9	1,7	5,6	< 44	116	0,38	1,3	0,18	0,20	0,64	1,0	< 0,44	0,28	< 0,12	0,28	< 44
117	0,67	2,5	0,47	0,44	1,7	2,6	< 0,44	0,53	0,67	1,2	< 43	117	1,0	1,7	0,27	0,30	0,93	1,5	< 0,44	0,68	< 0,12	0,68	< 43
118	0,84	5,5	0,71	1,6	2,4	4,7	< 0,44	3,8	2,3	6,1	< 43	118	0,76	2,5	0,36	0,37	1,3	2,0	< 0,44	0,42	< 0,12	0,42	< 43
119	1,0	4,6	0,87	0,61	1,7	3,2	< 0,44	interference	3,3	61	119	0,39	0,61	0,11	0,11	0,32	0,54	< 0,44	0,14	< 0,12	0,14	< 43	
120	2,2	24	2,7	2,8	11	17	< 0,44	2,8	4,5	7,3	310	120	0,50	2,4	0,34	0,37	1,3	2,0	< 0,44	0,43	< 0,12	0,43	< 43
121	0,56	75	1,6	2,1	6,4	10	< 0,44	interference	2,2	560	121	0,54	1,2	0,17	0,18	0,56	0,91	< 0,44	0,19	< 0,12	0,19	< 43	
122	1,2	17	0,46	0,48	1,5	2,4	< 0,44	interference	3,8	210	122	0,70	2,9	0,38	0,43	1,5	2,3	< 0,44	0,53	< 0,12	0,53	< 44	
123	0,40	3,2	0,16	0,16	0,45	0,77	< 0,44	interference	0,66	< 43	123	0,59	2,2	0,29	0,30	1,0	1,6	< 0,44	0,26	< 0,12	0,26	< 43	
124	6,4	13	2,8	2,1	8,7	14	< 0,56	1,8	1,7	3,5	< 55	124	0,52	< 0,55	< 0,12	< 0,12	0,29	0,29	< 0,56	< 0,13	< 0,15	< 0,15	< 55
125	0,41	3,8	0,25	0,23	0,69	1,2	< 0,45	0,47	0,32	0,8	< 45	125	0,71	3,5	0,28	0,30	1,0	1,6	< 0,45	0,15	< 0,12	0,15	< 45
126	0,92	12	0,89	1,0	3,1	5,0	< 0,44	1,8	1,1	2,9	< 43	126	0,22	0,55	0,12	0,13	0,41	0,66	< 0,44	0,34	< 0,12	0,34	< 43
127	0,38	0,88	0,34	0,15	0,45	0,94	< 0,45	interference	0,99	< 44	127	0,47	0,46	< 0,10	< 0,09	0,26	0,26	< 0,45	interference	< 0,12		< 44	
128	1,0	7,6	0,97	1,0	3,4	5,4	< 0,44	interference	2,0	64	128	0,49	0,47	< 0,10	< 0,09	0,25	0,25	< 0,44	0,10	< 0,12	0,10	< 44	
129	0,86	10	0,80	0,81	2,7	4,3	< 0,44	interference	2,0	37	129	0,44	0,55	< 0,10	0,10	0,27	0,37	< 0,44	< 0,10	< 0,12	< 0,12	< 44	
130	4,2	11	1,2	0,65	3,5	5,4	< 0,44	interference	4,8	300	130	0,47	0,98	0,14	0,19	0,47	0,8	< 0,44	interference	< 0,12		< 43	
131	0,63	5,2	0,44	0,45	1,5	2,4	< 0,44	0,84	0,48	1,3	< 43	131	2,2	3,7	0,47	0,50	1,8	2,8	< 0,44	0,48	< 0,12	0,48	< 43
132	0,67	4,9	0,39	0,37	1,1	1,9	< 0,44	1,2	0,76	2,0	41	132	0,73	1,5	0,21	0,22	0,65	1,1	< 0,44	0,30	< 0,12	0,30	< 43
133	0,91	18	0,87	0,99	2,7	4,6	< 0,44	interference	3,1	190	133	0,65	1,6	0,16	0,20	0,55	0,91	< 0,44	0,31	< 0,12	0,31	< 43	
134	0,44	1,9	0,38	0,20	0,58	1,2	< 0,44	0,53	0,58	1,1	< 44	134	0,68	1,3	0,19	0,23	0,64	1,1	< 0,44	0,36	< 0,12	0,36	< 44
137	2,7	8,1	1,2	0,99	3,4	5,6	< 0,39	1,8	1,1	2,9	220	137	0,77	1,9	0,31	0,36	1,1	1,8	< 0,39	0,37	< 0,10	0,37	< 39
140	1,3	9,6	0,46	0,31	0,98	1,8	< 0,44	interference	1,3	76	140	0,86	1,1	0,16	0,18	0,54	0,88	< 0,44	0,29	< 0,12	0,29	< 44	
143	0,81	3,5	0,39	0,40	1,1	1,9	< 0,44	interference	1,7	180	143												
144	5,9	4,2	0,60	0,57	1,9	3,1	< 0,44	1,8	0,81	2,6	61	144	0,69	0,69	0,11	0,12	0,34	0,57	< 0,44	< 0,10	< 0,12	< 0,12	< 43
145	2,5	7,0	2,8	2,5	9,5	15	< 0,44	0,78	0,81	1,6	< 43	145	0,53	1,1	0,66	0,63	2,3	3,6	< 0,44	0,26	< 0,12	0,26	< 43
146	2,3	25	4,0	5,5	15	25	< 0,44	10	3,0	13	< 43	146	0,43	1,3	0,20	0,25	0,64	1,1	< 0,44	0,27	< 0,12	0,27	< 43
147	1,3	6,0	0,54	0,71	1,7	3,0	< 0,44	2,0	1,3	3,3	< 43	147	0,54	1,7	0,29	0,37	0,95	1,6	< 0,44	0,45	< 0,12	0,45	< 43
148	1,4	18	0,30	0,19	0,57	1,1	< 0,43	2,2	3,4	5,6	170	148	0,40	< 0,43	< 0,10	< 0,09	0,23	0,23	< 0,43	0,12	< 0,11	0,12	< 43
149	0,40	2,5	0,25	0,19	0,41	0,9	< 0,39	0,84	0,92	1,8	< 39	149	0,52	0,90	0,17	0,17	0,54	0,88	< 0,39	0,42	< 0,10	0,42	< 39
150	0,49	5,0	0,17	0,16	0,41	0,7	< 0,39	4,5	3,4	7,9	96	150	0,49	0,64	0,12	0,12	0,33	0,57	< 0,39	< 0,09	< 0,10	< 0,10	< 39
151	2,7	12	1,8	1,8	4,5	8,1	< 0,41	3,6	2,2	5,8	130	151	0,54	0,52	0,11	0,10	0,28	0,49	< 0,41	0,11	< 0,11	0,11	< 41
152	0,83	1,8	0,27	0,26	0,65	1,2	< 0,44	0,88	1,2	2,1	< 43	152	0,68	0,59	0,11	0,09	0,30	0,50	< 0,44	< 0,10	< 0,12	< 0,12	< 43
153	0,57	2,0	1,2	0,48	2,4	4,1	< 0,43	interference	2,8	88	153	0,47	0,95	0,14	0,15	0,50	0,79	< 0,43	0,30	< 0,11	0,30	< 43	
154	0,41	1,4	0,51	1,1	1,7	3,3	< 0,42	2,9	0,89	3,8	55	154	0,47	0,90	0,15	0,16	0,52	0,83	< 0,43	0,31	< 0,11	0,31	< 43
155	0,59	3,5	0,31	0,28	0,86	1,5	< 0,47	interference	0,81	< 46	155	0,45	1,0	0,17	0,18	0,58	0,93	< 0,47	0,35	< 0,12	0,35	< 46	
157	0,57	3,2	0,46	0,53	1,2	2,2	< 0,43	interference	1,6	40	157	0,51	0,84	0,15	0,21	0,47	0,83	< 0,43	0,29	< 0,11	0,29	< 43	
159	1,4	13	6,4	4,7	21	32	< 0,44	3,6	1,3	4,9	< 43	159	0,50	1,8	0,28	0,28	0,98	1,5	< 0,44	0,39	< 0,12	0,39	< 43
161	0,53	20	2,8	1,8	5,3	9,9	< 0,44	interference	4,8	< 44	161	0,54	1,1	0,20	0,19	0,59	0,98	< 0,44	0,29	< 0,12	0,29	< 44	
162	0,55	3,5	0,26	0,25	0,75	1,3	< 0,44	0,78	interference	< 44	162	0,57	1,1	0,19	0,18	0,59	0,96	< 0,44	0,34	< 0,12	0,34	< 44	
163	0,52	4,3	2,3	0,40	1,3	4,0	< 0,44	3,7	4,0	7,7	< 44	163	0,43	1,0	0,19	0,19	0,59	0,97	< 0,44	0,25	< 0,12	0,25	< 44
164	0,70	1,5	0,38	0,30	1,1	1,8	< 0,44	interference	1,5	43	164	0,49	1,4	0,22	0,24	0,81	1,3	< 0,44	0,42	< 0,12	0,42	< 43	

Ude-4a	0,087	0,11	0,11	0,24	0,29	0,11	0,25	0,31	Ude-4a	0,10	0,12	0,12	0,25	0,20	0,26	0,12	0,27	Ude-5a	0,10	0,091	0,17	0,31	0,20	0,12	0,19	0,21
Ude-4b		0,11	0,11	0,20	0,32	0,11	0,21	0,37	Ude-4b		0,12	0,12	0,19	0,21	0,22	0,12	0,26	Ude-5b		0,091	0,18	0,31	0,21	0,12	0,17	0,23

TABLE A3.6. Raw Data – Detailed study of variation. Blue: under the detection limit, red: above the regulatory limit.

Residence C													
Position	nov-15	feb-16	apr-16	maj-16	jul-16	sep-16	nov-16	dec-16	jan-17	mar-17	maj-17	jun-17	sep-17
Ude-1	V-garage	V-garage	V-garage	V-garage	V-garage	V-garage	V-garage	V-garage	V-garage	V-hus	V-garage	V-garage	V-garage
Ude-2	S	S	S	S	S	S	S	S	S	S	S	S	S
Ude-3	E	E	E	E	E	E	E	E	E	E	E	E	E
Ude-4	N	N	N	N	N	N	N	N	N	N	N	N	N
Ude-5	V-house	V-house	V-house	V-house	V-house	V-house	V-house	V-house	V-house	V-house	V-house	V-house	V-house
Ude-6	Blind	Blind	Blind	Blind	Blind	Blind	Blind	Blind	Blind	Blind	Blind	Blind	Blind
Benzene (µg/m³)													
Indoor median	0,54	0,42	0,28	0,044	0,0077	0,28	0,32	0,51	0,75	0,31	0,17	0,036	0,29
Inde-a	0,54	0,42	0,28	0,044	0,0077	0,28	0,28	0,57	0,72	0,32	0,15	0,030	0,30
Inde-b							0,35	0,45	0,77	0,30	0,18	0,041	0,28
Outdoor median						0,24	0,42	0,50	0,97	0,32	0,18	0,053	0,26
Ude-2a							0,42	0,43	0,81	0,24		0,011	0,17
Ude-2b							0,34	0,52	0,96	0,37	0,18	0,082	0,22
Ude-3a						0,24	0,45	0,47	1,1	0,31	0,20	0,053	0,26
Ude-3b							0,52	0,49	0,97	0,40	0,20	0,076	0,23
Ude-4a						0,29	0,39	0,39	0,91	0,33	0,16	0,0077	0,25
Ude-4b							0,42	0,56	0,97	0,29	0,12	0,16	0,26
Ude-5a						0,21	0,42	0,52	0,99	0,30	0,14	0,0077	0,31
Ude-5b							0,41	0,50	1,0	0,35	0,15	0,052	0,33
Ude-1a	0,70	0,80	0,50	0,58	2,6	1,8	0,80	1,3	1,5	0,91	0,87	0,77	5,4
Ude-1b							0,85	1,1	1,8	1,1	0,77	0,84	4,7
TVOC (µg/m³)													
Indoor median	42	49	44	37	57	38	158	36	39	119	61	43	38
Inde-a	42	49	44	37	57	38	36	38	39	140	52	43	36
Inde-b							280	34	39	97	70	43	39
Outdoor median						38	34	34	39	40	43	43	34
Ude-2a							34	34	43	40		43	34
Ude-2b							34	34	39	40	43	43	34
Ude-3a						38	34	34	39	40	43	43	34
Ude-3b							34	34	39	40	43	43	34
Ude-4a						38	34	34	39	40	43	43	34
Ude-4b							34	34	39	40	43	43	34
Ude-5a						38	34	34	39	40	43	43	34
Ude-5b							34	34	39	40	43	43	34
Ude-1a	42	39	44	37	120	38	34	34	43	49	46	49	120
Ude-1b						38	34	34	39	40	43	66	110
Toluene (µg/m³)													
Indoor median	7,2	2,9	3,2	2,6	5,7	5,9	4,1	4,1	3,6	2,7	3,9	4,0	6,2
Inde-a	7,2	2,9	3,2	2,6	5,7	5,9	3,3	4,1	3,4	2,6	3,9	4,0	6,0
Inde-b							4,9	4,0	3,8	2,8	3,9	4,0	6,3
Outdoor median						1,1	0,48	1,2	1,9	1,0	1,6	1,6	1,8
Ude-2a							0,47	1,0	1,7	0,78		1,6	1,6
Ude-2b							0,35	1,1	1,9	1,3	1,7	1,6	1,8
Ude-3a						0,97	0,67	1,2	2,1	0,99	1,6	1,4	1,7
Ude-3b							0,65	1,3	1,9	1,3	1,6	1,5	1,6
Ude-4a							1,4	0,34	0,80	1,6	1,5	1,1	1,4
Ude-4b								0,42	0,94	1,8	1,0	1,3	1,5
Ude-5a						1,1	0,66	1,2	1,9	0,99	1,4	1,6	1,8
Ude-5b							0,48	1,2	2,0	0,93	1,5	1,6	1,9
Ude-1a	2,3	4,1	2,5	6,0	24	14	4,9	8,1	7,9	6,9	9,8	9,3	42
Ude-1b							5,1	7,4	10	7,6	8,5	11	37
Xylenes (µg/m³)													
Indoor median	2,7	1,5	2,6	1,7	2,9	2,8	2,0	2,1	2,2	1,6	2,2	2,3	3,3
Inde-a	2,7	1,5	2,6	1,7	2,9	2,8	1,7	2,2	2,2	1,6	2,3	2,2	3,1
Inde-b							2,4	2,0	2,3	1,6	2,1	2,4	3,4
Outdoor median						1,0	0,53	1,1	1,7	1,1	1,4	1,6	1,8
Ude-2a							0,50	1,0	1,5	0,74		1,5	1,6
Ude-2b							0,40	1,1	1,7	1,3	1,4	1,6	1,8
Ude-3a						0,91	0,75	1,3	2,0	1,1	1,4	1,5	1,8
Ude-3b							0,74	1,3	1,8	1,4	1,4	1,6	1,6
Ude-4a							1,3	0,42	0,83	1,4	1,6	1,1	1,3
Ude-4b								0,52	0,98	1,5	1,1	1,2	1,3
Ude-5a						0,97	0,63	1,1	1,8	1,1	1,2	1,5	1,7
Ude-5b							0,53	1,2	1,8	1,1	1,3	1,6	1,8
Ude-1a	2,0	3,3	2,7	5,1	18	12	4,4	6,4	6,3	5,9	9,7	9,2	32
Ude-1b							4,3	6,7	8,7	6,5	8,3	11	28
C9-aromatics (µg/m³)													
Indoor median	3,9	2,0	2,1	0,53	3,0		1,7	1,7	1,4				2,9
Inde-a	3,9	2,0	2,1	0,53	3,0		1,4	1,8	1,4				2,8
Inde-b							2,0	1,6					3,0
Outdoor median								0,46	0,69	0,67	0,39	0,64	0,69
Ude-2a								0,41	0,54	0,41			0,63
Ude-2b								0,44	0,59	0,67	0,39		0,72
Ude-3a								0,53	0,72	0,61	0,38		0,76
Ude-3b								0,53	0,78	0,70	0,41		0,69
Ude-4a								0,36	0,63	0,81	0,33	0,64	0,70
Ude-4b								0,48	0,66	0,52	0,36	0,79	0,64
Ude-5a								0,45	0,74	0,67	0,43	0,64	0,61
Ude-5b								0,46	0,78	0,74	0,38		0,69
Ude-1a	0,65	1,1	0,81	0,81	4,4	2,9	1,1	1,7	1,8	2,7	3,2	7,2	

Ude-1b							1,1	1,6	1,5	1,9	2,3	3,7	6,6
C10-aromatics (µg/m³)													
Indoor median	1,4	1,2	0,87	0,51	0,88	0,69	0,77	0,90	0,78	0,84	0,93	0,66	1,7
Inde-a	1,4	1,2	0,87	0,51	0,88	0,69	0,64	0,92	0,71	0,84	0,91	0,62	1,6
Inde-b							0,89	0,87	0,85	0,84	0,94	0,70	1,7
Outdoor median							0,10	0,091	0,18	0,29	0,19	0,13	0,25
Ude-2a							0,091	0,15	0,32	0,11		0,19	0,21
Ude-2b							0,091	0,14	0,35	0,17	0,13	0,21	0,23
Ude-3a						0,10	0,091	0,21	0,23	0,17	0,12	0,11	0,24
Ude-3b							0,091	0,22	0,22	0,20	0,11	0,13	0,25
Ude-4a						0,10	0,091	0,17	0,24	0,24	0,15	0,18	0,30
Ude-4b							0,091	0,20	0,26	0,12	0,17	0,22	0,27
Ude-5a						0,10	0,091	0,17	0,31	0,20	0,12	0,19	0,21
Ude-5b							0,091	0,18	0,30	0,21	0,12	0,17	0,23
Ude-1a	0,13	0,10	0,12	0,30	0,46	0,35	0,097	0,33	0,85	0,33	0,29	0,42	0,92
Ude-1b							0,096	0,30	0,32	0,34	0,22	0,72	0,97

TABLE A3.7. Raw Data – Chimney Study. **Blue:** under the detection limit, **red:** above the regulatory limit.

Residence B - Chimney								
Regulatory Limit		100	0,13	400		100		40
Sample	Position	TVOC	Benzene	Toluene	Xylenes	C9	C10	Naphthalene
µg/m ³								
Nov-16								
B-st.-2a	East	370	220	63	25			31
B-st.-2b	West	280	160	46	19		1,3	24
Jan-17								
B-st1-Ø-a	East	62	2,9	1,5	0,80	0,46	0,17	0,63
B-st1-Ø-b	East	62	1,7	0,82	0,41	0,14	0,17	0,63
B-st2-V-a	West	880	470	120	40			87
B-st2-V-b	West	660	430	110	38			78
Mar-17								
B-st-1a	West	190	29	13	6,8	2,9	0,53	2,1
B-st-1b	West	190	21	11	6,2	3,1	0,96	2,0
B-st-2a	East	360	120	38	15	4,3	0,62	8,5
B-st-2b	East	310	130	42	17	4,6	0,73	8,6
May-17								
B-st-1a	East	92	81	25	11		0,73	11
B-st-1b	East	110	150	51	22		1,2	11
B-st-2a	West	86	41	12	5,7	1,7	0,56	3,4
B-st-2b	West	86	27	8,9	4,2	1,3	0,51	2,0
Jun-17								
B-st-1	East	59	0,12	2,7	2,1	0,79	0,27	0,6
B-st-1	East	59	0,12	2,7	2,1	0,79	0,27	0,6
Sep-17								
B-st-1a	East	52	0,067	2,1	1,26	0,53	0,19	0,53
B-st-1b	East	52	0,0093	1,1	0,48	0,17	0,14	0,53
B-st-2a	West	52	4,1	1,6	0,87	0,29	0,14	0,53
B-st-2b	West	52	0,013	2,1	1,33	0,58	0,23	0,53
B-st-1a	East	52	0,067	2,1	1,26	0,53	0,19	0,53

TABLE A3.8. Raw Data – Coastal, Wind Turbine and Fir study. **Blue:** under the detection limit, **red:** above the regulatory limit.

Coastal Study								
Regulatory limit		100	0,13	400	100	100	40	
Name	Position	TVOC	Benzene	Toluene	Xylenes	C9	C10	Naphthalene
$\mu\text{g}/\text{m}^3$								
Vest1	West	43	0,0077	0,43	0,12		0,12	0,44
Vest2	West	43	0,17	0,43	0,22		0,12	0,44
Vest3	West	43	0,18	0,43	0,24		0,12	0,44
Vest4	West	43	0,20	0,43	0,26		0,12	0,44
Vest5	West	43	0,073	0,43	0,17		0,12	0,44
Vest6	West	43	0,0077	0,43	0,099		0,12	0,44
Vest7	West	43	0,12	0,43	0,15		0,12	0,44
Vest8	West	43	0,11	0,43	0,19		0,12	0,44
Vest9	West	43	0,18	0,43	0,21		0,12	0,44
Vest10	West	43	0,035	0,43	0,11	0,10	0,12	0,44
Vest11	West	43	0,22	0,43	0,099		0,12	0,44
VestA	West	43	0,0077	0,43	0,099	0,10	0,12	0,44
Øst1	East	43	0,41	0,43	0,21		0,12	0,44
Øst2	East	43	0,24	0,43	0,23		0,12	0,44
Øst3	East	43	0,28	0,43	0,24		0,12	0,44
Øst4	East	43	0,20	0,43	0,40		0,12	0,44
Øst5	East	43	0,017	0,43	0,15	0,10	0,12	0,44
Øst6	East	43	0,27	0,43	0,38		0,12	0,44
Øst7	East	43	0,17	0,43	0,24		0,12	0,44
Øst8	East	43	0,26	0,43	0,26		0,12	0,44
Øst9	East	43	0,10	0,43	0,11	0,10	0,12	0,44
Øst10	East	43	0,052	0,43	0,13		0,12	0,44
Øst11	East	43	0,22	0,43	0,17		0,12	0,44
Øst12	East	43	0,0077	0,43	0,099	0,10	0,12	0,44
ØstA	East	43	0,0077	0,43	0,099		0,12	0,44
Wind Turbine Study								
Vind-2	Mill 07 - 9 m DVR	36	0,20	0,36	0,14		0,096	0,37
Vind-4	Mill 74 - 71 m DVR	43	0,48	0,73	0,51		0,11	0,44
Vind-5	Mill 74 - 9 m DVR	43	0,38	0,53	0,24		0,11	0,44
Vind-6	Mill 02 - 71 m DVR	43	0,54	0,69	0,89		0,11	0,43
Vind-8	Mill 02 - 9 m DVR	43	0,45	0,66	1,1		0,11	0,43
Fir Study								
GranA		190.000	0,74	310	21		1.700	
GranB		210.000	0,73	310	21		1.700	
Kontrol	Office	150	0,056	0,84	0,82			0,46

TABLE A3.9. Raw Data – Pre-study (sampler deployment). **Blue:** under the detection limit, **red:** above the regulatory limit.

Pre-study on sampler deployment						
Reg. Limit	100	0.13	400	100	100	
Sample	TVOC	Benzene	Toluene	Xylenes	C9	C10
			$\mu\text{g}/\text{m}^3$			
A1	36	0,21	0,81	1,1	0,25	0,29
A2	36	0,23	0,89	1,2	0,25	0,29
A3	36	0,27	1,2	1,3	0,25	0,29
A4	36	0,22	0,82	1,1	0,25	0,29
A5	36	0,17	0,93	1,1	0,25	0,29
A6	36	0,32	1,6	1,5	0,25	0,29
B1	35	0,31	5,3	1,9	0,51	0,28
B2	35	0,31	5,9	1,8	0,52	0,28
B3	35	0,41	10,0	2,5	0,69	0,28
B4	35	0,42	8,9	2,2	0,69	0,28
B5	35	0,43	14	2,9	0,75	0,28
B6	35	0,40	12	2,7	0,78	0,28
C1	36	0,23	0,73	0,59	0,25	0,28
C2	36	0,23	0,69	0,58	0,25	0,28
C3	36	0,18	0,63	0,52	0,25	0,28
C4	36	0,26	0,78	0,64	0,25	0,28
C5	36	0,21	0,55	0,44	0,25	0,28
C6	36	0,19	0,63	0,54	0,25	0,28
D1	36	0,22	0,60	0,54	0,25	0,28
D2	36	0,21	0,53	0,51	0,25	0,28
D3	36	0,20	0,46	0,33	0,25	0,28
D4	36	0,22	0,52	0,49	0,25	0,28
D5	36	0,20	0,51	0,50	0,25	0,28
D6	36	0,19	0,52	0,48	0,25	0,28
E1	43	0,23	1,5	1,1	0,30	0,35
E2	43	0,29	1,9	1,3	0,30	0,35
E3	43	0,47	4,3	3,3	0,48	0,35
E4	43	0,42	2,9	2,1	0,30	0,35
E5	43	0,34	2,7	1,9	0,30	0,35
E6	43	0,80	7,1	5,2	0,62	0,35
A7	36	0,063	0,36	0,081	0,25	0,29
B7	35	0,061	0,35	0,079	0,24	0,28
C7	36	0,063	0,36	0,081	0,25	0,28
D7	36	0,063	0,36	0,081	0,25	0,28
E7	43	0,077	0,43	0,099	0,30	0,35
A7	36	0,21	0,81	1,1	0,25	0,29

Appendix 4. Basic data treatment

In the following section, the individual steps involved in the statistical data treatment of the results are presented.

Appendix 4.1 Data categorization

Data are categorized according to the answers to the questionnaires. For example, in the study where the effect of wood-burning stoves is investigated, there are two categories: residences with wood-burning stoves and residences without wood-burning stoves. All residences are put in one of the categories, and the specific effect of wood-burning stoves on the composition of contaminants in the indoor air can be investigated, e.g. the indoor benzene levels.

Appendix 4.2 Basic statistical data handling

The basic data examination is done by visual inspection of the empirical distribution function. Data from each group are ranked in ascending order and plotted against their empirical distribution function according to the following formula:

$$F_N(X)(\%) = \frac{i}{N}$$

Where i is the number of the observation after rank ordering, N is the number of the measurements, and $F_N(X)$ is the empirical distribution function, i.e. the percent of data that are less or equal to the value of the observation i .

An example of the empirical distribution function of the indoor benzene concentrations in the May 2016 measurement campaign is shown in the figure below, together with a box-and-whiskers plot for the same data.

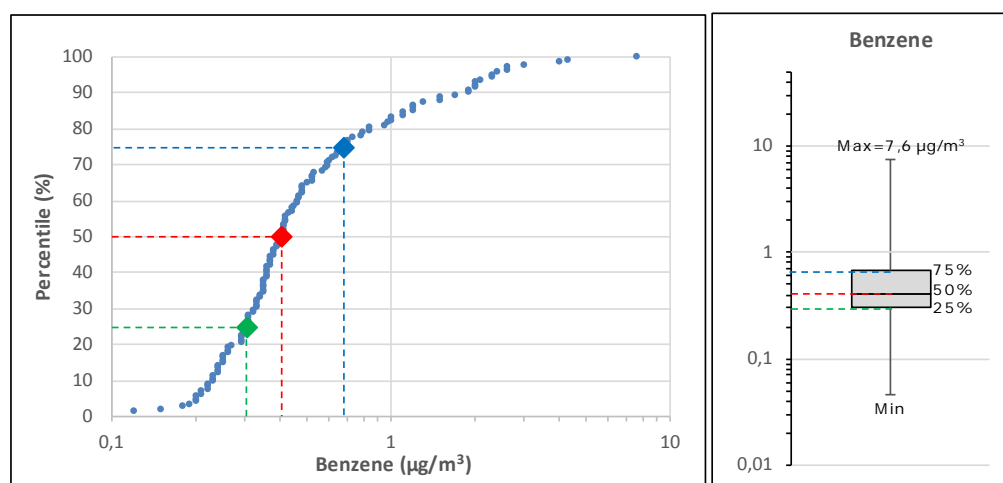


FIGURE A4.1. Empirical distribution function (left) of indoor benzene concentrations (log scale) in May 2016 and a box-and-whiskers plot for the same data (right).

The green point (left figure) and lines correspond to the 25th percentile (0,30 µg/m³), the red point (left figure) and lines mark the median (50th percentile) of the measurements (0,41 µg/m³), and the blue point (left figure) and lines mark 75th percentile (0,68 µg/m³).

The endpoints of an 80% confidence interval – meaning that 80 of the values fall within this interval – for the indoor benzene levels (May 2016) can be derived as the 10th percentile (0,23 µg/m³) and the 90th percentile (1,9 µg/m³).

Appendix 4.3 Further statistical data handling

Based on the empirical distribution function of the measurements and the answers to the questionnaire, visual/qualitative interpretation of the effects due to the tested parameters can be achieved.

For example, in the investigation of the effect of wood-burning stoves on the indoor benzene concentration (cf. section 5.3) in the November 2015 measurement campaign, the empirical distribution functions for residences with and without wood-burning stoves are plotted in the same graph, for a visual comparison of the results.

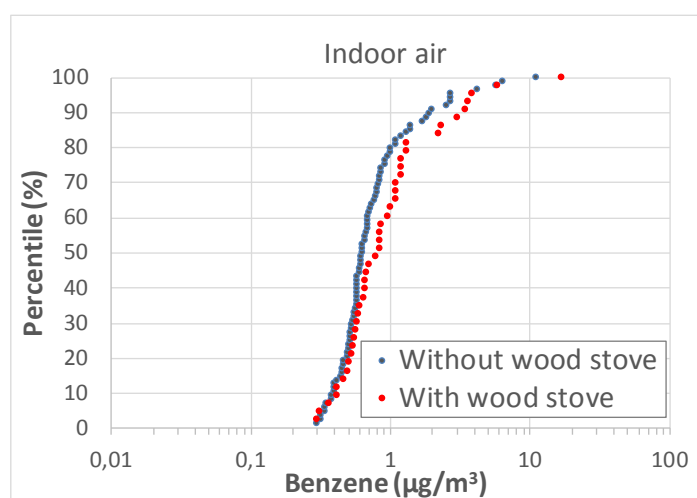


FIGURE A4.2. Empirical distribution functions of indoor benzene concentrations (log scale) in November 2015 for residences with (red) and without (blue) wood-burning stoves.

Based on the graph, there is a tendency for higher benzene concentrations in residences with wood-burning stoves. This can be seen from the fact that the red distribution function (residences with wood-burning stoves) is placed to the right of the blue distribution function (residences without wood-burning stoves).

From the graph, it can also be seen that the median values for the two distributions are close; i.e. the benzene concentrations (on the x-axis) corresponding the 50th percentile, are approx. the same for the two data-sets, with median values (50th percentiles) of 0,63 and 0,83 µg/m³, respectively.

In the above case, there is a clear tendency for the residences with wood-burning stoves to have higher indoor benzene concentrations in the November 2015 measuring campaign. With only a slight difference in median values, the difference is not that certain, but the trend can be tested statistically to determine if the difference is significant.

Appendix 4.4 Mann-Whitney test for equality of distributions

As seen above, wood-burning stoves seem to have an effect on the indoor benzene concentration. There is a tendency, but it is not entirely clear if the tendency is statistically significant. In such cases, the Mann-Whitney test for statistical comparison of the equality of two distributions can be a helpful tool.

The Mann-Whitney test is a rank based, non-parametric test that does not assume anything about the underlying data distribution. Therefore the data do not have to conform to a normal distribution, or any other well defined distribution. Another advantage of the Mann-Whitney test is that the two sets of data to be compared do not need to have the same number of observations (data points).

Basically, the test compares the empirical distribution functions for the two data-sets (as in figure A4.2), and one can decide if it is more likely that the two samples come from the same underlying distribution, or from two different distributions. In the latter case, the test can also tell us which distribution tends to be associated with higher values. We assign the "name" X to one of the distributions (e.g. residences with wood-burning stoves) and the "name" Y to the other (i.e. residences without wood-burning stoves).

We then apply the one-sided Mann-Whitney test, testing the hypothesis:

$H_0: F_N(X) = F_N(Y)$ – *the values in distribution X and Y tend to be the same*

Against the two alternative hypotheses:

$H_1: F_N(X) > F_N(Y)$ – *the values in distribution Y tend to be higher than in distribution X*

$H_2: F_N(X) < F_N(Y)$ – *the values in distribution X tend to be higher than in distribution Y*

The interpretation of the two alternative hypotheses (in italics) might seem counter intuitive. But as can be seen in figure A4.2, the empirical distribution functions demonstrate that if $F_N(Y)$ (without wood-burning stoves) tend to be higher than $F_N(X)$ (with wood-burning stoves) at a given benzene concentration (e.g. $1 \mu\text{g}/\text{m}^3$), this actually means that the Y-values tend to be lower.

For an actual walk-through of the test procedure, see /31/. Suffice it to say that the test is based on the fact that if the two data sets are mixed together, and ranked from the lowest to the highest number while keeping track of which values came from which data set to begin with, then the sum of the ranks for the X and Y data sets should be approx. the same for the two data sets if the distributions are likely to come from the same underlying distribution. If the sum of the ranks for distribution X is (significantly) higher than for distribution Y, then the values in data set X tend to be higher than in dataset Y.

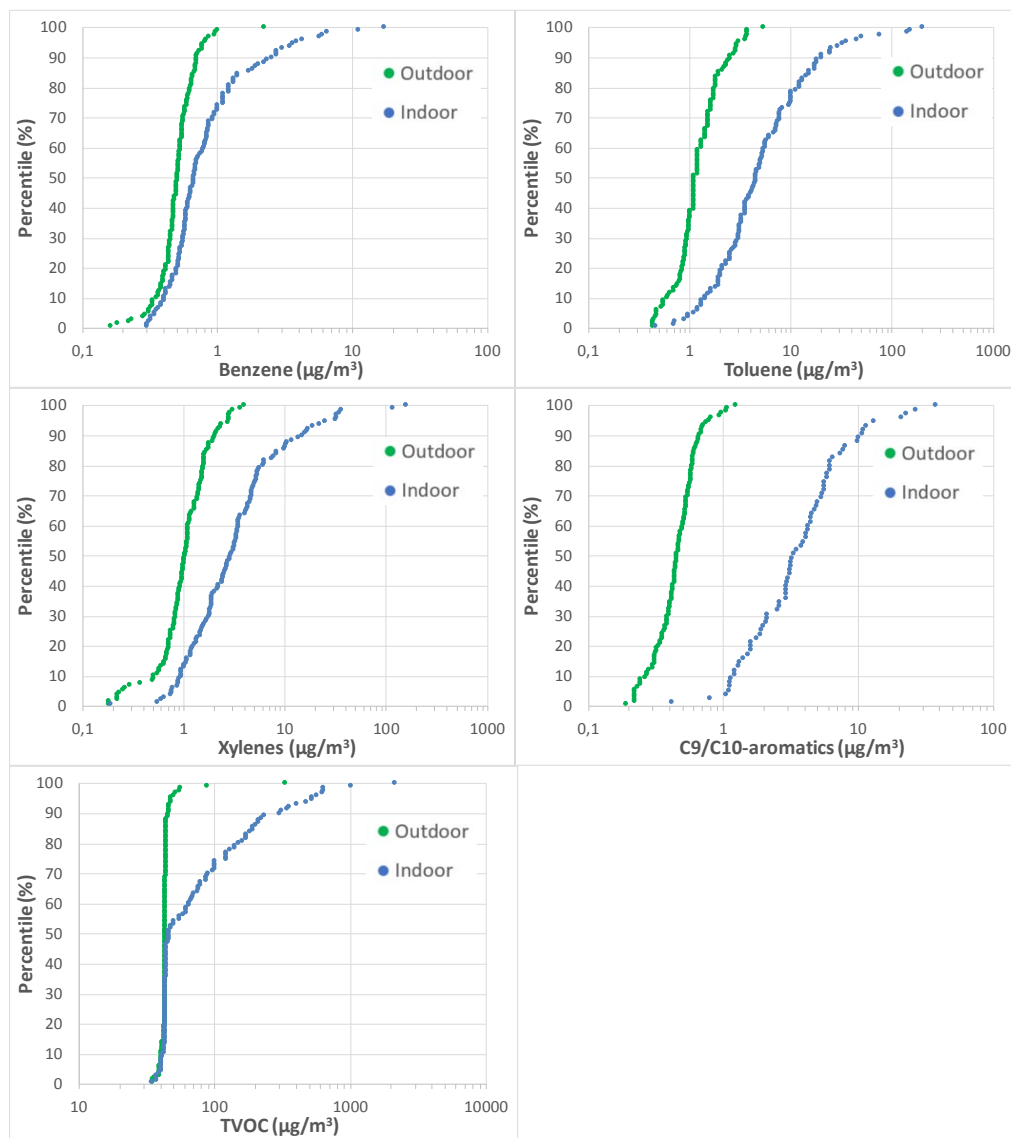
Throughout this project, the Mann-Whitney test is applied at the 5% significance levels. This means that we accept a 5% error of accepting H_0 (that the samples are from the same underlying distribution) even though it might be wrong. This is specified by the so-called alpha-value. In this case we apply $\alpha = 0,05$.

The Mann-Whitney allows us to calculate the p-value for each of the alternative hypotheses (H_1 and H_2) and tells us what the actual risk is of accepting a false hypothesis. A p-value of 0,03 means that there is a 3% risk of accepting the hypothesis, even though it might be false. Since we have decided on an alpha value of 0,05 and the p-value is $< \alpha$, we already have decided to accept the hypothesis. If the p-value is 0,07, we will not accept the hypothesis (the p-value $> \alpha$), since the risk of accepting a false hypothesis is defined as unacceptable for investigation in question.

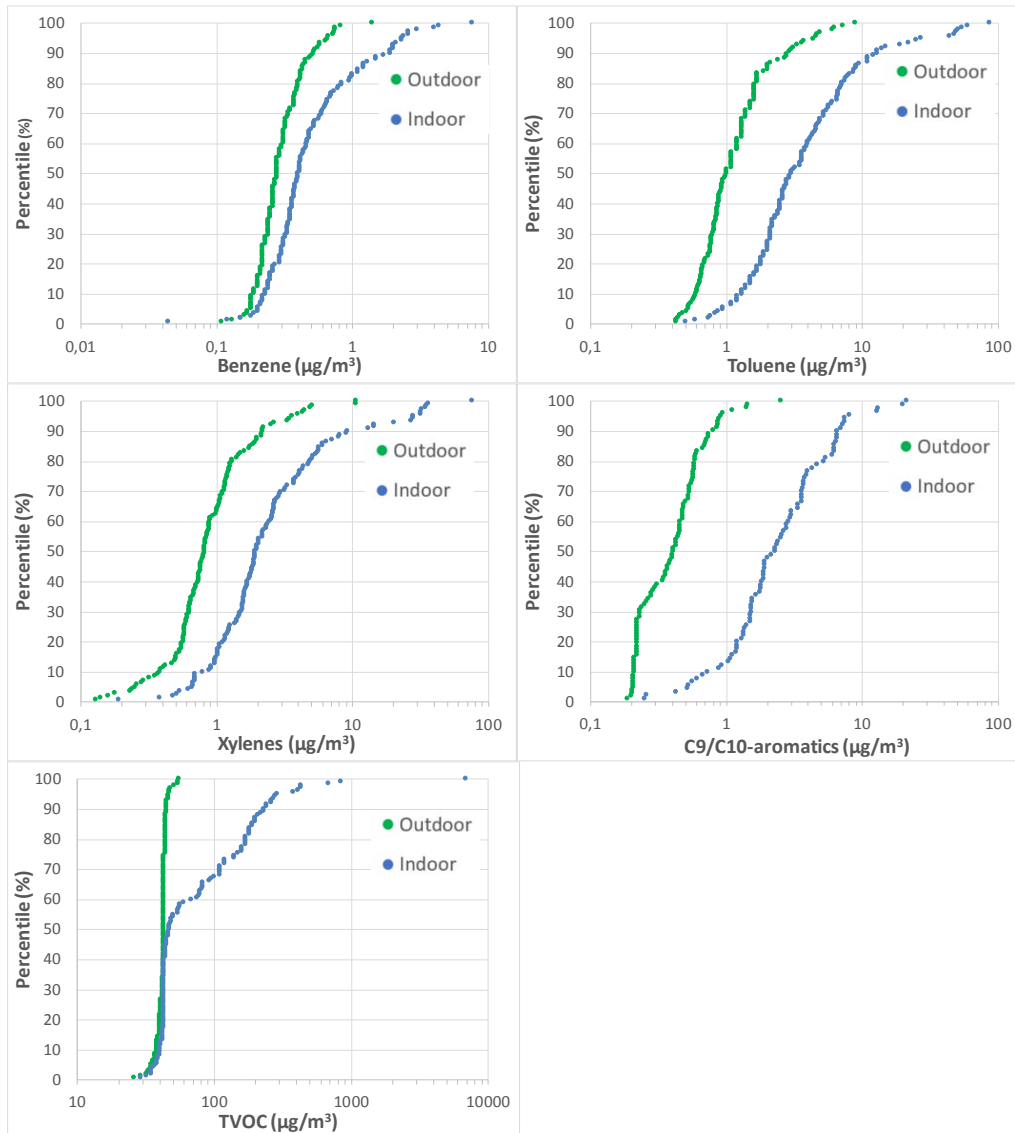
In the case of the wood-burning stove data in figure A4.2, the p-value of the Mann-Whitney test for H_2 (that residences wood-burning stoves have a higher indoor benzene concentration) is 0,058. Based on a decision on an alpha value of 0,05, we will reject the hypothesis that homes with wood-burning stove have higher indoor benzene concentrations, since the risk of the hypothesis being wrong is more than 5% (although quite close at 5,8%).

Appendix 5. Distribution functions

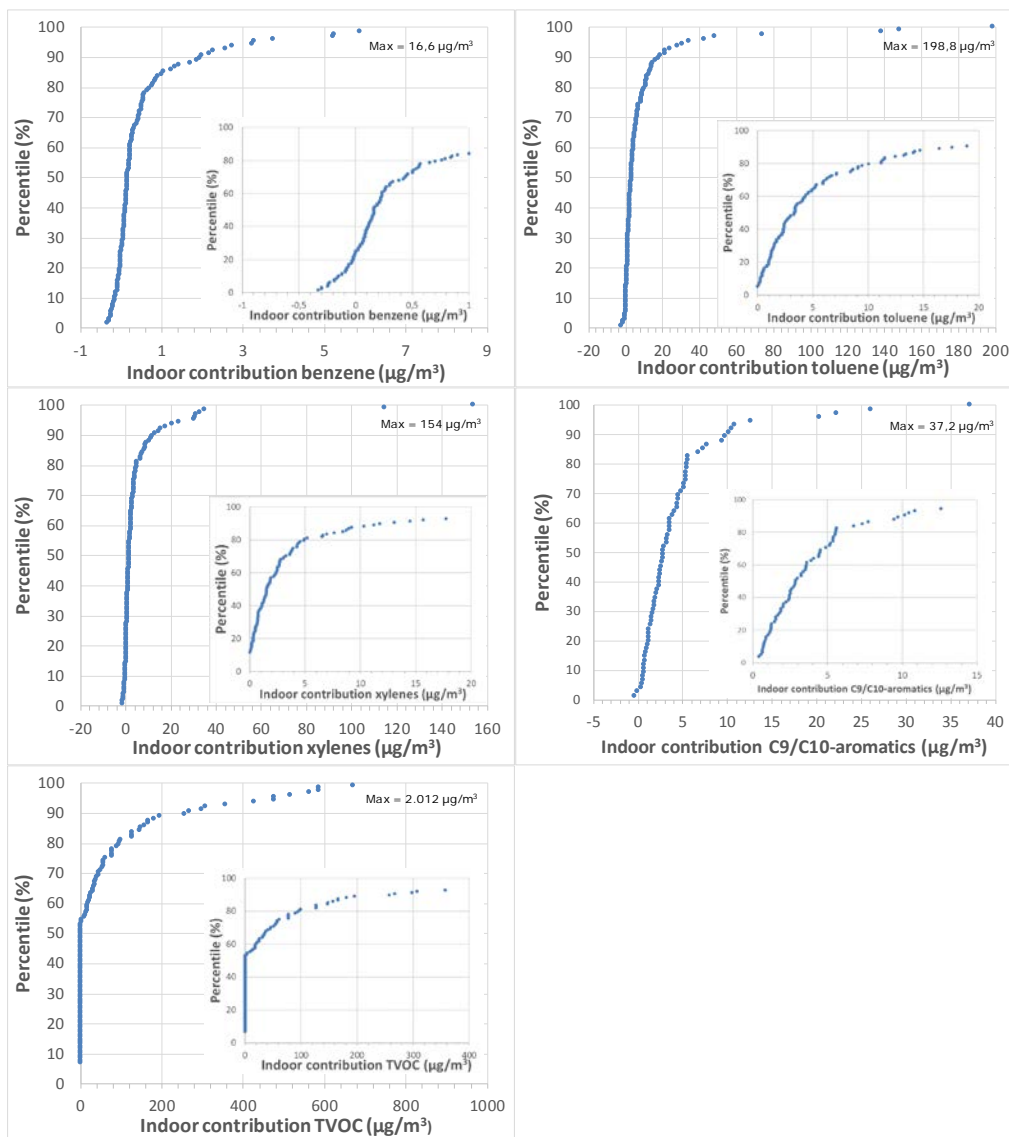
Appendix 5.1 General VOC levels, November 2015



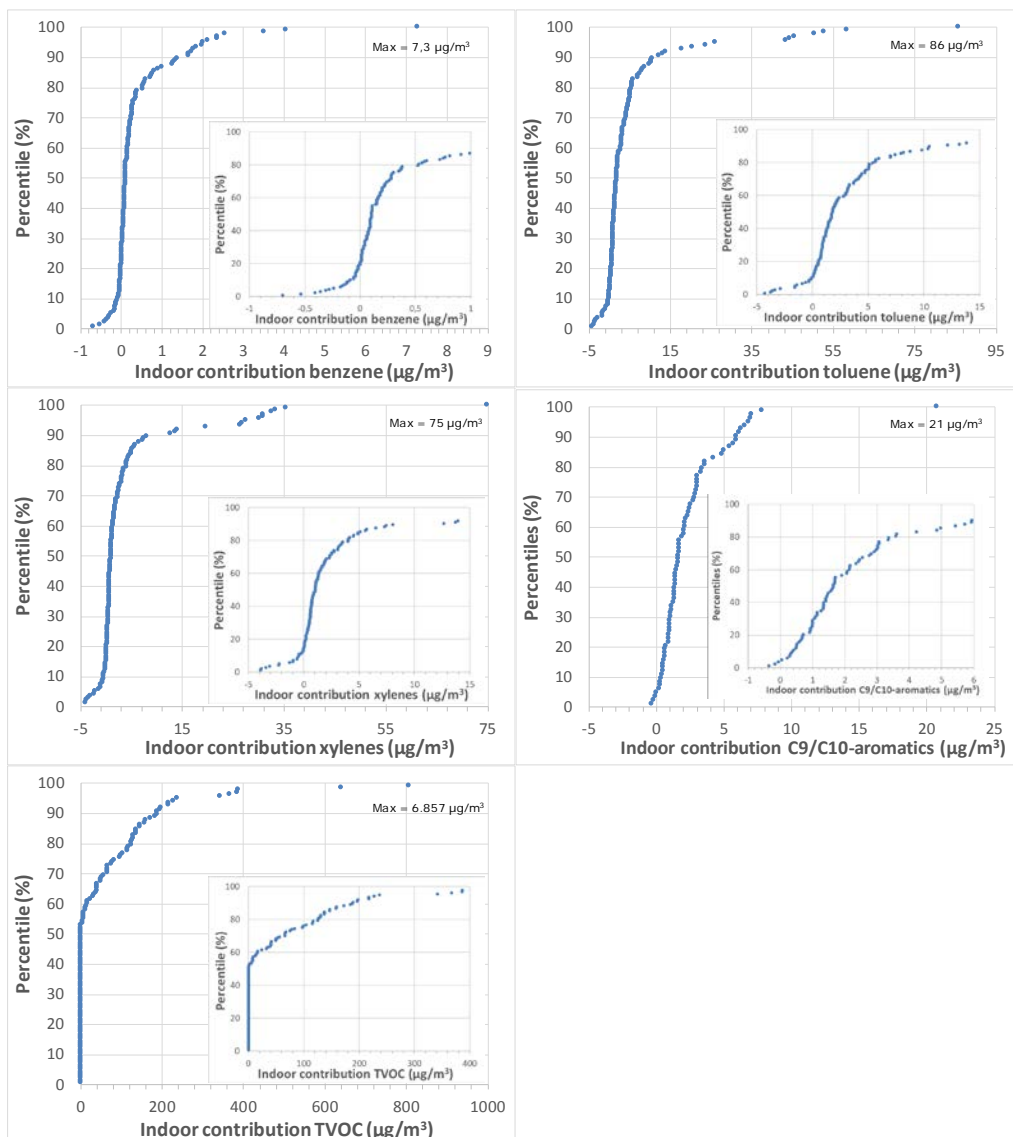
Appendix 5.2 General VOC levels, May 2016



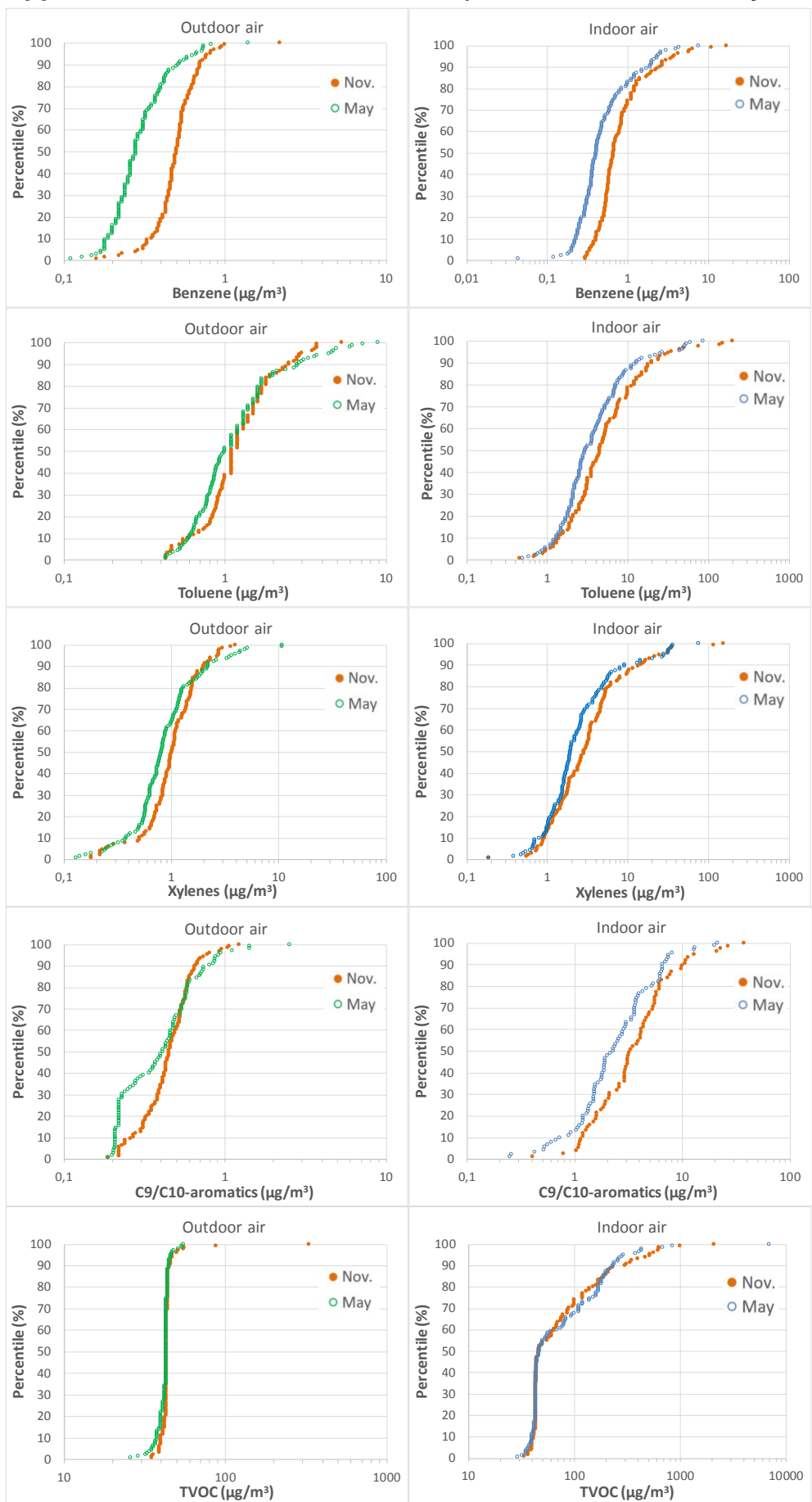
Appendix 5.3 Indoor internal contribution, November 2015



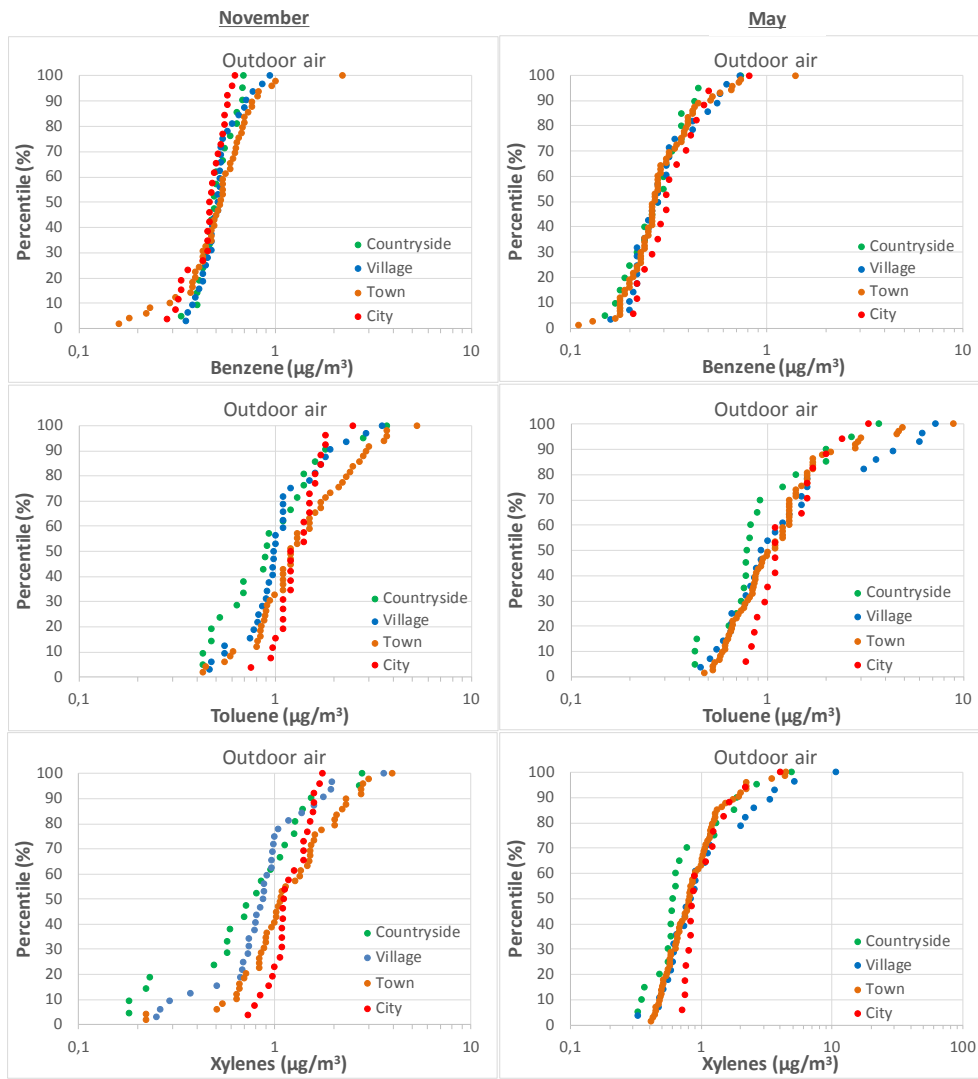
Appendix 5.4 Indoor internal contribution, May 2016



Appendix 5.5 Seasonal variations (November 2015 vs. May 2016)



Appendix 5.6 Population density



Appendix 5.7 Study of variability



At sites with a subsurface petroleum hydrocarbon contamination, many of the specific compounds detected in the indoor air can originate both from outdoor air, internal sources in the building and from the contaminated subsurface through vapor intrusion (VI). Hence, VI studies at hydrocarbon contaminated sites are challenged by the task of distinguishing between the contribution from ambient sources, and the contribution from vapor intrusion.

The objective of this study was to address a need for well-documented Danish background/ambient levels of hydrocarbon VOCs in outdoor and indoor air in residences, which can be used for VI studies.

It was concluded that background levels of benzene and TVOC have the potential to interfere with vapor intrusion studies, while background levels of other compounds are not expected to interfere with vapor intrusion studies.

A median value for the background indoor concentration of benzene was calculated to $0,66 \mu\text{g}/\text{m}^3$ in November and $0,41 \mu\text{g}/\text{m}^3$ in May. The corresponding value for outdoor air was calculated to $0,50 \mu\text{g}/\text{m}^3$ in November and $0,28 \mu\text{g}/\text{m}^3$ in May. Both outdoor and indoor background concentrations of benzene exceed the Danish regulatory limit for ($0,13 \mu\text{g}/\text{m}^3$) in >99% of the residences.

The indoor background concentration of TVOC can be expected to exceed the Danish regulatory limit ($100 \mu\text{g}/\text{m}^3$) in about 30% of uncontaminated Danish residences, but the general background concentration of TVOC in outdoor air cannot be expected to exceed the regulatory limit.



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